Environmental Science & Technology

Article

pubs.acs.org/est

Mitigating Greenhouse Gas and Ammonia Emissions from Swine Manure Management: A System Analysis

Yue Wang,[†,‡] Hongmin Dong,*[,†,‡] Zhiping Zhu,[†,‡] Pierre J. Gerber,[§,∥] Hongwei Xin,[⊥] Pete Smith,[#] Carolyn Opio,[§] Henning Steinfeld,[§] and Dave Chadwick[▽]

[†]Institute of Environment and Sustainable Development in Agriculture, Chinese Academy of Agricultural Sciences, Beijing 100081, China

[‡]Key Laboratory of Energy Conservation and Waste Treatment of Agricultural Structures, Ministry of Agriculture, Beijing 100081, China

[§]Animal Production and Health Division, Food and Agriculture Organization, 00153 Rome, Italy

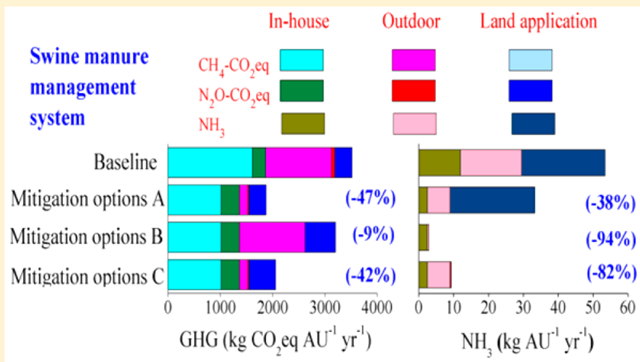
[∥]Animal Production Systems Group, Wageningen University, P.O. Box 338, Wageningen, The Netherlands

[⊥]Department of Agricultural and Biosystems Engineering, Iowa State University, Ames, Iowa 50011, United States

[#]Institute of Biological and Environmental Sciences, University of Aberdeen, 23 St. Machar Drive, Aberdeen AB24 3UU, United Kingdom

[▽]Environment Centre Wales, School of Environment, Natural Resources and Geography, Deiniol Rd., Bangor University, Bangor LL57 2UW, United Kingdom

Supporting Information



ABSTRACT: Gaseous emissions from animal manure are considerable contributor to global ammonia (NH_3) and agriculture greenhouse gas (GHG) emissions. Given the demand to promote mitigation of GHGs while fostering sustainable development of the Paris Agreement, an improvement of management systems is urgently needed to help mitigate climate change and to improve atmospheric air quality. This study presents a meta-analysis and an integrated assessment of gaseous emissions and mitigation potentials for NH_3, methane (CH_4), and nitrous oxide (N_2O) (direct and indirect) losses from four typical swine manure management systems (MMSs). The resultant emission factors and mitigation efficiencies allow GHG and NH_3 emissions to be estimated, as well as mitigation potentials for different stages of swine operation. In particular, changing swine manure management from liquid systems to solid–liquid separation systems, coupled with mitigation measures, could simultaneously reduce GHG emissions by 65% and NH_3 emissions by 78%. The resultant potential reduction in GHG emissions from China's pig production alone is greater than the entire GHG emissions from agricultural sector of France, Australia, or Germany, while the reduction in NH_3 emissions is equivalent to 40% of the total NH_3 emissions from the European Union. Thus, improved swine manure management could have a significant impact on global environment issues.

1. INTRODUCTION

Livestock production represents the largest anthropogenic source of methane (CH_4) and nitrous oxide (N_2O),[1,2] and contributes a range of critical environmental problems,[3,4] including greenhouse gas (GHG) emissions,[5−8] ammonia (NH_3) emissions and alteration of nitrogen cycles,[9−12] land and water use,[7] and misuse of antibiotics leading to antimicrobial resistance.[13] In China, for example, an estimated 42% of the national total chemical oxygen demand (COD) and 22% of the total nitrogen (TN) discharged to the environment arise from livestock production.[14]

Livestock produce large quantities of manure rich in nitrogen and organic matter that contribute considerably to global emissions of NH_3 and GHGs.[15] Approximately 40% of the global anthropogenic NH_3 and N_2O emissions are associated with livestock manures.[2,9,16] In China, as much as 78% of the N excreted from the animals are lost to the environment,[17] mainly through NH_3 emissions which can contribute to odor emanation, water eutrophication, soil acidification,[18,19] promote the formation of particulate matter (PM), and also increase climate change since NH_3 is a precursor of N_2O.[20,21] Pig manure is

Received: December 19, 2016
Revised: March 15, 2017
Accepted: March 20, 2017
Published: March 20, 2017

DOI: 10.1021/acs.est.6b06430
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

particularly important due to the rapid increase in pig production over recent decades[22] and the trend toward intensification of production. Pig manure contributes, respectively, 76%, 32%, and 44% of the national CH_4, N_2O, and NH_3 emissions from livestock manures in China.[23,24]

Gaseous emissions from manure management occur in three phases, namely, in-house handling, outdoor storage and treatment, and land application.[25] As emissions of NH_3, N_2O, and CH_4 result from microbiological, chemical, and physical processes, these emissions are influenced by a multitude of different factors, such as manure characteristics,[25] temperature,[26] O_2 availability,[27] trade-off between emissions of CH_4 and N_2O,[28] as well as interactions between N_2O and NH_3.[29] Studies have been conducted to address manure-related emissions, and various mitigation measures have been tested and developed. However, most studies have focused either on one specific gas, one individual manure management phase or influencing factor, or mitigation practice.[1,30,31] Yet it is now recognized that some mitigation measures can cause unintended environmental side effects on other gaseous emissions. For instance, shallow injection, while reducing NH_3 emissions from slurry spreading as compared to surface broadcasting, can result in greater N_2O emissions and may also increase the persistence of faecal indicator organisms in soil.[25,32] Therefore, radical rethinking is imperative to achieve comprehensive reductions in major environmental impacts through an entire manure management system assessment.

Four typical manure management systems (MMSs) associated with swine production throughout the world, namely, deep-pit, pull-plug, bedding, and solid–liquid separation, were analyzed in this study (Figure 1).

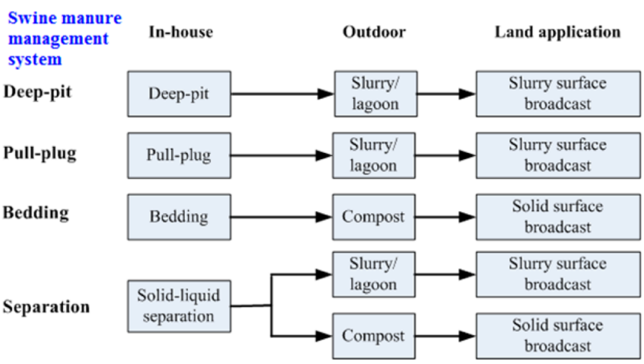


Figure 1. Representation of the baseline scenarios of four manure management systems.

Deep-Pit System. This is a liquid system, in which manure is collected and stored in the pit below a slatted floor for several months. Manure is usually thoroughly cleaned out from pit when a batch of pigs is finished, and the liquid slurry is stored in a lagoon or storage tank until the soil tillage season when it is land-applied.

