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Characteristics of Ambient Ammonia Levels Measured in Three Different Industrial Parks in Southern Taiwan

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ABSTRACT

To characterize concentrations of atmospheric ammonia (NH_3) at industrial parks in southern Taiwan, three representative industrial parks and two background locations were selected for this study. The sampling process for gaseous ammonia was performed in accordance with Taiwan EPA NIEA A426.71B. The results revealed that the mean NH_3 levels ranged from 70.5 to 153.9 ppb (mean = 100.2 ppb) in Neipu Industrial Park. Both highest mean levels in Neipu Industrial Park during 12:00–13:00 in the daytime and during 20:00–21:00 at night were about 50–150 times greater than ambient background levels. For the whole day, the mean NH_3 levels ranged from 43.0 to 114.6 ppb (mean = 72.8 ppb) in Pingtung Industrial Park. In Pingnan Industrial Park, the mean NH_3 levels for the whole day ranged from 45.0 to 122.6 ppb (mean = 84.9 ppb). Moreover, the ratio values (Rs) were categorized according to the cover-range (from minimum ratios to maximum ratios) at different sampling periods and the minimum separated boundary lines. The results indicate that the characteristics of ratio values normalized by the background levels can be divided into three main categories in southern Taiwan: (1) R > 200; (2) R > 60; (3) R > 10. Our research data reveal obvious reasons for this clear distinction of ambient NH_3 levels in southern Taiwan. Significantly, the results reveal that agricultural NH_3 emissions influence local ambient concentrations of NH_3.

Keywords: Ambient; Industrial park; Ratio; Ammonia; Background.

INTRODUCTION

Emission of ammonia (NH_3) is of concern because of its direct contribution to secondary photo- or smog reactions of air pollutants, and for its adverse influences on various ecosystems, such as bodies of water and soil. NH_3 may be either wet- or dry deposited as a gas, or react with sulfuric (H_2SO_4), nitric (HNO_3), and hydrochloric (HCl) acids to form ammonium sulfate $(NH_4)_2SO_4$, bisulfate (NH_4HSO_4), nitrate (NH_4NO_3), and chloride (NH_4Cl) aerosols, thereby contributing to inorganic ambient particulate matter (Walker *et al*., 2004). Like in many other countries, in Taiwan ammonia gas (NH_3) is defined as a toxic pollutant under the EPA's Air Pollution Control Act Enforcement Rules (promulgated by Environmental Protection Administration order on July 23, 2003) (EPA Act, 2010). Therefore, considerable attention is paid to emission of NH_3 in Taiwan's industrial parks in order to maintain air quality and reduce pollution.

Many different ammonia sources or sites have been studied to understand their harmful effects on air quality. Bouwman *et al*. (1997) reported a global high resolution emission inventory for ammonia. Their study indicates that domestic animals are the largest source (22 Tg N/yr, 1 Tg = 10^{12} g) of atmospheric NH_3, comprising approximately 40% of natural and anthropogenic emissions combined, while synthetic fertilizers and agricultural crops together contribute an additional 12.6 Tg NH_3-N/yr (23% of total emissions) (Bouwman *et al*., 1997).

Carmichael *et al*. (2003) reported their measurements of ammonia concentrations in Asia, Africa, and South America using passive samplers. Their monthly measurements were obtained at 50 stations in Asia, Africa, South America, and Europe. Their results indicated that median ammonia concentrations range from 20 ppb at Dhangadi, India, to 0.1 ppb at nine other stations. At 27 of the regional stations, the ambient ammonia levels were found to exceed 1 ppb.

Walker *et al*., (2004) presented one year of ambient ammonia (NH_3) and other ions' concentrations at three sites in the Coastal Plain region of North Carolina. The

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three sites, Clinton, Kinston, and Morehead City, are all located in counties with total NH_3 emission densities of 4800, 2280, and 320 kg NH_3-N/km^2-yr, respectively (Walker *et al*., 2004). Their study found that the average NH_3 concentrations were 5.32, 2.46, and 0.58 μg/m^3 at Clinton, Kinston, and Morehead City, respectively. In addition, their results showed that NH_3 concentrations were highest during the summer at all sites, with summer-to-winter concentration ratios of 2.40, 5.70, and 1.70 at Clinton, Kinston, and Morehead City, respectively. They also indicated that NH_3 concentrations were higher at night at the Clinton site, during the day at the Kinston site, and day vs. night concentrations were similar at the Morehead City site (Walker *et al*., 2004). To sum up, this earlier study demonstrated that agricultural NH_3 emissions influence local ambient concentrations of NH_3.

Wilson and Serre (2007a) reported that the examination of atmospheric ammonia levels near hog concentrated animal feeding operations (namely, CAFOs), homes, and schools in eastern North Carolina. The data for each phase of sampling was stratified by distance from the nearest hog CAFO. The mean Phase I levels were 16, 8, 7 and 5 ppb for distances < 0.5, 0.5–1, 1–2, and 2 km or more, respectively. The mean levels for Phase II were 29, 16, and 11 ppb for distances < 0.5, 0.5–1, and 1 km or more, respectively (Wilson and Serre, 2007a). The results of the distance stratification provide the strongest evidence that distance from one or more CAFOs is the key variable in controlling weekly NH_3 atmospheric concentration at the community level in Eastern NC (Wilson and Serre, 2007a).

In fact, agriculture and livestock activity contribute significantly to anthropogenic ammonia (NH_3) emission to air, the latter ranging from 2.8 to 5.2 Tg (based on N-NH_3) per year in Western Europe (ECETOC, 1994). Balsari *et al*. (2007) reported that their experimental data highlighted a strong effect of both slurry temperature and air velocity over the slurry surface on NH_3 emissions. However, they also indicated that the funnel system allowed a comparison of emissions from different sources (cattle and pig slurry) under different temperature regimes, but, due to the very low wind speed under the funnels, did not allow data to be obtained in a way that would be comparable to real environmental conditions (Balsari *et al*., 2007). Therefore, it is important that real sampling be performed and that it be undertaken close to the investigated target.

Over the past decade, NH_3 has been intensively studied in many countries (McCulloch *et al*., 1998; Aneja *et al*., 2000; Walker *et al*., 2000a, 2000b; Aneja *et al*., 2001; Pryor *et al*., 2001; Rattray *et al*., 2001; Robarge *et al*., 2002; Sather *et al*., 2008; Cao *et al*., 2009). However, research into NH_3 in different environmental matrices is still at an early stage in southern Taiwan. Therefore, a detailed and thorough environmental analysis of ammonia is needed to set a baseline for future research in Taiwan. In particular, two distinctive national features call for such an investigation to be conducted. Firstly, in southern Taiwan, many public disputes derive from malodorous toxic pollutants, such as ammonia gas. Moreover, ammonia gas is defined as a toxic pollutant in Taiwan EPA's Air Pollution Control Act Enforcement Rules (promulgated by Environmental Protection Administration order on July 23, 2003). In addition, according to Taiwan EPA's Air Pollution Control Act Enforcement Rules, Article 45 "If an air pollution injury incident in Article 80 of this Act overlaps two special municipalities, counties, or cities, or any combinations thereof, the victims may apply to the central competent authority for appraisal of the reason for their injuries." Therefore, a comprehensive environmental analysis of ammonia is required to improve the EPA's database that contains the characteristics of ambient ammonia levels measured in three different industrial parks in the Kao-Ping Area of Taiwan. Secondly, many unregistered farmyards raise hogs or fowl in southern Taiwan, and thus the lack of knowledge with regard to atmospheric NH_3 at the community level is an important problem, as without it is impossible to develop accurate exposure profiles for residents who live near such farmyards, making it difficult to adequately address any negative health responses in the exposed population.

One of the major preoccupations of environment researchers in southern Taiwan over the past decade has been investigating the characteristics of air-borne toxins. Building on this, the main objective of this study is to measure and characterize the concentrations of NH_3 during industrial activity in southern Taiwan. In addition, meteorological conditions, such as wind speed, wind direction, and temperature, were also measured during the sampling period. These results are expected to generate useful information with regard to evaluating the health effects of NH_3.

