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Conséquences de l'introduction de la méthanisation dans une exploitation de polyculture-élevage sur les cycles du carbone et de l'azote. Combinaison de l'expérimentation et de la modélisation à l'échelle de la ferme

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Thèse de doctorat de l'université Paris-Saclay

École doctorale n° 581 : agriculture, alimentation, biologie, environnement et santé (ABIES)
Spécialité de doctorat: Sciences de l'environnement
Unité de recherche : Université Paris-Saclay, INRAE, AgroParisTech, UMR ECOSYS, 78850, Thiverval-Grignon, France
Référent : AgroParisTech

Thèse présentée et soutenue à Paris-Saclay, le 18/11/2021, par

Victor MOINARD

Composition du Jury

Yves COQUET Professeur, AgroParisTech (Université Paris-Saclay)	Président
David FANGUEIRO Assistant Professor (eq. HDR), Université de Lisbonne (Portugal)	Rapporteur & Examinateur
Safya MENASSERI-AUBRY Maîtresse de Conférences (HDR), Institut Agro-AgroCampus Ouest	Rapporteur & Examinatrice
Anne TREMIER Directrice de Recherche, INRAE Bretagne Normandie	Examinatrice

Direction de la thèse

Sabine HOUOT Directrice de Recherche, INRAE IdF-Versailles-Grignon	Directrice de thèse
Jean-Marie PAILLAT Chercheur (HDR), CIRAD – ISTOM Angers	Co-Directeur & Examinateur
Florent LEVAVASSEUR Ingénieur de recherche, INRAE IdF-Versailles-Grignon	Invité

Thèse de doctorat

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Consequences of the anaerobic co-digestion of cattle effluents in a crop livestock farm on C and N cycles. Combination of experimentation and of modelling at the farm scale.

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Résumé

La méthanisation agricole des effluents animaux est une pratique en fort développement en France. Elle produit de l'énergie renouvelable (biogaz). La valorisation des digestats au champ, comme celle des effluents non méthanisés, permet le retour au sol de nutriments et de matière organique, ce qui diminue le besoin en engrais minéraux et entretient les stocks de C des sols. Le traitement et l'épandage de ces produits peut aussi induire l'émission de gaz à effet de serre et de contaminants. La méthanisation agricole influence ces impacts : pour les maîtriser, il faut comprendre comment la digestion des effluents avec des déchets importés modifie les cycles du C et du N à l'échelle de la ferme. Cette question a été traitée en s'appuyant sur un cas d'étude à l'INRAE de Nouzilly (Centre – Val de Loire) : une exploitation agricole avec un méthaniseur traitant les effluents de son élevage bovin et divers déchets organiques. Lors de l'essai au champ MétaMétha, nous avons comparé les flux d'azote au cours d'une rotation culturale fertilisée avec des engrais minéraux, des lisiers et fumiers bovins, ou des digestats issus de ces effluents. Les digestats se substituent bien aux engrais minéraux, mais ils sont sensibles à la volatilisation d'ammoniac (NH_3). Les vers de terre peuvent être négativement impactés juste après l'épandage de digestat ou de lisier, mais les effets sont similairement positifs après 2 ans d'apports de matière organique. Nous avons ensuite évalué les modèles STICS et SYS-Metha pour simuler respectivement l'essai au champ et le traitement des digestats. Ces modèles ont été couplés pour simuler les flux de C et N à l'échelle de la ferme. Avec de forts imports de déchets, la méthanisation favorise la substitution des engrais minéraux, le stockage de C dans les sols, mais aussi les émissions de NH_3. Ce travail permet de mieux évaluer les conséquences de l'introduction d'un méthaniseur dans une exploitation agricole et ainsi d'optimiser la filière.

Mots-clefs

Méthanisation ; Polyculture élevage ; Cycles du carbone et de l'azote ; Bilan environnemental ; Modélisation ; Vers de terre.

Abstract

The anaerobic digestion (AD) of animal effluents is strongly developing in France. It produces renewable energy (biogas). Like undigested effluents, the use of anaerobic digestates in the field enables the recycling of nutrients and organic matter in the soil, which decreases the need for synthetic fertilizers and maintains soil organic carbon stocks. However, the treatment and field application of those organic products can also cause greenhouse gas emissions and contaminants. The on-farm AD nfluences those impacts. In order to control them, we need to understand how the co-digestion of animal effluents with imported organic wastes influences C and N cycles at the farm scale. We studied this question with the support of a case study at INRAE in Nouzilly (Centre – Val de Loire, France): a crop and livestock farm where an anaerobic digester treats the cattle effluents together with imported wastes. During the MetaMetha field experiment, we compared N fluxes during a crop rotation that was fertilized with synthetic N fertilizers, undigested cattle slurry and farmyard manure, or digestates issued from their digestion. We found that digestates can substitute synthetic fertilizers, despite the risk of ammonia (NH_3) emissions. Earthworms can be negatively impacted just after the application of digestate of slurry, but the input of organic matter induced similar positive impacts after two years. We then evaluated the STICS and SYS-Metha models to simulate the field experiment, and digestate treatment and storage, respectively. Both models were coupled to simulate C and N fluxes at the farm scale. The models showed that when large amounts of digested wastes are imported, AD promotes substitution of synthetic N fertilizers and storage of soil organic C, but also NH_3 emissions. The study enabled us to better evaluate the consequences of the on-farm AD and therefore to optimize the sector.

Keywords

Anaerobic digestion; Crop-livestock farm; Carbon and nitrogen cycles; Environmental assessment; Modelling; Earthworms.

Liste des publications associées à cette thèse

Article scientifiques parus dans une revue à comité de lecture

Moinard, V., Redondi, C., Etiévant, V., Savoie, A., Duchene, D., Pelosi, C., Houot, S., Capowiez, Y., 2021. Short- and long-term impacts of anaerobic digestate spreading on earthworms in cropped soils. Applied Soil Ecology 168, 104149. https://doi.org/10.1016/j.apsoil.2021.104149

Bareha, Y., Affes, R., Moinard, V., Buffet, J., Girault, R., 2021. A simple mass balance tool to predict carbon and nitrogen fluxes in anaerobic digestion systems. Waste Management 135, 47–59. https://doi.org/10.1016/j.wasman.2021.08.020

Autre article scientifique paru dans une revue à comité de lecture

Moinard, V., Levavasseur, F., Houot, S., 2021. Current and potential recycling of exogenous organic matter as fertilizers and amendments in a French peri-urban territory. Resources, Conservation and Recycling 169, 11. https://doi.org/10.1016/j.resconrec.2021.105523

Posters et communications orales lors de congrès scientifiques français ou internationaux

Moinard, V., Redondi, C., Etiévant, V., Savoie, A., Houot, S., Pelosi, C., Capowiez, Y., 2019. Impacts à court et moyen terme de l'épandage de digestats de méthanisation en grandes cultures sur les populations de vers de terre. APIVALE 2019 (Approche Intégrée des filières de VALorisation des Effluents organiques), Rennes, France, 2019.

Savoie, A., Moinard, V., Pasquier, C., Houot, S., 2020. Impacts de l'introduction de la méthanisation à la ferme sur les bilans Carbone et Azote de la fertilisation des cultures : Résultats de 3 ans de mesures à Nouzilly en région Centre Val-de-Loire. JRI Biogaz méthanisation 2020, Toulouse, France, 2020.

Moinard, V., Levavasseur, F., Savoie, A., Pasquier, C., Génermont, S., Bareha, Y., Girault R., Paillat, J.-M., Houot, S., 2021. Evaluation of the impacts of anaerobic digestion on C and N cycles using an anaerobic digestion model coupled to a soil-crop model at the scale of a French crop-livestock farming system. 29th European Biomass Conference and Exhibition (EUBCE 2021), Marseilles, France, 2021.

Moinard, V., Savoie, A., Pasquier, C., Génermont, S., Goubard Delaunay, Y., Levavasseur, F., Paillat, J.-M., Houot, S., 2021. The integrated effects on C and N cycles of digestate use in arable cropping systems in France: a study coupling a three year field experiment and modelling. Eurosoil 2021, Genève, Suisse, 2021.

Table des matières

Financements 4
Remerciements 5
Résumé 8
Abstract 9
Liste des publications associées à cette thèse 10
Table des Matières 11
Introduction 16
I.1 La méthanisation: définitions 17
I.2 Diversité des procédés de méthanisation 17
I.3 Pourquoi la méthanisation ? 19
I.4 La méthanisation dans le monde, en Europe, et en France 19
I.5 Controverses liées à la méthanisation 22
I.6 Cadre de la thèse 23

Chapitre 1 Synthèse bibliographique : introduction de la méthanisation dans une ferme de polyculture élevage 25
1.1 Les digestats sont des Produits Résiduaires Organiques (PRO) 26
1.1.1 Les PRO : définitions et présentations 26
1.1.2 Étude de la valeur fertilisante des PRO 28
1.1.3 Étude de la valeur amendante des PRO 30
1.1.4 Les digestats parmi les PRO 31
1.2 Effet de l'épandage de digestat au champ à l'échelle de la parcelle 33
1.2.1 Digestat et fertilisation azotée 33
1.2.2 Digestat et volatilisation d'ammoniac 34
1.2.3 Effet des digestats sur la matière organique du sol 37
1.2.4 Effet des digestats sur la fertilité des sols à long terme 38
1.2.5 Effet des digestats sur la vie du sol 39
1.2.6 Digestat et émissions de gaz à effet de serre, dont le N_2O 39
1.2.7 Digestat et lixiviation de nitrate 41
1.3 Modification des flux de C et N à l'échelle de l'exploitation avec la méthanisation 42
1.3.1 Comment la méthanisation influe-t-elle sur les flux de C et N à l'échelle de l'exploitation ? 42
1.3.2 Bilans C et N à l'échelle de la ferme et effet de l'import de déchets extérieurs 43
1.3.3 Changement de pratiques agricoles liées à l'introduction d'un méthaniseur 44
1.3.4 Emissions gazeuses lors du stockage des PRO, émissions fugitives du méthaniseur 46
1.3.5 Intérêt de la prise en compte des émissions lors du stockage et de l'épandage des PRO 49
1.4 Evaluation environnementale de la méthanisation 50
1.4.1 Quelles limites ont les approches expérimentales lors de l'évaluation environnementale de la méthanisation ? 50
1.4.2 Les modèles : quelques définitions 51
1.4.3 Utilisation des modèles pour modéliser la digestion, le stockage et l'épandage de digestats 52
1.4.4 Les modèles permettent de synthétiser les impacts à l'échelle de l'exploitation agricole 53
1.4.5 Analyses de cycle de vie et méthanisation 57

1.5 Conclusion de la synthèse bibliographique et objectifs de la thèse 59
1.5.1 Conclusion de la synthèse bibliographique 59
1.5.2 Objectifs de la thèse 60

Chapitre 2 Présentation du site d'expérimentation à Nouzilly 65
2.1 Contexte régional 66
2.1.1 Localisation, agriculture et climat 66
2.1.2 La méthanisation en Centre Val-de-Loire 67
2.1.3 Enquête sur la méthanisation en Centre Val-de-Loire en 2016 67
2.1.4 Evolution de la méthanisation agricole en région Centre Val-de-Loire entre 2016 et 2020 68
2.2 Description du site de Nouzilly 69
2.2.1 Description générale 69
2.2.2 L'élevage 70
2.2.3 Le méthaniseur 70
2.2.4 L'exploitation agricole 72
2.3 Description du projet MétaMétha 72
2.3.1 Objectifs du projet 72
2.3.2 Présentation de l'essai agronomique 72
2.3.3 Caractérisation du sol sur l'essai agronomique 75
2.3.4 Itinéraire technique agricole 77
2.3.5 Suivi du site 82

Chapitre 3 Comment la méthanisation des effluents bovins modifie-t-elle le cycle de l'azote sur une rotation culturale de 3 ans ? 87
Abstract 89
3.1 Introduction 90
3.2 Material and Methods 92
3.2.1 Site and anaerobic digester 92
3.2.2 Field experiment design 93
3.2.3 Physico-chemical characteristics of the exogenous organic matters 94
3.2.4 Soil description 97
3.2.5 Field trial monitoring 97
3.2.6 Kinetics of C and N mineralization of EOMs during incubations in controlled conditions 99
3.2.7 Fertilizer N availability 100
3.2.8 Field N balance 102
3.2.9 Statistical analyses 103
3.3. Results 104
3.3.1 Characteristics of exogenous organic matters 104
3.3.2 Ammonia emissions after application of exogenous organic matters 107
3.3.3 Agronomical performances in the different treatments 109
3.3.4 Evolution of soil mineral N stocks during the 3-year experiment 112
3.3.5 N_2O emissions 113
3.3.6 Mineral N balance 118
3.3.7 Amendment properties of the EOMs 119
3.4. Discussion 120
3.4.1 Digestates have higher fertilizer properties and similar amendment properties than undigested animal effluents 120
3.4.2 Assessing the N balance in a field experiment: limits and strength 122
3.4.3 The use of exogenous organic matters influences N losses in the field 122
3.4.4 Digestates are able to substitute chemical fertilizers to grow crops in field conditions 124
3.5 Conclusion 125
Acknowledgment 126

Chapitre 4 Conséquences de la méthanisation des effluents d'élevage sur les flux d'azote lors d'une rotation culturale de 3 ans : évaluation du modèle sol-plante STICS 129
Abstract 130
4.1 Introduction 131
4.2. Material and Methods 133
4.2.1. 3-year field experiment and EOM characterization 133
4.2.2. Simulating the three-year crop sequence with STICS 136
4.2.3. Calibration of the STICS submodule modelling the C and N mineralization after application of EOMs 137
4.2.4. Simulation of ammonia volatilization after EOM application using STICS or ALFAM2 138
4.2.5 Evaluation of STICS soil-crop model 139
4.2.6 Sensitivity of STICS output to EOM parameters. 141
4.3 Results 142
4.3.1 Calibration and evaluation of STICS module for residue mineralization 142
4.3.2 Evaluation of volatilization submodule of STICS 145
4.3.3 STICS performances 146
4.3.4 STICS sensitivity to EOM parameters 151
4.3.5 Modelling nitrate leaching, N_2O emissions, and soil organic N storage during the crop rotation 153
4.4 Discussion 155
4.4.1 STICS performance to simulate EOM mineralization in controlled conditions 155
4.4.2 STICS performance to simulate EOM volatilization 156
4.4.3 STICS performance using organic fertilizers 157
4.4.4 Using STICS to simulate cropping system with organic fertilization 158
4.5 Conclusion 159
Acknowledgment 159

Chapitre 5 Évaluation du modèle de méthanisation SYS-Metha à l'aide des données opérationnelles d'un méthaniseur traitant des déchets variés 161
Abstract 162
5.1 Introduction 163
5.2 Material and Methods 165
5.2.1 Description of SYS-Metha, mass balance model of anaerobic digestion 165
5.2.2 Case study: an agricultural digester with high external inputs 167
5.2.3 Investigating periods of stationary functioning of the digester from operational data to evaluate the model 168
5.2.4 Mass balance of the digester based on the operational dataset 168
5.2.5 Evaluation of the SYS-Metha model to simulate biogas production, digestate production, and digestate characteristics 170
5.3. Results 171
5.3.1 Operational data: assessment of data consistency and selection of study period 171
5.3.2 Evaluation of SYS-Metha: fresh mass balance, nutrient flows, and digestate characteristics 175
5.3.3 Modelling gaseous emission during storage 180
5.4 Discussion 180
5.4.1 Mass balances of anaerobic digesters 180
5.4.2 Performances of SYS-Metha compared to other anaerobic digestion models 180
5.4.3. Further consideration: discussing the assumptions of the SYS-Metha model 181
5. Conclusion 184
Acknowledgement 184

Chapitre 6 La méthanisation des effluents d'élevage peut diminuer l'utilisation d'engrais de synthèse, augmenter le stockage de C dans les sols et augmenter la volatilisation d'ammoniac à l'échelle de la ferme : rôle clef de l'import de déchets et évaluation de différentes pratiques de gestion 187
Abstract 189
6.1 Introduction 190
6.2 Material and Methods 192
6.2.1 Description of the farm model 192
6.2.2 Case study and scenarios 197
6.2.3 Indicators for studying C and N fluxes at the farm scale 202
6.2.4 C footprint of anaerobic digestion with a consequential approach 202
6.3 Results 206
6.3.1 Agronomical use of EOM and biogas production 206
6.3.2 C fluxes in the farm with or without on-farm AD 208
6.3.3 N fluxes in the farm with or without on-farm AD 213
6.3.4 Influence of climatic year on C and N fluxes in the farm 216
6.3.5 Influence of external inputs on C and N fluxes in the farm 217
6.3.6 Influence of storage conditions and agronomical management of EOM on C and N fluxes in the farm 219
6.3.7 C footprint of diverse scenarios compared to "no AD" scenario in extending the system 222
6.4 Discussion 226
6.4.1 The input of external wastes is a major driver of the C and N fluxes in the farm 226
6.4.2 Quantifying the effect of good practices at the farm scale 228
6.4.3 C footprint of AD with a consequential approach 229
6.4.4 Uncertainties and variability of the outputs of the farm model 230
6.4.5 Assumptions of the model and perspectives 230
6.5. Conclusion 232
Acknowledgement 232

Chapitre 7 Effets à court et long termes de l'épandage de digestat en grande culture sur les vers de terre 235
Abstract 237
7.1 Introduction 238
7.2 Material and methods 240
7.2.1 Field and soil description 240
7.2.2 Fertilizers and chemical solutions used as treatments. 241
7.2.3 Earthworms 245
7.2.4 Experiment setup 245
7.2.5 Statistical analyses 249
7.3 Results 250
7.3.1 Long-term and short-term impacts on earthworms of applying organic products in the field 250
7.3.2 Short-term ecotoxicity of organic products in microcosms under controlled laboratory conditions 252
7.3.3 Microcosm experiment to investigate the causes of short-term ecotoxicity to L. terrestris 254
7.3.4 Short-term impact of organic products on earthworm behavior and survival in soil columns 255
7.4 Discussion 256
7.4.1 Long-term impact of digestates on earthworm populations 256
7.4.2 Comparison of short-term effects under field and laboratory conditions 258
7.4.3 Probable causes of liquid organic product toxicity in the short term 259
7.4.4 Earthworm behavior in the short term after organic product spreading 260

7.5 Conclusions 261
Acknowledgements 261

Chapitre 8 – Discussion générale et conclusion 263
8.1 Résumé des résultats majeurs de la thèse 265
8.2 Réflexions sur la démarche utilisée dans la thèse 270
8.3 Introduction d'un méthaniseur dans une exploitation agricole, quelles conséquences ? 273
8.4 Perspectives : quelle recherche pour mieux appréhender les enjeux de la méthanisation ? 275

Références 279

Annexes – Chapitre 3 : Supplementary Material 311
S. M. 1 Map of the experimental design 312
S. M. 2 Crop management 313
S. M. 3 Detailed methods: Agricultural performance of crops 314
S. M. 4 Detailed method: SMN analysis 315
S. M. 5 Detailed methodology: Incubations 315
S. M. 6 Individual analyses of cattle slurry 316
S. M. 7 Rapeseed growth from sowing to February 317

Annexes – Chapitre 4 : Supplementary Material 319
S. M. 1 Parameters and options used in STICS 320
S. M. 2 Calibration of EOM mineralization submodule of STICS 320
S. M. 3 STICS submodule for ammonia volatilization after spreading of EOM. 322
S. M. 4 Evaluation of the performance of STICS and ALFAM2 model to predict volatilization 324
S. M. 5 Evaluation of STICS in the reference parametrization (NH3_ALFAM2) and (EOM_cal) 325
S. M. 6 Analysis of sensitivity of STICS outputs induced by the use of different EOM mineralization submodules 330
S. M. 7 Analysis of sensitivity of STICS outputs induced by the use of different EOM volatilization submodules 332
S. M. 8 Analysis of sensitivity of STICS outputs induced by the variation of EOM characteristics (winter/spring) 334
S. M. 9 Analysis of sensitivity of STICS outputs induced by the variation of EOM characteristics (summer) 340

Annexes – Chapitre 6 : Supplementary Material 347
S. M. 1 Emission factors used to model EOM storage 348
S. M. 2 Parameters and options used in STICS 349
S. M. 3 Characteristics of climatic years 349
S. M. 3 Cropping system 350
S. M. 4 Hypothesis used for C footprint computing 354
S. M. 5 Modelling sewage sludge application at the farm scale 357
S. M. 6 Modelled fertilization on each crop 359
S. M. 7 Repartition of the use of EOMs in the different fields 364
S. M. 8 Yield evolution 366
S. M. 9 Evolution of indicators related to C and N cycle through the time depending on the used climatic year 370
S. M. 10 Indicators of reference scenarios, depending on climatic year 371
S. M. 11 Indicators for all scenario, on reference climatic year 375

Annexes – Chapitre 7 : Supplementary Material 379
S. M. 1 Treatments in the microcosm ecotoxicity tests, series #1 (2019) and #2 (2020) 380
S. M. 2 Biomass of earthworms in the field 382
S. M. 3 Weight change in microcosm ecotoxicity tests, series #1 (2019) and #2 (2020) 383
S. M. 4 Weight change in macrocosm experiments 385

Introduction

I.1 La méthanisation : définitions

La **méthanisation** correspond à la dégradation en conditions anaérobies de substrats organiques (déchets organiques, résidus de cultures, cultures dédiées) par des microorganismes, ce qui conduit à la production de **biogaz**, un mélange principalement composé de dioxyde de carbone (CO_2), de méthane (CH_4), et de vapeur d'eau (H_2O). Le **biométhane** extrait de ce biogaz peut être utilisé comme source d'énergie renouvelable. La matière résiduaire obtenue après la digestion est appelée **digestat**. C'est un substrat organique contenant tous les nutriments présents dans les intrants, et qui peut être valorisé de plusieurs façons, incluant sa valorisation en agriculture en tant que fertilisant ou amendement organique (section 1.1). La plateforme où se déroule la méthanisation est un **méthaniseur** (Moletta, 2015).

I.2 Diversité des procédés de méthanisation et des digestats

Il existe une grande diversité de types de méthanisation, ne répondant pas aux mêmes contraintes locales, ni aux mêmes enjeux. Cette section introduit les facteurs de variations entre les systèmes de méthanisation : la diversité des intrants, des tailles de digesteurs, des procédés de méthanisation, des types de valorisation du biogaz, et des modalités de gestion des digestats.

Un méthaniseur se caractérise d'abord par ses intrants (également nommés substrats). Les premiers intrants pouvant être méthanisés sont des déchets ou sous-produits organiques, de nature très variée (European Biogas Association, 2020; Guilayn et al., 2019a; Moletta, 2015). Parmi les intrants classiques, on peut citer : les effluents d'élevage (lisiers, fumiers), les biodéchets issus des agglomérations (fraction fermentescible des ordures ménagères, autres biodéchets collectés sélectivement tels que les invendus des petites et moyennes surfaces), les boues de station d'épuration des eaux usées (STEU), divers déchets issus des industries agro-alimentaires (IAA) (boues d'IAA, graisses), des déchets agricoles (issus de silos). Il est aussi possible d'utiliser des cultures, qu'il s'agisse de cultures dédiées (maïs ensilage, par exemple), ou de cultures intermédiaires récoltées pour être méthanisées (cultures intermédiaires à vocation énergétique, dites CIVE). Souvent, les mélanges d'intrants sont optimisés pour augmenter la production de biogaz ou répondre à des contraintes techniques telles que la teneur en matière sèche, le rapport C/N, des limitations des éléments inhibiteurs ou toxiques comme les acides gras volatils ou l'ammoniac (Esposito et al., 2012; Iacovidou et al., 2012; Mata-Alvarez et al., 2014). Chaque intrant a des caractéristiques variables, qui vont influencer la production de biogaz et la nature du digestat. Ces intrants ont des teneurs variables en matière sèche (MS), matière organique (MO), et différents macro- et micronutriments (N, P, K, éléments traces métalliques - ETM). Les intrants sont également caractérisés par leur vitesse de dégradation et leur potentiel à être digéré et à produire du méthane en lien avec leurs caractéristiques biochimiques (Bareha et al., 2019; Fisgativa et al., 2018). Une analyse classique des intrants est le potentiel méthanogène (BMP - *biochemical methane potential*), qui

détermine la proportion de MO qui pourra être transformée en méthane dans des conditions normées. Le BMP est très utilisé pour caractériser les intrants (Filer et al., 2019; Koch et al., 2020; Owen et al., 1979). Le prétraitement des intrants peut permettre de changer leurs caractéristiques physico-chimiques et améliorer la production de biogaz (traitements mécaniques, physico-chimiques, biologiques) (Esposito et al., 2012; Fisgativa et al., 2020b; Mata-Alvarez et al., 2014).

En plus de la nature des intrants, les quantités d'intrants traités dans un méthaniseur peuvent être très variées. Il existe des méthaniseurs traitant quelques tonnes de déchets par an, et d'autres traitant plus de 100 000 t de déchets par an. Les petits méthaniseurs à l'échelle d'un foyer peuvent produire moins de 10 m^3 de biogaz par an, les méthaniseurs agricoles peuvent avoir une production de 200 à plus 1000 m^3 de biogaz, tandis que des méthaniseurs territoriaux peuvent produire jusqu'à 8 000 m^3 de biogaz par an (Vasco-Correa et al., 2018).

Un méthaniseur peut être caractérisé par sa température de fonctionnement. Une température élevée favorise la digestion : on parle de méthanisation mésophile (30 – 40°C), ou thermophile (45 – 60°C). De plus, plusieurs procédés existent pour réaliser la digestion anaérobie. Le procédé le plus courant en Europe pour la méthanisation de déchets agricoles est la voie humide infiniment mélangée avec approvisionnement en continu (*continuous stirred tank reactor CSTR*) : dans ce cas, le résidu dans le digesteur est liquide. Il existe aussi des procédés en voie sèche, c'est-à-dire avec mélange d'intrants solides, et où l'approvisionnement peut être continu (piston, vis sans fin) (*plug-flow reactor*), ou discontinu (apports par charges, *batch*). Enfin, la digestion peut avoir lieu dans un unique digesteur, ou dans plusieurs digesteurs successifs (Moletta, 2015).

Le biogaz peut être utilisé pour produire de l'électricité et est alors le plus souvent associé à une production de chaleur (**cogénération**). Le biogaz peut aussi être purifié en biométhane, pour être soit utilisé en tant que carburant ou être injecté dans les réseaux de gaz naturel (**injection**).

Enfin, le digestat peut être post-traité avant sa valorisation. Le digestat dit brut subit souvent une séparation de phase (par presse à vis, centrifugation par exemple), amenant à avoir à la fois un digestat liquide, pauvre en matière sèche et souvent riche en azote minéral, et un digestat solide, plus riche en matière organique (Guilayn et al., 2019b). Les digestats peuvent la plupart du temps être simplement valorisés et épandus au champ en tant que fertilisants. Les digestats peuvent aussi subir différents traitements (Cesaro, 2021; Logan and Visvanathan, 2019) visant à améliorer ou modifier leurs qualités fertilisantes (compostage, séchage, traitements thermochimiques : gazéification, pyrolyse, carbonisation hydrothermale). Les nutriments des digestats peuvent également être extraits pour produire des engrais concentrés par stripping et précipitation de struvites (Palakodeti et al., 2021; Vaneeckhaute et al., 2017). La bioraffinerie (c'est-à-dire l'extraction de molécules d'intérêt économique) et les utilisations comme support de culture pour des micro-algues ou éventuellement d'autres organismes (insectes par exemple) sont aussi envisagés, mais en cours de développement (Cesaro, 2021).

En conclusion, nous pouvons dire que la méthanisation est un terme qui englobe des procédés très divers. En conséquence, en fonction des intrants et procédés, les digestats peuvent avoir des caractéristiques physico-chimiques très différentes (Beggio et al., 2019; Guilayn et al., 2019a; Stürmer et al., 2020).

I.3 Pourquoi la méthanisation ?

L'agriculture est confrontée à plusieurs défis globaux. L'agriculture (incluant l'élevage), avec la gestion forestière et les changements d'usages des sols, contribue à hauteur de 24% aux émissions de gaz à effet de serre (GES), favorisant le changement climatique (IPCC, 2015). L'agriculture utilise de plus en plus d'engrais de synthèse ou issus de ressources minières, par exemple en ce qui concerne les engrais azotés (Erisman et al., 2008) et les engrais phosphatés (Cordell et al., 2009). Leur surutilisation cause des problèmes de pollution de l'air et de l'eau et peut affecter les écosystèmes (Erisman et al., 2008). L'utilisation des engrais phosphatés repose sur des ressources fossiles non renouvelables, dont la réserve estimée est faible et très localisée (Cordell et al., 2009). L'agriculture est aussi la cause de nombreux risques de pertes de biodiversités (Dudley and Alexander, 2017). Dans ce contexte, le concept d'agriculture intelligente face au climat (*climate smart agriculture*) a été proposé par la FAO (FAO, 2021). À travers ce concept, la FAO identifie trois principaux défis pour l'agriculture : l'augmentation durable de la production agricole, l'adaptation face au changement climatique et la diminution des émissions de GES causées par l'agriculture.

Pour relever ces défis, de nombreuses solutions complémentaires sont utilisées. La méthanisation est considérée comme l'une d'elle. Les premiers méthaniseurs ont été construits dans les années 1940. La recherche sur ce domaine a augmenté dans les années 1970, lorsque le choc pétrolier a causé une augmentation des coûts de l'énergie (Moletta, 2015). Actuellement, les principaux avantages attribués à la méthanisation sont le traitement des déchets, la production d'énergie renouvelable et le recyclage des nutriments en agriculture. La méthanisation offre une source d'énergie, le biogaz, à partir des « ressources » renouvelables que peuvent être les déchets organiques et les cultures, dédiées ou non, dans le plein esprit de la bioéconomie (Hamelin et al., 2019; Tsui and Wong, 2019). Le biogaz est une énergie renouvelable. Comparé à d'autres énergies renouvelables, le biométhane a l'avantage de pouvoir être stocké. Un des débouchés du biométhane est son usage comme carburant pour le transport (Cong et al., 2017; Scarlat et al., 2018; Vasco-Correa et al., 2018). La méthanisation a aussi l'avantage de recycler les nutriments en les valorisant au champ (Stürmer et al., 2020).

I.4 La méthanisation dans le monde, en Europe, et en France

Depuis des années 2000, la production de biogaz a fortement augmenté dans la plupart des régions du monde (Scarlat et al., 2018). La production mondiale de biogaz a été multipliée par un facteur 4 à 5 entre 2004 et 2014 (d'après Eurostat, Scarlat et al., 2018). En 2015, l'Europe produisait la moitié du biogaz mondial, la Chine et les Etats-Unis étant les autres producteurs importants (Lu and Gao, 2021;

Scarlat et al., 2018). L'essor de la méthanisation est favorisé par des politiques publiques (objectifs de réduction des émissions de GES, objectifs de développement des énergies renouvelables, objectifs de développement territorial, objectifs de réduction des pollutions associées au traitement des déchets ou à la gestion des nutriments), et par des incitations (par exemple, des tarifs de rachat du biogaz ou des crédits alloués à la mise en place de la méthanisation) (Vasco-Correa et al., 2018).

En Afrique et en Asie, on retrouve beaucoup de petits méthaniseurs domestiques, à l'échelle d'un foyer. Sur ces deux continents, de nombreux pays développent des politiques pour promouvoir la méthanisation, à l'échelle domestique ou industrielle (Kemausuor et al., 2018; Mengistu et al., 2015; Rupf et al., 2017; Scarlat et al., 2018; Surendra et al., 2014; Tagne et al., 2021; Tolessa et al., 2020). La méthanisation s'est rapidement développée en Chine entre 2000 et 2015, avec un ralentissement de la croissance entre 2015 et 2017, et concerne les méthaniseurs domestiques (environ 40 millions en 2017) et les méthaniseurs industriels (environ 100 000 en 2017) (Lu and Gao, 2021). En Amérique latine, des pays comme la Bolivie, l'Equateur, le Mexique, le Nicaragua, ou le Pérou ont favorisé l'installation de méthaniseurs de petite taille, tandis que le Brésil, la Colombie, le Honduras, l'Argentine, ont plutôt été sujet à des installations de plus gros méthaniseurs industriels, notamment dans les grandes fermes et les palmeraies (Scarlat et al., 2018).

L'Europe est le continent où la production de biogaz est la plus importante. Le nombre de méthaniseurs en Europe était de 18 000 en 2019, dont 11 000 localisés en Allemagne (European Biogas Association, 2020). La production de biogaz européenne a été multipliée par 2,5 entre 2011 et 2016, avec une très faible évolution globale entre 2016 et 2019. Entre 2009 et 2019, le nombre de méthaniseurs a été multiplié par 2,9. Ce développement de la méthanisation correspond à des actions de la part des gouvernements comme la directive européenne pour l'énergie renouvelable de 2009 (Directive 2009/28/EC, 2009), la stratégie pour la bioéconomie de 2012 (European Commission. Directorate General for Research and Innovation, 2012) ou le récent pacte vert pour l'Europe (COM(2019) 640 final, 2019). Cette situation cache une grande hétérogénéité dans la mise en place de la méthanisation entre les pays. Entre autres, les intrants utilisés varient fortement d'un pays à l'autre (Figure I.1). En Allemagne et en Autriche, plus de 50% des intrants sont des cultures dédiées ; en France et en République Tchèque, 60% des intrants sont des déchets issus de l'agriculture ; la Suède utilise majoritairement des boues de STEU (> 90%) ; le Royaume Uni utilise des déchets issus des ordures ménagères ou de l'industrie agroalimentaire à hauteur de 50% (European Biogas Association, 2020). La plupart des méthaniseurs européens ont des tailles industrielles (Vasco-Correa et al., 2018). Les petits méthaniseurs, traitant quelques tonnes de déchets par an dans les zones urbaines européennes (Thiriet et al., 2020; Walker et al., 2017), sont parfois étudiés mais très peu répandus.

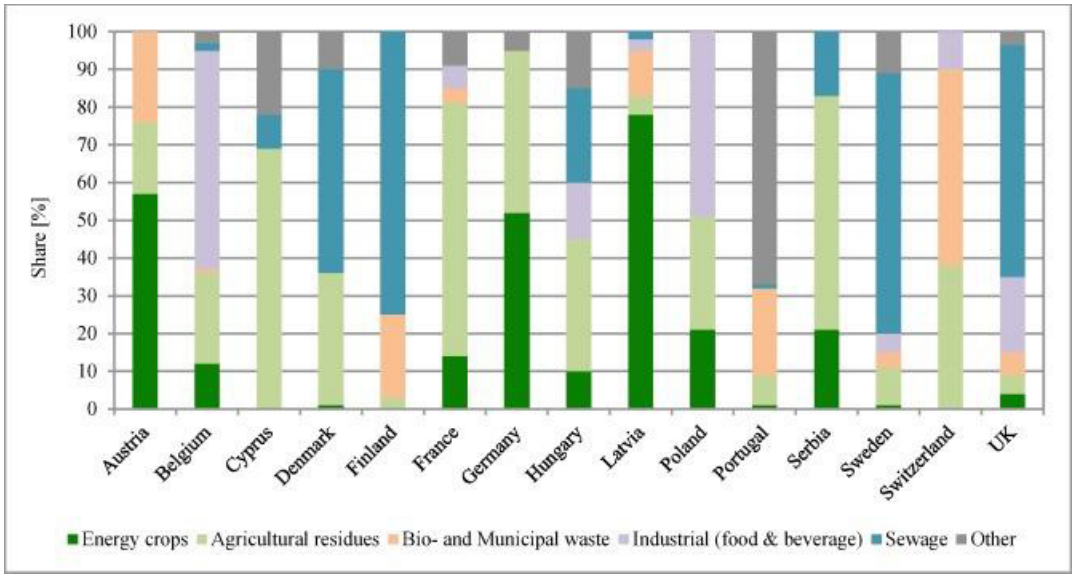

Figure I.1 – Intrants utilisés pour la méthanisation dans différents pays européens, en 2015. La figure est issue de Scarlat et al. (2018).

En France, l'agriculture est responsable de 19% des émissions de GES. Parmi ces émissions, 45% sont dues aux émissions de méthane (CH_4), originaires majoritairement de l'élevage, et 42% aux émissions de protoxyde d'azote (N_2O), majoritairement originaires de la fertilisation azotée. 89% et 68% des émissions nationales de N_2O et de CH_4 proviennent de l'agriculture (CITEPA, 2020). La méthanisation a été avancée comme une des manières de réduire les émissions de GES agricoles en France (Pellerin et al., 2013). Le plan Énergie Méthanisation Autonomie Azote (Ministère de l'agriculture, de l'agroalimentaire et de la forêt, 2013) a été lancé en 2013 pour promouvoir la méthanisation agricole, dans une optique de production d'énergie renouvelable, d'ancrage de l'agriculture dans le territoire et de diminution des imports d'engrais de synthèse dans les exploitations agricoles. En France, le nombre de méthaniseurs a ainsi augmenté : en 2010, il y avait 204 unités de méthanisation en France ; il y en avait 1 019 en 2020 (SINOE, 2021) (Figure I.2). Le nombre d'unités valorisant le biogaz en injection augmente fortement : il a par exemple doublé entre 2019 (93 unités) et 2020 (193 unités, la moitié des nouvelles unités installées cette année-là valorisent le biogaz en injection). Les méthaniseurs sont principalement des méthaniseurs agricoles à la ferme, gérés par un ou des agriculteurs, et dont la capacité moyenne de traitement des déchets est de 11 000 t an^{-1}. La particularité de la méthanisation en exploitation agricole est la forte importance de l'épandage des digestats au champ. Le digestat est vu comme un engrais qui peut bénéficier aux agriculteurs. Les méthaniseurs dit « territoriaux », ou « centralisés », utilisent en premier lieu des déchets ménagers, des déchets de collectivités ou des déchets issus des industries agro-alimentaires. Ils traitent en moyenne 44 000 t de déchets par an (SINOE, 2021). On trouve aussi des unités de méthanisation en STEP ou en industrie. Malgré cette augmentation de l'importance de la méthanisation, le biogaz ne représente en 2019 que 3.6% de la production d'énergie primaire renouvelable, soit 0.4% de la production d'énergie primaire totale en France (SDES, 2020).

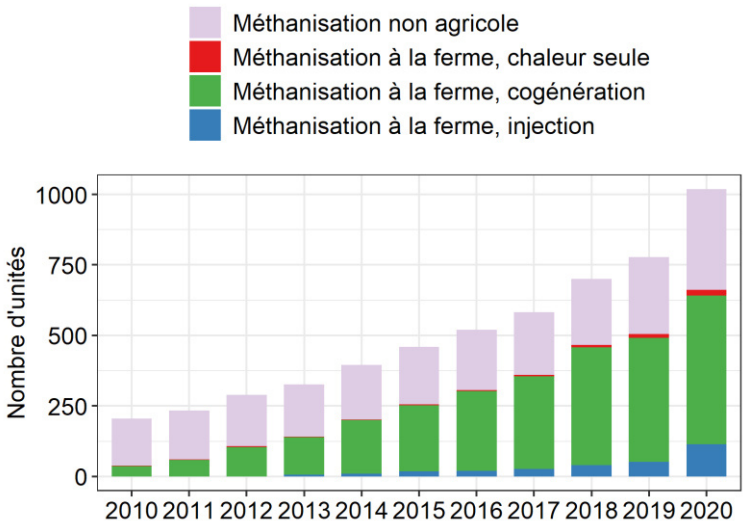

Figure I.2 – Évolution du nombre d'unité de méthanisation en France. Les données proviennent de SINOE (2021)

I.5 Controverses liées à la méthanisation

Actuellement, la méthanisation soulève aussi des questions de la part de la société civile, auxquelles des réponses doivent être apportées. Ces questions concernent, entre autres, les effets sur les agrosystèmes des digestats. Ce sont des engrais organiques de plus en plus fréquents, et le développement récent de la méthanisation fait qu'il y a encore un manque de recul sur certains effets des digestats sur les sols. Un besoin d'information existe sur l'effet des digestats sur la vie du sol (faune du sol, microbiologie) et sur leur capacité à enrichir le sol en matière organique. Comme tous les engrais organiques, l'épandage de digestat comporte aussi des risques de pollution azotée (nitrate, ammoniac) en cas de mauvaise utilisation. Le type d'intrant méthanisé est une question importante liée à la méthanisation. L'Allemagne est un pays où la méthanisation repose en grande partie sur les cultures énergétiques. Cette pratique pose des questions sur l'usage des terres agricoles et la concurrence avec les productions alimentaires et a été montrée comme moins performante d'un point de vue environnemental que l'utilisation de déchets (Hamelin et al., 2014). En France, cette pratique est proscrite et un méthaniseur ne peut pas utiliser plus de 15% de cultures principales dédiées dans ses intrants (Décret n° 2016-929 du 7 juillet 2016 pris pour l'application de l'article L. 541-39 du code de l'environnement). Cependant, il y a un développement de l'utilisation de CIVE destinées à être méthanisées, dont l'effet de leur incorporation dans les systèmes de cultures est en cours d'étude, par exemple en terme de stockage de C, de services associés aux cultures intermédiaires ou de consommation éventuelle de ressources. Ces interrogations sont visibles entre autres par l'augmentation des articles de presse traitant le sujet de la méthanisation durant ces dernières années. Il est nécessaire d'étudier ces questions pour identifier les risques et les minimiser, dans le but de promouvoir une méthanisation vertueuse pour l'environnement et la société.

I.6 Cadre de la thèse

Cette première section a permis de mettre en évidence la diversité des procédés de méthanisation. Dans le cadre de cette thèse, nous nous intéresserons à une implémentation particulière de la méthanisation : **la co-digestion à la ferme des effluents bovins et de déchets urbains et agro-industriels collectés localement**. Il s'agit d'une modalité de méthanisation qui a suscité un programme de développement important, et qui s'est fortement développée en France depuis 2012. Les systèmes basés spécifiquement sur la méthanisation de déchets non agricoles, ou ceux utilisant de façon importante des CIVE ne seront pas abordés. La méthanisation touche trois secteurs économiques : la production d'énergie, le traitement des déchets, l'agriculture. Les questions sur la production d'énergie, qui est un des buts principaux de la méthanisation, n'est pas au centre de cette thèse. En effet, notre centre d'intérêt est **l'aspect agronomique de la méthanisation agricole**, en particulier la gestion et l'épandage des digestats au champ. Malgré ce périmètre déjà restreint, tous les effets de l'épandage de digestats ne peuvent pas être étudiés. Nous nous concentrons dans cette thèse sur les effets de la méthanisation liés **aux cycles du C et du N à la ferme**. Ces deux éléments jouent un rôle de premier plan dans la performance environnementale de la méthanisation. En effet, ils sont impliqués dans de nombreux processus d'intérêt : émissions de GES, dépendance aux ressources renouvelables, émissions d'ammoniac, lixiviation de nitrates, fertilité des sols. Ces deux cycles biogéochimiques sont également très liés en agriculture (Guenet et al., 2021). Pour prendre en compte les effets de la méthanisation par l'import de nutriments via les co-substrats sur l'épandage des produits et sur les changements induits dans la gestion des effluents d'élevage, nous **considérons l'échelle du méthaniseur et de l'exploitation agricole (hormis l'activité d'élevage en elle-même)**.

La thèse traite donc la question de l'introduction d'un méthaniseur dans une exploitation en polyculture élevage sur les cycles du C et de N. Pour cela, nous allons utiliser des expérimentations au champ et au laboratoire. Dans un deuxième temps, ces expériences vont permettre de valider deux modèles. D'abord, le modèle sol-plante STICS (Brisson et al., 2008) permet de simuler la croissance des cultures et les cycles du C et de N au champ, incluant les pertes azotées vers l'environnement (ammoniac, N_2O, nitrates). Nous évaluons ici la capacité de STICS à simuler une rotation culturale fertilisée avec des digestats. Ensuite, le modèle de méthanisation SYS-Metha (Bareha et al., 2021b) permet de prédire la production de biogaz, les caractéristiques des digestats et les émissions gazeuses durant leur stockage. Le modèle est paramétré sur le cas d'étude du méthaniseur installé sur l'exploitation INRAE du centre de Nouzilly. Enfin, ces deux modèles sont couplés pour faire un bilan des cycles biogéochimiques du C et de N à l'échelle de l'exploitation agricole. Un focus particulier est aussi fait sur la vie du sol à travers le cas des vers de terre. La thèse débute par une synthèse bibliographique qui amène à la problématique et aux objectifs détaillés de la thèse.

Chapitre 1

Synthèse bibliographique : introduction de la méthanisation dans une ferme de polyculture élevage

1.1 Les digestats sont des Produits Résiduaires Organiques (PRO)

1.1.1 Les PRO : définitions et présentations

Les **Produits Résiduaires Organiques (PRO)** sont des matières organiques d'origine résiduaire (déchets ou sous-produits) valorisées en agriculture pour augmenter la fertilité des sols. Les PRO peuvent être d'origine et de natures très variées. On peut citer quelques grandes familles de PRO : les effluents d'élevages (lisiers, fumiers), les PRO d'origine urbaine (composts ou digestats de biodéchets, de déchets verts, boues d'épuration), les PRO d'origine industrielle (vinasse, marcs…) ; ils peuvent avoir subi différents traitements avant leur épandage (compostage, méthanisation, pyrolyse…) (Houot et al., 2014). Traités ou non traités, les PRO peuvent être épandus au champ, pour y apporter des éléments d'intérêts : de la matière organique et des nutriments (azote N, phosphore P, et potassium K entre autres). L'utilisation des PRO au champ permet donc d'augmenter la fertilité des sols et de recycler des nutriments, pour diminuer les besoins en engrais de synthèse ou d'origine fossile. Ils sont une des sources principales de nutriments en agriculture biologique. Pour les agriculteurs, les raisons principales d'utilisation des PRO sont une augmentation de la fertilité des sols (teneur en matière organique du sol, structure) et l'apport de nutriments. La disponibilité et le prix sont des critères importants à leur utilisation ou non. À l'inverse, les principales barrières sont les contraintes techniques d'épandage (logistique, temps de travail, portance des sols), l'incertitude sur les teneurs en nutriments qui complexifie le pilotage de la fertilisation, l'émission de mauvaises odeurs pour les riverains, et l'envie de ne pas prendre le risque de contaminer les sols avec des éléments provenant des déchets, en particulier pour les PRO d'origine urbaine (Case et al., 2017; Moinard et al., 2021).

Les différents PRO ont des caractéristiques physico-chimiques très variées. La première caractéristique est leur teneur en matière sèche : les PRO liquides ne sont pas épandus de la même façon que les PRO solides. Les PRO sont plus ou moins riches en matière organique. On considère que la matière organique des PRO est composée environ pour moitié de carbone C (en masse). Cette matière organique va être plus ou moins riche en azote organique (N_{org}), ce qui est mesuré par le rapport C/N_{org}. En plus de ce N organique, les PRO peuvent contenir plus ou moins d'azote sous forme minérale (souvent du NH_4^+ dissous ou adsorbé, ou des nitrates dans le cas de composts), facilement utilisable par les plantes. Les teneurs en autres nutriments (P, K, Ca,…) peuvent varier. Des exemples de caractéristiques sont données dans le Tableau 1.1.

Tableau 1.1 – Caractéristiques types de quelques PRO courants. Ces caractéristiques pourront être comparées à celles des digestats. (Chambre d'agriculture de Région Île-de-France, 2018; Gutser et al., 2005; Houot et al., 2014; Levasseur et al., 2019; Levavasseur et al., 2021a).

	Lisier bovin	Fumier bovin	Compost de déchets verts	Boue de STEP (épaissie, chaulée)	Fientes de volaille
MS (%MB)	3 – 11	14 – 30	36 – 75	20 – 30	40 – 85
MO (%MB)	4 – 8	14 – 18	16 – 33	8 – 17	24 – 63
N total (g $kgMB^{-1}$)	1 – 5	4 – 7	6 – 10	8 – 13	22 – 46
N ammoniacal (g $kgMB^{-1}$)	0,5 – 2,5	0,5 – 1,5	0 – 0,5	0,0 – 0,1	3 – 10
N ammoniacal (% N total)	40 – 60	5 – 20	~ 0	0 – 3	10
C/N	5 – 9	10 – 15	9 – 45	6 – 12	8
P (g $kgMB^{-1}$)	0,2 – 0,9	0,4 – 2,2	1,4 – 2,6	2,3 – 9,6	9 – 18
K (g $kgMB^{-1}$)	1,1 – 3,6	2,8 – 8,5	2,4 – 7,8	0,3 – 1,6	9 – 20
ISMO	45 – 57	46 – 62	73 – 87	32 – 66	6 – 48
K_{eq}	5 – 50%	5 – 30%	~ 0%	~ 10%	10 – 60%

MS : matière sèche. MB : matière brute. MO : matière organique. Keq : coefficient d'équivalence engrais. ISMO : indice de stabilité de la matière organique.

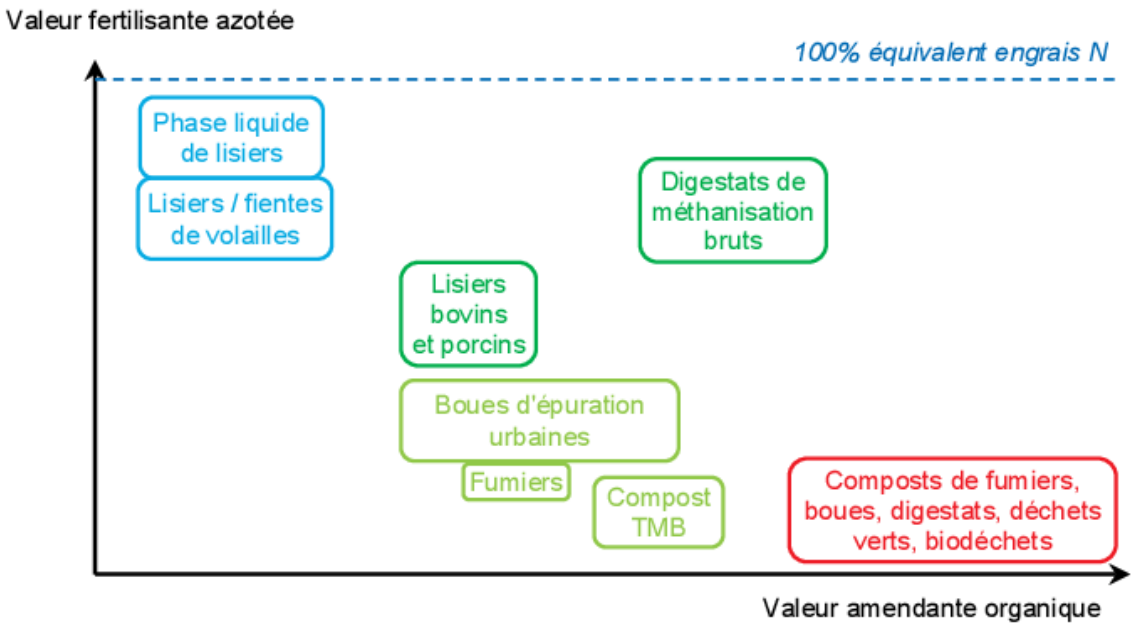

Figure 1.1 – Présentation de différents PRO, classés selon leurs propriétés amendantes ou fertilisantes. Extrait de Houot et al. (2014). TMB : tri mécano-biologique.

Houot et al. (2014) ont proposé de classer les PRO en deux catégories, selon leur utilité et leurs effets au champ (Figure 1.1). Les PRO **fertilisants** apportent de l'azote pouvant être valorisé à court terme par les plantes. Les PRO fertilisants vont pouvoir se substituer aux engrais de synthèse. Ils sont en général riches en azote minéral, une forme assimilable rapidement par les plantes. Les PRO fertilisants incluent, entre autres, les lisiers, les fientes de volailles, les digestats liquides. Les PRO **amendants** sont des PRO plus riches en matière organique, qui vont participer à l'augmentation de la matière organique du sol (MOS), ce qui va contribuer à améliorer la fertilité physique (amélioration de la structure, diminution de la densité apparente) et chimique (fourniture en azote accrue, plus grandes réserves de P et K) du sol après des apports répétés à long terme (plusieurs années). Les PRO amendants incluent les composts, les fumiers, les digestats solides.

1.1.2 Étude de la valeur fertilisante des PRO

En France, les besoins des plantes en terme de nutriments sont en premier lieu lié à l'azote, puis à d'autres nutriments (P, K, S). Nous nous concentrons ici sur la valeur fertilisante azotée des PRO, car l'azote est l'élément qui nous intéresse dans cette thèse.

La valeur fertilisante azotée des PRO peut se définir comme la capacité des PRO à apporter de l'azote pour la croissance des plantes et donc à se substituer aux engrais azotés de synthèse. On la mesure souvent grâce à des expériences au champ, en comparant l'absorption d'azote par une culture qui n'est pas fertilisée, avec celle d'une culture fertilisée. Le Coefficient Apparent d'Utilisation (CAU) d'un engrais est défini comme le rapport entre l'azote supplémentaire absorbée par la culture fertilisée, divisé par l'azote total apporté par l'engrais. En anglais, on parle de *Nitrogen use efficiency* (*NUE*) ou de *apparent N recovery* (*ANR*). Plus le CAU est élevé, plus la culture a utilisé efficacement l'azote de l'engrais. On peut ensuite définir le coefficient d'équivalence engrais (K_{eq}) d'un PRO comme le rapport entre le CAU de ce PRO et le CAU d'un engrais azoté de synthèse de référence. Si un lisier possède un K_{eq} de 50%, cela signifie qu'un apport de 100 kgN ha^{-1} avec ce lisier sera équivalent à un apport de 50 kgN ha^{-1} avec des engrais de synthèse. En anglais, on parle de *mineral fertilizer equivalence* (*MFE*), ou encore de *Nitrogen fertilizer replacement value* (*NFRV*). Le pouvoir fertilisant azoté des PRO peut être très faible (Keq proche de 0%, composts) à très fort (K_{eq} proche de 100%, certaines fientes ou produit à base d'urine) (Tableau 1.1). Les valeurs de K_{eq} dépendent des PRO, mais aussi du climat, de la culture, de la période et du mode d'apport.

Les processus associés à la fertilisation avec les PRO sont présentés sur la Figure 1.2. La valeur fertilisante azotée des PRO est d'abord liée à leur teneur en azote ammoniacal (NH_4^+, urée) (Gutser et al., 2005), qui est rapidement nitrifié (transformation sous forme de nitrate) puis utilisable par les plantes au champ. Cet azote ammoniacal est sujet à de la volatilisation sous forme d'ammoniac, un polluant atmosphérique, ce qui peut fortement limiter la valeur fertilisante des PRO (Frost et al., 1990).

À titre d'exemple, la volatilisation des lisiers concerne en général 15 à 20% et jusqu'à 100% de l'azote ammoniacal apporté (Hafner et al., 2018). Enfin, lorsque la matière organique des PRO est décomposée par les organismes du sol, l'azote organique peut être minéralisé (minéralisation) ou, à l'inverse, l'azote minéral peut être organisé (immobilisation), à l'échelle de quelques mois, ce qui influence la valeur fertilisante des PRO (Bhogal et al., 2016; Delin et al., 2012). Le rapport C/N des PRO explique ces différences d'effet. Un rapport C/N faible des PRO est statistiquement associé à une forte minéralisation d'azote (Delin et al., 2012). La caractérisation de ce phénomène se fait souvent grâce à des incubations au laboratoire : les PRO sont mélangés à du sol dans des conditions contrôlées et l'on suit l'augmentation (minéralisation) ou la diminution (immobilisation) de l'azote minéral du sol au cours du temps (Aoyama and Nozawa, 1993). Ces résultats en laboratoire peuvent ensuite être extrapolés au champ, grâce à des modèles par exemple. L'immobilisation ou la minéralisation à l'échelle d'une année concerne en général entre 0% et 50% de l'azote organique apporté (Levavasseur et al., 2021a).

Comme pour tous les engrais azotés, les apports de PRO peuvent être suivis d'émissions de N_2O (Zhou et al., 2017) ou de lixiviation de nitrate (NO_3^-) (Basso and Ritchie, 2005) (Figure 1.2). Cela s'explique entre autres par la difficulté d'ajuster les doses d'apport, à cause de la variabilité des PRO et des contraintes d'épandage : il y a donc un risque de surfertilisation ou d'apport d'azote en dehors des périodes de besoins des plantes.

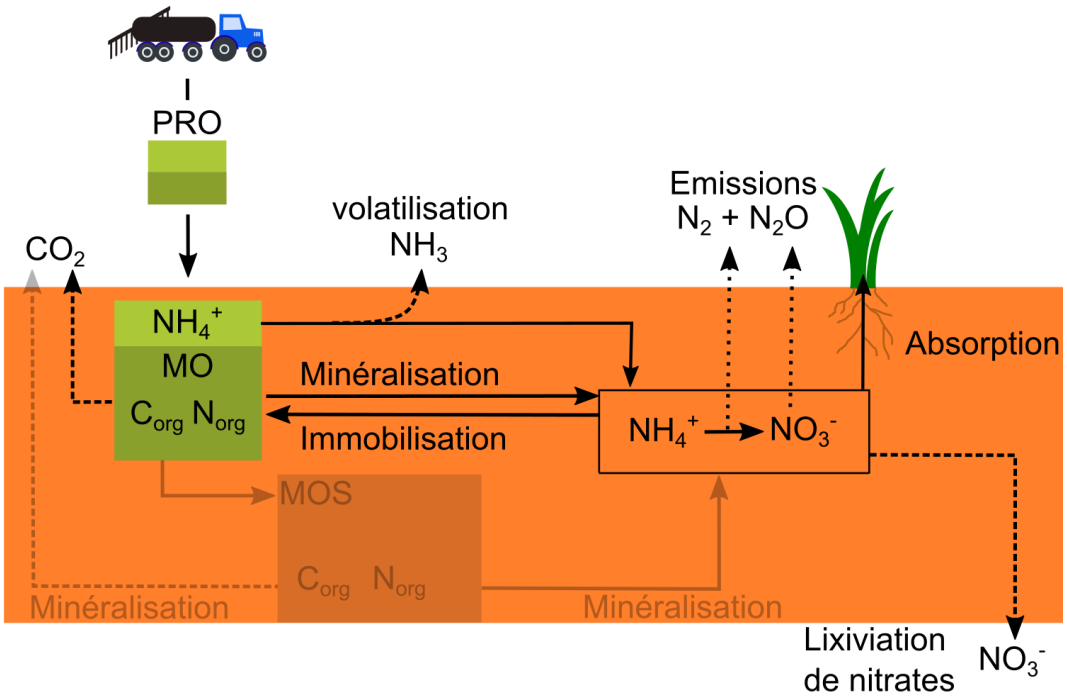

Figure 1.2 – Effets de l'épandage de PRO à court terme (1 an) sur les cycles du C et du N du sol (effet fertilisant)

1.1.3 Étude de la valeur amendante des PRO

Des essais au champ à long terme ont montré des aspects positifs liés à des apports répétés de PRO à long terme sur la qualité des sols (pouvoir amendant). Lors de leur épandage, les PRO apportent de la matière organique au champ, dont une partie est minéralisée et émise dans l'atmosphère sous forme de CO_2 (Figure 1.3). Contrairement au CO_2 issu de la combustion de ressources fossiles, Ce CO_2 est dit biogénique, car le carbone est issu de matière biologique et donc capté à l'origine dans l'atmosphère par photosynthèse. Ces émissions ne sont donc pas considérées comme des émissions de GES. Une partie de la matière organique peut s'intégrer plus durablement dans le sol. Cette augmentation des entrées de C dans le sol va promouvoir l'augmentation des stocks de C du sol (Chenu et al., 2019). Ainsi, les apports répétés de PRO augmentent la matière organique du sol (MOS) (Bhogal et al., 2018; Diacono and Montemurro, 2010; Edmeades, 2003; Haynes and Naidu, 1998; Maillard and Angers, 2014; Maltas et al., 2018; Zavattaro et al., 2017), ce qui se traduit par une augmentation de la fertilité physique des sols. Les indicateurs les plus consensuels sont la diminution de la densité apparente, et l'augmentation de la stabilité des agrégats, mais d'autres bénéfices ont parfois été notés : amélioration des propriétés hydriques du sol (rétention en eau, conductivité hydraulique), augmentation de la porosité du sol, augmentation de la capacité d'échanges cationiques (CEC), diminution de la croûte de battance (Annabi et al., 2011; Bhogal et al., 2018; Diacono and Montemurro, 2010; Edmeades, 2003; Haynes and Naidu, 1998; Oldfield et al., 2018; Sadet-Bourgeteau et al., 2018). Les apports de PRO augmentent aussi les teneurs en nutriments du sol comme le N, le P et le K (Bhogal et al., 2018; Sadet-Bourgeteau et al., 2018). Les apports de PRO répétés à long terme peuvent aussi favoriser la biomasse microbienne (Bhogal et al., 2018; Sadet-Bourgeteau et al., 2018), ainsi que la mésofaune du sol (D'Hose et al., 2018). L'augmentation de la MOS augmente la fourniture en azote par le sol (Bhogal et al., 2018; Diacono and Montemurro, 2010). En conséquence, on peut considérer que l'efficacité fertilisante azotée des PRO augmente lors d'apports répétés à long terme (Gómez-Muñoz et al., 2017 ; Gutser et al., 2005). La séquestration de C dans la MOS est vue comme une émission négative de GES ; cependant, dans le cas des PRO, cela concerne uniquement l'utilisation de nouvelles matières organiques qui n'était pas valorisées au champ ailleurs auparavant (Powlson et al., 2011).

Le potentiel des PRO à augmenter la teneur en matière organique du sol est corrélée à la quantité de carbone apportée au sol (Maltas et al., 2018), ainsi qu'à la récalcitrance de cette matière organique. En effet, la proportion de C apporté qui est incorporée à la MOS diffère en fonction des PRO. On distingue la matière organique labile des PRO, qui est minéralisée rapidement et émise sous forme de CO_2, et la matière organique récalcitrante, qui restera dans le sol plus longtemps (par exemple, plus d'un an). La proportion de C récalcitrant d'un PRO est souvent estimée avec des incubations de mélange sol-PRO en laboratoire. Il est ainsi possible de suivre les émissions de CO_2 au cours du temps et le C apporté qui reste au sol est considéré récalcitrant. Les PRO peuvent avoir en général 20% (faible pouvoir amendant) à 100% (très fort pouvoir amendant) de leur matière organique qui est récalcitrante

(Levavasseur et al., 2021a). Pour faciliter la détermination du pouvoir amendant des PRO, des analyses basées sur des fractionnements biochimiques sont utilisées. Elles permettent par exemple l'établissement d'indicateurs comme l'ISMO (Indice de Stabilité de la Matière Organique) (en anglais I_{ROC} : *Indice of recalcitrant organic carbon*) (Lashermes et al., 2009). L'ISMO est un prédicteur du pourcentage de matière organique récalcitrante, plus il est élevé, plus le PRO aura un bon pouvoir amendant. La connaissance de la récalcitrance de la matière organique des PRO permet ensuite d'évaluer leur effet amendant à long terme grâce à des modèles (Mondini et al., 2017; Peltre et al., 2012).

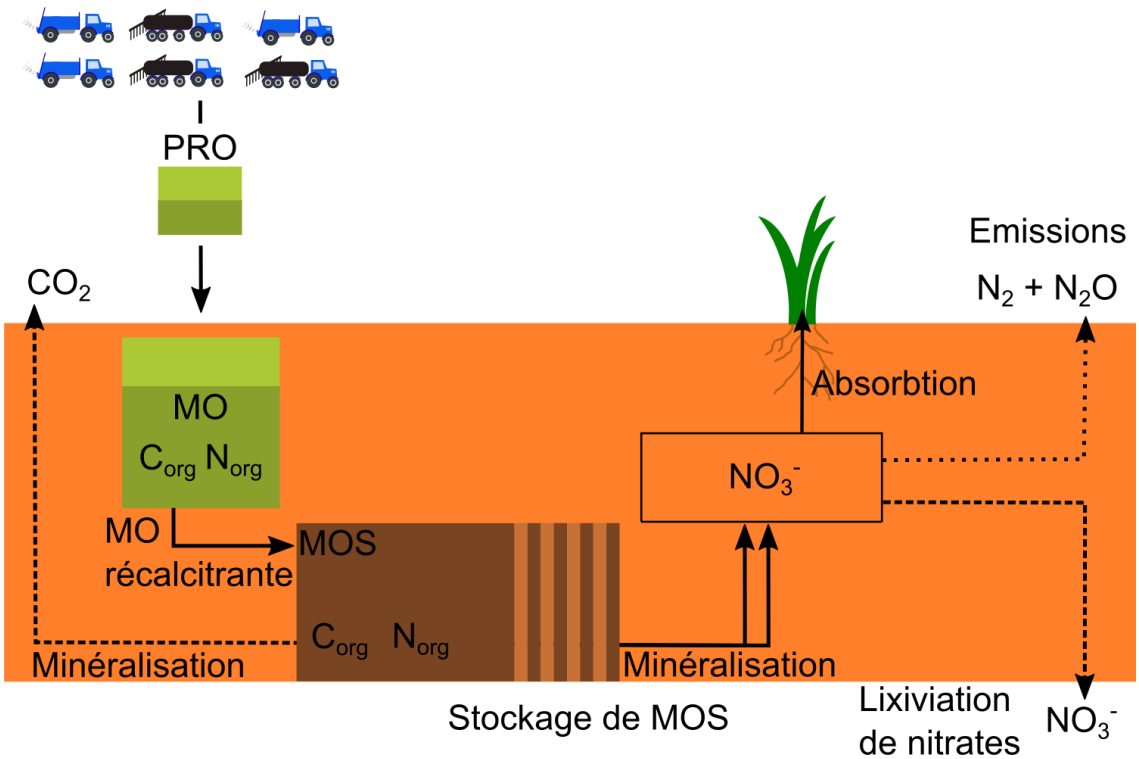

Figure 1.3 – Effets d'épandages répétés de PRO à long terme (10 ans) sur le cycle du C et du N du sol (effet amendant).

1.1.4 Les digestats parmi les PRO

De nombreuses études et essais au champ à long terme ont contribué à améliorer la connaissance et la gestion des PRO depuis plusieurs dizaines d'années, en particulier en ce qui concerne les effluents d'élevage et les composts (Bhogal et al., 2018; Diacono and Montemurro, 2010; Edmeades, 2003; Haynes and Naidu, 1998; Maltas et al., 2018; Zavattaro et al., 2017). Les digestats, utilisés de manière importante plus récemment, n'ont pas encore été étudiés avec le même recul.

Les digestats peuvent avoir des caractéristiques très variées (Tableau 1.2). Pendant la digestion anaérobie, de la matière organique est minéralisée. Du C est exporté sous forme de biogaz, et la matière organique résiduelle est plus stable après digestion. Concernant l'azote, une partie de l'azote organique est minéralisée. Les digestats issus d'effluents d'élevage ont ainsi un rapport C/N plus faible et un ratio azote minéral/azote organique plus fort que les effluents non digérés (Möller and Müller, 2012). Néanmoins, les autres intrants vont apporter des nutriments et vont affecter la composition des digestats (Guilayn et al., 2019a). Les digestats bruts issus de voies humides sont liquides, avec des teneurs en matière sèche variant entre 2 et 10%, assez semblables à des lisiers (Beggio et al., 2019; Guilayn et al., 2019a; Houot et al., 2014; Stürmer et al., 2020). Les digestats issus de méthanisation en voie sèche (plus rare) seront plus pâteux ou solides.

La séparation de phase des digestats est fréquente. On appellera par la suite **digestat brut** le digestat n'ayant pas subi de séparation de phase. Après séparation de phase, la phase liquide est dénommée **digestat liquide**, et la phase solide **digestat solide**. Le digestat liquide sera appauvri en matière solide (matière sèche, matière organique, P). Le digestat solide a des teneurs en matière sèche semblables à un fumier ou à un compost. En fonction de la technologie utilisée, la séparation de phase sera plus ou moins efficace. L'utilisation d'une presse à vis, peu efficace, va laisser des digestats liquides avec des teneurs relativement élevées en matière sèche (environ 30% de la matière sèche peut être exportée vers la phase solide). L'utilisation de technologies plus efficaces (centrifugation par exemple) va amener la grande majorité de la matière sèche (environ 80%) dans la phase solide (Guilayn et al., 2019b). L'azote ammoniacal, soluble, reste préférentiellement dans la phase liquide. D'autres post-traitments (par exemple compostage ou séchage) peuvent modifier les caractéristiques des digestats, mais ne seront pas étudiés en détail.

Tableau 1.2 – Caractéristiques types de digestats (Guilayn et al., 2019a).

	Digestat brut, voie humide	Digestat brut, voie sèche	Digestat liquide (voie humide, presse à vis)	Digestat solide (voie humide, presse à vis)
MS (%MB)	2.5 – 10	12 – 25	2 – 8	20 – 35
MO (%MB)	1 – 6	5 – 11	1 – 5	20 – 30
N total (g $kgMB^{-1}$)	2 – 8	3 – 6	3 – 8	4 – 8
N ammoniacal (g $kgMB^{-1}$)	0,5 – 4,0	1,0 – 2,5	2,0 – 3,0	0,7 – 3,0
N ammoniacal (% N total)	30 – 70%	10 – 40%	30 – 70%	20 – 40%
C/N	2 – 10	10 – 20	3 – 6	12 – 25
P (g $kgMB^{-1}$)	0,2 – 1,5	0,1 – 2,5	0,6 – 1,3	1,1 – 4,0
K (g $kgMB^{-1}$)	0,1 – 4,0	0,2 – 5,0	1,5 – 5,0	2,0 – 5,0

MS : matière sèche. MB : matière brute. MO : matière organique.

1.2 Effet de l'épandage de digestat au champ à l'échelle de la parcelle

La section 1.2 synthétise les connaissances actuelles sur les effets de l'apport de digestat au champ, et plus particulièrement sur les effets liés aux cycles du C et du N au champ.

1.2.1 Digestat et fertilisation azotée

Nous nous intéressons ici à la capacité des digestats à se substituer aux engrais azotés à court terme (un an). Les digestats bruts ou liquides issus de méthaniseurs en voie humide sont en général riches en azote (ordre de grandeur : 2 à 8 gN par kg de matière brute MB), avec une forte proportion d'azote minéral (l'ordre de grandeur est de 30 à 80 % de l'azote total) (Beggio et al., 2019; Guilayn et al., 2019a; Nkoa, 2014; Stürmer et al., 2020). Ces propriétés en font de bons fertilisants azotés. Beaucoup d'études ont montré que les digestats liquides ou bruts pouvaient remplacer les fertilisants azotés de synthèse sur des essais au champ, sur des cultures comme le blé, le maïs, l'orge ou sur prairie (Barłóg et al., 2019; Cavalli et al., 2016; Chantigny et al., 2008; Ferdous et al., 2020; Loria et al., 2007; Pastorelli et al., 2021; Verdi et al., 2019; Zicker et al., 2020). Des exemples de Keq de digestats sont données dans le Tableau 1.3. À l'échelle d'une année, on suppose souvent que l'azote ammoniacal correspond à l'azote assimilable par les plantes à court terme. Néanmoins, cette valeur fertilisante peut être modifiée par les phénomènes de volatilisation d'ammoniac au champ et de minéralisation de l'azote organique (Möller and Müller, 2012). La dynamique de l'azote minéral dans le sol après apport de digestat peut être suivie grâce à des incubations de mélanges sol-digestat en conditions contrôlées. Avec cette méthode, en fonction des digestats bruts ou liquides étudiés, il est possible d'observer une minéralisation d'azote organique (Müller-Stöver et al., 2016; Tampio et al., 2016) ou une immobilisation d'azote minéral (Alburquerque et al., 2012; de la Fuente et al., 2013). L'influence de ces deux phénomènes sur la teneur en azote inorganique total du sol reste souvent faible au regard des quantités d'azote ammoniacal apportées (Alburquerque et al., 2012; Cavalli et al., 2017; Müller-Stöver et al., 2016).

Les digestats solides sont plus souvent utilisés en tant qu'amendement. En effet leur teneur en matière organique est plus élevée et leur teneur en azote ammoniacal plus faible que les digestats liquides ou bruts (Guilayn et al., 2019b; Houot et al., 2014). Leur pouvoir fertilisant azoté risque donc d'être faible (Cavalli et al., 2016; Thomas et al., 2017) (Tableau 1.3). Chiyoka et al. (2014), de la Fuente et al. (2013) et Cavalli et al. (2017) ont montré que certains digestats solides immobilisaient de l'azote lors d'incubation avec du sol en conditions contrôlées, ce qui pourrait limiter leur valeur fertilisante. Néanmoins, certains digestats solides ont été montrés comme efficace pour remplacer des engrais azotés sur des légumes (Maucieri et al., 2017).

Actuellement, nous savons donc que les digestats bruts et liquides sont de bons fertilisants. Nous savons que les déterminants de cette fertilité sont d'abord la teneur en azote ammoniacal, et en second lieu la volatilisation de l'azote ammoniacal et la minéralisation de l'azote organique des digestats. L'efficacité fertilisante des digestats solides semble faible mais pourrait mériter une investigation supplémentaire.

Tableau 1.3 – Exemple de K_{eq} de digestats.

Référence	Culture	Produit	K_{eq} (%)
Cavalli et al. (2016)	Maïs	Digestat liquide ou brut	20 – 70
		Digestat solide	0 – 25
Baral et al. (2017)	Orge	Digestat brut	60
Valeurs proposées par la Chambre d'Agriculture de Bretagne (2017)	Blé, colza, Maïs, prairie	Digestat liquide ou brut	60 – 70
		Digestat solide composté	10

1.2.2 Digestat et volatilisation d'ammoniac

L'ammoniac (NH_3) est un gaz polluant. Il contribue à l'acidification et à l'eutrophisation des milieux, et est un précurseur de particules fines : il a donc des impacts sur l'environnement et la santé. En 2018, les émissions d'ammoniac agricoles représentaient 94% des émissions d'ammoniac totales en France. Au sein du secteur agricole, les émissions étaient dues à l'épandage des engrais azotés de synthèse (29%), à la gestion des effluents bovins (bâtiment et stockage, 26%), à l'épandage de PRO au champ (21%). Les autres sources incluent la pâture et la gestion des effluents animaux hors bovins (CITEPA, 2020). Les digestats peuvent donc être concernés par les émissions d'ammoniac lors de leur stockage et lors de l'épandage. Nous nous concentrons ici sur l'épandage des digestats au champ.

La volatilisation est un processus physico-chimique. Lorsqu'un PRO est épandu au champ, un équilibre physico-chimique a lieu entre les ions ammonium NH_4^+ en solution, les molécules d'ammoniac NH_3 en solution et les molécules de NH_3 à l'état gazeux. Sous cette dernière forme, à l'interface entre le PRO et l'atmosphère, le NH_3 peut être émis vers l'atmosphère (Sommer, 1997). C'est un processus rapide, qui dure quelques heures à quelques jours (Carozzi et al., 2013a; Riva et al., 2016). De nombreux facteurs influencent la volatilisation. La volatilisation est directement liée à la concentration en azote ammoniacal dans les PRO : plus la quantité d'azote ammoniacal est grande, plus la quantité d'ammoniac volatilisable sera grande. L'augmentation du pH du mélange PRO-sol et l'augmentation de la température favorisent le déplacement des équilibres thermochimiques vers la phase ammoniacale gazeuse, et favorisent donc la volatilisation. Des conditions climatiques venteuses et une absence de couverture végétale du sol diminuent la résistance atmosphérique au-dessus du sol et favorisent la volatilisation (Hafner et al., 2018). À l'inverse, les caractéristiques du sol, les caractéristiques des PRO,

et les pratiques agricoles qui permettent de limiter le contact avec l'atmosphère inhibent la volatilisation : facilité de l'infiltration des PRO dans le sol, pluie, enfouissement, modes d'apport limitant le contact entre PRO et atmosphère (Hafner et al., 2018; Sommer and Hutchings, 2001; Webb et al., 2010). Lors d'un épandage, les facteurs d'émissions d'ammoniac sont souvent exprimés comme une proportion de l'azote ammoniacal apporté qui est volatilisé.

La volatilisation des lisiers a été fortement étudiée dans la littérature, comme l'atteste la taille de la base de données de mesures d'émissions de NH_3 construite par Hafner et al. (2018). La volatilisation des digestats liquides ou bruts a également été étudiée. Les digestats liquides ou bruts sont sujets aux émissions d'ammoniac, à cause de leur forte concentration en azote ammoniacal et à leur pH relativement élevé (Nkoa, 2014). En fonction des digestats, des conditions et des méthodes d'épandage, des facteurs d'émissions très variés sont mesurés. Quelques exemples sont données dans le Tableau 1.4. La volatilisation concerne une partie importante des apports d'azote ammoniacal et peut donc diminuer la valeur fertilisante azotée des digestats.

La sensibilité des digestats à la volatilisation comparée à celles des lisiers donnent des résultats ambivalents dans la littérature (Möller, 2015). D'un côté, la méthanisation augmente la teneur en azote ammoniacal et le pH, ce qui favorise la volatilisation. De l'autre, elle diminue la teneur en matière sèche du PRO, ce qui peut favoriser l'infiltration des digestats et donc diminuer la volatilisation. Möller and Stinner (2009) ne voient pas de différence de facteurs d'émissions entre les digestats et les lisiers ; Nyord et al. (2012) et Nicholson et al., (2017) observent en général plus de volatilisation après épandage de digestats qu'après épandage de lisiers ; Neerackal et al., (2015) montrent une volatilisation plus importante des lisiers en comparaison avec celle des digestats en conditions de laboratoire. L'injection des digestats directement dans le sol a été démontrée efficace pour réduire fortement les émissions, de l'ordre de 70% (Maris et al., 2021; Riva et al., 2016). Comme pour les autres PRO, l'enfouissement rapide devrait limiter la volatilisation. Comme pour les lisiers, l'apport de digestats acidifiés au champ a été proposé (de França et al., 2021). Cette acidification pourrait limiter la volatilisation et augmenter le pouvoir fertilisant des digestats (Mackens et al., 2021).

La volatilisation de l'ammoniac après apports de produits solides, qui incluent les fumiers, a été moins étudiée dans la littérature. Les facteurs d'émissions des fumiers peuvent être très importants (supérieurs à 60%, voire 80% de l'azote ammoniacal apporté) (Hafner et al., 2018). Nous n'avons trouvé aucune donnée de volatilisation après épandage de digestats solides dans la littérature. Certains auteurs considèrent que la volatilisation après l'épandage des digestats solides doit être négligeable par rapport à l'épandage de digestats liquides (Holly et al., 2017; Nyord et al., 2012). Néanmoins, ceci ne repose sur aucune preuve expérimentale et des références sur la sensibilité des digestats solides à la volatilisation seraient nécessaires.

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Tableau 1.4 – Exemple de facteurs d'émissions d'ammoniac, après épandage de digestats liquides ou bruts, ou de lisiers.

Référence	PRO	Détail	Volatilisation (% N ammoniacal)	Volatilisation (% N total)
Hafner et al. (2018)	Lisiers bovins	~1000 observations	27% (médiane estimée) (0% – >100%)	13% (médiane estimée) (0 – 70%)
Nicholson et al. (2017)	Digestats de biodéchets	Epandage au pendillard. Moyenne sur 4 épandages.	48% (estimé)	38%
	Lisiers bovins et porcins non digérés	Epandage au pendillard. Moyenne sur 4 épandages.	30% (estimé)	24%
Räbiger et al. (2020)	Digestat liquide ou brut	Epandage au pendillard. 5 sites, 3 ans.	13% – 25%	8% – 18%
Riva et al. (2016)	Digestat liquide ou brut	Application en surface	30% – 46%	18% – 29%
	Digestat liquide ou brut	Injection	9% – 11%	5% – 7%
Maris et al. (2021)	Digestat liquide ou brut	Application en surface	17%	8%
	Digestat liquide ou brut	Injection	4%	2%

Comme les lisiers, les digestats liquides ou bruts sont sensibles à la volatilisation. Les déterminants de cette volatilisation et les méthodes de réduction des émissions sont connues. Mieux comprendre les déterminants de l'infiltration des PRO pourrait permettre de mieux comparer effluents non digérés et digestats, ainsi que les différents digestats entre eux. La sensibilité des digestats solides est une question d'intérêt pour la communauté scientifique et les praticiens.

1.2.3 Effet des digestats sur la matière organique du sol

Comme les autres PRO, on attend des digestats qu'ils apportent de la matière organique au champ, favorisant l'augmentation des stocks de MOS. La question du pouvoir amendant des digestats peut prendre deux points de vue. D'une part, on peut essayer de caractériser le pouvoir amendant des digestats en absolu, en étudiant la concentration et la stabilité de la matière organique qui les constitue. D'autre part, dans le cadre de la méthanisation agricole des effluents d'élevage, il est aussi pertinent de comparer le pouvoir amendant des digestats à celui des effluents non digérés.

Très peu d'essais au champ avec des apports de digestat à long terme ont été réalisés, ce qui limite les preuves directes de l'effet amendant de l'épandage des digestats au champ. Dans une approche intermédiaire, Wentzel et al. (2015) ont comparé des champs dans des exploitations utilisant des lisiers ou des digestats depuis plus de 20 ans : en moyenne, les champs fertilisés avec les lisiers ou les digestats ont des teneurs en MOS similaires, malgré des différences dans un sens ou dans l'autre selon les sites. Après 8 ans d'apports de digestats solides, Odlare et al. (2014) montrent une augmentation faible de la MOS en comparaison avec un contrôle sans amendement organique. Dans cet essai, d'autres PRO (fumiers, boues, composts) ont le même effet que les digestats solides. Après 9 ans d'apports de digestats, Zicker et al. (2020) observent une augmentation de la MOS par rapport à un traitement sans PRO. Les auteurs observent un effet des digestats similaire à celui des lisiers, malgré un plus faible apport de C via les digestats.

La méthanisation augmente la stabilité de la matière organique. À l'aide d'incubations en conditions contrôlées (Béghin-Tanneau et al., 2019; Cavalli et al., 2017; de la Fuente et al., 2013; Thomsen et al., 2013), de fractionnements biochimiques de type Van Soest (Van Soest and Wine, 1967) (Tambone et al., 2009), d'analyses spectrométriques (Tambone et al., 2009) ou de la demande en oxygène (Tambone et al., 2009, 2013), plusieurs études ont mis en évidence l'augmentation de la stabilité de la matière organique après méthanisation. Avec des incubations, il a été montré que la stabilité de la matière organique de différents digestats peut être très variable : de 0% à 90% de la matière organique des digestats est stable selon les études d'Alburquerque et al. (2012), Levavasseur et al. (2021a), et Nielsen et al. (2020). La variabilité de la stabilité des digestats est confirmée par des études liées au fractionnement biochimique de digestats issus de différents intrants (Jimenez et al., 2017). Après séparation de phase, la nature de la matière organique est différente entre les digestats liquides et solides. De la Fuente et al. (2013) montrent que les digestats liquides possèdent une matière organique moins

stable que les digestats solides. L'inverse a été trouvé par Askri et al. (2016). Tambone et al. (2019) montrent que les digestats liquides pourraient posséder une matière organique stable, comparable aux composts, et que l'effet amendant des digestats liquides pourrait ne pas être négligeable. Levavasseur et al. (2021a) trouvent un ISMO moyen des digestats de 66% (écart-type : 13%, n = 54), ce qui est supérieur aux effluents d'élevage et similaire ou inférieur aux composts.

La méthanisation peut influencer les propriétés amendantes des PRO via des apports de C par les co-substrats, des exports de carbone dans le biogaz et une augmentation de la stabilité de la matière organique. L'effet global de la méthanisation est donc difficile à évaluer. L'étude de Thomsen et al. (2013) semble montrer que la digestion n'aurait pas d'impact sur le stockage de C dans le sol, car l'export de C dans le biogaz serait compensé par une augmentation de la stabilité de la matière organique du digestat. Cette théorie, largement communiquée, demande toutefois à être validée, nuancée ou réfutée par d'autres études récentes. À partir d'indicateurs de stabilité de la matière organique, Bodilis et al. (2015) ont modélisé une augmentation légèrement moins forte des stocks de MOS sur des fermes utilisant des digestats, en comparaison avec des fermes utilisant d'autres PRO.

En résumé, les digestats permettent d'entretenir la MOS, comme d'autres PRO. Il n'y a pas de consensus pour déterminer si la méthanisation des effluents d'élevage favorisait ou défavorisait le stockage de MOS. Des essais à long terme pourraient compléter notre connaissance sur la capacité des digestats à entretenir la MOS. La caractérisation plus fine de la stabilité des digestats en fonction des processus de méthanisation et des intrants est intéressante, pour ensuite étudier l'effet d'apports répétés de digestats à long terme grâce à de la modélisation, comme suggéré par Jimenez et al. (2015).

1.2.4 Effet des digestats sur la fertilité des sols à long terme

Comme pour les autres PRO, l'augmentation de la MOS suite à l'épandage des digestats peut améliorer la fertilité des sols. L'amélioration de la fertilité physique du sol suite à l'épandage de digestats a également été montrée dans quelques études (Möller, 2015). Dans une étude italienne (Badagliacca et al., 2020), l'apport de digestat solide dans des vergers a induit une augmentation de la stabilité des agrégats à l'échelle de quelques semaines, avant que l'effet ne s'estompe un an après l'apport.

Les effets des digestats sur la fertilisation à long terme (> 5 ans) sont encore peu étudiés avec des essais au champ. Aucun effet de l'apport répété de digestats sur la fertilisation azotée n'a été observé pour des digestats solides (Odlare et al., 2014) et bruts (Zicker et al., 2020).

Des essais à long terme pourraient être très intéressants pour compléter notre connaissance sur les propriétés amendantes des digestats (amélioration de la fertilité physique du sol, augmentation de la fourniture en azote à long terme).

1.2.5 Effet des digestats sur la vie du sol

L'effet de l'application des digestats sur la mésofaune du sol est encore mal caractérisé (Möller, 2015). Seules quelques études se sont intéressées à l'effet de l'épandage de digestats, répétés ou non, sur les populations de vers de terre au champ. Pourtant, le changement de qualité de la matière organique pendant la digestion pourrait influencer sa qualité nutritive pour la faune et la microfaune du sol (Ernst et al., 2008; Onrust et Piersma, 2019; Sizmur et al., 2017). Butt et Putwain, (2017), Clements et al. (2012) et Koblenz et al. (2015) ont observé des abondances et/ou des biomasses de vers de terre plus fortes après épandage de digestats par rapport à un traitement sans apport de PRO. Les abondances de vers de terre sont en général similaires, en comparaison avec des traitements utilisant des effluents d'élevage non digérés, mais les communautés de vers de terre peuvent être différentes (Koblenz et al., 2015). D'autres études montrent une absence d'effet, voire une diminution de l'abondance de vers de terre, en comparaison aux traitements non amendés, après quelques mois ou quelques années d'apports répétés (Bermejo et al., 2010; Frøseth et al., 2014; Rollett et al., 2020). En plus de ces effets à long terme, une mortalité des vers de terre en surface a déjà été observée juste après épandage de digestat liquide (Burmeister et al., 2015; Johansen et al., 2015; Løes et al., 2014). Ces évènements ne semblent pas affecter les populations après quelques années d'apports répétés. Ce phénomène a également été observé après des apports de lisiers (Curry, 1976; Curry et al., 1980; Hansen, 1996; Van Vliet et de Goede, 2006). L'augmentation de la teneur en azote ammoniacal des digestats est une hypothèse possible sur sa toxicité à court terme (Curry, 1976; Hughes et al., 2008). Les effets des digestats sur les vers de terre à court et long terme restent à explorer.

De façon similaire, le changement de qualité de la matière organique des digestats pourrait affecter la vie du sol. À court terme, on observe des pics d'activité microbienne après application de digestat (Alburquerque et al., 2012; Odlare et al., 2014). À plus long terme, Wentzel et al. (2015) montrent une diminution de la biomasse microbienne rapportée à la quantité de C du sol lors de l'utilisation de digestat par rapport à l'utilisation d'effluents d'élevage. Pourtant, d'autres études semblent montrer l'inverse (Möller, 2015). L'effet des épandages répétés de digestat sur les microorganismes du sol reste encore peu caractérisé. Il est donc nécessaire de continuer de s'intéresser à l'effet des digestats sur la vie du sol.

1.2.6 Digestat et émissions de gaz à effet de serre, dont le N_2O

Les émissions de N_2O au champ sont liées à des processus microbiens. On présente souvent deux processus à l'origine des émissions de N_2O : la nitrification (transformation de NH_4^+ en NO_3^-) et la dénitrification (transformation de NO_3^- en N_2). La nitrification est essentiellement réalisée par des organismes autotrophes, en condition aérobie, même si des organismes hétérotrophes nitrificateurs existent. Les organismes dénitrificateurs sont en général hétérotrophes, la dénitrification ayant lieu en présence de matière organique et en condition anaérobie (Wrage et al., 2001). Néanmoins, il s'agit d'une vision simplifiée, car il existe d'autres voies métaboliques microbiennes qui peuvent en fait être responsables des émissions de N_2O (Butterbach-Bahl et al., 2013). Les deux principaux déterminants

des émissions de N_2O au champ sont la température et l'humidité du sol. L'humidité du sol influe sur la disponibilité en oxygène pour les microorganismes (Butterbach-Bahl et al., 2013). D'autres facteurs influençant les émissions de N_2O sont la texture, le pH et la densité apparente du sol, la quantité et la forme de l'azote dans le sol, la présence de polluant pouvant inhiber les réactions microbiennes. Les émissions de N_2O sont particulièrement variables spatialement et temporellement (Williams et al., 1999), ce qui rend leur estimation complexe.

Lors de l'application de PRO ou d'engrais azoté minéraux, on observe en général un pic d'émissions de N_2O. La fertilisation avec des PRO apporte de la matière organique facilement dégradable, ce qui pourrait favoriser les activités microbiennes et donc les émissions de N_2O (Butterbach-Bahl et al., 2013; Köster et al., 2011, 2015). Zhou et al. (2017) ont ainsi proposé que les effluents d'élevage auraient tendance à augmenter les émissions de N_2O, par rapport aux engrais azoté minéraux, mais que cela dépend du type de sol et pourrait dépendre de la nature des effluents. Skinner et al. (2014) montrent que les émissions de N_2O lors de fertilisations organiques sont inférieures à celles émises lors de l'utilisation d'engrais de synthèse pour une même surface, mais supérieures si on les rapportent à un même rendement. Les facteurs d'émissions associés à l'épandage de PRO, exprimés en proportion de l'azote total apporté, sont assez variables, et peuvent en moyenne varier de 0,03% à 2,4% (Charles et al., 2017). Un C/N faible et une quantité importante de C facilement dégradable semblent favoriser les émissions de N_2O. Ces deux points concernent les digestats, qui sont classés comme des PRO sensibles aux émissions de N_2O, de même que les lisiers, en opposition à d'autres PRO comme les composts (Charles et al., 2017). Des facteurs d'émissions de N_2O associés à l'épandage de digestat sont présentés dans le Tableau 1.5.

Les émissions de N_2O après apports de digestats ont été étudiées de façon assez importante à l'échelle de la parcelle (Häfner et al., 2021; Möller and Stinner, 2009; Nicholson et al., 2017; Verdi et al., 2019). Les émissions de N_2O au champ après apports de digestats ont été rapportées supérieures à celles suivant les apports d'engrais minéraux par Buchen-Tschiskale et al. (2020), Köster et al. (2011) et Verdi et al. (2019), et similaires par Köster et al. (2015) et Pezzolla et al. (2012). Toujours à l'échelle de la parcelle, les émissions de N_2O après apports de digestats liquides ou bruts ont été mesurées comme étant en général plus faibles ou similaires que celle des lisiers (Köster et al., 2015; Möller, 2015; Nkoa, 2014; ten Huf and Olfs, 2020), ce qui pourrait être expliqué par la présence de moins de carbone facilement dégradable dans le digestat.

Un certain nombre de pratiques peuvent influencer les émissions de N_2O. La séparation de phase des digestats a été rapportée comme une mesure qui diminue les émissions de N_2O au champ (Askri et al., 2016; Möller, 2015). Aucun effet n'a été mesuré par Köster et al. (2015). Au champ, la quantité d'azote apporté et les conditions pédo-climatiques semblent être les principaux facteurs explicatifs des émissions ; en comparaison, le type de digestat n'aurait qu'un effet limité sur les

émissions (Häfner et al., 2021). L'apport rapproché de digestats et d'engrais minéraux pourrait augmenter les facteurs d'émissions, comme montré sur différents PRO (Charles et al., 2017). L'injection des digestats liquides dans le sol, qui limite les émissions d'ammoniac, semble favoriser les émissions de N_2O (Herr et al., 2019). La date d'application du digestat impacte probablement les émissions de N_2O, comme montré par Bell et al. (2015) après des applications de lisier.

Les émissions de N_2O sont susceptibles d'évoluer à long terme en cas d'apports répétés de digestats. Les émissions de N_2O peuvent ainsi être influencées par les épandages antérieurs de PRO (Rosace et al., 2020). L'augmentation des stocks d'azote dans les sols à long terme pourraient aussi augmenter le potentiel des sols à émettre du N_2O, ce qui demande à être étudié (Guenet et al., 2021).

La thématique des émissions de N_2O est largement explorée dans la littérature. Une limite des connaissances actuelles est due au fait que les émissions de N_2O sont sensibles à beaucoup de pratiques, que cette sensibilité peut varier en fonction des études, et que cette sensibilité est souvent masquée par des effets pédoclimatiques. En conséquence, les résultats obtenus sur l'effet de la digestion des effluents, de la séparation de phase des digestats, des dates d'apports, des différents types de digestats, restent encore peu sûrs dans l'état des connaissances actuelles.

Enfin, quelques articles mesurent des émissions de méthane (CH_4) lors de l'épandage de digestats au champ. Les émissions de CH_4 sont en général très faibles, de l'ordre de 0.5 à 1% des émissions de CO_2 induites par l'épandage (Czubaszek and Wysocka-Czubaszek, 2018; Holly et al., 2017; Pezzolla et al., 2012; Rosace et al., 2020; Tiwary et al., 2015).

Tableau 1.5 – Quelques facteurs d'émissions de N_2O associées à l'épandage de digestats

Référence	Facteur d'émissions de N_2O (% N total)
Charles et al. (2017)	0,9
Nicholson et al. (2017)	0,3 – 0,5
Häfner et al. (2021)	0,2 – 0,7
Möller et Stinner (2009)	1,3 - 1,4
(IPCC, 2019) (cattle effluents)	0,6

1.2.7 Digestat et lixiviation de nitrate

L'azote sous forme de nitrate (NO_3^-) présent dans les sols agricoles peut être lixivié et être emporté dans les eaux de surface et souterraines. La lixiviation des nitrates est une des sources de l'eutrophisation des eaux de surface, avec, entre autres, des risques de perte de biodiversité. Une eau avec une trop forte concentration en nitrates est impropre à la consommation, ce qui est une cause de fermeture de captage d'eau potable (Pinay et al., 2017; SDES and OFB, 2020).

La lixiviation des nitrates est causée lors d'une période de drainage coïncidant avec une période où la concentration en nitrates dans les sols est importante. Dans les climats tempérés français, ceci est typiquement le cas en fin d'automne et début d'hiver. Les mesures qui limitent la lixiviation du nitrate consiste à diminuer la réserve en nitrates dans les sols avant les périodes de drainage, par exemple par la régulation et l'optimisation des apports d'azote, ou la mise en place d'un couvert végétal au lieu d'un sol nu pour capter l'azote du sol (Di and Cameron, 2002).

Comme avec les autres PRO, les digestats peuvent être à l'origine de lixiviation de nitrates (Nicholson et al., 2017). Celle-ci est principalement influencée par les pratiques agricoles liées à l'épandage de PRO (cultures, dates, doses), ainsi que par le type de sol et les conditions météorologiques (Möller, 2015; Räbiger et al., 2020). Avec une approche de modélisation, Räbiger et al. (2020) suggèrent qu'en cas d'apport d'azote minéral similaire avec des digestats et des engrais synthétiques, les digestats pourraient causer une lixiviation légèrement plus importante, entre autres, en raison de la minéralisation de l'azote organique des digestats. L'azote lessivé représente 6 à 18% de l'azote apporté dans la simulation.

1.3 Modification des flux de C et N à l'échelle de l'exploitation avec la méthanisation

1.3.1 Comment la méthanisation influe-t-elle sur les flux de C et N à l'échelle de l'exploitation ?

Dans la section 1.1 , nous avons vu que les digestats avaient des caractéristiques physicochimiques différentes de celles des effluents d'élevage non digérés, et que cela pouvait changer les cycles du N et du C lors de leur épandage au champ. Néanmoins, à l'échelle d'une exploitation agricole, l'import de déchets à digérer va influencer à la fois les caractéristiques des digestats, mais aussi leurs quantités. La gestion des PRO initiaux, et en particulier leur temps et conditions de stockage, peut être modifiée, entraînant une modification de leurs caractéristiques. Le système agricole peut aussi être influencé lors de l'introduction d'un méthaniseur, par exemple au niveau de la gestion des résidus de cultures, de la gestion de la fertilisation ou de l'assolement. Ces changements, présentés dans la Figure 1.4, vont influencer les cycles du C et du N. L'échelle de la parcelle agricole ne permet pas la considération de ces effets. Aussi, l'échelle de l'exploitation semble donc pertinente pour analyser les conséquences de la méthanisation. Cette section détaille les différents effets de l'introduction d'un méthaniseur sur les flux de C et N à l'échelle de l'exploitation.

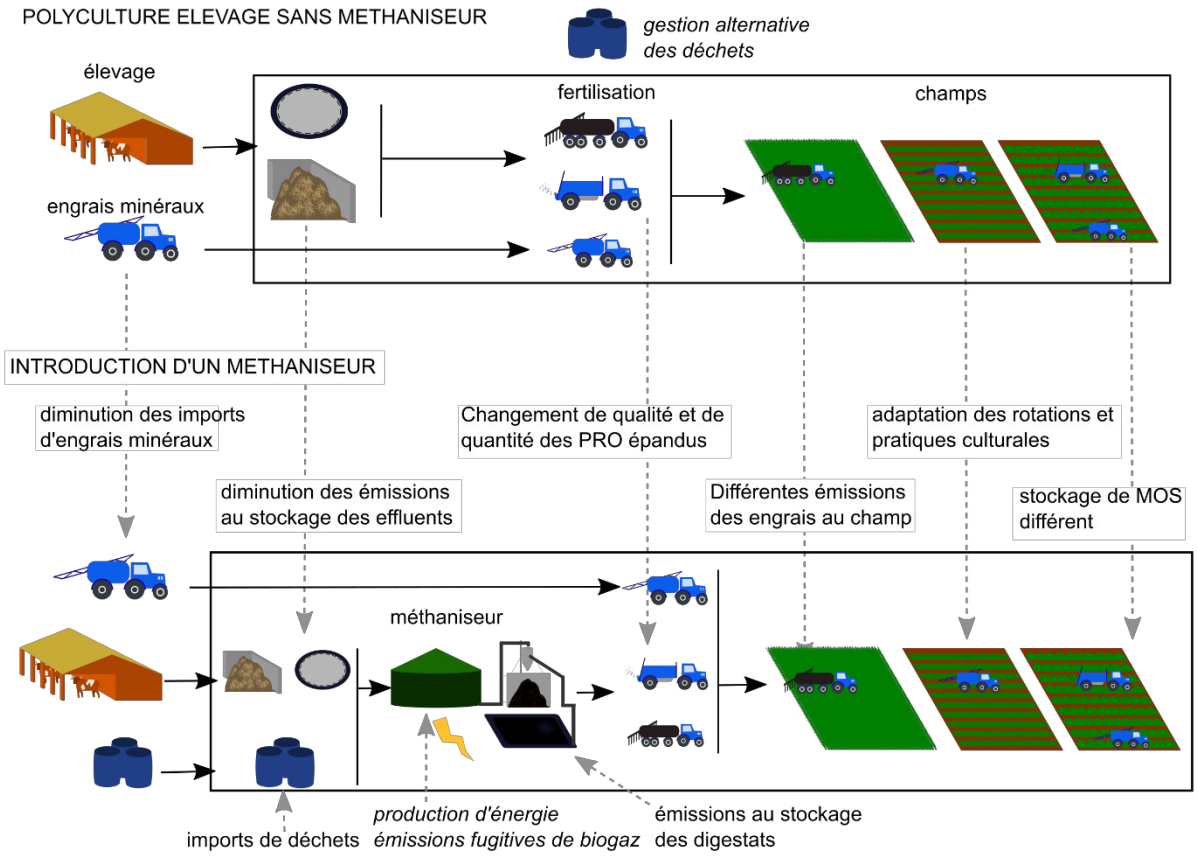

Figure 1.4 – L'introduction d'un méthaniseur dans une exploitation en polyculture élevage influence les cycles du C et du N. Le cadre représente les limites du système, c'est-à-dire l'exploitation, en excluant l'élevage que l'on suppose non modifié lors de l'installation d'un méthaniseur.

1.3.2 Bilans C et N à l'échelle de la ferme et effet de l'import de déchets extérieurs

Lors de la méthanisation agricole, les effluents sont souvent codigérés et des déchets extérieurs peuvent donc être importés sur la ferme pour la méthanisation. Cela a pour conséquence un import de nutriments avec ces matières organiques, et donc des flux supplémentaires de MO et N vers les sols via les digestats. À l'aide d'un bilan de masse sur un méthaniseur, Camarillo et al, (2013) montrent que ces apports de déchets extérieurs représentent une fraction non négligeable de la matière organique dans le digesteur et donc de la production totale de biogaz. Ils contribuent également à augmenter les nutriments présents dans le digesteur. La digestion est en général conservative pour les nutriments comme le N, le P, et le K, et la masse de digestat est très proche de la masse d'intrants (Banks et al., 2011; Camarillo et al., 2013; Schievano et al., 2011). On peut donc dire que ces déchets extérieurs causent une augmentation de la quantité de digestat à épandre, ainsi qu'une augmentation des nutriments disponibles dans les PRO à l'échelle de la ferme. L'importance de cet effet dépend en toute vraisemblance de la quantité de déchets importés, et n'a que très peu été caractérisée. Möller (2009) a réalisé des estimations de bilan de C et N à l'échelle d'une ferme en polyculture élevage, avec ou sans méthanisation. Il estime que la digestion

des effluents bovins seule ne va que peu affecter les bilans C et N à l'échelle de la ferme, mais que l'import de substrats extérieurs pourrait augmenter les apports de C et N au champ, avec comme conséquences plus de fuites azotées et plus de stockage de MOS. Cette augmentation de la quantité de nutriments peut contribuer à la diminution des achats d'engrais de synthèse. Ainsi, en France, l'installation d'un méthaniseur peut permettre une diminution des achats d'engrais synthétiques azotés de 20% dans certains cas (ADEME et SOLAGRO, 2018). L'étude ne permet par contre pas d'interpréter à quel point cette diminution de l'achat d'engrais est liée à l'import de nutriments sur la ferme, par rapport à d'autres effets possibles comme le changement de qualité des PRO (effluents d'élevage ou digestats) ou le changement de l'assolement.

Les flux de nutriments à l'échelle de la ferme ont été utilisés pour évaluer la performance environnementale de différentes fermes (Bassanino et al., 2007; Haas et al., 2007; van Leeuwen et al., 2019; Watson et al., 2002). Ils peuvent être utilisés comme outil de dialogue avec les agriculteurs pour améliorer les performances environnementales des exploitations (Fangueiro et al., 2008b). Ces approches pourraient être utilisées pour étudier les fermes pratiquant la méthanisation.

1.3.3 Changement de pratiques agricoles liées à l'introduction d'un méthaniseur

À l'échelle d'une exploitation agricole en polyculture élevage, l'introduction d'un méthaniseur peut causer un changement de pratiques agricoles. Tout d'abord, l'infrastructure de la ferme peut changer, en particulier la capacité de stockage des PRO et la quantité de PRO à épandre. Cela peut induire un changement des dates d'applications des PRO de par l'allègement ou le renforcement des contraintes logistiques. L'assolement peut être influencé par l'introduction d'un méthaniseur. Dans la région de Hesse en Allemagne, Lüker-Jans et al. (2017) ont ainsi montré une corrélation entre l'introduction de la méthanisation et l'augmentation des surfaces de maïs, récolté comme culture énergétique. Toujours en Allemagne, Möller et al. (2011) observent aussi une importante augmentation des cultures de maïs sur 14 fermes après l'implantation d'importants méthaniseurs à proximité ; près d'un tiers de l'assolement de ces fermes sont dédiés à la culture de maïs en tant que culture énergétique. En France, les cultures principales ne peuvent pas excéder 15% des intrants du méthaniseur ; le changement d'assolement reste donc plus limité. Néanmoins, des cultures intermédiaires à vocation énergétique (CIVE) peuvent être ajoutées à l'assolement (ADEME et SOLAGRO, 2018), ce qui peut tendre à modifier la rotation. Les effets des cultures intermédiaires (diminution de la lixiviation, de l'érosion et du drainage, demande en eau, compétition avec les adventices, stockage de carbone, engrais vert) sont également attendus avec des CIVE, mais l'augmentation du temps de couverture, les dates de semis différentes, et leur modalité de récolte peuvent modifier l'amplitude des effets (Constantin et Launay, 2020). Enfin, les résidus de cultures, comme les pailles, peuvent être exportés du champ et méthanisés, plutôt qu'enfouis.

Ces changements de pratiques culturales peuvent avoir des effets sur les cycles de l'azote et du carbone au champ. Du point de vue de l'azote, ils peuvent affecter la lixiviation de nitrate (Möller, 2015):

- La saison d'application des PRO peut être modifiée ; les apports d'automne favorisent la lixiviation par rapport aux apports de printemps.
- Le changement du type de culture intermédiaire peut influencer la lixiviation.
- En cas de changement d'assolement, les nouvelles cultures peuvent êtes plus ou moins sensibles à la lixiviation par rapport aux anciennes.

Lors de la méthanisation des cultures intermédiaires, leur export plutôt que leur enfouissement peut diminuer la minéralisation de l'azote. L'azote ainsi exporté est réappliqué via les digestats, à un autre moment voire sur un autre champ. Cela change les dynamiques de l'azote dans le sol, ce qui peut influer sur les émissions de N_2O (Brozyna et al., 2013). Ainsi, Möller and Stinner (2009) ont comparé des systèmes de culture sur une ferme avec ou sans méthanisation. Les auteurs montrent que la récolte des cultures intermédiaires diminue les émissions de N_2O juste après leur récolte et que le report d'application de cet azote via les digestats augmente les émissions de N_2O juste après la fertilisation : au total, la récolte et la méthanisation des cultures contribue globalement à diminuer les émissions de N_2O.

Un changement important de l'assolement peut affecter les quantités d'engrais de synthèse utilisées. Möller et al. (2011) observent, par exemple, une augmentation des achats d'engrais azotés après que les surfaces de maïs aient augmenté dans l'assolement. En ce qui concerne la volatilisation d'ammoniac, les rotations culturales peuvent jouer sur les dates d'apport et donc les conditions météorologiques et l'importance de la volatilisation (Quakernack et al., 2012).

Du point de vue du carbone, l'augmentation de production de CIVE peut augmenter le stockage de C du sol via les apports de C racinaire. À l'inverse, la méthanisation peut induire un nouvel export de biomasse auparavant enfouie (culture intermédiaire ou résidus de culture). La matière organique des végétaux est alors méthanisée puis retournée au sol sous forme de digestat. Des interrogations se posent pour savoir si cette pratique induit ou non une diminution des stocks de MOS (Constantin et Launay, 2020).

En conclusion, l'introduction d'un méthaniseur peut influencer le système agricole de l'exploitation. Pour de nombreuses raisons, ceci peut influencer les cycles du C et du N au champ. Ces changements ont des impacts multiples, qui sont encore difficiles à synthétiser. Il y a donc un intérêt fort à étudier l'effet du remplacement des effluents d'élevage par des digestats avec des approches qui considèrent l'ensemble de la rotation agricole et de la sole de l'exploitation, et de mener des études qui considèrent l'ensemble des flux d'azote et de carbone, qui interagissent fortement entre eux.

1.3.4 Emissions gazeuses lors du stockage des PRO, émissions fugitives du méthaniseur

Lors de leur stockage, les PRO sont sujets à des émissions gazeuses, incluant des émissions de CO_2, CH_4, N_2O, NH_3. Les émissions gazeuses issues du stockage des lisiers et fumiers ont été intensément étudiées, en différentes conditions, ce qui permet de connaître les ordres de grandeur de ces émissions, et les facteurs qui les influencent (Chadwick et al., 2011; Kupper et al., 2020; Vigan et al., 2019; Walling et Vaneeckhaute, 2020b). Ces émissions gazeuses sont dues à différents processus biologiques et physico-chimiques. Les microorganismes décomposent la matière organique des PRO, en conditions aérobies (émissions de CO_2) ou anaérobies (émissions de CO_2 et de CH_4), ce qui cause également la minéralisation de l'azote organique. Quand les conditions sont aérobies, par exemple lors du stockage des PRO solides, ou sur la croûte de surface lors du stockage des lisiers, l'azote minéral peut être transformé par les microorganismes par nitrification ($NH_4^+ \rightarrow NO_3^-$) et dénitrification ($NO_3^- \rightarrow N_2$), à l'origine d'émissions de N_2O et de N_2 (Chadwick et al., 2011). De la même façon qu'au sol après épandage, les ions ammonium sont en équilibre avec l'ammoniac gazeux, qui peut volatiliser (émissions de NH_3). Les PRO liquides (lisiers, digestats bruts, digestats liquides) sont donc soumis à des émissions de CO_2, CH_4, NH_3 (Amon et al., 2006; Owen and Silver, 2015). Des émissions de N_2O et de N_2 sont également possibles, même si elles ne sont pas toujours détectées (Clemens et al., 2006; Rodhe et al., 2015). Les PRO solides (fumiers, digestats solides) sont soumis à des émissions de CO_2, NH_3, N_2O et N_2, et dans une moindre mesure aux émissions de CH_4 (Amon et al., 2006; Owen and Silver, 2015). Des exemples de facteurs d'émissions de CH_4, N_2O, et NH_3 sont indiqués dans le Tableau 1.6

Différents facteurs peuvent influencer les émissions gazeuses lors du stockage des PRO. Lors du stockage des lisiers ou des digestats, une croûte naturelle peut se former, ce qui affecte les émissions gazeuses : il s'agit d'une couverture naturelle qui limite les échanges gazeux et d'une zone où se côtoient des conditions aérobies et anaérobies, ce qui peut aussi augmenter les émissions de N_2O (Chadwick et al., 2011). La couverture d'une lagune de stockage de PRO liquide diminue fortement les émissions d'ammoniac (60% à 90% de réduction) (Clemens et al., 2006; Kupper et al., 2020). Une couverture peut également diminuer les émissions de CH_4, mais l'efficacité est moins prononcée que pour le NH_3 (Clemens et al., 2006; Kupper et al., 2020; Rodhe et al., 2015). Il est possible de récupérer le méthane émis lors du stockage des digestats à l'aide d'une couverture spécifique (Balsari et al., 2013). Les effets sur les émissions de N_2O sont variables (Kupper et al., 2020). L'acidification ou la dilution des PRO peuvent également réduire les émissions (Kupper et al., 2020). La hausse de la température est connue pour favoriser les émissions gazeuses au stockage (Baldé et al., 2018; Gioelli et al., 2011; Kaparaju and Rintala, 2003; Kimura et al., 2019; Sommer, 1997). La vitesse du vent peut aussi impacter la volatilisation d'ammoniac (Baldé et al., 2018). Une des conséquences est un possible effet de la gestion des stocks de digestat sur les émissions : la période de l'année à laquelle les digestats sont stockés ou épandus pourrait influencer les émissions, comme montré par modélisation (Ericsson et al., 2020).

Malgré les nombreuses mesures rapportées dans la bibliographie, la diversité des PRO, de l'environnement de stockage (environnement, durée, température), des méthodes de mesures et des méthodes de gestion des effluents (retournement et/ou compaction pour les PRO solides, présence et type de couverture pour les PRO liquides, séparation de phase) complexifie la synthèse de ces émissions (Kupper et al., 2020; Vigan et al., 2019). Les émissions lors du stockage des effluents contribuent de manière importante aux émissions de GES à l'échelle d'une ferme (Baral et al., 2018). De par leur importance, ces émissions vont donc avoir un effet sur la qualité des digestats.

Lors de l'introduction d'un méthaniseur, les effluents d'élevage sont souvent utilisés directement dans le digesteur avec un stockage réduit. Il y a donc une réduction des émissions au stockage des effluents non digérés, mais en contrepartie des émissions ont lieu au stockage du digestat. Les émissions au stockage des digestats sont influencées par plusieurs facteurs :

- La méthanisation induit un changement de teneurs en C et en N et peut changer la masse totale de PRO. La quantité absolue de matière pouvant être soumise aux émissions sera donc différente en présence ou absence de méthaniseur.
- La méthanisation stabilise la matière organique et minéralise une partie de l'azote organique. Ce changement de qualité des PRO peut influencer les émissions.
- En fonction de la nature des effluents (lisier liquide et/ou fumier solide) et de la séparation de phase ou non du digestat (stockage de digestat brut ou de digestats liquides et solides), les conditions aérobies ou anaérobies de stockage sont modifiées, ce qui va donc influencer les émissions.
- La durée de stockage peut être modifiée.
- Les infrastructures de stockage peuvent être modifiées, ce qui impacte les émissions.

La méthanisation consomme une partie de la matière organique, le carbone est exporté sous forme de biogaz. Durant la méthanisation, la matière organique résiduelle se stabilise. Les émissions de méthane au stockage des digestats sont donc en général plus faibles que celles des effluents non digérés (Amon et al., 2006; Clemens et al., 2006; Maldaner et al., 2018), même si l'inverse a parfois été montré (Rodhe et al., 2015). Les émissions de méthane lors du stockage des digestats sont tout de même non négligeables. La concentration en azote ammoniacal est plus élevée dans les digestats que dans les effluents non digérés, ce qui peut causer un plus grand risque de volatilisation d'ammoniac (Baldé et al., 2018; Clemens et al., 2006; Neerackal et al., 2015), même si l'inverse a aussi été mesuré (Amon et al., 2006). La méthanisation semble augmenter les émissions de N_2O, ce qui pourrait être expliqué par la plus forte teneur en azote des digestats en comparaison avec les effluents d'élevage non digérés (Amon et al., 2006; Clemens et al., 2006). Aucun effet significatif n'a été trouvé par Kupper et al. (2020), en terme de facteur d'émission de N_2O.

Tableau 1.6 – Exemples de facteurs d'émissions au stockage des PRO.

PRO	Gaz			
	NH_3	N_2O	CH_4	CO_2
Fumier	6% N_{tot} [a]	3% N_{tot} [a]	2%C [a]	32%C [a]
Lisier bovin	14 – 19%N_{amm} [b] 16 – 50% [c]	0,1 – 0,2%N_{tot} [b]	0.2 - 0.4 gCH_4 $kgVS^{-1}$ (environ 3 – 12%C) [b] 9%C [d]	11% gCO_2 $kgMO^{-1}$ (~ 6%C) [b] 34% [d]
Digestat liquide ou brut	30 – 40% N_{amm} [c,e]	0%N_{tot} (uncovered) [f] 0,24%N_{tot} (covered) [f]	1% - 12% (% du CH_4 dans le biogaz) [g,h] 0,1 – 0,3 gCH_4 $kgMO^{-1}$ j^{-1} (~ 5 – 16% C) [g,i]	Même volume que CH_4 i
Digestat solide	10% - 12% (%N_{tot}) [d]	0,1% – 0,3% (%N_{tot}) [d]	0,5% - 4% (%C) [d]	30% - 40% (%C) [d]

[a] (Hassouna et al., 2008). [b] (Kupper et al., 2020). [c] (Baldé et al., 2018). [d] (Amon et al., 2006). [e] (Perazzolo et al., 2016). [f] (Rodhe et al., 2015). [g] (Baldé et al., 2016). [h] (Hrad et al., 2015; Liebetrau et al., 2013). [i] (Maldaner et al., 2018). [j] (Dietrich et al., 2021; Li et al., 2018). N_{tot} : azote total. N_{amm} : azote ammoniacal. MO : matière organique.

La séparation de phase des effluents non digérés ou des digestats affecte les émissions : la phase liquide est moins riche en matière organique, la probabilité de présence d'une croûte naturelle sur la lagune de stockage est plus faible, et la phase solide est stockée en condition aérobie. Sans tenir compte des émissions de la fraction solide, la séparation de phase des effluents non digérés augmente les émissions d'ammoniac (absence de croûte), diminue les émissions de N_2O (absence de croûte) (Kupper et al., 2020) et de CH_4 (moins de matière organique) (Gioelli et al., 2011; Kupper et al., 2020). De plus, de fortes émissions de N_2O peuvent être mesurées lors du stockage de la phase solide (Holly et al., 2017). Concernant les effluents animaux non digérés, la séparation de phase aurait plutôt tendance à augmenter les émissions totales de N_2O et diminuer celles de CH_4 (Fangueiro et al., 2008a), les mêmes conclusions sont obtenues par Holly et al. (2017) lors de la séparation de phase de digestats. Perazzolo et al. (2016) et Baldé et al. (2018) montrent un risque d'augmentation globale des émissions de NH_3 après séparation de phase des digestats. Baldé et al. (2018) explique ce résultat par l'absence de croûte sur la lagune de stockage du digestat liquide, comparé au digestat brut.

En plus des émissions au stockage des digestats, des émissions fugitives de biogaz peuvent avoir lieu sur les plateformes de méthanisation (fuites, combustion incomplète) (Flesch et al., 2011; Groth et al., 2015; Hrad et al., 2014, 2015; Reinelt et al., 2016). Hrad et al. (2015) résument des travaux précédents et estiment que les fuites de gaz peuvent représenter 1 à 6% de la production de biogaz, ce qui comprend les émissions au stockage des digestats.

1.3.5 Intérêt de la prise en compte des émissions lors du stockage et de l'épandage des PRO

La gestion des effluents est composée d'étapes successives (traitements, stockage, épandage). Chacune de ces étapes est soumise à des émissions et à des modifications des caractéristiques physico-chimiques des effluents, ce qui peut avoir des répercussions sur les émissions lors des étapes suivantes. Neerackal et al. (2015) montrent par exemple que le stockage influence la qualité des digestats (matière sèche, azote ammoniacal), ce qui pourrait influer sur leurs effets au champ. Ainsi, l'évaluation des effets d'une nouvelle pratique (séparation de phase, couverture de fosse de stockage) devrait considérer l'ensemble du « continuum de gestion des effluents » (« *manure management continuum* », (Chadwick et al., 2011). Les émissions gazeuses au stockage des digestats doivent être mises en regard des émissions au champ.

