

Greenhouse Gas and Ammonia Emissions from Different Stages of Liquid Manure Management Chains: Abatement Options and Emission Interactions

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Abstract

Farm livestock manure is an important source of ammonia and greenhouse gases. Concerns over the environmental impact of emissions from manure management have resulted in research efforts focusing on emission abatement. However, questions regarding the successful abatement of manure-related emissions remain. This study uses a meta-analytical approach comprising 89 peer-reviewed studies to quantify emission reduction potentials of abatement options for liquid manure management chains from cattle and pigs. Analyses of emission reductions highlight the importance of accounting for interactions between emissions. Only three out of the eight abatement options considered (frequent removal of manure, anaerobic digesters, and manure acidification) reduced ammonia (3–60%), nitrous oxide (21–55%), and methane (29–74%) emissions simultaneously, whereas in all other cases, tradeoffs were identified. The results demonstrate that a shift from single-stage emission abatement options towards a whole-chain perspective is vital in reducing overall emissions along the manure management chain. The study also identifies some key elements like proper clustering, reporting of influencing factors, and explicitly describing assumptions associated with abatement options that can reduce variability in emission reduction estimates. Prioritization of abatement options according to their functioning can help to determine low-risk emission reduction options, specifically options that alter manure characteristics (e.g., reduced protein diets, anaerobic digestion, or slurry acidification). These insights supported by comprehensive emission measurement studies can help improve the effectiveness of emission abatement and harmonize strategies aimed at reducing air pollution and climate change simultaneously.

Core Ideas

- Emission reductions of abatement options were quantified in the manure management chain.
- Significant emission interactions exist in manure management chains.
- Single-stage emission abatement needs to shift to a whole-chain perspective.
- Understanding variability of results minimizes uncertainty of emission reductions.
- A holistic management strategy is required to tackle air pollution and climate change.

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LIVESTOCK production and rearing is a major source of ammonia (NH₃) and greenhouse gas (GHG) emissions. In 2014, the livestock sector contributed a share of 53% methane (CH₄), 21% nitrous oxide (N₂O), and 75% NH₃ to total agricultural emissions in the European Union (EU-28) (UNECE, 2016; UNFCCC, 2016). The negative impacts of these emissions on the environment have been widely reported. Accumulation of GHGs in the atmosphere causes climate change, which leads to undesirable consequences like sea level rise, increase in extreme weather events, and losses in food production (Stocker et al., 2013). Ammonia, on the other hand, poses a threat to terrestrial and aquatic systems and contributes to the formation of particulate matter in the atmosphere, which is harmful for human health (Sutton et al., 2011). Ammonia volatilization and subsequent atmospheric deposition is also a source of indirect N₂O emissions (De Klein et al., 2006). Given the projected increase in demand for livestock products (Steinfeld et al., 2007), there is a pressing need to abate emissions from livestock.

Among the many sources associated with emissions from the livestock sector, emissions from manure management are prominent. Emissions from manure management include emissions from animal housing, during the handling and storage of manure, grazing, and the application of manure as a fertilizer to soils. Manure management alone accounts for 31% of GHGs and almost all of NH₃ emissions from livestock in the EU-28 (UNECE, 2016; UNFCCC, 2016). Policies are in place to tackle emissions from manure management in EU-28. In terms of NH₃, national policies reflect a response to obligations accepted with the Gothenburg Protocol under the Convention on Long-Range Transboundary Air Pollution, or the National Emission Ceilings Directive. Ammonia emissions are also affected by indirect policies that are not necessarily aimed at reducing NH₃ but influence them, such as the Directive on Integrated Pollution Prevention and Control, the Common Agricultural Policy, the Nitrate Directive, and the Water Framework Directive. The Kyoto Protocol and the 20-20-20 targets under the Climate and Energy Package focus on reducing GHG emissions. The Common Agricultural Policy and Nitrate

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Abbreviations: CO₂e, carbon dioxide equivalents; CP, Crude protein; EU, European Union; GHG, greenhouse gas.

Directive also influence agricultural decisions and thereby affect emissions of GHGs. These policies are informed by scientific research, highlighting their role in regulating emissions from manure management.

Many studies have analyzed and evaluated the potential of abatement options to reduce NH_3 and GHGs (Ivanova-Peneva et al., 2008; Sistani et al., 2010; Rodhe et al., 2012; Dai and Blanes-Vidal, 2013; Madrid et al., 2013). Although these studies are valuable in estimating the emission reduction potentials of abatement options, they do suffer from some drawbacks. The majority of literature focuses on the abatement of a single gas, be it NH_3 or GHGs (CH_4 and N_2O expressed in carbon dioxide equivalents [CO_2e]). This is problematic, since there are significant interactions between emissions and abatement options, whereby abating a single pollutant might lead to increases in the other (i.e., pollution swapping). Although, the idea of emission interactions and their effect on the ability of abatement strategies to reduce multiple pollutants has been discussed in literature (Brink et al., 2005; Anthony et al., 2008; Wagner et al., 2011; Winiwarter and Klimont, 2011; UNECE, 2015; Wagner et al., 2015), a systematic evaluation of emission interactions in the management of manure is still amiss. Second, abatement options are usually recommended for a single stage within the entire manure handling and management chain. Although this is important, it is often found lacking in a broader context, since reduction of emissions at one stage might lead to emissions further down the chain. Only a few studies address this issue. Such studies recommend a whole-chain or mass-flow approach in evaluating abatement strategies (Sommer et al., 2009; Chadwick et al., 2011; Del Prado et al., 2013). An analysis of literature related to emission abatement in manure management suggests that only some studies used a whole-chain approach inclusive of emission interactions (Gooday et al., 2014; De Vries et al., 2015; Hou et al., 2015, 2017; Wang et al., 2017).

The present study identifies potential abatement options to reduce GHGs and NH_3 emissions collectively. It also sheds light on the potential cobenefits and the issue of pollution swapping by determining the trends in interactions of CH_4 , N_2O , and NH_3 among the various abatement options identified. The deficiencies in single-pollutant, single-stage approaches when devising abatement strategies are also highlighted, along with potential solutions and the way forward in tackling these deficiencies. Such a comprehensive analysis is necessary to set realistic emission targets and to ensure the effectiveness of proposed emission abatement strategies in the management of manure.

Methodology

A meta-analytical approach was used to determine emission reductions for the different manure abatement options. The section below describes the methodology used to define the system boundaries, selection of data sources, and estimation of emission reductions.