Pull-Plug System. This is also a liquid system, but it differs from the deep-pit system in the length of manure storage period. In pull-plug mode, a shallow pit is used in-house to store slurry for 2–8 weeks and then drained, by gravity, to an outdoor storage facility, and the slurry is then land-applied. Liquid systems (including both the deep-pit system and pull-plug system), are widely used in confined animal feeding operations, accounting for 87%, 92%, and 100% of the swine MMSs in the United States, Germany, and The Netherlands, respectively.[33]

Bedding System. This is a solid manure system, in which the animal's excreta is deposited onto straw, sawdust or other bedding materials during the in-house phase. Solid manure is then removed from the pig house and either stockpiled or actively composted, then land-applied. Given that composting can prevent potential risks of pathogen transfer and reduce viable weed seeds compared to stockpiling manure, only the composting treatment is included in the analysis of gaseous emissions from the bedding system. Bedding systems are expected to increase in the future due to concerns about animal welfare under other systems.[34]

Separation System. This system refers to the separation of solid and liquid manure, in which solids are scraped or manually cleaned out from pig house daily or more frequently, and the liquid is separated. The liquid fraction contains a reduced nutrient burden and flows out of the animal house by gravity to an outdoor storage facility (lagoon or tank). The solid fraction would be composted. Finally, both solid and liquid manure will be land-applied. The separation system is particularly attractive for new facilities, and would be difficult to retrofit to existing buildings.

This study represents the first attempt to perform a system-level, comprehensive assessment of GHG and NH_3 emissions from four typical swine MMSs to demonstrate the potential influence of system choices on the magnitude of gaseous emissions. A comprehensive data set has been collated and developed on CH_4, N_2O, and NH_3 emission factors (EFs) for each stage of the MMSs, which included four in-house manure handling practices, three outdoor storage and treatment practices, and seven land application practices. This meta-analysis also quantifies the efficiencies of 17 mitigation strategies, including three in-house, eight outdoor storage and treatment, and six land application mitigation measures. System-level GHG and NH_3 emissions for the four MMSs, with or without mitigation measures were analyzed, and the most effective designs for simultaneous reduction of GHG and NH_3 emissions from each MMS were recommended.

2. MATERIALS AND METHODS

2.1. Data Sources and Selection Criterion. The ISI Web of Knowledge database (www.isiwebofknowledge.com) and the Chinese journal database (www.cnki.net) were used to search all published data sets as of January 2016. Specific search terms were combined and used, depending on animal categories (swine, pig, livestock, animal), manure, in-house manure management (slatted floor, pit, bedding, litter, pull-plug, discharge, scraper, separation), outdoor manure management (lagoon, slurry pond, storage tank, compost, solid storage, stockpile), land application (surface spreading, injection, incorporation, band spreading), gaseous emission (NH_3, CH_4, N_2O, and GHG gas), and mitigation measure (diet, biofilter, biogas, additive, cover, acid, cooling, nitrification inhibition). Literature sources used in this study were selected based on the following criteria: (1) The research object was swine; (2) The study included at least one of the CH_4, N_2O and NH_3 gases; (3) Gas emission flux or gas emission factor was available; (4) For literature related to mitigation, only studies that reported at least one control group were selected so that emission mitigation efficiency could be calculated.

Application of the selection criteria resulted in 142 peer-reviewed papers containing 958 effective observations which were used in the meta-analysis. Data were collected from both published tables and text for all the selected research articles, as

DOI: 10.1021/acs.est.6b06430
Environ. Sci. Technol. XXXX, XXX, XXX–XXX

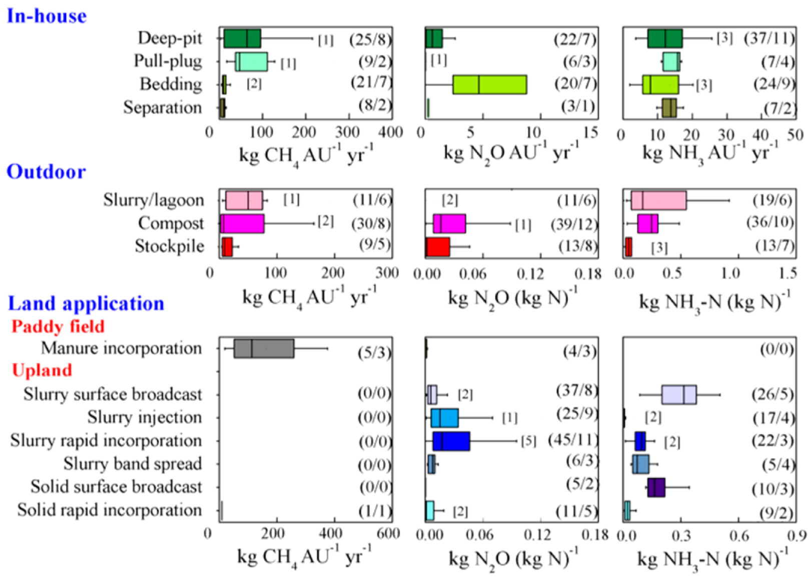


Figure 2. Box and whisker plots of the CH_4, N_2O and NH_3 emission factors for the various manure management practices in three phases (in-house, outdoor and land application) (see SI Table S3−S5 for numeric data). The vertical lines of the boxplots represent the median, upper and lower quartiles. The whiskers show values that extend to 1.5 orders of box length. The numbers in the square brackets represent the number of outliers (>1.5 orders of box length). Values in parentheses represent the number of observations on which the statistics were based and the number of studies from which the observations originated.

well as extracted from published figures using the GetData Graph Digitizer software (v. 2.22).[35] In addition to the gaseous emission data, related information allowing interpretation of the observations such as swine number, swine weight, area of the lagoon/storage tank, emission flux, and other gas emission relevant information such as study location, seasons, the manure property parameters, and soil properties were recorded (Data set S1, tabs for raw data). The location and distribution of the data used in this study are summarized in Supporting Information (SI) Figure S1. It can be seen that most studies were distributed in Europe, North American, and East Asia.

2.2. Data Analysis. *2.2.1. Calculation of Emission Factors (EFs) in the Different Phases.* To perform statistical analysis, the various units of gas emissions were converted into kg AU^{-1} yr^{-1} (1 AU [animal unit] = 500 kg) using the calculation method presented in SI Table S1. The NH_3 and N_2O EFs for outdoor manure management (storage and treatment) and land application phases in this paper were calculated as the percentage of total nitrogen (TN), that is, kg NH_3−N (kg TN)$^{-1}$ and kg N_2O−N (kg TN)$^{-1}$. When unit conversion was not possible due to lack of key information, the original emission data were excluded from the statistical analysis. The integrated EFs for each phase of MMS, including the median, mean value, standard error, and Interquartile Range (IQR), were calculated with SPSS software (v. 20.0, SPSS Inc., Chicago, IL). Results were not weighted according to sample size; therefore, all of the observations had equal impact on the results. Given the influence of a few measurements with very high values or very low values on the mean values, median values were used instead of means as the basis for subsequent calculations, since median values are quite robust to outliers.[36] The 95% confidence interval (95%CI) of the median was calculated using eq 1.

$$95\%\mathrm{CI} = 1.58 \times \frac{IQR}{\sqrt{N}} \tag{1}$$

where N represents the number of observations for each emission factor.

2.2.2. Calculation of GHG and NH_3 Emissions for the Baseline Scenarios of Four Swine Manure Management Systems. Integrated GHG and NH_3 emissions for the baseline scenarios of the four MMSs were calculated, based on the summation method for CH_4 and N mass flow method for NH_3 and N_2O, respectively. The indirect N_2O emissions arising from N deposition and N leaching or runoff were also considered. The detailed calculation process is presented in section 2 of the SI.

2.2.3. Calculation of Mitigation Efficiency of Each Measure. The efficiencies of individual mitigation measures for the corresponding manure management phases were assessed by comparing the result of control and treatment groups sourced from 347 observations, using the following formula:

$$E_m = \left(\frac{\mathrm{ER}_{trt}}{ER_{ctrl}} - 1\right) \times 100\% \tag{2}$$

where E_m is mitigation efficiency, ER_{trt} is gas emissions in the experimental group with mitigation measures, and ER_{ctrl} is gas emissions in the control group without mitigation measures. Thus, a negative or positive E_m value indicates that the selected measure can reduce or increase gas emissions, respectively. The median E_m values for each measure were calculated using an analytical approach adapted from Benayas et al.[37] and Tuomisto et al.[38] The normality of the data was tested using the Kolmogorov−Smirnov test. Not all of the E_ms for each mitigation measure were normally distributed; therefore, the Wilcoxon Signed-Rank test was used to determine if the median E_ms were significantly different from zero when there were sufficient results for specific measures. SPSS 20.0 software was used for the statistical analyses.