Cinq études consacrées aux émissions gazeuses lors du stockage puis de l'épandage ont été recensées (Amon et al., 2006; Baral et al., 2017, 2018; Dietrich et al., 2020, 2021; Holly et al., 2017; Neerackal et al., 2015). Comme vu précédemment, la méthanisation diminue les émissions de CH_4 au stockage, qui sont plus importantes que celles à l'épandage (Amon et al., 2006; Dietrich et al., 2020, 2021; Holly et al., 2017). La méthanisation peut impacter de façon antagoniste les émissions de N_2O et de NH_3 à l'épandage ou au stockage (Amon et al., 2006; Baral et al., 2017, 2018; Holly et al., 2017;

Neerackal et al., 2015). Aucune tendance n'est trouvée quant aux sens des effets de la méthanisation sur la volatilisation de NH_3 au stockage, à l'épandage, ou total (Amon et al., 2006; Holly et al., 2017; Neerackal et al., 2015). La méthanisation tend à favoriser les émissions de N_2O au stockage et à les diminuer à l'épandage, mais l'effet global résultant varie selon les études (Amon et al., 2006; Holly et al., 2017). Enfin, si les émissions de CH_4 sont toujours importantes dans le bilan des émissions de GES, l'importance des émissions de N_2O varie, et il n'y a pas de consensus pour dire si les émissions de N_2O au champ ou au stockage sont les plus importantes (Amon et al., 2006; Baral et al., 2017, 2018; Dietrich et al., 2020, 2021; Holly et al., 2017).

Dietrich et al. (2020, 2021) ont mesuré l'effet du compostage de digestats solides sur les émissions de GES. Les auteurs montrent que le compostage réduit les émissions de N_2O au champ, mais augmente celles de CH_4 lors du stockage, avec au final un effet variable sur les émissions totales de GES. Nous n'avons pas trouvé d'études expérimentales étudiant l'effet d'autres pratiques de gestion du digestat (séparation de phase, couverture de fosse) en considérant l'enchaînement de leur stockage et de leur épandage.

Si les ordres de grandeurs et les facteurs influençant les émissions lors du stockage des digestats et lors de leur application au champ sont connus (sections 1.2.2 , 1.2.6 et 1.3.4), la résultante de ces émissions n'est pas bien caractérisée. En effet, une évolution dans la gestion des digestats peut avoir des effets opposés au niveau des différentes sources d'émissions. Il est donc important de considérer les deux sources d'émissions (traitement – stockage et épandage) conjointement. Néanmoins, les études expérimentales considérant ces deux sources sont rares, voire insuffisantes pour bien comprendre l'effet global de la méthanisation sur les émissions gazeuses.

1.4 Evaluation environnementale de la méthanisation

1.4.1 Quelles limites ont les approches expérimentales lors de l'évaluation environnementale de la méthanisation ?

Les sections 1.1 et 1.3 ont permis de décrire les différents processus liés aux cycles du C et du N qui étaient affectés par l'introduction d'un méthaniseur dans une ferme en polyculture élevage. Néanmoins, l'évaluation environnementale d'une pratique comme la méthanisation nécessite de comparer ces impacts les uns par rapport aux autres, de les hiérarchiser, et de considérer toutes les interactions qui peuvent intervenir entre les différentes sources d'impacts sur la ferme.

Certaines études expérimentales peuvent proposer des approches pour répondre à ce besoin de synthèse et d'évaluation multicritère. Par exemple, en ce qui concerne l'épandage de PRO au champ, Obriot et al. (2016) comparent l'effet de différents PRO sur la fertilité des sols (fertilité physique, fertilité biologique, production agricole), la biodiversité du sol, ou encore la contamination du sol. Clements et al. (2012) s'intéressent ainsi à l'effet des digestats au champ sur la fertilisation des cultures et la macrofaune du

sol. La section 1.3.5 décrit des études qui synthétisent les émissions de GES et de NH_3 induites par la méthanisation des effluents d'élevage, en considérant le stockage et l'épandage des PRO. Néanmoins, l'utilisation de l'expérimentation seule reste limitée dans cet objectif d'évaluation synthétique d'une pratique. La prise en compte de nombreuses mesures, dont certaines peuvent être complexes (lixiviation de nitrates, stockage de carbone dans les sols), limite le nombre de traitements et de cas d'études. Il est donc difficile d'étendre les résultats de telles études à d'autres systèmes. Les études utilisant la modélisation permettent de résoudre ce problème. Les expérimentations permettent de calibrer des modèles, et ces modèles permettent de généraliser les résultats des expérimentations. Cette section s'intéresse donc aux approches de modélisation qui visent à évaluer les impacts de la méthanisation.

1.4.2 Les modèles : quelques définitions

Les modèles sont des outils qui, une fois conçus et calibrés sur des données expérimentales, peuvent permettre de répondre à des objectifs nouveaux. Il existe de multiples façons de caractériser et de différencier les modèles. Dans le cadre de cette synthèse bibliographique, on peut dire que les modèles sont caractérisés par (entre autres) :

- Le sujet modélisé. Nous décrivons ici des modèles qui s'intéresse à 4 types de sujets :
    - La digestion anaérobie (modèle de méthanisation)
    - Le stockage des PRO (modèle de stockage)
    - Le fonctionnement du sol et des cultures (modèle sol-plante)
    - L'exploitation agricole (modèle à l'échelle de l'exploitation).
- L'approche de modélisation utilisée. Certains modèles combinent plusieurs approches.
    - Les modèles empiriques modélisent les processus grâce à des lois empiriques, c'est-à-dire des relations issues d'observations, sans lien avec la théorie. Ces modèles utilisent en général peu de paramètres, mais ne peuvent pas être utilisés en dehors de leur domaine de calibration.
    - Les modèles mécanistes ou biophysiques modélisent des processus à l'aide d'équations issues de lois physiques ou biologiques. Ils sont donc plus facilement généralisables que les modèles empiriques.
    - Les modèles de type « boîte noire » utilisent un apprentissage « aveugle » sur un grand jeu de données observées pour pouvoir prédire les variables d'intérêt à partir de nouvelles données d'entrées. Ils incluent de nombreux types de modèles statistiques ou des réseaux de neurones. Leur objectif reste souvent la prédiction de variables d'intérêt.
- Leur complexité, qu'il s'agisse de la complexité du modèle ou de sa complexité d'utilisation. En général, un modèle complexe (grand nombre de paramètres et/ou de processus modélisés) est plus complexe d'utilisation.

- Leur objectif. Différents modèles peuvent avoir des objectifs différents pour un même sujet modélisé. Lors de la construction d'un modèle, l'objectif est un facteur clef de décision de l'approche et de la complexité utilisée.

La section 1.4.3 présente la nature et les objectifs de quelques modèles de méthanisation, de stockage ou de modèle sol-plante, qui peuvent être utilisés pour étudier la méthanisation. La section 1.4.4 présente l'intérêt des modèles à l'échelle de l'exploitation lors de l'étude de la méthanisation.

1.4.3 Utilisation des modèles pour modéliser la digestion, le stockage et l'épandage de digestats

Des modèles de méthanisation ont été développés pour, en premier objectif, prédire, comprendre et optimiser la production de biogaz lors de la méthanisation. Ces modèles peuvent être des modèles mécanistes, incluant entre autres des versions modifiées du modèle ADM1 ou des modèles de type « boîte noire » (Lauwers et al., 2013; Yu et al., 2013). Les modèles de méthanisation mécanistes peuvent être complexes et demandent, en général, une bonne description des intrants utilisés. Un autre objectif des modèles de méthanisation peut être la prédiction de la qualité des digestats, comme le modèle SYS-Metha (Bareha et al., 2021). Certaines versions d'ADM1 modifiées permettent la prédiction de certaines caractéristiques des digestats, mais souvent en se focalisant sur des caractéristiques influant la production de biogaz, comme la teneur et la nature de la matière organique ou la teneur en azote ammoniacal (Bareha et al., 2019; Lauwers et al., 2013).

D'autres modèles de méthanisation, plus simples et donc utilisables facilement, ont été créés, avec des objectifs plus opérationnels comme l'évaluation économique de l'introduction d'un méthaniseur, l'évaluation de la production potentielle de biogaz ou le dimensionnement du méthaniseur en fonction des intrants disponibles, ou l'estimation des impacts environnementaux associés à la méthanisation (Kythreotou et al., 2014). Les principales limites de ces modèles simples sont leur grande spécificité qui les rend peu généralisables, et leur simplicité, qui limitent leur utilisation pour résoudre des questions de recherche précises.

Des modèles de stockage des digestats ont été adaptés à partir de modèles de méthanisation pour estimer la production de biogaz dans le digesteur, ainsi que dans la cuve de stockage du digestat brut (Ericsson et al., 2020; Muha et al., 2015; Vergote et al., 2019). Il existe quelques modèles d'émissions de N_2O ou NH_3 lors du stockage de lisiers (Li et al., 2012). De nombreux modèles de compostage, dont certains peuvent modéliser les émissions gazeuses associées au compostage, ont été développés (Oudart et al., 2015; Walling et al., 2020). Ils pourraient être adaptés et utilisés pour modéliser le stockage des digestats solides. Les modèles de stockage permettent de comprendre les facteurs qui influencent les émissions au stockage. Ericsson et al. (2020) ont par exemple montré qu'une augmentation du temps de séjour du digestat dans le digesteur ou la vidange pluriannuelle de la lagune de stockage permettent de réduire les émissions de CH_4.

Si on s'intéresse au sujet du champ, plusieurs modèles ont été développés pour évaluer les flux de C et N dans le sol (Brilli et al., 2017). Ils peuvent considérer différentes échelles, allant du champ (modèle sol-plante par exemple) à la ferme. Les modèles sol-plante simulent la croissance de la plante, les caractéristiques du sol (par exemple, teneur en eau, teneur en azote minéral, MOS), et certains impacts environnementaux (par exemple, N_2O, NH_3, lessivage de nitrate). Certains parmi les plus utilisés dans le monde sont les modèles de la famille CERES (par exemple, Gabrielle et al., 1995), le modèle DayCent (Parton et al., 2015), le modèle DNDC (Li et al., 1992) et le modèle STICS (Brisson et al., 2008). Ce dernier est de plus particulièrement utilisé par la communauté scientifique française. Ces modèles permettent d'estimer ou d'extrapoler les flux non mesurables facilement, ou de prendre en compte l'impact d'un changement de pratiques à long terme (émissions de N_2O, lessivage de nitrates, stockage de MOS) (Jégo et al., 2012; Lugato et al., 2018; Plaza-Bonilla et al., 2018b). Les modèles sol-plante peuvent aussi servir comme outil de dialogue avec les agriculteurs pour proposer des améliorations de pratiques (Loussouarn et al., 2016).

Quelques études ont utilisé des modèles pour estimer les effets de l'épandage de digestats. Räbiger et al. (2020) ont, par exemple, modélisé la lixiviation des nitrates au champ pour évaluer l'effet de la fertilisation du colza avec du digestat à long terme (voir section 1.2.7). Häfner et al. (2021) utilisent le modèle de volatilisation d'ammoniac après épandage de lisier ALFAM2 (Hafner et al., 2019) pour estimer la volatilisation après épandage de digestat dans le cas d'une étude où ces émissions n'ont pas été mesurées, et estiment ainsi que les émissions indirectes de N_2O causées par la volatilisation de NH_3 représentent entre 2 et 6% des émissions de GES émis lors de l'épandage de digestats. Comme dernier exemple, Launay et al. (2021) ont utilisé le modèle sol-plante STICS pour estimer l'effet de l'augmentation de l'utilisation de PRO urbains, incluant des digestats : cela favoriserait fortement localement le stockage de MOS. Des pertes azotées supplémentaires sont aussi prédites. Les surfaces impactées seraient restreintes à l'échelle nationale.

Les modèles sol-plante peuvent être des outils utiles dans l'évaluation des effets de l'épandage de digestats, mais ils sont encore peu utilisés dans cette optique. La capacité de ces modèles à simuler des systèmes de culture utilisant des digestats devra également être évaluée.

1.4.4 Les modèles permettent de synthétiser les impacts à l'échelle de l'exploitation agricole

La section précédente (1.4.3) décrit comment il est possible d'utiliser des modèles pour étudier certains aspects de la méthanisation et de l'utilisation des digestats au champ. Néanmoins, le sujet d'un modèle peut aussi être l'ensemble de l'exploitation agricole.

Les modèles à l'échelle de l'exploitation agricole sont très courants pour étudier les exploitations de polyculture élevage (Del Prado et al., 2013; Petersen et al., 2007; Rotz, 2018; Schils et al., 2007). Certains modèles sont relativement simples et ont vocation à être utilisés comme outils d'aide à la

décision, tandis que d'autres sont plus complexes et plus mécanistes et ont pour but de répondre à des questions de recherche (Rotz, 2018). Selon Petersen et al. (2007), ces modèles à l'échelle de l'exploitation agricole permettent de prendre en compte les interactions entre différents aspects de l'exploitation de polyculture élevage : alimentation du bétail, qualité des effluents, gestion des effluents, par exemple. Cet article ne s'intéresse pas à l'application des effluents au champ, mais cet aspect s'intègre également dans la même réflexion. Cette aptitude est particulièrement intéressante, nous avons vu que ces questions étaient difficiles à répondre à l'aide de l'expérimentation seule (section 1.3.5). Pour Del Prado et al. (2013), un autre avantage des modèles à l'échelle de la ferme est qu'ils représentent un système à l'échelle duquel les décisions de gestion sont prises, et où il est donc possible de changer les pratiques pour améliorer le bilan environnemental de la filière.

Les modèles à l'échelle de l'exploitation sont généralement constitués de plusieurs modules, pouvant inclure l'élevage, le traitement et le stockage des effluents, la croissance des cultures au champ. Certains modèles utilisent un module de « règle de décisions » représentant les décisions des exploitants, ce qui permet de faire interagir ces différents modules entre eux de façon cohérente (Chardon et al., 2012; Vayssières et al., 2009). Des exemples de modèles à l'échelle de l'exploitation incluant la méthanisation des effluents d'élevage sont présentés dans le Tableau 1.7.

Les objectifs et les approches utilisés dans ces études sont variables. L'épandage des PRO peut être plus ou moins finement considéré, allant d'une non considération à l'utilisation de modèle sol-plante pour représenter les surfaces cultivées. Certains modèles sont très complexes et utilisent de nombreux modules mécanistes (Li et al., 2012; Rotz, 2018). Néanmoins, l'objet d'intérêt des modèles décrits n'est pas le méthaniseur, mais l'élevage : la méthanisation est vue comme une pratique possible à l'échelle de la ferme, et il n'est pas forcément possible de comparer différentes pratiques associées à la méthanisation. La co-digestion des effluents n'est pas forcément considérée. D'autres études ont considéré l'introduction de la méthanisation à la ferme en plaçant le méthaniseur au centre de leur système (Baral et al., 2018; Boldrin et al., 2016; Costa Junior et al., 2015; Vergote et al., 2019). Elles sont associées à une modélisation plus fine de la méthanisation et de la gestion des PRO. En revanche, l'épandage des digestats est modélisé simplement. Dans certains cas, la modélisation n'est qu'une façon de compléter le bilan GES après l'acquisition de mesures d'émissions au stockage ou au champ (Baral et al., 2018; Costa Junior et al., 2015). De plus, ces études se concentrent surtout sur le bilan carbone de la méthanisation, et peu sur ses autres aspects.

La méthanisation semble induire une réduction des émissions de GES au stockage des PRO (Baral et al., 2017; Rigolot et al., 2010; Rotz, 2018; Weiske et al., 2006). Vergote et al. (2019) montrent que l'effet de la diminution du stockage des effluents au bâtiment, causée par l'intérêt de méthaniser les PRO les plus frais possible, est un facteur très important de cette réduction des émissions de GES. Costa Junior et al. (2015) nuancent le propos en estimant des impacts au stockage des PRO similaires

pour une ferme avec ou sans méthaniseur. Il n'y a pas de consensus sur l'effet de la méthanisation sur les émissions de NH_3 (Rigolot et al., 2010; Veltman et al., 2018)

La méthanisation diminue les émissions totales de GES. Les principaux facteurs qui influencent le bilan carbone de la méthanisation sont la production de biogaz, les émissions au stockage des digestats, les fuites de biogaz (Baral et al., 2018; Boldrin et al., 2016; Costa Junior et al., 2015; Vergote et al., 2019; Weiske et al., 2006). L'épandage de digestats à la place des lisiers (changement des émissions de N_2O au champ, changement du stockage de MOS, diminution des engrais de synthèse) semble être moins important en terme de bilan carbone (Baral et al., 2018; Boldrin et al., 2016; Vergote et al., 2019). Néanmoins, ceci doit être vérifié à l'aide de modèles plus précis sur l'effet de l'épandage de digestats.

Les intrants extérieurs permettent d'augmenter la production de biogaz et donc d'améliorer le bilan carbone (Boldrin et al., 2016; Weiske et al., 2006). La méthanisation des cultures a un effet variable : en cas de bon rendement, le bénéfice issu du surplus de production de biogaz est plus important que les aspect négatifs liés à la production des cultures ; l'inverse peut également être constaté en cas de moins bon rendement (Weiske et al., 2006).

En résumé, les études présentées dans cette section nous montrent d'abord l'intérêt que peuvent avoir les modèles à l'échelle de la ferme pour évaluer l'effet de l'introduction de la méthanisation (hiérarchie des impacts, prise en compte d'interactions entre les différents éléments de la chaîne de traitement). Ces études se sont surtout concentrées sur la gestion des effluents (bâtiment, digestion, stockage). L'import de déchets extérieurs et les effets agronomiques de la méthanisation n'ont été que peu étudiés avec des modèles à l'échelle de l'exploitation.

On peut également évoquer l'existence de modèles orientés vers la gestion des déchets, et qui ont pour but de renseigner des études par analyse de cycle de vie (ACV) (Gentil et al., 2010). Ces modèles sont souvent spécifiques de la zone géographique dans laquelle ils sont utilisés. Différents modèles ont des hypothèses différentes, ce qui fait nécessairement varier les résultats. ORWARE est un exemple de modèle qui considère la méthanisation du point de vue des déchets (Dalemo et al., 1998; Eriksson et al., 2002). Ce modèle considère l'épandage de digestat en utilisant des facteurs d'émissions (émissions gazeuses, substitution aux engrais chimiques). ORWARE a pu être utilisé, par exemple, pour estimer l'intérêt de la méthanisation des effluents équins (Hadin and Eriksson, 2016). Ce type de modèles n'a pas pour but de s'intéresser aux aspects agronomiques de la méthanisation et paraît plus pertinent pour comparer différents traitements de déchets à une échelle plus vaste que la ferme.

Tableau 1.7 – Exemples de modèles à l'échelle de l'exploitation, considérant la méthanisation des effluents d'élevage. FE : Facteurs d'émissions

Modèle et référence	Simulation de l'élevage	Simulation de la méthanisation	Simulation du stockage des PROs	Simulation des champs et de l'épandage des PRO.	Objectif de recherche
FarmGHG Olesen et al. (2004) Weiske et al. (2006)	Oui	Equations simples. Possible co-digestion de déchets extérieurs ou de cultures	FE	Substitution aux engrais : coefficient d'équivalence. FE. Pas de considération du N_2O ou de la MOS.	Bilan GES de l'exploitation (bovin)
Rigolot et al. (2010)	Oui	Equations simples. Mono-digestion seulement	FE, équations mécanistes.	Non	Comparaison de différentes pratiques de gestions des effluents porcins
IFSM (*Integrated Farm System Model*) Rotz et al. (2018) Veltman et al. (2018)	Oui	Equations simples. Mono-digestion seulement	Modèle mécaniste	Cycles du C et du N au champ, grâce au modèle sol-plante DayCent	Comparaison de différentes pratiques de gestion de l'exploitation (élevage, effluents, champs)
Manure-DNDC Li et al. (2012)	Oui	Equations simples. Mono-digestion seulement	Modèle mécaniste	Cycles du C et du N au champ, grâce au modèle sol-plante DNDC	Comparaison de différentes pratiques de gestion de l'exploitation (élevage, effluents, champs)
Boldrin et al. (2016)	Non	Bilan de masse, Equations simples. co-digestion.	FE.	N_2O au champ : FE.	Bilan GES et analyse économique de la méthanisation
Baral et al. (2018)	Non	Equations simples. co-digestion.	Mesures	Mesures et FE. Inclue une estimation du stockage de MOS	Bilan GES de la méthanisation
Costa Junior et al. (2015)	Non	Bibliographie.	Mesures et FE.	Mesures et FE..	Bilan GES de la méthanisation
Vergote et al. (2019)	Non	Modèle mécaniste	Modèle mécaniste et FE	FE	Effet de la méthanisation et de la gestion des effluents sur le bilan GES

1.4.5 Analyses de cycle de vie et méthanisation

La section précédente (1.4.4) présente des études utilisant des modèles à l'échelle de la ferme, qui s'intéressent en particulier aux émissions de GES, parfois en regard d'analyses économiques (Boldrin et al., 2016; Weiske et al., 2006). Néanmoins, d'autres impacts environnementaux peuvent également être étudiés. Les Analyses de Cycle de Vie (ACV) sont des outils largement utilisés pour l'évaluation environnementale, selon une méthode multicritère normalisée ISO. Les ACV permettent également de regarder les impacts d'un méthaniseur au-delà des limites de la ferme (Rotz, 2018).

De nombreuses études par ACV ont été menées pour estimer le bilan environnemental de la méthanisation. Plusieurs revues de la littérature sur les ACV de la méthanisation montrent une grande diversité au sein de ces études (Bacenetti et al., 2016b; Hijazi et al., 2016; Vasco-Correa et al., 2018). Tout d'abord, les ACV peuvent répondre à des objectifs différents, comme l'évaluation de la méthanisation en fonction des dernières avancées scientifiques, l'amélioration des pratiques de méthanisation, ou la comparaison de la méthanisation avec d'autres manières de traiter les déchets ou de produire de l'énergie. Étant donné la multifonctionnalité de la méthanisation (production d'énergie, traitement des déchets, application de fertilisants organiques au champ) et la variabilité des systèmes utilisant un méthaniseur, les systèmes étudiées dans les ACV sont aussi variables. Les ACV peuvent ainsi considérer des limites différentes, traiter des intrants différents, valoriser le biogaz différemment, se placer à des échelles différentes allant de la ferme (Styles et al., 2015) à l'échelle nationale (Styles et al., 2016). Les unités fonctionnelles utilisées varient. Elles représentent en général une unité liée à la production d'énergie (en MJ, kWh, m^3 de biogaz) et/ou une unité liée au traitement des déchets (en t de déchets traités) (Hijazi et al., 2016). Des unités fonctionnelles basées sur l'apport de nutriments (Avadí, 2020) ou la production de matière agricole (Bacenetti et al., 2016a; Knudsen et al., 2014) sont également utilisées quand la question traite de l'utilisation de digestats en tant que fertilisants. Cette multifonctionnalité induit de nombreuses questions techniques dans l'application des ACV, notamment en ce qui concerne les limites du système et les allocations d'impacts utilisées. Des difficultés méthodologiques ont également été identifiées pour réaliser des ACV en lien avec l'utilisation de PRO (définition des facteurs d'équivalence aux engrais de synthèse, considération ou non de la substitution aux engrais N, P, et K, effet sur la MOS, choix des émissions au champ considérées) (Brockmann et al., 2018) ou avec la production agricole (prise en compte de l'interaction entre deux cultures successives, difficulté d'attribution des émissions au champ à une culture, définition d'unités fonctionnelles de production qui s'appliquent à différentes cultures) (Goglio et al., 2018). En fonction des systèmes considérés, ces difficultés peuvent également concerner les ACV concernant la méthanisation.

Cette diversité dans les ACV induit des résultats variables, mais des tendances générales dans les conclusions peuvent quand même être identifiées (Bacenetti et al., 2016b; Hijazi et al., 2016; Vasco-Correa et al., 2018). Les principales externalités positives de la méthanisation sont une diminution des émissions de GES et la diminution des besoins en ressources non renouvelables. La

valorisation du biogaz et l'amélioration des pratiques de traitement des déchets sont les principales sources de bénéfices sur ces indicateurs, suivi par la substitution des engrais chimiques. Les émissions fugitives de biogaz augmentent les émissions de GES. Quand ils sont considérés, le changement d'usage des sols induit par la méthanisation des cultures jouent aussi négativement sur le bilan carbone. L'effet de la méthanisation sur l'acidification et l'eutrophisation varient en fonction du système (amélioration ou détérioration des émissions lors du traitement des effluents d'élevage, substitution d'engrais par le digestat, fertilisation des cultures dédiées le cas échéant). Un meilleur bilan environnemental est trouvé lors de la méthanisation de déchets en comparaison avec celle de cultures énergétiques (Bacenetti et al., 2016b; Hamelin et al., 2014; Siddiqui et al., 2020). En ce qui concerne la méthanisation des effluents animaux, Miranda et al. (2015) proposent une revue de littérature des ACV s'intéressant au bilan carbone de la méthanisation (mono-digestion) des lisiers d'une ferme avec un élevage laitier. La méthanisation des effluents animaux diminue les émissions de GES, ce qui est largement dû à la diminution d'émissions de GES lors du traitement des PRO (stockage), puis par la production de biogaz ; les effets positifs concernant les émissions au champ de la méthanisation sont du même ordre de grandeur que les effets négatifs dus aux émissions fugitives de biogaz. Néanmoins, l'étude montre une grande diversité de résultats entre les systèmes et montre que les ACV ne considèrent pas toutes les mêmes sources d'émissions. En France, très peu d'ACV portant sur la méthanisation ont été menées. Garrigues et al. (2014) ont réalisé une ACV sur une exploitation en polyculture élevage (élevage porcin). Les auteurs montrent qu'en contexte français, où le biogaz est utilisé en cogénération pour remplacer de l'électricité provenant majoritairement de l'industrie nucléaire, les effets de la méthanisation sur les émissions de GES sont faibles. L'étude ne considère aucun import de déchets extérieurs.

Des ACV récentes se sont intéressées avec plus de détails aux aspects agronomiques liés à l'introduction de la méthanisation, en utilisant des modèles d'émissions au champ ou des modèles à l'échelle de la ferme pour réaliser leurs inventaires de cycle de vie. La prise en compte de façon plus précise de la substitution aux engrais, des émissions gazeuses après apports de digestats au champ ou du stockage de MOS, ou du changement d'usage des sols, est ainsi possible (Hamelin et al., 2014; Vaneeckhaute et al., 2018). Ces études montrent un impact important positif (substitution aux engrais, stockage de MOS) ou négatif (émissions gazeuses après apports de digestats au champ) de ces effets au champ sur le bilan environnemental général de la méthanisation. Néanmoins, la considération des effets de l'épandage de digestats au champ demande une expertise fine des impacts réels de la méthanisation : par exemple, Bacenetti et al. (2016b) revendiquent le fait de ne pas tenir compte de la substitution des engrais P et K par les digestats si ceux-ci sont appliqués dans des champs où le besoin en ces nutriments est faible. À l'inverse, Zhang et al. (2021) réalisent une ACV de la méthanisation de lisier porcin et prennent en compte la richesse des sols en P et K pour limiter les apports de digestat. En fonction de l'ampleur de ces limites, les digestats doivent être épandus sur des surfaces plus grandes, ce qui augmente leurs impacts sur l'environnement, dans cette étude. L'effet de l'import de déchets à l'échelle d'une ferme

peut être exploré. Par exemple, Michel et al. (2010) montrent qu'à l'échelle d'une ferme, l'import de déchets extérieurs augmente à la fois la production de biogaz et donc le bilan positif de la méthanisation, mais aussi les effets négatifs sur l'acidification et l'eutrophisation liés à la gestion des digestats. Il y a donc une intensification locale des impacts.

En conclusion, nous pouvons dire que les modèles et les ACV permettent une synthèse des impacts de la méthanisation. La méthanisation peut avoir des impacts différents en fonction des systèmes. Elle a en général un impact positif en terme de bilan carbone et d'utilisation des ressources fossiles. Néanmoins, l'effet global de la méthanisation sur les pertes azotées restent encore mal caractérisé. Les effets agronomiques de la méthanisation pourraient également être mieux pris en compte.

1.5 Conclusion de la synthèse bibliographique et objectifs de la thèse

1.5.1 Conclusion de la synthèse bibliographique

Au cours de l'introduction de cette thèse, nous avons vu que la méthanisation est une pratique en expansion dans le monde, et en France de façon relativement récente. Il s'agit en fait d'une technologie qui se traduit par une diversité de pratiques. Par conséquent, les références en terme de bénéfices et d'impacts sur l'environnement sont difficiles à acquérir, car elles doivent couvrir de très nombreuses situations.

La synthèse bibliographique permet de montrer que les effets de l'épandage des digestats à l'échelle de la parcelle sont raisonnablement bien caractérisés. Nous avons cependant mis en évidence un manque de recul sur les effets des apports répétés de digestats au champ à long terme, comme sur le stockage de MOS ou sur la vie du sol. De plus, les différents effets de l'épandage des digestats au champ sont souvent étudiés séparément, et peu mis en regard les uns par rapport aux autres.

L'introduction de la méthanisation dans une exploitation en polyculture élevage a de nombreux effets sur la gestion des effluents, l'épandage de PRO, les systèmes de cultures, l'import de déchets extérieurs pour la méthanisation. Nous avons mis en évidence l'importance de caractériser à la fois les effets de la méthanisation sur l'aspect du traitement des PRO (méthanisation, post-traitement, stockage), et l'aspect agronomique lié à l'épandage des PRO. En effet, ils ne sont pas indépendants, entre autres parce que le traitement des effluents bovins et la présence de co-substrats dans le méthaniseur influence la qualité et la quantité de PRO épandus au champ. L'échelle de la ferme semble être très pertinente pour étudier la méthanisation agricole.

À cause de tous ces effets qui interagissent les uns avec les autres, il n'est pas évident de conclure de façon synthétique sur les effets de la méthanisation. Les mesures expérimentales peuvent difficilement étudier tous les aspects modifiés par l'introduction d'un méthaniseur, surtout si la volonté est de

généraliser les résultats à différents contextes. Elles sont cependant nécessaires pour valider des modèles, qui sont des outils utiles pour dépasser ces problèmes. Les modèles à l'échelle de la ferme, permettent d'étudier simultanément plusieurs aspects de l'introduction d'un méthaniseur. Ils permettent de généraliser les résultats obtenus par les mesures et d'explorer des scénarios à plus long terme, et selon des pratiques plus nombreuses que ce qui peut être réalisé simplement avec les mesures. Ils permettent d'identifier des pratiques à conseiller ou à proscrire pour améliorer le bilan environnemental de la filière.

Ces approches montrent que, à l'échelle de la ferme, la valorisation du biogaz et la diminution des émissions de CH_4 lors du stockage des PRO sont des éléments essentiels au bénéfice de la méthanisation en terme d'empreinte carbone. Les aspects agronomiques de la méthanisation agricole, comme l'effet de la substitution aux engrais chimiques (positif), les effets sur la MOS (mal caractérisés) ou les effets des émissions des digestats lors de leur application au champ (négatifs), peuvent impacter le bilan environnemental de la méthanisation. Ces questions sont au cœur de la thématique de la méthanisation agricole, mais n'ont encore que peu été étudiées avec de telles approches.

1.5.2 Objectifs de la thèse

La méthanisation agricole dans une exploitation de polyculture élevage a d'abord pour rôle la production d'énergie renouvelable et le traitement des déchets. Néanmoins, elle a également de fortes conséquences agronomiques, sur lesquelles nous nous concentrerons dans cette thèse. Les cycles du C et du N sont liés à une grande partie des effets agronomiques de la méthanisation : fertilisation, pertes azotées, émissions de GES, stockage de MOS. Les effets de la méthanisation sur la vie du sol peuvent aussi être liés à ces cycles, entre autres via l'apport de matière organique comme source de nourriture, et l'épandage de produits riches en azote ammoniacal qui pourrait impacter certains organismes. Dans cette optique, cette thèse veut donc répondre à la question générale suivante :

Comment la méthanisation agricole en polyculture élevage modifie-t-elle les cycles du carbone et de l'azote à l'échelle de la ferme, c'est-à-dire sur l'ensemble de la filière du traitement des effluents jusqu'à l'épandage des digestats au champ ?

Cette question générale regroupe en réalité plusieurs questions plus spécifiques :

- Comment les caractéristiques des digestats, influencées par les conditions de stockage ou de post-traitement, déterminent-elles leurs propriétés lors de leur épandage au champ ?
- Peut-on quantifier globalement les effets de la méthanisation sur les différents flux azotés d'intérêt, comme la substitution des engrais chimiques par les digestats, la volatilisation d'ammoniac, les émissions de N_2O ou la lixiviation des nitrates, en considérant les différentes sources d'émissions ?
- Quel est l'effet de la méthanisation sur le stockage de MOS, la qualité des sols, la fourniture en azote du sol et la vie du sol ?

- Quel est l'effet de l'import de déchets extérieurs sur la qualité et la quantité des digestats et donc les bénéfices et risques de la méthanisation ?
- Peut-on quantifier et comparer les effets de différentes pratiques lors du traitement, du stockage, ou de l'épandage des digestats ?

Pour répondre à ce questionnement, nous avons utilisé une démarche utilisant l'expérimentation et la modélisation. Un cas d'étude, incluant un essai au champ, a permis d'acquérir de bonnes références sur les effets d'un système de fertilisation utilisant des digestats, au lieu d'effluents bovins ou d'engrais de synthèse, sur le cycle de l'azote du champ et sur la vie du sol. Ce cas d'étude nous a permis de construire un modèle à l'échelle de la ferme, composé de deux parties, l'une traitant de l'aspect traitement des déchets de la méthanisation (méthanisation et stockage du digestat, modèle SYS-Metha), l'autre de l'aspect agronomique (application des digestats au champ, modèle STICS). La qualité et la quantité de digestats est au cœur du couplage entre ces deux parties. Le cas d'étude a permis d'évaluer et de calibrer chacune des sous-parties du modèle. Des scénarios ont enfin été simulés pour répondre aux questions de la thèse.

L'organisation de la thèse est détaillée dans la

Figure 1.5, ainsi que dans le paragraphe suivant.

Chapitre 2 – La thèse s'appuie sur un cas d'étude original : le site d'expérimentation de l'INRAE à Nouzilly (Centre Val-de-Loire, France). Ce site est basé sur une exploitation de polyculture élevage de l'INRAE. Un élevage bovin fournit ses effluents à un méthaniseur exploité par Cap Vert Energie. Les digestats sont ensuite épandus au champ sur l'exploitation agricole. Le chapitre présente le cas d'étude, ainsi que le protocole d'un essai au champ de 3 ans qui compare des systèmes de fertilisation utilisant des effluents bovins ou des digestats issus de leur méthanisation.

Chapitre 3 – Les résultats de cet essai au champ introduit dans le chapitre 2 sont présentés dans ce chapitre 3. Nous mettons en regard les effets de la méthanisation des effluents bovins sur les propriétés fertilisantes des digestats et sur les pertes azotées au champ.

Chapitre 4 – Cet essai au champ a permis d'évaluer la capacité du modèle sol-plante STICS à simuler le cycle de l'azote lors d'une rotation culturale fertilisée avec des digestats, ainsi qu'à prédire les différences entre les systèmes de fertilisation étudiés. STICS sera donc utilisé comme modèle pour simuler les épandages de digestats.

Chapitre 5 – Le méthaniseur du cas d'étude, traitant des déchets variés, a permis d'évaluer la capacité du modèle de de méthanisation SYS-Metha à prédire la qualité des digestats et la production de biogaz. SYS-Metha sera utilisé pour simuler la digestion et le stockage des digestats.

Chapitre 6 – Enfin, nous couplons les modèles STICS et SYS-Metha pour quantifier l'effet de l'introduction de la méthanisation sur les flux de C et N. Nous simulons différents scénarios pour quantifier les effets de l'import de déchets extérieurs et de différentes pratiques de gestion du digestat, du post-traitement à l'épandage.

Chapitre 7 – Néanmoins, cette quantification des flux de C et N ne permet pas d'évaluer les autres effets de l'introduction de la méthanisation. Pour illustrer cette limite, et explorer une dernière question de recherche liée à l'introduction de la méthanisation agricole, nous présentons une étude sur l'effet de l'épandage de digestat sur la biologie du sol au travers de l'exemple des vers de terre.

Chapitre 8 – Enfin, l'ensemble des résultats obtenus dans cette thèse fait l'objet d'une discussion générale

Conséquences de l'introduction de la méthanisation dans une exploitation de polyculture-élevage sur les cycles du carbone et de l'azote

Chapitre 2 – Présentation du site d'expérimentation à Nouzilly

Combinaison de l'expérimentation et de la modélisation à l'échelle de la ferme pour quantifier les flux de C et N

Chapitre 3 – Comment la méthanisation des effluents bovins modifie-t-elle le cycle de l'azote sur une rotation culturale de 3 ans ?
Quels sont les propriétés fertilisantes des digestats par rapports aux effluents bovins ? Quels sont les effets de la méthanisation des effluents bovins sur les pertes azotées au champ ?

Chapitre 4 – Conséquences de la méthanisation des effluents d'élevage sur les flux d'azote lors d'une rotation culturale de 3 ans : évaluation du modèle sol-plante STICS
Peut-on prédire les impacts de l'épandage de digestat sur le cycle de l'azote au champ en utilisant le modèle sol-plante STICS ?

Chapitre 5 – Évaluation du modèle de méthanisation SYS-Metha à l'aide des données opérationnelles d'un méthaniseur traitant des déchets variés
Peut-on prédire la qualité des digestats et la production de biogaz d'un méthaniseur complexe à l'aide du modèle de méthanisation SYS-Metha ?

Chapitre 6 – La méthanisation des effluents d'élevage peut diminuer l'utilisation d'engrais de synthèse, augmenter le stockage de C dans les sols et augmenter la volatilisation d'ammoniac à l'échelle de la ferme : rôle clef de l'import de déchets et évaluation de différentes pratiques de gestion
Peut-on quantifier l'effet de l'introduction de la méthanisation sur les cycles du C et du N à l'échelle de la ferme ? Quel est l'effet de l'import d'intrants extérieurs sur ces flux ? Peut-on quantifier et comparer les effets de différentes pratiques de gestion du digestat ?

Approfondissement : aller au-delà des flux de C et N

Chapitre 7 – Effets à court et long termes de l'épandage de digestat en grande culture sur les vers de terre
Quels sont les impacts à court et long termes de l'épandage de digestat sur les populations de vers de terre par rapport aux effluents bovins ?

Chapitre 8 – Discussion générale et conclusion

Figure 1.5 – Plan de la thèse

Chapitre 2
Présentation du site d'expérimentation à Nouzilly

Cette thèse est construite autour du cas d'étude de l'exploitation en polyculture élevage du centre INRAE à Nouzilly (Centre – Val de Loire) et du méthaniseur localisé sur ce centre. Ce chapitre présente le cas d'étude, le projet de recherche MétaMétha qui l'a étudié, et l'essai au champ qui a eu lieu dans le cadre de ce projet. Les résultats de l'essai au champ seront présentés dans le chapitre 3.

2.1 Contexte régional

2.1.1 Localisation, agriculture et climat

Le site de Nouzilly est situé dans le département d'Indre-et-Loire (37), région Centre Val-de-Loire, France. Il est localisé à environ 20 km au nord de Tours (coordonnées : 47°32' N, 0°48' E) (Figure 2.1).

D'après l'enquête AGRESTE 2017, en région Centre Val-de-Loire, la surface agricole utile (SAU) représentait 60% de la surface totale en 2016. La culture de céréales domine. Elle représente environ la moitié de la SAU. Néanmoins, dans la région, l'agriculture est diversifiée, avec des oléagineux, du vignoble, des vergers de pommes et de poires, du maraîchage, ainsi que de l'élevage (bovin viande et lait, ovin, porcin, avicole, caprin) (AGRESTE, 2017), (centre-valdeloire.chambres-agriculture.fr/) (Figure 2.2).

En Indre-et-Loire, à l'exception des zones de cultures spécialisées des bords de Loire, l'agriculture est dominée par les grandes cultures, associées à l'élevage. La superficie totale est de 615 000 ha, et la SAU de 351 000 ha. En 2018, les principales cultures sont :

- les céréales (46% de la SAU), dont le blé (31% de la SAU), l'orge (7%) et le maïs (5%),
- les oléagineux (19%) qui incluent le tournesol,
- les prairies artificielles et temporaires (7%), et les surfaces toujours en herbe (10%) (AGRESTE, 2020).

L'élevage est dominé par l'élevage bovin laitier et l'élevage caprin (AGRESTE, 2020). Le climat est océanique (classification Cfb selon la classification de Köppen). A Tours, entre 1981 et 2010, la température moyenne annuelle est de 11,8 °C et la pluviométrie annuelle moyenne est de 696 mm (Météo-France).

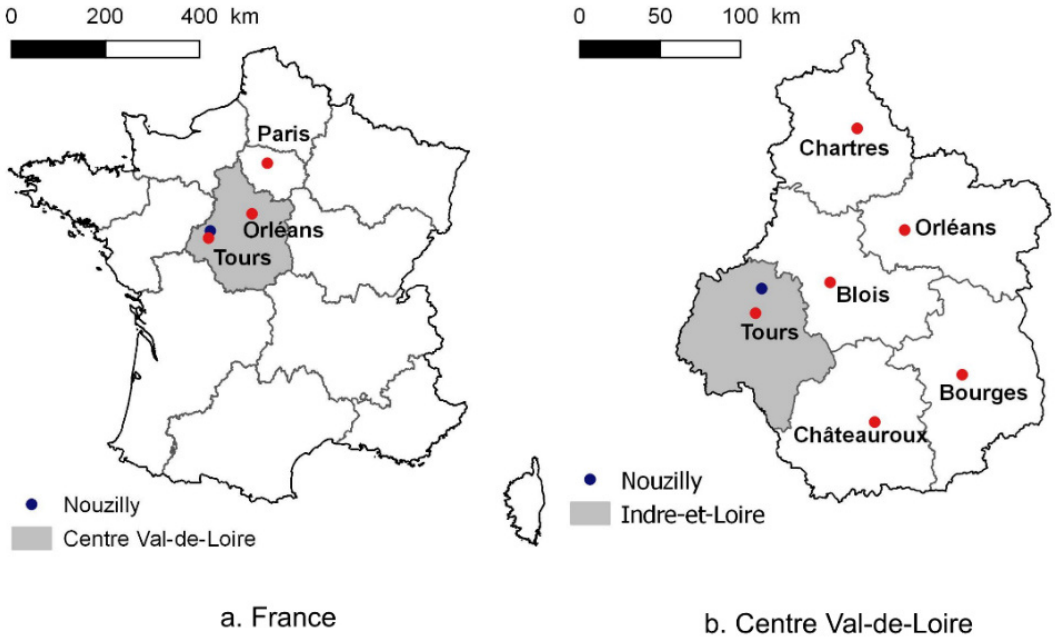

Figure 2.1 – Localisation du site de Nouzilly, a) en France, et b) en région Centre Val-de-Loire.

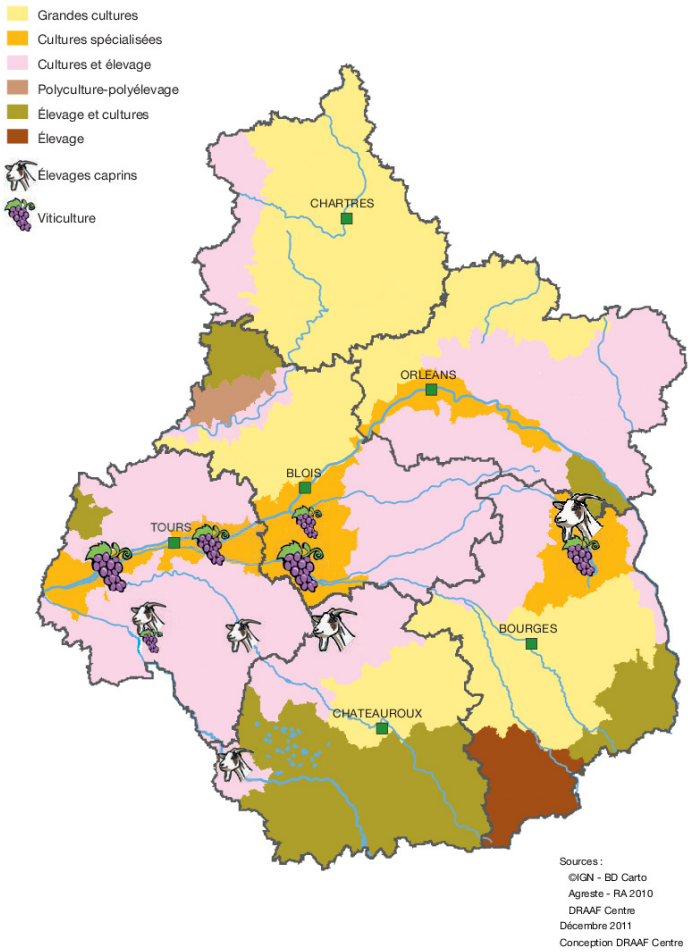

Figure 2.2 – Type d'agriculture en région Centre Val-de-Loire. Extrait des Agri'repères AGRESTE (2017).

2.1.2 La méthanisation en Centre Val-de-Loire

En Centre Val-de-Loire, en 2021, on dénombre 39 unités de méthanisation, dont 22 à la ferme (16 en cogénération, 6 en injection), 11 centralisées (4 en cogénération, 7 en injection), 2 unités utilisant seulement une couverture de fosse à lisier avec valorisation de la chaleur uniquement, 1 unité en station d'épuration des eaux usées (STEP) avec valorisation de la chaleur uniquement, et 3 unités en industrie avec valorisation de la chaleur uniquement (SINOE, 2021). Elles étaient 20 en 2017.

2.1.3 Enquête sur la méthanisation en Centre Val-de-Loire en 2016

En 2016, une enquête a été menée auprès de 13 unités de méthanisation à la ferme par la chambre d'agriculture du Centre Val-de-Loire, dans le cadre du projet MétaMétha (présenté en section 2.3). Le but était de répertorier les pratiques agricoles liées à la gestion des digestats. Nous en faisons une synthèse ici. Les unités enquêtées utilisent toutes un procédé en voie humide mésophile. Elles traitent entre 5 500 et 22 500 t de déchets par an (moyenne : 10 200 t an^{-1}). Sur les 13 unités, 10 post-traitent le digestat par séparation de phase, par presse à vis. Les unités n'utilisant pas de séparation de phase produisent des digestats bruts avec des teneurs en matière sèche relativement faibles (5 à 10%), ce qui permet de les épandre malgré tout à l'aide d'une rampe à pendillard sans bouchage des buses. Cinq unités sont équipées d'une couverture de fosse de stockage des digestats liquides ou bruts.

Les digestats solides représentent des volumes relativement faibles. Ils sont le plus souvent épandus avec des épandeurs à fumier. Ils sont épandus soit en fin d'été (devant colza, blé, ou cultures

intermédiaires à vocation énergétique – CIVE), soit au printemps (prairies, ou avant les semis de maïs). Les digestats solides sont typiquement apportés avec une dose de 10 à 15 t ha^{-1}.

Les digestats liquides ou bruts représentent des volumes importants. La plupart des digestats liquides ou bruts sont épandus avec une tonne à lisier et une rampe à pendillard. Quelques agriculteurs testent des systèmes d'épandage sans tonne (enrouleur, cordon ombilical), souvent en prestation de service, pour éviter les problématiques de tassement de sol. L'utilisation des digestats est guidée par des contraintes de volume de stockage et la volonté de valoriser leur capacité fertilisante. En conséquence, les deux périodes d'épandage sont l'automne (gestion du stock) et au début du printemps (fertilisation des cultures). En fin d'été ou automne, les digestats sont apportés devant colza ou sur prairie, parfois devant une céréale ou une CIVE. Au printemps, période qui semble être favorisée, les digestats sont apportés sur céréale (blé en particulier), devant maïs, sur prairie, et très peu sur colza. Les doses d'apports de digestats liquides ou bruts varient de 20 à 35 t ha^{-1} en fin d'été ou en automne, et de 25 à 50 t ha^{-1} au printemps. Lors d'apports sur les cultures en fin d'hiver ou au printemps, l'épandage avec tonne à lisier et rampe à pendillard ne permet pas de passer trop tôt (sol humide et peu portant), ni trop tard (risque d'abîmer les cultures). Sur maïs, les apports se font surtout juste avant le semis ou à la place du premier apport d'engrais minéral (stade 2-3 feuilles). Sur blé, les apports sont réalisés à la place du premier ou deuxième apport, mais jamais en troisième apport. Dans les rares épandages sur colza au printemps, les digestats sont apportés au premier apport, mais pas au deuxième. Les prairies peuvent recevoir du digestat à de nombreux moments dans l'année, dès que le sol est portant : en février pour avoir le temps que la pluie lave les feuilles avant la pâture ou la récolte, après une coupe au printemps pour favoriser la repousse ou en novembre. La prairie permet d'épandre le digestat à de nombreux moments dans l'année en raison de l'absence de problème de portance des sols, ce qui facilite la gestion des stocks. Pour les agriculteurs, les avantages des digestats par rapport aux effluents non digérés sont un meilleur pouvoir fertilisant et moins de problèmes d'odeurs. En 2017, la fertilisation avec les digestats est encore mal maîtrisée (recul encore faible sur leur utilisation), et beaucoup d'agriculteurs s'interrogent sur les problèmes de tassement de sol.

2.1.4 Evolution de la méthanisation agricole en région Centre Val-de-Loire entre 2016 et 2020

Depuis 2016, le nombre de méthaniseurs agricoles augmente, et la gestion du digestat évolue avec les retours d'expérience. J'ai pu réaliser plusieurs réunions avec des membres des chambres d'agriculture du Loiret, du Cher et de l'Indre, pour suivre les évolutions des pratiques des agriculteurs méthaniseurs. Les projets de méthanisation ont tendance à être plus importants : parmi les méthaniseurs recensés sur la base de données SINOE, les méthaniseurs agricoles mis en service avant 2017 traitent en moyenne 9 700 t de déchets par an (n = 11) contre 11 900 t pour ceux mis en service entre 2017 et 2020 (n = 8). Il s'agit toujours d'unités en voie humide infiniment mélangée. La méthanisation en voie sèche discontinue n'est pas très développée dans la région. Avant 2017, les méthaniseurs agricoles étaient souvent installés sur des exploitations en polyculture élevage, pour gérer les effluents. Sur ce type d'installation, les intrants moyens sont : 70% d'effluents (lisier + fumier), 10% d'intercultures, 20% de déchets extérieurs (déchets de tonte, déchets d'industrie agroalimentaire – IAA, déchets de cantine, par exemple). Un autre type de projet a vu le jour, avec des méthaniseurs dont la ration est composée par environ 60% de culture (CIVE, quelques cultures dédiées) et des produits extérieurs (pouvant inclure des effluents animaux). Ce nouveau système concerne des grandes surfaces de cultures céréalières.

La gestion des digestats n'a que très légèrement évoluée. Les agriculteurs essaient de valoriser les digestats le mieux possible d'un point de vue agronomique, mais les contraintes logistiques sur le choix des dates et doses d'épandage restent très fortes. Les épandages avec une tonne à lisier posent des problèmes de logistique (nombreux allers-retours), et de tassement des sols. Les épandages se font de

plus en plus avec des systèmes plus adaptés : épandage sans tonne, rampe à pendillard large de 36 m qui permet de faire moins de traces dans la culture. Nous n'avons pas eu d'information sur l'enfouissement des digestats. La gestion de la charge de travail (méthaniseur, transport et épandage des digestats, culture des CIVE) et du coût de l'épandage sont des questions d'actualité pour les agriculteurs.

2.2 Description du site de Nouzilly

2.2.1 Description générale

L'Unité Expérimentale de Physiologie Animale de l'Orfrasière (UE PAO – INRAE, Nouzilly, France) est une unité expérimentale de recherche qui contribue à des travaux en production animale, ainsi qu'en agroécologie (www6.val-de-loire.inrae.fr/uepao). L'UE PAO possède ainsi des élevages bovins, ovins, caprins, équins et porcins. L'équipe « Domaine » de l'UE PAO a pour mission d'exploiter les terres agricoles de l'unité pour produire les fourrages nécessaires aux élevages ce qui inclue la valorisation des effluents au champ, entre autres. C'est sur ses terres que se déroulent les projets de recherche sur l'agroécologie (autonomie fourragère, itinéraire techniques sans pesticides, agroforesterie). En 2014, un méthaniseur, exploité par la société Cap Vert Energie, a été construit à proximité des installations de l'UEPAO. Ce méthaniseur recycle une grande partie des effluents d'élevage de l'INRAE, et le digestat est majoritairement épandu sur les terres agricoles de l'INRAE. La proximité de l'élevage, du méthaniseur et des terres agricoles a contribué à la mise en place du projet MétaMétha et de son essai au champ. L'emplacement de ces infrastructures est montré en Figure 2.3.

Figure 2.3 – Photographie aérienne du site de Nouzilly. Coordonnées : 47,545377°N, 0,789024°E. Source : www.geoportail.gouv.fr.

2.2.2 L'élevage

L'INRAE possède plusieurs élevages. Seuls ceux dont les effluents sont traités dans le méthaniseur du site sont présentés : les élevages bovin, ovin et équin.

L'élevage équin comporte environ 150 chevaux dont 120 juments. Le fumier équin frais est un intrant du méthaniseur. L'élevage ovin est constitué de 1600 bêtes. La plupart des animaux pâturent au printemps et en été, et restent en bergerie en hiver. Le fumier ovin frais est un intrant du méthaniseur. L'élevage bovin est un élevage bovin laitier, composé d'environ 65 vaches laitières et 100 génisses et vaches taries. Seules les génisses pâturent 4 mois de l'année. Les vaches laitières et taries restent en bâtiment toute l'année sur aire paillée (un couloir équipé d'un racleur évacue le lisier en bout de bâtiment dans une pré-fosse non couverte). Le lisier est ensuite pompé vers le méthaniseur. Les fumiers bovins sont assez pailleux. Quand ils sont frais, ils sont un intrant du méthaniseur. Alternativement à la gestion par méthanisation, les fumiers bovins sont stockés en fumière couverte comme les fumiers ovins et équins. Ces fumiers sont ensuite compostés en andain sur une plateforme non couverte puis épandus sur les terres agricoles.

2.2.3 Le méthaniseur

Le méthaniseur de Nouzilly (Cap Vert Bioénergie Nouzilly) a été mis en service en 2014, à proximité du site de l'INRAE. Il est exploité par la société Cap Vert Energie. Il s'agit d'un méthaniseur en voie humide infiniment mélangée mésophile. Le biogaz est valorisé en cogénération. Le méthaniseur est une Installation Classée pour la Protection de l'Environnement (ICPE) soumise à autorisation. La capacité de traitement est de 12 000 t de déchets par an, mais la quantité réelle de déchets est comprise entre 7 500 et 8 000 t an^{-1}. Une partie de la chaleur est valorisée avec le chauffage du méthaniseur, une autre partie est vendue à l'INRAE pour le chauffage des bâtiments en hiver, et pour un séchoir à fourrage agricole en été. La chaleur vendue à l'INRAE reste faible, de l'ordre de 100 MWh an^{-1} thermique, au regard de la production électrique qui se situe autour de 1 300 MWh an^{-1} électrique.

Le méthaniseur est constitué d'une préfosse pour mélanger la ration journalière, d'un digesteur principal (1 600 m^3) et d'un post digesteur (700 m^3). Le temps de séjour total est de 100 jours environ. À sa sortie, le digestat brut subit une séparation de phase dans une presse à vis. Le digestat solide est stocké sur place puis envoyé à l'INRAE tous les quinze jours où il est usuellement co-composté avec des effluents d'élevage. Le digestat liquide est stocké dans une lagune non couverte (3 500 m^3, 1 340 m^2) (Figure 2.4).

Les exploitants du méthaniseur ont pu nous transmettre des données d'imports de déchets sur la plateforme, de déchets entrant dans la ration, de production de biogaz, et d'export de digestats, à l'échelle de la semaine, entre le 01/01/2017 et le 31/12/2019. Sur ces trois années, les intrants ont été stables. En moyenne, le méthaniseur a traité 7 500 t de déchets. Les déchets traités sont variés : on en dénombre une trentaine sur cette période. Il s'agit de lisiers bovins de l'INRAE (11% des intrants en masse), de fumiers équin, bovin, et ovin (17%), de boues d'épuration issues de stations d'épurations urbaines (29%), de divers boues et déchets issus des IAA (18%), de graisses (8%), d'eaux sales (9%), d'issues de silos de céréales (8%) ainsi que quelques déchets autres comme des déchets de tonte de bords de route (Figure 2.5).

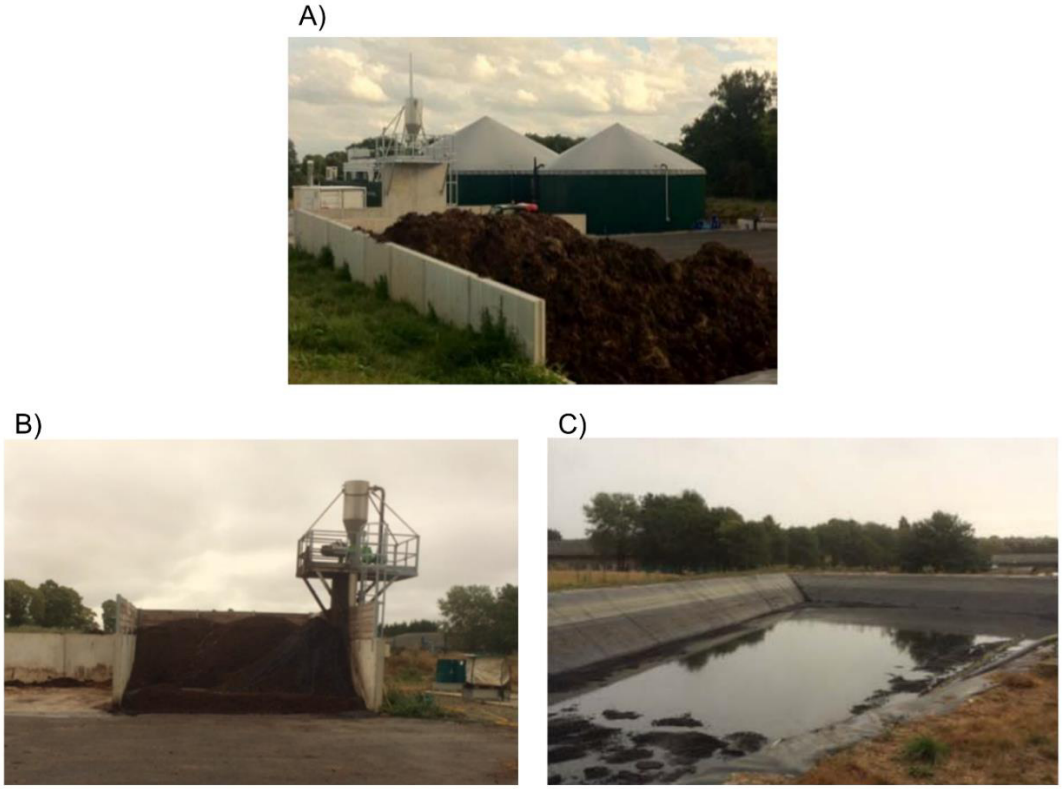

Figure 2.4 – Photographie A) du méthaniseur de Nouzilly, B) de la presse à vis et du stockage de digestat solide, C) de la lagune de stockage de digestat liquide. © INRAE.

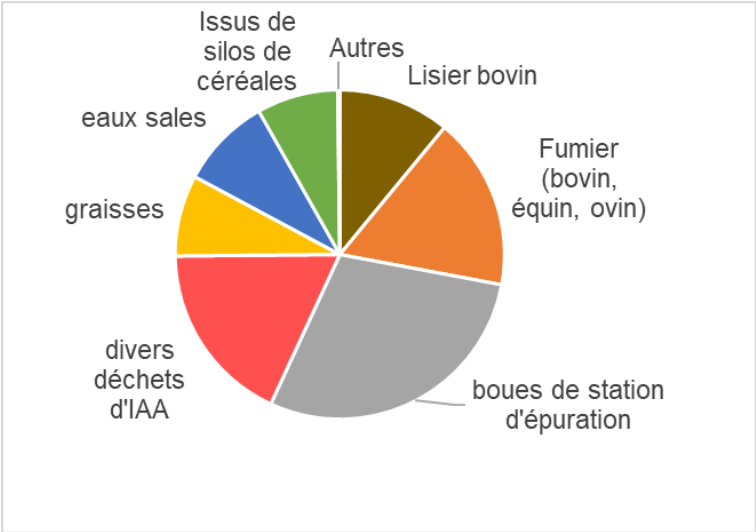

Figure 2.5 – Ration moyenne du méthaniseur

2.2.4 L'exploitation agricole

Les terres agricoles de l'INRAE sont exploitées par l'UE PAO. Il s'agit d'une exploitation agricole de grandes cultures en gestion conventionnelle de 407 ha. L'exploitation produit de l'alimentation pour les élevages de l'INRAE et permet de valoriser les effluents des animaux au champ. L'assolement est constitué de prairies (32%, dont 4% de prairies permanentes), de blé (22%), de maïs (17%, dont maïs grain 10% et maïs ensilage 7%), d'orge (6%), de luzerne (5%), et d'une diversité d'autres cultures, souvent fourragères (pois, soja, moha, fèverole, méteil, triticale, avoine). Les prairies restent en moyenne 4 ou 5 ans. Les prairies sont en général des associations de graminées (ray-grass hybride, dactyle, fétuque), avec des trèfles. Sur les prairies fauchées, on réalise 2 à 3 coupes dans l'année : la première coupe sert à faire de l'ensilage ou de l'enrubannage, les coupes suivantes font du foin. La plupart des prairies ne sont pas pâturées.

Le compost de fumier et le digestat solide sont majoritairement apportés devant maïs ou sur prairie. Le digestat liquide est apporté majoritairement sur prairie, blé ou maïs. Le lisier porcin est principalement épandu sur prairie. Les périodes et doses d'apport dans l'année sont précisées dans le Tableau 2.1.

Tableau 2.1 – Apports type de PRO sur l'exploitation de Nouzilly

PRO	Culture	Période d'apport	Dose d'apport
Fumier composté, digestat solide	Maïs	Printemps	15 – 25 t ha^{-1}
	Prairie	Automne	15 – 25 t ha^{-1}
	Méteil	Automne	15 – 25 t ha^{-1}
Digestat liquide	Prairie	Fin d'hiver à fin de printemps	20 – 30 t ha^{-1}
	Prairie	Automne	10 – 20 t ha^{-1}
	Blé	Printemps	15 – 25 t ha^{-1}
	Maïs	Printemps	20 – 25 t ha^{-1}
	CIPAN avant maïs	Automne	10 – 20 t ha^{-1}
Lisier porcin	Prairie	Automne	20 – 35 t ha^{-1}

2.3 Description du projet MétaMétha

2.3.1 Objectifs du projet

Le projet MétaMétha est un projet de recherche coordonné par l'UE PAO (INRAE, Nouzilly) auquel participent l'UR SOLS (INRAE, Orléans) et l'UMR ECOSYS (INRAE, Thiverval-Grignon), qui s'est déroulé entre 2017 et 2019. Les chambres d'agriculture du Loiret et de l'Indre-et-Loire, la société de conseil Evea et la société Cap Vert Energie sont partenaires du projet. Le projet est financé par la région Centre Val-de-Loire. Son objectif est de comprendre les implications environnementales de l'introduction d'un méthaniseur sur une exploitation en polyculture élevage. Le projet a permis de réaliser une enquête auprès des agriculteurs méthaniseurs de la région Centre Val-de-Loire (voir la section 2.1.2), un essai agronomique et une analyse du cycle de vie (ACV). Cette section s'intéresse à l'essai agronomique.

2.3.2 Présentation de l'essai agronomique

L'essai agronomique MétaMétha avait pour objectif de mesurer une diversité d'impacts et de bénéfices associés à la fertilisation organique (effluents animaux ou digestat) : rendement des cultures, volatilisation d'ammoniac, émissions de N_2O, lixiviation de nitrates. Un deuxième objectif était d'étudier la possibilité de mener une rotation culturale avec une fertilisation entièrement organique (pas d'apport d'engrais minéraux). L'essai a duré 3 ans (2017 à 2019), avec des cultures de blé (2016-2017),

colza (2017-2018) et blé (2018-2019). Cette rotation simplifiée a été choisie car elle permet de s'intéresser au blé, sujet à l'épandage de digestat, et au colza : l'essai avait pour volonté de regarder la valorisation des apports de fin d'été et de printemps lors d'une culture de colza. L'essai allégé se poursuit encore aujourd'hui.

L'essai compare cinq systèmes de fertilisation. Trois systèmes utilisent une fertilisation organique, avec des lisier et fumier bovin, du digestat brut (sans séparation de phase), ou des digestats liquides et solides (séparation de phase). Les deux autres traitements consistent en un système témoin avec une fertilisation minérale, et un système contrôle sans fertilisation (Tableau 2.2). Les traitements organiques ont reçu deux types d'apports :

- des apports de lisiers, de digestats liquides, ou de digestat bruts en sortie d'hiver ou printemps (produits liquides dont l'épandage est plutôt raisonné comme un fertilisant), pour apporter l'azote nécessaire aux cultures. Le digestat brut peut aussi être apporté après la récolte en fin d'été.
- des apports de produits solides (fumier ou digestat solide) en été, ayant pour but d'apporter de la matière organique au sol.

Chaque traitement correspond à une parcelle de 1 800 m^2 (75 x 24 m). Les parcelles sont suffisamment grandes pour pouvoir épandre les PRO à l'aide de machines agricoles comme en conditions réelles. De plus, les cinq parcelles sont chacune centrées sur un drain, qui a été utilisé pour estimer la lixiviation des nitrates (Figure 2.6).

L'originalité de cet essai repose sur une combinaison de facteurs :

- Une approche « système », qui mesure les effets de différents types de fertilisation, en couplant différents PRO considérés comme amendants (apport de matière organique) ou fertilisants (apport d'engrais azoté), à l'échelle d'une succession de culture.
- Des essais à grande échelle spatiale, autorisant l'utilisation de machines agricoles en condition réelles.
- Un suivi de nombreux flux d'azote au champ pendant 3 ans, décrit en section 2.3.5.

Tableau 2.2 – Traitements étudiés sur l'essai agronomique MétaMétha

Systèmes de production schématisés par les traitements	Code	Apport au printemps	Apport en été
Ferme en polyculture élevage	CSM	Lisier bovin	Fumier bovin
Ferme en polyculture élevage avec un méthaniseur, sans séparation de phase	RD	Digestat brut	Digestat brut
Ferme en polyculture élevage avec un méthaniseur, avec séparation de phase	LSD	Digestat liquide	Digestat solide
Témoin, fertilisation minérale	MN	Engrais minéraux	/
Contrôle, absence de fertilisation	0N	/	/

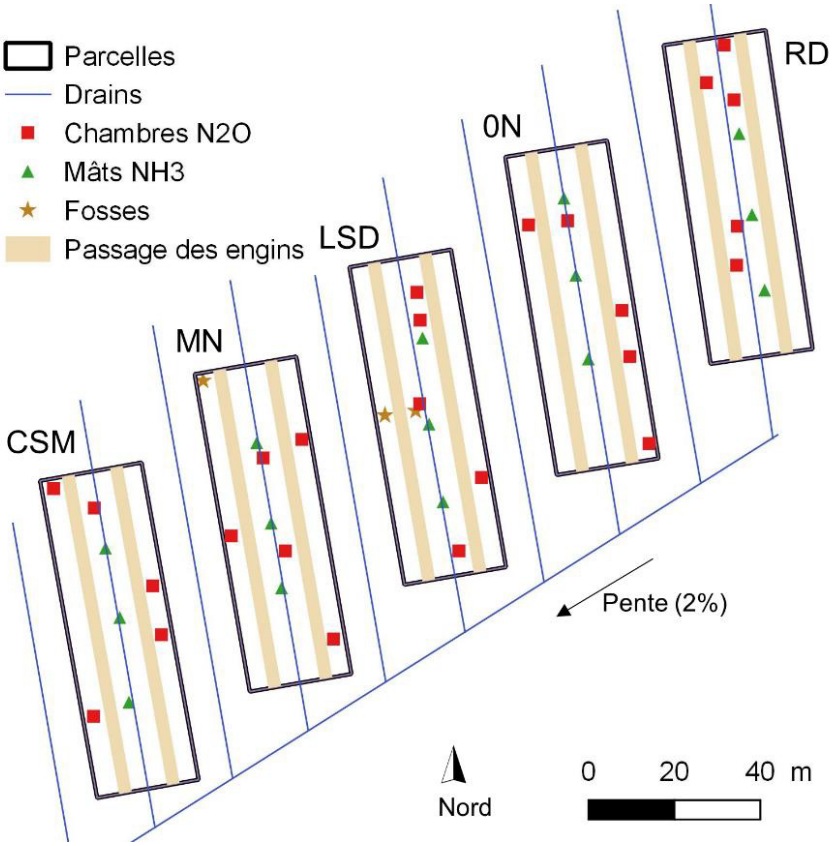

Figure 2.6 – Plan d'expérimentation de l'essai agronomique MétaMétha

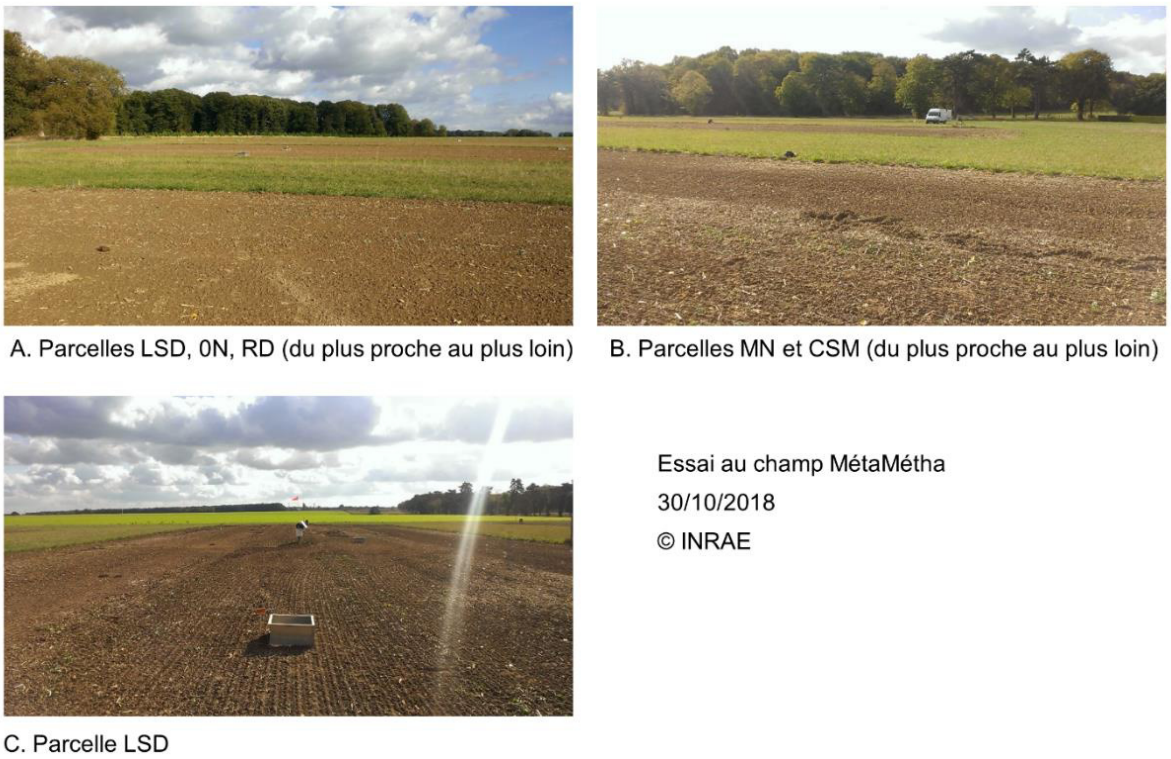

Figure 2.7 – Photographies de l'essai au champ MétaMétha, le 30/10/2018. © INRAE.

2.3.3 Caractérisation du sol sur l'essai agronomique

Le sol de l'essai a été caractérisé précisément début 2017 par l'UR SOLS (INRAE, Orléans). Cette section résume les principales analyses et conclusions sur l'étude des sols de l'essai MétaMétha.

En janvier 2017, le sol a été prélevé sur 9 points par parcelle et à 4 profondeurs par point. Les teneurs en C et N total (combustion sèche), $CaCO_3$ (acidification et dosage du CO_2 émis), la capacité d'échange cationique (CEC, méthode de Metson), ainsi que la texture (sédimentation) ont été analysées par le Laboratoire d'Analyse des Sols (LAS, INRAE, Arras). Des analyses de densité de sol (méthode au cylindre) ont été faites dans les différents horizons et au cours des trois années. En plus de cette caractérisation initiale, une campagne de mesure de la résistivité électrique a été conduite par l'entreprise CIBLAGRO en octobre 2016 ; les données ont été analysées par l'UR SOLS. Les trainées électriques sont espacées de 6 m, sur trois « profondeurs » (0 – 50 cm, 0 – 100 cm, 0 – 150 cm). L'acquisition s'est faite par des trainés électriques espacés de 6 m. À humidité constante, la résistivité est corrélée à la teneur en argile. Ces mesures ont permis d'analyser l'homogénéité des sols dans la parcelle et de différencier des classes de sols. Trois fosses pédologiques ont été creusées et caractérisées (Figure 2.6), pour déterminer le type de sol du site.

Le sol de la majeure partie de la surface de l'essai est un Luvisol typique redoxisol (estimation 75%, UR SOLS), mais il y a aussi présence de Planosol sédimorphe (estimation 25%, UR SOLS), définis selon le référentiel pédologique (Association française pour l'étude des sols et al., 2008). La profondeur du sol est d'environ 100 cm. La pente sur le site est de 2% (Figure 2.6). Les caractéristiques des sols de surface dans les différents traitements sont présentées dans le Tableau 2.3. Les densités de sols utilisées dans les calculs de reliquats (entre autres) sont les suivantes : 1,37 g cm^{-3} (0 – 20 cm), 1,55 g cm^{-3} (20 – 40 cm) et 1,60 g cm^{-3} (40 – 100 cm). Les teneurs en $CaCO_3$ sont très faibles (< 2 g kg^{-1}). Sur la surface de l'essai, on observe un léger gradient dans la nature du sol. De l'ouest vers l'est, la texture s'enrichit légèrement en argile et en limon et s'appauvrit en sable (Tableau 2.3). Tous les prélèvements appartiennent à la même texture (limon fin). La teneur en argile du sol est plus élevée en profondeur qu'en surface. En profondeur, on voit un appauvrissement en argile de l'ouest vers l'est. Ceci est interprété comme une possibilité de surface plus importante de Planosol à l'ouest qu'à l'est. On observe aussi un léger gradient de CEC et de pH. On observe donc des différences entre les parcelles, même si les sols restent de même nature. Cette limite a été assumée dès le montage du projet, au regard des avantages de configuration du plan expérimental (section 2.3.2).

Tableau 2.3 – Caractéristiques des sols de surfaces (0 – 20 cm). Les résultats sont indiqués sous la forme moyenne ± écart type (n=9). Les parcelles sont ordonnées de l'ouest (CSM) vers l'est (RD). L'absence de lettre communes indique que les teneurs sont significativement différentes (ANOVA et Tukey tests, $p < 0.05$).

	Argile (%)	Limon (%)	Sable (%)	pH	COS (g kg^{-1})	C/N	CEC (meq 100 g^{-1})
CSM	15,4 ± 1,5a	57,3 ± 1,3a	27,2 ± 1,4a	6,4 ± 0,2ab	14,6 ± 1,3a	10,3 ± 0,1a	8,5 ± 0,6a
MN	16,1 ± 1,5a	59,2 ± 1,3a	24,7 ± 0,7b	6,7 ± 0,3a	14,6 ± 1,3a	10,1 ± 0,2b	8,9 ± 0,7ab
LSD	17,1 ± 1,4a	62,3 ± 1,0b	20,6 ± 2,0c	6,5 ± 0,4ab	13,3 ± 1,0ab	10,1 ± 0,1b	8,9 ± 0,9ab
0N	20,2 ± 3,0b	63,5 ± 3,0b	16,4 ± 0,9d	6,4 ± 0,2bc	13,4 ± 0,6ab	10,1 ± 0,2b	10,0 ± 1,5b
RD	15,8 ± 1,7a	70,6 ± 1,6c	13,6 ± 0,7e	6,1 ± 0,2c	12,8 ± 1,3b	10,4 ± 0,2a	6,9 ± 1,1c

COS : Carbone Organique du Sol (COS).

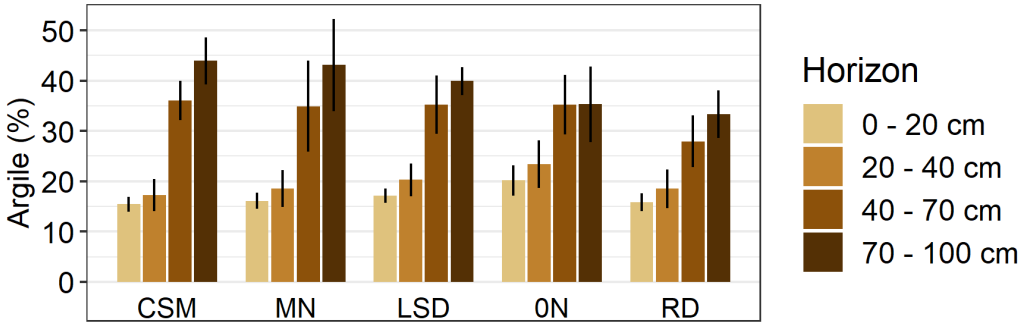

Figure 2.8 – Teneur en argile du sol selon les différents horizons. Les barres d'erreurs indiquent un écart-type (n = 9). Adaptée d'une figure de Catherine Pasquier.

2.3.4 Itinéraire technique agricole

L'essai agronomique est géré de manière conventionnelle avec une rotation simplifiée blé – colza – blé. Les opérations agricoles sont décrites dans le Tableau 2.4. Excepté au 01/05/2019, les engrais de synthèse azotés sont de la solution azotée (30% d'azote, c'est-à-dire 15% d'azote uréique, 7,5% d'azote nitrique et 7,5% d'azote ammoniacal), épandue à l'aide d'un pulvérisateur. Le lisier, le digestat brut et le digestat liquide sont épandus à l'aide d'une tonne à lisier équipée d'une rampe à pendillard (12 m). Les fumiers et digestats solides sont épandus à l'aide d'un épandeur à fumier à hérisson verticaux (Figure 2.9). Les PRO épandus ont été prélevés avant et au moment de chaque épandage, puis analysés par un laboratoire externe (Auréa, Orléans, France). Les caractéristiques des PRO mesurées sont les teneurs en matière sèche, azote total, azote ammoniacal, matière organique, P, K et le pH. La teneur en C, utilisée pour calculer le rapport C/N, est supposée être égale à la moitié de la teneur en matière organique. Le nombre de dates d'analyse est de 3 pour le digestat solide et le fumier, de 7 pour le digestat liquide et le lisier, et de 10 pour le digestat brut. Les doses d'apports et les analyses des PRO sont indiquées dans ce chapitre (Tableau 2.5, Tableau 2.6). Le Tableau 2.5 est également présenté et commenté dans le Chapitre 3. Le Tableau 2.6 est présenté de façon synthétique et commenté dans le Chapitre 3.

a. Épandage du digestat liquide b. Épandage du digestat solide

Figure 2.9 – Matériel agricole utilisé pour l'épandage a) des PRO liquides, et b) des PRO solides. Victor Moinard, UMR ECOSYS, INRAE

Tableau 2.4 – Itinéraire technique de l'essai agronomique MétaMétha

Culture	Date	Action
2016 – 2017 Blé	26/10/2016 et 27/10/2016	Préparation du sol : déchaumeur à disque (15 cm),
	27/10/2016	covercrop (10 cm), suivi du semis de blé (variété Syllon, 260 gr m^{-2})
	22/11/2016	Herbicide (FOSBURI 0.50 l ha^{-1}, HERBAFLEX 1.56 l ha^{-1})
	22/03/2017	Fertilisation
	19/04/2017	Fertilisation
	04/05/2017	Herbicide (NICANOR PREMIUM 30 g ha^{-1})
	15/05/2017	Fongicide (CERIAX 1.24 l ha^{-1})
	20/07/2017	Récolte du grain (récolte machine et prélèvements manuels)
	23/07/2017	Récolte de la paille (récolte machine)
2017 – 2018 Colza	02/08/2017	Apport des PRO solides
	03/08/2017	Enfouissement et préparation du sol : labour (20 cm), covercrop (10 cm), rouleau (2 cm)
	21/08/2017	Semis de colza (variété Fernando, 40 gr m^{-2})
	21/08/2017	Herbicide (CENTIUM 0.20 l ha^{-1}, RAPSAN TDI 2.00 l ha^{-1})
	25/10/2017	Insecticide (PROTEUS 0.63 l/ha, EMULSOL 0.05 l l ha^{-1})
	21/03/2018	Fertilisation
	23/03/2018	Fongicide insecticide (MYSTIC EXTRA 0,6 l ha^{-1}, DASKOR 440 0,595 l ha^{-1})
	04/07/2018	Récolte (récolte machine et prélèvements manuels)
2018 – 2019 Blé	19/09/2018	Apport des PRO solides
	20/09/2018	Enfouissement : covercrop (10 cm)
	24/10/2018	Préparation du sol : covercrop (10 cm), suivi du semis (variété Descartes, 300 gr m^{-2})
	26/10/2018	Herbicide (ARANDA 1,11 l ha^{-1}, COMPIL 0,167 l ha^{-1}, ROXY 800 EC 1,11 l ha^{-1})
	19/02/2019	Fertilisation
	12/03/2019	Fertilisation
	26/04/2019	Herbicide (DAYTONA 0.02 kg ha^{-1}, HARMONY EXTRA SX 0.019 kg ha^{-1})
	01/05/2019	Fertilisation (traitements MN et CSM, ammonitrate)
	03/05/2019	Fongicide (COGITO 0.427 l ha^{-1})
	07/05/2019	Fongicide (VELDIG 0.769 L/ha)
	22/07/2019	Récolte du grain (récolte machine et prélèvements manuels)
	23/07/2019	Récolte de la paille (récolte machine)

Tableau 2.5 – Doses d'engrais épandus sur l'essai au champ MétaMétha. Les doses sont indiquées en t ha^{-1} (matière brute). Ce tableau est aussi présenté dans le chapitre 3 (1/2)

Culture	Date		Traitements			
			MN	CSM	RD	LSD
Blé 2017	22/03/2017	fertilizer	Solution azotée	Lisier bovin	Digestat brut	Digestat liquide
		t ha^{-1}	/	37	38	36
		kg N_{min} ha^{-1}	86	23	86	74
		kg N_{tot} ha^{-1}	86	40	169	137
	19/04/2017	fertilizer	Solution azotée	Lisier bovin	Digestat brut	Digestat liquide
		t ha^{-1}	/	60	31	32
		kg N_{min} ha^{-1}	43	46	63	63
		kg N_{tot} ha^{-1}	43	59	129	123
	Total 2017	kg N_{min} ha^{-1}	129	69	149	137
		kg N_{tot} ha^{-1}	129	99	298	260
Colza 2018	02/08/2017	fertilizer	/	Fumier bovin	Digestat brut	Digestat solide
		t ha^{-1}	/	35	32	33
		kg N_{min} ha^{-1}	/	20	66	43
		kg N_{tot} ha^{-1}	/	276	133	207
	21/03/2018	fertilizer	Solution azotée	Lisier bovin	Digestat brut	Digestat liquide
		t ha^{-1}	/	27	32	36
		kg N_{min} ha^{-1}	99	34	82	98
		kg N_{tot} ha^{-1}	99	88	166	198
	Total 2018	kg N_{min} ha^{-1}	99	54	148	141
		kg N_{tot} ha^{-1}	99	364	299	405

N_{min}: azote mineral. N_{tot}: azote total.

Tableau 2.5 – Doses d'engrais épandus sur l'essai au champ MétaMétha. Les doses sont indiquées en t ha^{-1} (matière brute). Ce tableau est aussi présenté dans le chapitre 3 (2/2)

Culture	Date		Traitements			
			MN	CSM	RD	LSD
Blé 2019	19/09/2018	fertilizer	/	Fumier bovin	Digestat brut	Digestat solide
		t ha^{-1}	/	12	16	16
		kg N_{min} ha^{-1}	/	10	44	28
		kg N_{tot} ha^{-1}	/	105	86	110
	19/02/2019	fertilizer	Solution azotée	Lisier bovin	Digestat brut	Digestat liquide
		t ha^{-1}		24	12	23
		kg N_{min} ha^{-1}	40	47	48	56
		kg N_{tot} ha^{-1}		64	57	99
	12/03/2019	fertilizer	Solution azotée	Lisier bovin	Digestat brut	Digestat liquide
		t ha^{-1}	/	18	20	18
		kg N_{min} ha^{-1}	80	19	44	39
		kg N_{tot} ha^{-1}	80	40	87	75
	01/05/2019	fertilizer	Ammonitrate	Ammonitrate	/	/
		t ha^{-1}	/	/	/	/
		kg N_{min} ha^{-1}	41	24	/	/
		kg N_{tot} ha^{-1}	41	24	/	/
	Total 2019	kg N_{min} ha^{-1}	161	100	136	123
		kg N_{tot} ha^{-1}	161	233	230	284
3 years	Total	kg N_{min} ha^{-1}	389	223	433	401
		kg N_{tot} ha^{-1}	389	696	827	949

N_{min}: azote mineral. N_{tot}: azote total.

Tableau 2.6 – Analyses physico-chimiques des PRO épandus sur l'essai au champ MétaMétha. Quand disponible, l'écart type est indiqué (n = 2 pour le 19/09/2018, n = 3 pour le 12/03/2019)

PRO	Date d'apport	MS (%MB)	N_{min} (g $kgMB^{-1}$)	N_{tot} (g $kgMB^{-1}$)	MO (g $kgMB^{-1}$)	pH	P (g $kgMB^{-1}$)	K (g $kgMB^{-1}$)
Lisier bovin	22/03/2017	1,6	0,6	1,1	11,3	7,7	0,2	0,9
	19/04/2017	0,8	0,8	1,0	4,8	7,5	0,1	0,8
	21/03/2018	7,5	1,3	3,3	56,1	6,6	0,4	3,3
	19/02/2019	4	2,0	2,7	27,6	7,5	0,5	1,4
	12/03/2019	3,2 ± 0,1	1,1 ± 0,5	2,2 ± 0,0	22,1 ± 1,1	7,9 ± 0,1	0,2 ± 0,0	2,9 ± 0,1
Fumier bovin	08/08/2017	35,0	0,6	8,0	264,4	10,0	2,0	12,9
	19/09/2018	30,1 ± 2,4	0,8 ± 0,0	8,6 ±0,3	195,4 ± 14,6	9,5 ± 0,3	2,1 ± 0,1	9,5 ± 0,2
Digestat brut	22/03/2017	6,5	2,3	4,4	43,8	7,9	1,1	2,5
	19/04/2017	6,9	2,1	4,2	47,7	8,0	0,9	2,8
	08/08/2017	6,1	2,1	4,2	40,2	8,1	0,8	3,7
	21/03/2018	7,1	2,6	5,2	46,6	8,1	1,0	3,6
	19/09/2018	7,6 ± 0,1	2,8 ± 0,4	5,5 ± 0,0	51,9 ± 1,1	8,2 ± 0,0	1,2 ± 0,1	4,5 ± 0,6
	19/02/2019	6,1	3,9	4,6	42,9	7,9	0,8	3,8
	12/03/2019	6,0 ± 0,2	2,2 ± 0,1	4,3 ± 0,1	43,1 ± 1,7	7,9 ± 0,1	0,8 ± 0,0	4,2 ± 0,1
Digestat liquide	22/03/2017	3,7	2,0	3,8	22,8	7,8	0,6	2,3
	19/04/2017	4,1	2,0	3,8	24,5	7,7	0,8	2,5
	21/03/2018	6,6	2,7	5,5	37,5	8,1	1,4	4,2
	19/02/2019	4,9	2,4	4,3	30	7,9	0,9	3,9
	12/03/2019	4,8 ± 0,0	2,2 ± 0,4	4,2 ± 0,1	29,3 ± 0,1	8,1 ± 0,1	0,9 ± 0,0	4,5 ± 0,0
Digestat solide	08/08/2017	30,7	1,3	6,2	269	9,3	2,3	4,1
	19/09/2018 ±	23,7 ± 0,7	1,8 ± 0,1	7,0 ± 0,2	240,7 ± 2,6	9,4 ± 0,1	2,7 ± 0,0	3,2 ± 0,0

2.3.5 Suivi du site

Les données mesurées pendant l'essai ont fait l'objet d'un dataverse (Pasquier et al., 2019). Le suivi du site est synthétisé ici, et est détaillé plus précisément dans le Chapitre 3. La Figure 2.10 résume les dates de mesure des différents indicateurs.

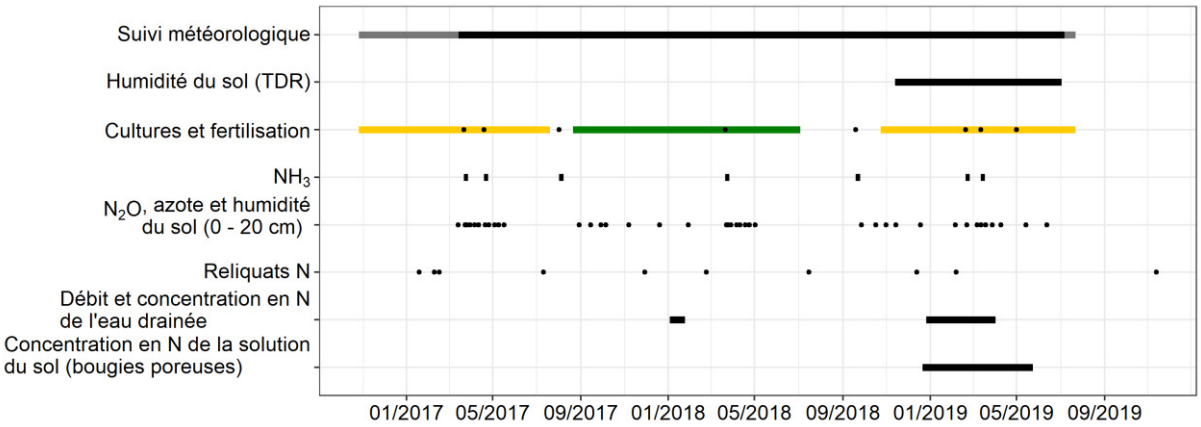

Figure 2.10 – Suivi de l'essai au champ MétaMétha. Les données météorologiques issues de la station climatique sur l'essai ou des données Météo-France sont indiquées en noir et en gris, respectivement. Cultures et fertilisation : le blé est indiqué en jaune, le colza en vert, et les dates de fertilisation par des points noirs.

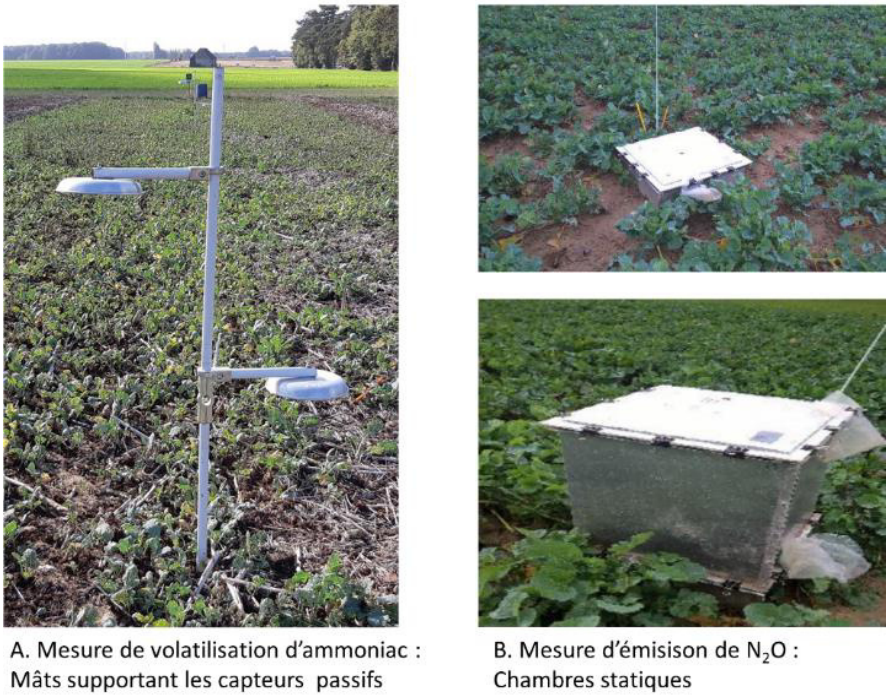

Figure 2.11 – Dispositifs de mesures A) de la volatilisation de NH_3. et B) des émissions de N_2O. Les chambres statiques sont rehaussées lors de la croissance de la culture. A) INRAE. B) Catherine Pasquier, UR SOLS, INRAE.

2.3.5.1 Conditions climatiques

Une station climatique a permis de suivre la météo du site du 15/03/2017 au 04/07/2019. Les données météorologiques avant et après cette période ont été obtenues via le jeu de données SAFRAN (Météo-France). En 2019, des sondes TDR ont permis de suivre à un pas de temps horaire l'humidité du sol. Elles sont placées à 3 emplacements correspondant aux fosses pédologiques (Figure 2.6) et à 3 profondeurs (15 cm, 30 ou 45 cm, 75 cm).

2.3.5.2 Récolte et absorption d'azote par les plantes

La récolte de chaque culture se fait à la machine sur toute la parcelle. Le grain et la paille exportés sont pesés pour déterminer le rendement machine sur l'ensemble de la parcelle. Au préalable, des prélèvements manuels sont réalisés pour analyser les teneurs en azote des grains et des résidus de culture.

2.3.5.3 Mesure de la volatilisation d'ammoniac

La volatilisation d'ammoniac a été mesurée après chaque épandage (5 au printemps, 2 en été), pendant 6 jours. Des capteurs passifs d'ammoniac (badge ALPHA©) sont installés sur des mâts à deux hauteurs (3 mâts par parcelle) (Figure 2.11). Ces capteurs sont prélevés et remplacés régulièrement pendant la période de mesure. Dans le même temps, un anémomètre sonique mesure finement la vitesse et la direction du vent. L'analyse des badges reflète la concentration en NH_3 gazeux à leur emplacement, intégrée sur la période d'exposition. Les flux d'ammoniac sont ensuite estimés à partir de modélisation inverse, à l'aide du modèle FIDES, basé sur des équations d'advection-diffusion (Loubet et al., 2010 ; Carozzi et al., 2013). Les analyses ont été menées par la BU EnVisAGES, l'UMR Ecosys et l'UE PAO.

2.3.5.4 Mesure des émissions de N_2O, suivi des teneurs en azote et en eau du sol de surface

Les émissions de N_2O ont été mesurées à l'aide de chambres statiques à prélèvement manuel (50 cm par 50 cm) (Figure 2.11). Cinq chambres sont positionnées dans chaque parcelle (Figure 2.6). Leurs positions restent identiques durant les trois ans, ces positions ont été déterminées pour représenter toutes les classes de sols (résistivité électrique). Lors de chaque mesure, les chambres sont fermées hermétiquement. Des prélèvements d'air sont réalisés régulièrement pendant 1h30. Leur concentration en N_2O est déterminée par chromatographie en phase gazeuse et détecteur par capture d'électrons. L'accumulation du N_2O dans la chambre permet de déterminer le flux. Les mesures de N_2O ont été réalisées lors de 42 dates (Figure 2.10). Les prélèvements sont plus fréquents après les épisodes de fertilisation (deux fois par semaine), puis s'espacent (jusqu'à une fois par mois environ).

À chaque date de mesure, la teneur en eau (perte de masse à 105°C pendant 48h), la teneur en azote minéral (extraction au KCl et dosage par colorimétrie) et la température du sol (sondes) sont aussi déterminées au niveau de chaque chambre. Les analyses sont menées par l'UR SOLS, l'UE PAO, et l'UMR Ecosys.

2.3.5.5 Reliquats azotés

Les reliquats azotés ont été mesurés lors de 8 dates en entrée d'hiver (novembre ou décembre), en sortie d'hiver (février) et après la récolte (juillet). Les prélèvements de sols sont réalisés à 4 profondeurs, sur 9 points par parcelle. Les teneurs en azote sont analysées par extraction au KCl et dosage par colorimétrie. Les densités de sol considérées sont 1,37 g cm^{-3} (0-20 cm), 1,55 g cm^{-3} (20-40 cm), et 1,60 g cm^{-3} (40-70 cm et 70-100 cm). Les prélèvements et analyses ont été réalisées par l'UR SOLS et l'UE PAO.

2.3.5.6 Suivi de la lixiviation en nitrates

Des débitmètres électromagnétiques (ABB AquaMAster 3 DN40) ont permis de mesurer les débits d'eau dans le drain, à la sortie de chaque parcelle (traitements MN, CSM, RD, LSD). Des préleveurs d'eau automatiques (ISCO 3700, 24 bouteilles) ont également pu permettre de collecter l'eau des drains (toutes

les 8h), et d'analyser leur concentration en nitrate. Ces équipements ont été installés en février 2017. Durant l'année 2017, très peu d'eau est prélevée, permettant la mesure de peu d'échantillons. Durant l'hiver 2017 – 2018, le faible débit d'eau et des problèmes techniques sur les préleveurs ont également limité le nombre d'échantillons prélevés. Durant l'hiver 2018 – 2019, des mesures de débits et de teneur en nitrate dans l'eau des drains a pu permettre de faire des calculs de lixiviation de nitrates. Néanmoins, plusieurs questions ont émergé et ont mis en cause la qualité de ces mesures :

- Le débit d'eau n'est pas identique dans les drains d'une parcelle à l'autre. Une partie de cette différence est expliquée par le fait qu'un des drains ne serait pas aussi long que les autres (longueur estimée à partir d'un ancien plan de drainage), mais la correction de cet effet n'explique pas toute la différence.
- Les débits en eau mesurés ne représentent qu'une faible proportion (environ 20%) des débits d'eau attendus à l'aide d'un bilan en eau simple basé sur la pluviométrie et l'évapotranspiration potentielle.
- La mise en route relativement tardive des préleveurs a induit une possible sous-évaluation de la lixiviation, une proportion non négligeable des flux d'eau ayant eu lieu avant leur mise en route.
- Les valeurs de lixiviation mesurées sont très faibles, et sont homogènes entre les parcelles. Cela n'est pas cohérent avec les reliquats d'azote en entrée et sortie d'hiver. Des essais de prise en compte de l'eau lixiviée non collectée dans les drains ne permettent pas de retrouver des valeurs plus cohérentes. Les données hydriques mesurées ne permettent pas de connaître les dynamiques de la nappe d'eau.

Les explications de ces mesures peuvent être la présence de flux d'eau latéraux (Planosol, légère pente). En conséquence, nous n'avons pas étudié ces données de lixiviation dans cette thèse.

En 2019, des bougies poreuses ont été installées, comme deuxième moyen d'estimation possible de la lixiviation. 4 bougies poreuses ont été installées sur chaque parcelle en décembre 2018 (un emplacement par parcelle, 3.5 m du drain, 70 cm de profondeur). Des prélèvements en eau ont été réalisés de façon hebdomadaire. Malheureusement, les analyses obtenues ont montré une forte variation intra-parcellaire et une faible variation inter-parcellaire. Les estimations de lixiviation possible n'ont pas non plus donné de résultats cohérents avec les reliquats azotés du sol (entrée et sortie d'hiver). En conséquence, ces données n'ont pas été non plus étudiées dans cette thèse.

Chapitre 3

Comment la méthanisation des effluents bovins modifie-t-elle le cycle de l'azote sur une rotation culturale de 3 ans ?

Nous avons vu dans le Chapitre 1 que, si les effets de l'épandage au champ sont caractérisés individuellement, peu d'études étudient ces effets de façon simultané. Ce chapitre présente donc un essai au champ pour comparer les effets des épandages d'effluents bovins ou de digestats sur l'ensemble du cycle de l'azote au champ.

Ce chapitre est écrit sous la forme d'une publication scientifique, qui a vocation à être soumise dans une revue scientifique, après adaptation et validation de la stratégie de publication par les tous auteurs.

How does anaerobic digestion of manures modify N behaviour during a 3-year crop rotation?

Victor Moinard[1], Adeline Besnault[2], Marco Carozzi[1,3], Sophie Génemont[1], Yolaine Goubard-Delaunay[1], Catherine Hénault[1], Florent Levavasseur[1], Benjamin Loubet[1], Jean-Marie Paillat[4,5], Catherine Pasquier[1], Antoine Savoie[6], Polina Voylokov[1,3], Sabine Houot[1]

1: UMR Ecosys, Université Paris Saclay, INRAE, AgroParisTech, Thiverval-Grignon, France

2: UR SOLS, INRAE, Orléans, France

3: BU EnVisAGES, INRAE, Thiverval-Grignon, France

4: UPR Recyclage et risques, CIRAD, Montpellier, France

5: UPR ADI-Suds, ISTOM, Angers, France

6: UE PAO, INRAE, Nouzilly, France

Highlights

- We compared the use of digested or undigested cattle effluents to synthetic fertilizers
- Crop N uptake was explained by mineral N input, volatilization, and organic N mineralization.
- Digestates induced similar yield and N_2O emissions than synthetic fertilizers
- Systems with digestates were the most sensitive to ammonia emissions.
- Organic applications in summer induced N losses but also likely SOM storage

Abstract

Anaerobic digestion of animal effluents at the farm is increasing in Europe to promote biogas production as a renewable energy. The use of digestates as organic fertilizers and soil amendment is expected to promote nutrient recycling, decreasing the need in synthetic fertilizers. Digestates have different fertilizer and amendment properties than undigested animal effluents, and are subjected to different N losses. We evaluated those differences in a field experiment located in the center of France, during a three-year crop succession (wheat - rapeseed - wheat). Crops were managed with five different fertilization systems : no fertilization, synthetic N fertilizers, cattle manure and slurry, raw digestate, or separated solid and liquid digestates. Five applications of exogenous organic matters (EOMs) occurred in winter and spring (chemical fertilizer, cattle slurry, liquid or raw digestates) and two applications in summer (cattle manure, solid or raw digestates). Ammonia volatilization and N_2O emissions were measured after each fertilizer application, additionally to crop N uptake and soil mineral N stocks. The dynamics of C and N mineralization from EOMs were assessed during incubations in soil under controlled conditions. Thanks to this complete N monitoring, we had a good understanding of mineral N cycle in the field. In spring, the surplus N uptake by crops compared to unfertilized treatment was well explained by the mineral N content of fertilizers, ammonia volatilization and to a lesser extent by the mineralization of organic N from EOMs. Compared to synthetic fertilizers (urea ammonium nitrate solution), the use of digestates induced similar yields, similar N_2O emissions, and 2 to 3 times higher ammonia volatilization. In the treatments with organic fertilization, 40% to 53 % of total ammonia emissions occurred during summer amendment applications. The N_2O emissions were explained by soil nitrate and ammonium contents, soil moisture, and temperature without influence of the nature of the fertilizers. Because of the imports of N with co-digested wastes, the use of digestate should also be compared to the use of undigested effluents at the farm scale.

Keywords Digestates, NH_3, N_2O, nitrate, fertilizer efficiency, N cycling

3.1 Introduction

Anaerobic digestion (AD) is a process in which organic substrates are degraded by microorganisms under anaerobic conditions to produce biogas, further used to produce energy. Various organic substrates can serve as input, such as biowastes, animal effluents, sewage sludge, or energy crops. The residual degraded matter, called digestate, is often valued in agriculture as an organic fertilizer. AD is an interesting technology to produce renewable energy and mitigate climate change (Cong et al., 2017; Hijazi et al., 2016; Lyng et al., 2018). The anaerobic digestion of biowastes can also improve nutrient cycling (Klinglmair and Thomsen, 2020; Moinard et al., 2021). Therefore, several countries in Europe and worldwide promote AD (Scarlat et al., 2018; Stürmer et al., 2020). The use of digestates as fertilizers is currently increasing, with a need to better characterize their positive and negative effects after field application.

Most of AD plants in Europe are located on farms (European Biogas Association, 2019), where the main inputs are animal effluents, which can be co-digested with other external wastes or agricultural products. In this case, raw digestates are often liquid products, similar to classical slurries (Beggio et al., 2019; Guilayn et al., 2019a; Stürmer et al., 2020). During the anaerobic digestion, organic matter is mineralized, with C removed as biogas (CO_2, CH_4), while organic N is mineralized as NH_4^+ form (Möller and Müller, 2012). Therefore, digestates often have a lower C:N ratio and a higher Total Ammoniacal Nitrogen to Total Nitrogen (TAN:TN) ratio than undigested effluents (Möller and Müller, 2012). The digestates have a more stable organic matter than the input effluents (Béghin-Tanneau et al., 2019; de la Fuente et al., 2013; Thomsen et al., 2013). The raw digestate may be post-treated with a phase separation (Guilayn et al., 2019b). The solid phase, called solid digestate, has high dry matter and volatile solid contents. It is used primarily as a soil amendment, i.e. a product that maintains or increase soil organic matter (SOM) stocks (Houot et al., 2014). The liquid phase, called liquid digestate, has high TAN content: it is used as a N fertilizer, i.e. a product that can substitute synthetic N fertilizers (Houot et al., 2014). For farmers, the introduction of AD thus implies the use of digestates instead of animal effluents with different fertilizer and amendment properties, as well as different impacts.

Multiple studies demonstrated that liquid or raw digestates substitute synthetic N fertilizers (Barłóg et al., 2019; Cavalli et al., 2016; Ferdous et al., 2020; Loria et al., 2007; Möller et al., 2008; Verdi et al., 2019), or P fertilizers (Zicker et al., 2020). However, their efficiency as N fertilizer may be influenced by ammonia volatilization and mineralization of organic N, which still needs to be better characterized (Möller and Müller, 2012; Sharifi et al., 2019). Solid digestates are often applied before sowing (Houot et al., 2014; Thomas et al., 2017). They have been sometimes demonstrated to be efficient short term N fertilizer as well (Maucieri et al., 2017). Moreover, as other exogenous organic

matters (EOMs) such as compost or solid manures, the repeated use of solid or liquid digestates could increase soil N release at the long term after repeated applications (Gómez-Muñoz et al., 2017; Gutser et al., 2005), which has been poorly explored concerning digestates. Such repeated applications could increase soil organic matter contents compared to mineral fertilizers (Odlare et al., 2011), although differences could be very small (Thomas et al., 2019). When comparing the incorporation of digested and undigested substrates, the total C remaining in soil is similar or slightly lower (Bodilis et al., 2015; Thomsen et al., 2013; Wentzel et al., 2015). Due to the diversity of digestate characteristics (Guilayn et al., 2019a), more studies are needed to better value digestate with their efficiency at increasing soil organic matter.

Liquid digestates are subjected to ammonia emissions due to high TAN contents and high pH (Nkoa, 2014), participating to air pollution and decreasing the fertilizing potential of digestate. Liquid digestate can be either more or less sensitive to volatilization than undigested slurries (Möller, 2015; Nicholson et al., 2017). Ammonia emissions from solid digestates has been poorly explored in the literature. When assessing environmental performances of manure digestion using life cycle assessments, ammonia emissions are major contributors to the acidification potential indicator, which can be a drawback (Vaneeckhaute et al., 2018; Zhang et al., 2021), even if the contrary was also found (Lyng et al., 2015; Styles et al., 2015; Wang et al., 2021). More field references are still needed to better predict ammonia emissions and improve environmental evaluation of AD. As all fertilizers, the use of digestate may trigger N_2O emissions from soils. At the plot scale, lower N_2O emissions have been reported after digestate application than after undigestated slurries (Möller, 2015; Nkoa, 2014), even if the contrary was also measured (Verdi et al., 2019). Fertilization practices, including spreading periods (e.g. autumn or spring) and management of crop residues could influence soil N cycle and N_2O emissions (Möller and Stinner, 2009; Nicholson et al., 2017). N_2O emissions could also be increased in case of repeated applications (Rosace et al., 2020). Nitrate leaching has been shown more sensitive to agricultural system changes induced by AD than to digestate physico-chemical properties (Möller, 2015).

Numerous positive and negative outcomes of the use of digestates are interacting and it is important to consider the N cycle at the scale of the crop rotation to correctly understand these interactions (Möller, 2009; Möller et al., 2008; Sieling et al., 1997; Stinner et al., 2008). If numerous references have been acquired on N fertilizing efficiency, ammonia volatilization, and N_2O emissions after digestate application, there is still a need to characterize their impacts on the whole N cycle to better identify synergies and tradeoffs induced by the digestate application. Most published field studies were done either at the scale of one year and often with a single digestate applications, only focused on fertilizing and amendment properties (Cavalli et al., 2016; Odlare et al., 2014), or on N losses without considering crop production (Nicholson et al., 2017). Solid or liquid products are often applied in different treatments (Cavalli et al., 2016; Möller et al., 2008; Nicholson et al., 2017).

In this study, we aimed at investigating two linked questions: what are the fertilizing properties of digestates compared to undigested effluents? Simultaneously, do the digestate induce higher N losses in the field? More specifically, we investigated these questions: what are the factors that explain the different N fertilizer efficiencies of the different EOMs induced by AD introduction? Can digestates entirely substitute synthetic N fertilizer for annual crops? What are the amendment properties (potential increase of soil organic C) of digestates compared to animal effluents? Do digestates emit more NH_3 and N_2O compared to undigested effluents? Here, we present a three-year field experiment, where we measured N flows during a wheat-rapeseed-wheat rotation. Five treatments represented five fertilization systems, with either undigested effluents (solid farmyard manure and liquid slurry), digested effluents with (liquid and solid phase) or without separation phase (raw digestate), chemical fertilizers, or a control without any fertilization. The kinetics of EOM organic N and C mineralization were assessed during soil-EOM incubations in laboratory controlled conditions to assess their contribution to soil mineral Nitrogen (SMN) supply and the stability of their organic matter. The N balance was assessed considering the entire crop succession.

3.2 Material and Methods

3.2.1 Site and anaerobic digester

The field experiment was realized from September 2017 to July 2019 at INRAE, Nouzilly, Centre – Val de Loire, France (47°32’ N, 0°48’ E) (Pasquier et al., 2019). The climate is temperate (oceanic climate, Cfb) with a mean annual temperature of 11.9°C and a total rainfall of 650 mm. Monthly temperature, rainfall, and windspeed during the experiments were collected from a local meteorological station and additional data from Météo France (SAFRAN) (Figure 3.1).

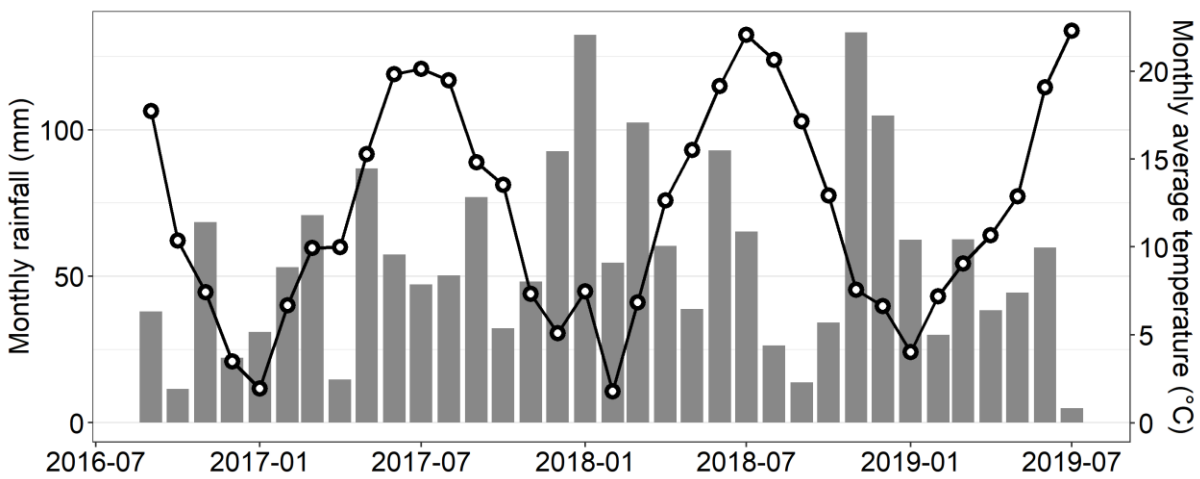

Figure 3.1 – Monthly average temperature (line, °C), and rainfall (bars, mm) during the field experiment.

The experimental unit at INRAE (UE PAO) could be considered as a crop-livestock farm with dairy cattle. Before 2014, the cattle slurry was usually stored in a covered pit before application, and the solid cattle manure on an opened platform. In 2014, an anaerobic digester was installed with a continuous wet mesophilic process, with one main digester and one post-digester. The digester treats 7,500 t of wastes per year (fresh weight), consisting of cattle slurry (11% of fresh weight), cattle, sheep, and horse manure (17%), sewage sludge (29%), agroindustrial wastes (18%), grease (8%), and cereal middlings (8%). The remaining inputs consist in site water runoff (9%) and minor fraction of other wastes (e.g. grass silage, beet pulp). The total retention time is 100 days in the 2 digesters. At the output of the post-digester, the raw digestate is post-treated with phase separation using a screw press. The liquid phase is stored in an open lagoon, and the solid phase on an open storage platform. All the EOMs are approximately stored six months before their application in the field.

3.2.2 Field experiment design

Five treatments (five fertilization systems, Table 3.1) were tested in five plots of 24 x 75 m (1800 m^2, one plot per treatment) (Supplementary Material 1). The crop succession was cultivated under conventional management (Supplementary Material 2). Winter wheat (*Triticum aestivum*) (var. Syllon) was harvested in 2017, rapeseed (*Brassica napus*) (var. Fernando) in 2018, and winter wheat (var. Descartes) in 2019. The wheat straws were exported and crop protection products were applied. The soil was tilled prior to each sowing, including ploughing (20 cm) before rapeseed sowing but not before wheat sowing.

The five treatments consisted in: (MN) fertilization with only mineral N solution in late winter and spring, and chemical P fertilizer when needed; (CSM) fertilization with cattle farmyard manure in summer and cattle slurry in late winter and spring; (RD) application of raw digestate (without phase separation) in both summer and late winter/spring; (LSD) application of solid digestate in summer and liquid digestate in late winter and spring; and (0N) no fertilization.

Mineral fertilizer, cattle slurry, raw digestate and liquid digestate were applied during late winter and spring, twice on each wheat crop, and once on rapeseed. The synthetic fertilizers were mostly Urea Ammonium Nitrate (UAN) solution with 30% N (%w/w) (15% urea, 7.5% ammonium, 7.5% nitrate). We used ammonium nitrate once, which was a solid N fertilizers with 33.5% N (w/w) (16.8% nitrate, 16.8% ammonium). They were both applied with dedicated sprayers. The liquid EOMs were applied using a trailing hose. The doses of N were determined in function of N demand by the crop, soil mineral nitrogen (SMN) in the whole soil profile in late winter, and crop N uptake in late winter. The doses of EOMs were then determine based on this N demand and their total ammoniacal N (TAN) contents. Variations to calculated doses occurred, due to the difficulty to apply a precise homogeneous dose with

a trailing hose, and to small variations in EOM analyses. We also tried not to apply too high doses. The fertilization dates were based on crop development and capacity of the soil to bear spreading engines.

Cattle manure, raw digestate, and solid digestate were applied as soil amendments at the end of summer, once before rapeseed sowing, and once before the second wheat sowing. Raw digestate was applied with a trailing hose, solid digestate and cattle manure were scattered with a manure spreader. The five plots were tilled 24 h after amendment application to bury the EOMs and prepare the soil for sowing.

3.2.3 Physico-chemical characteristics of the exogenous organic matters

All EOMs were sampled for analysis during each application. Thus, the number of analysis were 2 for cattle manure and solid digestate, 5 for liquid digestate and cattle slurry, and 7 for raw digestate.

The dry matter (DM) content was determined after 48 h of drying at 105°C (NF EN 13040). The total volatile solids (VS) (mass loss after combustion) and total nitrogen (TN) contents (total Kjedhal N) were determined by standard methodology (NF EN 13039 and NF EN 13654-1, respectively). The mineral nitrogen (N_{min}) was extracted with a 1 M KCl solution then analyzed by colorimetry on a continuous flow analyzer (Skalar, The Netherlands). In the EOMs, the nitrate content was very low and the mineral N content (N_{min}) was approximately equal to TAN content. The C:N ratio was computed assuming that C content were half of VS content. Total P and total K contents were dosed by inductively coupled plasma optical emission spectrometry (NF EN ISO 11885) after dissolution in agua regia (NF EN 13346).

I_{ROC} is an indicator proposed by Lashermes et al. (2009) to represent the proportion of EOM organic matter remaining in soil after each EOM application. It is based on the biochemical fractionation of EOM (Van Soest and Wine, 1967) and on the carbon mineralized after three days during an incubation of dried EOM. I_{ROC} has been demonstrated to be a good predictor of EOM amendment properties, i.e. its capacity at contributing to soil OM stocks (Levavasseur et al., 2020; Peltre et al., 2012). I_{ROC} was measured on the incubated EOMs following Lashermes et al. (2009) and using the equation:

(equation 3.1) $I_{ROC} = 44.5 + 0.5\ SOL - 0.2\ CEL + 0.7\ LIC - 2.3\ C3d$

where I_{ROC} was expressed in %VS, SOL, CEL, and LIG were the soluble fraction, cellulose-like fraction, and lignin-like fractions of organic matter (%VS), and C3d was the proportion of C mineralized from the dried EOM in soil after three days. The proportion of EOM C contributing to soil C stocks (recalcitrant fraction) was computed by multiplying their C content and I_{ROC}.

