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Concentrations and Emissions of Particulate Matter from Intensive Pig Production at a Large Farm in North China

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ABSTRACT

Particulate matter (PM) pollution from pig farms is of increasing environmental concern due to the rapid development of intensively managed pig farms worldwide. However, information on this topic is scarce in China to date. Therefore a two-year study to monitor the dynamics of total suspended particles (TSP) and particulate matter with diameter less than 10 µm (PM_{10}) and their soluble ions was conducted at an industrial pig farm near Beijing. In total, two fattening pig houses were sampled during three fattening periods from 2010 to 2011. Daily mean indoor TSP and PM_{10} concentrations were 1.85 and 0.63 mg m^{-3} on average. The emissions of PM per livestock unit (LU unit =500 kg), and per unit area (m^2) from the pig house averaged 467.5 mg h^{-1} LU^{-1} and 38.6 mg h^{-1} m^{-2} for TSP, while those for PM_{10} averaged 256.7 mg h^{-1} LU^{-1} and 16.8 mg h^{-1} m^{-2}, respectively. Particulate NH_4^+ in both TSP and PM_{10} showed significant positive correlations with particulate NO_3^- and SO_4^{2-} inside and outside the pig houses. Daily mean indoor concentrations of secondary inorganic aerosol (sum of NH_4^+, NO_3^- and SO_4^{2-}) ranged from 10.8 to 368.8 µg m^{-3} (average 116 µg m^{-3}) in TSP and from 5.1 to 220.1 µg m^{-3} (average 58.6 µg m^{-3}) in PM_{10}. Contributions of secondary inorganic PM to TSP and PM_{10} emissions varied from 0.3 to 50.8% and from 1.0 to 48.7%, respectively, with averages of 12.0 and 13.9%. Our results suggest elevated PM and secondary inorganic PM concentrations and substantial emissions of PM from the fattening pig houses in China.

Keywords: Particulate matter; Secondary inorganic aerosol; Air quality; Livestock emissions; Pig farm particulates.

INTRODUCTION

Particulate matter (PM) from pig houses is a component of aerial pollution and has received worldwide attention in recent years due to health and environmental concerns. A close relationship between PM air pollution and respiratory and cardiovascular diseases and mortality, has been identified (Pope *et al.*, 2002). Particulate matter has detrimental effects on both indoor air quality and on the external environment via exhaust outlets. For example, PM from pig houses contains micro-organisms and their components as well as other bioactive components (bio-aerosols) which, through atmospheric transmission, increase the prevalence of respiratory infections (e.g., chronic bronchitis and coughs) to people living in neighboring communities (Bakutis *et al.*, 2004; Cai *et al.*, 2006). Emitted PM can also impact the environment causing vegetation stress and ecosystem change (Grantz *et al.*, 2003). Small PM, including PM_{10} and $PM_{2.5}$ (particulate matter with an aerodynamic diameter less than 10 and 2.5 µm), is implicated in climate change issues, such as cloud formation and radiative forcing and contributes to reduction in atmospheric visibility reduction (IPCC, 2005); furthermore it also affects human and animal health.

Several studies have focused on the identification and quantification of PM sources in order to evaluate the impact of pig fattening facilities on human and animal health and the environment. Particulate matter in pig houses originates primarily from feed stuffs, manures, bedding, and pig skin and hair (Donham *et al.*, 1986; Cambra-López *et al.*, 2011). In addition, secondary particulate matter (e.g., $(NH_4)_2SO_4$, NH_4NO_3 and NH_4Cl) is formed in livestock buildings as a result of chemical reactions between NH_3 and acid gases (e.g., H_2SO_4, HNO_3 and HCl) (Roumeliotis and Van Heyst, 2008; Cambra-López *et al.*, 2010). Secondary PM is the main component of livestock PM (Robarge *et al.*, 2002) and depends on the type of animal and the way they are housed and managed (type of bedding, feed, etc.). Roumeliotis and Van Heyst (2008) indicated that the contribution of secondary PM accounted for more than 50% of the total

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$PM_{2.5}$ in a layer hen house. On the other hand, PM concentrations (0.73 mg m^{-3} for PM_{10}) and emissions (108–250 mg h^{-1} LU^{-1} for PM_{10}) within pig houses have also been investigated by some researchers (e.g., Haeussermann *et al.*, 2008; Costa *et al.*, 2009). In spite of the intensive research that has been conducted, the contribution of secondary PM to total PM emissions from livestock houses is still unclear (Cambra-López *et al.*, 2010).

Situation in China/on the North China Plain

Intensive large-scale pig husbandry in China has grown rapidly due to the increasing demand for pork. Chinese annual production of pork increased from 31.6 Tg in 1996 to 50.5 Tg in 2011 (CNBS, 2011), and accounted for almost 50% of world production (FAO, 2008). Meeting this demand is closely connected with a sharp increase in intensive pig production. This, in turn, has resulted in the growth of aerial pollutant emissions (e.g., NH_3 and PM) emissions from pig farms in China. Gao *et al.* (2013) calculated a national mean NH_3 emission intensity of 26.6 g NH_3-N kg^{-1} of pork for China in 2009. The North China Plain (NCP), one of the most intensive livestock production regions in the country, contributes about 30% of national animal products (Zhang *et al.*, 2010). Previous studies by some of the present authors on NH_3 emission and deposition confirm that the NCP is experiencing high levels of NH_3 pollution caused by livestock production and application of mineral nitrogen (N) fertilizers (Shen *et al.*, 2011; Liu *et al.*, 2013; Luo *et al.*, 2013). According to Zhang *et al.* (2010), on the NCP pig husbandry is the biggest livestock NH_3 contributor, accounting for 60% of total livestock emissions. We therefore suspect that there is a large amount of secondary inorganic aerosol being formed in pig buildings due to reactions between NH_3 and acid gases in ambient air. Huaitalla *et al.* (2013) reported similar high concentrations of PM at the same pig farm as we investigated. However, little information is available about PM and secondary inorganic PM emissions from pig houses in China. The main objectives of the present study were therefore to quantify the concentration and emission levels of TSP (total suspended particles) and PM_{10} in two naturally ventilated fattening pig houses at an industrial pig farm on the northern edge of the NCP and to identify the relationships between particulate NH_4^+, NO_3^-, SO_4^{2-} and Cl^- and TSP and PM_{10} levels. After determining the main ionic components of secondary inorganic PM, their emissions and contributions to total PM emissions from the pig houses were estimated.

