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Greenhouse gas emissions from inorganic and organic fertilizer production and use: A review of emission factors and their variability

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Highlights

The primary emissions sources are from production and in-field post-application.

Transportation is generally very low compared to all other sources.

Variability in emission factors for all sources is high.

Sub-optimal composting and anaerobic digestion can lead to high GHG emissions.

Case specific data is necessary for accurate emissions assessment.

Abstract

Fertilizers have become an essential part of our global food supply chain and are necessary to sustain our growing population. However, fertilizers can also contribute to greenhouse gas (GHG) emissions, along with other potential nutrient losses in the environment, e.g. through leaching. To reduce this environmental impact, tools such as life cycle assessments and decision support systems are being used to aid in selecting sustainable fertilization scenarios. These scenarios often include organic waste-derived amendments, such as manures, composts and digestates. To produce an accurate assessment and comparison of potential fertilization scenarios, these tools require emission factors (EFs) that are used to estimate GHG emissions and that are an integral part of these analyses. However, such EFs seem to be very variable in nature, thereby often resulting in high uncertainty on the outcomes of the analyses. This review aims to identify ranges and sources of variability in EFs to provide a better understanding of the potential uncertainty on the outcomes, as well as to provide recommendations for selecting EFs for future studies. As such, an extensive review of the literature on GHG emissions from production, storage, transportation and application of synthetic fertilizers (N, P, K), composts, digestates and manures was performed. This paper highlights the high variability that is present in emissions data and confirms the great impact of this uncertainty on the quality and validity of GHG predictions related to fertilizers. Variability in EFs stem from the energy source used for production, operating conditions, storage systems, crop and soil type, soil nutrient content, amount and method of fertilizer application, soil bacterial community, irrigation method, among others. Furthermore, a knowledge gap exists related to EFs for potassium fertilizers and waste valorization (anaerobic digestion/composting) processes. Overall, based on this review, it is recommended to determine EFs on a case by case basis when possible and to use uncertainty analyses as a tool to better understand the impact of EF variability.

Keywords: compost, anaerobic digestion, digestate, synthetic, manure, waste valorization

1. Introduction

Over the course of the past century, the human population has nearly quintupled from about 1.6 billion people in 1900 to nearly 7.6 billion in 2017 (United Nations, 2017). This explosion in world population has resulted in a drastically increased demand for food and has put the agricultural sector under heavy stress to provide the necessary sustenance. Though the rapid growth in population has likely plateaued, the population is still expected to surpass 11 billion by the end of the century (United Nations, 2017). As a consequence of this continued expansion (predominantly from developing countries in Africa) and of the increase in affluence in developing nations, we will continue to demand more from our resource-limited planet. Current estimates predict an increase in agricultural production of about 1.5% per year, totalling a growth of about 15% over the next ten years (OECD, 2018) and 50 to 100% by 2050 (Baulcombe et al., 2009; Tilman et al., 2011).

Alongside the increasing crop production, we also find an equally impressive 200-300% increase in synthetic fertilizer use between 1970 and 2010 (FAOSTAT, 2013; Smith et al., 2014), while China increased its nitrogen (N) fertilizer production by a factor of 39 between 1963 and 2015 (Luo et al., 2018). It is believed that, during this ground-breaking period in agriculture, at least 30-50% of the crop yield was attributed to the use of fertilizers (Baligar et al., 2001; Stewart et al., 2005). Consequently, it has been estimated that nearly 50% of the world's population is now dependent on nitrogen fertilizers for their sustenance (Erisman et al., 2008; Smil, 2002). Furthermore, this propensity towards fertilizer use isn't waning, as the International Fertilizer Association (IFA) reported a 46% increase in urea production between 2003 and 2013 (Heffer and Prud'homme, 2016) and the Food and Agriculture Organization of the United Nations (FAO) predicted an annual increase in fertilizer nutrient demand of 1.5, 2.2 and 2.4% for nitrogen, phosphorus (P) and potassium (K) between 2016 and 2020 (FAOSTAT, 2017).

However, though fertilizers have allowed us to keep up with the growing demand for agricultural products, their historical overuse (Byrareddy et al., 2019; Kurdi et al., 2020; Lu and Tian, 2017; Sun et al., 2019; Withers et al., 2015; Zulfiqar and Thapa, 2017) has introduced an environmental crisis in many parts of the world. Indeed, improper use of fertilizers can have a detrimental effect on terrestrial, marine and freshwater ecosystems by causing soil nutrient depletion (through unbalanced fertilization), soil acidification, eutrophication, nutrient run-off, reduced biological diversity and greatly increased greenhouse gas (GHG) emissions from agricultural practices (Horrigan et al., 2002; Sutton et al., 2013; Vitousek et al., 1997).

Today, the International Panel on Climate Change (IPCC) estimates that agricultural production accounts for about 12% of man-made (anthropogenic) GHG emissions, which can go up to 24% when considering land use and change activities. It is the largest contributor to anthropogenic non-CO_2 GHG emissions (IPCC, 2014), while production of synthetic nitrogen fertilizers alone accounts for approximately 2% of the world's energy use (Kongshaug, 1998; Sutton et al., 2013). The most prevalent GHG emissions are in the form of nitrous oxide (N_2O), methane (CH_4) and carbon dioxide (CO_2), accompanied by high emissions of ammonia (NH_3). It is estimated that agriculture is responsible for more than 80% of the anthropogenic emission of N_2O and 70% of the anthropogenic NH_3 emissions, which originate predominantly from the application of livestock manure and inorganic fertilizer, as well as around 40% of anthropogenic CH_4, which comes mostly from enteric fermentation (Bouwman et al., 1997; Davidson and Kanter, 2014; IPCC, 2014; Zhang et al., 2010).

One of the most useful metrics to evaluate the environmental impact of fertilizers and to undertake accurate environmental and life cycle assessments (LCAs) is emission factors (EFs, i.e. amount of GHG/reference unit). However, EFs are also currently one of the most limiting factors to the validity of these analyses, being one of the main sources of variability on the outcomes (Basset-Mens et al., 2009; Brodt et al., 2014; Brown et al., 2010; Chen and Corson, 2014; Flysjö et al., 2011; Meyer-Aurich et al., 2012; Milne et al., 2014; Wolf et al., 2017b).

Indeed, even though proper emissions accounting is integral to accurate environmental assessments, awareness of the factors influencing EFs and their impact on these evaluations is grossly lacking. When looking through work on environmental and life cycle assessments, we find that the scope of considered factors varies drastically from one study to the next. This can have serious implications on the results of these studies and can greatly complexify the generalization and comparison of different works. For example, the assessment by Nakamura et al. (2014) determined that 67% of total emissions from the use of digestate as an organic fertilizer came from transportation, while a similar assessment by Timonen et al. (2019) found that transportation was only responsible for 2% of total emissions. This great disparity in findings stems from the scope of emissions considered in these studies, with Nakamura et al. (2014) neglecting emissions from production, storage, and considering more limited in-field emissions, highlighting how much of an issue this can be.

Given the highly multidisciplinary nature of many of these environmental assessments, especially when waste valorization and agriculture come together, as is the case with organic fertilizers, it is understandable that the scope of what emission sources should be considered can be nebulous. When assessing the overall fertilizer production and use chain, many factors can come into play. For example, the source of energy used to power these processes could potentially have an important impact, while different types of organic matter/waste may lead to varying emission profiles from processes such as manure management and composting. Soils are also complex ecosystems and, given their integral role in the (de)nitrification process, it stands to reason that many factors can influence emissions following the application of fertilizers, such as soil type, climate, soil nutrient content, etc.

However, despite a great deal of work on EFs and although essential to performing accurate and objective LCA and decision-support studies, a comprehensive overview of EFs and their sources of variability for various types of fertilizers is lacking in the scientific literature. This is especially true for the larger fertilizer life cycle, including production, storage, transportation and post-application in-field emissions, previous reviews having only focused on a single type of fertilizer and a single stage of the cycle (Broucek, 2017;

Snyder et al., 2009; Wood and Cowie, 2004). Such work is particularly important given the growth of circular economies, oftentimes creating a strong link between waste management and agriculture.

Therefore, the objective of this review is twofold: in the aim of capturing and better understanding the variability of GHG emissions from fertilizer production and use (1) we seek to review and inventory emission factors for production, storage, transportation and post-application in-field emissions of synthetic fertilizers, composts, digestates and manures; (2) based on these factors, we hope to identify the main sources of variability and highlight the impact of the various amendments in terms of GHG emissions and how we can best mitigate them. Note that this paper does not target a comparison of fertilizer alternatives, but instead aims at highlighting areas that environmental assessments should focus on and precautions that should be taken when selecting and using emission factors. This paper is based on a review of the literature, of which the methodology is presented in section 2, on GHG emissions related to fertilizers and is divided by emissions associated to production (section 3), transportation (section 4), storage (section 5) and post-application (section 6) of fertilizers, followed by a discussion and concluding remarks in sections 7 and 8.

2. Methodology

This review was done using the Web of Science database and complemented with results from Google Scholar, as well as technical reports that the authors were aware of. Multiple advanced searches were used for every fertilizer type and emission source using combinations of the following search terms: emission*, N_2O, nitrous oxide, CH_4, methane, emission factor*, fertilizer, inorganic, mineral, synthetic, organic, compost*, manure*, digest*, phosphorus, P, potassium, K, production, storage, transportation, *field, *application. Articles were then assessed based on their title, abstracts and keywords. Despite the large number of articles identified for every step (between 20 and 500 for each search), a majority were discarded due to a lack of pertinence. In all, about 600 articles were investigated, more than 200 of which ended up being used in this review.

In many cases throughout this review, emission factors will be given as CO_2 equivalents. These values, when not provided directly by studies, have been obtained using global warming potentials of 25 for methane and 298 for nitrous oxide (Solomon et al., 2007). In some cases, emission factors were not provided directly in the reviewed papers, but the information necessary to calculate them was.