Manure Management Chain and Emission Abatement Options

Livestock production and management is a source of emissions. In this paper, we focus on those activities that are related to manure management, as these processes are strongly related. The

manure management chain consists of various stages, from animal feeding to the application of manure to soils. Animal feeding is included, since it alters manure characteristics and subsequently affects emissions through the entire manure management chain. Each stage is associated with emissions of the relevant gases (CH_4 , N_2O , or NH_3) in varying amounts and rates. However, in each stage, emissions can be influenced by management practices (e.g., frequent removal of manure, covering manure, etc.) or by technological intervention (e.g., low protein animal diets, manure acidification, installation of scrubbers, etc.). Here, we employed a literature review to identify prominent abatement options associated with different manure management stages. The manure management chain, along with the abatement options considered, is illustrated in Fig. 1. In our selection of abatement options, we took advantage of the United Nations Economic Commission for Europe's (UNECE) recommendations (Bittman et al., 2014) and the recent literature (Del Prado et al., 2013; Montes et al., 2013; Oenema et al., 2014), which allowed us to arrive at a set of consistent abatement options. Manure-related emissions include emission fluxes from urine and feces excreted in animal housing, during storage and further treatment, and after application to agricultural soils. Direct CH_4 emissions from animals, specifically "enteric fermentation," are not included in the current analysis, as these would not entail manure management. Also, since the focus of this study pertains to housed animals, emissions from grazing were excluded. Furthermore, we take advantage of the clear separation between liquid and solid manure systems. This paper covers the interrelations of the liquid manure systems for the animal types (pigs and cattle) that contribute to a majority of manure emissions, but the analysis did not distinguish between them. Limiting the scope to only liquid manure management rules out abatement strategies such as slurry separation, as this would entail managing emissions from the solid part of the separated slurry in downstream stages during storage and application of solid manure. Although this is sufficient for the objectives of this study, a comprehensive analysis inclusive of all abatement options for both solid and liquid manure with a greater detail of separation on the basis of animal type is important to fully understand the potential of emission abatement options for manure management.

Selection of Data Sources

Emission reduction potentials for the identified abatement options were estimated using effective observations from published literature according to the methods described in recent studies that focus on emission abatement in manure management systems (Hou et al., 2015, 2017; Wang et al., 2017). The selection criteria were as follows: (i) the animal category was either cattle or pigs; (ii) the study was subject to at least one of the eight chosen abatement options; (iii) the study measured and reported either NH_3 and/or GHG emissions for at least one of the manure management stages of housing, treatment, storage, or application; (iv) the study included data on reference treatments and base emissions; and (v) the article was peer reviewed and available in English. This resulted in a selection of 89 peer-reviewed papers and reports measuring NH_3 and/or GHG emissions from on-farm and experimental farm settings. This corresponded to 465 individual observations, which were used in the analysis. The reference systems, abatement options,

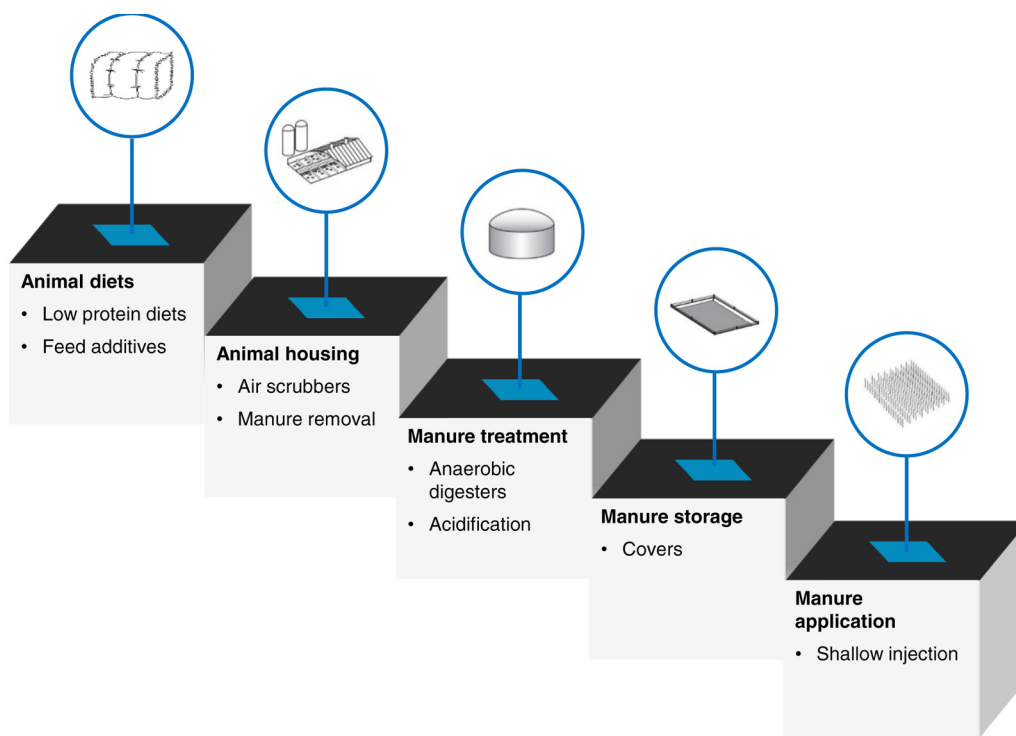


Fig. 1. Stages in the manure management chain and associated abatement options considered in the meta-analytical approach.

and number of emission observations are reported in Table 1. A full list of studies separated by manure management stage and animal type for all abatement options, along with the emission reductions and references, is provided in the supplemental information (Supplemental Tables S1 to S5).

Estimation of Emission Reduction Potentials and Interactions

Emission reductions for abatement options were calculated relative to a reference system. The emission reductions were derived according to Eq. [1]:

$$AE = (E_{\text{abt}}/E_{\text{ref}} - 1)100 \quad [1]$$

where AE is the emission abatement efficiency (%), E_{abt} refers to emissions with abatement measures, and E_{ref} is the emissions under the reference scenario. The units for the emissions corresponding to the reference scenario and with abatement measures vary depending on the measurement methods and units used in individual studies. This allowed for a normalization of emission reductions in percentage terms to arrive at relative changes caused by an abatement option to the different gases, even if only

few studies considered the effect of an abatement option on all three of them (CH_4 , N_2O and NH_3). A negative value of the emission abatement efficiency indicates a decrease in emissions due to the abatement option used. It is worth mentioning that even though this study aims to quantify the emission reductions over the whole manure management chain, the overall emission reductions calculated for a particular abatement option is an average of a multitude of emission reduction estimates typically measured at a single manure management stage or occasionally covering multiple stages. For example, in the case of feeding, the overall emission reduction is an average of measurements reported at housing, storage, and application stages. Since variability in emission trends could arise between stages, the emission reductions from individual stages from which the overall emission reductions are calculated while using a particular abatement option has been explicitly mentioned in the text.

