Article

Meta-Analysis of Strategies to Reduce NH₃ Emissions from Slurries in European Agriculture and Consequences for Greenhouse Gas Emissions

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Abstract: The intensification of livestock production, to accommodate rising human population, has led to a higher emission of ammonia into the environment. For the reduction of ammonia emissions, different management steps have been reported in most EU countries. Some authors, however, have criticized such individual measures, because attempts to abate the emission of ammonia may lead to significant increases in either methane, nitrous oxide, or carbon dioxide. In this study, we carried out a meta-analysis of experimental European data published in peer-reviewed journals to evaluate the impact of major agricultural management practices on ammonia emissions, including the pollution swapping effect. The result of our meta-analysis showed that for the treatment, storage, and application stages, only slurry acidification was effective for the reduction of ammonia emissions (−69%), and had no pollution swapping effect with other greenhouse gases, like nitrous oxide (−21%), methane (−86%), and carbon dioxide (−15%). All other management strategies, like biological treatment, separation strategies, different storage types, the concealing of the liquid slurry with different materials, and variable field applications were effective to varying degrees for the abatement of ammonia emission, but also resulted in the increased emission of at least one other greenhouse gas. The strategies focusing on the decrease of ammonia emissions neglected the consequences of the emissions of other greenhouse gases. We recommend a combination of treatment technologies, like acidification and soil incorporation, and/or embracing emerging technologies, such as microbial inhibitors and slow release fertilizers.

Keywords: ammonia reduction; climate change mitigation; greenhouse gases; liquid manure management; meta-analysis; pollution swapping

1. Introduction

One-third of the earth's arable land resources have already been severely degraded due to pollution through eutrophication, carbon and nutrient losses through soil management, and contamination with agrochemicals [1]. Ammonia (NH₃) and its salts are major contributors to eutrophication, and corresponding abatement strategies might lead to increased greenhouse gas (GHG) emissions [2]. Agriculture, mainly livestock production, is the greatest contributor to the emission of ammonia (NH₃) [2–4]. The GHGs from animal husbandry are nitrous oxide and methane. The latter is mainly produced by enteric fermentation and manure storage, and has an effect on global warming 28 times higher than carbon dioxide [5]. The general demand for measures to abate emissions of NH₃ due to the

rise in livestock production has led to the development of various manure management strategies, at different stages of the manure management chain (animal diets and housing, manure treatment, storage, and application) [3,6]. Livestock production is projected to double in the next century to accommodate the growing world population [7], which is expected to reach 9 billion by 2050 [8]. The consequence of these projected assumptions is a continued depletion of the environment due to NH_3 and GHG emissions. NH_3 is one of the major atmospheric pollutants leading to the deterioration of human health and ecosystems [9]. For example, apart from the eutrophication and acidification tendencies of NH_3 , the nitrification of ammonium (NH_4^+), which can be formed from NH_3 , may also contribute to ground water contamination [9], through nitrate leaching [10], nitric oxide (NO), and nitrous oxide (N_2O) emissions [11]. Large parts of the NH_3 emitted into the atmosphere form particulate matter (PM) [12] which have the potential to reduce human life expectancy via respiratory and cardiovascular diseases [13]. The estimated reduction of life expectancy due to exposure to particulate matter, in the Benelux region in 2005, accrued to an average of 9.0 months and 8.5 months in the EU-28 [14].

Due to the transboundary nature of NH_3 and GHGs, international cooperation is required to meet the air quality guideline values of the World Health Organization (WHO) [15], e.g., 20% of the nitrogen deposition on nature areas in the Netherlands emanates from Germany [14]. The sensitivity of agricultural practices to NH_3 and GHG emissions has paved the way for new policies, leading to changing trends and technologies compared to the old practices. New policies are still emerging [3,16]. For example, the new Danish regulation on pig production mandates pig farm owners to implement NH_3 abatement strategies, such as acidification and air-scrubbing techniques [17]. In France, various manure treatments and land application techniques are part of the national implemented nitrate and water framework directive [18]. For Spain, a study by Sanz-Cobena et al. (2014) showed that the implementation of the most effective region-specific mitigation strategy led to 63% NH_3 mitigation at the country level, compared to the reference situation of standard practices for the year 2008 [19]. Results for the UK were presented by Misselbrook et al. (2010), showing 231.8kt of NH_3 from livestock production and fertilizer used for 2009 [20].

