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# Assessing Ammonia (NH<sub>3</sub>) Emissions, Precursor Gas (SO<sub>2</sub>, NO<sub>x</sub>) Concentrations, and Source Contributions to Atmospheric PM<sub>2.5</sub> from a Commercial Manure Composting Facility

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Article

# Assessing Ammonia (NH<sub>3</sub>) Emissions, Precursor Gas (SO<sub>2</sub>, NO<sub>x</sub>) Concentrations, and Source Contributions to Atmospheric PM<sub>2.5</sub> from a Commercial Manure Composting Facility

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**Abstract:** Increased ammonia (NH<sub>3</sub>) emissions from intensive agriculture negatively affect environmental and ecosystem health, contributing to particulate matter (PM) formation and the potent greenhouse gas, N<sub>2</sub>O. Better understanding NH<sub>3</sub> emissions from the manure composting process and their behavior as a constituent of the atmospheric aerosol load is a crucial element in creating better farm management systems, improved public health outcomes and mitigating the broader environmental and climatic impacts of agriculture. Retarded generation of PM with major constituent source of NH<sub>3</sub> is a primary mechanism evaluating the effects of agricultural contribution to PM. This study aims to quantify NH<sub>3</sub> emissions, examine the influence of environmental factors, and investigate the relationship between precursor gases (SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub>) and PM<sub>2.5</sub> at a modern manure composting facility in Paju, South Korea. Over 35 days, average internal concentrations of NH<sub>3</sub>, SO<sub>2</sub>, and NO<sub>x</sub> were significantly higher than external levels. NH<sub>3</sub> concentrations reached  $3.64 \pm 0.06$  mg m<sup>-3</sup> at 3m height and  $2.43 \pm 0.16$  mg m<sup>-3</sup> at ground level, while the total NH<sub>3</sub> flux from the facility was  $24.47 \pm 1.39$  NH<sub>3</sub>-N kg d<sup>-1</sup>. Internal PM<sub>2.5</sub> concentrations ( $36.9 \pm 2.6$  μg m<sup>-3</sup>) were about 50% higher than external levels ( $23.7 \pm 2$  μg m<sup>-3</sup>), with a moderate correlation ( $r = 0.341$ ) suggesting some contribution of external PM<sub>2.5</sub> to internal levels. Despite large internal emissions, the facility's sealed design with a negative pressure ventilation system effectively minimized external emissions. These results suggest that while manure composting facilities are significant sources of NH<sub>3</sub> and PM<sub>2.5</sub>, advanced systems like high-volume ventilation and scrubbing technologies can effectively reduce their impact on regional air pollution, contributing to better environmental management in agriculture.

**Keywords:** Particle Matter; Greenhouse gas; Precursor gases; Manure composting facility

## 1. Introduction

Agriculture-related emissions, including greenhouse gases (GHGs), ammonia (NH<sub>3</sub>), particulate matter precursors, and toxic pollutants, can travel across regions, adversely affecting air quality and posing significant risks to human health (XXX). These far-reaching impacts highlight the urgent need for effective emission management strategies in agricultural systems to mitigate their environmental and health consequences (XX).

In 2017, agricultural activities were estimated to be responsible for approximately 80% of ammonia (NH<sub>3</sub>) emissions from the Republic of Korea [1] in line with similar figures from the United States, China and the European Union [2-5]. Livestock manure is thought to be the single largest contributor to NH<sub>3</sub> emissions [6], estimated to account for around 50% of the total in the US and China [7,8]. Increased NH<sub>3</sub> emissions as a result of intensive agriculture are known to negatively affect environmental and ecosystem health through eutrophication [9], and as an indirect source of the potent greenhouse gas, nitrous oxide (N<sub>2</sub>O) [3,10].

Per capita meat consumption in South Korea is projected to continue increasing over the next decade, with consumption of chicken and pork having doubled since the year 2000, and beef consumption increasing by more than 40% [11]. Available grazing and arable land is limited in South Korea [12], and pressure on agriculture production means that concentrated animal feeding

operations (CAFOs) are common. Effectively managing the manure produced in these facilities is important to controlling  $\text{NH}_3$  and odour pollution [8,13]. Manure composting is an appealing management technique, involving the growth of aerobic micro-organisms within the manure that break it down into a nutrient rich organic fertilizer that can be applied to agricultural land [5]. When composted, biological degradation of the manure results in the production of Sulfur Dioxide ( $\text{SO}_2$ ) and Nitric Oxide ( $\text{NO}_x$ ) gasses, as well as greenhouse gasses (GHGs) such as Carbon Dioxide ( $\text{CO}_2$ ), Methane ( $\text{CH}_4$ ) and  $\text{N}_2\text{O}$  [14,15]. Ammonium ( $\text{NH}_4^+$ ) is also created through microbial processes within the manure and although not volatile itself, exists in equilibrium with  $\text{NH}_3$  gas [10]. Factors that affect  $\text{NH}_3$  emissions from manure compost include temperature, moisture content, pH, initial nitrogen content and air turbulence [5,10]. Aerating compost (via forced aeration, mechanical turning, and the use of organic bulking agents such as wood chips) has also been found to reduce direct emissions of GHGs  $\text{CH}_4$  and  $\text{N}_2\text{O}$ , lowering the impact of manure composting on climate change, but this causes increases in the pH and temperature of the manure, shifting the equilibrium towards gaseous  $\text{NH}_3$ , increasing emissions, and leading to interest in capturing and scrubbing  $\text{NH}_3$  from the air [16-18].

$\text{NH}_3$  emissions are thought to be an important contributor to the formation of secondary particulate matter.  $\text{NH}_3$  gas reacts with acidic compounds (such as  $\text{SO}_2$  and  $\text{NO}_x$ ) in the atmosphere resulting in  $\text{PM}_{2.5}$  formation [19,20]. Studies have investigated the role of  $\text{NH}_3$  interactions with  $\text{SO}_2$  and  $\text{NO}_x$  in contributing to severe  $\text{PM}_{2.5}$  episodes in both South and East Asia [4,21,22], as well as in Europe [23,24], and North America [7,25]. Poor air quality is among the greatest issues currently facing East Asian societies, negatively affecting health outcomes [26,27] and economic productivity [28] across the region.

Better understanding  $\text{NH}_3$  emissions from the manure composting process and their behavior as a constituent of the atmospheric aerosol load is a crucial element in creating better farm management systems, improved public health outcomes and mitigating the broader environmental and climatic impacts of agriculture. This study aims to characterize and quantify  $\text{SO}_2$ ,  $\text{NO}_x$ ,  $\text{NH}_3$  (SNA) and  $\text{PM}_{2.5}$  emissions from a modern manure composting facility in South Korea in order to a) calculate  $\text{NH}_3$  flux, b) assess the role that physical and environmental variables play in  $\text{NH}_3$  emission, c) investigate any relationship between SNA and  $\text{PM}_{2.5}$  formation within the facility. In addition, this study aims to evaluate whether this modern facility, equipped with an  $\text{NH}_3$  scrubbing system, is able to minimize external emissions that may contribute to regional air pollution.