MATERIALS AND METHODS

Site Description

The present study was conducted at a large commercial pig farm located in the peri-urban Shunyi District of Beijing (39°55′N, 116°25′E). The centralized pig raising plant was set up in 1995 and had an annual stock of 12,000 breeding boars and 20,000 fattening pigs (porkers).The plant consisted of 56 east-west oriented pig barns with an average capacity of 200–300 pigs (including weaning, fattening, gestation, and farrowing barns). It was characterized by the manure management system referred to as "gan qing fen" in Chinese or "cleaning the manure in dry conditions" in English (Schuchardt *et al*, 2011). Two identical fattening pig barns (houses I and II), with the same structure, feeding and manure removal systems, were used for PM measurements during a total of three fattening periods. Each barn, 8 m × 50 m, was divided into 16 pens (each with 13–18 fattening pigs) with a fully solid concrete floor (see Fig. S1 in the Supporting Information, SI). The ventilation for each barn was primarily via passive air exchange (natural ventilation) by means of 16 (3 × 1.5 m) controlled window openings on the north and south sides of each barn, respectively. In order to maintain a suitable indoor temperature, 50, 100 and 25% of the windows were generally opened in late spring, summer and early autumn, respectively, and all of the windows were closed in winter. The arrival live weight of the pigs was of the order of 20 kg and the pigs were normally fattened to 100 kg. The monthly animal mortality rate was recorded and taken into account for total weight estimations. The monthly animal mortality rate was documented and taken into account for total weight estimations. The pigs were fed twice daily by hand (8:00–8:30; 16:30–17:00, before each manure removal), with dry feed and drinking water was provided with a nipple attached to the fence of the pen. Pig diets consisted of corn, wheat, soybean meal, fish powder, compound premix and lysine. The workers in the fattening houses used a shovel or scraper to manually collect the pig manure from the concrete floors twice a day and then flushed the pig pens with 6–9 L of water per pig per day (up to 25 L in small units) afterwards ("gan qing fen" system). Remaining urine and some feces flowed freely to a gutter outside the house due to the slightly sloped pen floor. A more detailed description of the farm and the experimental pig houses has been given in a previous paper (Xu *et al.*, 2014).

Measurement Strategy

Particulate matter (i.e., TSP and PM_{10}) measurements were performed 2nd June 2010 and 11th August 2011 within three fattening period. Measurements of TSP were carried out in house I from 2nd June–1st September 2010 (first fattening period) and from 15th September 2010 to 7th January 2011 (second fattening period), while PM_{10} measurements were performed in house II from 1st April to 11th August 2011 during the third fattening period. During each sampling period, daily mean indoor concentrations of PM (24 h sampling (8:00 AM until 8:00 AM of next day) per sample) were measured randomly, but normally covering each full and/or incomplete month (1/3 at the beginning, 1/3 at the middle and 1/3 at the end). The typical wind direction was southwesterly outside the selected pig farm as shown in detail in Fig. S2. Thereby the PM concentrations outside the houses, as background readings, were also measured with the same sampling strategy as indoor PM. Because of the limited number of PM samplers, indoor and outdoor PM measurements were performed sequentially. For the two fattening houses, both indoor and outdoor air temperature and relative humidity were measured continuously throughout the experiment.

Indoor concentrations of PM were measured in the central position of the pen with a protective iron cage at 1.6 m height, which was representative of the ventilation exhaust height. It should be noted that this height is also in the farmer's breathing zone in the two selected pig barns. The instrument for outdoor PM measurements was positioned at 2 m height, 6 m away from the southern wall of the barns. Indoor temperature and relative humidity were measured in the center of the barn at 2 m height, while outdoor temperature and relative humidity were monitored at 5 m from the south walls of the barns and 2 m above the ground. The layout of sampling locations is shown in Fig. S1.

Measuring Equipment and Analytical Procedures

Indoor and outdoor (background) TSP and PM_{10} samples were collected using a medium-volume air sampler (TH-150A, Tianhong Co., Tianjin, China) at a flow rate of 100 L min^{-1}. To sample TSP and PM_{10}, the inlet system of the instrument was configured and the relative TSP and PM_{10} collectors equipped with Teflon filters (47 mm, Whatman) were placed on the instrument for 24 h sampling, respectively. After sampling, the filters were sealed in aluminum foil and frozen inside the on-site freezer (–17°C) until delivery in iceboxes to the analytical laboratory by routine monthly site-maintenance visit. In the laboratory the filters were equilibrated for 24 h in a room with a controlled relative humidity (50%) and temperature (22°C) to measure the pre- and post-sampling weights both before and after sampling. The filters were weighted on an electronic balance (Sartorius, precision: 10 µg), and all the filters collected were weighted in less than a month. The concentrations of PM were calculated by the dust mass collected on the filter by sampling air flows. After weighing, the indoor and outdoor particle-loaded Teflon filters were ultrasonically extracted with 100 and 20 mL high-purity water (18.2 MΩ) for 30 min, respectively, and the extract filtered using a syringe filter (0.45 µm, Tengda Inc., Tianjin, China). The extraction solutions were stored in a refrigerator at 4°C until analysis within one month of extraction. Concentrations of particulate soluble inorganic ions (NO_3^-, SO_4^{2-}, Cl^-, NH_4^+, K^+, Ca^{2+}, Mg^{2+}, and Na^+) from the exposed filters were determined by ion chromatography (DX-120, Dionex Ltd, USA). Three field blank samples were taken each month at each site and analyzed following the same procedure and methods.

