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Short Communication

Different characteristics of greenhouse gases and ammonia emissions from conventional stored dairy cattle and swine manure in China



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HIGHLIGHTS

GRAPHICAL ABSTRACT

- Quantifying GHGs and NH₃ emissions of solid stored manure from dairy cattle and swine
- Dairy cattle manure emits similar amounts of GHGs but lower NH₃ compared to swine manure.
- Physicochemical characteristics of manure significantly influence gases emissions.



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ABSTRACT

Livestock manure emits considerable amounts of greenhouse gases (GHGs) and ammonia (NH₃), inducing climate change and air pollution. However, there remains a lack of knowledge in the literature related to GHGs and NH₃ emissions from the manure of various livestock species. This study reports on a field observation we conducted to analyze GHGs and NH₃ emissions of solid stored manure from dairy cattle and swine, which represent the two main livestock species raised in China. Results showed that although dairy cattle manure emitted 521.9% more methane (CH₄) than swine manure, they separately emitted 50.8% and 40.9% less nitrous oxide (N₂O) and carbon dioxide (CO₂) emissions, respectively. With respect to their global warming potential, the GHGs emission from dairy cattle manure was similar to that from swine manure. NH₃ emissions from swine manure were significantly higher, namely, greater by a factor of 2.4 compared to dairy cattle manure. Differences in gas emissions between dairy cattle and swine manure can be explained by differences in the physicochemical characteristics of their manure and their associated microbiological, chemical, and physical processes that produce gas during

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Solid manure storage Mitigation strategies storage periods. Based on our results, this study highlights the necessity for prospective mitigation strategies to simultaneously decrease GHGs and NH₃ emissions from livestock manure. Our findings provide useful implications for understanding GHGs and NH₃ emissions, which can be used to develop corresponding mitigation strategies for livestock manure management in China.

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1. Introduction

In response to the increasing demand for milk and meat, dairy cattle and swine populations in China have increased by 191.7% and 15.1% between 2000 and 2015, respectively, with a corresponding increase in the manure produced (China Agricultural Statistical Yearbook, 2000–2015). China produces a significant amount of manure, having reached 3.8 billion tons in 2017 (MOA, 2017). Livestock manure is rich in nutrients (Bai et al., 2016), such as organic matter, nitrogen (N), phosphorus (P), and potassium (K), which could potentially provide numerous benefits in maintaining crop nutrient demands (Chen et al., 2014), subsequently increasing soil fertility (Wang et al., 2015). In addition, substituting livestock manure for chemical fertilizers could reduce reactive N emissions (Zhou et al., 2016; Xia et al., 2017).

In China, only 43% of manure is applied to the cropland, and most of the amount is discarded (Gu et al., 2015). Accordingly, such improper manure management has caused a substantial amount of greenhouse gases (GHGs) emissions to be released into the atmosphere, including carbon dioxide (CO_2) , nitrous oxide (N_2O) , and methane (CH_4) (Herrero et al., 2016; Wang et al., 2017; Zhuang et al., 2019a, 2019b). Livestock manure is also an important source of ammonia (NH₃), a compound that can react with acids (e.g., nitric acid (HNO₃) and sulfuric acid (H_2SO_4)) to promote the formation of fine particulate matter (PM) (Seinfeld and Pandis, 2006; Fang et al., 2009), leading to soil acidification (Bouwman et al., 2002), eutrophication (Bergström and Jansson, 2006), and environmental degradation (e.g., biodiversity loss) (Matson et al., 2002). It is estimated that livestock manure contributes approximately 11%, 29%, and 52% of agriculturally-derived CH₄, N₂O, and NH₃ emissions in China, respectively (Zhuang et al., 2019a, 2019b; Kang et al., 2016).

GHGs and NH₃ emissions from livestock manure are produced during four phases (Chadwick et al., 2011): i) livestock housing, ii) manure storage, iii) treatment, iv) land application. Understanding livestock manure emissions during all four phases is a prerequisite for their successful mitigation. Nevertheless, most relevant studies have focused on emissions sourced from livestock housing (Philippe et al., 2013; Borhan et al., 2013; McGinn and Flesch, 2018), manure treatment (Zhang et al., 2017; Cao et al., 2019; Mao et al., 2019; Jiang et al., 2018), and land application phases (Cai et al., 2017; Jeong et al., 2018; Zhang et al., 2019). In addition, mitigation strategies for each phase have been suggested (Hou et al., 2015; Wang et al., 2017, 2018). However, while studies have tended to ignore emissions from the manure storage phase, this phase plays a crucial role as it pertains to the whole manure management system (Masse et al., 2008; Hou et al., 2015; Li, 2016; Bai et al., 2017; Maldaner et al., 2018). Unlike the other phases, manure storage management can alter the nutritional constituents of manure, thus leading to emissions from its treatment and application (Evans et al., 2018; Holly et al., 2017) while influencing plant nutrient uptake (Zhang et al., 2019). Although a few studies on GHGs and NH₃ emissions have been conducted during the livestock manure storage phase, no comprehensive investigation of emission patterns during this phase has been conducted (Bai et al., 2017). For instance, although some studies have focused on single gas source emissions from manure, no studies have reported on simultaneous GHGs and NH₃ estimations (McGinn and Flesch, 2018). This hinders our understanding of certain gases that may result from ineffectual mitigation strategies given that such strategies could reduce emissions from one specific gas while simultaneously increasing another (Chadwick et al., 2011; Holly et al., 2017). Wang et al. (2018) reported that during the manure storage phase, composting manure could decrease CH₄ emissions while increasing N₂O emissions compared to stockpiling manure. In addition, other studies reported on the existence of a trade-off between NH₃ and N₂O emissions (Sommer et al., 2004; Wang et al., 2017). Moreover, existing studies on gas emissions during the manure storage phase have mostly focused on "individual" livestock species rather than livestock "categories" (Dai et al., 2015). Indeed, a direct comparison between characteristics of gases emissions from the manure of different livestock species remains scarce in the literature (Dai et al., 2015). However, temperature, the carbon-to-nitrogen ratio (C:N), pH, water content, oxygen (O_2) availability, and environmental factors have all been shown to influence manure storage emissions differently (Dai et al., 2015). Hence, ignoring differences in livestock categories may result in a significant deviation from real world conditions when relative mitigation strategies are implemented (Zhuang et al., 2019a, 2019b). Therefore, it is essential to simultaneously quantify and compare GHGs and NH₃ emissions from different livestock categories during the manure storage phase to develop appropriate mitigation policies.