Statistical Analyses

Descriptive statistics were used to quantify and describe the emission reductions. Means and SDs were calculated and reported for all existing datasets. High variability occurred in several instances, with SDs exceeding the mean value such that

Table 1. Number of emission observations, reference system, and list of abatement options included in the meta-analytical approach

Abatement stage	Reference system	Abatement options	Sample size (observations)		
			N_2O	CH_4	NH_3
Feeding	High protein feeds	Reduced crude protein	11	10	72
		Additives	3	6	19
Housing	Liquid manure housing systems	Air scrubbers	5	5	23
		Frequent removal	4	4	5
Treatment	No treatment	Anaerobic digesters	26	7	17
		Acidification	6	18	33
Storage	No cover	Covers	18	27	50
Application technique	Surface spreading	Shallow injection	22	–	77

reported trends may be merely indicative. The use of complex statistical models was not possible due to a small sample size. This is in line with the primary focus of the study to compile descriptive arguments for observed trends and interactions between various abatement options on GHGs and NH₃, rather than estimate emission reductions on an absolute scale.

Results

The section below quantifies the emissions reductions from various abatement options for the manure management chain. The abatement options and emission reductions are categorized according to various stages involved in manure management, as illustrated in Fig. 1. As outlined above, emission reductions were estimated using either single-stage or multistage measurements corresponding to the functioning of the abatement option and included the analysis of all three gases (CH₄, N₂O, and NH₃) collectively. Table 2 reports the average emission reductions, along with the SDs, for the selected abatement options. The emission reduction estimates indicate variability, which in some cases can be attributed to specific reasons and should not be taken as blanket uncertainty. Variations exist, as measurements have been made at different stages of the manure management chain, using different methods, and as the reduction estimates are derived from various scales ranging from laboratory to field. Other influencing factors related to specific abatement options that affect emission reductions and induce variability will be discussed in the sections below.

Feeding Strategies

Manipulating animal diets has implications on emissions from livestock manure (Oenema et al., 2014). Reducing the dietary crude protein (CP) content of animal diets and the inclusion of feed additives such as fiber, enzymes, and acids are some of the abatement options reported to lower gaseous emissions from the manure management chain.

Reduced Crude Protein

Ammonia emissions decrease with a reduction in dietary CP levels. (Oenema et al., 2014). Reduced CP in animal diets decreases the amount of N in manure (Agle et al., 2010), along with lowering the manure pH. This leads to higher incorporation of N into a stable state as microbial protein (i.e., organic N), thereby decreasing the ratio of ammonium (NH₄⁺) to total N in manure, and consequently NH₃ emissions (Le et al., 2009).

Some studies used external amino acid supplementation along with reduced CP to balance the feed N intake. This exogenous supplementation improves N utilization and reduces N excretion (Panetta et al., 2006; Madrid et al., 2013; Montalvo et al., 2013). It also leads to a decrease in NH₃ emissions, but the implications of amino acid supplementation are beyond the scope of this work. In the present study, the CP levels for reference diets before a reduction in CP were averaged at 17.2 ± 2% (maximum = 22%, minimum = 11%) for both pigs and cattle. The results from the analysis indicate a reduction in NH₃ emissions by 42 ± 21% (Table 2) relative to conventional diets with higher CP. This corresponds to a 13 ± 7% reduction in NH₃ emissions per %-point decrease in CP, where %-point is the difference between two CP levels (e.g., a change in CP level from 16 to 14% is expressed as a reduction of two %-points). The emission reductions were consistent irrespective of the specific manure management stage. Ammonia emissions decreased by 41 ± 22, 41 ± 19, and 47 ± 24% with reduced CP when measured at the housing, storage, and application stages, respectively.

In contrast with NH₃, CH₄ emissions increased with a reduction in dietary CP. At lower CP levels, the digestibility of fiber is lower, and when this undigested fiber is excreted with manure, it provides additional carbon for methanogenesis, generating CH₄ during manure storage (Külling et al., 2001). The estimates from the data collected indicate an overall increase in CH₄ emissions by 71 ± 131% (Table 2), mainly from manure storage. In terms of a unit reduction in %-point CP, this equates to an increase in CH₄ emissions of 20 ± 30%. Nitrous oxide emissions tend to decrease with reduced CP in animal diets (Külling et al., 2001; Lee et al., 2012). Nitrous oxide emissions were negligible at the housing stage, but emissions were reduced by 52 ± 39 and 17% (only one observation) at the storage and application stages, respectively. Averaged over all manure management stages, lowering dietary CP reduced N₂O emissions by 30 ± 38% (Table 2), which is a decrease in N₂O emissions by 9 ± 12% per %-point reduction in CP. This is primarily due to the lower N content in the manure. The variability in emission reductions is due to certain factors associated with reduced CP that could contribute to an increase in N₂O emissions during manure storage. As described above for CH₄, the digestibility of fiber in the digestive tract of animals decreases with a reduction in CP. At the manure storage stage, this enhances crust formation, which may lead to higher N₂O emissions due to the creation of aerobic and anaerobic zones, leading to nitrification and denitrification (Külling et al., 2003).

Table 2. Emission changes (relative to reference). Compilation from a literature survey comprising peer reviewed studies (see explanation and text in the following sections).

Abatement stage	Reference scenario	Abatement options	Emission changes†		
			N ₂ O	CH ₄	NH ₃
			%		
Feeding	High protein feeds	Reduced crude protein	-30 (38)	+71 (131)	-42 (21)
		Additives	+9 (9)	+20 (33)	-9 (34)
Housing	Liquid manure housing systems	Air scrubbers	+164 (270)	-6 (21)	-59 (39)
		Frequent removal	-41 (17)	-55 (5)	-22 (29)
Treatment	No treatment	Anaerobic digesters	-23 (44)	-29 (116)	-3 (45)
		Acidification	-55 (45)	-74 (22)	-60 (28)
Storage	No cover	Covers	>+500	-11 (34)	-65 (29)
Application technique	Surface spreading	Injection	+259 (416)	-	-71 (25)

† Numbers marked in bold with a "+" sign indicates an increase in emissions relative to the reference.