2.2.4. Calculation of Gas Emissions under Mitigation Scenarios for Four Manure Management Systems. The integrated mitigation scenarios were set with individual mitigation options included into the corresponding phases of the MMS, and these scenarios are displayed in SI Table S2. The gas emissions under mitigation scenarios for the four MMSs were

DOI: 10.1021/acs.est.6b06430
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

the sum of the emissions from each phase, and were based on the numerous calculation schemes described in section 3 of SI. The calculations are presented in Data set S1 (DeepPitSystem, PullPlugSystem, BeddingSystem, and SeparationSystem tabs; select the dynamic links to other tabs to view the raw data).

2.2.5. Uncertainty Analysis. Monte Carlo simulations (1000 runs) with R (version 3.3.1) were applied to estimate the uncertainty of the system level emissions. The calculated median values of the gas emission factors, mitigation efficiency factors, as well as their 95% confidence intervals (CI) were included in the uncertainty analysis. The probability density functions (PDF) were assumed as normal distributions for each input data.[39]

As there is a total of 101 designed scenarios for the four systems, quantifying the uncertainty for all the systems would be quite complex, considering the upstream and downstream relations of N. Therefore, a partial uncertainty analysis[22] for the four baseline systems and the 12 recommended systems was conducted to illustrate the likely uncertainty ranges in the results.

3. RESULTS AND DISCUSSION

3.1. Gaseous Emission Factors (EFs) for Different Phases of the Swine Manure Management Systems. Emission factors for each phase of the MMSs were assessed from 611 observations by meta-analysis, including four in-house manure handling practices, three outdoor storage and treatment practices, and seven land application practices (detailed description in SI text) (Figure 2).

3.1.1. In-House Phase. The results show that different in-house manure collection methods have a significant impact on gas emissions, especially for CH_4 and N_2O. The CH_4 EF is largest for the deep-pit mode (median value of 64.37 kg CH_4 AU^{-1} yr^{-1}, SI Table S3), because manure in deep-pits with long storage periods is conducive to generation of CH_4 due to anaerobic conditions. The pull-plug mode with manure regularly removed has the next highest CH_4 EF of 47.09 kg CH_4 AU^{-1} $year^{-1}$. In comparison, CH_4 emissions for separation mode are much lower with an EF of 10.93 kg CH_4 AU^{-1} yr^{-1}. The bedding mode has comparatively the lowest CH_4 EF (10.63 kg CH_4 $^{-1}AU^{-1}$ yr^{-1}) but the highest N_2O EF (4.70 kg N_2O AU^{-1} yr^{-1}) due to the nitrification and denitrification processes, which are facilitated by the coexistence of aerobic and anaerobic areas in the continuously accumulating manure on the animal house floor.[40] The IQR for N_2O EF of bedding is high at 15.16, with the high variation of the N_2O EF likely due to the complex emission mechanism of N_2O. For NH_3 emissions, the bedding mode shows the lowest median value of 8.05 kg NH_3 AU^{-1} yr^{-1}; whereas for deep-pit, pull-plug and separation modes, the median NH_3 EFs are higher, in the range of 11.99−14.98 kg NH_3 AU^{-1} yr^{-1}. There are only three studies available for separation mode (SI Table S3), indicating more research is needed.

3.1.2. Outdoor Manure Storage and Treatment Phase. Slurry/lagoon storage has the largest median CH_4 EF of 50.4 kg CH_4 AU^{-1} yr^{-1}, which is much greater than that for composted manure (11.1 kg CH_4 AU^{-1} yr^{-1}) or stockpiled manure (9.4 kg CH_4 AU^{-1} yr^{-1}), as the liquid slurry storage maintains anaerobic conditions compared to solid manure storage. Slurry/lagoon storage emits almost no N_2O (Figure 2, SI Table S4), but Harper et al.[41] showed one outlier with an N_2O EF of 0.012 kg N_2O−N $(kg\ N)^{-1}$. Harper et al.[41] indicated that the NO_3^- content in the top 0.5m of lagoon can be 0−34.0 mg N kg^{-1} which may be supported by the O_2 released from algae in the slurry surface. The N_2O EF for composted manure is 0.017 kg N_2O−N $(kg\ N)^{-1}$, compared to 0.0017 kg N_2O−N $(kg\ N)^{-1}$ for manure that is statically stockpiled. Meanwhile, NH_3 EFs for the slurry/lagoon storage, composted, and stockpiled manure are 0.170, 0.249, and 0.047 kg NH_3−N $(kg\ TN)^{-1}$, respectively. Compared with solid stockpile, the consecutive air exchange, in combination with the elevated temperature due to aerobic fermentation, leads to the higher N_2O and NH_3 EFs during active composting.[42]

3.1.3. Land Application Phase. Manure contains a large quantity of C which can be converted to CH_4 when applied to flooded paddy field soils (113.4 kg CH_4 AU^{-1} yr^{-1}) (Figure 2, SI Table S5).For upland cropping systems, CH_4 emissions are low and the cropping system is usually seen as a sink for CH_4.[43] As such CH_4 emissions during manure upland application are not considered in the following system-level emission calculations.

N_2O emission from land application is approximately 0.0058 kg N_2O−N $(kg\ N)^{-1}$ for surface broadcast slurry and 0.0001 kg N_2O−N $(kg\ N)^{-1}$ for surface broadcast solid manure. Liquid slurry broadcast had a notably higher N_2O EF compared to solid manure. Liquid slurry provides nitrogen, moisture and a source of easily degradable C to the soil, and the increase in heterotrophic activity due to C turnover may provide oxygen-deficient conditions stimulating N_2O emissions for extended periods.[44] Slurry injection and rapid incorporation increased the N_2O emission factor to 0.0150 and 0.0170 kg N_2O−N $(kg\ N)^{-1}$, respectively (SI Table S5).

Compared with N_2O−N, NH_3−N loss is larger from manure land application. Surface broadcast slurry and solid manure results in high NH_3 emission factors of 0.3177 and 0.1800 kg NH_3−N $(kg\ TN)^{-1}$, respectively (Figure 2 and SI Table S5). The usually larger surface area for air contact with slurry may cause higher NH_3 volatilization than solid manure during the land application process. But the NH_3 EF of solid manure land application is lower than that during the solid manure composting process (0.249 kg NH_3−N $(kg\ TN)^{-1}$), since a large proportion of TAN is removed during the aerobic fermentation process of compost. The NH_3 emission factors for slurry injection and rapid incorporation were 0.0049 and 0.0955 kg NH_3−N $(kg\ TN)^{-1}$, respectively (Figure 2 and Table S5).

