

Article

Source-Directional and Micrometeorological Influences on Short-Term NH₃ Variability in a Livestock- and Agriculture-Influenced Peri-Urban Environment

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Abstract

Atmospheric ammonia (NH₃) is an important alkaline precursor of secondary inorganic aerosols, but its variability in livestock- and agriculture-influenced peri-urban environments remains poorly constrained. In this study, atmospheric NH₃ was measured at a peri-urban site in Chuncheon, South Korea and its variability was examined in relation to micro-meteorology, source direction, and surface–atmosphere exchange. Mean NH₃ concentrations were 200.8 ± 91.1 ppb during the April campaign, 83.0 ± 34.4 ppb during the May campaign, and 27.9 ± 16.2 ppb during the December campaign, indicating higher NH₃ levels during the April and May campaigns than during the December campaign. Campaign-specific correlation analyses showed that the relationships between NH₃ and micrometeorological variables differed among the observation periods, with robust associations observed in April and May but not in December. Moreover, the higher NH₃ concentration in the April campaign than in the May campaign, despite the lower mean temperature, indicates that the observed variability was not controlled by temperature alone. Conditional probability function analysis showed that elevated NH₃ concentrations in the April and May campaigns were mainly associated with southwesterly winds, suggesting the influence of nearby livestock and agricultural sources. The Penman–Monteith-derived latent heat flux further showed that daytime NH₃ enhancement coincided with evaporative surface-exchange conditions potentially favorable for volatilization, although it did not directly quantify manure-derived NH₃ emissions. In contrast, the December campaign showed lower NH₃ concentrations, weaker source-directional patterns, and limited latent heat flux influence, suggesting suppressed volatilization and intermittent local accumulation under stable conditions. These results indicate that the conditions associated with short-term NH₃ variability differed among the selected campaigns, reflecting complementary influences of source direction and campaign-specific micrometeorological and surface-exchange conditions, highlighting the need for concurrent gas- and particle-phase measurements to assess potential implications for PM_{2.5} formation.



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Keywords: ammonia (NH_3); livestock emissions; agricultural sources; latent heat flux; surface-atmosphere exchange

1. Introduction

Atmospheric ammonia (NH_3) is the most abundant alkaline gas in the atmosphere and a key regulator of aerosol acidity. It readily reacts with acidic species such as nitric acid (HNO_3) and sulfuric acid (H_2SO_4) to form ammonium nitrate (NH_4NO_3) and ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$), which often account for a substantial fraction of $\text{PM}_{2.5}$ mass, particularly during severe haze episodes [1,2]. In addition to its importance in secondary aerosol formation, NH_3 is also a major odor-causing compound associated with livestock facilities and manure management and is regulated as a designated odorant in South Korea.

Despite its importance for air quality and nitrogen cycling, NH_3 mitigation has historically received less attention than controls on other $\text{PM}_{2.5}$ precursors such as NO_x and SO_2 . In South Korea, Atmospheric NH_3 is often present in excess relative to HNO_3 , and nitrate formation is therefore commonly viewed as being limited primarily by NO_x [3,4]. However, this perspective overlooks the role of NH_3 in controlling the partitioning and atmospheric lifetime of reactive nitrogen. While gas-phase HNO_3 is removed rapidly by dry deposition, particulate NH_4NO_3 generally has a lower deposition velocity and longer residence time [5]. Thus, abundant NH_3 can prolong the persistence and transport of reactive nitrogen by stabilizing nitrate in the particle phase. Even under NH_3 -rich conditions, reducing NH_3 emissions may shift nitrate partitioning toward gas-phase HNO_3 and shorten the atmospheric lifetime of reactive nitrogen. This highlights that NH_3 remains an important control variable for secondary aerosol formation and nitrogen cycling, even in environments where it is not conventionally treated as the limiting precursor.

This issue is particularly important in South Korea, where NH_3 emissions are dominated by diffuse agricultural and livestock-related sources characterized by strong spatial heterogeneity and pronounced temporal variability. According to the Clean Air Policy Support System (CAPSS), national NH_3 emissions reached 242,227 tons in 2023, of which approximately 83% originated from agriculture. A substantial fraction of agricultural NH_3 emissions is associated with livestock activities, including animal housing, manure storage, manure handling, and land application of livestock waste, in addition to fertilizer application to croplands. Unlike NO_2 and SO_2 , which have declined under strengthened emission controls, NH_3 emissions have remained largely unchanged. Recent satellite observations have also indicated increasing NH_3 column abundances, implying a growing atmospheric NH_3 reservoir and a greater potential for secondary inorganic aerosol formation [6]. At the local scale, however, atmospheric NH_3 exhibits strong spatiotemporal variability because its concentrations are controlled not only by emission strength but also by source proximity, wind direction, bidirectional surface exchange and wind-dependent transport, dilution, or local accumulation [7–9].