Table 3.1 – Application of mineral and organic fertilizers in the different treatments. Doses are reported in t ha^{-1} (Fresh Weight).(1/2)

Culture	Date		Treatment			
			MN	CSM	RD	LSD
Wheat 2017	22 March 2017	fertilizer	UAN solution	Cattle slurry	Raw digestate	Liquid digestate
		t ha^{-1}	/	37	38	36
		kg N_{min} ha^{-1}	86	23	86	74
		kg N_{tot} ha^{-1}	86	40	169	137
	19 April 2017	fertilizer	UAN solution	Cattle slurry	Raw digestate	Liquid digestate
		t ha^{-1}	/	60	31	32
		kg N_{min} ha^{-1}	43	46	63	63
		kg N_{tot} ha^{-1}	43	59	129	123
	Total 2017	kg N_{min} ha^{-1}	129	69	149	137
		kg N_{tot} ha^{-1}	129	99	298	260
Rapeseed 2018	2 August 2017	fertilizer	/	Manure	Raw digestate	Solid digestate
		t ha^{-1}	/	35	32	33
		kg N_{min} ha^{-1}	/	20	66	43
		kg N_{tot} ha^{-1}	/	276	133	207
	21 March 2018	fertilizer	UAN solution	Cattle slurry	Raw digestate	Liquid digestate
		t ha^{-1}	/	27	32	36
		kg N_{min} ha^{-1}	99	34	82	98
		kg N_{tot} ha^{-1}	99	88	166	198
	Total 2018	kg N_{min} ha^{-1}	99	54	148	141
		kg N_{tot} ha^{-1}	99	364	299	405

N_{min}: mineral N. N_{tot}: total N.

Table 3.1 – Application of mineral and organic fertilizers in the different treatments. Doses are reported in t ha^{-1} (Fresh Weight).(2/2)

Culture	Date		Treatment			
			MN	CSM	RD	LSD
Wheat 2019	19 September 2018	fertilizer	/	Manure	Raw digestate	Solid digestate
		t ha^{-1}	/	12	16	16
		kg N_{min} ha^{-1}	/	10	44	28
		kg N_{tot} ha^{-1}	/	105	86	110
	19 February 2019	fertilizer	UAN solution	Cattle slurry	Raw digestate	Liquid digestate
		t ha^{-1}		24	12	23
		kg N_{min} ha^{-1}	40	47	48	56
		kg N_{tot} ha^{-1}		64	57	99
	12 March 2019	fertilizer	UAN solution	Cattle slurry	Raw digestate	Liquid digestate
		t ha^{-1}	/	18	20	18
		kg N_{min} ha^{-1}	80	19	44	39
		kg N_{tot} ha^{-1}	80	40	87	75
	1 May 2019	fertilizer	Ammonium nitrate	Ammonium nitrate	/	/
		t ha^{-1}	/	/	/	/
		kg N_{min} ha^{-1}	41	24	/	/
		kg N_{tot} ha^{-1}	41	24	/	/
	Total 2019	kg N_{min} ha^{-1}	161	100	136	123
		kg N_{tot} ha^{-1}	161	233	230	284
3 years	Total	kg N_{min} ha^{-1}	389	223	433	401
		kg N_{tot} ha^{-1}	389	696	827	949

N_{min}: mineral N. N_{tot}: total N.

3.2.4 Soil description

The soil was characterized at the beginning of the experiment in 2017, with nine soil sampling in each plot. It was mainly a Stagnic Luvisol, with minor surface described as a Haplic Planosol (WRB, 2015). Soil depth was 100 cm. We analyzed the following properties on 9 samples per plot in 2017: particle size distribution to determine texture (NF X 31-107), total C and N contents by dry combustion, gas chromatography, and thermal conductivity detector (NF ISO 10694 and NF ISO13878), pH in a water suspension (NF ISO 10390), cation-exchange capacity (CEC) (NF X 31-130), $CaCO_3$ content by acidification and measurement of released CO_2 volume (NF ISO 10693), mass loss after combustion for SOM (1100 °C), corrected by $CaCO_3$ content. The bulk density was measured in the upper layer on fifteen locations with the core method (NF X31-501). The characteristics of the topsoil (0-20 cm depth) are described in Table 3.2 for each plot. We observed a slight gradient of texture between the plots. The topsoil was silty loamy. The bulk density was 1.37 g cm^{-3} (standard deviation: 0.05). The $CaCO_3$ content was low (1.4 g kg^{-1}, standard deviation: 0.3).

Table 3.2: Soil physico-chemical characteristics in the upper layer (0-20 cm) of each treatment plot. Results are expressed as mean ± standard deviation (n = 9).

	Clay (% w/w)	Silt (% w/w)	Sand (% w/w)	pH	SOC (g kg^{-1})	C:N	CEC (meq 100 g^{-1})
MN	16.1 ± 1.5	59.2 ± 1.3	24.7 ± 0.7	6.7 ± 0.3	14.6 ± 1.3	10.1 ± 0.2	8.9 ± 0.7
CSM	15.4 ± 1.5	57.3 ± 1.3	27.2 ± 1.4	6.4 ± 0.2	14.6 ± 1.3	10.3 ± 0.1	8.5 ± 0.6
RD	15.8 ± 1.7	70.6 ± 1.6	13.6 ± 0.7	6.1 ± 0.2	12.8 ± 1.3	10.4 ± 0.2	6.9 ± 1.1
LSD	17.1 ± 1.4	62.3 ± 1.0	20.6 ± 2.0	6.5 ± 0.4	13.3 ± 1.0	10.1 ± 0.1	8.9 ± 0.7
0N	20.2 ± 3.0	63.5 ± 3.0	16.4 ± 0.9	6.4 ± 0.2	13.4 ± 0.6	10.1 ± 0.2	10.0 ± 1.5

SOC: soil organic carbon.

3.2.5 Field trial monitoring

3.2.5.1 Aboveground biomass, grain yield and N content

The crops were harvested on the whole plot surface (1800 m^2) with classical machinery. The whole harvested grain was weighted to measure grain yield. The uncertainty in yield was computed considering the uncertainty measurement of the balance. Just before harvest, the crops were sampled in 3 pooled subplots (total 0.2 m^2) for wheat 2017, 3 subplots (total 3 m^2) for rapeseed 2018, 5 subplots (total 1.5 m^2) for wheat 2019. We measured total aboveground and grain biomass. N content in grain and straw, and protein content in wheat grain were analyzed by a specialized laboratory. We used these measures to compute the N export in grain at harvest, and the total aboveground N uptake (including straw and

grain). Detailed calculations are in Supplementary Material 3. Their uncertainties were computed using error propagation formulas.

Aerial biomass and N uptake of rapeseed were also evaluated similarly on 3 subplots per treatment (total 3 m^2) on 20th of November 2017 and 20th of February 2018 to drive N fertilization.

3.2.5.2 Ammonia volatilization

The volatilization of ammonia was measured after each fertilizer applications, using passive ALPHA samplers (Adapted Low-cost Passive High Absorption). On each event, immediately after the application of the EOMs or chemical fertilizers, 3 masts were installed in each plot uniformly distributed in the middle line of each plot. On each mast, three passive samplers were installed at two heights (30 cm and 1 m above soil or vegetation). Three masts with three samplers each (3 m - high) were also installed in three opposite directions at 300 m from experimental field, to measure environmental NH_3 concentration. Samplers were changed regularly, every 12 h during 24 to 48 h, then once a day to once every two days until day 4 to 6. During the whole measurement period, wind speed and 3-dimensional wind direction were measured with a sonic anemometer installed at 2 m height on the field site with a frequency of 20Hz (Gill Instruments, United Kingdom). Total cumulated NH_3 volatilization (kgN ha^{-1}) after 6 days and associated uncertainty, were estimated from samplers concentrations and meteorological data with reverse modelling using the FIDES model (Carozzi et al., 2013b; Loubet et al., 2010).

3.2.5.3 Soil mineral Nitrogen

The stocks of soil mineral Nitrogen (SMN) over the whole soil profile were measured in February to drive fertilizer doses, after harvest to calculate the mineral N left by the crops, and in November (second and third year), to assess potential winter nitrate leaching in winter. At each date and on each treatment, 9 soil cores were sampled and pooled in 1 to 3 pools, at four depths (0-20 cm, 20-40 cm, 40-70 cm, 70-100 cm). The N-NO_3^- and N-NH_4^+ contents were analyzed by colorimetry after KCl extraction. SMN contents were calculated as the sum of NO_3^--N and NH_4^+-N contents. SMN stocks (kgN ha^{-1}) were computed using soil bulk density (1.37 g cm^{-3} on 0-20 cm, 1.55 g cm^{-3} on 20-40 cm, 1.60 g cm^{-3} on 40-100 cm).

3.2.5.4 N_2O emissions

N_2O emissions were measured with the static chamber method (Gu et al., 2013; Jeuffroy et al., 2013). Each plot was equipped with 5 static chambers. The position of each chamber was semi-randomly chosen to be representative of the whole plot surface. They were located more than 1.5 m from the plot border, avoiding wheel tracks. After a spring fertilizer application, N_2O emissions were measured twice a week during 2 weeks, then once a week for one month and once a month during late spring and summer. After a summer fertilizer application, N_2O emissions were measured twice a month, and once a month in late autumn and winter before the next application event. In total, N_2O emissions were

measured at 42 dates. The static aluminum chambers measured 50 cm x 50 cm. They were 25 cm high in autumn and winter, and 75 cm high in spring when vegetation was higher. They were buried 10 cm within the soil. At each measurement date, chambers were sealed, and air was sampled at time 0 min, 30 min, 60 min, and 90 min. N_2O emissions started between 10 am and 2 pm. N_2O in air samples was analyzed by Gas Chromatography with Electron Capture Detector (Trace GC Ultra, Thermo Scientific). When the N_2O concentration in the chamber was positively correlated with time ($r > 0.9$), there was an accumulation of N_2O and the slope of the regression line was used to compute N_2O emission rate. When the N_2O did not accumulate in the chamber (correlation between N_2O content and time $r < 0.9$), N_2O emissions were set to 0.

Topsoil sampling (0-20 cm) were realized in the proximity of each chamber at each N_2O emission measurement date. Gravimetric soil water content (SWC) (% w/w) was measured by weighting and drying soil at 105°C, and converted to fraction of water-filled pore space (%WFPS) using topsoil bulk density (1.37 g cm^{-3}) and assuming that soil particle density is equal to 2.65 g cm^{-3}. SMN contents (i.e, N-NH_4^+ and N-NO_3^- contents) were measured by colorimetry after KCl extraction (see also Supplementary Material 4).

To summarize, we estimated the total emissions by integrating punctual N_2O emissions along the time during 50 days, in late summer/autumn (2017, 2018), and late winter/spring (2017, 2018, 2019). The 5 periods (250 days on three years) were then summed. This computation did not aim to estimate the right absolute N_2O emission value nor emission factors, but enabled to compare fertilization systems integrating emissions during the periods following applications.

3.2.6 Kinetics of C and N mineralization of EOMs during incubations in controlled conditions

Each EOM was incubated to follow the kinetics of mineralization of C and N. The EOM were incubated in the same soil as the field experiment (plot 0N, 0 - 20 cm). The soil was sieved fresh to 4 mm. The field capacity of the sieved soil was measured with a Richards press (30,990 Pa) and was 25% w/w. Raw digestate, solid digestate, and cattle manure were sampled on September 19th, 2018 during field application. Cattle slurry and liquid digestate were sampled on March 12th, 2019 during field application. Solid digestate and cattle manure were chopped by hand for homogenization. The EOMs were stored at 4°C in plastic containers during two months before the incubations. The EOMs were incubated fresh, in non-limiting conditions for moisture, SMN, and temperature. They were mixed homogeneously into 500 g of soil in the proportion of 2000 mg C_{org} kg DS $^{-1}$. After soil mixing, KNO_3 solution or deionized water was added when necessary to the mixture to reach a minimum SMN content of 35 mgN_{min} $kgDS^{-1}$, and a soil moisture of 25% w/w (field capacity). Mixtures were incubated in darkness at 28 ± 1 °C for 175 days.

The mineralization of organic Carbon was measured by following soil CO_2 emissions. Four replicates per EOM were used, as well as an unamended soil control treatment and blank treatment without any material. The mixture of each replicate was placed in a 3 L airtight vessel, along with a plastic vessel containing 20 mL of NaOH (1 mol L^{-1}) solution to trap CO_2 emissions. CO_2 was measured by colorimetry on a continuous flow analyser (Skalar, The Netherlands). We computed the cumulated C mineralization, which was then expressed in % C_{org} initially present in the EOM, by subtracting CO_2 emissions in control treatment (soil respiration). We evaluated the proportion of recalcitrant organic C as the remaining C in the soil after 175 days.

The behaviour of mineral N during EOM decomposition was measured with destructive replicates. Four replicates per treatment and per date of measurement were used. At dates 0, 3, 7, 14, 25, 49, 91, 175 days, we extracted and analyzed SMN as described before. At each date, to avoid accounting for initial mineral N, mineralized N-NH_4^+ and N-NO_3^- contents were computed by subtracting total N-NH_4^+ and N-NO_3^- contents by their contents at t=0 day in each treatment. Net N mineralization or immobilization was computed by subtracting mineralized N-NH_4^+ and N-NO_3^- contents in soil by the one in unamended control (soil mineralization). Net N mineralization or immobilization was then expressed in % N_{org} initially present in the EOM. We also computed the available mineral N brought by the EOM, expressed in kg N_{min} in soils per kg FW of EOM brought, taking into account both mineralization/immobilization cited above and initial TAN content in the EOM. Details in methods and computations are given in Supplementary Material 5.

3.2.7 Fertilizer N availability

For each cropping season, we evaluated the N availability (%N_{total}) of the different fertilizers. It is defined as the fraction of total applied N which can be used on the short term by the crop. It was computed considering mineral N input, volatilization, mineralization from EOMs during crop growth. The period considered in this analysis ranged from winter (February) to harvest. It was computed as follow:

(equation 3.2)

$$\text{N availability} = \frac{N_{min}\text{fertilizer input} - \text{volatilized N} + \text{mineralized N from EOM}}{fertilizer\ N_{total}\ input}$$
$$= TAN{:}TN - EF_{volat} + fertilizer\ mineralization\ .\ N_{org}{:}TN$$

N_{min} fertilizer input (kg N ha^{-1}) was computed from EOM analysis. Volatilized N (kg N ha^{-1}) was measured as described in section 3.2.5.2. The mineralized N from EOMs (kg N ha^{-1}) was estimated from their percentage of mineralized organic N during laboratory incubation (section 3.2.6.3) at day 49. Indeed, we used the function of EOM mineralization used in the STICS model (Brisson et al., 2008) and daily air temperature to estimate that the mineralization between first fertilizing event and harvest were equivalent to 60 to 70 days of incubation at 28°C, in 2017, 2018, and 2019. Fertilizer N_{total} input was the

total N applied in the field (kgN ha^{-1}). TAN:TN is the TAN:TN ratio of the fertilizer. EF_{volat} was the emission factor of volatilization relatively to applied TN. Fertilizer mineralization was the ratio of mineralized N to applied organic N, and N_{org}:TN was the ratio between applied organic N and total applied N.

We computed surplus N uptake by plant, relatively to 0N treatment (surplus N uptake, kgN ha^{-1}). Surplus N uptake was the difference of N uptake by crops (in aboveground organs) between fertilized and unfertilized treatments, from February to harvest.

(equation 3.3)

$$\text{surplus N uptake} = (crop\ N_{harvest} - crop\ N_{winter}) - \left(crop\ N_{harvest}^{0N} - crop\ N_{winter}^{0N}\right)$$

Where crop N was the N uptake in aboveground organs of the crops (kgN ha^{-1}). N uptake were not measured on wheat in February in 2017 and 2019. However, N uptake by wheat at that stage is low, it was thus neglected. The time of analysis is indicated in subscript. The indication 0N in superscript indicate that the value refers to unfertilized control treatment.

The surplus N uptake was compared to the mineral N supply, to verify whether the crop growth was correlated to the fertilizer N availability. The mineral N supply was defined as the mineral N available for plant in each plot compared to control, from February to harvest. We computed the mineral N supply from the fertilizer N availability. We also considered the difference of variation of SMN stocks in each treatment compared to control plot, which enabled to consider the differences in SMN stocks in February between the treatments. We assumed that mineralization of SOM was similar in all treatments and that the effect of incorporation of former crop residues and organic amendment on SMN stocks occurred before opening the N balance in February. Therefore, we neglected the N supply from previously applied organic matter. At last, we computed the mineral N supply (kgN ha^{-1}) with equation 3.4:

(equation 3.4)

$$\begin{aligned} Mineral\ N\ supply \\ &= N\ availability\ .\ fertilizer\ N_{total}\ input + (SMN_{winter} - SMN_{harvest}) \\ &- \left(SMN_{winter}^{0N} - SMN_{harvest}^{0N}\right) \end{aligned}$$

Where N availability was computed from equation 3.2 (%N_{total}), fertilizer N_{total} input was the total N applied with the fertilizers (kgN ha^{-1}), and SMN was the SMN stocks in the whole soil profile (kgN ha^{-1}). The time of analysis is indicated in subscript. The indication 0N in superscript indicates that the value refers to unfertilized control treatment.

We also computed the N use efficiency (NUE) (%N_{total}) of each fertilizers was then computed as:

(equation 3.5) $$NUE = \frac{surplus\ N\ uptake}{fertilizer\ N_{total}\ input}$$

NUE considered only the N applied in late winter and spring.

3.2.8 Field N balance

3.2.8.1 Mineral N balance

To verify the consistency of all N flux measurements, we computed the mineral N balance in the soil. We computed the balance once, during a period of three years that started from February 2017 for the first fertilizing event of the first wheat until the harvest of the second wheat in July 2019. We first computed the sum of mineral N inputs, i.e. the processes that supplied mineral N to crops: the mineral N input from fertilizers, the decrease in SMN stocks between the end and the beginning of the experiment, the mineralization of organic N from applied EOMs, and the mineral N supply from SOM and crop residues. Atmospheric deposition was neglected. Then, we compared the total input to the total outputs of mineral N. The outputs of mineral N are all the processes that remove mineral N from the soil : nitrate leaching, ammonia volatilization, N immobilization in SOM induced by EOM decomposition, N uptake by crops (aboveground and belowground biomass). The emissions of N_2O and N_2 were neglected.

The mineral N input from fertilizers and ammonia volatilization were measured. So were the N uptake in plant (aboveground organs) that considered exported grains and crop residues. The organic N mineralization or immobilization following EOMs application were deduced from the incubation in controlled conditions, as previously described, from application to the end of the experiment. We estimated nitrate leaching every year by computing the difference of SMN stocks at the beginning and at the end of winter. The N uptake in wheat roots was considered to be 15% of total N uptake (Allard et al., 2013). N uptake in rapeseed roots and in dead leaves, which was not measured, were considered to be 6% and 11% of N uptake in aerial rapeseed at harvest, respectively (Malagoli et al., 2005). The difference in SMN stocks was computed once between the end and the start of the experiment. Total N uptake in 0N control treatment were assumed to be a proxy of the net soil N supply via SOM and crop residues and roots decomposition. When the soil was bare in summer, the mineralized N was assumed to stay in the soil and to be used by the following crop.

3.2.8.2 Total N balance

We also computed the balance of total N during the crop rotation to estimate the incorporation of organic N into the SOM. The only input of total N came from the fertilizers and EOMs. Total N outputs were N in exported crops (measured), ammonia volatilization (measured), and nitrate leaching (estimated, see above). Emissions of N_2O and N_2 were neglected. The balance between input and output was considered an input of soil organic N.

3.2.9 Statistical analyses

Statistical analyses were realized with the R software (v4.0.2) (R Core Team, 2020).

The uncertainty on agricultural performance was low, because the whole plots were measured: we knew precisely what the crop yields were. However, the variabilities of the crop growth inside each plot were not characterized and no statistical test could be performed to distinguish the agricultural performance of the treatments.

At each date, we used a Kruskal-Wallis test to verify whether N_2O emissions were different between treatments. If the treatments had significantly different N_2O emissions, ($p < 0.05$), a post-hoc Dunn's test was used to find which treatment was different from one another ($p < 0.05$). We used the kruskal.test function, and the dunn.test function from the *dunn.test* package (Dinno, 2017). Similarly, we verified whether NH_3 volatilization (total and emission factors) differed between treatments for each application event.

Ammonia volatilization was obtained from a reverse modelling approach that provided a mean and standard deviation of the measure for each treatment and event. Therefore, no statistical tests were realized to compare the treatments. The cumulated NH_3 volatilization and N_2O emissions cannot be realized for each replicates, ad no statistical tests were realized to differentiate the treatments.

We analyzed the major drivers of NH_3 volatilization after application of liquid EOMs using a linear model. Solid digestate and cattle manure were excluded from the analysis. The analyzed datasets contained 17 points, i.e. 5 fertilization dates and 3 EOMs in winter/spring, 2 dates with raw digestate in summer. The response variable of the model was the volatilization emission factor, i.e. the percentage of applied TAN which volatilized. Tested explanatory variables were: mean air temperature (°C, 24h, 72h, or 6 days), total rainfall (mm, after 24h, 72h, or 6 days), mean wind speed (m s^{-1}, during 24h, 72h, or 6 days), pH of EOMs, DM content of EOMs (g kg^{-1}), TAN content of EOMs (gN $kgFW^{-1}$), type of EOMs, and a Boolean variable indicating whether the EOM was burrowed. Different models were built successively, using the lm function of the R software (R Core Team, 2020). We used the stepAIC function from *MASS* package (Venables and Ripley, 2002) to select the best model, using an automatic "stepwise" approach that iteratively add or remove one explanatory variable at a time. The successive models were compared using the Aikake Indicator Criterion (AIC), and the model with the lower AIC was selected. The consistency of the model was verified by hand. The assumptions of the linear model were visually verified.

We analyzed the major determinant of N_2O emissions. The analyzed datasets contained 185 points, i.e. 5 treatments and 37 dates. The response variable of the model were the logarithm of N_2O emissions. Tested explanatory variables were: topsoil water content (% WFPS), $N\text{-}NH_4^+$ topsoil content (mg kg DS^{-1} (dry soil)), $N\text{-}NO_3^-$ topsoil content (mg kg DS^{-1}), SMN topsoil content (mg $kgDS^{-1}$), mean

daily air temperature (°C), season (was the last application of fertilizer in winter/spring or summer?), treatment, date expressed in days from the beginning of the experiment (to evaluate any adaptation effect). The best model was investigated as described previously.

3.3 Results

3.3.1 Characteristics of exogenous organic matters

Solid digestate and cattle manure are solid products, with high DM and VS contents (Table 3.3). They had similar basic pH and similar DM, VS, and P contents. The main differences were a higher TAN:TN ratio in solid digestate (24%) compared to manure (8%), and a higher K content in manure (11.3 gK $kgFW^{-1}$) compared to solid digestate (3.6 gK $kgFW^{-1}$), and a slightly higher TN content in manure (8.3 gN $kgFW^{-1}$) compared to solid digestate (6.6 gN $kgFW^{-1}$). Cattle slurry, raw digestate and liquid digestate are liquid products, with high TAN contents, and similar high TAN:TN ratio (50% to 60%). Raw and liquid digestates were similar, with only a slightly higher DM and VS contents in raw digestate than in liquid digestate. The cattle slurry had DM, VS, and K contents that were similar or lower than the one in digestates. The slurry pH was lower than the one of digestates, and the TN and TAN contents were more than 30% lower in slurry (2.1 gTN $kgFW^{-1}$) compared to liquid digestate (4.3 gTN $kgFW^{-1}$) or raw digestate (4.6 gTN $kgFW^{-1}$). Slurry also had a lower P content than digestates.

The chemical characteristics (DM, VS, TN, TAN, P, K contents) of solid digestates, cattle manure, raw digestates, and liquid digestates varied from one year to another, the coefficient of variations of these characteristics were up to 25%. Only pH were less variable with coefficients of variation lower than 3%. The characteristics of cattle slurry were much more variable. This was partly explained because the slurry was less concentrated in 2017 compared to 2018 and 2019; however, the variability of the slurry was higher than the one in digestate even if we considered only 2018 and 2019 (Supplementary Material 6). Anaerobic digestion seemed thus to lower the variation of the characteristics of the liquid EOM through the time. The coefficient of variation for TAN content were 47%, 25%, and 13% for slurry, raw digestate, and liquid digestate, respectively.

The decomposition of the EOMs induced a net N mineralization for liquid digestate (27% of the initial organic N N_{org} at day 49), a net N immobilization for cattle slurry (-19%) and solid digestate (-28%), and a variable to neutral N release for raw digestate and cattle manure (Figure 3.2.A). The N availability of the EOMs, that considered the net mineral N due to TAN inputs and N mineralization, is displayed in Figure 3.2.B. At the beginning of the incubation, the liquid digestate had a slightly lower TAN content than the raw digestate. The N availability of the liquid digestate increased with the time due to the mineralization of the organic N, and became similar or higher than the N availability of raw digestate. Compared to liquid and raw digestate, cattle slurry would have lower mineral N availability for crops, mainly because of lower TAN content and, on a lesser extent, of N immobilization. Solid digestate had

a higher TAN content than manure, but the induced N immobilization after addition to soil decreased the mineral N availability from solid digestate and both solid digestate and cattle farmyard manure had equivalent potential N availability.

All EOMs presented rather large rates of organic C mineralization (Figure 3.2.C) with intense mineralization during 28 days then decreasing and reaching a plateau at the end of the incubation. The raw digestate presented the lowest mineralization kinetics with unexpected larger intensity of mineralization for both solid and liquid phases. The remaining organic C in soil was calculated as the difference between added and mineralized organic C after 175 days (Table 3.3). The data were consistent with the I_{ROC} value measured except for the raw digestate for which residual C during incubation was higher than measured through the I_{ROC} value ($r^2 = 0.98$, excluding raw digestate). We observed an intense C mineralization on the first day of incubation for liquid digestate and slurry, indicating the presence of highly degradable organic matter (Figure 3.2.C). This peak was not observed for raw digestate.

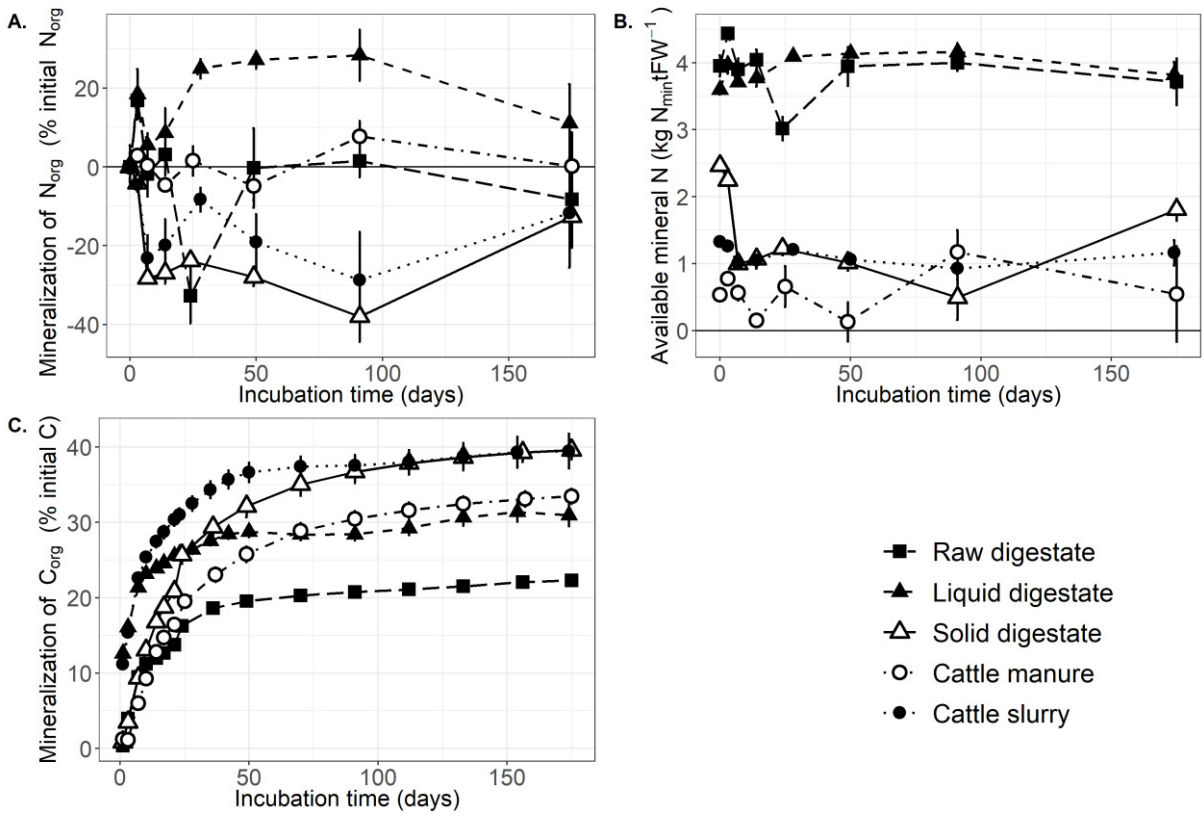

Figure 3.2: Kinetics of C and N mineralization from EOMs during incubations in soils under controlled conditions. For each subfigure, error bars show one standard error. A: Cumulated C released under CO_2 form, expressed in percentage of initial organic C from the EOM. B: Cumulated N mineralization or immobilization, expressed in percentage of initial organic N in the EOM. C: Cumulated mineral N supply by the EOM, taking into account mineral N input together with organic N mineralization.

Table 3.3: Average physicochemical characteristics of the EOMs, calculated from the analysis done for each application. Results are indicated as: mean ± standard deviation (coefficient of variation). Stable C fraction during incubation and I_{ROC} were analyzed on one sample; the mean was written followed by the standard error between square brackets (standard error).

	Cattle farmyard manure	Solid digestate	Cattle slurry	Liquid digestate	Raw digestate
DM (%FW)	32.5 ± 3.4 (11%)	27.2 ± 4.9 (18%)	3.4 ± 2.6 (77%)	4.8 ± 1.1 (23%)	6.6 ± 0.6 (9%)
pH	9.7 ± 0.3 (3%)	9.3 ± 0.1 (1%)	7.4 ± 0.5 (7%)	7.9 ± 0.2 (2%)	8.0 ± 0.1 (2%)
VS (g $kgFW^{-1}$)	253 ± 17 (7%)	232 ± 52 (22%)	24.4 ± 19.9 (81%)	28.8 ± 5.7 (20%)	45.2 ± 3.9 (9%)
Stable C (%C_{org}) from incubations	66.5 [1.0]	60.4 [1.2]	60.5 [2.3]	69.0 [1.5]	77.7 [0.4]
I_{ROC} (%C_{org})	67.0 [0.3]	58.0 [1.1]	56.0 [0.7]	72.6 [2.4]	59.7 [3.5]
TAN (g $kgFW^{-1}$)	0.7 ± 0.2 (22%)	1.6 ± 0.4 (24%)	1.1 ± 0.5 (47%)	2.3 ± 0.3 (13%)	2.6 ± 0.6 (25%)
TN (g $kgFW^{-1}$)	8.3 ± 0.4 (5%)	6.6 ± 0.5 (8%)	2.1 ± 1.0 (49%)	4.3 ± 0.7 (16%)	4.6 ± 0.5 (11%)
TAN:TN (%)	8 ± 1.0 (17%)	24 ± 4 (16%)	59 ± 16 (28%)	53 ± 3 (5%)	55 ± 13 (24%)
$C:N_{total}$	15.3 ± 1.8 (11%)	17.8 ± 5.4 (30%)	5.2 ± 2.2 (42%)	3.3 ± 0.2 (6%)	4.9 ± 0.4 (8%)
P (g $kgFW^{-1}$)	2.1 ± 0.1 (5%)	2.5 ± 0.3 (12%)	0.3 ± 0.2 (59%)	0.9 ± 0.3 (34%)	1.0 ± 0.2 (17%)
K (g $kgFW^{-1}$)	11.3 ± 2.4 (21%)	3.6 ± 0.6 (17%)	1.8 ± 1.2 (63%)	3.5 ± 1.0 (29%)	3.6 ± 0.7 (21%)

Organic matter in the undigested farmyard manure (I_{ROC} = 67%VS) were slightly more stable than solid digestate (I_{ROC} = 60%VS), confirmed by a higher I_{ROC} and residual C during incubation. On the contrary, the organic matter stability of the slurry was slightly lower (I_{ROC} = 61%VS) than the one of liquid digestate (I_{ROC} = 69%VS). According to I_{ROC}, the stable organic matter remained preferentially in the liquid phase during the phase separation. However, the differences between these organic matter stabilities were rather small.

3.3.2 Ammonia emissions after application of exogenous organic matters

In summer, 99% to 100% of the total volatilization occurred during the first 2 days then stopped because of the EOM burying (data not shown). The quick and important volatilization for all EOMs were attributed mainly to high temperature (Figure 3.3.A). The very high emission factors for cattle manure and solid digestate (up to 100% of TAN) represented 5 to 24 kg of N losses despite of the low TAN content of those products.

After application of EOMs in late winter and spring, 64% to 100% of the total volatilization occurred during the first 2 days (data not shown). The EOMs were not buried. Across the five events, absolute volatilization values and emissions factors varied according to EOM doses, meteorological conditions, and product characteristics (Figure 3.3.B). The volatilization varied from 0 to 5 kgN ha^{-1} for chemical N solution (0 to 7% of TAN brought); from 0 to 8 kg N ha^{-1} for cattle slurry (0 to 40% of TAN applied); from 7 to 38 kg N ha^{-1} for raw digestates (13 to 60% of TAN applied); and from 2 to 16 kg N ha^{-1} for liquid digestate (3 to 29% of TAN applied). Considering the five applications, the EOMs had higher emission factors than chemical solution. Raw digestate was the most sensitive liquid EOM to volatilization, regarding both emission factors or total volatilization. Liquid digestate had similar or slightly higher emission factors than cattle slurry; however, the total volatilization was higher for liquid digestate due to higher doses of applied TAN.

The best linear regression model (adjusted $R^2 = 0.63$) found to explain the percentage of TAN volatilized after the application of liquid EOMs was:

(equation 3.6) $EF_{volat} = - 245.1 – 17.1\ TAN + 6.8\ DM + 25.8\ pH + 8.8\ T - 1.3\ R - 76.0\ BURIED.$

where EF_{volat} was the emission factor of ammonia after 6 days (% applied TAN), TAN was the TAN content of the organic product (gN $kgFW^{-1}$), DM was the DM content of the EOM (g $kgFW^{-1}$), pH was the pH of EOM, T was the mean air temperature during 6 days (°C), R was the total rain during 6 days (mm), and BURIED was 1 if the EOM was buried and 0 if not. Wind speed, application dose (tFW ha^{-1}), and type of EOM were not selected in the model. The climatic variables were better explanatory variables when considered on 6 days than when they were considered on 1 or 3 days. The model showed a positive correlation ($p < 0.05$) of volatilization to DM content, pH, air temperature, and a negative correlation ($p < 0.05$) of volatilization to rainfall and TAN content, and burying.

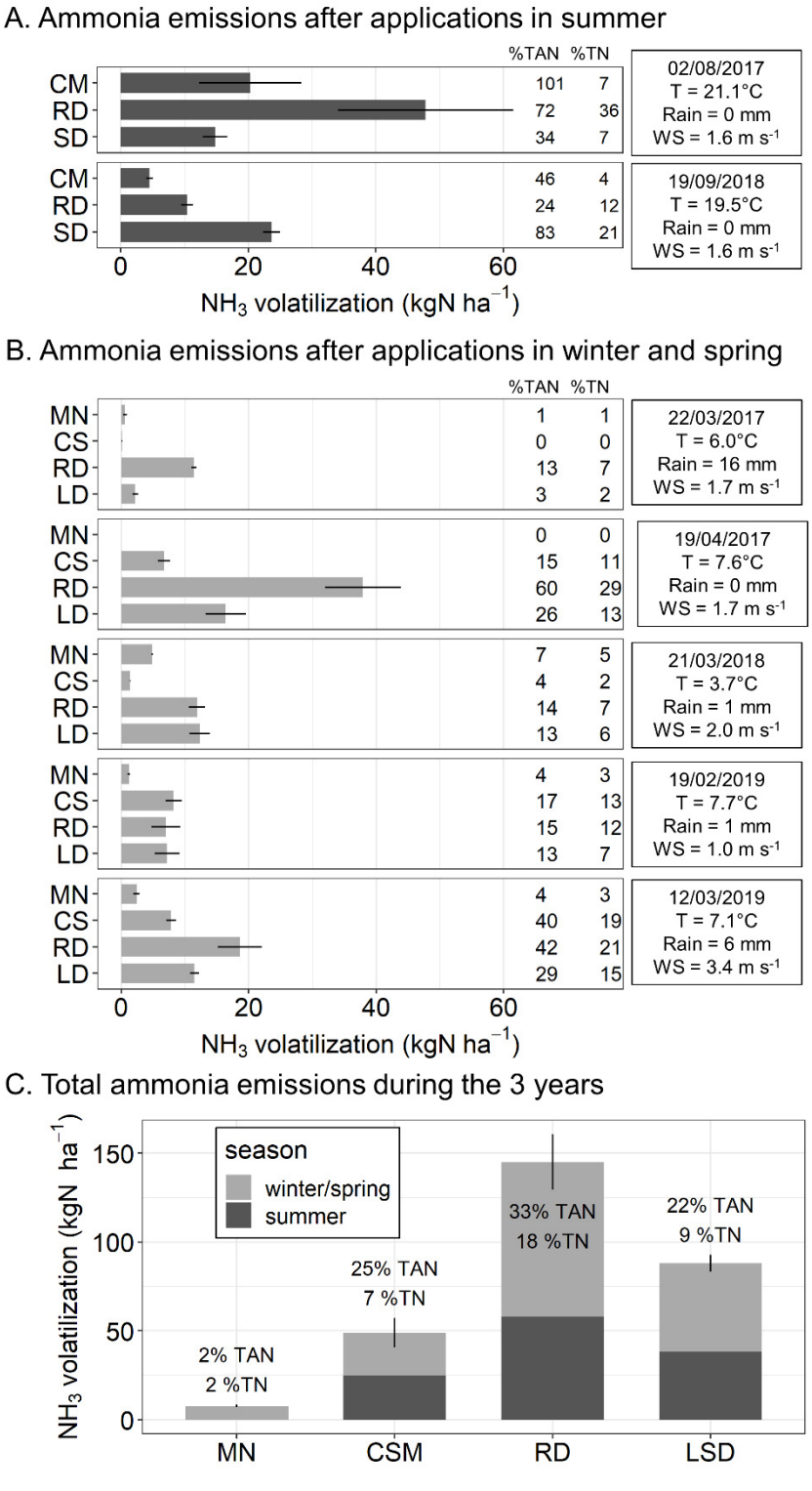

Figure 3.3: Ammonia emissions 6 days after the applications of fertilizers in the field, expressed in kgN ha^{-1}, % of applied TAN, and % of applied TN. In each subfigure, error bars show one standard error. For each dates, mean air temperature (T), rainfall, and wind speed (WS) during 6 days are indicated (A, B). A) NH_3 volatilization during winter and spring applications. MN: Mineral N. CS: Cattle slurry. RD: Raw digestate. LD: Liquid digestate B) NH_3 volatilization during summer applications. CM: Cattle manure. RD: Raw digestate. SD: Solid digestate. C) Total emissions during the three-year period, in each treatment. No statistical tests were realized to differentiate the treatments. MN: mineral N. CSM: cattle slurry and manure. RD: raw digestate. LSD: liquid and solid digestate.

In 2017, ammonia emissions factors for slurry were low, because the applied slurry had a very low DM content (< 2% FW). In 2018 and 2019, slurry and liquid digestate had similar DM contents. The liquid digestate had a higher pH that promoted high emission factors, but higher TAN content that decreased the emission factors. Therefore, emission factors were similar for both products. A negative correlation was observed between TAN content and emission factors. This could be due to a larger difficulty to reach a high emission factor when the TAN content is high, because it requires the volatilization of a high absolute amount of ammonia. The liquid digestate had similar or lower emission factors than raw digestate. We attributed it to its lower DM content that enabled a faster infiltration of digestate in the soil and therefore reduced volatilization. Considering raw digestate, the meteorological conditions were more favorable to volatilization in summer than in winter or spring, but the raw digestate was buried. Therefore, the emission factors were similar in both conditions.

The total volatilization of the different treatments during the three years depended on emission factors and doses of applied TAN. The treatments with EOMs presented higher volatilization than mineral fertilizer (Figure 3.3.C). In CSM, RD, and LSD treatments, the summer volatilization accounted for 51%, 40%, and 44% of total volatilization, respectively.

Phase separation decreased volatilization: the proportion of volatilized TAN was 33% lower in LSD than in RD treatment. It was first explained by volatilization reduction in winter, mainly due to the faster infiltration of liquid digestate in the soil compared to raw digestate. There was also less TAN applied in summer with solid digestate compared to raw digestate, which is a period where emission factors were high. Total emission factors were similar for CSM and LSD treatments. However, because of the higher TAN input, total volatilization was 1.8 times higher in LSD than in CSM treatment.

3.3.3 Agronomical performances in the different treatments

Agronomical performances provided by the different treatments were assessed through crop yields and N uptake by crops (Figure 3.4). For each year, crop N uptake (aboveground organs) were strongly correlated to grain yield (Pearson's $r^2 = 0.97$ in 2017, $r^2 = 0.99$ in 2018, $r^2 = 0.96$ in 2019) as well as to grain N export ($r^2 = 0.93$ in 2017, $r^2 = 0.99$ in 2018, $r^2 = 0.90$ in 2019) (data not shown). Because we did not assess the variability of the yield inside each plot, we could not assess if the crop yields were significantly different in the different treatments. However, we observed some tendencies. Excluding control treatment 0N, the highest yield and N uptake were always achieved in MN and LSD treatments, and the lowest yield and N uptake were always found in CSM and RD treatment (Figure 3.4.A and 3.4.B). The highest and lowest yields differed from 15 to 20%. The wheat grain quality target is 10% of protein in the grain, enabling for higher valorization and better sale. In 2017, this goal was only achieved in RD and LSD treatments (higher TAN input), but not in LSD (lower TAN input) (Figure 3.4.C). In MN treatment, lower N inputs compared to RD and LSD treatments and the lack of a third fertilization event during the upstream stage (recommended fertilization practice in France) probably prevented a

good grain quality. On the contrary in 2019, only MN treatment reached a good grain quality, because of higher winter and spring mineral N inputs than in CSM, RD, and LSD treatments.

The crop N export in the different fertilization systems was possibly influenced by the total applied N doses and the N availability in the fertilizers applied in winter and spring, and by possible effects of EOMs applied in summer. The investigation of surplus N uptake and mineral N supply enabled to differentiate these different effects. The of surplus N uptake and mineral N supply were well correlated ($r^2 = 0.84$) (Figure 3.5.A). A first conclusion is that the mineral N supply in winter explained the crop N export, and therefore that the application of EOMs in summer had no detectable effects on crop N export between February and harvest. As a second conclusion, because the mineral N supply was mostly described by the N availability of EOM, our computation of the N availability in the EOMS reflected well their properties as N fertilizers. In other terms, the N availabilities in the EOMs were correctly described by their TAN content, ammonia volatilization, and mineralization of organic N. The mineral N supply was 30% lower than the surplus N uptake. Therefore, we were able to investigate the contribution of TAN content, volatilization, and mineralization of organic N to total N availability (Figure 3.5.B). TAN content was the major driver of the fertilizing properties of the fertilizer N availability. The TAN:TN ratio in cattle slurry were highly variable. The volatilization and N immobilization during slurry decomposition decreased the total N availability in the same order of magnitude. Volatilization significantly reduced the N availability of raw digestate. For liquid digestate, the decrease of N availability due to volatilization was compensated by the organic N mineralization. Therefore, volatilization and N mineralization had similar orders of magnitude.

The NUE of the organic fertilizers were lower than the one of the synthetic fertilizers (Figure 3.5.C). The NUE of the cattle effluents were slightly higher than the NUE of the liquid digestate. The NUE of the raw digestate were lower than the one of liquid digestate and cattle slurry.

To conclude, the good N availability and high total N input with liquid digestate enabled to achieve correct crop yield and N uptake. The crop yield and N uptake were lower with cattle slurry and raw digestate because of lower doses of applied N (cattle slurry) or lower N availability (raw digestate). We did not highlight any effect of the summer application of EOMs on SMN stocks in February (data not shown). It is possible that the summer application could have influenced the early growth of rapeseed; however, our experiment design cannot conclude on that point (Supplementary Material 7).

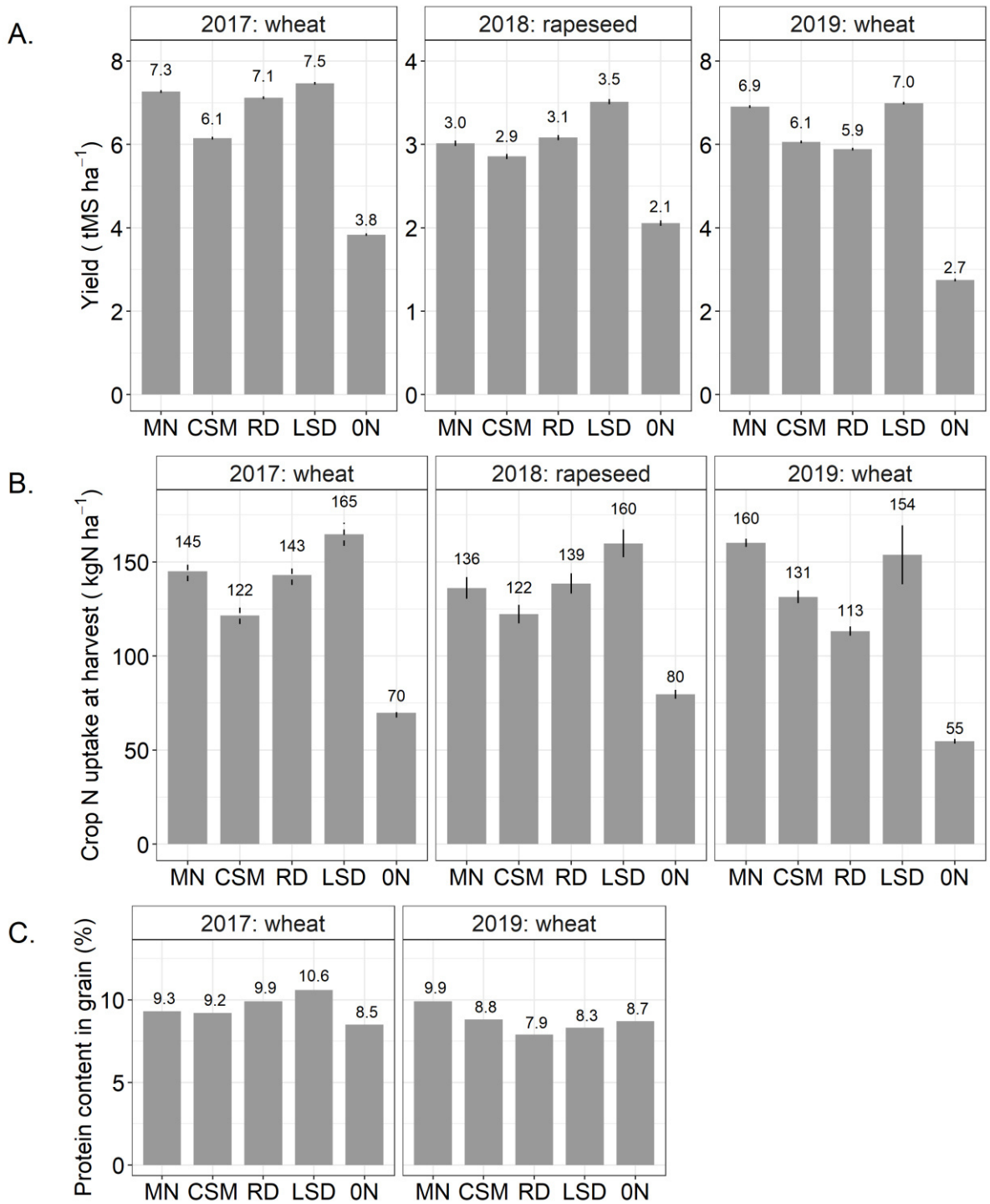

Figure 3.4: Agronomical performances during the field experiment. Error bars show measure uncertainty. A) Grain yield. B) N uptake by crop (aboveground organs), at harvest date. In 2017, uncertainty could not be computed due to no replication of N content measurement. The shown dotted error bars represent an estimated uncertainty with the same relative uncertainty in N content as for wheat in 2019. C) Protein content in wheat grain (2017 and 2019). The measure were not repeated and no uncertainty could have been estimated

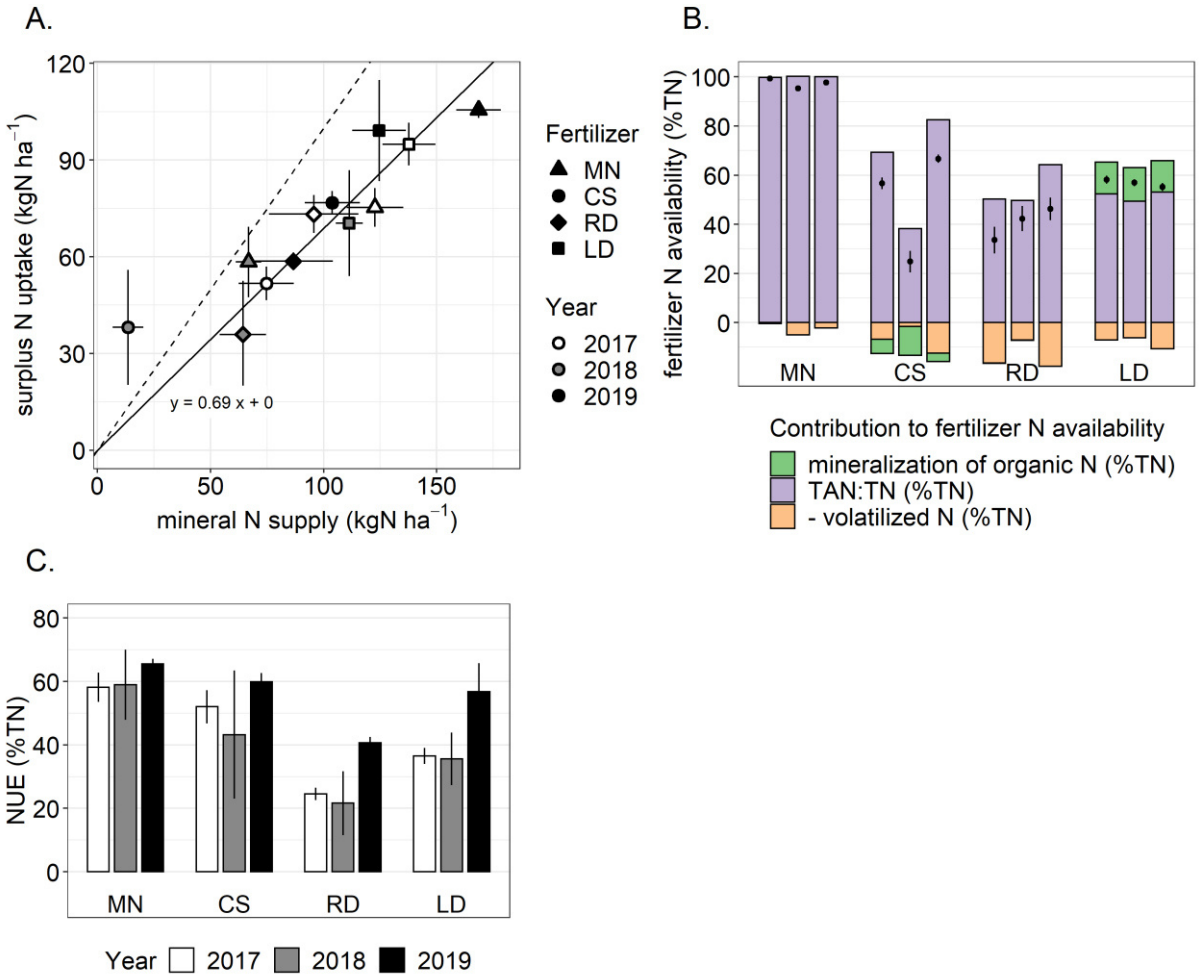

Figure 3.5: N availability of fertilizers applied in late winter and spring. MN: Mineral N fertilizer. CS: Cattle slurry. RD: Raw digestate. LD: Liquid digestate. A) Correlation between surplus N uptake and the mineral N supply that were computed between February and harvest. Solid lines represent the best regression line passing through the origin. Dotted lines are 1:1 line. Error bars represent one measure uncertainty. Pearson's r^2 was 0.84. B) Decomposition of fertilizer N availability into its three components, TAN:TN ratio, mineralization of the EOMs, and volatilization. For each treatment, the bars represent the N availability in 2017, 2018, and 2019, from left to right. C) Nitrogen Use Efficiency of the fertilizers in the different treatments.

3.3.4 Evolution of soil mineral N stocks during the 3-year experiment

In 2017, after wheat harvest and before drainage period in November, the SMN stocks were similar within the treatments, even after the application of EOMs in summer. This was interpreted by a non-excessive fertilization of wheat and the early N uptake by rapeseed crop. During winter 2017/2018, the SMN decreased a little similarly in all the treatments. At the end of the drainage period in February 2018, the SMN remained similar in all treatments (Figure 3.6). High nitrate leaching was unlikely.

In July 2018, after rapeseed harvest, the SMN stocks were relatively high (Figure 3.6). After the application of organic fertilizer and before the drainage period, the SMN stocks increased in the fertilized treatment (CSM, RD, LSD). In RD and CSM treatments, the SMN increase was consistent with mineral N input from EOMs, lower volatilization than in the previous year, and N mineralization

from organic amendments. However, the observed SMN increase in LSD treatment was unexpectedly high and the N immobilization observed in controlled conditions unlikely occurred. During the winter drainage period between December 2018 and February 2019, the N absorption by wheat was likely low and the high decrease of SMN were likely explained by nitrate leaching.

To conclude, the investigation of SMN stocks highlighted the absence of detectable effect of fertilizer application before rapeseed sowing, the leaching induced by fertilizer application before wheat sowing, and some inconsistency between the incubation of solid digestate in controlled conditions and the SMN stocks in the field.

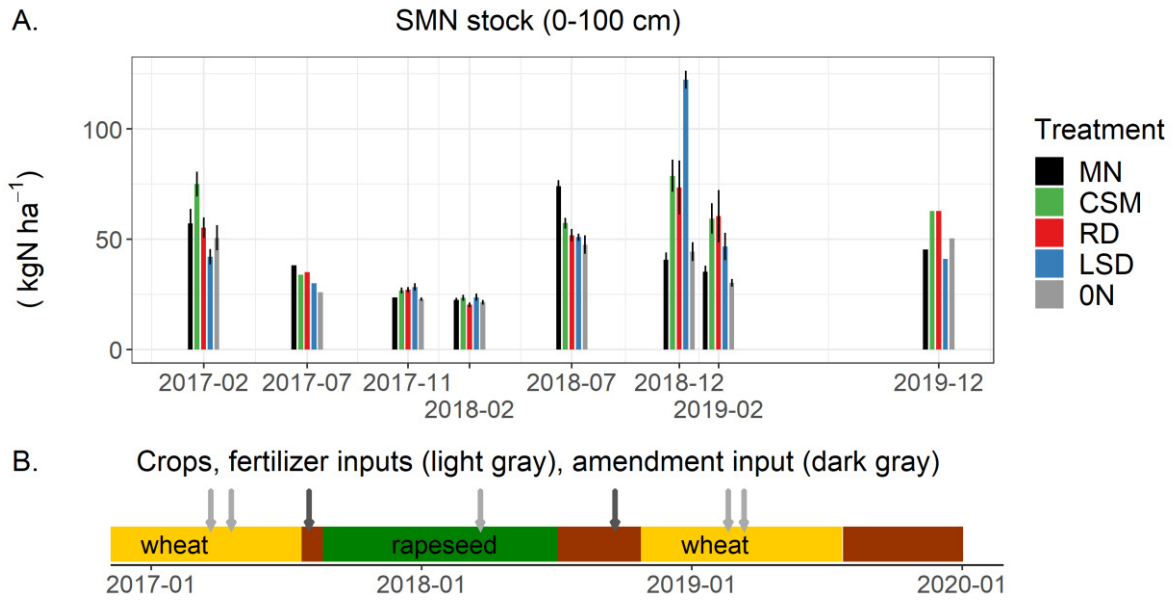

Figure 3.6: A) Soil mineral N (SMN) stocks (0-100cm), on the same x-scale as B) Crop management. Arrows indicate the fertilization dates in late winter and spring (light gray, MN, CSM, RD, and LSD treatment), and in summer (dark gray, CSM, RD, and LSD treatments).

3.3.5 N_2O emissions

In all treatments, we observed a peak of N_2O emission in spring during one month after application events (Figure 3.7). Emissions in both treatments using digestates (LSD and RD) were similar. The N_2O emissions of cattle effluents (CSM treatment) were slightly lower. The use of synthetic fertilizers induced N_2O emissions that were strongly higher in spring 2018 and similar than in other treatment, and were similar to the one of LSD and RD treatment in spring 2017 and spring 2019. In autumn, the N_2O emission were low in all treatments.

We summed the emissions during the five periods following fertilizers applications (50 days each) (Figure 3.8). The surplus N_2O emissions compared to 0N during the 250 measurement dates were 0.67% ± 0.07%, 0.11% ± 0.03%, 0.22% ± 0.03%, and 0.21% ± 0.02% of total N applied for MN, CSM, RD, and LSD treatments, respectively. They also were 0.67% ± 0.06%, 0.38% ± 0.11%, 0.41 ± 0.05%, and 0.49% ± 0.06% of mineral N applied for MN, CSM, RD, and LSD treatments, respectively. The emissions were not measured during the whole year and therefore these percentages cannot be considered as emission factors. The EOMs (CSM, RD, and LSD treatments) had similar ratios of measured N_2O emissions to mineral N input. The synthetic fertilizer (MN treatment) resulted in higher N_2O emissions for similar mineral N input. Therefore, the cumulated N_2O emissions were the highest on MN treatment (Figure 3.8). Cumulated N_2O emissions were similar on RD and LSD treatments; they had the same order of magnitude as in the MN treatment. Cumulated N_2O emissions were twice lower on CSM treatment, because of the lower dose of mineral N applied. The emissions in autumn after summer application only represented 7%, 34%, 19%, and 16% of total emissions for MN, CSM, RD, LSD treatment, respectively, although it represented 40% of measured time.

We investigated the driver of the N_2O emissions with a linear regression model. The best model (adjusted $R^2 = 0.42$) to explain the N_2O emissions was:

(equation 3.7) $\log(N_2O) = -5.379 + 0.026\ NH_4^+ + 0.154\ NO_3^- + 0.048\ SWC + 0.048\ T + 1.441\ FERTILIZED + 1.017\ SEASON$

where $\log(N_2O)$ was the logarithm of N_2O emissions expressed in $gN_2O\text{-}N\ ha^{-1}\ day^{-1}$, NH_4^+-N was the content of NH_4^+ in the topsoil ($mgNH_4^+\text{-}N\ kgDS^{-1}$) , NO_3^--N was the content of NO_3^- in the topsoil ($mgNO_3^-\text{-}N\ kgDS^{-1}$), SWC was the soil water content (%WFPS), T was the air temperature (°C), FERTILIZED was equal to 0 if the treatment was (0N) and 1 else, and SEASON was equal to 1 if the last application of fertilizer was in winter or spring, and 0 else. The model was better when using both soil NO_3^- and NH_4^+ contents instead of total mineral N content. All explanatory variables had a slope significantly different to 0 ($p < 0.05$). Date, treatment, and last fertilizer used were not selected in the model.

The model confirmed and partly explained the previous observations. The contribution of autumn emissions to cumulated emissions were low: it was first attributed to lower SWC in autumn than in spring, but also to slightly lower SMN in soil. During late winter and spring, we detected no effect of fertilizer type on EOM. The low N_2O emissions in CSM treatment was induced by low SMN content.

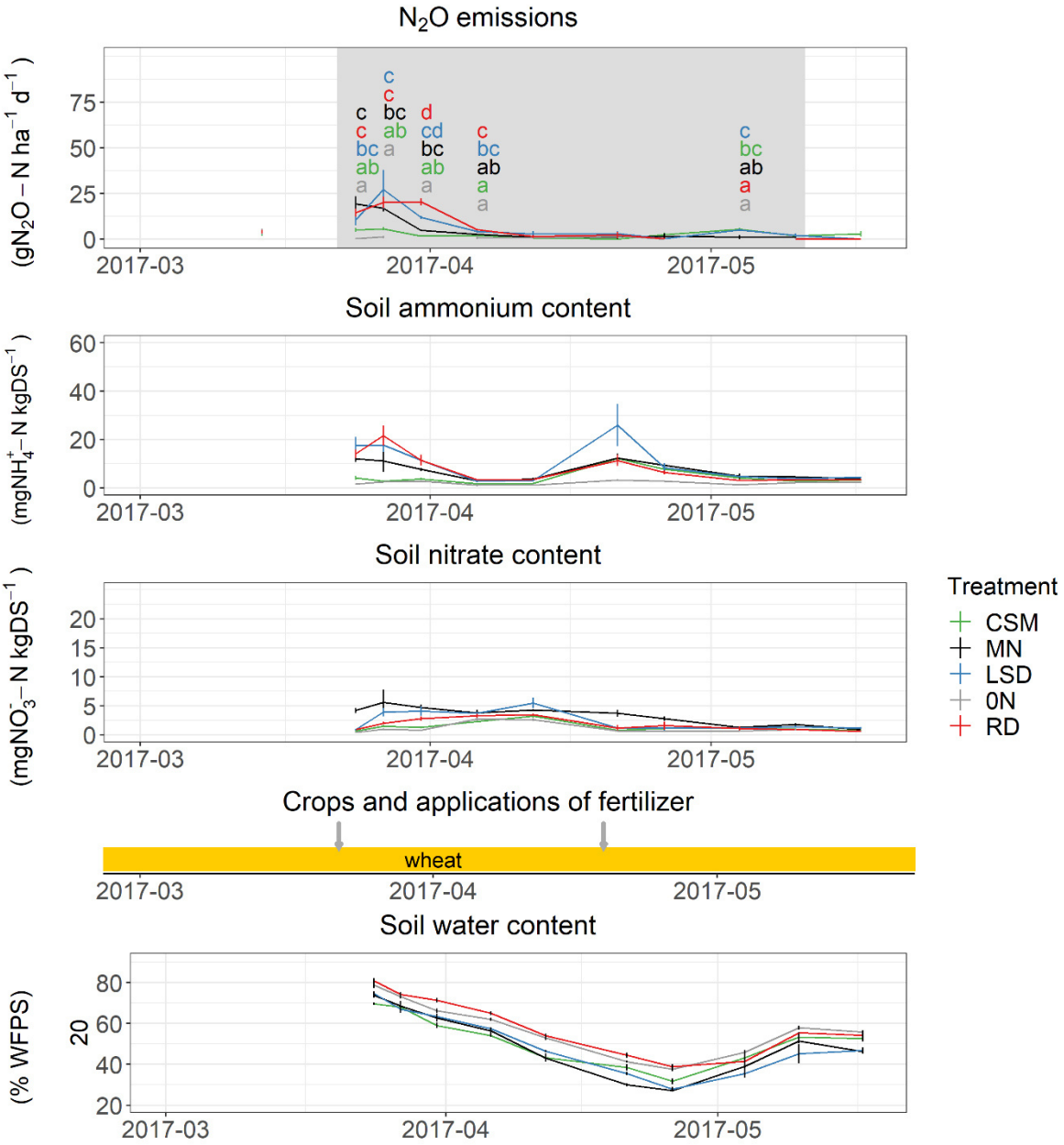

Figure 3.7.A: N_2O emissions, evolution of total mineral N and nitrate stocks in the upper soil layer (0 – 20 cm) and evolution of topsoil water content as expressed by the water filled pore space during wheat cropping season in 2017. In each subfigure, error bars show one standard error. All subfigures share the same x-axis (time): punctual N_2O emissions, topsoil mineral N content, topsoil nitrate content, crops and fertilization dates, soil water content. Only for N_2O emissions, at each date, a Kruskal-Wallis test followed by a post-hoc Dunn's test was realized to test whether the emissions were significantly different from one another. When no difference were highlighted between treatments, no letters are drawn. When treatment are shown different by Kruskal-Wallis test (p-value < 0.05), letters are drawn for each treatment and treatment sharing no letters are significantly different (Dunn's test, p-value <0.05). The period that were considered for total emissions were highlighted in gray.

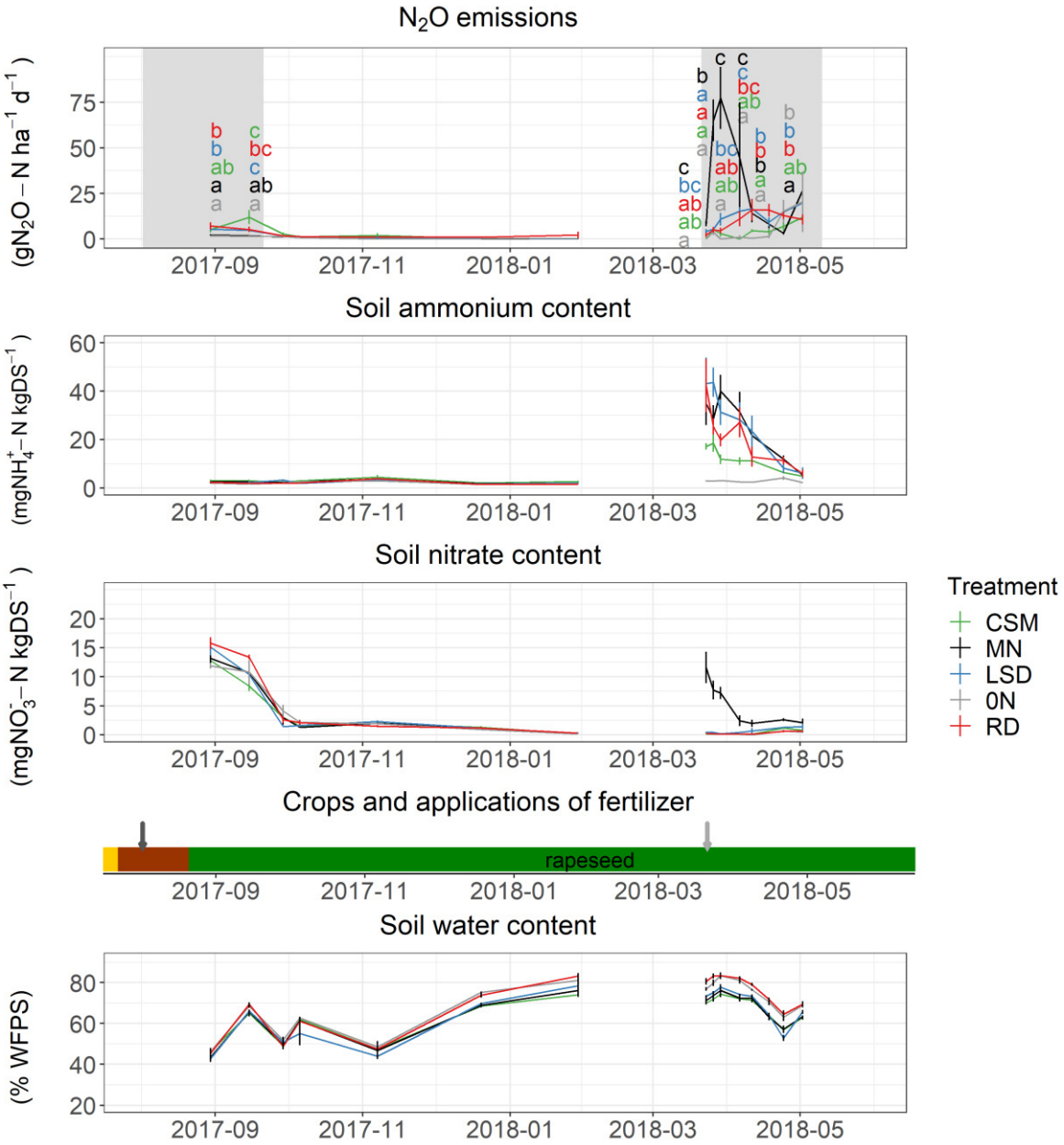

Figure 3.7.B: N_2O emissions, topsoil mineral N content and water content (rapeseed 2017-2018). See legend of Figure 3.7.A for more details.

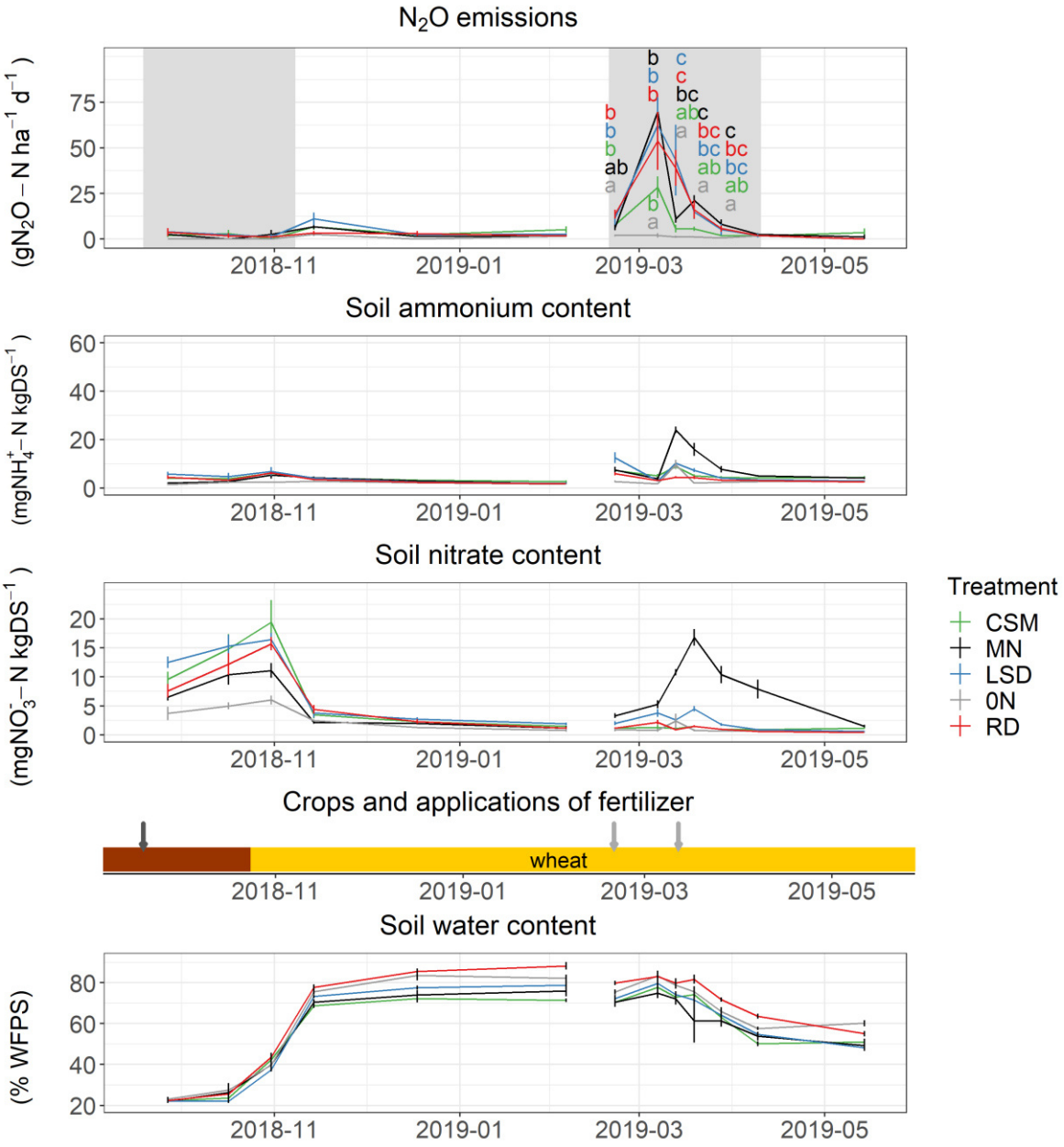

Figure 3.7.C: N_2O emissions, topsoil mineral N content and water contents (wheat 2018-2019). See legend of Figure 3.7.A for more details.

3.3.1 Mineral N balance

The mineral N balance in the soil showed a correct accuracy at the scale of the three cropping seasons. The difference between outputs and inputs of soil mineral N were -89 kgN ha^{-1} (13% of total inputs), 50 kgN ha^{-1} (10 %), -5 kgN ha^{-1} (1 %) and 105 kgN ha^{-1} (15 %) in MN, CSM, RD, and LSD treatments, respectively (Figure 3.9). These imbalance were low relatively to uncertainties of measurement (N uptake, volatilization, leaching, mineralization from EOMs), and neglected processes (e.g., inhomogeneous crop residue mineralization and N_2 and N_2O losses). The mineral N fluxes in the soil were the highest with digestates (RD and LSD treatments), compared to CSM treatment. It induced higher N losses as well as higher agronomical performances in LSD treatments. The N losses (leaching and volatilization) were equivalent to 4 % of crop aerial N uptake in the MN treatment, which was lower than in CSM (18 %), RD (41 %), and LSD (34 %) treatments.

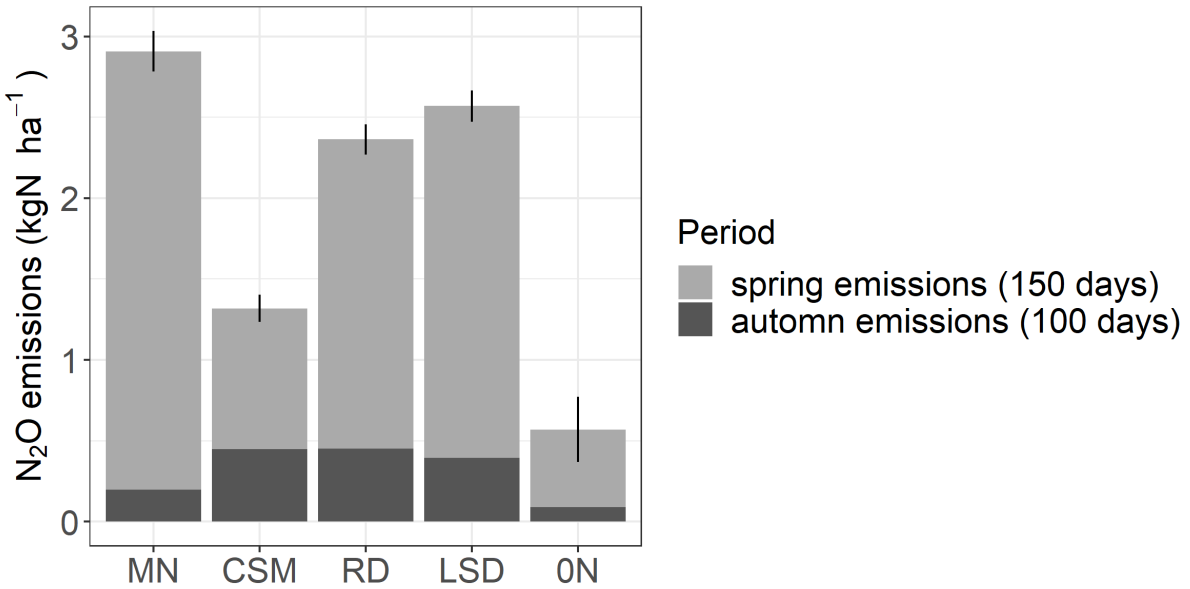

Figure 3.8: Sum of the estimated N_2O emissions. Fluxes represent 250 days of measurement for three years and did not represent the real absolute value of N_2O emissions. Error bars show one standard error.

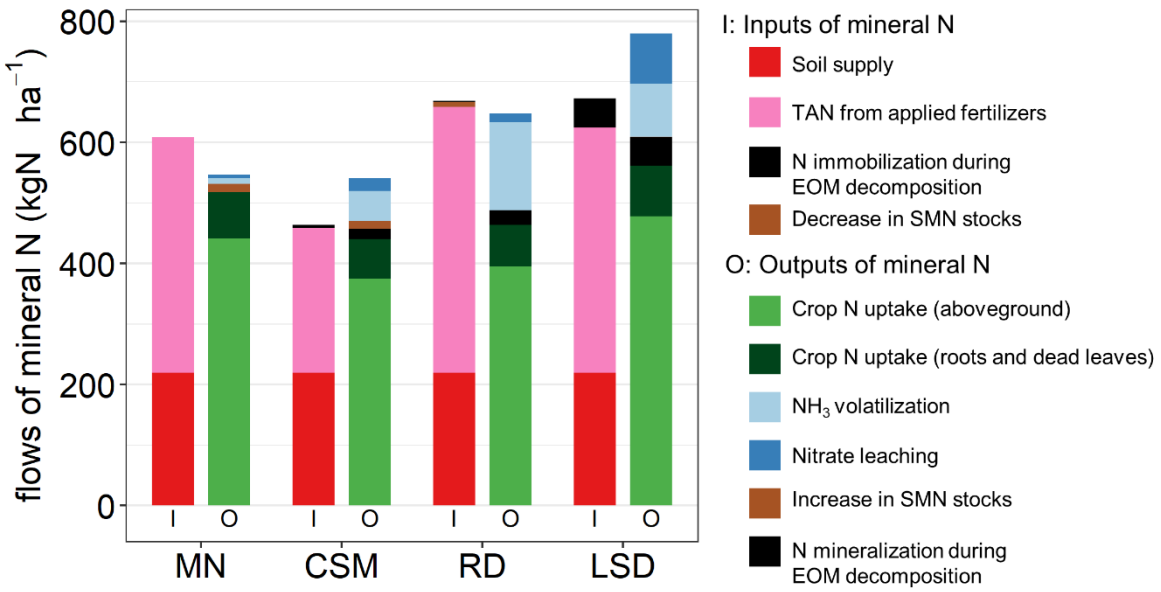

Figure 3.9: Mineral N balance in the field at the scale of the crop rotation during three cropping seasons. (I): Inputs, i.e. processes that supply mineral N to the soil. (O): Outputs, i.e. possible behaviours of soil mineral N.

3.3.2 Amendment properties of the EOMs

There was no surplus of total N in MN treatment (Table 3.4), indicating no supplementary soil organic nitrogen (SON) storage. In the treatment using organic fertilizers, there were more inputs than outputs of total N (Table 3.4). This balance was assumed to represent the input of organic N in soil during the rotation. This high input represented c.a. 10% of initial soil organic N stock (3.7 t ha^{-1} on average), and reflected the potential contribution to soil organic matter from organic fertilization, so-called amendment properties of the EOM. Note that this corresponded to an input and not to a net storage, because the additional mineralization of SOM caused by these inputs was not considered.

Simultaneously, we estimated the inputs of stable organic C in soil from the EOM based on their VS content and I_{ROC} value (Table 3.4). The input of stable C was lower in RD treatment than in CSM and LSD treatment, due to lower C inputs (no solid EOM). Manure and solid digestates represented 81% and 71% of total stored C for CSM and LSD treatment, respectively. It highlighted their higher amendment effect than liquid EOMs, even if liquid digestates stored more C than slurry.

Table 3.4: Total N balance in the field at the scale of the crop rotation during the three cropping seasons. Inputs include total N applied by fertilizers. Outputs include crop export, leaching, and volatilization. Inputs of C from EOMs come from application doses and analysis. The I_{ROC} were used to estimate the C incorporated to SOM.

Flux of total N and C		Treatment			
		MN	CSM	RD	LSD
Total N fluxes (kgN ha^{-1})	Input of organic N	0	423	423	537
	Input of total N	389	662	863	942
	Output of total N	380	373	491	558
	Balance of total N: estimation of input of N inside SOM	9	290	372	384
C fluxes (tC ha^{-1})	Input of C	0	7389	4064	8455
	Input of stable C: estimation of input of C inside SOM	0	4770	2425	5205

3.4 Discussion

3.4.1 Digestates have higher fertilizer properties and similar amendment properties than undigested animal effluents

Guilayn et al. (2019a) proposed a typology of digestates. Based on chemical characteristics, raw digestate fitted type 2 characteristics (Sewage sludge, biowastes, food agro industry residues, pig slurry). Our results confirmed that the large proportions of non-agricultural inputs like sewage sludge seemed to import high quantities of nutrients and to drive the digestate chemical characteristics more than slurry and manure incorporated at relatively low input doses. Therefore, when looking at the properties of the digestates, both effect of the digestion and of the import of nutrients have to be considered. The liquid and solid phases of digestate fitted well the expected typology associated with low-performance phase separation. According to the analysis of digestates, separation phase removed only 27% of DM content from the raw digestate. Despite the slightly lower DM content, the chemical characteristics of liquid digestate were similar to raw digestate. This is common for low efficiency separation phase (Guilayn et al., 2019b).

With the exception of raw digestates, I_{ROC} value was consistent with the mineralized C during the incubations. The stable fraction of organic C of digestate were consistent with the one found by several authors (Alburquerque et al., 2012; Askri et al., 2016; Cavalli et al., 2017; de la Fuente et al., 2013; Nielsen et al., 2020). As liquid and solid phases come from raw digestate, we expected a degradation rate of raw digestate between solid and liquid digestates, as found by Askri et al. (2016). It was not the

case in our experiment, as well as in the study from Cavalli et al. (2017). Therefore, it was difficult to conclude if this was explained by a difference induced by the different sampling dates between raw and liquid digestate, or another explanation such as a change in liquid phase properties during storage. The application of slurry and liquid digestate to soil during the incubation in controlled conditions triggered a quick and important C mineralization, which was already reported in literature (Alburquerque et al., 2012; de la Fuente et al., 2013; Iocoli et al., 2019; Risberg et al., 2017). Liquid digestate had a higher fraction of stable organic C than solid digestate, resulting in a higher I_{ROC} value for liquid digestate than raw and solid digestates. It suggests that the stable C from raw digestate remained preferentially in the liquid phase. In the literature, the liquid phase of different digestates had a higher (Askri et al., 2016), similar (Cavalli et al., 2017; Nielsen et al., 2020) or lower (de la Fuente et al., 2013) organic matter stability than the corresponding solid phase.

In laboratory conditions, the mineralization of N varied during 50 days and stabilized after. This dynamics was similar to the one observed by Cavalli et al. (2017). We observed N mineralization after liquid digestate application but no N mineralization after raw digestate application to soil in controlled conditions. Observations varied in the literature. N mineralization (Tampio et al., 2016), no N mineralization (Cavalli et al., 2017), or N immobilization (de la Fuente et al., 2013) were observed during the incubations of liquid or raw digestates in the soil. Alburquerque et al. (2012) suggested that the less digestates caused C mineralization, the more they induced N-mineralization. Our digestate had a similar C-mineralization as the more stable digestate in the latter study, which is consistent with the observed N mineralization, on the contrary to other digestates that could exhibit N immobilization. Consistently to our results, Cavalli et al. (2017) observed immobilization following solid digestate application to soil, However, contrary to the present study, it was followed by mineralization. De la Fuente et al. (2013) also found N immobilization associated to solid digestate incubation. Chiyoka et al. (2014) showed N immobilization and N mineralization during solid digestate and solid manure incubation, respectively. In a more synthetic approach, Levavasseur et al. (2021a) highlighted the variability of the N behaviour following each type of EOMs. They highlighted a neutral N mineralization for digestates and bovine effluents on average. Our study was consistent with this result.

We confirmed the different expected effects for solid and liquid EOMs. Here, solid digestates and manure were poor fertilizers, as their TAN content was low and because they did not induce N mineralization. However, they brought high amount of recalcitrant C in the soil. The stability of organic C in manure made it a slightly better amendment than solid digestate. On the contrary, the input of organic C with liquid EOMs was low. The fertilizing potential of slurry and liquid digestates were primarily explained by their TAN content. However, N mineralization increased the fertilizer potential of liquid digestate compared to slurry and raw digestate.

3.4.2 Assessing the N balance in a field experiment: limits and strength

In this study, we compared different fertilization systems using full-scale machinery. We used a single large plot per treatment for this experiment, rather than classical bloc-designed experiment with smaller plots. We assumed that the main drivers in the different crop performances were the N supply by the fertilizer. The good correlation between the surplus crop N uptake and the N availability from the fertilizers partly verified this assumption.

Variation of EOM characteristics explained that the N applied differed from the targeted amounts. We also avoided applying very high dose of liquid EOM that would not have been consistent with classical management. Consequently, the applied N doses differed among the treatments. N losses and crop N uptake therefore have to be interpreted in two ways. First, we compared the N fluxes relatively to the applied N, to compare the quality of the different EOMs. Then, it was also interesting to interpret the total N fluxes, to consider the fact that it is easier to apply high total N dose with digestates than cattle effluents.

Despite these limits, thanks to numerous measurement of N fluxes, we were able to propose a consistent soil mineral N balance in the field at the scale of the rotation. This enables to better understand the effects of fertilization with digestates. In our study, the output of mineral N seemed underestimated. One possible explanation is the immobilization of more N due to crop residue degradation, or an underestimated nitrate leaching. On the contrary, the inputs of mineral N were lower than outputs in LSD and CSM treatments. Probable explanations could be the underestimation of manure and cattle slurry mineralization, or the overestimation of nitrate leaching.

3.4.3 The use of exogenous organic matters influences N losses in the field

Due to high N inputs, the use of digestates induced N losses (N_2O, NH_3, nitrate). Möller (2009) also highlighted the importance to reduce N losses to improve N efficiency of the different fertilization systems using digestates thanks to N cycle at the field scale.

Ammonia emission is driven by temperature, wind speed, rainfall, application method, pH, TAN content (Hafner et al., 2019), but also crop height (Thorman et al., 2008) and infiltration in soil (Pedersen et al., 2021). Our findings were consistent with this knowledge. Depending on meteorological conditions and variation in digestates characteristics, we measured highly variable ammonia emission factors ranging from 2% to 60% of TAN for liquid and raw digestates. This was consistent with previous measurement (Nicholson et al., 2017; Räbiger et al., 2020; Riva et al., 2016). The emissions factors for cattle manure and cattle slurry were consistent with the value reported in the ALFAM2 database (Hafner et al., 2018). For solid digestate, it was similar to that of cattle manure. Considering the high volatilization rate of solid digestate and despite their lower TAN content, we argue that these emissions should not be neglected, as it is sometimes done (Amon et al., 2006; Holly et al., 2017). To our knowledge, nobody else already published ammonia emission factors for solid digestate. The phase separation of digestate

decreased the ammonia emission factors and the total ammonia emissions, during the three years. The lower ammonia emissions from liquid digestate compared to raw digestate was attributed to higher infiltration rate. Such reduction of ammonia emission with phase separation was also found, but for undigested slurry (Amon et al., 2006; Nyord et al., 2012). The use of liquid and solid digestates induced similar ammonia emission factors than the use of cattle effluents. Digestates had sometimes lower and sometimes higher ammonia emission factors than undigested effluent in the literature (Möller, 2015). Due to the higher TAN applied, the use of digestate resulted in higher total ammonia losses. Digestate injection could be an efficient way to reduce volatilization and improve digestate N fertilizer efficiency when applied on grasslands, or before sowing of annual crops (Herr et al., 2019; Maris et al., 2021; Webb et al., 2010). However, for rapeseed and wheat, the digestate was mainly applied on the crop and other solution could be proposed to reduce volatilization, such as acidification (de França et al., 2021).

N_2O emissions peaked around 75 gN_2O-N ha^{-1} day^{-1} in late winter and spring. This was consistent with measures reported in the literature, even if N_2O fluxes were sometimes three to four times higher for digestates (Möller and Stinner, 2009; Nicholson et al., 2017; Verdi et al., 2019) or undigested effluents (Bell et al., 2015; Williams et al., 1999). We measured less emission in summer and autumn, often below 10 gN_2O-N ha^{-1} day^{-1}, which was attributed to soil dryness. This was consistent with the findings of Pezzolla et al. (2012), who used digestates, and Parnaudeau et al. (2009), who used other EOMs. On the contrary, other studies measured higher emissions in autumn or summer compared to spring with animal effluents (Bell et al., 2015, 2016), which were explained by relative high rainfall and soil moisture. Using a linear model, we highlighted the effect of soil moisture, soil mineral N content, and temperature on N_2O emissions, which were known as important variables explaining N_2O emissions (Butterbach-Bahl et al., 2013). We did not highlight any additional effect of the nature of the fertilizer. The type of digestate was also found as a minor factor to explain N_2O emissions (Häfner et al., 2021). In our field conditions, the effect of SMN could have masked any quality effect of the different EOMs that have been reported in other studies (Charles et al., 2017). We did not find any effect of phase separation. On the contrary, Askri et al. (2016) and Möller (2015) observed that the phase separation of digestate decreased N_2O emissions. One impact of fertilizer type was the form of mineral N applied, i.e. ammonium or nitrate. This likely influenced N_2O emissions if the conditions favored N_2O production through nitrification or denitrification. N_2O emission was lower in CSM treatment than in other treatment, mainly due to lower N inputs (similar N_2O emissions to applied TAN ratio to RD and LSD treatments). At the scale of the crop rotation, N_2O emissions were similar in LSD, RD, and MN treatments. A higher amount of N_2O from digestate compared to mineral fertilizers was found by Buchen-Tschiskale et al. (2020) and Köster et al. (2011). Other publications described a lower N_2O emission with digestate than undigested slurry (Köster et al., 2015; Möller, 2015; Nkoa, 2014). The strong emission peak in spring 2018 in MN treatment occurred in a period with higher soil nitrate content than in other treatments. The soil moisture was very high, a condition where N_2O emissions are usually

mainly attributed to denitrification than nitrification (Senbayram et al., 2009), and therefore the high soil nitrate content could have induced strong N_2O emissions. In spring 2019, N_2O emissions in MN treatment were not higher than other treatments, despite a higher nitrate content, similarly to 2018. However, the conditions may have been less favourable for denitrification in 2019 than in 2018.

We found that applying EOMs in autumn before wheat sowing both promoted nitrate leaching and did not improve wheat yield, in accordance to Sieling et al. (1997). During the crop rotation, estimated leaching represented 1%, 3%, 2%, and 8 % of total N inputs for MN, CSM, RD, and LSD treatments, respectively. This was lower than measure by Nicholson et al. (2017), who estimated a leaching representing 15% of total N for slurry or digestate applications. It was consistent with the modelling from Räbiger et al. (2020), who estimated a leaching of 5 to 18% of total N input, with more leaching using digestate than mineral fertilizers.

3.4.4 Digestates are able to substitute chemical fertilizers to grow crops in field conditions

The application of EOMs had to occur when the soil was dry enough to prevent soil compaction due to heavy spreading engine, adding a constraint on fertilization date. Doses could be less easily fractionated, and no third fertilization occurred on wheat (but in 2019 for MN and CSM), as it is classically recommended to improve protein content in grain. The use of combined digestate and mineral fertilizers at different dates could solve this problem, as already proposed (de França et al., 2021; Odlare et al., 2014). We observed a slightly higher NUE for cattle slurry than for liquid digestate. It was mostly attributed to its slightly higher TAN:TN ratio. However, because of their respective TN content, it should be easier to apply the right dose of N with digestates than with slurry, and therefore to achieve the targeted yields. These results are similar to the study from Baral et al. (2017) on barley. The NUE of raw digestate was slightly lower, due to higher volatilization.

In this study, we estimated the NUE of cattle slurry and digestate. The repetition of EOM application in spring and in summer possibly affects NUE. Estimated NUE ranged from 22% to 41% of N applied for raw digestate, and 37% to 57% for liquid digestate. The NUE of the raw digestate was consistent with the NUE of raw digestates measured on barley by Thomas et al. (2017) and Baral et al. (2017) (19% and 37%). We did not find NUE digestates on rapeseed in the literature.

We here explained crop yield only with fertilizer inputs, without considering amendment application in autumn. It is possible that the application in summer before rapeseed could have improved early rapeseed growth in RD treatment, and, to a lesser degree, in LSD and CSM treatments, but this effect was not highlighted. The rapeseed N uptake in CSM treatment was higher than expected, which could also be an effect of manure application in late summer before sowing.

The TAN content of digestates is often used as a proxy of their fertilizer efficiency (de França et al., 2021; Iocoli et al., 2019; Tampio et al., 2016). However, it is a simplified understanding of the fertilizer potential of digestate which could also be influenced by ammonia emissions and the mineralization of digestate organic nitrogen (Möller and Müller, 2012). Ammonia volatilization could decrease its fertilizer efficiency (Frost et al., 1990; Quakernack et al., 2012), whereas the mineralization of organic N in digestate could increase N supply for crops (Abubaker et al., 2012; Cavalli et al., 2017) and could make digestate efficient EOMs for N fertilization in the short-term. The potential of digestate to fertilize crops was often investigated without consideration of volatilization or mineralization. Here, we highlighted the importance of these two processes to describe digestate fertilizer potential. TAN content was the main factor driving fertilizer efficiency, followed by volatilization and then mineralization/immobilization. A lower volatilization and a higher mineralization explained the better agronomical results in LSD treatment compared to RD treatment. It also implied that reducing volatilization could have both environmental and agronomical benefits.

We found a good correlation between the estimated mineral N supply that included data from the incubations, and N uptake. Cavalli et al. (2017) also showed a good correlation of N availability calculated from incubation or field experiment. The evolution of mineral N stocks in soils during autumn and winter was consistent with incubation results of raw digestate and manure, which highlighted limited mineralization of organic N. However, the N immobilization during solid digestate incubation was not observed in the field with both high N uptake in late winter by rapeseed, and high increase of SMN stocks during autumn and early wheat growth. This result was unexpected, because of the use of the same soil, and the use of fresh digestate in the incubation. SMN were high, and did not inhibit the decomposition of solid digestate. One explanation could be that during autumn, the soil is less moist than during incubation, and therefore would have limited the decomposition.

The use of EOMs instead of mineral fertilizers induced higher total N input in the field. The main results was the probable SOM storage. However, such effect should be studied at a longer-term than our experiment (Bhogal et al., 2018).

3.5 Conclusion

We presented here the results of a three years field experiment using different fertilization strategies with cattle effluent, digestates, or synthetic N fertilizers. The main N flows in the field were investigated, and we were able to estimate a consistent soil mineral N balance at the scale of the crop rotation, highlighting a good understanding of mineral N fluxes at the field scale. The use of digestates induced similar N_2O emission factors as cattle effluents. Concerning the liquid EOMs, slurry and liquid digestate had similar NH_3 emission factors. The NH_3 emission factor of raw digestates was higher. The NH_3 volatilization after application of solid EOMs were significant at the scale of the three years. The liquid EOMs were efficient N fertilizer. Their NUE were well explained by TAN content, ammonia emissions,

and then mineralization of EOMs. Liquid digestate and slurry had similar NUE. The NUE of raw digestate was lower, due to higher volatilization. We did not find any effect of EOM applied in summer on rapeseed or wheat yield. The application of EOM in summer before sowing wheat induced nitrate leaching. However, the use of solid EOMs likely provided high input of organic matter in the soil. In practice, because of higher TAN contents, the use of digestates induced higher input of mineral N in the soil. Therefore, we were able to reach similar yields using digestate compared to mineral fertilizers, as well as to bring organic matter in the soil. In the same time, the N_2O emissions were similar and the ammonia emissions were higher when we used digestates instead of mineral fertilizers. Phase separation decreased ammonia volatilization, and improved agronomical performance of digestates. The use of slurry as N fertilizer did not enable to achieve similar yields. AD transforms the organic fertilizers, but also imports nutrient via external co-substrates. Therefore, the real impact of the introduction of AD in a dairy farm should be realized at the farm scale, to consider the change of treatment, quality, and quantity of the EOMs.

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Chapitre 4

Conséquences de la méthanisation des effluents d'élevage sur les flux d'azote lors d'une rotation culturale de 3 ans : évaluation du modèle sol-plante STICS

Lors du Chapitre 3, nous avons étudié, dans un essai au champ original, les effets de l'épandage des digestats au champ sur le cycle de l'azote. Ce type d'étude participe à l'acquisition de références sur l'utilisation agronomique des digestats et à la meilleure compréhension de leurs effets, en étudiant tous les impacts associés liés aux flux d'azote. Néanmoins, il s'agit d'un cas d'étude, dont les résultats ne peuvent être généralisés sans précaution. La modélisation des systèmes sol-plante est un outil qui peut être utilisé pour évaluer des pratiques agricoles, après une étape nécessaire de validation de ses capacités de prédiction. Ce Chapitre 4 a pour but de valider la capacité du modèle de culture STICS à prédire les résultats agronomiques et les impacts environnementaux mesurés sur l'essai présenté au Chapitre 3.

Ce chapitre évalue, entre autre, le module de volatilisation d'ammoniac des PRO de STICS. Ce travail a permis de d'isoler et de faire remonter à l'équipe de développement de STICS un bug dans ce module. Au moment de la soumission du manuscrit de thèse, ce bug venait d'être validé et était en cours de correction : la volatilisation comptée comme journalière serait en fait horaire. La performance de STICS à prédire la volatilisation d'ammoniac des PRO devra donc être mis en regard de ce fait. Néanmoins, la démarche de la thèse reste valide, et permet même de souligner l'importance de la validation des modèles.

Le Chapitre 4 est écrit sous la forme d'une publication scientifique, qui a vocation à être soumise dans une revue scientifique après adaptation. Le cas échéant, la version corrigée de STICS sera évaluée dans la publication.

Consequences of anaerobic digestion of cattle effluents on N fluxes in a 3-year crop rotation: evaluation of the STICS soil-crop model.

Victor Moinard[1], Florent Levavasseur[1], Antoine Savoie[2], Jean-Marie Paillat[3,4], Sabine Houot[1]

1: UMR Ecosys, Université Paris Saclay, INRAE, AgroParisTech, Thiverval-Grignon, France

2: UE PAO, INRAE, Nouzilly, France

3: UPR Recyclage et risques, CIRAD, Montpellier, France

4: UPR ADI-Suds, ISTOM, Angers, France

Abstract

The short-term and long-term impacts of anaerobic digestate application in the field as organic fertilizer need to be further characterized. Soil-crop models are an interesting tool to evaluate cropping practices. We tested the capacity of the STICS soil-crop model to predict crop growth as well as soil mineral N content and ammonia volatilization during a three-year arable crop rotation (France) with five fertilization systems: no fertilization, chemical N fertilizers, undigested cattle effluents (slurry and farmyard manure), raw digestate, or solid and liquid digestates. The C and N mineralization from organic fertilizers were parametrized from laboratory incubation of soil – organic fertilizer mixtures under controlled conditions. Two models predicting ammonia volatilization after organic fertilizer applications were compared: STICS, and ALFAM2 (for slurry, liquid and raw digestates) associated with emission factors (for manure, solid digestates). The ALFAM2 model coupled with emission factors performed better (rRMSE = 70%) than the STICS volatilization submodule (rRMSE = 109%). STICS predicted crop yield and N content at harvest and soil mineral N with similar accuracy than for other systems without digestates (e.g., rRMSE = 83%, 15%, and 25% for topsoil mineral N content, grain yield, and N uptake, respectively). STICS was sensitive to the ammoniacal N content of the organic fertilizers. The simulations confirmed and completed the results from the observations: the use of digestates instead of cattle effluents increased total N input, leading to higher crop exports, higher N losses, and similar soil organic N storage. STICS can be used to simulate other scenarios using digestate as fertilizers.

Keywords

Anaerobic digestion; Exogenous Organic Matters; soil-crop model; STICS; ALFAM2; ammonia.

4.1 Introduction

Exogenous organic matters (EOMs) are organic wastes or by-products (e.g., animal effluents) issued from human activities that are applied on cropped soils. Some EOMs such as animal slurries can efficiently substitute synthetic N fertilizers, because of their input of ammoniacal N readily available for plant at a very short-term and because of the possible release of mineral N during EOM degradation (Bhogal et al., 2016; Gutser et al., 2005). They are used as organic fertilizers. Other EOMs (e.g. farmyard manures) have higher organic matter contents. In the long term, the repeated applications of such EOMs on soils promote soil organic matter (SOM) storage, which is often associated with an increase in physical soil fertility and nutrients release (Bhogal et al., 2018; Diacono and Montemurro, 2010; Edmeades, 2003; Haynes and Naidu, 1998). They are used as soil amendments. On the other side, the use of EOMs is subjected to environmental risks, such as ammonia (NH_3) volatilization (Minoli et al., 2015; Sommer and Hutchings, 2001), N_2O emissions (Charles et al., 2017; Zhou et al., 2017) or nitrate leaching (Basso and Ritchie, 2005).

Anaerobic digestion of livestock effluents and biowastes has been promoted in many places in the world including Europe (Scarlat et al., 2018), as a way to produce biogas, a renewable source of energy, and as a way to recycle the nutrients from unvalued wastes. The introduction of anaerobic digestion in a farm induces the substitution of animal effluents by the residual digested organic matters, named digestates, which are seen as valuable EOMs (Case et al., 2017). Digestates are increasingly being used in the field (Stürmer et al., 2020). They have different properties than the undigested effluents and are thus associated with different advantages or risks.

During anaerobic digestion, the organic matter is degraded and a fraction of the initial organic C is exported through biogas. Therefore, digestates have a lower dry matter (DM) and volatile solid (VS) contents, a lower C:N ratio and a higher total ammoniac N (TAN) content compared to undigested effluent (Möller and Müller, 2012). Digestates have their organic matter more recalcitrant than undigested effluents (Möller, 2015). Animal effluents are often co-digested with external wastes. Depending on the characteristics of the digested effluents and co-substrates, digestates can have very diverse properties and nutrient contents (Beggio et al., 2019; Guilayn et al., 2019a; Stürmer et al., 2020). Digestates can also be post-treated, which influence its characteristics. In particular, digestate can be treated with a phase separation. The resulting liquid phase, named “liquid digestate”, has good fertilizer properties (e.g., high TAN content). The solid phase, named “solid digestate”, has good amendment properties (high VS contents) (Guilayn et al., 2019b). Digestates (raw or liquid) are usually efficient N fertilizers (Barłóg et al., 2019; Cavalli et al., 2016; Ferdous et al., 2020), partly due to high TAN content. The mineral N release after digestate decomposition in the soil can increase the fertilizing properties of digestates, while ammonia volatilization may decrease fertilizing properties (Möller and Müller, 2012). Indeed, because of the high TAN and high pH, raw or liquid digestates are sensitive to ammonia

volatilization (Nkoa, 2014; Räbiger et al., 2020; Riva et al., 2016). The digestates are often more sensitive to ammonia volatilization compared to undigested effluents (Nicholson et al., 2017; Nyord et al., 2012), even if the contrary was also found due to a faster infiltration into the soil (Möller, 2015; Neerackal et al., 2015). Field N_2O emissions from digestate could be lower than the one from undigested effluent, even if this can vary from one study to another (Möller, 2015). Apart from environmental conditions, the origin of the digestate could be a second-order factor to explain N_2O emissions (Häfner et al., 2021).

A few short-term studies argued that digestate could improve soil physical fertility compared to unamended soils (Badagliacca et al., 2020; Möller, 2015). However, few studies with mid- to long-term digestate application (e.g. five years or more) are reported. In a 8-year field experiment, Odlare et al. (2014) found a similar positive effects of digestate on SOM compared to other EOMs. Zicker et al. (2020) described a field experiment where digestate or slurry were applied in the field during 6 years. They measured a similar increase in SOM content in both treatments. Digestate led to higher risks of P losses, and a slightly lower soil pH compared to slurry. Wentzel et al. (2015) investigated farms using slurry or digestate for more than 15 years, and found no difference in soil quality when they were fertilized with one EOM or the other. Considering the numerous combinations of types of digestates, agricultural practices associated to their use, and environmental conditions of fertilized fields, the long-term amendment efficiency of digestates (potential C storage in soil after repeated applications of digestate) has not been well characterized and we cannot conclude on the better amendment properties of digestates compared to undigested effluents.

Recently, a 3-year field experiment was designed to explore the integrated effect of fertilizing a crop succession with digestates instead of untreated cattle effluent on the N cycle in the field (Chapter 3). The study showed that the use of digestates was associated to higher N applications in the field, inducing higher yields together with higher N losses (NH_3, N_2O) compared to undigested cattle effluents. Such study was limited to describe some N fluxes that are difficult to assess (N_2O emissions, nitrate leaching), and did not allow to compare the amendment properties of EOMs due to the relatively short-term scale. The use of modelling can be useful to generalize and deepen results obtained from former field experiments using digestates as fertilizers. The use of modelling can help to estimate variables that are difficult to measure: for example, Räbiger et al. (2020) used a model to estimate nitrate leaching occurring in fields fertilized with digestates. Modelling can also simulate the effect of a practice on the long term: as a second example, Bodilis et al. (2015) evaluated the long-term SOM storage induced by repeated applications of digestates.

STICS (Simulateur mulTIdisciplinaire pour des Cultures Standard) is a common one-dimensional crop and soil model running at the daily time step (Brisson et al., 2008). It has been designed to predict crop growth and soil characteristics (water, N, and C contents) under different soil and climatic conditions,

as well as different crop managements (e.g. tillage, fertilization, irrigation). STICS simulates various processes from soil C and N cycles: soil C and N mineralization, nitrate leaching, ammonia volatilization, N_2O emissions, and soil organic carbon (SOC) storage. It has been already extensively used to evaluate the environmental performances of diverse cropping systems such as nitrate leaching at a territorial scale (Jégo et al., 2012), N leaching reduction induced by catch crops (Constantin et al., 2015), N losses induced by the use of legumes (Plaza-Bonilla et al., 2018a), greenhouse gas emissions in different cropping systems (Autret et al., 2020), or potential C storage at the French national scale (Launay et al., 2021).

STICS has been able to simulate multiple arable crops in a variety of pedoclimatic conditions (Coucheney et al., 2015), in a comparable way than other soil-crop models (Sansoulet et al., 2014; Yin et al., 2020). With a new EOM mineralization submodule, STICS has been also able to simulate crop yields and SOC storage in a 20 years long experiment using organic amendments (urban composts and manure) (Levavasseur et al., 2021b). However, it would be interesting to evaluate the ability of STICS to simulate cropping systems that strongly rely on the use of organic fertilizers such as digestates. We can also wonder if STICS is sensitive enough to represent the difference between undigested slurry and digestate. In such case, STICS could be used to simulate crop rotations to quantify the effects of the introduction of anaerobic digestion in a crop livestock farm.

Therefore, we propose here to evaluate the ability of the soil-crop model STICS at simulating the 3-year field experiment described in Chapter 3, which compared the N cycle in crop succession with different fertilization systems based on digestates or undigested effluents. This presented work addresses three objectives. First, we aimed to evaluate two submodules of STICS related to digestate behavior in soil (the EOM mineralization and ammonia volatilization submodules) and to quantify the influence of the parametrization of those submodules on the other outputs of STICS. Then, we aimed to evaluate the ability of STICS model to simulate the crop and soil variables in crop rotations based on organic fertilizers. At last, we aimed at highlighting the advantages of the use of a crop model to estimate indicators such as N_2O emissions, nitrate leaching, and SOM storage that are difficult to measure in the field.

4.2 Material and Methods

4.2.1 3-year field experiment and EOM characterization

We used a 3-year field experiment located on a crop-livestock farm, at Nouzilly, Centre-Val-de-Loire, France (47°32’ N, 0°48’ E), from the 1st of September 2016 to the 31st of July 2019 (Pasquier et al., 2019). The climate is temperate oceanic, with an average annual mean temperature of 11.9 °C and annual total rainfall of 650 mm. The soil is mainly a Stagnic Luvisol, with a minor surface described as a Haplic Planosol (WRB, 2015), with an average depth of 100 cm. It was a silty loamy soil with the following

characteristics in the upper horizon (0-20 cm) (mean ± standard deviation): 17% ± 3% clay, 63% ± 5% silt, 20% ± 5% sand, pH 6.4 ± 0.3, 23.8 ± 2.3 g kg^{-1} SOM, C:N ratio 10.2 ± 0.2, $CaCO_3$ content 1.4 ± 0.3 g kg^{-1}, bulk density 1.37 g cm^{-3}.

The crop rotation was winter wheat (*Triticum aestivum*) (2016/2017), rapeseed (*Brassica napus*) (2017/2018), and winter wheat (2018/2019), with conventional management (ploughing at 20 cm, use of crop protection products) and with wheat straw removed. The goal of the experiment was to characterize the N cycle under different fertilization systems. The treatments were tested in plots of 24 x 75 m each. The five treatments consisted of (MN) fertilization with only Urea Ammonium Nitrate (UAN) solution at the end of winter or early spring, and chemical P fertilizer when needed; (CSM) application of solid bovine manure in summer and liquid cattle slurry in late winter or early spring; (RD) application of raw digestate in summer and in late winter or early spring; (LSD) application of solid digestate in summer and liquid digestate in late winter or early spring; and (0N) no fertilization.

The used EOMs were either undigested dairy cattle effluents, or digestates. The raw digestate came from a local anaerobic digester (continuous stirred tank reactor, mesophilic process) that treated 7,500 t of wastes per year (cattle slurry 11%, manures 17%, sewage sludge 29%, agroindustrial wastes 18%, grease 8%, cereal middlings 8 %, water runoff 9%, minor others). The raw digestate was post-treated by phase separation (screw press) to obtain liquid digestate and solid digestate.

UAN solution (MN treatment), cattle slurry (CSM treatment), raw digestate (RD treatment), and liquid digestate (LSD treatment) were applied at the end of winter or early spring, twice on wheat and once on rapeseed (Table 4.1). Their doses of application were calculated to provide similar available mineral N to crops as mineral fertilizer. Cattle manure, raw digestate, and solid digestate were applied in late summer before crop sowing (once in 2017 before rapeseed and once in 2018 before wheat) (Table 4.1). The objective was mainly to bring organic matter in the field to improve soil fertility at a longer-term.

Table 4.1 – Average characteristics of EOM during the three years, and application doses. Characteristics are presented as mean ± standard deviation.

Treatment	EOM or fertilizer	DM (%FW)	pH	VS (g kg^{-1}FW)	C:N_{total}	TN (g kg^{-1}FW)	TAN (g kg^{-1}FW)	Period of application	Average dose (t ha^{-1})	$N_{mineral}$ applied (N_{total} applied) during 3 years (kgN ha^{-1})
MN	UAN solution							winter/spring		389 (389)
CSM	Cattle slurry	4.6 ± 3.7	7.4 ± 0.5	24 ± 20	5.2 ± 2.2	2.3 ± 1.0	1.1 ± 0.5	winter/spring	33	193 (315)
CSM	Cattle manure	30.8 ± 3.9	9.7 ± 0.3	253 ± 17	15.3 ± 1.8	7.4 ± 1.5	0.7 ± 0.2	summer	24	30 (381)
RD	Raw digestate	6.7 ± 1.0	8.0 ± 0.1	45 ± 4	4.9 ± 0.4	5.1 ± 1.2	2.8 ± 0.8	winter/spring	27	323 (608)
								summer	24	110 (219)
LSD	Liquid digestate	4.7 ± 1.0	7.9 ± 0.2	29 ± 6	3.3 ± 0.2	4.4 ± 0.7	2.4 ± 0.5	winter/spring	29	330 (630)
LSD	Solid digestate	25.5 ± 4.5	9.3 ± 0.1	232 ± 52	17.8 ± 5.4	6.6 ± 0.4	1.8 ± 0.5	summer	25	71 (317)

FW : fresh weight.

Total aerial and grain biomass and their N content were measured at harvesting for each crop. Climatic data were followed with an on-site meteorological station, completed with daily data from Météo-France (SAFRAN). The topsoil water content, and topsoil mineral N content (0-20 cm) were assessed on 45 dates during the three years, with time intervals from three days (after EOMs or fertilizer applications) to one month. Soil water content (SWC) was continuously monitored during the third year of the experiment using time-domain reflectometry (TDR) moisture sensors at depth 15 cm, 35 (or 45) cm, and 75 cm, at two locations. The mean volumetric humidity from the sensors were used to estimate the water content (in mm) in the 0 – 25 cm, 25 – 60 cm, and 60 – 100 cm horizons. Soil mineral nitrogen stocks (SMN) on the whole soil profile (0 – 20 cm, 20 – 40 cm, 40 – 70 cm, 70 – 100 cm) were measured 7 times in three years: in early winter before drainage, at the end of winter after drainage, after harvesting. Ammonia volatilization was measured after each application event with passive samplers and reverse modelling (Carozzi et al., 2013b; Loubet et al., 2010). The N_2O emissions were measured on 39 dates with manual static closed chambers, at the same dates than topsoil water content and topsoil mineral N content.

The EOMs were sampled and analyzed at each spreading date: DM, TAN, Total N (TN), VS, pH (Table 4.1). The EOMs were also incubated in soil in laboratory controlled conditions: mixture of sieved soil and fresh EOMs were incubated during 6 months at 28°C with optimal SWC and SMN conditions, to measure the kinetics of C and N mineralization induced by their decomposition.

More details about field experiment monitoring, and EOMs analyses have been described in Chapter 3.

4.2.2 Simulating the three-year crop sequence with STICS

We used the STICS model, version 10.2751, to simulate the field experiment in Nouzilly. Input meteorological data, soil data (soil depth, bulk density, texture, $CaCO_3$, C:N, SOM, pH), initial conditions (SWC, SMN), and crop management (date and depth of tillage, date and density of sowing, date and dose of fertilizer applications, characteristics of fertilizers, date of harvest) came from field experiment monitoring (Chapter 3). The characteristics of EOM included C content, $C:N_{org}$ ratio (organic C : N_{org}), TAN content, DM content. The soil water contents (SWC) at field capacity and at wilting point were estimated for each horizons (0 cm – 20 cm, 20 cm – 40 cm, 40 cm – 70 cm, 70 cm – 100 cm) from the highest and lowest SWC measured by TDR moisture sensor during the cropping season, as suggested in STICS documentation (Supplementary Material 1). Other soil parameters were fixed from soil characteristics as proposed in Brisson et al. (2008).

Some soil parameters (the maximal soil depth affected by soil evaporation and a parameter defining the soil contribution to evaporation versus depth) were adjusted to improve soil water content prevision in the (MN) and (0N) treatments. The density of volunteer rapeseed were also adjusted by hand to improve topsoil mineral N modelling on (MN) and (0N) treatments. We selected the crop cultivars that provided the most accurately simulated yields and plant N uptake for each year on (MN) and (0N) treatments.

The other parameters (including plant parameters) were kept at their default values, to preserve model consistency.

Additionally, the simulation of EOMs required to calibrate the EOM mineralization submodule (section 4.2.3), and to investigate the ammonia volatilization model (section 4.2.4).

4.2.3 Calibration of the STICS submodule modelling the C and N mineralization after application of EOMs

STICS models the SOM with two compartments: inert SOM, and active SOM. Inert SOM (c.a. 65% of SOM) is assumed to not evolve with the time at the scale of few decades. The active SOM decomposition is modelled by a first-order kinetics (constant K_a). K_a depends on topsoil $CaCO_3$ and clay contents, and is influenced by soil temperature and moisture.

STICS includes a submodule to simulate the dynamics of evolution of organic C and N from EOMs and crop residues. For crop residues, the submodule represents the applied organic product within one pool of organic matter, as described in Justes et al. (2009). For EOM, a new submodule has been proposed recently by Levavasseur et al. (2021b, 2021a) to improve the simulation of the kinetics of C and N release from EOMs. The submodule uses eight parameters to describe EOM degradation. Levavasseur et al. (2021a) proposed default sets of parameters for different EOMs, but also argued that the use of the incubations of EOM in soil in controlled conditions could improve the simulation of EOM decomposition. Therefore, we compared two sets of parameters for each EOM, to evaluate the consequences of the calibration from laboratory incubations. The first set of parameters, named (EOM_cal), was obtained with the calibration of four parameters, using the incubation of EOM in soil in controlled conditions, similarly to Levavasseur et al. (2021a). The second set of parameters, named (EOM_def), was a default set of parameters only depending on the type of EOM (Levavasseur et al., 2021a).

The submodule that simulates the dynamics of organic C and N in soils considers two organic pools for EOMs, i.e. a labile organic fraction and a recalcitrant organic fraction (Supplementary Material 2). The proportion of labile organic C (*RES1*) is defined for each EOM. The repartition of organic N in both pools is defined by the parameter a_{CN1}, which is the ratio between $C:N_{org}$ ratio (organic N) from labile pool of organic matter and total $C:N_{org}$ ratio from the EOM. The labile organic fraction is degraded following a first-order kinetics (constant of degradation K_{res1}). The degraded carbon is either incorporated into a pool of zymogenous microbial biomass (proportion *Y*), or released in the atmosphere as CO_2. The pool of zymogenous microbial biomass is also degraded following a first order kinetics (constant K_{bio}) and the C is either incorporated inside the SOM (proportion *H*), or released in the atmosphere as CO_2. The N from the EOM labile fraction is first incorporated into zymogenous microbial biomass, then to SOM following C fluxes. Exchanges of N between zymogenous microbial biomass and SMN can occur to respect the constraints of C:N ratios of zymogenous microbial biomass (CN_{bio}) and

SOM. The recalcitrant fraction is also degraded following a first order kinetics (constant K_{res2}), with C entirely released as CO_2 and N mineralized. Finally, each EOM was parametrized using eight parameters: RES_1, a_{CN1}, K_{res1}, Y, K_{bio}, H, CN_{bio}, and K_{res2}. Values of H, K_{bio}, CN_{bio} were fixed and constant for all EOM. K_{res2} was set to be equal to K_a.

The calibration of the parameters in the (EOM_cal) set was realized independently from the whole STICS model, using only the EOM mineralization submodule and the incubations of EOMs in soil. It was realized in three steps with the R software (R Core Team, 2020), similarly to Levavasseur et al. (2021a). On the first step, K_a was calibrated using kinetics of C and N mineralization during soil incubations without EOM. The optimization used the optim function with the "Brent" method (Brent, 1973) (Supplementary Material 2). On the second step, the other parameters of the EOM submodel (RES_1, a_{CN1}, K_{res1}, Y) were calibrated using the "optim" function with the "L-BFGS-B" method (Byrd et al., 1995), which use lower and upper bounds for each parameter. These bounds were chosen according to Levavasseur et al. (2021a) and are described in Supplementary Material 2. The calibration parameters minimized the sum of RMSE (root mean squared error, section 4.2.6) of mineralized C (% of added organic C with EOM) and RMSE of mineralized N (% of added organic N with EOM), as described in Justes et al. (2009). Once calibrated, the found values of RES_1, a_{CN1}, K_{res1}, Y were used in the general STICS model.

4.2.4 Simulation of ammonia volatilization after EOM application using STICS or ALFAM2

STICS uses a semi-empirical submodule to simulate ammonia (NH_3) volatilization. Briefly, the model considers two pools of ammoniacal N, one that is close to the surface and can be volatilized, and one that cannot. The proportion of ammonium within each pool depends on the DM content of EOM and recent soil tillage: the more liquid the EOM is, the faster it infiltrates the soil, and infiltration decreases volatilization. The pH of surface soil changes just after EOM application, assuming that all EOMs have a pH equal to 8.6, because the pH of EOM is not an input of STICS model. The pH then regularly return to its original value at a rate proportional to NH_3 volatilization. Then, the equilibrium between three ammoniacal forms (NH_4^+ in aqueous phase, NH_3 in aqueous phase, NH_3 in gaseous phase) is calculated using the equilibria equations, whose constants of equilibrium depend on soil temperature, SWC, bulk density, and soil pH. Volatilization is computed with a resistive approach, aerodynamical resistances depend among others on wind speed and crop development. Emission is stopped after a tillage event. More detailed equations are described in Supplementary Material 3. This submodule was designed to simulate volatilization of liquid slurry after spreading. However, it is used by default for all solid or liquid EOM. Because of such limit, we also estimated ammonia volatilization of EOM with a second method.