Air temperature and relative humidity inside and outside the pig barn were measured at 30 min intervals using HOBO sensors (Onset Computer Corporation, MA) which were both housed in a PVC shelter to shield them from direct sunlight. The measurement range of the sensor was –20 to 70°C with an accuracy of ± 0.35°C and its maximum measurement time was 60 days. The recorded data were downloaded and treated as daily-based averages using Microsoft Excel (2010) in a desktop computer at China Agricultural University, Beijing. Daily mean wind velocity and direction data during each sampling period and the entire experimental period were obtained from a local meteorological station nearby (about 9 km from the pig farm).

All instruments were calibrated by the manufacturer prior to start of the measurements. The results on climate conditions and pig growth characteristics of the two selected pig houses are presented in Table S1 and also discussed in Text S1 (see the Supporting Information (SI)).

Estimates of PM Emissions

The emission rate (ER) is defined as the product of the ventilation rate and the pollutant concentration, expressed as:

$$ER = (C_{in} - C_{out}) \times V_i \quad (1)$$

where ER = pollutant emission rate at time i, V_i = the ventilation rate at time i, C_i. = pollutant concentration at time i (indoor concentration minus outdoor concentration), and i = time in minutes of the monitored parameter.

In the present study the ventilation rate was calculated on a 24-hour basis (8:00 AM until 8:00 AM of the next day) based on the heat balance method (CIGR, 2002) which depends on indoor and outdoor temperature, sensible heat and transmission heat loss. The detailed calculation process of ventilation rate has been described previously (Xu *et al.*, 2014) and is also given in Text S2 (see the Supporting Information, SI). An error is inevitably involved in the calculation of the ventilation rate but was at an acceptable level, e.g., an error of 9.5% corresponding a difference of 1°C in the measurement of temperature but a 10% increase in the coefficient of heat transmission, solar radiation, and live mass can arise with inaccuracies of 1.0, 0.1 and 1.4% in the estimates, respectively (Blanes *et al.*, 2005). Since there were no side-by-side measurements for indoor and outdoor PM, the daily PM emission rate was therefore calculated by multiplying the daily mean ventilation rate by the difference (daily mean indoor PM concentration minus mean outdoor PM concentration of the adjacent days), and further normalized by expressing either on a per livestock unit (LU, equivalent to 500 kg body weight) or per area (m^2) basis. Total weight of pigs in the pig building was determined by multiplying the directly counted pig numbers by the averaged weight of one pig as weighed by the stockman. Total area of the pig building was measured with a tapeline. The emission rate (mg h^{-1} LU^{-1}) was calculated by dividing the total emission rate by the total weight of pigs and then multiplying by 500 kg, and PM emission per area from the pig barn was obtained by multiplying the emission rate per LU by the live mass density (LU m^{-2}) in the barns. The emission of secondary inorganic PM was estimated by multiplying PM emission rate by contribution from the secondary inorganic PM.

Statistical Analysis

Statistical differences in TSP and PM_{10} concentrations between seasons within and outside the fattening houses were indicated by Duncan's multiple range tests following analysis of variance. The relationships between daily mean concentrations of particulate NO_3^-, SO_4^{2-}, Cl^- and NH_4^+ in TSP and PM_{10} were analyzed by calculating the Pearson correlation coefficient with a significance level of 0.05.

All statistical analysis was performed using the SPSS 13.0 statistical package (SPSS Inc., Chicago, IL).

RESULTS AND DISCUSSION

PM Concentrations and Emissions

Daily mean concentrations of PM ranged from 0.24 to 4.5 mg m^{-3} (average 1.85 mg m^{-3}) for TSP inside house I and from 0.13 to 2.3 mg m^{-3} (average 0.63 mg m^{-3}) for PM_{10} inside house II, with large seasonal variations at both sites (Table 1). For indoor TSP concentrations, fattening periods which started during the autumn and winter months had significantly higher (both $P < 0.05$) values than fattening periods which started during the summer months. The average concentration of indoor PM_{10} during a single fattening period was three times higher (significantly, $P < 0.05$) in spring than in summer. In general the seasonal variation of indoor PM concentrations was clearly influenced by ventilation rate, determined by environmental conditions. The present results show that the indoor PM concentration decreased at increasing ventilation rate and indoor relative humidity, which is consistent with the findings of previous studies (Gustafsson, 1999; Haeussermann *et al.*, 2008). Many other factors such as feeding operations, animal activity, number of animals, and animal weight, which affect PM generation and levels, are also considered to affect the seasonal variances (Cambra-López *et al.*, 2010). Daily mean particle concentrations ranged from 0.13 to 0.58 mg m^{-3} (average 0.32 mg m^{-3}) for TSP outside house I and from 0.04 to 0.97 mg m^{-3} (average 0.32 mg m^{-3}) for PM_{10} outside house II. In the present study there was no obvious seasonal variation in outdoor TSP or PM_{10} concentrations. This is likely due to the changes in wind velocity and wind direction between different sampling seasons (Fig. S2). Outdoor PM concentrations, except PM emissions from the barn, were likely influenced by outdoor environmental factors (e.g., resuspended PM, wind speed and direction). For example, we obsersed that frequent dust storms occurred in Beijing in the spring of year 2011. In addition, there were roads and large tracts of agricultural land to the south (upwind) of the pig farm and this can also contribute to the outdoor particles on windy days during the experimental period.