In the EU-27 more than 1400 million tons of slurry is produced yearly, with a high percentage applied as fertilizers to agricultural lands [17,21]. A study by Ma et al. [22] on N use efficiency in China, reported that only 12% of the applied N was utilized by plants; 47% was lost to atmospheric emissions, and 41% to ground water, through leaching [23].

Research efforts are in place for the abatement of NH_3 emissions at the various stages of the manure management chain, namely; the feeding, housing, treatment, storage, and application stages to ensure a whole-farm management model [2,7]. However, the impact of these mitigation strategies on the emission of other GHGs has not received equal attention. Most studies have just focused on a single gas mitigation effort [2,11]. A few studies, where such impacts have been highlighted, pose contradictory results, and hence need to be accurately assessed [24]. A comprehensive analysis of the effects of NH_3 abatement strategies on emissions of different GHGs is so far missing. Therefore, our meta-analysis can contribute to the question of how the environmental impact of accepted mitigation strategies for NH_3 influences the emissions of N_2O , CH_4 , and CO_2 .

Furthermore, Webb et al. [23] highlighted that abatement efforts to suppress the emission of gases at initial stages of manure management do not guarantee complete abatement, but will only transfer the conserved gases to a later stage, thereby making the management of manure increasingly important as one moves down the chain. Seidel et al. [25] and Sajeev et al. [6] reported that manure storage, manure treatment, and field application are the major contributors to the total NH_3 emitted in European agriculture. Furthermore, at these last three stages the pollutant swapping effect plays a major role. Therefore, we investigated the consequences of the most effective measures to decrease NH_3 emissions, for the emission of other GHG (swapping effect).

2. Materials and Methods

2.1. Selection of Studies

A systematic literature search was conducted on the databases of Scopus, Google scholar, and gray literature, and from the websites of governmental/non-governmental agencies by combining the following keywords: ammonia emission, ammonia abatement, Europe, abatement strategies, integrated assessment, systematic review, meta-analysis, air quality, long-range transboundary air pollution (LRTAP), livestock, livestock management, animal housing, manure treatment, animal diet, Europe, France, Belgium, Germany, Netherlands, Spain, UK, Sweden, Finland. The keyword search and screening of the abstracts led to the identification of 289 documents with information on NH_3 abatement strategies, and a subset of 142 articles with information on abatement strategies across 11 EU countries in the period from 2008 to 2018. Due to the incomplete description in some papers, and lack of response from contacted authors (only 4 out of 19 contacted authors responded), only the papers listed in Annex I were included in the meta-analysis. A list of all studies used for the different treatment strategies can be found in the Table S1.

2.2. Slurry Treatment Methods

Based on the results of the literature research the following treatment methods were most common, and they are briefly described in this section. Slurry acidification is mainly achieved using sulfuric acid, with a pH-value between 5.1 and 6.5 [18]. Biological treatment means the conversion of slurry to biogas for energy purposes (anaerobic digestion) [2]. By separation slurry components are separated into the liquid and solid fractions, mainly by mechanical measures [6]. The use of covers as a further mitigation strategy is carried out to reduce the overall emitting surface, by the concealment of the liquid slurry with different materials like plastic, straw, or saw-dust [18]. Field application techniques of slurry can be divided into different categories, injection, incorporation, and band application [23]. Injection is mainly realized at a depth of 15 cm below the surface, while in incorporation the slurry is only covered by a few centimeters of soil [23]. With band spreading application technique, liquid manure, or fertilizers in general, are applied below crops in a concentrated layer in the soil, commonly 8–15 cm below the surface. This technique is considered as more efficient because of the uniform application of manure compared to surface application [23]. Furthermore, band application means application of fertilizers in a “band”, often 5 cm over and 5 cm below from the seed during planting [26].