ALFAM2 is a simple semi-empirical model (Hafner et al., 2019) to simulate volatilization after field application of slurry. Two pools of ammoniacal N are represented ("fast" and "slow"), with two volatilization rates. It was parametrized on a complete database of ammonia volatilization measurements from slurries (Hafner et al., 2018). Therefore, volatilization can be predicted from EOM characteristics (TS, pH, TAN), meteorological conditions (air temperature, wind speed, rainfall), application rate, technic of application (open slot, broadcast, trailing hose, trailing shoe), injection (deep or shallow). Compared to STICS, it only required one additional input variable, i.e. EOM pH. We used ALFAM2 model to predict ammonia emissions from slurry, raw digestates, and liquid digestates, whose DM content are below 15% (Hafner et al., 2019). We used "ALFAM2" R package (Hafner et al., 2019) to predict NH_3 emissions after 3 days and assumed that it corresponded to the total volatilization. ALFAM2 was not designed to predict volatilization from solid EOMs. Therefore, we used the mean emission factor measured in the field experiment to estimate the volatilization of solid manure and solid digestates, i.e. 66% of applied TAN (Chapter 3). This emission factor is consistent with other emission factors measured after solid manure application in the field with similar burying (ALFAM2 database) (Hafner et al., 2018).

At last, we also forced the values of ammonia volatilization to be the same as the measured values. It enabled to evaluate the performance of STICS model without considering the errors on ammonia volatilization.

In summary, we compared three submodules of ammonia volatilization inside the crop simulation with STICS: 1) (NH3_meas) used measured volatilization in the field experiment (Chapter 3), that were injected into STICS model; 2) (NH3_STICS) simulated volatilization of both liquid and solid EOM with STICS submodel; 3) (NH3_ALFAM2) simulated volatilization of liquid EOM by ALFAM2 and estimated volatilization of solid EOM by an emission factors then modelled values were injected into STICS model. The volatilization of mineral fertilizers were simulated specifically in STICS, it was not modified and only briefly evaluated.

4.2.5 Evaluation of STICS soil-crop model

Six classical statistical indicators were used to evaluate STICS model, as well as the EOM mineralization submodule and the volatilization submodule: root mean squared error (RMSE), relative root mean squared error (rRMSE), mean difference (MD), relative mean difference (rMD), squared Pearson correlation coefficient (r^2), and model efficiency (EF). MD and rMD indicated the bias in the simulation. r^2 indicated if the difference between years or treatments were well reproduced. RMSE, rRMSE, and EF were indicators of the global error of the simulations.

They were computed as:

(equation 4.1)

$$RMSE = \sqrt{\frac{1}{n} \cdot \sum_{i=1}^{n} (O_i - S_i)^2}$$

(equation 4.2)

$$rRMSE = \frac{RMSE}{\bar{O}}$$

(equation 4.3)

$$MD = \frac{1}{n} \cdot \sum_{i=1}^{n} (S_i - O_i)$$

(equation 4.4)

$$rMD = \frac{1}{\bar{O}} \cdot MD$$

(equation 4.5)

$$r^2 = \left(\frac{\sum_{i=1}^{n} \left((O_i - \bar{O}) \cdot (S_i - \bar{S}) \right)}{\sqrt{\sum_{i=1}^{n} (O_i - \bar{O})^2} \cdot \sqrt{\sum_{i=1}^{n} (S_i - \bar{S})^2}} \right)^2$$

(equation 4.6)

$$EF = 1 - \frac{\sum_{i=1}^{n} (O_i - S_i)^2}{\sum_{i=1}^{n} (O_i - \bar{O})^2}$$

Where O_i are the observed values, S_i the simulated values, $\bar{O}$ the mean of observed values, and $\bar{S}$ the mean of simulated values.

EOM mineralization submodule was evaluated directly with incubations of EOM in controlled conditions. Two simulations of the submodule (i.e. one per set of parameters) were run for each EOM. The simulated C and N mineralization of organic product during incubation were compared to the observed one. The five EOMs were pooled to compute the indicators of performance.

Volatilization of ammonia after EOM applications simulated with both (NH3_STICS) and (NH3_AFLAM2) were compared to measured volatilization (7 dates and 3 organic fertilizers per date). The evaluations were made considering the volatilization of both liquid and solid organic fertilizers, as well as the volatilization of liquid fertilizers only. Volatilization of synthetic fertilizers, simulated with a separate submodule, was evaluated separately.

Finally, STICS was evaluated on the five treatments and the three years. The evaluated variables were: SWC (0 – 20 cm), SWC (0 – 100 cm), SMN (0 – 20 cm), SMN (0 – 100 cm), aerial plant biomass at harvest, aerial plant N content at harvest, grain biomass at harvest, grain N content at harvest.

We analyzed four series of simulation runs. The reference one used (EOM_cal) EOM mineralization submodule and (NH3_ALFAM2) volatilization submodule. The effect of EOM calibration on STICS performance was quantified with a second series of simulation runs that used (NH3_ALFAM2) volatilization submodule and (EOM_def) set of parameters for EOM mineralization submodule. The effect of the choice of volatilization submodule on STICS performance was evaluated with the last two series of simulation runs, using (EOM_cal) set of parameters for mineralization submodule, and (NH3_STICS) and (NH3_MEAS) volatilization submodule, respectively.

4.2.6 Sensitivity of STICS output to EOM parameters.

We assessed how much the parametrization of digestates and cattle effluent influenced the main outputs of STICS. We realized a local one-at-a-time sensitivity analysis, as defined by Monod et al. (2006). The reference simulation runs used (EOM_cal) and (NH3_ALFAM2).

We investigated the following inputs: application date, DM content (without change in VS or TAN applied), TAN content, $C:N_{org}$ ratio, VS content (constant $C:N_{org}$ ratio). These parameters were first modified one at a time for all EOM applications in winter/spring during the crop rotation on one side (five applications). They were then modified for all EOM applications in summer (two applications).

We used six additional simulation runs for each studied input parameter. Application date was modified by -4, -2, -1, +1, +2, and +4 days. The other parameters were modified by -20%, -10%, -5%, +5%, +10%, +20%. Indeed, Chapter 3 showed that EOM characteristics could vary up to 20% between two applications, and application date could easily vary of one week around ideal application date due to meteorological data and logistical constraints.

The impact of these changes were assessed on the following outputs: yearly average SMN in the upper horizon (0 – 20 cm), yearly average SMN in the whole profile (0 – 100 cm), aerial plant biomass at harvest, N content in plant aerial organs at harvest, grain yield, N in grain at harvest, yearly ammonia volatilization, yearly N_2O emissions, yearly nitrate leaching. We analyzed the consequences of the change of each input on each output, considering each cropping season (i.e. 2016/2017, 2017/2018, and 2018/2019) and each organic treatment (CSM, RD, and LSD treatments). Nitrate leaching was measured

only in 2018/2019, as leaching were null or very low during the first two years. The impact of EOM characteristics applied in summer was not evaluated in 2016/2017 (no EOM application in summer 2016).

We draw the response profiles of outputs to varying inputs for each treatment and cropping season. Then, we computed a sensitivity indicator for each couple of input X and output Y. Because some response profile were not linear, the classically use of local derivatives were not adapted, and we used an indicator based on the range of output values across the different runs (Monod et al., 2006). We used the index proposed by Bauer and Hamby (1991) computed as:

(equation 4.7) $$\mathrm{I}_X^Y(\mathrm{y},\mathrm{t}) = \frac{\max_i(Y_X(i,y,t)) - \min_i(Y_X(i,y,t))}{\max_i(Y_X(i,y,t))}$$

where $\mathrm{I}_X^Y(\mathrm{y},\mathrm{t})$ is the index of sensitivity of output Y in response to input X, on treatment t and year y, and $Y_X(i,y,t)$ is the value of the output for the simulation testing the variation of input X at a rate i, on year y and treatment t. Here, the sensitivity is thus expressed as the normalized range that the output Y experience in response to a change of input X of ± 20% (± 4 days for application date). For each couple of input X and output Y, we analyzed the mean $\mathrm{I}_X^Y(\mathrm{y},\mathrm{t})$ across cropping seasons and treatments, which is named I. It is interpreted as following one example. Considering the effect of a change of TAN content of the EOMs applied in winter on crop yield, I = 4% means that if we change the TAN content of EOM applied in winter by ± 20%, the simulated yield will change of 4% on average depending on the year and treatment.

In addition, the output of the simulation runs of STICS using (EOM_def), as well as the simulation runs using (NH3_meas) or (NH3_STICS), were compared to the output of reference simulation runs. We studied the same six outputs. For each simulation run, we computed the normalized change of output variables, compared to the reference instance of STICS, for each year and each of CSM, RD, and LSD treatments. We also summarized those changes in computing the mean absolute normalized change of each output across the three years and the three treatments.

4.3 Results

4.3.1 Calibration and evaluation of STICS module for residue mineralization

The incubations has been already described in Chapter 3 and are briefly summarized here. The mineralization of C in EOM reached a plateau at the end of the incubations. The amount of mineralized C is an indicator of the stability of the organic matters of the EOM. The proportions of mineralized C was similar for all EOMs (between 30% and 40%) except for the raw digestate (20%) (Figure 4.1.A). The solid digestate and cattle slurry induced an immobilization of mineral N in the soil, the

decomposition of the raw digestate and the cattle manure had no impact on SMN, and only the decomposition of the liquid digestate induced a release of mineral N.

The two sets of parameters used in the EOM mineralization submodule are detailed in Supplementary Material 2.

In the (EOM_def) set, the proportion of C in the labile pool RES_1 was 0.24 for all three digestates and cattle manure, and 0.42 for cattle slurry. In the (EOM_cal) set, RES_1 were 0.27, 0.57, 0.27, 0.22, 0.51 for cattle manure, cattle slurry, raw digestate, liquid digestate, and solid digestate, respectively. Default values of RES_1 were close to the parametrized one for cattle slurry, cattle manure, and raw digestate. It revealed that during phase separation, the solid digestate was enriched in labile C (higher RES_1 than raw digestate), and the liquid digestate in recalcitrant C (lower RES_1 than raw digestate). In the (EOM_cal) set, the proportions of organic N in the labile pool, defined from RES_1 and a_{CN1}, were 0.03, 0.06, 0.03, 0.13, 0.05 for cattle manure, cattle slurry, raw digestate, liquid digestate, and solid digestate, respectively. They were different than the one in the (EOM_def) set that were 0.08 for cattle manure, 0.20 for cattle slurry, and 0.02 for all three digestates. The high N quantity in the labile pool of liquid digestate could be one explanation of the early N mineralization after being applied in the soil, in particular compared to raw digestate. The degradation rate of labile pool (K_{res1}) and the proportion of C incorporation in zymogeneous microbial biomass (Y) had similar values between (EOM_cal) and (EOM_def) for all EOMs, except for liquid digestate. The labile organic pool of liquid digestate was characterized by a fast degradation rate (K_{res1} = 0.34 day^{-1}) compared to other EOMs (K_{res1} < 0.06 day^{-1}). The labile organic pool of liquid digestate was poorly incorporated into the biomass and C was thus mainly emitted under the CO_2 form (low Y = 0.1). This was related to the very quick CO_2 emission after liquid digestate incorporation in the soil. The parametrization indicated that organic matter inside the liquid and solid digestates had different properties, the single "digestate" default set of parameters might not be appropriate. The incubations also enabled to better simulate the behaviour of SMN after the application of EOMs in the soil.

The (EOM_cal) set of parameters enabled a good simulation of C mineralization with RMSE = 3.3% of organic C from EOM mineralized (% added C_{org}), MD = 0.1% added C_{org}, r^2 = 0.93, and EF = 0.90 (Figure 4.1.A). The plateau observed at the end of the incubation was not reproduced, because the mineralization of the stable organic pool occured at the same rate than the active SOM ($K_{res2} = K_a$ = 3.78 10^{-4} year^{-1}). However, the (EOM_def) set of parameters also enabled to model the C mineralization of EOM with a "reasonable accuracy" (Figure 4.1.A), with RMSE = 5.8% added C_{org}, MD = -1.4% added C_{org}, r^2 = 0.76 and EF = 0.70.

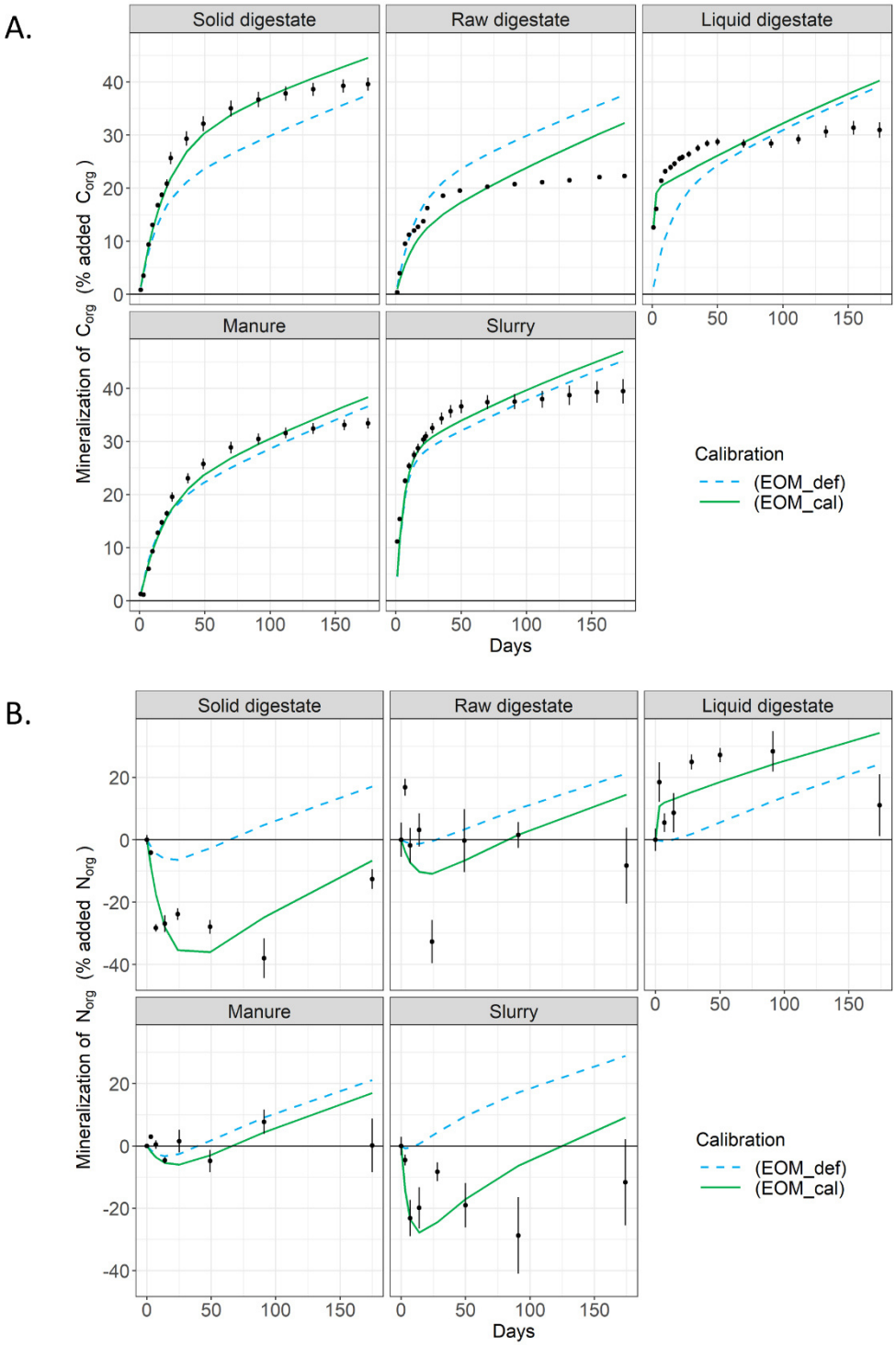

Figure 4.1 – A) C mineralization from EOMs during incubation in soil in controlled conditions. B) N mineralization from EOMs during incubation in soil in controlled conditions. Measures are displayed with dots, and simulated values with lines. Two sets of simulations are represented with the set of calibrated parameters (EOM-cal) or with the default values proposed in STICS (EOM-def). Error bars show one standard error.

The (EOM_cal) set of parameters successfully simulated the N mineralization of all EOMs (Figure 4.1.B), with RMSE = 11% of added organic N mineralized from EOM (% added N_{org}), MD = 0.4% added N_{org}, r^2 = 0.59, and EF = 0.54. Similarly as C mineralization, because of K_{res2} being equal to K_a, the N mineralization increased linearly from day 50 for all EOM, even if this phenomenon was not measured during the incubations. (EOM_def) failed to correctly reproduce the N mineralization of EOMs during the incubation, as shown by high RMSE (19.4% added N_{org}), low r^2 (0.01), and negative EF (-0.45). It caused a strong overestimation of the mineralization of N from EOMs (MD = 8.2% added N_{org}).

4.3.2 Evaluation of volatilization submodule of STICS

The ammonia volatilization following EOM application was already described in Chapter 3 and is briefly summarized here. Concerning the liquid EOMs, the raw digestate was the most sensitive to volatilization (high TAN content, low infiltration), followed by liquid digestate then by cattle slurry (lower TAN content). The ammonia volatilization after solid EOM applications was important (up to 25 kgN ha^{-1}), despite their burrowing 24h after their application.

STICS were able to simulate ammonia volatilization from liquid digestate and slurry with a reasonable accuracy (RMSE = 5 kgN ha^{-1}, rRMSE=69%), slightly underestimating it (rMD = -34%). However, STICS strongly underestimated the volatilization from raw digestate, solid digestate, and cattle manure. Considering all EOM, STICS failed to reproduce the trends of volatilization (RMSE = 14.7 kgN ha^{-1}, rRMSE = 109%, rMD = -67%, r^2 = 0.03, EF = -0.59) (Figure 4.2). Additionally, the timing of simulated volatilization with STICS did not reproduce the measurements: in the model, emissions could occur during two months or more (data not shown), when it was measured to last less than one week (Chapter 3).

ALFAM2 simulated ammonia volatilization from slurry and liquid digestates, with a good accuracy (RMSE = 3.8 kgN ha^{-1}, rRMSE = 52%, rMD = -10%, r^2 = 0.62, EF = 0.58), superior to STICS (Figure 4.2). The ammonia volatilization of raw digestate was underestimated. However, the evaluation of ALFAM2 to simulate the volatilization of all liquid EOM (slurry, liquid digestates, and raw digestate) remained correct (RMSE = 9.7 kgN ha^{-1}, rRMSE = 75%, r^2 = 0.47, EF = 0.37). The measured emission factors after application of solid EOM were used to represent ammonia volatilization after solid EOM application, thus explained their good accuracy. The volatilization from synthetic fertilizers simulated with STICS was underestimated (rMD = 2.9kgN ha^{-1}) and were simulated with a RMSE of 4.1 kgN ha^{-1}. The evaluation of the ammonia volatilization is further detailed in Supplementary Material 4.

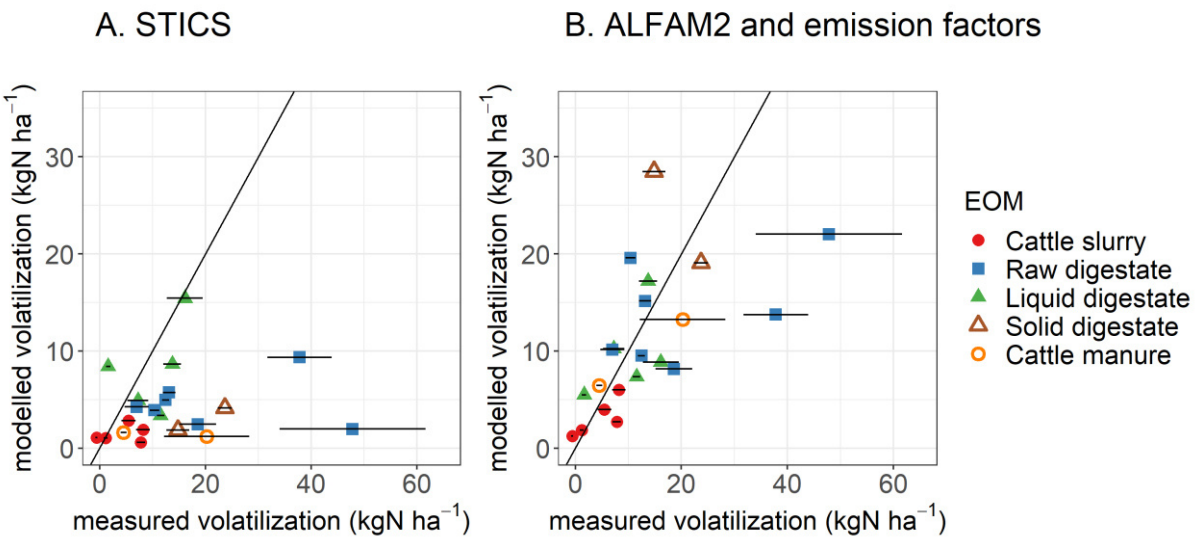

Figure 4.2 - Comparison of measured volatilization to simulated volatilization by A) STICS model, and B) ALFAM2 model (cattle slurry, raw digestate, liquid digestate) and emission factors (manure, solid digestate). Error bars show one standard error.

Considering the 3 years, total volatilization using (NH3_ALFAM2) submodule were 36 kgN ha^{-1}, 98 kgN ha^{-1}, 97 kgN ha^{-1} for CSM, RD, and LSD treatment, respectively. Only volatilization in RD treatment were significantly underestimated compared with measured total volatilization (46 kgN ha^{-1}, 147 kgN ha^{-1}, 89 kgN ha^{-1} for CSM, RD, and LSD treatments, respectively). On the contrary, total volatilization using (NH3_STICS) submodule were strongly underestimated (11 kgN ha^{-1}, 32 kgN ha^{-1}, 47 kgN ha^{-1} for CSM, RD, and LSD treatment, respectively).

4.3.3 STICS performances

The agronomical results of the field experiments were described in Chapter 3. Briefly, the use of digestate (LSD and RD) had globally similar yields that the (MN) treatment using synthetic fertilizers. The yields obtained with raw digestate (RD) were slightly lower than the one using liquid and solid digestates (LSD) treatment, which was attributed to high volatilization. The use of cattle effluents (CSM) resulted in lower yields than with other fertilizers, because of the lower inputs of mineral N.

In the reference simulation runs, SWC of the topsoil along the three years were correctly simulated (RMSE = 3.58% w/w, rRMSE = 17%, rMD = 7%, r^2 = 0.70, EF = 0.62). The water stocks in the whole soil profile were also well reproduced in 2019 (RMSE = 39 mm, rRMSE = 13%, rMD = 9%, r^2 = 0.89, EF = 0.50) (Supplementary Material 5). STICS consistently simulated the variations of SMN content in the topsoil (Figure 4.3), the RMSE were 22 kgN ha^{-1} (Table 4.2). SMN on the whole soil profile were underestimated (MD = -17 kgN ha^{-1}, ARE = -34%), for all treatments (Supplementary Material 5).The plant aerial and grain biomass and N uptake at harvest were reasonably well predicted (Table 4.2, Figure 4.4). RMSE were 2.90 t ha^{-1} for aerial plant DM, 32.0 kgN ha^{-1} for aerial N uptake, 0.74 t ha^{-1}

for grain yield, and 15.5 kgN ha^{-1} for N export in grain. STICS slightly overestimated wheat aerial biomass (rMD = 20%) and N uptake (rMD = 25%), and underestimated rapeseed aerial biomass (rMD = -25%) and grain yield (rMD = -19%) (Figure 4.4, Supplementary Material 5). The dry biomasses returned to soil with crop residues at harvest were overestimated: RMSE = 2.02 t ha^{-1} (rRMSE = 51%), MD = 1.25 t ha^{-1} (rMD = 32%), EF = -0.30, r^2 = 0.22. Similar performance was obtained for N returned to soil with crop residues at harvest: RMSE = 11.2 kgN ha^{-1} (rRMSE = 50%), MD = 4.7 kgN ha^{-1} (rMD = 21%), EF = 0.34, r^2 = 0.62. The general performances of STICS were similar when considering all treatments, or only organic treatments CSM, RD, and LSD treatments (Supplementary Material 5): only topsoil SMN (rRMSE = 90%, r^2 = 0.31) and aerial N uptake (r^2=0.20) were slightly less well predicted considering only those three amended treatments.

In summary, STICS was able to simulate the crop and soil variable with a correct accuracy. The sensitivity of STICS enabled to differentiate the treatments with important differences (Figure 4.4). The treatments were well differentiated in 2017 and 2018, and slightly less in 2019. The application of a high dose of TAN in (RD) treatment before rapeseed sowing led to an overestimation of rapeseed growth and yield.

The use of (NH3_STICS) or (NH3_ALFAM2) submodule did not degraded the overall STICS performances compared to the use of measured values of volatilization (NH3_meas) (Table 4.2); only the prediction of topsoil SMN was slightly degraded (Table 4.2, Supplementary Material 5). Therefore, the change of volatilization submodule mainly influenced the prediction of NH_3 volatilization. The use of (EOM_def) instead of (EOM_cal) parametrization increased N mineralization from EOM, and thus increased the SMN content in the entire soil layer. Consequently, the use of default values (EOM_def) slightly decreased the prediction quality of aerial N uptake and N in grains, and slightly the prediction quality of grain yield compared to (EOM_cal) (Table 4.2).

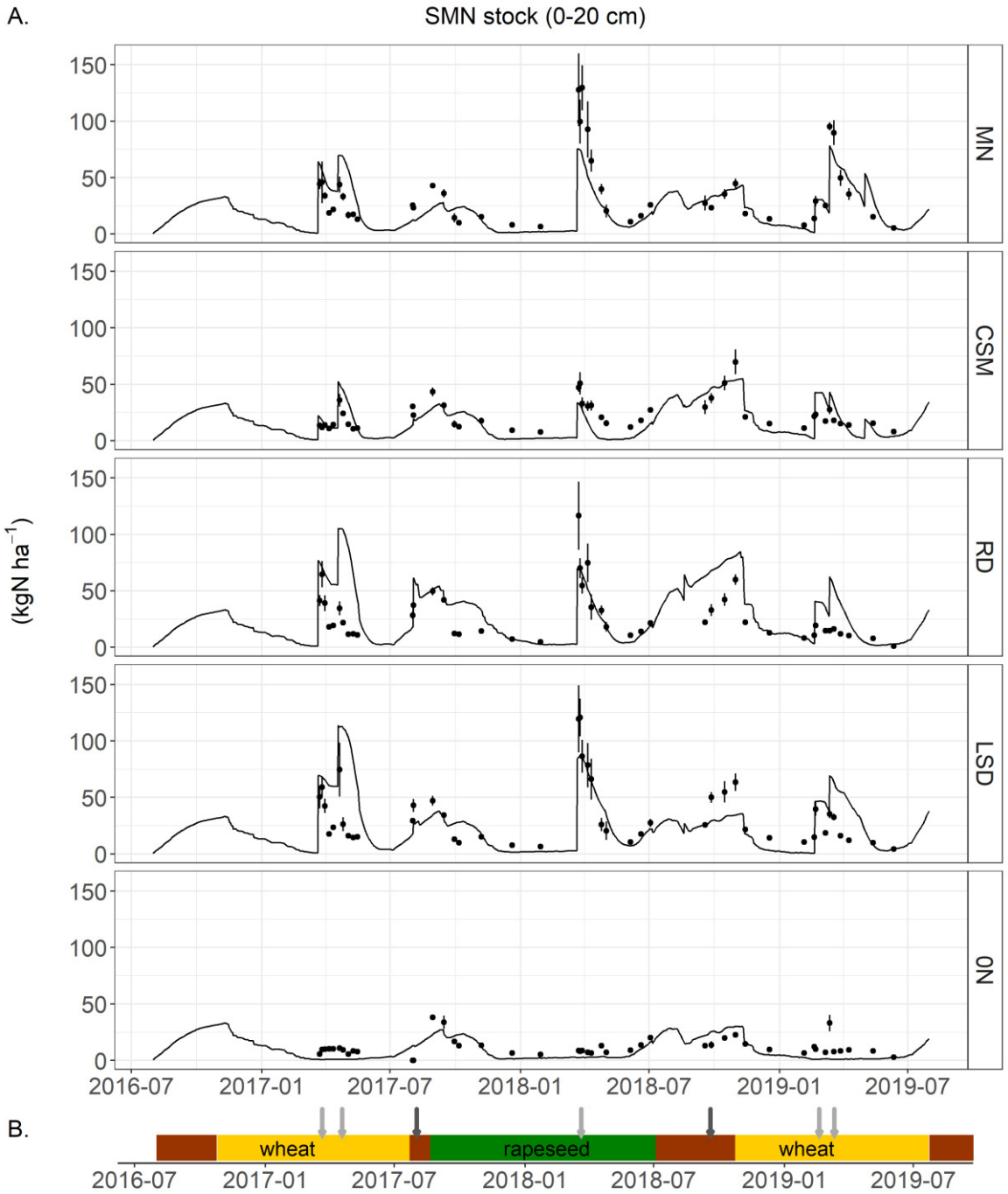

Figure 4.3 – Simulated and measured SMN content in topsoil (0 – 20 cm). STICS simulations used (NH3_ALFAM2) submodule and (EOM_cal) parametrization. Solid line represent the simulated SMN content. Dots represent measured SMN content, error bar represent one standard error.

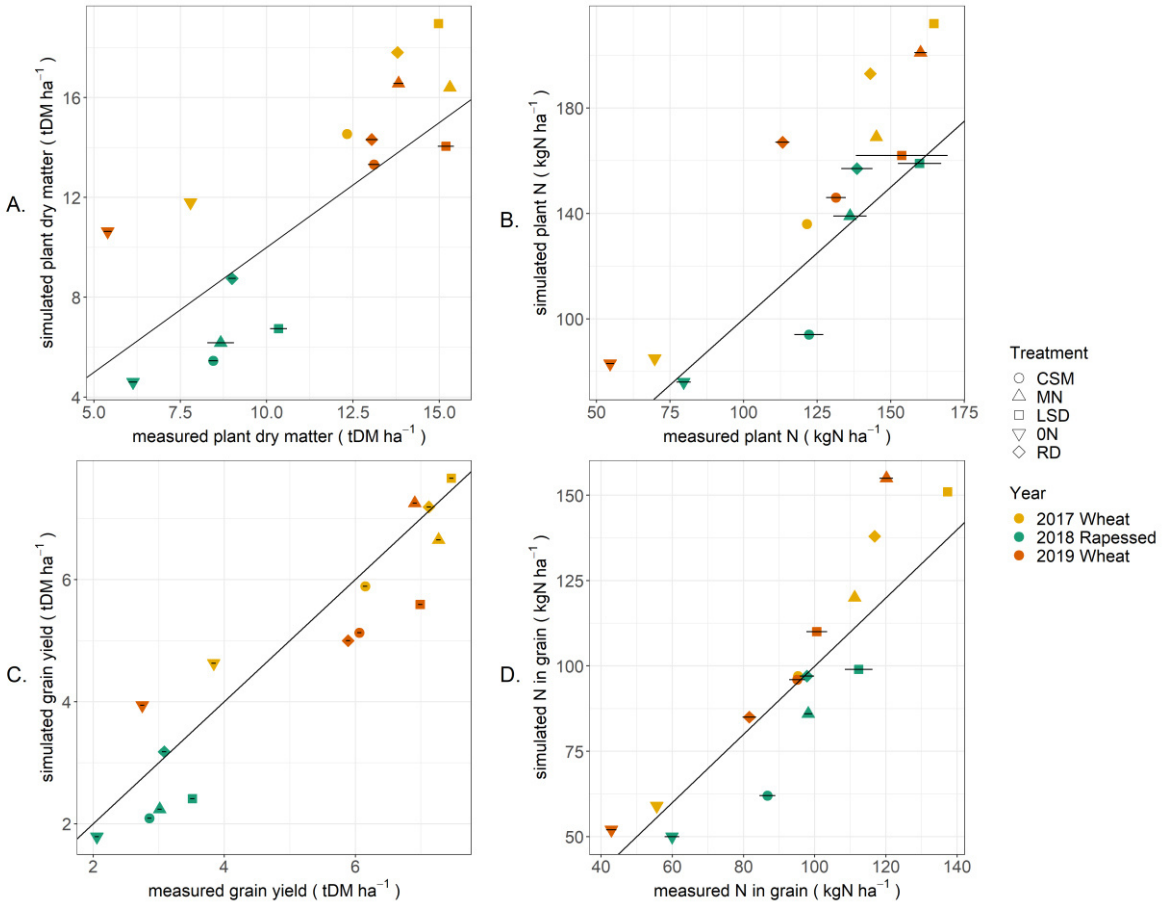

Figure 4.4 – Performance of STICS to model. A) aerial plant dry matter, B) aerial plant N uptake, C) grain yield, and D) N exported in grain. For A) and C) error bars represent measurement uncertainty (type B uncertainty). For B) and D), error bars show one standard error. STICS simulations used (NH3_ALFAM2) submodule and (EOM_cal) parametrization.

Table 4.2 – Evaluation of STICS model, among the different parametrizations.

STICS calibration	variable	RMSE	rRMSE (%)	MD	rMD (%)	EF	r^2
Reference	Aerial dry matter (t ha^{-1})	2.89	26	0.92	8	0.22	0.65
(NH3_ALFAM2)	Aerial N uptake (kgN ha^{-1})	32.0	25	20.1	16	0.05	0.67
(EOM_cal)	Grain yield (t ha^{-1})	0.74	15	-0.24	-5	0.85	0.88
	N in grain (kgN ha^{-1})	15.5	16	3.3	4	0.61	0.79
	SMN (0-20 cm) (kgN ha^{-1})	22.0	83	4.0	15	0.16	0.41
	SMN (0-100 cm) (kgN ha^{-1})	27.1	54	-19.7	-39	-0.33	0.46
(NH3_STICS)	Aerial dry matter (t ha^{-1})	2.74	25	1.21	11	0.29	0.69
(EOM_cal)	Aerial N uptake (kgN ha^{-1})	37.0	29	26.6	21	-0.27	0.69
	Grain yield (t ha^{-1})	0.76	15	-0.18	-4	0.85	0.86
	N in grain (kgN ha^{-1})	16.1	17	5.8	6	0.58	0.78
	SMN (0-20 cm) (kgN ha^{-1})	21.4	81	5.2	20	0.20	0.47
	SMN (0-100 cm) (kgN ha^{-1})	26.3	52	-17.2	-34	-0.26	0.45
(NH3_MEAS)	Aerial dry matter (t ha^{-1})	2.74	25	0.74	7	0.29	0.66
(EOM_cal)	Aerial N uptake (kgN ha^{-1})	29.0	23	17.4	14	0.22	0.69
	Grain yield (t ha^{-1})	0.76	15	-0.24	-7	0.85	0.88
	N in grain (kgN ha^{-1})	14.4	16	1.2	1	0.65	0.78
	SMN (0-20 cm) (kgN ha^{-1})	20.3	77	3.2	12	0.28	0.46
	SMN (0-100 cm) (kgN ha^{-1})	27.1	54	-19.7	-39	-0.35	0.47
(NH3_ALFAM2)	Aerial dry matter (t ha^{-1})	2.84	25	1.29	12	0.24	0.70
(EOM_def)	Aerial N uptake (kgN ha^{-1})	35.4	28	26.8	21	-0.17	0.72
	Grain yield (t ha^{-1})	0.58	12	-0.07	-1	0.91	0.92
	N in grain (kgN ha^{-1})	18.4	20	8.6	9	0.45	0.75
	SMN (0-20 cm) (kgN ha^{-1})	21.6	82	5.3	20	0.18	0.43
	SMN (0-100 cm) (kgN ha^{-1})	23.7	47	-15.9	-31	-0.02	0.58

4.3.4 STICS sensitivity to EOM parameters

The change of outputs in function of the parametrization of the EOM mineralization submodule are shown in Supplementary Material 6. The response of outputs in function of the choice of EOM volatilization submodule are shown in Supplementary Material 7. The response of outputs in function of the variation of inputs linked to EOM characteristics are shown in Supplementary Materials 8 and 9. The Figure 4.5.A summarized the change of output induced by a change of submodule (EOM mineralization or volatilization); Figure 4.5.B summarized the change of outputs induced by the variation of the EOM characteristics (sensitivity indicators I).

The parametrization of EOM mineralization submodule significantly affected STICS outputs (Figure 4.5.A). The effect of the parametrization method was higher in 2017/2018 and 2018/2019, compared to 2016/2017. It was also higher for CSM and LSD treatment compared to RD treatments (Supplementary Material 6). This could be related to the higher amount of organic matter applied in the field, including during summer: a similar relative variation of the mineralization had higher absolute consequences. Compared to (EOM_cal), (EOM_def) generally increased SMN content in soil (change of 15%) and thus crop characteristics at harvest (7% to 11%), N_2O emissions (14%), and leaching (39%) (Figure 4.5.A).

The choice of EOM volatilization submodule affected volatilization, which was negatively correlated to SMN contents, crop biomass and N uptake at harvest, N_2O emissions, and leaching (Supplementary Material 7). Compared to the use of measured values of volatilization (NH3_meas), the use of (NH3_STICS) submodule underestimated volatilization for every year and treatment, excepted for LSD treatment in 2016/2017. The relative changes in volatilization were very high (65%), inducing changes of SMN contents, N_2O emissions, and leaching of 14 to 17%, and crop characteristics at harvest from 9 to 11% (Figure 4.5.A). Compared to (NH3_meas), the use of (NH3_ALFAM2) submodule could over- or underestimate volatilization depending on treatments and year. The mean absolute changes in STICS output induced by (NH3_ALFAM2) were about twice lower than the one induced by (NH3_STICS). For both (NH3_STICS) and (NH3_ALFAM2), RD treatment was more impacted than LSD or CSM treatments.

Considering the two investigated submodules, a change in the parametrization method or equations can affect output variables. These changes were low relatively to the error of the model; rRMSE was up to c.a. 25% for crop characteristics and 80% for SMN contents. Therefore, the fine parametrization of these submodules did not improve the performance of STICS on crop growth. However, it was important to estimate N losses consistently.

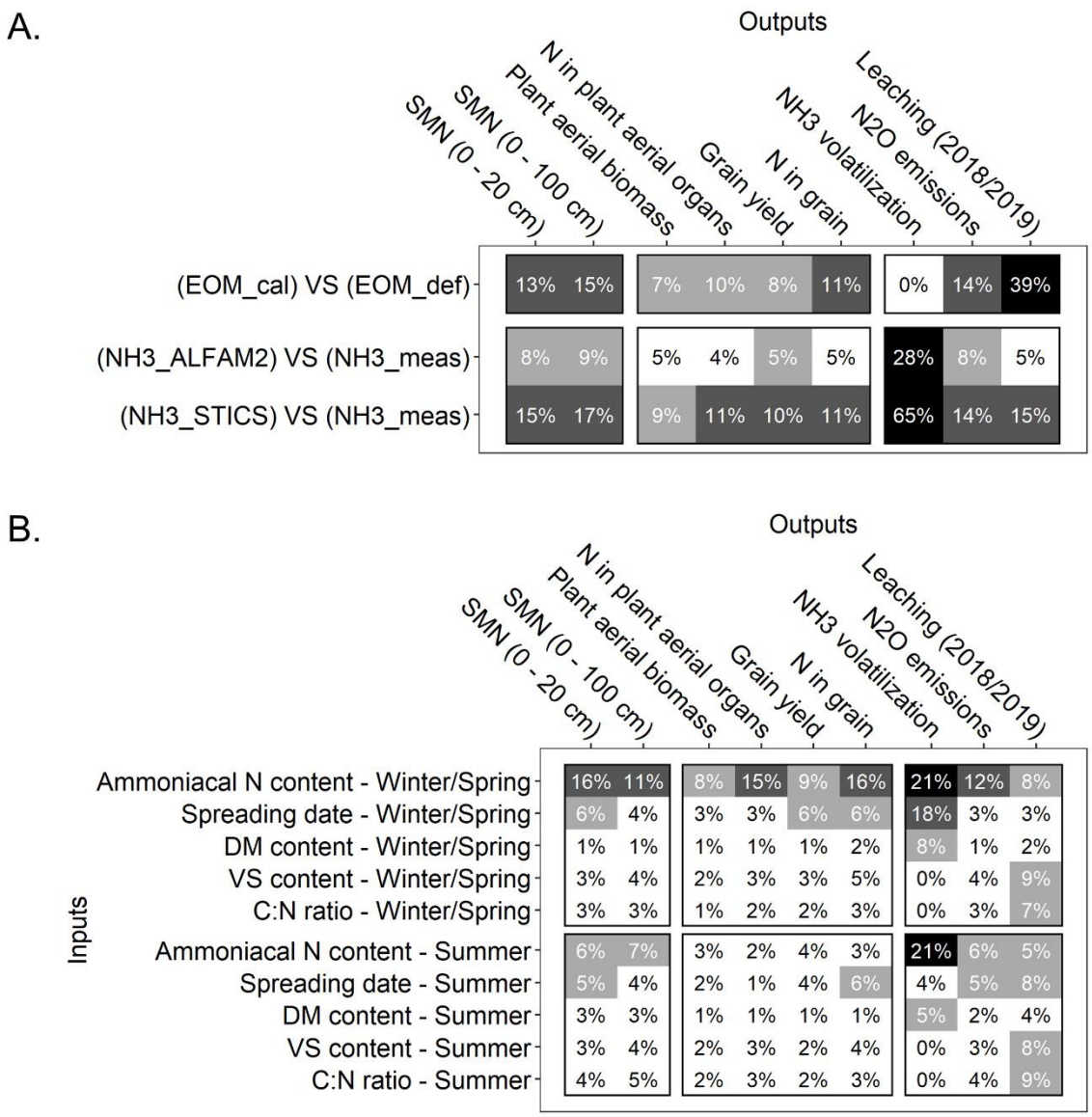

Figure 4.5 – Sensitivity analysis of EOM modelling and characteristics on STICS outputs. A) Changes in STICS output induced by the parametrization of EOM mineralization submodule, and by the choice of EOM volatilization submodule. 10% indicate that a change of submodule affect the output of ± 10% on average. B) Sensitivity indices I for each couple of outputs and EOM characteristics. I = 10% indicate that a variation of input of ±20% (±4 days for spreading dates) will induce on average a change of 10% in the output. For B), STICS simulations used (NH3_ALFAM2) submodule and (EOM_cal) parametrization.

The change of TAN content in EOM applied in winter or spring was positively correlated to all considered outputs. The effect of its change was homogeneous in all treatments and years (Supplementary Material 8). A change of up to ± 20% of TAN content induced on average sensitivity indices (I) comprised between 8 and 21% depending on the considered output (Figure 4.5.B). The application date of EOM applied in winter or spring strongly impacted EOM volatilization (I = 18%), and more moderately grain yield (I = 6%) and N in grain (I = 6%) (Figure 4.5.B). This was attributed to changes in meteorological conditions, and changes in crop development at fertilization date. A change in DM content was positively correlated to volatilization in winter and spring, with a relatively high sensitivity (I = 8%). In summer, volatilization was not impacted by application date nor DM content because of the use of emission factors for solid digestate and manure. The change of TAN content in EOM applied in summer importantly increased annual volatilization (I = 21%) and increased more moderately SMN content (I = 6% in the topsoil and 7% in soil profile), N_2O emissions (5%), and leaching (I = 6%) (Figure 4.5.B, Supplementary Material 9). Changes in VS or $C:N_{org}$ ratio of EOM applied at both season had only low impact on all considered outputs (Figure 4.5.B).

4.3.5 Modelling nitrate leaching, N_2O emissions, and soil organic N storage during the crop rotation

We analyzed the N balance in the different treatments with the reference simulation (NH3_ALFAM2 and EOM_cal) (Figure 4.6). The use of digestates (LSD and RD) instead of cattle effluent (CSM) induced higher N inputs, leading to higher N export in the crops, higher soil organic N storage, but also higher N losses. The three organic treatments (CSM, RD, and LSD) induced an increase in soil organic N storage compared to (MN) treatment. In (RD) treatment, the input of organic matter was lower than in (CSM) and (LSD) treatments. This conclusion was in line with the conclusion from the field experiment alone (Chapter 3). Thanks to modelling, we simulated leaching, N_2O emissions, and organic N storage, that were only approximately estimated in the field experiment.

Simulated total N_2O emissions during the three years were 9.4 kgN ha^{-1}, 8.1 kgN ha^{-1}, 13.2 kgN ha^{-1}, and 10.6 kgN ha^{-1} for MN, CSM, RD, and LSD treatments, respectively. The proportion of total yearly N_2O emissions occurring in summer and autumn (from 21st of June to 21st of December) were 62 %, 81%, 77%, and 63%, for MN, CSM, RD, and LSD treatments, respectively. CSM treatment emitted lower N_2O emissions compared to the other treatment because of lower input of mineral N, which was a conclusion of the field experiment (Chapter 3). However, the conclusion from modelling and field observation diverged concerning the MN treatment: MN treatments emitted less N_2O than RD and LSD treatments in the simulation when the emissions were similar in the 3 treatments for the field observation (Chapter 3). With static chambers, punctual N_2O emissions were measured. This cannot easily be compared to the daily simulated N_2O emissions, as the amplitude of the two measured and simulated emissions do not represent the same thing. However, we compared the dynamics of N_2O emissions (Supplementary Material 5). The emission peaks observed in winter and spring after the application of

the fertilizers were well reproduced. The simulations predicted several peaks of N_2O emissions during the summer and autumn. We did not observed the N_2O emission during all those periods. However, during some of them, there were experimental evidence of the absence of N_2O emissions. STICS could therefore overestimate N_2O emissions in this field experiment. As a result, the proportion of N_2O emissions between the summer/autumn season compared to winter/spring season was higher than the observed measurements in the field experiment (Chapter 3). This could be a reason explaining why the MN treatment emitted similar N_2O emissions than RD and LSD in the observations but less N_2O emissions than RD and LSD in the simulation.

During the 2016/2017 and 2017/2018 cropping seasons, simulated nitrate leaching was lower than 2 kgN ha^{-1} in each treatments, excepted for RD treatment in 2018 where leaching reached 12 kgN ha^{-1}. In 2018/2019, nitrate leaching was 33 kgN ha^{-1}, 38 kgN ha^{-1}, 58 kgN ha^{-1}, and 29 kgN ha^{-1}, for MN, CSM, RD, and LSD treatments, respectively. Therefore, the simulation confirmed the low leaching in 2016/2017 and 2017/2018, and that the use of organic amendments in summer before wheat in summer 2018 promoted leaching (Chapter 3). The difference between treatments in simulated leaching were lower than the one from estimated leaching from measured SMN stocks (Chapter 3). This was partly explained by the simulation of less N mineralization from solid digestate, than the one that can be estimated from the measured SMN stocks.

STICS simulated a change of soil organic N (SON) along the three cropping season of -137 kgN ha^{-1}, 212 kgN ha^{-1}, 109 kgN ha^{-1}, and 287 kgN ha^{-1}, for MN, CSM, RD, and LSD treatments, respectively. The simulated changes of soil organic C (SOC) were -1.99 tC ha-^{1}, 2.64 tC ha^{-1}, 0.42 tC ha^{-1}, and 2.81 tC ha^{-1}, for MN, CSM, RD, and LSD treatments, respectively. The use of EOM increased the SON and SOC storage. In RD treatment, the absence of solid EOM application did not allow to store as many SOC as CSM and LSD treatments.

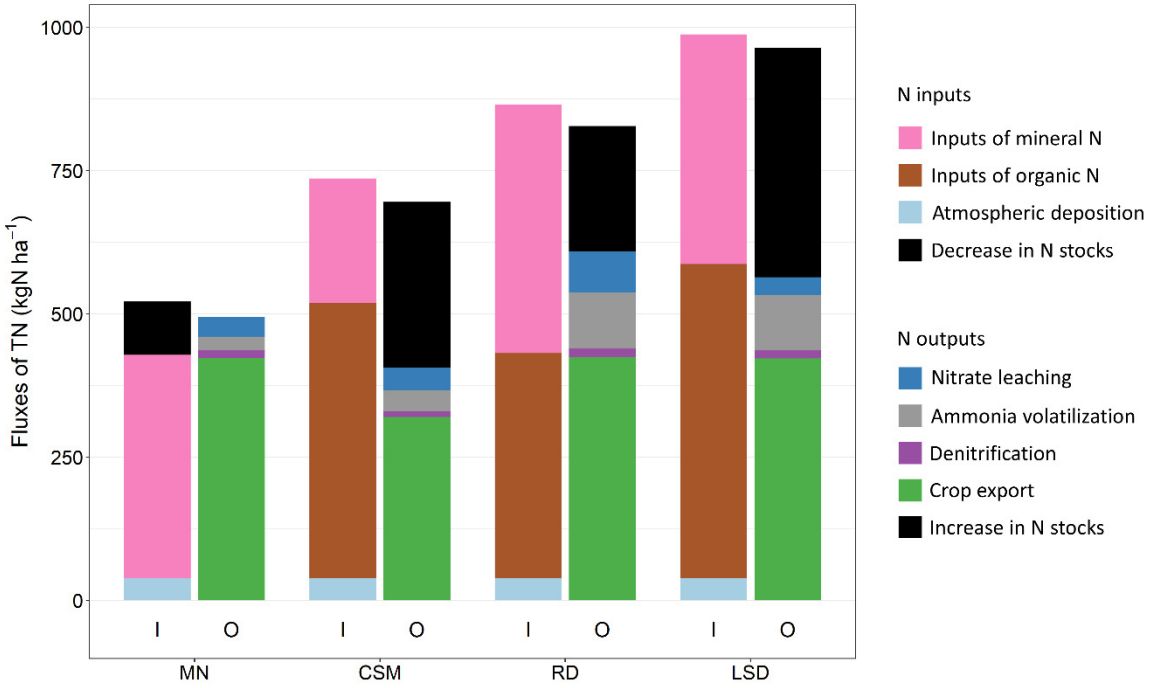

Figure 4.6 – Simulated fluxes of N in the field, during the 3-year crop rotation. Inputs of total N are on the the left (I), and output of N in the right (O). The variation of N stocks include soil organic N, SMN, and N in fresh crop residues. We observed a decrease in N stocks in MN treatment, and an increase in N stocks in CSM, RD, and LSD treatments (higher inputs than outputs).

4.4 Discussion

4.4.1 STICS performance to simulate EOM mineralization in controlled conditions

(EOM_cal) and (EOM_def) sets of parameters did not perform as well as in the previous study from Levavasseur et al. (2021b) where eight parameters were parametrized, instead of four in the present work. However, the RMSE were comparable to the one reported by Levavasseur et al. (2021a). The performance of STICS using incubation-based calibration (EOM_cal), was similar to the one obtained with RothC for C mineralization (Mondini et al., 2017), and NCSOIL model for C and N mineralization (Noirot-Cosson et al., 2016). All calibration were comparable to the performance of simulation of crop residues mineralization (Justes et al., 2009) (5% added C_{org}).

The use of laboratory experiments to calibrate soil-crop models in order to simulate field experiments assume an adequacy between results obtained in laboratory and field conditions. This hypothesis is assumed in many studies and gave good results, as deeply discussed by Mondini et al. (2017) for C mineralization. Regarding N mineralization from EOM, Cavalli et al. (2017) found a good adequacy between incubation in controlled conditions and mineralization of digestate in the field (Chapter 3). We also found a good adequacy between the EOM N mineralization and field N balance in the same field

experiments as the one used in the present study. The only exception being solid digestate that immobilized N in the incubation, although it was probably not the case in the field (Chapter 3).

Both the use of default parameters and the calibration of some parameters with EOM incubation allowed to simulate topsoil SMN contents with an acceptable accuracy. STICS was not sensitive enough to detect any improvement in the overall simulation. The parametrization of the EOM mineralization submodule allowed to differentiate some EOM, and influenced the output with a higher amplitude than most of the other EOM characteristics. We would recommend parametrizing EOM if they are an important factor of the treatments, e.g. to compare the use of different EOMs on SOM storage. Otherwise, the use of default parameters might be sufficient.

4.4.2 STICS performance to simulate EOM volatilization

In this study, we found that STICS strongly underestimated the NH_3 volatilization from raw digestate, solid digestate, and farmyard manure. The simulation of ammonia volatilization after EOM application was evaluated once on swine slurry, and the error of modelling were approximately 10 kgN ha^{-1} (Jing et al., 2017), slightly lower compared to the present evaluation of STICS.

ALFAM2 successfully simulated the NH_3 emissions of liquid and raw digestate. ALFAM2 model was selected because of its simplicity, its calibration based on a large dataset, and the ability to consider different type of slurry application method for further modelling experiments. ALFAM2 performed slightly better than the volatilization submodule of the soil-crop model DNDC (Congreves et al., 2016) (RMSE: 12 kgN ha^{-1}). The simplicity of ALFAM2 did not enable to consider variables such as soil humidity or canopy development, which can influence volatilization and are available in STICS (Brisson et al., 2008). Gericke et al. (2012) found a better prediction of volatilization after digestate applications, with a more complex model (RMSE = 2.0 kgN ha^{-1}). Such more complex model with finer time step could perform slightly better to simulate volatilization from liquid EOM.

Both STICS and ALFAM2 underestimated the volatilization from raw digestate. One hypothesis is that the raw digestate did not efficiently infiltrate soil after application, as observed in the field experiment (Chapter 3). Digestate could have a different viscosity than slurries used to calibrate ALFAM2 model. Better understanding liquid infiltration could improve volatilization model (Pedersen et al., 2021).

One issue in this study was the modelling of ammonia emission from solid EOMs. There were less studies interested in the volatilization from solid EOMs compared to liquid EOMs. To our knowledge, no mechanistic model to predict volatilization after field application of solid EOMs has been developed. Misselbrook et al. (2005) designed an empirical linear model that could be used to predict ammonia emissions. The model importantly overestimated the volatilization in the present field experiment and was not kept (data not shown). Currently, emission factors may be a robust way to predict field

volatilization of poorly investigated EOM such as composts, sewage sludge, or solid digestates (Houot et al., 2014).

STICS simulated the volatilization from EOM with a low accuracy, in particular with EOMs with medium or high DM contents (raw digestate, farmyard manure, solid digestate). The dynamics of the emissions were not satisfactory. We recommend to not use this submodule to estimate NH_3 volatilization from EOM. Instead, we recommend to use emission factors or another model, ALFAM2 being a good candidate for liquid EOM. These recommendations do not concern the NH_3 volatilization from synthetic N fertilizers.

4.4.3 STICS performance using organic fertilizers

In our study, STICS performance for the prediction of water content (topsoil and soil profile), SMN (topsoil and soil water content), and plant characteristics at harvest were similar than the one reported in Coucheney et al. (2015), who evaluated STICS model for different crops and soil and climate conditions in France. Only the SMN content in the topsoil was highly underestimated in our study in all treatments. The underestimation of rapeseed yield and aerial biomass by STICS was already highlighted by Coucheney et al. (2015). Levavasseur et al. (2021b) evaluated STICS in a long-term experiment using EOMs as soil amendment. Comparing only cropping systems using EOMs (Supplementary Material 4), our simulations were of similar quality compared to the one they obtained (grain yield, aerial biomass, SMN content in soil profile and aerial N uptake). They underestimated SMN content in the soil profile by 17 kgN ha^{-1}, similarly to our results. Guest et al. (2017) also found that STICS underestimated SMN in soil profile compared to the other soil-crop models DNDC and DayCent. The performance of STICS to predict SMN content in topsoil was similar to the one predicted by Jing et al. (2017) who simulated timothy growth under mineral and organic fertilization with STICS. STICS performed as well in our study to simulate cropping systems using digestates and effluents than in other studies.

Few other soil-crop models were evaluated under cropping systems using EOMs. Li et al. (2015) evaluated DSSAT-CSM model, on SOC, SON, and yield, one treatment being fertilized with manure. Wheat yield under manure fertilization was predicted with a rRMSE of 22%, which was consistent to our findings with STICS. Begum et al. (2017) evaluated DailyDayCent on Rothamsted site, with manure additions. Yields were correctly simulated (rRMSE ranged from 30 to 40%), which is still a less good prediction than in the present study. With CERES-EGC, Noirot-Cosson et al. (2016) obtained good fits for yields (rRMSE ranged from 7 to 27%) and N exports (rRMSE ranged from 16 to 35%); however SMN was less accurately predicted (rRMSE > 100%). Räbiger et al. (2020) developed a plant-soil-atmosphere model to simulate N leaching under rapeseed, wheat, and barley fields fertilized with digestates. The mineralization rate of digestate was calibrated on field data, no ammonia volatilization nor N_2O emission were modelled. SMN in topsoil was similarly simulated in the present study compared

to the modelling of Räbiger et al. (2020) (r^2 ranged from 0.08 to 0.38, RMSE ranged from 16 to 30 kgN ha^{-1}).

Here, we studied the sensitivity of the outputs in response to a change in inputs associated to EOM characteristics. We concluded that the TAN content of EOM were the main parameter of interest when considering crop growth and N losses. The ammonia volatilization was also sensitive to application date and DM content of the EOMs. Variations of C content or $C:N_{org}$ content could however have impacts at longer-term, e.g. on SOC content. A more sophisticated global sensitivity analysis would give more insight in the EOM characteristics interacting with each other (Monod et al., 2006). Such analysis would also benefit to investigate the interaction between EOM characteristics and diverse soil and climate conditions.

Compared to the literature, we concluded that STICS provided a good representation of EOM. We argued that STICS is able to simulate a crop rotation based on fertilization with EOMs, if the ammonia volatilization model is changed. The main parameters that should be assessed precisely are the TAN contents of EOM applied during crop growth. The mineralization of EOM can be parametrized, but the default values should lead to consistent simulations. Slight differences between two similar EOMs may not lead to different simulation outputs.

4.4.4 Using STICS to simulate cropping system with organic fertilization

STICS performance allowed comparing N fluxes including ones not measured in the field experiment. It included N_2O emissions during the whole cropping season, nitrate leaching, and the SOM storage. The use of the simulation together with the field experiment strengthened our conclusion on the use of digestates as fertilizers.

Compared to synthetic fertilizers, the use of EOM increased the N content in the soil. This effect was already highlighted (Bhogal et al., 2018). The use of EOMs may also increase ammonia volatilization depending of their TAN contents. Here, the nitrate leaching was only increased due to the application of EOMs just before wheat sowing, consistently to the findings of Sieling et al. (1997). The effects of the use of EOM on N_2O emissions were contradictory. The importance of N_2O emissions during summer is crucial. EOMs can bring N to the soil during summer, and could lead to high N_2O emissions because of high temperature and when the soil is moist enough. However, if the soil is too dry, the application of EOM in summer can have very low effect on N_2O emissions. Indeed, soil moisture and temperature are the two main drivers of N_2O emissions (Butterbach-Bahl et al., 2013). Both low (Parnaudeau et al., 2009; Pezzolla et al., 2012) and high (Bell et al., 2015, 2016) N_2O emission during summer after EOM application have been reported in the literature and this question could be further investigated.

The use of digestate instead of cattle effluents induced a higher TAN input, and therefore higher yields simultaneously to higher N losses (NH_3, N_2O). This was consistent with the findings in the literature,

although the different effects were often studied independently (e.g., Möller, 2015; Möller and Müller, 2012; Nicholson et al., 2017). The use of raw digestate instead of liquid and solid digestates increased the ammonia volatilization, although it was not well simulated. Raw digestates also induced lower input of organic matter in the field.

4.5 Conclusion

In this study, we calibrated and evaluated the STICS model on a field experiment using organic fertilizers and amendments, including cattle effluents and digestates. We investigated the role of the parametrization of EOM mineralization submodule, and the role of the choice of EOM volatilization submodule. We showed that STICS performed reasonably well to predict plant yield and N content at harvest, and SMN content. The choice of the parametrization method for the EOM mineralization submodule did not affect the ability of STICS to simulate a consistent crop rotation, despite the fact that the absolute value of the outputs can be impacted. The submodule of STICS predicting ammonia volatilization from EOM underestimated volatilization, especially for solid EOMs or raw digestates, ALFAM2 model performed better (for liquid EOMs). A reasonable uncertainty on EOM characteristics should not affect importantly STICS outputs. Further use of STICS should include the effect of using digestates with different characteristics, at different doses, or on different crops. STICS could thus be used to estimate impacts that are difficult to measure (e.g. N_2O emissions, leaching, volatilization, C storage) at a longer-term, especially with digestate for which long-term experiments are still scarce.

Acknowledgement

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Chapitre 5
Évaluation du modèle de méthanisation SYS-Metha à l'aide des données opérationnelles d'un méthaniseur traitant des déchets variés

Le Chapitre 4 a permis de montrer que le modèle sol-plante STICS, associé au modèle de volatilisation ALFAM2, possède des capacités de prédiction suffisante pour être utilisé comme outil d'évaluation environnementale de pratique agronomique. Comme présenté au Chapitre 1, l'impact environnemental de la méthanisation dépend des effets de l'épandage de digestat au champ, mais également de l'effet des impacts du traitement des déchets, au niveau de la plateforme de méthanisation. Le but de cette thèse est ainsi d'étudier en quoi un changement de pratique lors du traitement des effluents bovins peut avoir un effet sur la qualité agronomique des digestats et avoir des effets indirects sur les effets au champ. Pour atteindre ce but, un modèle de prédiction de la qualité des digestats est nécessaire et doit également être évalué. Dans ce Chapitre 5, nous présentons un modèle de méthanisation simple, SYS-Metha (UR OPAALE, INRAE, Rennes), prédisant les caractéristiques des digestats, les productions de biogaz, et les émissions gazeuses lors du stockage des digestats. Nous évaluons ce modèle sur le cas d'étude du méthaniseur de Nouzilly. Ce Chapitre est écrit dans le format d'une publication scientifique, mais il n'a pas vocation à être publié. Le travail présenté dans ce chapitre a participé à un travail plus large d'évaluation de SYS-Metha sur plusieurs méthaniseurs, coordonnées par les membres de l'UR OPAALE, qui a fait l'objet d'une publication scientifique.

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Evaluation of the SYS-Metha model of anaerobic digestion on a territorial digester processing a large diversity of organic wastes

Victor Moinard[1], Younès Bareha[2], Romain Girault[2] Jean-Marie Paillat[3,4], Sabine Houot[1]

1: UMR Ecosys, Université Paris Saclay, INRAE, AgroParisTech, Thiverval-Grignon, France

2: UR OPAALE, INRAE, Rennes, France

3: UPR Recyclage et risques, CIRAD, Montpellier, France

4: UPR ADI-Suds, ISTOM, Angers, France

Abstract

Farms equipped with anaerobic digestion benefit from the production of digestates as organic fertilizers. The physico-chemical characteristics of digestates influence both their fertilizing potential, as well as their associated environmental risks. SYS-Metha is a simple anaerobic digestion model that simulates the biogas production, the gaseous emissions during storage, and the digestate characteristics from operational data. The purpose of this study was to evaluate the model on a territorial digester processing multiple agricultural and urban inputs. Operational data were obtained from the managers of the anaerobic digestion plant. We realized a mass balance to assess the consistency of the dataset, and selected an adequate period to evaluate SYS-Metha during stationary functioning. A period of 56 weeks was selected, with homogeneous inputs, biogas production, hydraulic retention time (HRT), and consistent analyses of the digestate. During digestion, the export of digestate fitted the predicted production of digestate from inputs and biogas production, with a slight overestimation of 12%. The phase separation and storage were found conservative for P and mineral matters. During this period, SYS-Metha consistently simulated flows of nutrients and digestate characteristics (relative errors often lower than 20%). The simulated biogas production was 61% higher than measured biogas consumption. Slight adjustments in some input characteristics led to decrease this overestimation, with a predicted biogas production that was 41% higher than measured biogas consumption. The digestate characteristics were simulated reasonably well. Considering its simplicity and assumptions, the simulation of the anaerobic digester by SYS-Metha was considered as successful. This work has been integrated in a broader scientific publication aiming at evaluating SYS-Metha in regards to twelve digesters. SYS-Metha is a promising simple tool for further works aiming at evaluating the environmental performance of anaerobic digestion.

Keywords

Anaerobic digestion; mass balance; storage; modelling; digestate; emissions.

5.1 Introduction

On-farm anaerobic digestion is developing in France. Farms equipped with anaerobic digestion produce energy and benefit from the production of digestate as organic fertilizer. The physico-chemical characteristics of digestate influence both its fertilizing potential, as well as its associated environmental risks (Chapter 1, Chapter 3). Moreover, anaerobic digestion is subject to biogas leakage (Hrad et al., 2015) as well as gaseous emissions during storage of wastes and digestates (e.g., CH_4, N_2O, NH_3) (Baldé et al., 2018, 2016; Daniel-Gromke et al., 2015), which influence the environmental performance of anaerobic digestion (e.g., Hijazi et al., 2016; Vaneeckhaute et al., 2018). The modelling of digestate characteristics and emission during storage could be a relevant tool to evaluate the environmental impacts of anaerobic digestion.

During the last decades, the scientific community developed a diversity of anaerobic digestion models (Lauwers et al., 2013). Among them, ADM1 (Batstone et al., 2002) is a complex mechanistic model which has been the most used in the scientific community. Numerous contributions developed "modified ADM1 models" to include new chemical process inside the digester (Lauwers et al., 2013). One major issue of these mechanistic complex models is the need to deeply characterize the inputs (Fisgativa et al., 2018; Girault et al., 2012; Jimenez et al., 2016; Li et al., 2021), and often to calibrate some parameters of the model to consistently simulate the observed data (Donoso-Bravo et al., 2011). Other modelling approaches included simpler mechanistic models based on first-order kinetics (Ericsson et al., 2020; Linke et al., 2013; Muha et al., 2015), or the use of data based models such as artificial neural network (González-Fernández et al., 2011). All those models were primarily used to predict biogas production at a fine time scale in order to improve the digestion process (Lauwers et al., 2013), to evaluate the co-digestion of diverse effluents (Mata-Alvarez et al., 2014), or to evaluate the benefits of the pre-treatment of inputs (Esposito et al., 2012). Other simple anaerobic digestion models have also been developed to easily predict the biogas production or the economical consequences of anaerobic digestion at the farm level (Kythreotou et al., 2014). Such models often focused on very specific types of plants or geographic area, and they rarely consider environmental assessment, or quality of digestate (Kythreotou et al., 2014).

With the increasing interest for the environmental performance of anaerobic digestion, the anaerobic digestion models were sometimes used to predict the pollutants or greenhouse gas emissions from digestate during storage, or to predict the digestate characteristics. Ericsson et al. (2020), Linke et al. (2013) and Muha et al. (2015) used simple mechanistic models to predict methane emissions from raw digestate storage. Vergote et al. (2019) modified ADM1 to predict biogas emission from digestate during and after digestion. They explored the impact of hydraulic retention time (HRT), temperature, and frequency of digestate removal on biogas emissions during storage. To our knowledge, no model predicting the gaseous emissions from solid digestate originating from solid/liquid phase separation has

been developed. Many composting models has been developed, which could sometimes predict gaseous emissions (Walling et al., 2020). They could be adapted for the modelling of solid digestate storage. Some authors focused on the prediction of digestate characteristics. Although ADM1 was initially not focusing on digestate quality (Lauwers et al., 2013), volatile solid (VS) content, total ammoniacal nitrogen (TAN) content, or pH can be predicted by some versions of ADM1 (Donoso-Bravo et al., 2020; Galí et al., 2009), because they are linked to biogas production. More recently, Bareha et al. (2018) and Fisgativa et al. (2020a) investigated the mineralization of organic N during digestion. The prediction of the stable fraction of organic matter in digestate has also been investigated, which could be used in the future to predict the digestate capacity at contributing to carbon storage in soils (Jimenez et al., 2015). Finally, some other types of models were specifically dedicated to provide information for life cycle assessment (LCA) (Gentil et al., 2010). Among them, the ORWARE model aimed at predicting environmental impacts of diverse waste treatments, including anaerobic digestion (Carlos-Pinedo et al., 2020; Dalemo et al., 1997; Eriksson et al., 2002). Such models consider a large spatial scale and is relevant for environmental assessment, e.g. to compare different waste treatments. However, it relies on many hypothesis that are sometimes difficult to verify, and does not aim at exploring the effect of different processes on the environmental performance of anaerobic digestion. ORWARE assumptions are for example based on a single plant, based in Sweden (Eriksson et al., 2002).

SYS-Metha (Bareha et al., 2021b, 2021a) is a recently developed anaerobic digestion model (continuous stirred tank reactor, mesophilic conditions). It is based on a mass balance approach, to predict biogas production and digestate characteristics from a waste database including physico-chemical characteristics, potential biomethane production and N mineralization. The model aims to be a robust tool to predict digestate characteristics, and gaseous emissions during storage using emission factors. The advantages of SYS-Metha compared to the previously presented models are 1) the ability to simulate different initial mixtures with a broad waste description, 2) the simplicity of its input data, and 3) the consideration of all major gas emissions (NH_3, CH_4, CO_2, N_2O) during all steps of the process. Therefore, it could be used for environmental assessment, considering different initial mixtures used for anaerobic digestion. The aim of this study was to evaluate the ability of the SYS-Metha model to predict digestate characteristics for an agricultural digester with large external inputs, using the operational dataset from the plant manager. This work has been included in a broader evaluation of SYS-Metha model (Bareha et al., 2021b).

5.2 Material and Methods

5.2.1 Description of SYS-Metha, mass balance model of anaerobic digestion

The SYS-Metha model (Bareha et al., 2021a, 2021b) aims at predicting digestate characteristics, biogas production, and gaseous emissions during digestate storage, in an anaerobic digestion plant, based on a simple operational dataset. It is a mass balance model. The model assumes a stationary functioning, and a wet mesophilic process. The data needed to run SYS-Metha model are the inputs of the digester (masses of fresh weight), the hydraulic retention time (HRT), information on digestate phase separation (screw press, centrifugation, or none), and about the cover or not of liquid or raw digestate tanks. The model predicts the production of CO_2 and CH_4 in the biogas, the quantity of digestate produced, the total solid (TS), VS, carbon (C), total nitrogen (TN), and TAN contents in digestates for all steps (with or without phase separation, before and after storage), as well as gaseous emissions during storage (NH_3, N_2O, CO_2, CH_4). Briefly, it is constituted of 5 steps.

Step 1) All inputs have to be listed in an organic waste database in which the characteristics of 48 types of inputs are described by their contents in TS, VS, C, TN, TAN. Each input is also characterized by its bio-methanogenic potential (BMP, in Nm^3CH_4 tTS^{-1}) which represents the quantity of methane that could be emitted, and the CH_4:(CH_4+CO_2) volumetric ratio inside biogas produced during the digestion of the input . Each input is also characterized by its apparent nitrogen mineralized potential (ANMP, in $gN\text{-}NH_4^+$ $kgTS^{-1}$), which represents the quantity of organic N that could be mineralized under ammoniacal N form during input digestion. ANMP considers organic N mineralization, biomass growth and biomass decay. BMP, CH_4:(CH_4+CO_2) ratio, and ANMP are computed as described and defined in Bareha et al. (2018). Standard errors are associated to each characteristics.

Step 2) For each input, CH_4 and CO_2 production as well as organic N mineralization are computed. CH_4 production is computed as:

(equation 5.1) $CH_4 = TS \,.\, BMP \,.\, PMR_{BMP}$

where CH_4 is the methane produced (Nm^3), TS is the quantity of total solid within the input in the digester (tTS). PMR_{BMP} (potential mineralization rated obtained) is a factor varying from 0 to 1.05 used to adjust BMP in function of HRT, BMP, and TS and C contents in inputs. PMR_{BMP} is thus an increasing empirical function of HRT (Bareha et al., 2021b). CO_2 production is deduced from CH_4 production and CH_4:(CH_4+CO_2) ratio. TN, water, mineral matter (MM) of each inputs are conserved during digestion. The C emitted as biogas is removed (mass balance). VS is degraded with the same proportion as C. It induces a slightly higher mass of outputs (digested matter and gas) compared to inputs, because the C content inside the volatile solid is different in the inputs and in the biogas. Additionally, the mineralization of organic N is computed as:

(equation 5.2) $N_{org \rightarrow amm} = N_{org} \cdot ANMP_{\%Norg} \cdot PMR_{ANMP}$

$N_{org \rightarrow amm}$ (tN) is the total mass of organic N mineralized into ammoniacal N. N_{org} is the initial mass of organic N from the input in the digester (tN). $ANMP_{\%Norg}$ is ANMP expressed in percentage of organic N in the input. Like PMR_{BMP}, PMR_{ANMP} is a factor used to adjust ANMP in function of HRT, ANMP, and TAN and TN contents in inputs. PMR_{ANMP} is thus an increasing empirical function of HRT (Bareha et al., 2021b).

Step 3) The biogas production is computed by summing the biogas produced by each input. The biogas production can be used to predict the electric power of the plant, assuming that the generator runs 8000 h $year^{-1}$ with an energy yield of 0.38 (kW electricity per kW of primary energy). The raw digestate production is computed by mass balance, summing the elements brought by each input, considering the mass loss in biogas. There is no interactions between the inputs. The TS, VS, C, TN, and TAN contents in raw digestate are also computed by mass balance, considering inputs characteristics, biogas production, and mineralization of organic N.

Step 4) After phase separation, the characteristics and quantities of solid and liquid digestates are computed using separation efficiency ratio for each element (fresh weight (FW), TS, VS, C, TN, TAN). These ratios come from (Guilayn et al., 2019b). Phase separation is conservative for all elements.

Step 5) During digestate storage, the simulated processes are the anaerobic and aerobic degradation of organic matter that emit CO_2 and CH_4 (solid, liquid, and raw digestates), the organization of ammoniacal N that affect TAN:TN ratio and emit N_2 and N_2O from solid digestate, and the volatilization of NH_3 from solid, liquid, and raw digestates. Storage is modelled using degradation ratios and gaseous emission factors, described in Table 5.1. Considering solid digestate, organic matter is decomposed, leading to a decrease of TS, VS, and C contents. 1% of the lost C is emitted under CH_4 form, the other part being CO_2. Part of the TN is also lost through emissions of N_2, N_2O, NH_3. 51% of TAN is lost or organized: 26% is emitted under NH_3 form, 8% is emitted under the N_2O form, and the last part (17%) is organized or lost under other forms such as N_2. Considering liquid and raw digestate, organic matter is also decomposed, leading to a decrease of TS, VS, and C contents. The emission factors were similar with or without storage cover. The lost C was emitted under CH_4 and CO_2 forms, with the same ratio as the one found in the digester. The NH_3 volatilization depends on the presence of storage cover. Organic N was conserved, N organization, N mineralization, and other N emissions, including N_2O, were assumed to be null. For solid, liquid, and raw digestates, total mass of digestate is deduced from initial mass and losses.

SYS-Metha provides uncertainties associated to each outputs, coming from the uncertainties of wastes characteristics.

Table 5.1 – Degradation ratios and gaseous emission factors used in SYS-Metha to simulate the digestate transformation during storage.

Reference	Liquid digestate	Raw digestate	Solid digestate
Water loss ($kgH_2O\ kgH_2O^{-1}$)	0	0	0
TS loss ($kgTS\ kgTS^{-1}$)	0.08	0.05	0.22
TN loss ($kgTN\ kgTN^{-1}$)	equal to NH_3 emission	equal to NH_3 emission	0.09
TAN loss ($kgTAN\ kgTAN^{-1}$)	equal to NH_3 emission	equal to NH_3 emission	0.51
VS loss ($kgVS\ kgVS^{-1}$)	0.13	0.07	0.30
C loss ($kgC\ kgC^{-1}$)	0.15	0.08	0.34
NH_3 emissions (with cover) (% initial TAN)	9%	7%	26%
NH_3 emissions (without cover) (% initial TAN)	44%	36%	26%
N_2O emissions (% initial TAN)	0	0	8%
CH_4 emissions	Similar proportion of C_{loss} as in the biogas	Similar proportion of C_{loss} as in the biogas	1% C_{loss}

TS: total solid. TN: total N. TAN: total ammoniacal N. VS: volatile solid.

5.2.2 Case study: an agricultural digester with high external inputs

The present case study is based on an agricultural anaerobic digester located in the Centre – Val de Loire region of France. A continuous wet mesophilic process (continuous stirred-tank reactor) is performed, with one main digester (1600 m^3) and one post digester (700 m^3). The digester has the capacity to treat 12,000 t of waste per year. However, recently, it only treated 7,500 t of wastes per year, consisting in cattle slurry (11%), cattle, sheep, and horse manures (17%), sewage sludge (29%), agroindustrial wastes (18%), grease (8%), and cereal middlings (8%). The remaining inputs consisted in site water runoff (9%) and other wastes (e.g. grass silage, beet pulp). The biogas is used to produce electricity with a combined heat and power unit. For this study, the retention time was 100 days. The raw digestate is post-treated by phase separation using a screw press. The liquid phase is stored in an open lagoon, the solid phase is stored outdoor, for about six months. They were sampled during their application in the field. We sampled the raw digestate immediately after going out of the post-digester (no storage).

We interviewed the managers of the digester in order to acquire operational management data. The following dataset used in this study was obtained from 2017 to 2019: biogas consumption (volume, proportion of CH_4 and CO_2, on a weekly time step), digester inputs (weekly time step), digestate exports (mass of solid and liquid digestates, on a weekly time step). From 2017 to 2019, digestates were sampled and analyzed regularly. Overall, the raw digestate was analyzed on 7 dates (22 March 2017, 19 April 2017, 21 March 2018, 2 August 2017, 11 January 2019, 19 February 2019, 12 March 2019), liquid digestate on 7 dates (8 February 2017, 22 March 2017, 19 April 2017, 21 March 2018, 11 January 2019, 19 February 2019, 12 March 2019), solid digestates on 4 dates (8 February 2017, 2 August 2017, 19 September 2019, 11 January 2019). The following digestate characteristics were analyzed in a specialized laboratory (Aurea, Ardon, France): TS, TN, TAN, VS, P, and K contents. C content was assumed equal to half of VS content. Mineral matters (MM) were computed as the difference between TS and VS. Those analysis included the one from Chapter 3 and additional analyses given by the managers of the anaerobic digester and the farm.

5.2.3 Investigating periods of stationary functioning of the digester from operational data to evaluate the model

As described in section 5.2.1, SYS-Metha assumed a stationary functioning of the digester. Therefore, a consistent period of time had to be found, during which this assumption is probable, to correctly evaluate the model. First, we identified candidate periods by analyzing the following variables: weekly mass inputs, weekly biogaz production and HRT. We identified candidate periods, where those three variables were as constant as possible. Moreover, the candidate periods had to be positioned between two exports of liquid digestate. Indeed, liquid digestate was stored continuously without indication on production rate, until spreading period, which occurred twice, or three times a year. After a spreading period, we assume that the lagoon is close to empty. This assumption enabled to assess absolute mass of liquid phase digestate production. The solid phase of digestate is exported regularly every two weeks, thus no constraints were needed to assess its export. HRT (days) was computed every four weeks with the equation:

(equation 5.3) $$HRT = \frac{V}{F} = \frac{V}{\frac{m}{\rho \cdot \Delta t}} = \frac{V \cdot \rho \cdot \Delta t}{m}$$

where V is the volume of digester and post-digester ($V = 1600 + 700\ m^3 = 2300\ m^3$), F is the input flux ($m^3\ d^{-1}$), Δt is the considered period (here, $\Delta t = 4$ weeks = 28 days), m is the mass of inputs (t_{FW}) during the time Δt, and ρ is the mean density of inputs, which is assumed to be equal to water density ($\rho = 1\ t_{FW}\ m^{-3}$).

5.2.4 Mass balance of the digester based on the operational dataset

We realized a mass balance of the digestion step and post-treatment step during the candidate periods, in order to select the period with the most consistent operational data for SYS-Metha evaluation. During

the digestion step, due to the diversity of inputs (26 denominations) and the lack of chemical analyses of inputs, only the fresh weight mass balance was assessed. This step was useful to evaluate the consistency of inputs, biogas production, and digestate exports. We evaluated the production of raw digestate both from the inputs and the outputs:

(equation 5.4) $m_{RD}^{in} = m_{inputs} - m_{biogas}$

(equation 5.5) $m_{RD}^{out} = m_{LD} + m_{SD}$

Where m_{RD}^{in} is the mass of raw digestate estimated from inputs (ton of fresh weight tFW), m_{RD}^{out} is the mass of raw digestate estimated from outputs (tFW), m_{inputs}, m_{biogas}, m_{LD}, m_{SD} are the mass of inputs, biogas, exported liquid digestate, and exported solid digestate, respectively (tFW). m_{biogas} was estimated from the consumed volume of gas using the ideal gas law. We assumed that biogas was only constituted by CO_2 and CH_4, and that all biogas was consumed:

(equation 5.6) $m_{biogas} = m_{CH_4} + m_{CO_2} = \frac{P \cdot M_{CH_4} \cdot V_{CH_4}}{R \cdot T \cdot 10^6} + \frac{P \cdot M_{CO_2} \cdot V_{CO_2}}{R \cdot T \cdot 10^6}$

where P is the gas pressure in normal condition (c.a. 101325 Pa), R in the molar gas constant (c.a. 8.314 J K^{-1} mol^{-1}), T is the temperature in normal conditions (273.15 K), M_{CH_4} and M_{CO_2} are the molar masses (g mol^{-1}) of CH_4 ad CO_2, respectively, and V_{CH_4} and V_{CO_2} are the consumed volume (Nm^3) of CH_4 and CO_2, respectively.

We used the analyses of digestates to verify the consistency of TS, VS, C, MM, TN, TAN, P, and K mass balances during post-treatment (phase separation and storage), and to evaluate the consistency of these analyses. For both candidate periods, digestates were assumed to have constant average physico-chemical characteristics, computed as the average characteristics of digestates analysis realized during each period. It was a moderate assumption, because digestate characteristics were globally homogenous during the three years (Chapter 3, data not shown). We used the mass balance approach described by Guilayn et al. (2019b). First, we computed the ratio of liquid fraction after phase separation (R_{LF}) with the following equation :

(equation 5.7) $R_{LF} = \frac{m_{LF}}{m_{SF} + m_{LF}}$

Where m_{LF} and m_{SF} are the masses of the exported liquid and solid digestates, respectively (tFW). This equation assumed that the exported liquid and solid digestates correspond to the same period of production. The ratio of solid fraction is computed as:

(equation 5.8) $R_{SF} = 1 - R_{LF}$

It was then possible to estimate errors in mass distribution ($Error_{MD}$) on an element X, which was the excess ($Error_{MD} > 0$) or deficit ($Error_{MD} < 0$) of the element X found in post-treated digestates (solid and

liquid), compared to raw digestate. It was expressed in percentage of the flow of element X in the raw digestate. We computed $Error_{MD}$ with equation 5.9:

(equation 5.9) $$Error_{MD} = \frac{R_{SF} \cdot [X]_{SF} + R_{LF} \cdot [X]_{LF}}{[X]_{RF}} - 1$$

where $[X]_{SF}$, $[X]_{LF}$, and $[X]_{RF}$ were the content of element X (kgX tFW^{-1}) in solid, liquid, and raw fractions, respectively. The following elements were investigated: DM, VS, C, MM, TN, TAN, P, and K. Theoretically, the perfect conservation of an element during phase separation and storage would induced a null $Error_{MD}$. Guilayn et al. (2019b) considered that $Error_{MD}$ lower than 10% were small and indicated a conservative mass balance. Here, we mainly verified the conservation of MM, P, and K to assess the quality of digestate analysis, because those elements should not be degraded or lost during phase separation and storage.

The interpretation of the mass balances at both digestion and post-treatment steps enabled the selection of a single study period, during which we evaluated SYS-Metha.

5.2.5 Evaluation of the SYS-Metha model to simulate biogas production, digestate production, and digestate characteristics

To evaluate SYS-Metha, the operational data (inputs, biogas production, digestate export, digestate analyses) were restrained to only one study period. Because they were not present in the SYS-Métha database, some waste inputs had to be grouped in the simulation. Thus, 15 types of inputs were considered in the model: mixed liquid sludge (38% of inputs, included liquid sewage urban sludge and some liquid effluent from agro-industry), cattle slurry (11%), waste from cereal storage (9%), solid secondary sewage sludge (8%), ovine manure (8%), cattle manure (7%), grease (7%, included solid and liquid grease), brown water (6%), equine manure (3%), grass silage (2%), whey permeate (1%), corn silage, vegetable wastes, cattle feed waste, waste from yeast production (<1% each). The absolute mass of inputs were proportionally adjusted from operational data to correspond to one year of functioning. HRT was set to 100 days, as indicated by managers.

We first ran the model using the model database, without any calibration step. We then evaluated a second version of the model, called adjusted model to determine if adjustment of the waste characteristics enabled to improve model performances. In the adjusted model, we performed the following changes:

- The DM content of cattle slurry was adjusted from 9% to 6%, based on measured data. VS, C, TN, TAN content, expressed in g $kgTS^{-1}$, remained unchanged, as well as BMP, ANMP, and CH_4:CO_2 ratio in biogas.
- Based on data collected from the managers, DM content of grease were also adjusted from 36% to 17%, similarly to slurry. The BMP of the grease was adjusted, from 975 Nm^3 tTS^{-1} to 359

Nm^3 tTS^{-1}. Indeed, the grease included in the database is solid grease, whereas the input greases were a mix of liquid and solid ones.

- The CH_4:(CH_4+CO_2) volumetric ratio of the waste from cereal storage was adjusted from 58% to 45%, which corresponded to the ratio from other cereal residues in the database. To conserve the C degradation from the cereal wastes, we adjusted its BMP from 450 Nm^3 tTS^{-1} to 350 Nm^3 tTS^{-1}. This last value was more consistent with the BMP analyzed by the managers of the anaerobic digester for one of the main type of cereal waste (c.a. 300 Nm^3 tTS^{-1}). Indeed, the correspondence between real cereal wastes inputs and cereal waste described in the database was uncertain. Moreover, this waste produced half of methane from the digester, being a very sensitive waste that had to be calibrated to obtain a consistent biogas composition.

The following outputs were compared to operational data: raw digestate characteristics before storage, liquid and solid digestate characteristics after storage, total fluxes of FW, DM, VS, C, TN, and TAN exported from the digester, biogas production and electrical power of the cogeneration facility. We evaluated the model by computing, for each variable of interest, the model error and relative error:

(equation 5.10) $error = S - O$

(equation 5.11) $relative\ error = \frac{S-O}{O}$

where S is the simulated value, and O the observed value.

5.3 Results

5.3.1 Operational data: assessment of data consistency and selection of study period

In 2017 and early 2018, we observed some variations of inputs, biogas production, and HRT (Figure 5.1). Two candidate periods were selected. The first one strictly respected the stationary conditions; it lasted from 24th of December 2018 to 7th of April 2019 (15 weeks). The number of analyses during that period were 2 for liquid digestate, 1 for solid digestate, and 2 for raw digestate. The second candidate period lasted from 20th of August 2018 to 15th of September 2019 (56 weeks). It respected less strictly the stationary functioning; however, it integrated a longer period and more digestate analyses (3 for liquid phase, 2 for solid phase, and 3 for raw digestates).

Despite the variation of inputs mass and biogas production, the proportions of the different categories of inputs were homogeneous during the three years (data not shown) and the digestate characteristics did not vary extensively (Table 5.2).

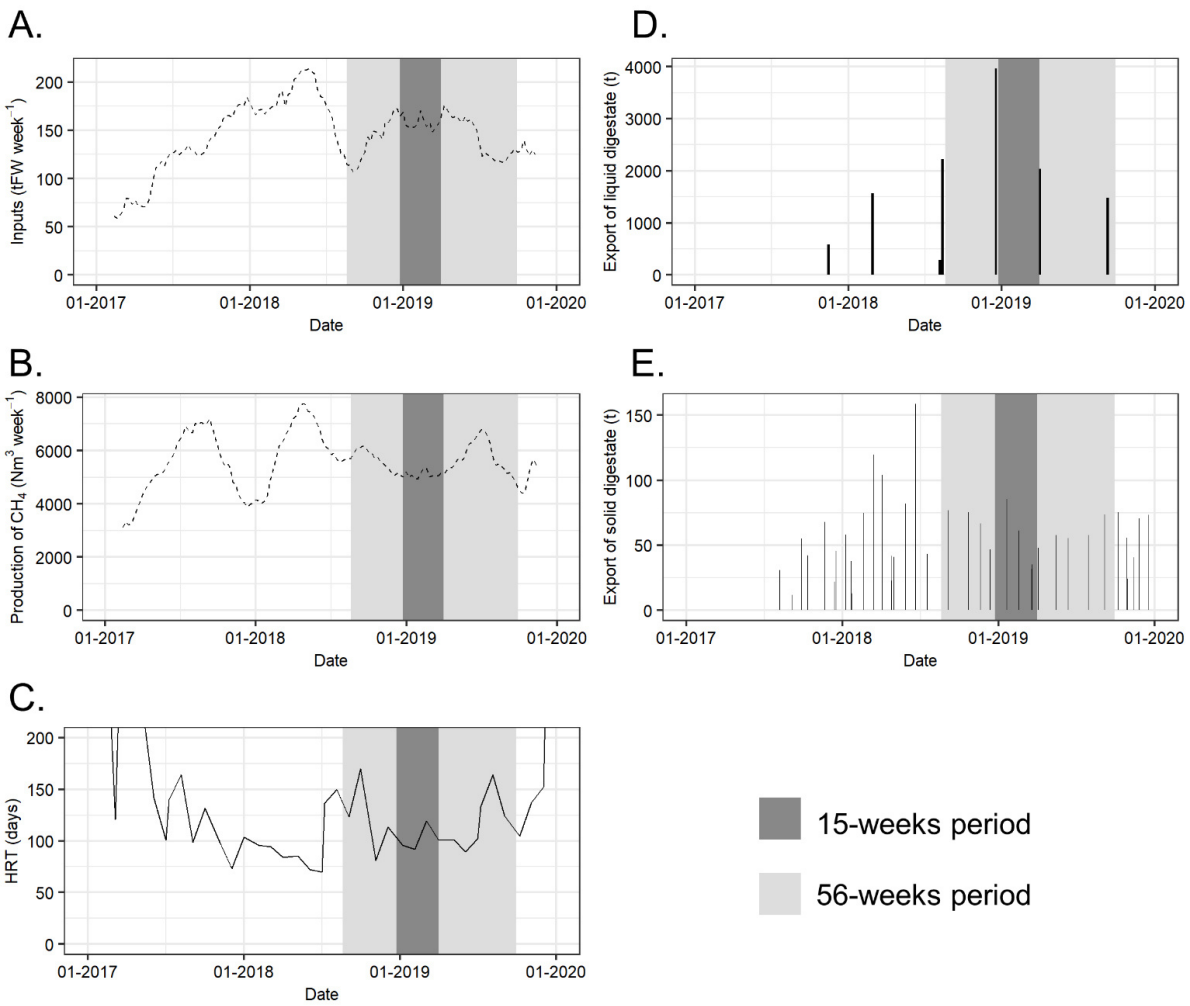

Figure 5.1 – Investigation of period of stationary functionary using operational data from the digester. The 15-weeks candidate period (dark grey) and 56-weeks candidate period (light grey) were indicated in each subfigure. A) Weekly inputs in the digester. Dotted line represented the moving average of inputs along the 13 weeks. B) Weekly biogas consumption. Dotted line represented the moving average of biogas consumption along the 13 weeks. C) HRT, computed every 28 days. D) Exports of liquid digestate (fresh weight). E) Exports of solid digestates (fresh weight).

Table 5.2 – Digestate characteristics during the three years, and during the two candidate periods. Variation coefficient are indicated between brackets. VS, TAN, TN, P, and K content are given in g per kg of fresh weight.

Period	Digestate	Number of analyses	DM (%FW)	VS (g kg^{-1})	TAN (g kg $^{-1}$)	TN (g kg $^{-1}$)	P (g kg $^{-1}$)	K (g kg $^{-1}$)
15 weeks	raw	2	6.1 (1%)	43 (<1%)	3.0 (38%)	4.5 (3%)	0.8 (1%)	4.0 (7%)
	liquid	2	4.7 (7%)	29 (3%)	2.2 (14%)	4.4 (6%)	0.8 (22%)	3.7 (21%)
	solid	1	19	152	0.8	6.1	2.8	3.4
56 weeks	raw	3	6.6 (14%)	46 (11%)	3.0 (28%)	4.8 (13%)	0.9 (26%)	4.1 (8%)
	liquid	3	4.7 (7%)	29 (3%)	2.2 (14%)	4.4 (6%)	0.8 (2%)	3.7 (21%)
	solid	2	21 (15%)	174 (17%)	1.3 (54%)	6.6 (9%)	2.8 (3%)	3.3 (5%)
3 years	raw	7	6.6 (9%)	45 (9%)	2.6 (25%)	4.6 (11%)	1.0 (17%)	3.6 (21%)
	liquid	7	4.8 (20%)	29 (17%)	2.2 (14%)	4.4 (13%)	0.9 (32%)	3.4 (26%)
	solid	4	24 (20%)	203 (24%)	1.1 (57%)	6.4 (6%)	2.5 (12%)	3.3 (17%)

Table 5.3 – Mass balance during the two candidate digestion periods.

	15-weeks period	56 weeks period
m_{inputs} (tFW)	2488	8105
m_{biogas} (t)	207	755
m_{RD}^{in} (tFW)	2281	7330
m_{LD} (tFW)	2040	7490
m_{SD} (tFW)	261	772
m_{RD}^{out} (tFW)	2301	8262
Mass balance error (t) $m_{RD}^{in} - m_{RD}^{out}$	-20	-932
Relative mass balance error (%) $2\frac{m_{RD}^{in} - m_{RD}^{out}}{m_{RD}^{in} + m_{RD}^{out}}$	-1%	-12%

FW: fresh weight. LD: liquid digestate. RD: raw digestate. SD: solid digestate

Table 5.4 – Errors in mass distribution ($Error_{MD}$, %) during phase separation, for each element. Results are reported as the mean ± one standard error. During the 15-week period, uncertainties were unavailable (not enough digestate analysis).

	Period 15 weeks	56 weeks
TS	4.2	-5.0 ± 8.6
VS	0.6	-7.1 ± 8.8
MM	13.1	0.4 ± 10.2
TN	4.1	-3.6 ± 10.6
TAN	-34.0	-31.2 ± 14.9
P	27.2	4.7 ± 14.0
K	-7.8	-11.9 ± 11.8

The fresh mass balance was perfectly closed during the 15-weeks period (Table 5.3). During the 56 weeks period, there was more export of digestates than predicted from inputs and biogas production. The order of magnitude of the difference was 12%. The possible variations of stocks of digestates inside the lagoon or the digester between the beginning and the end of the period could explain the difference.

The ratio of liquid fraction R_{LF} was similar when considering both periods, and was 0.886 for the 15 weeks period, and 0.907 for the 56 weeks period. Errors in mass distribution are indicated in Table 5.4. Because the number of analysis were low, $Error_{MD}$ was associated to high uncertainties. During the 56-week period, MM and P balances were close to 0: those two elements were probably better conserved during phase separation and storage. This result indicated a good consistency between raw digestates and liquid and solid digestates analysis. In that regard, the mass balance based on the 15-week period was of lower quality, with high surplus of MM and P, without estimated uncertainty. DM, VS, TN, and K were conserved for both candidate periods. We observed a loss of TAN, which could be explained by N organization, or gaseous N losses (NH_3, N_2O, N_2).

Because of the good conservation of P and MM during phase separation and storage (Table 5.4), and despite larger error in fresh mass balance during digestion (that remained reasonable) (Table 5.3), the 56-week period was selected to evaluate SYS-Metha. During this period the mean HRT was 115, which was consistent with the value of 100 days used in SYS-Metha. The characteristics of digestates used for SYS-Metha evaluation were the average of the characteristics measures during the 56-week period.

5.3.2 Evaluation of SYS-Metha: fresh mass balance, nutrient flows, and digestate characteristics

The model was evaluated on the 56-week period. The simulated organic load rate was 1.8 kgVS m^{-3} d^{-1} in the unadjusted model, and 1.6 kgVS m^{-3} d^{-1} in the adjusted model. The unadjusted model overestimated by 61% the biogas production (439 t). The proportion of CH_4 inside biogas was also overestimated. The adjusted model still overestimated the biogas production by 40%, however the proportion of CH_4 in biogas was better simulated.

The simulated amount of produced raw digestate were well estimated (Table 5.5). The simulated amounts of exported liquid and solid digestates were underestimated from 12% to 15% for each phase (adjusted and unadjusted model), compared to observed data (Table 5.5). This error was in the same order of magnitude as the mass balance error in fresh weight (Table 5.3).

The mass of outputs (raw digestate and biogas) exceeded inputs by 203 tFW (2.7% of inputs) and 221 tFW (2.9% of inputs) for adjusted and unadjusted model, respectively (Table 5.5). This was in the same order of magnitude as model uncertainty. According to model hypothesis, the mass of gas was correct, and the surplus mass was located in the digestate.

Regarding total element flows, TS, VS, and TN were predicted with a correct accuracy of less than 20% error for both phases by the unadjusted model (Table 5.6). This error was comparable to the coefficients of variation of EOM characteristics (Chapter 3). The mineral matter was overestimated. Total TAN flows were well simulated in liquid digestate, and overestimated by 36% in solid digestate: this overestimation was low in absolute value, and not so important regarding model uncertainty. Adjusted model showed similar performance. However, TN flow was underestimated by 24% in the liquid phase.

Considering unadjusted model, for raw digestate, TS, VS, TN, and TAN contents were overestimated by 25% to 30% (Table 5.7). However, it was consistent regarding model uncertainties. The simulated C:VS ratio of the digestates was greater than our assumption (C:VS = 0.5). As a results, $C:N_{org}$ ratio was slightly overestimated. However, we did not measure the real C content in digestates and we cannot conclude if this is an error of the model or our own assumption. TAN:TN ratio was well simulated. The performance of the unadjusted model to simulate liquid digestate was similar to the one of raw digestate. TN and TAN contents in liquid digestate were well modelled. Considering solid digestate, TS, and VS contents were well modelled (relative errors below 8%). TN content was slightly overestimated by 19%, which was consistent with model uncertainty. TAN content was highly overestimated, causing the overestimation of TAN:TN contents.

The adjusted model simulated the digestate characteristics with similar performance compared to unadjusted model (Table 5.7). As a result of the decrease in the BMP of some wastes, the overestimation of TS, and VS contents in raw and liquid digestate increased. The TS and VS contents in raw and liquid digestate were inside the range of uncertainties of the model. Those results illustrated the difficulty to adjust biogas production without affecting digestate characteristics, in a context where information on inputs is limited.

Globally, the model had a good performance to simulate N content, a medium performance to simulate TS and VS content in digestates.

Table 5.5 – Observed and simulated fresh mass flows during the 56-week period: inputs, digestates, and biogas. Simulated variables are indicated as: mean ± uncertainty. Relative errors (RE) are indicated on the second line, in percentage of observed data.

Type	Inputs (tFW an^{-1})	Biogas (tFW an^{-1})	CH_4 in biogas (10^3 Nm^3)	CO_2 in biogas (10^3 Nm^3)	Raw digestate, before post-treatment (tFW an^{-1})	Liquid digestate, export (tFW an^{-1})	Solid digestate, export (tFW an^{-1})	All digestates, export (tFW an^{-1})
Measure	7547	722	287	274	6825	6974	719	7693
Unadjusted model RE	7546	1161 ± 121 61%	617 ± 60 115%	367 ± 58 34%	6588 ± 131 -3%	5899 ± 178 -15%	619 ± 60 -15%	6518 ± 239 -15%
Adjusted mode REl	7547	1021 ± 90 41%	396 ± 43 38%	376 ± 43 37%	6747 ± 120 -1%	6041 ± 163 -13%	633 ± 55 -12%	6673 ± 218 -13%

Table 5.6 – Measured and simulated nutrient and mass flows in digestates during the 56-week period, expressed in t. Observed values are indicated as mean ± standard error: standard errors were derived from digestate characteristics, no error could be estimated on fresh mass flows. Simulated values are reported as simulation results ± uncertainty. Relative errors (RE) are indicated on the second line, in percentage of observed data.

Digestate	Type	Fresh mass	TS	VS	MM	TN	TAN
Liquid digestate	measure	6974	325 ± 13	204 ± 11	121 ± 8	31 ± 3	15 ± 1
	unadjusted model	5899 ± 178	352 ± 89	202 ± 70	151	25 ± 5	13 ± 3
	RE	-15%	8%	-1%	25%	-20%	-14%
	adjusted model	6040 ± 163	371 ± 82	222 ± 64	149	24 ± 5	12 ± 3
	RE	-13%	14%	9%	23%	-24%	-17%
Solid digestate	measure	719	154 ± 18	125 ± 15	29 ± 4	4.7 ± 0.6	0.9 ± 0.3
	unadjusted model	619 ± 60	141 ± 36	99 ± 35	41	4.8 ± 1.1	1.3 ± 0.3
	RE	-14%	-9%	-20%	42%	2%	36%
	adjusted model	633 ± 55	148 ± 33	109 ± 31	39	4.6 ± 1.0	1.2 ± 0.3
	RE	-12%	-4%	-12%	33%	-2%	31%

Table 5.7 – Simulated characteristics of digestates during the 56-week period, on a fresh weight basis (simulation result ± uncertainty). Observed values are indicated as mean ± standard error. Relative errors (RE) are indicated in percentage of observed data.

Digestate	Type	TS (%FW)	VS (g kgFW^{-1})	C :VS ratio	TN (g kgFW^{-1})	TAN (g kgFW^{-1})	C:N_{org} ratio	TAN:TN ratio
Raw digestate	observed	6.6 ± 0.5	46 ± 3		4.8 ± 0.4	3.0 ± 0.5	13 ± 7	0.62 ± 0.11
	unadjusted model	8.6 ± 2.1	57 ± 19	0.62	6.1 ± 0.3	3.9 ± 1.0	16	0.64 ±0.16
	RE	30%	23%		26%	29%	25%	2%
	adjusted model	8.8 ± 1.9	61 ± 17	0.64	5.7 ± 0.3	3.7 ± 0.8	19	0.64 ±0.16
	RE	34%	32%		18%	22%	49%	3%
Liquid digestate	observed	4.7 ± 0.2	29 ± 0.4		4.4 ± 0.2	2.2 ± 0.2	6 ± 1	0.48 ± 0.05
	unadjusted model	6.0 ± 1.5	34 ± 12	0.61	4.2 ± 0.9	2.2 ± 0.5	10	0.52 ± 0.18
	RE	28%	17%		-6%	2%	63%	8%
	adjusted model	6.1 ± 1.4	37 ± 11	0.62	3.9 ± 0.8	2.1 ± 0.5	12	0.53 ± 0.17
	RE	32%	25%		-12%	-4%	93%	9%
Solid digestate	observed	21 ± 2	174 ± 21		6.6 ± 0.4	1.3 ± 0.5	16 ± 2	0.20 ± 0.6
	unadjusted model	23 ± 6.2	161 ± 58	0.59	7.8 ± 2	2.0 ± 0.5	16	0.26 ± 0.13
	RE	6%	-8%		19%	58%	-1%	33%
	adjusted model	23 ± 5.6	173 ± 52	0.60	7.3 ± 2	1.9 ± 0.5	19	0.26 ± 0.11
	RE	9%	-1%		11%	49%	17%	34%

5.3.3 Modelling gaseous emission during storage

The gaseous emissions during storage are indicated in Table 5.8. CH_4 and NH_3 emissions mostly occurred during storage of liquid digestate. CH_4 emissions during storage of liquid digestate corresponded to 4% to 6% of the produced biomethane for unadjusted and adjusted model, respectively. Gaseous emissions were similar for unadjusted and adjusted models. Without phase separation, the storage of raw digestate would emit 11 t CH_4-C and 9 t NH_3-N.

The used emission factors for NH_3 and N_2O relatively to initial TAN are displayed in Table 5.1. For solid digestate, the emission of NH_3 and N_2O were 13% and 4% of initial total N before storage, respectively (adjusted model).

Table 5.8 – Simulated gaseous emissions during storage

	Liquid digestate			Solid digestate		
	CH_4 (tC)	NH_3 (tN)	N_2O (tN)	CH_4 (tC)	NH_3 (tN)	N_2O (tN)
Unadjusted model	14 ± 27	10 ± 3	0	0.2 ± 0.1	0.7 ± 0.3	0.20 ± 0.10
Adjusted model	13 ± 19	10 ± 2	0	0.3 ± 0.1	0.6 ± 0.3	0.20 ± 0.09

5.4 Discussion

5.4.1 Mass balances of anaerobic digesters

Several studies realized a mass balance during anaerobic digestion, to assess digester performance (Banks et al., 2011; Chen et al., 2014; Pognani et al., 2012; Schievano et al., 2011), or to evaluate struvite precipitation (Marti et al., 2008). The mass balance concerned fresh matter, but also TS, VS, C, N, P, K, or even Mg, often with a few number of well characterized inputs (Banks et al., 2011; Marti et al., 2008). Here, due to missing information about the inputs, we were only able to evaluate a fresh mass balance during digestion. The fresh weight mass balance error obtained in this study was similar to the one reported in other studies (Banks et al., 2011; Schievano et al., 2011).

5.4.2 Performances of SYS-Metha compared to other anaerobic digestion models

Bareha et al. (2021b) evaluated the SYS-Metha model on several pilot-scale and real-scale digesters. They found on average good agreements between measured biogas production and digestate characteristics, with r^2 equal to 0.76, 0.92, 0.91, 0.72, and 0.78, for the evaluation of TS, VS, C, TN and TAN contents, respectively, with absolute bias lower than 10%. This chapter aimed at investigating the evaluation of SYS-Metha model on a single anaerobic digester, with numerous poorly characterized inputs. Thus, the results are much less powerful than the validation across several digesters.

In the present case study, biogas production was overestimated. Input adjustments had to be done to have a more realistic simulation of biogas production, although it remained overestimated by 41%. The simulated biogas production model was compared to gas consumption data, which was likely lower than gas production. The consumption of biogas given by the managers of the plant did not consider the possible flaring caused by the surplus biogas production. Secondly, one model assumption was an optimal degradation of organic inputs. In the digester, microorganism activity could be limited by diverse possible inhibitions, e.g. due to ammonia or volatile fatty acids (Esposito et al., 2012). Globally, SYS-Metha simulated biogas production with an error of 18% (normalized root mean squared error) (Bareha et al., 2021b). As a comparison, other mechanistic models were able to predict the biogas production from full scale digesters with error ranging from 0 to 23% (Carlos-Pinedo et al., 2020; Donoso-Bravo et al., 2020; Page et al., 2008). Those last studies used a calibration step and used a limited number of inputs that were well described. A more consistent simulation of biogas production in SYS-Metha could be improved by better characterizing the inputs in digesters similar to the one studied here, or with the enrichment of the waste database.

The model predicted digestate production and characteristics with a correct accuracy. The simulated C:VS ratio was higher than 0.5, which could possibly represent an overestimation of C content of digestates. We found a limited number of study predicting such digestate characteristics. Galí et al. (2009) predicted VS and TAN contents of a digestate in a lab-scale digester with a 10% error on one case study using a modified and adjusted ADM1 model. Bareha et al. (2018) dynamically simulated TAN content in digestates originating from different substrates with very low errors ($r^2 = 0.99$), with a modified and calibrated ADM1 model. Donoso-Bravo et al. (2020) was able to predict dynamically TAN content and VS content of two digestates within a 20% and 10% error, respectively. Page et al. (2008) underestimated TAN contents of 83% and 36% in two digestates. The consistent simulation of the digestate characteristics was a real strength of SYS-Metha model.

5.4.3 Further considerations: discussing the assumptions of the SYS-Metha model

This section aimed at discussing the hypothesis and emission factors used in SYS-Metha in regards to the literature.

5.4.3.1 Methane emissions from liquid and raw digestates

In SYS-Metha, a decrease in HRT implies a higher VS content in digestate and thus a higher CH_4 emission during storage. The stability of the organic matter is correlated to HRT (Akhiar et al., 2021). The degradability of digestate is not modelled in SYS-Metha, which could impact methane emissions during storage (Ericsson et al., 2020) but also the efficiency of digestate at increasing soil organic matter after field application, as suggested by Jimenez et al. (2015).

SYS-Metha assumed that the proportion of CO_2 and CH_4 in gas emission during liquid or raw digestate storage was similar to the one in produced biogas (54% in our study). This hypothesis was consistent with experimental measurements (Baldé et al., 2016; Gioelli et al., 2011; Kimura et al., 2019).

SYS-Metha estimated the methane emissions from liquid digestate storage to be between 4% and 6% of production of methane in biogas. This was consistent with the survey of Liebetrau et al. (2010) on 10 plants (from 0.2 to 11 %), but higher than the 1.0% to 1.2% measured by Hrad et al. (2015, 2014)

SYS-Metha simulated 19% more methane emissions during storage of separated digestates after post treatment with solid/liquid phase separation, compared to no phase separation, due to a lower C degradation ratio of raw digestate during storage (8%) compared to the one of liquid digestate (15%). This was not consistent with the findings of Gioelli et al. (2011), or the one of Amon et al. (2006) and Kupper et al. (2020) on slurries. The higher VS content in raw digestate could increase the amount of matter that can be degraded and therefore the emissions of methane (Gioelli et al., 2011). Poor justification of different CH_4 emission factor on a VS basis for raw and liquid storage were found in the literature; one argument could be a higher ability of raw digestate to form a crust, which was shown to reduce CH_4 emissions (Kupper et al., 2020).

5.4.3.2 Ammonia emissions from liquid and raw digestates

Kupper et al. (2020) reported emission factors of 9 to 23% of initial TAN during pig and cattle slurry storage. Baldé et al. (2018) estimated ammonia emission factors of 33% and 42% of TAN for liquid and raw digestates assuming some hypothesis for winter emissions and duration of storage. Perazzolo et al. (2016) found TAN emission factors of 6% for liquid and raw digestates during a three-month winter storage, and 32% and 41% emission factors for raw and liquid digestates, respectively, during a three-month summer storage. We concluded that SYS-Metha model used high but consistent emission factors (Table 5.1) for ammonia volatilization from liquid and raw digestate storage.

When a crust is formed on a digestate lagoon, ammonia emissions are reduced (Kupper et al., 2020). The solid-liquid separation phase usually decrease digestate ability to form a crust and to decrease emissions, as shown by Baldé et al. (2018) where the ammonia emissions of liquid digestate were 50% higher than the one of raw digestate. This was indirectly considered in SYS-Metha that used a lower emission factor for raw digestate compared to liquid digestate.

The reduction of NH_3 emissions due to storage cover was comprised between 50% and 75% for slurries (Kupper et al., 2020), the emission factors of SYS-Metha were consistent with these results.

5.4.3.3 N2O emissions from liquid and raw digestates

SYS-Metha predicted no N_2O emission from liquid or raw digestate. Sneath et al. (2006) detected no N_2O emission from covered slurry storage, and low N_2O emission from uncovered slurry storage (0.1 – 0.2 gN m^{-3} d^{-1}), approximately corresponding to an annual emission of 0.13 to 0.26 tN_2O-N for a

3500 m^3 lagoon in our case study. Some N_2O emission was measured by Holly et al. (2017) during a six-month liquid digestate storage. However, it was small in regard to the emission during storage of solid digestate in the same study. Kupper et al. (2020) proposed an emission factor of N_2O of 0.13% of initial N for cattle slurry storage, with no important difference for digestate. If SYS-Metha used these emission factors, it would correspond to an emission of 0.04 tN_2O-N. Therefore, it would be possible that a significant N_2O emission occur from liquid or raw digestate, even if the current assumption from SYS-Metha is also consistent. If N_2O emission would be considered, the coverage of the lagoon could increase this N_2O emission (Kupper et al., 2020).

5.4.3.4 Emissions from solid digestate

During solid digestate storage, emissions of N_2O (0.1% to 0.3% of total N), NH_3 (10% to 12% of total N), CH_4 (0.4% to 4% of initial C), and CO_2 (30 to 50% of initial C) were measured by Dietrich et al. (2021) and Li et al. (2018). The emission factors used by SYS-Metha (Table 5.1 and section 5.3.3) were similar for CO_2, CH_4, and NH_3, and slightly higher for N_2O. It was also consistent with emission factors of farmyard manures (Amon et al., 2006; Hassouna et al., 2008; Petersen et al., 1998). More research could be needed on the gaseous emissions during the storage of solid digestate.

5.4.3.5 Discussing the use of emission factors

The use of emission factors is a simple approach to approximate gaseous emissions during digestate storage. However, those emissions are affected by numerous factors, which are not taken into account with the emission factor approach. Ambient temperature is known to affect gaseous emissions during storage. A higher temperature promote gaseous emissions during storage (Baldé et al., 2018; Gioelli et al., 2011; Kaparaju and Rintala, 2003; Kimura et al., 2019; Sommer, 1997). As a consequence, the emission factors described in the model could depend on the location of the anaerobic digester, as well as the dates and frequency of digestate removal. For these reasons, various emission factors were measured in the literature, and high uncertainties may be caused by the use of emission factors (Walling and Vaneeckhaute, 2020b). SYS-Metha uses an emission factor approach that increases the robustness and the simplicity of the model. However those uncertainties should to be considered.

5.5 Conclusion

SYS-Metha is a promising tool that was validated on a complex real case study. The quantity of produced digestates as well as their TS, VS, TN, and TAN contents were correctly predicted. One limit to consider was the overestimation of biogas. The gaseous emissions during storage were estimated with correct assumptions in regard to the current scientific knowledge. The CH_4 emission during raw digestate storage compared to the one of liquid digestate, and the absence of N_2O emission during the liquid and raw digestate storages, are two assumptions that can be discussed. SYS-Metha finally did not assess the increase in organic matter stability in digestate compared to the inputs. However, the simplicity of SYS-Metha, and the fact that it is one of the only model designed to predict digestate characteristics, were strong arguments to use it inside a broader farm-scale model to explore the impact of the anaerobic digestion at the farm scale.

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Chapitre 6

La méthanisation des effluents d'élevage peut diminuer l'utilisation d'engrais de synthèse, augmenter le stockage de C dans les sols et augmenter la volatilisation d'ammoniac à l'échelle de la ferme : rôle clef de l'import de déchets et évaluation de différentes pratiques de gestion

Les Chapitres 4 et 5 ont individuellement évalué un modèle sol-plante (STICS), et un modèle de méthanisation (SYS-Metha). L'utilisation de modèles peut permettre de généraliser les connaissances obtenues grâce aux mesures sur des cas d'étude. Dans ce Chapitre 6, nous couplons ces deux modèles, pour simuler une exploitation agricole de polyculture élevage avec un méthaniseur. Ces simulations permettent et comprendre comment la méthanisation influence des flux de C et de N à l'échelle de l'exploitation. Ce Chapitre 6 est écrit sous la forme d'un article scientifique, qui aura vocation à être soumis à une revue scientifique après adaptation.

The anaerobic digestion of cattle effluents may increase synthetic fertilizer substitution, soil organic matter storage, and ammonia volatilization at the farm scale: key role of imported wastes and evaluation of different management practices

Victor Moinard[1], Antoine Savoie[2], Younès Bareha[3], Romain Girault[3], Florent Levavasseur[1], Jean-Marie Paillat[4,5], Sabine Houot[1]

1: UMR ECOSYS, Université Paris-Saclay, INRAE, AgroParisTech, Thiverval-Grignon, France

2: UE PAO, INRAE, Nouzilly, France

3: UR OPAALE, INRAE, Rennes, France

4: UPR Recyclage et risques, CIRAD, Montpellier, France

5: UPR ADI-Suds, ISTOM, Angers, France

Abstract

Anaerobic digestion (AD) has been strongly developing in Europe during the last decade. The introduction of AD in a crop-livestock farm has multiple consequences on the treatment of animal effluents and on the management of crop fertilization. These consequences need to be considered in an integrative way. In this study, we built a farm scale model to simulate the C and N fluxes in a French crop-livestock farm, with or without anaerobic digestion. The farm model first consisted in an exogenous organic matter (EOM) management module based on the AD model SYS-Metha and on emission factors during EOM storage. The soil-crop model STICS was used during 20 years to simulate the C and N dynamics in the different crop successions of the farm, in response to organic and mineral fertilization predefined with various decision rules. We simulated different scenarios to explore the impact of the types and quantities of external wastes used in the digester, and the impact of digestate management such as phase separation, cover of digestate storage, and different strategies of digestate applications on the crops including its injection in the soil. We quantified the emissions of greenhouse gas (GHG - N_2O and CH_4) and other pollutant emissions (NH_3, NO_3^-), the use of synthetic fertilizer, and the soil organic carbon storage. Finally, we estimated the net GHG emissions induced by AD in considering the farm management, the energy production, and the alternative treatment of wastes. When the imports of external wastes were high, AD increased SOC from 0.4 to 1.0 tC ha^{-1} after 20 years, decreased the use of synthetic fertilizers from 25% to 33%, and increased ammonia volatilization from 80% to 102%. Within our assumptions of biogas valuing as electricity (no heat valuing, electric mix based on nuclear energy), AD slightly increased the net GHG emissions. Inputs and AD processes were the main drivers of the AD environmental balance, but the management practices also had some effects. In particular, an uncovered digestate lagoon storage strongly increased ammonia emission during storage (from 51% to 65%) and subsequent use of synthetic fertilizers. This study highlighted the interest of the farm scale approach to synthetize the effects of on-farm AD. Such approach could be used to evaluate the introduction of AD in territories with different resources in organic matter, or to evaluate the effects the change of cropping systems induced by AD.

Keywords

Exogenous organic matter; Ammonia volatilization; Nitrate leaching; Nitrous oxide emissions; Soil organic matter; Anaerobic digestion; Crop-livestock farm.

6.1 Introduction

Anaerobic digestion (AD) is a waste treatment where microorganisms degrade organic wastes, animal manures, or dedicated crops to produce biogas used as a renewable energy. AD also produces digestate that is the residual organic product. Digestate is often applied on crops as organic fertilizer (Stürmer et al., 2020), promoting nutrient recycling. In the world and in particular in Europe, AD has been thus promoted by many policies that aimed at promoting the production of renewable energy, at decreasing the environmental impact of agriculture, or at improving waste treatment (Scarlat et al., 2018; Vasco-Correa et al., 2018). As a consequence, the number of digesters in Europe was multiplied by three between 2009 and 2019 reaching 18,000 digesters in Europe in 2019 (European Biogas Association, 2020). AD generally induces a decrease in total greenhouse gas (GHG) emissions, but can be associated to other environmental risks including a possible increase in acidification and eutrophication (Hijazi et al., 2016). In this regard, C and N are two elements of major interest that are linked to numerous benefits and risks of AD, such as biogas production, gaseous emissions during digestate storage and field applications, substitution to synthetic fertilizers, or soil organic matter (SOM).