3. Production

3.1 Synthetic fertilizers

3.1.1 Nitrogen fertilizers

The main production method for N-fertilizers is through the Haber-Bosch process (see Haber and Le Rossignol (1916)). It concerns an energy intensive process that allows for the transformation of hydrogen, usually stemming from the CH_4 from natural gas, and nitrogen extracted from air into ammonia. This ammonia can then be used in the synthesis of N-fertilizers, most notably urea ($CO(NH_2)_2$) and ammonium nitrate (NH_4NO_3), which make up approximately 75% of the world's straight N-fertilizer consumption according to the International Fertilizers Industry Association (IFA, 2019). As a consequence of the high energy requirements of ammonia synthesis, the energy demand for crop systems using synthetic nitrogen tends to be dominated by the energy requirements to produce these fertilizers. Hoffman et al. (2018) estimated that 45% of the total energy use for a conventional grain crop production stems from synthetic N production.

Emission factors for urea and ammonium nitrate production are compiled in Table 1. Looking at urea production, we can see a relatively large variability in the reported EFs, ranging from 1.3 to 5.5 kg CO_2-eq./kg of N. This variability is most often associated to differences in feedstock and the maturity of the practice between countries. The comparative analysis provided by Brentrup et al. (2016) highlights the wide differences in EFs reported in Europe (generally lower) versus the United States and China (generally higher). Indeed, many developing countries, with China at the forefront, use coal for the steam reformation step instead of natural gas, leading to significantly higher emissions. In fact, in 2012, 86% of the energy used for ammonia synthesis in China came from coal, leading to a fivefold increase in energy requirements compared to a process operating with natural gas (Zhang et al., 2013). EFs for ammonium nitrate production appear to be at least twice as high and up to five times higher than those of urea, but the range from 3.5 to 10.3 kg CO_2-eq./kg of N is still very variable. These higher emissions are due to the increased N_2O emissions from nitric acid production required for ammonium nitrate that greatly affect the overall emissions of the fertilizer production process (Brentrup et al., 2004).

Table 1. Emission factors for N-fertilizer production.

	Emission factor (kg CO_2-eq./kg of N)	Country/Region	Reference
Urea	1.3	Western Europe	Kongshaug (1998)
	1.6	Europe	Skowrońska and Filipek (2014)

	1.9/2.7/5.5	Europe/Russia, USA/China	Brentrup et al. (2016)
	3.1	Southeastern United States	Albaugh et al. (2012)
	3.5	United Kingdom	Williams et al. (2010)
	4	Sweden and Western Europe	Davis and Haglund (1999)
Ammonium nitrate	6.2	Europe	Skowrońska and Filipek (2014)
	6.5	United Kingdom	Elsayed et al. (2003)
	6.8	Western Europe	Kongshaug (1998)
	7	Sweden and Western Europe	Davis and Haglund (1999)
	7.1	Netherlands	Kramer et al. (1999)
	7.2	United Kingdom	Williams et al. (2010)
	3.5/8/10.3	Europe/Russia, USA/China	Brentrup et al. (2016)

3.1.2 Phosphorus fertilizers

Moving onto the second major macronutrient, phosphorus (P) fertilizers are generally produced from the chemical treatment of phosphate rocks obtained through mining (Syers et al., 2011). These rocks are treated with sulfuric acid (H_2SO_4) to form phosphoric acid (H_3PO_4), which is then used in the synthesis of many common P fertilizers such as monocalcium phosphate ($Ca(H_2PO_4)_2$), monoammonium phosphate ($NH_4H_2PO_4$) and diammonium phosphate ($(NH_4)_2HPO_4$).

In terms of P-fertilizer consumption, ammonium phosphates are by far the most popular P-fertilizer, representing 48.7% of global P-fertilizer consumption (IFA, 2019), while the straight (single nutrient) P-fertilizer market is mostly made up of single superphosphate (45.6% straight, 8.2% total) and triple superphosphate (31.7% straight, 5.7% total). All three of these products are obtained through what is known as the wet route of P-rock transformation (da Silva and Kulay, 2005). In all these cases, the P-rock is reacted with sulfuric acid to produce single superphosphate or phosphoric acid. Triple superphosphate can then be produced by reacting phosphate concentrate with phosphoric acid, while ammonium phosphate is obtained by reacting ammonia with phosphoric acid (da Silva and Kulay, 2005).

Table 2 presents the emission factors for P-fertilizer production. We can start by noting once again the significant variability in reported EFs. EFs for ammonium phosphates vary from 1.3 to 8.9 kg CO_2-eq./kg of P_2O_5, while emissions for the super phosphates are significantly lower, though they still present an important variation. In general, it is expected that ammonium phosphates will have a higher emission factor due to the need for ammonia in the process.

Table 2. Emission factors for P-fertilizer production.

	Emission factor (kg CO_2-eq./kg of P_2O_5)	Country/Region	Reference
Ammonium phosphates	1.3-1.8	Sweden and Western Europe	Davis and Haglund (1999)
	1.4/1.7/ 2.89	Europe/Russia, USA/China	Brentrup et al. (2016)
	6.4	Southeastern United States	Albaugh et al. (2012)
	7.8-8.9	China	Zhang et al. (2017)
Single superphosphate	0.6	United Kingdom	Williams et al. (2010)
	1	Sweden and Western Europe	Davis and Haglund (1999)
Triple superphosphate	0.4-0.54	Europe, Russia, USA, China	Brentrup et al. (2016)
	1	Sweden and Western Europe	Davis and Haglund (1999)
	1.1	Brazil	da Silva and Kulay (2005)
	1.2	United Kingdom	Williams et al. (2010)
	1.6	Europe	Skowrońska and Filipek (2014)

3.1.3 Potassium fertilizers

For the final of the major macronutrients, potassium (K) is generally provided to crops in the form of potash, which encompasses a variety of K-bearing salts, such as potassium chloride (KCl), potassium sulphate (K_2SO_4) and potassium nitrate (KNO_3) (Fixen and Johnston, 2012). As is the case for phosphorus, the most common production method of potash is through mining.

A recent life cycle assessment of KCl production in China, undertaken by Chen et al. (2018), estimated that the production of potassium chloride resulted in the emission of 0.19 kg CO_2-eq./kg of K_2O, equivalent to 0.11 kg CO_2-eq./kg of KCl, with an uncertainty range of 0.14 kg CO_2-eq./kg of K_2O to 0.25 kg CO_2-eq./kg of K_2O within a 95% confidence interval. This EF is presented in Table 3 for ease of identification. Of these emissions, the vast majority stemmed from energy generation, accounting for 72.61% of the total, as well as on-site emissions of CO_2 and N_2O during the manufacturing process (25.70%). Due to the importance of energy needs and the generally coal based sources used in China, the authors highlighted the potential to greatly reduce emissions by using greener energy sources. This underlines the potential for these EFs to have a significant variability across the world. The emissions emanating from potassium fertilizer production appear to have been neglected in the literature, seeing how we are unable to find any other studies on the subject. As such, it is important to be mindful when using these values. Further corroboration is necessary. It would also be pertinent to have an analysis of Canadian potash production due to its dominance in global production and the high purity of the minerals found there (USGS, 2019).

Table 3. Emission factor for K-fertilizer production.

	Emission factor (kg CO_2-eq./kg of K_2O)	Country/Region	Reference
Potassium chloride	0.14-0.25	China	Chen et al. (2018)

3.2 Manures

Looking at organic fertilizers, the production of manures results in important GHG emissions from the enteric fermentation from livestock, as well as emissions from manure management. EFs for enteric fermentation are presented in Table 4, while manure management encompasses a variety of phases that will be explored throughout the following sections. These phases include manure storage (sections 3.3 and 3.4) and treatment (section 4). The EFs used in Table 4 stem from the United Nation's Food and Agriculture Organisation based on the IPCC tier 1 methodology.

Table 4. Emission factors for enteric fermentation (Tubiello et al., 2013).

Agriculture	Emission factor (kg CO_2 /head/yr)
Dairy cattle	1050-3200
Non-dairy cattle	675-1500
Pigs	25-37.5
Sheep/goats	125-200
Buffalo	1375
Mules/asses/horses	250-450
Lamas	200
Camels	1150

These EFs can be extremely variable depending on the type of animal, their age, their weight, the type of feed, the acidity of the rumen contents, the variability in production operations, the country of production, the season and the time of day (Bannink et al., 2011; Huang and Guo, 2018; Lee et al., 2018; Wolf et al., 2017a; Woods and Yan, 2010; Yan et al., 2010). This has proven to be an important and longstanding issue in the literature. Dong et al. (2004), comparing the various methods presented in the IPCC's guidelines from 1996, found differences in emissions of up to 33% from one method to another. These results have been consistently validated an expanded upon over time for subsequent guidelines by many other researchers. Indeed, Woods and Yan (2010) and Yan et al. (2010) compared the IPCC's guidelines (IPCC, 2006) to in-fied data obtained in Ireland, concluding that "Tier 1 default factors over-estimated CH_4 emissions for dairy cows, young cattle and sheep, while they under-predicted CH_4 emissions for beef, cattle, heifers and sheep at age between 1 and 2 years old" (Yan et al., 2010). They highlighted that "actual EF data that is representative of the age and diet of the animal" (Woods and Yan, 2010) is necessary and that these findings indicate that the development of emission inventories from the IPCC guidelines, notably the Tier 1 method, "can result in considerable and systematic errors". This has led other researchers to revise these standard EFs, such as Wolf et al. (2017a) who developed new EFs resulting in estimates 11% higher than those using the IPCC's factors. Some researchers have also focused on specific sources of variability, such as the impact of feed, the age of the animal, or variability of EFs through time. For instance, Lee et al. (2018) developed new EFs for dairy cow enteric fermentation in Korea based on the body weight (age) of the animals. In all cases, they found EFs greater than those of the IPCC, ranging from 2.3% to 78.5% greater. This variability in EFs can be problematic when undertaking environmental assessments, especially given that EFs for enteric fermentation are the main factor in the uncertainty of methane emissions (Milne et al., 2014).