3.2. GHG and NH_3 Emissions from Baseline Scenarios of Four Manure Management Systems. Of the four MMSs, the deep-pit system has the greatest GHG emissions, reaching 3517 ± 67 (95%CI) kg CO_2-eq AU^{-1} yr^{-1}, followed by the pull-plug system (2879 ± 88 kg CO_2-eq AU^{-1} yr^{-1}), and the bedding system (2809 ± 108 kg CO_2-eq AU^{-1} yr^{-1}). The separation system has the lowest GHG emission of 1400 ± 41 kg CO_2-eq AU^{-1} yr^{-1}, which is only 40% of the emissions of the deep-pit system (Figure 3. Detailed calculations are presented in SI

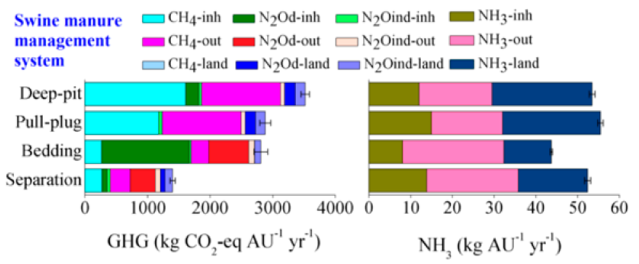


Figure 3. GHG and NH_3 emissions of baseline scenarios for deep-pit, pull-plug, bedding and separation systems as defined in Figure 1 (see Tab SummBaseEmi in Data set S1 for numeric data). N_2Od = direct N_2O emission; N_2Oind = indirect N_2O emission; in = in-house; out = outdoor; land = land application; AU = animal unit (1 AU= 500 kg).

DOI: 10.1021/acs.est.6b06430
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

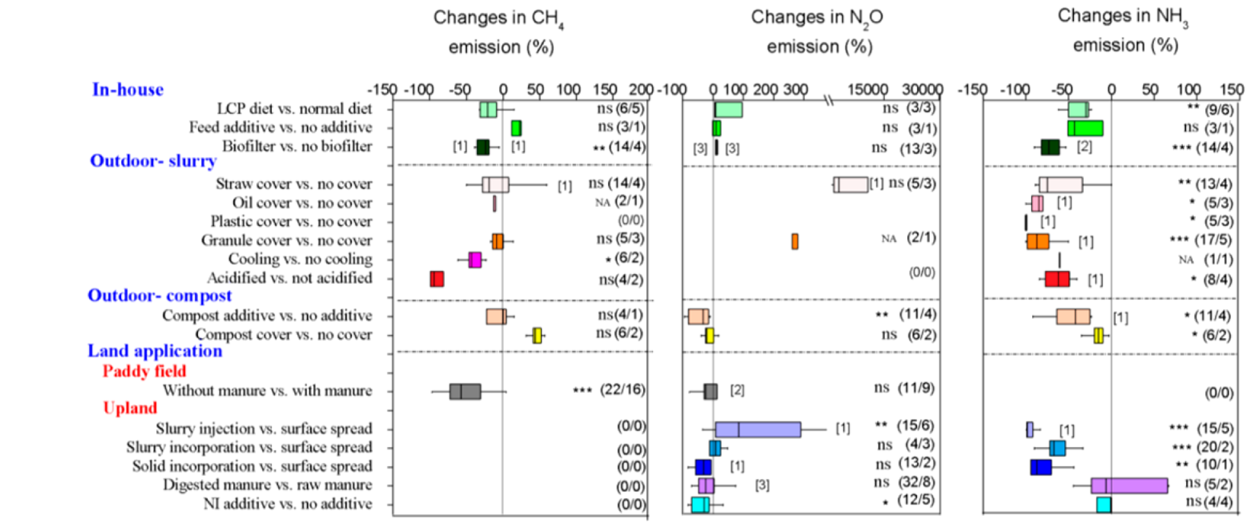


Figure 4. Box and whisker plots of the efficiency of mitigation strategies for CH_4, N_2O and NH_3 emissions (see SI Table S6−S8 for numeric data). Vertical lines of the boxplot represent the median, upper and lower quartiles. The whiskers show values that extend to 1.5 orders of box length. The numbers in the square brackets represent the number of outliers (>1.5 orders of box length). Values in parentheses indicate the number of observations for the statistical analysis, and the number of studies from which the observations originated. Wilcoxon Signed Rank test: ***$P < 0.001$; **$P < 0.01$; *$P < 0.05$; ns = not significantly different from zero; NA= not applicable. LCP= low crude protein; NI = nitrification inhibitor.

section 2, and results are presented in tab SummBaseEmi of Data set S1). The results are consistent with the life cycle analysis (LCA) study by De Vries et al.[39] which reported that separation reduced GHG emission by 66−82%. However, the relative uncertainty of the results in this study is comparatively lower than that of De Vries et al.[39] The improvement may result from using the computed median value and its 95% CI as the input parameter in this analysis, instead of the use of one point value and the high uncertainty range represented by observed min to max values.

The relative contributions of different GHGs are quite different between the four baseline systems, in that CH_4 dominates the GHG emissions of both liquid systems (deep-pit and pull-plug), but accounts for smaller GHG emissions for the pull-plug system. The reason for the lower CH_4 emission of the pull-plug system lies in its less anaerobic environment and a shorter in-house storage period than the deep-pit system. For the bedding system, N_2O is the major GHG contributor due to occurrence of nitrification and denitrification in the solid manure at different phases of the MMS, with N_2O emissions from in-house manure handling and outdoor phases representing 50% and 23% of the total GHG emissions, respectively. For the separation system, the in-house CH_4 and N_2O emissions are both relatively low because the solid fraction of the manure is removed from the house soon after excretion. Land application represents a relatively small source of the total GHG emissions from MMSs, contributing less than 9% of the whole-system emissions. Since there are no CH_4 emissions during upland manure application process, only N_2O emissions were included in the calculation of GHG emissions. In addition, the lower manure N preserved in the final stage, combined with the low direct N_2O EF factors of 0.0001−0.017 kg N_2O−N (kg N)$^{-1}$, and the low indirect N_2O EF of 1% for NH_3−N to N_2O−N, as well as 0.75% for N leaching/runoff to N_2O−N,[21] contributed to the low GHG emissions from this land application stage.

NH_3 emissions for both liquid systems of deep-pit and pull-plug are comparable at 53.4 ± 0.7 and 55.4 ± 0.7 kg AU^{-1} yr^{-1}. The bedding system has the lowest NH_3 emission factor of 43.7 ± 0.3 kg AU^{-1} yr^{-1} (Figure 3), because the NH_3 EF for surface broadcasting of solid manure is only half of that for liquid manure (Figure 2). For the two liquid systems, the land application phase dominates the NH_3 emissions for the whole system; whereas for the bedding and separation systems, the outdoor manure storage and treatment phase contributed the most, as the solid fraction has a higher NH_3 emission during the composting phase than the land application phase.

3.3. Effect of Mitigation Measures. Various mitigation practices have been developed for reducing NH_3 and GHG emissions at each phase of MMS; but only practices with available measurement data on the mitigation effect are included in this analysis. The definitions of each mitigation measure chosen here are detailed in the SI text. The changes in NH_3, N_2O and CH_4 emissions under different mitigation practices at each phase are presented in Figure 4.

3.3.1. Effect of in-House Mitigation Measures. A low crude protein (LCP) diet is highly beneficial as it limits N at source, resulting in lower N content of the excreta (17.0%, SI Table S9) and thus reduces N-related gaseous emissions during the subsequent manure management phases. This delivers a mitigation potential for NH_3 emissions during the in-house phase (30%, $p < 0.01$) and provides other environmental cobenefits, such as reduced N losses in runoff and eutrophication. Some experiments show that LCP diets may increase manure N_2O emissions,[45] although the amount is not appreciable (Figure 4).