The livestock sector of China has undergone a rapid transition over the past few decades. These changes will inevitably have profound effects on GHGs and NH₃ emissions from livestock manure, being the main source of such emissions (Zhuang et al., 2019a). Dairy cattle and swine, respectively, belong to ruminant and monogastric animal species, representing two inherently different livestock types, making them an interesting case study. Furthermore, no studies to date have analyzed GHGs and NH₃ emissions during the manure storage phase of these two livestock categories, particularly in China. This study accordingly measures CH₄, N₂O, CO₂, and NH₃ emission characteristics of traditional solid dairy cattle and swine manure storage practices in China based on a field experiment and laboratory analysis, and then compares GHGs and NH₃ emissions between the two. Finally, we discuss our results and propose potential mitigation strategies regarding manure sourced from these two livestock types.

2. Materials and methods

2.1. Experiment sites

Our experiment was conducted at the Changsha Research Station for Agricultural and Environmental Monitoring, Institute of Subtropical Agriculture, Chinese Academy of Sciences, in Jinjing Town, Changsha County, Hunan Province, China (28°33' N, 113°19' E). This region is in a subtropical monsoon zone that is hot and rainy in summer and mild and drier in winter. The mean annual temperature and the average annual precipitation are 17.5 °C and 1330 mm, respectively. Table S1 provides data on the temperature and precipitation during the experimental period. The topography is hilly and mountainous in the north and northwest of Jinjing Town, and plains dominate in the east and the south of the region. The four main land-use types in this region are paddy fields, dry land, forestland, and tea gardens. See Liang et al. (2015) for detailed information of the study area.

2.2. Experimental material and design

We collected fresh swine and dairy manure from local swine and dairy cattle farms. According to the traditional practices of local farmers, swine or dairy cattle manure and rice straw were mixed at the ratio of 7:1 on a basis of their wet weight and then stacked into cones (1.5 m width by 0.8 m height). The initial pile weight of each cone was 500 kg with three replications. Each pile was stored in the open air. Fig. 1 shows the overall storage arrangement. According to the conventional storage period used by farmers, the storage period for this study began on July 25, 2015, and ended on August 26, 2015, namely, a cumulative period of 33 days.

We measured the physicochemical characteristics of raw materials and piles, and results showed that the C:N ratio and the moisture content of solid mixed swine manure were 14 and 61.4%, respectively (Table 1).

2.3. GHGs, NH₃ and environmental factor measurements

We measured NH₃ emissions using the Dräger-Tube method (DTM) as used by Pacholski et al. (2006). The measurement system included a gas capture hood, a Teflon indicator tube, and a hand pump. The measuring device (Drägerwerk AG, Lübeck, Germany) acted as a hand pump. Each hood covered an area of 104 cm^2 , with a 0.5 mm air inlet and an air outlet on top of the hood to allow for the movement of gas. Each outlet was fastened to a Teflon tube to connect four gas collecting hoods together. The four gas capture hoods were placed on the surface of manure to collect NH₃. By extracting the surface gas of manure semicontinuously using a hand pump, the gas was free to pass through the Teflon tube and enter into the ammonia detection tube (Drägerwerk AG, Lübeck, Germany). The ammonia detection tube changed the color of the gas blue, and the concentration of NH₃ volatilization on the surface of manure was that which corresponded to the blue band (Roelcke et al., 2002; Pacholski et al., 2006). At the same time, we recorded the total duration (seconds) of gas extractions, the number of gas extractions, as well as air temperature, air pressure, and wind speed (Gericke et al., 2011; Pietzner et al., 2017). Following this, we collected three gas samples from each treatment. We analyzed NH₃ concentrations using the NH₃ concentration detection tube.

Furthermore, CO₂, N₂O, and CH₄ emissions were measured using static-chamber-gas chromatography as described by Zhuang et al. (2019b). Chambers with dimensions of 1.5 m × 1.5 m × 1 m (length × width × height) were placed on a fixed polyvinyl chloride (PVC) frame wherein three extractor vents were added for gas sampling. For each chamber, a circulating fan was installed inside the chamber

headspace to ensure the uniform mixing of chamber air. Moreover, each chamber was wrapped in a layer of insulating material to minimize changes in internal air temperature. Gas measurements were taken daily at 09:00–11:00 throughout the experimental period. Gas samples (60 ml) were collected at 0, 15, and 30 min after chamber closure. We collected three gas samples from each treatment. We used gas chromatography (Agilent 7890A, USA) to analyze CO₂, N₂O, and CH₄ sample concentrations, while we used nonlinear regression to calculate CH₄, N₂O, and CO₂ flux (Wang et al., 2013).

Samples were also collected from the surface, middle, and bottom layers of piles, and then they were evenly mixed. Uniform samples were then divided into two parts: the first part was air-dried to measure water content, total C and N, and pH, while the second part was immediately frozen to be used as fresh samples to measure nitrate nitrogen (NO_3-N) and ammonium nitrogen (NH_4^+-N) . We adopted the weighing method, elemental analyzer, pH detector, and flow analyzer described by the industrial standard for agriculture of the People's Republic of China (NO. NY 525-2002) to measure water content, total C and N. pH, and NO_3^--N and NH_4^+-N , respectively. Solid manure samples were oven dried at 75 °C to a constant weight to obtain the water content. Total C and N were measured by a CN elemental analyzer (Vario MAX CN, Elementar, Germany). NO₃⁻-N and NH₄⁺-N were extracted with 2 mol L^{-1} KCl (1:10, w/v) and then analyzed by a flow injection analyzer (FIA Star 5000, Foss, Hillerød, Denmark). pH was determined using 5 g of the air-dried subsample and 2 mm of sieved manure (at a water ratio of 1:10 (w/v)). In addition, we used a temperatureindicating instrument (JM624, Jiming Instrument Co., Ltd., China) to record the temperature of piles when collecting solid samples.