3.3 Composts

The composting process is undertaken by biodegrading organic matter in an aerobic environment. As such, it is expected that a properly operated composting system should release its GHGs in the form of CO_2, with most of the methane being oxidized. However, despite the frequency of this assumption, CH_4 emissions were found to be very prevalent in our review. Nitrous oxide emissions are also very likely, N_2O being produced either through incomplete ammonium oxidation or (de)nitrification (Beck-Friis et al., 2000). This being said, emissions from composting are often considered as being biogenic, in that they do not contribute to global warming. As such, there is a limited amount of work seeking to quantify the emissions related to the process.

The literature on these emissions has provided some very varied results, which are presented in Table 5. In terms of emission factors, Hao et al. (2005) obtained CH_4 emissions between 0.2 and 6.1% of initial total carbon, with N_2O emissions between 0.06 and 0.2% of initial total nitrogen, these ranges being about double of what was found during the experiments of Amlinger et al. (2008), but fitting within and below the bounds reported by Hellebrand (1998) and the review by Zeman et al. (2002) who reported N_2O and CH_4 EFs of between 1 and 6% of initial nitrogen/carbon. A detailed report on composting emissions was published by the French Environment and Energy Management Agency investigating emissions based on feedstock (ADEME, 2012). The report found very wide-ranging emissions depending on feedstock, with methane emissions varying from 0.01 to 10% of initial carbon and nitrous oxide emissions between 1 and 50% of initial nitrogen. This data truly highlights the variability in EFs from this process, seeing how no trend is apparent amongst operating conditions or waste types. Indeed, in the ADEME's report, the highest and lowest emissions were associated to processes treating the same substrate (pig manure) for similar lengths (>6 weeks) with forced aeration and turning at the same dates.

The IPCC's guideline for composting of 4.0 kg CH_4/tonne of waste and 0.3 kg N_2O/tonne of waste (IPCC, 2006), equating to about 190 kg CO_2-eq. per tonne in total, seems to concord with only a few of the studies, especially considering that CO_2 emissions are considered biogenic and therefore not accounted for. Furthermore, many country specific emission factors, such as those of Australia, Austria, Belgium, Germany, Italy and the Netherlands have significantly lower recommended EFs than the IPCC, varying between 32 and 57 kg CO_2-eq. per tonne (Jeong et al., 2019). Overall, there is clear variability in EFs used for composting which is due to the variable composition of the waste materials used and to the type and duration of the composting process that is not being considered in many environmental assessments.

Table 5. Emission factors for compost production.

Emission factor (kg CO_2-eq./tonne of waste) excluding CO_2	Emission factor (kg CO_2-eq./tonne of waste) including CO_2	Waste type	Reference
3-5	45-82	Hen carcasses and manure	Zhu et al. (2014)
28-44	145-173	Dairy manure	Ahn et al. (2011)
34.05-43.49	-	Food waste	Jeong et al. (2019)
-	323	General	White et al. (2012)
143	380	Grass and green waste	Hellebrand (1998)
183	-	Pulp and paper mill sludge	Jackson and Line (1997)

193	-	General	Jakobsen (1994)
250	400	Cattle manure	Hao et al. (2004)
284	-	Mixed waste	Lou (2008), Lou and Nair (2009)
2-380	46-942	Garden and biowaste	Boldrin et al. (2009)
0-521/1-228/0-1715/0-106	173-1873/89-298/475-2307/286-363	Biowaste/sludges/ livestock waste/Municipal waste	ADEME (2012)

Assuming a N content of 2% and P content of 0.4% in the final compost (compost usually varying between 0.4 to 3.5% total N and 0.2 to 1.5% total P (Harrison, 2008)) and a loss in mass of 45% (Tiquia et al., 2002), this would leave us with a range in emissions from production of 0.25 to 170 kg CO_2-eq./kg of N and 1 to 850 kg CO_2-eq./kg of P_2O_5 (excluding CO_2 emissions), these emissions going upwards to 850 kg CO_2-eq./kg of N and 1700 kg CO_2-eq./kg of P_2O_5 for composts with lower nutrient contents. It is important to understand that waste valorization through composting still results in GHG emissions, even though they may be less than other waste management alternatives such as landfilling.

This high variability in emissions from composting stem from the complex and inter-related nature of the various parameters influencing the process and how they can impact GHG emissions. Many operating parameters can have a drastic impact on GHG emissions during composting, such as the temperature, moisture content, aeration rate, pH, and carbon to nitrogen (C/N) ratio of the substrate. For example, CH_4 emissions were found to be seven times higher at 40°C then they were at 67°C by Ermolaev et al. (2015), while Cui et al. (2019) obtained a 90% reduction in cumulative N_2O emissions by using hyperthermophilic composting compared to traditional composting. Ermolaev et al. (2015) also noted other impacting factors, such as higher initial nitrate content leading to greater N_2O emissions due to denitrification. In a similar vein, Jiang et al. (2019) found a significant correlation between the C/N ratio of the substrate and the activity of the methanogens in the composting system, NH_4-N also inhibiting CH_4 oxidation by methanotrophs (Tlustos et al., 1998), highlighting the need to optimize C/N ratio to decrease CH_4 emission. This can be further impacted by the pH of the composting environment that has a direct link to ammonia volatilization and microbial activity (de Bertoldi et al., 1983; DeLaune et al., 2004), thus globally influencing emissions. Aeration also plays an important role, higher oxygen levels leading to greater oxidation of CH_4 and N_2O, but also increasing ammonia and CO_2 emissions (Oliveira et al., 2018; Shen et al., 2011). This relation being further complexified by the aeration method that has a drastic impact on the mass balances and biochemical transformations occurring within the system (Fernandes and Sartaj, 1997). When taking all of this together, and considering that many of these parameters impact one-another, it is clear how emissions from composting are, without a doubt, extremely variable in reality. Trying to capture this variability with a general emission factor will most likely misrepresent the actual emissions from the process, assessment of which should either be approached through uncertainty analysis or mathematical modelling (Walling et al., 2020).

3.4 Digestates

Digestates are nutrient-rich substances that can be used as organic fertilizers. They are a residual product from anaerobic digestion, also known as biomethanation or biogas production, which is the anaerobic degradation of organic matter in a controlled environment. Biogas is a gas that is rich in methane and carbon dioxide, being generally composed of between 40-75% CH_4 and 25-60% CO_2 (Ryckebosch et al., 2011). This gas can be used as a source of energy, or as a precursor for other chemical compounds, though currently most of it is used to produce energy.

The IPCC recommends a general emission factor of 1 kg CH_4/tonne of waste (5% generated CH_4) and considers N_2O emissions to be negligible (IPCC, 2006), though no references were found on how these values were obtained. Some countries also have their own EFs, though these are often based off the IPCC's guidelines and often vary around 5% of CH_4 production as well (Jeong et al., 2019). Most of the literature on emissions related to the anaerobic digestion process are from LCAs and not from actual emissions data, making it somewhat hard to paint a clear picture of emissions. Such works include those of Moller et al. (2009), Kaparaju and Rintala (2011), Bacenetti et al. (2013) and Li et al. (2018), providing ranges of -47 to -2900 kg CO_2-eq/tonne of waste. These negative ranges are due to most researchers considering emissions related to the production and combustion of the biogas as being biogenic or by comparing them to non-biogenic activities, such as landfilling. These comparative analyses thus allow for the obtention of "negative" emissions. Though this type of analysis can be useful to compare different alternatives, it does not provide actual emission factors. For example, Moller et al. (2009) estimated emissions from biogas combustion of 20 to 70 kg CO_2-eq./tonne of waste, while recent on-site studies at anaerobic digestion facilities have found that this value can be significantly higher. Indeed, Jeong et al. (2019) measured on-site emissions of about 180 kg CO_2-eq./tonne of waste, most of which came from the contribution of N_2O (0.57 kg N_2O/tonne of waste), significantly higher than the estimates used in many LCAs. In either case, CO_2 emissions were considered to be biogenic and not taken into account.

Furthermore, most of these works tend to minimize or completely neglect fugitive emissions from the reactors. These emissions can be an important source of GHGs and have been estimated as being anywhere between 3 and 4% of methane production (Flesch et al., 2011; Groth et al., 2015; Hrad et al., 2015). In the case of Jeong et al. (2019), who did not measure or take into account fugitive emissions, based on their data, these emissions could add another 50 kg CO_2-eq./tonne of waste to their total, bringing it up to approximately 230 kg-CO_2 eq./tonne of waste.

There is sadly an important lack of real and comprehensive on-site data for emissions associated to digestate production. Inventorying of emissions from anaerobic digestion have been neglected since process related emissions are considered biogenic. This limits our understanding of the actual production of GHGs. Table 6 presents the emission factors discussed in this section.

Table 6. Emission factors for digestate production.

Emission factor (kg CO_2-eq./tonne of waste)	Reference
25	IPCC (2006)
180	Jeong et al. (2019)
230	Based on Jeong et al. (2019) with added fugitive emissions

A summary of the factors influencing emission from organic fertilizer production is presented in Figure 1.