2.3. Data Analysis

The treatment methods for NH_3 abatement included in this study were acidification, biological treatments (anaerobic digestion), and separation. They were compared to the worst practices, to identify the reduction potential (in percentage) attributed to each treatment strategy. In addition, the meta-analysis covered the aspects of storage (use of covers) and field application techniques. The number of selected papers and corresponding repetitions in relation to the different management strategy are shown in Table 1. The effectiveness of the treatments, reported as mean percentage reduction, was considered the primary outcome of interest for both NH_3 and GHGs. CO_2 released through biodegradation was considered carbon neutral. Biogenic carbon, which is not degraded in soil, increases the soil carbon pool and can be considered a carbon sink [6,23]. The considered time frame is of high importance, because, e.g., in 20 years less carbon will be degraded compared to 100 years. To assess the results of the different studies, the considered time frame must be the same. As these time frames are not indicated in most of the articles used in this study, a deeper assessment of total relevant greenhouse gas emission fluxes is not possible. Due to the higher global warming potential of CH_4 and N_2O , the release of those substances has a higher impact on global warming (Table S2).

All treatments were compared to a reference (no slurry treatment, no storage cover, and band spread application) to quantify the effect (Table 2). The percentage reduction in emissions (outcome variable) for each of the reported gases was calculated by comparing the differences between the abatement

strategy/treatment relative to the reference strategy/control. Negative values indicate a reduction, while positive values indicate an increased emission. To analyze the combined effect of different measures, taking sample sizes into account, testing for moderators, and obtaining corresponding forest plots, the meta-analytical software OpenMEE was used (<http://www.cebm.brown.edu/openmee>). A random effect model was used to aggregate the collected data into a meta-analysis, to satisfy the assumption of variance heterogeneity. We refrained from calculating the combined effects of the different abatement strategies, because in some of the studies only relative values were published and it was not possible to obtain the absolute values. The efficiency of greenhouse gases must be combined with their lifetimes to form a global warming potential (GWP) e.g., 20 or 100 years. A table with the different GWPs and atmospheric lifetimes is provided in the Table S2.

Table 1. Number of selected publications and number of observations per management strategy included in the meta-analysis. The numbers in brackets represent the total number of observations from all studies.

Strategy	Reference Method	Number of Studies (Number of Observations)			
	(worst practice)	NH ₃	N ₂ O	CH ₄	CO ₂
Treatment	No Treatment				
Acidification		14 (103)	5 (15)	9 (33)	9 (41)
Biological		4 (16)	7 (26)	7 (30)	3 (14)
Separation		8 (42)	6 (25)	6 (29)	8 (36)
Storage					
Cover	No cover	3 (25)	3 (10)	5 (30)	3 (28)
Application					
Injection	Surface incorporation	3 (12)	2 (7)	2 (5)	2 (16)
Injection	Surface application	3 (8)	2 (19)		
Injection	Band spread	2 (30)		1 (3)	
Band Application	Surface application	3 (5)	3 (6)	1 (2)	1 (2)
Incorporation	Surface application	4 (28)	4 (40)	3 (12)	2 (12)

Table 2. Results of the meta-analysis of greenhouse gas (GHG) emission changes, following reduction strategies across different manure treatment strategies (numbers in brackets represent the 95% confidence intervals). Negative values represent emission reduction, and positive values show increase in emission.

Strategy	Reference	Mean Emission Changes in Percentage (%)			
	Method	NH ₃	N ₂ O	CH ₄	CO ₂
Treatment					
Acidification	no treatment (raw slurry)	−77 (−81, −73)	−21 (−27, −15)	−86 (−92, −80)	−15 (−19, −11)
Biological	no treatment (raw slurry)	−14 (−27, −1)	+10 (−10, +30)	−46 (−65, −27)	−26 (−42, −10)
Separation	no treatment (raw slurry)	−1 (−13, +11)	+24 (−4, +52)	−27 (−46, −8)	+59 (−23, +95)

Table 2. Cont.

Strategy	Reference	Mean Emission Changes in Percentage (%)			
Storage					
Cover	no cover	−68 (−81, −55)	+13502 (−25, +39600)	−16 (−33, +1)	−31 (−45, −17)
Application					
Injection	Surface application	−61 (−89, −31)	+196 (−39, +1365)		
Injection	Surface incorporation	−19 (−100, +155)	+133 (−54, +300)	−23 (−34, −13)	−20 (−50, +7)
Incorporation	Surface application	−68 (−96, +3)	+61 (−77, +412)	−37 (−167, +117)	+4 (−50, +33)
Injection	Band spread	−51 (−84, −14)	+3 (−50, +36)		
Band application	Surface application	−33 (−53, −4)	+25 (−2, +57)	+153 (+108, +197)	+18 (+10, +25)

3. Results and Discussion

3.1. Impact of Acidification in Reference to Raw Slurry

The acidification of slurry material during storage and soil application has been reported by various authors to decrease NH_3 emissions by shifting equilibrium between NH_3 and NH_4^+ , with low pH favoring NH_4^+ dominance [16,21,26]. Furthermore, acidification is used to reduce the emissions of CH_4 [16,27,28], N_2O [29,30], and CO_2 [24,26,31,32].