Table 2 presents mean emission rate per pig and normalized emission rate (expressed as per LU and per m^2) for TSP in house I and for PM_{10} in house II during each monitoring period. Mean TSP emission rates per LU and area were in the ranges 41.7–1544.4 mg h^{-1} LU^{-1} (average 467.5 mg h^{-1} LU^{-1}) and 3.74–114.9 mg h^{-1} m^{-2} (average 38.6 mg h^{-1} m^{-2}), respectively. Mean PM_{10} emission rates per LU and area were in the ranges 19.0–1028 mg h^{-1} LU^{-1} (average 256.7 mg h^{-1} LU^{-1}) and 1.43–88.1 mg h^{-1} m^{-2} (average 16.8 mg h^{-1} m^{-2}), respectively. In the present study a large seasonal variation in TSP emissions was observed, mainly attributable to changes in PM concentration and ventilation rate of the barn. Mean TSP emissions were highest in autumn, and about equal in summer and winter. In the autumn the ventilation rates, which were at a high level in early autumn (average 892 m^3 h^{-1} LU^{-1}) could be an explanatory factor. The relatively high TSP emissions in winter likely resulted from a significant ($p < 0.05$) increase in indoor PM concentration (Table 1) casued by conditions including the completely closed windows and coal burning-based domestic home heating in the manual workers' room (from the middle of November through the middle of March the following year), even though the ventilation rates in winter were markedly lower than those in summer and autumn due to a large difference between indoor and outdoor temperatures. Because of the similar temperature adjustment management between the spring and autumn seasons, it was assumed that the mean emission rates in spring and summer

Table 1. Seasonal mean, median, minimum and maximum and standard deviations of PM concentrations (mg m^{-3}) inside and outside the pig houses from June 2010 to August 2011.

Site	Year		Indoor TSP			Outdoor TSP		
			Summer	Autumn	Winter	Summer	Autumn	Winter
			6/2–8/31	9/15–11/30	12/1–1/7	6/2–8/31	9/15–11/30	12/1–1/7
Fattening house I	2010/2011	Mean[a]	0.59a	2.34b	3.94c	0.31a	0.32a	0.34a
		Median	0.47	2.45	4.00	0.32	0.27	0.33
		Min	0.24	0.37	3.00	0.16	0.13	0.16
		Max	1.89	4.25	4.50	0.51	0.58	0.52
		s.d.	0.32	1.03	0.57	0.09	0.14	0.11
		n	30	22	13	18	22	6

Site	Year		Indoor PM_{10}		Outdoor PM_{10}	
			Spring	Summer	Spring	Summer
			4/1–5/30	6/1–8/11	4/1–5/30	6/1–8/11
Fattening house II	2011	Mean[a]	0.96b	0.34a	0.42a	0.22a
		Median	0.60	0.33	0.33	0.12
		Min	0.38	0.13	0.21	0.04
		Max	2.30	0.64	0.97	0.74
		s.d.	0.63	0.14	0.25	0.21
		n	20	24	7	11

[a] Different letters in the same row denote differences in TSP or PM_{10} concentrations among seasons in and/or outside fattening houses are significant at the 5% level.

Table 2. Barn measurements of TSP and PM_{10} emission rates for monitoring periods during different seasons.

Season	Site	Monitoring period	No.[c]	Live mass density	Mean Emission rate[a]	Mean normalized emission rate[a, b]	
				LU m^{-2}	(mg h^{-1} pig^{-1})	(mg h^{-1} LU^{-1})	(mg h^{-1} m^{-2})
					TSP	TSP	TSP
Summer 2010	Fattening house I	6/2–8/31	30	0.083	57.5 (23.6)	406.6 (237.7)	32.1 (16.1)
Autumn 2010		9/15–11/30	22	0.085	72.6 (41.7)	626.8 (421.8)	50.3 (29.8)
Winter 2010		12/1–1/7	13	0.099	57.3 (14.0)	339.4 (96.8)	34.1 (9.9)
Mean						467.5 (315.6)	38.6 (24.4)
					PM_{10}	PM_{10}	PM_{10}
Spring 2011	Fattening house II	4/1–5/30	20	0.051	20.7 (16.4)	266.6 (223.2)	13.9 (11.1)
Summer 2011		6/1–8/11	24	0.072	38.3 (36.7)	248.5 (230.2)	19.3 (16.8)
Mean						256.7 (236.2)	16.8 (16.4)

[a] Numbers in parenthesis denote standard deviation.
[b] Based on 500 kg live animal weight.
[c] Numbers of indoor PM samples used for estimate of emission.

were the same. Therefore, an annual emission factor of 0.6 kg TSP^{-1} pig^{-1} yr^{-1} for fattening pigs was obtained based on middle -term emission measurements in house I from 2010 to 2011. Due to water shortages in north China, the "gan qing fen" ("cleaning the manure dry") manure removal system is obligatory for pig fattening farms in the Beijing region (Schuchardt *et al.*, 2011), and most intensive pig farms in Beijing and north China have adopted this system. About 3.14 million (head) fattening pigs were breed in the Beijing Municipality in 2010 (CNBS, 2010). Based on these figures, a total of 1.9 million tonnes of dust or particles were emitted from fattening pig production in Beijing, posing serious environmental and health threats. PM_{10}, emission rates in spring were not much lower than those in summer. This was likely due to high ventilation rates (average 1328.5 m^3 h^{-1} LU^{-1}) and high levels of indoor PM_{10} concentrations (average 0.96 mg m^{-3}) in the spring.

The reported PM emissons from the fattening barns were commonly expressed as per livestock unit (LU) for an objective comparison among them (See Tables 3 and 4 and reference therein). However, another form of expression for the PM emission rate, expressed per floor area, has been widely used in recent studies (Kim *et al.*, 2008a; Van Ransbeeck *et al.*, 2012). In order to make a further comprehensive comparison, the average value of live mass density of 0.077 LU m^{-2} obtained in our study, which covered all seasons throughout the year for the fattening barn, was used to convert the reported PM emissions (expressed per LU) from previous studies into an area basis. It should be noted that the values derived from this conversion are considered to be rough estimates. Based on results reported in the literature by several other researchers (Table 3), values of the average, minimum and maximum TSP concentrations in fattening pig barns were 3.81, 0.03 and 21.0 mg m^{-3}, respectively. The mean emission rates of TSP per LU and area were 728.2 mg h^{-1} LU^{-1} and 56.1 mg h^{-1} m^{-2}, respectively. Concentrations of PM_{10} in the fattening pig barns reported by other researchers averaged 0.95 mg m^{-3} and ranged from 0.02 to 6.41 mg m^{-3}. Emissions of PM_{10}, expressed per LU and area, from fattening pig barns reported from several studies were on average 184.6 mg h^{-1} LU^{-1} and 14.2 mg h^{-1} m^{-2} (Table 4). According to previous reports, only Takai *et al.* (1998) and Kim *et al.* (2008a) estimated TSP emissions from fattening pig buildings, whereas the TSP concentration measurements were well performed in all the earlier studies. In contrast, numerous recent studies in pig fattening facilities have focused on the concentrations and emissions of PM_{10}. Considerable differences in concentration and emission of PM were found between studies. Variations in the concentration and emission of PM between the previous reports were likely due to a combination of different factors such as external climatic conditions and the degree of indoor cleanliness when taking samples.