Various types of AD can be found, including the digestion of sewage sludge in wastewater treatment plants, or territorial digesters that treat urban or agro-industrial wastes. In this study, we will focus on on-farm AD, where animal effluents are co-digested with other wastes (Vasco-Correa et al., 2018). The use of digestates instead of animal effluents as organic fertilizers will have strong implications on C and N fluxes at the farm scale. First, the import of external organic wastes influences the biogas production (Boldrin et al., 2016; Weiske et al., 2006) and the nutrient balance in the farm (Möller, 2009). AD often improves manure handling and reduces CH_4 emissions during manure storage (Baral et al., 2018; Miranda et al., 2015). This can be partly counterbalanced by biogas leakage from the digester (Hrad et al., 2015). AD can also increase (Baldé et al., 2018; Clemens et al., 2006; Neerackal et al., 2015) or decrease (Amon et al., 2006) ammonia (NH_3) emission during digestate storage. Both digestates and animal effluents are exogenous organic matters (EOMs) that are valued in the field. However, they have different characteristics. During the digestion, part of the input organic matter is mineralized, producing an export of C as biogas and inducing an increase in inorganic N content. Digestates are therefore richer in total ammoniacal N (Möller, 2015) and characterized by a more stable organic matter than undigested animal effluents (Béghin-Tanneau et al., 2019; Cavalli et al., 2017; Thomsen et al., 2013). However, depending on digester inputs and digestate post-treatment, digestates can have variable characteristics (Guilayn et al., 2019a; Stürmer et al., 2020). Digestates often have good N fertilizer properties and can substitute synthetic N fertilizers (Möller and Müller, 2012). AD could also possibly impact SOC storage after repeated applications in the long term (Bodilis et al., 2015; Wentzel et al., 2015), but the impact of digestate on soil organic matter (SOM) and soil life compared to undigested animal effluents still remains unclear (Möller, 2015). As other EOMs, the repeated application of digestates in the long term could improve soil N supply and decrease N fertilizer

requirements (Gutser et al., 2005). However, the digestates can be more sensitive to ammonia volatilization after application in the field than undigested effluents (Nkoa, 2014).

These multiple consequences of the introduction of on-farm AD can interact. For example, both EOM storage and field application contribute to GHG and NH_3 emissions, and considering both together is important to understand the real impact of the digestion of animal effluents (Amon et al., 2006; Holly et al., 2017). Because of the variety of practices associated with on-farm AD and the numerous effects that need to be accounted for, it is difficult to realize the synthesis of the effects of on-farm AD only with experiments. Farm scale models can be useful tools to consider interactions between all the components of the farms and therefore to understand the environmental impacts of AD and its diverse management practices (Petersen et al., 2007; Rotz, 2018; Schils et al., 2007). Veltman et al. (2018) used a whole-farm model that simulated the livestock management and the cropping systems in dairy farms in the USA. The authors showed that cattle effluent mono-digestion could decrease GHG emissions while only slightly increasing N losses in the environment. With a model including swine breeding and manure handling, Rigolot et al. (2010) highlighted the ability of AD (mono-digestion) to reduce gaseous emissions during EOM storage. Vergote et al. (2019) estimated the GHG emissions of an on-farm digester using a digestion model and additional emission factors. The authors highlighted the importance of a good manure handling (i.e., short storage inside the livestock buildings) to reduce GHG emissions at the farm. Weiske et al. (2006) also used a whole-farm modelling approach to evaluate the GHG emissions of AD at the farm level. The authors highlighted the importance of the amount and type of the organic inputs co-digested with animal effluents (biowastes or dedicated crops) on net GHG emissions of AD. Boldrin et al. (2016) also confirmed the importance of the import of wastes on biogas production with a mass balance model. These studies helped to understand the factors that drive the good environmental performance of AD at the farm scale, in particular concerning the GHG emissions. However, in most studies, GHG emissions after digestate application in the field was only considered with emission factors and the cropping systems at the farm were rarely simulated. The use of a whole-farm model considering a detailed cropping system could help to refine our understanding on the impact of AD on GHG emissions, SOC change, fertilizer substitution, NH_3 volatilization, or nitrate leaching, considering both short- and long-term effects after one or repeated applications of digestates in the field. Indeed, all these factors can be important in AD environmental performance (Vaneeckhaute et al., 2018).

The objectives of this study was to investigate the effects of the introduction of AD in a French cattle dairy farm with croplands. We focused on C and N fluxes. We aimed at answering four questions. 1) What are the effects of the introduction of AD on C and N fluxes at the farm scale, and in particular in the fields, considering the long-term impacts? 2) What is the effect of the inputs of external wastes on digestate quality and quantity, and therefore on the C and N balance at the farm scale? 3) Can we quantify the effect of different digestate management strategies on C and N fluxes in order to compare them? 4) What is the importance of field digestate management in the GHG emissions related with AD?

To answer those questions, we built a farm scale model. We first used the AD model SYS-Metha (Bareha et al., 2021b) predicting biogas production, gaseous emissions during digestate storage, and digestate characteristics. Then, the predicted digestates were applied on different plots every year, using an algorithm to follow consistent fertilization decision rules. The C and N dynamics were finally simulated with the soil-crop model STICS (Brisson et al., 2008) for the different plots of the farm. This farm model was able to simulate C and N fluxes at the farm scale considering different practices associated to AD. The system was then extended, and we assessed the C footprint of AD with a consequential approach. The originality of the approach locates in the fine modelling of the cropping system with realistic fertilization practices, and in the use of an AD model that is able to predict the digestate characteristics coming from various distinct inputs.

6.2 Material and Methods

6.2.1 Description of the farm model

6.2.1.1 General description

We built a farm model that simulated a cattle dairy farm producing farmyard manure and slurry. The farm model considered the anaerobic co-digestion of cattle effluents and imported wastes, the management of EOMs, and the cropped plots of the farm. The cattle breeding was considered unchanged after the introduction of AD and was excluded from the limits of the system. The farm model consisted in an EOM management module and a crop-soil module that simulated different plots. The EOM management module predicted the quantities and characteristics of EOM that were applied in the fields every year. It also predicted gaseous emissions during EOM storage and biogas production during AD. In case of no on-farm AD, the module consisted in emission factors to simulate the gaseous emissions and mass losses of cattle effluents during their storage. In case of on-farm AD, the module consisted in the SYS-Metha model (Bareha et al., 2021b) which simulated the outputs cited above, together with biogas production. Then, the crops were simulated in several plots. Each plot was defined by a crop rotation and a surface. Crop growth, C cycle, and N cycle in each plot were simulated with the soil-crop model STICS (Brisson et al., 2008) during 20 years at a daily time step. A single soil was simulated for all plots. A single climatic year (September to August) was replicated 20 times, so that the impacts of EOM application on the long term was not modified by the variability of meteorological data. Note that the meteorological data only influenced the simulation in the fields, and not the EOM management module. Each year, an algorithm determined the doses of EOM and synthetic N fertilizer that are applied in each field. The available EOM is allocated and applied in the different plots, and the doses of N synthetic fertilizers are adapted in each plot. The system is described in Figure 6.1.

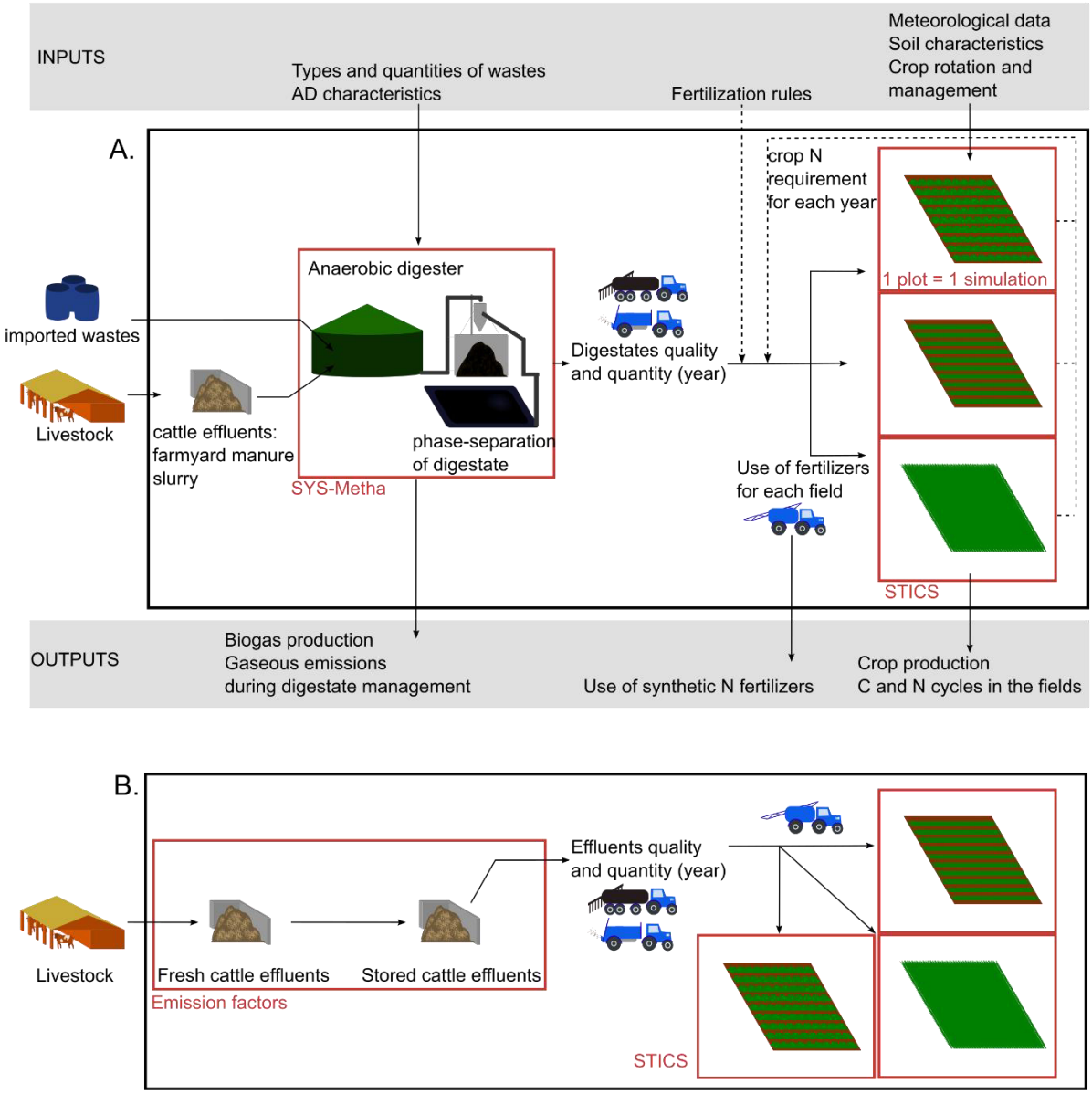

Figure 6.1 – Schematic representation of the farm model. The black box represents the limit of the system. The red boxes represent the use of individual models. A) Simulation of a farm with on-farm AD. B) Simulation of a farm without AD. Only the EOM management module differed from A).

6.2.1.2 EOM management module: AD mass balance model SYS-Metha

We used an EOM management submodel to estimate the yearly available amounts and characteristics of EOMs to be applied in the fields, as well as the biogas production from AD.

When we simulated a crop livestock farm without AD, fresh cattle effluents were stored on a dedicated platform for solid manure, and in a covered pit for slurry. Gaseous emissions (NH_3, N_2O, CH_4, CO_2), N transformation, and total solid and total mass losses during storage were simulated using emission factors for slurry (Amon et al., 2006) and for farmyard manure (Hassouna et al., 2008) (Supplementary Material 1). We deduced the amount and characteristics of cattle slurry and cattle solid manure applied in the field with a mass balance approach.

When cattle effluents were digested, they were supposed to be put fresh inside the digester and no emissions were considered before AD. The amount and characteristics of digestates, the gaseous emissions during the storage of digestates, and the biogas production were simulated with SYS-Metha model (Bareha et al., 2021b, 2021a) (Chapter 5). The SYS-Metha model was selected for its ability to predict digestate characteristics from the digestion of a variety of different inputs. It is a simple static AD model designed to predict digestate production and characteristics, biogas production, and gaseous emissions during digestate storage. The inputs of the model are the main digester characteristics (hydraulic retention time HRT, type of phase separation of digestate, presence of a cover over liquid or raw digestate storage), and the input types and quantities. Briefly, SYS-Metha includes a waste database that is used to estimate the waste characteristics (contents of total solid TS, volatile solid VS, C, total N TN, total ammoniacal N TAN; biogas production potential; N mineralization potential). The waste characteristics and HRT are used to predict biogas production and N mineralization inside the digester. The amount and characteristics (TS, VS, C, TN, TAN contents) of raw digestate are deduced with a mass balance approach. When a phase separation occurs, liquid and solid digestates quantities and characteristics are simulated using separation efficiency coefficients. During storage, gaseous emissions (NH_3, N_2O, CH_4, CO_2), and changes in digestate characteristics are simulated using emission factors.

6.2.1.3 Field simulation: use of the soil-crop model STICS and of the ammonia volatilization model ALFAM2

In the farm model, we simulated several farm plots with each their own crop rotation, during 20 years. The soil-crop system of each plot was simulated at the daily time step using the STICS model (Brisson et al., 2008). The ability of STICS to predict the growth of multiple crops has been evaluated in diverse French pedological and meteorological conditions (Coucheney et al., 2015; Yin et al., 2020). Recently, STICS was calibrated to predict soil-grassland system (Graux et al., 2020) and to simulate the effects of EOMs in the long term (Levavasseur et al., 2021b). The ability of STICS to simulate a cropping system using digestate was evaluated on similar soil and weather conditions as in this present

study (Chapter 4). We used the default parameters for EOM mineralization from (Levavasseur et al., 2021a). Indeed, SYS-Metha does not model the organic matter stability of digestate.

We used a research version of STICS (v10.2751). STICS was not able to represent correctly ammonia volatilization from EOM (Chapter 4). Therefore, the ammonia volatilization following EOM application was simulated using ALFAM2 model (Hafner et al., 2019) for liquid EOMs, and emission factors for solid EOMs. Considering liquid EOMs, the ALFAM2 model is a simple volatilization model that predicts ammonia emissions after slurry application using EOM characteristics (DM content, pH, TAN content), application rate, method of application, wind speed, air temperature, and rain rate as inputs. The use of ALFAM2 enabled to simulate various types of application, i.e. trailing hose application or shallow injection. Considering solid EOMs (farmyard manure and solid digestate), we used common emission factors, i.e. 66% and 28% of applied TAN when the EOM was buried after 24 h and 4 h, respectively. Those emission factors were taken from Chapter 3 (24 h), and a reduction of volatilization of 43% was assumed when the solid EOMs are burrowed sooner (ALFAM2 database) (Hafner et al., 2018).

Crop management parameters such as soil tillage, sowing date, sowing density, fertilization dates, were determined for each crop based on local practices of farmers and kept unchanged from one year to another. The TS, VS, C, TN, TAN contents of EOM were deduced from the EOM management module. The pH of EOMs were set as constant and were 8.0, 7.9, 9.3, 7.3, and 9.7 for raw digestate, liquid digestate, solid digestate, cattle slurry and cattle manure, respectively (Chapter 3). The doses of EOM and N synthetic fertilizer were determined every year by the algorithm described in the following section.

6.2.1.4 Algorithm used to determine the applied dose of organic and synthetic fertilizers in each plot

The doses of EOM and synthetic N fertilizer that were applied on each plot were adjusted every year. Solid EOMs (cattle manure, solid digestate) were considered as soil amendments, i.e. they did not directly substitute synthetic N fertilizer but rather bring high amount of organic matter in the soil and influence SOC storage. Liquid EOMs (raw digestate, liquid digestate, cattle slurry) were considered as organic fertilizers with high TAN contents, and their application substitutes an application of synthetic fertilizer. The applied doses depended of the crop N requirement, to ensure a constant yield as far as possible. We used an approach based on the fertilization recommendation of the French comity of study and development of sustainable fertilization (COMIFER, 2021).The application doses were determined iteratively every year with an algorithm that consisted in four steps:

1) Determination of the plot where and dates when solid EOMs were applied (predefined doses)

Solid EOMs were applied with constant fixed doses. Fertilization rules defined an ordered list of possible application events (crops and dates of application). Solid EOMs were allocated to these events one by one until no solid EOM remained. When two plots had the same priority (e.g., two maize crops), the

priority was given first to the plot that did not received EOM for the longer time, and then to the plot that received the less organic matter from EOM since the beginning of the experiment.

2) Estimation of the N demand from the crops in each plot. The crop N requirement was computed for each plot, depending on the soil N supply:

   2.1) We simulated all the years that preceded the year of interest, with previously computed fertilization. It was followed by the simulation of the year of interest, without any synthetic fertilizers nor liquid EOM (eventual solid EOM were applied).

   2.2) We simulated the crop N uptake before the fertilization period (N crop), the soil mineral N content before the fertilization period (SMN), the soil N supply during the fertilization period until harvest (soil N supply), and the atmosphere N deposition during the fertilization period until harvest (atmosphere N deposition).

   2.3) The inorganic N needed by the crop to reach a specific target (crop N target) was deduced with a N balance approach (COMIFER, 2021):

   total crop N requirement = crop N target – N crop – SMN – soil N supply – atmosphere N deposition

   The crop N target depends on yield target and thus on case study (COMIFER, 2021).

   2.4) The total crop N requirement was split in different fertilization dates, using the guidance of agricultural advisors. Therefore, each plot was subject to several fertilization dates that were each defined by a date-specific crop N requirement.

3) Determination of the plot, dates, and doses where liquid EOMs are applied: the doses depended on crop N requirement.

Concerning liquid EOMs, fertilization rules defined an ordered list of possible application events (crops and dates of application). Liquid EOM was allocated to these events one by one until no liquid EOM remained. When two plots had the same priority (e.g., two maize crops), the priority was given first to the plot that did not received EOM for the longer time, and then to the plot that received the less organic matter from EOM since the beginning of the experiment. The dose of liquid EOM was defined according to N requirement from the crop and date of application, based on EOM TAN content. Volatilization or mineralization of organic N were not considered; indeed preliminary work showed that it complicated the model and that TAN content was a good proxy of EOM N availability in the simulation. EOM were always applied at a dose comprised between 30 and 40 t ha^{-1}. Indeed, lower doses were rarely used by farmers (logistical constraints), and higher doses were considered possibly harmful for the environment. When calculated dose was lower than 20 t ha^{-1}, the dose was considered too low and no liquid EOM was applied. When the calculated dose was between 20 t ha^{-1} and 30 t ha^{-1}, the liquid EOM was applied at a 30 t ha^{-1} dose. When the calculated dose was higher than 40 t ha^{-1}, the liquid EOM was applied at a 40 t ha^{-1} dose. Then, when the TAN applied with EOM was not equal to date-specific N requirement, the

consequent N surplus or N default was reported to another fertilization event in the same plot. The last applications of liquid EOM always concerned grassland, where liquid EOM was applied whatever the dose to ensure that all available liquid EOM was applied. Indeed, farmers use grasslands as a buffer, where digestate can be applied at any dose and on multiple dates, to facilitate digestate management. When the fertilization date was earlier than March 1st, the fertilization occurred on March 1st to ensure that the soil was able to bear the spreader tank.

4) Determination of the plot, dates, and doses for applied synthetic N fertilizers: the doses depended on crop N requirement and liquid EOMs applications.

Synthetic N fertilizers were applied in each plot at the dates when no liquid EOM were applied, at a dose corresponding to date-specific crop N requirement.

This method enabled to compute dynamically the synthetic N fertilizer substituted by digestate, considering the increase of SOM with the time due to repeated application of EOM. The consumption of chemical fertilizer was thus an output of the model, and not an assumption. This method helped to simulate similar yields through time and between scenarios, even if a constant yield was difficult to achieve due to different dynamics of N release between organic and synthetic fertilizers.

6.2.2 Case study and scenarios

The simulations were globally based on the case study of the anaerobic digester at INRAE of Nouzilly, Centre Val-de-Loire, France (Chapters 2, 3, 4, et 5) (47°32’ N, 0°48’ E). However, several adjustments and simplifications were done in the simulated systems. Several scenarios were simulated with the farm model (Table 6.1).

All scenarios considered a single soil representative from the Nouzilly case study. The soil was a Luvisol (WRB, 2015) with an average depth of 100 cm. It was a silty loamy soil with the following characteristics in the upper horizon (0-20 cm depth): 20% clay, 64% silt, 16% sand, pH 6.7, Soil Organic Matter (SOM) content 23.3 g kg^{-1}, C:N ratio 10.1, $CaCO_3$ content 1.1 g kg^{-1}, bulk density 1.37 g cm^{-3} (Chapter 3). The other soil horizons are described in Supplementary Material 2. We used the climatic dataset from Météo-France (SAFRAN), from 01/2002 to 12/2014. A climatic year was defined between 1st of September and 31st of August. The simulation only use one climatic year that was repeated 20 times. Preliminary work showed that the set of climatic years 2003, 2004, 2006, 2007, 2010, and 2011 were a good representation of the variety of the climate and of the range of the outputs of the model. Yearly mean temperature ranged from 11.5°C to 12.8°C. Yearly total rainfall ranged from 459 mm to 805 mm (Supplementary Material 3). All scenarios were simulated six times, i.e. one for each of these six climatic years. Climatic year 2010 was found to be a correct representative year because it had an average yearly mean temperature, an average yearly rainfall, and because the outputs of the model while simulating this year were often in the middle of the range of the outputs found while

simulating the other climatic years. Yearly mean temperature was 11.9°C and yearly total rainfall was 592 mm. 2010 was used as the default climatic year.

All scenarios represented the same crop successions. We assumed that AD did not affect the crop successions. Cropping area was 299 ha, which corresponded to a consistent area regarding livestock dimension and quantity of digestate according to local experts. Three simplified crop rotations were implemented (ten plots), which was a tradeoff between system representation and numerical costs of the simulation. We simulated three plots of 46 ha each with a rotation rapeseed – wheat – wheat, two plots of 23 ha each with a rotation maize – wheat, and five plots of 23 ha each with rotation of maize – wheat – 3-years temporary grassland. In total, every year, we thus simulated 138 ha of winter wheat (*Triticum aestivum*) (46% of the area), 46 ha of rapeseed (*Brassica napus*) (15%), 46 ha of maize (*Zea mays*) (15%), and 69 ha of temporary grassland (fescue, *Festuca*) (23%). These four crops are the main crops where digestates are applied in the region according to local experts. It corresponds to a simplification of a typical local cropping-system oriented toward dairy production and large cultivated area without irrigation as described by local experts (Chambre d'agriculture de Région Centre – Val de Loire, 2021). The simplifications included the simulation of wheat instead of barley. The simulations of wheat and maize with STICS have been largely validated. Rapeseed was less studied, but it was tested in Chapter 4 in the same case study of Nouzilly. All crops were managed conventionally, with tillage. Maize was used as maize silage, and was preceded by a mustard cover crop. Grasslands were cut two to three times per year. Wheat straws were exported. Crop N target (N requirement) were 300 kgN ha^{-1}, 301 kgN ha^{-1}, 196 kgN ha^{-1} and 220 kgN ha^{-1} for wheat, rapeseed, maize and grassland, respectively, based on local yield targets. Preliminary work showed that rapeseed was very sensitive to ammoniacal N addition in autumn in STICS, which resulted in a too strong and unrealistic yield gain. Therefore, only solid EOMs were applied in summer or autumn before rapeseed sowing, despite farmers could also use liquid EOM in that case. More details on the technical management are presented in Supplementary Material 4.

All scenarios considered the same dairy cattle farm that produced 845 t $year^{-1}$ of fresh cattle slurry and 532 t $year^{-1}$ of fresh cattle solid manure. The characteristics of manure and slurry are in Table 6.2. When the cattle effluents were not digested, solid manure was stored outside, and cattle slurry was stored in a covered pit. The cattle effluents were co-digested with a mixture of wastes. Two types of mixture of other wastes were considered:

- Mixture "A": a mixture of municipal and agricultural wastes. It is considered as a classical mixture for the on-fam digesters in the region of the case study. It consisted in wastes from cereal storage (17% of the mixture), maize silage (17%), grass clippings (22%), kitchen wastes (22%), and wastes from supermarket (22%).

- Mixture "V": a mixture of sewage sludge and various wastes. It is similar to the mixture used in the case study, which is more territorial than the classical on-farm digesters of the region. It consisted in liquid urban and industrial sludge (62% of the mixture), wastes from cereal storage (14%), solid urban sludge (13%), grease (5%), grass clippings (3%), whey permeate (1%), maize silage (1%), wastes from supermarkets (1%), cattle feed wastes (<1%), wastes from yeast production (<1%).

We mainly used the default parameters of SYS-Metha to benefit from the robustness of the model. However, cattle fresh effluents characteristics were adjusted to those from the Nouzilly case study (Table 6.2), and the characteristics of grease and wastes from cereal storage were adjusted so they produced less biogas. Indeed previous analysis showed that default parameters of these wastes led to overestimate biogas production (Chapter 5). HRT was set to 100 days in all scenarios. When the digestate was post-treated by phase separation, a screw press was used.

The scenarios were defined by the presence or the absence of digester, the type and quantities of digested wastes, EOM management practice (storage, post-treatment), and practices associated to EOM application in the field. Each scenario was compared to two default scenarios. The first scenario served as the reference and considered the farm without AD ("no AD"). The second scenario (baseline AD, ADb) considered the co-digestion of cattle effluents with a mixture of sewage sludge and various wastes and was similar to the Nouzilly case study. It was used as a baseline, in the sense that we compared every following scenario to this one to evaluate the impact of the different practices.

"no AD" and ADb shared the same characteristics. These characteristics were based on farmer practices in the studied region, considered as "good practices":

- The liquid EOMs was stored in a covered lagoon
- The liquid EOMs was applied in late winter and spring. It was applied preferentially on wheat and before maize sowing, and then on grasslands. Indeed, wheat and maize can value the N inputs.
- Application doses of liquid EOM was comprised between 30 t ha^{-1} and 40 t ha^{-1}.
- The liquid EOMs was band applied. It was buried after 24 h when applied before maize sowing.
- The solid EOMs were applied in summer before rapeseed sowing, or in spring before maize sowing. Applied doses were 15 and 20 t ha^{-1} for solid digestate and farmyard manure, respectively, due to farmer practices and simulated quantities of available EOM. The solid EOMs were buried 24 h after application.

Additionally, in ADb scenario, the cattle effluents (30%) were co-digested with a mixture of sewage sludge and various wastes (mixture "V", 70%). This high import of various wastes is similar to Nouzilly case study.

First, the simulation outputs of these two scenarios were studied thoroughly to understand the change in C and N fluxes caused by the introduction of AD in the farm. Then, several scenarios were used to evaluate the influence of the type and quantity of external inputs in the digester in the farm C and N fluxes.:

- EI0 scenario consisted in mono-digestion of animal effluent (inexistent in the regional context, realized as a simulation exercise).
- EI30V scenario consisted used the same external wastes as ADb but in lower quantities: the digested wastes consisted in 70% of cattle effluents and 30% of sewage sludge and various wastes (mixture "V") in the digester (simulation exercise). EI30V stand for External Input 30% mixture "V".
- EI30A scenario used low imports the mixture of municipal and agricultural wastes: the digested wastes consisted in 70% of cattle effluents and 30% of mixture "A". This scenario is a classical type of on-farm digester in the region of the case study.
- EI70A used high imports the mixture of municipal and agricultural wastes: the digested wastes consisted 30% of cattle effluents and 70% of mixture "A" (simulation exercise).

Because we considered the treatment of the same quantity of cattle effluents, increasing the share of external wastes increased the quantity of produced digestate. The size of the digester was similar for ADb and EI70A scenarios, lower for EI30V and EI30A scenario, and lower again for EI0 scenario. Using this writing code, ADb would exactly correspond to an "EI70V" scenario.

At last, several scenarios were used to evaluate the influence of different practices in AD or in digestate application on the farm C and N fluxes. They were all based on ADb scenario. The investigated practices were:

- the absence of phase separation (PS)
- the absence of storage cover on digestate lagoon (SC)
- the low storage capacity that led to spread liquid digestate during all seasons of the year (Y)
- the application of liquid digestate on maize and grassland in priority, where soil injection was possible (C).
- the injection of liquid digestate and fast burying of solid digestate, alone (I) or in combination with other practices (Y+I, C+I)
- a change in minimal dose of digestate field application (D20, D40)

Table 6.1 – Simulated scenarios. All scenarios consider a farm with anaerobic digestion, excepted "no AD" scenario.

Scenario	Changes between each scenario, and the baseline scenario (ADb)
No AD	Crop-livestock farm without anaerobic digestion
ADb	Crop-livestock farm with anaerobic digestion, baseline scenario.
EI0	Mono-digestion. Digested wastes: Cattle effluents (100%).
EI30A	Low import of wastes. Digested wastes: Cattle effluents (70%), municipal and agricultural wastes (30%).
EI30V	Low import of wastes. Digested wastes: Cattle effluents (70%), sewage sludge and various wastes (30%).
EI70A	High import of wastes. Digested wastes: Cattle effluents (30%), municipal and agricultural wastes (70%).
PS	No phase separation of digestate.
SC	No cover on liquid digestate storage
I	Liquid digestate was injected in the soil when possible, instead of being band spread. Solid digestate was buried after 4 h, instead of 24 h.
Y	Low storage capacity: multiple liquid digestate spreading along the year. The liquid digestate was applied during end of summer (33%, grassland), late autumn (17%, grassland), late winter (33%, wheat, then grassland), and spring (17%, maize).
Y+I	Combination of Y and I scenarios.
C	Use of digestates on plots where injection was possible. Liquid digestate was applied preferentially before maize, then on grassland, then on wheat.
C+I	Combination of C and I scenarios.
D20	Application dose of digestate between 20 t ha^{-1} and 30 t ha^{-1}
D40	Application dose of digestate between 40 t ha^{-1} and 50 t ha^{-1}

6.2.3 Indicators for studying C and N fluxes at the farm scale

First, we considered C and N fluxes inside the farm, without any extension of the system. This view was interesting to help finding the best practices that could be implemented by the farmers, regarding their constraints. We investigated the following indicators: yearly biogas production, yearly use of synthetic N fertilizers, yearly gaseous emissions of CH_4, N_2O, and NH_3, yearly nitrate leaching. The indicators were averaged during the simulated period of 20 years. SOC storage in all the plots of the farm was also computed as the difference in the SOC stocks before and at the end of the simulated period (20 years). The yearly SOC storage was also computed by dividing the total SOC storage by 20.

6.2.4 C footprint of anaerobic digestion with a consequential approach

6.2.4.1 Goal and scope

Environmental impacts of AD are not only due to the emissions at the farm scale, but also depend on external practices such as the previous treatment of external inputs and the type of energy which is substituted by the biogas. Such environmental evaluations of AD were often studied with Life Cycle Assesment (LCA), using consequential approaches that is a way to deal with the multi-functionality of AD (Hamelin et al., 2014; Styles et al., 2015; Vaneeckhaute et al., 2018). Here, our goal was not to present a LCA study; however, it was interesting to investigate how a detailed farm model can benefit to global environmental assessment of AD. Therefore, we estimated the impact of each scenario described in Table 6.1 on climate change (C footprint). We used the indicator "Global Warming Potential after 100 years" (GWP_{100}) defined by IPCC (2015). Therefore, we estimated the C footprint of each scenario by the sum of all GHG emissions, expressed in t CO_2eq. We used a consequential approach to consider the C footprint of each scenario inside and beyond the farm. The system was extended for each scenario so that they:

- treat of similar amount of cattle effluents, sewage sludge, and other wastes (t fresh weight, tFW)
- produce similar amount of electricity (kWh)
- consider a similar agricultural activity. Two units were used: either a similar crop surface (ha), or similar crop production (t of dry matter of all exported crops, tDM).

GHG emissions were computed on the extended systems for each scenario. Finally, we computed the difference between the GHG emissions each scenario with AD on one side, and the GHG emissions of the "no AD" scenario on the other side, considering either a similar crop surface or a similar crop production. The net GHG emissions expressed the C footprint of AD. Data about GHG emissions were mainly taken from two French databases that were designed to account for GHG emissions, the Base Carbone ® (ADEME, 2021) and GES'TIM+ (ARVALIS Institut du Végétal et al., 2021). The sections 6.2.4.2 to 6.2.4.4 describe how we computed the GHG emissions considering a similar crop surface in all the scenarios. GHG emissions considering a similar crop production in all the scenarios were computed with additional emissions, described in section 6.2.4.5.

6.2.4.2 Impacts coming from the farm

We considered the yearly emissions of CH_4, N_2O, NH_3, and NO_3^- (average emissions during 20 years). The GHG emissions inside the farm gate included:

- CH_4 emissions during EOM storage and biogas leakage and N_2O emissions during EOM storage and in the field. We assumed that leakage corresponded to 3% of biogas production (Hrad et al., 2015). The global warming potential of CH_4 and N_2O were 28 and 265, respectively (IPCC, 2015).
- indirect N_2O emissions from NH_3 and NO_3^- emissions. 1% of NH_3-N and 1.1% of leached NO_3^--N were assumed to be reemitted under N_2O form (GES'TIM+).
- EOM transport from farm to field. The transport distance was assumed to be 5 km. The emission factor used was 0.0715 $kgCO_2eq$ per t km, taken from a local LCA expertise.
- EOM and synthetic fertilizers applications (machinery and fuel). Emissions were computed following GES'TIM+ recommendation and emission factors, considering direct and indirect emissions. Hypotheses are described in Supplementary Material 5. The fertilizer spreading emitted 53.7, 61.0, 4.3, or 6.6 kg $CO_2eq\ ha^{-1}$ when the fertilizer was a liquid EOM, a solid EOM, Urea Ammonium Nitrate (UAN) solution, or ammonium nitrate, respectively (i.e., only considering machinery use and diesel consumption).
- Synthesis and use of of N fertilizers. The emission factors were 3.97 and 4.99 $kgCO_2eq\ kgN^{-1}$ for ammonium nitrate and UAN solution, respectively (GES'TIM+)
- Other agricultural operation (tillage, production of seeds, sowing, production and application of plant protection products, harvest, grain transport to silo). Emissions were computed following GES'TIM+ recommendation and emissions factors, considering direct and indirect emissions. Crop management is detailed in Supplementary Material 4. The assumptions made to compute C footprint are described in Supplementary Material 5. On average, those operations emitted 397.9 $kgCO_2eq\ ha^{-1}$ for wheat, 544.3 $kgCO_2eq\ ha^{-1}$for maize, 412.7 $kgCO_2eq\ ha^{-1}$ for rapessed, and 172.2 $kgCO_2eq\ ha^{-1}$ for grassland.
- We considered that a positive net SOC storage was equivalent to a negative net GHG emission. The average yearly stored C was computed as descried in section 6.2.3. It was then converted into avoided CO_2 emissions.
- P and K synthetic fertilizers. The demand of P and K nutrients were assumed to be 40 $kgP_2O_5\ ha^{-1}$ and 40 $kgK_2O\ ha^{-1}$ based on local experts. P and K applied by EOM were deduced from this demand. P and K contents in EOM were described in Supplementary Material 5. The GHG emissions associated to the P and K synthetic fertilizers were 1.45 $kgCO_2eq\ kgP_2O_5^{-1}$, and 0.71 $kgCO_2eq\ kgK_2O^{-1}$ (GES'TIM+).

The emissions that occurred during the construction of the biogas facility were not considered.

6.2.4.3 Energy production

AD was supposed to use biogas to produce electricity in a combined heat and power unit. Partial heat was used in the digestion process. The supplementary heat energy production was considered as lost. The SYS-Metha model gave the production of electricity. In all scenarios, the electricity consumption by the digester was assumed to be 47.2 kWh per ton of waste treated in one year (data from the case study). In the scenarios where the alternative production of electricity was needed, we considered an average French electricity production. This electricity had a low C footprint (0.066 $kgCO_2eq\ kWh^{-1}$) (Base Carbone) because of the importance of nuclear energy in the French energy mix. Those assumptions were close to a worst-case scenario for energy production with AD: the valorization of heat produced by AD, or the valorization of biogas to replace fossil energies would lead in higher benefits in terms of GHG emissions. Exploring these assumptions were out of the scope of the study, in which we wanted to focus on digestate use.

6.2.4.4 Waste treatment

In all scenarios, the similar amount of wastes were treated, either by AD, or by alternative treatments.

Animal effluents (cattle slurry and manure) were spread in the field, either raw ("no AD" scenario), or digested. All scenarios treated the same quantity of animal effluents, so no additional GHG emissions were accounted in either case.

Liquid and solid sludge are mainly recycled in agriculture as fertilizers in France. We considered that the alternative treatment of sludge was the direct application in the field (without composting). In the scenarios where the sludge were digested, no additional emissions was computed. In the scenarios where the sludge were not digested, we considered the net emissions resulting from their direct spreading. To this end, we simulated their direct spreading using a similar farm model approach, using a single rotation with rapeseed and wheat. Sewage sludge were applied in summer before either wheat or rapeseed at fixed doses (10 to 20 t ha^{-1} depending on sludge type and crop). Emissions during storage were considered. GHG emissions of the farm that used sewage sludge were computed as described in the previous sections. The GHG emissions of a similar farm without sewage sludge were also computed. The difference between the two latest farms was the GHG emissions attributed to the direct spreading of sewage sludge. Two types of sewage sludge were considered (solid and liquid sludge). More details are given in Supplementary Material 6.

Waste from supermarkets and kitchen wastes were considered as biowastes. In the scenarios where the sludge were not digested, we considered the net emissions resulting from their treatment. In France, biowastes are mainly buried, or incinerated with energy recovery. We considered an average biowastes treatment scenario, where 36% of biowaste were incinerated (-172 $kgCO_2eq\ tFW^{-1}$, energy production), and 64% were buried (216 $kgCO_2eq\ tFW^{-1}$, CH_4 emissions). This ratio came from Base Carbone. Therefore, we assumed that the treatment of one ton of biowastes emitted 76 $kgCO_2eq$. The alternative

treatment of cattle feed wastes were considered to be either incineration or burying, therefore the same emission factor was used.

Maize silage was produced in the farm. Alternatively, it was used as feed or sold. When we considered the C footprint per cropped surface, no emissions were accounted in either case.

We assumed grass clippings were co-composted when they were not digested. The co-composting process would have required too much hypothesis and efforts regarding the low proportion that this waste represent in total AD inputs. Therefore, the GHG emissions due to alternative grass clippings treatment were estimated with general emission factors for composting process from Base Carbone. We considered that the treatment of grass clippings emitted 3.1 kg CH_4 t^{-1}, 0.063 kgN_2O t^{-1}, and saved 26 kgCO_2eq t^{-1} due to fertilizing and soil amendment properties of composts. In total, the treatment of grass clippings emitted 77 kgCO_2eq t^{-1}.

A possible alternative treatment of whey permeate and wastes from yeast production is their use as co-products in animal feeding (ORDIF, 2016). Therefore, the digestion of these inputs implied the production of additional similar feed which emitted some GHG emissions therefore computed in the AD scenarios. We used emission factors of 1.89 kgCO_2eq kg^{-1} for whey permeate (GES'TIM+), and 0 kgCO_2eq kg^{-1} for wastes of cereal storage and wastes from yeast production (no available information).

According to our knowledge, the alternative uses of grease could vary (e.g. animal feed, use agro-industrial processes, incineration), however we found no reliable information about their treatment. Therefore, we assumed that their alternative treatment emitted no GHG emissions.

The transport of external wastes from location of their production to the farm was not considered, we assumed that these wastes were likely transported on similar distance when digested or treated with their reference treatment.

6.2.4.5 Compensating differences in agricultural production

When the agricultural activity considered a similar crop production, new GHG emissions had to be accounted to equalize the crop production in all scenarios, in addition to GHG emissions described in the sections 6.2.4.2 to 6.2.4.4. The difference of crop production between the scenarios had three origins. First, the different scenarios showed similar, however slightly different yields. To consider these differences, we simulated a farm without any animal effluents using only synthetic fertilizers. GHG emissions were computed as in section 6.2.4.2. We used the ratio of GHG emissions per ton of exported biomass (sum of all crops) to consider the possible required additional crop production. Secondly, the direct spreading of sewage sludge can affect the yields in the farm where they are applied. This difference in crop production was considered as describe previously. Lastly, the quantity of digested maize silage was deduced from the farm agricultural production. This deficit in crop production was taken into account when the agricultural activity was considered for a similar crop production.

6.3 Results

6.3.1 Agronomical use of EOM and biogas production

In the "no AD" scenario, the quantities of cattle slurry and manure were rather low regarding the cropped area. Therefore, the EOMs only slightly accounted for the total fertilization (Supplementary Material 7). AD (ADb scenario) resulted in much higher application of EOMs in the farm (Table 6.2). The EOMs characteristics were also different (Table 6.2): liquid digestate had higher TN and TAN contents than cattle slurry, and solid digestate had lower VS and TN contents than cattle manure. In ADb scenario, the digestates accounted for an important part of wheat and maize fertilizations and were applied on grassland in small quantities (Supplementary Material 7). Because of the higher surface of wheat, the majority of liquid digestate was applied on wheat (Supplementary Material 8). Using climatic year 2010, yields were constant with the time, and were similar between ADb and "no AD" scenarios (Supplementary Material 9); only maize yields varied between the different rotations in both scenarios (Supplementary Material 9). Without AD ("no AD" scenario), the average yields were 7.1 tDM ha^{-1}, 2.9 tDM ha^{-1}, 16.8 tDM ha^{-1} and 7.2 tDM ha^{-1} for wheat, rapeseed, maize silage, and grassland, respectively. In ADb scenario, the average yields were 7.4 tDM ha^{-1}, 3.0 tDM ha^{-1}, 15.3 tDM ha^{-1} and 6.9 tDM ha^{-1} for wheat, rapeseed, silage maize silage, and grassland, respectively. The slightly higher rapeseed yield in ADb scenario was due to the higher N input in autumn with solid digestate compared to the one with manure in "no AD" scenario. The slightly higher maize and grassland yields in "no AD" scenario were attributed to the lower N efficiency of digestate compared with synthetic N fertilizer. When simulating the other climatic years, the crop yields were also constant with time and similar in both scenarios (Supplementary Material 9). In ADb scenario, the liquid digestate was preferentially applied on wheat or before maize sowing, but because of the larger area cropped with wheat, two third of the digestate was applied on wheat. Overall, we simulated a consistent crop management, and the yields were similar enough between both reference scenarios and along 20 years so that we can consistently interpret the simulation results.

In the ADb scenario, biogas production was 463.10^3 Nm^3, including 236.10^3 Nm^3 of methane (51%). The electricity production was 872.10^3 kWh.

Table 6.2 – EOM characteristics after modelling.

EOM	Scenarios	Flow (tFW year^{-1})	TS (%FW)	VS (g kgFW^{-1})	C (g kgFW^{-1})	TN (g kgFW^{-1})	TAN (g kgFW^{-1})
Cattle slurry, fresh	All	845	7.4%	61	30	3.2	1.2
Cattle manure, fresh	All	532	26.3%	221	111	6.7	0.8
Cattle slurry, stored	no AD	822	4.9%	35	18	2.7	1.4
Cattle manure, stored	no AD	308	32.5%	253	126	8.3	0.7
Liquid digestate	ADb, I, Y, Y+I, C, C+I, D20, D40	3793	4.7%	29	15	4.8	3.3
	SC	3793	4.7%	29	15	3.5	2.0
	EI0	1415	5.8%	37	18	3.7	1.7
	EI30A	1811	6.1%	37	19	4.9	2.9
	EI30V	1872	5.0%	34	17	4.1	2.4
	EI70A	3485	6.5%	37	20	6.9	4.7
Solid digestate	ADb, I, Y, Y+I, C, C+I, D20, D40	403	17.8%	133	69	6.5	1.9
	SC	403	17.8%	133	69	6.5	1.9
	EI0	149	22.1%	175	82	5.8	1.0
	EI30A	190	23.2%	175	86	7.1	1.6
	EI30V	198	20.4%	156	77	6.1	1.3
	EI70A	364	25.0%	174	92	9.5	2.7
Raw digestate	PS	4218	6.5%	44	24	5.1	3.4

6.3.2 C fluxes in the farm with or without on-farm AD

In this section, we present the C fluxes of the farm (no system extension) for scenarios "no AD" and ADb, using climatic year 2010 (Figures 6.2 and 6.3). In the "no AD" farm, the fresh effluents are degraded during storage inducing some C losses (CO_2 or CH_4) (Figure 6.2). The amount of C in stored slurry was low, leading to very low methane emissions. The stored effluents are applied in the soil. The input of C in soil from EOM represented 8% of the C from returned crop residues that were simulated with STICS. Cattle manure was a more important source of C than cattle slurry (Figure 6.2). The majority of the organic matter is degraded and emitted in the atmosphere on the form of CO_2. A small proportion of C remains in the SOM.

The introduction of AD (ADb) led to large import of C with organic wastes (+327% compared to "no AD") of the C from cattle effluents (Figure 6.3). A large proportion (74%) of the waste C was turned into biogas. ADb decreased the C emissions (CO_2 and CH_4) during EOM storage (-20%), but the total C losses during digestate treatment considering the additional biogas leakage were similar to "no AD" scenario. In the ADb scenario, the C in stored liquid digestate was much higher (+130%) than the C in the slurry in "no AD" scenario (Figures 6.2 and 6.3). Despite the reduced CH_4 emission factor, the total CH_4 emission from EOM storage was thus higher after the co-digestion of the effluents (+49%) (Figure 6.4.A). CH_4 emission was even increased by biogas leakage, which represented half of the total CH_4 emission in the farm.

Despite the high difference in C import that entered the farm with the external wastes, the C applied to soils with EOM was only 62% higher in ADb scenario than in "no AD" scenario (Figures 6.2 and 6.3). On the contrary to no AD, the very large amount of liquid digestate make it a greater source of C than solid digestate in the ADb scenario (Figure 6.3). This influenced the soil C content in the different plots where the organic matter was applied: the amount of C applied with EOM increased by 14%, 36%, and 173% in the fields with the maize-wheat-grassland rotation, the maize-wheat rotation, and the rapeseed-wheat-wheat rotation, respectively (Supplementary Material 8). Therefore, in ADb scenario, the SOC storage increased by c.a. 0.0 tC ha^{-1} in the plots with grassland, and 1.0 tC ha^{-1} in the others plots, compared to no AD. Indeed, the wheat fields benefitted from regular and high inputs of liquid digestate (Figure 6.5.A). As a results, the SOC stock in the cropped soils increased of 0.6 tC ha^{-1} in 20 years on average in ADb scenario, compared to "no AD" scenario (Figure 6.5.A).

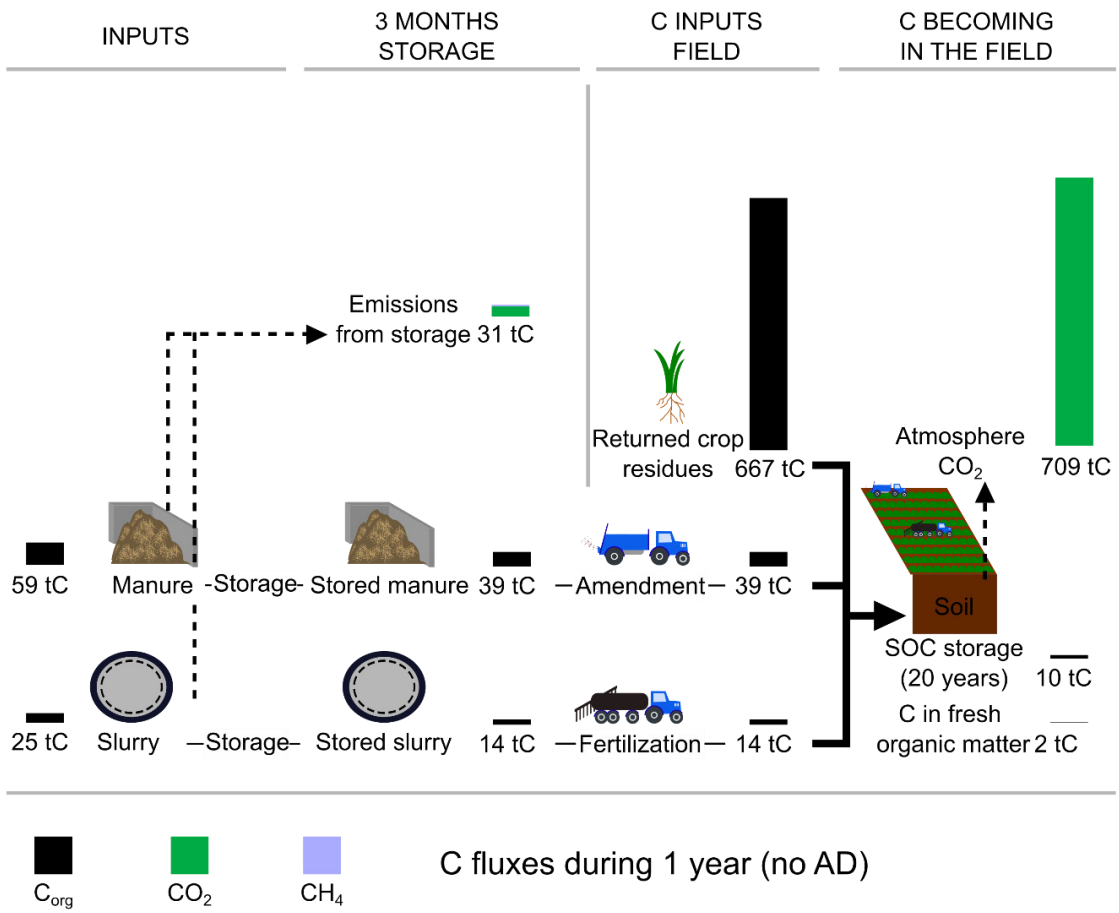

Figure 6.2 – C fluxes at the farm scale, "no AD" scenario. Flows of C are the average yearly flow on the 20 year period, expressed in tC year-1, it includes organic C, CO2, and CH4. Bars height are proportional to the flows. The colors of the bars represent the form of the C. Corg: organic C. Climatic year is 2010. The cropped area is 299 ha.

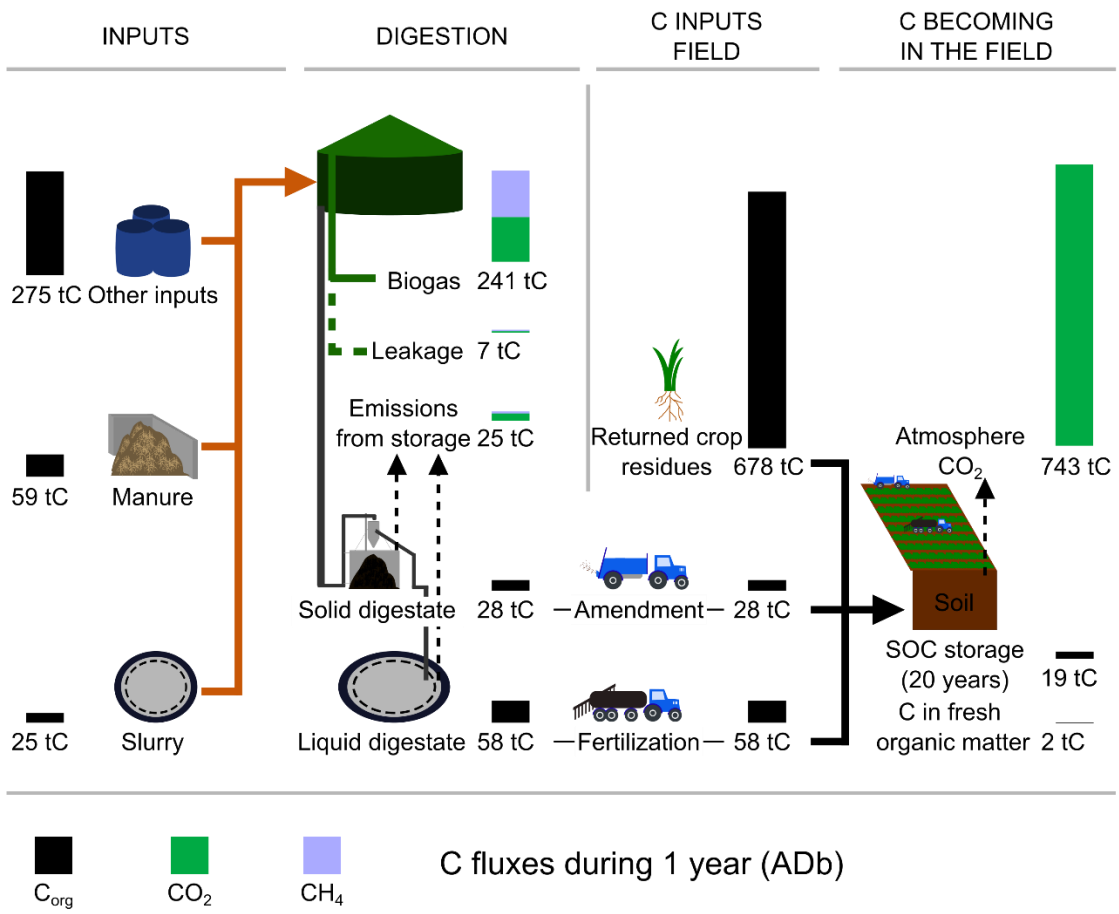

Figure 6.3 – C fluxes at the farm scale, in the baseline scenario with on-farm AD (ADb). Flows of C are the average yearly flow on the 20 year period, expressed in tC year^{-1}, it includes organic C, CO_2, and CH_4. Bars height are proportional to the flows. The colors of the bars represent the form of the C. C_{org}: organic C. Climatic year is 2010. The cropped area is 299 ha.

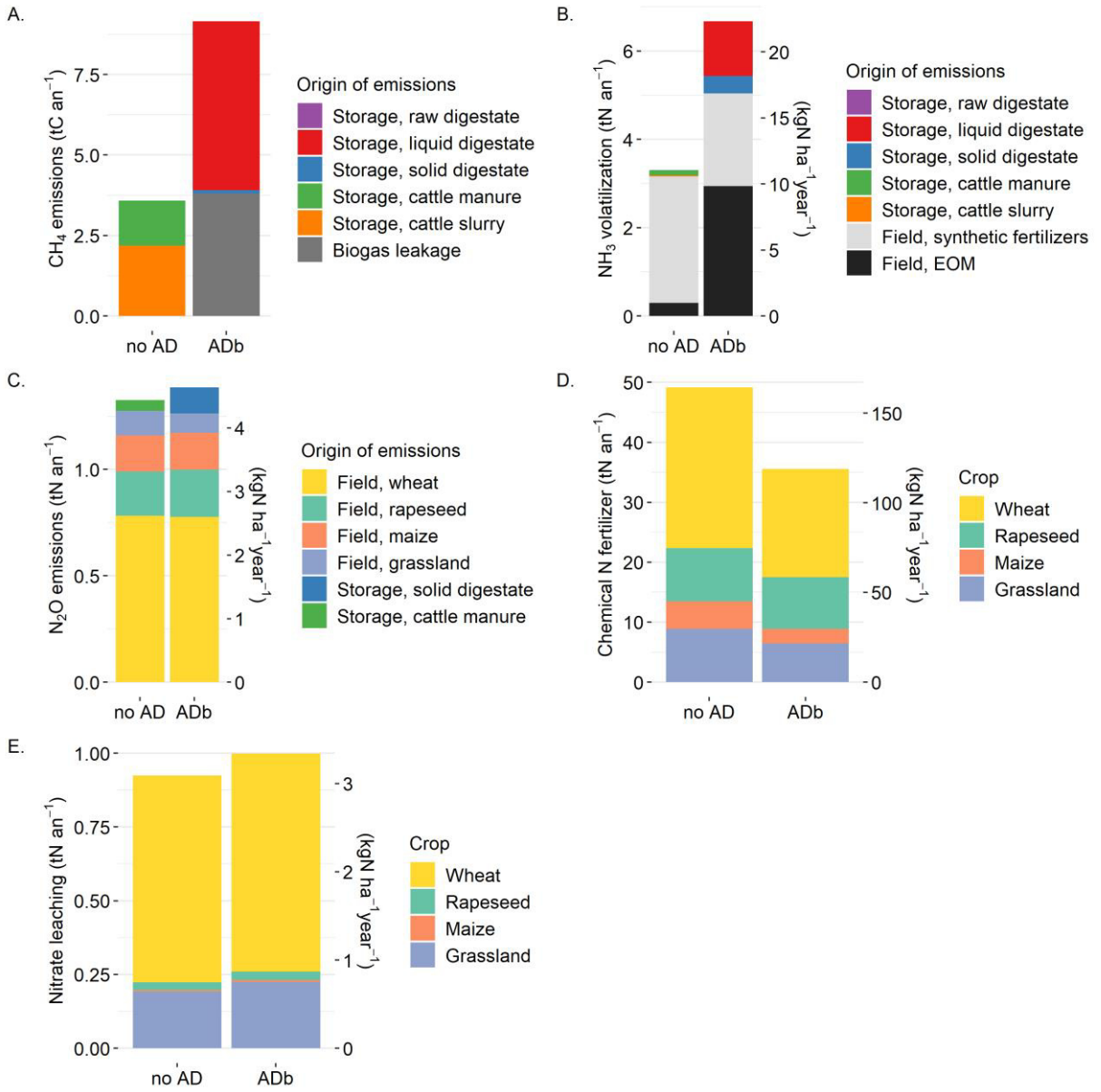

Figure 6.4 – Comparison of environmental indicators of the farm from ADb and "no AD" scenarios, using climatic year 2010. The indicators consider the emissions within the farm, without any system extension. Each indicator is the yearly indicator averaged along the 20 years of simulations A) Total CH_4 emissions, according to the source of emission. B) Total NH_3 emissions, according to the source of emissions. C) Total N_2O emissions, according to the source of emission. D) Use of chemical N fertilizers, according to crop where these fertilizers are applied. E) Nitrate leaching, according to the crop where leaching occur.

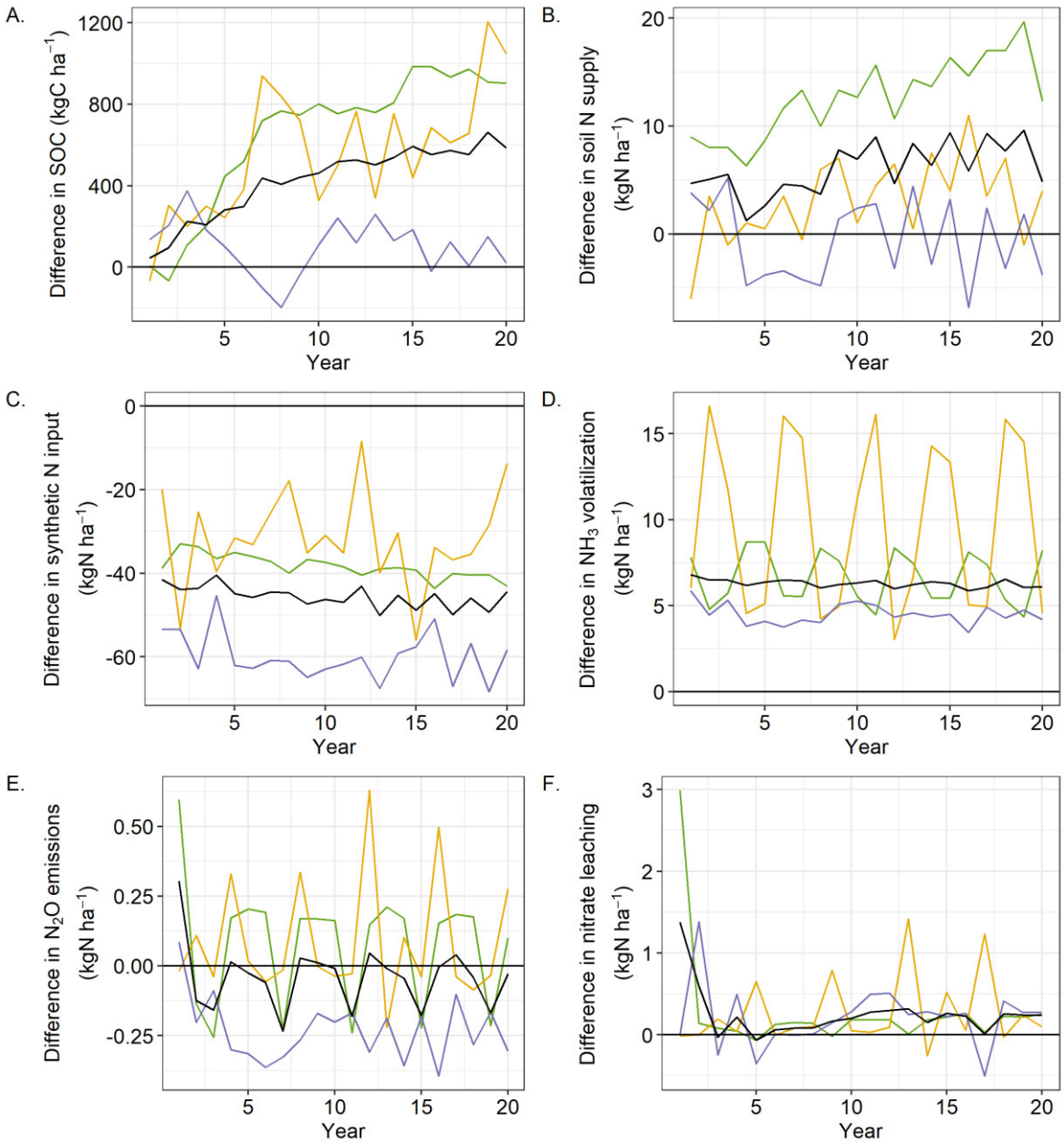

Figure 6.5 – Simulated difference of indicators between ADb and "no AD" scenarios, across the time. A positive value indicate that indicator is higher in the fields of ADb scenario compared to the one of "no AD" scenario. Black lines represent the average difference of impacts in the entire farm, colored line considered one type of field each (similar crop rotation). Green: Rapeseed – Wheat – Wheat. Yellow: Maize – Wheat. Blue: Maize – Wheat – 3-year grassland. Climatic year is 2010. A) SOC stocks; B) soil N supply; C) synthetic N input; D) ammonia volatilization; E) N_2O emissions; F) nitrate leaching.

6.3.3 N fluxes in the farm with or without on-farm AD

In this section, we present the N fluxes of the farm (no system extension) for scenarios "no AD" and ADb, using climatic year 2010 (Figures 6.6 and 6.7). In the "no AD" farm (Figure 6.6), a part of the N in the cattle effluents is lost during storage. Most of the simulated N losses were simulated to be N_2 emissions originating from the organic matter decomposition. The cattle effluents were applied on the soil, they represented a very low fraction (9%) of the N inputs in the fields. Most of the applied N (86%) is taken and exported by the crops. Alternatively, N can be emitted in the environment (NO_3^-, N_2, N_2O, NH_3) or stored in the soil (Figure 6.6).

As for C fluxes, the introduction of AD led to the import of N from organic wastes. This external N import represented 596% of the N from cattle effluents (Figure 6.7). The total N losses during EOM storage were not increased with the introduction of AD (Figures 6.6 and 6.7). In the AD scenario, the ammonia volatilization during storage was multiplied by 10 compared to "no AD" (Figure 6.4.B), due to higher TAN quantities in digestates compared to cattle effluents (Figures 6.6 and 6.7), and higher emission factors. The N_2O emission during solid EOM storage increased by 142%, because these emissions were computed from TAN content that was higher for solid digestate than cattle manure (Figure 6.4.C).

The N applied with solid digestate was similar to the amount of N applied with cattle manure. However, the imports of N induced a very high amount of N inside liquid digestate (Figures 6.6 and 6.7). Therefore, the application of liquid digestate induced a reduction in the use of synthetic N fertilizers of 46 kgN ha^{-1} $year^{-1}$ on average, which was a 28% reduction (Figures 6.4.D, 6.6 and 6.7). This reduction mostly occurred on wheat plots, which received the highest quantities of liquid digestates. After 20 years, due to SOM storage the soil N supply tended to increase in ADb scenario compared to "no AD" scenario, the increase was about 10 kgN ha^{-1} $year^{-1}$. This effect was only apparent in the fields under rapeseed-wheat-wheat rotation (Figure 6.5.B). As a result, the consumption of synthetic N fertilizers also slightly decreased during the 20 years (Figure 6.5.C). Due to the use of low quantity of slurry, ammonia volatilization in the fields in "no AD" scenario mainly originated from mineral fertilizers and was low. AD increased the ammonia volatilization in the fields by 56% (+6.3 kgN ha^{-1}) after the introduction of AD in the farm (Figures 6.4.B, 6.6 and 6.7). The NH_3 volatilization did not evolve during the time (Figure 6.5.D). The field N_2O emissions were comparable between both scenarios (-1% in ADb compared to "no AD") and did not evolve with time (Figures 6.4.C and 6.5.E). Nitrate leaching were low (inferior to 0.5 kgN ha^{-1}) (Figures 6.6 and 6.7) and did not evolve with time (Figures 6.4.E and 6.5.F).

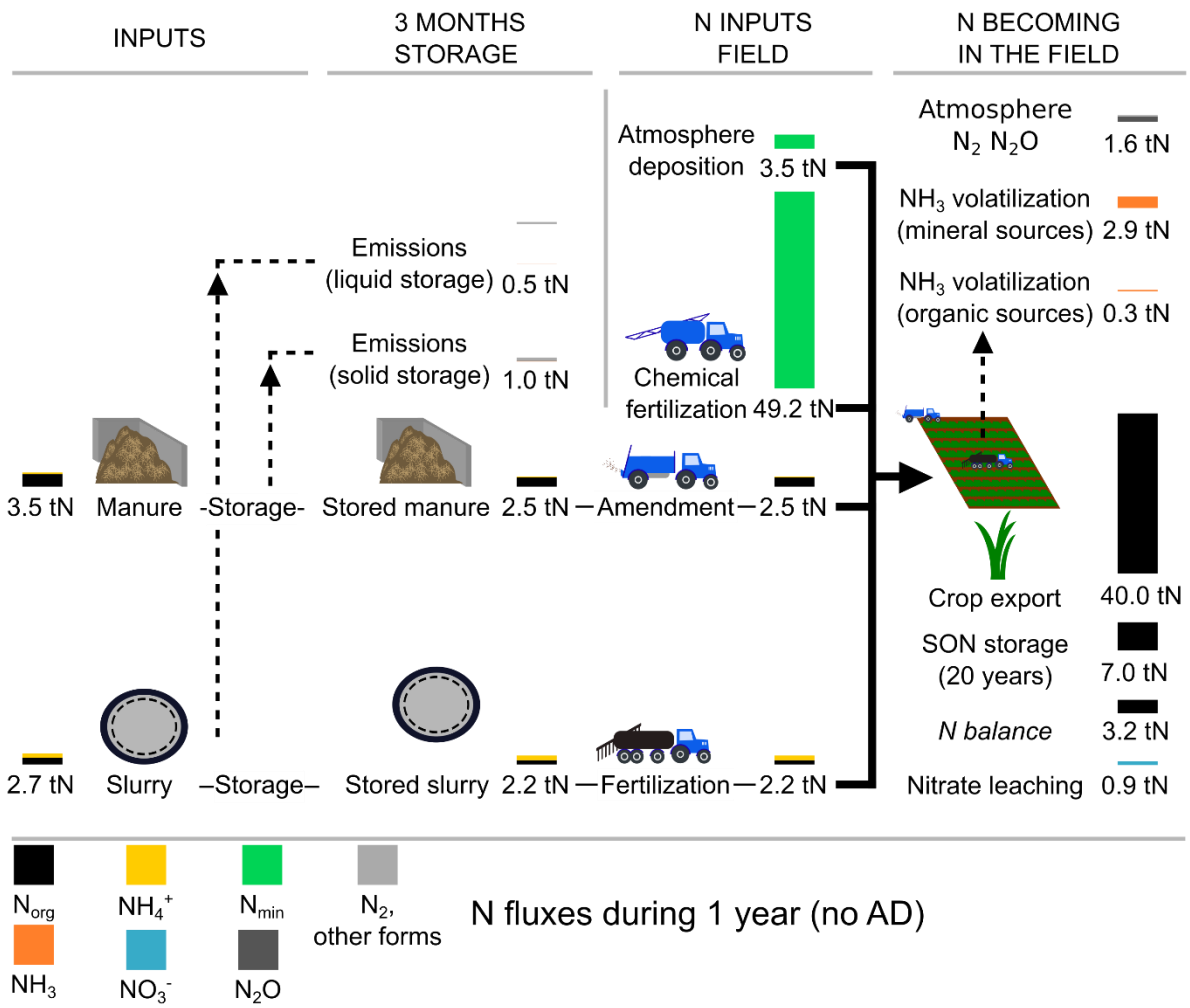

Figure 6.6 – N fluxes at the farm scale, "no AD" scenario. Flows of N are the average yearly flow on the 20 year period, expressed in tN year^{-1}. Bars height are proportional to the flows. The colors of the bars represent the form of the N. N_{org}: organic N. N_{min}: mineral N. Climatic year is 2010. The cropped area is 299 ha.

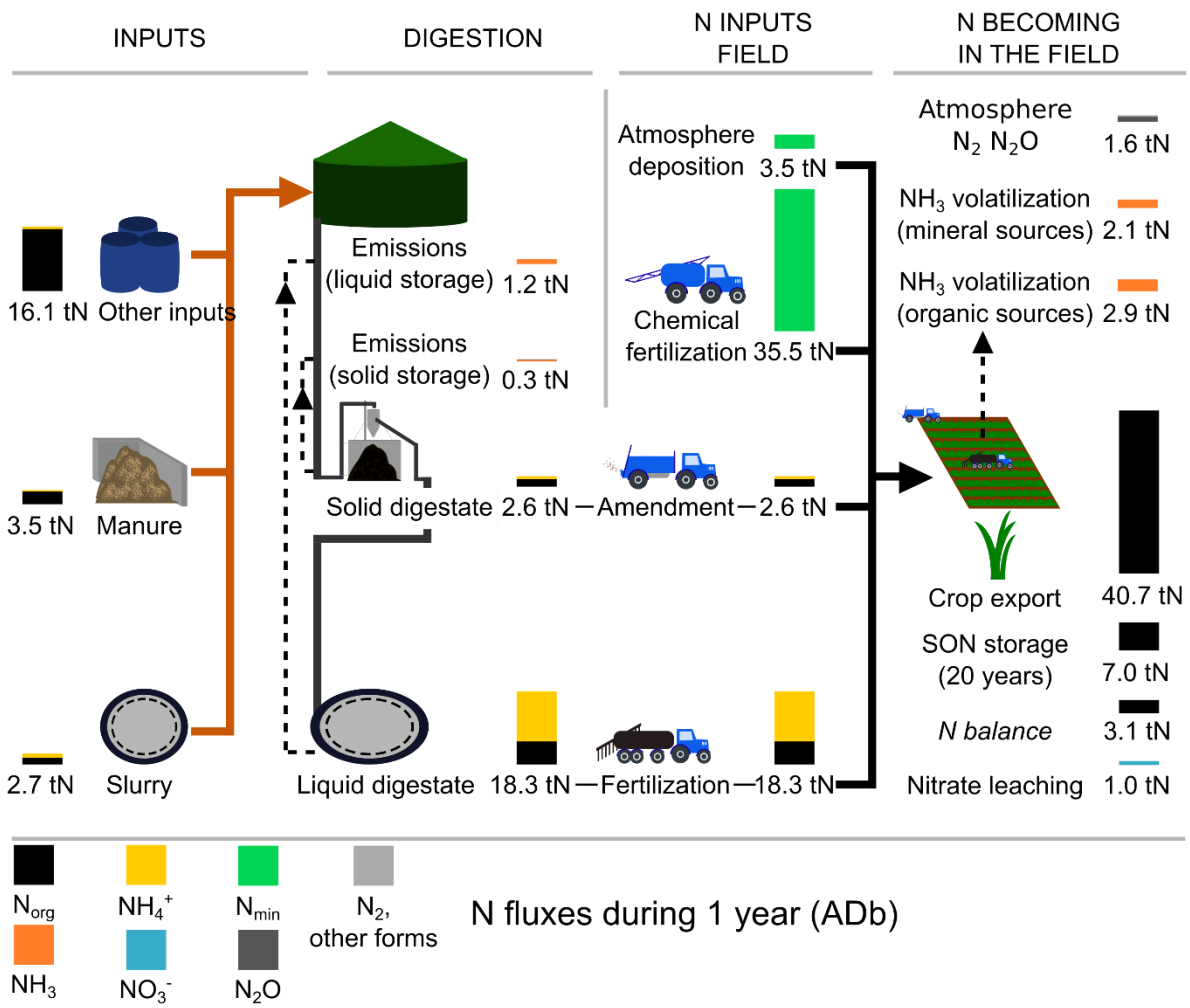

Figure 6.7 – N fluxes at the farm scale, ADb scenario. Flows of N are the average yearly flow on the 20 year period, expressed in tN year^{-1}. Bars height are proportional to the flows. The colors of the bars represent the form of the N. N_{org}: organic N. N_{min}: mineral N. Climatic year is 2010. The cropped area is 299 ha.

Considering all sources of ammonia emissions, ADb scenario was subject to 102% higher emissions than "no AD" scenario (Figure 6.4.B). About two-third of the NH_3 emissions occurred in the fields and one-third during EOM storage. However, half of the surplus NH_3 emission in ADb compared to no AD originated from EOM storage, and half of it originated from field application. Considering all sources of N_2O emissions, the difference of emissions between both scenarios was mostly due to the difference of N_2O emissions during EOM storage; emissions were higher in ADb scenario than in "no AD" scenario (+59 kgN_2O-N, i.e. +0.2 kgN_2O-N ha^{-1}) (Figure 6.4.C).

6.3.4 Influence of climatic year on C and N fluxes in the farm

The simulated climatic years slightly influenced the effects of the introduction of AD, without changing the conclusion described in sections 6.3.2 and 6.3.3 (Supplementary Material 10 and 11). In all climatic years, we compared ADb scenario to "no AD" scenario:

- AD always increased the average SOC in the in ADb compared to "no AD" scenario (from 0.4 tC ha^{-1} to 1.0 tC ha^{-1} in 20 years, i.e. from 114 tC to 296 tC). The years with the lowest yields induced a lower production of crop residues and a lower C returned to soil. Therefore, using those years, the relative importance of organic inputs from EOMs was higher and difference between both scenario was more accentuated.
- AD always decreased the use of synthetic N fertilizers (from -43 to -49 kgN ha^{-1} $year^{-1}$, i.e. from -25% to -33%)
- AD always increased NH_3 volatilization in the field (from +4.9 to +7.7 kgN ha^{-1} $year^{-1}$, i.e. +42% to +63%). It was correlated to temperature at application date. AD increased total NH_3 volatilization (from +80% to +102%) (only the volatilization in the field was influenced by climate in our simulations).
- AD slightly decreased the field N_2O emission (from -0.0 to -0.1 kgN ha^{-1} $year^{-1}$, i.e. from -1% to -4%). AD increased total N_2O emissions (from +40 to +65 kgN_2O-N $year^{-1}$) (only the emissions in the field was influenced by climate in our simulations).

The increase in soil N supply was similarly low using all six climatic years (from 0 to10 kgN ha^{-1} $year^{-1}$ after 20 years). In 2004, 2007, 2010, and 2011, total nitrate leaching was low (< 4 kgN ha^{-1}), and the difference in nitrate leaching between "no AD" and ADb scenarios was low (< 0.5 kgN ha^{-1}). In 2003, leaching was more important (9 kgN ha^{-1}) because of important rains between late winter and early spring, but the difference in nitrate leaching between "no AD" and ADb scenarios was also low. In 2006, rain was important, in particular from September to February, total nitrate leaching was high (18 kgN ha^{-1}), and nitrate leaching was higher in ADb compared to "no AD" scenario (+1.8 kgN ha^{-1}, i.e. +11%).

6.3.5 Influence of external inputs *on C and N* fluxes *in the farm*

This section concerns the influence of the import of different quantities and types of wastes on the C and N fluxes within the farm, without extending the system (Figure 6.8).

Compared to "no AD" scenario, the mono-digestion of the same quantities of cattle effluents (scenario EI0) decreased the use of synthetic N fertilizers (from -2.6% to -5.1% compared to no AD depending on climatic year). It also produced a small amount of biogas, but slightly increased NH_3 emissions (from +8.7% to +12.9%) and decreased SOC storage (-0.3 to -0.6 tC ha^{-1} in 20 years).

The municipal and agricultural wastes (scenarios EI30A and EI70A) were richer in C and N than the sewage sludge and various wastes (scenarios EI30V and ADb). Therefore, the quality of the digestates were different in those scenarios (Table 6.2): the digestate coming from animal effluents and agricultural wastes had higher TS, TN and TAN contents. For all indicators, the scenarios where the digester used 30% of external wastes (EI30V and EI30A) were in intermediate position between mono-digestion (EI0), and the scenario where the digester used 70% of external wastes (ADb and EI70A, respectively) (Figure 6.8). The quantity of imported nutrients in external wastes had a much greater impact than the change in digestate characteristics. The increase of N input induced an almost linear increase in net NH_3 emissions, and net substitution to synthetic N fertilizers compared to "no AD" scenario. In 2006 and 2007, which were two years with high rain in spring, nitrate leaching could also increase with the imports of higher quantities of N. In the two concerned scenarios (EI70A, and ADb), the digestate had high TAN content, and the minimal dose of 30 t ha^{-1} induced a high TAN input at one date, and therefore a higher risk of leaching. However, the net difference of nitrate leaching compared to no AD, which used mainly synthetic N fertilizers, remained relatively low (5 kgN ha^{-1} in the worst case). Similarly, the increase of C input induced a linear increase in biogas production and SOC storage. However, we also highlighted some effect of the change in digestate quality. The use of agricultural wastes led to a slightly higher NH_3 emissions for similar N import, which was attributed to the higher TS and TAN content (Table 6.2). Because the use of agricultural wastes was more easily turned into biogas, more imports of C were required to reach a similar net SOC storage.

Altogether, we estimated that, on average, depending on the scenarios, 64% to 80% of the N import with external wastes was used to substitute synthetic N fertilizer, 18% to 23% was volatilized as NH_3, 2% to 5% was stored in the SOM. The (exported N):(input N) ratio slightly decreased with the increase of N imports in external wastes, showing that the N imported with external wastes was used less efficiently than the one applied with synthetic fertilizers. However, the decrease was small compared to the variation of this ratio due to meteorological variations. We estimated that, on average, 75% to 80% of the C import with external wastes was valued as biogas, 5% to 7% were stored in the soil, and 1% to 2% were emitted as CH_4.

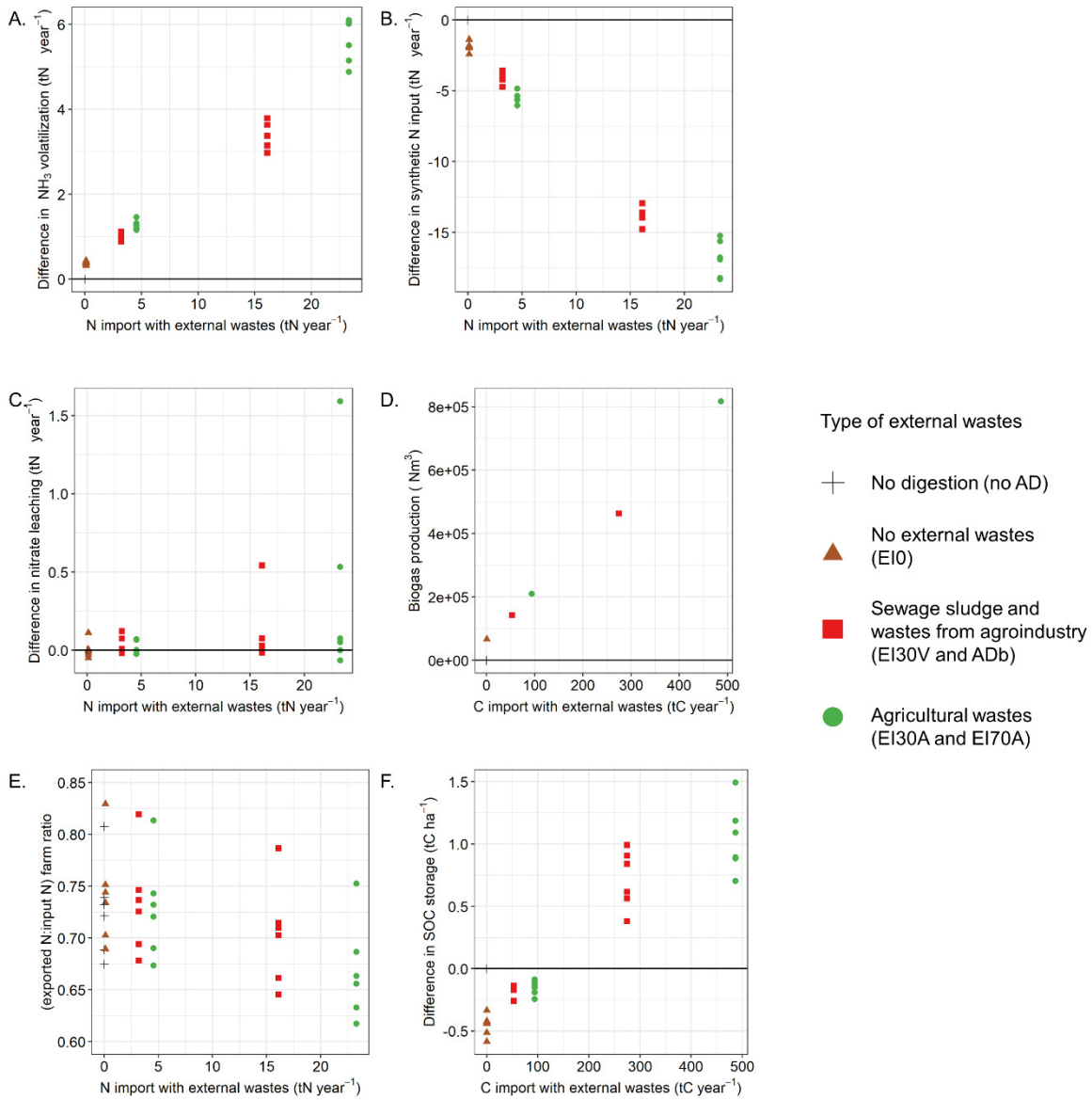

Figure 6.8 – Consequences of the digestion of cattle effluents with different types and quantities of external wastes. Each point represents the net impact of one scenario for one climatic year (each scenario is compared to "no AD" scenario). For each indicator, the yearly average value on the whole farm is considered, without extending the system. The six selected climatic years were investigated for each scenario. A) Difference in NH_3 emissions at the farm scale. B) Difference in synthetic N fertilizer use. C) Difference in nitrate leaching. D) Biogas production. E) Ratio of exported N in crop to total imported N (synthetic N fertilizers, cattle effluents, imported organic wastes). F) Difference in average SOC storage.

The absolute value of the indicators varied depending on the simulated climatic year, but the conclusions and the ranks of the different scenarios were unchanged.

More details are shown in Supplementary Material 12.

6.3.6 Influence of storage conditions and agronomical management of EOM *on C and N* fluxes *in the farm*

The impact of different practices on C and N fluxes were investigated within the farm, without extending the system (Figure 6.9). The different practices did not change C content in the digestate, therefore the SOC change was poorly impacted by the change of these practices.

The absence of phase separation (PS scenario compared to ADb scenario) decreased NH_3 emission during EOM storage (-0.6 tN, i.e. –35%), and slightly increased NH_3 volatilization after field application of raw digestate (from +0.6 to +1.1 tN, i.e. from +12% to +19%, depending on the climatic year). Overall, it led to a small increase on total NH_3 emissions (from +0.0 to +0.5 tN, i.e. from +1% to +7%). TAN contents of raw and liquid digestates were similar (Table 6.2). The use of raw digestate allowed to substitute slightly higher amount of N, despite the higher NH_3 volatilization after field application. Indeed, the ammonia emission during digestates storage was lower in PS scenario than in ADb scenario, and no TAN was exported toward the solid phase. The digestate was also applied on slightly different crops and dates than liquid digestate (Supplementary Material 7). As a result, there was more TAN applied with raw digestate than with liquid digestate (Supplementary Material 8). Because no solid EOM were stored, no N_2O emissions were simulated (-0.12 tN_2O-N). The N_2O emissions in the field were increased or decreased depending on the simulated climatic year (from -0.04 tN_2O-N to +0.06 tN_2O-N). The mineralization of organic N from raw digestates in winter slightly increased the risk of nitrate leaching, and increase N_2O emissions in case of a warm spring season. CH_4 emission during EOM storage decreased by 16% due to lower emission factor for raw digestate than for liquid digestate.

The absence of liquid storage cover (SC scenario compared to ADb scenario) strongly increased total ammonia emissions (from +3.8 to +4.2 tN, i.e. from +51% to +65%). This was due to the increase of ammonia emission during liquid digestate storage (+4.8 tN, i.e. +294%). It was from far not compensated by the slight decrease in NH_3 volatilization in the field after digestate applications induced by the lower TAN content. This lower TAN content in liquid digestate decreased the substitution of synthetic fertilizers: the use of chemical fertilizer were 13% to 18% higher (+4.2 to +5.6 tN). It also decreased the (exported N):(input N) ratio.

The use of different doses of digestates (D20 and D40) only marginally affected the C and N balance of the farm.

The use of digestates in priority for maize and grassland (scenario C) instead of wheat and maize (scenario ADb) did not affect the C and N fluxes at the farm scale. Only the use of synthetic fertilizers

were slightly higher (from -0.2 to 3.7 tN, i.e. from –1% to +9%), because of a less advantageous use of liquid digestate.

The logistical constraints that forced the application of digestate at four seasons during the year (Y) moderately influenced the N fluxes at the farm. Compared to a target agronomical use of digestate (ADb), there were slightly more ammonia volatilization in the field (from -0.2 to +0.7 tN, i.e. from -4% to +14%), and therefore at the farm scale (from -3% to +11%). The N_2O emissions in the field (from +0.08 to +0.13 tN, i.e. from +6% to +17%) and the use of synthetic N fertilizers (change of application crops and dates) (from +4.9 to +6.6 tN, i.e. from +12% to +22%) were also slightly increased. The change in plots where digestate was used could slightly increase or decrease nitrate leaching, depending on the climatic year.

The injection of liquid digestate and fast burying of solid digestate (I versus ADb scenarios) decreased NH_3 volatilization from EOM from 12% to 15%, and total NH_3 emissions from 9% to 11%. This low impact was due to the fact that most of the liquid digestate was applied on wheat and could not be injected. The fast burying of solid digestate was efficient, but only concerned a small proportion of the total NH_3 emissions. The injection was more efficient to decrease NH_3 volatilization when the digestate was applied in priority on crop where the injection is possible (C+I versus C from -9% to -16%, and Y+I versus Y from -21 to -25%).

The absolute value of the indicators varied importantly depending on the simulated climatic year. The variations of the indicators due to climate could be more important than the difference between the different practices. However, the conclusions and the ranks of the different scenarios were unchanged from one year to another.

More details are shown in Supplementary Material 12.

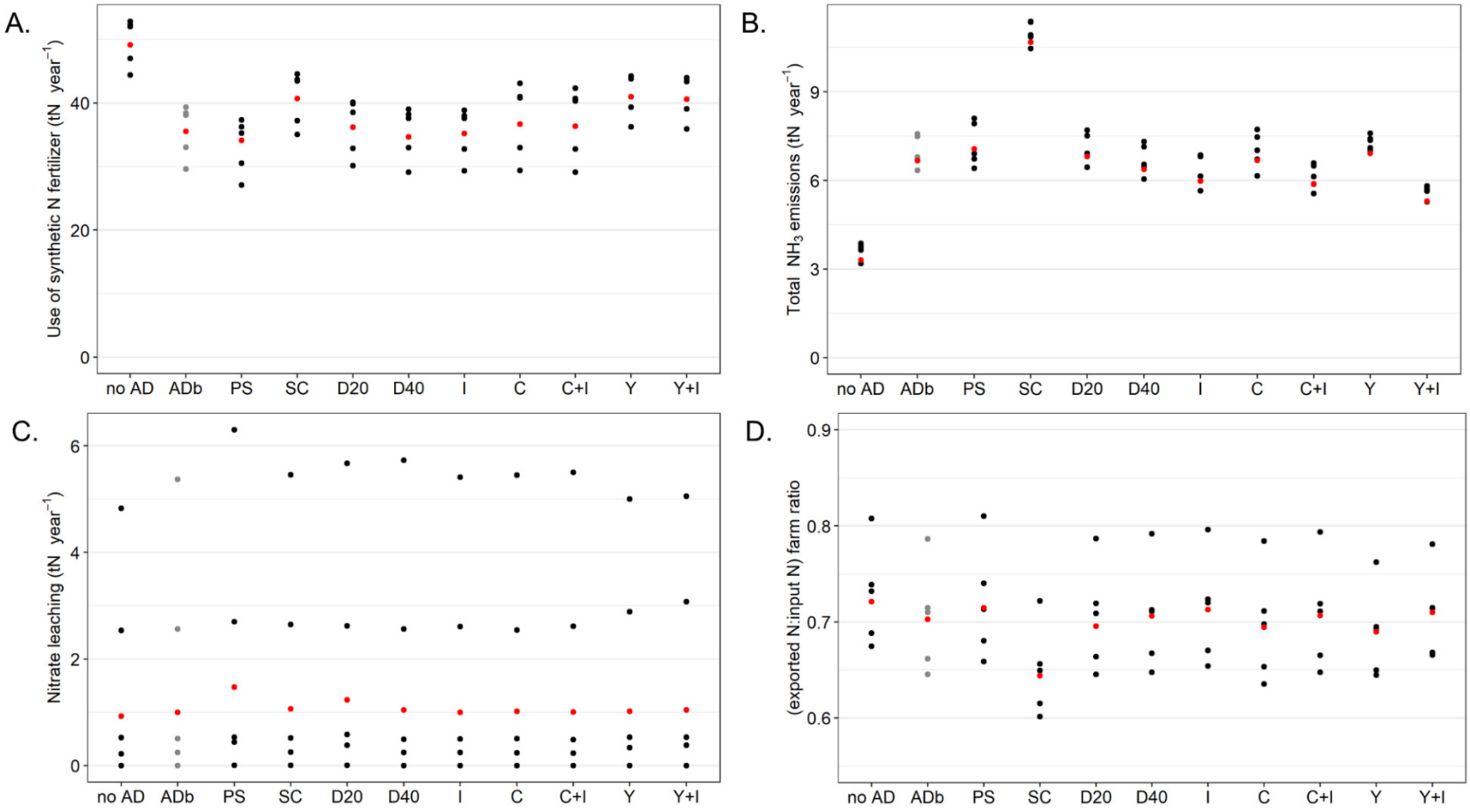

Figure 6.9 – Indicators for different scenario of AD at the farm scale, without extending the system. Average indicators on the 20 years simulation period were presented. Each dot represent one scenario using one climatic year, the six climatic years were investigated. ADb scenario is represented with grey dots, simulations using climatic year 2010 are represented with red dots. A) Substitution to synthetic fertilizers, compare to "no AD" scenario. B) Total NH_3 emissions at the farm. C) Total nitrate leaching at the farm. D) (exported N):(input N) ratio.

6.3.7 C footprint of diverse scenarios compared to "no AD" scenario in extending the system

This section presents the C footprint of the simulated scenarios, considering the extended system (section 6.2.4). Total GHG emissions of ADb and "no AD" scenarios using climatic year 2010 is detailed in Table 6.3 for similar cropped area and similar crop production. When we considered the same cropped area, the main source of increase in net GHG emissions induced by AD were biogas leakage and the increase in GHG emissions during EOM storage. The main sources of net decrease in GHG emissions were the substitution of fertilizers (N at first, but also P and K), the net decrease of GHG emissions due to waste treatment, the decreased electricity consumption due to biogas production, and SOC storage. The change of GHG emissions in the field and in GHG emissions due to EOM transport and spreading (machinery) were less important. Considering a similar cropped area, AD induced a net increase of farm GHG emissions (+26 tCO_2eq, i.e +2.1% of the total emissions of "no AD" scenario). The crop production was lower in ADb scenario compared with "no AD" scenario, because of slightly lower yields and the digestion of a part of silage maize. The sludge increased the crop production in the fields where they are directly applied ("no AD" scenario). Therefore, when we considered a similar crop production, the digestion of sewage sludge also induced GHG emissions in the ADb scenario due to the additional crop production. Therefore, when we considered a similar crop production, AD also induced a net increase of farm GHG emissions (+79 tCO_2eq, i.e. +6.4% of the total emissions of "no AD" scenario).

Using each of the six climatic years, similar conclusions were obtained about the important drivers of the global warming potential of AD (Supplementary Material 11). Considering a similar cropped area, AD induced a net increase of farm GHG emissions from +22 to +67 tCO_2eq, or from +2.1% and +5.2% of the total emissions of "no AD" scenario, depending on the simulated climatic year (similar cropped area). GHG emissions of ADb (similar crop production) ranged from +39 to +79 tCO_2eq, or from 3.6% to 7.1% of the total emissions of "no AD" scenario, depending on the simulated climatic year. The origin of the variations were the SOC storage, the crop production, the emissions induced by the direct spreading of sewage sludge, and, in a lesser extent, the use of synthetic fertilizers.

The mono-digestion of cattle effluents (EI0) decreased the GHG emissions when considering a similar cropped area compared to no AD. These GHG emissions were unchanged by mono-digestion when considering a similar crop production (Figure 6.10). The beneficial effect was mostly due to an improved EOM management. The main sources of GHG emissions increase was the decrease in SOC storage when the effluents were digestes, biogas leakage, and GHG emissions associated to supplementary crop production.

The import of external wastes increased GHG emissions from EOM storage and from biogas leakage, and decreased the GHG emissions from electricity production, net SOC storage, alternative treatment

of wastes, and need of synthetic fertilizers (EI30A, EI30V, EI70A, ADb). AD decreased the net GHG emissions from EOM storage when the import of external wastes were low (EI30A, EI30V), increased them when the import of external wastes were high (EI70A, ADb). AD increased GHG emissions due to a decrease in SOC storage when the import of external wastes were low compared to no AD (EI30A, EI30V), and decreased them when the import of external wastes were high (EI70A, ADb). The digestion of imported wastes increased both biogas production and GHG emissions associated to the management of the additional digestate. Considering our assumptions (valuing of biogaz as electricity, no heat valuing, electric mix based on nuclear energy), it induces a net increase in GHG emissions. Overall, AD either did not influence the total net GHG emissions when the external inputs were low (EI30A, EI30V), and increased them when the external inputs were high (EI70A, ADb). The consideration of the alternative treatment of the imported waste was an important factor of the C footprint.

Interestingly, the total GHG emissions were strongly reduced when there was no phase separation (Figure 6.10). The GHG emissions of PS scenario were lower than the "no AD" scenario (from -97 to -146 tCO_2eq for a similar cropped area, from -34 to -149 tCO_2eq for a similar crop production). This was mainly induced by a strong decrease in GHG emissions during EOM storage. Because no solid EOM were stored, no N_2O emissions were simulated. Additionally, CH_4 emission during EOM storage decreased by 16%. Because of the higher use of synthetic N fertilizer, the absence of cover on liquid digestate (SC) increased the GHG emissions compared to ADb. The GHG emissions compared to "no AD" scenario ranged from +27 to +48 tCO_2eq for a similar cropped area, and from +31 to +54 tCO_2eq for a similar crop production. The use of low digestate dose (D20) increased field GHG emissions compared to ADb, which was attributed to the different dates of application of digestates (higher N_2O emission in the field). The logistical constraints that forced the application of digestate at four seasons during the year (Y) increased GHG emissions due to the increase in field N_2O emissions and to the use of synthetic N fertilizer. This was compensated by a higher crop export, mainly due to grasslands that benefited from high application of digestates: the grassland yield was 9.5 tDM ha^{-1} in climatic year 2010 (6.9 tDM ha^{-1} in ADb scenario). Therefore, the net GHG emissions was higher than ADb scenario when considering a similar cropped area (from +51 to +89 tCO_2eq, compared to no AD), but similar or lower to ADb scenario when considering a similar crop export production(from -74 to +29 tCO_2eq, compared to "no AD").

The high variability in GHG emissions in EI70A and PS scenarios was attributed to the variability of N_2O emissions in the field. In those two scenarios, high TAN inputs were applied with digestate in spring. In case of warm conditions in spring, it induced higher N_2O emissions in the field than the other scenarios. Globally, the absolute value of GHG emissions indicators varied depending on the simulated climatic year, but the conclusions and the ranks of the different scenarios were unchanged.

Table 6.3 – GHG emissions at the farm scale. The simulated climatic year is 2010. GHG emissions are indicated in tCO_2eq. For "no AD" and ADb scenarios, the proportion of total net emissions (similar area) is indicated (r). The difference in GHG emissions between ADb and "no AD" scenarios indicates the net effect of the introduction of AD in the farm; a positive value indicate an increase in GHG emission due to AD.

Origins of GHG emissions	no AD		ADb		Difference in GHG emissions
	tCO_2eq	%	tCO_2eq	%	tCO_2eq
GHG emissions during EOM storage	156	13%	258	20%	102
GHG emissions in the field	549	44%	552	44%	3
SOC storage	-35	-3%	-69	-5%	-34
Biogas leakage (3%)	0	0%	142	11%	142
Electricity consumption	58	5%	15	1%	-43
N fertilizer production	236	19%	170	13%	-66
P and K fertilizer production	18	1%	5	0%	-13
Transport of EOM (5 km)	<1	0%	2	0%	1
Spreading of EOM	8	1%	15	1%	7
Waste treatment, production of digester inputs	137	11%	64	5%	-73
Tillage, plant protection products, treatment, seeding, harvest	111	9%	111	9%	0
Total net emissions (similar cropped area)	1236	100%	1263	100 %	26
Additional crop production (crop-livestock farm)	0	0%	18	1%/	18
Additional crop production (application or not of sewage sludge)	0	0%	32	3%	32
Additional crop production (digested silage maize)	0	0%	2	0%	2
Total net emissions (similar crop production)	1236	100%	1314	104 %	79

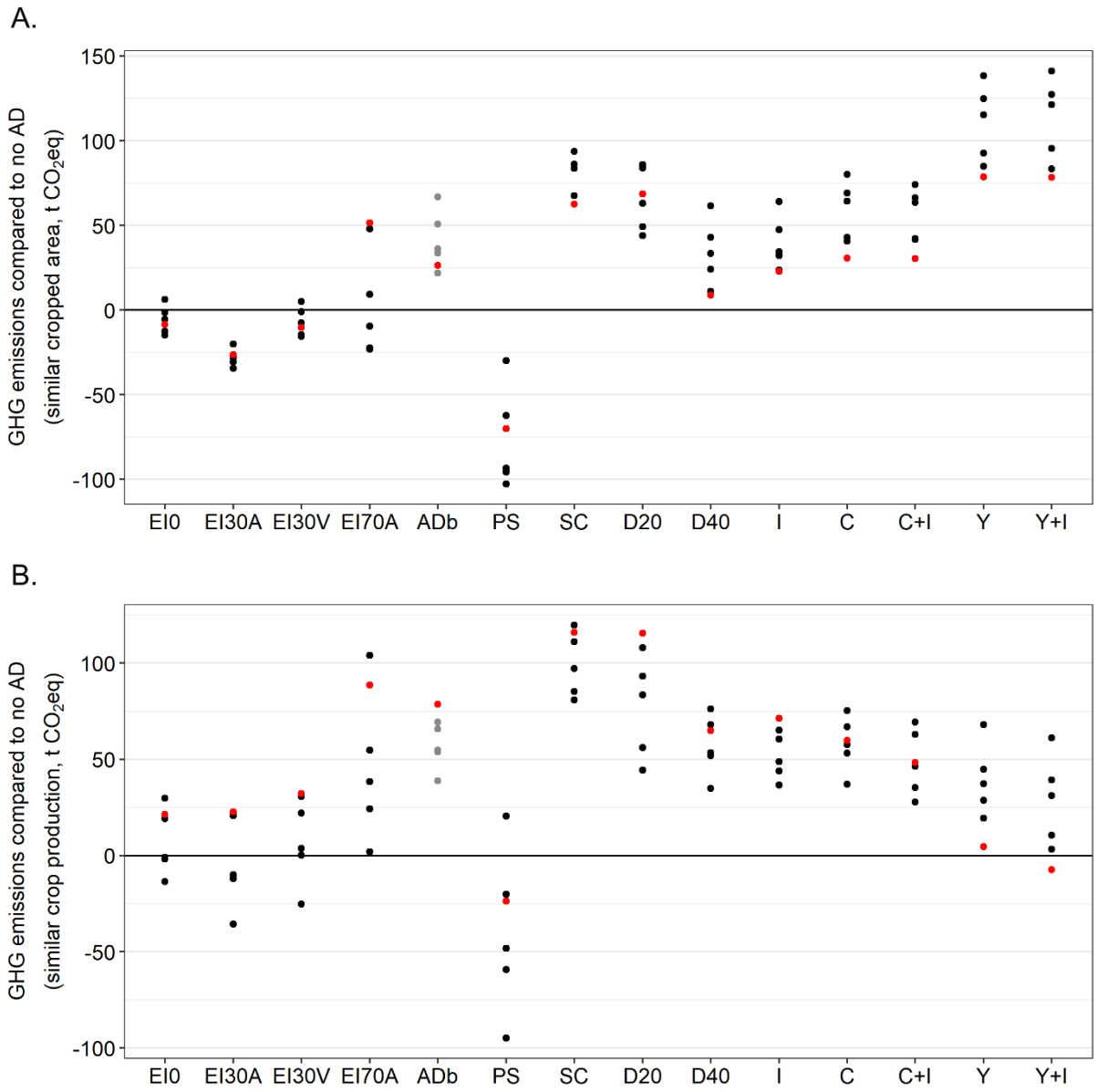

Figure 6.10 – GHG emissions of the different scenarios, compared to "no AD" scenario. A positive value indicates that the GHG emissions is higher in the considered scenario than in the "no AD" scenario. ADb scenario is represented with grey dots, simulations using climatic year 2010 are represented with red dots. A) GHG emissions, similar cropped area. B) GHG emissions, similar crop production.

When the sewage sludge are directly applied instead of being digested, they emit GHG and other pollutants (NH_3, NO_3^-), substitute synthetic N fertilizers, and affect SOC stocks. These effects were accounted in the C footprint of AD, but they have additional impact on environment pollution, use of non-renewable resources, and soil fertility, that was not accounted here. In the sections 6.3.2 to 6.3.6, we only consider the effect of AD inside the farm. Here, we briefly detail the impacts of AD on C and N fluxes outside the farm due to the digestion of sewage sludge. Outside the farm, ADb scenario increased the use of N synthetic fertilizers (from +4.0 to +5.9 tN year^{-1}), decreased SOC stocks (from -209 to -313 tC after 20 years), decreased NH_3 emissions (-0.2 to -0.4 tN year^{-1}), and decreased nitrate leaching (-0.0 to -1.4 tN year^{-1}) (similar cropped area, "no AD" scenario as reference). The comparison between these fluxes outside the farm with the flux inside the farm (sections 6.3.2 and 6.3.3) showed that the large digestion of sewage sludge induced a spatial shift in C and N fluxes: C was stored and synthetic N fertilizers were applied in different soils. Considering the C and N fluxes beyond the farm would induce a slight decrease in the global NH_3 emissions attributed to AD, an important decrease of the global N fertilizer substitution, and a global negative SOC storage.

More details are shown in Supplementary Material 12.

6.4 Discussion

6.4.1 The input of external wastes is a major driver of the C and N fluxes in the farm

The effects of the mono-digestion of cattle effluents were relatively small: decrease in the use of synthetic N fertilizer, decrease of N_2O and CH_4 emissions during EOMs storage, decrease in SOC storage, and increase in NH_3 emission. On the contrary, the import of organic waste was a major driver of C and N fluxes at the farm scale. The import of C mainly affected biogas production, SOC storage, and CH_4 emissions during digestates storage. The import of N firstly decreased the use of synthetic fertilizer, but increased the ammonia volatilization (during storage and after field application). It also increased N_2O emission during solid digestate storage, but not in the field. Here, we concluded that the C and N contents of the wastes could influence the C and N fluxes in the farm; however, the quantity of external wastes had higher impacts than their characteristics. This importance of the import of wastes was consistent with the study from Möller (2009), who estimated that the mono-digestion of animal effluent would only poorly affect C and N fluxes, contrary to the import of external wastes and their application on soil as digestates. Boldrin et al. (2016) also highlighted the importance of external inputs in term of VS fluxes in the farm.

We found that mono-digestion decreased SOC storage, but that the import of wastes induced an increase in SOC storage. The effect of AD on SOC storage at the farm scale was only poorly explored in the literature. Möller (2009) estimated that the import of external wastes and their application on soil as

digestates could increase SOC storage. On the contrary, with a modelling approach in France, Bodilis et al. (2015) found that AD decreased SOC in 8 upon 10 farms. The difference with our results could be attributed to the different proportions of imported wastes in the digester. In our study, the amplitude of SOC variation was low: in ADb scenario, the increase in SOC compared to "no AD" is c.a. 3% of initial C stock after 20 years, i.e. 0.15% of increase per year.

We simulated an important saving of synthetic fertilizers in the farm thanks to AD, which was surprisingly not well investigated in the literature. A French survey also found an important decrease of the use of synthetic N fertilizer (-20%) in several farms after the introduction of AD (ADEME and SOLAGRO, 2018) that supported our findings.

Our simulations highlighted a higher risk of ammonia volatilization with AD. This risk was supported by some studies separately during storage (Baldé et al., 2018) or in the fields (Möller, 2009). Liebetrau et al. (2013) reported ammonia emissions from digestate storage corresponding to 0.01 to 3.68 gNH_3-N kWh^{-1}. Consistently, we simulated ammonia emission from digestates storage of 1.9 gNH3-N kWh^{-1} (ADb scenario).

We simulated that, in the case of no or low import of external wastes (EI0, EI30V, AI30A), AD decreased CH_4 emission during EOM storage. This result was similar to other studies that considered either the mono-digestion of cattle effluents (Amon et al., 2006), or a similar quantity of animal effluents or digestate applied on fields (Clemens et al., 2006; Holly et al., 2017; Rigolot et al., 2010). Maldaner et al. (2018) also measured a lower CH_4 emission from liquid digestate compared to manure with a whole farm approach (digester used 35% of external wastes). However, in addition to previous literature, we also highlighted that the high import of wastes could finally increase the CH_4 emissions (ADb compared to no AD). In ADb scenario, the CH_4 emission from EOM storage was 4% of the biomethane production, which was comparable to the values found by Baldé et al. (2016), Liebetrau et al. (2013), and Vergote et al. (2020) (from 0% to 12%). The total CH_4 emissions (EOM storage and fugitive emissions) was 7% of the biomethane production, which was higher than the value reported by Hrad et al. (2015) and Groth et al. (2015) from 2% to 6%.

In conclusion, the import of wastes locally increased the benefits and risks of AD. It confirmed the results of previous LCA studies (Michel et al., 2010; Zhang et al., 2021). Therefore, the import of wastes could be interesting in areas where the resources of animal effluents are scarce, as a mean to bring organic matter in the field and maintain soil fertility. On the contrary, the import of external wastes on a farm that is already saturated with organic fertilizers in a sensitive zone could increase the risks of N losses.

6.4.2 Quantifying the effect of good practices at the farm scale

In our study, the farm scale modelling approach enabled to quantify the effect of different management practices. Therefore, we confirmed the need to consider a continuous management of EOM from their production to their application in the field, for good environmental assessment. The fine modelling of the cropping systems enabled to study the digestate management in the field.

We highlighted a strong effect of the cover of the storage lagoon (ADb compared to SC scenarios). The presence of a cover did not only decreased ammonia emission, and consequently increased TAN content in digestate and indirectly improved the substitution to synthetic fertilizers. It also indirectly increased the ammonia volatilization following digestate application in the field: this highlighted the importance to also consider good practices of digestate application, to avoid losing in the field the N that was saved during storage.

In our simulation, phase separation increased N_2O and CH_4 emissions during storage (ADb compared to PS scenario). However, this strong simulated effect of phase separation should to be nuanced. Studying undigested effluents (Fangueiro et al., 2008a) and digestates (Gioelli et al., 2011; Holly et al., 2017), previous studies found that phase separation increased N_2O emissions but decreased CH_4 emissions during storage. These studies were not consistent with the hypothesis of SYS-Metha that applied lower CH_4 and CO_2 emission factors to raw digestate (8% of C lost during storage) compared to liquid digestate (15% of C lost during storage). If we had considered a C loss of 15% in raw digestate similarly to slurry, that could have induced an increase in CH_4 of 7.7 tCH_4-C $year^{-1}$, and a surplus of GHG emissions of 147 tCO_2eq. Therefore, the conclusion about the promotion or mitigation of GHG emissions by the phase separation depended on this emission factor (Figure 6.10).

Phase separation of digestates was found to increase NH_3 volatilization during storage (Baldé et al., 2016) and decrease it in the field (Chapter 3). Its global effect is unsure. Here, we support that the phase separation decreases the ammonia emissions due to a higher effect on volatilization in the field. Because the NH_3 volatilization after the application of raw digestate in the field is possibly underestimated by STICS (Chapter 4), the benefits of phase separation concerning this indicator could be even higher.

We found that the constraints in digestate use, such as constraints caused by a low storage capacity, induced a slight increase in ammonia volatilization and N_2O emissions in the field (Y scenario). We did not consider the effect of the dates and frequency of digestate application in the fields on gaseous emissions during storage, although the more frequent removal of digestate could decrease CH_4 emissions during storage (Ericsson et al., 2020; Paillat et al., 2014).

Digestate injection in soils was shown very effective to decrease ammonia volatilization in the field (Maris et al., 2021). However, this efficiency was reduced at the farm scale because some crops cannot be subject to injection. It could be more useful when digestate is often applied on grassland or all along

the year. Here, we suggest that the effect of injection should be also investigated in other cropping systems.

6.4.3 C footprint of AD with a consequential approach

Energy production, biogas leakage, and emissions due to alternative treatment of wastes were important parts of the C footprint of AD. These parts were already highlighted by LCA studies (Bacenetti et al., 2016b; Hijazi et al., 2016; Vasco-Correa et al., 2018). In our study, the low C footprint of the French electric mix induced a low benefit of AD in term of C footprint. AD even induced an increase of C footprint for most of the scenarios. This conclusion was already given by Garrigues et al. (2014), who studied the AD at the scale of a swine farm. However, the valuing of heat together with electricity, the valuing of biogas as biomethane with injection into the gas network, or the decrease in biogas leakage could importantly improve the GHG emissions balance of AD. The reduction of biogas leakage would strongly reduce GHG emissions from AD. Our study did not aim to explore different scenarios of alternative waste treatment or of biogas valuing.

In addition, we highlighted that the field application of digestate could significantly affect its C footprint, through SOC storage and substitution to synthetic fertilizers. These effects had similar importance as electricity production in our study. The importance of the cropping system in the environmental assessment of AD was sometimes reported in the literature (Baral et al., 2018; Vaneeckhaute et al., 2018). The effect of the difference in crop production cannot be neglected. It is difficult to say if this yield difference would happen in practice because farmers could try to compensate the difficulty to adjust digestates doses by higher N fertilizer application. We also highlighted the possible effect of the management practices of digestate on AD C footprint. Phase separation could have an important but still unsure impact on C footprint (PS). The absence of storage cover (SC), and the sub-optimal use of digestate in the field (Y), could also increase the C footprint of AD.

We used the consequential approach only to study the C footprint of AD. SOC storage, NH_3 emissions, NO_3^- emissions, use of synthetic N fertilizers were only assessed at the farm scale. Depending on the type of digested wastes, these indicators can also be impacted beyond the farm scale. The digestion of wastes such as animal effluents or sewage sludge could improve their treatment and help produce energy. However, if these wastes were already valued in the field, their digestion would likely only slightly improve the global nutrient recycling, and could decrease the SOC storage. On the other side, the use of wastes that were not valued in the field could improve soil quality and nutrient recycling at the territorial scale (Moinard et al., 2021). The importance of the type of digested waste regarding the environmental performance of AD was supported by previous studies (e.g., Hamelin et al., 2014; Vaneeckhaute et al., 2018; Vasco-Correa et al., 2018). We support that farm models could be the base of LCA to consider not only GHG emissions, but also other environmental factors such as acidification or eutrophication of the environment, or use of non-renewable resources (Avadí et al., 2021). There is

an important need to conduct LCA studies about AD in France to explore its impact in the different national or regional contexts.

6.4.4 Uncertainties and variability of the outputs of the farm model

The characteristics of the digested wastes could vary, inducing an uncertainty on digestates characteristics and biogas production. SYS-Metha proposes to consider this uncertainty on the inputs and computes an uncertainty about the results. The order of magnitude of the standard variations were 12% for biogas production, 4% for digestate quantities, 33% for C fluxes from the digestate, and 10% for N fluxes from the digestate. The variation coefficients of liquid digestate TS, TN, TAN, TC contents were comprised between 20% and 30%. This uncertainty should be propagated inside the farm and STICS models. Because of the numerical cost of the farm modelling (high required time of computation), no analysis of propagation errors has been assessed. As an order of magnitude, it was shown that the uncertainty in digestate characteristics of 20% would lead to an error of up to 20% of NH_3 volatilization, 12% of N_2O emissions, and 7% for nitrate leaching in the fields fertilized with liquid digestate (Chapter 4). STICS was also shown to possibly overestimate N_2O emissions during summer (Chapter 4).

We showed that climatic conditions had a strong effect on absolute environmental indicators. However, the important drivers or practices affecting C and N fluxes at the farm scale with AD were similar for each of the simulated climatic years. The simulation of repeated climatic years allowed to assess the variability of the indicator due to climate while simultaneously isolate the individual effect of each practice. We did not consider the impact of climatic conditions on emissions during EOM storage, even if they can influence these emissions (e.g., Baldé et al., 2018, 2016; Maldaner et al., 2018). Together with the uncertainties associated to emission factors or other model inputs, this variability has to be considered to relativize some of the impacts of AD. Costa Junior et al. (2015) also found high uncertainties associated to net GHG emissions of AD in a Brazilian beef cattle farm. Such uncertainty is often not realized in LCA of AD (e.g., Hamelin et al., 2014; Vaneeckhaute et al., 2018), or C footprint of AD (e.g., Boldrin et al., 2016; Vergote et al., 2019). The estimation of uncertainties could be an advantage of model-based environmental assessment of AD.

Emission factors for gaseous emissions at storage are associated to strong uncertainties (Kupper et al., 2020; Walling and Vaneeckhaute, 2020b). Lyng et al. (2015) reported that the change of such parameters could change the conclusion of the environmental assessment of AD they described. Therefore, a sensitivity analysis of these factors could be important.

6.4.5 Assumptions of the model and perspectives

The model used several assumptions that are summarized here together with some perspectives to explore their importance.

We did not model any N_2O emissions from liquid EOM storage. In the literature, N_2O emissions from liquid storage were low, but sometimes still existed. Kupper et al. (2020) reported an average N_2O emission factor of 0.13% of initial TN for cattle slurry. If we had used this emission factor in our model, it would have increased N_2O emissions by 2.9 kgN_2O-N year^{-1} in "no AD" scenario, and by 23.8 kgN_2O-N year^{-1} in ADb scenario. The resulting net increase GHG emissions would have been 8.7 tCO_2eq in ADb compared to "no AD" scenario. This change was not important but could still be considered regarding the net GHG emissions induced by AD. The simulation of N_2O emissions during raw digestate and liquid digestate storage would have induced a change in N_2O emissions of 3.9 kgN_2O-N year^{-1} (1.6 tCO_2eq) in PS scenario compared to ADb scenario. This effect would have remained low.

Some authors reported low CH_4 emissions after field application of liquid EOM (Czubaszek and Wysocka-Czubaszek, 2018; Holly et al., 2017; Rosace et al., 2020; Tiwary et al., 2015). We neglected them in our study. According to Rosace et al. (2020), the order of magnitude of CH_4 emission could be 1% of CO_2 emission, during less than 10 days. The laboratory analyses of EOM corresponding to the same case study (Chapter 3) showed that the order of magnitude of CO_2 emission after that period could be at maximum 25% of organic C for liquid EOM. Within these hypotheses, we concluded that the CH_4 emission in the field could be 35 kgCH_4-C year^{-1} in "no AD" scenario, and 145 kgCH_4-C year^{-1} in ADb scenario which seemed low (from 1% to 2% of CH_4 emissions in noAD and ADb scenarios). The resulting net GHG emissions for ADb scenario would be +4.1t CO_2eq induced by AD, which was also low.

One strength of our model was the module of decision rules. This module enabled to accurately predict the substitution of synthetic N fertilizer by digestates. The model implicitly assumed a good knowledge on digestate fertilization properties. However, the fertilization using EOM is often more difficult to use in practice. Indeed, the digestate can vary along the time, requiring frequent analysis to be properly managed. Moreover, the mineralization of EOM can induce a net N release or capture, which is difficult to predict.

A higher HRT could decrease CH_4 emission during digestates storage through stabilization of EOM (Muha et al., 2015), which could be due to VS decrease and stabilization of organic matter in the digestate (Maldaner et al., 2018). Here, SYS-Metha only considered the decreased in VS content during digestion. The stability of organic matter was not simulated. Therefore, we used default values that were not related to the characteristics of the digester to parametrize the stability of the digestates in STICS. The digestate stability could increase with HRT (Akhiar et al., 2021). In the future, the modelling of increasing organic matter stability with digestion could be used to predict the SOM storage, as suggested by Jimenez et al. (2015).

Finally, the introduction of AD can modify crop rotation or crop residues handling, in order to produce biomass to feed the digester. This change in agricultural practices can affect soil N fluxes, such as NH_3

volatilization (Quakernack et al., 2012), N_2O emissions (Möller and Stinner, 2009), or nitrate leaching (Möller, 2015). Our model could further be a tool to study the effect of AD on different cropping systems or soil type, which would enable to investigate for example nitrate leaching in other soils.