The use of biofilters is seen as one of the most effective mitigation measures for limiting NH_3 emissions from animal houses (72%, $P < 0.001$) (Figure 4). However, some studies suggest that biofilters may increase N_2O emissions because the absorbed NH_3 from the exhaust air may be nitrified and denitrified, generating N_2O.[46] Biofilters are also effective at removing CH_4 (24%, $P < 0.01$) via oxidation.[47]

3.3.2. Effects of Outdoor Manure Storage and Treatment Mitigation Measures. For mitigation from slurry storage, almost all types of covers have proven to be effective in reducing NH_3 emissions with median mitigation efficiencies of >75%. Floating plastic cover is the most effective option with a mitigation efficiency of 99.5% ($P < 0.05$), because the plastic covering with secure sealing characteristics could help to avoid gas emissions. Floating straw and granule covers are not recommended since they may increase N_2O emissions by 29 and 2.7 times, respectively, due to nitrification and denitrification processes occurring within the slurry/additive crusts that develop,[48] although only the effect of straw cover is statistically significant (Figure 4; $P < 0.05$). Petersen et al.[49] also indicated that cumulative N_2O emission from swine slurry storage can reach 20.6−39.7 g N_2O m^{-2} with a straw cover, compared to 0−0.1 g N_2O m^{-2} without a straw cover during a 58 day summer measurement period. Meanwhile, a straw cover showed a CH_4 mitigation effect with a median value below 0, with the large IQR of 46.50%. Some studies have reported that the decomposition of straw, if used for a prolonged period, may serve as an additional carbon source for methanogens.[50] Acidification is effective in NH_3 mitigation, with a reduction efficiency of 56% ($P < 0.05$). It also results in a high CH_4 mitigation efficiency (88%, $P = 0.068$) as methanogenesis is inhibited in the acidified slurry.[51,52]

For mitigation of emissions during active composting, additives have proven to be effective in reducing NH_3 (42%, $p < 0.05$) and N_2O (32%, $p < 0.01$) emissions and improving the compost nutrient value. The only outlier that occurred for NH_3 mitigation was for the forsterite compost additive,[53] which increased NH_3 emissions by 86%, but delivered a low N_2O emission of 0.65% kgN_2O−N (kg N)$^{-1}$ (a 94% reduction of N_2O from control), since forsterite can inhibit the process of conversion of NH_3 to N_2O during composting. Bautista et al.[54] reported that the NH_4^+-N ions of compost with alum and zeolite amendment were three times greater than those of compost without the additives.

Biogas recovery and utilization exhibited a high GHG mitigation potential. However, according to 2006 IPCC guideline,[21] approximately 10% of the CH_4 generated from biogas digesters may subsequently leak to the air. Meanwhile, CH_4 loss from digestate storage is not negligible,[55] and 5−15% additional biogas yield from digestate storage has been reported.[56] All of these emissions should be taken into account when assessing the mitigation effect of biogas digesters. Unfortunately, there is no literature reporting a direct comparison of biogas digester vs. the baseline scenario. Therefore, we could not give quantitative data on the mitigation efficiency of biogas digester. A detailed calculation method was developed and presented in SI section 2.4.

3.3.3. Effects of Mitigation Measures for Land Application. Avoiding manure application to rice paddy fields is an effective GHG mitigation option, with CH_4 and N_2O mitigation efficacy of 57% ($p < 0.001$) and 23% ($p = 0.575$), respectively. Emissions from paddy fields, with vs. without manure application, could be 105−353 vs 31−108 kg ha^{-1}for CH_4, and 0.44−0.97 vs. 0.31−0.74 kg ha^{-1} for N_2O.[57] Compared with pig manure application, use of chemical fertilizers proved to be 50% lower in GHG emissions from paddy fields;[58] thus use of chemical fertilizers instead of animal manure is recommended for paddy fields. But, the emission from manufacture process of chemical fertilizers should be included in future LCA analyses.

For manure application to other crops in upland, the specific loss of NH_3−N can be reduced significantly by changing the application method from surface broadcast to injection or incorporation. Mitigation efficiency is usually higher than 70%, and the highest NH_3−N (TN)$^{-1}$ abatement (99%, $p < 0.001$) is observed for slurry injection with a low IQR of 6.90%, meaning a notable agreement between cases available. Reducing NH_3 loss means that more nitrogen is available for crop uptake, with reduced requirement for commercial fertilizers, but the increased soil mineral N pool could potentially cause higher N_2O emissions. Slurry injection may increase N_2O−N (TN)$^{-1}$ by 84% ($p < 0.01$); nevertheless, the increase of N_2O emission may still be deemed as an acceptable trade-off for the reduction in NH_3 losses[44] due to the low N_2O−N loss to TN ratio (median value of 0.7% as indicated in Figure 2). It can be seen that almost all measures used in land application showed a variety of effects on N_2O emission with the IQRs being in the range of 49% to 282% (Figure 4). The complex N_2O production processes, the variable manure and soil properties in each study lead to the variability among results for these measures.[59]

3.4. Emissions of Four Manure Management Systems under Mitigation Scenarios. GHG and NH_3 emissions corresponding to the mitigation scenarios for the four MMSs are shown in SI Figure S2. The GHG mitigation potentials for bedding and separation systems are always lower than 24%, while for the two liquid systems (deep-pit and pull-plug), some combinations of effective mitigation options can have significant GHG mitigation potentials of 47−51% (Figure 5). However, the

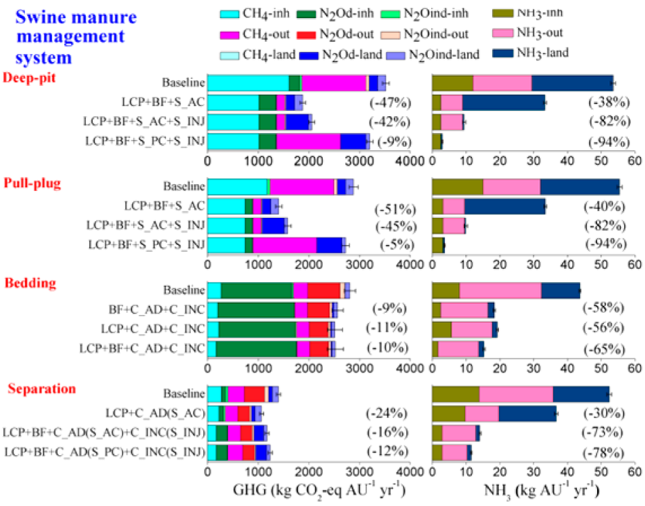


Figure 5. GHG and NH_3 emissions of baseline scenarios and recommended mitigation scenarios for deep-pit, pull-plug, bedding and separation systems, with baseline scenarios defined in Figure 1; the numbers in parentheses indicate the mitigation efficiency (see DeepPitSystem tab, PullPlugSystem tab, BeddingSystem tab and SeparationSystem tab in Data set S1 for numeric data). N_2Od = direct N_2O emission; N_2Oind = indirect N_2O emission; in = in-house; out = outdoor; land = land application; LCP = low crude protein; BF = biofilter; S_AC = slurry acidification; S_PC = slurry plastic cover; S_INJ = slurry injection; C_AD = compost additive; C_INC = compost incorporation; AU = animal unit (1 AU= 500 kg).

baseline GHG emissions from the separation system without any mitigation measures, are still lowest when compared with GHG emissions using the mitigation scenarios for the other three MMSs. The largest NH_3 reduction potential for the four MMSs could be 65−94%. The major reductions in NH_3 stem from use of plastic storage covers and changing manure application from surface broadcast to injection or rapid incorporation (Figure 5).

3.4.1. Emission Mitigation in the Deep-Pit System. Of all the mitigation strategies, the most effective GHG mitigation design for the deep-pit system is the combination of LCP diet, biofilters, and slurry acidification (LCP+BF+S_AC; 1877 kg ±54.2 CO_2-eq AU^{-1} yr^{-1}, a 47% reduction from the baseline, Figure 5; Scenario DPS-S18 in DeepPitSystem tab in Data set S1, SI Figure

DOI: 10.1021/acs.est.6b06430
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

S2A). The largest mitigation potential comes from CH_4 emissions during the outdoor (manure storage and treatment) phase. As a final step in the manure management chain, the NH_3 mitigation potential from the land application process was critical for NH_3 control, thus adding slurry injection (S_INJ) could increase the NH_3 mitigation potential from 38% to 82% compared with the LCP+BF+S_AC scenario (Figure 5). The most effective NH_3 mitigation system design is the combination of LCP diet, biofilters, plastic cover on slurry storage, and injection of slurry (LCP+BF+S_PC+S_INJ; 2.9 ± 0.1 kg NH_3 AU^{-1} yr^{-1}, a 94% reduction, Figure 5; Scenario DPS-S21 in DeepPitSystem tab in Data set S1, SI Figure S2A). The combined design of LCP diet, biofilters, slurry acidification and slurry injection (LCP+BF+S_AC+S_INJ, Scenario DPS-S19 in DeepPitSystem tab in Data set S1) would achieve both low GHG (2057 ± 55 kg CO_2-eq AU^{-1} yr^{-1}) and NH_3 (9.4 ± 0.5 kg NH_3 AU^{-1} yr^{-1}) emissions (Figure 5).