2.4. Data analysis

Eqs. (1) and (2) were used to calculate NH_3 and GHGs flux, respectively, as follows:

$$F_{Ng} = V \times |conc| \times 10^{-6} \times pNH_3 \times U_N \times U_F \times U_Z \tag{1}$$

where F_{Ng} is the NH₃ flux (mg NH₃-N m⁻² h⁻¹); *V* is the extraction volume (L); |conc| is the NH₃ concentration (µg·L⁻¹); pNH₃ is the NH₃ density (mg·L⁻¹); U_N is the conversion factor from NH₃ to N; U_F is the conversion factor of the surface area (m²); U_Z is the conversion factor of time.

$$F = \rho H \frac{dc}{dt} \frac{273}{273 + T P_0}$$
(2)



Fig. 1. Design and pattern of the experimental solid manure storage plot.

Table 1

Ph	vsicochemical	characteristics	of	manure	from	swine	and	dairv	cattle.

Livestock types	Raw materials	Water content (%)	C:N	$NO_{3}^{-}-N (mg \cdot g^{-1})$	$NH_{4}^{+}-N (mg \cdot g^{-1})$	pH
Swine	Swine manure	73.0	12.0	0.097	2.77	6.48
	Rice straw	12.0	34.6	-	-	-
Dairy cattle	Dairy cattle manure	71.5	13.4	0.18	4.10	8.48
	Rice straw	12.0	34.6	-	-	-

where F is the gas flux $(g m^{-2} h^{-1})$; ρ is the gas density $(g \cdot L^{-1})$; H is the height of the chamber (m); T is the temperature (°C); P is the atmospheric pressure under sampling (mmHg); P_0 is the standard atmospheric pressure (mmHg); $\frac{dc}{dt}$ is the rate of gas accumulation in the

chamber.

Eq. (3) was used to calculate cumulative $\rm NH_3,$ CO_2, CH_4, and $\rm N_2O$ as follows:

$$Q = \frac{\sum t_i (F_t + F_{t+1})}{2}$$
(3)

where Q is the cumulative gas flux; i is the sampling frequency; F is the gas flux (g $m^{-2} h^{-1}$); t is the interval (time) between two adjacent measurements.

The global warming potential (GWP) was used to convert CO_2 (1), CH_4 (28), and N_2O (298) to CO_2 equivalents over a 100-year time horizon (Myhre et al., 2013). Eq. (4) was used to express the total GHGs emissions as follows:

$$GHGs (CO_2 equivalent) = CO_2 + 28CH_4 + 298N_2O$$
(4)

Analysis of variance was conducted using SPSS statistical software (version 19) to test differences in physicochemical characteristics prior to and following our experiment as well as cumulative gases emissions between swine manure and dairy cattle manure during the solid storage phase. Effects of different livestock manure types were considered significantly different if P < 0.05.

3. Results and discussion

3.1. Physicochemical characteristics of solid swine and dairy cattle manure storage

Various physicochemical indicators from solid swine and dairy cattle manure differed prior to and following our experiment (Table 2). Compared to pre experimental values, the water content of swine manure after 32 days of solid storage decreased significantly (by 16.4%) while the temperature increased (by 22.2%). By contrast, the water content and temperature of dairy cattle manure during solid storage remained consistent prior to and following our experiment. Water content differences between swine and dairy cattle manure could be attributed to their different properties. On the one hand, the bulk density of dairy cattle manure (748.67 \pm 16.12 g·cm⁻³). On the other hand, dairy cattle

manure contained much more cellulose and hemicellulose in comparison to swine manure, which are beneficial for absorbing and storing water (Dai et al., 2015; Zhang, 2018). In addition, no significant changes were observed in pH levels from the solid manure of either swine or dairy cattle prior to and following our experiment. However, NH₃ release has been shown to decrease pH, while CO₂ emissions have been shown to increase pH (Dai et al., 2015). This could be attributed to a trade-off between NH₃ release and CO₂ emissions from manure (Dai et al., 2015).

The C:N ratio decreased by 32.1% for swine manure and by 22.2% for dairy cattle manure over the storage period. This decrease in the C:N ratio during the solid storage phase of swine and dairy cattle manure could be explained by changes in degradation rates, namely, the C degradation rate exceeded the N degradation rate (Table S2). Moreover, NO_3^- -N content in the solid manure of swine and dairy cattle decreased significantly by 32.1% and 22%, respectively, which was mainly attributed to denitrification processes throughout the experimental period. In addition, we also observed a 25% increase in NH_4^+ -N content of swine manure and a 40% decrease of dairy cattle manure, which further affected the differences in N_2O and NH_3 released between swine and dairy cattle manure.

3.2. Characteristics of gases emissions

3.2.1. N₂O

N₂O emission flux ranged from 0.08 to 6.34 mg N m⁻² h⁻¹ for swine manure and from 0.06 to 7.16 mg N m⁻² h⁻¹ for dairy cattle manure (Fig. 2A). As a whole, N₂O emission flux from swine manure was higher than that from dairy cattle manure, which is consistent with comparative results from two meta-analysis studies sourced by Wang et al. (2017, 2018). This phenomenon is largely explained by differences in initial NO₃⁻-N content that acted as the primary denitrification substrate between swine manure and dairy cattle manure. In addition, we also observed that the majority of N₂O emission flux from swine and dairy cattle manure occurred during the early manure storage phase (Fig. 2A). High NO₃⁻-N content, an appropriate water content and C:N ratio, and a suitable environment that favors microbial activity together helped stimulate denitrification, thus resulting in a large amount of N₂O emissions (Monteny et al., 2006; Nelson et al., 2007; Chadwick et al., 2011; Awasthi et al., 2018). By contrast, along with an extension in the duration of the manure storage phase, particularly during the latter period (i.e., after 25 days), N₂O emission flux remained relatively low and more stable. This could be attributed to a decrease in the C:N ratio and anaerobic environmental conditions that will occur under

Table 2

Physicochemical characteristics of swine and dairy cattle manure prior to and following our experiment.