## 2. Materials and Methods

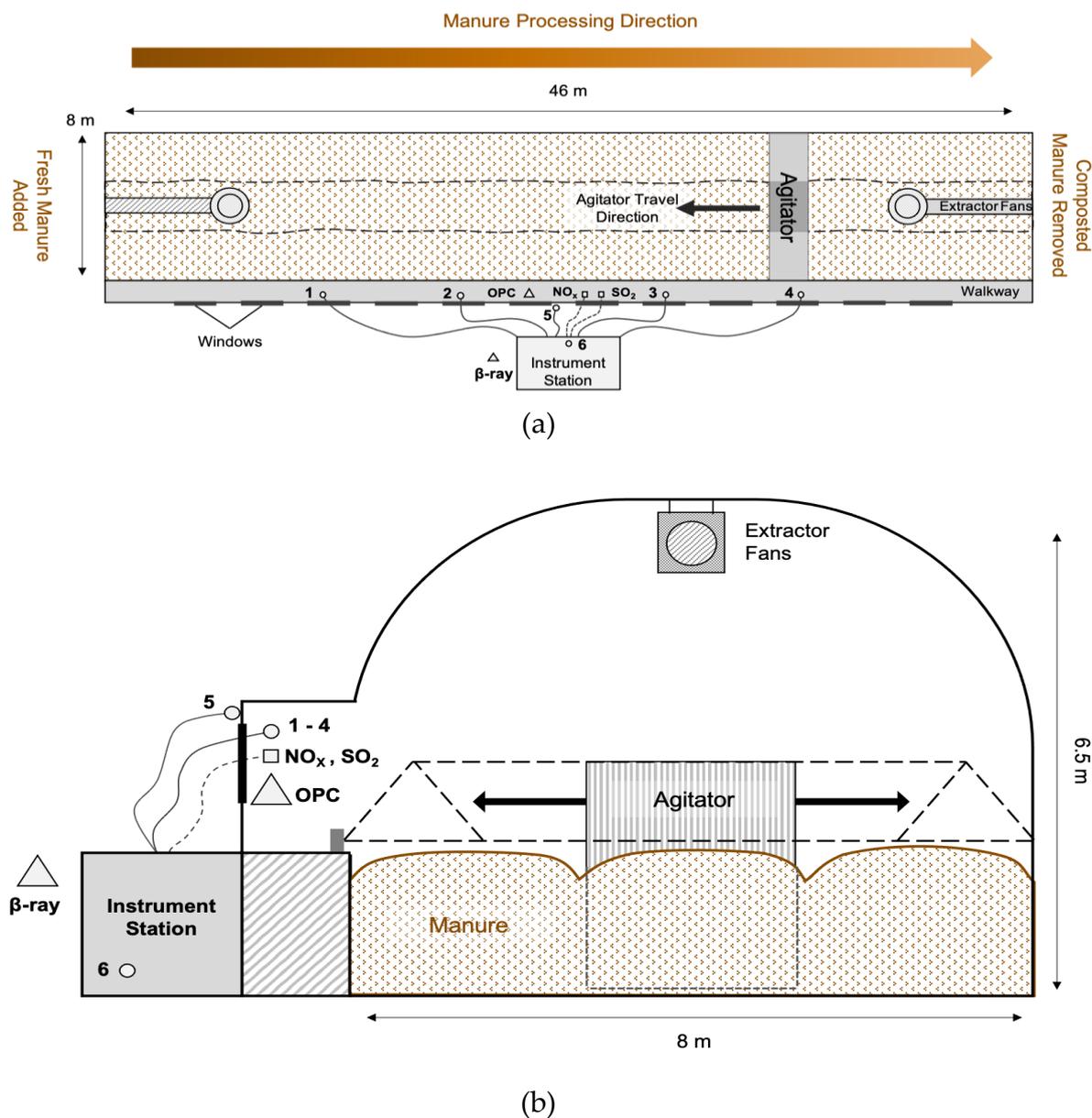
A multi-instrument monitoring system was set-up to monitor SNA emissions and PM concentrations at a manure composting facility located in Papyeong-myeon, Paju City, Gyeonggi Province, South Korea. The chosen facility began operations in 2017, and is a purpose built, modern manure composting facility installed with a high-volume ventilation and scrubber system. The monitoring instruments were operated over a 5-week period between 11<sup>th</sup> June – 16<sup>th</sup> July 2020. Internal concentrations of SNA and external concentrations of  $\text{NH}_3$  and  $\text{PM}_{2.5}$  were monitored continuously. In addition, internal  $\text{PM}_{2.5}$  concentrations were monitored for shorter durations during the monitoring period.

### 2.1. Monitoring Site

The Paju manure composting facility (PMCF) is designed to process animal manure from animal feeding operations (AFOs) into organic fertilizer for agricultural use. The facility is composed of several distinct sections. Fresh manure is delivered from local poultry and livestock farms and stored for 25 days in an atrium sectioned into ten large bays for bulk manure storage prior to composting. The manure is then mixed at a manure 85 : 15 sawdust ratio and added to one of two halls designed to agitate the manure during the composting process. In each hall, the manure is mixed daily by a conveyor-type agitator that lifts, aerates and deposits the manure along three windrows. During the daily agitation the manure is gradually moved from the input to the extraction end of each hall over a period of 18 days. Following the composting agitation process, the manure is extracted and taken

to a composted manure storage hall where it is allowed to rest for approximately 33 days, before being moved to a facility for packaging composted manure for sale as organic fertilizer. The facility also contains a separate underground tank for processing the liquid / slurry component of manure. This study focused on monitoring precursor gas and PM emissions from one of the manure composting agitation halls (Fig. 1). It is expected that the greatest  $\text{NH}_3$  losses occur during this stage of the manure composting process due to the regular agitation and aeration of the manure.

The building is installed with two  $1400 \text{ m}^3 \text{ min}^{-1}$  capacity extraction fans which remove the air in the facility, maintaining a negative pressure internally, and pass it through scrubbers designed to remove precursor gasses and PM before it is emitted externally. Two ceiling-mounted vents leading to the extractor fans are located in each manure composting agitation hall.



**Figure 1.** a) a birds-eye view of the facility showing the location of sampling points, and b) a side on view.

## 2.2. Monitoring System

A weatherproof station to house the monitoring instruments was installed outside of the PMCF, adjacent to one of the manure composting agitation halls. The instruments were housed externally to reduce the possibility for contamination and corrosion of the instruments due to high precursor gas SNA concentrations inside the facility. Filtered gas samplers were placed along a walkway inside the manure composting agitation hall. The instruments were connected to the filtered gas samplers with 10-meter lengths of 4 mm diameter Teflon tubing, wrapped in a heated trace to reduce condensation, and run through holes in the buildings window fittings. Filtered gas samplers were used to ensure that the tubing did not become blocked with dust particles entrained during the daily manure agitation cycles.

### 2.2.1. Precursor gas measurement

NH<sub>3</sub> gas concentrations were monitored via a 6-channel gas sampling doser (INNOVA1403) connected to an INNOVA1512i photoacoustic NH<sub>3</sub> gas analyzer (INNOVA1403 & 1512i, LumaSense Technologies, Ballerup, Denmark). Four channels were used to monitor NH<sub>3</sub> concentrations along the length of the manure composting agitation hall. In addition, two gas sampling channels were installed outside the facility, one located at a height of 3 m above ground level adjacent to a window on the outer wall of the manure composting agitation hall and one located at ground level next to the instrument station. The INNOVA gas sampling system provided NH<sub>3</sub> concentrations (measurement range: 0 - 500 ppm, sensitivity: 1 ppb) sequentially across the 6 channels at approximately 45 second intervals. Except where explicitly mentioned, internal NH<sub>3</sub> emissions are an average of the data from the four internal channels.

Sulphur Dioxide (SO<sub>2</sub>) and Nitrogen Oxide (NO<sub>x</sub>) gas concentrations (measurement range: 0 - 500 ppb, sensitivity: 0.1 ppb) were sampled at five-minute intervals at single points inside the manure composting agitation hall using KENTEK gas analyzers (KENTEK CO. LTD., Daejeon, South Korea). NO<sub>x</sub> was measured on a KENTEK Mezus 210 instrument via chemiluminescence. SO<sub>2</sub> was measured by a KENTEK MEZUS 110 instrument, using an ultraviolet fluorescence method. NO<sub>x</sub> gas concentrations are only available prior to the 4<sup>th</sup> July 2020. While heated traces were used to avoid moisture condensation inside the teflon sampling tubes, condensation buildup after this date caused abnormal NO<sub>x</sub> data readings after 4<sup>th</sup> July.

The presented precursor gas concentration datasets measured at the facility have been transformed to 15-minute averages to reduce noise and to aid cross comparison between data sampled at different time intervals. For comparison with Environmental Variables and Regional PM and gas concentration measurements, precursor gas concentrations have been converted to hourly average values to match the data resolution of this externally sourced data

Regional hourly SO<sub>2</sub> and Nitrogen Dioxide (NO<sub>2</sub>) gas concentration data measured at local government operated air quality monitoring sites in Paju were downloaded from the Air Korea government website for the duration of the study (<https://www.airkorea.or.kr/index>).

This study investigates the difference in gas concentrations during day time and night time, and between weekdays and weekends. This is because manure agitation only occurs during the day time on weekdays. Day time is defined in this study as between 7am – 7pm, while night time is from 7pm to 7am. Weekends are defined as the 48 hours from 7am on Saturday until 7am on Monday.

### 2.2.2. Particulate Matter measurement

A Beta-ray absorption method ultrafine dust automatic measuring device was installed at ground level outside the facility next to the instrument station to monitor external PM concentrations. The β-ray instrument conducted hourly air samples for the duration of the sampling period.

A GRIMM 11-D Aerosol Spectrometer Optical Particle Counter (GRIMM OPC 11-D, Grimm Aerosol Technik Ainring GmbH & Co.KG, Ainring, Germany) instrument was used to sample internal PM concentrations in the composting conveyor hall for two separate periods, from 24-26 June and from the 8-16 July 2020.

Regional particulate matter concentration data measured at local government operated air quality monitoring sites in Paju were downloaded from the Air Korea government website at an hourly resolution for the duration of the study (<https://www.airkorea.or.kr/index>).

### 2.2.3. Environmental Variables

Hourly local temperature and humidity data measured at a government operated meteorological station in Paju were downloaded from the Korean Meteorological Association website for the duration of the study (<https://data.kma.go.kr/>).

### 2.3. Ammonia Emission Flux Calculations

For calculation of the flux of NH<sub>3</sub> emissions from manure being processed inside the manure composting agitation hall, the extraction fans were disabled, allowing gaseous NH<sub>3</sub> concentrations to rise until they reach peak concentration, likely as a result of equilibrium in the concentration gradient at the manure – air boundary. The difference in normal (ventilated) NH<sub>3</sub> concentrations and saturated NH<sub>3</sub> concentrations was divided by the time taken for this increase to occur, and the results were averaged across the four internal NH<sub>3</sub> sampling channels.

$$\text{Emission Rate}_{\text{NH}_3} = (C_{\text{NH}_3(\text{sat.})} - C_{\text{NH}_3(\text{vent.})}) / \Delta t \quad (1)$$

Equation 1 demonstrates how NH<sub>3</sub> emission rate was calculated, where  $C_{\text{NH}_3(\text{sat.})}$  is the point at which NH<sub>3</sub> concentrations in the air reach equilibrium with NH<sub>3</sub> concentrations at the manure surface (i.e. there is no longer a concentration gradient leading to a stabilization of NH<sub>3</sub> concentrations in the air),  $C_{\text{NH}_3(\text{vent.})}$  is the ambient concentration of NH<sub>3</sub> under ventilated conditions (i.e. when there is negative pressure, and emitted NH<sub>3</sub> is being constantly removed and scrubbed by the ventilation system during ordinary operation), and  $\Delta t$  is the change in time in minutes between these two states following the disengagement of the ventilation system.