3.4.2. Emission Mitigation in the Pull-Plug System. The recommended integrated mitigation options under the pull-plug system are the same as those under the deep-pit system (Figure 5). The lowest GHG emission and NH_3 emission achieved by the mitigation combinations would be 1404 ± 63 kg CO_2-eq AU^{-1} yr^{-1} and 3.6 ± 0.2 kg NH_3 AU^{-1} yr^{-1}, respectively (SI Figure S2B).

3.4.3. Emission Mitigation in the Bedding System. The system-level GHG mitigation efficiencies of all mitigation scenarios are less than 11% from the bedding system, resulting from the high baseline N_2O emissions and a low corresponding in-house N_2O mitigation potential (see Figure 5 and SI Figure S2C). Meanwhile, the uncertainty of the GHG emission value from the designed mitigation system with LCP was greater compared with the baseline (Figure 5), due to the high uncertainty of mitigation efficiency of LCP (8% ± 42%, median ±95%CI, K31 in MitigationEffect tab in Data set S1). The combination of LCP and biofilters, compost additives and incorporation of manure in land application (LCP+BF+C_AD +C_INC) resulted in the lowest system NH_3 emission of 15.3 ± 0.3 kg AU^{-1} yr^{-1}, a 65% reduction (Figure 5; Scenario BDS-S15 in BeddingSystem tab in Data set S1).

3.4.4. Emission Mitigation in the Separation System. The separation system has the lowest baseline GHG emissions, and the GHG mitigation potentials for all the mitigation scenarios are less than 24% (Figure 5, SI Figure S2D). This phenomenon is caused by the major fraction of VS in raw manure being separated into the solid fraction (usually higher than 90%) with low CH_4 emissions. However, the mitigation potential for NH_3 could reach 78% leading to a final emission of 11.5 ± 0.2 kg NH_3 AU^{-1} yr^{-1} through use of LCP, biofilters, compost additives and incorporation of the separated solid fraction, plastic cover and injection for the separated liquid fraction (LCP+BF+C_AD-(S_PC)+C_INC(S_INJ), Figure 5; scenario SGS-S26 in SeparationSystem tab in Data set S1) since both the liquid and solid manure could achieve high NH_3 mitigation potential.

3.5. Mitigation of Gaseous Emissions by Changing the Swine Manure Management System. Liquid MMSs are widely used in large-scale confined swine operations because of simplicity in the building structure, reduced labor requirements and advanced mechanization, for example, for pumping the slurry between different manure management phases. Based on our meta-analysis, changing MMS may be advantageous for some countries, for example, with a high proportion of liquid systems, such as in The Netherland with 100% liquid production systems. In the case of The Netherlands, the national GHG emissions could be reduced by 1.3−1.8% on 1990 levels if conventional liquid pig manure systems were transferred to separation systems. This emission reduction would be significant considering the reduction for The Netherlands, as a member of EU which submitted a pledge to reduce its GHG emissions by 2020 by 20% compared to 1990 levels.[60] Furthermore, with 50% of global pork production, it is estimated that GHG emissions from China's swine industry would be 213 Tg and 85 Tg CO_2-eq in 2014 using the assumptions of all deep-pit systems and separation systems, respectively. Substituting the deep-pit system with a separation system would lead to a GHG emission reduction of 128 Tg, representing a 15.6% reduction in China's total agricultural GHG emissions, or a 1.8% reduction in China's total GHG emissions from all sources (2005 value).[23] Putting this into perspective, such GHG emission reductions in China's pig production sector, would be greater than GHG emissions for the entire agricultural sector of France, Australia, or Germany, or the total national GHG emissions of New Zealand.

With reference to NH_3 mitigation, the effect of a simple change from a deep pit system to a separation system would not be so substantial (only 1.0 kg NH_3 AU^{-1} $year^{-1}$), but changing manure application from a surface broadcasting practice to injection or incorporation is recommended. The NH_3 emissions from China's swine industry would be 3.24 Tg and 1.82 Tg NH_3 in 2014 using the assumptions of all deep-pit systems and separation systems plus injection/incorporation method, respectively. Substituting the deep-pit system with a separation system plus injection/incorporation method would lead to a NH_3 emission reduction of 1.42 Tg, representing a 14.0% reduction in China's total national NH_3 emissions (2005−2008 value).[24] Putting this into perspective, such NH_3 emission reduction in China's pig production sector would be equivalent to 40% of total NH_3 emissions from the European Union.[24]

Although this study is based on a large number of reported observations, they may or may not represent emission factors for the whole world as well as some individual countries, because of the large variety of influence factors, including climate, weather, availability of oxygen, the chemical composition of the manure (e.g., Carbon/Nitrogen-ratio), and soil properties in different locations. The application of EFs or recommended mitigation strategies should take into account these local circumstances.

In addition, economic viability will largely determine the selection and implementation of a mitigation system or measure. However, such an economic analysis is beyond the scope of this study. In addition, data are currently lacking about the economic effectiveness of various systems and mitigation measures. Future work should focus on collection of these data which will allow such economic viability analysis to occur.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b06430.

A brief description of some manure management terms, also the detailed methods, equations and assumptions for calculating the emissions for baseline and mitigation scenarios of each phase and whole systems. They are unit conversion method (Table S1); detailed set of the baseline scenario and the mitigation scenarios for each MMS (Table S2), calculated gas emission factors for pig manure management in three stages (Tables S3−5), gas mitigation efficiency of each mitigation option (Tables S6−8), and

DOI: 10.1021/acs.est.6b06430
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

other parameters used in gas emission calculation (Tables S9−12). In addition, Figure S1 shows the location and distribution of the data used in this study, and Figure S2 shows the GHG and NH_3emissions in baseline and mitigation scenarios for each MMS (PDF)

Data set 1 includes the gas emissions calculation process, the parameters used for calculation, as well as raw data from literature (XLSX)

■ AUTHOR INFORMATION

Corresponding Author

*Phone/fax: 86-10-82109979; e-mail: donghongmin@caas.cn.

ORCID

Yue Wang: 0000-0002-4194-7386

Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We thank all our colleagues for their recommendations and support during this extensive study. Funding for the study was provided by the National Basic Research Program of China (2012CB417104), the Non-Profit Research Foundation for Agriculture (201303091), China Agriculture Research System (CARS-36), and UK-China Virtual Joint Centres on Nitrogen "N-Circle" and "CINAg" funded by the Newton Fund via UK BBSRC/NERC (BB/N013484/1 and BB/N013468/1, respectively).

■ REFERENCES

(1) Gerber, P. J.; Steinfeld, H.; Henderson, B.; Mottet, A.; Opio, C.; Dijkman, J.; Falcucci, A.; Tempio, G. *Tackling Climate Change through Livestock — A Global Assessment of Emissions and Mitigation Opportunities*. Food and Agriculture Organization of the United Nations (FAO): Rome, 2013.

(2) Davidson, E. A. The contribution of manure and fertilizer nitrogen to atmospheric nitrous oxide since 1860. *Nat. Geosci.* **2009**, *2* (9), 659−662.