Supplementing animal diets with feed additives such as fiber, acids, and enzymes generally led to a decrease in NH_3 emissions. In the case of additives, the reference CP levels were $17.6 \pm 2.2\%$ (maximum = 22%, minimum = 12%) on average for both animal types. Analysis of emission reduction estimates indicates a reduction of $27 \pm 20\%$ at the housing level and an increase of $5 \pm 37\%$ during storage. Combined estimates for all feed additives aggregated over housing and storage show a NH_3 reduction of $9 \pm 34\%$ (Table 2). Although there is a difference in emission trends between housing and storage stages, the variability in emission reductions mainly stems from the effect of different feed additives on NH_3 emissions. Enzyme addition to boost nutrient digestibility in pig diets may lead to higher NH_3 emissions. Addition of enzymes leads to the degradation of β -glucan in barley (*Hordeum vulgare* L.)-based diets, leading to more urinary N. Furthermore, enzyme addition reduces microbial fermentation and increases the slurry pH (O'Connell et al., 2006). Both these factors enhance NH_3 generation. On the other hand, inclusion of acids and fiber as feed additives leads to a reduction in NH_3 emissions (Hansen et al., 2007; Galassi et al., 2010; Montalvo et al., 2013). Acids reduce urinary pH, which leads to decreased NH_3 emissions, and fibers affect the manure composition by shifting the N from urine to feces, which is less susceptible to NH_3 volatilization (Hansen et al., 2007; Lynch et al., 2007, 2008; Montalvo et al., 2013). Moreover, addition of certain fibers such as inulin leads to a lower manure pH, resulting in a subsequent decrease in NH_3 emissions (Hansen et al., 2007; Lynch et al., 2007). However, addition of some fibers such as wheat (*Triticum aestivum* L.) bran was not as effective as other fibers (inulin and sugar beet [*Beta vulgaris* L. ssp. *vulgaris*] pulp) (Galassi et al., 2010). This is because wheat bran contains low levels of soluble nonstarch polysaccharides, which lead to a smaller shift of N from urine to feces. In addition, wheat bran is somewhat resistant to microbial fermentation; thus, the pH values of slurries were not reduced (Galassi et al., 2010).

Feed additives led to slight increases in overall CH_4 emissions by $20 \pm 33\%$ (Table 2). Estimates show an increase in CH_4 emissions of $17 \pm 33\%$ from housing and $21 \pm 42\%$ during storage. Addition of fiber led to higher CH_4 emissions, which could be due to an increase in degradable fiber in manure (Montalvo et al., 2013). Although lower pH generally leads to decreased CH_4 emissions, the addition of benzoic acid did not have a significant impact on CH_4 emissions (Montalvo et al., 2013). The effect of feed additives (both acids and fiber) on N_2O emissions during the housing stage indicated an increase in emissions by $9 \pm 9\%$ (Table 2).

Animal Housing

Cattle and pig housing also contribute to manure emissions. Better management of manure and installation of appropriate abatement technology could help abate these emissions. In this section, the abatement potential of air scrubbers and the frequent removal of manure are discussed in terms of NH_3 and GHG emissions. It is important to note that air scrubbing systems are mainly applicable to pig and poultry housing, which usually use forced ventilation (required for scrubber techniques), in contrast with naturally ventilated cattle housing systems.

Chemical and biological scrubbers have been used to reduce NH_3 and odor emissions from pig and poultry housing facilities (Feilberg and Sommer, 2013). Both chemical and biological scrubbers have the potential to reduce NH_3 emissions. Overall estimates indicate reduction in emissions from housing by $59 \pm 39\%$ (Table 2) with the use of air scrubbers. However, chemical scrubbers are more effective in reducing NH_3 emissions than biological scrubbers. In the case of chemical scrubbers, the addition of acid (usually sulfuric acid) reduces the pH of washing water, which absorbs the dissolved NH_3 , forming an ammonium salt solution (Van der Heyden et al., 2015). Reduction estimates show that chemical scrubbers can reduce NH_3 emissions by $90 \pm 8\%$. On the contrary, biological scrubbers have lower NH_3 reductions of $46 \pm 40\%$. In biological scrubbers, the washing liquid captures NH_4^+ and converts it into nitrate (NO_3^-) with the help of ammonia-oxidizing bacteria. The low NH_3 removal efficiencies in biological scrubbers, as compared with chemical scrubbers, is caused by the release of untreated air into the environment from operational inadequacies and inhibition of biological material from accumulation of NH_4^+ and NO_3^- (Martens et al., 2001).

Greenhouse gases cannot be removed using chemical scrubbers, as these gases have lower water solubility than NH_3 . In biological scrubbers, CH_4 emission reductions were negligible, with reductions of $6 \pm 21\%$ (Table 2). However, the use of biological scrubbers might lead to an increase in N_2O emissions. The longer bed residence time, along with the nitrification and denitrification processes in the presence of nitrifying bacteria, may lead to a conversion of $\text{NH}_3\text{-N}$ to NO_3^- and then N_2O (Melse and Mosquera, 2014; Van der Heyden et al., 2015). All the observations analyzed in this study pertaining to biological scrubbers revealed an increase in N_2O emissions of $164 \pm 270\%$ (Table 2).

Frequent Removal of Manure

Ammonia emissions from animal housing are dependent on the surface area of manure in contact with air. A major share of the NH_3 emissions is generated from fresh manure, especially urine. Hence, regular removal of fresh manure can reduce NH_3 emissions (Amon et al., 2007; Ivanova-Peneva et al., 2008). Results from the analysis show a reduction in NH_3 emissions of $22 \pm 29\%$ (Table 2) from animal housing facilities with daily or weekly removal of manure. Methane and N_2O emissions from animal housing systems are also dependent on the total amount of manure (Amon et al., 2007). Frequently removing manure from animal houses reduces the accumulation of manure and decreases CH_4 and N_2O emissions by 55 ± 5 and $41 \pm 17\%$ (Table 2), respectively. Lower temperatures during outside storage of the removed manure are likely to reduce CH_4 emissions, compared with manure stored indoors (Amon et al., 2007). Hence, the overall emission balance may be improved with frequent manure removal to outside storage.

Manure Treatment

Manure treatment refers to techniques by which manure can be handled to ensure lower emissions and, in some cases, generate energy. Among the different manure treatment options available, anaerobic digestion and acidification of manure are discussed here.

Anaerobic Digestion

Anaerobic digestion involves the degradation of organic matter to generate CH_4 , CO_2 , and other gaseous byproducts. The generated gases, particularly CH_4 , can act as an additional renewable energy source when captured. Hence, anaerobic digestion is not only a direct emission abatement technique but can also produce cogeneration credits by substituting conventional energy sources (Roos et al., 2004).