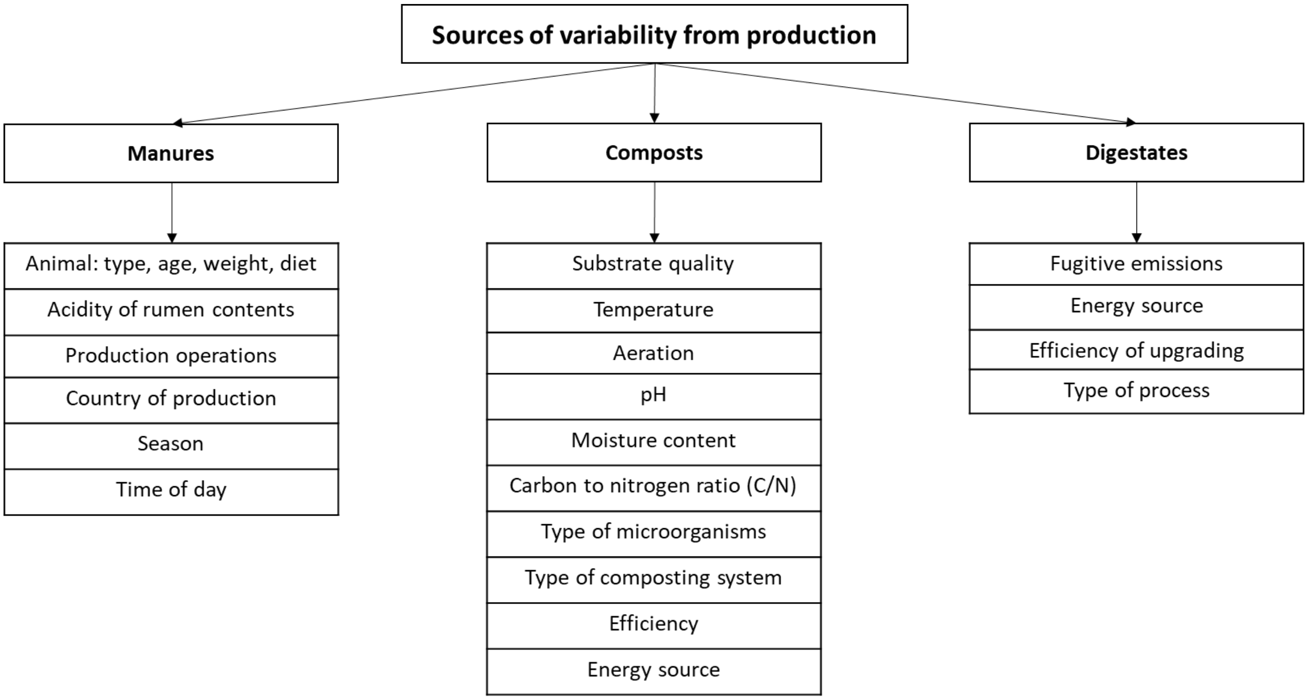

Figure 1. Sources of GHG emissions variability for organic fertilizer production.

4. Transportation

Transportation of fertilizers plays a major part in the global nutrient supply chain, though the reality of synthetic fertilizers is vastly different to the one of organic fertilizers. In the case of synthetics, the majority of P and K fertilizers are produced and exported from only a very few countries. Indeed, based on the information provided by the United States Geological Survey (USGS), China is the main global phosphorus producer, accounting for more than half of P-rock mining in 2018, followed by Morocco and the United States with about 10% each (in spite of Morocco having over 70% of the world's phosphate rock reserves) (USGS, 2019)**.** For potassium, Canada is responsible for 30% of potash production, followed by Russia, Belarus and China who produce between 14 and 17% each (USGS, 2019). Therefore, the supply chain for P and K fertilizers is a global one and will oftentimes lead to very large shipping distances. This can also be the case for N fertilizers, but the distances tend to be much shorter due to the more widespread production of these products. Indeed, shipping distances for N fertilizers have been reported as being around 500-1000 km in the United States (Smil, 2012) and Germany (Kathrin et al., 2017), and 1500 km in Brazil and China (de Lima et al., 2016; Zhang et al., 2013) for land-based transportation. Overseas shipping for P and K fertilizers can exceed distances of 10 000 km when considering potash exports from Canada to Brazil and China for example.

However, despite the narrative regarding the importance of transportation-related emissions, which is often used to promote alternatives to synthetic fertilizers, we did not manage to find information supporting this. Recent life cycle assessments of NPKs have found that emissions from transportation of these fertilizers are minimal when compared to production and application. The analysis by Hasler et al. (2015) determined that these emissions were only responsible for 1 to 3% of total emissions, while Kathrin et al. (2017) found even more conservative values of 0.5 to 0.9%. Both of these analyses focused on fertilizer use in Germany, but nevertheless include shipment over long distances by all three major modes of transportation (truck, train, sea). This is in line with the results of the analysis by Zhang et al. (2013) who determined that transportation was responsible for 0.75% of emissions related to N-fertilizer manufacture and use in China. Similar results have also been obtained for P-fertilizers (Albaugh et al., 2012). Therefore, the literature consistently indicates that transportation is a low contributor to the GHG emissions of the global synthetic fertilizer chain. This is especially apparent when compared to production and post-application emissions that often make up the near totality of the emissions profiles (Hasler et al., 2015; Kathrin et al., 2017; Zhang et al., 2013). Going forward, these emissions profiles are subject to change with the advent of greener production and transportation technologies, highlighting once again the benefit of taking variability into consideration.

This worldwide distribution network is, however, in stark contrast to the transportation chain for organic fertilizers that, given economic and technical restrictions, tend to be limited to a small radius around the production facilities. Estimates for feasible transportation distances for these amendments are around 30 km for manures (Paudel et al., 2009; Wiens et al., 2008) and up to 80 km for composts and digestates (Danso et al., 2006; Drosg et al., 2015). Despite the much lower nutrient content of organic amendments, which translates to the need for longer cumulative transportation distance on a nutrient basis, transportation was not found to be an important source of emissions and tends to be very low when compared with emissions from the other stages (Brown et al., 2010; Piippo et al., 2018; Timonen et al., 2019). Indeed, Timonen et al. (2019) report that emissions from the transportation of digestates are of about 2% of total emissions, while Piippo et al. (2018) report a range of 0 to around 5% of total emissions in the case of composting. This is consistent with Brown et al. (2010) concluding that "transportation [of biosolids] had relatively minor effects on overall emissions". As mentioned in the introduction, some studies have found relative important emissions from transportation (Nakamura et al., 2014), though this is more likely the result of a limited-scope analysis, neglecting emissions from production, storage and proper in-field emissions.

5. Storage

Another major step in the fertilizer production and application chain is the storage of fertilizing materials. This is negligible for inorganic fertilizers, which do not emit GHGs due to their chemical stability, but can be an important source of emissions for organic fertilizers. In the latter case, emissions will stem from the degradation of carbon and (de)nitrification of nitrogen to produce CH_4 and N_2O and is therefore highly variable and dependent on a variety of factors such as C/N content, temperature, and humidity. N_2O emission factors for manure storage can be found in the work of Broucek (2017), though they are not on a nutrient basis.

This variability is apparent when looking at table 7. Different storage strategies are obviously one of the main factors determining emissions. However, emissions are also affected by the type of organic matter, the temperature, the weather and the time of year. For example, Clemens et al. (2006) found that CH_4 emissions from uncovered liquid digestate were nearly 8 times higher over the summer than the winter, while Balde et al. (2016) obtained emissions 52 times higher in July than in January. These emissions are also variable within the same season, as highlighted by VanderZaag et al. (2010) who obtained a range of 23 to 35 g CH_4 m^{-3} d^{-1} for raw dairy manure during the summer. Furthermore, Maldaner et al. (2018) found daytime emissions to be twice as high as nighttime emissions during their study. Despite the potential benefits of different storage scenarios, this also remains uncertain. Closed systems are often presented as being preferable to mitigate emissions than opened ones, but studies such as that of VanderZaag et al. (2010) found no significant difference in GHG emissions between open and covered manure storage over a 162 day period. The researchers even noted a slight increase in CH_4 emissions, alongside a significant decrease in N_2O emissions in the covered system, cancelling out the benefits.

One trend does seem to be apparent, and it is that raw manures have the highest EFs. A study by Amon et al. (2006) investigating N_2O and CH_4 emissions from various storage scenarios for dairy cattle manure demonstrates the importance of storage on the emission profiles of these fertilizers. Their results indicated that digested slurry decreased emissions of about 60% when compared to untreated slurry, from 26.5 kg CO_2-eq./kg of N to 10.3 kg CO_2-eq./kg of N over an 80 day period (based on the initial N content provided by Amon et al. (2006) and assuming a slurry density of 1041 kg/m^3 (Lorimor et al., 2004)), mostly due to a major reduction (≈65%) in methane emissions. Digested slurry was found to have the lowest emissions compared to untreated, separated, straw covered and aerated slurry storage. Similar results have since been obtained, Maraseni and Maroulis (2008) noting a reduction of 60 to 85% in CH_4 emissions when comparing raw and digested manure storage, Balde et al. (2016) finding a reduction of volatile solids of 74%, and Maldaner et al. (2018) noting a decrease of 85% in these emissions. Lower mitigations have been observed by Kaparaju and Rintala (2011) and Holly et al. (2017), ranging from 12 to 27%, but the latter's digesters were poorly optimised, underscoring the uncertainty associated to the process efficiency. It is important to note though that the reduction in methane emissions is often accompanied by an increase in N_2O emissions that can mitigate the CH_4 reduction, presenting a further source of variability (Amon et al., 2006; Moller, 2015; Wang et al., 2014). Overall, it seems that digestate, from a storage perspective, offers significant benefits when compared to untreated manure storage, although it remains a significant source of emissions (Vaneeckhaute et al., 2018). It is also important to note that the design and implementation of the storage systems also have an important impact on the emissions from storage (Wang et al., 2014).

With respect to composting, the composting process is often used as a way to both store and treat manures and digestates, but these emissions are the same as the ones presented in Section 3. Only one study on the subject of emissions from matured compost storage was found. The study, conducted by Hao (2007), determined that CH_4 and N_2O were only released in trace amounts, between 0.001 and 0.053% of total nitrogen (at the beginning of storage) for N_2O and 0.00017% of total carbon for CH_4 emissions, while CO_2 emissions were much more significant at 2.9 to 10% of total carbon. Nevertheless, given the variability of results presented throughout this paper, further validation would be beneficial.