Indicator	Solid swine manure storage		Solid dairy cattle manure storage			
	Prior to the experiment	Following the experiment	Prior to the experiment	Following the experiment		
Water content (%)	$61.4 \pm 4.0a$	$51.3 \pm 2.4b$	65.6 ± 2.7a	$73.0\pm0.8a$		
Temperature (°C)	$29.9 \pm 0.1b$	$35.8\pm0.8a$	$33.8 \pm 0.6a$	$32.1 \pm 0.2a$		
рН	$6.6 \pm 0.1a$	$7.3 \pm 0.1a$	$8.5\pm0.1a$	$8.8\pm0.2a$		
C:N	$14.0 \pm 0.4a$	$9.5 \pm 0.1b$	$20.0 \pm 1.8a$	15.6 ± 0.2b		
$NO_3^-N (mg \cdot g^{-1})$	9.7 ± 1.3a	$6.7 \pm 0.3b$	$3.8\pm0.6a$	$1.9 \pm 0.2b$		
NH_4^+ -N (mg·g ⁻¹)	$0.5~\pm~0.1b$	$0.6\pm0.0a$	$0.5\pm0.1a$	$0.3 \pm 0.0b$		

Note: Different letters indicate a significant difference prior to and following our experiment at a level of 0.05.



Fig. 2. Characteristics of N₂O (A), CH₄ (B), CO₂ (C), and NH₃ (D) emissions from dairy cattle and swine manure.

physiochemical conditions, which jointly inhibit microbial activity associated with denitrification processes (Maeda et al., 2011; Lovanh et al., 2014).

3.2.2. CH₄

CH₄ emission flux from dairy cattle manure exhibited large variations, ranging from 29.5 to 849.4 mg C m⁻² h⁻¹, with an average of 281.4 mg C m⁻² h⁻¹. This was significantly higher than the 45.3 mg C $m^{-2} h^{-1}$ average measured from swine manure, ranging from 4.1 to 66.0 mg C m⁻² h⁻¹ (Fig. 2B). Li (2016) derived similar results. Previous studies demonstrated that a lower C:N ratio reduced CH₄ emissions by inhibiting the activity of methane-producing bacteria (Xu and Li, 2012). Observed differences in the C:N ratio between swine manure and dairy cattle manure in this study could provide a possible explanation for the abovementioned phenomenon (Table S2). Moreover, we found that the majority of CH₄ emission flux from dairy cattle manure occurred during the middle storage period. With an extension in the duration of manure storage, manure storage processes were able to consume more oxygen, which subsequently formed an anaerobic environment. This suitable environment in combination with the availability of rich organic matter were beneficial for CH₄ generation, resulting in higher CH₄ emission (Wang et al., 2018).

3.2.3. CO₂

 CO_2 emission flux from swine and dairy cattle manure exhibited similar emission patterns. However, the average CO_2 emission flux from swine manure (13.4 g C m⁻² h⁻¹, ranging from 4.2 to 41.6 g C m⁻² h⁻¹) was significantly higher than that from dairy cattle (7.9 g C m⁻² h⁻¹, ranging from 3.2 to 34.7 g C m⁻² h⁻¹) (Fig. 2C), which agreed with results obtained by Dai et al. (2015) and Li et al. (2016). Li et al. (2016) reported that a negative correlation between CO_2 emissions and the C:N ratio was the main factor that influenced the abovementioned differences, indicating that the lower the C:N ratio is, the higher the CO_2 emission rate will be. In our study, the C:N ratio of swine manure was lower than that of dairy cattle manure (Table S2), thus resulting in higher CO_2 emissions from swine manure.

3.2.4. NH3

NH₃ released from swine and dairy cattle manure varied with an average of 0.4 to 270.0 mg N m⁻² h⁻¹ and 0.7 to 90.2 mg N m⁻² h⁻¹, respectively (Fig. 2D). Swine manure had a higher NH₃ release rate than dairy cattle manure, which is consistent with results from previous studies (Liu et al., 2013; Dai et al., 2015; Li et al., 2016; Wang et al., 2017, 2018). The high C:N ratio and the low NH₄-N content in dairy cattle manure compared to swine manure observed in this study (Table S2) could be a potential reason for the low NH₃ emissions observed from dairy cattle manure (Liu et al., 2013; Li et al., 2016; Awasthi et al., 2018; Mao et al., 2019). In addition, we also found that the majority of NH₃ released occurred during the early and middle stages of storage for both swine and dairy cattle manure, which could be explained by the higher NH₄-N content, the relatively better aerobic conditions, and the rising temperatures observed at the start of solid manure fermentation in comparison to the latter stage of solid manure storage (Chaoui et al., 2009; Saha et al., 2011; Li et al., 2016; Yuan et al., 2014; Dai et al., 2015).