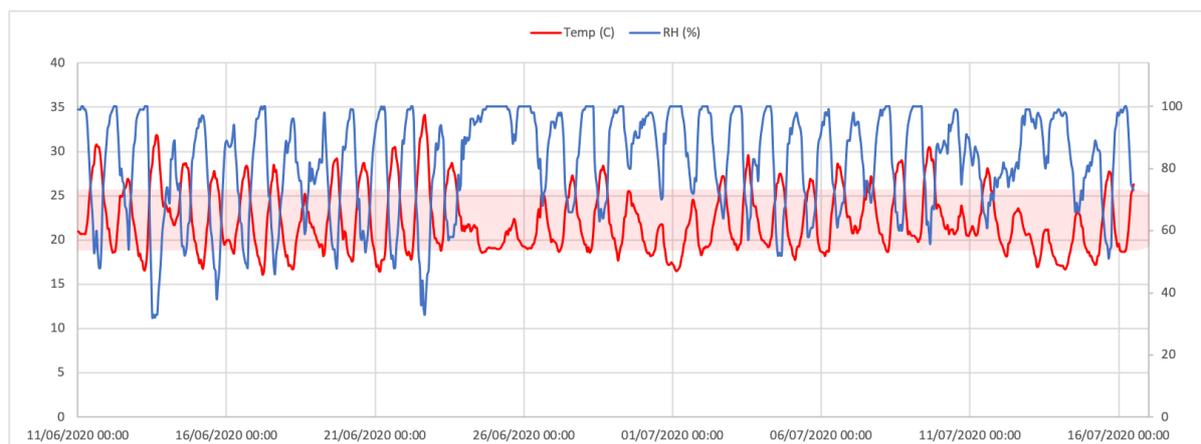
### 2.4. Statistical Analyses

The bivariate correlations between gas concentrations and environmental variables were tested using Pearson's correlation coefficients, while the significance of differences in precursor gas concentrations between day, night, weekday and weekend conditions were tested using a one-way ANNOVA test at a 95% confidence level using the package R version 4.0.2.

## 3. Results and Discussion

### 3.1. Environmental Variables: Temperature and Relative Humidity

Data for environmental variables shows that temperature and relative humidity maintained a relatively consistent diurnal pattern throughout the monitoring period (Fig. 2). While separate internal data for temperature and humidity was not measured, it is expected to be closely related to local meteorological conditions. The agitation hall is not climate controlled, but the air is constantly ventilated to maintain a negative pressure situation ensuring that the internal environment remains similar to prevailing local environmental conditions.



**Figure 2.** Consistent diurnal pattern of temperature and relative humidity throughout the monitoring period.

The average temperature during the monitoring period was 22.3°C, with a coldest temperature of 16.1°C degrees in the early morning of 17<sup>th</sup> June and an overall high of 34°C on the afternoon of 22<sup>nd</sup> June (Table 1). Daily minimum temperatures (mean = 18.3°C, standard deviation = 1.3°C) varied slightly less than Daily maximum temperatures (mean = 27.1°C, standard deviation = 2.9°C). As expected relative humidity showed an inverse pattern compared to temperature, with lower humidity during the day and higher humidity during cooler nights (Fig. 2). Relative humidity reached dew point (100% RH) most nights. Relative Humidity data attests to two periods of rain during the monitoring period from 23-26<sup>th</sup> June and 12-14<sup>th</sup> July. The environmental variable data shows that the main variation was diurnal, with no discernible trends over the whole monitoring period.

**Table 1.** Summary of average, minimum, maximum, and standard deviation values of temperature and relative humidity along with daily maximum and minimum averages.

	Temperature (°C)	Relative Humidity (%)
Average	22.3	82
Min.	16.1	32
Max.	34.1	100
St. Dev.	3.6	15.9
Daily Max. Average	27.1	98.4
St. Dev.	2.9	2.9
Daily Min. Average	18.3	60.2
St. Dev.	1.3	13.7

### 3.2. Gas Concentrations

#### 3.2.1. Ammonia monitoring results

NH<sub>3</sub> was the most abundant of the monitored gasses, with internal NH<sub>3</sub> concentrations averaging 57.1 ± 1.3 mg m<sup>-3</sup> across the entire monitoring period (excluding the two occasions when the extractor fans were disabled – these periods will be discussed separately). There was substantial variation in NH<sub>3</sub> concentrations depending on the timing of the measurements. The highest average

NH<sub>3</sub> concentrations were recorded during weekend day time at  $66.3 \pm 8.2 \text{ mg m}^{-3}$  (Table 2), although this was not significantly different ( $p < 0.05$ ) to average weekday day time concentrations of  $64 \pm 2.5 \text{ mg m}^{-3}$  (Fig. 3). Average weekday night time NH<sub>3</sub> concentrations were found to be slightly lower, at  $54.2 \pm 2.1 \text{ mg m}^{-3}$ , and statistically significantly different to day time concentrations. Weekend night time average NH<sub>3</sub> concentrations were measured to be less than 60% of average day time levels at  $37.9 \pm 2.6 \text{ mg m}^{-3}$ . The differences in average concentrations of NH<sub>3</sub> during separate time periods likely reflect differences in environmental variables (i.e. temperature and humidity) and manure agitation activity between night and day, and the absence of manure agitation activities on the weekend..

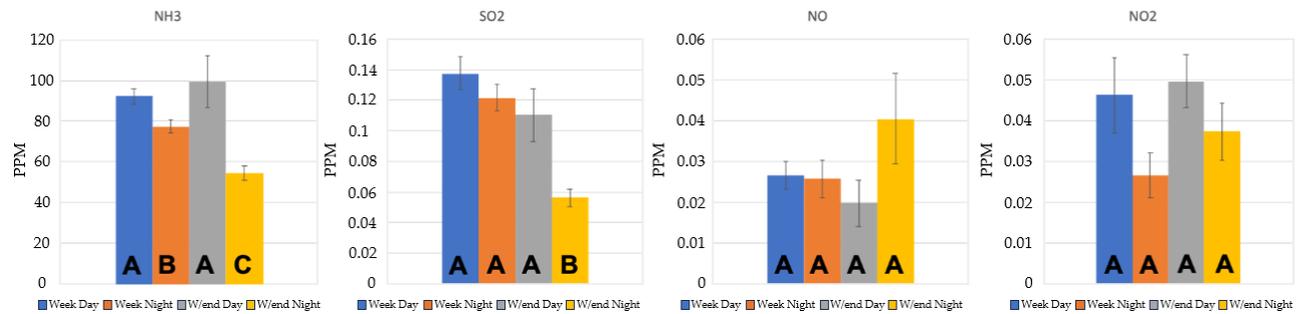
**Table 2.** Average internal and external concentrations of monitored SNA gasses and PM<sub>2.5</sub>.

		NH <sub>3</sub> Int	NH <sub>3</sub> Ext Hi	NH <sub>3</sub> Ext Lo	NO Fac	NO <sub>2</sub> Fac	SO <sub>2</sub> Fac	PM <sub>2.5</sub> Int	PM <sub>2.5</sub> Ext
		mg m <sup>-3</sup>			µg m <sup>-3</sup>				
Week Day	Mean	63.951	3.850	3.155	34.007	9.000	371.004	-	23.570
	Std.								
	Err.	2.530	0.070	0.366	4.052	1.846	30.452	-	3.832
Week Night	Mean	54.217	3.555	2.053	31.928	4.991	321.118	-	22.171
	Std.								
	Err.	2.074	0.084	0.092	5.421	1.052	23.671	-	3.277
W / end Day	Mean	66.278	3.793	2.079	27.939	10.332	264.985	-	22.337
	Std.								
	Err.	8.157	0.179	0.092	6.616	1.363	45.646	-	3.529
W / end Night	Mean	37.911	3.180	1.736	49.723	7.032	147.155	-	23.319
	Std.								
	Err.	2.558	0.112	0.056	13.629	1.328	15.669	-	2.644
Over all	Mean	57.063	3.642	2.425	33.350	7.276	307.224	36.901	23.713
	Std.								
	Err.	1.316	0.056	0.159	3.153	0.846	17.426	2.599	1.959