MATERIALS AND METHODS

Sampling Strategy

Southern Taiwan is located on the lee side of the central mountain range of Taiwan. This region has a stable atmospheric, clear skies and a distinct pattern of diurnal land-sea breeze circulations (Hsieh *et al*., 2005). To examine the spatial distribution of NH_3 in different industrial parks, five locations in southern Taiwan were chosen for NH_3 monitoring, three industrial locations plus two reference ones. Firstly, three industrial park sites (namely Neipu, Pingtung, and Pingnan Industrial Parks) were chosen based on both their different industrial and manufacturing characteristics. Secondly, two reference sampling sites near Lao-Pi village, NPUST dormitory (as a background site #1) and a bamboo grove (as a background site #2), were also chosen because there are no registered industrial plants nearby and they are also far from any other registered industrial or agriculture sources. More detailed descriptions of the five sampling locations are presented below.

Neipu Industrial Park: This is an industrial park with a very low population and traffic density. The total area of the park is approximately 99 ha, and the park contains a total of 61 factories, with the buildings few and widely scattered. The types of industry in the park can be divided into three categories: (1) food processing, (2) food manufacturing, and (3) manufacturing electrical appliances (Hsieh *et al*., 2005; PCIC, 2010).

Pingtung Industrial Park: This is an important industrial park near Pingtung City's main commercial area. This park has high traffic density and there are numerous high rise commercial buildings located nearby. The total area of the park is approximately 113.2 ha, and 146 factories are located within it. The types of industry in this park can be divided into three different categories: (1) machinery manufacturing, (2) metalworking, and (3) food manufacturing (Hsieh *et al*., 2005; PCIC, 2010).

Pingnan Industrial Park: This is a newly developed area which has slowly industrialized over recent years, close to both Fangliao and Donggang. The area contains a mixture of old low-rise residential buildings and newly built low-rise factory buildings. The total area of the park is approximately 278 ha, and the park contains some 97 factories. The park has two major types of industry: (1) iron and steel and (2) refrigerator manufacturing (Hsieh *et al*., 2005; PCIC, 2010).

NPUST Dormitory: The National Pingtung University of Science and Technology (NPUST) campus is situated in near Lao-Pi village in Ping-Tung County, with minimal anthrogenic pollution and adverse environmental impact. In this study, NPUST dormitory is considered as background monitoring station #1. In general, motor vehicles in the parking lots around the dormitory are the only potential sources of ammonia emission.

A Bamboo Grove: The bamboo grove is within an area of primeval forests near Lao-Pi village, and driving is strictly forbidden in this area. In this study, the bamboo grove is considered as background monitoring station #2. The major difference between background monitoring stations #1 and #2 is the amount of traffic they experience.

Sampling and Instruments

Fig. 1 displays the locations of five ammonia sampling sites. Field sampling was conducted on two consecutive days per week from September 2003 to December 2004, with rainy days excluded. On each sampling day, samplers were deployed for 14–15 hours at sites at Neipu Industrial Park (the number of samples n, n = 120), Pingtung Industrial Park (n = 120), Pingnan Industrial Park (n = 112), NPUST dormitory (n = 120) and the bamboo grove (n =120). The sampling system operated on 09:00–24:00 daily-cycle at all the sampling sites, with the exception of the site at Pingnan Industrial Park, which was on a 10:00–24:00 daily-cycle. According to EPA NIEA A426.71B (Taiwan EPA, 1997), ambient air should into a sampling duct by using a pump at flow rates under 5 L/min. For NH_3 sampling, the ambient air flows through a pre-filter, a front-impinger bottle, a rear-impinger bottle, and a dehumidifier bottle in sequence. During each sampling hour, each impinger bottle contains ~25 mL of 0.1 N H_2SO_4. In the field, after starting the pump the ambient air flows through a pre-filter, then the both particle-free and gaseous NH_3 enters the two impinger bottles (including a front-impinger bottle and a rear-impinger bottle) and NH_3 is absorbed in 0.1 N H_2SO_4 solution. NH_3 concentration is a summation of the levels in the front and rear impinger

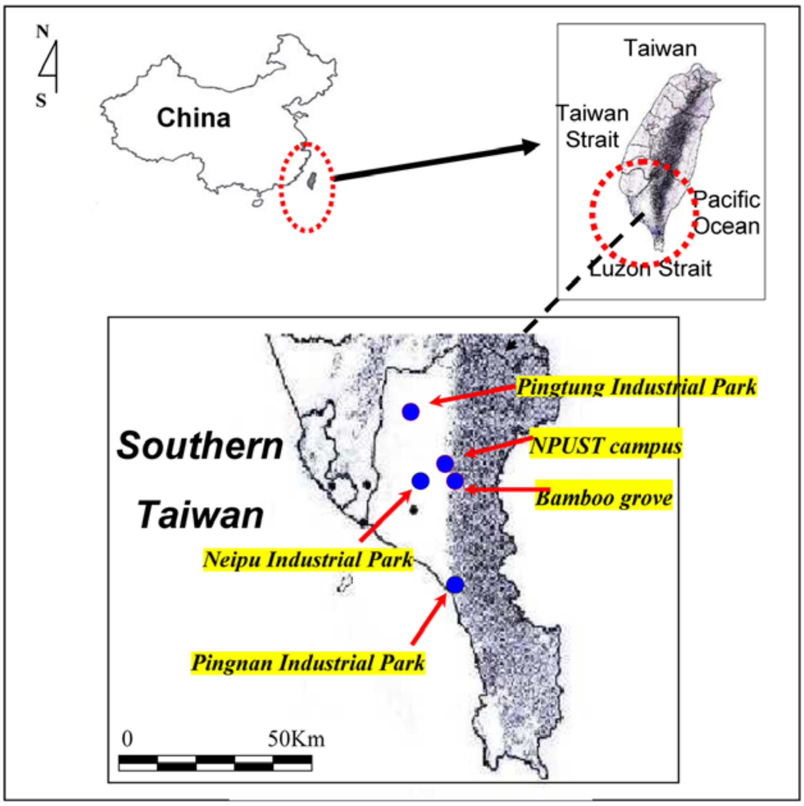

Fig. 1. Sampling locations for ammonia in southern Taiwan.

bottles. In this study, SKC pump samplers were operated at flow rate of 2 L/min for a duration of 1 hr, and then for another 1 hr duration after the replacement of two impinger bottles. Each hour we gathered two samples (by using two sampling instruments side by side) and averaged them to yield 1-h average concentrations for all runs.

The collected NH_3 samples were stored in airtight boxes at 0°C and later analyzed in our EP207 laboratory by the Indophenol-UV Method (Taiwan EPA, 1997). All the chemicals used in this study were of analytical reagent grade, and water purified with a Milli Q Labo (Millipore) was used for the preparation of all solutions. The reagents were added and the mixtures were allowed to react at room temperature (about 25°C) for 30 minutes, after which the absorbance at 630 nm of each sample was measured repeatedly using an UV spectrophotometer (Type: Hitachi 2100). The calculation of the actual ammonia concentrations in the samples was done using a calibration curve based on an ammonia standard solution.

RESULTS AND DISCUSSION

Ambient Ammonia Concentrations in Different Industrial Parks

Fig. 2 presents the daily variation in both meteorological parameters records (Fig. 2(a)–2(d)) and the average ambient NH_3 concentrations (Fig. 2(e)) in Neipu Industrial Park in southern Taiwan. For the Neipu Industrial Park, the mean relative humidity level for the nighttime was approximately 1.5 times higher than the mean level for the daytime (Fig. 2(a)). Fig. 2(b) shows the mean wind speed level for the daytime was approximately 2.7 times higher than the mean level for the nighttime. Fig. 2(d) shows the mean ambient temperature level for the daytime was approximately 1.3 times higher than the mean level for the nighttime. For increased accuracy, we use the value of measured sunlight strength (Fig. 2(e)) to distinguish between the day and night. If the measured sunlight strength is more than zero, then the sampling period is part of the daytime, and otherwise is part of the nighttime. Therefore, from the point of view of the whole day, the mean NH_3 levels ranged from 70.5 to 153.9 ppb (mean = 100.2 ppb) in Neipu Industrial Park (Fig. 2(e)).