In most of the studies acidification was achieved using sulfuric acid, with a pH-value between 5.1 and 6.5. Slurries amended with acids affected N-dynamics by delaying nitrification, and consequently N_2O emission [21,33]. Acidified slurries applied to agricultural lands inhibited the emission of NH_3 , but also, released concurrently large amounts of NH_4^+ [21]. Due to the delay in the nitrification activities of microbes as a result of acidification, which would have otherwise led to an increased formation of NO_3^- (with high leaching tendency), a successful NH_3 and N_2O inhibition was observed [7,21]. The activities of methanogens were also hampered when slurries were amended with acid, which reduced the possibility for their formation and consequently CH_4 and CO_2 emission [21,34].

In this meta-analysis results for the acidification of animal manure from 37 studies, with 192 measurement events, showed that acidification treatment was effective for the reduction of NH_3 , and had no pollution swapping effect with other GHGs (Table 2). For NH_3 , an average reduction in emission of −77% (CI −81, −73) was observed. Methane showed reductions of up to −86% (CI −80, −92), N_2O was reduced by −21% (CI −15, −27), and CO_2 had a reduction of up to −15% (CI −11, −19) (Figure 1).

Figure 1. Forest plot of the impact of the acidification of animal slurry with reference to raw slurry. The black square boxes show the individual mean points of each gas. Horizontal bars indicate the 95% confidence intervals.

3.2. Impact of Biological Treatment Relative to Raw Slurry

The conversion of slurry to biogas for energy purposes leads to the production of CH_4 and CO_2 prior to the storage of manure, leading to a reduction in GHG emission potentials in the subsequent management chain [35]. The available studies, however, show contrasting results. Some reports have related the emission of CH_4 to the quantity of total solid (TS) and volatile solid (VS) present in manure [18,36]. Digestate, the material remaining after the anaerobic digestion of a biodegradable feedstock, contains less TS and VS compared to raw slurries, and hence has less easily degradable carbon (C), compared to raw undigested slurry materials [37]. Rodhe et al. [38] attributed high production of CH_4 in digestate, relative to non-digestate, to the presence of more anaerobes, present as a result of TS conversion during and after anaerobic digestion, converting lignocellulose materials present in the soil to CH_4 . Perazzolo et al. [37] also associated the reduced CO_2 emission to low carbon content (TS and VS), by comparing solid separated manure (with a higher TS and VS) to the liquid manure (with low TS and VS), and concluded that low CO_2 emission can be expected from digestate with low TS and VS, compared to non-digestate. Results from this present study showed an average reduction of -46% (CI -65 , -27) in CH_4 , and -26% (CI -42 , -10) in CO_2 (Figure 2). It is also important to note that subsequent production of CH_4 and CO_2 can be provoked during storage when full methanogenesis is not achieved in anaerobic digestion, and that this can also lead to even more CH_4 and CO_2 in digestate compared to non-digestate during the land application of manure [39,40].

Figure 2. Forest plot of the impact of biological treatment of animal slurry with reference to raw slurry. The black square boxes show the individual mean points of each gas. Horizontal bars indicate the 95% confidence intervals.

Changes in manure characteristics are also expected following digestion of manure, which may provoke simultaneous emissions of other gases. Digestion is known to increase slurry pH [37,38], temperature [41], and N mineralization [37], leading to possible increase in NH_3 and N_2O emissions. Crust formation (which can mitigate NH_3 emission) is decreased by the gasification and removal of CH_4 and CO_2 , which may lead to an increased NH_3 emission [2,7].

For biological treatment the results showed an aggregate NH_3 emission reduction of -14% (CI -27 , -1) (4 studies and 16 observations). In contrast, Nyord et al. [41] attributed an increased NH_3 emission following anaerobic digestion to the increase of pH and total ammoniacal nitrogen (TAN), and backed up their findings with similar earlier reports [42,43]. For N_2O , biological treatment leads to an average increase of 10% (CI -10 , 30) (7 studies and 26 observations). The result of the experiment by Regueiro et al. [32], showed a significant decrease in NH_3 and N_2O emissions when the digestate was acidified prior to storage. The activities of microbes are also reduced at low pH-values, and thus acidification may prove to be useful for the hindrance of methanogens (pH-value below 6); moreover, one of the requirements for nitrification is a neutral to slight alkaline condition. For both CH_4 (-46%) and CO_2 (-26%) biological treatment leads to a pronounced reduction of gas emissions (Figure 2).