Compared to the reported data (averaged data from references in Table 3 and 4, see Table S2), indoor concentrations of both TSP and PM_{10} were both lower in the present study. This could be explained by two factors. Firstly, in many cases pig buildings with natural ventilation have higher ventilation rates than those that are mechanically ventilated (Kim *et al.*, 2008b), which can partially explain the lower PM concentrations found in the present study. The results from previous studies were mostly obtained in mechanically ventilated pig barns. Secondly, several best management practices are included in the "cleaning the manure dry" system, such as feeding twice a day with dry meal, manure removal twice a day and pit flushing, all of which can reduce PM generated inside pig buildings. For example, Bundy and Hazen (1975) showed that feeding twice a day with a dry feeder resulted in lower PM concentrations compared to free access to feed by pigs. Manure and feed are regarded as the main sources of PM in the pig house (Cambra-López *et al.*, 2011) so twice a day manure removal shortens the retention time of manure in the pig house, thereby substantially reducing the PM produced from manure. In addition, fogging, spraying or sprinkling clean water may reduce the concentration of dust in the air (Takai and Pedersen, 2000; Aarnink *et al.*, 2011; Costa *et al.*, 2014). Therefore, the lower PM concentrations inside pig houses operated with the "cleaning the manure dry" manure removal system are expected and reasonable. Nevertheless, daily concentrations of indoor TSP in early autumn (3.37 ± 0.65 mg m^{-3}) and in winter (3.95 ± 0.57 mg m^{-3}) as well as indoor

Table 3. Concentrations (unit: mg m^{-3}) and emissions (unit: mg h^{-1} LU^{-1} and mg h^{-1} m^{-2}) of TSP in pig buildings as reported in the literature.

Concentration					
Country	Housing type	Animal type	Mean	Range	References
U.S.	Slats	Fatteners	8.00	6.4–9.6	(Curtis *et al.*, 1975)
	Slats	Fatteners	15.30	—	(Donham *et al.*, 1986)
	Slats	Fatteners	2.00	1.3–2.7	(Meyer and Manbeck, 1986)
	Slats	Fatteners	7.85	6.9–8.8	(Heber and Stroik, 1988)
	Slats	Fatteners	2.41	—	(Zhang *et al.*, 1998)
	Slats	Fatteners	2.75	—	(Wang *et al.*, 2002)
E.U.	Slats	Fatteners	2.82	0.47–9.55	(Attwood *et al.*, 1987)
	Slats	Fatteners	—	1.66–21.04	(Crook *et al.*, 1991)
	Slats	Fatteners	3.19	0.4–47.00	(Pederson, 1993)
	Slats	Fatteners	2.40	1.00–5.00	(Hinz and Linke, 1998)
	Slats	Fatteners	2.42	—	(Takai *et al.*, 1998)
	Litter	Fatteners	1.30		
Canada	Slats	Fatteners	2.20	1.60–2.74	(Barber *et al.*, 1991)
	Slats	Fatteners	3.54	2.15–5.60	(Duchaine *et al.*, 2000)
Taiwan	Slats	Fatteners	0.25	0.03–1.11	(Chang *et al.*, 2001)
S Korea	Slats	Fatteners	1.62	0.18–1.32	(Kim *et al.*, 2008a)
	Litter	Fatteners	2.94	0.52–1.68	
Mean			3.81	0.03–21.04	

Emission					
Country	Housing type	Animal type	Mean		References
			mg h^{-1} LU^{-1} [a]	mg h^{-1} m^{-2} [b]	
E.U.	Slats	Fatteners	612.3	47.1	(Takai *et al.*, 1998)
	Litter	Fatteners	725.5	55.9	
S Korea	Slats	Fatteners	556.5	42.9	(Kim *et al.*, 2008a)
	Litter	Fatteners	1018.3	78.4	
Mean			728.2	56.1	

[a] Based on 500 kg live animal weight.
[b] Obtained by multiplying 0.077 LU m^{-2} by emission rate per LU.

Table 4. Concentrations (unit: mg m^{-3}) and emissions (unit: mg h^{-1} LU^{-1} and mg h^{-1} m^{-2}) of PM_{10} in pig buildings as reported in the literature.

Concentration					
Country	Housing type	Animal type	Mean	Range	References
E.U.	Slats	Fatteners	0.73	0.06–6.41	(Haeussermann *et al.*, 2008)
	Slats	Fatteners	0.69	—	
	Litter	Fatteners	—	0.19–0.39	(Haeussermann *et al.*, 2007)
	Slats	Fatteners	0.72	—	(Van Ransbeeck *et al.*, 2013a)
	—	Fatteners	—	0.02–2.29	(Van Ransbeeck *et al.*, 2013b)
	Slats	Fatteners	0.62	0.03–1.49	(Van Ransbeeck *et al.*, 2012)
	Slats	Fatteners	1.97	—	(Ulens *et al.*, 2014)
Mean			0.95	0.02–6.41	

Emission					
Country	Housing type	Animal type	Mean		References
			mg h^{-1} LU^{-1} [a]	mg h^{-1} m^{-2} [b]	
E.U.	Slats	Fatteners	399.6	30.8	(Emission Inventory Guidebook, 2007)
	Slats	Fatteners	249.6	19.2	(Haeussermann *et al.*, 2008)
	Slats	Fatteners	107.5	8.3	
	Litter	Fatteners	58.3	4.5	(Haeussermann *et al.*, 2007)
	Slats	Fatteners	107.9	8.3	(Costa *et al.*, 2009)
Mean			184.6	14.2	

[a] Based on 500 kg live animal weight.
[b] Obtained by multiplying 0.077 LU m^{-2} by emission rate per LU.