Literature on the effects of anaerobic digestion on NH_3 emissions reveals mixed results (Chantigny et al., 2007) with considerable variability. Estimates here showed an increase in NH_3 emissions by $13 \pm 76\%$ during the storage of digestate and a decrease of $8 \pm 34\%$ when applied to the soils. Overall estimates averaged over the storage and application stages indicate a tendency for a small decrease in NH_3 emissions by $3 \pm 45\%$ (Table 2) for digestates relative to raw slurry. Some studies claim that anaerobic digestion decreases the dry matter content of manure, allowing for greater infiltration of digestate when applied to the soils, which could reduce NH_3 emissions from the soil surface (Chantigny et al., 2009). Other studies suggest that the process of anaerobic digestion increases total ammoniacal nitrogen concentration and the pH of digested manure, which might lead to an increase in NH_3 emissions during storage and application of digestate (Petersen and Sommer, 2011). Ammonia emissions are also dependent on site-specific factors and management practices during the storage and application of digested manure. For example, an increase in NH_3 emissions was observed when digestate was stored without a cover (Amon et al., 2006). Injection of digested slurry during field application can also reduce NH_3 emissions, relative to surface spreading (Nyord et al., 2012). This highlights the importance of properly handling anaerobic digestates during storage and field application to optimize the NH_3 emission reduction potential.

In the case of CH_4 emissions, estimates from the present study show a reduction in emissions by $29 \pm 116\%$ (Table 2), primarily during the storage of digestate. Variability in emissions exists due to varying CH_4 recovery rates from manure of different animal types and the influence of external factors such as storage temperature, duration, and structure of manure storage systems (Massé et al., 2011). Care should also be taken to prevent CH_4 losses from improper operation, uncontrolled losses, and leaks from anaerobic digestion systems, which could make the system ineffective in reducing CH_4 and other GHGs (Montes et al., 2013).

Nitrous oxide emissions increased by $20 \pm 41\%$ during digestate storage and decreased by $29 \pm 43\%$ during the application of digestate. Averaging the N_2O emission reductions from treatment, storage, and application of digestate indicates reductions of $23 \pm 44\%$ (Table 2). Clemens et al. (2006) reported higher N_2O emissions from digested slurry relative to untreated slurry during summer storage. They associated the increase in N_2O emissions to the differences in $\text{NH}_4\text{-N}$ content, which could influence nitrification and denitrification processes. Another study (Wang et al., 2014b) also revealed elevated emissions of N_2O during the storage of digestate. The authors attributed the increase in emissions to the high ventilation rate, high storage temperature, and low depth of manure storage. During manure application, the decomposition of organic matter after anaerobic digestion has a twofold effect on reducing N_2O emissions. First,

it makes a higher share of N directly available to plants due to the mineralization of organic N to ammonium (Walsh et al., 2012), and second, it limits the activity of N_2O -forming microorganisms dependent on fresh organic matter for their energy supply (Montes et al., 2013). However, Thomsen et al. (2010) suggested that factors such as manure composition and external conditions, such as water-filled pore space and the balance between oxygen supply and demand, could lead to increases in N_2O emissions when digestate was applied to soils relative to untreated slurry.

Acidification of Manure

Acidification refers to the addition of acids to manure. Acids can be introduced into the manure management chain right from the feeding stage, as discussed above. Here, we only investigate the effect of acids during housing and storage, and also further downstream during the application of manure to soils.

Acidification has not been widely adopted in the United States and some European countries due to acid hazard, foam formation, and costs that were attributed to this technology until recently (Fangueiro et al., 2015). However, considerable progress has been made in manure acidification, and it is recognized as one of the best available technologies to reduce NH_3 emissions in Denmark (Kai et al., 2008). Moreover, it has recently been assigned as a category-one abatement strategy in the UNECE guidance document to reduce NH_3 emissions from manure (Bittman et al., 2014). Addition of acids reduces the pH level of manure, thereby inhibiting the activity of urease-producing bacteria, shifting the equilibrium from $\text{NH}_3\text{-N}$ to favor NH_4^+ and consequently lowering NH_3 emissions (Jensen, 2002). Additionally, as a result of retaining N in the manure, manure acidification is valuable during land application since the acidified manure has more plant available N (Montes et al., 2013). Ammonia emissions decreased by $77 \pm 11\%$ for housing, $63 \pm 30\%$ during storage, and $43 \pm 18\%$ after the application of acidified slurry. Averaged over all manure management stages, NH_3 emissions were reduced by $60 \pm 28\%$ (Table 2). All the studies used here pertaining to manure acidification revealed consistent reductions in NH_3 , highlighting the potential of acidification as a promising NH_3 abatement technique. However, the results were sensitive to the final pH level achieved with the addition of acids. Former assessments indicate that pH levels lower than six were effective in reducing NH_3 emissions (Wang et al., 2014a).

Although acidification shows promise as a NH_3 abatement option, its implication on GHGs is not well documented. Our evaluation indicates that acidification of manure can reduce CH_4 emissions by $74 \pm 22\%$ (Table 2) during the storage of manure. Studies have reported that methanogenesis is limited by addition of acids, particularly sulfates (Petersen et al., 2012, 2014). Methanogenic activity leading to CH_4 emissions is prevalent between pH levels of 6.5 and 8.5. Acidification of manure generally reduces the pH to lower levels (below six), inhibiting methanogenesis and subsequently reducing CH_4 emissions (Wang et al., 2014a). Effects of acidification on N_2O emissions were mixed. Addition of sulfuric acid led to overall reductions in N_2O emissions by $17 \pm 30\%$ (Table 2) during the storage and application of acidified slurry. The reduction in N_2O emissions was due to the decreased activity of bacterial nitrifiers with the lowering of slurry pH (Owusu-Twum et al., 2017). One of the studies,

however, reported no change in N_2O emissions during storage of acidified slurry (Petersen et al., 2014). The authors suggested that they may have disturbed surface crust formation, a prime source of N_2O emissions, during acid mixing in the experiment, leading to no change in emissions. Lactic acid reduced N_2O emissions by 90% or more due to the lowering of slurry pH (Berg et al., 2006b). The use of nitric acid, on the other hand, adds N to the system and also stimulates denitrification, leading to large increases in N_2O emissions (Berg et al., 2006b). As nitric acid is not currently considered a practical acidification technique, it was omitted from our analysis. Overall estimates indicate a N_2O reduction of $55 \pm 45\%$ (Table 2), comprising N_2O reductions of $56 \pm 51\%$ during storage of acidified manure and 52% when applied to the soils. The variations in emission reduction stem from the selection of appropriate acid and application rates, which are acknowledged in the published literature (Petersen and Sommer, 2011).