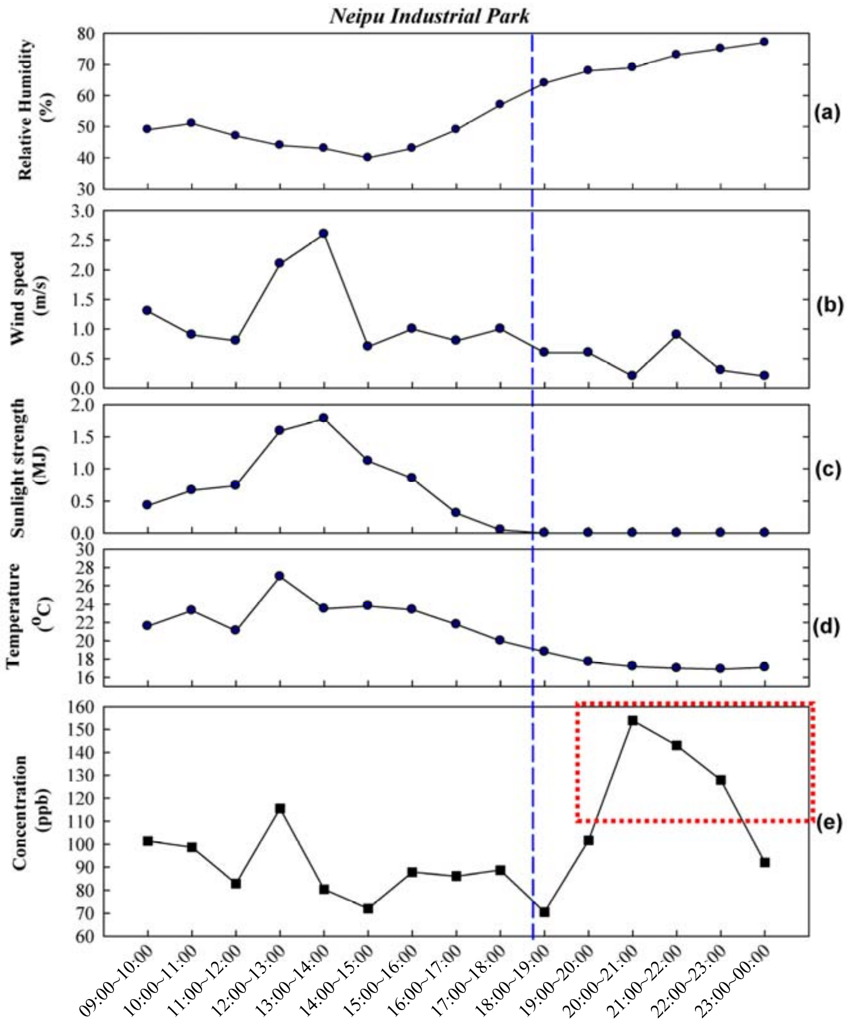

Fig. 2. Daily variation in both meteorological parameter records (from (a) to (d)) and average ambient ammonia concentrations (e) in Neipu Industrial Park in southern Taiwan.

Fig. 2 shows that the mean NH_3 levels ranged from 72.0 to 115.6 ppb (mean = 90.4 ppb) and from 70.5 to 153.9 ppb (mean = 114.9 ppb) in the day and at night, respectively. For all the sampling duration, the highest mean level of 153.9 ppb was obtained from 20:00 to 21:00. Regarding the maximum level, the highest mean levels of 115.6 and 153.9 ppb were obtained from 12:00 to 13:00 and 20:00 to 21:00, respectively. Both these highest mean levels were about 50–150 times greater than the ambient background levels (1–3 ppb) measured in several previous reports (Sickles *et al*., 1990; Aneja *et al*., 1998; Leaderer *et al*., 1999; Lefer *et al*., 1999; McCurdy *et al*., 1999; Walker *et al*, 2004; Wilson and Serre, 2007a, 2007b).

For the whole day, the mean NH_3 levels ranged from 43.0 to 114.6 ppb (mean = 72.8 ppb) in Pingtung Industrial Park (Fig. 3). In addition, the highest mean level of 114.6 ppb was obtained between 09:00 and 10:00. Moreover, the mean NH_3 levels ranged from 67.3 to 114.6 ppb (mean = 88.5 ppb) and from 43.0 to 59.3 ppb (mean = 49.4 ppb) in the daytime and at night, respectively. For the daytime and night, the highest mean levels of 114.6 and 59.3 ppb were obtained from 09:00 to 10:00 and 19:00 to 20:00, respectively. These two highest mean levels in the daytime and at night were approximately 20–110 times greater than ambient background levels (1–3 ppb) measured in several previous reports (Sickles *et al*., 1990; Aneja *et al*., 1998; Leaderer *et al*., 1999; Lefer *et al*., 1999; McCurdy *et al*., 1999; Walker *et al*, 2004; Wilson and Serre, 2007a, 2007b).

Fig. 4 shows that the mean NH_3 levels for the whole day ranged from 45.0 to 122.6 ppb (mean = 84.9 ppb) in Pingnan Industrial Park. For the duration of sampling, the highest mean level of 122.6 ppb was obtained from 12:00 to 13:00. Moreover, the mean NH_3 levels ranged from 73.0 to 122.6 ppb (mean = 105.2 ppb) and from 45.0 to 79.0 ppb (mean = 58.0 ppb) in the daytime and at night, respectively. For the daytime and night, the highest mean levels of 122.6 and 79.0 ppb were obtained from 12:00 to 13:00 and 18:00 to 19:00, respectively. Like the case in Pingtung, these two highest mean levels in day and night were about 25–120 times greater than ambient background levels (1–3 ppb) measured in several previous reports (Sickles *et al*., 1990; Aneja *et al*., 1998; Leaderer *et al*., 1999; Lefer *et al*., 1999; McCurdy *et al*., 1999; Walker *et al*, 2004; Wilson and Serre, 2007a, 2007b).