PM_{10} in early spring (average 1.65 ± 0.40 mg m^{-3}) exceeded the Chinese daily exposure limits for humans (implemented nationwide in 1999 for pig husbandry) of 3 mg m^{-3} for TSP and 1 mg m^{-3} for PM_{10} (MEPC, 1999), respectively. Thus, more comprehensive PM mitigation techniques (e.g., liquid feeding and oil spraying) by the farm manager should be implemented in order to improve air quality in pig buildings.

The PM emission, values were generally lower for TSP and slightly higher for PM $_{10}$ in our study compared to data reported by other studies (Table S2). This phenomenon could be explained by the differences in indoor PM concentration and ventilation rate. As mentioned above, ventilation rate is usually higher in a naturally ventilated system such as in the present experimental farm compared to a mechanically ventilated system. So the much lower TSP concentrations, in comparison with the literature, would be one possible explanation for lower TSP emissions in our study. For PM_{10}, we found slighter higher emissions in comparison to other studies. This was likely linked to the higher ventilation rate under conditions in which the PM_{10} concentrations were only slightly lower than reported elsewhere.

Emission rates of PM are generally calculated as the multiplication of pollutant concentration and the ventilation rate recorded in the same minute. Concentrations of PM in the pig buildings can be determined simply, but it is very difficult to estimate accurately the ventilation rate of the pig building. According to Gay *et al.* (2003), tracer gas, heat balance, or carbon dioxide measurements for naturally ventilated pig buildings provide, at best, rough estimates of ventilation rate. Notwithstanding the fact that the emission rate in this study was obtained using an appropriate method, it should be noted that the precision of the derived emission rate is still disputable, i.e., a ventilation rate based on a heat balance will always produce errors when the true sensible heat production does not correspond exactly with the daily mean value (Pedersen *et al.*, 1998). In addition, concentrations of indoor and outdoor PM were not monitored simultaneously due to a lack of PM samplers. The correction made for incoming PM was therefore approximate only. Given the above two points it becomes quite clear that a more accurate method needs to be developed to determine the emission rate. For the present study it can be noted that the corrections for TSP and PM_{10} should be acceptable (at least partly). For example, the background concentrations used for correction of PM_{10} in spring and summer seasons averaged 0.29 mg m^{-3}. This value is comparable to put own measured results at a rural site in Beijing Municipality where PM_{10} concentrations averaged 0.34 mg m^{-3} in the spring and summer seasons (Lu, 2014).

Particulate NH_4^+, NO_3^-, SO_4^{2-} and Cl^- Concentrations and Their Correlations

Concentrations of gases (NH_3, SO_2, NO_2) and inorganic ions (particulate NH_4^+, NO_3^-, SO_4^{2-} and Cl^-) inside and outside the fattening pig houses are presented in Table S3. Higher concentrations of particulate NH_4^+ in TSP and PM_{10} were observed inside the houses compared to outside, on average 18.0 vs. 12.6 µg m^{-3} for TSP and 11.3 vs. 8.1 µg m^{-3} for PM_{10}. This can be explained by excess NH_3 and high relative humidity inside the houses combined with acid gases (e.g., HNO_3, H_2SO_4 and HCl) from the external environment, favouring the formation of particulate ammonium (Robarge *et al.*, 2002). In TSP the differences between indoor and outdoor concentrations of NO_3^- and SO_4^{2-} were not significant (both $p > 0.05$) except Cl^-. In PM_{10} there were no significant differences (all $p > 0.05$) between indoor and outdoor concentrations of particulate NO_3^-, SO_4^{2-} or Cl^-. It should be noted that part of the measured inorganic ions in PM may be ascribed to road and soil dust emissions as mentioned above.

Concentrations of particulate NH_4^+, SO_4^{2-}, NO_3^- and Cl^- in TSP showed large seasonal variations both inside and outside house I (Figs. 1(a) and 1(b)). Concentrations of particulate NH_4^+, SO_4^{2-} and NO_3^- were much higher in summer than in autumn or winter. The increased ventilation rates and climatic parameters (e.g., temperature and relative humidity) in summer (Table S1) can enhance the amount of NH_3 emitted and incoming acid gases, resulting in efficient mixing of air containing ammonia and acid gases (precursors of particulate NH_4NO_3 and $(NH_4)_2SO_4$). Moreover, increases in particulate SO_4^{2-} and NO_3^- in ambient air were favored in summer due to favorable external environmental factors (e.g., high rate of photochemical activity and high temperature) (Khoder *et al.*, 2002), which can directly increase their indoor concentrations via air exchange. Indoor concentrations of particulate Cl^- were comparable and highest in autumn and winter, and lowest in summer. Increased use of coal (for domestic heating starting from November to March the following year) in the manual workers' room may be the main cause, as coal combustion contributes to particulate Cl^- (Sun *et al.*, 2004). Indoor concentrations of particulate inorganic ions in PM_{10} were higher in a similar fashion to TSP, the season when windows were fully opened with the exception of particulate Cl^- (Fig. 1(c)). Surprisingly, seasonal variations in outdoor concentrations of NH_4^+, SO_4^{2-} , NO_3^- and Cl^- in PM_{10} not as large as those in TSP (Fig. 1(d)).