Manure Storage

Manure is typically stored after removal from animal houses or after manure treatment for various reasons (e.g., appropriate timing of soil application, limited land near the livestock operation for application, time restraints of work staff, etc.). Improper storage of manure leads to physical and chemical degradation. A result of this degradation is emissions, odors, loss of fertilizer value, and also high hauling cost when exposed to rain. A feasible emission abatement option during manure storage is the use of covers. Covers include the use of chopped straw, wooden lids, granules, floating films, plastic covers, roofs, and also the use of other materials like oil, peat, etc. (Amon et al., 2014).

Similar to animal housing, the extent of NH_3 emissions depend on the surface area of exposed manure. Since covers form a barrier and can retain the N without exposing it to air, they can reduce emissions of NH_3 . Straw covers, granules, wooden lids, plastic sheets, natural crust, and peat can reduce NH_3 emissions when used as covers (Amon et al., 2014). Overall reductions in NH_3 from using covers were $65 \pm 29\%$ (Table 2). Among the different covering materials analyzed, the use of plastic films, granules, and peat were more effective in reducing NH_3 emissions than other covering materials like straw, wooden lids, and the natural formation of surface crusts. Some studies also used covers in conjunction with acids (lactic acid), which also yielded favorable reductions in NH_3 emissions (Berg et al., 2006a).

Results from the analysis indicate a reduction in CH_4 emissions by $12 \pm 33\%$ (Table 2) with the use of covers. Oxidation of CH_4 , limiting variations in moisture content, maintaining a stable CH_4 concentration above the stored manure, and avoiding CH_4 losses from ebullition (i.e., bubbling or foaming) could reduce CH_4 emissions from stored manure (Rodhe et al., 2012). Although the use of covers generally favors mechanisms that reduce CH_4 emissions (Amon et al., 2007; Rodhe et al., 2012), the emission patterns vary among individual studies. These studies argue that the microbial processes governing CH_4 production with certain covering materials (straw, formation of surface crusts) and the influence of manure storage conditions (time, weather, etc.) affect CH_4 emissions (Hansen et al., 2009; Petersen et al., 2013).

The use of covers led to large increases in N_2O emissions ($>500\%$, Table 2). This estimate was accompanied by high variability, due to contrasting emission trends among all the covering materials analyzed. Certain covering materials such as formation of surface crust, addition of straw, and the use of granules tended to increase emissions of N_2O (Berg et al., 2006a; Hansen et al., 2009; VanderZaag et al., 2009; Petersen et al., 2013). These types of covering materials create both aerobic and anaerobic zones, providing suitable conditions for the nitrification and denitrification processes responsible for N_2O emissions (Clemens et al., 2006). Some studies argue that the use of impermeable covers like wooden lids or plastic films could reduce N_2O emissions through the elimination of oxygen in the headspace (Clemens et al., 2006). External factors such as moisture content of the crust, manure storage time, and weather conditions during storage may also influence the emission patterns of N_2O (Rodhe et al., 2012; Petersen et al., 2013). The combined use of covering materials or covers with manure acidification also produced contradictory results in terms of N_2O emissions (Berg et al., 2006a; Clemens et al., 2006). Further research is needed to understand the mechanism of N_2O emissions with the use of covers and possible interactions with other emission abatement options.

Manure Application

Animal manure is rich in micro- and macronutrients essential for plant growth. Addition of manure to soils increases the organic matter, along with enhancing numerous other soil properties (Hristov et al., 2013). However, the application of manure is associated with NH_3 and N_2O emissions. Methane emissions from manure application are generally low due to carbon uptake by the soils and the existence of an aerobic environment in most cropping systems (Goulding et al., 1996). A study by Sherlock et al. (2002) estimated CH_4 losses to be 0.08% of the carbon applied, as compared with 22.5 and 2.1% of the nitrogen applied lost as NH_3 and N_2O after manure application. In terms of warming potentials, this translated to 3700 kg CO_2e for N_2O compared with 29 kg CO_2e for CH_4 . Hence, CH_4 emissions from manure application can be considered negligible for the purposes of this study (Montes et al., 2013). Although many modes of manure application are available, this study analyzed only shallow injection relative to surface spreading as the reference.

On average, shallow injection reduced NH_3 emissions by $71 \pm 25\%$ (Table 2) as compared with surface spreading of manure. These high reductions in NH_3 emissions were due to the reduced contact of manure with the atmosphere (Huijsmans et al., 2003). Manure injection led to major increases in N_2O emissions by $259 \pm 416\%$ (Table 2). This could be due to creation of high moisture anaerobic zones that promote denitrification (Vallejo et al., 2005; Dell et al., 2011; Duncan et al., 2017). However, there was high variability in estimates of increased N_2O emissions from shallow injection. A review study by Webb et al. (2010) underlines some of the reasons for this variability. Oxygen availability, preexisting N concentrations in the soil, soil moisture content, and the length of the diffusion path to the soil surface could influence the impact of shallow injection on the amount of N_2O emissions. In some cases, this could even reduce or nullify the impact of shallow injection on N_2O emissions.

Implication on Emission Abatement Strategies

The results discussed in this study may play an important role in developing consistent procedures in the design of emission abatement strategies. Some of the key elements that could aid in selection and implementation of abatement options, along with prospects to further improve effectiveness of emission abatement strategies, are described below.