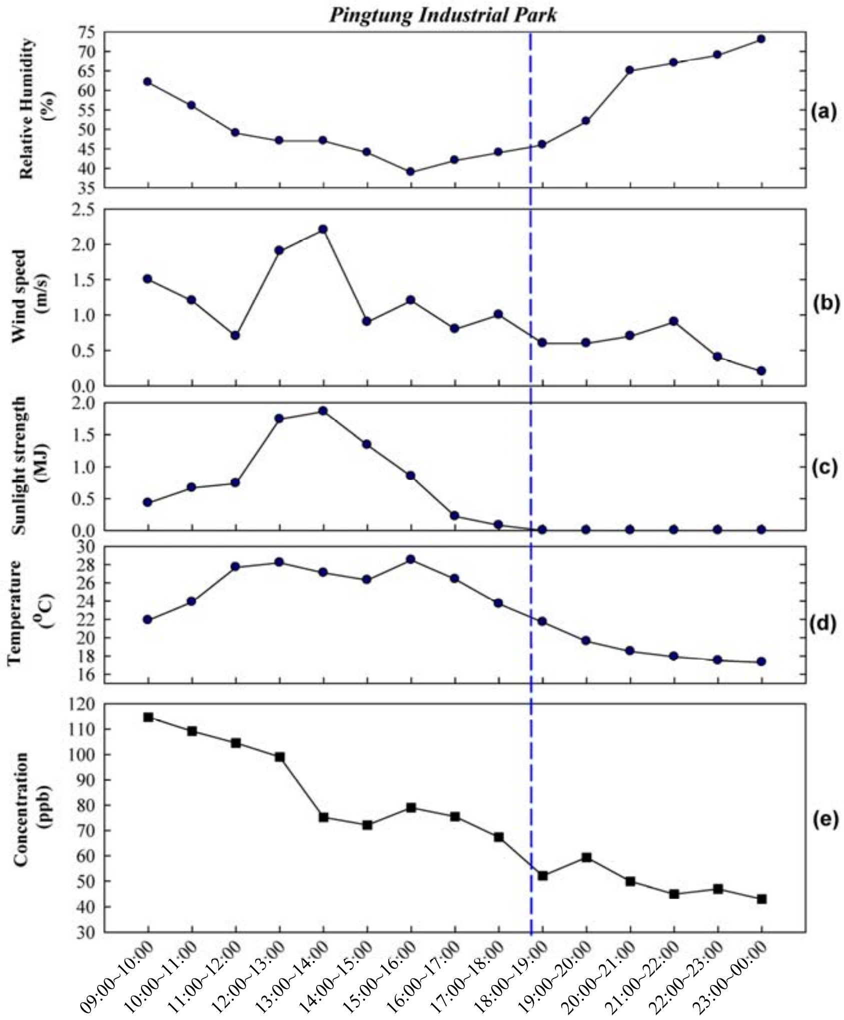

Fig. 3. Daily variation in both meteorological parameter records (from (a) to (d)) and average ambient ammonia concentrations (e) in Pingtung Industrial Park in southern Taiwan.

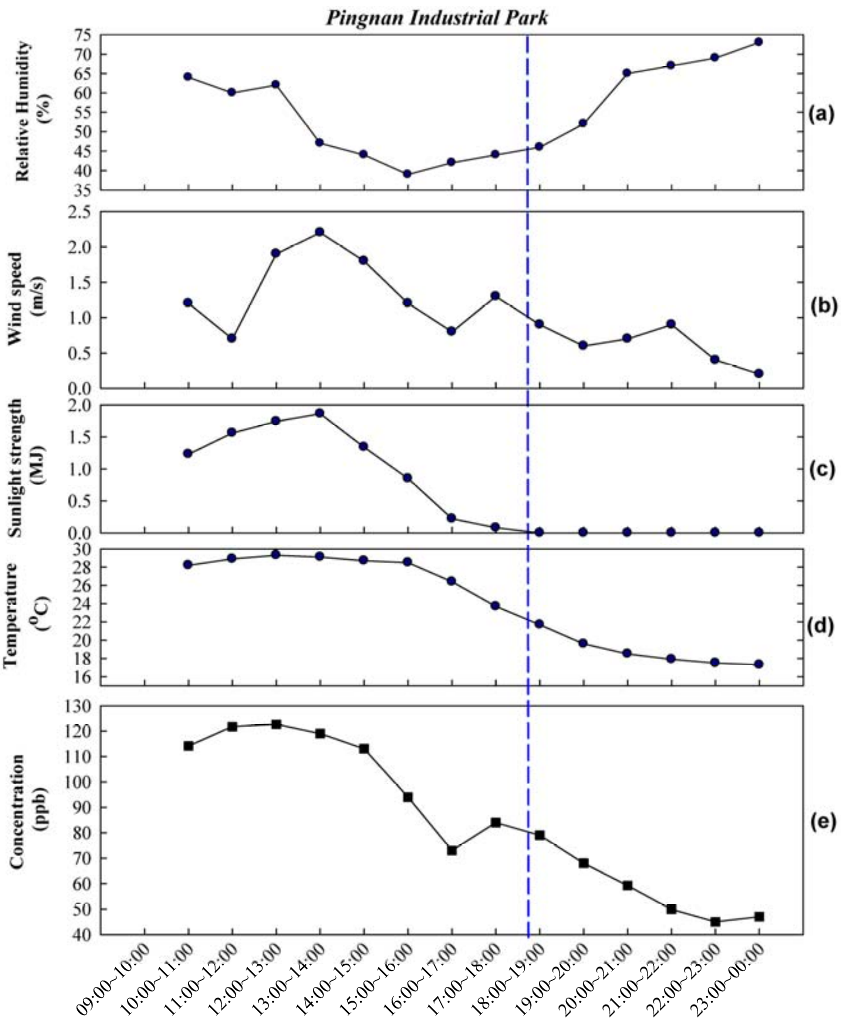

Fig. 4. Daily variation in both meteorological parameter records (from (a) to (d)) and average ambient ammonia concentrations (e) in Pingnan Industrial Park in southern Taiwan.

To discern the difference between day and night, the mean daytime NH_3 concentration was divided by the mean nighttime NH_3 concentration. The mean daytime to night ratios were 1.8, 1.8, 4.2 and 2.6 in Pingtung, Pingnan, NPUST dormitory, and the bamboo grove, respectively. However, the ratio of 0.79 in Neipu Industrial Park is significantly different to the others being the only one that is under 1, which indicates that there is either an increased emission under the same sources as daytime, or there is a new emission source only at night. At this stage of the investigation, one explanation is NH_3 sampling occurred in the Neipu area with multiple sources, and, while the nearest source does have an impact, we cannot ignore the overall elevation of ammonia in the sampling area due to the contribution of the other sources.

Ambient Ammonia Concentrations in Background Levels

Fig. 5 presents the daily variation in both meteorological parameter records and the average ambient NH_3 concentrations near Ren-Jhai Dormitory on the campus of National Pingtung University of Science and Technology (NPUST) in Neipu Township, in a location that is generally classified as a rural area. The sampling site near the NPUST dormitory is representative of the ambient level of a residential area without industrial sources nearby. Therefore, the NPUST dormitory was selected as the location #1 for the background site without industrial sources. Indeed, the only possible emission of NH_3 is from the transoms of toilets.

For the whole day, the mean NH_3 levels ranged from 13.0 to 101.7 ppb (mean = 52.2 ppb) in the ambient air of NPUST dormitory (background site #1). For the duration of sampling, the highest mean level of 101.7 ppb was obtained from 11:00 to 12:00. Moreover, Fig. 5 also shows that the mean NH_3 levels ranged from 17.5 to 101.7 ppb (mean = 69.4 ppb) and from 13.0 to 25.1 ppb (mean = 16.6 ppb) in the daytime and at night, respectively. For the day and night, the highest mean levels of 101.7 and 25.1 ppb were obtained from 11:00 to 12:00 and 19:00 to 20:00, respectively. These two highest mean levels in day and night were 25–100 times greater than the ambient background levels (1–3 ppb) measured in several previous reports (Sickles *et al*., 1990; Aneja *et al*., 1998; Leaderer *et al*., 1999; Lefer *et al*., 1999; McCurdy *et al*., 1999; Walker *et al*, 2004; Wilson and Serre, 2007a, 2007b).