There were significant positive correlations between molar concentrations of NH_4^+ and NO_3^- as well as NH_4^+ and SO_4^{2-} at the four sampling sites, and the positive correlations between NH_4^+ and Cl^- were also significant outside houses I and II for both TSP and PM_{10} (Table S4). However, the correlation coefficients between molar concentrations of NH_4^+ and the sum of NO_3^- and SO_4^{2-} at the four sites were higher than those between NH_4^+ and the sum of NO_3^-, SO_4^{2-} and Cl^-. This indicates that particulate NH_4^+ was mainly combined with particulate NO_3^- and SO_4^{2-}, and so the particulates are likely to be present as NH_4NO_3, $(NH_4)_2SO_4$ and/or NH_4HSO_4, which are secondary products of reactions between gaseous NH_3, SO_2 and HNO_3.

Further analysis shows that mean molar ratios of NO_3^-/$NH_4^+$and (NH_4^+-NO_3^-)/SO_4^{2-} varied from 0.37 to 0.65 and from 0.71 to 1.62 across all sites, respectively (Table S5). Mean molar ratios of NO_3^-/NH_4^+ were higher at the outdoor sampling sites than the indoor sampling sites. This is because the NH_3 concentrations outside the barn were much lower than inside (Table S3), and gaseous NO_2 in

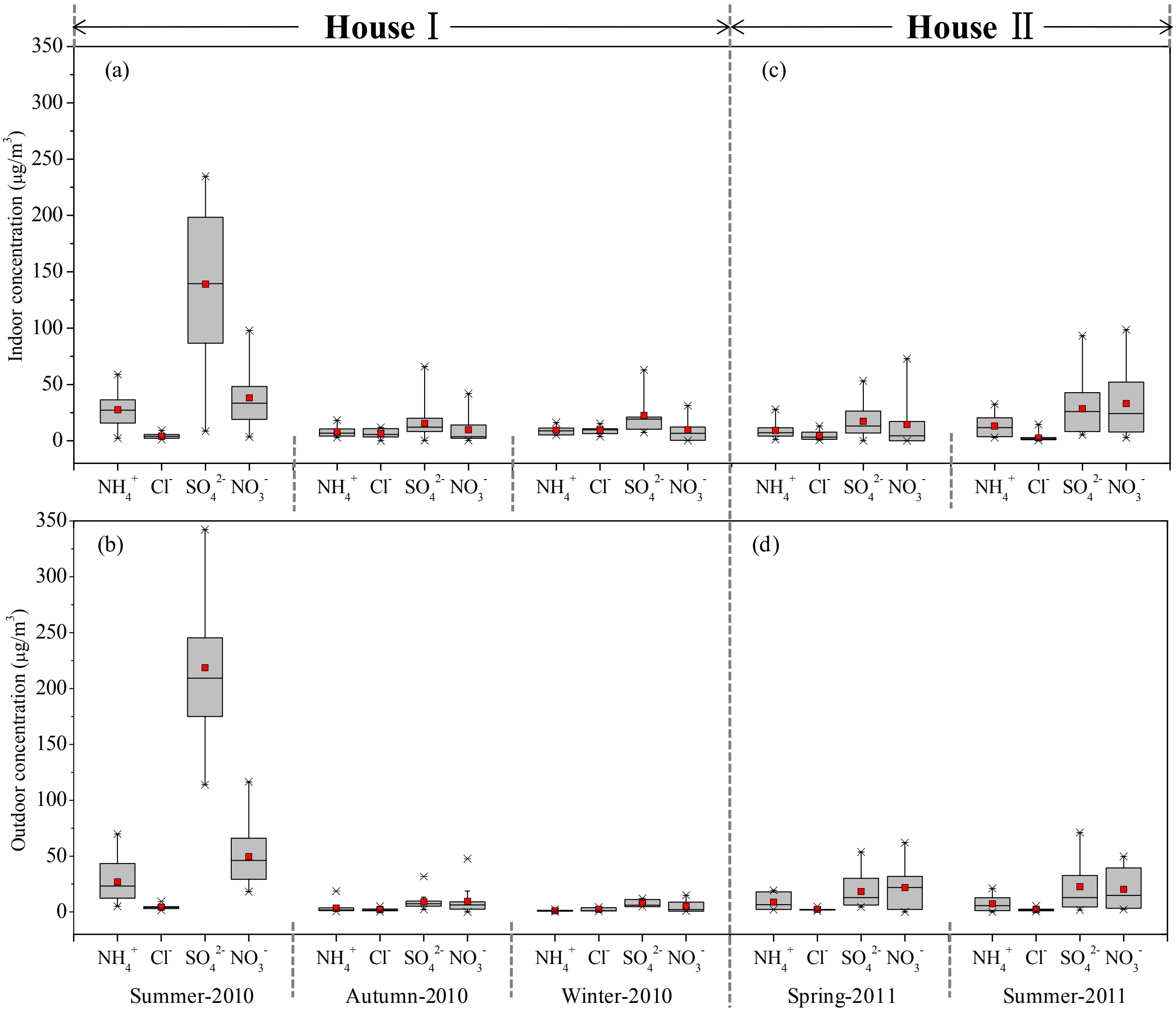

Fig. 1. Seasonal mean concentrations of particulate NH_4^+, NO_3^-, SO_4^{2-} and Cl^- in (a) TSP inside house I, (b) TSP outside house I, (c) PM_{10} inside house II and (d) PM_{10} outside house II. The black lines and red squares within the box mark the median and the mean, respectively. Whiskers above and below the box indicate the maximum and minimum values.

the external environment might be at a higher level compared to the indoor environment without emission sources. Mean molar ratios of $(NH_4^+ - NO_3^-)/SO_4^{2-}$ ((ammonium minus nitrate)/sulfate) were close to 1.0 at the outdoor sampling sites but were larger than 1.0 at the indoor sampling sites. This indicates that secondary particles existed mainly as NH_4NO_3 and NH_4HSO_4 at the outdoor sampling sites and as NH_4NO_3, NH_4HSO_4 and $(NH_4)_2SO_4$ at the indoor sampling sites.