3.3. Total GHGs and NH₃ emissions in swine and dairy cattle manure

As illustrated in Fig. 3, results showed that total GHGs emissions from dairy cattle manure (7222.1 g CO₂-eq.) during the storage phase were similar to that of swine manure (6793.7 g CO₂-eq.), while total NH₃ emissions from swine manure (880.9 g) were significantly higher, namely, greater by a factor of 2.4 compared to dairy cattle manure (381.5 g). CH₄ emissions from dairy cattle manure were significantly higher than that from swine manure, while N₂O and CO₂ emissions from dairy cattle manure were significantly lower that swine manure. The CH₄ increment in dairy cattle manure compared to swine manure offset the decrement of both N₂O and CO₂ emissions, which explains the balance of GHGs emissions between dairy cattle manure and swine manure. In addition, we analyzed the contribution of N₂O, CH₄, and CO₂ emissions to total GHGs emissions. N₂O, CH₄, and CO₂ emissions from dairy cattle manure accounted for 1.1%, 49.3%, and 49.6% of the total GHGs emissions (Fig. 4A), respectively, while corresponding



Fig. 3. Total GHGs and NH₃ emissions from dairy cattle and swine manure. Note: Different letters indicate significant differences at a level of 0.05.

emission values from swine manure were 2.3%, 8.4%, and 89.3%, respectively (Fig. 4B), indicating that both CH_4 emissions and CO_2 emissions dominated total GHGs emissions from swine and dairy cattle manure.

4. Conclusions and implications

Under the various challenges to meet increasing population and crop demands for livestock products under conditions of climate change and issues related to environmental pollution, it is crucial to be able to quantify and compare GHGs and NH3 emissions from different livestock categories during the manure storage phase to develop effective mitigation strategies. Therefore, this study comprehensively estimated GHGs and NH₃ emissions from conventional solid dairy cattle and swine manure storage based on field measurements. Results showed that GHGs emissions from swine manure were similar to that from dairy cattle manure, but NH₃ emissions of the former were higher overall. This could be attributed to the different physicochemical characteristics of manure and may also be associated with the microbiological, chemical, and physical processes that produce gas during the storage phase. As it pertains to GHGs emissions, CO₂ emissions from swine manure accounted for 89.3% of total GHGs emissions, which was the largest overall gas source. By contrast, CH₄ and CO₂ were the two largest gas sources from dairy cattle manure, representing 49.3% and 49.6%, respectively. Along with the increasing predicted demand for milk and meat in the future, a correspondingly large amount of manure will be excreted by dairy cattle and swine, which will result in much higher GHGs and NH₃ emissions unless effective mitigation strategies are taken.

Our results not only provide insight into the simultaneous evaluation of GHGs and NH₃ emissions from the manure of swine and dairy cattle, but they also offer useful information for the simultaneous mitigation of GHGs and NH₃ emissions from two inherently different livestock species when strategies are to be adopted in making relevant mitigation policies. Controlling the increase in the population of dairy cattle and swine is regarded as the most direct and effective strategy to mitigate GHGs and NH₃ emissions; however, this strategy is somewhat unrealistic and challenging (Zhuang et al., 2019a).

The question of how to simultaneously mitigate GHGs and NH₃ emissions from swine and dairy cattle manure has drawn increasing attention in China in recent years (Wang et al., 2017, 2018; Bai et al., 2017). CO_2 emissions have become the largest gas source from swine manure, while CH₄ and CO₂ emissions are the two largest gas sources that derive from dairy cattle manure. However, to avoid trade-offs between different gases emissions, we should comprehensively evaluate the effects of mitigation strategies on overall gas emissions rather than a single gas (Wang et al., 2017). In this context, some potential mitigation strategies have been proposed, such as applying selective additives (e.g., biochar, alum, and zeolite) (Lim et al., 2017; Cao et al., 2019; Mao et al., 2019; Zhang et al., 2019) or focusing on aeration (Chowdhury et al., 2014; Zeng et al., 2017), pile turning (Li et al., 2016; Zeng et al., 2018), or coverage strategies (Wang et al., 2017, 2018) to mitigate solid manure gases emissions. For instance, Zhang et al. (2019) reported that swine manure composting applying biochar reduced total N₂O and NH₃ emissions by 39.1% compared to conventional solid swine manure practices, while Lim et al. (2017) found that composting with phosphogypsum and zeolite significantly decreased NH₃ and CH₄ emissions by



Fig. 4. Contributions of a single gas to total GHGs emissions in dairy cattle manure (A) and swine manure (B).

30% and 97.0%, respectively. In addition, manure coverage practices may be another useful mitigation solution to reduce GHGs and NH₃ emissions. Previous studies have shown that coverage strategies can reduce NH₃ emissions from swine manure by 40–98% and CH₄ emissions by 38-86%, respectively, which depend on the coverage management practices used, such as the cover material and the cover method (Sommer et al., 2000; Portejoie et al., 2003; Zhu et al., 2015). However, selecting inappropriate cover material and cover methods could also increase GHGs and NH₃ emissions from manure (Amon et al., 2006). Based on the above analysis, we found that the mitigation effect on GHGs and NH₃ emissions varied among these different mitigation strategies. Thus, although the combination and optimization of the various mitigation strategies may be effective, they warrant further research. Information such as that provided in this study is crucial for devising relevant mitigation policies for livestock manure on a national scale into the future.

Although we systemically analyzed and compared GHGs and NH₃ emissions of solid stored manure from dairy cattle and swine over the short-term, we did not investigate long-term gas emissions from manure in this study, which could to a certain extent lead to uncertainty in the effectiveness of the mitigation strategies discussed. Therefore, continuous long-term field observations of GHGs and NH₃ emissions from the manure of various livestock animals should be carried out in the future, which will allow us to better understand seasonal and interannual variation in gases emissions and their associated influencing factors, while also helping to develop much more effective mitigation strategies.

CRediT authorship contribution statement

Minghao Zhuang: Conceptualization, Methodology, Writing - original draft, Writing - review & editing. Nan Shan: Formal analysis, Investigation, Resources. Yingchun Wang: Methodology, Data curation, Resources. Dario Caro: Formal analysis, Writing - review & editing. Rachael Marie Fleming: Writing - review & editing. Ligang Wang: Conceptualization, Methodology, Writing - review & editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi. org/10.1016/j.scitotenv.2020.137693.