Table 7. Emission factors for storage (kg CO_2 eq./kg N/day)*

References	Waste type	Untreated manure	Separated manure	Straw cover	Aerated	Digestate	Compost	Storage time (days)
Amon et al. (2006)	Dairy slurry	0.33	0.14	0.42	0.19	0.13	-	80
Holly et al. (2017)	Dairy manure	<0.01	<0.01	-	-	<0.01	-	186
Maldaner et al. (2018)	Dairy manure	0.14	-	-	-	<0.01	-	365
Clemens et al. (2006)	Cattle slurry	0.01-0.02	-	-	-	<0.01	-	240
VanderZaag et al. (2010)	Dairy slurry	0.02	-	-	-	-	-	162
Balde et al. (2016)	Dairy manure and food waste	-	-	-	-	<0.01-0.04	-	730
Hao (2007)	Cattle manure	-	-	-	-	-	negligible	233

*Calculations assuming a nitrogen content of 3.25% for digestates when non-available.

6. Post-application

The final source of GHG emissions that we will focus on in this paper is the emissions following application of the fertilizing material to the soil. The in-field emissions of GHGs related to fertilizer application mainly stem from two processes, both of which are microbial in nature. The first major source of emissions is the (de)nitrification process that emits N_2O. These processes are controlled by many factors, including soil temperature, moisture, pH and the availability of mineral N and organic C (Beauchamp, 1997; Butterbach-Bahl et al., 2013). The other source of emissions are the anaerobic conditions that can be exacerbated by irrigation or rainfall, leading to the production of CH_4 by microorganisms. Methane emissions are especially prominent among semi-aquatic crops, such as rice paddies, with some estimates placing their contribution to global anthropogenic methane emissions as high as 17% (Bridgham et al., 2013). Despite the focus that is placed on production-related emissions when evaluating GHG emissions from fertilizers (especially valorized waste), post-application emissions can be the largest contributor to GHG emissions from crop systems, inorganic or organic, as was observed by Hasler et al. (2015) and Hoffman et al. (2018).

Though these processes occur naturally, the addition of fertilizers will impact the environment and can lead to significant changes in emissions. As will be explored in this section, the addition of nitrogen to the soil will provide a greater quantity of N that can be reduced during the denitrification process to N_2O, while addition of carbon through organic amendments can increase bacterial activity (energy), as well as being a substrate for methanogens (CH_4 producers).

There is also the impact of soil amendments on soil respiration and soil organic carbon sequestration. The addition of fertilizers, both inorganic and organic, has been linked to soil carbon in a variety of ways, though consensus is still far off. The meta-analysis by Jiang et al. (2018) determined that synthetic fertilizers greatly increased grain yield (91 to 184%), while having limited impact on soil organic carbon sequestration (4 to 16%) in China over the past 30 years, while organic amendments had a significantly lesser impact on grain yields (6 to 19%) but increase soil organic carbon by up to 39%. However, this benefit can potentially be mitigated by a decreased capacity for the soil to store methane (Gregorich et al., 2005) and increased N_2O emissions (Cui et al., 2012; Gu et al., 2017). The soil/carbon system is extremely complex and deserves a full review and analysis to itself, being far beyond the scope of this work, but we can point to some interesting literature for those who want a better understanding of the situation (Agegnehu et al., 2016; Banger et al., 2012; Eberwein et al., 2015; Khan et al., 2007; Liu and Greaver, 2010; Mahal et al., 2019; Mukumbuta et al., 2017; Oertel et al., 2016; Ramirez et al., 2010; Reid, 2008; Wang et al., 2018a; Yang et al., 2015; Zhang et al., 2016; Zhou et al., 2014). The following section will seek to explore post-application emissions from synthetic fertilizers (section 6.1) and organic amendments (section 6.2), while examining some of the causes behind their variability (section 6.3).

6.1 Synthetic fertilizers (nitrogen)

Following the IPCC guideline, and the one most commonly used throughout the literature, N_2O emissions from all fertilizers should be estimated at 1% of applied N (IPCC, 2006). However, the literature shows some varied results, our investigation finding emission factors as low as 0.03% and as high as 14% (see Table 8). Furthermore, whether these emissions increase linearly or non-linearly with respect to application rate remains a contentious area in the literature.

Indeed, literature investigating the linearity of post-application emissions has provided some very differing results. For example, many meta-analyses have produced linear models (Liu et al., 2013; Wang et al., 2011; Wang et al., 2018b; Yi et al., 2017), while many others have found emissions to increase non-linearly with application rates (Davis et al., 2019; Jiang et al., 2017; Kim et al., 2013; McSwiney and Robertson, 2005; Shcherbak et al., 2014). Furthermore, these models have been subject to the inherent variability witnessed throughout this paper; the global meta-analysis by Kim et al. (2013) concluding that "overall, across all the ranges of N input and agricultural land-use types […] examined in [their] study, no general pattern was identified to collectively describe the observed nonlinear dependency or linear dependency of direct N_2O emissions on N input rates", with similar comments from many of the other studies.

Table 8. N_2O emission factors for post-application of N-fertilizers.

Emission factor (% N_2O/kg N)	Emission factor (kg CO_2 eq./kg N)	Crop type	Soil type	Country/Region	Reference
0.03	0.09	Maize	Clay loam	Northeast China	Li et al. (2013)
0.04-0.06	0.11-0.18	Cotton	Sand	Northwestern China	Kuang et al. (2018)
0.03-0.12	0.09-0.36	Silage corn	Silt loam	Pacific Northwest	Collins et al. (2011)
0.15	0.45	Rice	Clay loam	South China	Chen et al. (2013)
0.17	0.51	Spring barley	Sandy loam	Denmark	Baral et al. (2017)
0.06-0.19	0.11-0.57	None	Silty-clay	Italy	Verdi et al. (2018)
0.15-0.21	0.45-0.63	Wheat and maize	Sandy loam	South China	Meng et al. (2005)
0.15-0.31	0.45-0.92	Almond	Sandy loam	California	Wolff et al. (2017)
0.1-0.4	0.30-1.19	Ryegrass	Sandyclay loam/clay loam	United Kingdom	Jones et al. (2005)
0.24	0.72	Rice	Sandy loam	Japan	Singla and Inubushi (2014)
0.26	0.77	Potato, sweet corn, winter wheat, sugar beet	Andisol	Japan	Koga (2013)
0.3	0.89	Pac choi, winter wheat and rice	Andisol, unspecified	Japan, South China	Akiyama and Tsuruta (2003), Yuan et al. (2017)
0.3-0.5	0.89-1.49	Winter wheat	Silt	France	Gu et al. (2013)
0.48	1.43	Welsh onion, winter wheat, summer maize	Cambisol, sandy loam	North China Plain	Yao et al. (2017), Ding et al. (2013)

0.5	1.49	Various, Rice	Sandy clay loam	Mediterranean, India	Cayuela et al. (2017)*, Das and Adhya (2014)
0.6	1.78	None	Silty clay loam	-	Saunders et al. (2012)
0.62	1.85	None	Sandy loam	-	Akiyama et al. (2004)
0.40-0.67	1.19-2.00	Cereals, pulses, millets and oilseed crops	Sandy loam and sandy clay loam	Northwest India	Jain et al. (2016)
0.17-0.69	0.51-2.06	Timothy grass	Loam and sandy loam	Eastern Canada	Chantigny et al. (2007)
0.68	2.03	None	Sandy loam	Northern Germany	Senbayram et al. (2009)
0.69	2.06	Various vegetables	Various soils	China	Wang et al. (2018b)*
0.7	2.09	Barley	Unspecified	Western Canada	Lemke et al. (2012)
0.82	2.44	Barley	Calcaric Cambisol	Central Spain	Meijide et al. (2009)
0.9	2.68	None	Loam	-	Bertora et al. (2008)
0.92	2.74	Various	Various	China	Lu et al. (2006)*
1	2.98	Various	Various	Germany, Global, China	Kaiser and Ruser (2000), Bouwman et al. (2002)*, Yang et al. (2015)
1.46	4.35	Cotton	Calcaric Fluvisol	Xinjiang	Tao et al. (2018)
1.49	4.44	Maize	Sandy loam	Central Spain	Meijide et al. (2007)
1.6	4.77	Maize, alfalfa	Silt loam	Pennsylvania	Adviento-Borbe et al. (2010)
1.80	5.36	Maize, none	Sandy loam	Central Spain	López-Fernández et al. (2007)
1.1-2.1	3.28-6.26	Spring barley	Sandy loam, loam, silt loam	Eastern Canada	Zebarth et al. (2008)
1.7	5.07	Various	Various	Eastern Canada	Gregorich et al. (2005)
1.9	5.66	Various vegetables	Silty clay loam	Southeastern China	Zhang et al. (2016)
2.07	6.17	Potato	Clay loam	Central Spain	Vallejo et al. (2006)
3.24	9.66	Corn	Silt loam	Northwestern Italy	Alluvione et al. (2010)
2.96-3.64	8.82-10.84	Komatsuna	Sand	Japan	Singla et al. (2013)
2.1-4	6.26-11.92	None	Sand	Netherlands	Velthof et al. (2003)
0.4-6.5	1.19-19.37	Various	Various	United Kingdom	Dobbie and Smith (2003)
2-7	5.96-20.86	Maize	Fine and coarse loamy	Northern United States	McSwiney and Robertson (2005)

1.55-8.93	4.62-26.61	Corn	Clay, loam	Eastern Canada	Chantigny et al. (2010)
0.03-10	0.09-29.80	Various	Various	Global	Kim et al. (2013)*
3.5-12.9	10.43-38.44	Corn	Andosol	Japan	Mukumbuta et al. (2017)

*meta-analyses

The studies presented in Table 8 were carried out for a variety of crops and soil types, without any apparent pattern emerging in terms of emissions, regardless of crop rotation, soil type, temperature, country, application method or climate. One point that we want to highlight is the potential importance of post-application emissions when compared to the other sources discussed in this paper. If we consider the IPCC recommended 0.01 kg N_2O-N/kg applied N, that is a CO_2 equivalent of approximately 3 kg CO_2 eq./kg applied N, we have a nearly identical emission factor from post-application as for production of a fertilizer like urea (see Table 1).