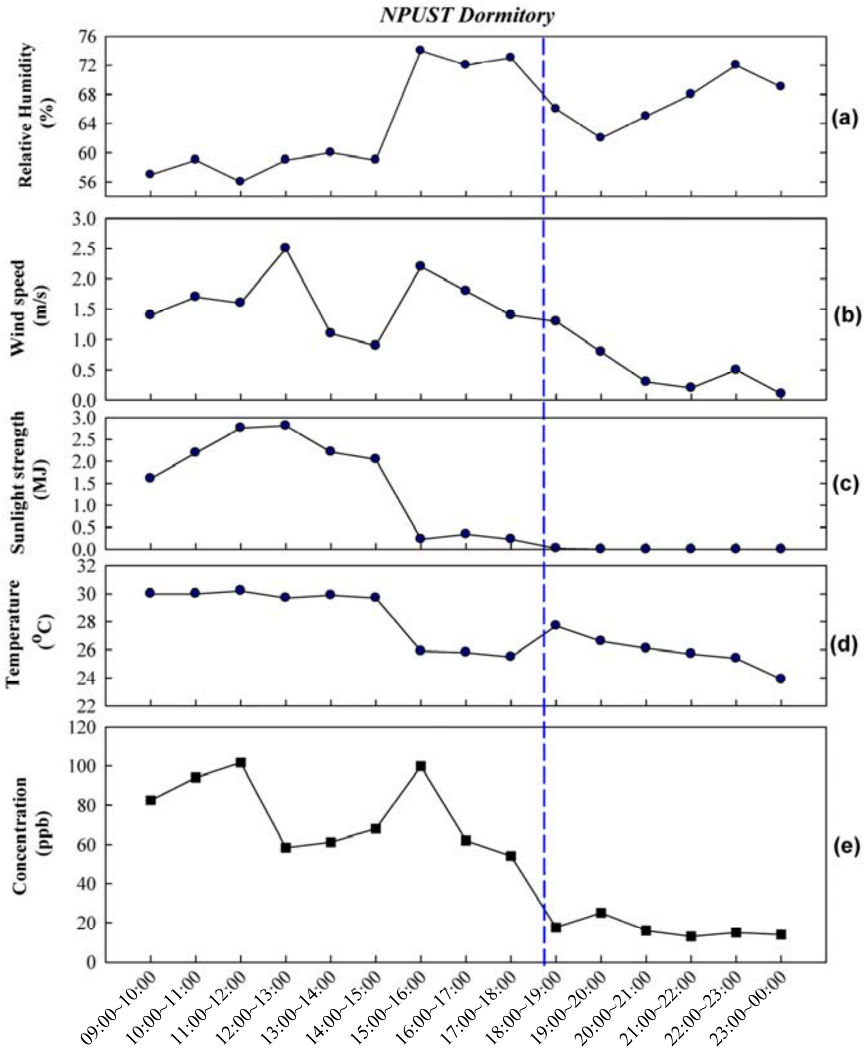

Fig. 5. Daily variation in both meteorological parameter records (from (a) to (d)) and average ambient ammonia concentrations (e) in NPUST dormitory in southern Taiwan.

Fig. 6 presents the daily variation in both meteorological parameters records and the average ambient NH_3 concentrations in a bamboo grove (the background site #2) in southern Taiwan. The NH_3 levels measured in this site compare well with those in other studies on the levels of atmospheric NH_3 at or near hog concentrated animal feeding operations in eastern North Carolina. The mean values obtained for background site #2 in this study were 4.6 ppb (daylong), 5.8 ppb (daytime) and 2.2 ppb (night). These values are comparable to those obtained by Langford *et al*. (1992), Buijsman *et al*. (1998) and Wilson and Serre (2007a). When NH_3 values from our study are compared to the mean levels from other studies, it can be seen that the data corresponds well to these earlier measurements. In addition, the data for background site #2 are consistent with what is expected for low ambient NH_3 emission or agricultural sites (Buijsman *et al*., 1998). The comparison between the data from our work and previous studies also indicates that background site #2 can serve as a reasonable reference to judge the elevated ambient NH_3 measured in the Pingtung area.

The results show that NH_3 concentrations were higher at night in Neipu industrial park, during the day in Pingtung and Pingnan industrial parks, and the day vs. night concentrations were similar in the bamboo grove background site. Table 1 lists the ammonia concentration correlation coefficients (R) during daytime, night time and daylong sampling periods at the five sampling sites. During the daylong periods, significant correlations were found between Pingtung and Pingnan industrial parks and between Pingtung and NPUST dormitory. During the daytime sampling periods, no significant correlations were found among the five sites. However, during night time periods, a significant correlation was found between the Pingtung industrial park and NPUST dormitory. These results can be seen more clearly in Fig. 7.

Ammonia Levels vs. Meteorological Parameters

Table 2 gives the correlations matrix for the NH_3 levels versus their corresponding meteorological values at the five sampling locations during the daylong sampling periods. In addition, Fig. 8 presents the visual interaction

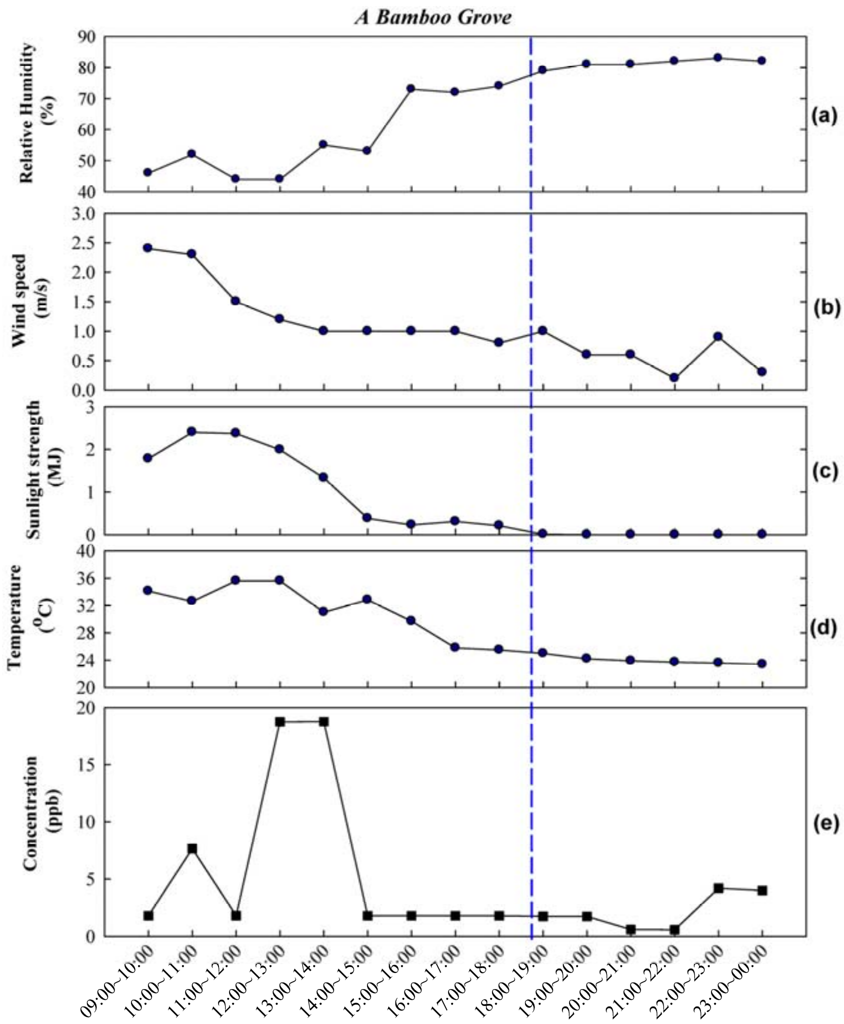

Fig. 6. Daily variation in both meteorological parameter records (from (a) to (d)) and average ambient ammonia concentrations (e) in the bamboo grove in southern Taiwan.