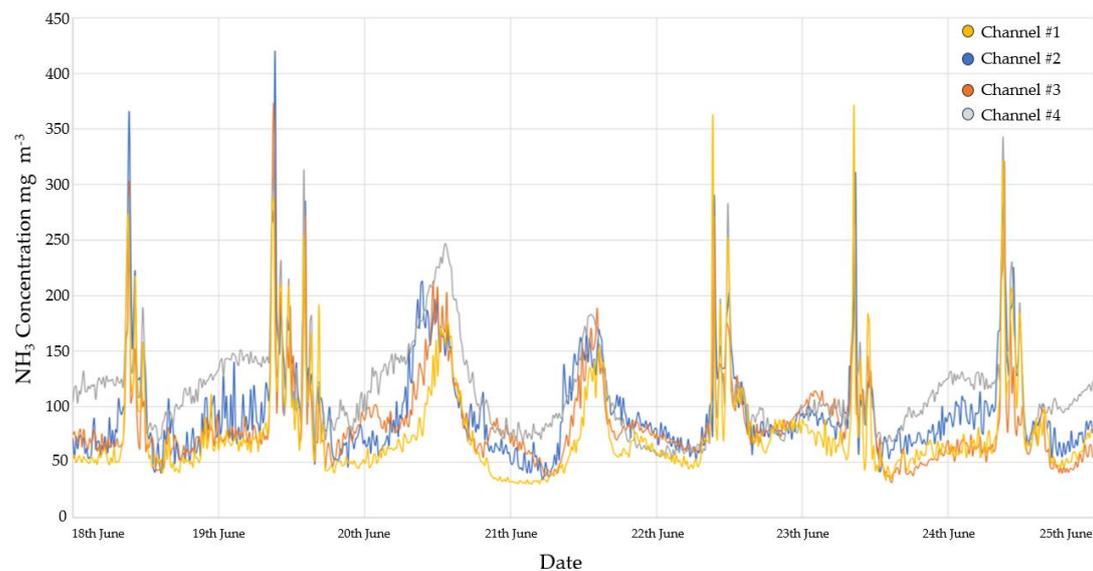
The diurnal fluctuations in NH<sub>3</sub> concentrations and the difference in concentration patterns between weekday and weekends can be clearly seen across all four internal NH<sub>3</sub> sampling points (Fig. 4). On weekdays, all internal sampling points show concentrations peaking sharply, three times, during the daily agitation events, matching the travel of the agitator along the three manure windrows (Fig. 1). On weekends a single broader NH<sub>3</sub> concentration peak is observed. It is likely that agitation itself results in shorter, higher magnitude NH<sub>3</sub> concentrations that are quickly cleared by the ventilation system, reducing peak NH<sub>3</sub> emissions outside of the agitation period. By contrast, on weekends there is a lower but broader NH<sub>3</sub> concentration peak during the day (Fig. 4). On weekdays, the lowest concentrations of NH<sub>3</sub> are usually recorded immediately after manure agitation has ceased, while on weekends the lowest concentrations of NH<sub>3</sub> are observed overnight, in the early morning prior to sunrise. The pattern of emissions on the weekend may suggest a greater role of temperature in NH<sub>3</sub> emissions at this time.

Turning of the manure during agitation cycles aerates the compost and provides a supply of oxygen to the microorganisms that are performing aerobic digestion [18]. The biological oxidation of C to CO<sub>2</sub> releases heat that raises the temperature of the compost into a thermophilic (>40°C) state [18]. In addition, aeration exposes the manure to air allowing the release of dissolved CO<sub>2</sub>, which raises the pH of the manure [10]. Increases in either temperature or pH promote the emission of NH<sub>3</sub> by increasing the dissociation of NH<sub>4</sub><sup>+</sup> [10,16,17]. The process of turning / aeration itself increases the ability for NH<sub>3</sub> volatilization through air turbulence across the greatly expanded manure surface area [10], leading to the high NH<sub>3</sub> concentrations recorded during agitation events. However, alongside volatilizing the reservoir of NH<sub>3</sub> gas in the manure, it also temporarily lowers the temperature of the

manure, likely leading to the lower  $\text{NH}_3$  concentrations that are recorded directly after agitation. Following aeration, the increased availability of oxygen leads to temperature increases within the compost over the next day [15]. Following aeration during the week, during the weekends, the temperature of the undisturbed manure likely remains higher leading to increased  $\text{NH}_3$  concentrations during the warmer day time and lower  $\text{NH}_3$  concentrations during the cooler night time temperatures.



**Figure 3.** Average concentrations and standard errors of measured pre-cursor gasses. The letters denote the results of a one-way ANNOVA test showing whether there is a significant difference in concentrations measured during separate periods: Weekday Day, Weekday Night, Weekend Day, and Weekend Night.

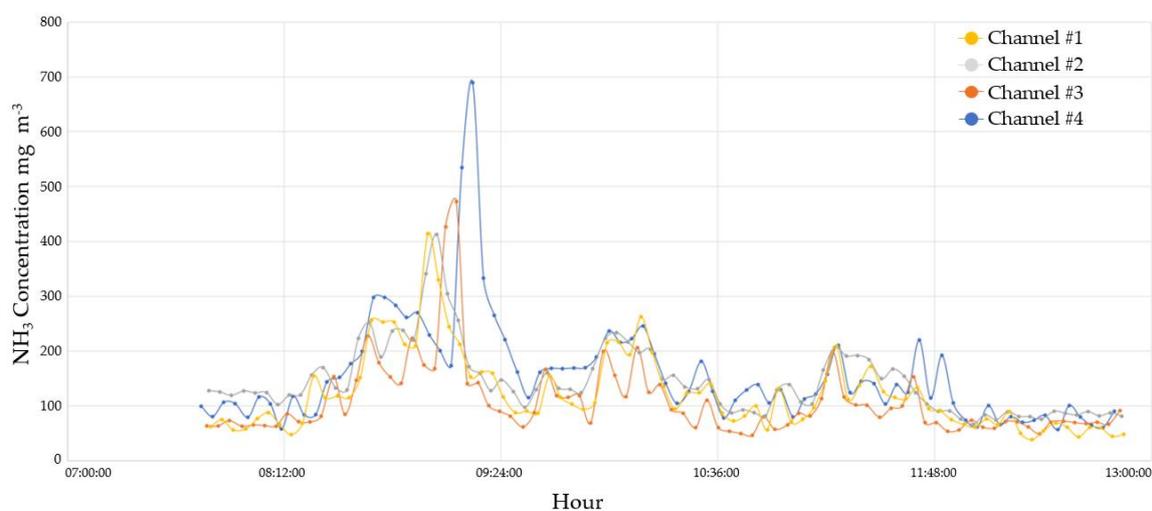


**Figure 4.** Concentration trends along with a weekend event (20-21, June, 2020) of single broader  $\text{NH}_3$  concentration peak during 1 week time series (18 June to 25 June, 2020).

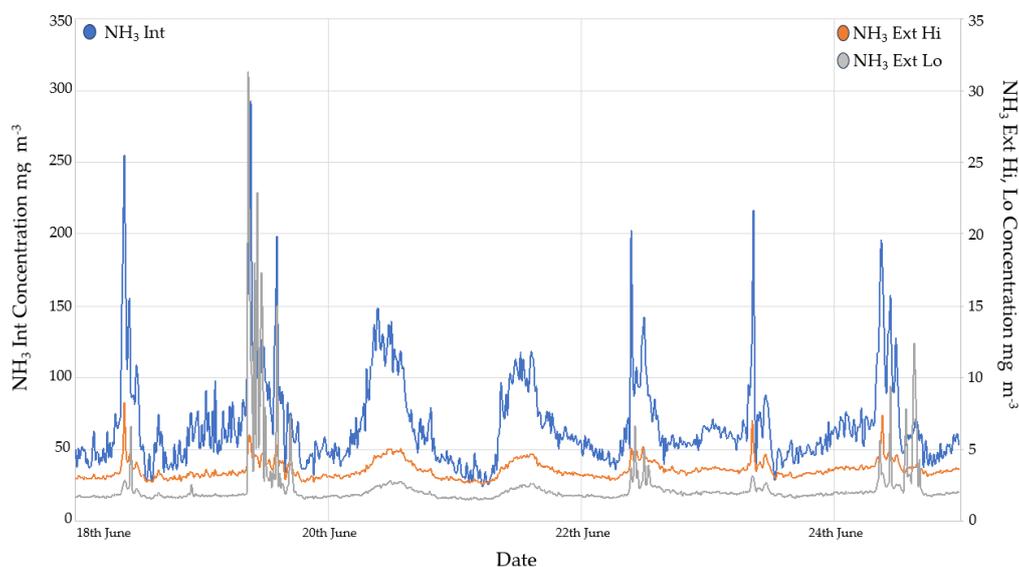
The agitation events can be clearly recognized in the raw  $\text{NH}_3$  concentration dataset for the internal sampling points (Fig. 5). Three periods of raised concentrations are apparent, representing the agitator travelling along each of the three windrows. The first period of raised concentrations with the highest peaks (at 275 – 500  $\text{mg m}^{-3}$ ) shows the agitation of the windrow closest to the internal sampling points, and distinctly displays sequential concentration peaks as the agitator passes each of the four sampling points.