Though much of the focus on in-field emissions is placed on N_2O emissions, application of synthetic fertilizers can also increase methane emissions. Table 9 presents the data of a few studies on the subject. Note that these emission factors are in addition to the biogenic CH_4 emissions from soils and crops.

Table 9. CH_4 emission factors following N-fertilizer application.

Emission factor (kg CO_2 /kg of N)	Crop type	Reference
2.5	Rice	Yuan et al. (2017)
7.5	Rice	Das and Adhya (2014)
9.5	Rice, wheat	Yang et al. (2015), Zhao et al. (2015)

6.2 Organic amendments

The organic nature of manures, composts and digestates leads to some added complexity when considering emissions from their application. Organic fertilizers are reputed for their ability to increase soil nutrient availability, microbial activity and biodiversity by providing a wider range of compounds, such as humic acids and carbon, which are lacking in synthetic fertilizers (Jannoura et al., 2014; Yuan et al., 2017). These properties are generally seen as being beneficial for the soil and crops, though the increased microbial activity can also lead to an increase in emission.

This is especially the case when organic amendments are applied to paddy fields. Though the scope of the increase varies, studies have shown a significant increase in CH_4 emissions by a factor of about 2 to 3 when comparing manures to urea (Das and Adhya, 2014; Ren et al., 2017; Yang et al., 2015; Yuan et al., 2017; Zhang et al., 2019; Zhao et al., 2015). Though less pronounced, increased CH_4 emissions have also been observed in grasslands, being associated with more humid weather conditions (Jones et al., 2005; Ren et al., 2017; Wulf et al., 2002; Yang et al., 2015; Zhang et al., 2016). Composts and digestates also increase methane emissions compared to inorganic fertilizers, higher increases being related to higher carbon contents (Chadwick and Pain, 1997; Collins et al., 2011; Sasada et al., 2011; Severin et al., 2015; Singla and Inubushi, 2014; Verdi et al., 2018; Win et al., 2010). Emission factors for manures applied to paddy fields are presented in Table 10, in which it is again obvious that EFs are highly uncertain.

Table 10. CH_4 emission factors for manure application in paddy fields.

Emission factor (kg CO_2 /kg of N)	Manure type	Reference
6.5	Fowl manure	Yuan et al. (2017)
23.5	Poultry manure	Zhao et al. (2015)
0.9-46/0.35-60	Pig slurry/ dairy slurry	Chadwick et al. (2000)

For N_2O emissions, the situation is more complex. There is currently little consensus throughout the literature as to whether application of organic amendments increases or decreases nitrous oxide emissions when compared to synthetic fertilizers. Though this comparison is not very suitable given the different roles of mineral and organic fertilizers, it is still often undertaken in the literature. These studies highlight how variable information on emission factors is and the dangers of considering EFs as deterministic. Indeed, there is a division between studies reporting reduced N_2O emissions from fields after manure application and those reporting an increase in emissions compared to fields using synthetic fertilizers (see Table 11 and the supporting information for detailed information on emissions from manures, composts and digestates). Further confounding results have shown no difference between the two, which would be in-line with the IPCC's guidelines. There have also been mixed results, such as those of Cayuela et al. (2017) who's meta analysis found organic-liquid fertilizers had a much higher emission rate, but organic-solid fertilizers had the lowest emission rate. Such variation in results was previously addressed by Rochette et al. (2008) who attribute this difference to the variable composition of the manure and not its physical form. Another example is that of Chantigny et al. (2010) who obtained 60% higher emissions for N-fertilizers in clay soil crops, but 225% higher emissions for manures in loam soil tests. Table 11 presents results from studies comparing post-application emissions for three or more of the different types of fertilizers, while individual inventories of EFs for manures, composts and digestates can be found in the supporting information. There is once again no clear pattern or EF arising from the reviewed literature.

Table 11. N_2O emission factors from studies comparing emissions from N-fertilizers and organic fertilizers.

Emission factor (kg CO_2 /kg N) for synthetic N	Emission factor (kg CO_2 /kg N) for manure/slurry	Emission factor (kg CO_2 /kg N) for compost	Emission factor (kg CO_2 /kg N) for digestate	Application method	Reference
1.85	0.60-3.70	0.60	-	Broadcasting and incorporation	Akiyama et al. (2004)
5.36	1.37-3.78	1.50	-	Broadcasting and incorporation	López-Fernández et al. (2007)
9.66	6.88	0.33	-	Incorporation	Alluvione et al. (2010)
2.44	1.07	1.82	0.72	Subsurface	Meijide et al. (2009)
4.44	3.87	1.97-5.60[a]	2.68	Broadcast and incorporation	Meijide et al. (2007)
6.17	2.98	4.62	1.70	Broadcast	Vallejo et al. (2006)
0.09-0.36	0.27-0.33	-	0.15-0.30	Incorporation	Collins et al. (2011)
0.51	0.48-1.07	-	0.30	Subsurface	Baral et al. (2017)
0.51/2.06	0.89/3.67	-	0.42/1.19	Broadcast	Chantigny et al. (2007)
1.8	1.19-1.79	-	7.2-8.9	Broadcast and subsurface	Saunders et al. (2012)
2.1	11.0-13.4	-	3.3-6.0	Subsurface	Lemke et al. (2012)
2.7	7.7-14.3	-	5.4	Broadcasting and incorporation	Bertora et al. (2008)
4.62/26.61	10.51/16.27	-	8.3/17.6	Side-dressed	Chantigny et al. (2010)
0.18-0.57	-	0.06-0.75	1.9-15.2	Incorporation	Verdi et al. (2018)

[a]With added urea

6.3 Sources of post-application emissions variability

As demonstrated throughout the tables in section 6 and further emphasized with the information found in the supporting information, emissions from post-application in-field emissions of fertilizers can vary wildly and oftentimes without a clear trend. Such behavior has also been noted in some meta-analyses, such as Davis et al. (2019) whose results "highlight[ed] an inherent challenge to fully assessing N_2O

emissions” due to the variability in soil conditions, while Jiang et al. (2017) pointed out that their model was limited by “the large variance” of the data. Indeed, a plethora of sources can impact the variability of emission factors and greatly complexify their use in obtaining accurate representations of GHG emissions.

The complexity of these soil systems mirrors in many ways those highlighted during the composting process, both environments having many similarities. This includes the impact of temperature, moisture content, aerobic and anaerobic environments within the system, pH, and the physical and chemical characteristics of the amendments, among others. However, there are also added considerations regarding the application methods of fertilizers, the farming practices and general location specific factors, such as soil and climate conditions, that can all impact these emissions. The impact of these factors has been best demonstrated in meta-analyses, but even then, they remain uncertain. For example, Cayuela et al. (2017) found significant differences in N_2O EFs for crops in the Mediterranean, identifying several parameters that impacted these EFs. These included irrigation methods, with differences of up to 340% between the lowest (rain-fed, 0.27% of applied N) and the highest (sprinkler irrigation, 0.91% of applied N). Significant differences were also noted between extensive and intensive crops, while little differences were found between the types of fertilizer applied, with the exception of liquid slurries. Precipitation has also been identified as one of the main factors in EF variability, as focused on in the study by Dobbie and Smith (2003) and the meta-analysis by Lu et al. (2006). Other factors include tillage, soil type and soil temperature, as well as the timing of fertilizer application (Chadwick et al., 2011; Chantigny et al., 2010; Omonode et al., 2011; Rochette et al., 2008)

Despite certain tendencies in the scientific community, such as expecting broadcasted slurries to have the highest EFs, the data presented in tables S2 to S4 demonstrate how variable these emissions are, without any clearly visible trends. For example, Baral et al. (2017) reported an EF of 0.48 to 1.07 kg CO_2 eq/kg N for the injection of pig and cattle slurry, while Jones et al. (2005) reported an EF of 1.5 to 7.7 kg CO_2 eq/kg N for broadcasting of solid pig manure. This is a prime example of how variable these emissions can be, given how, generally speaking, injection of liquid manures is expected to have significantly higher emissions than broadcasting of solid manures. All of this once again demonstrates the difficulty of properly representing emissions through general emission factors.

7. General Discussion

7.1 The global picture

Through our research, the following has been highlighted regarding GHG emissions:

- EFs for synthetic N production range from 1 to 10 kg CO_2-eq./kg of N depending on the type of fertilizer and the source of hydrogen used by the steam reforming process. EFs for phosphorus fertilizer production are generally around 1-1.5 kg CO_2-eq./kg of P_2O_5, while knowledge on emissions from potassium fertilizer production is greatly lacking.
- Emissions for compost production can be very variable, ranging from 1 to 2300 kg CO_2-eq./tonne of waste, or about 1 to 850 kg CO_2-eq./kg of N or 1 to 1700 kg CO_2-eq./kg of P_2O_5. Information on emissions from anaerobic digestion is greatly lacking and needs further study. The generally assumed emission factor is 5% of produced CH_4, with another estimated 3 or 4% in fugitive emissions and some N_2O emissions that are often omitted in assessments.
- Emissions from transportation were found to be generally very low for all types of fertilizers, especially relative to production, storage and post-application.
- Emissions from storage of digestates and manures can be high, especially for longer storage times. Storage of liquid digestates vs liquid manures significantly decreases CH_4 emissions, though the increase in N_2O emissions can mitigate the benefit. Emissions from the storage of synthetic fertilizers or compost were found to be negligible to non-existent, though information on compost storage is extremely limited.
- For post-application emission, EFs for synthetic fertilizers were found to range between 0.03 and 12.9% of applied nitrogen (0.1 - 40 kg CO_2 eq./kg of N) and 0 to 10 kg CO_2 eq./kg of N for methane emissions. Emissions from manures varied between 0.05 and 13.9% of applied N, generally being higher than those of synthetics, and had higher methane emissions. Due to these ranges, manure was found to have the highest potential emissions of any of the studied amendments. EFs for composts were between 0.11 and 1.55% of applied N, while digestates ranged from 0 to 5.1% of applied N.