Table 1. Summary of ammonia concentration correlation coefficients (R) during daytime, nighttime and daylong sampling periods in the five sites

Daylong	*Neipu Industrial Park*	*Pingtung Industrial Park*	*Pingnan Industrial Park*	*NPUST dormitory*	*A bamboo grove (background site)*
Neipu Industrial Park	1.000				
Pingtung Industrial Park	–0.298	1.000			
Pingnan Industrial Park	–0.493	**0.867****	1.000		
NPUST dormitory	–0.477	**0.876****	**0.822****	1.000	0.152
A bamboo grove (background site)	–0.071	0.307	0.490	0.152	1.000
Daytime					
Neipu Industrial Park	1.000				
Pingtung Industrial Park	0.614	1.000			
Pingnan Industrial Park	0.218	0.585	1.000		
NPUST dormitory	–0.044	0.552	0.264	1.000	–0.391
A bamboo grove (background site)	0.398	0.068	0.512	–0.391	1.000
Night time					
Neipu Industrial Park	1.000				
Pingtung Industrial Park	–0.255	1.000			

Table 1. *(continued).*

Daylong	*Neipu Industrial Park*	*Pingtung Industrial Park*	*Pingnan Industrial Park*	*NPUST dormitory*	*A bamboo grove (background site)*
Pingnan Industrial Park	–0.539	0.748	1.000		
NPUST dormitory	–0.339	**0.958****	0.620	1.000	–0.165
A bamboo grove (background site)	–0.364	–0.349	–0.469	–0.165	1.000

** Correlation is significant at the 0.01 level (2-tailed).

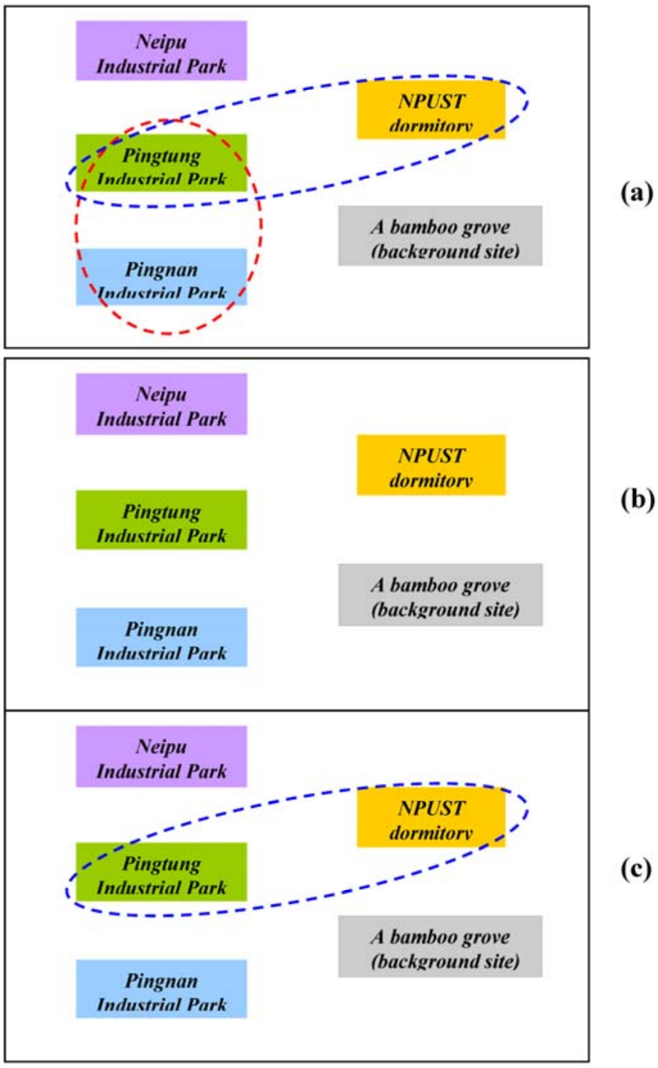

Fig. 7. Correlations in the five different sampling sites during the daylong (a), daytime (b), and nighttime (c) periods in southern Taiwan.

correlations. The information contained in both Table 2 and Fig. 8 is summarized, as follows.

(i) A significant correlation between the NH_3 level and relative humidity was observed in Neipu Industrial Park (Table 2 and Fig. 8(a)). Moreover, high positive correlations between measured ambient temperature, wind speed, and sunlight strength were also observed, although there was a high negative correlation between the ambient temperature level and relative humidity (Table 2 and Fig. 8(a)).

Significant positive correlations between the NH_3 level and ambient temperature, wind speed, and sunlight strength were also observed in Pingtung Industrial Park (Table 2 and Fig. 8(b)), and it is likely that these have significant theoretical importance (for example, in relation to the dispersion of NH_3). Furthermore, high positive correlations between the measured ambient temperature level and wind speed and sunlight strength were also observed. These trends are fairly consistent across a wide variety of meteorological parameters. In contrast, high negative correlations between the ambient temperature level and relative humidity and atmospheric pressure were observed (Table 2 and Fig. 8(b)).

(ii) Significant positive correlations between the NH_3 level and ambient temperature, wind speed, and sunlight strength were also observed in Pingnan Industrial Park (Table 2 and Fig. 8(c)). Moreover, a significant negative correlation between the NH_3 level and atmospheric pressure was also observed. In contrast, the results showed a high negative correlation between the ambient temperature level and atmospheric pressure.

(iii) Significant positive correlations between the NH_3 level and ambient temperature, wind speed, and sunlight strength were found at NPUST dormitory (Table 2 and Fig. 8(d)). In contrast, the results showed a high negative correlation between the ambient temperature level and relative humidity.

(iv) In the bamboo grove, no significant correlation between the NH_3 level and others was found (Table 2 and Fig. 8(e)). This result is quite different from those obtained for the other four sampling sites in this study (please see Fig. 8(a)–(d)). Moreover, on the basis of different meteorological parameters, high positive correlations between measured ambient temperature

Table 2. Correlation matrix for NH_3 levels vs. the meteorological values in the five sampling sites.

	NH_3 level	Ambient Temp.	RH.	Atmospheric Pressure	Wind speed	Sunlight strength
Neipu Industrial Park						
NH_3 level	1					
Ambient temp.	–0.425	1				
RH.	**0.546***	**–0.923****	1			
Atmospheric pressure	0.422	–0.476	0.5	1		
Wind speed	–0.211	**0.715****	**–0.639***	–0.373	1	
Sunlight strength	–0.336	**0.880****	**–0.815****	**–0.545***	**0.824****	1
Pingtung Industrial Park						
NH_3 level	1					
Ambient temp.	**0.667****	1				
RH.	–0.369	**–0.852****	1			
Atmospheric pressure	–0.351	**–0.800****	**0.573***	1		
Wind speed	**0.586***	**0.615***	–0.397	**–0.568***	1	
Sunlight strength	**0.535***	**0.770****	–0.488	**–0.826****	**0.804****	1
Pingnan Industrial Park						
NH_3 level	1					
Ambient temp.	**0.941****	1				
RH.	–0.38	–0.532	1			
Atmospheric pressure	**–0.736****	**–0.652***	–0.213	1		
Wind speed	**0.765****	**0.739****	–0.466	–0.311	1	
Sunlight strength	**0.927****	**0.872****	–0.163	**–0.740****	**0.752****	1
NPUST dormitory						
NH_3 level	1					
Ambient temp.	**0.625***	1				
RH.	–0.371	**–0.883****	1			
Atmospheric pressure	0.214	0.255	–0.365	1		
Wind speed	**0.744****	0.476	–0.156	0.275	1	
Sunlight strength	**0.679****	**0.905****	**–0.803****	0.083	**0.520***	1
A bamboo grove (background site)						
NH_3 level	1					
Ambient temp.	0.446	1				
RH.	–0.483	**–0.978****	1			
Atmospheric pressure	–0.351	–0.398	0.343	1		
Wind speed	0.162	**0.732****	**–0.748****	0.087	1	
Sunlight strength	0.498	**0.877****	**–0.906****	–0.066	**0.798****	1

* Correlation is significant at the 0.05 level (2-tailed).
** Correlation is significant at the 0.01 level (2-tailed).