6.5 Conclusion

In this study, we designed and used a farm model to quantify the changes in C and N fluxes caused by the introduction of AD in a crop-livestock farm, in an integrative way. At the farm scale, AD decreased the use of synthetic fertilizers, increased SOC storage, and increased NH_3 emissions. These three effects were mainly driven by the input of external wastes. Therefore, the import of external wastes could be interesting in areas with low resources of organic matter, or it could increase the risks of N losses if the initial N input were already important. In this local French context, AD increased the net GHG emissions, due to an electric mix with low C footprint (nuclear energy), and due to the valuing of biogas as electricity without heat valuing; this conclusion could be different if the biogas was valued as biomethane or if the heat was better valued. Using such a model, we highlighted and prioritized the best practices to improve the environmental performance of AD at the farm scale. The storage cover strongly reduced ammonia emissions. The application of digestate on agronomically adequate dates and crops, and the injection of digestate into the soil both had moderate impact on the total NH_3 emissions. Phase separation moderately decreased NH_3, its effect on GHG emissions was uncertain. Such model could help to investigate the effect of AD in other territorial contexts (other wastes, different N availability in animal effluents, other cropping systems). AD with a stronger change of cropping systems could also be investigated with such an approach. It could also provide reliable information and hypothesis that can be considered for further LCA of AD to assess its environmental impacts beyond the farm scale. At last, the effect of AD on other variables of interest, such as social or economical impacts of AD, workload, or other impacts on soil such as soil compaction or soil biodiversity could be further explored.

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Chapitre 7
Effets à court et long termes de l'épandage de digestat en grande culture sur les vers de terre

En se basant sur le cas d'étude de Nouzilly (Chapitres 2, 3, et 5), nous avons validé le modèle sol-plante STICS (Chapitre 4) et le modèle de méthanisation SYS-Metha (Chapitre 5). Ces deux modèles ont été couplés pour réaliser des simulations à l'échelle de l'exploitation (Chapitre 6). Cette approche a permis de mieux comprendre les effets directs et surtout indirects de différentes pratiques lors du traitement des déchets ou de la gestion des digestats. Les Chapitres 3, 4, 5, et 6 se sont concentrés sur les impacts de l'introduction de la méthanisation liés aux changements de flux de carbone et d'azote. Le modèle développé dans le Chapitre 6 est une vision simplifiée de la réalité. Lors de l'évaluation de la méthanisation, il est primordial de considérer d'autres facettes de la méthanisation : aspects techniques et logistiques, problématiques socio-économiques, charge de travail, vérification de l'innocuité des digestats, pour n'en citer que quelques-uns. Dans cette thèse, nous avons voulu ouvrir la vision induite par l'utilisation des modèles, et explorer une de ces autres questions liées à l'introduction de la méthanisation : l'effet de l'épandage de digestats sur la vie du sol, en prenant le cas des vers de terre. Le choix de ce thème est en partie justifié par le besoin de connaissance sur cette thématique qui a récemment émergé de la filière. Son intégration dans cette thèse est aussi justifiée par le lien entre les vers de terre, et les éléments C et N apportés par les digestats et effluents. En effet, la matière organique sert de nourriture aux vers de terre, et les émissions d'ammoniac ont été proposé comme pouvant causer une toxicité sur ces organismes. Ce Chapitre 7 est écrit sous la forme d'une publication scientifique, publiée dans la revue Applied Soil Ecology. Ce travail a été réalisé avec des chercheurs de l'UMR EMMAH (INRAE, Avignon).

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Short- and long-term impacts of anaerobic digestate spreading on earthworms in cropped soils

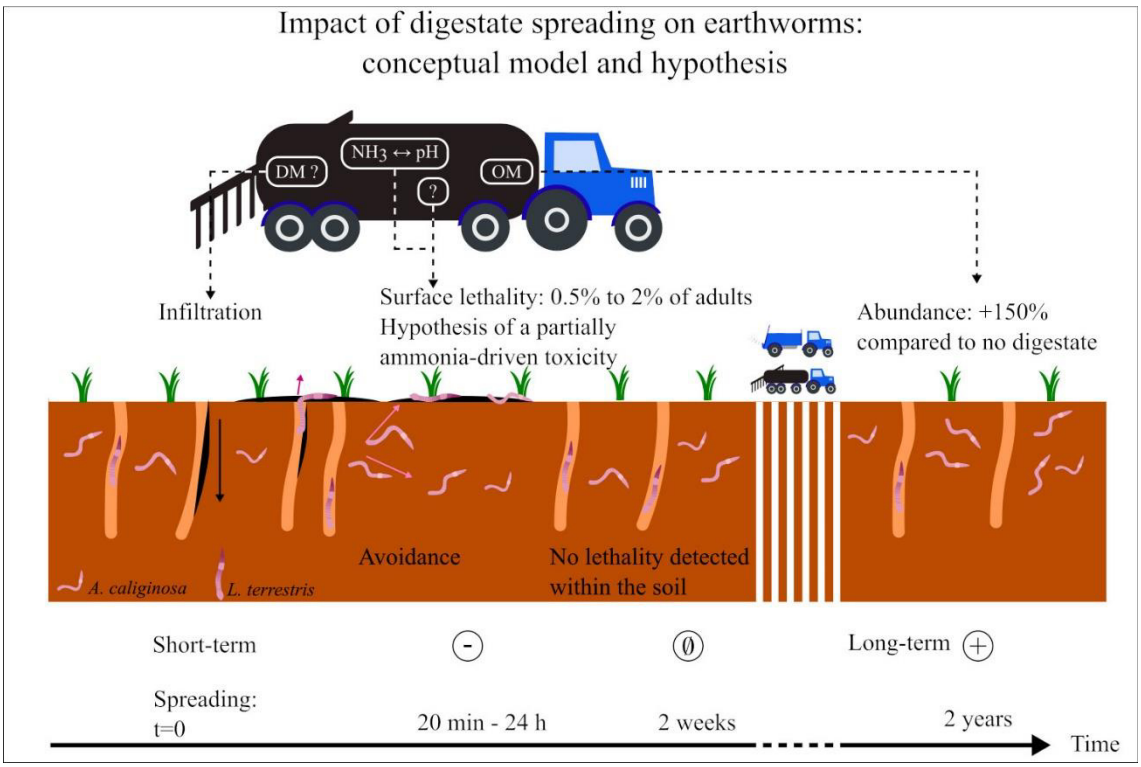
Victor Moinard[1], Clément Redondi[1], Véronique Etievant[1], Antoine Savoie[2], David Duchenne[2], Céline Pelosi[3], Sabine Houot[1], Yvan Capowiez[3]

1: UMR Ecosys, Université Paris Saclay, INRAE, AgroParisTech, Thiverval-Grignon, France
2: UE PAO, INRAE, Nouzilly, France
3: UMR EMMAH, INRAE, Avignon, France

Highlights

- Contact with high-dose liquid digestates or slurry was lethal to earthworms.
- Ammonia in the organic products contributed to this short-term mortality.
- After organic product spreading, a few earthworms died at the soil surface.
- No mortality was observed within the soil after a few days in field and soil cores.
- Digestate or manure application increased the earthworm population after two years.

Graphical Abstract



Abstract

Anaerobic digestion is increasingly used in Europe to treat organic substrates and produce biogas as a renewable energy source. The residual matter (digestate) is used in agriculture as an organic fertilizer. The study aims at assessing the impact of digestate application in the field on earthworms from the short term (few hours) to the long term (two years), and at investigating under laboratory conditions the role of ammonia and earthworm behavior on digestate toxicity in the short term. First, we studied earthworm communities in fields fertilized with digestates, cattle effluents, or chemical fertilizers for two years. Earthworm abundance was assessed before and after the fertilization event of the third year. Earthworm mortality at the soil surface was also assessed immediately after fertilization. Next, the toxicity of digestate or ammonia solutions on *Aporrectodea caliginosa* and *Lumbricus terrestris* was measured in microcosms (110 cm^3) to better understand the short-term toxicity (two weeks). Finally, we spread digestate (40-80 t ha^{-1}) on soil columns (5300 cm^3) and used X-ray tomography after two weeks to assess the burrowing behavior of earthworms in the cores. Earthworm abundance was 150% higher in the fields treated for two years with digestates or cattle effluents compared to the field treated with chemical fertilizers. 0.5 to 2% of adult earthworms died at the soil surface a few hours after liquid digestate and cattle slurry spreading (18 to 24 t ha^{-1}). The digestate (10% to 20% (fresh digestate / dry soil)) and ammonia were also lethal to earthworms in the microcosms within two weeks. In contrast, no mortality occurred inside soil columns two weeks after digestate spreading; *A. caliginosa* avoided the soil surface with high digestate inputs. This case study highlighted the potential short-term toxicity of digestate (a few hours), which evolved towards a neutral to positive impact in the field in the longer term (from two weeks to two years). Further research is needed to understand the impact of diverse solid and liquid digestates on soil macrofauna in different soils.

Keywords

Exogenous organic matter; Organic fertilizer; Ammonia-driven toxicity; Earthworm avoidance-behaviour; X-ray tomography; Soil ecotoxicology.

7.1 Introduction

Anaerobic digestion is a waste treatment process in which organic materials (e.g., animal effluents, biowastes, wastes from agriculture and agroindustry) are digested under anaerobic conditions to produce biogas, which is considered a renewable energy source. Anaerobic digestion could thus contribute to climate change mitigation (Hijazi et al., 2016). After digestion, the residual organic material (digestate) is usually spread in the field as a valuable organic fertilizer and amendment to promote nutrient recycling (Bachmann et al., 2014; Coelho et al., 2020). For these reasons, anaerobic digestion has been promoted in recent years in France (Ministère de l'Écologie du Développement Durable et de l'Énergie and Ministère de l'Agriculture, de l'Agroalimentaire et de la Forêt, 2013; Pellerin et al., 2013) as well as in multiple regions in Europe and worldwide (Scarlat et al., 2018), leading to an increase in digestate use in agriculture. Digestates can originate from diverse inputs and be used in agriculture as-is, or after multiple post-treatments, including phase separation or composting. For these reasons, digestates have diverse physicochemical characteristics (Guilayn et al., 2019a; Möller, 2015; Nkoa, 2014) and are thus applied for different purposes. Some digestates are liquid and used primarily as fertilizers for short-term N fertilization, in a similar fashion to animal waste slurries. Other digestates are solid and used as soil amendments to increase soil organic matter content and long-term nutrient release, like composts or solid manure (Houot et al., 2014). Many studies have confirmed the interest in digestates as fertilizers for crop production, but they have also highlighted the potential impacts of digestate application on greenhouse gas and ammonia emissions (Möller, 2015; Nkoa, 2014). However, their effects on living soil organisms still need to be better characterized.

Earthworms are key soil organisms that can be promoted by organic matter inputs in cropping systems (D'Hose et al., 2018). Thus, repeated applications of digestates could have a positive effect on earthworm populations. However, anaerobic digestion generally increases C stability (Béghin-Tanneau et al., 2019; de la Fuente et al., 2013; Thomsen et al., 2013) and modifies the nature of the spread organic matter, which may influence earthworm populations in soils. Indeed, the quality of organic matter applied to soils (energy content, particle sizes) influences earthworm growth (Sizmur et al., 2017). Only a few studies have focused on the effects of digestates on earthworm communities under field conditions after one or several applications (from a few weeks to a couple of years after spreading, i.e. in the mid- to long-term). Butt and Putwain (2017), Clements et al. (2012), and Koblenz et al. (2015) found higher earthworm biomass and abundance in treatments amended with solid or liquid digestate compared to unamended soils. The impacts of digestates and undigested animal effluents were generally similar. Digestate application was also shown to influence community composition (Koblenz et al., 2015). In contrast, other studies reported no change or a decrease in earthworm populations compared with unfertilized arable or grass fields from a few months to a couple of years after liquid or dry digestate applications (Bermejo et al., 2010; Frøseth et al., 2014; Rollett et al., 2020). In regards to the physico-

chemical diversity of the digestates applied, as well as the different earthworm species involved, their contrasting impacts still need to be further investigated.

In contrast to these long-term neutral to beneficial effects of soil amendment on earthworm abundance, short-term negative impacts of digestates on earthworm communities were also reported. Indeed, during anaerobic digestion, organic substrates are degraded to produce biogas, leading to N mineralization and an associated increase in the ammonium concentration in the liquid digestates (Möller, 2015; Nkoa, 2014). Several studies highlighted short-term toxicity in earthworms after application of high slurry doses (Curry, 1976; Curry et al., 1980; Hansen, 1996; Van Vliet and de Goede, 2006). According to Curry (1976) and Hughes et al. (2008), urine degradation products, such as ammonia, benzoic acid, or sulphides, could be responsible for short-term toxicity to earthworms after slurry spreading. These compounds, including ammonia, can be present in liquid digestate (Ghidotti et al., 2018; Qiu et al., 2019). In Germany, Burmeister et al. (2015) observed dead earthworms at the surface immediately after liquid digestate or slurry were applied. A Norwegian study reported a similar observation after spreading of undigested and digested slurry (Johansen et al., 2015; Løes et al., 2014). However, earthworm populations were not affected after three years of repeated application. Such phenomena could depend on soil moisture (Van Vliet and de Goede, 2006). These results suggest that digestates influence earthworm behavior (surface foraging). The toxicity of solid or liquid digestate toxicity was assessed in the laboratory for diverse soil organisms (Tigini et al., 2016), including the epigeic earthworm *Eisenia fetida* (Krishnasamy et al., 2014; Pivato et al., 2016; Renaud et al., 2017) and the endogeic earthworm *Aporrectodea caliginosa* (Ross et al., 2017). Although numerous hypotheses have been proposed to explain digestate toxicity (ammonia, salinity, oxygen deficiency, heavy metals), none has been completely verified or rejected. Moreover, an insufficient number of studies has been carried out to distinguish the effects of solid or liquid digestates. The understanding and quantification of the short-term toxicity of digestates and slurries on earthworms remain incomplete.

The aim of this study was to investigate the short-term (a few hours to two weeks) and long-term (after two years in the field) effects of digestate application in the field on earthworms. We investigated four hypotheses. (H1) In the long term, we expect a positive effect of digestates because of organic matter inputs, despite possible short-term negative effects. (H2) Digestate is toxic to earthworms in the short term, with differences in toxicity at the surface or within the soil. (H3) Ammonia explains short-term digestate toxicity. (H4) Digestate affects earthworm behavior (e.g., surface foraging and/or avoidance) in the short term, which can influence exposure to organic products. To explore these hypotheses, we conducted a total of three experiments under field or controlled laboratory conditions. To assess the long-term effects on earthworm abundance, we performed a two-year field experiment with different fertilization strategies based on mineral fertilizer, undigested slurry and manure or digestates with or without phase separation (H1). The short-term toxicity related to liquid digestate and slurry spreading was assessed in the same field experiment after a fertilization event by sampling the earthworm

population and counting dead earthworms at the soil surface after spreading (H2). The short-term ecotoxicity of the same organic products was further tested in two sets of microcosm experiments (110 cm^3) under laboratory conditions with a focus on the role of ammonia in short-term toxicity (H2, H3). In microcosm ecotoxicological tests, earthworms cannot avoid the digestate as it is possible in the field. Therefore, we explored the short-term impact of digestate and slurry application on earthworm behavior and survival under conditions closer to field conditions in soil columns (5300 cm^3) analyzed with X-ray tomography (H2, H4).

7.2 Material and methods

7.2.1 Field and soil description

The field study was carried out at Nouzilly, Centre-Val de Loire, France (47°32' N, 0°48' E) (Pasquier et al., 2019). It is a temperate oceanic climate, with an average annual mean temperature of 11.9 °C and total annual rainfall of 650 mm. Prior to this experiment, the field has been homogeneously managed for more than ten years under conventional arable cropping systems, including wheat, barley, maize, sunflower and fertilization with mineral N with occasional inputs of cattle manure (c.a. once every five years). The soil was sampled in 2017 at the beginning of the experiment (45 samples), and the following properties/characteristic were analyzed by a specialized laboratory (US Laboratoire d'Analyses des Sols - INRAE, Arras, France): sedimentation to measure texture (NF X 31-107), total C and N content by dry combustion, gas chromatography, and thermal conductivity detector (NF ISO 10694 and NF ISO13878), pH in a water suspension (NF ISO 10390), $CaCO_3$ content by acidification and measurement of released CO_2 volume (NF ISO 10693), mass loss after combustion for SOM (1100°C), corrected from $CaCO_3$ content. Bulk density of the upper layer was measured on fifteen locations with the core method (NF X31-501). The soil was a silty loam with the following characteristics in the ploughed layer (0-20 cm depth): 20% clay, 64% silt, 16% sand, pH 6.7, SOM content 23.3 g kg^{-1}, C:N ratio 10.1, $CaCO_3$ content 1.1 g kg^{-1}, bulk density 1.37 g cm^{-3}. The water holding capacity (WHC) was estimated in the field to be 30% (gravimetric humidity), which corresponded to the maximal soil moisture measured in the field (44 measurement dates in three years, five replicates per treatments).

The soil was sampled once in winter to conduct the laboratory experiments. The soil was sieved fresh to a size of 4 mm and stored in a closed container at 5°C before being used. The WHC of the sieved soil was measured by saturating 25 g of dry sieved soil with water in elevated cylinders and waiting 48h at 5°C for the excess water to runoff. The WHC of sieved soil was 40% (gravimetric humidity). When needed, the gravimetric soil moisture was measured by mass loss after 48 h of drying at 105°C. When needed, soil pH was measured in a water suspension (1:5 volume ratio, i.e., 5 g of dry soil in 24 mL): after addition of the reverse osmosis-purified water, the pH was measured with an electrode after agitation for 1 hour.

7.2.2 Fertilizers and chemical solutions used as treatments.

All experiments used digestates from the same anaerobic digester located in the Centre-Val de Loire region of France, and referred to as 'territorial digestate' here. In the digester, a continuous wet mesophilic process is performed, with one main digester and one postdigester. The digester treats up to 12,000 t of waste per year, consisting of cattle slurry, cattle and horse manure, sewage sludge, grease, agroindustrial wastes, cereal middlings, and grass silage. The retention time is 100 days. The digestate can be applied raw or after phase separation using a screw press. In the latter case, the liquid digestate is stored in an open lagoon and the solid digestate is stored in dedicated outdoor platform. The experiments mainly used the liquid territorial digestate, but also the raw and solid ones. We also used aerated liquid territorial digestate. Aeration was performed in a 2 L container with an aquarium oxygenizer (1.5 L_{air} min^{-1}) under agitation for 24 h. Aeration promoted the volatilization of diverse volatile compounds and decreased ammonia concentrations. Finally, we used the undigested cattle effluents. We used both cattle slurry and cattle manure, which came from a dairy farm.

A second digestate resulting from the digestion of cover crops was studied for comparison. This digestate from cover crops was sampled in 2020 (Île-de-France, France). This second digester uses a continuous wet mesophilic process to treat 28,000 t of waste per year. Animal manure is excluded from the feedstock, which mainly includes silage maize or other cereals, completed with soy wastes and agroindustrial wastes. The retention time is 90 days. The digestate is not post-treated and is stored in covered lagoons. Only the raw digestate was used in the experiments for this second digestate.

Ammonia solutions were used as control treatments in the laboratory experiments. They were prepared by diluting a commercial ammonia solution to reach the target ammonia concentration. An ammonium chloride solution was also used as another control and was produced from the ammonia solution by the addition of concentrated hydrochloric acid to reach a target pH value. Ammonia and hydrochloric acid were obtained from Carlo-Erba (Val-de-Rieu, France).

Before each laboratory experiment, organic products were sampled and stored in closed containers at 5°C for less than one month before use. Aeration of liquid territorial digestate was performed one day before the laboratory experiment. Ammonia and ammonium solutions were prepared less than one hour before the experiment to avoid volatilization.

Table 7.1 – Physico-chemical characteristics of the organic products and chemical solutions. Their use in the different experiments is also summarized. Missing values are indicated by N.A. (not available).

Organic products			Physico-chemical characteristics							Use					
Type	Origin	Sampling date	DM (% FW)	[N_{amm}] (g $kgFW^{-1}$)	[N_{tot}] (g $kgFW^{-1}$)	pH	Moistened power (% FW)	VS (% FW)	Conductivity (mS cm^{-1})	LTFE	STFE#1	STFE#2	ME#1	ME#2	CE
Cattle slurry	Nouzilly, Centre-Val de Loire	2017-2018	3.3	0.9	1.8	7.3	N.A.	2.4	2.0	X					
		February, 2019	4.0	2.0	2.7	7.6	59	2.8	3.2		X		X		X
		March, 2019	3.2	1.1	2.2	7.9	N.A.	2.2	2.4			X			
		2020	4.9	1.0	2.0	6.9	46	3.5	2.7					X	
Cattle manure	Nouzilly, Centre-Val de Loire	2017-2018	32.5	0.7	8.3	9.7	N.A.	25.3	6.1	X					
Territorial digestate, liquid	Nouzilly, Centre-Val de Loire	2017-2018	4.8	2.2	4.4	7.9	N.A.	2.8	3.9	X					
		February , 2019	5.1	3.2	4.3	8.2	81	3.0	4.3		X		X		X
		March, 2019	4.8	2.2	4.2	8.1	N.A.	2.9	4.4			X			
		2020	5.5	2.4	4.1	8.0	58	3.8	4.7					X	
Territorial digestate, liquid, aerated	Nouzilly, Centre-Val de Loire	2019	5.0	1.9	N.A.	9.3	81	N.A.	N.A.				X		X
		2020	5.3	1.9	N.A.	8.9	58	N.A.	N.A.					X	
Territorial digestate, raw	Nouzilly, Centre-Val de Loire	2017-2018	6.8	2.4	4.7	8.0	N.A.	4.6	4.3	X					
		February, 2019	6.1	3.9	4.6	7.9	N.A.	4.3	4.5		X				
		March, 2019	6.0	2.2	4.2	7.9	N.A.	4.3	4.2			X			
Territorial digestate, solid	Nouzilly, Centre-Val de Loire	2017-2018	27.2	1.6	6.6	9.3	N.A.	23.2	3.1	X					
Digestate from intermediate crop, raw	Île-de-France	2020	6.9	4.2	4.9	8.3	51	5.2	5.3					X	
Ammonia (NH_3) solution			0.0	10.0	10.0	11.8	100	0	N.A.					X	
Ammonium (NH_4Cl) solution			0.0	10.0	10.0	8.0	100	0	N.A.					X	

LTFE: Long-term field experiment (2017-2018). STFE#1: First short-term field experiment (February, 2019). STFE#2: Second short-term field experiment (March, 2019). ME#1: Microcosm experiment, series #1 (2019). ME#2: Microcosm experiment, series #2 (2020). CE: Column experiment.

The dry matter (DM) content of all organic products was determined after 48 h of drying at 105°C. The volatile solid (VS) content (mass loss after combustion, NF EN 13039), total Kjedhal N content (NF EN 13654-1), and pH and conductivity (measured in a water suspension NF EN 13037 and NF EN 13038) were analyzed in fresh products by specialized laboratories (AUREA, Ardon, France, and AUREA, La Rochelle, France). Their ammoniacal nitrogen (N_{amm}) contents were determined after the extraction of 25 g of a fresh sample with 100 mL of 1 mol L^{-1} KCl for 1 h, after which the liquid phase was analyzed by colorimetry on a continuous flow analyzer (Skalar, The Netherlands). The concentration accounted for both NH_3 and NH_4^+ forms. In the microcosm experiments, the liquid digestate and slurry applied to soils contained large amounts of water, which was taken into account to achieve common soil moisture levels in all treatments. Because of the large amount of water associated with organic matter in the organic products, the DM content of the organic products is not relevant for this experiment. Thus, we defined the "moistening power" of an organic product as the quantity of water that significantly humidified soils in the microcosm experiments. It was determined by weighing free water after centrifugation of the organic products for 10 min at 614 x g (three replicates). The physico-chemical characteristics of all organic products and chemical solutions are summarized in Table 7.1, which also includes their use in the different experiments.

Heavy metal (Cd, Cr, Cu, Ni, Pb, Zn) contents were analyzed in one replicate in organic products at the CIRAD Laboratory of Water, Soil and Plant Analysis (US Analyses, CIRAD, Montpellier, France). The organic products were mineralized (NF ISO 14869-1) and heavy metal contents were measured using inductively coupled plasma mass spectrometry (ICP-MS) (Table 7.2). Le Bars et al. (2018) included some of these heavy metals analyses in their study and gave more details in the methods. Seven polychlorinated biphenyls (PCB), including PCB 028, PCB 052, PCB 101, PCB 118, PCB 138, PCB 153 and PCB 180, were measured by an external laboratory (AUREA, Ardon, France) (Table 7.2). The method consisted in an accelerated solvent extraction (ASE) followed by a dosage with Gas Chromatography - Tandem Mass Spectrometry (GC-MS-MS) (XP X 33-012). Thirteen polycyclic aromatic hydrocarbons (PAH) were measured (Table 7.2). Sertillanges et al. (2020) included these PAH analyses in their study and gave more details in the methods.

Table 7.2 – Heavy metals, polycyclic aromatic hydrocarbons (PAH), and polychlorinated biphenyls (PCB) content in the organic fertilizer and amendments spread in the field experiment. Results are expressed in mg kgDM-1. NA: not available (no measures). ND: not detected (below the threshold detection content). For 13 PAH, results are indicated as mean ± standard deviation (n=4). Heavy metals were analysed only on one sample; when it is available, the uncertainty of analysis (inter laboratory comparison) is indicated as mean ± uncertainty.

	Cd	Cr	Cu	Ni	Pb	Zn	7 PCB	13 PAH
Cattle slurry	ND	2	36 ± 3	2	2	180 ± 22	ND	191 ± 75
Territorial liquid digestate	1.0	59 ± 3	249 ± 20	21	22 ± 0.8	663 ± 79	ND	741 ± 27
Territorial raw digestate	1.0	34 ± 2	266 ± 22	16	23 ± 0.8	701 ± 84	ND	762 ± 125
Cattle solid manure	ND	3	22	2	1	83 ± 10	ND	89 ± 33
Territorial solid digestate	0.1	14	64 ± 5	5	8 ± 0.3	178 ± 21	ND	508 ± 49

7.2.1 Earthworms

In the laboratory experiment, we considered two earthworm species, *L. terrestris* and *A. caliginosa*. *L. terrestris* were bought in a fishery store (Decathlon®). *A. caliginosa* were sampled from an orchard, that was pesticide free for more than five years, located close to Avignon, France. All earthworm individuals were acclimated in the experimental soil for two weeks before the experiments. Before and after all laboratory experiments, the earthworms were weighed after a fasting period (Fründ et al., 2010). For this purpose, they were left in Petri dishes with moistened filter paper, which was changed every day. After three days, the earthworms with an empty gut were weighed. *L. terrestris* is an epi-anecic earthworm (Bouché, 1972). *A. caliginosa* is an endogeic species and a good model for studying agricultural practices (Bart et al., 2018). *L. terrestris* and *A. caliginosa* were chosen because they are both common in agricultural fields of temperate soils.

7.2.1 Experiment setup

7.2.1.1 Field experiment setup

The field experiment started in 2017. Four treatments are tested in plots of 24 x 75 m under conventional management (wheat - rapeseed - wheat succession, export of wheat straw, ploughing). The treatments consisted of (A) fertilization with only mineral N solution in winter. It did not receive any organic amendment; (B) fertilization with solid bovine farmyard manure in summer and liquid cattle slurry in spring; (C) application of raw territorial digestate in both summer and spring (no phase separation); and (D) application of solid territorial digestate in summer and liquid territorial digestate in spring (Table 7.3). Prior to the experiment, we assumed similar earthworm populations in all plots due to similar management practices in the long-term and plot proximity. Treatment (A) was assumed to be representative of the existing initial earthworm populations.

In February 2019, after two years of the different fertilization schemes, we assessed their long-term effects. On the 12th of February 2019, seven days before applying the fertilizer treatments, we excavated the soil to assess earthworm populations. Fertilizers were first applied on the 19th of February 2019, and one hour later, earthworms that came up to the surface and died were counted. A second evaluation of the earthworm population was carried out two weeks later, on the 5th of March 2019, to assess the short-term effect of fertilization with liquid mineral N or the organic products. Fertilizers were applied for a second time on the 12th of March 2019. The number of earthworms that came to the surface and died was assessed after 1 h and 24 h (Table 7.3).

Table 7.3 – Field assessment of the impact of the use of organic products on earthworms: summary of the 4 treatments and of the experimental procedure. The described experiment is a 3-year field experiment with 4 fertilization treatments that started in October, 2016 (undifferentiated fields).

Date	Event	Treatments			
		Mineral N	Slurry and manure	Raw territorial digestate	Liquid and solid territorial digestates
		A	B	C	D
January, 2017 to April, 2017 (wheat)	fertilization (2 events)	mineral fertilization 86 + 43 kgN ha^{-1}	cattle slurry 37 + 60 t ha^{-1}	raw digestate 38 + 31 t ha^{-1}	liquid digestate 36 + 32 t ha^{-1}
August, 2017	amendment	/	cattle manure 35 t ha^{-1}	raw digestate 32 t ha^{-1}	solid digestate 33 t ha^{-1}
March, 2018 (rapeseed)	fertilization	mineral fertilization 99 kgN ha^{-1}	cattle slurry 27 t ha^{-1}	raw digestate 32 t ha^{-1}	liquid digestate 36 t ha^{-1}
September, 2018	amendment	/	cattle manure 12 t ha^{-1}	raw digestate 16 t ha^{-1}	solid digestate 16 t ha^{-1}
February 12, 2019 (wheat)	earthworm population evaluation				
February 19, 2019	fertilization	mineral fertilization 40 kgN ha^{-1}	cattle slurry 24 t ha^{-1}	raw digestate 12 t ha^{-1}	liquid digestate 23 t ha^{-1}
1 h after spreading	surface mortality evaluation				
March 5, 2019	earthworm population evaluation				
March 12, 2019	fertilization	mineral fertilization 80 kgN ha^{-1}	cattle slurry 18 t ha^{-1}	raw digestate 20 t ha^{-1}	liquid digestate 18 t ha^{-1}
1 h after spreading	surface mortality evaluation				
24 h after spreading	surface mortality evaluation				

Earthworm populations were assessed by chemical extraction, soil excavation (three replicates per treatment, 40 x 40 x 20 cm), hand sorting and formaldehyde conservation, as described in Pelosi et al. (2009). The number of earthworms at the surface after fertilizer application was counted in six 1 m^2 subplots. Earthworm morbidity was tested according to their reaction to physical stimuli (earthworms that did not react were considered dead; Langdon et al., 1999). At the same date, soil moisture was measured (0-20 cm, five replicates per treatment).

7.2.1.2 Laboratory ecotoxicity tests in microcosms

Ecotoxicity tests were carried out in microcosms to evaluate the mortality and weight change of earthworms living in a mixture of soil and organic products. For each treatment, sieved soil and the organic product were mixed and then the soil moisture was adjusted to reach 90% of the sieved soil-WHC considering the "moistening power" of the organic products (see section 7.2.2). Seven replicates were used for each treatment . For each replicate, one fasted and weighed (as in section 7.2.3) *L. terrestris* or *A. caliginosa* adult was placed in a Petri dish (diameter: 14 cm) with 150 g of the mixture. During the experiment, microcosms were placed in a dark chamber at 15°C (±2°C) for 2 weeks. We assessed mortality every day during the first five days and then every two or three days (earthworms that did not react to physical stimuli were considered dead). At the end of the incubation, living earthworms were fasted and weighed. Treatments with fewer than three living earthworms were excluded from the data analysis because of the low sample size. These methods were adapted from Leveque et al. (2013) and Olvera-Velona et al. (2008).

We conducted two series of ecotoxicity microcosm experiments. Series #1 (2019) was designed to compare *L. terrestris* and *A. caliginosa* mortality in the presence of liquid digestate and slurry. In each species, we tested the eight following treatments (Supplementary Material 1): four doses of liquid territorial digestate (2.5%, 5%, 10%, 20%, in g of fresh organic product per 100 g of dry soil); two doses of cattle slurry (10%, 20%); one control without organic products; and one dose of aerated liquid territorial digestate (20%). Series #2 (2020) was designed to explore the hypothesis that ammonia was responsible for the toxicity of the organic products in an experiment conducted only in *L. terrestris*, which was the species on which digestate had the greatest impact in the field. We tested the following treatments (Supplementary Material 1): one control without any organic products; four doses of liquid territorial digestate (5%, 10%, 15%, 20%); four doses of aerated liquid territorial digestate (5%, 10%, 15%, 20%); five doses of digestate from cover crops (2.5%, 5%, 10%, 15%, 20%); and 3 doses of cattle slurry (5%, 10%, 20%). We tested five doses of ammonia (NH_3) solution (5% D_{eq}, 9% D_{eq}, 18% D_{eq}, 27% D_{eq}, 36% D_{eq}) and four doses of ammonium chloride (NH_4Cl) solution (5% D_{eq}, 9% D_{eq}, 18% D_{eq}, 27% D_{eq}, 36% D_{eq}). The X% D_{eq} doses correspond to an amount of ammoniacal N similar to a dose of X% of the liquid territorial digestate. In both series, aerated liquid territorial digestate was used to test whether a decrease in the ammonia concentration would decrease mortality. Digestate from cover crops was used to test whether the inputs could influence digestate toxicity. NH_3 and NH_4Cl solutions were

used to test the toxicity of ammonia according to its chemical form. We calculated the theoretical contents of ammoniacal N in the mixture at the beginning of the experiment, expressed in mg of ammoniacal N per g of dry soil (mgN_{amm} $kgDS^{-1}$), on the basis of the ammoniacal N concentration in organic products. These contents are presented in Supplementary Material 1. In Series #2, because of its impact on $NH_3 \rightleftarrows NH_4^+$ equilibrium, we analyzed the pH of each mixture: one part of the mixtures was left for 24 h without earthworms, after which pH was analyzed as described in section 7.2.1. Ammoniacal N together with pH was a relevant proxy for NH_3 concentration in soil mixture, as it was difficult to discriminate between NH_3 and NH_4^+ forms in soil mixtures.

7.2.1.3 Laboratory ecotoxicity tests in soil columns.

We compared earthworm biomass and burrowing behavior in soil columns before and after spreading organic products on the top of the column. The soil columns consisted of PVC cylinders (40 cm in height and 15 cm in diameter) filled with sieved soil to a height of 30 cm. Columns were iteratively filled with sieved soil in 2 cm-high layers, which were individually moistened and compressed to control the homogeneous bulk density at 1.3 g cm^{-3} following the methodology of Capowiez et al. (2015). Prior to the experiment, the earthworms were weighed. At t=0 days, five *A. caliginosa* and two *L. terrestris* were added to each column. We covered the columns with plastic grids to prevent the earthworms from escaping. The columns were placed in a chamber under controlled conditions with a 12 h light and 12 h dark cycle to reproduce the diurnal cycle. The temperature was controlled to ensure a constant soil temperature of 15°C (±1°C). The soil moisture content in the columns was adjusted every three days by weighing them and adding water at the top of the columns.

The experimental setup first consisted of 14 days of incubation, after which the macrocosms were imaged by X-ray tomography to assess the 3D burrowing systems. Then, at t = 15 days, we spread the organic products corresponding to each treatment on the top of each column. The columns were constantly observed for 4 h and were then regularly observed for the next two days to report the occurrence of any earthworms at the soil surface. Columns were observed regularly and the presence of casts recorded. Two weeks later, at t = 28 days, the macrocosms were imaged again by X-ray tomography. The soil columns were opened at t = 32 days to retrieve the earthworms, and all living earthworms were fasted and weighed. For both imagery sessions, the X-ray tomography (CT Siemens Somatom® Definition AS 128, Siemens, Germany) used the same parameters: 120 kV, 50 mA, resolution of 0.7 mm x 0.7 mm x 0.7 mm. Tomography images were analyzed using ImageJ software (Schindelin et al., 2012). We first binarized the images to separate soils and burrows using a single manually fitted threshold. Macropores with a section less than to 5.0 mm^2 were not considered (noise). Then, we used the 3D diameter of the burrows to discriminate the burrows of *L. terrestris* and *A. caliginosa* using the Local Thickness plugin (the threshold for the diameter was set to 7.57 mm). For each macrocosm and each species, we computed the volume of macropores at t = 14 days, the difference of macropores volume between t = 28 days and t = 14 days, and the mean burrow diameter at t = 28 days.

We computed the volume of macropores on four soil layers of 7.5 cm each per macrocosm (0 - 7.5 cm, 7.5 – 15 cm, 15 – 22.5 cm, 22.5 – 30 cm).

We compared five treatments (with five replicates each) run at the same soil humidity of 70% WHC as measured for sieved soil: (C) control treatment, in which 40 t ha^{-1} of water was spread; (LD40) 40 t ha^{-1} of liquid territorial digestate; (S40) 40 t ha^{-1} of slurry; (LD80) 80 t ha^{-1} of liquid territorial digestate; (ALD40) 40 t ha^{-1} of aerated liquid territorial digestate. A supplementary treatment was added (LD40-H+) with 40 t ha^{-1} of liquid territorial digestate, and a higher soil moisture content of 80% WHC. We also included two control columns without earthworms (70% and 80% WHC), reaching a total of 32 columns. The (ALD40) treatment was used to test whether a decrease in the ammonia concentration affects earthworm behavior. The (LD40-H+) treatment was used to test whether soil moisture can change the impact of digestate according to worm locations.

7.2.2 Statistical analyses

Statistical analyses were performed with R software (v.4.0.2) (R Core Team, 2020). The statistical significance among treatments of all variables was tested using a significance level of 0.05. In the field experiment, the difference in the abundance and biomass of earthworms within each sampling date among treatments was assessed using Kruskal-Wallis (kruskal.test function from stats package, R Core Team, 2020) and Dunn's tests (dunn.test function from the *dunn.test* package, Dinno, 2017). The evolution of earthworm abundance and biomass within each treatment between both sampling dates was tested using one-sided Wilcoxon-Mann-Whiney tests (wilcox.test function from *stats* package, R Core Team, 2020). We also simultaneously examined the effects of the treatment and date on earthworm abundance in the field (grouped data of both sampling dates) with the Sheirer-Ray-Hare test. The Sheirer-Ray-Hare test is a nonparametric test conducted in lieu of two-way ANOVA and an extension of the Kruskal-Wallis test (Sokal and Rohlf, 1995), followed by a post hoc Dunn's test. We used the scheirerRayHare function from the *rcompanion* package (Mangiafico, 2020). In the microcosm experiments, the difference in earthworm weight change among treatments was tested using Kruskal-Wallis with Dunn's tests. The difference in earthworm mortality among treatments was tested with a Fischer exact test followed by a pairwise Fischer exact test as a post hoc test (fisher.bintest function from RVAideMemoire package, Hervé, 2020). We examined the correlation between mixture pH and earthworm mortality using the cor.test function (*stats* package, R Core Team, 2020), with the "pearson" method. In the column experiment, the difference in earthworm weight change and burrow system characteristics among treatments were tested using Kruskal-Wallis with Dunn's tests. Fischer exact tests and post hoc pairwise Fischer exact tests were used to test a difference in the presence of casts at the surface of the columns among treatments.

7.3 Results

7.3.1 Long-term and short-term impacts on earthworms of applying organic products in the field

In February 2019, after two years of the different amendments, the main species found in the soil in the field were *L. terrestris*, *A. caliginosa*, *Aporrectodea rosea* and *Allolobophora chlorotica* in all treatments.

Following both spreading events (19th of February 2019 and 12th of March 2019), we observed earthworm mortality at the soil surface immediately after applying the liquid digestate and raw digestate. The observed mortality rates were 0.1 to 2.7 earthworms m^{-2} (Table 7.4). Mortality appeared in the first 20 min after spreading, the only exception was the first application of slurry after which mortality was reported after 24 h (data not recorded) (Table 7.4). In all treatments, most dead earthworms were adult *L. terrestris*. Some large endogeic earthworms, such as *A. caliginosa*, also died after the second application (12th of March 2019). Juveniles as well as smaller endogeic earthworms, such as *A. chlorotica* or *A. rosea,* were not observed at the soil surface. Soil moisture was 26% (gravimetric, i.e. 85% of WHC) on 19th of February 2019, and 27% (gravimetric, i.e. 89% of WHC) on 12th of March 2019.

Table 7.4 – Evaluation of short-term mortality in earthworms in the field experiment: the numbers of earthworms found dead at the soil surface after the spreading of the organic products are indicated as the mean ± standard error. Adult earthworm abundance is expressed in earthworms m-2 (± standard error)

Date	Liquid digestate	Raw digestate	Cattle slurry	Mineral fertilization
First spreading event, 1 h after spreading	0.6 ± 0.3	0.1 ± 0.2	0	0
Second spreading event, 1 h after spreading	2.7 ± 1	1.5 ± 0.6	1.7 ± 0.7	0
Second spreading event, one day after the spreading	1.8 ± 1.2	0.7 ± 0.3	1.0 ± 0.6	0
Mean adult abundance (both sampling dates)	138 ± 21	82 ± 9	126 ± 15	68 ± 17

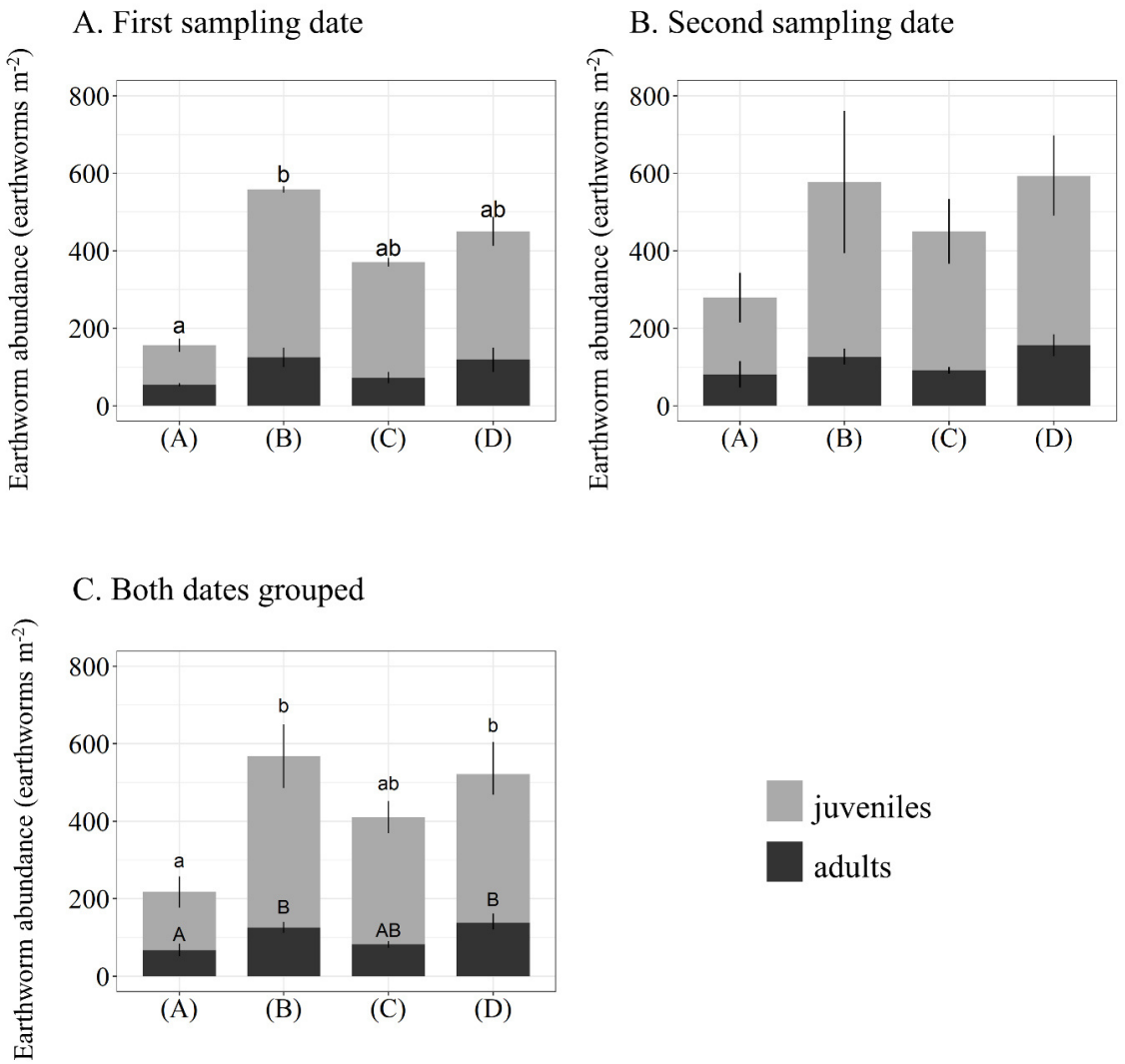


Figure 7.1 – Abundance of earthworms in fields before and after application of organic products. Error bars indicate standard errors. Two treatments with different lower case (or capital) letters indicate that the total (or adult, respectively) earthworm abundance in those treatments was significantly different ($p < 0.05$). No letter means that the treatment did not have a significant impact on adult or total abundance ($p > 0.05$). Each treatment is a fertilization system, using (A) chemical N fertilizers, (B) slurry and manure, (C) raw territorial digestate, (D) liquid and solid territorial digestates. A. Long-term effect of repeated application of organic products: first sampling date, in plots which received the different amendments for two years, seven days before spreading. B. Second sampling date, 14 days after spreading. Comparison with subfigure 1.A shows the impact of fertilization after two weeks on soil earthworm populations. C. Two sampling dates grouped together.

Adult earthworm abundance did not differ before and after the first application (19th of February 2019) of any treatments ($p > 0.05$). During the same period, total earthworm abundance only increased in the mineral N treatment ($p = 0.05$) (Figure 7.1). Considering biomass, the only change was an increase in total biomass in the slurry and manure treatment ($p < 0.05$) (Supplementary Material 2).

For the two years of the different fertilization treatments, before the first spreading in 2019, the inputs of organic matter were 0 t VS ha^{-1}, 13.7 t VS ha^{-1}, 6.7 t VS ha^{-1}, and 15.7 t VS ha^{-1} for mineral N, slurry and manure treatment, raw digestate treatment, and liquid and solid digestate treatment, respectively. At the first sampling date, the total abundance of earthworms in the slurry and manure treatment was significantly higher than under the mineral N treatment without organic products ($p < 0.05$) (Figure 7.1). Treatment did not have a significant effect on the abundance of adult earthworms ($p > 0.05$). None of the treatments had a significant effect on biomass ($p > 0.05$) (Supplementary Material 2). At the second sampling date, the treatments had no effect on total or adult abundance ($p > 0.05$). Thus, because earthworm abundance did not change before and after fertilization, the data were grouped. Considering both dates, total and adult abundance were significantly higher under the slurry and manure treatment (+161% and +87%) and in the territorial solid and liquid digestates treatment (+147% and +109%) than under the mineral N treatment ($p < 0.05$). Total and adult abundances were not influenced by the sampling date ($p > 0.05$). Earthworm biomass was not impacted by treatment ($p > 0.05$).

7.3.2 Short-term ecotoxicity of organic products in microcosms under controlled laboratory conditions

In the microcosm ecotoxicity experiment, the mortality rate was more than 75% when territorial digestate was mixed in soils at doses of 10% (series #1) and 15% (series #2) for *L. terrestris*, and 20% for *A. caliginosa* (Figure 7.2). As a comparison, slurry caused a mortality rate of more than 75% at a 20% dose only for series #1 (both species), but not for series #2. A 15 % dose of digestate from cover crops was also lethal to 75% of *L. terrestris*. Mortality occurred rapidly: in all treatments in both series, 64% and 94% of deaths occurred within the first 24 h then within the first four days for *L. terrestris* (48% and 83% for *A. caliginosa*, respectively).

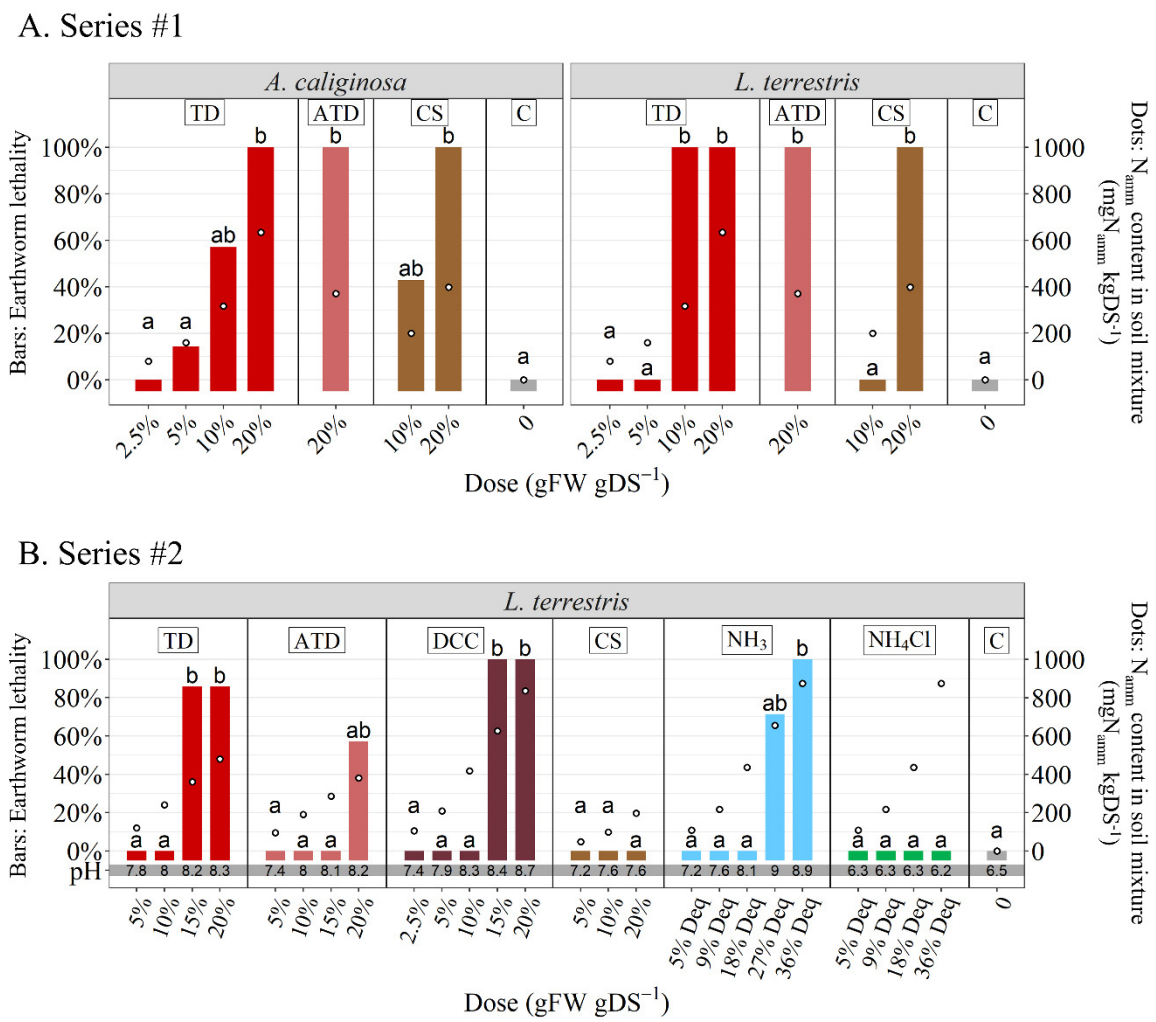


Figure 7.2 – Ecotoxicity tests of organic products in microcosms, series #1 (A) and series #2 (B). Bars represent the proportion of dead earthworms within the seven replicates for each treatment. Bars are still presented in the figure for treatments with no mortality. Dots indicate the ammoniacal N content in the (soil + organic products) mixtures, expressed in mg of ammoniacal N per g of dry soil (mgNamm kgDS-1). The organic product mixed with soil is indicated in boxes: (TD) liquid territorial digestate, (ATD) aerated liquid territorial digestate, (DCC) digestate from cover crops, (CS) cattle slurry, (NH3) ammonia solution, (NH4Cl) NH4Cl solution, (C) control. The dose of organic products in the mixture is expressed in g of fresh weight of organic product per g of dry soil (gFW gDS-1). The doses of the ammonia (NH3) and ammonium (NH4Cl) treatments are expressed in the territorial digestate equivalent dose (Deq): a dose of 5% Deq corresponds to an amount of ammoniacal N similar to a dose of 5% of the territorial digestate (gFW gDS-1). Within each series and species, different letters indicate that two treatments showed a significant difference ($p < 0.05$).

Considering only the treatments with at least three living earthworms, the mean weight change of *L. terrestris* was +6% and +4% in the control treatments of series #1 and #2, respectively. It ranged from -1% (slurry 10%) to +6% (liquid territorial digestate 2.5%) in series #1 (Supplementary Material 3), from +8% (cattle slurry at 20%) to -9% (aerated liquid territorial digestate at 15%) in series #2 (Supplementary Material 3). It was not affected by the treatments (series #1: $p > 0.05$; series #2: Kruskal-Wallis test $p < 0.01$, but Dunn's test $p > 0.05$). The mean weight change of *A. caliginosa* was -14% in the control treatment, ranging from -34% (10% slurry treatment, which was lethal to 42% of earthworms) to -7% (5% territorial digestate). The weight change was significantly different only between this 10% slurry treatment and the 5% territorial digestate treatment ($p < 0.05$). The experimental procedure was valid because earthworm weight loss in the control treatment was less than 20% (Fründ et al., 2010).

7.3.3 Microcosm experiment to investigate the causes of short-term ecotoxicity to L. terrestris

Three observations from our experiments were consistent with the hypothesis that ammonia is partly responsible for digestate toxicity. First, the ammonia solution was toxic to more than 75% earthworms at a dose of 723 mg N_{amm} kg DS^{-1} (0.9 g NH_3 kg DS^{-1}), which corresponded to the ammoniacal N content of the soil mixture containing 27% of territorial digestate (Figure 7.2B). Moreover, the lethal doses of the digestate from cover crops (15% and 20%) corresponded to 630 mg N_{amm} kg DS^{-1} (0.8 g NH_3 kg DS^{-1}) and 840 mg N_{amm} kg DS^{-1} (1.1 g NH_3 kg DS^{-1}), which were similar to lethal ammoniacal N concentrations in ammonia solutions (Figure 7.2B). Finally, as a partial negative control, aeration of the territorial digestate, reduced the ammoniacal N concentration (Table 7.1) and decreased toxicity in series #2. Aerated territorial digestate caused the same mortality rate at similar doses as territorial digestate in series #1 (both species), and lower mortality rates in series #2, with 0% at the 15% dose and 60% at the 20% dose (Figure 7.2B). Regarding the hypothesis that ammonia drove toxicity, soil mixed with the aerated territorial digestate (20%) had a similar soil N_{amm} content to the soil mixed with unaerated territorial digestate (10%), which was lethal to *L. terrestris* (Figure 7.2A). However, the ammonia content alone could not explain territorial digestate toxicity. The ammoniacal N content of lethal mixtures containing territorial digestate (15% and 20%) were equal or lower than 380 mg N_{amm} kg DS^{-1}. A similar ammoniacal N concentration in the NH_3 treatment was not lethal (Figure 7.2B). The territorial digestate was lethal at the same dose as the digestate from cover crops (15%), which had a similar pH but lower ammoniacal N concentration (Figure 7.2B).

The pH of the soil + organic product mixtures correlated with mortality (Pearson's $r = 0.65$, $p < 0.001$). All lethal treatments showed a mixture pH greater than 8.2 (Figure 7.2B). NH_4^+ was never lethal, demonstrating the importance of the ammoniacal N form (NH_4^+/NH_3) in toxicity.

7.3.4 Short-term impact of organic products on earthworm behavior and survival in soil columns

In the soil columns, despite spreading the organic products at high (40 t ha^{-1}) and very high (80 t ha^{-1}) doses, we did not observe mortality at the surface or inside the soil. The mortality recorded at the end of the experiment was low and was not significantly different between treatments (3% for *L. terrestris* and 6% for *A. caliginosa* on average). The treatments did not significantly affect the changes in earthworm weight observed over the course of the experiment ($p > 0.05$ for both *L. terrestris* and *A. caliginosa*) (Supplementary Material 4). In the control treatment, earthworm weight decreased by 18% and 16% for *L. terrestris* and *A. caliginosa*, respectively. This validated the experimental procedure (Fründ et al., 2010).

L. terrestris casts were found at the surface in all columns in the S40 treatment, in two columns for ALD40, LD80, and LD40-H+, and in no columns in the LD40 and control treatments. We found that a treatment had a significant effect on the presence of casts ($p < 0.01$). The addition of slurry resulted in some solids, including straw, remaining on the soil surface, which was burrowed into the soil by the earthworms within the week after spreading. In contrast, the digestate infiltrated the soil during the first day following spreading. Traces of digestate were found in large *L. terrestris* burrows when the columns were destroyed at the end of the experiment, suggesting the quick preferential infiltration of digestate.

Using image analysis, we could satisfactorily distinguish between *A. caliginosa* and *L. terrestris* burrows (Figure 7.3). Before applying organic matter, there was no difference in burrow excavation at any depth for the two species in any of the standard humidity conditions ($p > 0.05$; Figure 7.4). In the LD40-H+ treatment, in the presence of high soil moisture content, *A. caliginosa* burrowed significantly more in the first 15 cm and less below 15 cm compared to in the other treatments ($p < 0.05$). While *A. caliginosa* burrowed homogenously within the first 22.5 cm before inputs, these endogeic earthworms burrowed less in the first and fourth soil layers after application of the digestates (LD80, LD40). In contrast, in the S40 and C treatments, endogeic earthworms still burrowed homogeneously at all depths (Figure 7.4). Considering the entire soil core, *A. caliginosa* burrowed significantly less after spreading in the LD80 treatment than in the slurry and control treatments ($p < 0.05$). Except for LD40-H+, *L. terrestris* burrowing activity was not affected by treatment at any depth. None of the treatments had a significant impact on burrow mean diameter for either species ($p > 0.05$).

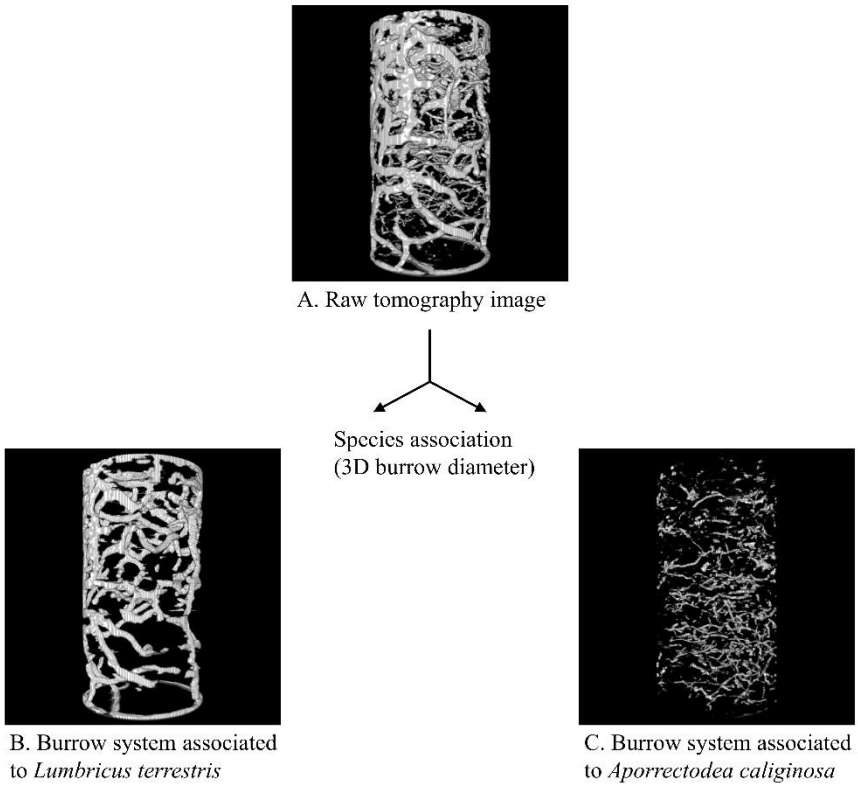


Figure 7.3 – Example tomography images before (A) and after the separation of the different burrow systems built by L. terrestris (B) or A. caliginosa (C).

7.4 Discussion

7.4.1 Long-term impact of digestates on earthworm populations

Here, we confirmed that (H1) digestates can have a positive effect on earthworms in the long term, despite short-term negative effects. This is in accordance with previous studies. In organic grassland, Clements et al. (2012) observed a similar earthworm biomass but higher earthworm abundance in plots fertilized with slurry (+125%) or liquid digestate (+53%) six weeks after application compared with unamended plots. Four months after spreading slurry or liquid digestate, Koblenz et al. (2015) found similar positive effects on earthworm abundance (+33% to +126% and +34% to +100%, respectively) and biomass (+49% to +208%, and +42% to +143%, respectively) compared with unamended treatments. In a restored colliery soil, Butt and Putwain (2017) observed a higher abundance (+150% to +232%) and biomass (+120% to +195%) of earthworms in plots amended with solid digestate than in the untreated control plots after two years of application.

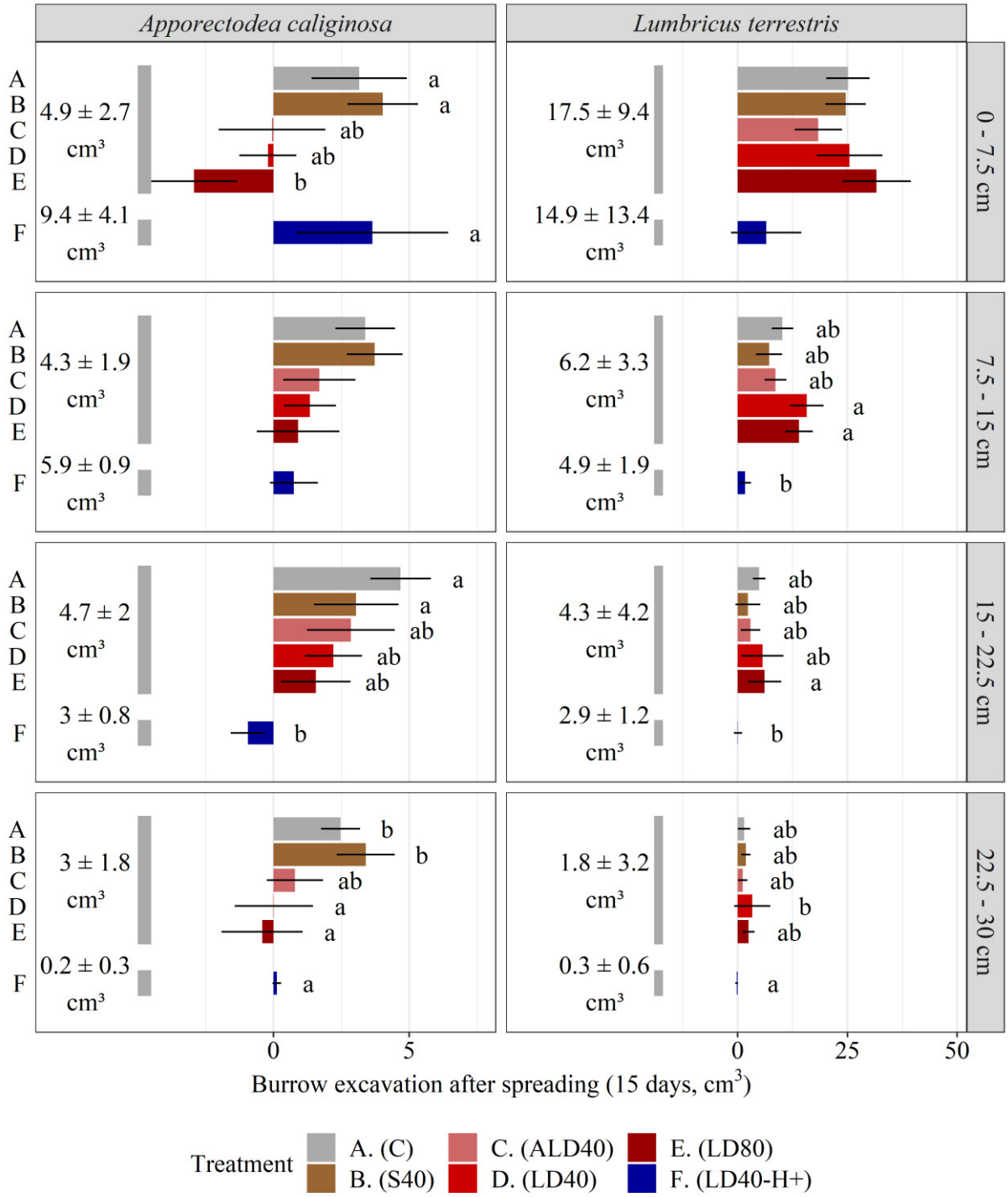


Figure 7.4 – Difference in burrow volume after the application of organic products between day 14 and day 28 (15 days) for both earthworm species and for all four soil layers. For each species and soil layer, different letters indicate that the treatments showed significant differences ($p < 0.05$). The absence of letters indicates that there was no significant difference between the treatments ($p > 0.05$). In the left part of each subgraph, we indicate the burrow volume associated with each soil moisture content during the first 14 days of the experiment (day 0 to day 14) before spreading (± standard deviation). Treatments are: (C) control, (S40) cattle slurry (40 t ha-1), (ALD40) aerated liquid territorial digestate (40 t ha-1), (LD40) liquid territorial digestate (40 t ha-1), (LD80) liquid territorial digestate (80 t ha-1), (LD40-H+) liquid territorial digestate (40 t ha-1), high SWC.

This positive effect of digestate and animal effluents was likely due to the input of organic matter, which can serve as a food source for earthworms (D'Hose et al., 2018; Ernst et al., 2008; Sizmur et al., 2017). After two years, the lower observed earthworm abundance in plots treated with raw digestate was most likely due to the overall lower input of organic matter contained in the raw digestate compared to liquid and solid digestate or slurry and manure treatments (see section 7.3.1). Solid organic amendments (manure or solid digestate) and liquid organic fertilizers (slurry or liquid digestate) could promote earthworm populations differently, we cannot determine their relative impact from the field experiment described in this study. The different organic products may have different feeding properties due to their C and N availability and calorific value (Ernst et al., 2008; Sizmur et al., 2017) and thus benefit earthworm species differently (Ernst et al., 2008; Onrust and Piersma, 2019). Replacing animal effluents with digestate may affect earthworm populations, depending on primarily the quantity but also the quality of organic matter contained in the products (Abail and Whalen, 2018).

Another hypothesis to explain the evolution of earthworm populations in the long-term was a change in the soil C:N ratio due to the organic product inputs. Earthworm abundance appeared to increase in soil with a low C:N ratio (De Wandeler et al., 2016; Nieminen et al., 2011). Although the C:N ratio was determined only at the beginning of the field experiment, several other studies showed no change in C:N ratios in long-term field experiments with repeated digestate inputs similar to ours (Barłóg et al., 2020; Głowacka et al, 2020; Pastorelli et al., 2021; Persson et al., 2020; Zicker et al., 2020). Thus, this scenario appears to be less probable than one where the amount of organic matter brought to field explains the observed differentiation in earthworm populations.

7.4.2 Comparison of short-term effects under field and laboratory conditions

Consistent with our findings, Burmeister et al. (2015) found that digestate toxicity had a low effect on earthworm populations in the field, recording less than one dead earthworm per m^2 after liquid digestate spreading (0.3% of the population). A Norwegian study (Johansen et al., 2015; Løes et al., 2014) observed a higher mortality of *L. terrestris*, *Lumbricus rubellus*, *A. caliginosa*, and *A. rosea* (between two and 19 dead earthworms m^{-2}, respectively, corresponding to 1.3% and 17% of the population) after undigested and digested slurry was applied at different doses in a grass-clover field.

Several studies focused on solid digestates (Pivato et al., 2016; Renaud et al., 2017; Ross et al., 2017; Sizmur et al., 2017) that were later buried in the soil, so these authors did not assess earthworm surface foraging immediately after spreading. To our knowledge, this is only the second study (after Natalio et al. (2021)) to propose a microcosm experiment for assessing liquid digestate toxicity. We found lower lethal concentrations of digestate compared to that reported by Pivato et al. (2016) (40% solid digestate on a dry matter basis, corresponding to 145% on a fresh matter basis) and Krishnasamy et al. (2014) (80% fresh solid digestate together with 20% sawdust) for *E. fetida*. This can be explained

by the relatively lower sensitivity of this species (Pelosi et al., 2013) and the nature of the digestates applied (solid versus liquid). In contrast to the findings from microcosm experiments, we detected no mortality within the soil two weeks after slurry and digestate was spread in either the field or in the soil columns. This can be explained by avoidance leading to a lower probability of contact between the earthworms and the products. Only earthworms that came to the surface immediately after spreading were in direct contact with digestate concentrations close to or higher than the lethal concentration observed in our laboratory experiments.

We thus confirmed that the (H2) digestate and slurry can be lethal to earthworms in the short term and that this mortality appears to be limited to the soil surface.

7.4.3 Probable causes of liquid organic product toxicity in the short term

The presence of ammonia in the liquid organic products likely explains some of their ecotoxicity to earthworms. Hughes et al. (2008) measured an LC_{50} of 1.5 g NH_3 per kg of soil for *E. fetida*, which was slightly higher than the ammonia concentration that was lethal to *L. terrestris* in our microcosm experiment. Once more, the difference may be explained by the different sensitivity of the two species (Pelosi et al., 2013). Krishnasamy et al. (2014) found that concentrations of a solid digestate that were toxic to *E. fetida* in soils corresponded to ammonium concentrations between 0.9 and 1.2 g NH_3 kg Dry $Soil^{-1}$ (g NH_3 $kgDS^{-1}$), which is consistent with the lethal dose of digestate from cover crops. Furthermore, aeration of the digestate decreased its toxicity in our experiment, suggesting that at least one toxic compound in the digestate is volatile, which is a well-established characteristic of ammonia. Ammonia toxicity can also explain why we observed differences in the toxicity of slurry in the microcosms between series #1 and series #2. In the first series, the slurry was toxic and had a higher N_{amm} concentration than in the second series where no toxicity was observed. Consistent with our results, Hughes et al. (2008) also found that the form of ammoniacal N (NH_3 or NH_4^+) influenced its toxicity. The $NH_4^+ \rightleftarrows NH_3 + H^+$ acid-base equilibrium depends on the pH. A high pH promotes a high NH_3 concentration, with a pKa of 9.25 being observed at 25°C in ideal aqueous solution. Both digestates and soil pH may thus influence ammoniacal N toxicity.

At the lethal doses of the territorial digestate in the microcosms, the ammoniacal N soil content was too low to completely explain the observed toxicity: this suggests that other compounds originating from the inputs may also affect digestate toxicity to earthworms. Urine degradation products such as benzoic acids or sulphides (Curry, 1976) could also be toxic, as could diverse ionic compounds through salinity or CEC (Natalio et al., 2021; Pivato et al., 2016; Tigini et al., 2016). However, it is unlikely that heavy metal contamination was involved in the observed short-term toxicity. In the lethal 20% dose of the territorial digestate, the concentrations of heavy metals were: 2.5 mg Cu kg DS^{-1}, 6.6 mg Zn kg DS^{-1}, and 0.2 mg Pb kg DS^{-1} (Table 7.2). This is much lower than the estimated LC_{50} values for *Aporrectodea tuberculata* found by Lukkari et al. (2005) (134 mg Cu kg DS^{-1} and 234 mg Zn kg DS^{-1}) or the

concentration of heavy metals in soils that was not lethal soils to *L. terrestris* (Kennette et al., 2002). Similarly, the PAH content in this treatment was 7.4 mg kg DS^{-1}, much lower than the LC_{50} for PAH towards *E. fetida* estimated in Eom et al. (2007).

In summary, we confirmed that (H3) ammonia is partly responsible for digestate toxicity, but further studies are required to understand other potential toxicity mechanisms.

7.4.4 Earthworm behavior in the short term after organic product spreading

When liquid organic products, i.e. slurry and digestates, were spread on the soil, earthworms came up to the surface. This is a key mechanism for understanding field ecotoxicity. Mortality was only observed 24 h after the first application of slurry in the field. As the solid phase of slurry could not infiltrate the soils very rapidly, we assume that anecic earthworms came to the surface at night, as commonly observed in epi-anecic species such as *L. terrestris* (Bastardie et al., 2005), and then came in contact with pools of slurry. Similar to our experiment, Brauckmann and Broll (2007) spread a digestate within soil cores (30 cm high, 6800 cm^3) and did not observe surface mortality. Because the phenomenon affected only a small percentage of earthworms in the field, it makes sense that it was not detected in the column experiments where the total number of earthworms involved was only 35 per treatment.

In contrast to the slurry treatments, no *L. terrestris* surface activity (casts) was observed in columns amended with digestate. This suggests that the digestate and thus organic matter infiltrated efficiently within the column and that *L. terrestris* did not need to come to the surface to feed. *A. caliginosa* burrowed less in the presence of digestate than slurry, particularly in the surface soil layer. A first hypothesis is that *A. caliginosa* avoided the first soil layer and decreased its activity due to possible digestate toxicity. A second hypothesis is that the infiltration of the digestate within the soil increased the presence of accessible food, allowing these endogeic earthworms to decrease their activity (Frazão et al., 2019; Hughes et al., 1996). The lack of significant differences in weight change between the treatments as well as the toxic effect shown in the microcosm experiments tends to favor the first hypothesis of a toxic effect. Indeed, Ross et al. (2017) observed that *A. caliginosa* avoided soil layers containing digestate and Ernst et al. (2008) found that *A. caliginosa* could not feed on raw digestate (weight loss was observed in these 1500 mm^3 mesocosms). This avoidance of digestate could explain why some *A. caliginosa* came to the surface in the field after digestate spreading, which is not a common behavior for endogeic species. The negative burrowing activity of *A. caliginosa* seen in some treatments (e.g., LD80) was due to large quantities of digestate or the worms own casts filling burrows, or *L. terrestris* activity destroying the endogeic burrows. We confirmed that the (H4) digestate has an impact on earthworm behavior in the short term, with different possible outcomes, to either limit toxicity (avoidance) or trigger mortality (surface foraging).

According to Burmeister et al. (2015) and Van Vliet and de Goede (2006), digestate toxicity only occurs under humid soil conditions, which was the case when we observed earthworm mortality in the field. When the soil is moist, earthworms stay closer to the surface and may be more exposed to the applied organic products. However, we did not observe surface mortality after liquid digestate spreading in soil columns with high soil humidity (LD40-H+), where the earthworms were close to the surface; therefore, we could not reach a conclusion regarding this point.

7.5 Conclusions

To promote sustainable agricultural practices, the impacts of using digestates and slurry need to be better assessed in the soil and on the associated living organisms. This case study provided new insights regarding the short- and long-term effects of anaerobic digestates on earthworms. In the short term, liquid organic products may be toxic to earthworms mainly when direct contact occurs. This toxicity depends on the ammonia concentration, which is promoted by high pH, and other elements that could not be clearly identified here and will require further research. In the field, mortality likely occurred shortly after application because earthworms foraged at the soil surface, where the products were highly concentrated. In the longer term, we found that earthworm populations increased after two years of regular input of organic products. Further research is needed to better identify the causes of the toxicity of digestates from different substrates, to highlight the parameters explaining why earthworms come to the surface after spreading, to compute the frequency of lethal events in the field, and to estimate the feeding properties of digestates in the long term. The impact of different solid and liquid digestates on earthworm ecology (e.g., species, behavior, functions) still needs to be investigated in different soils.

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Chapitre 8

Discussion générale et conclusion

8.1 Résumé des résultats majeurs de la thèse

La méthanisation agricole est une pratique en développement dans le monde et en particulier en France. L'évaluation des effets de l'introduction d'un méthaniseur sur l'environnement dans une exploitation est une question complexe, car elle entraine de nombreux effets interagissant les uns avec les autres. Dans cette thèse, nous cherchions donc à répondre à la question :

Comment la méthanisation agricole en polyculture élevage modifie-t-elle les cycles du carbone et de l'azote à l'échelle de la ferme, c'est-à-dire sur l'ensemble de la filière du traitement des effluents jusqu'à l'épandage des digestats au champ ?

L'état de l'art (Chapitre 1) a montré que, si l'épandage de digestat avait fait l'objet de nombreuses études, nous manquions de travaux prenant en compte de multiples effets simultanément. Il y a aussi un manque d'informations sur l'effet des digestats à long terme sur la vie du sol et le stockage de matière organique du sol. Ces questions ont été traitées à l'aide d'un cas d'étude en région Centre – Val de Loire (Chapitre 2). Dans un essai au champ permettant de comparer les flux d'azote dans une succession de cultures fertilisées avec des effluents d'élevage non traités ou méthanisés (avec ou sans post traitement par séparation de phase) ou avec des engrais de synthèse, nous avons confirmé le bon pouvoir fertilisant des digestats (forte teneur en azote ammoniacal accompagnée parfois de la minéralisation de l'azote organique), ainsi que leur sensibilité à la volatilisation, y compris pour les digestats solides (Chapitre 3). Sur le même cas d'étude, nous avons montré qu'il était possible de simuler avec une précision correcte les dynamiques du C et du N dans ces différentes situations avec le modèle sol-plante STICS (Brisson et al., 2008) (Chapitre 4), et la qualité des digestats avec le modèle de méthanisation SYS-Metha (Bareha et al., 2021b) (Chapitre 5). Nous avons donc utilisé ces deux modèles pour comparer différents scénarios de gestion des effluents à l'échelle d'une ferme en incluant traitement amont et épandage au champ. Nous avons ainsi simulé les flux de C et N d'une exploitation en polyculture-élevage avec un méthaniseur. Ces flux sont en premier lieu impactés par les apports de déchets extérieurs. Dans le périmètre du plan d'épandage du méthaniseur, la méthanisation permet de diminuer l'utilisation d'engrais azotés de synthèse, de maintenir ou d'augmenter la teneur en matière organique dans les sols, mais augmente aussi la volatilisation d'ammoniac (Chapitre 6). Néanmoins, la méthanisation n'impacte pas uniquement les flux de C et N. À titre d'exemple, nous avons étudié l'effet des digestats et d'effluents animaux sur les vers de terre au champ (Chapitre 7). Nous montrons que les vers de terre peuvent être négativement impactés à très court terme après épandage de digestat liquide ou de lisier, mais que les effets de l'apport de matière organique sont positifs après 2 ans d'apports. Ce cas d'étude est loin de permettre une conclusion généralisée sur l'effet des digestats sur la biologie du sol, il s'agit d'un premier pas proposant une base de réflexion pour des études à venir sur cette question.

Beaucoup d'expérimentations liées à l'étude de l'épandage des digestats se situent à l'échelle de la parcelle expérimentale, et comparent ainsi la qualité des digestats à celle des effluents animaux (Chapitre 1). Cette thèse appelle à considérer les effets agronomiques de l'introduction d'un méthaniseur à l'échelle d'une exploitation ou d'un plan d'épandage, et donc de dépasser l'échelle de la parcelle expérimentale. L'échelle de l'exploitation permet d'abord de considérer à la fois les effets de la méthanisation sur le traitement des effluents, et sur les effets des PRO après épandage aux champs. Ceci a déjà été évoqué dans la littérature (Amon et al., 2006; Holly et al., 2017; Neerackal et al., 2015). De plus, l'échelle de l'exploitation permet aussi de considérer à la fois un changement de qualité, mais aussi un changement de quantité des produits résiduaires organiques (PRO) disponibles en lien avec les types et quantités d'intrants dans le méthaniseur. Cela permet d'élargir et de compléter les messages sur l'effet des digestats émanant d'études qui s'intéressent à l'échelle de la parcelle. L'échelle de l'exploitation pourrait aussi permettre d'analyser les effets d'un changement d'allocation des PRO sur des cultures différentes.

Ainsi, la thèse apporte des réponses à ces questions, introduites dans le Chapitre 1, qui s'adresse à la méthanisation des effluents bovins à la ferme. Pour ces questions, nous illustrerons nos conclusions par des chiffres issus de la simulation de deux types de méthaniseurs à la ferme (Chapitre 6) : un méthaniseur agricole, valorisant en grande majorité des effluents d'élevage avec quelques déchets agricoles (scenario EI30A, 2000 t de déchets par an), et un méthaniseur agricole et territorial, deux fois plus important, valorisant de nombreux déchets agroindustriels et urbains en plus des effluents d'élevage (scenario ADb, 4500 t de déchets par an).

Comment les caractéristiques des digestats, influencées par les conditions de stockage ou de post-traitement, déterminent-elles leurs propriétés lors de leur épandage au champ ?

Les PRO de notre cas d'étude suivent la règle générale issue de la littérature selon laquelle les digestats liquides et bruts sont plus riches en azote ammoniacal que les lisiers dont ils sont issus (Chapitre 3). Les digestats sont donc plus faciles à utiliser pour fertiliser les cultures : l'azote est plus facilement valorisable par les plantes, et il est possible d'apporter des doses importantes d'azote minéral plus facilement.

Lors d'une séparation de phase par presse à vis, la phase liquide est très semblable au digestat brut, en termes de teneur en nutriments (Chapitre 3). Néanmoins, lors de leur épandage pour fertiliser les cultures, les digestats bruts s'infiltrent moins facilement, ce qui va augmenter les risques de volatilisation d'ammoniac et diminuer leur efficacité fertilisante. Au champ, nous estimons ainsi que le coefficient d'équivalence aux engrais azotés de synthèse (Keq) des digestats liquides se situent entre 0,60 et 0,85, et celui des digestats bruts entre et 0,35 et 0,60 (Chapitre 3). Ces Keq sont calculés avec des apports répétés de produits sur un même champ, et ne correspondent donc pas exactement aux Keq indiqués par les Chambres d'Agriculture, par exemple. La couverture de fosse permet de réduire les pertes d'azote

ammoniacal lors du stockage et donc d'enrichir significativement les digestats en azote. Cela les rendra meilleurs en tant que fertilisants azotés, si les pratiques d'épandage permettent de limiter la volatilisation au champ (Chapitre 6).

Peut-on quantifier globalement les effets de la méthanisation sur les différents flux azotés d'intérêt, comme la substitution des engrais chimiques par les digestats, la volatilisation d'ammoniac, les émissions de N_2O, ou la lixiviation des nitrates, en considérant les différentes sources d'émissions ?

Dans cette thèse, nous validons l'hypothèse que les principaux effets de la méthanisation concernant le cycle de l'azote sont dirigés par l'import d'azote sur l'exploitation, et sa minéralisation partielle durant la digestion. Cela influe d'abord sur la substitution des engrais de synthèse. Le méthaniseur agricole diminue l'utilisation d'engrais azotés de synthèse d'environ 10% et les flux d'azote entrant sur l'exploitation de 2% (engrais de synthèse et produits organiques) ; le méthaniseur plus territorial diminuent l'utilisation d'engrais d'environ 30% en augmentant les entrées d'azote de 4% (Chapitre 6). Le deuxième flux le plus impacté est la volatilisation d'ammoniac, qui est augmentée au stockage et lors de l'épandage. Le méthaniseur agricole et le méthaniseur plus territorial augmentent la volatilisation de NH_3 de 35% et 95%, respectivement (Chapitre 6). Ceci est principalement dû aux digestats liquides : si à l'échelle du champ les digestats solides sont émetteurs d'importantes quantités d'ammoniac (Chapitre 3), la faible masse de digestat solide comparée à celle de digestat liquide rend ces émissions beaucoup moins significatives à l'échelle de la ferme (Chapitre 6).

Dans le cas où les digestats sont principalement épandus en hiver, les émissions de N_2O seraient peu impactées. Ceci est confirmé par nos observations au champ (Chapitre 3). Le méthaniseur agricole ne change pas non plus les émissions de N_2O au stockage des PRO. À l'inverse, en considérant nos hypothèses et facteurs d'émissions, le méthaniseur agricole et territorial augmente les émissions de N_2O au stockage de 141% à cause de la plus grande présence de digestat solide. Cela représente l'équivalent de 0,2 kgN par hectare cultivé, l'impact reste donc modéré. Les flux de N_2O sont moins facilement évaluables précisément, et ces données sont soumises à de plus fortes incertitudes. Enfin, dans notre cas d'étude, la lixiviation de nitrate reste faible, et nous ne détectons pas d'effet de la méthanisation sur ces pertes. Nous rappelons seulement que les apports de PRO devant blé favorisent la lixiviation de nitrate (Chapitres 3 et 4). Cette question reste importante à traiter dans d'autres conditions pédoclimatiques.

Les chiffres indiqués sont des ordres de grandeur qui vont varier en fonction des situations et des pratiques. Ils concernent uniquement le périmètre de la ferme, et ne prennent pas en compte d'éventuels effets déplacés, dû à la valorisation de déchets qui étaient déjà épandus au champ par exemple.

Quel est l'effet de la méthanisation sur le stockage de MOS, la qualité des sols, la fourniture en azote du sol, et la vie du sol ?

À masse fraîche égale, les digestats liquides possèdent beaucoup moins de matière organique que les digestats solides (30 g kg^{-1} et 230 g kg^{-1} avec nos analyses, respectivement) (Chapitre 3). Les digestats liquides ont donc de faibles propriétés d'amendement organique, c'est-à-dire de capacité à augmenter les stocks de MOS (Chapitre 3), malgré une matière organique assez récalcitrante (ISMO = 73%) (Chapitre 3). Néanmoins, à l'échelle de l'exploitation, la quantité de C dans les digestats liquides est deux fois plus importante que celle des digestats solides. Les digestats liquides sont donc un atout majeur pour l'entretien des stocks de MOS dans les exploitations avec un méthaniseur (Chapitre 6). Les simulations montrent que le méthaniseur agricole et celui plus territorial induisent une diminution de 0,1 tC ha^{-1} et une augmentation de 0,7 tC ha^{-1} des stocks de MOS à l'échelle de l'exploitation, respectivement (Chapitre 6). Ces variations sont d'une ampleur faible à moyenne. Ce changement d'échelle de la parcelle à la ferme n'a pas pu être réalisé pour les résultats sur la biologie du sol : dans le Chapitre 7, nous n'avons regardé l'effet de l'épandage de digestat sur les vers de terre qu'à l'échelle du champ. Il s'agit d'une question importante, car, si nous avons identifié que les digestats liquides pouvait avoir de faibles effets négatifs à très court terme sur les vers de terre, l'effet positif du mode de fertilisation avec digestats sur les populations de vers de terre n'a pas permis de distinguer les effets des digestats liquide ou solide, et les effets différenciés de ces deux PRO reste à étudier.

Quel est l'effet de l'import de déchets extérieurs sur la qualité et la quantité des digestats et donc les bénéfices et risques de la méthanisation ?

L'import de déchets entraîne des apports de nutriments et de matière organique sur l'exploitation. Dans nos simulations du Chapitre 6, environ 75% à 80% du C importé est transformé en biogaz, 1% à 2% est émis dans l'atmosphère sous forme de méthane, et 5% à 7% est stocké dans les sols après 20 ans. En ce qui concerne le N, 64% à 80% du N importé se substitue aux engrais azotés, 18% à 23% est émis dans l'atmosphère sous forme de NH_3, et 2% à 5% est stocké dans la MOS après 20 ans. Il s'agit bien évidemment d'ordres de grandeur, ces chiffres vont varier en fonction de la nature des intrants et des situations.

L'import des déchets va donc favoriser la substitution des engrais de synthèse et le stockage de matière organique dans les sols, mais également favoriser les émissions de gaz à effets de serre et les pertes azotées. Au regard de ces résultats, il est probable que l'intérêt de l'import de déchets soit plus grand dans des territoires où on observe un manque de ressources organiques. À l'inverse, si les plans d'épandage sont déjà saturés par les effluents d'élevage et dans le cas où les meilleures pratiques d'épandage sont appliquées pour réduire la volatilisation ammoniacale, l'import de déchets risque d'augmenter les pertes azotées, et en particulier la lixiviation des nitrates que nous n'avons que peu observée dans nos conditions.

Peut-on quantifier et comparer les effets de différentes pratiques lors du traitement, du stockage, ou de l'épandage des digestats ?

La couverture de la fosse de stockage des digestats liquides est importante ; nous estimons qu'elle permet de réduire la volatilisation totale de NH_3 de 40% environ, et de diminuer la consommation d'engrais de synthèse de 15% environ, dans les simulations du méthaniseur agricole et territorial (Chapitre 6).

L'injection des digestats est une manière très efficace au champ pour réduire la volatilisation (Maris et al., 2021), à l'échelle de la parcelle. Néanmoins, l'injection est en fait contrainte par les cultures sur lesquelles le digestat est épandu, diminuant légèrement son efficacité réelle (Chapitre 6). Dans nos simulations, l'injection permet néanmoins de réduire les émissions de NH_3 de l'ordre de 10 à 15% à l'échelle de l'exploitation.

La séparation de phase des digestats augmente la volatilisation de NH_3 au stockage (Baldé et al., 2018) et la diminue à l'épandage (Chapitre 3). Les simulations montrent que la résultante de ces deux effets est une légère diminution de la volatilisation (5%). Cette diminution est du même ordre de grandeur pour une valorisation des digestats au printemps, au lieu de toute l'année, et en majorité sur prairie. La séparation de phase semble affecter les émissions de gaz à effet de serre lors du stockage : elle induit plus d'émissions de N_2O à cause du digestat solide stocké en condition aérobie (Chapitre 6), et possiblement moins d'émissions de méthane (Gioelli et al., 2011). À cause de l'incertitude sur les facteurs d'émissions, nous n'avons pas pu conclure sur l'effet global de la séparation de phase sur les émissions de gaz à effet de serre.

Bilan

En répondant à ces questions, cette thèse propose également des résultats opérationnels. Les « bonnes pratiques » associées à la méthanisation agricole sont en grande partie connues (épandage des digestats avec un matériel adapté, sur des cultures et à des doses appropriées, couverture de fosse de stockage), comme le montre par exemple un récent « guide des bonnes pratiques » associées à l'épandage de digestat (AgroParisTech, 2021). Néanmoins, cette thèse permet de quantifier l'effet de plusieurs de ces pratiques et de les hiérarchiser. Par exemple, nous confirmons l'importance de la couverture des fosses de stockage des digestats. Dans ce même Chapitre 6, nous utilisons les dernières informations disponibles en terme de modélisation des stocks de matière organique du sol (MOS) en cas d'apport de digestat et par rapport à des apports d'effluents non méthanisés, pour montrer que l'effet sur la MOS dépendrait en grande partie du type et de la quantité de déchets extérieurs méthanisés. Ensuite, nous proposons des éléments de réponses à des questions posées par les professionnels et les riverains, sur l'effet des digestats sur la vie du sol et sur le stockage de la matière organique dans les sols. Ces questions peuvent aussi transparaître à travers des articles de presse, par exemple avec l'article de Mandard (2019).

Comme nous l'avons vu dans le Chapitre 1, nous manquons d'études pour répondre à ces questions, en particulier à cause du manque d'essais appropriés à long terme ou à large échelle. Le Chapitre 7 propose une première vision de la question sur l'impact des digestats sur les vers de terre. Enfin, les études plus intégrées comme le propose cette thèse pourraient servir d'outils de synthèse avec un rôle en communication ou pédagogique, par exemple en représentant des scénarios-types contrastés, pour montrer les effets de différentes pratiques.

8.2 Réflexions sur la démarche utilisée dans la thèse

La démarche utilisée dans cette thèse a consisté à obtenir et à analyser des données expérimentales, à utiliser ces données pour valider les modèles SYS-Metha et STICS dont le couplage a ensuite permis de simuler des scénarios à l'échelle de l'exploitation agricole. Si le choix de cette échelle d'étude a déjà été justifié dans la section 8.1, nous voulons discuter ici de l'utilisation conjointe de l'expérimentation et des modèles.

Les expérimentations (Chapitre 3, Chapitre 7) permettent d'obtenir des données pour répondre à une ou plusieurs questions de recherche spécifiques. Il s'agit d'approches nécessaires pour identifier les processus en jeu lors de l'épandage de digestats. Les expérimentations que nous avons mises en place ont pour limite d'être restreintes à un cas d'étude (une condition pédoclimatique, un digestat). Un avantage est que nous pouvons considérer les impacts d'un système que nous avons décrit sous plusieurs facettes, et que les impacts peuvent être considérés les uns en rapport avec les autres. À l'inverse, il est difficile de généraliser ces résultats en les appliquant à d'autres situations pédoclimatiques. Cet essai est non usuel par sa taille, et la non réplication des traitements. Les intérêts et limites de cette démarche ont été présentés au Chapitre 2 : cela rend possible une utilisation d'épandeurs réels pour l'épandage de digestat, et de considérer des grandes surfaces, mais l'absence de répétitions en blocs nécessite d'interpréter les données avec précaution. Ces limites s'appliquent aux Chapitres 3 et 7.

En agronomie, la multiplicité des conditions pédoclimatiques, la diversité des pratiques agricoles, la difficulté de mesure de certains indicateurs, ou encore la présence d'effets à long terme d'une pratique notamment sur la qualité des sols, rendent difficile l'évaluation des systèmes agricoles. Une première approche pour résoudre ce problème est l'utilisation de méta-analyses, qui permettent de synthétiser l'effet d'une pratique à partir de nombreuses études expérimentales, comme par exemple l'effet des PRO sur le stockage de C dans les sols ou la fertilisation azotée (Zavattaro et al., 2017), ou sur les émissions de N_2O (Zhou et al., 2017). Néanmoins, ces études demandent de grands jeux de données expérimentales. Comme évoqué par Jones et al. (2017), les modèles permettent également de généraliser les résultats expérimentaux. Ils permettent aussi d'explorer des systèmes difficilement instrumentables, et d'observer des interactions entre des effets qui seraient difficilement isolables. Ici, nous avons utilisé deux modèles pour prendre en compte les interactions entre d'une part la production, le post-traitement,

et le stockage des digestats, et d'autre part l'utilisation des digestats au champ (Del Prado et al., 2013). Cela permet de transposer les résultats du champ à l'échelle de l'exploitation. Les modèles mécanistes peuvent avoir différentes complexités, en fonction de leurs objectifs d'utilisation : simple pour l'aide à la décision, plus complexe pour répondre à des questions de recherche (Rotz, 2018). Ici, nous avons développé un modèle à l'échelle de la ferme plutôt complexe, capable de tenir compte de plusieurs interactions. Son rôle est de répondre à des questions de recherche et de proposer une analyse intégrée des effets agronomiques de la méthanisation agricole.

Une des originalités fortes du couplage de ces deux modèles est la routine d'allocation du digestat sur les différents champs en fonction de règles de décision préétablies (Chapitre 6). Les modules simulant les règles de décisions des acteurs peuvent être importants dans les modèles à l'échelle de la ferme, où ils peuvent permettre de considérer les contraintes des acteurs et donc simuler les pratiques de façon plus réalistes, allant de la gestion des effluents (Guerrin, 2001; Lopez-Ridaura et al., 2008), jusqu'à un ensemble conséquent des activités d'une ferme en polyculture élevage (choix des assolements, modalité de fertilisation et d'épandage des PRO, récolte,…) (Chardon et al., 2012; Vayssières et al., 2009). Ici, nous utilisons un module de règles de décision relativement simple. Ces règles de décision permettent le choix des doses et types d'engrais utilisés en fonction des besoins des cultures dans chaque parcelle, de la fourniture en azote du sol, en prenant en compte des contraintes simples de doses d'épandage réalistes et éventuellement de fréquences d'épandage de digestats plus grandes, attribuées à des contraintes de stockage du digestat (scenario Y). Ces règles de décision permettent de réellement simuler les doses d'engrais utilisées en fonction des cultures et de l'augmentation de la fourniture en azote du sol avec le temps. Le développement de ce module a nécessité des ressources de travail relativement importantes, entre autres sur :

- le recensement, le choix, et la simplification des pratiques agricoles à considérer,
- la recherche d'algorithmes efficaces pour maintenir un rendement constant dans le temps et entre les traitements,
- l'ajustement, la recherche des paramètres et la validation des sorties du modèle pour simuler une gestion cohérente de l'exploitation,
- l'implémentation informatique du couplage entre les différents modules.

Ce travail est justifié car il permet de mieux appréhender des questions de recherche liées aux différentes gestions agronomiques possibles du digestat (Chapitre 6).

La validation des modèles est nécessaire à leur utilisation pour en généraliser les résultats. Un modèle à l'échelle de la ferme est complexe, voire impossible à valider, car cela demande beaucoup d'informations, idéalement sur plusieurs exploitations. Comme suggéré par Del Prado et al. (2013), nous avons ici procédé à une validation par parties. Le Chapitre 4 évalue le modèle STICS à simuler une rotation utilisant des digestats comme fertilisants. Ce Chapitre illustre le fait que cette étape de validation

est nécessaire : la volatilisation de l'ammoniac après apport des digestats n'était pas simulée correctement, ce qui a conduit à l'introduction d'un autre sous modèle de volatilisation, ALFAM2 (Hafner et al., 2019). L'intérêt de valider le modèle STICS indépendamment est aussi d'enrichir l'expérience sur ce modèle qui peut être utilisé pour de nombreux autres objectifs, comme estimer l'effet du changement climatique sur la production agricole (Falconnier et al., 2020), estimer le potentiel de production agricole à l'échelle nationale, par exemple pour la prairie (Graux et al., 2020), ou estimer le stockage de C dans les sols et les pertes azotées par l'agriculture à l'échelle nationale (Launay et al., 2021).

Le choix du modèle STICS s'est fait de par son aptitude à modéliser de nombreuses cultures pour des conditions pédoclimatiques françaises (Coucheney et al., 2015; Yin et al., 2020). STICS possède un module pouvant mesurer le stockage de MOS, les émissions d'azote (volatilisation de NH_3, émission de N_2O, lixiviation du nitrate). De plus, la communauté STICS française développe activement le modèle. Récemment, STICS a montré sa capacité à simuler une rotation culturale avec apports de PRO à long terme (Levavasseur et al., 2021b), et des versions de recherche de STICS permettent de simuler des prairies (Graux et al., 2020) ou autres cultures pérennes (Strullu et al., 2020) par exemple. Ainsi, les incubations de digestats présentées aux Chapitres 3 et 4 ont aussi participé à la nouvelle paramétrisation des PRO par défaut dans STICS (Levavasseur et al., 2021a). La présence d'une communauté de développement est un argument important dans le choix du modèle STICS. Cette communauté permet le dialogue avec des scientifiques spécialistes de différents flux ou de différents types de cultures. Dans cette thèse, grâce à cela, nous avons par exemple pu avoir accès à une version de recherche de STICS afin de pouvoir appliquer le nouveau modèle de minéralisation des PRO, de changer le module de volatilisation d'ammoniac, et de simuler l'application de digestat sur prairie.

SYS-Metha utilise une base de données de déchets avec leurs caractéristiques, pour prédire la qualité des digestats par bilan de masse, en considérant la minéralisation d'une partie de la matière organique. Les transformations des digestats au stockage sont estimées avec des facteurs de conversion et d'émissions. SYS-Metha a été choisi pour sa capacité à prédire simplement les caractéristiques des digestats à partir de déchets variés. À notre connaissance, cette capacité est unique dans la littérature (Chapitre 5). Il ne paraissait pas nécessaire de modéliser le processus de digestion finement comme le fait un modèle de digestion anaérobie mécaniste tel qu'ADM1 (Batstone et al., 2002) qui est l'un des plus utilisés (Lauwers et al., 2013). La variabilité temporelle de biogaz n'était pas une variable recherchée. De plus, ADM1 demande une caractérisation fine des intrants ou l'ajustement de paramètres par calibration mathématique (Donoso-Bravo et al., 2020; Li et al., 2021). La faible caractérisation des nombreux intrants du méthaniseur de Nouzilly aurait été une limite à l'utilisation de ce type de modèles, et la généralisation sans nouvelle paramétrisation à d'autres types d'intrants aurait été limitée. Cependant, un avantage possible aurait été une éventuelle modélisation de la stabilisation de la matière organique dans le digesteur, mais le lien entre la stabilité de la matière organique dans le digesteur et

celle au champ est encore une question en développement. Dans le contexte de cette thèse, une des limites de SYS-Metha est l'utilisation d'hypothèses implicites liées au choix et au mode d'application des facteurs d'émissions, et qui doivent pouvoir être remises en cause si nécessaire (Chapitre 5). En effet, ces facteurs d'émissions semblent fortement conditionner les impacts liés au stockage des PRO (Chapitre 6). Une des manières de valider ces facteurs d'émissions aurait été de mesurer des émissions gazeuses spécifiques à notre cas d'étude ce qui avait été envisagé lors du montage du projet de thèse. Ces mesures n'auraient pas été réalisables parce qu'elles auraient demandé un effort et des ressources trop grandes par rapport aux objectifs de modélisation de la thèse.

Lors de la phase de construction des scénarios, nous avons échangé avec les Chambres d'Agriculture du Cher, et de l'Indre, et du Loiret. Ceci a permis de mieux comprendre les pratiques actuelles en terme de méthanisation agricole dans cette région, et de proposer des scénarios de modélisation réalistes pouvant répondre à certaines questions qui n'auraient pas été identifiées auparavant. Par exemple, en région Centre – Val de Loire, comme ailleurs, l'épandage des digestats est fortement contraint par des questions logistiques, ce qui a mené à considérer cette question dans le Chapitre 6 (scenario Y). Ce contact avec les professionnels a également un autre avantage. Le travail de modélisation est un travail de synthèse et de simplification. Le modèle est donc une représentation simplifiée de la réalité, selon un point de vue particulier. Comme évoqué par Legay (1997), un des risques de la modélisation est de déformer la réalité dans l'esprit du modélisateur. Le dialogue avec des professionnels de l'agriculture et de la méthanisation permet de garder une vision moins simpliste que celle des modèles sur nos objets d'études, et de garder à l'esprit les questions actuelles que se posent la filière.

8.3 Introduction d'un méthaniseur dans une exploitation agricole, quelles conséquences ?

L'objet d'étude de cette thèse est la méthanisation agricole, utilisant comme intrants principaux des effluents animaux et des déchets organiques. Cette pratique est toujours en fort développement en Europe et en France (European Biogas Association, 2020). Les conséquences de l'introduction d'un méthaniseur peuvent être réfléchies de deux points de vue. On peut d'abord s'intéresser au point de vue « interne », en étudiant les conséquences de l'implantation d'un méthaniseur pour les agriculteurs de l'exploitation ou des exploitations concernées par la méthanisation (par exemple, « la méthanisation diminue-t-elle l'utilisation d'engrais de synthèse sur l'exploitation ? »). On peut aussi se demander quelles sont les conséquences selon un point de vue « global », c'est-à-dire l'ensemble des conséquences directes ou indirectes en termes d'impacts sur l'environnement (par exemple, « la méthanisation permet-elle la réduction totale de l'utilisation d'engrais de synthèse sur le territoire ? »). Ces deux points de vue posent des questions différentes et n'amènent pas exactement les mêmes réponses sur les bénéfices et risques associées à la méthanisation.

Selon le point de vue « intra exploitation », l'introduction d'un méthaniseur permet une diversification des activités et l'intégration de digestats dans les pratiques de fertilisation des systèmes de culture. Elle a aussi de nombreux impacts économiques, sociaux, et de charge de travail (ADEME and SOLAGRO, 2018). La quantité de déchets importés semble être le déterminant principal de l'amplitude des effets de la méthanisation. L'import de nutriments permet d'augmenter significativement la substitution des fertilisants de synthèse au champ, parfois au risque d'émissions d'ammoniac plus importantes. Ces importations de matières organiques permettent de compenser les pertes de carbone via le biogaz et de mener à un bilan C neutre voir positif pour les sols du plan d'épandage. La mise en place de pratiques telles que la couverture de fosse ou le choix agronomique optimisé des parcelles épandues peut aider à réduire les effets négatifs et augmenter les effets positifs de la méthanisation. L'augmentation des flux de nutriments dans une ferme ne doit pas s'accompagner d'effets négatifs, avec par exemple une augmentation potentielle de la lixiviation des nitrates. Des contraintes peuvent aussi apparaître relativement aux apports de P et K, qui ne doivent pas être excédentaires. La question de la taille des méthaniseurs paraît donc être une question importante à considérer au regard des sols du plan d'épandage et des ressources en PRO et en nutriments déjà présentes. L'augmentation de la quantité de PRO à gérer peut impliquer des contraintes de stockage des intrants ou des digestats, des contraintes logistiques sur la gestion des épandages en veillant à ne pas apporter des doses excédentaires par rapport aux besoins des cultures ou de faire les épandages à des dates où les risques de tassement des sols sont importants. Le changement de qualité des digestats en lien avec les types d'intrants peut également influencer leurs teneurs en contaminants (Lehmann and Bloem, 2021; Sertillanges et al., 2020) ou la biodiversité du sol. Ces questions sont en cours d'investigation dans la communauté scientifique.

Selon le point de vue « global », la méthanisation a trois objectifs principaux : proposer une filière de traitement des déchets, la production d'énergie renouvelable, et le recyclage des nutriments au sol (Ministère de l'agriculture, de l'agroalimentaire et de la forêt, 2013). La méthanisation peut aussi être un outil de développement de projets territoriaux, qui peuvent promouvoir la valorisation locale des déchets et l'autonomie des territoires en terme de nutriments (Moinard et al., 2021). Nous n'avons que très rapidement abordé l'échelle territoriale à travers le bilan carbone présenté au Chapitre 6. La nature des déchets valorisés semble déterminante pour savoir quels sont les déchets valorisés à cette échelle. Pour des déchets comme les boues ou les effluents d'élevage, parce qu'ils sont le plus souvent déjà valorisés dans des plans d'épandage (Houot et al., 2014), la méthanisation tendraient à n'augmenter le recyclage des nutriments que faiblement, via l'augmentation de la richesse en ammoniac dans les digestats. Leur méthanisation produit de l'énergie, peut diminuer les effets négatifs liés à leur traitement, mais peut aussi augmenter les émissions d'ammoniac au stockage et lors de l'épandage et diminuer légèrement le stockage de C dans le sol. À ce propos, l'étude de Thomsen et al. (2013), largement citée dans la littérature, émet l'hypothèse que les PRO stockent la même quantité de matière organique qu'ils soient méthanisés ou non : la minéralisation d'une partie de la matière organique par la digestion

anaérobie serait compensée par l'augmentation de la stabilité de la matière organique résiduelle. Nous n'obtenons pas ces résultats dans nos travaux de modélisation, basés sur des paramètres dérivés d'études en laboratoire. Cette théorie est loin d'être confirmée et l'effet de l'épandage des digestats sur la MOS comparé à celui d'autres PRO reste une question importante. D'autre part, certains déchets organiques (municipaux ou agro-industriels par exemple), qui ne sont actuellement pas recyclés en agriculture, peuvent présenter un intérêt à être méthanisés. Leur méthanisation peut permettre le recyclage de leur nutriments et l'apport de matière organique supplémentaire au champ (Launay et al., 2021). L'effet positif ou négatif de leur méthanisation dépend également fortement du type de déchets et de leur traitement alternatif en vigueur actuellement (ex. enfouissement, incinération …), comme le montre les analyses de cycles de vie (ACV) (Vaneeckhaute et al., 2018; Vasco-Correa et al., 2018).

Les ACV sont des outils utiles, qui permettent d'agréger les impacts globaux d'une pratique, selon plusieurs indicateurs. Les indicateurs revenant fréquemment pour parler de la méthanisation sont l'impact sur le changement climatique, mais aussi l'acidification et l'eutrophisation des milieux, et l'utilisation de ressources non renouvelables (Hijazi et al., 2016). De nombreuses ACV sur la méthanisation ont été réalisées, dans des contextes et prenant en compte des périmètres très différents. Globalement, elles montrent que les principaux bénéfices de la méthanisation sont la diminution des émissions de gaz à effet de serre et de la demande en ressources non renouvelables. Les risques incluent une possible augmentation de l'acidification et de l'eutrophisation des milieux (Hijazi et al., 2016; Vasco-Correa et al., 2018). Dans le contexte français, la valorisation du biogaz en cogénération sans valorisation de la chaleur pourrait ne pas mener à des bénéfices d'un point de vue des émissions de gaz à effet de serre, en raison du faible impact carbone de la production d'électricité française. Ceci reste à étudier plus en détail avec des ACV plus spécifiques à la méthanisation française.

8.4 Perspectives : quelle recherche pour mieux appréhender les enjeux de la méthanisation ?

En premier lieu, le modèle à l'échelle de l'exploitation que nous avons développé au cours de cette thèse pourrait évidement être complété ou raffiné, si les questions de recherche le justifient. En termes de compréhension scientifique, une étude plus poussée sur la sensibilité des résultats à différents paramètres incluant les facteurs d'émissions des PRO au stockage serait nécessaire. L'étude de la propagation des incertitudes au sein du modèle pourrait être intéressante, mais une réflexion devrait au préalable être menée pour savoir si son coût (ressources temporelle et humaine) est justifié par rapport à ses apports scientifiques. Nous n'avons pas eu le temps d'aborder ces questions dans cette thèse. L'implémentation de modèles plus mécanistes pour simuler les émissions au stockage des digestats pourrait permettre d'étudier l'effet de différentes gestions du digestat. Néanmoins, il ne faut pas que cette complexité conduise à une dégradation de la prédiction des caractéristiques des digestats qui sont essentielles pour

le couplage avec l'utilisation au champ. Enfin, l'ajout de nouvelles contraintes dans le module logistique organisant l'utilisation des digestats dans un plan d'épandage (temps de travail, import de P ou de K par exemple), serait aussi une implémentation utile pour garantir une gestion plus optimale des digestats.

Ce type d'approche semble également pertinent pour étudier les questions de la méthanisation à l'échelle de l'exploitation, en faisant varier des paramètres comme la surface du plan d'épandage, l'assolement, les contraintes logistiques, les types et proportions de déchets, la méthanisation des lisiers mais pas des fumiers. Par exemple, il serait possibe d'étudier la méthanisation basée sur la digestion de cultures dédiées (culture principales, cultures intermédiaires à vocation énergétique CIVE). En effet, la méthanisation a alors un effet sur l'assolement de l'exploitation, la modélisation explicite des champs pourrait permettre de prendre en compte ces effets.

Le modèle développé ici n'a pas vocation à être utilisé comme outil d'aide à la décision. Il est trop complexe, et ne répond pas à une demande opérationnelle précise de la filière (Walling et Vaneeckhaute, 2020a). De plus, il n'est pas non plus destiné à être mis à disposition librement. Ceci impliquerait un travail de développement informatique, de construction d'interfaces, de mise en place de garde-fous relatifs aux hypothèses faites, qui n'est pas prévu et n'est pas justifié en termes d'applications. Les modèles mis à disposition d'un large public sont souvent soutenus par une communauté qui réfléchit aux enjeux de transfert, qui corrige, simplifie et complète les modèles, et qui peuvent conseiller les utilisateurs.

Néanmoins, les perspectives de ces travaux sont loin de ne concerner que la modélisation. La construction d'un modèle amène à réfléchir à formuler certaines hypothèses, qui peuvent affecter sensiblement les conclusions. La construction et l'utilisation d'un modèle mènent donc à des réflexions sur les connaissances actuelles et appellent à mener de nouvelles expériences. Cet aller-retour entre l'expérimentation et la modélisation fait partie du processus de recherche scientifique (Legay, 1997). Ainsi, sur une thématique proche de celle de cette thèse, plusieurs études ont amené les dernières années à se demander si le bénéfice du stockage de carbone des sols ne serait pas compensé par l'augmentation des émissions de N_2O du sol. Cette problématique a, entre autres, été formulée par Lugato et al., (2018), qui montrent de possibles risques dans un travail de modélisation. Ceci a amené la communauté scientifique à étayer ou réfuter cette hypothèse via la synthèse de données expérimentales (Guenet et al., 2021). Des travaux sont aussi en cours sur le sujet, par exemple sur les réseaux d'essais à long-terme SOERE ACBB et SOERE PRO.

Dans cette thèse, les réflexions sur les hypothèses du modèle, ajoutées aux résultats expérimentaux, permettent de mettre en évidence un besoin de davantage d'études sur les digestats. Nous manquons d'information sur les émissions de gaz à effet de serre produites par les digestats lors de leur stockage, et en particulier en ce qui concerne les digestats solides. Parmi les questions auxquelles ces études pourraient répondre: les émissions gazeuses lors du stockage ou du compostage de digestat solide sont-

elles comparables à celles des fumiers ou des composts ? Dans quels cas la séparation de phase des digestats augmente-t-elle ou diminue-t-elle les émissions de gaz à effet de serre ? Une autre question en cours d'investigation est la prédiction de la stabilité de la matière organique des digestats au champ (par exemple, la prédiction de l'ISMO) en fonction des intrants et procédés de méthanisation. Cette question pourrait apporter des réponses plus fines sur l'effet des digestats sur la MOS que celle utilisées actuellement, comme suggéré par Jimenez et al. (2015). La volatilisation d'ammoniac lors de l'épandage de digestat solide n'est que peu documentée. Cela parait être une question moins importante, car les techniques limitant les émissions au champ sont bien connues (enfouissement rapide), et les digestats solides sont en quantités faibles relativement aux digestats liquides à l'échelle d'un méthaniseur. Néanmoins, explorer cette question pourrait être plus importante pour des méthaniseurs utilisant la digestion anaérobie par voie sèche. Une meilleure caractérisation de l'infiltration des digestats dans le sol pourrait aussi affiner notre compréhension de l'augmentation ou de la diminution des émissions d'ammoniac relativement à celles des lisiers (Pedersen et al., 2021). L'effet des digestats sur la vie du sol requiert plus d'investigation (Möller, 2015). Une perspective dans la continuité logique de la thèse serait de réaliser une ACV de la méthanisation dans notre contexte régional. En effet, nous ne considérons l'impact global de la méthanisation que sur un unique critère, le potentiel de réchauffement climatique. Les modèles peuvent permettre de documenter des ACV qui considèrent alors les principales contraintes des acteurs et du territoire (Paillat et al., 2009), qui explorent différentes pratiques, ou qui montrent une variabilité dans les indicateurs. Cette perspective serait intéressante, car les ACV de la méthanisation, à une échelle fine ou générale, sont encore peu présentes dans le contexte français. Enfin, dans le Chapitre 7, nous avons mis en lumière plusieurs de ces questions relativement aux vers de terre, qui peuvent être élargies. Les digestats ont-ils la même qualité nutritive pour les êtres vivants que les autres PRO ? Dans quelles conditions les digestats sont-ils bénéfiques on néfastes pour la vie du sol ? L'effet positif du digestat est-il dû aux apports de digestats liquides et/ou solides ? Si l'effet des apports de digestat est plutôt positif à moyen terme, peut-on préciser les conditions dans lesquelles on observe une mortalité partielle des vers ? Quel est l'effet de l'apport répété de digestats sur les autres organismes du sol au sens large ?

La méthanisation est un procédé qui propose des solutions intéressantes en termes de traitement des déchets, d'économie circulaire, et de production de gaz renouvelable. Entre autres, la méthanisation conserve les nutriments, ce qui optimise leur recyclage. Le biogaz est une énergie renouvelable avec des avantages comme sa capacité à être stocké, et son utilisation possible dans le secteur du transport. La bonne connaissance des risques environnementaux liés au développement de la méthanisation dans les territoires est nécessaire pour réduire ses risques et maximiser ses impacts positifs, dans le but d'optimiser la filière.

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ÉCOLE DOCTORALE
Agriculture, alimentation, biologie, environnement, santé (ABIES)

Titre : Conséquences de l'introduction de la méthanisation dans une exploitation de polyculture-élevage sur les cycles du carbone et de l'azote. Combinaison de l'expérimentation et de la modélisation à l'échelle de la ferme.

Mots clés : Méthanisation ; Polyculture élevage ; Cycles du carbone et de l'azote ; Bilan environnemental ; Modélisation ; Vers de terre

Résumé : La méthanisation agricole des effluents animaux est une pratique en fort développement en France. Elle produit de l'énergie renouvelable (biogaz). La valorisation des digestats au champ, comme celle des effluents non méthanisés, permet le retour au sol de nutriments et de matière organique, ce qui diminue le besoin en engrais minéraux et entretient les stocks de C des sols. Le traitement et l'épandage de ces produits peut aussi induire l'émission de gaz à effet de serre et de contaminants. La méthanisation agricole influence ces impacts : pour les maitriser, il faut comprendre comment la digestion des effluents avec des déchets importés modifie les cycles du C et du N à l'échelle de la ferme. Cette question a été traitée en s'appuyant sur un cas d'étude à l'INRAE de Nouzilly (Centre – Val de Loire) : une exploitation agricole avec un méthaniseur traitant les effluents de son élevage bovin et divers déchets organiques. Lors de l'essai au champ MétaMétha, nous avons comparé les flux d'azote au cours d'une rotation culturale fertilisée avec des engrais minéraux, des lisiers et fumiers bovins, ou des digestats issus de ces effluents. Les digestats se substituent bien aux engrais minéraux, mais ils sont sensibles à la volatilisation d'ammoniac (NH_3). Les vers de terre peuvent être négativement impactés juste après l'épandage de digestat ou de lisier, mais les effets sont similairement positifs après 2 ans d'apports de matière organique. Nous avons ensuite évalué les modèles STICS et SYS-Metha pour simuler respectivement l'essai au champ et le traitement des digestats. Ces modèles ont été couplés pour simuler les flux de C et N à l'échelle de la ferme. Avec de forts imports de déchets, la méthanisation favorise la substitution des engrais minéraux, le stockage de C dans les sols, mais aussi les émissions de NH_3. Ce travail permet de mieux évaluer les conséquences de l'introduction d'un méthaniseur dans une exploitation agricole et ainsi d'optimiser la filière.

Title : Consequences of the anaerobic co-digestion of cattle effluents in a crop livestock farm on carbon and nitrogen cycles. Combination of experimentation and of modelling at the farm scale.

Keywords : Anaerobic digestion ; Crop-livestock farm; Carbon and nitrogen cycles; Environmental assessment; Modelling; Earthworms

Abstract : The anaerobic digestion (AD) of animal effluents is strongly developing in France. It produces renewable energy (biogas). Like undigested effluents, the use of anaerobic digestates in the field enables the recycling of nutrients and organic matter in the soil, which decreases the need for synthetic fertilizers and maintains soil organic carbon stocks. However, the treatment and field application of those organic products can also cause greenhouse gas emissions and contaminants. The on-farm AD influences those impacts. In order to control them, we need to understand how the co-digestion of animal effluents with imported organic wastes influences C and N cycles at the farm scale. We studied this question with the support of a case study at INRAE in Nouzilly (Centre – Val de Loire, France): a crop and livestock farm where an anaerobic digester treats the cattle effluents together with imported wastes. During the MetaMetha field experiment, we compared N fluxes during a crop rotation that was fertilized with synthetic N fertilizers, undigested cattle slurry and farmyard manure, or digestates. We found that digestates can substitute synthetic fertilizers, despite the risk of ammonia (NH_3) emissions. Earthworms can be negatively impacted just after the application of digestate of slurry, but the input of organic matter induced similar positive impacts after two years. We then evaluated the STICS and SYS-Metha models to simulate the field experiment, and digestate treatment and storage, respectively. Both models were coupled to simulate C and N fluxes at the farm scale. When large amounts of digested wastes are imported, AD promotes substitution of synthetic N fertilizers and storage of soil organic C, but also NH3 emissions. The study enabled us to better evaluate the consequences of the on-farm AD and therefore to optimize the sector.