Importance of Proper Clustering and Classification

The estimated emission reductions for the different abatement strategies are characterized by variability. Although inherent uncertainty due to experimental design, measurement technologies, and site-specific factors contributes to this variability, in some cases, this variability can be explained and disentangled. For example, in the case of dietary manipulation, lowering CP in animal diets reduced overall NH_3 emissions by $42 \pm 21\%$. However, a closer analysis indicates an influence of the level of CP reductions on NH_3 emissions. Reducing CP levels by less than two %-points decreases NH_3 emissions by $26 \pm 21\%$, whereas a reduction in CP levels greater than five %-points reduces NH_3 emissions by $51 \pm 21\%$. Feed composition could also affect NH_3 emissions from reduced CP in animal diets. For example, a CP reduction from 22 to 16% for barley-based diets in pigs showed a more pronounced effect on lowering NH_3 emissions, rather than for wheat. This is because the relatively lower β -glucan content in wheat led to more N in urine than barley, generating higher NH_3 emissions (O'Connell et al., 2006). As far as the difference between cattle and pigs is concerned, estimates indicate a greater reduction in NH_3 by $16 \pm 6\%$ per unit percentage decrease in CP for cattle, compared with $11 \pm 6\%$ for pigs. A similar comparison distinguishing between animal types for reduced dietary CP and feed additive supplementation on GHG and NH_3 emissions could not be conducted due to limited data availability. An assessment of the results also highlights the importance of accounting for the differences in biological and physical aspects of animals when quantifying emission reductions from lowering CP in animal diets. For example, the effect of reduced CP on NH_3 emissions was magnified in growing pigs as compared with pigs in the finishing stage, highlighting the influence of growth stage on NH_3 emissions (Hernández et al., 2011). Additionally, emission measurement studies show that animal category (e.g., gilts, barrows, boars, etc.) could also have an impact on the level of NH_3 reductions that can be achieved with reduced CP in pigs (Lynch et al., 2007, 2008). This suggests that even though reducing CP in animal diets is an effective NH_3 abatement option, influencing factors such as level of CP reductions, feed composition, animal type, growth stage, and animal category must be accounted for while estimating emission reductions (Sajeev et al., 2017). Improved classification and reporting of influencing factors are necessary to ensure that the proposed emission reductions can be achieved. The supplemental section in the UNECE guidance document on NH_3 abatement strives toward this objective by accounting for and categorizing the effect of influencing factors on NH_3 emissions from a reduction in CP levels (Bittman et al., 2014).

Another example is related to the acidification of manure and its impact on N_2O emissions. The results show that lactic acid reduced N_2O emissions by $93 \pm 3\%$, whereas the addition of sulfuric acid had lower N_2O reductions of $17 \pm 30\%$. Nitric acid, on the other hand, led to a significant increase in N_2O emissions ($>100\%$) (Berg et al., 2006b; Petersen et al., 2012; Owusu-Twum et al., 2017). Although nitric acid is currently not used as an abatement option, it does reduce NH_3 and CH_4 emissions. This underlines the implication of different acid choices on emission reduction trends. Clearly specifying the choice of acid to be added can improve the effectiveness of acidification as an abatement option.

Similarly, overall estimates suggest that the use of covers during manure storage led to an increase in N_2O emissions. However, on assessing the effects of different types of covers on N_2O emissions, the addition of chopped straw and formation of natural crusts were two options that significantly increased N_2O emissions ($>500\%$). Other covering options such as wooden lids, plastic sheets, and floating films were successful in reducing N_2O emissions in the range of 30%. Clustering these different covering materials together without understanding the differences in the way they operate can severely undercut potential emission reductions and make the whole option unviable. The observations and insights discussed above call for researchers to perform experiments where emission reductions from different abatement options are compared, along with explicitly describing assumptions, reporting influencing factors, highlighting caveats, and properly classifying the results associated with the abatement options they analyze and recommend. This will help to correctly understand the dynamics of implementing an abatement option, ensuring desired efficiency.

Accounting for Emission Interactions

Our results also highlight and quantify the extent of interactions between the three gases along the manure management chain for different stages and for various abatement options. Current policies on manure management adopt a uniform approach that is limited to a single pollutant to abate either air pollution from increased NH_3 emissions or climate change impacts due to GHGs. Given the extent of interactions between multiple pollutants such one-dimensional policy initiatives may prove to be suboptimal. The results presented here indicate that although a particular abatement option is effective in reducing emissions of a single gas, it might not be as effective for other gases and may even lead to an increase in emissions. This was evident in various stages of the manure management chain. In the case of housing, the use of air scrubbers was promising when it comes to NH_3 emissions. However, the use of biological scrubbers could lead to an increase in N_2O emissions ($>100\%$). Similarly, in the case of manure storage, the use of chopped straw and formation of surface crust were effective NH_3 abatement options, with reduction estimates of 66 and 57%, respectively. However, these options showed a significant increase in N_2O emissions ($>500\%$) and a negligible to moderate abatement (12%) of CH_4 emissions.

Another example where the effects of interactions were visible was during the application of manure to soils. Although shallow injection was capable of reducing NH_3 emissions by 71%, it can lead to increases in N_2O emissions ($>200\%$) relative to surface

spreading of manure. The net impact of abatement options must therefore take into account the result on all gaseous emissions to determine their actual effectiveness in abating air pollution and climate change at the same time. Hence, it is vital to understand the extent of the role of emission interactions, as it may have important policy implications. This understanding can inform multipollutant optimization approaches that minimize tradeoffs associated with regulating NH_3 and GHGs.

Shifting to a Whole Chain Perspective

Most of the abatement research has focused on individual abatement options that only influence certain emissions in a particular manure management stage. However, this merely transfers the saved emissions downstream along the chain and also discounts for possible side effects due to emission interactions. For example, the use of covers during manure storage can lower NH_3 emissions by $65 \pm 29\%$ as compared with uncovered manure. However, surface spreading of the stored manure during the application stage may release all the NH_3 retained during the previous stage (increase of 71% relative to shallow injection). Additionally, in the case of anaerobic digesters, we observed an increase in NH_3 and N_2O emissions during digestate storage, followed by a decrease in NH_3 and N_2O when applied to soils. Hence, even though abatement options targeting specific stages and particular gases in the manure management chain are useful, a whole-chain perspective is vital to accurately ascertain the overall effectiveness of an abatement option.

An overall assessment of single-stage abatement measures illustrating the best abatement pathways that can achieve maximum emission reductions for individual gases is presented in Fig. 2. For instance, acidification of manure is a promising abatement option, reducing NH_3 emissions by $60 \pm 28\%$, CH_4 by $74 \pm 22\%$, and N_2O by $55 \pm 45\%$ when averaged over housing, storage, and application stages relative to untreated raw slurry. Frequent removal of manure from animal housing and anaerobic digestion were two other abatement options that reduced all three gases collectively. Whole-chain thinking also accounts for emission interactions. This is clearly observed in the manure application stage, where shallow injection was the best option for NH_3 abatement, whereas surface spreading was preferred for N_2O abatement. This was also visible in the feeding stage, where reduced CP in animal diets lowered emissions of NH_3 by $42 \pm 21\%$ and N_2O by $30 \pm 38\%$ but could lead to an increase in CH_4

by $71 \pm 131\%$ relative to conventional diets with high CP content. The concept and need for proper clustering and classification was also evident and demonstrated in the manure storage stage, where covers that were deemed unviable as a N_2O abatement option due to large increases in N_2O emissions (Table 2) could actually be effective with the proper selection of covering material. Hence, shifting to a whole-chain perspective that tracks the fate of manure and gases along the entire manure management chain could help in choosing effective strategies that reduce the risk of emission leakages and pollution swapping.