Secondary Inorganic PM Concentrations and Emissions

On the basis of the findings of the present study regarding correlations between particulate NH_4^+, SO_4^{2-}, NO_3^- and Cl^- (Table S4), it can be concluded that secondary inorganic PM inside the two fattening houses were both formed mainly by particulate NH_4^+, NO_3^- and SO_4^{2-}. The daily mean concentrations of secondary inorganic PM (here referring to the sum of NH_4^+, SO_4^{2-} and NO_3^-) varied from 10.8 to 369 μg m^{-3} (average 116 μg m^{-3}) inside house I and from 5.1 to 221 μg m^{-3} (average 58.6 μg m^{-3}) inside house II, accounting for 0.3–50.8% (average 12.0%) and 1.0–48.7% (average 13.9%) of the corresponding indoor PM concentration. The concentration of secondary inorganic particles in PM_{10} (average of two sampling sites, 54.1 μg m^{-3}) in this study is comparable to the average value of 59.6 μg m^{-3} measured at six sites with secondary inorganic particle pollution on the North China Plain (Shen *et al.*, 2011). The daily mean emissions per LU and area varied from 1.2 to 286.3 mg h^{-1} LU^{-1} (average 68.6 mg h^{-1} LU^{-1}) and from 0.1 to 25.6 mg h^{-1} m^{-2} (average 5.66 mg h^{-1} m^{-2}) for secondary inorganic PM in TSP, respectively, while those for secondary inorganic PM in PM_{10} varied from 0.43 to 127.9 mg h^{-1} LU^{-1} (average 28.6 mg h^{-1} LU^{-1}) and from 0.02 to 10.4 mg h^{-1} m^{-2} (average 2.11 mg h^{-1} m^{-2}) (Table 5).The corresponding secondary PM emissions for TSP and PM_{10} were both distinctly higher in summer than at

Table 5. Mean secondary inorganic PM (SIPM) concentrations (referring to sum of NH_4^+, SO_4^{2-} and NO_3^-) and emission rates in TSP and PM_{10}.

Pollutant	Season	Site	Monitoring period	Mean concentration[a]	Mean emission rate[b]	Mean normalized emission rate[a,b]	
				($\mu g\ m^{-3}$)	($mg\ h^{-1}\ pig^{-1}$)	($mg\ h^{-1}\ LU^{-1}$)	($mg\ h^{-1}\ m^{-2}$)
SIPM in TSP	Summer 2010	Fattening house I	6/2–8/31	198.8 (95.5)	16.3 (8.9)	115.3 (57.7)	9.28 (4.90)
	Autumn 2010		9/15–11/30	33.1 (25.4)	1.45 (2.20)	9.36 (10.1)	0.77 (0.80)
	Winter 2010		12/1–1/7	47.5 (37.1)	0.79 (0.70)	5.08 (4.81)	0.50 (0.45)
SIPM in PM_{10}	Spring 2011	Fattening house II	4/1–5/30	41.1 (40.0)	1.07 (1.49)	12.3 (15.4)	0.69 (0.93)
	Summer 2011		6/1–8/11	74.6 (58.2)	6.99 (6.91)	42.2 (40.1)	3.29 (3.06)

[a] Numbers in parenthesis denote one standard deviation.
[b] Based on 500 kg live mass weight.

other times of year, resulting from higher concentrations of particulate NH_4^+, NO_3^- and SO_4^{2-} and increased ventilation rates in the summer (Fig. 1 and Table S1). Comparing emissions of secondary PM is difficult due to the absence of comparable studies.

The high concentrations and emissions, in particular of secondary PM in TSP and PM_{10} formed mainly at the source inside the pig barns and emitted due to the high natural ventilation rate are direct evidence for a major portion of the ammoniacal N (NH_3-N + NH_4^+-N) emitted from the pig farm being in the form of particulate matter. This may be subject to medium- and long-term transport and thus supports the evidence for very high atmospheric N deposition rates measured by our group not only in Beijing Municipality but also in more outlying regions of the NCP, including those with less intensively managed animal husbandry operations (Luo *et al.*, 2013, 2014).

CONCLUSIONS

This study is the first time to estimate PM and secondary inorganic PM emissions from pig houses based on measurements at an industrial pig farm in northern China. Two similar fattening pig houses were sampled during three fattening periods.

Significant seasonal variations were observed for indoor, but not outdoor, PM concentrations. Indoor TSP concentrations were higher in winter (3.94 ± 0.57 mg m^{-3}) than in summer (0.59 ± 0.32 mg m^{-3}) or autumn (2.34 ± 1.03 mg m^{-3}), while indoor PM_{10} concentrations were higher in spring (0.96 ± 0.63 mg m^{-3}) than in summer (0.34 ± 0.14 mg m^{-3}). TSP emissions were higher in autumn than summer or winter, while comparable values were found between spring and summer for PM_{10} emissions.

The indoor TSP and PM_{10} concentrations were usually lower than the concentrations found in the literature, whereas emissions were lower for TSP but higher for PM_{10} compared to the literature. This was likely a result of the combined influences of PM concentrations and ventilation rates in the pig houses in the present study.

The secondary inorganic PM inside and outside the pig houses was mainly formed by particulate NH_4^+, NO_3^- and SO_4^{2-}. The concentrations and emissions of secondary inorganic PM in TSP and PM_{10} were both higher in summer compared to the respective monitoring period in other seasons, mainly due to an increased level of interaction between NH_3 and acid gases associated with the ventilation rate. It may be feasible to reduce secondary inorganic PM levels by directly targeting ammonia and/or the acid gases.

Our results demonstrate that, as in the case of gaseous NH_3, the so-called "gan qing fen" solid and liquid excreta separation and handling system has proven beneficial for controlling concentrations and emissions of PM and also in comparison to other manure handling systems e.g. with slatted floors. Given the comparable PM_{10} emissions in the present study compared to research worldwide as well as the rapidly expanding number and size of intensively managed animal husbandry operations in China, more efforts are required towards on devising future pig house construction and manure handling strategies.

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SUPPORTING INFORMATION

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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