References

- Amon, B., Kryvoruchko, V., Amon, T., Zechmeister-Boltensten, S., 2006. Methane, nitrous oxide and ammonia emissions during storage and after application of dairy cattle slurry and influence of slurry treatment. Agric. Ecosyst. Environ. 112 (2), 153–162.
- Awasthi, M.K., Wang, Q., Awasthi, S.K., Wang, M., Chen, H., Ren, X., Zhao, J., Zhang, Z., 2018. Influence of medical stone amendment on gaseous emissions, microbial biomass and abundance of ammonia oxidizing bacteria genes during biosolids composting. Bioresour. Technol. 247, 970–979.
- Bai, Z.H., Ma, L., Jin, S.Q., Ma, W.Q., Velthof, G.L., Oenema, O., Liu, L., Chadwick, D., Zhang, F.S., 2016. Nitrogen, phosphorus, and potassium flows through the manure management chain in China. Environ. Sci. Technol. 50 (24), 13409–13418.

- Bai, Z.H., Li, X.X., Lu, J., Wang, X., Velthof, G.L., Chadwick, D., Luo, J.F., Ledgard, S., Wu, Z.G., Jin, S.Q., Oenema, O., Ma, L., Hu, C.S., 2017. Livestock housing and manure storage need to be improved in China. Environ. Sci. Technol. 51 (15), 8212–8214.
- Bergström, A.N.N.K., Jansson, M., 2006. Atmospheric nitrogendeposition has caused nitrogen enrichment and eutrophication oflakes in the northern hemisphere. Glob. Change Biol. 12 (4), 635–643.
- Borhan, M., Gautam, D., Engel, C., Anderson, V., Rahman, S., 2013. Effects of pen bedding and feeding high crude protein diets on manure composition and greenhouse gas emissions from a feedlot pen surface. J. Air Waste Manage. Assoc. 63 (12), 1457–1468.
- Bouwman, A.F., Boumans, L.J.M., Batjes, N.H., 2002. Estimation of global NH3 volatilization loss from synthetic fertilizers and animal manure applied to arable lands and grasslands. Glob. Biogeochem. Cycles 16 (2), 1024.
- Cai, Y.J., Chang, S.X., Cheng, Y., 2017. Greenhouse gas emissions from excreta patches of grazing animals and their mitigation strategies. Earth-Sci. Rev. 171, 44–57.
- Cao, Y.B., Wang, X., Bai, Z.H., Chadwick, D., Tom Misselbrook, T., Sommer, S.G., Qin, W., Lin Ma, L., 2019. Mitigation of ammonia, nitrous oxide and methane emissions during solid waste composting with different additives: a meta-analysis. J. Clean. Prod. 235, 626–635.
- Chadwick, D., Sommer, S., Thorman, R., Fangueiro, D., Cardenas, L., Amon, B., Misselbrook, T., 2011. Manure management: Implications for greenhouse gas emissions. Anim. Feed Sci. Tech. 166/167, 514–531.
- Chaoui, H., Montes, F., Richard, C.A.R.T.L., 2009. Volatile ammonia fraction and flux from thin layers of buffered ammonium solution and dairy cattle manure. T. ASABE 52 (5), 1695–1706.
- Chen, X.P., Cui, Z.L., Fan, M.S., Vitousek, P.M., Zhao, M., Ma, W.Q., Wang, Z.L., Zhang, W.J., Yan, X.Y., Yang, J.C., Deng, X.P., Gao, Q., Zhang, Q., Guo, S.W., Ren, J., Li, S.Q., Ye, Y.L., Wang, Z.H., Huang, J.L., Tang, Q.Y., Sun, Y.X., Peng, X.L., Zhang, Y.M., He, M.R., Zhu, Y.J., Xue, J.Q., Wang, G.L., Wu, L., An, N., Wu, L.Q., Ma, L., Zhang, W.F., Zhang, F.S., 2014. Producing more grain with lower environmental costs. Nature 514, 486–489.
- China Agricultural Statistical Yearbook (CASY): 2000–2015. China Agriculture Press, Beijing.
- Chowdhury, M.A., de Neergaard, A., Jensen, L.S., 2014. Potential of aeration flow rate and biochar addition to reduce greenhouse gas and ammonia emissions during manure composting. Chemosphere 97, 16–25.
- Dai, X.R., Saha, C.K., Ni, J.Q., Heber, A.J., Blanes-Vidal, V., Dunn, J.L., 2015. Characteristics of pollutant gas releases from swine, dairy, beef, and layer manure, and municipal wastewater. Water Res. 76, 110–119.
- Evans, L., Vanderzaag, A.C., Sokolov, V., Baldé, H., MacDonald, D., Wagner-Riddle, C., Gordon, R., 2018. Ammonia emissions from the field application of liquid dairy manure after anaerobic digestion or mechanical separation in Ontario, Canada. Agric. For. Meteorol. 258, 89–95.
- Fang, M., Chan, C.K., Yao, X.H., 2009. Managing air quality in a rapidly developing nation: China. Atmos. Environ. 43 (1), 79–86.
- Gericke, D., Pacholski, A., Kage, H., 2011. Measurement of ammonia emissions in multiplot field experiments. Biosyst. Eng. 108, 164–173.
- Gu, B.J., Ju, X.T., Chang, J., Ge, Y., Vitousek, P.M., 2015. Integrated reactive nitrogen budgets and future trends in China. Proc. Natl. Acad. Sci. U. S. A. 112, 8792–8797.
- Herrero, M., Henderson, B., Havlik, P., Thornton, P.K., Conant, R.T., Smith, P., Wirsenius, S., Hristov, A.N., Gerber, P., Gill, M., Butterbach-Bahl, K., Valin, H., Garnett, T., Stehfest, E., 2016. Greenhouse gas mitigation potentials in the livestock sector. Nat. Clim. Chang. https://doi.org/10.1038/nclimate2925.
- Holly, M.A., Larson, R.A., Powell, J.M., Ruark, M.D., Aguirre-Villegas, H., 2017. Greenhouse gas and ammonia emissions from digested and separated dairy manure during storage and after land application. Agric. Ecosyst. Environ. 239, 410–419.
- Hou, Y., Velthof, G.L., Oenema, O., 2015. Mitigation of ammonia, nitrous oxide and methane emissions from manure management chains: ameta-analysis and integrated assessment. Glob. Chang. Biol. 21, 1293–1312.
- Jeong, S.T., Kim, G.W., Hwang, H.Y., Kim, P.J., Kim, S.Y., 2018. Beneficial effect of compost utilization on reducing greenhouse gas emissions in a rice cultivation system through the overall management chain. Sci. Total Environ. 613/614, 115–122.
- Jiang, J.S., Kang, K., Wang, C.J., Sun, X.J., Dang, S., Wang, N., Wang, Y., Zhang, C.Y., Guangxuan Yan, G.X., Li, Y.B., 2018. Evaluation of total greenhouse gas emissions during sewage sludge composting by the different dicyandiamide added forms: mixing, surface broadcasting, and their combination. Waste Manag. 81, 94–103.
- Kang, Y.N., Liu, M.X., Song, Y., Huang, X., Yao, H., Cai, X.H., Zhang, H., Kang, L., Liu, X., Yan, X.Y., He, H., Zhang, Q.W., Shao, M., Zhu, T.J., 2016. High-resolution ammonia emissions inventories in China from 1980–2012. Atmos. Chem. Phys. 15 (19), 26959–26995.
- Li, LL, 2016. Study on the Characteristic and Reduction for Greenhouse Gases and Ammonia Emissions During the Storage of Manure and Wastewater. Master thesis. Chinese Academy of Agricultural sciences. Beijing. China (in Chinese with English abstract).
- Liang, F.C., Liu, L.M., Shuang, W.Y., Zou, L.L., 2015. Zoning control and regulation of agricultural production landscapes based on natural suitability—a case study of Jinjing Town of Changsha County in Hunan Province. Soil 47, 142–147 (Chinese with English abstract).
- Lim, S.S., Park, H.J., Hao, X., Lee, S.I., Jeon, B.J., Kwak, J.H., Choi, W.J., 2017. Nitrogen, carbon, and dry matter losses during composting of livestock manure with two bulking agents as affected by co-amendments of phosphogypsum and zeolite. Ecol. Eng. 102, 280–290.
- Liu, M., Giard, D., Barrington, S., 2013. Ammonium dissociation for swine and dairy cattle manures. J. Environ. Prot. 4, 6–15.
- Lovanh, N., Loughrin, J., Cook, K., Silva, P., Oh, B., 2014. Effect of windrow management on ammonia and nitrous oxide emissions from swine manure composting. Int. Sci. Index 8, 369–373.
- Maeda, K., Hanajima, D., Toyoda, S., Yoshida, N., Morioka, R., Osada, T., 2011. Microbiology of nitrogen cycle in animal manure compost. Microb. Biotechnol. 4, 700–709.