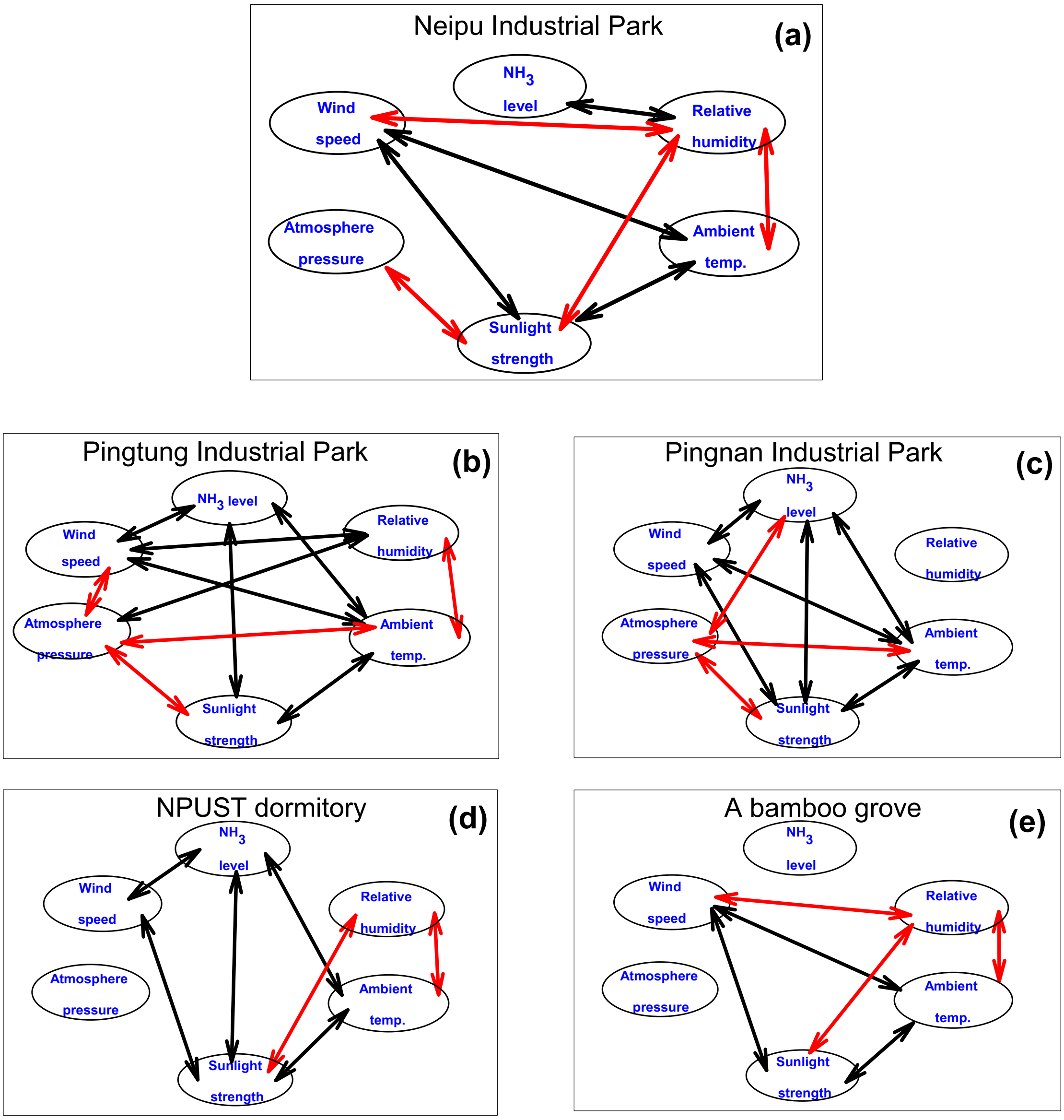

Fig. 8. The visual interaction correlations of the different measured parameters in the five different sampling sites in southern Taiwan.

and wind speed, and sunlight strength were observed at this site, while a high negative correlation between measured ambient temperature level and relative humidity was also found (Table 2 and Fig. 8(e)).

(v) These different correlations highlight the need for further research to investigate many of the ambient meteorological factors, and especially variations between different sampling sites.

Characteristics of Ratio Values Normalized by the Background Levels

It is difficult for local governments to address illegal agricultural practices and the related nighttime emissions in southern Taiwan. Specifically, the facts that there are many unregistered farmyards in this region make it impossible to accurately estimate the emission of NH_3 or odors (such as H_2S) from such sources. Therefore, we use the characteristics of ratio value based on the standard mathematical normalization process to obtain better information. In this study, ratio values were categorized according to the cover-range (from minimum ratios to maximum ratios.) at different sampling periods and the minimum separated boundary lines. Fig. 8 shows that the characteristics of the ratio values normalized by the background levels can be divided into three main categories: (1) $R > 200$; (2) $R > 60$; (3) $R > 10$.

Fig. 9 shows that the ratio values were highest ($R > 200$) from both 20:00 to 21:00 and 21:0 to 22:00 in Neipu Industrial Park. The difference at different sampling locations in Fig. 9 is most likely due to the different ammonia sources, such as industrial processes, agriculture, and livestock. The value of R was determined mostly by the effects of the multiple sources at each sampling location, indicating that different distribution zones might

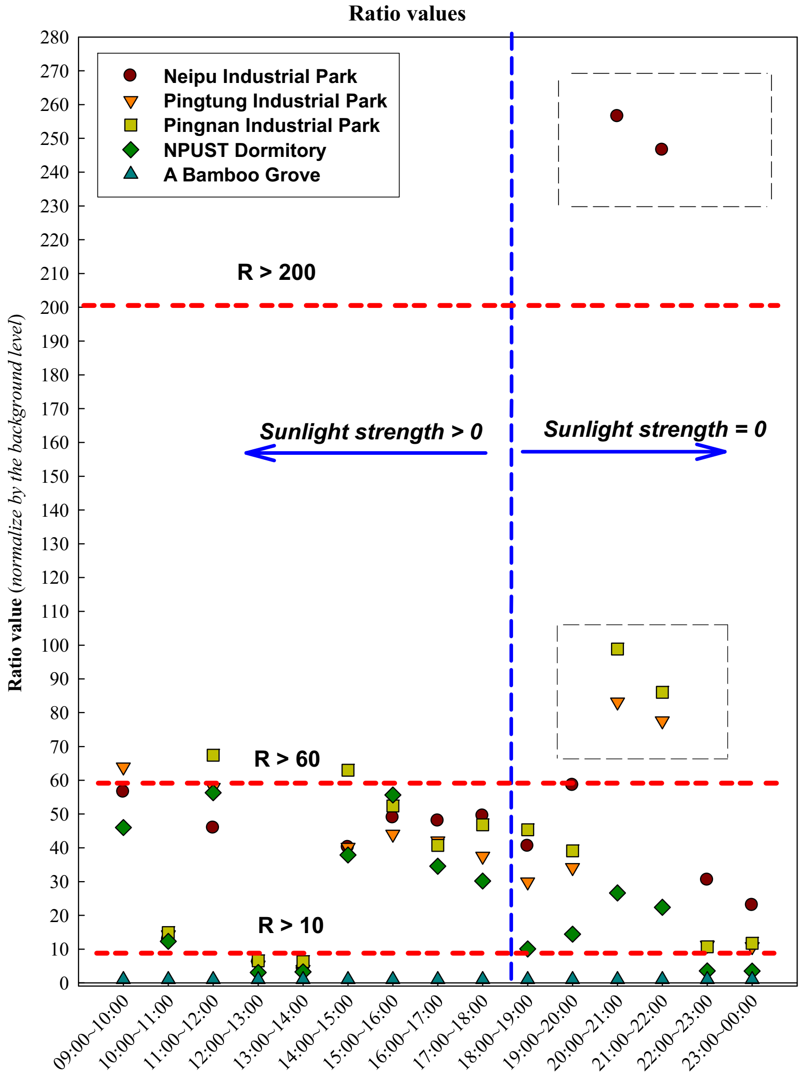

Fig. 9. Ratio values in the different sampling location in southern Taiwan.

the result of both cover and accumulate in their NH_3 source profile. This trend is similar to that found by Walker *et al.* (2004), that agricultural NH_3 emissions influence local ambient concentrations of NH_3 and $PM_{2.5}$ Therefore, illegal or unregistered NH_3 sources could play a critical role with regard to the higher R levels in Neipu country. However, to the best of our knowledge the validation of such hidden NH_3 sources is very difficult due to the point-source release during nighttime or off-duty periods. However, it is particularly interesting to compare the industrial parks and background NH_3 levels. Such high times in Neipu Industrial Park gave us a clear understanding of NH_3 level in different industrial parks. In this study, our research data reveal obvious reasons for this clear distinction of ambient NH_3 levels in southern Taiwan.