Overall, the main and most consistent finding is the widespread variability between the emission factors that are used to undertake environmental assessments. Though we have focused on the IPCC’s guidelines, many studies also found major differences with country or even locally specific emissions data. The reality is that predicting GHG emissions from generalized EFs has not proven to be accurate. The production and supply chain explored throughout this paper are highly variable and it appears that emission factors are not sufficient to represent them without taking the risk of having important errors in emissions estimates.

Awareness of this variability and its sources can be the first step to ensuring proper and accurate environmental impact analysis. Figure 2 presents a conceptual overview of the various factors influencing emissions from the different sources discussed throughout this paper. This includes factors such as the source of energy used and the type of fertilizer being produced for synthetic fertilizers, while emissions from organic fertilizer production and storage are highly dependent on the waste source used, the operating conditions (aeration, pH, temperature, moisture) and the process efficiency. Most complex of all are post-application in-field emissions which are affected by the crop and soil type, previous crop rotations and fertilizer regimes, the soil nutrient and organic matter content, the time and weather, the amount of fertilizer applied (potentially non-linear), the method of fertilizer application, the soil ecosystem, interactions with other amendments, soil pH, and irrigation (though this list is likely not an exhaustive list). There is also the added dimension of crop yield that can greatly impact the results of a fertilization scenario analysis (Linquist et al., 2012). Beyond this awareness though, it will be important to take measures to either avoid variability or mitigate its impact. Using long-term case-specific readings (data from the actual case being studied) would obviously be the best solution, though it would be unfeasible for many analyses, especially those aiming at assessing new alternative fertilizers. Undertaking field work and getting case by case measurements would be costly and time consuming, but it might be worth it. This is especially highlighted by works such as that of Brodt et al. (2014) that performed an LCA with direct field measurements and the IPCC Tier 1 methodology; the Tier 1 methodology overpredicting emissions by nearly 250% in their case. Given these limitations, this review can serve as a guide for potential ranges of EFs that can be useful for uncertainty analyses.

Indeed, at a minimum, uncertainty analyses should be undertaken to provide awareness and some level of information on the potential ranges of emissions and their consequences; proper variability and scenario analysis being key to environmental decision-making (Walling and Vaneeckhaute, 2020). These analyses must be undertaken on a case by case basis and the emissions assessments should not be generalized given how case-specific the sources of variability are. This has been supported by a growing body of work that has demonstrated the impact of these approaches when assessing emissions. Multiple studies have shown that conclusions of analyses and, in some cases, resulting decisions can be completely changed by taking this uncertainty into account. This includes an evaluation of biosolids processing and end-use by Brown et al. (2010) that found certain scenarios transitioned from significant net carbon sinks to net emitters when comparing low and high ranges of emissions. Similarly, Chen and Corson (2014) noted that the uncertainty and variability in emissions data had enough of an impact to change decisions between certain alternatives for manure management in their LCA of French dairy farms.

Despite the potentially confusing conclusions that can arise from these analyses, they are necessary to get a more realistic portrait of the situation. Furthermore, some recent works have highlighted the benefits that uncertainty assessments can bring, notably to future research. This has been especially pertinent to highlight areas that should be of particular focus when undertaking environmental assessments. For example, Wolf et al. (2017b) combined a local and global sensitivity analyses on an LCA to determine which parameters were essential to assessing GHG emissions from milk production, while Meyer-Aurich et al. (2012) noted that despite the uncertainty of certain emissions "dominat[ing] the variability of GHG emissions of the whole process", this knowledge could be used to gain understanding of the process and focus on these highly variable areas to minimize their impact.

Finally, modelling the emissions from these different fertilizers could provide a more valuable estimate, though these models require the ability to represent many of the interconnected and poorly understood phenomena taking place.

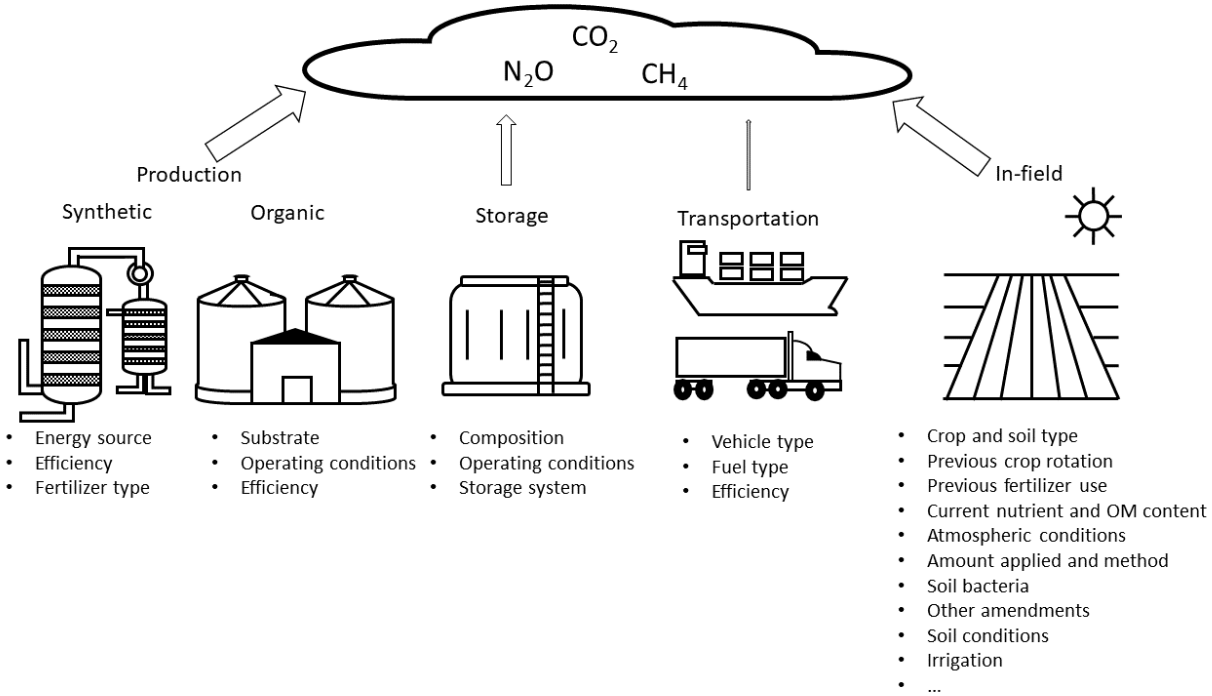

Figure 2. Factors influencing GHG emissions form fertilizer production and use. The arrows represent the general importance of each stage to the global emissions profile for fertilizer production and use. Production and post-application in-field emissions will make up the large majority of total emissions, followed by storage and transportation, the latter being generally very low. OM represents organic matter.

7.2 Mitigation and management practices

Despite the variability in emissions data, important amounts of GHGs are nevertheless being emitted and will contribute to global warming. Given the necessity of fertilizers in modern agriculture, it is important to find ways to mitigate emissions related to their production and use.

Regarding production of nutrients, the main source of emissions is the high energy requirement for ammonia synthesis and the energy required for P and K production. This could potentially be reduced by a variety of ways, such as through improving heat transfer and energy efficiency (Flórez-Orrego and de Oliveira Junior, 2017; Panjeshahi et al., 2008; Rafiqul et al., 2005), using alternative, non-fossil, energy sources (Arora, 2017; Tallaksen et al., 2015; Tock et al., 2015; Tunå et al., 2014), or even transitioning towards emerging photocatalytic and electrocatalytic ammonia synthesis processes (Chen et al., 2017; Michalsky et al., 2012; Montoya et al., 2015; Zhou et al., 2017a).

Better energy management can also reduce emissions from composting and anaerobic digestion. Tracking and reducing methane leaks from anaerobic digesters could also greatly mitigate CH_4 emissions. These processes can further be supplemented by CO_2 capture and sequestration processes (Walling et al., 2019), especially for biogas production, seeing how it already implements carbon capture for biogas upgrading (see Salomoni et al. (2011) and Vo et al. (2018) for examples). Furthermore, nutrients in digestates can be concentrated or used to produce mineral fertilizers (Vaneeckhaute et al., 2017), potentially reducing emissions on a nutrient basis. Though not thoroughly discussed, emissions from manure production can also be reduced by modifying the diets of the livestock and through genetic selection of low emission producing breeds (Boadi et al., 2004; Herrero et al., 2016; Thornton and Herrero, 2010).

Though relatively low, emissions from transportation can also be decreased using renewable energy sources, as well as optimized delivery routes to reduce distance and redundancies. For storage, composting or digesting manures to reduce CH_4 emissions is recommended. Maintaining digested manures in airtight environments can also reduce N_2O emissions. Closed storage systems for both manures and digestates can also be supplemented with biogas capture and valorization to significantly reduce methane emissions (Chianese et al., 2009).