External  $\text{NH}_3$  concentrations were generally an order of magnitude lower than internal concentrations. Average  $\text{NH}_3$  concentrations at 3 m height above ground level adjacent to the manure agitation hall window were  $3.64 \pm 0.06 \text{ mg m}^{-3}$  across the whole monitoring period, while average

external  $\text{NH}_3$  concentrations at ground level were slightly lower at  $2.43 \pm 0.16 \text{ mg m}^{-3}$  (Fig. 6; Table 2). There was no substantial diurnal / weekday – weekend variation in external  $\text{NH}_3$  concentrations at 3 m height, although at ground level weekday day  $\text{NH}_3$  concentrations ( $3.1 \text{ mg m}^{-3}$ ) were approximately 50% higher than during the other time periods ( $1.7 - 2.1 \text{ mg m}^{-3}$ ). Although the data shows that the  $\text{NH}_3$  emitted by the facility is low compared to internal concentrations, increases in external  $\text{NH}_3$  concentrations were detected contemporaneous to increases in internal concentrations, demonstrating that some leakage does occur. In addition, external  $\text{NH}_3$  concentrations at 3m height are comparable to average concentrations of  $3.7 - 5 \text{ mg m}^{-3}$  measured inside a Chinese commercial manure-belt layer house (Wang et al., 2020), and as such they are not negligible. It is expected that the external sampling point at 3m height would record higher average  $\text{NH}_3$  concentrations than the ground-level sampling point due to the potential for emissions through the window fitting. However, external ground-level  $\text{NH}_3$  concentrations, although lower on average, regularly displayed greater peak concentrations of  $\text{NH}_3$  during manure agitation activity (Fig. 6), although it is unclear why this might be the case.



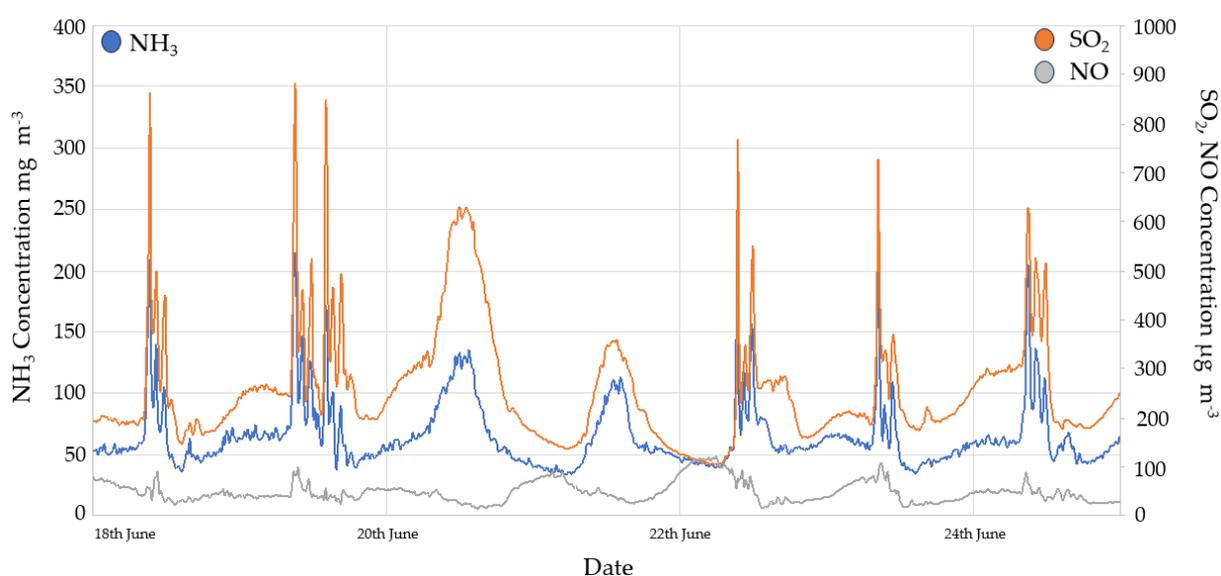
**Figure 5.** Spikes of  $\text{NH}_3$  trends followed by agitation practice from four sampling points (18 June 2022).



**Figure 6.** Comparison of average and peak  $\text{NH}_3$  concentrations inside the facility (internal) and outside at 3m height and ground level (external) adjacent to manure agitation hall.

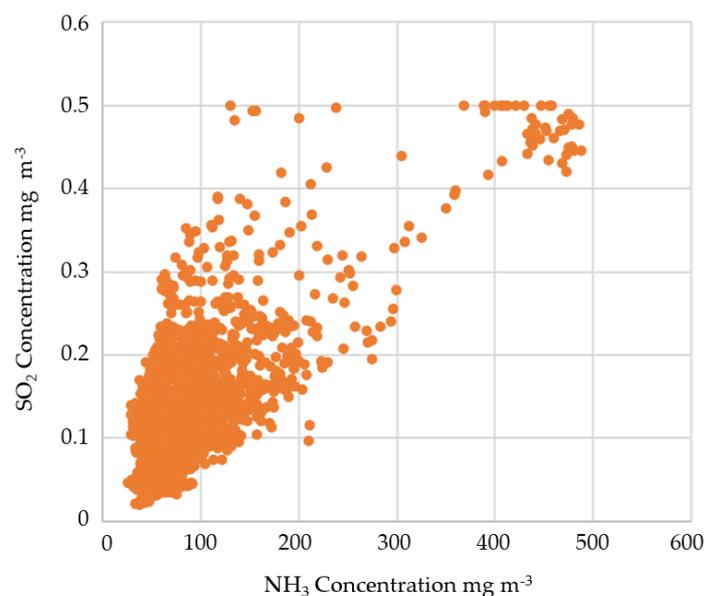
### 3.2.2. SO<sub>2</sub>

Recorded concentrations of SO<sub>2</sub> gas shared a similar pattern to NH<sub>3</sub> concentrations (Fig. 7), displaying high but variable concentrations during manure agitation and more broadly elevated concentrations during weekend day time when agitation did not occur. This similarity is reflected by a strong positive correlation ( $r = 0.751$ ,  $p < 0.001$ ) in the concentration of NH<sub>3</sub> and SO<sub>2</sub> (Fig. 8, Table 3). Although sharing a similar profile, SO<sub>2</sub> concentrations were consistently two orders of magnitude lower than NH<sub>3</sub> concentrations, averaging  $307.2 \pm 17.4 \mu\text{g m}^{-3}$  over the monitoring period. Unlike NH<sub>3</sub>, the average concentrations of SO<sub>2</sub> during weekday days ( $371 \pm 30.5 \mu\text{g m}^{-3}$ ) and nights ( $321.1 \pm 23.7 \mu\text{g m}^{-3}$ ) were not found to be significantly different ( $p < 0.05$ ). In addition, unlike NH<sub>3</sub>, average weekend day SO<sub>2</sub> concentrations ( $265 \pm 45.6 \mu\text{g m}^{-3}$ ) were lower than both weekday day and night concentrations, although were also not statistically different ( $p > 0.95$ ). However, average SO<sub>2</sub> concentrations on weekend nights ( $147.2 \pm 15.7 \mu\text{g m}^{-3}$ ) were found to be significantly lower ( $p < 0.05$ ) than SO<sub>2</sub> concentrations at all other times. During weekday manure agitation events SO<sub>2</sub> concentrations share a similar pattern with NH<sub>3</sub> of three main concentration spikes related to the sequential agitation of the three manure windrows. Weekday SO<sub>2</sub> concentrations were consistently measured to be lowest in the period directly after agitation activity ceased. However, again showing similarities to NH<sub>3</sub> concentration patterns, weekend day time concentrations of SO<sub>2</sub> were elevated for longer durations throughout the day and lowest concentrations occurred during the night, prior to sunrise.



**Figure 7.** Comparative analysis of SO<sub>2</sub>, NH<sub>3</sub>, and NO concentrations around a manure facility that showed patterns of emission peaks during agitation activities and weekend variability.