- Maldaner, L., Wagner-Riddle, C., Vanderzaag, A.C., Gordon, R., Duke, C., 2018. Methane emissions from storage of digestate at a dairy manure biogas facility. Agric. For. Meteorol. 258, 96–107.
- Mao, H., Zhang, H.Y., Fu, Q., Zhong, M.Z., Li, R.H., Zhai, B.N., Wang, Z.H., Zhou, L.N., 2019. Effects of four additives in pig manure composting on greenhouse gas emission reduction and bacterial community change. Bioresour. Technol. 292, 121896.
- Masse, D.I., Masse, L., Claveau, S., Benchaar, C., Thomas, O., 2008. Methane emissions from manure storages. Am. Soc. Agric. Biol. Eng. 51, 1775–1781.
- Matson, P., Lohse, K.A., Hall, S.J., 2002. The globalization of nitrogen deposition: consequences for terrestrial ecosystems. Ambio 31, 113–119.
- Mcginn, S.M., Flesch, T.K., 2018. Ammonia and greenhouse gas emissions at beef cattle feedlots in Alberta Canada. Agric. For. Meteorol. 258, 43–49.
- Ministry of Agriculture and Rural Affairs of the People's Republic of China (MOA), 2017. Accelerate the utilization of livestock and poultry waste resources and promote the green development of animal husbandry. http://www.moa.gov.cn/xw/zwdt/ 201706/t20170614_5680105.htm.
- Monteny, G.J., Bannink, A., Chadwick, D., 2006. Greenhouse gas abatement strategies for animal husbandry. Agric. Ecosyst. Environ. 112, 163–170.
- Myhre, G., Shindell, D., Breon, F.M., Collins, W., Fuglestvedt, J., Huang, J., Koch, D., Lamarque, J.F., Lee, D., Mendoza, B., Nakajima, T., Robock, A., Stephens, G., Takemura, T., Zhang, H., 2013. Anthropogenic and natural radiative forcing. In: Stocker, T.F., Qin, D., Plattner, G.K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P.M. (Eds.), Climate Change 2013: the Physical Science Basis. Contribution of Working Group I to the Fifth AssessmentReport of the Intergovernmental Panel on Climate Change. Cambridge University Press, New York.
- Nelson, M.I., Marchant, T.R., Wake, G.C., Balakrishnan, E., Chen, X.D., 2007. Self-heating in compost piles due to biological effects. Chem. Eng. Sci. 62 (17), 4612–4619.
- Pacholski, A., Cai, G., Nieder, R., Richter, J., Fan, X., Zhu, Z., Roelcke, M., 2006. Calibration of a simple method for determining ammonia volatilization in the field comparative measurements in Henan Province, China. Nutr. Cycl. Agroecosyst. 74, 259–273.
- Philippe, F.X., Laitat, M., Wavreille, J., Nicks, B., Cabaraux, J.F., 2013. 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. 155 (2), 397–406.
- Pietzner, B., Rücknagel, J., Koblenz, B., Bednorz, D., Tauchnitz, N., Bischoff, J., Köbke, S., Meyrer, K.H.E., Meißner, r., Christen, O., 2017. Impact of slurry strip-till and surface slurry incorporation on NH₃ and N₂O emissions on different plot trials in Central Germany. Soil Tillage Res. 169, 54–64.
- Portejoie, S., Martinez, J., Guiziou, F., Coste, C.M., 2003. Effect of covering pig slurry stores on the ammonia emission processes. Bioresour. Technol. 87 (3), 199–207.
- Roelcke, M., Li, S.X., Tian, X.H., Gao, Y.J., Richter, J., 2002. In situ comparisons of ammonia volatilization from N fertilizers in Chinese loess soils. Nutr. Cycl. Agroecosyst. 62, 73–88.
- Saha, C.K., Zhang, G., Ni, J.Q., Ye, Z., 2011. Similarity criteria for estimating gas emission from scale models. Biosyst. Eng. 108 (3), 227–236.
- Seinfeld, J.H., Pandis, S.N., 2006. Atmospheric Chemistry and Physics: From Air Pollution to Climate Change. 2nd ed. John Wiley, Hoboken, N. J 1203 pp.
- Sommer, S., Petersen, S., Søgaard, H., 2000. Greenhouse gas emission from stored livestock slurry. J. Environ. Qual. 29 (3), 744–751.
- Sommer, S.G., Petersen, S.O., Moller, H.B., 2004. Algorithms for calculating methane and nitrous oxide emissions from manure management. Nutr. Cycl. Agroecosyst. 69, 143–154.