Finally, for in-field emissions, the first recommendation is obviously to follow fertilizer recommendations for the respective crop and soil type and not to overapply fertilizers, which leads to increased emissions and other environmental consequences. Furthermore, a slew of recommendations were provided by

Snyder et al. (2009) and we highly recommend readers consult this work. These recommendations include properly timing the application of fertilizers with crop uptake demand, considering the specific nutrient needs for different crops, using inhibitors to decrease NH_3 and N_2O emissions, and evaluating NO_3-N levels in field drainage to adjust application, just to name a few. There is also a common guideline to improve nutrient use efficiency, the 4R Nutrient Stewardship guideline, that focuses on promoting the use of the right fertilizer source at the right rate at the right time and in the right place (Johnston and Bruulsema, 2014)

8. Conclusion

Following this review of emissions from organic and inorganic fertilizer production and use, the main observation is a high and pervasive variability of emissions data throughout the literature. Despite the common and widespread use of general emission factors, a large portion of the evaluated studies reported significantly varying results. This variability has many causes and highlights the complexity of the production and use chain of fertilizers. The most important recommendation for future analyses resulting from this review is the need to have case-specific emission factors of GHGs from waste management and agricultural sectors, where possible. Uncertainty analyses on the results can also be performed. Beyond this, simply being aware of this variability and the uncertainty it can cause for environmental assessments is a good first step. Nevertheless, it is still possible to provide recommendations for GHG mitigation, notably through the use of cleaner energy sources and CO_2 valorization upon fertilizer production, proper storage practices, and better management of fertilizer application.

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Supporting information for: Greenhouse gas emissions from inorganic and organic fertilizer production and use: A review of emission factors and their sources of variability

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The following tables provide a more comprehensive overview of emission factors for post-application emissions from manures, composts and digestate individually. Studies comparing manures and synthetic N-fertilizers are also more common than the ones comparing digestates or composts, the results of some of these studies are presented in table S1.

Table S1. Results from studies comparing post-application in-field N_2O emissions from N-fertilizers and manures.

Emission factor (kg CO_2 eq/kg N) for synthetic N	Emission factor (kg CO_2 eq/kg N) for manures/slurries	Reference
0.09	0.21-0.66	Li et al. (2013)
0.11-0.18	0.33-0.45	Kuang et al. (2018)
0.09-0.36	0.27-0.33	Collins et al. (2011)
0.51	0.48-1.07	Baral et al. (2017)
0.89	1.22-3.67	Akiyama and Tsuruta (2003)
0.89	0.60	Yuan et al. (2017)
0.3-1.19	0.60-7.75	Jones et al. (2005)
1.43	4.02	Yao et al. (2017)
1.49	0.57-2.53	Cayuela et al. (2017)*
1.78	1.19-1.79	Saunders et al. (2012)
1.85	0.6	Akiyama et al. (2004)
2.09	11.03-13.41	Lemke et al. (2012)
0.51/2.06	0.89/3.67	Chantigny et al. (2007)
2.44	1.07	Meijide et al. (2009)
2.68	7.75-14.51	Bertora et al. (2008)
2.98	2.38	Bouwman et al. (2002)*
2.98	8.05	Kaiser and Ruser (2000)
4.35	4.29	Tao et al. (2018)
4.44	3.87	Meijide et al. (2007)
4.77	7.15	Adviento-Borbe et al. (2010)
5.07	0.89-5.07	Gregorich et al. (2005)
5.36	1.37-3.78	López-Fernández et al. (2007)
6.17	2.98	Vallejo et al. (2006)
9.66	6.88	Alluvione et al. (2010)
6.26-11.92	1.5-41.4	Velthof et al. (2003)
4.62/26.61	10.51/16.27	Chantigny et al. (2010)

Table S2. Post-application in-field N_2O emission factors for manure

Emission factor (kg CO_2 eq/kg N) for manures/slurries	Manure type	Crop type	Soil type	Application method	Reference
0.27-0.33	Dairy manure	Corn	Silt loam	Incorporation	Collins et al. (2011)
0.33-0.45	Composted cattle and sheep manure	Cotton	Sand	Broadcasting and incorporation	Kuang et al. (2018)
0.21-0.66	Pig manure	Maize	Clay loam	Unspecified	Li et al. (2013)
0.60	Fowl manure	Winter wheat, rice	Unspecified	Unspecified	Yuan et al. (2017)
0.72	Green manure	Wheat, maize	Sandy loam	Broadcast and incorporation	Meng et al. (2005)
0.48-1.07	Pig slurry, cattle slurry	Spring Barley	Loamy sand	Subsurface	Baral et al. (2017)
1.07	Pig slurry	Barley	Calcaric Cambisol	Subsufrace	Meijide et al. (2009)
1.19-1.79	Dairy manure	None	Silty clay, silt loam	Broadcast and subsurface	Saunders et al. (2012)
2.38	Various	Various	Various	Various	Bouwman et al. (2002)*
0.57-2.53	Various	Various	Various	Various	Cayuela et al. (2017)*
0.15-2.71	Cattle slurry	Ryegrass	Sandy loam	Unspecified	Rubæk et al. (1996)
0.36-1.31/0.36-2.89	Pig slurry/dairy slurry	Ryegrass	Sandy loam	Broadcasting	Chadwick et al. (2000)
1.22/3.67	Swine manure/poultry manure	Pac choi	Andisol	Broadcasting and incorporation	Akiyama and Tsuruta (2003)
2.98	Pig slurry	Potato	Clay loam	Broadcasting	Vallejo et al. (2006)
0.60-3.7	Poultry litter, cattle manure	None	Sandy loam	Broadcasting and incorporation	Akiyama et al. (2004)
1.37-3.78	Sheep manure, pig slurry	Maize, none	Sandy loam	Broadcasting and incorporation	López-Fernández et al. (2007)
0.89/3.67	Liquid swine manure	Timothy grass	Loam and sandy loam	Broadcast	Chantigny et al. (2007)

3.87	Pig slurry	Maize	Sandy loam	Broadcasting with incorporation	Meijide et al. (2007)
4.02	Poultry manure	Welsh onion, winter wheat	Cambisol	Incorporation	Yao et al. (2017)
4.29	Cattle manure	Cotton	Calcaris Fluvisol	Broadcasting with incorporation	Tao et al. (2018)
0.89-5.07	Various	Various	Various	Various	Gregorich et al. (2005)
3.19/4.02/5.16	Poultry manure/cattle manure/pig manure	Various	Various	Various	Zhou et al. (2017b)*
6.88	Winter leguminous green manure	Corn	Silt loam	Incorporation	Alluvione et al. (2010)
7.15	Liquid dairy manure	Maize, alfalfa	Silt loam	Broadcast	Adviento-Borbe et al. (2010)
0.6-1.5/1.5-7.7	Cattle slurry/poultry manure	Ryerass	Sandy clay loam, clay loam	Broadcast	Jones et al. (2005)
8.0	Various	Various	Various	Unspecified	Kaiser and Ruser (2000)
9	Poultry manure	Sweet corn	Sandy loam	Incorporation	Heller et al. (2010)
11.0-13.4	Swine manure	Barley	Unspecified	Subsurface	Lemke et al. (2012)
7.7-14.3	Pig slurries	None	Loam	Broadcasting with incorporation	Bertora et al. (2008)
10.52/16.27	Liquid swine manure	Corn	Clay, Loam	Side-dressed	Chantigny et al. (2010)
1.5-5.6/5.4-9/21.7-41.4	Poultry manures/cattle slurry/liquid pig manure	None	Sand	Incorporation	Velthof et al. (2003)

Table S3. Post-application in-field N_2O emission factors for composts.

Emission factor (kg CO_2 eq/kg N) for compost	Compost type	Crop type	Soil type	Reference
0.33	Cattle manure, Municipal solid waste	Potato, sweet corn, winter wheat, sugar beat, soybean	Clay loam, silt loam	Koga (2013), Alluvione et al. (2010)
0.60	Plant residues	None	Sandy loam	Akiyama et al. (2004)
0.06-0.75	Municipal solid waste	None	Silty-clay	Verdi et al. (2018)
1.16	Agricultural waste	Winter wheat, summer maize	Silt loam	Ding et al. (2013)
1.49	Municipal solid waste	Maize, none	Sandy loam	López-Fernández et al. (2007)
1.82	Municipal and garden waste	Barley	Calcaric Cambisol	Meijide et al. (2009)
4.62	Pig slurry	Potato	Clay loam	Vallejo et al. (2006)
1.97-5.60[a]	Pig slurry	Maize	Sandy loam	Meijide et al. (2007)

[a]With added urea

Table S4. Post-application in-field N_2O emission factors for digestates.

Emission factor kg CO_2 eq/kg N) for digestate	Biogas influent	Crop type	Soil type	Application method	Reference
0.15-0.30	Dairy manure	Corn	Silt loam	Incorporation	Collins et al. (2011)
0.21	Unspecified	Rice	Sand	Broadcast	Singla and Inubushi (2014)
0.3	Unspecified	Spring Barley	Loamy sand	Subsurface	Baral et al. (2017)
0.72	Pig slurry	Barley	Calcaric Cambisol	Subsurface	Meijide et al. (2009)
0.00-1.04	Cattle slurry	Ryegrass	Sandy loam	Unspecified	Rubæk et al. (1996)
0.42/1.19	Liquid swine manure	Timothy grass	Loam and sandy loam	Broadcast	Chantigny et al. (2007)
1.70	Pig slurry	Potato	Clay loam	Broadcast	Vallejo et al. (2006)

2.7	Pig slurry	Maize	Sandy loam	Broadcast and incorporation	Meijide et al. (2007)
3.84-5.33	Pig slurry	Melon	Sandy clay loam	Incorporation	Sanchez-Martin et al. (2010)
5.36	Pig slurries	None	Loam	Broadcasting and incorporation	Bertora et al. (2008)
3.3-5.96	Swine manure	Barley	Unspecified	Subsurface	Lemke et al. (2012)
7.63	Maize	None	Sandy loam	Subsurface	Senbayram et al. (2009)
0.77	Pig slurry	Rice	Clay loam	Irrigation	Chen et al. (2013)
7.2-8.9	Dairy Manure	None	Silty clay, silt loam	Broadcast and subsurface	Saunders et al. (2012)
9.98-11.59	Unspecified	Komatsuna	Sand	Unspecified	Singla et al. (2013)
1.90-15.23	Pig slurry	None	Silty-clay	Incorporation	Verdi et al. (2018)
8.28/17.6	Liquid swine manure	Corn	Clay, Loam	Side-dressed	Chantigny et al. (2010)

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