Maison du doctorat de l'Université Paris-Saclay
2ème étage aile ouest, Ecole normale supérieure Paris-Saclay
4 avenue des Sciences,
91190 Gif sur Yvette, France

Conséquences de l'introduction de la méthanisation dans une exploitation de polyculture-élevage sur les cycles du carbone et de l'azote. Combinaison de l'expérimentation et de la modélisation à l'échelle de la ferme.

Annexes

Thèse de doctorat de l'université Paris-Saclay

École doctorale n° 581 : agriculture, alimentation, biologie, environnement et santé (ABIES)
Spécialité de doctorat: Sciences de l'environnement
Unité de recherche : Université Paris-Saclay, INRAE, AgroParisTech, UMR ECOSYS, 78850, Thiverval-Grignon, France
Référent : AgroParisTech

Thèse présentée et soutenue à Paris-Saclay,
le 18/11/2021, par

Victor MOINARD

Composition du Jury

Yves COQUET	Professeur, AgroParisTech (Université Paris-Saclay)	Président
David FANGUEIRO	Assistant Professor (eq. HDR), Institut d'Agronomie de l'Université de Lisbonne	Rapporteur & Examinateur
Safya MENASSERI-AUBRY	Maîtresse de Conférences (HDR), Institut Agro-AgroCampus Ouest	Rapporteur & Examinatrice
Anne TREMIER	Directrice de Recherche, INRAE Bretagne Normandie	Examinatrice
Sabine HOUOT	Directrice de Recherche, INRAE IdF-Versailles-Grignon	Directrice de thèse
Jean-Marie PAILLAT	Chercheur (HDR), CIRAD – ISTOM Angers	Co-Directeur & Examinateur
Florent LEVAVASSEUR	Ingénieur de recherche, INRAE IdF-Versailles-Grignon	Invité

Annexes – Chapitre 3
Supplementary Material

S. M. 1 Map of the experimental design

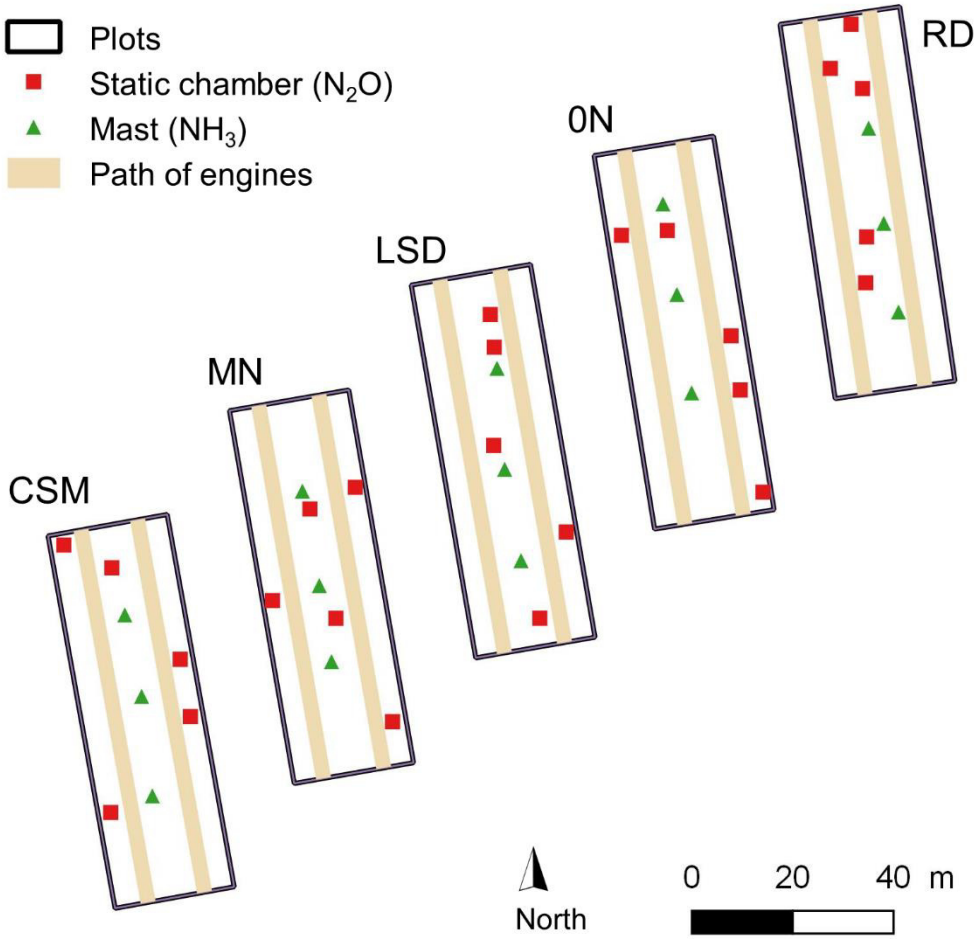


Figure S3.1 – Map of the experiment design

S. M. 2 Crop management

Table S3.1 – Technical management of the crops.

Crop	Date	Action
2016 – 2017 Wheat	26 October 2016 and 27 October 2016	Soil tillage: disc harrow (15 cm),
	27 October 2016	Covercrop (10 cm), followed by wheat sowing (cultivar Syllon, 260 gr m^{-2})
	22 November 2016	Herbicide
	22 March 2017	Fertilization
	19 April 2017	Fertilization
	04 May 2017	Herbicide
	15 May 2017	Fongicide
	20 July 2017	Grain harvest
	23 July 2017	Straw harvest
2017 – 2018 Rapeseed	02 August 2017	Application of EOMs
	03 August2017	Burying and soil tillage : ploughing (20 cm), covercrop (10 cm), roller (2 cm)
	21 August 2017	Rapeseed sowing (cultivar Fernando, 40 gr m^{-2})
	21 August 2017	Herbicide
	25 October 2017	Insecticide
	21 March 2018	Fertilization
	23 March 2018	Fongicide and insecticide
	04 July 2018	Harvest
2018 – 2019 Wheat	19 September 2018	Application of EOMs
	20/09/2018	Burying : covercrop (10 cm)
	24 October 2018	Soil tillage : covercrop (10 cm), followed by wheat sowing (cultivar Descartes, 300 gr m^{-2})
	26 October 2018	Herbicide
	19 February 2019	Fertilization
	12 March 2019	Fertilization
	26 April 2019	Herbicide
	01 May 2019	Fertilization (MN and CSM treatments only, ammonium nitrate)
	03 May 2019	Fongicide
	07 May 2019	Fongicide
	22 July 2019	Grain harvest
	23 July 2019	Straw harvest

S. M. 3 Detailed methods: Agricultural performance of crops

Yield uncertainty

The balance that measured grain and straw yield had a precision of ±10 kg. Therefore, the uncertainty of the mass of grain was $\pm\frac{10}{\sqrt{3}} = 5.8$ kg for 1800 m^2 (rectangular probability distribution, type B evaluation of uncertainty. It was then scaled in t ha^{-1}.

Computing of N uptake

In each subplot, we computed two intermediary variables. The N concentration in the whole aboveground crop ($[N]_{aerial}$, gN $kgDM^{-1}$), including grain and straw, was computed with the equation:

(equation S3.1) $[N]_{aerial} = ([N]_{grain} \cdot m_{grain} + [N]_{straw} \cdot m_{straw}) / (m_{grain} + m_{straw})$

where $[N]_{grain}$ is the N content in grain (gN $kgDM^{-1}$), m_{grain} is the mass of grain in the subplot (kgDM), $[N]_{straw}$ is the N content in straw (gN $kgDM^{-1}$), m_{straw} is the mass of straw in the subplot (kgDM).

The mass ratio of grain to aboveground crop organs ($ratio_{grain:aerial}$) was computed with the equation:

(equation S3.2) $ratio_{grain:aerial} = m_{grain} / (m_{grain} + m_{straw})$

At last, we computed the N uptake by the crop in the whole plot. N export in grain at harvest (N_{grain}, kgN ha^{-1}) were computed as:

(equation S3.3) $N_{grain} = yield \cdot [N]_{grain}$

where yield is grain yield (t DM ha^{-1}). Total aboveground N uptake (including straw and grain) (N_{aerial}, kgN ha^{-1}) was computed as:

(equation S3.4) $N_{aerial} = yield \cdot [N]_{aerial} / ratio_{grain:aerial}$

Uncertainty on N_{grain} and N_{uptake}, $N_{strawexp}$, and $N_{returned}$ were computed using error propagation formulas from yield measurement uncertainty and standard error of $[N]_{grain}$, $[N]_{aerial}$, $[N]_{straw}$, and $ratio_{grain:aerial}$.

N exported in straw $N_{strawexp}$ (kgN ha^{-1}) was computed as:

(equation S3.5) $N_{strawexp} = straw\ yield \cdot [N]_{straw}$

Where the straw yield is expressed in t DM ha^{-1} and $[N]_{straw}$ is the concentration of N in straws (gN $kgMS^{-1}$). N returned to soil through aerial crop residues ($N_{returned}$, kgN ha^{-1}) were then computed as:

(equation S3.6) $N_{returned} = N_{aerial} - N_{strawexp} - N_{grain}$

Uncertainty on N_{grain} and N_{uptake}, $N_{strawexp}$, and $N_{returned}$ were computed using error propagation formulas from yield measurement uncertainty and standard error of $[N]_{grain}$, $[N]_{aerial}$, $[N]_{straw}$, and $ratio_{grain:aerial}$.

S. M. 4 Detailed method: SMN analysis

SMN (i.e. NO_3^- and NH_4^+ contents in soil) were analyzed by colorimetry after extraction with a KCl solution. 25 g of soil were mixed with 100 mL of KCl solution (1 mol L^{-1}), then agitated during 1h. Supernatant is filtrated on glass-fiber filter (pores: 1.2 µm) and frozen at -20°C before dosage. Pure KCl solution were also filtrated and stored on each date to evaluate any exogeneous N contamination (blank). N-NH_4^+ and N-NO_3^- contents were dosed by colorimetry on a continuous flow analyser (Skalar, The Netherlands). N-NH_4^+ and N-NO_3^- contents were corrected by subtracting mean N-NH_4^+ and N-NO_3^- contents in blank sample.

S. M. 5 Detailed methodology: Incubations

The soil used in the experiments was sampled once in summer (incubations of manure, raw digestate, solid digestate) and once in winter (incubations of liquid digestate and slurry), in the field experiment on the 0N control treatment. The soil was sieved fresh to a size of 4 mm. The field capacity of the sieved soil was measured with a Richards press (30,990 Pa) and was 25% w/w. Soil was stored in a closed container at 5°C before being used.

Raw digestate, solid digestate, and cattle manure were sampled on 19th of September 2018 during amendment. Cattle slurry and liquid digestate were sampled on 12th of March 2019 during fertilization. Solid digestate and cattle manure were chopped by hand for homogenization. The EOMs were stored at 4°C in plastic containers at most two months before the incubations.

The EOMs were incubated fresh, in non-limiting conditions for moisture, SMN, and temperature. They were mixed homogeneously into 500 g of soil in the proportion of 2000 mg C_{org} kg DS $^{-1}$, i.e. 16.6, 20.5, 77.1, 106.1, and 145.1 g FW kg DS^{-1} (mass of Fresh Weight per mass of dried soil) for cattle manure, solid digestate, raw digestate, liquid digestate, and cattle slurry, respectively. After soil mixing, KNO_3 solution or deionized water was added to the mixture to reach a minimum SMN content of 35 mgN_{min} $kgDS^{-1}$, and a soil moisture of 25% w/w, corresponding to soil moisture under a 30,990 Pa pressure (pF=2.5). The addition of the liquid digestate and slurry to the soil provided large amounts of water, which had been considered in the incubation experiments to achieve a common soil moisture level in all treatments. Because of the large amount of water linked to organic matter in the EOMs, the DM content of the EOMs is not relevant for this adjustment of soil moisture. Thus, we used a “moistening power” of an EOM as the quantity of water significantly humidifying soils in microcosm experiments. It was determined by weighing free water after the centrifugation of the EOMs for 10 min at 614 x g (3 replicates). This method was previously described on the same soil by Moinard et al. (2021). Soil moisture was maintained constant during the incubation, with regular addition of deionized water. Mixtures were incubated in darkness at 28 ± 1 °C for 175 days. Two sets of incubations were followed, to measure both mineralization of C; and mineralization or immobilization of N.

The mineralization of organic Carbon were measured by following soil CO_2 emissions. Four replicates per EOM were used, as well as an unamended soil control treatment and blank treatment without any material. The mixture of each replicate was placed in a 3 L airtight vessel, along with a plastic vessel containing 20 mL of NaOH (1 mol L^{-1}) solution to trap CO_2 emissions. At time 1, 3, 7, 10, 14, 17, 21, 24, 36, 49, 70, 91, 112, 133, 156, 175 days, plastic bottles with NaOH solutions with trapped CO_2 were removed and closed. It was stored in a closed dessicator together with open 6 mol L^{-1} NaOH solution, to trap for eventual atmospheric CO_2, before dosage. Fresh NaOH was added into the replicates, enabling to measure CO_2 emissions by mixture for all date intervals. CO_2 was measured by colorimetry on a continuous flow analyser (Skalar, The Netherlands). CO_2 concentrations were corrected by subtracting

mean CO_2 content in the blank treatment, to evaluate any exogenous CO_2 contamination. For each date interval, net mineralization from EOM was computed for each replicate by subtracting total C-CO_2 emissions by the mean of C-CO_2 emissions in control treatment (soil respiration). Net C mineralization was summed on each replicate at each date interval to compute cumulated C mineralization, which was then expressed in % C_{org} initially present in the EOM.

The following of N mineralization or immobilization used destructive replicates. Four replicates per treatment and per date of measurement were used. Mixture was added in an open 1 L plastic vessel, covered by a perforated plastic film, in order to allow oxygen and air circulation while limiting soil water evaporation. At dates 0, 3, 7, 14, 25, 49, 91, 175 days, SMN was extracted with KCl solution. 120 g of mixture were mixed with 400 mL of KCl solution (1 mol L^{-1}), then agitated during 1h. Supernatant is filtrated on glass-fiber filter (pores: 1.2 µm) and frozen at -20°C before dosage. Pure KCl solution was also filtrated and stored on each date to evaluate any exogeneous N contamination (blank). N-NH_4^+ and N-NO_3^- contents were dosed by colorimetry on a continuous flow analyser (Skalar, The Netherlands). N-NH_4^+ and N-NO_3^- contents were corrected by subtracting mean N-NH_4^+ and N-NO_3^- contents in blank sample. At each date, to avoid accounting for initial mineral N, mineralized N-NH_4^+ and N-NO_3^- contents were computed by subtracting total N-NH_4^+ and N-NO_3^- contents by their contents at t=0 day in each treatment. Net N mineralization or immobilization was computed by subtracting mineralized N-NH_4^+ and N-NO_3^- contents in soil by the one in unamended control (soil mineralization). Net N mineralization or immobilization was then expressed in % N_{org} initially present in the EOM. We also computed the available mineral N brought by the EOM, expressed in kg N_{min} in soils per kg FW of EOM brought, taking into account both mineralization/immobilization cited above and initial TAN content in the EOM.

Moinard, V., Redondi, C., Etiévant, V., Savoie, A., Duchene, D., Pelosi, C., Houot, S., Capowiez, Y., 2021. Short- and long-term impacts of anaerobic digestate spreading on earthworms in cropped soils. Applied Soil Ecology 168, 104149. https://doi.org/10.1016/j.apsoil.2021.104149

S. M. 6 Individual analyses of cattle slurry

Table S3.2 – Individual analyses of cattle slurries.

EOM	Application date	TS (%FW)	TAN (g $kgFW^{-1}$)	TN (g $kgFW^{-1}$)	VS (g $kgFW^{-1}$)	pH
Cattle slurry	22/03/2017	1.6	0.6	1.1	11.3	7.7
	19/04/2017	0.8	0.8	1.0	4.8	7.5
	21/03/2018	7.5	1.3	3.3	56.1	6.6
	19/02/2019	4	2.0	2.7	27.6	7.5
	12/03/2019	3.2	1.1	2.2	22.1	7.9

TS: total solid. FW: Fresh weight. TAN: ammoniacal N. TN: total N.

S. M. 7 Rapeseed growth from sowing to February

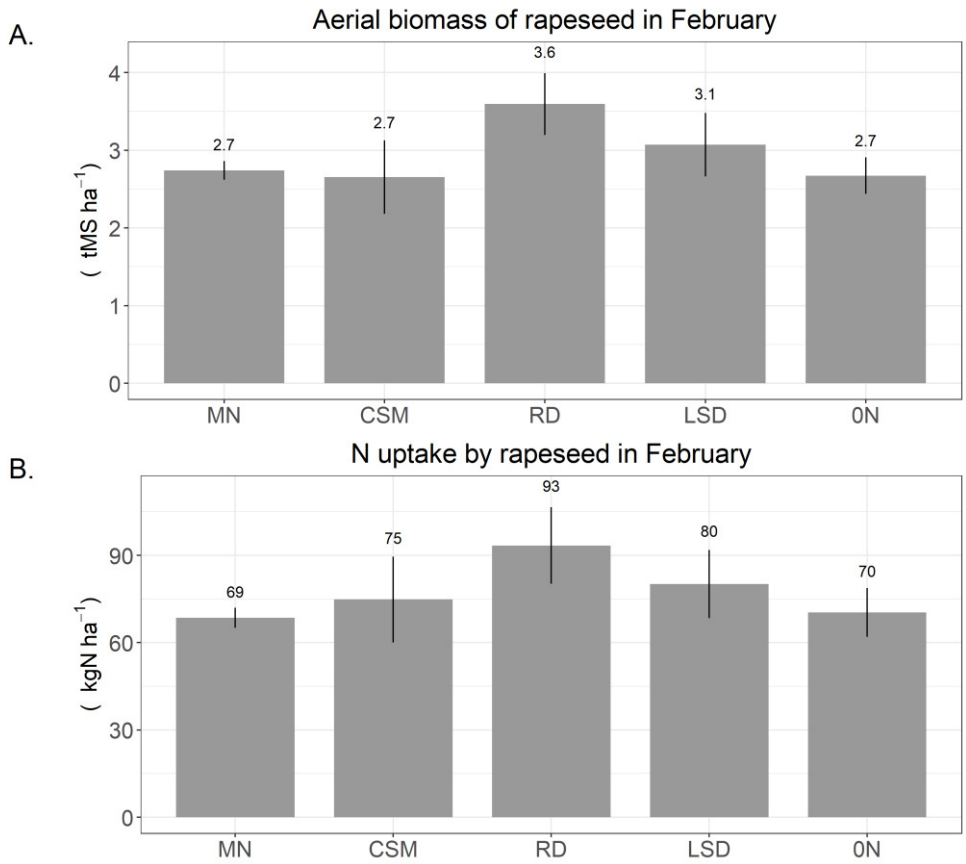


Figure S3.2: A) Aerial biomass and B) N uptake from rapeseed at the end of winter (February). Error bars show one standard error.

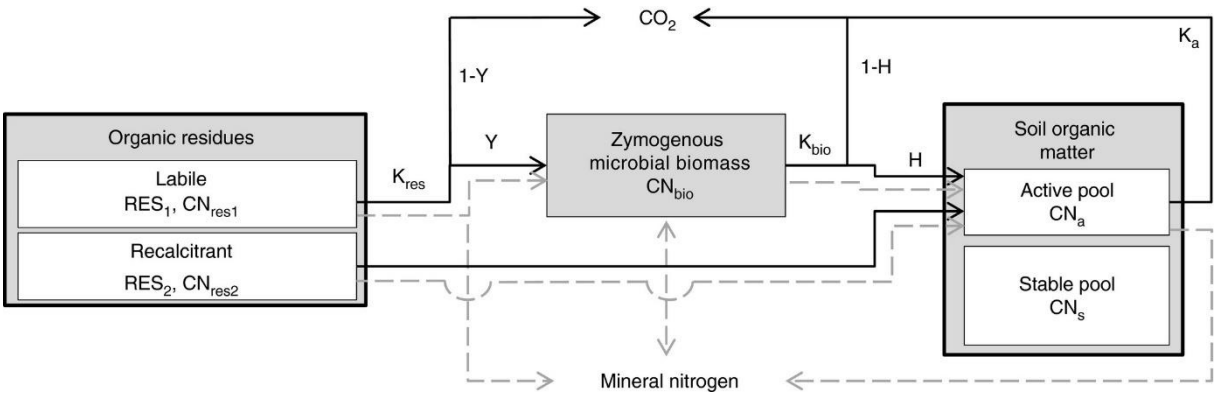
Annexes – Chapitre 4
Supplementary Material

S. M. 1 Parameters and options used in STICS

Supplementary Table S1 - The soil is described by 4 horizons. The first two horizons described a single horizon in the field, which was split to compare modelled soil characteristics to the one observed in the 0 – 20 cm depth. SWC: soil water content.

Soil depth	Bulk density ($g\ cm^{-3}$)	SWC at field capacity (% w/w)	SWC at wilting point (% w/w)
0 – 20 cm	1.37	26	8
20 – 25 cm	1.55	26	8
25 – 40 cm	1.55	23	10
40 – 60 cm	1.60	22	9
40 – 100 cm	1.60	21	15

S. M. 2 Calibration of EOM mineralization submodule of STICS



Supplementary Figure S2 – EOM mineralization submodule of STICS. Figure and captions were taken from Levavasseur et al., 2021a. "Decomposition model of EOM in the modified version of the STICS model. The C fluxes are represented as solid black lines, and the N fluxes are represented as dashed grey lines. RES1 and RES2 are the proportions of C from the EOMs in the labile pool and recalcitrant pools, respectively, with RES2 = 1−RES1. CNres, CNres1 and CNres2 are the C:N ratios of the EOM and the labile and recalcitrant pools, respectively, and aCN1 is the ratio CNres1:CNres. Kres1 is the decomposition rate of the labile pool. Y is the C assimilation yield of the labile pool. CNbio, CNa and CNs are the C:N ratios of the microbial biomass, and active and stable pools, respectively. Kbio and Ka are the decay rates of the zymogenous biomass and the active soil organic matter pools, respectively. H is the C humification yield"

At the beginning of the simulation, the stable pool of SOM was 65% of total SOM. C:N ratio of active (CN_a) and stable (CN_s) pools of SOM were set as equal at the beginning of the simulation, based on measured soil characteristics. K_a was calibrated using kinetics of C and N mineralization during soil incubations without EOM. The optimization used the optim function with the "Brent" method (Brent, 1973). Lower and upper bounds were 0.0005 day^{-1} and 0.05 day^{-1}. The function to minimize was the sum of rRMSE (relative root mean squared error, section 2.6) of mineralized C (mass of C per mass of Dry Soil, gC $kgDS^{-1}$) and rRMSE of mineralized N (gN $kgDS^{-1}$). In the soil that we considered in this work, the kinetics of degradation constant of labile SOM K_a is equal to 3.78 10^{-4} day^{-1} according to STICS equations. For all EOMs, CN_{bio} = 7, K_{bio} = 0.0076 day^{-1}, and H = 0.88.

Supplementary Table S2.A – Bounds used to calibrate parameters in (EOM_cal_full) and (EOM_cal_part) sets for EOM mineralization submodule of STICS. K_{bio}, H, and CN_{bio} were fixed.

Parameter	Lower bound		Higher bound
K_a		3.78 10^{-4} day^{-1}	
RES_1	0		1
a_{CN1}	0.1		10
	Additionally, the OM in every pool had to be lower than 1000 and superior than 0.		
K_{res1}	0.005 day^{-1}		0.7 day^{-1}
Y	0.1		0.6
CN_{bio}		7	
H		0.88	
K_{bio}		0.0076 day^{-1}	
K_{res2}	10^{-5} day^{-1}		10^{-3} day^{-1}

Table S2.B – Value of parameters inside the two sets of parameters used in the EOM mineralization submodule of STICS. N1 is the fraction of N in the labile pool (N1= RES1/a_{CN1}).

Set	EOM	RES_1	a_{CN1}	K_{res1} (day^{-1})	Y	$N1$
(EOM_cal)	Cattle manure	0.265	10	0.0190	0.296	0.03
	Cattle slurry	0.574	10	0.0590	0.522	0.06
	Raw digestate	0.265	10	0.0282	0.600	0.03
	Liquid digestate	0.217	1.66	0.3422	0.100	0.13
	Solid digestate	0.507	10	0.0179	0.475	0.05
(EOM_def)	Cattle manure	0.24	3.09	0.025	0.32	0.08
	Cattle slurry	0.42	2.14	0.062	0.39	0.20
	Raw digestate	0.24	10	0.024	0.25	0.02
	Liquid digestate	0.24	10	0.024	0.25	0.02
	Solid digestate	0.24	10	0.024	0.25	0.02

Levavasseur, F., Lashermes, G., Mary, B., Morvan, T., Nicolardot, B., Parnaudeau, V., Thuriès, L., Houot, S., 2021a. Quantifying and simulating carbon and nitrogen mineralization from diverse exogenous organic matters. Soil Use and Management sum.12745. https://doi.org/10.1111/sum.12745

S. M. 3 STICS submodule for ammonia volatilization after spreading of EOM.

The STICS submodule for ammonia volatilization after spreading of EOM is partly described in Brisson et al. (2008), and its source code is available on http://www6.paca.inra.fr/stics_eng. We present here the equations used in the module. We renamed variables.

Daily inputs

T surface temperature (K)

w mean soil moisture in the 0 – 2 cm depth, expressed in volumetric water content (% v/v)

Wind speed

Leef Area Index (LAI)

Crop height

Other inputs

DM_{EOM} is the DM content of EOM (% w/w).

AN_{EOM} is the ammoniacal N brought by the EOM (kgN ha^{-1})

RecentTillage is equal to -1 if the soil was tilled during the last 7 days and +1 else.

pH_{soil} is the pH of soil

BD is the bulk density of the first soil layer (g cm^{-3}). It is used to compute soil porosity (Porosity = 1 – BD /2.66)

Parameters

The layer of soil considered is z = 0.01 m

$[NH_3(g)]_{atm}$ is the reference concentration of NH_3 in the atmosphere, is default value is 0 µgN m^{-3}

General functionning

Volatilization is supposed to occur within the first 0-1 cm of soil. When EOM is applied, ammoniacal N is brought inside the first layer of soil. A pool of N that can be volatilized ($AN_{surface}$) is computed initially, to model the infiltration of EOM inside the soil (Step 1). Soil pH is modified by EOM application (Step 2). At the daily timestep, volatilization occurs (Steps 3 and 4), and then ammoniacal N on the first 0 – 1 cm of soil is adjusted. Ammoniacal N can also be nitrified, leached, taken by plants, or emmited in the atmosphere as N_2 or N_2O, as described in Brisson et al. (2008). At the end of each day, the content of N inside the first 0 – 1 cm of soil and the pool of N that can be volatilized are adjusted. If the soil is tilled, volatilization stops.

Step 1

Initially, when EOM is applied, a pool of ammoniacal N that is close to the surface and that can be volatilized is computed.

(equation S1) $$AN_{surface}^{initial} = AN_{EOM} \cdot (0.37 + 0.029 \cdot DM_{EOM} + 0.117 \cdot RecentTillage)$$

Where $AN_{surface}^{initial}$ is the pool of ammoniacal N at the surface that can be volatilized at application date (kgN ha^{-1}).

Step 2

When EOM is applied, pH of soil surface ($pH_{surface}$) is changed:

(equation S2) $$pH_{surface} = pH_{soil} + \Delta pH_{surface}$$

with

(equation S3) $$\Delta pH_{surface} = \begin{cases} \min(0.005 \cdot AN_{EOM}, 1.0), & \text{if } pH_{soil} \leq 7.0 \\ \min(0.005 \cdot AN_{EOM}, 1.0) \cdot \frac{8.6 - pH_{soil}}{8.6 - 7}, & \text{if } 7.0 \leq pH_{soil} \leq 8.6 \\ 0, & \text{if } 8.6 \leq pH_{soil} \end{cases}$$

During the following dates, at the daily time step, $pH_{surface}$ returns to its original value with:

(equation S4) $$pH_{surface} = pH_{soil} + \Delta pH_{surface} \cdot \frac{AN_{surface}}{AN_{surface}^{initial}}$$

where $AN_{surface}$ is the ammoniacal N at the surface that can be volatilized for the considered day (kgN ha^{-1}).

Step 3

At the hourly timestep, assuming homogenous environmental conditions through the day, three forms are considered inside the pool of ammoniacal N that could be volatilized: NH_4^+ in aqueous solution, NH_3 in aqueous solution, and NH_3 in the gaseous phase of soil. NH_4^+ adsorbed on solid phase is supposed to be null. Initially, the concentration NH_4^+ and NH_3 in aqueous solution is computed as:

(equation S5) $$[AN(aq)] = [NH_4^+\ (aq)] + [NH_3(aq)] = AN_{surface} \cdot \frac{1}{z \cdot 1000 \cdot w}$$

Where [AN(aq)] is expressed in mol L^{-1}, $AN_{surface}$ is expressed in mol m^{-2}, w is the mean soil moisture in the 0 – 2 cm depth (%v/v), and z is the considered layer of soil (0.01 m)

The content of each of the three forms of ammoniacal N are computed with an iterative algorithm, with the constraints to respect two equations of equilibriums, equation S6 and equation S8. The acid-base equilibrium $NH_3(aq) + H^+ \rightleftarrows NH_4^+(aq)$ is represented by the equation:

(equation S6) $$[NH_3(aq)] = [AN(aq)] \cdot \frac{1}{1 + \frac{[H_3O^+]}{Ka}}$$

Where $[NH_4^+(aq)]$ is the concentration of aqueous NH_4^+ (mol L^{-1}), $[NH_3(aq)]$ is the concentration of aqueous NH_3 (mol L^{-1}), $[H_3O^+]$ (mol L^{-1}) is deduced from $pH_{surface}$, and Ka is a constant depending on surface temperature T (K):

(equation S7) $$Ka = \exp\left(-177.953 - \frac{1843.22}{T} + (31.4335 \cdot \log(T)) - (0.0544943 \cdot T)\right)$$

The equilibrium of aqueous and gaseous forms of NH_3 $NH_3(g) \rightleftarrows NH_3(aq)$ is described by the following equation, from Beutier and Renon (1978):

(equation S8) $$[NH_3(g)]_{soil} = [NH_3(aq)] \cdot Kh \cdot (porosity - w) \cdot 1000 \cdot z \cdot 1.2 \cdot 10^9$$

Where $[NH_3(g)]$ is the concentration of NH_3 in the gaseous phase of soil expressed in µgN m^{-3}. Kh is the constant of Henry's law, which depend on surface temperature:

(equation S9) $$Kh = \exp\left(160.559 - \frac{8621.06}{T} - (25.6767 \cdot \log(T)) + (0.0544943 \cdot T)\right)$$

Step 4: Volatilization

At the hourly time step, within a dedicated subroutine, the volatilization is computed with a resistive approach:

(equation S10) $$Volat_{pot} = \frac{[NH_3(g)]_{soil} - [NH_3(g)]_{atm}}{AR_{S-C} + AR_{C-A}}$$

(equation S11) $$Volat = \min(AN_{surface}, Volat_{pot})$$

Where $Volat_{pot}$, Volat, and $AN_{surface}$ are expressed in kgN ha^{-1}. AR_{S-C} is the aerodynamical resistance between soil and top of the canopy, AR_{C-A} is the aerodynamical resistance between the top of the canopy and reference level in the atmosphere. AR_{C-A} and AR_{S-C} are expressed in s m^{-1} and are computed inside STICS model, depending on LAI, crop height, and wind speed.

Finally, all N pools are adjusted.

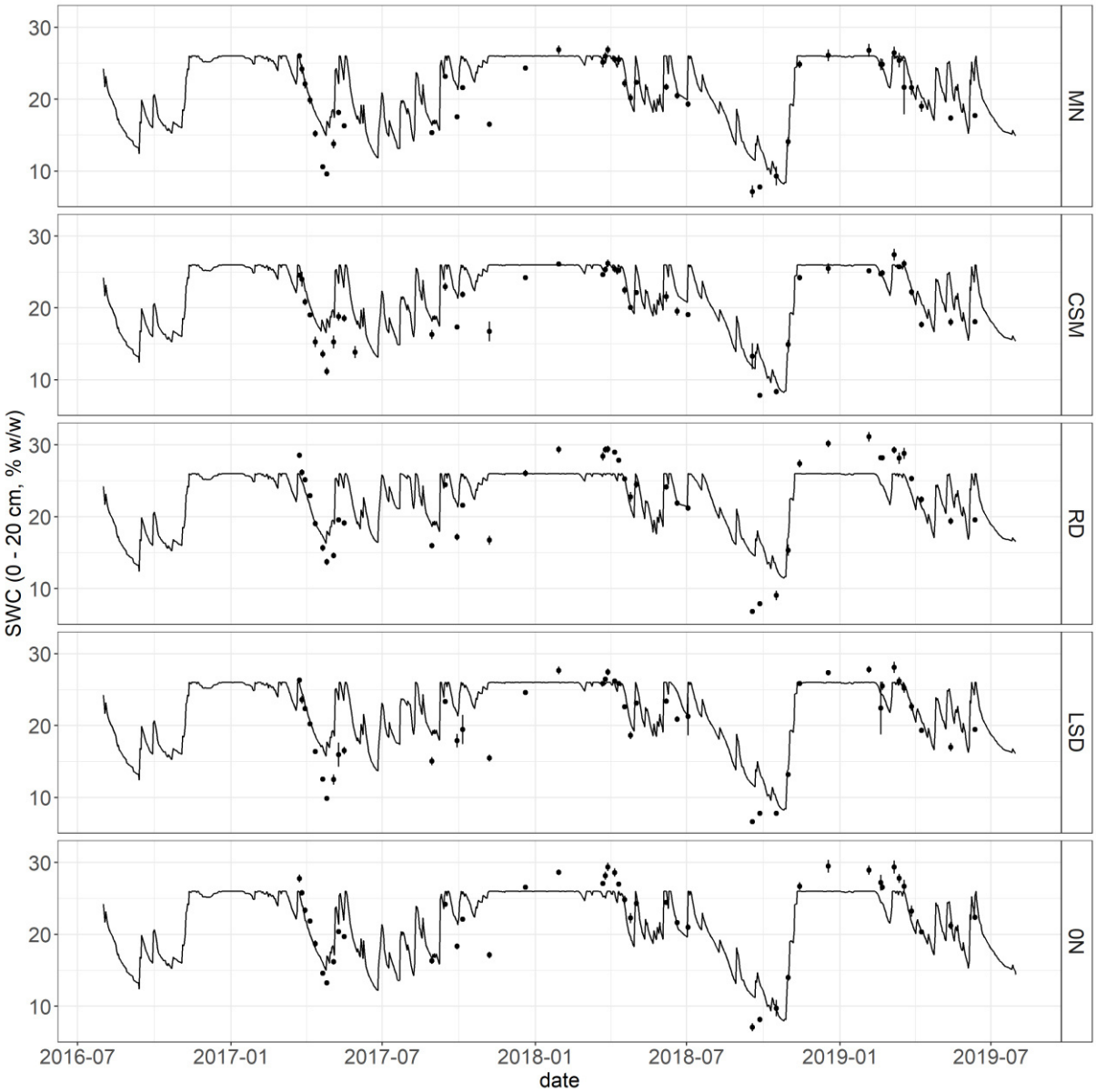
Brisson, N., Launay, M., Mary, B., Beaudoin, N., 2008. Conceptual basis, formalisations and parameterization of the STICS crop model. Editions Quae, Paris.

S. M. 4 Evaluation of the performance of STICS and ALFAM2 model to predict volatilization

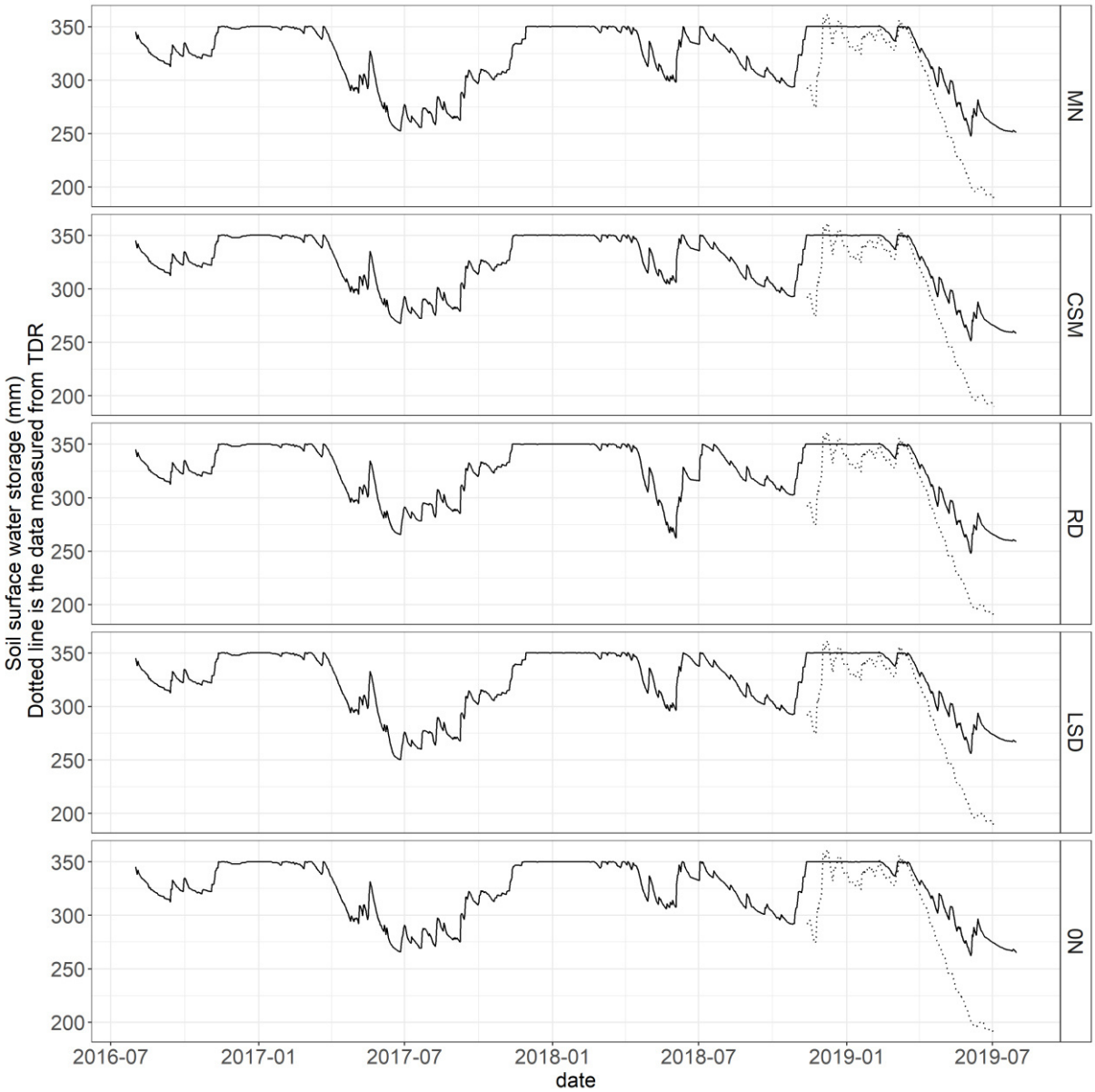
Table S4: Evaluation of the performance of STICS and ALFAM2 model to predict volatilization. RMSE and MD are indicated in kgN ha^{-1}.

EOM considered	STICS				ALFAM (liquid) and emission factors (solids)			
	RMSE (rRMSE)	MD (rMD)	EF	r^2	RMSE (rRMSE)	MD (rMD)	EF	r^2
All	14.7 (109%)	-9.2 (-67%)	-0.59	0.03	9.4 (70%)	-2.5 (-19%)	0.34	0.37
Liquid EOM	14.5 (113%)	-8.2 (-63%)	-0.66	0.04	9.7 (75%)	-3.3 (-27%)	0.37	0.47
Slurry, liquid digestate	5.0 (69%)	-2.4 (-34%)	0.44	0.37	3.8 (52%)	-0.7 (-10%)	0.68	0.52

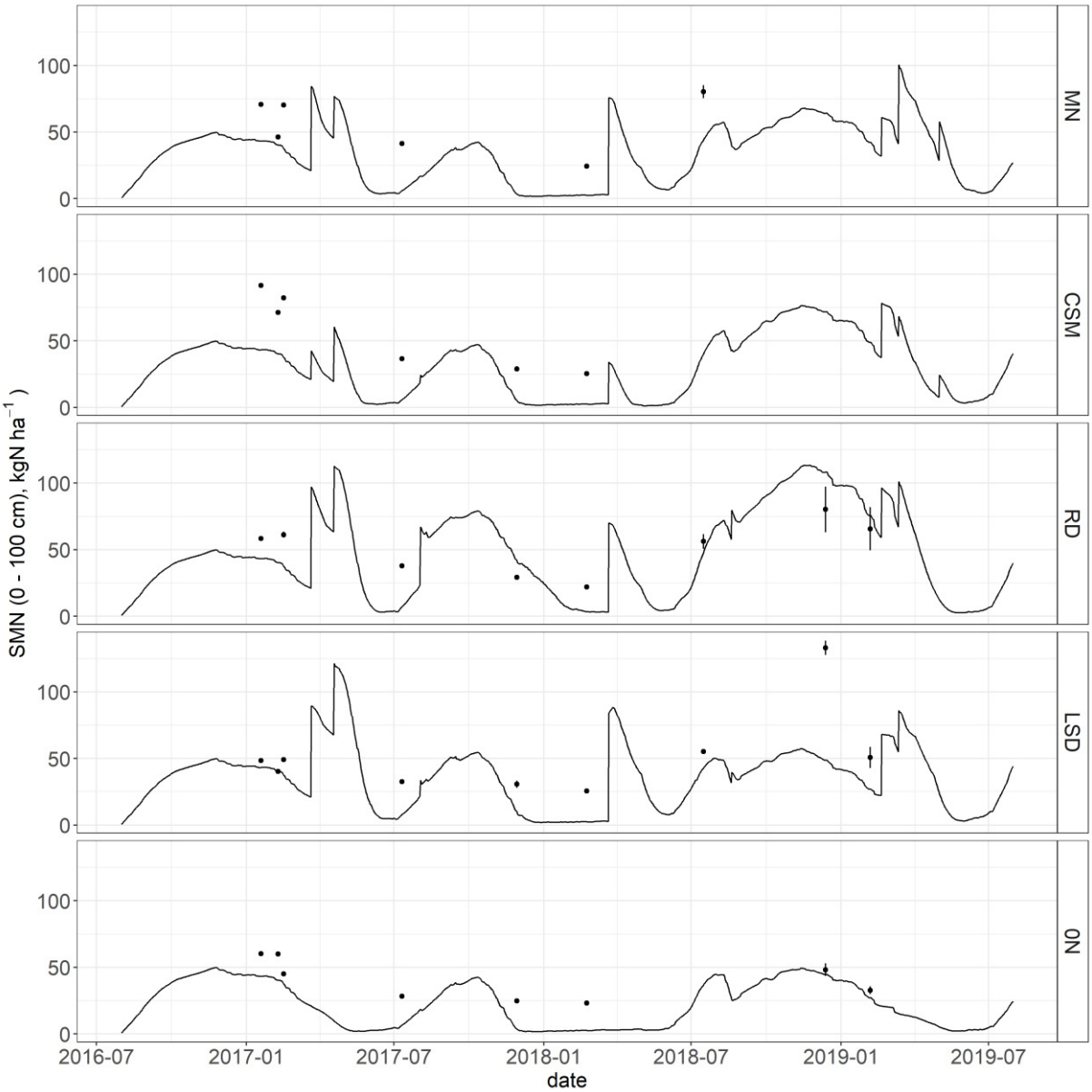
S. M. 5 Evaluation of STICS in the reference parametrization (NH3_ALFAM2) and (EOM_cal)



Supplementary Fig. S3.A – Observed and modelled SWC, on the topsoil (0-20cm). Solid lines represent modelled SWC. Dots represent measured SWC; error bars show one standard error. STICS instance used (EOM_cal) and (NH3_ALFAM2).



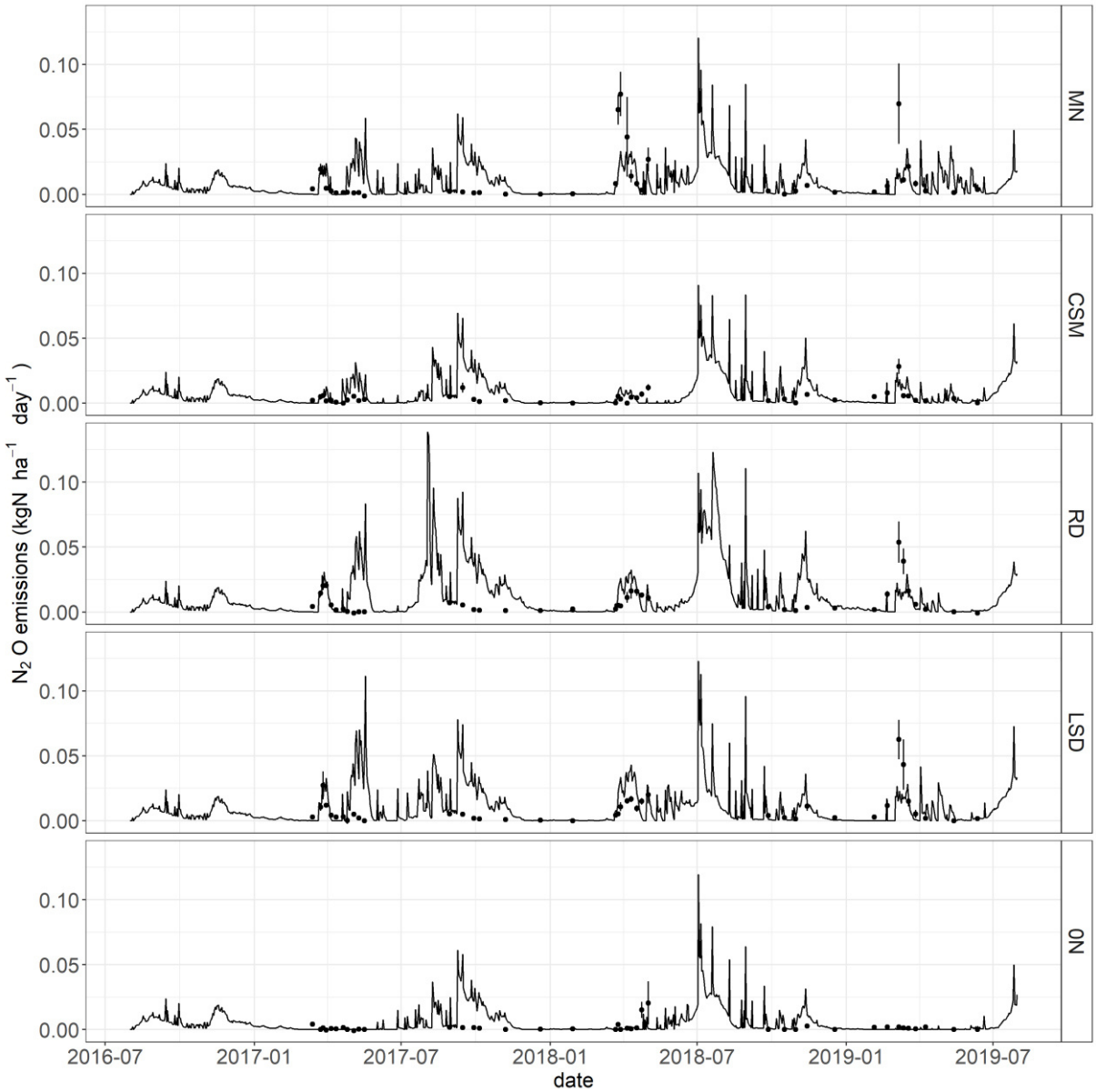
Supplementary Fig. S5.B – Observed and modelled SWC, on soil profile (0-100cm). Solid lines represent modelled SWC. Dotted lines represent measured SWC with TDR. STICS instance used (EOM_cal) and (NH3_ALFAM2).



Supplementary Fig. S5.C – Observed and modelled SMN stocks, on soil profile (0-100cm). Solid lines represent modelled SMN. Dots represent measured SWC, error bars show one standard error. STICS instance used (EOM_cal) and (NH3_ALFAM2).

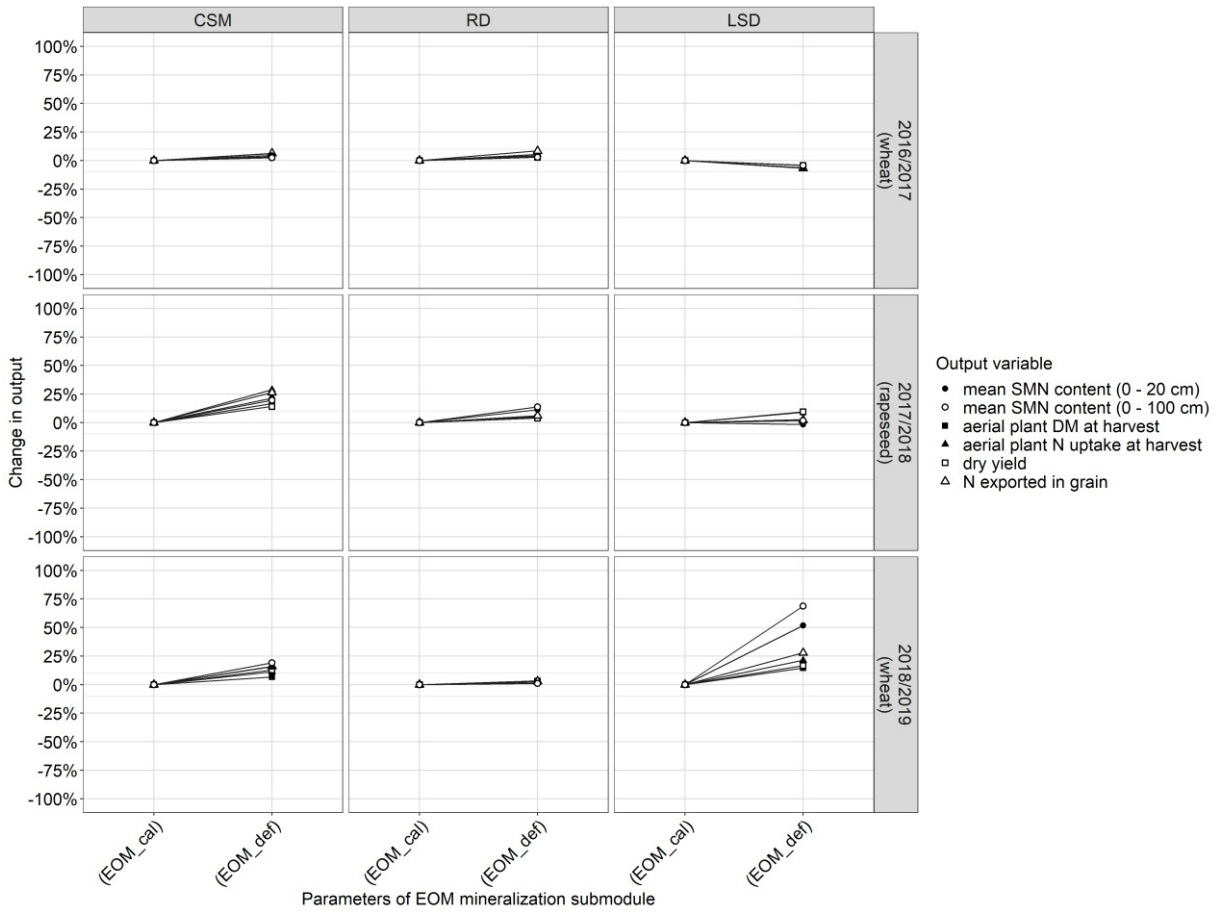
Supplementary Table S5 – Evaluation of STICS on main outputs, in the reference parametrization. STICS instance used (EOM_cal) and (NH3_ALFAM2).

Points considered	variable	RMSE	rRMSE (%)	ME	ARE (%)	EF	r^2
Three years CSM, RD, and LSD treatments	Aerial dry matter (t ha^{-1})	2.68	22	0.53	4	-0.33	0.78
	Aerial N uptake (kgN ha^{-1})	36.6	26	21.5	16	-3.57	0.20
	Grain yield (t ha^{-1})	0.74	14	-0.47	-9	0.81	0.91
	N in grain (kgN ha^{-1})	14.7	14	1.7	2	0.16	0.62
	SMN (0-20 cm) (kgN ha^{-1})	25.4	90	8.5	30	-0.27	0.31
	SMN (0-100 cm) (kgN ha^{-1})	29.6	56	-19.7	-37	-0.31	0.40
All treatments Wheat: from 1st of August 2016 to 28th of July 2017, and from 6th of July 2018 to 31st of July 2019	Aerial dry matter (t ha^{-1})	3.05	24	2.46	20	0.05	0.67
	Aerial N uptake (kgN ha^{-1})	36.8	29	31.8	25	-0.08	0.81
	Grain yield (t ha^{-1})	0.74	12	-0.08	-1	0.75	0.77
	N in grain (kgN ha^{-1})	14.9	16	11.4	12	0.71	0.92
	SMN (0-20 cm) (kgN ha^{-1})	25.4	110	11.2	48	-1.09	0.37
	SMN (0-100 cm) (kgN ha^{-1})	28.8	49	-20.4	-35	-0.77	0.29
All treatments Rapeseed: from 29th of July 2017 to 5th of July 2018	Aerial dry matter (t ha^{-1})	2.55	30	-2.16	-25	-2.48	0.34
	Aerial N uptake (kgN ha^{-1})	19.1	15	-3.3	-3	0.49	0.73
	Grain yield (t ha^{-1})	0.73	25	-0.55	-19	-1.36	0.30
	N in grain (kgN ha^{-1})	16.5	18	-13.0	-14	0.11	0.75
	SMN (0-20 cm) (kgN ha^{-1})	16.0	52	-5.9	-19	0.71	0.77
	SMN (0-100 cm) (kgN ha^{-1})	20.8	80	-17.8	-68	-54.24	0.30

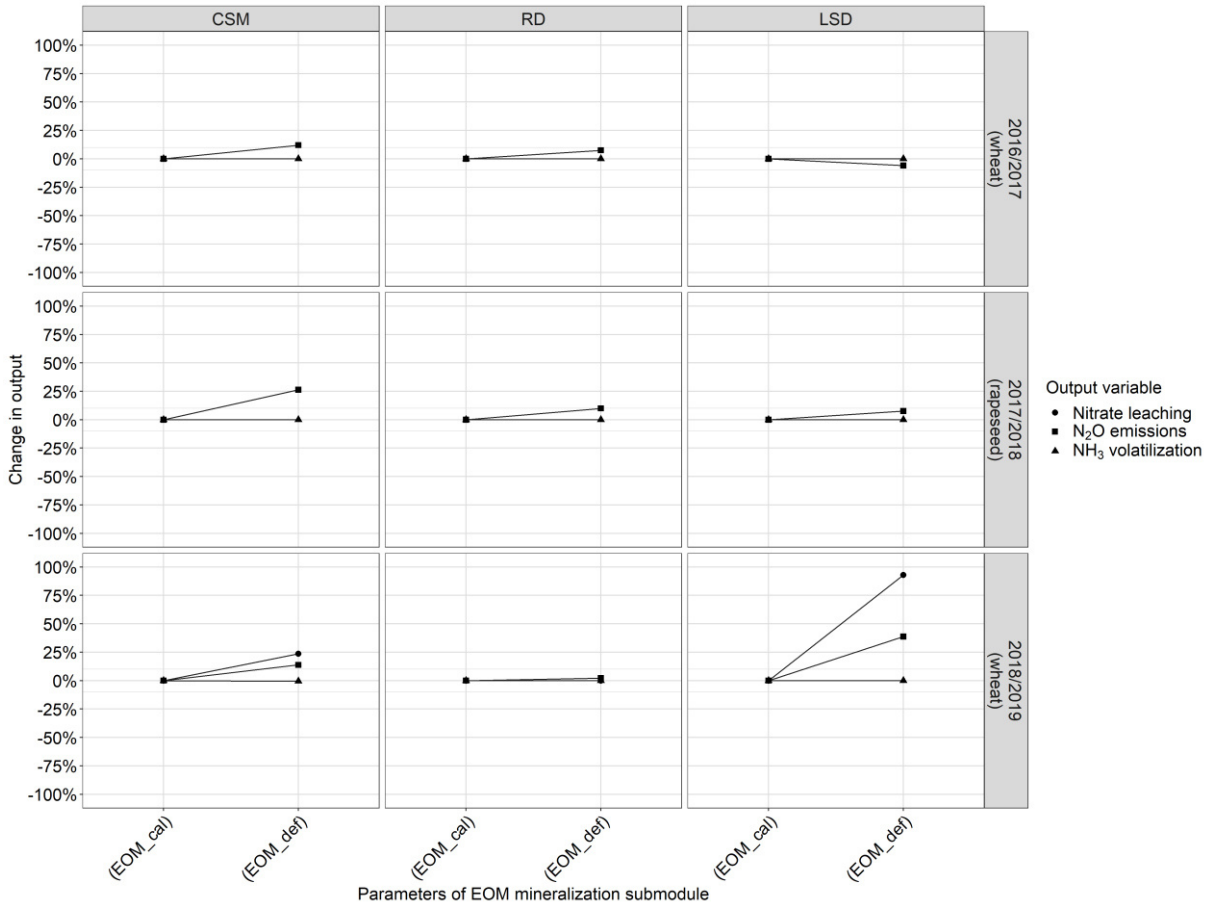


Supplementary Fig. S5.D – Modelled and measured N_2O emissions. Solid line represent the modelled daily emissions. Dots represent measured punctual emissions, error bar represent one standard error.

S. M. 6 Analysis of sensitivity of STICS outputs induced by the use of different EOM mineralization submodules

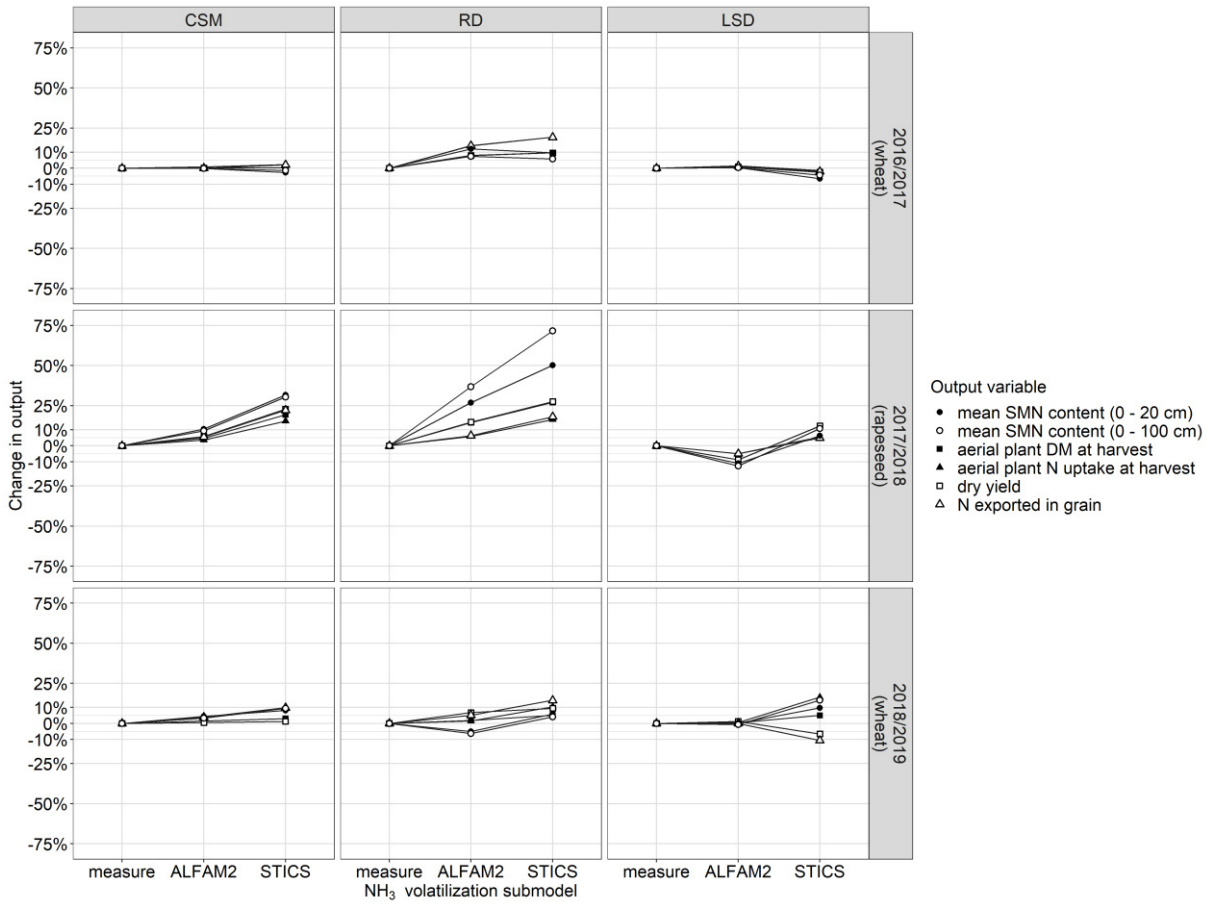


Supplementary Fig. S6.A – Relative change of STICS outputs induced by the use of different EOM mineralization submodules (1/2)

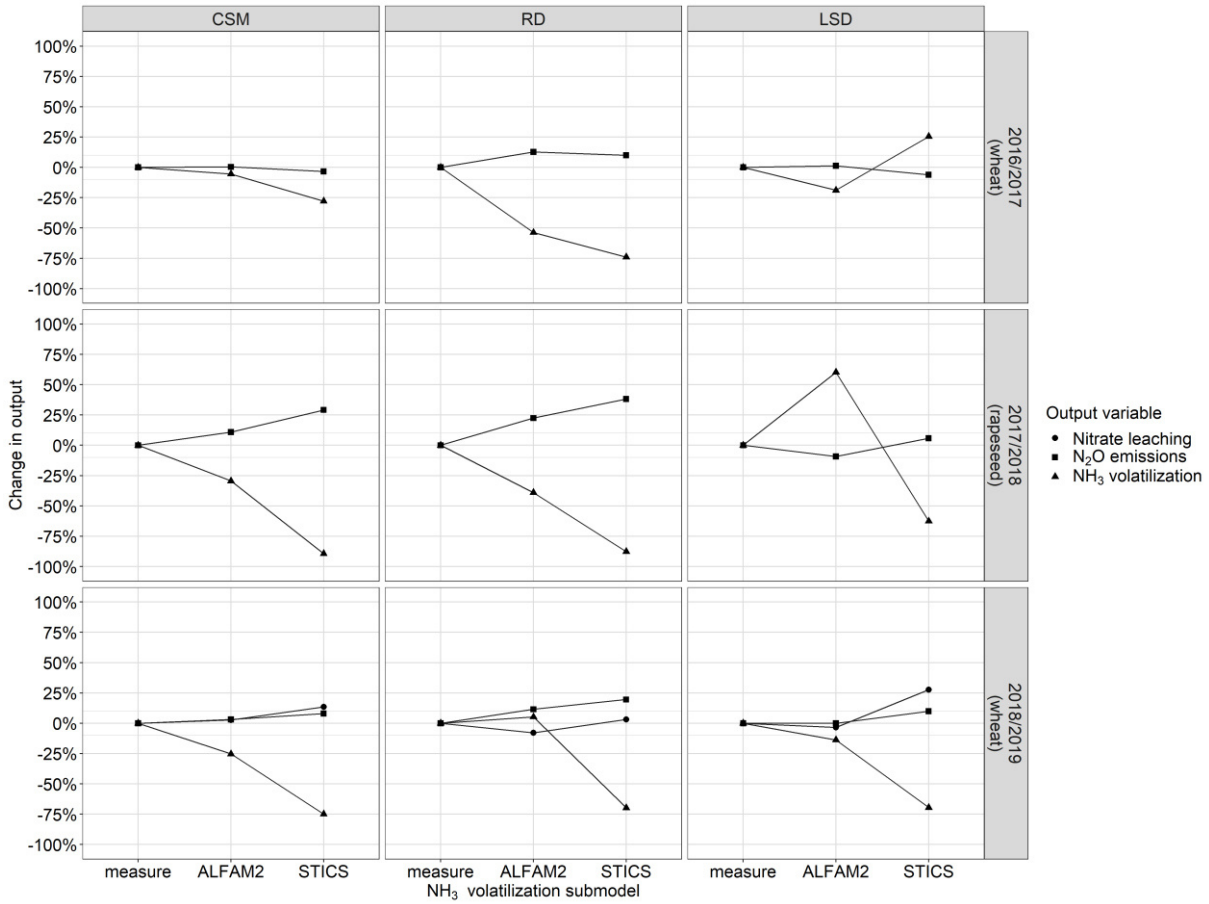


Supplementary Fig. S6.B – Relative change of STICS outputs induced by the use of different EOM mineralization submodules (2/2)

S. M. 7 Analysis of sensitivity of STICS outputs induced by the use of different EOM volatilization submodules

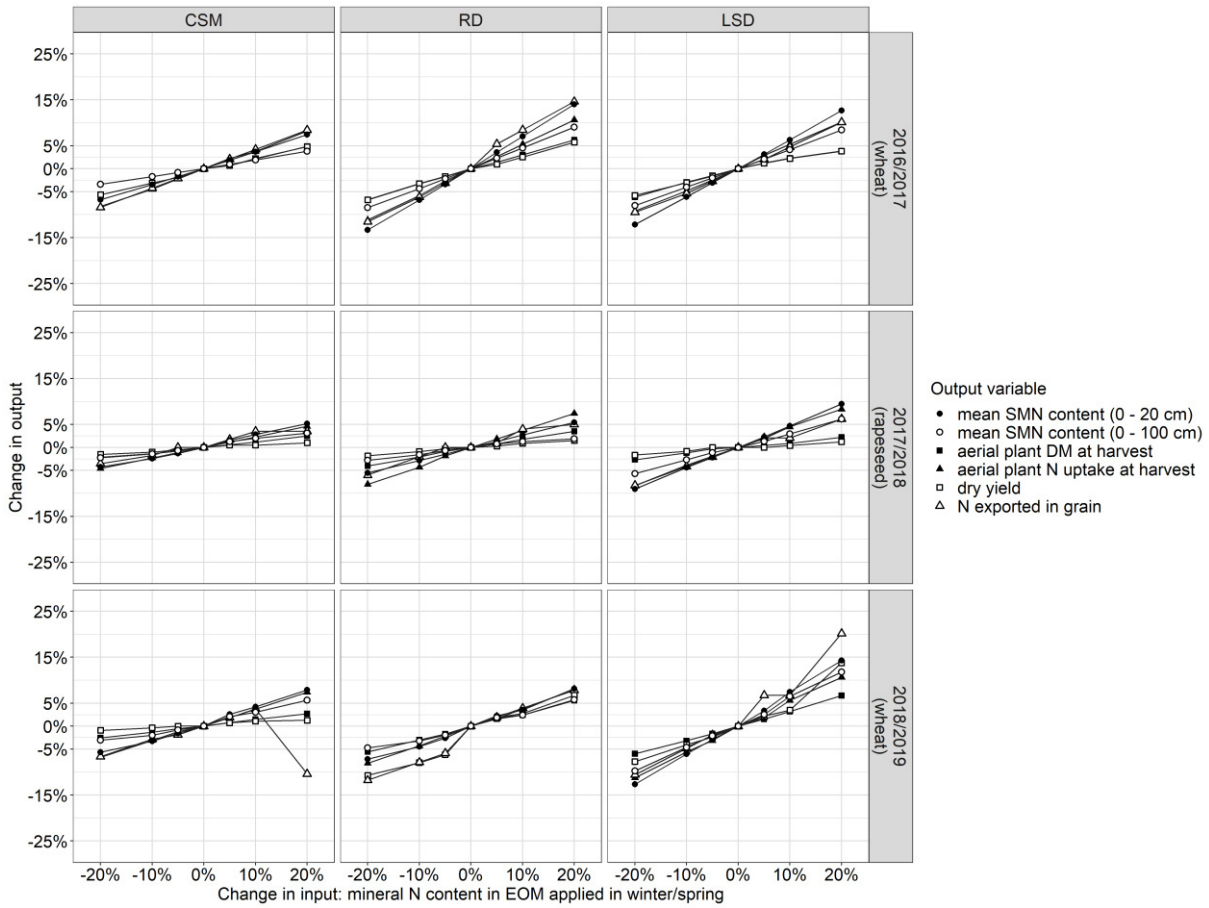


Supplementary Fig. S7.A – Relative change of STICS outputs induced by the use of different EOM volatilization submodules (1/2)

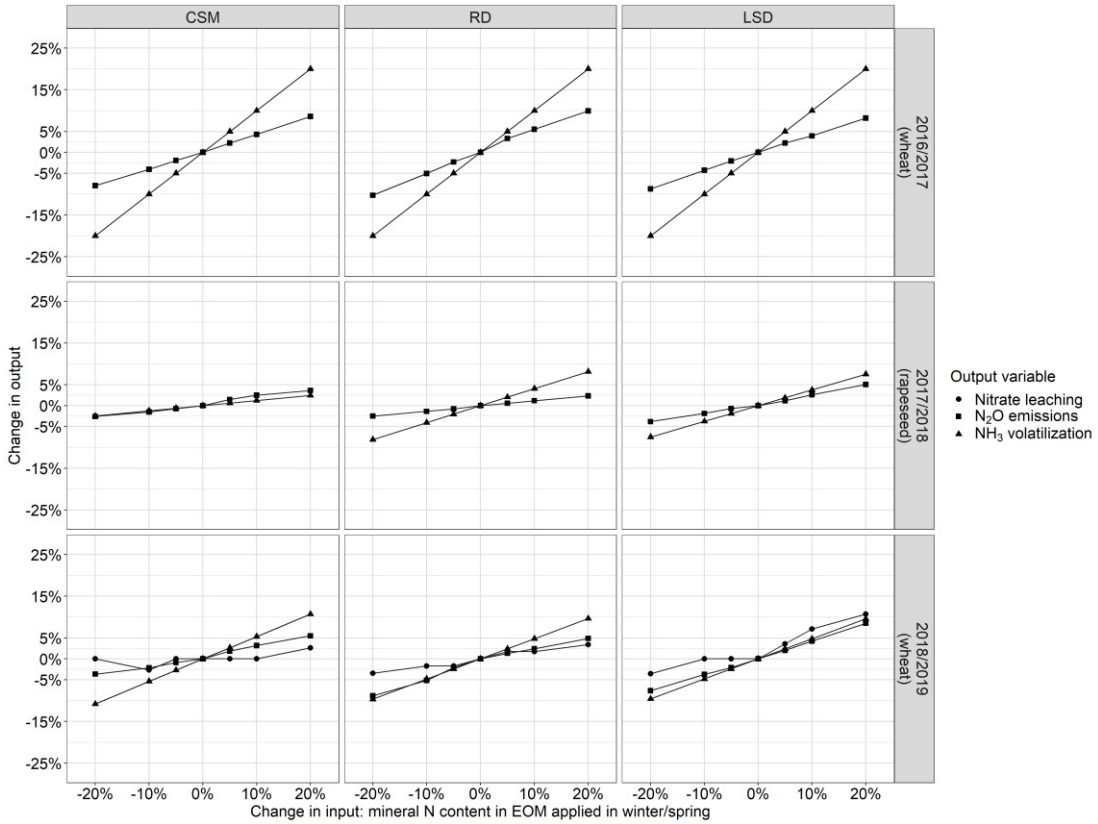


Supplementary Fig. S7.B – Relative change of STICS outputs induced by the use of different EOM volatilization submodules (2/2)

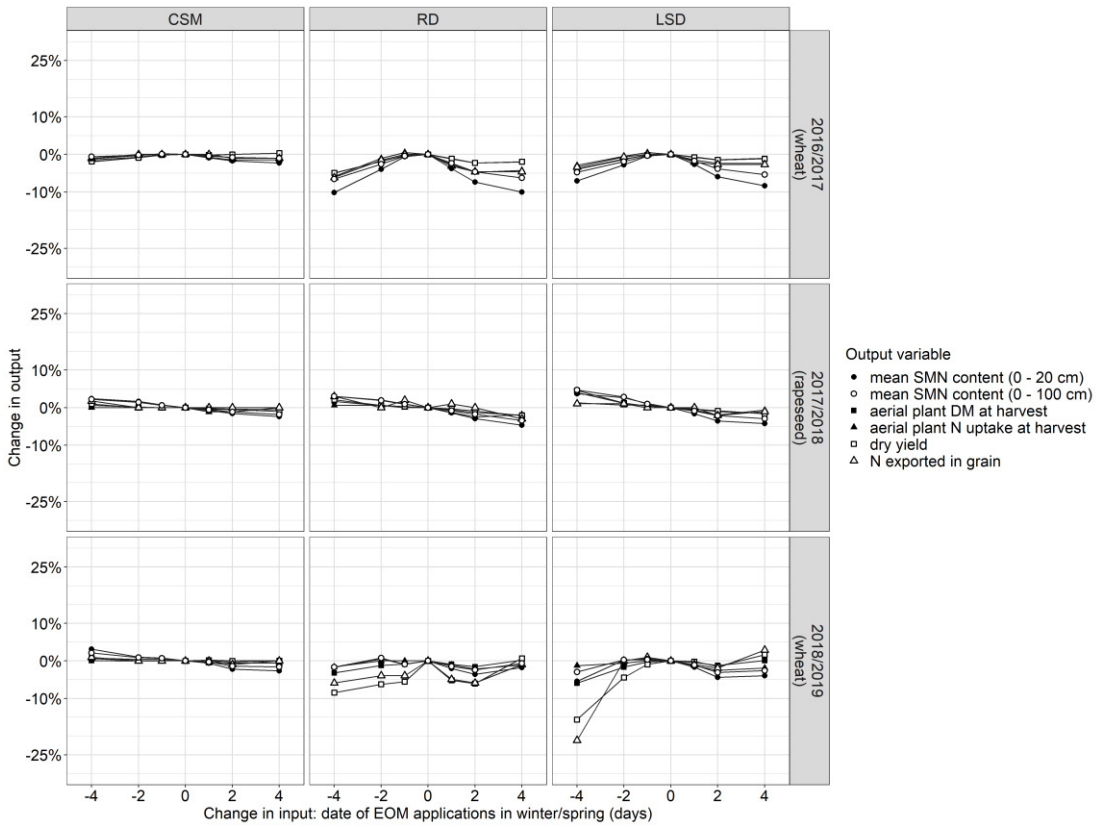
S. M. 8 Analysis of sensitivity of STICS outputs induced by the variation of EOM characteristics (winter/spring)



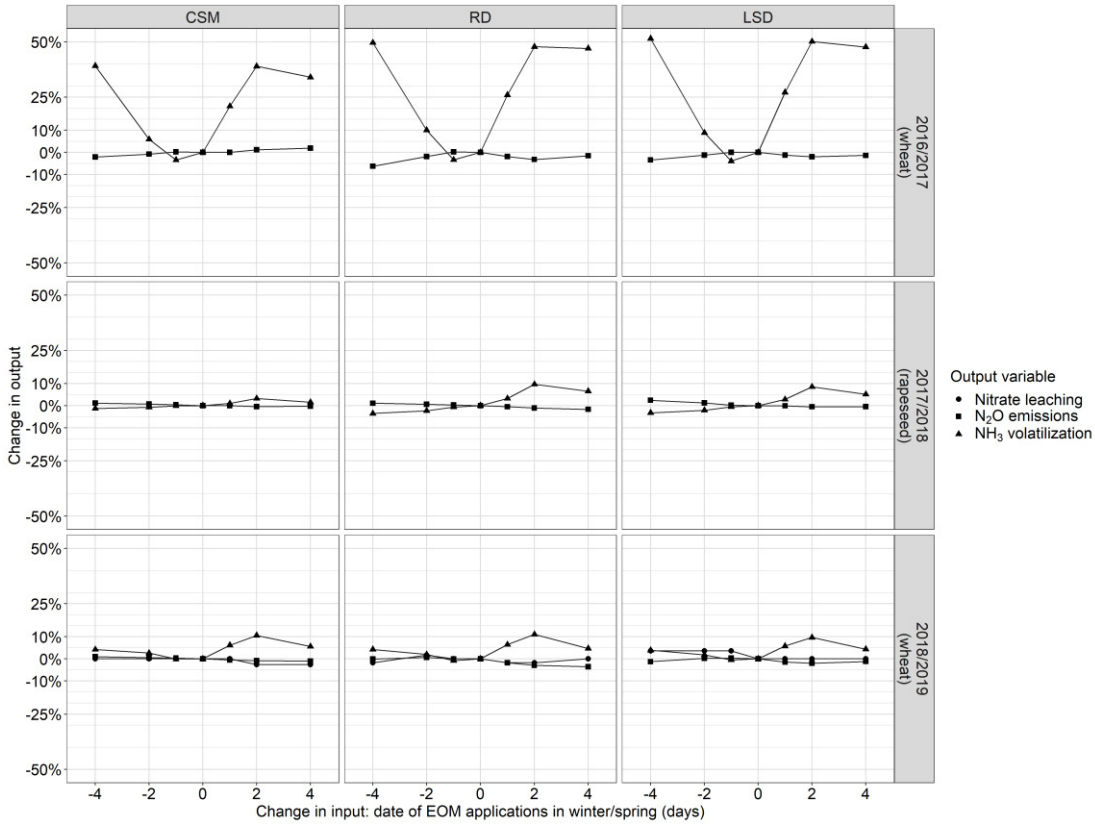
Supplementary Fig. S8.A – Relative change of STICS outputs induced by the variation of TAN content of EOM (winter/spring) (1/2)



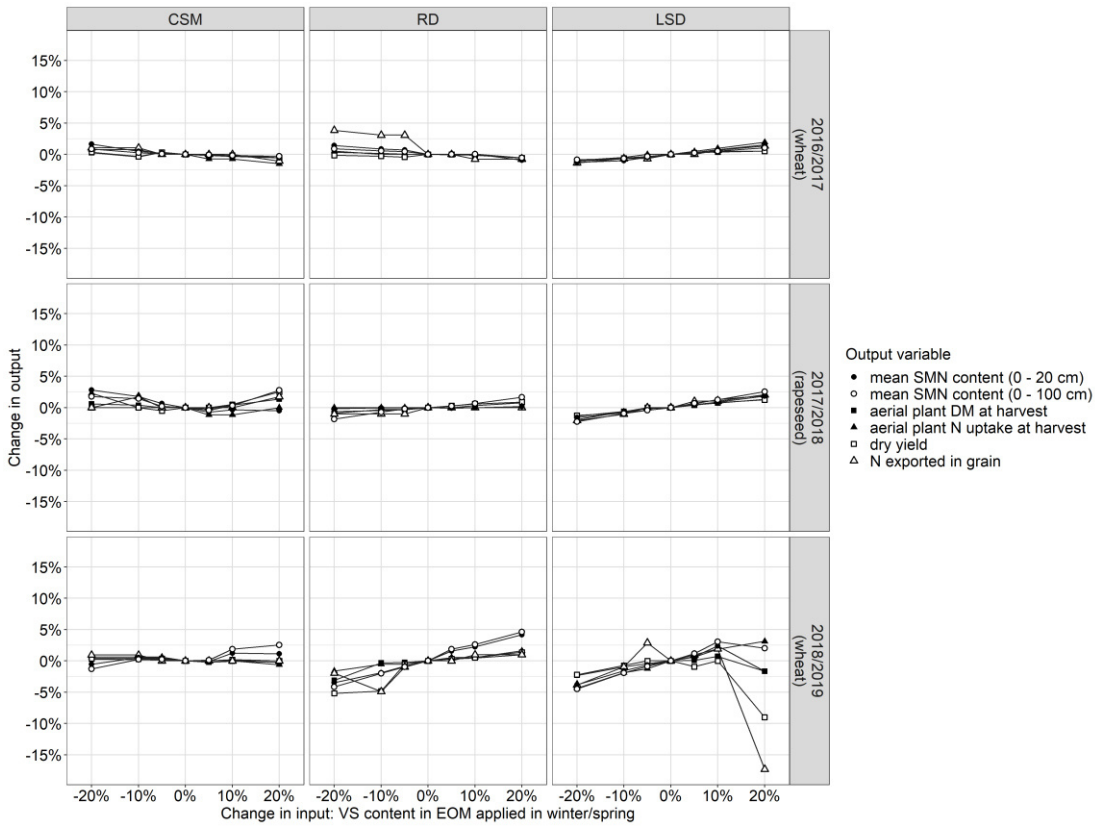
Supplementary Fig. S8.B – Relative change of STICS outputs induced by the variation of TAN content of EOM (winter/spring) (2/2)



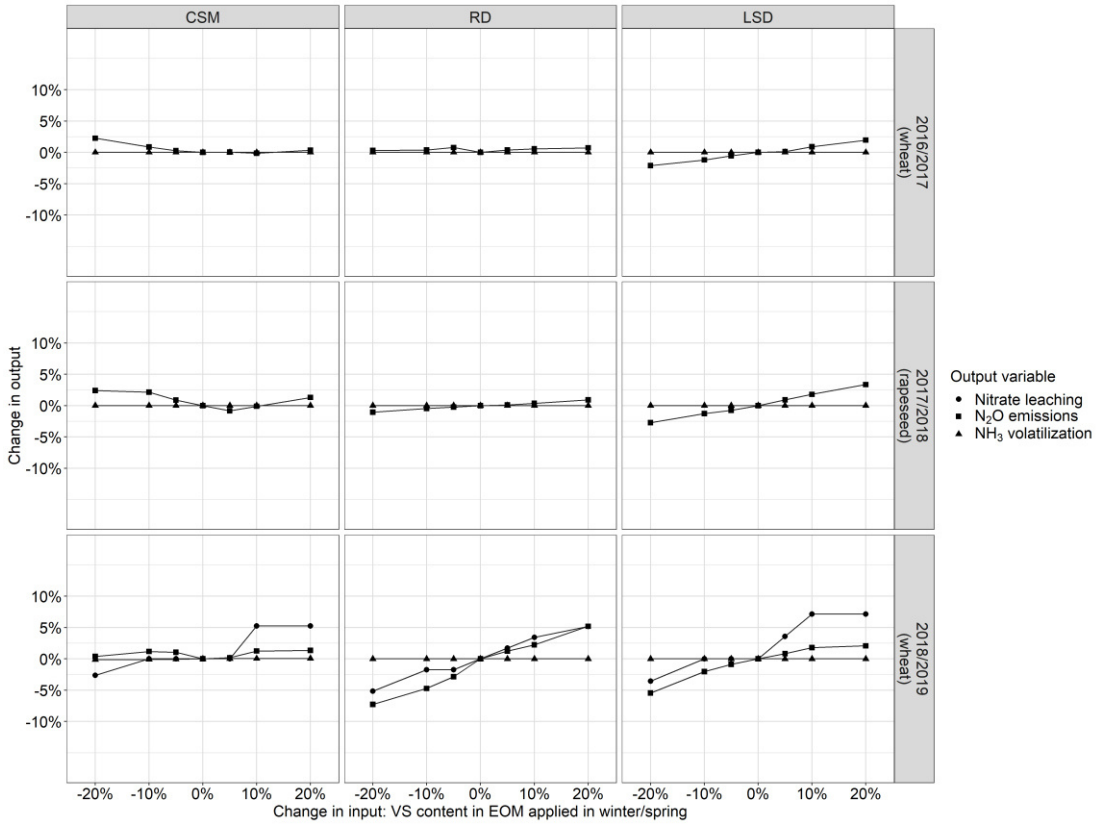
Supplementary Fig. S8.C – Relative change of STICS outputs induced by the variation of application date of EOM (winter/spring) (1/2)



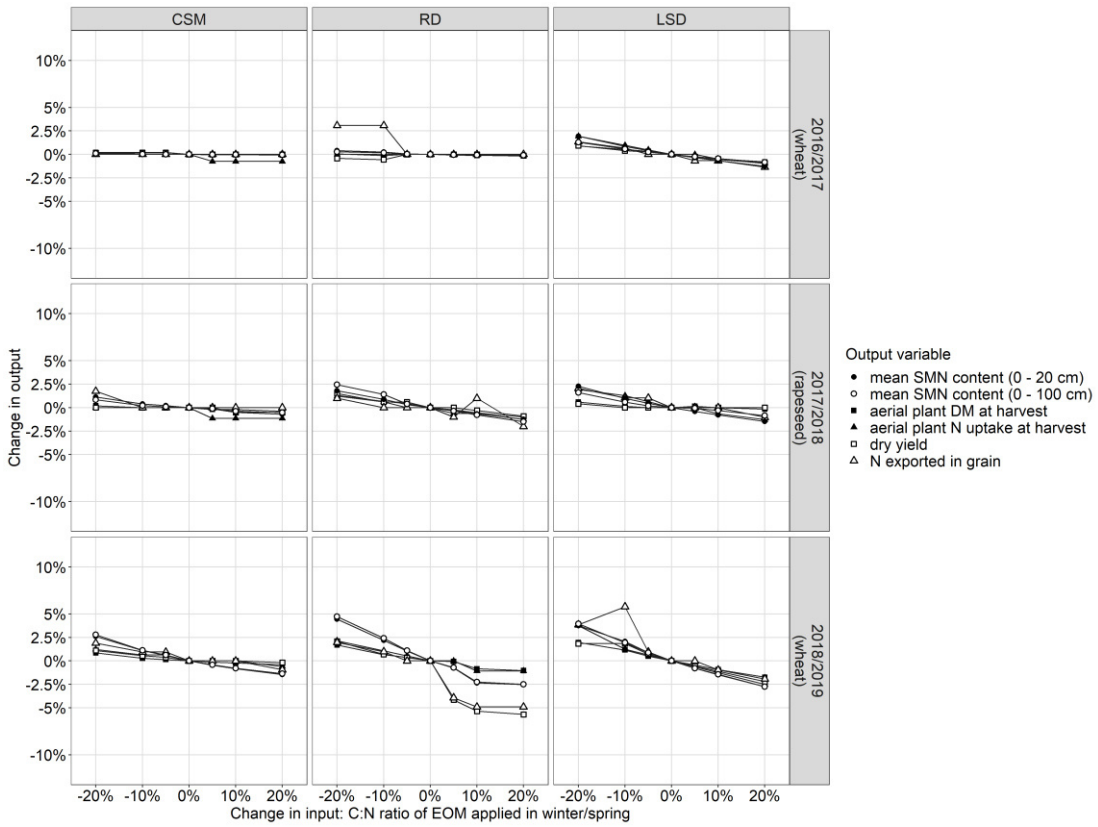
Supplementary Fig. S8.D – Relative change of STICS outputs induced by the variation of application date of EOM (winter/spring) (2/2)



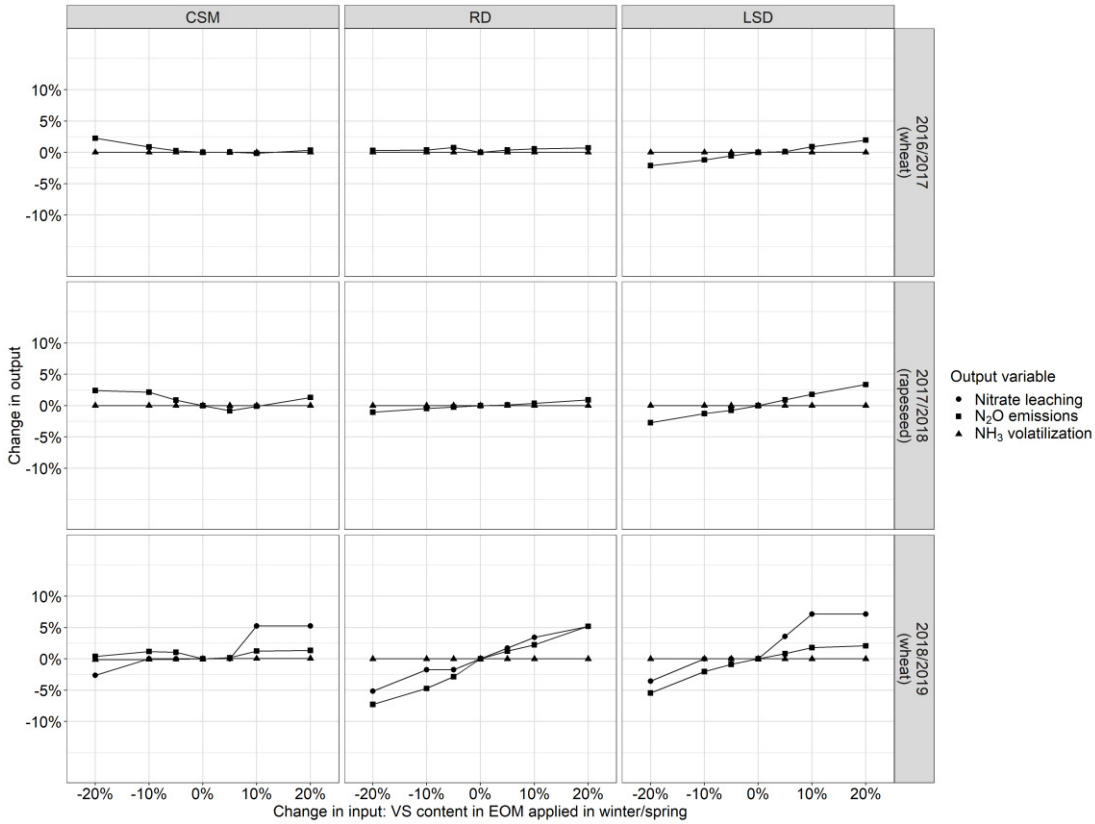
Supplementary Fig. S8.E – Relative change of STICS outputs induced by the variation of VS content of EOM (winter/spring) (1/2)



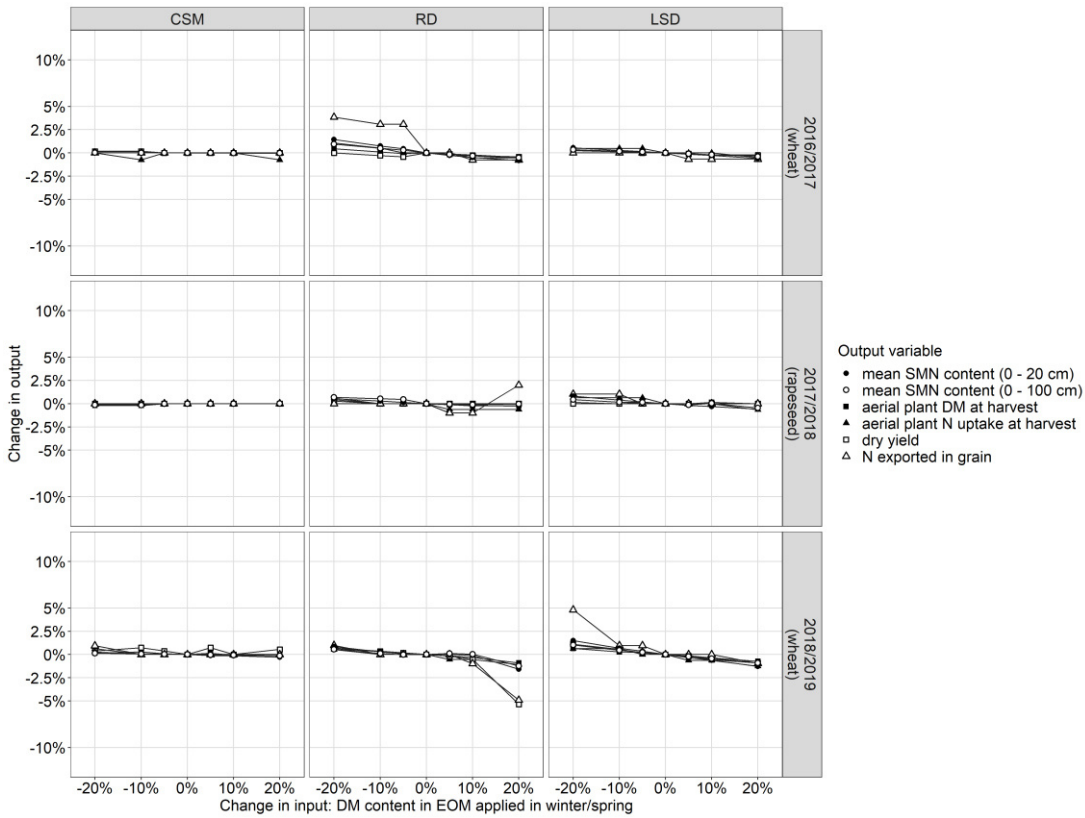
Supplementary Fig. S8.F – Relative change of STICS outputs induced by the variation of VS content of EOM (winter/spring) (2/2)



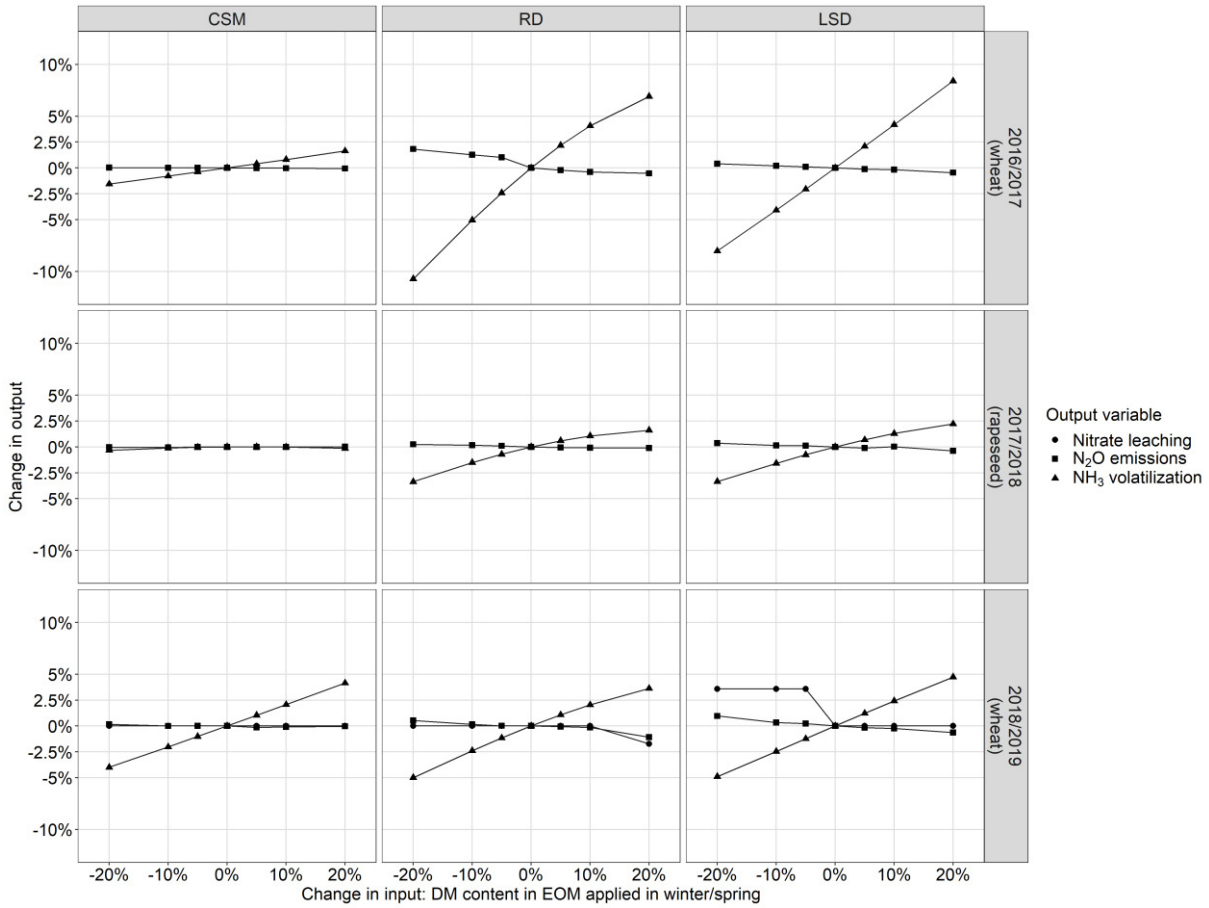
Supplementary Fig. S8.G – Relative change of STICS outputs induced by the variation of C:N_{org} ratio of EOM (winter/spring) (1/2)



Supplementary Fig. S8.H – Relative change of STICS outputs induced by the variation of C:N_{org} ratio of EOM (winter/spring) (2/2)

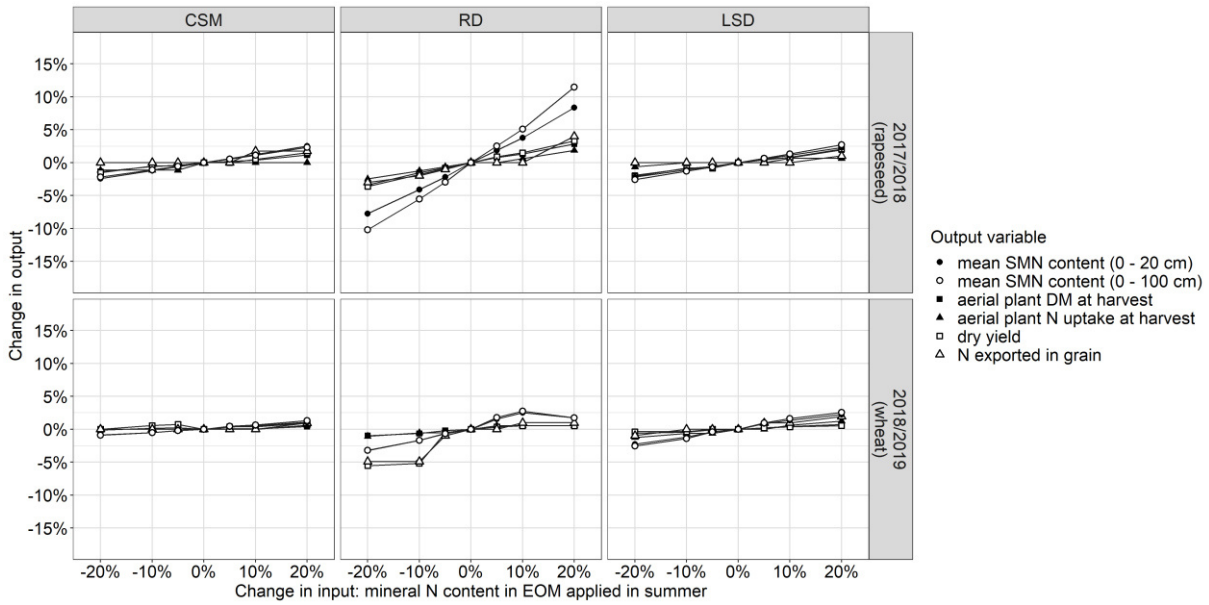


Supplementary Fig. S8.I – Relative change of STICS outputs induced by the variation of DM content of EOM (winter/spring) (1/2)

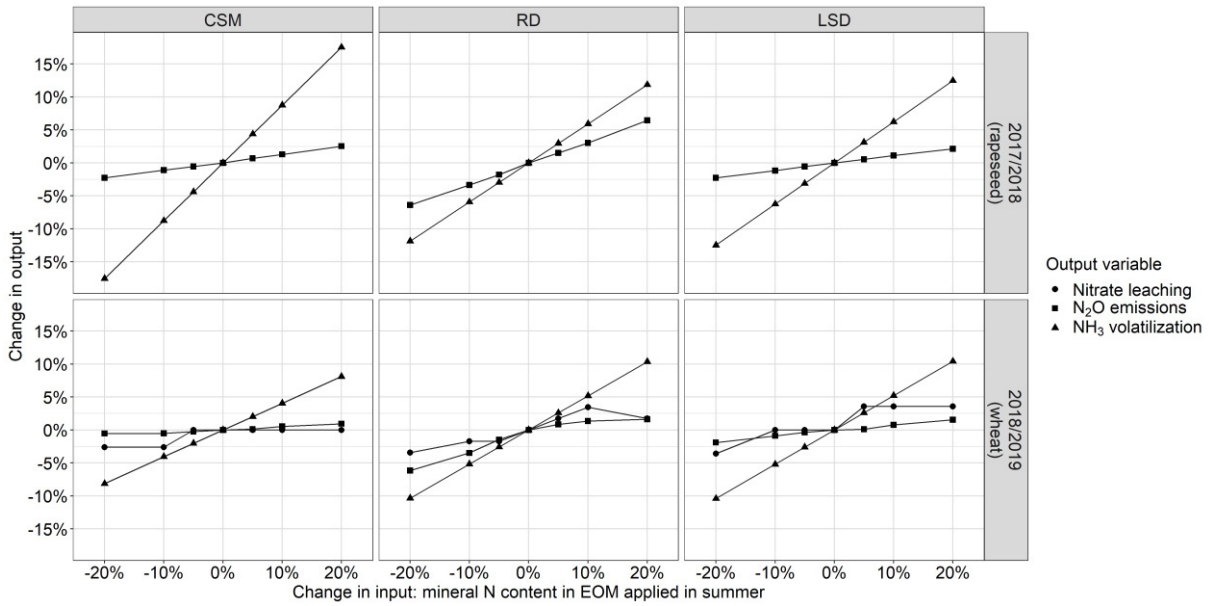


Supplementary Fig. S8.J – Relative change of STICS outputs induced by the variation of DM content of EOM (winter/spring) (2/2)

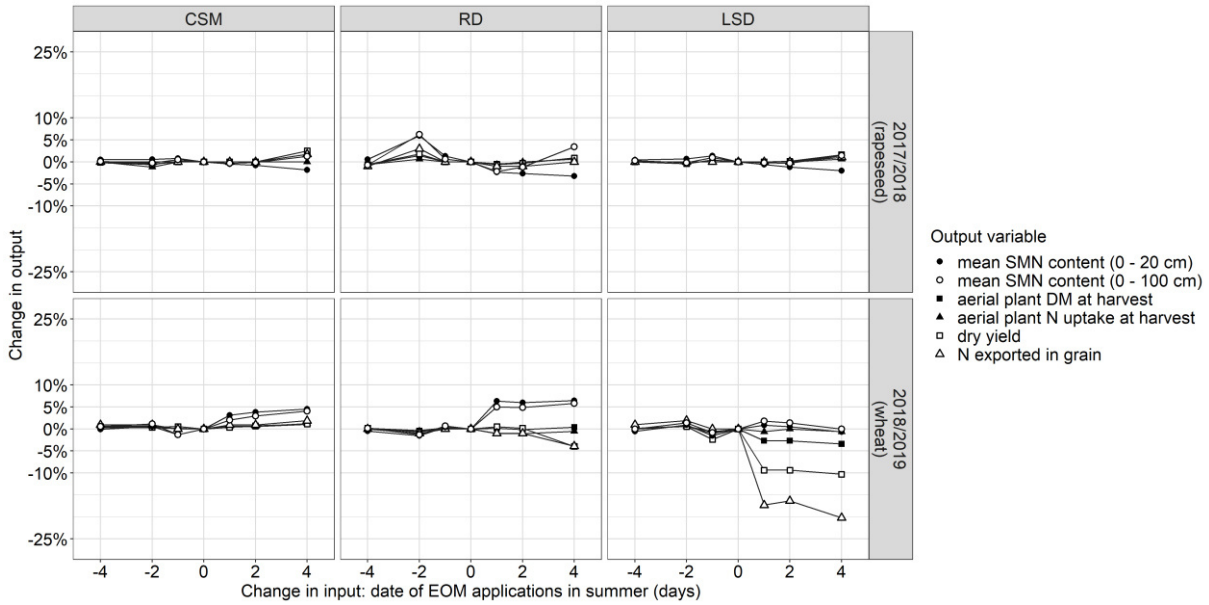
S. M. 9 Analysis of sensitivity of STICS outputs induced by the variation of EOM characteristics (summer)



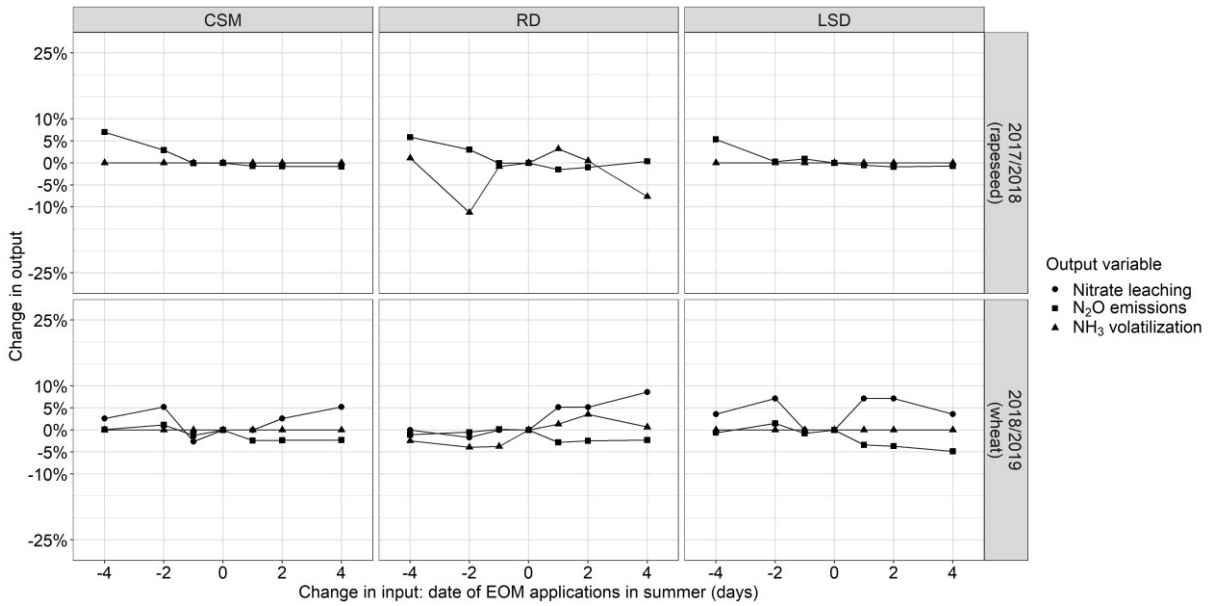
Supplementary Fig. S9.A– Relative change of STICS outputs induced by the variation of TAN content of EOM (summer) (1/2)



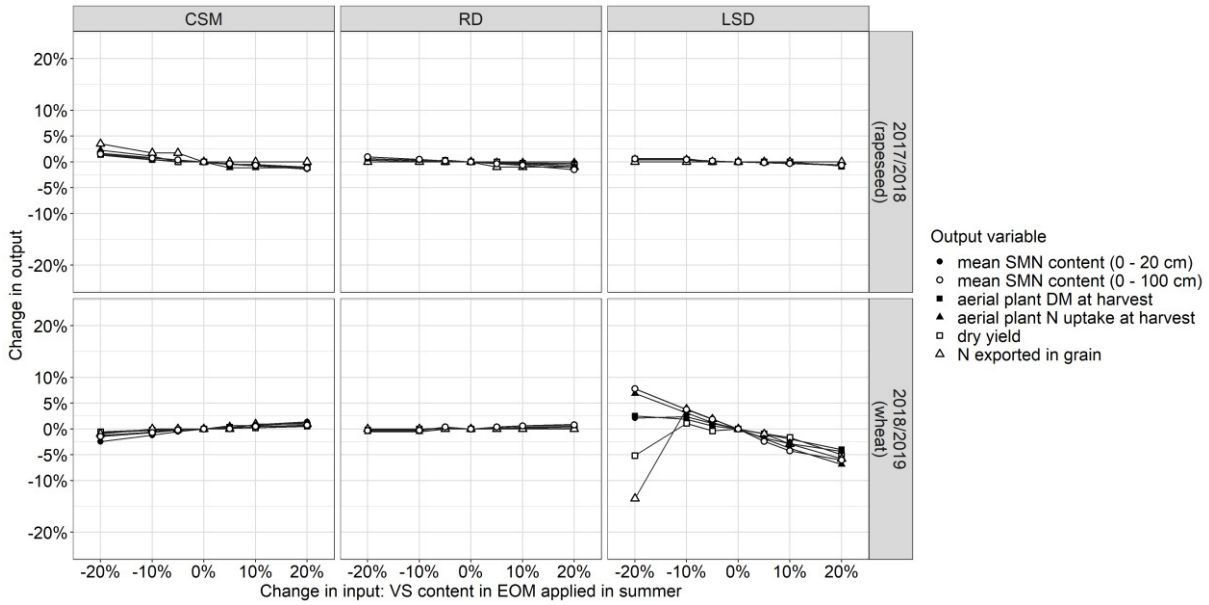
Supplementary Fig. S9.B – Relative change of STICS outputs induced by the variation of TAN content of EOM (summer) (2/2)



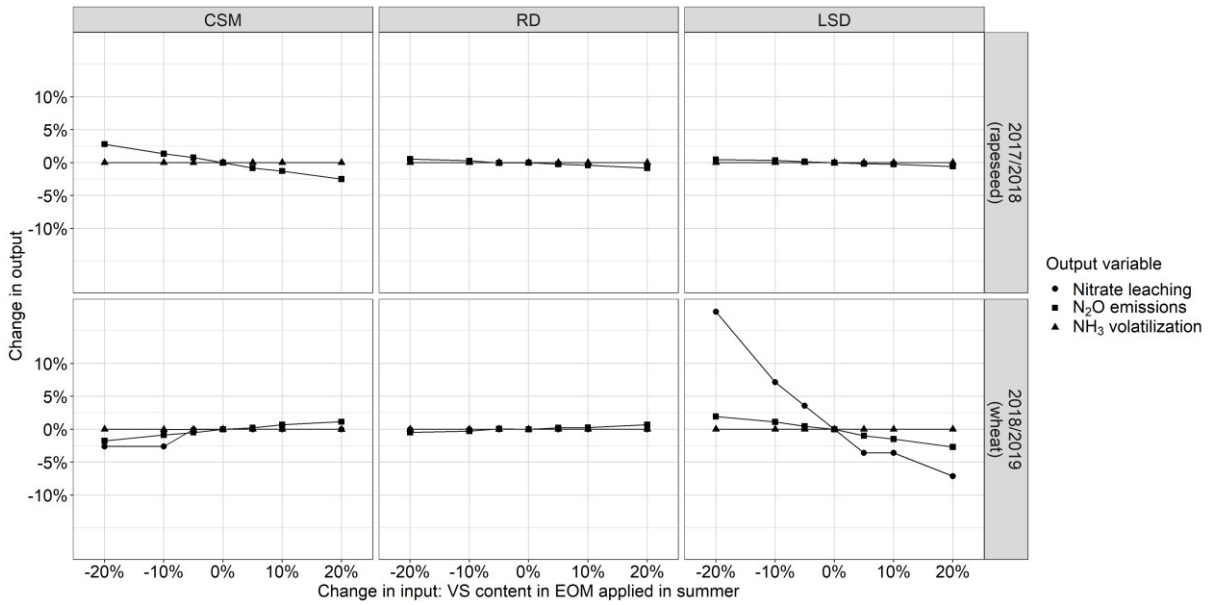
Supplementary Fig. S9.C – Relative change of STICS outputs induced by the variation of application date of EOM (summer) (1/2)



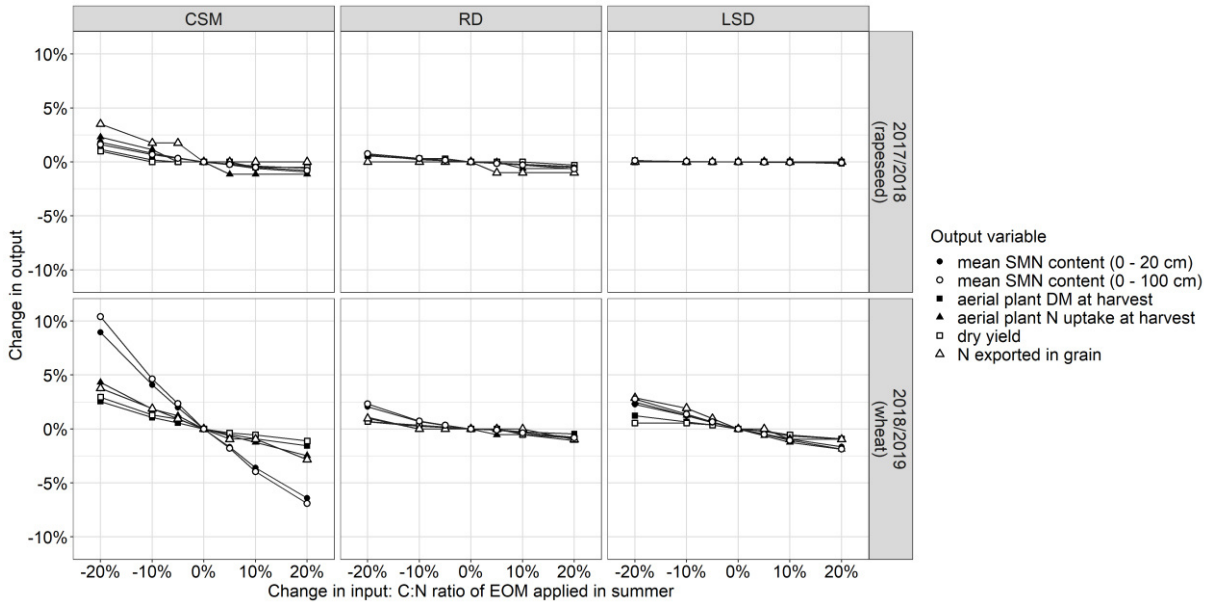
Supplementary Fig. S9.D – Relative change of STICS outputs induced by the variation of application date of EOM (summer) (2/2)



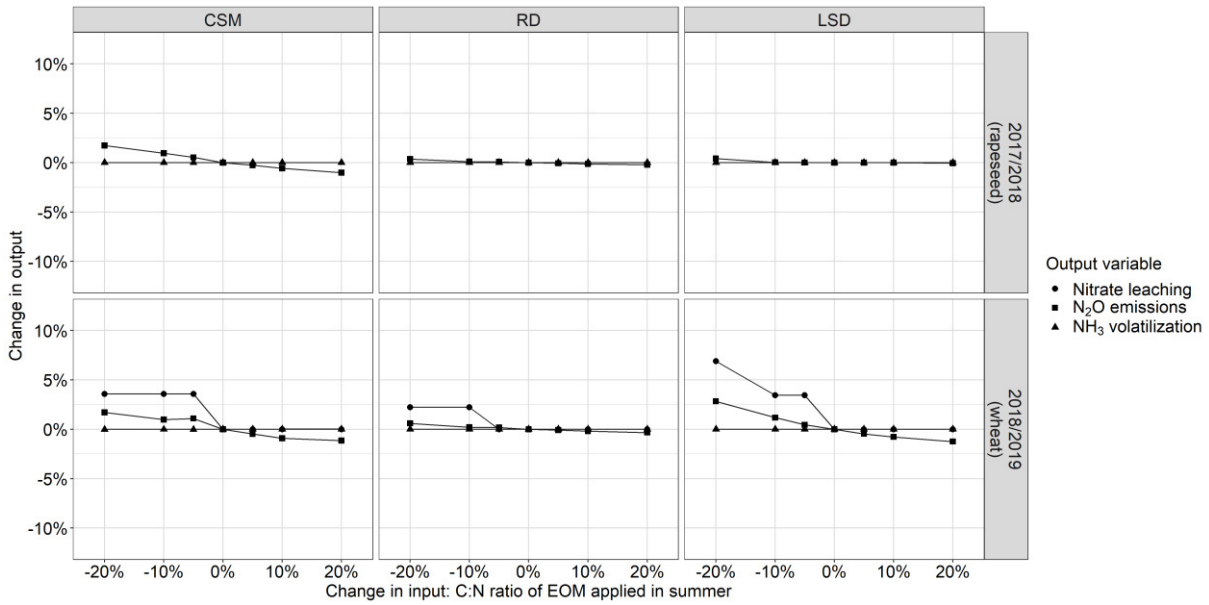
Supplementary Fig. S9.E – Relative change of STICS outputs induced by the variation of VS content of EOM (summer) (1/2)



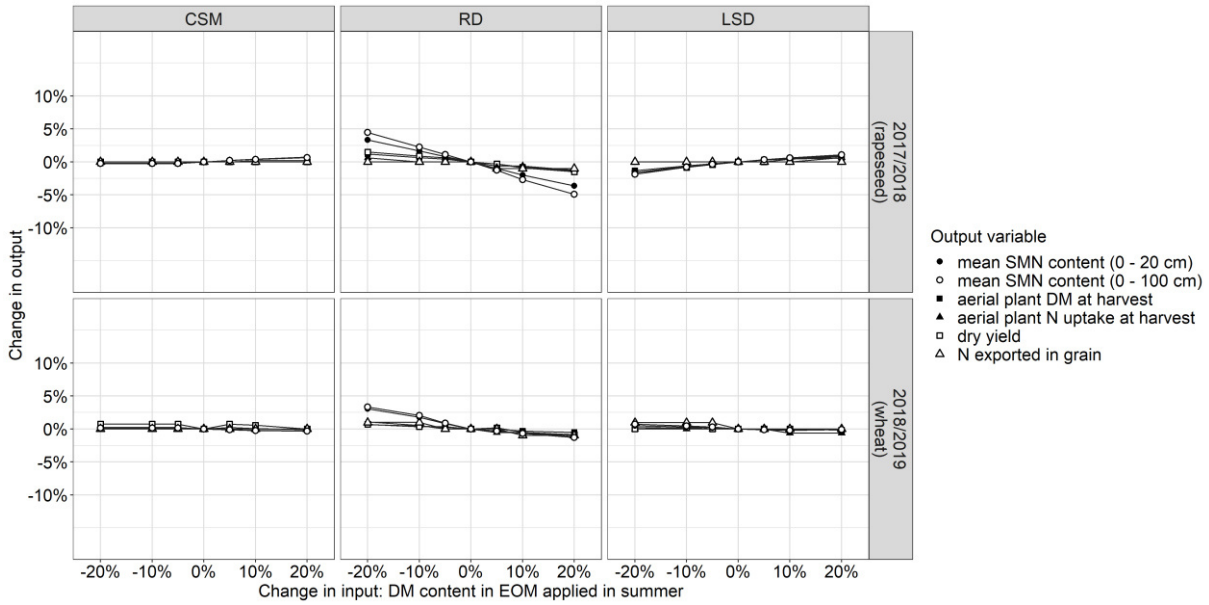
Supplementary Fig. S9.F – Relative change of STICS outputs induced by the variation of VS content of EOM (summer) (2/2)



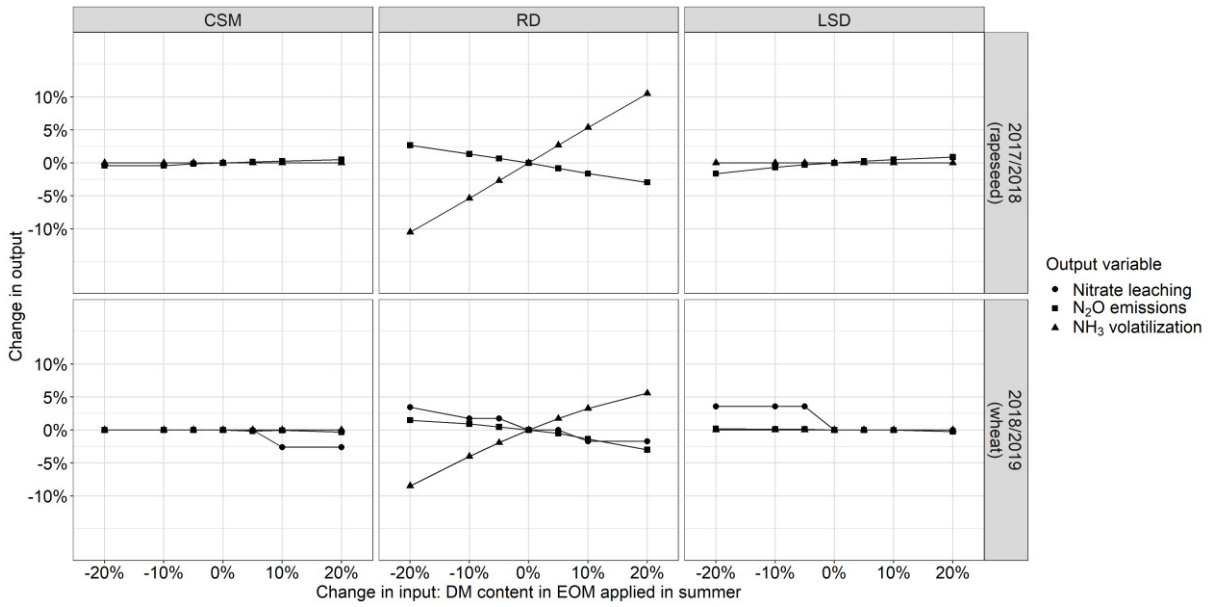
Supplementary Fig. S9.G – Relative change of STICS outputs induced by the variation of C:N_{org} ratio of EOM (summer) (1/2)



Supplementary Fig. S9.H – Relative change of STICS outputs induced by the variation of C:N_{org} ratio of EOM (summer) (2/2)



Supplementary Fig. S9.I – Relative change of STICS outputs induced by the variation of DM content of EOM (summer) (1/2)



Supplementary Fig. S9.J – Relative change of STICS outputs induced by the variation of DM content of EOM (summer) (2/2)

Annexes – Chapitre 6
Supplementary Material

S. M. 1 Emission factors used to model EOM storage

Supplementary Table S1 – Emission factors used to model EOM storage

	Cattle slurry in a covered pit	Cattle manure	Liquid digestate	Solid digestate	Raw digestate
Reference	Amon et al. (2006)	Hassouna et al. (2008)	SYS-Metha	SYS-Metha	SYS-Metha
Water loss ($kgH_2O\ kgH_2O^{-1}$)	0	0.34	0	0	0
TS loss ($kgTS\ kgTS^{-1}$)	*Mass balance*	*Mass balance*	0.08	0.22	0.05
TN loss ($kgTN\ kgTN^{-1}$)	0.18	0.28	*Mass balance*	0.09	*Mass balance*
TAN loss ($kgTAN\ kgTAN^{-1}$)	-0.16	0.47	0.09	0.51	0.07
VS loss ($kgVS\ kgVS^{-1}$)	0.44	0.34	0.13	0.30	0.07
C loss ($kgC\ kgC^{-1}$)	0.43	0.34	0.15	0.34	0.08
NH_3 emissions (% initial TAN)	2.2%	30%	9%	26%	7%
N_2O emissions (% initial TAN)	0*	13%	0	16%	0
CH_4 emissions	20% C_{loss}	7% C_{loss}	*Similar proportion of C_{loss} as in the biogas*	1% C_{loss}	*Similar proportion of C_{loss} as in the biogas*

* (hypothesis to be consistent with SYS-Metha)

Amon, B., Kryvoruchko, V., Amon, T., Zechmeister-Boltenstern, S., 2006. Methane, nitrous oxide and ammonia emissions during storage and after application of dairy cattle slurry and influence of slurry treatment. Agriculture, Ecosystems & Environment 112, 153–162. https://doi.org/10.1016/j.agee.2005.08.030

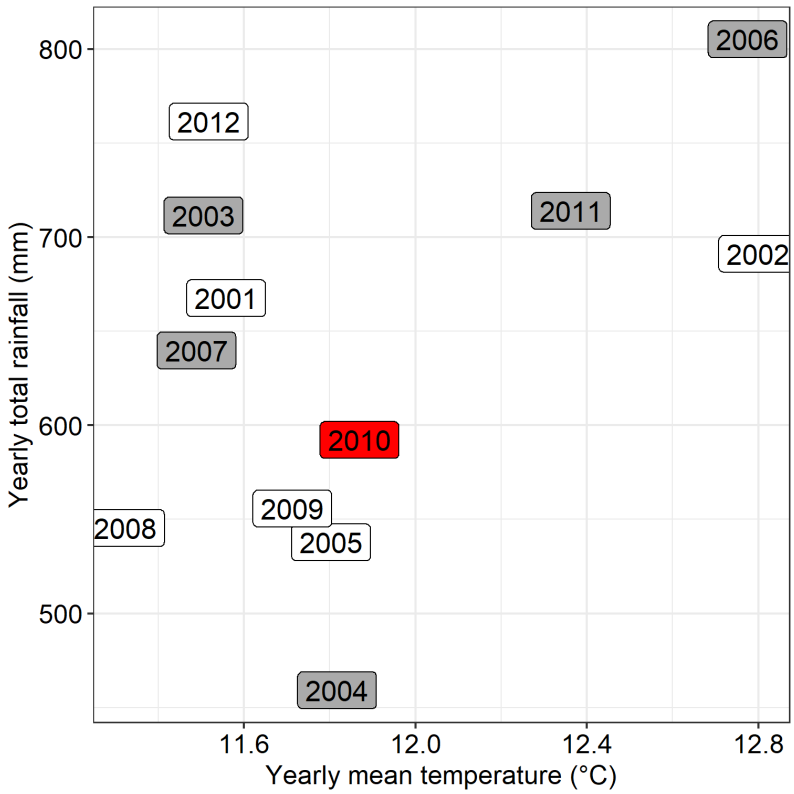
Hassouna, M., Espagnol, S., Robin, P., Paillat, J.-M., Levasseur, P., Li, Y., 2008. Monitoring NH_3, N_2O, CO_2 and CH_4 emissions during pig solid manure storage — effect of turning. Compost Science & Utilization 16, 267–274. https://doi.org/10.1080/1065657X.2008.10702388

S. M. 2 Parameters and options used in STICS

Supplementary Table S2 – The soil is described by 4 horizons. The first two horizons described a single horizon in the field, which was split to compare modelled soil characteristics to the one observed in the 0 – 20 cm depth.