(3) Pelletier, N.; Tyedmers, P. Forecasting potential global environmental costs of livestock production 2000−2050. *Proc. Natl. Acad. Sci. U. S. A.* **2010**, *107* (43), 18371−18374.

(4) Steinfeld, H.; Gerber, P. J. Livestock production and the global environment: Consume less or produce better? *Proc. Natl. Acad. Sci. U. S. A.* **2010**, *107* (43), 18237−18238.

(5) Steinfeld, H.; Gerber, P.; Wassenaar, T.; Castel, V.; Rosales, M.; De Haan, C. *Livestock'S Long Shadow*; Food and Agriculture Organization of the United Nations (FAO): Rome, 2006.

(6) Ripple, W. J.; Smith, P.; Haberl, H.; Montzka, S. A.; McAlpine, C.; Boucher, D. H. Ruminants, climate change and climate policy. *Nat. Clim. Change* **2014**, *4* (1), 2−5.

(7) Eshel, G.; Shepon, A.; Mskov, T.; Milo, R. Partitioning United States' feed consumption among livestock categories for improved environmental cost assessments. *J. Agric. Sci.* **2015**, *153*, 432−445.

(8) The Economist. Swine in China: Empire of the pig. http://www.economist.com/news/christmas-specials/21636507-chinas-insatiable-appetite-pork-symbol-countrys-rise-it-also (accessed January 1, 2016).

(9) Clarisse, L.; Clerbaux, C.; Dentener, F.; Hurtmans, D.; Coheur, P. F. Global ammonia distribution derived from infrared satellite observations. *Nat. Geosci.* **2009**, *2* (7), 479−483.

(10) Galloway, J. N.; Townsend, A. R.; Erisman, J. W.; Bekunda, M.; Cai, Z.; Freney, J. R.; Martinelli, L. A.; Seitzinger, S. P.; Sutton, M. A. Transformation of the nitrogen cycle: recent trends, questions, and potential solutions. *Science* **2008**, *320* (5878), 889−892.

(11) Bai, Z. H.; Ma, L.; Qin, W.; Chen, Q.; Oenema, O.; Zhang, F. S. Changes in pig production in China and their effects on nitrogen and phosphorus use and losses. *Environ. Sci. Technol.* **2014**, *48* (21), 12742−12749.

(12) Stokstad, E. Ammonia pollution from farming may exact hefty health costs. *Science* **2014**, *343* (6168), 238−238.

(13) Larson, C. China's lakes of pig manure spawn antibiotic resistance. *Science* **2015**, *347* (6223), 704−704.

(14) The First National Pollution Census Bulletin of The People's Republic of China (in Chinese). China National Environmental Protection Agency (CNEPA): Beijing, 2010; http://www.stats.gov.cn/tjsj/tjgb/qttjgb/qgqttjgb/201002/t20100211_30641.html (accessed June 1, 2016).

(15) Hou, Y.; Velthof, G. L.; Oenema, O. Mitigation of ammonia, nitrous oxide and methane emissions from manure management chains: a meta-analysis and integrated assessment. *Glob. Change Biol.* **2015**, *21* (3), 1293−1312.

(16) Galloway, J. N.; Dentener, F. J.; Capone, D. G.; Boyer, E. W.; Howarth, R. W.; Seitzinger, S. P.; Asner, G. P.; Cleveland, C. C.; Green, P. A.; Holland, E. A.; Karl, D. M.; Michaels, A. F.; Porter, J. H.; Townsend, A. R.; Vorosmarty, C. J. Nitrogen cycles: past, present, and future. *Biogeochemistry* **2004**, *70* (2), 153−226.

(17) Bai, Z.; Ma, L.; Jin, S.; Ma, W.; Velthof, G. L.; Oenema, O.; Liu, L.; Chadwick, D.; Zhang, F. S. Nitrogen, phosphorus, and potassium flows through the manure management chain in China. *Environ. Sci. Technol.* **2016**, *50* (24), 13409−13418.

(18) Bouwman, A. F.; Boumans, L. J. M.; Batjes, N. H. Estimation of global NH3 volatilization loss from synthetic fertilizers and animal manure applied to arable lands and grasslands. *Glob. Biogeochem. Cyles* **2002**, *16* (2), 8-1−8-14.

(19) Bergström, A. N. N. K.; Jansson, M. Atmospheric nitrogen deposition has caused nitrogen enrichment and eutrophication of lakes in the northern hemisphere. *Glob. Change Biol.* **2006**, *12* (4), 635−643.

(20) Barsanti, K. C.; McMurry, P. H.; Smith, J. N. The potential contribution of organic salts to new particle growth. *Atmos. Chem. Phys.* **2009**, *9* (9), 2949−2957.

(21) Intergovernmental Panel on Climate Change (IPCC). *IPCC 2006 Guidelines for National Greenhouse Gas Inventories Vol. 4: Agriculture, Forestry, and Other Landuse*; Institute for Global Environmental Strategies: Hayama, Japan, 2006.

(22) MacLeod, M.; Gerber, P.; Mottet, A.; Tempio, G.; Falcucci, A.; Opio, C.; Vellinga, T.; Henderson, B.; Steinfeld, H. *Greenhouse gas emissions from pig and chicken supply chains — A global life cycle assessment*; Food and Agriculture Organization of the United Nations (FAO): Rome, 2013.

(23) Second National Communication on Climate Change of the People's Republic of China. China National Development and Reform Commission (NDRC): Beijing, 2013; http://qhs.ndrc.gov.cn/zcfg/201404/t20140415_606980.html (accessed June 1, 2016).

(24) Paulot, F.; Jacob, D. J.; Pinder, R. W.; Bash, J. O.; Travis, K.; Henze, D. K. Ammonia emissions in the United States, European Union, and China derived by high-resolution inversion of ammonium wet deposition data: Interpretation with a new agricultural emissions inventory (MASAGE_NH_3). *J. Geophys Res.* **2014**, *119* (7), 4343−4364.

(25) Chadwick, D.; Sommer, S.; Thorman, R.; Fangueiro, D.; Cardenas, L.; Amon, B.; Misselbrook, T. Manure management: implications for greenhouse gas emissions. *Anim. Feed Sci. Technol.* **2011**, *166*, 514−531.

(26) Wang, Y.; Dong, H.; Zhu, Z.; Li, L.; Zhou, T.; Jiang, B.; Xin, H. CH_4, NH_3, N_2O and NO emissions from stored biogas digester effluent of pig manure at different temperatures. *Agric., Ecosyst. Environ.* **2016**, *217*, 1−12.

(27) Chan, Y. C.; Sinha, R. K.; Wang, W. Emission of greenhouse gases from home aerobic composting, anaerobic digestion and vermicomposting of household wastes in Brisbane (Australia). *Waste Manage. Res.* **2011**, *29* (5), 540−548.

DOI: 10.1021/acs.est.6b06430
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

(28) Sommer, S. G.; Petersen, S. O.; Møller, H. B. Algorithms for calculating methane and nitrous oxide emissions from manure management. *Nutr. Cycling Agroecosyst.* **2004**, *69* (2), 143−154.

(29) Petersen, S. O.; Sommer, S. G. Ammonia and nitrous oxide interactions: Roles of manure organic matter management. *Anim. Feed Sci. Technol.* **2011**, *166*, 503−513.

(30) Gerber, P. J.; Henderson, B.; Makkar, H. P., Eds.; *Mitigation of Greenhouse Gas Emissions in Livestock Production − A Review of Technical Options for Non-CO_2 Emissions*; Food and Agriculture Organization of the United Nations (FAO): Rome, 2013.