- Wang, K., Zheng, X.H., Pihlatie, M., Vesala, T., Liu, C.Y., Haapanala, S., Mammarella, I., Rannik, Ü., Liu, H.Z., 2013. Comparison between static chamber and tunable diode laser-based eddy covariance techniques for measuring nitrous oxide fluxes from a cotton field. Agric. For. Meteorol. 171–172, 9–19.
- Wang, J., Zhu, B., Zhang, J.B., Müller, C., Cai, Z.C., 2015. Mechanisms of soil N dynamics following long-term application of organic fertilizers to subtropical rain-fed purple soil in China. Soil Biol. Biochem. 91, 222–231.
- Wang, Y., Dong, H.M., Zhu, Z.P., Gerber, P.J., Xin, H.W., Smith, P., Opio, C., Steinfeld, H., Chadwick, D.R., 2017. Mitigating greenhouse gas and ammonia emissions from swine manure management: a system analysis. Environ. Sci. Technol. 51 (8), 4503–4511.
- Wang, Y., Li, X.R., Yang, J.F., Tian, Z., Sun, Q.P., Xue, W.T., Dong, H.M., 2018. Mitigating greenhouse gas and ammonia emissions from beef cattle feedlot production - a system meta-analysis. Environ. Sci. Technol. 52, 11232–11242.
- Xia, L.L., Lam, S.K., Yan, X.Y., Chen, D.L., 2017. How does recycling of livestock manure in agroecosystems affect crop productivity, reactive nitrogen losses and soil carbon balance? Environ. Sci. Technol. 51 (13), 7450.
- Xu, F.Q., Li, Y., 2012. Solid-state co-digestion of expired dog food and corn stover for methane production. Bioresour. Technol. 118 (4), 219–226.
- Yuan, Y.L., Wang, L.G., Li, H., Ding, G.Q., Han, S.H., Wei, J.H., 2014. Nitrogenous gas emissions from solid swine manure under natural composting conditions. J. Agro-Environ. Sci. 33, 1422–1428 (in Chinese with English Abstract).
- Zeng, J.F., Yin, H.J., Shen, X.L., Liu, N., Ge, J.Y., Han, L.J., Huang, G.Q., 2017. Effect of aeration interval on oxygen consumption and GHG emission during pig manure composting. Bioresour. Technol. 250, 214–220.
- Zeng, J.F., Shen, X.L., Sun, X.X., Liu, N., Han, L.J., Huang, G.Q., 2018. Spatial and temporal distribution of pore gas concentrations during mainstream large-scale trough composting in China. Waste Manag. 75, 297–304.
- Zhang, Z.H., 2018. Experimental Study on Preparation of Carbon Base Fertilizer by Mixing Animal Manure and Straw. Huazhong Agricultural University (Master's Degree Dissertation).
- Zhang, D.F., Luo, W.H., Yuan, J., Li, G.X., Luo, Y., 2017. Effects of woody peat and superphosphate on compost maturity and gaseous emissions during pig manure composting. Waste Manag. 68, 56–63.
- Zhang, J., Zhuang, M.H., Shan, N., Zhao, Q., Li, H., Ligang Wang, L.G., 2019. Substituting organic manure for compound fertilizer increases yield and decreases NH₃ and N₂O emissions in an intensive vegetable production systems. Sci. Total Environ. 670, 1184–1189.
- Zhou, M., Zhu, B., Brüggemann, N., Dannenmann, M., Wang, Y., Butterbach-Bahl, K., 2016. Sustaining crop productivity while reducing environmental nitrogen losses in the subtropical wheat-maize cropping systems: a comprehensive case study of nitrogen cycling and balance. Agric. Ecosyst. Environ. 231, 1–14.
- Zhu, H.S., Zuo, F.Y., Dong, H.M., Luan, D.M., 2015. Effects of covering materials and sawdust covering depths on ammonia and greenhouse gases emissions from cattle manure during storage. T. CSAE 31 (6), 223–229 (Chinese with English abstract).
- Zhuang, M.H., Lu, X., Caro, D., Gao, J., Zhang, J., Cullen, B., Li, Q.W., 2019a. Emissions of non-CO₂ greenhouse gases from livestock in China during 2000–2015: magnitude, trends and spatiotemporal patterns. J. Environ. Manag. 242, 40–45.
- Zhuang, M.H., Lam, S.K., Zhang, J., Li, H., Shan, N., Yuan, Y.L., Wang, L.G., 2019b. Effect of full substituting compound fertilizer with different organic manure on reactive nitrogen losses and crop productivity in intensive vegetable production system of China. J. Environ. Manag. 243, 381–384.