Soil depth	Bulk density (g cm^{-3})	SWC at field capacity (% w/w)	SWC at wilting point (% w/w)
0 – 20 cm	1.37	26	8
20 – 25 cm	1.55	26	8
25 – 40 cm	1.55	23	10
40 – 60 cm	1.60	22	9
40 – 100 cm	1.60	21	15

S. M. 3 Characteristics of climatic years



Supplementary Figure S3 – Characteristics of the different climatic years. The simulated are shown in gray. The reference climatic year (2010) is shown in red.

S. M. 4 Cropping system

Supplementary Table S4.A – Modelled wheat management

Management	Date	Details	Used in STICS	Used in C footprint
Stubble ploughing	18 October (after wheat or maize). 16 September (after rapeseed)	Use of a disk harrow, 15 cm deep.	Yes	Yes
Soil preparation	20 October	Use of rotary harrow, 10 cm deep. Combined with sowing	Yes	Yes
Sowing	20 October	260 grain m^{-2} (151 kg ha^{-1}). 4 cm deep.	Yes	Yes
Plant protection		Three events.	No	Yes
First fertilization event	15 February (synthetic fertilizer), or 1 March (EOM)	Synthetic fertilizer (UAN solution), or EOM	Yes	Yes
Second fertilization event	15 March	Synthetic fertilizer (UAN solution), or EOM	Yes	Yes
Third fertilization event	15 May	Only with synthetic fertilizer (ammonium nitrate)	Yes	Yes
Harvesting	When grain moisture content is lower than 15%. Not after 30 July	Straws are exported. Grain is transported to storage.	Yes	Yes

Supplementary Table S4.B – Modelled maize management

Management	Date	Details	Used in STICS	Used in C footprint
Stubble ploughing	1 August (after wheat)	Use of a disk harrow, 15 cm. Only before mustard sowing (after wheat).	Yes	Yes
Mustard sowing	2 August (after wheat)	Cover crop. Only after wheat. 100 grains m^{-2} (7 kg ha^{-1}). Direct seeding.	Yes	Yes
Mustard crushing	30 November (after wheat)	Only after wheat.	Yes	Yes
Solid EOM application	31 March	Not mandatory	Yes	Yes
Ploughing	1 April	20 cm deep.	Yes	Yes
Soil preparation	30 April	Use of rotary harrow, 10 cm. Combined with sowing	Yes	Yes
Sowing	30 April	10 grain m^{-2} (32 kg ha^{-1}). 4 cm deep.	Yes	Yes
Plant protection		One event.	No	Yes
First fertilization event	1 May (synthetic fertilizer), or 29 April (EOM, before sowing)	Synthetic fertilizer (UAN solution), or EOM	Yes	Yes
Second fertilization event	21 May	Synthetic fertilizer (UAN solution)	Yes	Yes
Harvesting	16 September	Silage maize. Fodder is transported to storage. Maize is compacted in the silo.	Yes	Yes

Supplementary Table 4.C – Modelled rapeseed management

Management	Date	Details	Used in STICS	Used in C footprint
Solid EOM application	2 August	Not mandatory	Yes	Yes
Ploughing	3 August	20 cm deep.	Yes	Yes
Soil preparation	19 August	Tine stubble cultivator, 15 cm deep	Yes	Yes
Soil preparation	20 August	Use of rotary harrow, 10 cm deep. Combined with sowing	Yes	Yes
Sowing	20 August	40 grains m^{-2} (2 kg ha^{-1}). 2 cm deep.	Yes	Yes
Plant protection		Five events.	No	Yes
First fertilization event	15 February(synthetic fertilizer)	Synthetic fertilizer (UAN solution)	Yes	Yes
Second fertilization event	1 March	Synthetic fertilizer (UAN solution)	Yes	Yes
Third fertilization event	5 April	Synthetic fertilizer (ammonium nitrate). Not mandatory	Yes	Yes
Harvesting	When grain moisture content is lower than 8%. Not after 5 July	Grain is transported to storage.	Yes	Yes
Volunteer rapeseed		From harvest to stubble ploughing (wheat)	Yes	No

Supplementary Table 4.D – Simulated grassland management (3 years)

Management	Date	Details	Used in STICS	Used in C footprint
Soil preparation	31 August. First year only.	Tine stubble cultivator, 15 cm deep	Yes	Yes
Soil preparation	1 September. First year only.	Use of rotary harrow, 10 cm. Combined with sowing	Yes	Yes
Sowing	1 September. First year only.	950 grain m^{-2} (19 kg ha^{-1}).	Yes	Yes
Plant protection		One event in three years.	No	Yes
EOM application	15 September (not on the first year)	Not mandatory. Used if scenarios Y, or Y+I.	Yes	Yes
EOM application	5 November	Not mandatory. Used if scenarios Y, or Y+I.	Yes	Yes
First fertilization event	15 February	Synthetic fertilizer (UAN solution), or EOM	Yes	Yes
First cut	10 April		Yes	Yes
Second fertilization event	15 April	Synthetic fertilizer (UAN solution), or EOM	Yes	Yes
Second cut	15 June		Yes	Yes
EOM application	16 June	Not mandatory. Used if scenarios Y, C, Y+I, or C+I.	Yes	Yes
Destruction	1 April. Third year only	Destruction by ploughing, see maize management.	Yes	Taken into account in maize management

S. M. 5 Hypothesis used for C footprint computing

The hypotheses used to compute C footprint came from Base Carbone ® (ADEME, 2010) and GES'TIM+ (ARVALIS et al., 2021) databases.

The emissions from all machineries were computed considering fuel consumption (direct and indirect emissions), and machinery production. We assumed that all machinery consumed diesel fuel, which emitted 0.657 $kgCO_2eq\ L^{-1}$ (direct emissions) plus 2.51 $kgCO_2eq\ L^{-1}$ (indirect emissions) (GES'TIM+). The emissions caused by machinery production were estimated with the total emissions of machinery production, adjusted in function of total lifetime and total time of use of the machinery. The production of the machinery was estimated to emit 5.5 tCO_2eq per t of machinery (Base Carbone). Finally, the total GHG emissions of one operation (kg $CO_2eq\ ha^{-1}$) were computed as:

$$Operation\ GHG\ emissions = consumption * (0.657 + 2.51) + operation\ time * \sum_i \frac{tool\ mass_i * 5.5}{tool\ lifetime_i}$$

Where consumption is the fuel consumption of the operation (L ha^{-1}), operation time is the duration of the operation (h ha^{-1}), i indicate each tool implicated in the operation, tool $mass_i$ is the mass of tool i (kg), and tool $lifetime_i$ is the lifetime of tool i (h). Consumption, duration, and tools implicated in each operation are summarized in Supplementary Table S5.A.

The GHG emissions from seeds production was estimated from the mass of seeds, and was 0.694 $kgCO_2eq\ kg^{-1}$ (wheat), 1.841 $kgCO_2eq\ kg^{-1}$ (rapeseed), 0.607 $kgCO_2eq\ kg^{-1}$ (mustard, hypotheses: similar to spring barley), 1.249 $kgCO_2eq\ kg^{-1}$ (maize), and 0.607 $kgCO_2eq\ kg^{-1}$ (grassland, hypotheses: similar to spring barley) (GES'TIM+).

The GHG emissions from plant protection products were 9.861 $kgCO_2eq$ per kg product (GES'TIM+). We assumed that on average, 3.0 kg ha^{-1} $year^{-1}$ of active substance of plant protection products were applied. The figure came from plant protection product sell data, on the city of Nouzilly, from 2015 to 2018 (Office Français de la Biodiversité, http://www.data.eaufrance.fr/jdd/a69c8e76-13e1-4f87-9f9d-1705468b7221, accessed August 22, 2021).

Supplementary Table S5.A – Characteristics used to compute GHG emissions of different cropping operations. Data come from GES'TIM+ database.

Operation	Fuel consumption (L ha^{-1})	Operation duration (h ha^{-1})	Tool	Tool mass (t)	Tool lifetime (h)
Soil preparation, disk harrow	10	0.5	Tractor	12	6440
			Harrow	2.1	3000
Ploughing	25	1.33	Tractor	12	6440
			Plough	3	1000
Soil preparation, tine stubble	20.83	0.83	Tractor	12	6440
			Harrow	1.57	1029
Sowing, combined with rotary	14.03	0.83	Tractor	12	6440
			Planter	1.2	1000
			Harrow	1.8	1380
Sowing, combined with rotary	14.03	0.83	Tractor	10	5300
			Planter	1.2	1000
			Harrow	1.8	1380
Mustard sowing, direct seeding	6	0.42	Tractor	10	5300
			Planter	1.2	5300
Liquid EOM application	5	1	Tractor	10	5300
			Spreader	5	1000
Solid EOM application	11.6	1.33	Tractor	10	5300
			Spreader	5	3500
UAN solution application	1.04	0.08	Tractor	7.2	3000
			Sprayer	3	2600
Application of ammonium nitrate	1.57	0.12	Spreader	7.2	3000
Application of plant protection	1.04	0.08	Tractor and	7.2	3000
				3	2600
Mustard crushing	10	0.5	Tractor	10	5300
			Crusher	2.3	2100
Harvest (Wheat, rapessed)	17.07	0.67	Harvester	4	10000
Fodder harvest (maize)	42	0.5	Silage	4	9000
Grassland cutting	11.7	0.9	Tractor	10	5300
			Bale presser	1.8	1773
Straw bale making (wheat)	11.7	0.9	Tractor	10	5300
			Bale presser	1.8	1773
Grain transport (wheat, rapessed)	8.94	0.67	Tractor	12	6440
			Wagon	6	5883
Fodder transport (maize)	15.13	2.5		7.2	3000
Silo compacting (maize)	7.0	0.61		/	/

Supplementary Table S5.B – P and K contents of EOM. For digestates, the total amounts of P and K were computed based and raw digestate characteristics and quantities, before phase separation.

EOM	P_2O_5 (g $kgDM^{-1}$)	K_2O (g $kgDM^{-1}$)	Reference
Cattle slurry (spread)	16	62	Nouzilly case study (Chapitre 3)
Cattle solid manure (spread)	15	42	Nouzilly case study (Chapitre 3)
Raw digestate, before phase separation and storage. All scenarios but not EI0, EI30A, EI70A	30	60	Guilayn et al. (2019) Type 3 digestate (digestate from urban wastes, agro-industrial wastes, and slurry).
Raw digestate, before phase separation and storage. Scenarios EI0, EI30A, EI70A	23	60	Guilayn et al. (2019) Type 1 digestate (digestate from fibrous feedstock).
Liquid urban sludge (spread)	82	16	Houot et al. (2014)
Solid Urban sludge (spread)	51	5	Houot et al. (2014)

ADEME, (Agence de l'environnement et de la maîtrise de l'énergie), 2021. Base Carbone ®.

ARVALIS Institut du Végétal, IDELE, CTIFL, IFIP, IFV, ITAVI, Terres Inovia, 2021. GES'TIM+.

Guilayn, F., Jimenez, J., Martel, J.-L., Rouez, M., Crest, M., Patureau, D., 2019. First fertilizing-value typology of digestates: A decision-making tool for regulation. Waste Management 86, 67–79. https://doi.org/10.1016/j.wasman.2019.01.032

Houot, S., Pons, M.-N., Pradel, M., Tibi, A., Aubry, C., Augusto, L., Barbier, R., Benoit, P., Brugère, H., Caillaud, M.-A., Casellas, M., Chatelet, A., Dabert, P., De Mareschal, S., Doussan, I., Etrillard, C., Fuchs, J., Génermont, S., Giamberini, L., Hélias, A., Jardé, E., Le Perchec, S., Lupton, S., Marron, N., Ménasseri, S., Mollier, A., Morel, C., Mouguin, C., Nguyen, C., Parnaudeau, V., Patureau, D., Pourcher, A.-M., Rychen, G., Savini, I., Smolders, E., Topp, E., Vieublé, L., Viguié, C., 2014. Valorisation des matières fertilisantes d'origine résiduaire sur les sols à usage agricole ou forestier, impacts agronomiques, environnementaux, socio-économiques (Expertise scientifique collective, rapport, INRA-CNRS-Irstea). France.

S. M. 6 Modelling sewage sludge application at the farm scale

We studied the application of sludge with the simulation of a dedicated type of farm. For each climatic year, we modelled three farms: one using solid sludge, one using liquid sludge, one using no EOM. These farms were composed of nine fields of 30 ha each. The crop rotation was rapeseed – wheat – wheat. In other terms, the farm total area was 270 ha, including 180 ha of wheat (67%) and 90 ha of rapeseed (33%). Agronomical management of wheat and rapeseed corresponded to the one described in Supplementary Material 4, with no EOM application. According to local experts, sludge were applied in the fields in summer or autumn, before a cereal or before rapeseed. All sewage sludge applications occurred on 2 August. Sewage sludge are buried 1 h after application. On the farm using solid sludge, 900 tons of sludge were spread every year. It consisted in an application on 60 ha before wheat (10 t ha^{-1}), and an application on 30 ha before rapeseed (10 t ha^{-1}). On the farm using liquid sludge, 1500 tons of sludge were spread every year. It consisted in an application on 30 ha before wheat (20 t ha^{-1}), an application on 30 ha before wheat (15 t ha^{-1}), and an application on 30 ha before rapeseed (15 t ha^{-1}). The emission factors used to simulate the covered storage of solid sewage sludge came from Willén et al. (2016b), and Willén et al. (2017). We assumed that the emission factors used to simulate the covered storage of liquid sewage sludge storage were the same as the one measured on cattle slurry by Amon et al. (2006). They are described in Supplementary Table 6.A. The sludge characteristics came from SYS-Metha model. Only pH came from Houot et al. (2014). Sludge characteristics were described in Supplementary Table 6.B and Supplementary Table 5.B. Ammonia volatilization after field application of liquid sludge was modelled with ALFAM2 model. Ammonia volatilization after field application of liquid sludge was modelled using an emission factor of 0.22 $kgNH_3$-N $kgTAN^{-1}$ (Average emission factors for burying times of 0 h and 4 h, Willén et al., 2016a).

The total C footprints of each of the three farms were computed similarly to the main simulated farm, as described in the article. The C footprint of the spreading of 900 t of solid sludge (1500 t of liquid sludge, respectively) was computed as the difference of GHG emissions of the farm using solid sludge (liquid sludge, respectively), and the farm using no sludge. This C footprint was computed for a similar cropped area, and a similar crop production (tDM). The GHG emission of the alternative sludge treatment was then computed proportionally to the amount of treated sludge.

Supplementary Table S6.A – Emission factors used to simulate the storage of sewage sludge.

Reference	Liquid sludge	Solid sludge
Water loss ($kgH_2O\ kgH_2O^{-1}$)	0	*mass balance*
TS loss ($kgTS\ kgTS^{-1}$)	*mass balance*	0.12
TN loss ($kgN\ kgN^{-1}$)	0.18	*mass balance*
TAN loss ($kgTAN\ kgTAN^{-1}$)	-0.16	-0.2
VS loss ($kgVS\ kgVS^{-1}$)	0.44	0.25
C loss ($kgC\ kgC^{-1}$)	0.43	0.25
NH_3 emissions	2.2% initial TAN	100 gNH_3-N kgN^{-1}
N_2O emissions	0 (hypothesis to be consistent with SYS-Metha)	3.4 gN_2O-N kgN^{-1}
CH_4 emissions	20% C_{loss}	11 gCH_4-C kgC^{-1}

Supplementary Table S6.B – Sewage sludge characteristics used. FW: fresh weight.

EOM	FW (t)	TS (%FW)	VS (g $kgFW^{-1}$)	C (g $kgFW^{-1}$)	TN (g $kgFW^{-1}$)	TAN (g $kgFW^{-1}$)	pH
Liquid sludge, fresh	1523	4.3%	35	19.5	2.6	0.15	
Solid sludge, fresh	920	18%	137	72	13.9	0.18	
Liquid sludge, stored	1500	2.8%	20	11	2.16	0.18	7.0
Solid sludge, stored	900	16.2%	105	55	12.74	0.22	7.1

Amon, B., Kryvoruchko, V., Amon, T., Zechmeister-Boltenstern, S., 2006. Methane, nitrous oxide and ammonia emissions during storage and after application of dairy cattle slurry and influence of slurry treatment. Agriculture, Ecosystems & Environment 112, 153–162. https://doi.org/10.1016/j.agee.2005.08.030

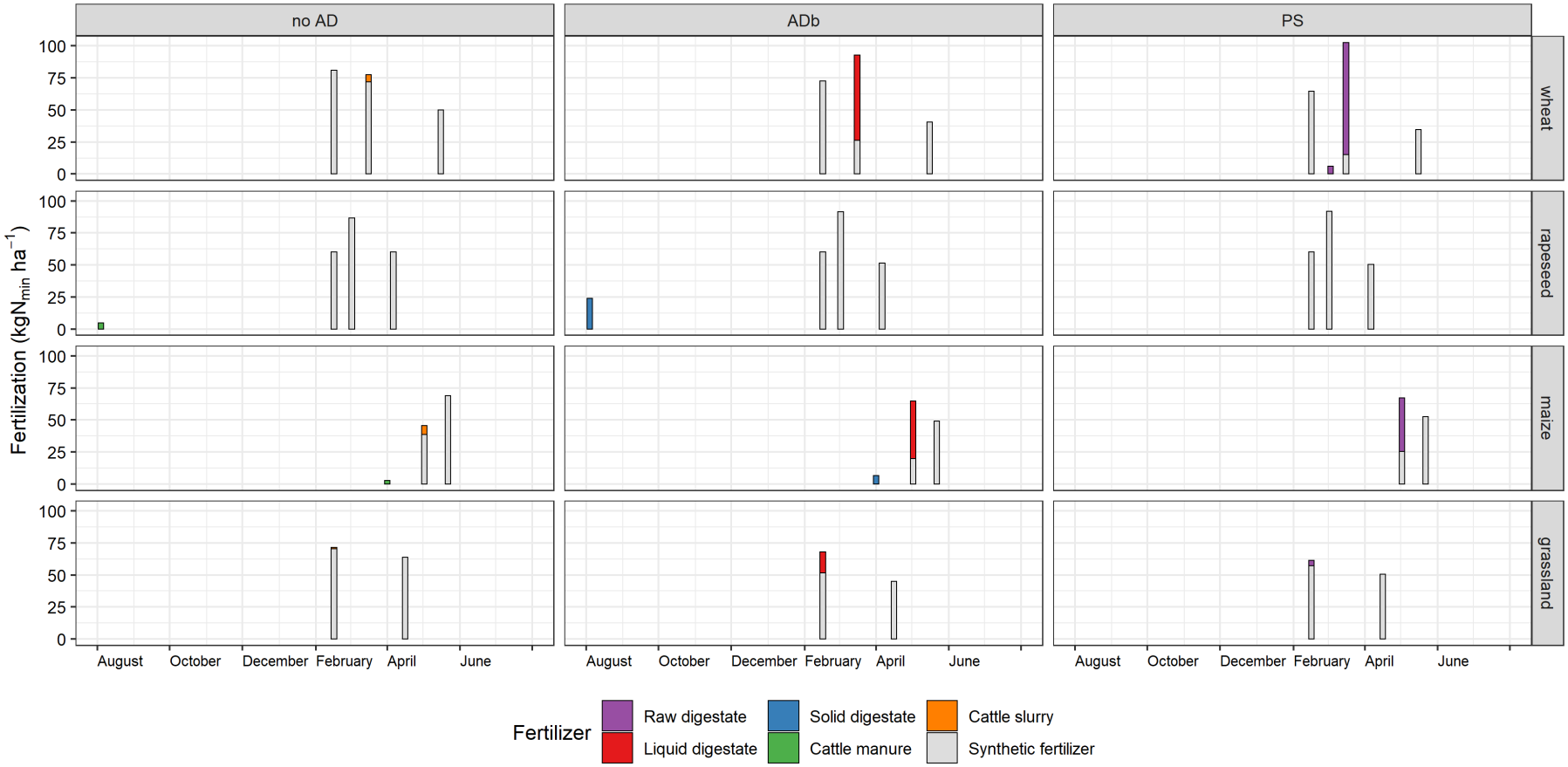
Houot, S., Pons, M.-N., Pradel, M., Tibi, A., Aubry, C., Augusto, L., Barbier, R., Benoit, P., Brugère, H., Caillaud, M.-A., Casellas, M., Chatelet, A., Dabert, P., De Mareschal, S., Doussan, I., Etrillard, C., Fuchs, J., Génermont, S., Giamberini, L., Hélias, A., Jardé, E., Le Perchec, S., Lupton, S., Marron, N., Ménasseri, S., Mollier, A., Morel, C., Mouguin, C., Nguyen, C., Parnaudeau, V., Patureau, D., Pourcher, A.-M., Rychen, G., Savini, I., Smolders, E., Topp, E., Vieublé, L., Viguié, C., 2014. Valorisation des matières fertilisantes d'origine résiduaire sur les sols à usage agricole ou forestier, impacts agronomiques, environnementaux, socio-économiques (Expertise scientifique collective, rapport, INRA-CNRS-Irstea). France.

Willén, A., Jönsson, H., Pell, M., Rodhe, L., 2016a. Emissions of nitrous oxide, methane and ammonia after field application of digested and dewatered sewage sludge with or without addition of urea. Waste Biomass Valor 7, 281–292. https://doi.org/10.1007/s12649-015-9456-2

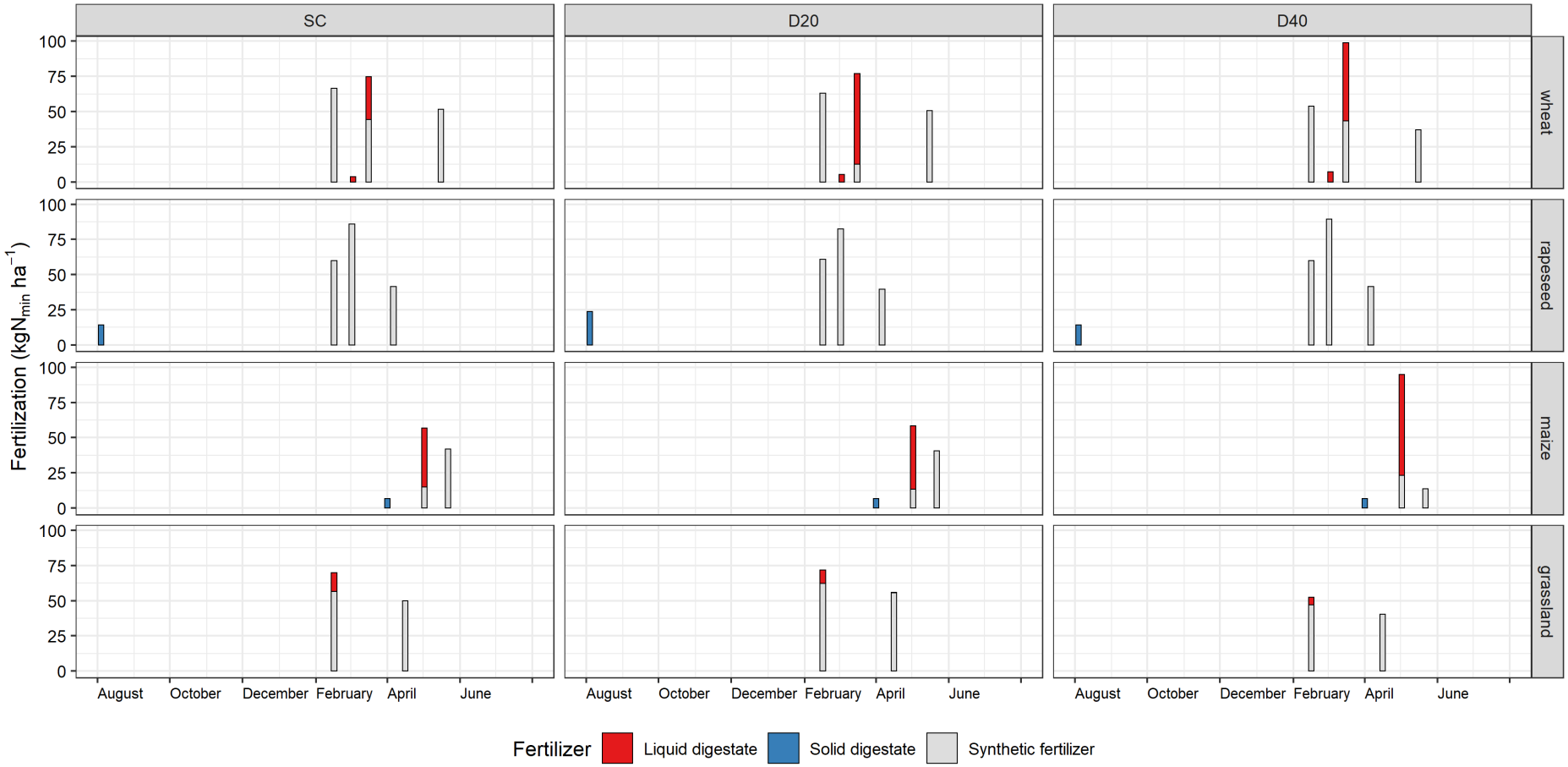
Willén, A., Junestedt, C., Rodhe, L., Pell, M., Jönsson, H., 2017. Sewage sludge as fertiliser – environmental assessment of storage and land application options. Water Science 17.

Willén, A., Rodhe, L., Pell, M., Jönsson, H., 2016b. Nitrous oxide and methane emissions during storage of dewatered digested sewage sludge. Journal of Environmental Management 184, 560–568. https://doi.org/10.1016/j.jenvman.2016.10.025

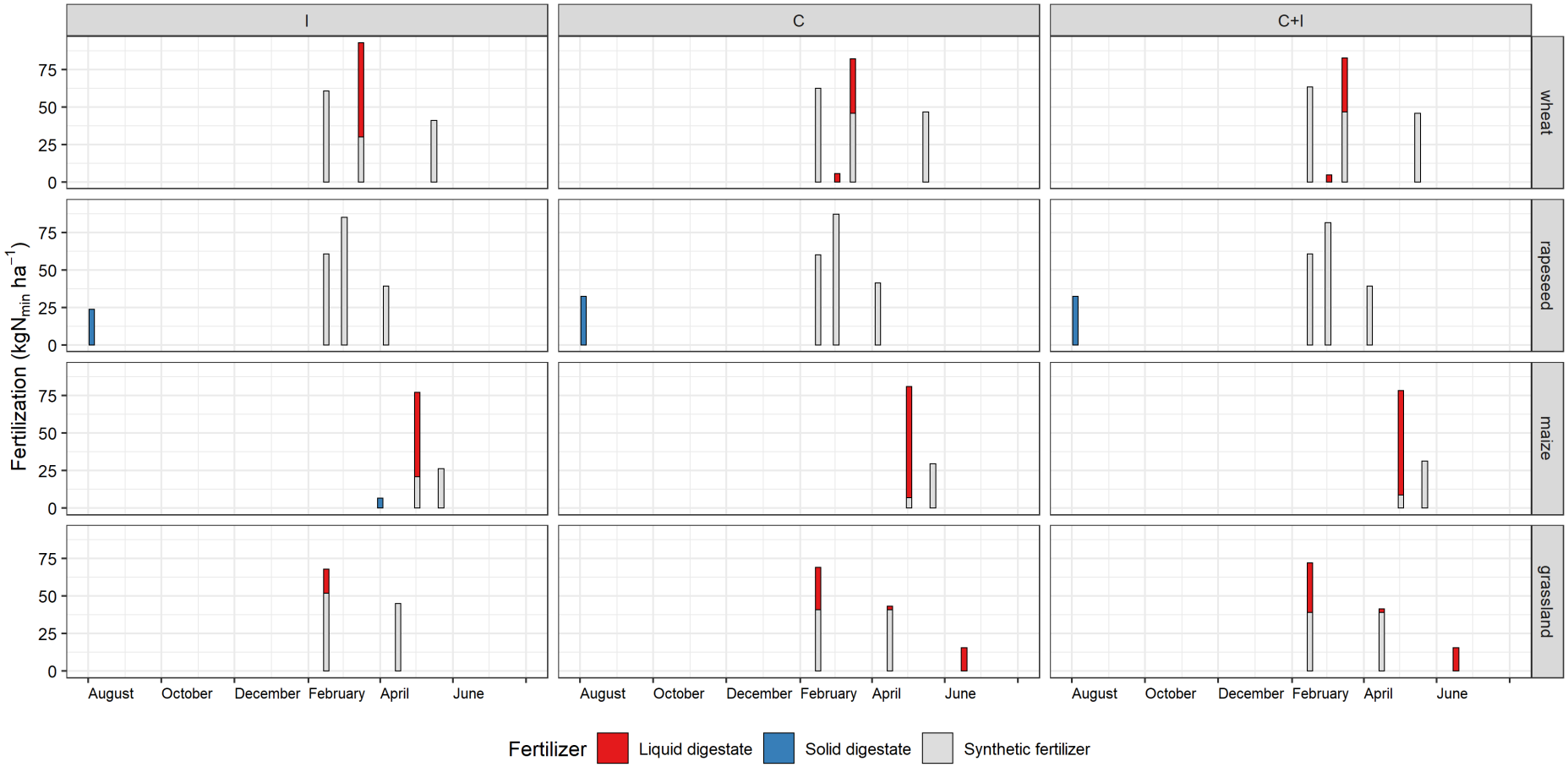
S.M. 7 Modelled fertilization on each crop



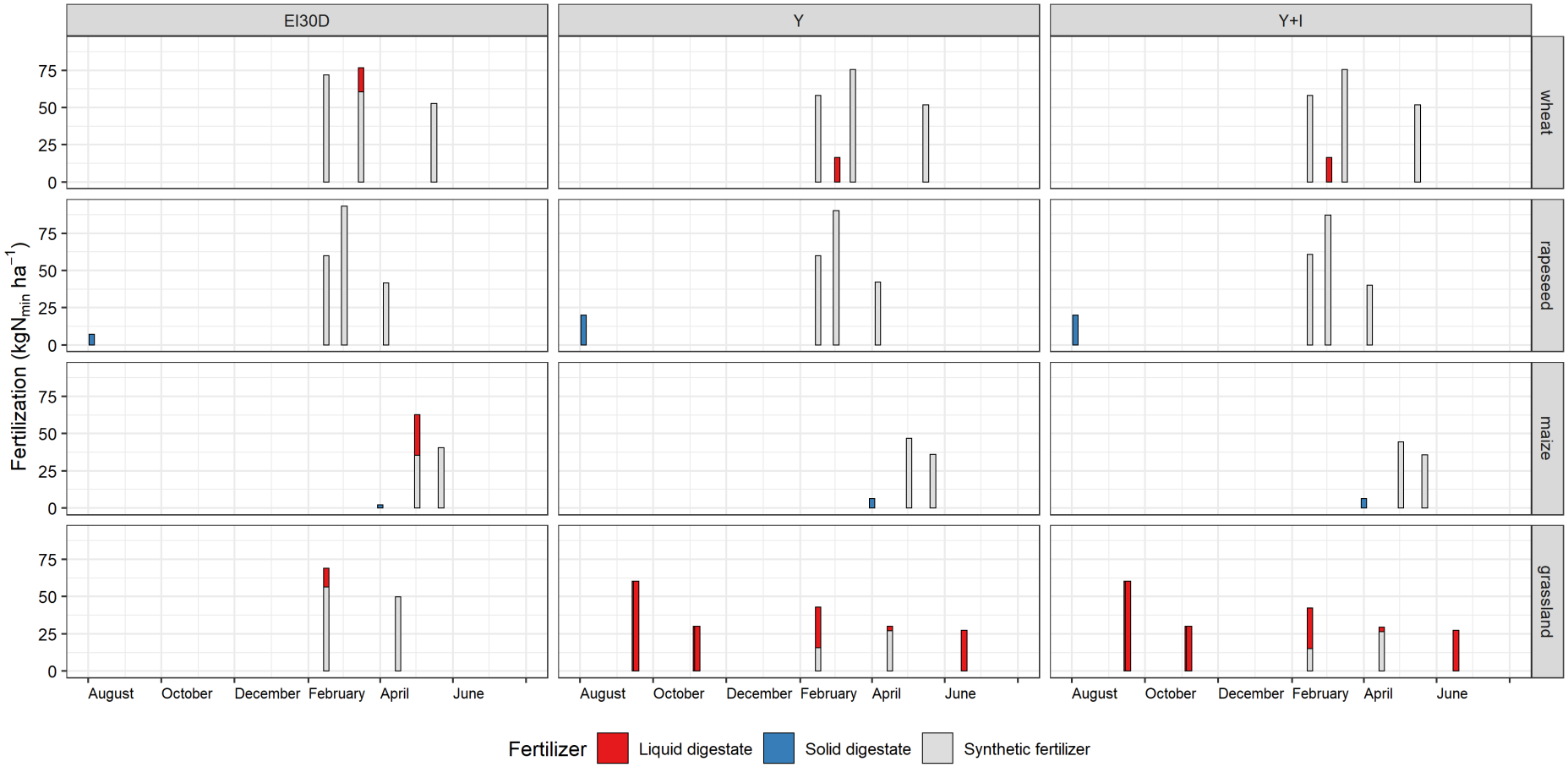
Supplementary Figure S7.A – Average fertilization of each crops in different scenario. Climatic year is 2010. (1/5)



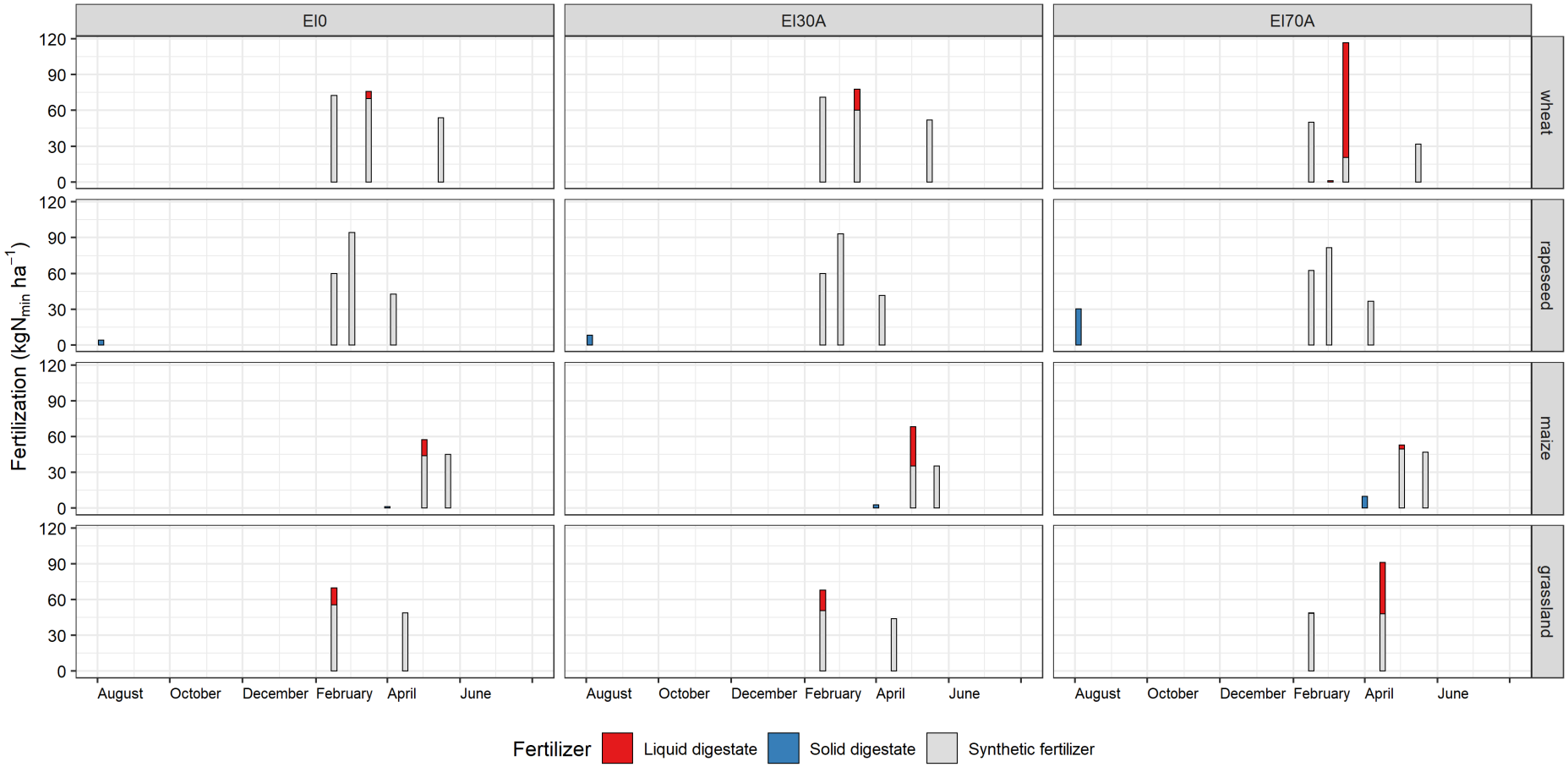
Supplementary Figure S7.B – Average fertilization of each crops in different scenario. Climatic year is 2010. (2/5)



Supplementary Figure S7.C – Average fertilization of each crops in different scenario. Climatic year is 2010. (3/5)

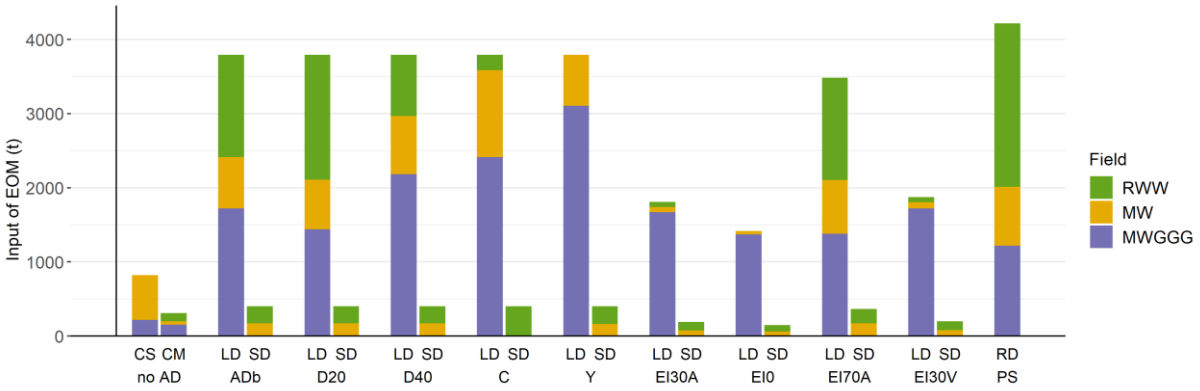


Supplementary Figure S7.D – Average fertilization of each crops in different scenario. Climatic year is 2010. (4/5)

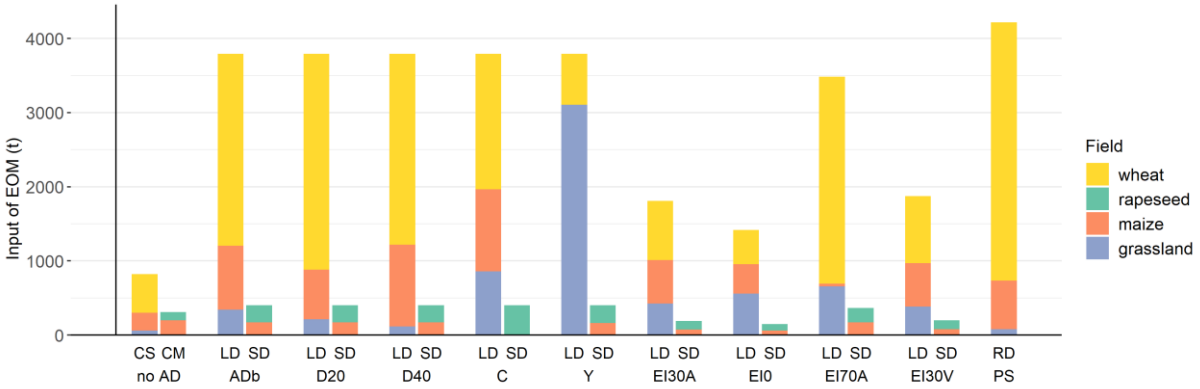


Supplementary Figure S7.E – Average fertilization of each crops in different scenario. Climatic year is 2010. (5/5)

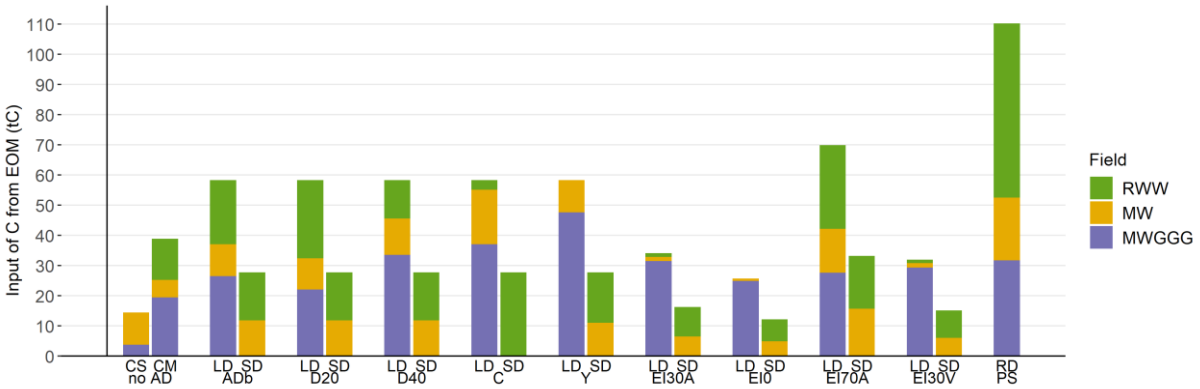
S.M. 8 Repartition of the use of EOMs in the different fields



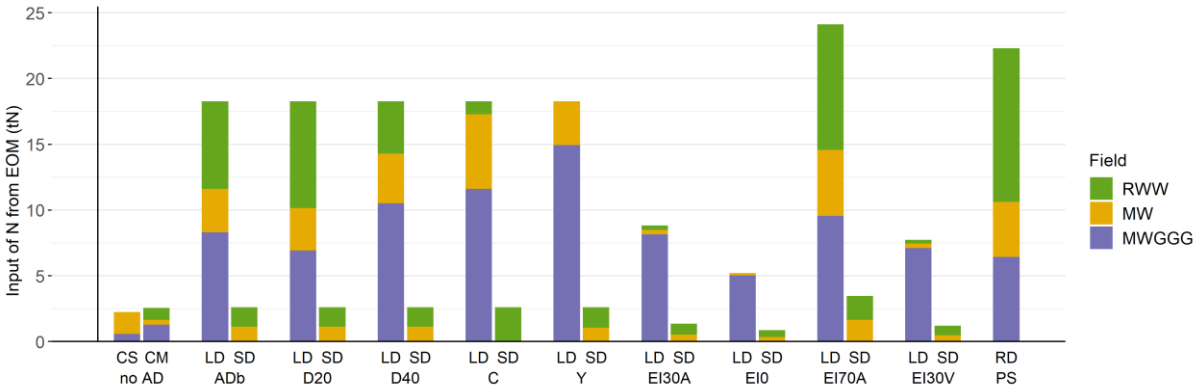
Supplementary Figure S8.A – Average use of EOM across the different fields, during the 20 years, for all scenarios (tFW). Climatic year is 2010. Color indicate the crop rotation on which the EOM is applied. RWW: Rapeseed – Wheat – Wheat. MW: Maize – Wheat. MWGGG: Maize – Wheat – 3-year grassland. CS: cattle slurry. CM: cattle manure. LD: liquid digestate. SD: solid digestate. RD: raw digestate.



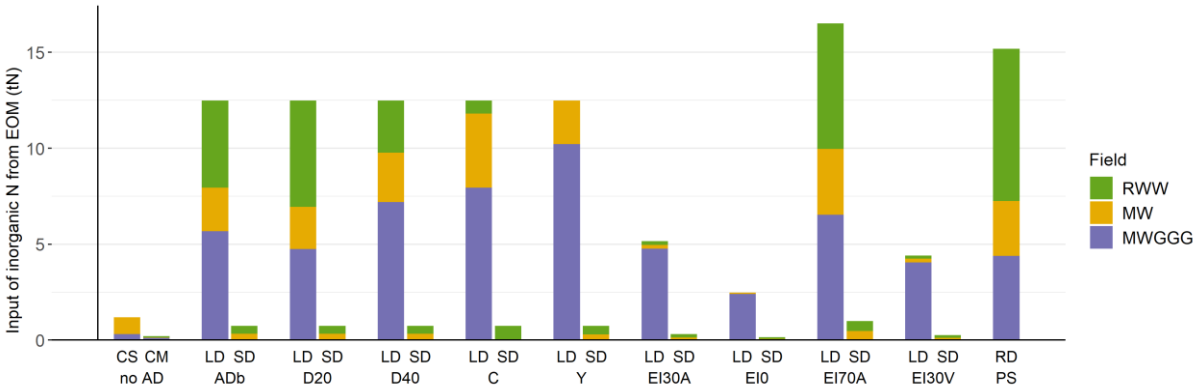
Supplementary Figure S8.B – Average use of EOM across the different fields, during the 20 years, for all scenarios (tFW). Climatic year is 2010. Color indicate the crop on which the EOM is applied. CS: cattle slurry. CM: cattle manure. LD: liquid digestate. SD: solid digestate. RD: raw digestate.



Supplementary Figure S8.C – Average use of EOM across the different fields, during the 20 years, for all scenarios (tC). Climatic year is 2010. Color indicate the crop rotation on which the EOM is applied. RWW: Rapeseed – Wheat – Wheat. MW: Maize – Wheat. MWGGG: Maize – Wheat – 3-year grassland. CS: cattle slurry. CM: cattle manure. LD: liquid digestate. SD: solid digestate. RD: raw digestate.

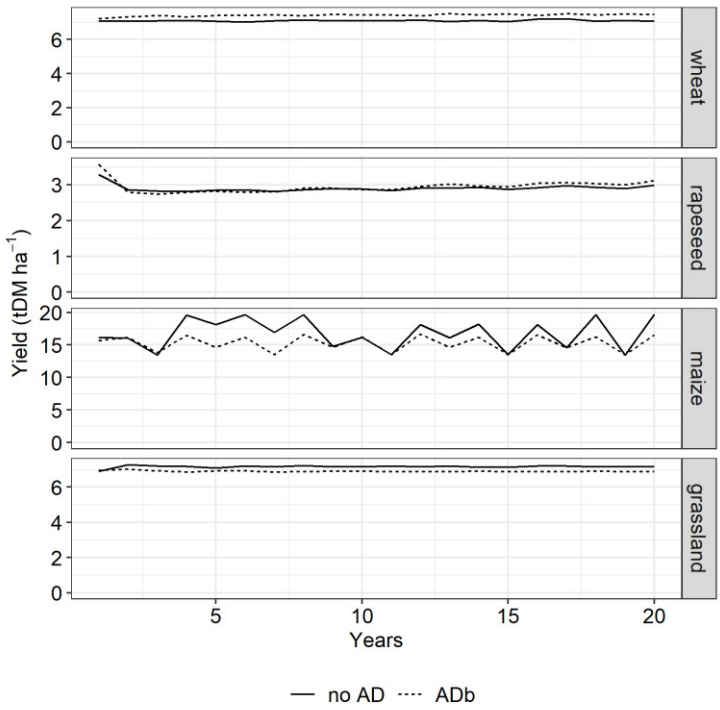


Supplementary Figure S8.D – Average use of EOM across the different fields, during the 20 years, for all scenarios (tN). Climatic year is 2010. Color indicate the crop rotation on which the EOM is applied. RWW: Rapeseed – Wheat – Wheat. MW: Maize – Wheat. MWGGG: Maize – Wheat – 3-year grassland. CS: cattle slurry. CM: cattle manure. LD: liquid digestate. SD: solid digestate. RD: raw digestate.

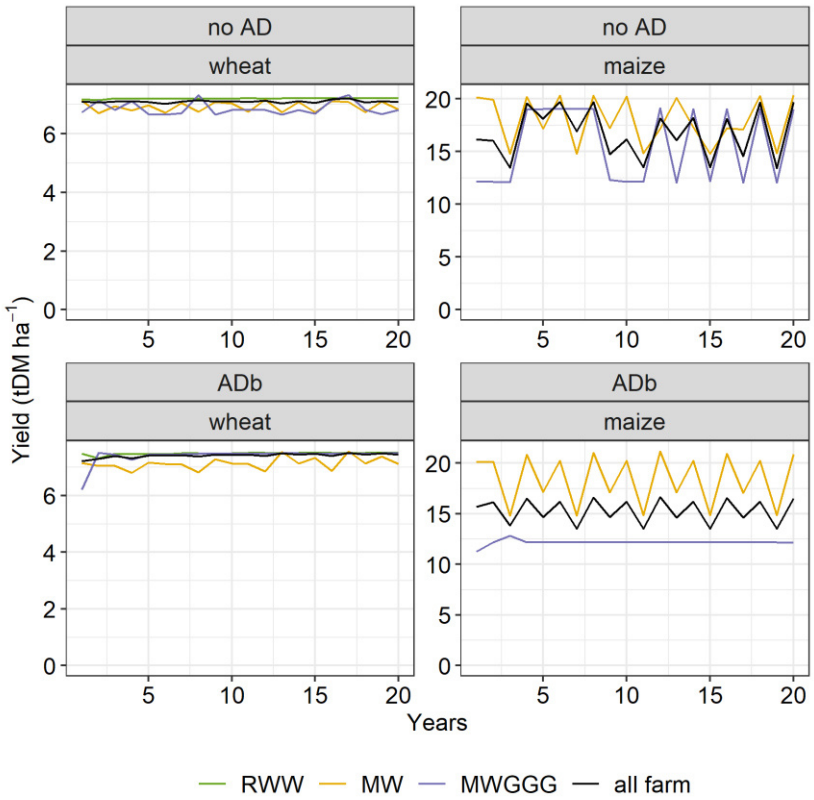


Supplementary Figure S8.E – Average use of EOM across the different fields, during the 20 years, for all scenarios (inorganic N, tN). Climatic year is 2010. Color indicate the crop rotation on which the EOM is applied. RWW: Rapeseed – Wheat – Wheat. MW: Maize – Wheat. MWGGG: Maize – Wheat – 3-year grassland. CS: cattle slurry. CM: cattle manure. LD: liquid digestate. SD: solid digestate. RD: raw digestate.

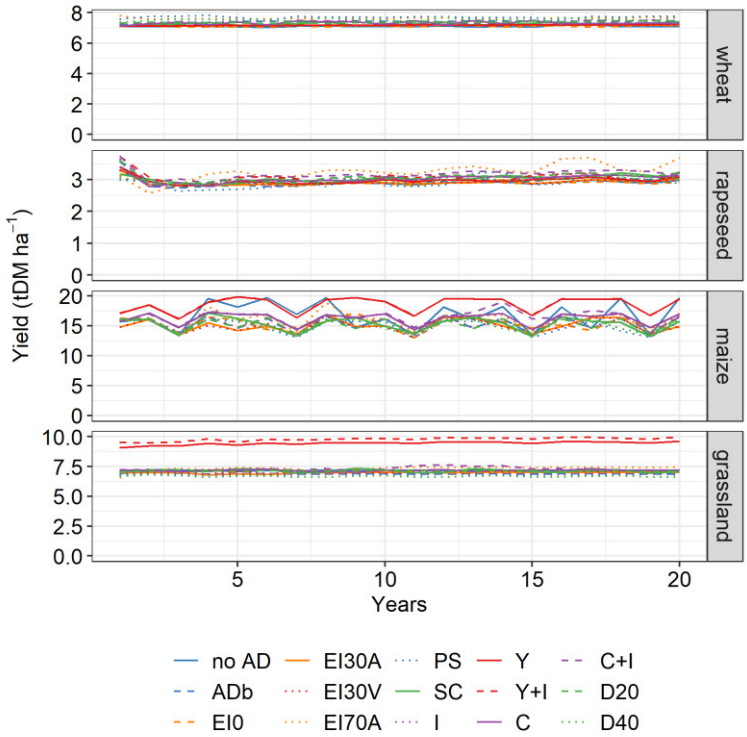
S.M. 9 Yield evolution



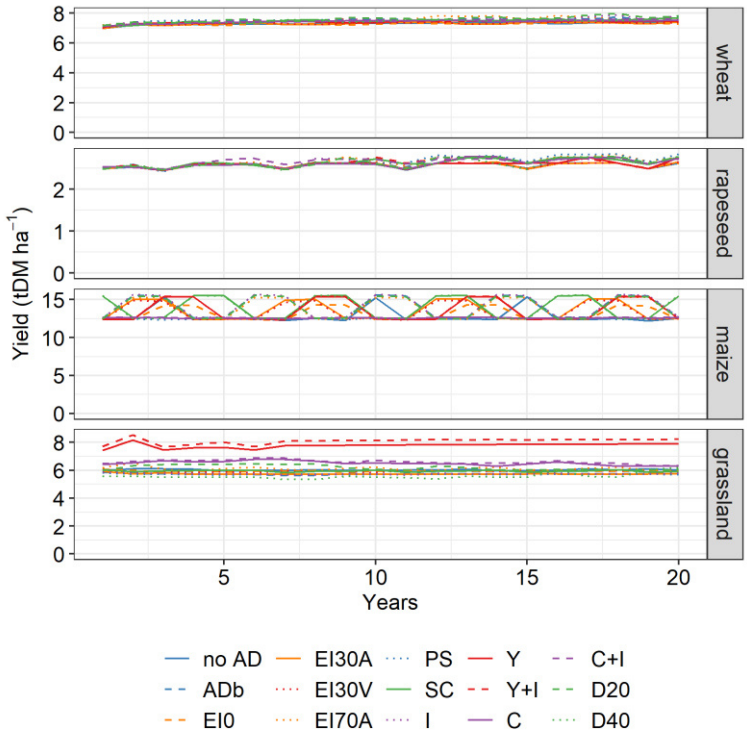
Supplementary Figure S9.A – Evolution of the yield of each crop through the time, for ADb and "no AD" scenarios. Climatic year is 2010.



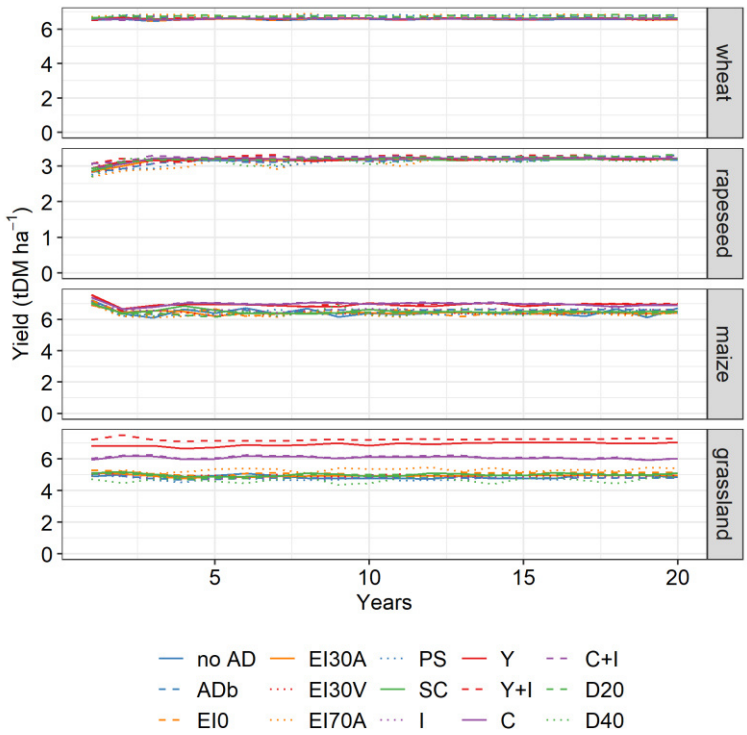
Supplementary Figure S9.B – Evolution of the yield of each crop through the time in function of the crop rotation, for ADb and "no AD" scenarios. Climatic year is 2010.



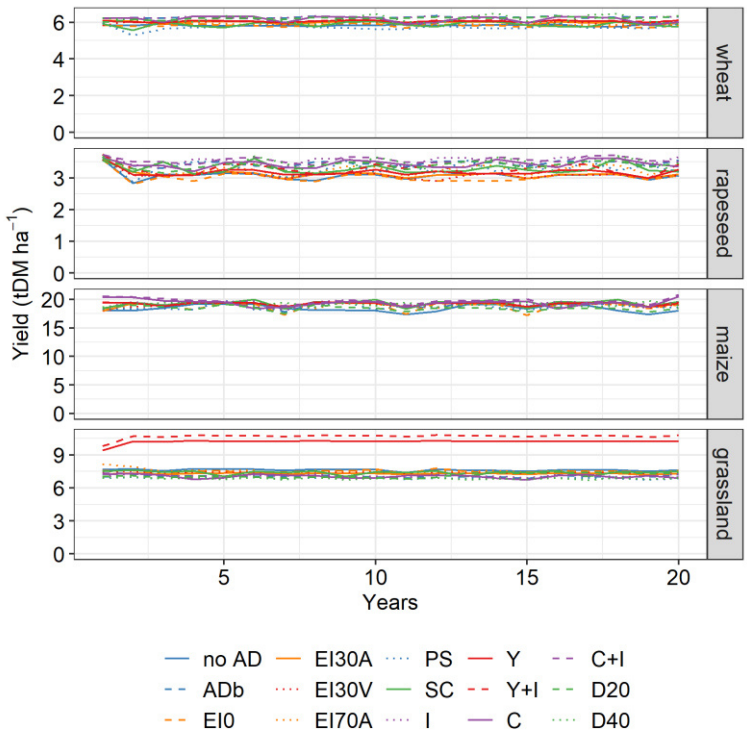
Supplementary Figure S9.C – Evolution of the yield of each crop through the time, for all scenarios. Climatic year is 2010.



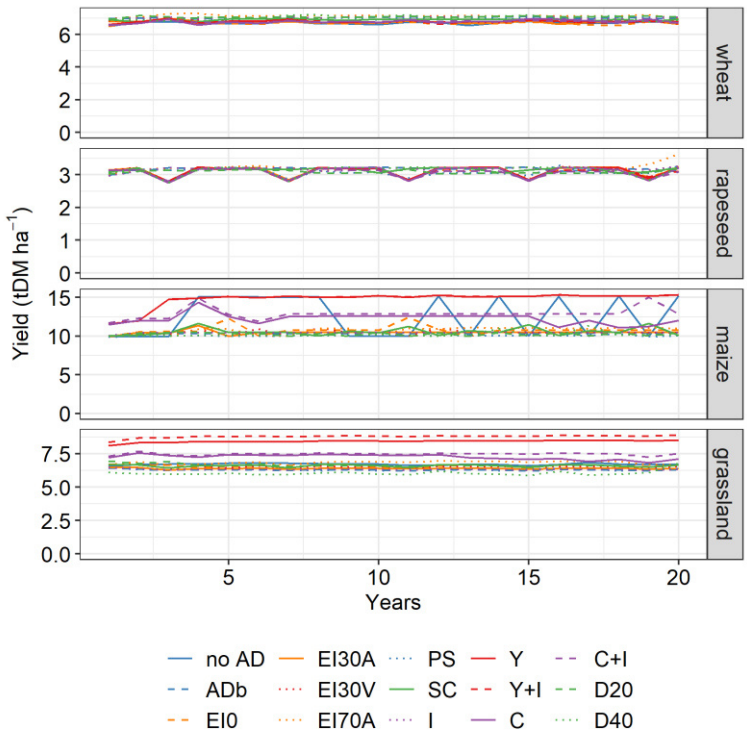
Supplementary Figure S9.D – Evolution of the yield of each crop through the time, for all scenarios. Climatic year is 2003.



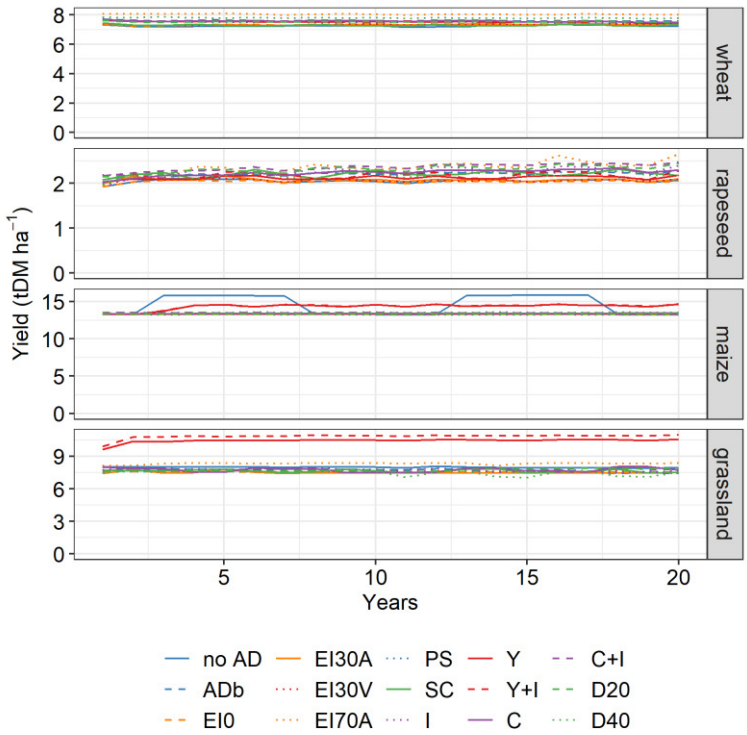
Supplementary Figure S9.E – Evolution of the yield of each crop through the time, for all scenarios. Climatic year is 2004.



Supplementary Figure S9.F – Evolution of the yield of each crop through the time, for all scenarios. Climatic year is 2006.

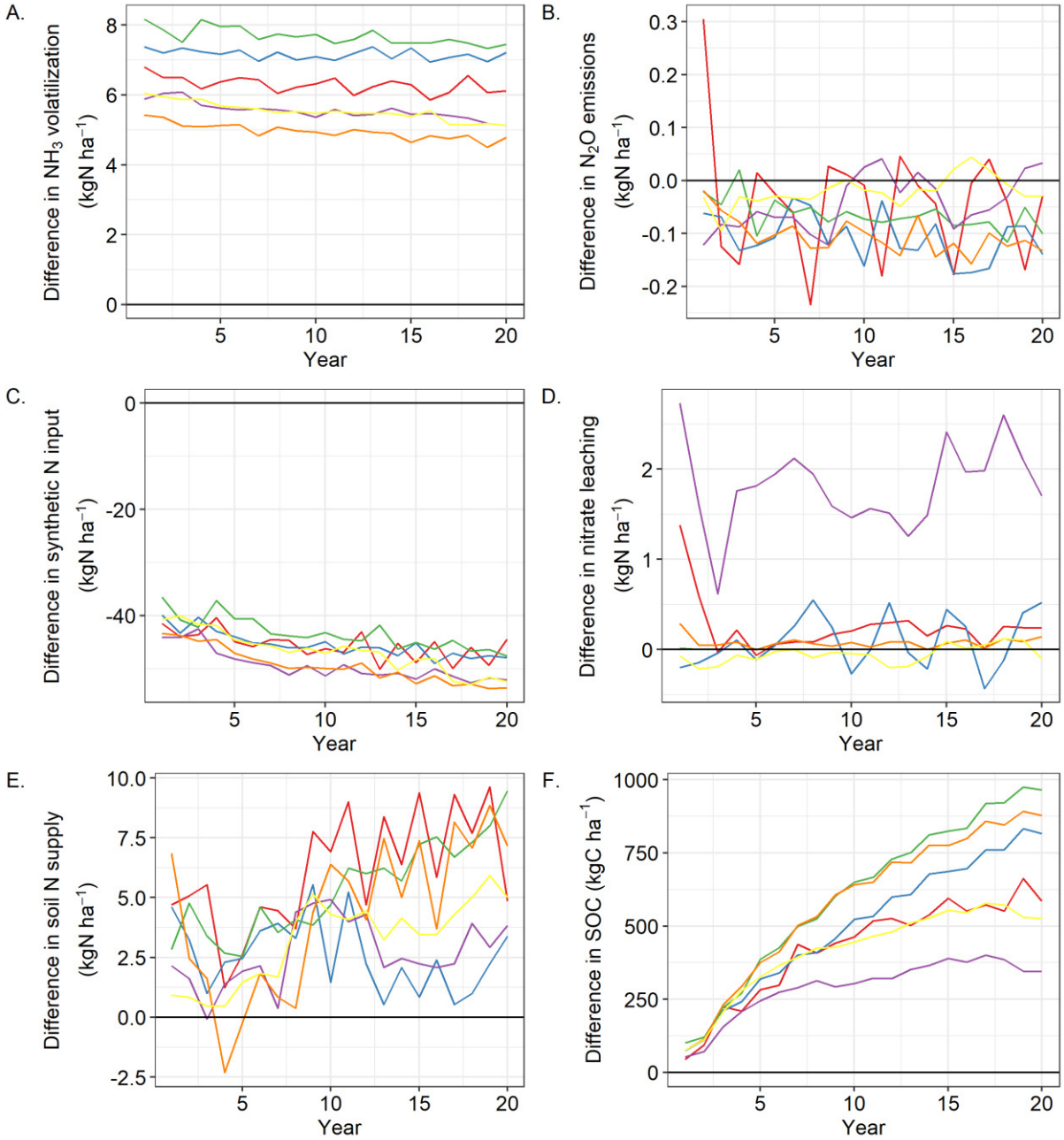


Supplementary Figure S9.G – Evolution of the yield of each crop through the time, for all scenarios. Climatic year is 2007.



Supplementary Figure S9.H – Evolution of the yield of each crop through the time, for all scenarios. Climatic year is 2011.

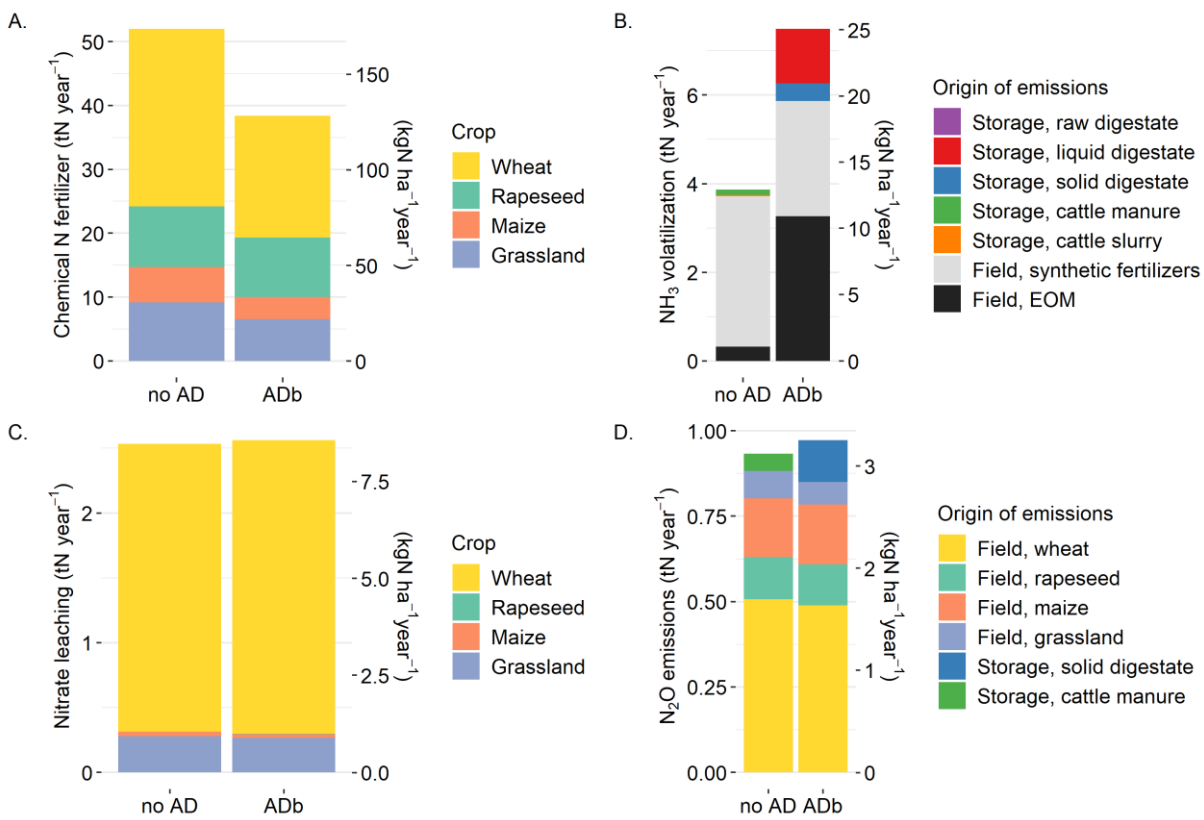
S.M. 10 Evolution of indicators related to C and N fluxes through the time depending on the used climatic year



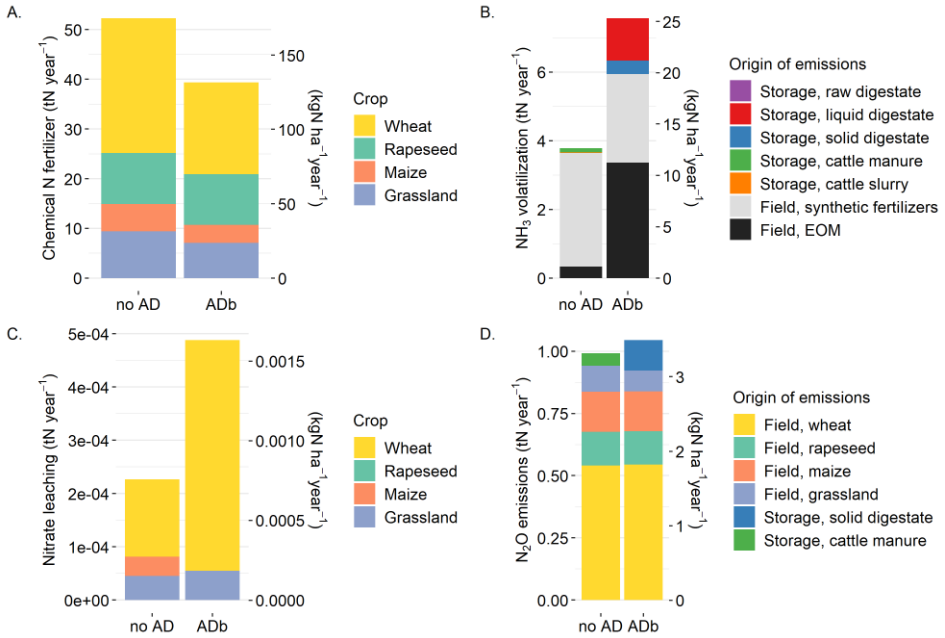
Supplementary Figure S10 – Simulated difference of A) ammonia volatilization; B) N_2O emissions; C) synthetic N input; D) nitrate leaching; E) soil N supply; SOC stocks, between ADb and "no AD" scenarios, across the time. A positive value indicate that the variable of interest is higher in the fields of ADb scenario compared to the one of "no AD" scenario. The average difference of impacts in the entire farm is represented. The color of the line indicate the used climatic year. Red: 2010. Blue: 2003. Green: 2004. Purple: 2006. Orange: 2007. Yellow: 2011.

S.M. 11 Indicators of reference scenarios, depending on climatic year

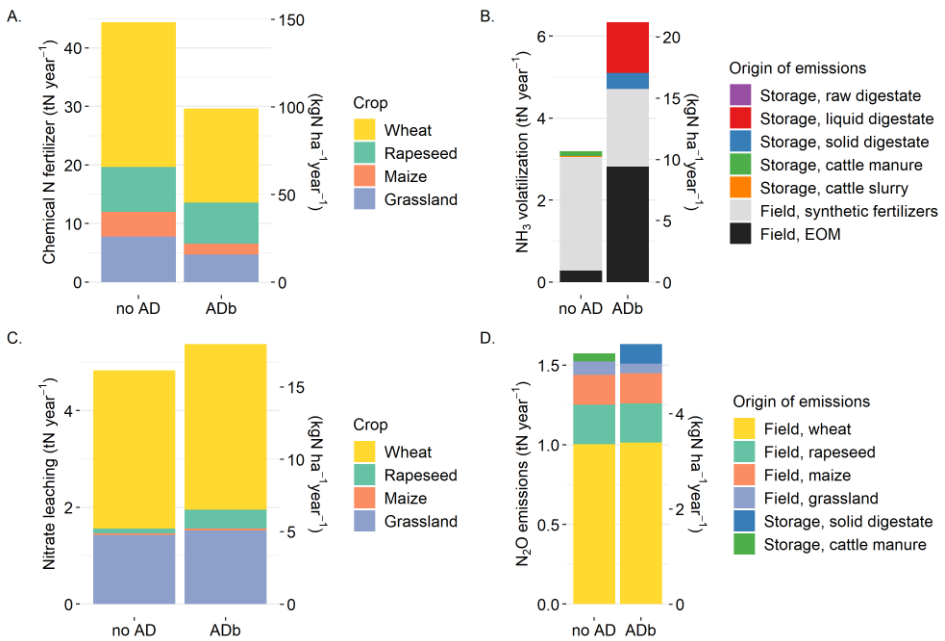
Because no methane emission was modelled in the field, CH_4 emissions did not vary in function of the climatic year.



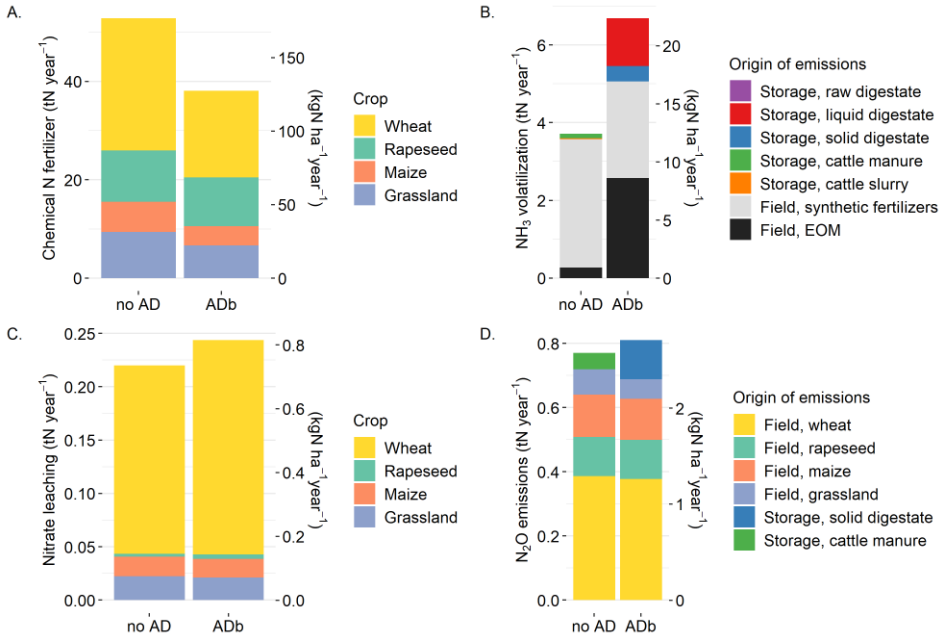
Supplementary Figure S10.A – Comparison of environmental indicators of the farm for ADb and "no AD" scenarios. Each indicator is the yearly indicator averaged along the 20 years of simulations A) Use of chemical N fertilizers, according to crop where these fertilizers are applied. B) Total NH_3 emissions, according to its source of emissions. C) Nitrate leaching in the farm, according to the crop where leaching occur. D) Total N_2O emissions, according to the source of emission. Climatic year is 2003



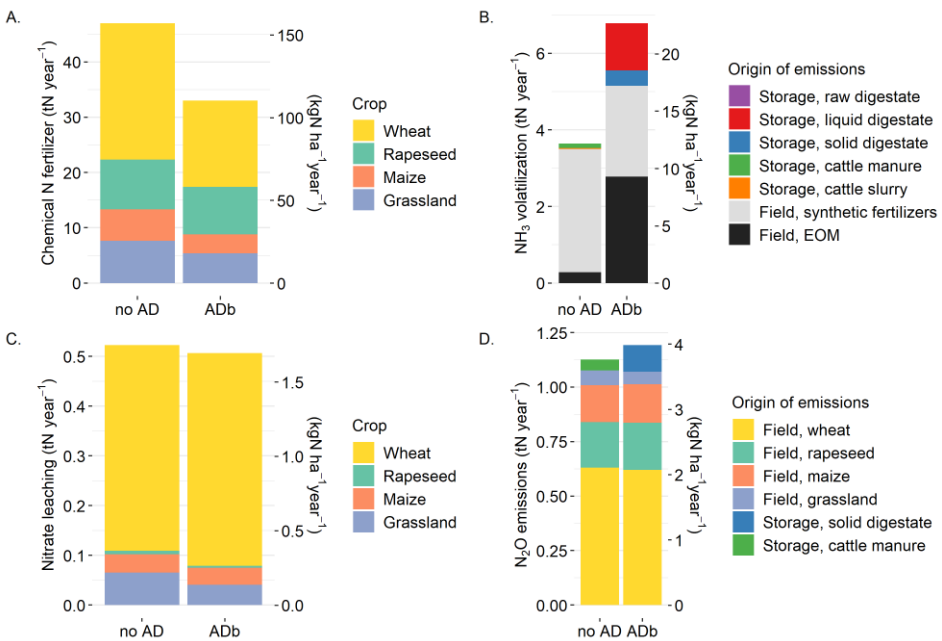
Supplementary Figure S10.B – Comparison of environmental indicators of the farm for ADb and "no AD" scenarios. Each indicator is the yearly indicator averaged along the 20 years of simulations A) Use of chemical N fertilizers, according to crop where these fertilizers are applied. B) Total NH_3 emissions, according to its source of emissions. C) Nitrate leaching in the farm, according to the crop where leaching occur. D) Total N_2O emissions, according to the source of emission. Climatic year is 2004



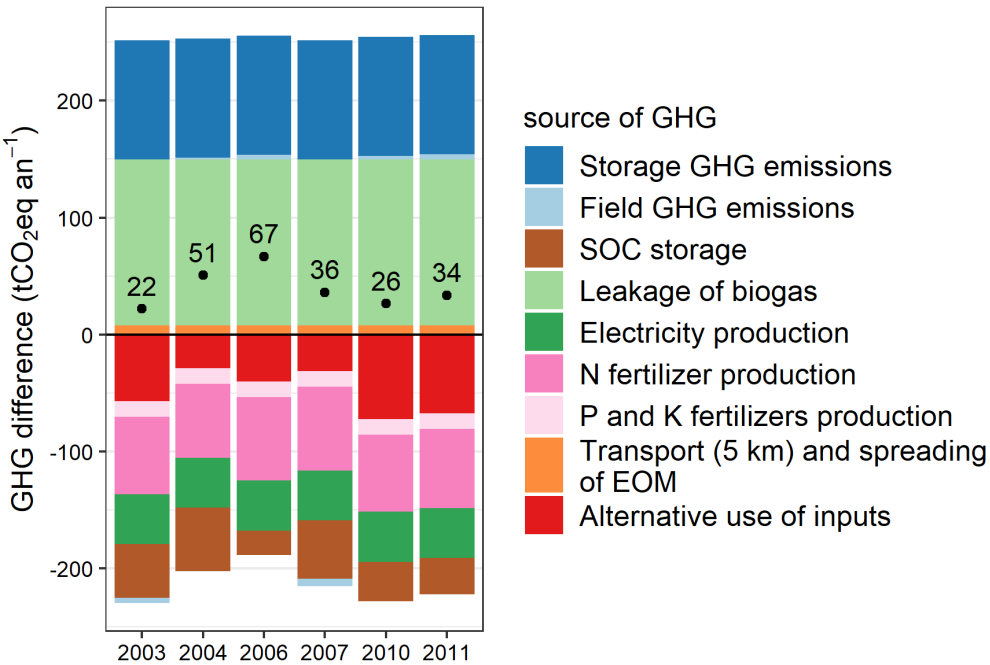
Supplementary Figure S10.C – Comparison of environmental indicators of the farm for ADb and "no AD" scenarios. Each indicator is the yearly indicator averaged along the 20 years of simulations A) Use of chemical N fertilizers, according to crop where these fertilizers are applied. B) Total NH3 emissions, according to its source of emissions. C) Nitrate leaching in the farm, according to the crop where leaching occur. D) Total N2O emissions, according to the source of emission. Climatic year is 2006.



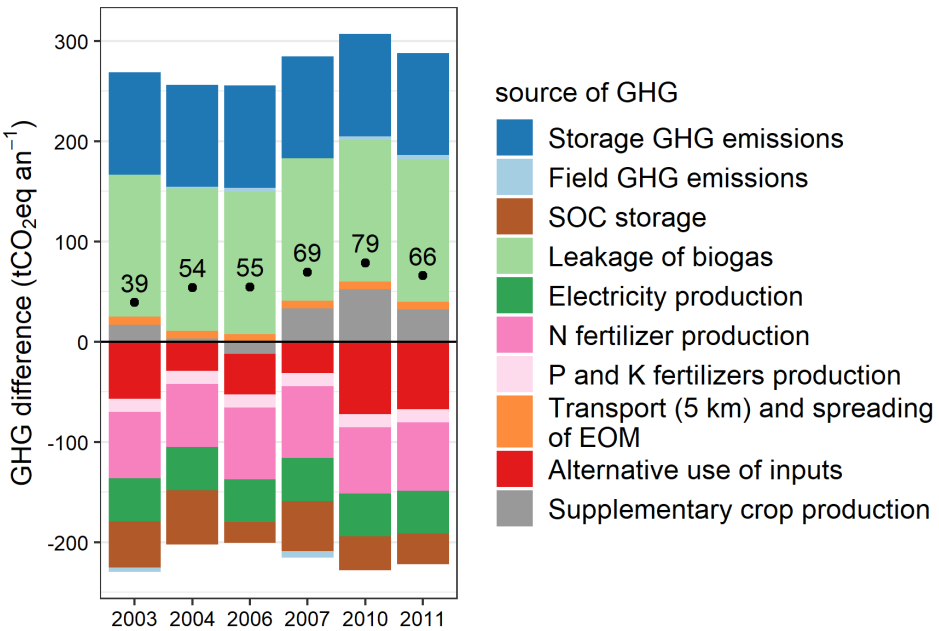
Supplementary Figure S10.D – Comparison of environmental indicators of the farm for ADb and "no AD" scenarios. Each indicator is the yearly indicator averaged along the 20 years of simulations A) Use of chemical N fertilizers, according to crop where these fertilizers are applied. B) Total NH_3 emissions, according to its source of emissions. C) Nitrate leaching in the farm, according to the crop where leaching occur. D) Total N_2O emissions, according to the source of emission. Climatic year is 2007.



Supplementary Figure S10.E – Comparison of environmental indicators of the farm for ADb and "no AD" scenarios. Each indicator is the yearly indicator averaged along the 20 years of simulations A) Use of chemical N fertilizers, according to crop where these fertilizers are applied. B) Total NH_3 emissions, according to its source of emissions. C) Nitrate leaching in the farm, according to the crop where leaching occur. D) Total N_2O emissions, according to the source of emission. Climatic year is 2011.

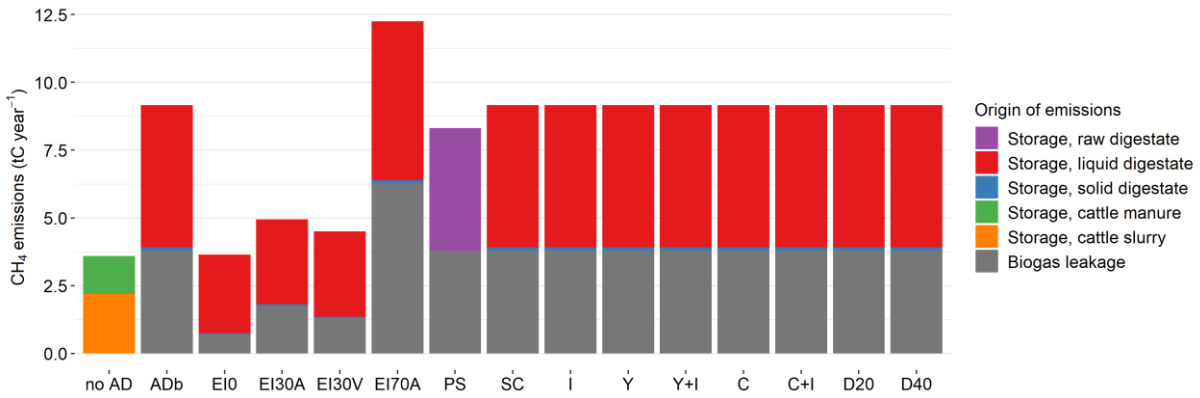


Supplementary Figure S10.F – Net GHG emissions of ADb scenario, compared to "no AD" scenario, in function of the climatic year simulated. The system consider a similar cropped area. The dots represent the net GHG emissions.

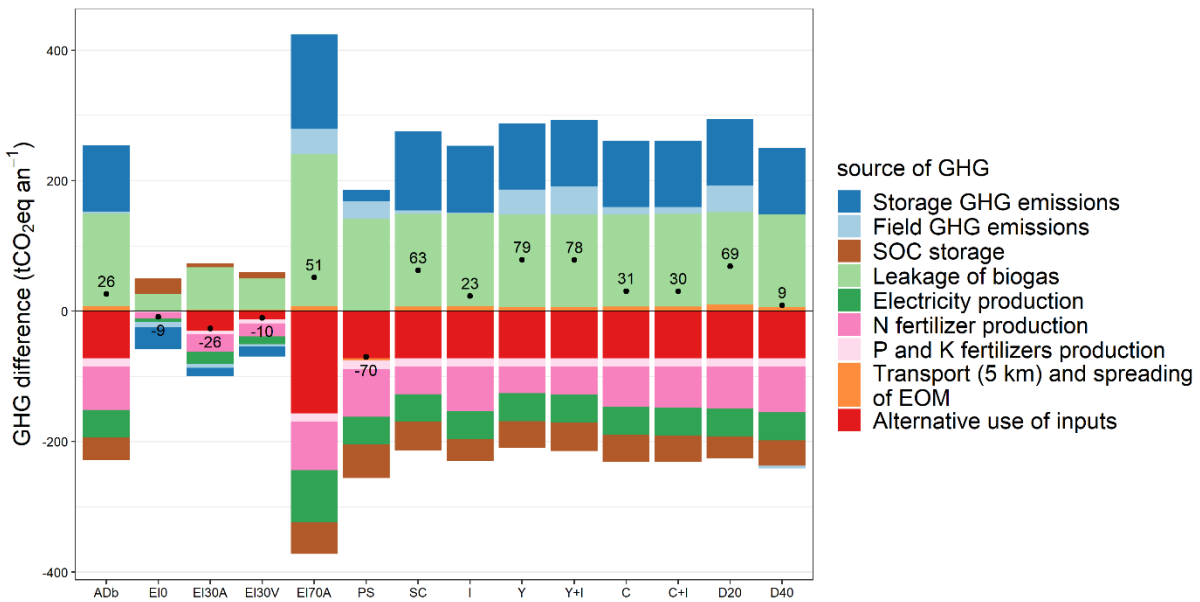


Supplementary Figure S10.G – Net GHG emissions of ADb scenario, compared to "no AD" scenario, in function of the climatic year simulated. The system consider a similar crop production. Supplementary crop production include yield difference in the main farm, yield difference in the farm where sewage sludge were applied, and supplementary production required by the use of maize silage as input of the digester. The dots represent the net GHG emissions.

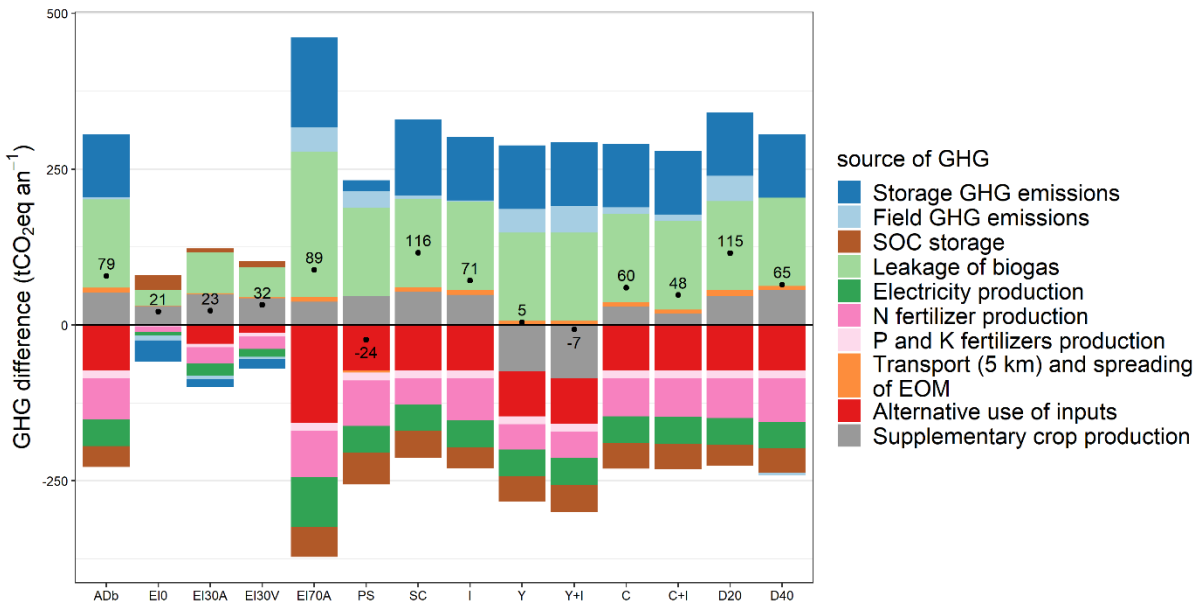
S.M. 12 Indicators for all scenario, on reference climatic year



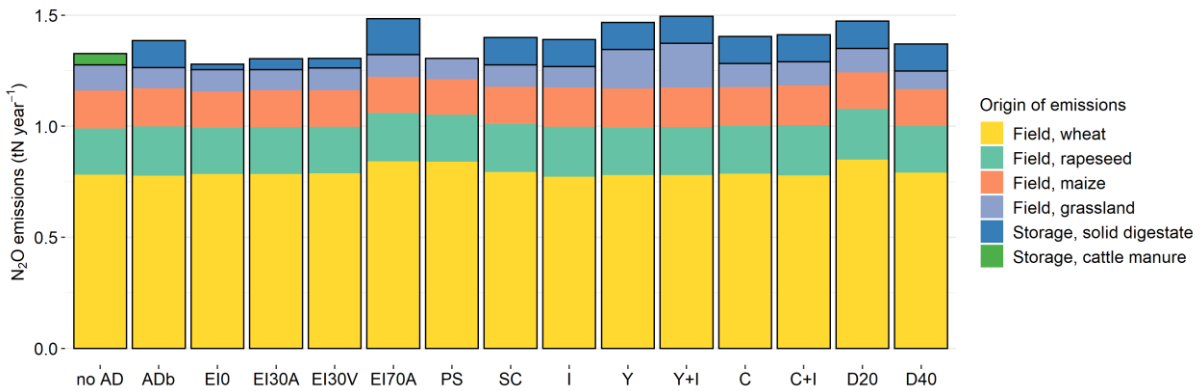
Supplementary FigureS12.A – Farm CH_4 emissions of all scenarios, compared to "no AD" scenario.



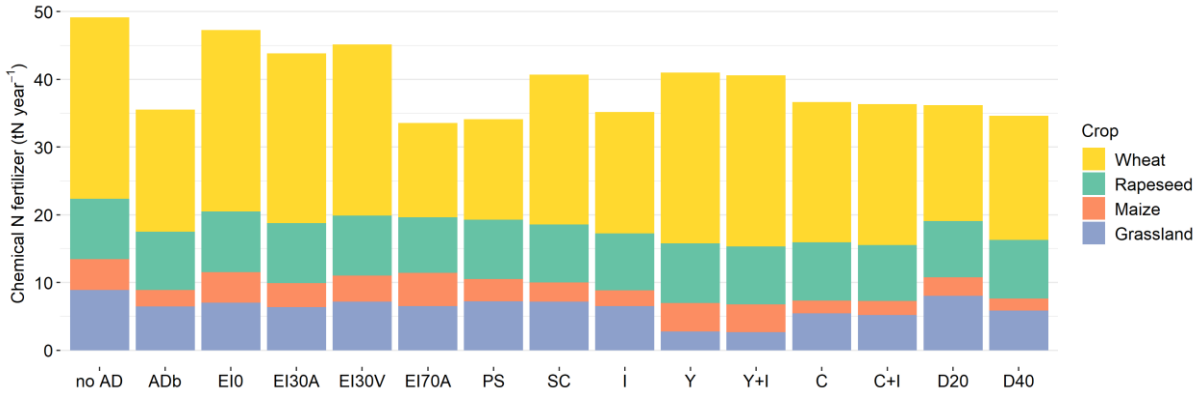
Supplementary Figure S12.B - Net GHG emissions of all scenarios, compared to "no AD" scenario. The system consider a similar cropped area. Climatic year was 2010. The dots represent the net GHG emissions.



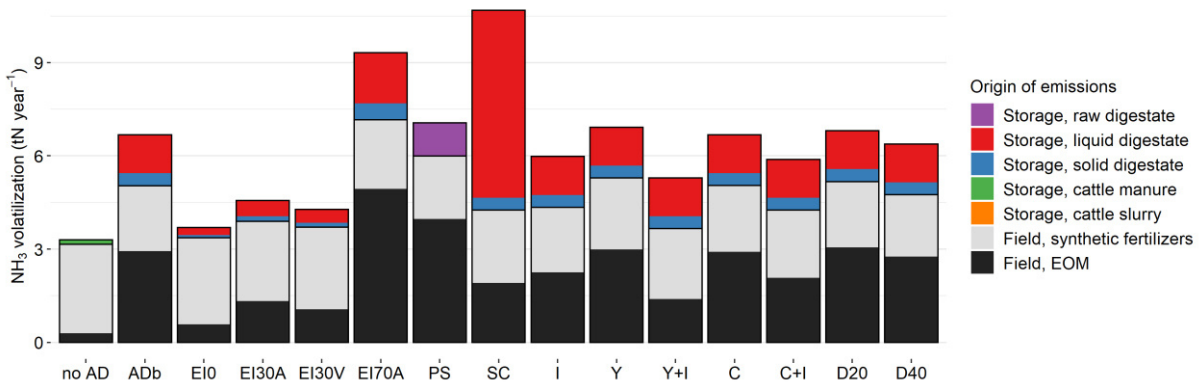
Supplementary Figure S12.C – Net GHG emissions of all scenarios, compared to "no AD" scenario. Climatic year was 2010. The system consider a similar crop production. Supplementary crop production include yield difference in the main farm, yield difference in the farm where sewage sludge were applied, and supplementary production required by the use of silage maize as input of the digester. The dots represent the net GHG emissions.



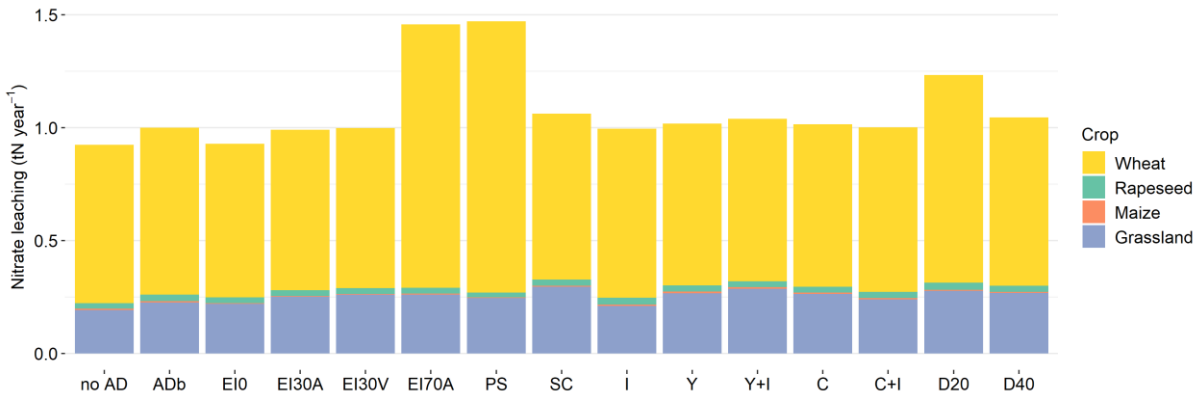
Supplementary Figure S12.D – Farm N_2O emissions of all scenarios, compared to "no AD" scenario. Climatic year was 2010.



Supplementary Figure S12.E – Use of synthetic N fertilizers in all scenarios, compared to "no AD" scenario. Climatic year was 2010.



Supplementary Figure S12.F – Farm NH_3 emissions of all scenarios, compared to "no AD" scenario. Climatic year was 2010.



Supplementary Figure S12.G – Farm nitrate leaching of all scenarios, compared to "no AD" scenario. Climatic year was 2010.

Annexes – Chapitre 7
Supplementary Material

S. M. 1 Treatments in the microcosm ecotoxicity tests, series #1 (2019) and #2 (2020)

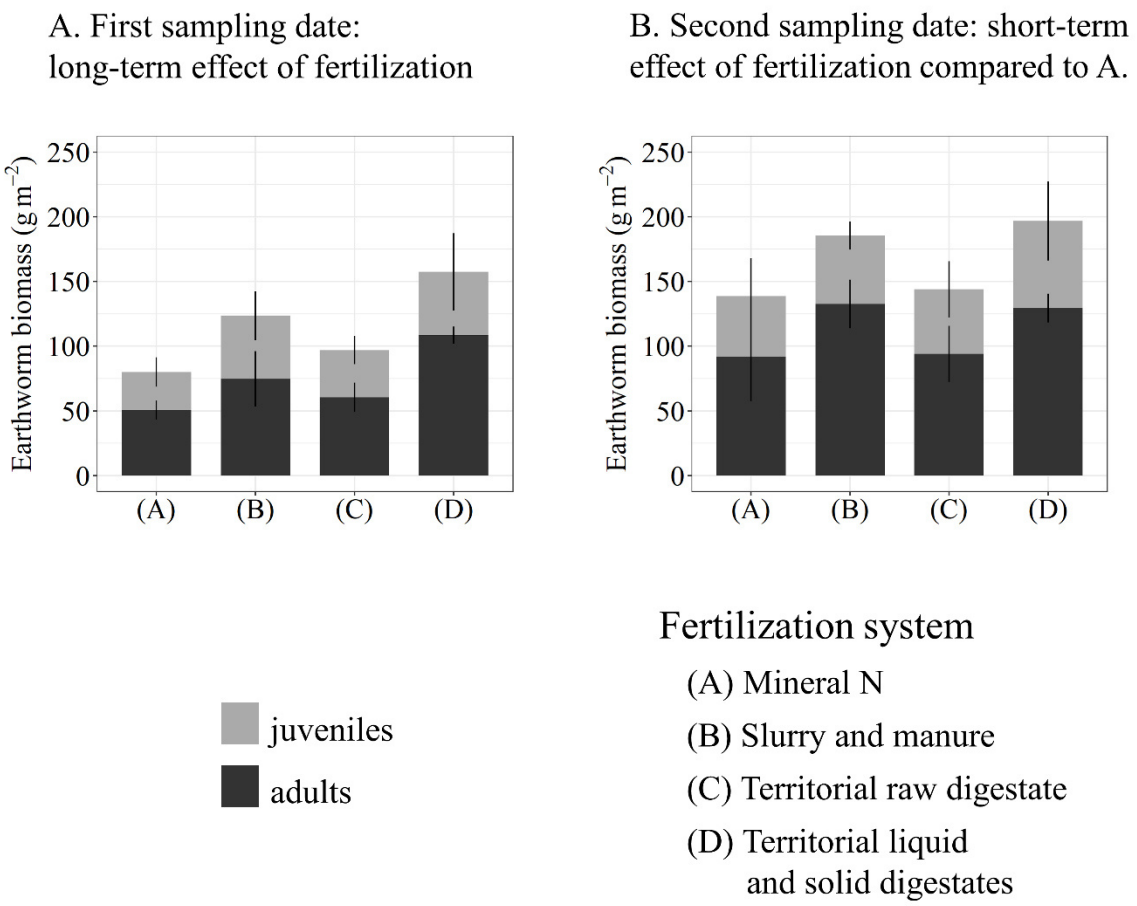
Supplementary Table S1 – Description of the treatments in the microcosm ecotoxicity tests, series #1 (2019)

Series	Species	Organic products	Dose (mass of fresh organic products): (mass of fresh soil)	soil N_{amm} content (mgN_{amm} $kgDS^{-1}$)
#1	*L. terrestris*	territorial digestate	2.5%	79
			5%	159
			10%	317
			20%	634
		territorial digestate, aerated	20%	370
		cattle slurry	10%	199
			20%	398
		control - no products		0
	A. caliginosa	territorial digestate	2.5%	79
			5%	159
			10%	317
			20%	634
		territorial digestate, aerated	20%	370
		cattle slurry	10%	199
			20%	398
		control - no products		0

Supplementary Table S2 – Description of treatments in the microcosm ecotoxicity tests, series #2 (2020)

Series	Species	Organic products	Dose (mass of fresh organic products): (mass of fresh soil)	soil N_{amm} content (mgN_{amm} $kgDS^{-1}$)
#2	*L. terrestris*	territorial digestate	5%	120
			10%	240
			15%	360
			20%	480
		territorial digestate, aerated	5%	95
			10%	191
			15%	286
			20%	382
		Digestate from intermediate crops	2.5%	105
			5%	209
			10%	418
			15%	627
			20%	836
		cattle slurry	5%	49
			10%	98
			20%	196
		Ammonia solution (NH_3)	equivalent to territorial digestate 5%	109
			equivalent to territorial digestate 9%	217
			equivalent to territorial digestate 18%	437
			equivalent to territorial digestate 27%	723
			equivalent to territorial digestate 36%	874
		Ammonium solution (NH_4Cl)	equivalent to territorial digestate 5%	109
			equivalent to territorial digestate 9%	217
			equivalent to territorial digestate 18%	437
			equivalent to territorial digestate 36%	874
		control - no products		0

S. M. 2 Biomass of earthworms in the field



Supplementary Fig. S1- Biomass of earthworms in the field before and after the spreading of organic products. Error bars indicate one standard error. Considering the two dates together, the date significantly influenced total and adult earthworm biomass (Scheirer-Ray-Hare tests, p-values < 0.05). A) Long-term effect of repeated organic product applications: first sampling date, in plots differentiated for 2 years, 7 days before spreading. Adult and total biomasses were not found impacted by treatment (Kruskal-Wallis test, p-value > 0.05). B) Second sampling date, 14 days after spreading. Comparison with subfigure 1.A show the impact of fertilization after 2 weeks on soil earthworm populations. Adult and total biomasses were not found impacted by treatment (Kruskal-Wallis test, p-value > 0.05).

S. M. 3 Weight change in microcosm ecotoxicity tests, series #1 (2019) and #2 (2020)

Supplementary Table S3 – Weight change in the microcosm ecotoxicity tests in series #1 (2019) after 15 days of the experiment. Weight change is expressed as a percentage of the initial body mass. A positive value indicates weight gain, and a negative value indicates weight loss. The number of living earthworms after 15 days is indicated in parentheses (7 earthworms initially).

Series	Species	Organic products	Dose (mass of fresh organic products): (mass of fresh soil)	Weight change in % ± standard error (sample size)
#1	*L. terrestris*	territorial digestate	2.5%	5.8 ± 4.3 (7)
			5%	3.4 ± 3.6 (7)
			10%	(0)
			20%	(0)
		territorial digestate, aerated	20%	(0)
		cattle slurry	10%	-0.5 ± 4.3 (7)
			20%	(0)
		control - no products		5.6 ± 3.6 (7)
	A. caliginosa	territorial digestate	2.5%	-17.1 ± 2.9 (7)
			5%	-7.3 ± 2.0 (6)
			10%	-14.6 ± 7.5 (3)
			20%	(0)
		territorial digestate, aerated	20%	(0)
		cattle slurry	10%	-34.7 ± 10.3 (4)
			20%	(0)
		control - no products		-14.2 ± 7.5 (7)

Supplementary Table S4 – Weight change in microcosm ecotoxicity tests in series #2 (2020) after 15 days of the experiment. Weight change is expressed as a percentage of the initial body mass. A positive value indicates weight gain, and a negative value indicates weight loss. The number of living earthworms after 15 days is indicated in parentheses (7 earthworms initially).

Series	Species	Organic products	Dose (mass of fresh organic products): (mass of fresh soil)	Weight change in % ± standard error (sample size)
#2	*L. terrestris*	territorial digestate	5%	1.6 ± 5.1 (7)
			10%	3.5 ± 2.9 (7)
			15%	-5.3 (1)
			20%	-14.8 (1)
		territorial digestate, aerated	5%	-4.5 ± 2.2 (7)
			10%	2.5 ± 4.9 (7)
			15%	-9.1 ± 2.5 (7)
			20%	-4.3 ± 2.8 (3)
		Digestate from intermediate crops	2.5%	-1.4 ± 1.7 (7)
			5%	-2.1 ± 2.2 (7)
			10%	-0.6 ± 4.2 (7)
			15%	(0)
			20%	(0)
		cattle slurry	5%	3.2 ± 1.9 (7)
			10%	5.6 ± 1.3 (7)
			20%	8.1 ± 4.9 (7)
		Ammonia solution (NH_3)	equivalent to territorial digestate 5%	-3.3 ± 2.7 (7)
			equivalent to territorial digestate 9%	-5.0 ± 4.3 (7)
			equivalent to territorial digestate 18%	-3.9 ± 1.7 (7)
			equivalent to territorial digestate 27%	-8.6 ± 1.9 (2)
			equivalent to territorial digestate 36%	(0)
		Ammonium solution (NH_4Cl)	equivalent to territorial digestate 5%	-2.4 ± 3.8 (7)
			equivalent to territorial digestate 9%	0.5 ± 2.7 (7)
			equivalent to territorial digestate 18%	6.1 ± 2.9 (7)
			equivalent to territorial digestate 36%	3.6 ± 3.2 (7)
		control - no		3.9 ± 2.2 (7)

S.M.4 Weight change in macrocosm experiments

Supplementary Table S5 – Weight change in the macrocosm tests after 28 days of the experiment. Weight change is expressed as a percentage of the initial body mass. A positive value indicates weight gain, and a negative value indicates weight loss.

Treatment	Weight change as % ± standard error	
	L. terrestris	*A. caliginosa*
(C) Control, 40 $t.ha^{-1}$ of water spreading	-17.7 ± 7.1	-15.9 ± 2.0
(LD40) 40 $t.ha^{-1}$ of liquid territorial digestate spreading	-9.7 ± 6.0	-9.4 ± 2.4
(S40) 40 $t.ha^{-1}$ of slurry spreading	-4.3 ± 7.6	-5.0 ± 5.2
(LD80) 80 $t.ha^{-1}$ of liquid territorial digestate spreading	1.5 ± 2.8	-10.7 ± 3.5
(ALD40) 40 $t.ha^{-1}$ of aerated liquid territorial digestate spreading	-6.2 ± 1.1	-7.9 ± 1.6
(LD40-H+) 40 $t.ha^{-1}$ of liquid digestate spreading and a higher soil humidity of 80% WFPS	-11.9 ± 11.1	-6.9 ± 2.8

ÉCOLE DOCTORALE
Agriculture, alimentation, biologie, environnement, santé (ABIES)

Titre : Conséquences de l'introduction de la méthanisation dans une exploitation de polyculture-élevage sur les cycles du carbone et de l'azote. Combinaison de l'expérimentation et de la modélisation à l'échelle de la ferme.

Mots clés : Méthanisation ; Polyculture élevage ; Cycles du carbone et de l'azote ; Bilan environnemental ; Modélisation ; Vers de terre

Résumé : La méthanisation agricole des effluents animaux est une pratique en fort développement en France. Elle produit de l'énergie renouvelable (biogaz). La valorisation des digestats au champ, comme celle des effluents non méthanisés, permet le retour au sol de nutriments et de matière organique, ce qui diminue le besoin en engrais minéraux et entretient les stocks de C des sols. Le traitement et l'épandage de ces produits peut aussi induire l'émission de gaz à effet de serre et de contaminants. La méthanisation agricole influence ces impacts : pour les maitriser, il faut comprendre comment la digestion des effluents avec des déchets importés modifie les cycles du C et du N à l'échelle de la ferme. Cette question a été traitée en s'appuyant sur un cas d'étude à l'INRAE de Nouzilly (Centre – Val de Loire) : une exploitation agricole avec un méthaniseur traitant les effluents de son élevage bovin et divers déchets organiques. Lors de l'essai au champ MétaMétha, nous avons comparé les flux d'azote au cours d'une rotation culturale fertilisée avec des engrais minéraux, des lisiers et fumiers bovins, ou des digestats issus de ces effluents. Les digestats se substituent bien aux engrais minéraux, mais ils sont sensibles à la volatilisation d'ammoniac (NH_3). Les vers de terre peuvent être négativement impactés juste après l'épandage de digestat ou de lisier, mais les effets sont similairement positifs après 2 ans d'apports de matière organique. Nous avons ensuite évalué les modèles STICS et SYS-Metha pour simuler respectivement l'essai au champ et le traitement des digestats. Ces modèles ont été couplés pour simuler les flux de C et N à l'échelle de la ferme. Avec de forts imports de déchets, la méthanisation favorise la substitution des engrais minéraux, le stockage de C dans les sols, mais aussi les émissions de NH_3. Ce travail permet de mieux évaluer les conséquences de l'introduction d'un méthaniseur dans une exploitation agricole et ainsi d'optimiser la filière.

Title : Consequences of the anaerobic co-digestion of cattle effluents in a crop livestock farm on carbon and nitrogen cycles. Combination of experimentation and of modelling at the farm scale.

Keywords : Anaerobic digestion ; Crop-livestock farm; Carbon and nitrogen cycles; Environmental assessment; Modelling; Earthworms

Abstract : The anaerobic digestion (AD) of animal effluents is strongly developing in France. It produces renewable energy (biogas). Like undigested effluents, the use of anaerobic digestates in the field enables the recycling of nutrients and organic matter in the soil, which decreases the need for synthetic fertilizers and maintains soil organic carbon stocks. However, the treatment and field application of those organic products can also cause greenhouse gas emissions and contaminants. The on-farm AD influences those impacts. In order to control them, we need to understand how the co-digestion of animal effluents with imported organic wastes influences C and N cycles at the farm scale. We studied this question with the support of a case study at INRAE in Nouzilly (Centre – Val de Loire, France): a crop and livestock farm where an anaerobic digester treats the cattle effluents together with imported wastes. During the MetaMetha field experiment, we compared N fluxes during a crop rotation that was fertilized with synthetic N fertilizers, undigested cattle slurry and farmyard manure, or digestates. We found that digestates can substitute synthetic fertilizers, despite the risk of ammonia (NH_3) emissions. Earthworms can be negatively impacted just after the application of digestate of slurry, but the input of organic matter induced similar positive impacts after two years. We then evaluated the STICS and SYS-Metha models to simulate the field experiment, and digestate treatment and storage, respectively. Both models were coupled to simulate C and N fluxes at the farm scale. When large amounts of digested wastes are imported, AD promotes substitution of synthetic N fertilizers and storage of soil organic C, but also NH3 emissions. The study enabled us to better evaluate the consequences of the on-farm AD and therefore to optimize the sector.

Maison du doctorat de l'Université Paris-Saclay
2ème étage aile ouest, Ecole normale supérieure Paris-Saclay
4 avenue des Sciences,
91190 Gif sur Yvette, France