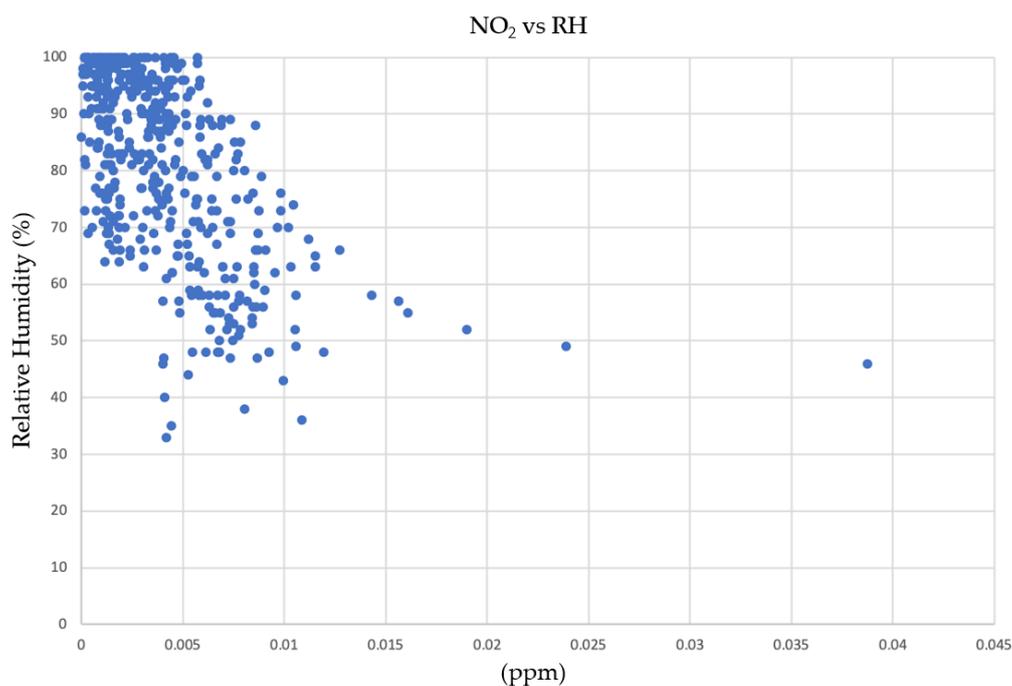
This data suggests that both NH<sub>3</sub> and SO<sub>2</sub> are emitted during the manure composting process, and the similarity in the pattern of recorded concentrations may suggest similar emission mechanisms. SO<sub>2</sub> can result from incomplete oxidation during manure breakdown by microorganisms activity, perhaps as a result of intermittently aerobic conditions [15]. However, unlike this study, research by Wang [29] found that indoor SO<sub>2</sub> concentrations at a belt-layer house Beijing were predominantly from the ambient air. While that study found a positive correlation between internal and regional SO<sub>2</sub> concentrations, data from this study rules out any substantial affect of ambient air on internal SO<sub>2</sub> concentrations at the PMCF (Table 3). Indeed, the strong positive correlation between NH<sub>3</sub> and SO<sub>2</sub> concentrations suggests that manure composting is the predominant source of both. Two existing studies both found that SO<sub>2</sub> is emitted during the manure composting process, however they produced widely different results for Sulphur mass loss ranging from <1% [30] to approximately 20% [15], and the SO<sub>2</sub> emission mechanism is not well understood.



**Figure 8.** Correlation analysis of  $\text{NH}_3$  and  $\text{SO}_2$  concentrations which is evidence of strong positive association during emission events.

### 3.2.3. $\text{NO}_x$

$\text{NO}_x$  are the total combination of  $\text{NO}$  and  $\text{NO}_2$ , which were both measured during this study. In this study  $\text{NO}$  was found to be the predominant component of  $\text{NO}_x$  (Table 2), with average concentrations ( $33.4 \pm 3.2 \mu\text{g m}^{-3}$ ) four and a half times greater than those of  $\text{NO}_2$  ( $7.3 \pm 0.8 \mu\text{g m}^{-3}$ ). Unfortunately,  $\text{NO}_x$  concentration data was contaminated after 4<sup>th</sup> July 2020, due to a build-up of condensation in the sampling tube, meaning that  $\text{NO}_x$  data for the final 12 days of the study has not been used. Weekday Day ( $34 \pm 4.1 \mu\text{g m}^{-3}$ ) and Night ( $32 \pm 5.4 \mu\text{g m}^{-3}$ ) concentrations of  $\text{NO}$  were similar, and weekend day concentrations were slightly lower ( $28 \pm 6.6 \mu\text{g m}^{-3}$ ). Average  $\text{NO}$  concentrations were greatest on weekend nights ( $49.7 \pm 13.6 \mu\text{g m}^{-3}$ ), however these difference were not found to be statistically significant ( $p < 0.05$ ). Similar to  $\text{NH}_3$  and  $\text{SO}_2$ ,  $\text{NO}$  concentrations recorded increases during manure agitation (Fig. 7), however during weekends  $\text{NO}$  concentrations appear to display an approximately inverse pattern to  $\text{NH}_3$  and  $\text{SO}_2$ . That is, when  $\text{NH}_3$  and  $\text{SO}_2$  concentrations are elevated during the weekend day time,  $\text{NO}$  concentrations are depleted, and this situation reverses during the weekend nights.  $\text{NO}$  is produced as a result of nitrification and denitrification processes during composting [15,29]. Similar to the increased concentrations of  $\text{NH}_3$  and  $\text{SO}_2$  during agitation,  $\text{NO}$  concentrations may be increased by manure turning by exposing more manure to the air, increasing diffusion potential [15]. The lower concentrations of  $\text{NO}$  during weekend days may be explained by the inhibition of nitrobacteria activity by high temperatures [15,17], with greater concentrations of  $\text{NO}$  overnight when temperatures are lower. Fillingham [16] reported an inverse relationship between  $\text{NH}_3$  emissions and  $\text{NO}$  emissions, with  $\text{NH}_3$  emissions greater in well aerated and higher temperature conditions, while  $\text{NO}$  emissions increased during manure storage. Previous research has found that  $\text{NO}$  emissions substantially increase only later in the composting process (beyond 15 – 30 days), after the thermophilic phase, when mesophilic nitrifying bacteria were able to grow [31,32].



**Figure 9.** Negative correlation with the relative humidity on NO<sub>2</sub> concentrations.

Unlike NO, concentrations of NO<sub>2</sub> display a pattern more similar to NH<sub>3</sub>, with elevated concentrations during weekday and weekend day time, and lower concentrations at night time (Table 2). Internal NO<sub>2</sub> concentrations were not correlated with ambient regional NO<sub>2</sub> concentrations (Table 3), suggesting that the main source of NO<sub>2</sub> in the facility was the composting manure. However, analysis showed that NO<sub>2</sub> concentrations had a strong negative correlation with Relative Humidity ( $r = -0.6$ ,  $p < 0.001$ ) (Fig. 9, Table 3), and a moderate positive correlation with temperature ( $r = 0.476$ ,  $p < 0.001$ ), although the latter likely only reflects the relationship between temperature and humidity. It is possible that greater humidity could cause the poor relationship between NO<sub>2</sub> and NO concentrations by providing water for the reaction from NO<sub>2</sub> to nitric acid and NO<sub>x</sub> (Eq. 2), and thus also explain the negative correlation with humidity.



**Table 3.** 15 Min / 1 hour Correlations Table - NH<sub>3</sub> Int, NH<sub>3</sub> Ext 1, NH<sub>3</sub> Ext 2, SO<sub>2</sub>, NO, NO<sub>2</sub>, Temp, RH, Beta-Ray PM, Paju PM.

	NH <sub>3</sub> Int	NH <sub>3</sub> Ext wi	NH <sub>3</sub> Ext gr	NO Fac	NO <sub>2</sub> Fac	SO <sub>2</sub> Fac	PM <sub>2.5</sub> Int	PM <sub>2.5</sub> Ext
Temp	0.101	0.086	0.071	-0.191	0.476	0.073	0.277	0.033
RH	-0.072	-0.012	-0.063	0.059	-0.600	-0.047	-0.322	0.078
PM <sub>2.5</sub> Paju	0.191	0.154	-0.139	0.072	0.096	0.053	0.341	0.666
NO <sub>2</sub> Paju				0.023	-0.025		0.084	0.412
SO <sub>2</sub> Paju						-0.204	-0.017	0.404
NH <sub>3</sub> Int								
NH <sub>3</sub> Ext wi	0.831							
NH <sub>3</sub> Ext gr	0.429	0.345						
NO Fac	0.191	0.140	0.107					
NO <sub>2</sub> Fac	-0.084	-0.139	-0.102	0.290				
SO <sub>2</sub> Fac	0.751	0.702	0.400	-0.008	-0.201			

\*=0.05

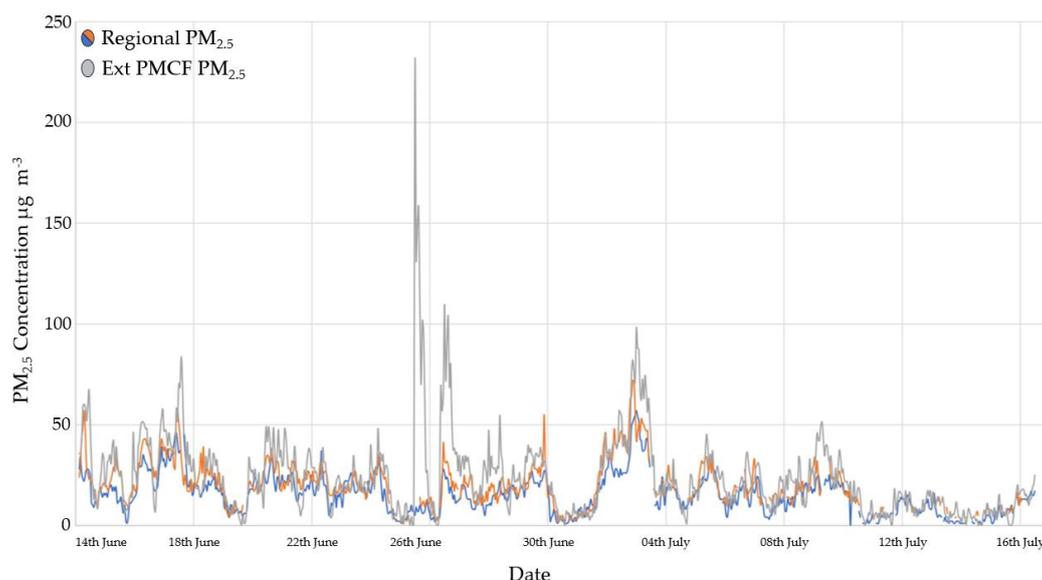
\*\*=0.01

\*\*\*=0.001

PM <sub>2.5</sub> Int	0.047	0.117	0.026	0.063	0.051	0.155	
PM <sub>2.5</sub> Ext	0.096	0.072	-0.110	-0.030	0.028	-0.065	0.088

### 3.3. Particulate Matter Concentrations

Average external PM<sub>2.5</sub> concentrations during the monitoring period were  $23.7 \pm 2 \mu\text{g m}^{-3}$  (Table 2), and unlike internal NH<sub>3</sub> concentrations, there was no significant variability between day-night and weekday-weekend periods. The hourly external PM<sub>2.5</sub> concentrations measured at the PMCF closely matched regional PM<sub>2.5</sub> concentrations (Fig. 10), with which they had a strong positive correlation ( $r = 0.67$ ,  $p < 0.001$ ).