CONCLUSIONS

Ammonia measurement investigations play a critical role in air pollution and atmospheric chemistry studies. In Neipu Industrial Park, the highest average 153.9 ppb was in the period from 20:00 to 21:00 and lowest average 70.5 ppb was from 18:00 to 19:00. In Pingtung Industrial Park, the highest average 114.7 ppb was from 09:00 to 10:00 and lowest average 70.5 ppb was recorded from 18:00 to 19:00. The results of this study indicate that NH_3 concentrations were higher at night in Neipu Industrial Park, during the day at in Pingtung and Pingnan Industrial Parks, and day vs. night concentrations were similar in the bamboo grove background site. In this study, the high levels of NH_3 at the Neipu site in southern Taiwan were dominated by various factors and sources which may be related to non-industrial emissions. Finally, it is important to note that widely distributed NH_3 emissions from local point sources are more likely to impact the local air quality in southern Taiwan, and are less likely to transport to the long distance area.

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REFERENCES

Aneja, V.P., Bunton, B., Walker, J.T. and Malik, B.P. (2001). Measurement and Analysis of Atmospheric Ammonia Emissions from Anaerobic Lagoons. *Atmos. Environ.* 35: 1949–1958.

Aneja, V.P., Chauhan, J.P. and Walker, J.T. (2000). Characterization of Atmospheric Ammonia Emissions from Swine Waste Storage and Treatment Lagoons. *J. Geophys. Res.* 105: 11535–11545.

Balsari, P., Airoldi, G., Dinuccio, E. and Gioelli, F. (2007). Ammonia Emissions from Farmyard Manure Heaps and Slurry Stores—Effect of Environmental Conditions and Measuring Methods. *Biosystems Eng.* 97: 456–463.

Bouwman, A.F., Lee, D.S., Asman, W.A.H., Dentener, F.J., van der Hoek, K.W. and Olivier, J.G.J. (1997). A Global High Resolution Emission Inventory for Ammonia. *Global Biogeochem. Cycles* 11: 561–587.

Buijsman, E., Aben, J.M.M., van Elzakker, B.G. and Mennen, M.G. (1998). An Automatic Atmospheric Ammonia Network in The Netherlands: Setup and Results. *Atmos. Environ.*32: 317–324.

Cao, J.J., Zhang, T., Chow, J.C., Watson, J.G., Wu, F. and Li, H. (2009) Characterization of Atmospheric Ammonia over Xi'an, China. *Aerosol Air Qual. Res.* 9: 277–289.

Carmichael, G.R., Ferm, M., Thongboonchoo, N., Woo, J.H., Chan, L.Y. and Murano K. (2003). Measurements of Sulfur Dioxide, Ozone and Ammonia Concentrations in Asia, Africa and South America Using Passive Samplers. *Atmos. Environ.* 3: 1293–1308.

ECETOC. (1994). *Ammonia Emission to Air in Western Europe*. Technical Report No. 62: 196.

Hsieḥ, L.T., Yang, H.H. and Chen, H.W. (2005). Characterization of Both MTBE and BTEX in the Ambient Air of Night Markets in Southern Taiwan. *Aerosol Air Qual. Res.* 15: 154–170.

Langford, A.O., Fehsenfeld, F.C., Zachariassen, J. and Schimel, D.S. (1992). Gaseous Ammonia Fluxes and Background Concentrations in Terrestrial Ecosystems of the United States. *Global Biogeochem. Cycles* 6: 459–483.

Leaderer, B.P., Naeher, L., Jankun, T., Balenger, K., Holford, T.R., Toth, C., Sullivan, J., Wolfson, J.M. and Koutrakis, P. (1999). Indoor, Outdoor, and Regional Summer and Winter Concentrations of PM_{10}, $PM_{2.5}$, SO_4^{2-} , H^+, NH_4^+, NO_3^-, and Nitrous Acid in Homes with and without Kerosene Space Heaters. *Environ. Health Perspect.* 107: 223–231.

Lefer, B.L., Talbot, R.W. and Munger, J.W. (1999). Nitric Acid and Ammonia at a Rural Northeastern US Site. *J. Geophys. Res.* 104: 1645–1661.

McCulloch, R.B., Few, G.S., Murray Jr., G.C. and Aneja, V.P. (1998). Analysis of Ammonia, Ammonium Aerosols and Acid Gases in the Atmospheric at a Commercial Hog Farm in Eastern North Carolina, USA. *Environ. Pollut.* 102: 263–268.

McCurdy, T., Zelenka, M.P., Lawrence, P.M., Houston, R.M. and Burton, R. (1999). Acid Aerosols in the Pittsburgh Metropolitan Area. *Atmos. Environ.* 33: 5133–5145.

PCIC (2010). http://invest.pthg.gov.tw/CmsShow.aspx, updated date on 2010-06-2.

Pryor, S.C., Barthelmie, R.J., Sorenson, L.L. and Jensen, B. (2001). Ammonia Concentrations and Fluxes over a Forest in the Midwestern USA. *Atmos. Environ.* 35: 5645–5656.

Rattray, G. and Sievering, H. (2001). Dry Deposition of Ammonia, Nitric Acid, Ammonium, and Nitrate to Alpine Tundra at Niwot Ridge, Colorado. *Atmos. Environ.* 35: 1105–1109.

Robarge, W.P., Walker, J.T., McCullock, R.B. and Murray, G. (2002). Atmospheric Concentrations of Ammonia and Ammonium at an Agricultural Site in the Southeast United States. *Atmos. Environ.* 36: 1661–1674.

Sather, M.E., Mathew, J., Nguyen, N., Lay, J., Golod, G., Vet, R., Cotie, J. and Geasland, F. (2008). Baseline Ambient Gaseous Ammonia Concentrations in the Four Corners Area and Eastern Oklahoma, USA. *J. Environ. Monit.* 10: 1319–1325.

Sickles II, J.E., Hodson, L.L., McClenny, W.A., Paur, R.J., Ellestad, T.G., Mulik, J.D., Anlauf, K.G., Wiebe, H.A., Mackay, G.I., Schiff, H.I. and Bubacz, D.K. (1990). Field Comparison of Methods for the Measurement of Gaseous and Particulate Contributors to Acidic Dry Deposition. *Atmos. Environ.* 24A: 155–165.

Taiwan EPA (1997). NIEA A426.71B: Indophenol-UV Method, http://www.niea.gov.tw/niea/AIR/A42671B.htm, updated date on 2010-06-2.

Walker, J.T., Aneja, V.P. and Dickey, D. (2000a). Atmospheric Transport and Wet Deposition of Ammonia in North Carolina. *Atmos. Environ.* 34: 3407–3418.

Walker, J.T., Nelson, D. and Aneja, V.P. (2000b). Trends in Ammonium Concentration in Precipitation and Atmospheric Ammonia Emissions at a Coastal Plain Site in North Carolina, USA. *Environ. Sci. Technol.* 34: 3527–3534.

Walker, J.T., Whitall, D.R., Robarge, W. and Paerl, H.W. (2004). Ambient Ammonia and Ammonium Aerosol across a Region of Variable Ammonia Emission Density. *Atmos. Environ.* 38: 1235–1246.

Wilson S.M. and Serre M.L. (2007a). Examination of Atmospheric Ammonia Levels near Hog CAFOs, Homes, and Schools in Eastern North Carolina. *Atmos. Environ.* 41: 4977–4987.

Wilson S.M. and Serre M.L. (2007b). Use of Passive Samplers to Measure Atmospheric Ammonia Levels in a High-density Industrial Hog Farm Area of Eastern North Carolina. *Atmos. Environ.* 41: 6074–6086.

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