Prioritization of Abatement Options

An additional aspect that is often ignored in whole-chain emission estimations is the difference in the functioning of abatement options. Oenema et al. (2009) addressed this topic, suggesting that priority should be given to abatement measures that (i) reduce N losses, (ii) increase the N content in useful products such as fertilizers, and (iii) decrease N input into the system. Emission estimates in this study indicated that abatement options such as the use of covers and frequent removal of manure can reduce N losses. However, these options only keep the emissions in check at a particular management stage. If the manure is not properly handled, the saved emissions can be emitted elsewhere in other manure management stages. On the other hand, options such as reduced CP, acidification, and anaerobic digestion affect emissions by altering manure characteristics, the implication of which is then reflected along the rest of the manure management chain. In the case of reduced CP in animal diets, measurement studies showed that NH_3 emission reductions could be achieved along the entire manure management chain (Portejoie et al., 2004; Lee et al., 2012). Emission measurements during storage and application of manure after acidification also suggest that significant emission reductions can be attained when acidification leads to pH levels <6 . Furthermore, acidified and anaerobically digested manure may have more plant available N as compared with untreated slurry (Montes et al., 2013). Hence, abatement options that alter manure characteristics could be more effective at overall emission reductions than other options that do not change manure properties. Prioritizing such abatement options could be an important input for researchers and policymakers in determining low-risk options in achieving effective emission reductions in the management of manure.

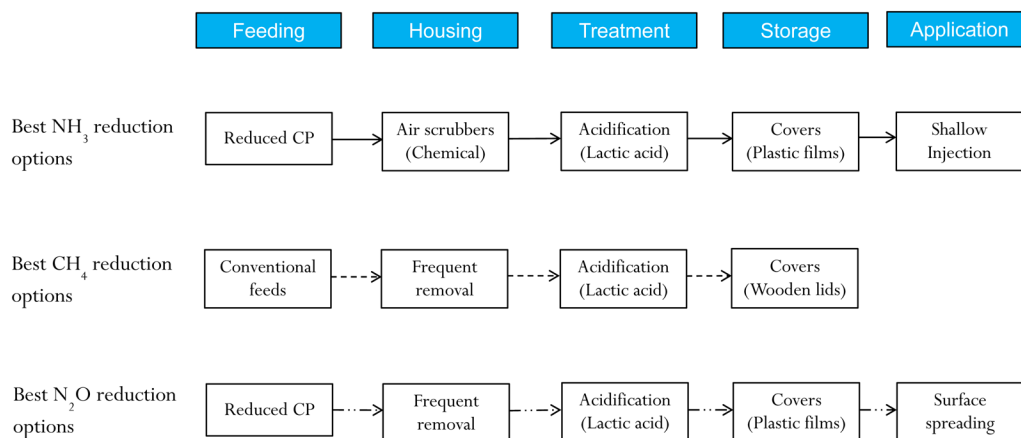


Fig. 2. Best emission abatement options along the manure management chain for ammonia (NH_3), methane (CH_4), and nitrous oxide (N_2O).

Need for Comprehensive Emission Measurements

Some studies have estimated the implications of abatement options on emission reductions for the whole manure management chain (Hou et al., 2015, 2017; Wang et al., 2017). These studies are useful in charting out abatement pathways that optimize both NH₃ and GHG emissions for the whole chain. In this study, however, we avoid these types of estimations due to the following reasons. First, the emission reduction estimates reported in this study, aggregated from a multitude of existing studies, are mostly limited to a single manure management stage. Thus, the specific effects of an abatement option on other manure management stages were not fully clear. For example, although frequent removal of manure from animal housing is a promising abatement option that reduces all three gases at the housing level, the removed manure must be handled properly down the whole manure management chain to prevent elevated emissions at later stages. Assessments that track the fate of manure through all stages would offer more realistic emission estimates, as opposed to emission trends accumulated from individual studies. Second, only a few studies among the many considered here measured the effect of an abatement option on all three gases simultaneously. Such studies are important in reducing variability in emission estimates, since the assumptions and site-specific conditions would be the same. In contrast, populating emission estimates from multiple studies with varying experimental conditions could increase variability. For example, with some abatement options, such as reduced CP content in animal diets, feed additives, air scrubbers, or acidification, research has nearly exclusively focused on NH₃ emissions. The impact of these measures on GHG emissions must be further investigated before an overall impact can be determined. Additional research is also needed to understand the mechanism of emissions under the influence of external factors. This was highlighted during the use of covers where studies show that storage conditions and external factors affect N₂O emissions (Rodhe et al., 2012; Petersen et al., 2013). Furthermore, combining different abatement options might lead to enhanced emission reductions. For example, combining frequent removal of manure and air scrubbers in housing may further reduce emissions compared with using just a single option. However, measurement studies that combine different abatement options and employ a whole-chain view are rare. These highlighted shortcomings call for comprehensive, reliable, and consistent emission measurement studies that measure the effect of abatement options on all three gases along the manure management chain while allowing for combination of different abatement options. These measurements can then help populate and correctly assess the effectiveness of abatement options for the entire manure management chain.

Conclusions

This study provides a review of abatement options for both NH₃ and GHGs pertaining to the management of liquid manure across different stages. Analysis of the emission reduction estimates highlights the importance of accounting for interactions between emissions. Some abatement options, such as manure acidification, anaerobic digesters, and frequent removal of manure, can reduce both NH₃ and GHG emissions, whereas other options are associated with tradeoffs or pollution swapping. The results call for a shift from assessment of single-stage abatement to a whole-chain

perspective to ensure overall emission reduction across the entire manure management chain. The analysis also identified some key elements, such as proper clustering, reporting of influencing factors, and explicitly describing assumptions associated with abatement options, which can reduce variability in emission reduction estimates and, in some cases, make previously unviable options applicable. Furthermore, prioritization of abatement options that change manure characteristics throughout the manure management chain (e.g., low-protein diets, anaerobic digestion, or slurry acidification) can help in determining low-risk emission reduction options. Such assessments, supported by reliable and comprehensive emission measurement studies, can help pave the way toward consistent and integrated manure management strategies that reduce the impact of livestock production on both air quality and climate change.

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