3.3. Effect of Separation of Slurry into Solid and Liquid Fractions

The calculated emission changes reflect an aggregate of both, solid and liquid fractions to obtain an overall impact of separation on gaseous emissions. Slurry separation, as a treatment strategy, aims at trapping nutrients (nitrogen, phosphorous) in the various separated fractions [44,45] for an enhanced management and storage capacity (easy infiltration of liquid fraction, and a concentration of organic matter in the solid fraction) [23].

Some studies reported a small to medium (about -10%) reduction of NH_3 emissions due to manure separation during storage [24,37,42]. Regarding manure separation during application, Dinuccio et al. [2,46,47] and Regueiro et al. [32] highlighted an increased emission. The mean reduction in NH_3 emission for the present meta-analysis was very limited, i.e., -1% (CI -13 , $+11$). N_2O and CO_2 emissions were increased by $+24\%$ (CI -4 , $+52$) and $+59\%$ (CI -23 , $+95$), respectively, while a reduction of -27% (CI -46 , -8) was achieved for CH_4 (Figure 3). Although soils amended with liquid manure tend to induce a higher crop yield, due to increased N-infiltration compared to untreated soils [23,48], some authors did not recommend separation as an effective treatment strategy without combining it with other treatment strategies, for the simple reason of increased net GHG emissions [47].

Figure 3. Forest plot of the impact of separation of animal slurry with reference to unseparated slurry. The black square boxes show the individual mean points of each gas. Horizontal bars indicate the 95% confidence intervals.

To highlight the importance of additional measures to this strategy, Hjorth et al. [43] showed a combination of separation and anaerobic digestion as an optimized version for manure management; NH_3 emission was largely reduced as well as the amount of VS due to separation. Acidification of slurry following separation has also proven to be more effective than simple separation in reducing the amount of N losses [32,37,49].

Increase in the emission of N_2O [50] and CO_2 [37] due to separation of manure, especially from the liquid fraction relative to the untreated slurry and solid fraction, has been reported. These reports are in line with the outcomes of this present study. Separating manure is perhaps the abatement measure with the lowest impact on GHG mitigation potential [51].

3.4. Use of Covers for Manure Storage

The use of covers as a mitigation strategy is carried out to reduce the overall emitting surface [52]. The concealing of animal slurry with different material (plastic, straw, saw-dust, etc.) has been identified as an efficient mitigation strategy for the abatement of NH_3 emission [53,54]. Emission reductions for NH_3 , CH_4 , and CO_2 were achieved by -68% (CI -81 , -55), -16% (CI -33 , 1), and -31% (CI -45 , -17) (Figure 4), respectively, while an increase in emission was noticed for N_2O , by several orders of magnitude ($+13502\%$ (CI -25 , $+39600$)).

It is also important to note that other environmental factors, such as temperature, can impact the emission potentials of these gases [2,31]. In addition, soil structure [6], crop type [42,43], and the composition of the slurry [35,39] strongly influence the emissions.

Figure 4. Forest plot of the impact of the use of covers during storage of animal slurry. The black square boxes show the individual mean points of each gas. Please note that N₂O is missing in the forest plot due to the extreme values +13502% (CI −25, +39600). Horizontal bars indicate the 95% confidence intervals.

Similar results were found in a meta-analysis carried out by Hou et al. [2] and Sajeev et al. [6] for NH₄, CH₄, and N₂O, although their results did not include CO₂ emissions.

Covering material allowed for nitrification/denitrification activities, which may have led to higher N₂O emission in the overhead space of storage materials, which subsequently bled out upon exposure [35,55,56]. The high confidence interval of N₂O emissions can possibly be explained by different cover materials, from non-permeable (plastic) to semi-permeable (e.g., saw dust), but the real explanation of the process behind this is not extractable from the studies.