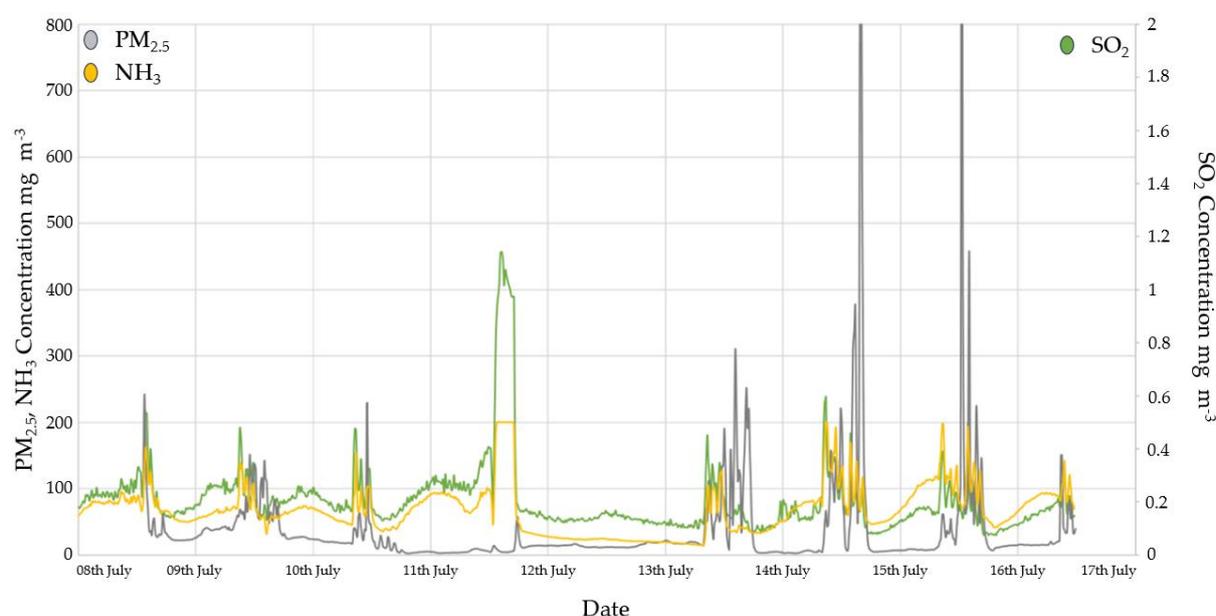
**Figure 10.** Comparison with PM<sub>2.5</sub> results between Beta-ray measurements and data from Paju weather station.

There were two exceptions, on the 25th and 26th June 2020, respectively, when external PMCF PM<sub>2.5</sub> concentrations greatly diverged from regional concentrations (Fig. 10). However, internal NH<sub>3</sub>, SO<sub>2</sub> and NO<sub>x</sub> concentration data on these two dates do not appear to show any large anomalies that might correspond with the high PM<sub>2.5</sub> concentrations. This suggests that the elevated PM<sub>2.5</sub> concentrations measured outside the facility on the 25th and 26th June did not originate from the manure composting function of the facility and were from a different source. Together with the high correlation with regional PM<sub>2.5</sub> concentrations, this data suggests that the PMCF is unlikely to be a significant local or regional source of PM<sub>2.5</sub>.

Average internal PM<sub>2.5</sub> concentrations were  $36.9 \pm 2.6 \mu\text{g m}^{-3}$ . Similar PM<sub>2.5</sub> concentrations ( $39 - 56 \mu\text{g m}^{-3}$ ) were found during monitoring of slatted floor hog houses in China [33], and are slightly lower than PM<sub>2.5</sub> concentrations ( $40 - 80 \mu\text{g m}^{-3}$ ) at a commercial laying hen house in Canada [34]. However, they are much lower than the PM<sub>2.5</sub> concentration ( $100 - 140 \mu\text{g m}^{-3}$ ) measured in Chinese manure-belt laying hen houses by Wang [29], and in a range of swine ( $75 - 177 \mu\text{g m}^{-3}$ ) and poultry houses ( $233 - 389 \mu\text{g m}^{-3}$ ) in the mid-western US [35]. Elevated periods of PM<sub>2.5</sub> concentrations appear to coincide with manure agitation (Fig. 11), and could be a result of mechanical lofting of fine fraction dust particles and / or secondary aerosol formation from elevated precursor gas concentrations. Secondary aerosol formation can occur when NH<sub>3</sub> gas reacts with acidic species SO<sub>2</sub> and NO<sub>x</sub> to form NH<sub>4</sub><sup>+</sup> salts like (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>NO<sub>3</sub> [7,19]. Reaction between NH<sub>3</sub> and sulfuric acids will occur preferentially over nitric acids under normal atmospheric conditions [19], and the reaction rate between NH<sub>3</sub> and sulfuric acids is faster than for nitric acids [20]. Peak PM<sub>2.5</sub> concentrations were recorded at  $889 \mu\text{g m}^{-3}$  and  $1479 \mu\text{g m}^{-3}$  on the 14th and 15th July 2020 respectively (Fig. 11), showing that very high internal PM<sub>2.5</sub> concentrations occasionally occur for brief durations coincident with

agitation activity. However,  $PM_{2.5}$  concentrations rarely rose above  $200 \mu\text{g m}^{-3}$  and it is likely that the high-volume ventilation system at the PMCF was an important factor in the relatively low internal  $PM_{2.5}$  concentrations, removing fine-fraction dust particles and precursor gasses before  $PM_{2.5}$  formation was able to occur.

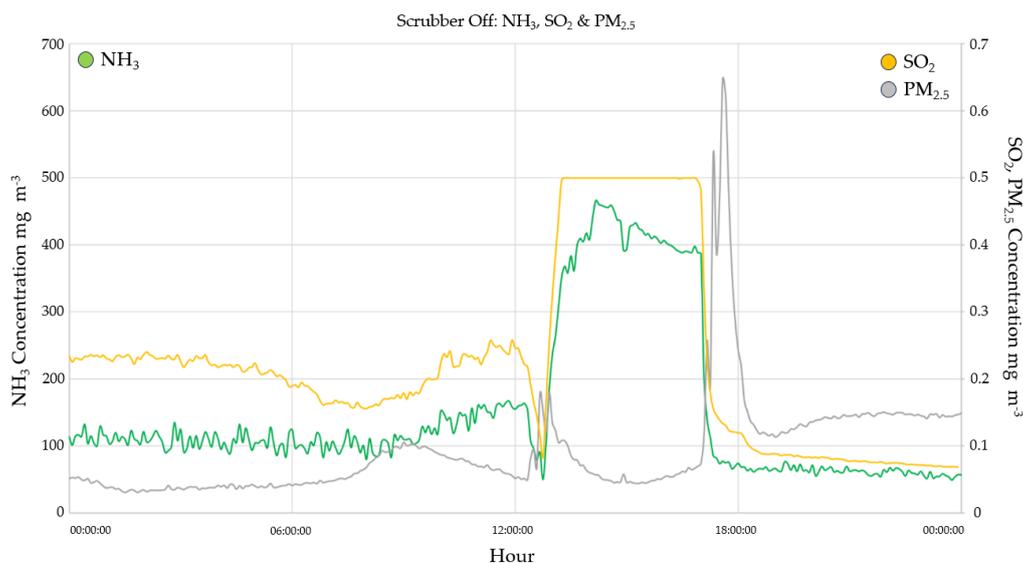
Due to the shorter duration (approx. 10 days) over which internal  $PM_{2.5}$  was measured it was not possible to assess statistical differences between weekend and weekday concentrations. Although internal  $PM_{2.5}$  was observed to reach much higher concentrations on weekday days following manure agitation, concentrations appeared to show comparatively little variability on weekends. A weak but significant correlation was found between regional and internal  $PM_{2.5}$  concentrations ( $r = 0.341$ ,  $p < 0.001$ ), suggesting that  $PM_{2.5}$  in the ambient air was responsible for a portion of internal  $PM_{2.5}$  concentrations, however no correlation existed between external and internal  $PM_{2.5}$  (Table 3).