(31) Gerber, P. J.; Hristov, A. N.; Henderson, B.; Makkar, H.; Oh, J.; Lee, C.; Meinen, R.; Montes, F.; Ott, T.; Firkins, J.; Rotz, A.; Dell, C.; Adesogan, A. T.; Yang, W. Z.; Tricarico, J. M.; Kebreab, E.; Waghorn, G.; Dijkstra, J.; Oosting, S. Technical options for the mitigation of direct methane and nitrous oxide emissions from livestock: a review. *Animal* **2013**, *7* (s2), 220−234.

(32) Hodgson, C. J.; Oliver, D. M.; Fish, R. D.; Bulmer, N. M.; Heathwaite, A. L.; Winter, M.; Chadwick, D. R. Seasonal persistence of faecal indicator organisms in soil following dairy slurry application to land by surface broadcasting and shallow injection. *J. Environ. Manage.* **2016**, *183*, 325−332.

(33) The United Nations Framework Convention on Climate Change (UNFCCC). National GHG Emission Inventory Database. 2014. http://unfccc.int/ghg_data/ghg_data_unfccc/items/4146.php.

(34) Bittman, S.; Dedina, M.; Howard, C. M.; Oenema, O.; Sutton, M. A. *Options for Ammonia Mitigation: Guidance from the UNECE Task Force on Reactive Nitrogen*. Centre for Ecology and Hydrology, Edinburgh, UK, 2014.

(35) Fedorov, S. GetData Graph Digitizer version 2.2.4. 2002. www.getdata-graph-digitizer.com.

(36) Larson-Hall, J.; Herrington, R. Improving data analysis in second language acquisition by utilizing modern developments in applied statistics. *Appl. Linguist.* **2010**, *31* (3), 368−390.

(37) Benayas, J. M. R.; Newton, A. C.; Diaz, A.; Bullock, J. M. Enhancement of biodiversity and ecosystem services by ecological restoration: a meta-analysis. *Science* **2009**, *325*, 1121−1124.

(38) Tuomisto, H. L.; Hodge, I. D.; Riordan, P.; Macdonald, D. W. Does organic farming reduce environmental impacts?−A meta-analysis of European research. *J. Environ. Manage.* **2012**, *112*, 309−320.

(39) De Vries, J. W.; Aarnink, A. J.; Groot Koerkamp, P. W.; De Boer, I. J. Life cycle assessment of segregating fattening pig urine and feces compared to conventional liquid manure management. *Environ. Sci. Technol.* **2013**, *47* (3), 1589−1597.

(40) Philippe, F. X.; Laitat, M.; Wavreille, J.; Nicks, B.; Cabaraux, J. F. Influence of permanent use of feeding stalls as living area on ammonia and greenhouse gas emissions for group-housed gestating sows kept on straw deep-litter. *Livest. Sci.* **2013**, *155* (2), 397−406.

(41) Harper, L. A.; Sharpe, R. R.; Parkin, T. B. Gaseous nitrogen emissions from anaerobic swine lagoons: Ammonia, nitrous oxide, and dinitrogen gas. *J. Environ. Qual.* **2000**, *29* (4), 1356−1365.

(42) Nelson, M. I.; Marchant, T. R.; Wake, G. C.; Balakrishnan, E.; Chen, X. D. Self-heating in compost piles due to biological effects. *Chem. Eng. Sci.* **2007**, *62* (17), 4612−4619.

(43) Le Mer, J.; Roger, P. Production, oxidation, emission and consumption of methane by soils: a review. *Eur. J. Soil Biol.* **2001**, *37* (1), 25−50.

(44) Thomsen, I. K.; Pedersen, A. R.; Nyord, T.; Petersen, S. O. Effects of slurry pre-treatment and application technique on short-term N_2O emissions as determined by a new non-linear approach. *Agric., Ecosyst. Environ.* **2010**, *136* (3), 227−235.

(45) Le, P.; Aarnink, A.; Jongbloed, A. Odour and ammonia emission from pig manure as affected by dietary crude protein level. *Livest. Sci.* **2009**, *121* (2), 267−274.

(46) Maia, G. D.; Gates, R. S.; Taraba, J. L. Ammonia biofiltration and nitrous oxide generation during the start-up of gas-phase compost biofilters. *Atmos. Environ.* **2012**, *46*, 659−664.

(47) Melse, R. W.; van der Werf, A. W. Biofiltration for mitigation of methane emission from animal husbandry. *Environ. Sci. Technol.* **2005**, *39* (14), 5460−5468.

(48) Berg, W.; Brunsch, R.; Pazsiczki, I. Greenhouse gas emissions from covered slurry compared with uncovered during storage. *Agric., Ecosyst. Environ.* **2006**, *112* (2), 129−134.

(49) Petersen, S. O.; Dorno, N.; Lindholst, S.; Feilberg, A.; Eriksen, J. Emissions of CH_4, N_2O, NH_3 and odorants from pig slurry during winter and summer storage. *Nutr. Cycling Agroecosyst.* **2013**, *95* (1), 103−113.

(50) Amon, B.; Kryvoruchko, V.; Amon, T.; Zechmeister-Boltenstern, S. Methane, nitrous oxide and ammonia emissions during storage and after application of dairy cattle slurry and influence of slurry treatment. *Agric., Ecosyst. Environ.* **2006**, *112* (2), 153−162.

(51) Petersen, S. O.; Højberg, O.; Poulsen, M.; Schwab, C.; Eriksen, J. Methanogenic community changes, and emissions of methane and other gases, during storage of acidified and untreated pig slurry. *J. Appl. Microbiol.* **2014**, *117* (1), 160−172.

(52) Bastami, M. S. B.; Jones, D. L.; Chadwick, D. R. Reduction of methane emission during slurry storage by the addition of effective microorganisms and excessive carbon source from brewing sugar. *J. Environ. Qual.* **2016**, *45* (6), 2016−2022.

(53) Zheng, J.; Wei, Y. S.; Wu, X. F.; Zeng, X. L.; Han, S. H.; Fang, Y. Nutrients conservation of N & P and greenhouse gas reduction of N2O emission during swine manure composting. *Environ. Sci.* **2011**, *32* (7), 2047−2055 (in Chinese with English abstract).

(54) Bautista, J. M.; Kim, H.; Ahn, D. H.; Zhang, R.; Oh, Y. S. Changes in physicochemical properties and gaseous emissions of composting swine manure amended with alum and zeolite. *Korean J. Chem. Eng.* **2011**, *28* (1), 189−194.

(55) Gioelli, F.; Dinuccio, E.; Balsari, P. Residual biogas potential from the storage tanks of non-separated digestate and digested liquid fraction. *Bioresour. Technol.* **2011**, *102*, 10248−10251.

(56) Weiland, P. Production and energetic use of biogas from energy crops and wastes in Germany. *Appl. Biochem. Biotechnol.* **2003**, *109* (1−3), 263−274.

(57) Pandey, A.; Mai, V. T.; Vu, D. Q.; Bui, T. P.; Mai, T. L.; Jensen, L. S.; Neergaard, A. Organic matter and water management strategies to reduce methane and nitrous oxide emissions from rice paddies in Vietnam. *Agric., Ecosyst. Environ.* **2014**, *196*, 137−146.

(58) Qin, X. B.; Li, Y. E.; Liu, K. Y.; Wan, Y. F. Methane and nitrous oxide emission from paddy field under different fertilization treatments. *T. CSAE* **2006**, *22* (7), 143−148 (in Chinese with English abstract).

(59) Velthof, G. L.; Kuikman, P. J.; Oenema, O. Nitrous oxide emission from animal manures applied to soil under controlled conditions. *Biol. Fertil. Soils.* **2003**, *37* (4), 221−230.

(60) European Commission. Climate action: Commission proposes ratification of second phase of Kyoto Protocol. http://europa.eu/rapid/press-release_IP-13-1035_en.htm (accessed January 20, 2017).

DOI: 10.1021/acs.est.6b06430
Environ. Sci. Technol. XXXX, XXX, XXX−XXX