Airtight coverage mitigated both NH₃ and N₂O emission. Findings by Sommer and Dahl [57] stated that compaction leads to increased temperature, and hence doubling of the emission of NH₃, in composting of deep litter like buried crop residues. Air tight covers can mitigate N₂O emissions in slurries with longer slurry age where nitrification has occurred. A possible scenario analysis for reduced N₂O emission would be to prolong air-tight coverage, to ensure that the denitrification of already nitrified gas has gone to completion, wherefore, only N₂ gas will be emitted. A major concern in this case would be the effect of N removal from slurry material on crop yield when slurries are used as fertilizers.

CO₂ was also reduced in all included studies (−31% (CI −45, −17)). This reduction has been attributed to an increased resistance to mass flow of CO₂ by covering the materials [31].

3.5. Effects of Different Field Application Methods

Field application techniques were divided into different categories, injection (normally 15 cm depth), band application, and incorporation, to allow for the inclusion of some important findings in the literature, and to pitch various application techniques against one another (Table 2). A range of 30–50% of NH₃ emissions in the EU emanates from the application of livestock slurry [58–60]. Land application of organic fertilizers increases the amount of metabolizable-C in the soil. This leads to increased microbial activity, as evidenced by increased CO₂ due to microbial respiration [23,24], and the subsequent consumption of oxygen due to the increase in density of microbial biomass. This eventually leads to anaerobic denitrification whereby N₂O is produced [24].

While the emission of NH₃ is reduced after land application of liquid manure by injection, relative to surface application, N₂O is increased due to soil microbial N-mineralization and nitrification/denitrification processes. This is evidenced by the high concentration of NH₄⁺ and subsequent oxidic NO₃⁺ increase as NH₄⁺ depletes, ultimately leading to an increasing amount of N₂O production under anaerobic conditions [48]. Some of the pre-requisite processes enabling these nitrification/denitrification activities include; (i) the presence of metabolizable fatty acids and degradable C in slurry material, which increases the activities of soil micro-organisms [60,61]; and (ii) O₂ depletion with increasing application depth, or as a result of an increased microbial activity, leading to an anaerobic environment which favors denitrifiers [60]. The results of the meta-analysis showed a reduction in NH₃ emission by −61% (CI −89, −31) and an increased N₂O emission by +196% (CI −39, +1365) when injection was compared to surface application.

A similar emission trend was also noticed for NH_3 and N_2O , with changes in the emissions of -19% (CI -100 , $+155$) and $+133\%$ (CI -54 , $+300$), respectively, when injection of manure was compared to surface incorporation. There is no information in the individual studies explaining the wide range of results, presumably caused by different soil types or meteorological conditions. The same technique leads to an average decrease of CH_4 (-23% (CI -34 , -13)) and CO_2 (-20% (CI -50 , $+7$)).

Surface application followed by immediate or delayed incorporation follows the same nitrification/denitrification process as mentioned above. The importance of application depth for the reduction of N_2O has also been pointed out for the concurrent reduction of both NH_3 and N_2O . Velthof and Mosquera [61] highlighted that with increasing depth, complete denitrification of N_2O to N_2 will be achieved before the gas reaches the surface. Nevertheless, full denitrification is complicated, and might lead to additional N losses in form of N_2O instead of N_2 . A comprehensive review by Almaraz et al. (2020) showed that for a better understanding of how denitrification rates vary across the soil profile, there is a need to more precisely investigate ecosystem N budgets. Therefore, surface and subsoil measurements should be included in future studies to determine controls on denitrification in surface soils vs. subsoils [62]. Fangueiro et al. [24] detected no N_2O emission when slurry material was injected to 30 cm depth. Furthermore, the authors also pointed out that a further N_2O reduction can be achieved for manure incorporation when soils are amended with acidified slurry in order to trigger nitrification inhibition as a result of slurry acidification. Manure incorporation, with reference to surface application with no incorporation, showed an emission reduction potential for NH_3 of -68% (CI -96 , $+3$), and an increase in N_2O emissions of $+61\%$ (CI -77 , $+412$), highlighting how no-incorporation affected N-dynamics. Infiltration of manure into soil was reduced in no-tillage management compared to a conventional tillage system, where surface exposure to air is minimized, resulting in a better N use efficiency (NUE), due to minimized environmental loss of N [62]. Moreover, urease activity is higher in the topsoil, and manure-soil contact and interaction is optimal, when the manure component is incorporated into the soil [63,64].