**Figure 11.** Trailing internal concentrations of  $PM_{2.5}$  along with  $NH_3$  and  $SO_2$ .

### 3.4. Ammonia Emission Rate

An experiment was undertaken during a 5-hour period between 12:30 – 17:30 on Saturday 11<sup>th</sup> July 2020 in which the high-volume ventilation / scrubber system was turned off in order to measure the flux of  $NH_3$  from the composting manure (Fig. 12). Prior to the experiment, under ventilated conditions, internal  $NH_3$  concentrations were stable at approximately  $100 \text{ mg m}^{-3}$ , rising to around  $150 \text{ mg m}^{-3}$  as all entrances and exits to the facility were closed. After the ventilation system was disabled,  $NH_3$  concentrations were recorded to drop to an average of  $\sim 30 \text{ mg m}^{-3}$  initially, before rising quickly to reach values of  $\sim 250 \text{ mg m}^{-3}$  within 30 minutes. Peak concentrations of  $\sim 325 \text{ mg m}^{-3}$  were reached after approximately 40 minutes. Concentrations of  $SO_2$  followed a similar pattern to  $NH_3$ , initially dropping before exceeding the monitoring instrument's detection limit at  $1.31 \text{ mg m}^{-3}$  (500 ppb). It is not clear why  $NH_3$  concentrations dropped after the ventilation system was disabled, but we hypothesise that the negative pressure environment created by the ventilation system causes a greater flux of  $NH_3$  from the manure surface due to the pressure gradient, and therefore when the ventilation system is disabled,  $NH_3$  is emitted at a reduced rate. Alternatively, the decrease could be related to lower air turbulence across the manure surface which would be expected to reduce convective mass transfer of  $NH_3$  from the manure surface to the air [10].



**Figure 12.** Emission flux response to high volume ventilation and scrubber system off during a 5-hour period between 12:30 – 17:30 on Saturday 11 July 2020.

The emission rate of  $\text{NH}_3$  was calculated according to Equation 1, and based on the time taken for  $\text{NH}_3$  concentrations to rise from their minima at  $\sim 30 \text{ mg m}^{-3}$  to the inflection point at  $\sim 250 \text{ mg m}^{-3}$  after which concentration increases slow down. The results show that under stable non-ventilated conditions  $\text{NH}_3$  emission rate inside the manure agitation hall is between  $6.16 - 8.02$  (mean =  $7.28 \pm 0.41$ )  $\text{NH}_3\text{-N mg min}^{-1} \text{ m}^{-3}$  (Table 4), and that extrapolated, the total mass of  $\text{NH}_3$  emitted daily is between  $20.7 - 26.96$  (mean =  $24.47 \pm 1.39$ )  $\text{NH}_3\text{-N kg d}^{-1}$ .

**Table 4.**  $\text{NH}_3$  emission rates inside the Paju Manure Composting Facility under non-ventilated conditions.

	Emission Rate		Scaled Emission Rates	
	$\text{mg min}^{-1} \text{ m}^{-3}$	$\text{g h}^{-1} \text{ m}^{-2} \text{ manure}$	$\text{g h}^{-1} \text{ m}^{-3} \text{ manure}$	$\text{g kg}^{-1} \text{ manure}^*$
Mean	7.28	2.77	1.85	1.52
Std. Error	0.41	0.16	0.1	0.09

\* over entire 18 day composting period

There is little standardisation of the units used to report  $\text{NH}_3$  emission rates across various studies. The data from this study is compared to that of other studies in the unit in which those studies published their data. The scaled  $\text{NH}_3$  emission rates in the PMCF are high compared to studies investigating emissions from AFOs. Hristov [10] present a compilation of data from multiple dairy farms and beef feed lots ( $n=29$ ), where all but one study found  $\text{NH}_3$  emissions between  $0.01 - 0.9 \text{ g h}^{-1} \text{ m}^{-2}$ , compared to  $2.77 \pm 0.41 \text{ g h}^{-1} \text{ m}^{-2} \text{ NH}_3\text{-N}$  in this study. Daily  $\text{NH}_3$  emissions from a composting facility at a manure belt poultry house were estimated to be much greater at  $596 \text{ kg d}^{-1}$  [5], approximately 20 times higher than this study. However, Zhao [5] also relay data from conference proceedings presented by Matsusada [36] reporting  $\text{NH}_3$  emissions of  $0.32 - 2.84 \text{ g kg}^{-1}$ ,  $0.02 - 2.74 \text{ g kg}^{-1}$ , and  $0.04 - 0.46 \text{ g kg}^{-1}$ , for composting of swine, poultry and dairy manure respectively. These figures are comparable to  $\text{NH}_3$  emissions of  $1.52 \pm 0.09 \text{ g kg}^{-1}$  from the mixed manure at the PMCF. A separate study of cattle manure composting reported  $\text{NH}_3$  emissions of  $0.07 - 0.19 \text{ g d}^{-1} \text{ kg}^{-1}$  during the first 50 days [37], again similar to a figure of  $0.084 \pm 0.005 \text{ g d}^{-1} \text{ kg}^{-1}$  for this study. As expected, the  $\text{NH}_3$  emission rate at the PMCF is more similar to measurements from other manure composting operations than from AFOs.

Internal PM<sub>2.5</sub> concentrations were also measured during the ventilation-off experiment, and were found to initially rise from approximately 3 – 10 µg m<sup>-3</sup> under ventilated conditions to 18 µg m<sup>-3</sup> when the ventilation system was disengaged before decreasing again to ~5 µg m<sup>-3</sup> during the experiment. At the end of the experiment, when the ventilation system was re-engaged PM<sub>2.5</sub> concentrations peaked at 65 µg m<sup>-3</sup>. It was expected that the high concentrations of NH<sub>3</sub> and SO<sub>2</sub> during the ventilation-off experiment would lead to secondary inorganic particulate formation and higher PM<sub>2.5</sub> concentrations. The presence of PM<sub>2.5</sub> concentration peaks at the beginning and end of the experiment, but not during it when NH<sub>3</sub> and SO<sub>2</sub> concentrations were highest, suggests that the processes affecting PM<sub>2.5</sub> formation are more complex than the co-existence of precursor gasses alone. This could be a productive area for future research.

#### 4. Conclusions

The concentrations of SNA precursor gasses and PM<sub>2.5</sub> were continuously measured at a modern manure composting facility in Paju, South Korea over a period of 35 days in June and July 2021. Average internal NH<sub>3</sub> concentrations were 57.1 ± 1.3 mg m<sup>-3</sup>, compared to external NH<sub>3</sub> concentrations of 3.64 ± 0.06 mg m<sup>-3</sup> at 3m-height, and 2.43 ± 0.16 mg m<sup>-3</sup> at ground level. Internal concentrations of NH<sub>3</sub> were found to differ significantly between day time (higher) and night time (lower), and weekend nights had concentrations that were significantly lower than weekday nights. These concentration differences are likely caused by manure agitation activity, which occurs during the weekday day time, and temperature differences between day and night. Sulfur Dioxide (SO<sub>2</sub>) concentrations followed a similar pattern with NH<sub>3</sub>, and it was found to be the second most abundant precursor gas, with average concentrations of 307.2 ± 17.4 µg m<sup>-3</sup>. Weekend night time concentrations of SO<sub>2</sub> were found to be significantly lower than at all other times. The predominant component of NO<sub>x</sub> gasses, was determined to be NO with average concentrations of 33.4 ± 3.2 µg m<sup>-3</sup>, compared to NO<sub>2</sub> concentrations of 7.3 ± 0.8 µg m<sup>-3</sup>. Although there was no significant difference between NO<sub>x</sub> concentrations at different times, the NO<sub>x</sub> concentration profile was markedly different to both NH<sub>3</sub> and SO<sub>2</sub>, suggesting that the processes governing emission are different. Average PM<sub>2.5</sub> concentrations inside the facility (36.9 ± 2.6 µg m<sup>-3</sup>), were about 50% higher than external concentrations (23.7 ± 2 µg m<sup>-3</sup>), and a moderate correlation (r = 0.341, p < 0.001) between the two is suggestive that ambient PM<sub>2.5</sub> contributes part of the internally measured concentrations. The NH<sub>3</sub> emission rate from the facility's manure agitation hall was calculated to be 7.28 ± 0.41 NH<sub>3</sub>-N mg min<sup>-1</sup> m<sup>-3</sup>, corresponding to scaled emissions rates of 1.85 ± 0.1 NH<sub>3</sub>-N g h<sup>-1</sup> m<sup>-3</sup> of manure compost, or 1.52 ± 0.09 NH<sub>3</sub>-N g kg<sup>-1</sup> of manure compost over the full 18-day composting period. Based on these figures, total daily NH<sub>3</sub> flux from the manure agitation hall is approximately 24.47 ± 1.39 NH<sub>3</sub>-N kg d<sup>-1</sup>. The data from this study suggests that despite the large fluxes of NH<sub>3</sub> and other precursor gasses emitted inside the manure composting facility, the sealed building design with a negative pressure high-volume ventilation / scrubber system is effective at minimizing NH<sub>3</sub> and PM<sub>2.5</sub> emissions to the atmosphere.

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