While promoting injection of fertilizers as the best manure application method for reduced NH_3 emissions [6,25,41], surface application using splash plates has been banned in most European countries because of its strong impact on NH_3 emission [23]. However, injection is accompanied by other flaws, such as the cutting of roots (especially of perennial crops) by injection materials, its impracticability on stony and sloped lands without the use of stronger tractors [23], and the great draught force hindering working perimeter [25,41].

In the band spreading application technique, fertilizers are applied below crops in a concentrated layer in the soil, commonly 8–15 cm below the surface, and are more efficient because of the uniform application of manure compared to surface application. Comparing the emission of gases from band application (applications of fertilizers in a “band” often 5 cm over and 5 cm down from the seed during planting) with reference to injection, the results of the meta-analysis showed an NH_3 reduction of -51% (CI -84 , -14), and an increase in N_2O by $+3\%$ (CI -50 , $+36$). Webb et al. [23] similarly concluded that the band application of fertilizers can be an efficient compromise when injection cannot be used. Bourdin et al. [65] showed that the band application of fertilizer, while conserving and elevating the concentration of NH_4^+ in the soil, may at the same time inhibit CH_4 oxidation due to competition for methanogenic enzymes by both NH_4^+ and CH_4 . They concluded that when N is not volatilized, it remains in the soil where it carries out its inhibitory actions on CH_4 oxidation. Moreover, anaerobic sites are created and, consequently, CH_4 gas is emitted when fertilizers are applied in bands. These findings are in line with the outcomes of this study. A reduction of -33% (CI -53 , -4) was achieved for NH_3 while increased N_2O , CH_4 , and CO_2 were noticed, $+25\%$ (CI -2 , $+57$), $+153\%$ (CI $+108$, $+197$), and $+18\%$ (CI $+10$, $+25$), respectively. Hansen et al. [55] stated that the use of additional draught to enable injection led to an increased CO_2 emission. Bourdin et al. [65] and Fangueiro et al. [24] attributed the high production of CO_2 to an initial increase in microbial respiration, following the application of metabolizable-C present in manure material (Table 2).

4. Conclusions

The results of the meta-analysis showed a reduction in NH_3 emission in all management strategies, but revealed mixed results for other GHGs; highlighting further that the management strategies were aimed at reducing the emission of NH_3 , while not considering the effect on GHGs. Thus, apart from acidification of manure, all management strategies affect N_2O negatively. In addition, strategies to conceal manure (e.g., covering during storage, injecting, and incorporating manure during application), led to increased N_2O emissions. This is possibly a result of the denitrification of already nitrified manure N-components, when oxygen is in limited supply as manure is concealed. Additional procedures, such as acidification, or the use of nitrification and urease inhibitors, are effective ways of ensuring reduced N_2O emission, so that nitrification is delayed, or by acidification which obstructs the activities of microbial communities. A positive effect (reduced emission) was noticed in all strategies for CH_4 , except during band application, with reference to the surface application of fertilizers. Band application, compared to surface application, may also affect CO_2 negatively.

The effect of all application processes on CO_2 emission have remained unclear so far, as authors have attributed its emission to the initial increase in the activities and population of soil microbial communities in response to the introduction of substrates present in manure. Although the review by Webb et al. [23] claimed that readily degradable C will be in short supply after the storage of solid manure, and hence will have less impact on microbial metabolism. We attempted to relate CO_2 emission to soil profile (microbial presence with application depth), but a comparison between incorporation and surface application foiled the attempt, as an increased emission is noticed. Although it can still be argued that the incorporation depth, when not as deep as the injection depth, may have an increased presence of microorganisms, compared to the presence at the soil surface.

The separation of manure, although effective for CH_4 reduction, had a negligible impact on NH_3 , while N_2O and CO_2 emissions were increased. We therefore conclude that although this treatment strategy is effective for crop-manure response and yield, this treatment is the least effective for NH_3 emission reduction, and at the same time has a negative impact on other GHGs (N_2O and CO_2) [47], and should only be used with an additional strategy, like slurry acidification, that is effective for the reduction of NH_3 emissions and has no pollution swapping effect with other greenhouse gases.

Supplementary Materials: The following are available online at www.mdpi.com/xxx/ Table S1: List of the studies used in the meta-analysis, Table S2: Greenhouse Gas Global Warming Potentials and Atmospheric Lifetimes

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