In livestock- and agriculture-influenced environments, these processes are tightly coupled. Livestock facilities can act as persistent local NH_3 sources through emissions from animal housing and manure-related surfaces, whereas fertilization and manure application can generate strong episodic NH_3 emissions [10]. NH_3 volatilization from manure and fertilized surfaces is strongly affected by temperature, pH, total ammoniacal nitrogen, surface moisture, exposed surface area, and wind-driven transfer [5,11,12]. After sunrise, increasing solar radiation and temperature can enhance volatilization and surface drying, while changes in surface moisture can influence NH_3 uptake and re-emission [13]. Accord-

ingly, short-term NH_3 variability near livestock and agricultural sources is likely to reflect multiple interacting local conditions rather than a single meteorological factor.

Although the effects of meteorological conditions, source proximity, and surface exchange on atmospheric NH_3 have been widely investigated, short-term NH_3 variability near closely located livestock and agricultural sources remains difficult to interpret because meteorological and source-related factors associated with NH_3 concentration changes may vary among short observation periods. In particular, it remains unclear whether similar relationships with meteorological variables or source direction persist under different local emissions and surface conditions. This uncertainty is important because temperature-dependent volatilization, wind-directional transport, local source activity, and surface drying or moisture accumulation can occur simultaneously and may reinforce or offset one another over short timescales. We therefore hypothesize that short-term atmospheric NH_3 variability at such a site is not governed by a single dominant factor, but by campaign-specific combinations of source-directional transport, local source conditions, and micrometeorological processes.

This study therefore investigates short-term NH_3 variability at a peri-urban site in a medium-sized city influenced by nearby livestock facilities and agricultural fields, focusing on differences among three selected short-term measurement periods. Using high-temporal-resolution observations, we examined whether the meteorological and source-directional conditions associated with NH_3 variability were consistent among the campaigns or differed according to campaign-specific local conditions. Campaign-specific correlation analyses were used to characterize the relationships between NH_3 and micrometeorological variables, while conditional probability function (CPF) analysis was applied to identify wind-directional characteristics associated with elevated NH_3 concentrations and to examine the potential influence of nearby livestock and agricultural source sectors. In addition, Penman–Monteith-derived latent heat flux was used only as an auxiliary meteorological diagnostic of relative evaporative and moisture-accumulating conditions, not as a direct proxy for NH_3 emissions, manure-surface wetness, soil moisture, or fertilizer volatilization. By combining these complementary lines of evidence, this study aims to provide a campaign-specific interpretation of short-term NH_3 variability rather than attributing the observed concentration changes to a single generalized controlling factor.

2. Method

2.1. Sampling Site and Period

The sampling site (37.940° N, 127.784° E) was situated in Chuncheon, Republic of Korea—a mid-sized city characterized by a high forest cover (~74%) and an absence of major industrial point sources (Figure 1a). Given these regional features, non-agricultural anthropogenic NH_3 emissions are expected to be relatively minor. To specifically examine the atmospheric NH_3 variability associated with local agricultural and livestock activities, the sampling station was positioned immediately east of a university-affiliated livestock facility, outside the facility boundary but in very close proximity to the animal housing and manure-management area (Figure 1). Forage crop fields were located to the north and south of the livestock facility, and an agricultural field was situated further south. The facility housed 113 animals in total, including 93 Korean cattle, 12 beef cattle, 1 donkey, 4 sheep, and 3 horses. Although the farm was modest in scale, the close spatial coupling between the livestock facility, surrounding forage crop fields, and the sampling inlet provided a suitable setting for capturing atmospheric NH_3 variability influenced by local livestock metabolism, manure handling, and agricultural activities.

Field measurements were conducted during three intensive 24 h campaigns in 2024: 11–12 April, 23–24 May, and 10–11 December. The April and May campaigns were con-

ducted during the active growing and fertilization period, when NH_3 emissions from soil fertilization and livestock-related sources were expected to be elevated. Specifically, the April campaign was conducted during the active fertilization period, whereas the May campaign was conducted later in the growing season. In contrast, the December campaign was conducted during a non-fertilized dormant period with limited agricultural activity in the surrounding field, allowing atmospheric NH_3 variability near the livestock facility to be examined under cold-season, non-fertilization conditions.

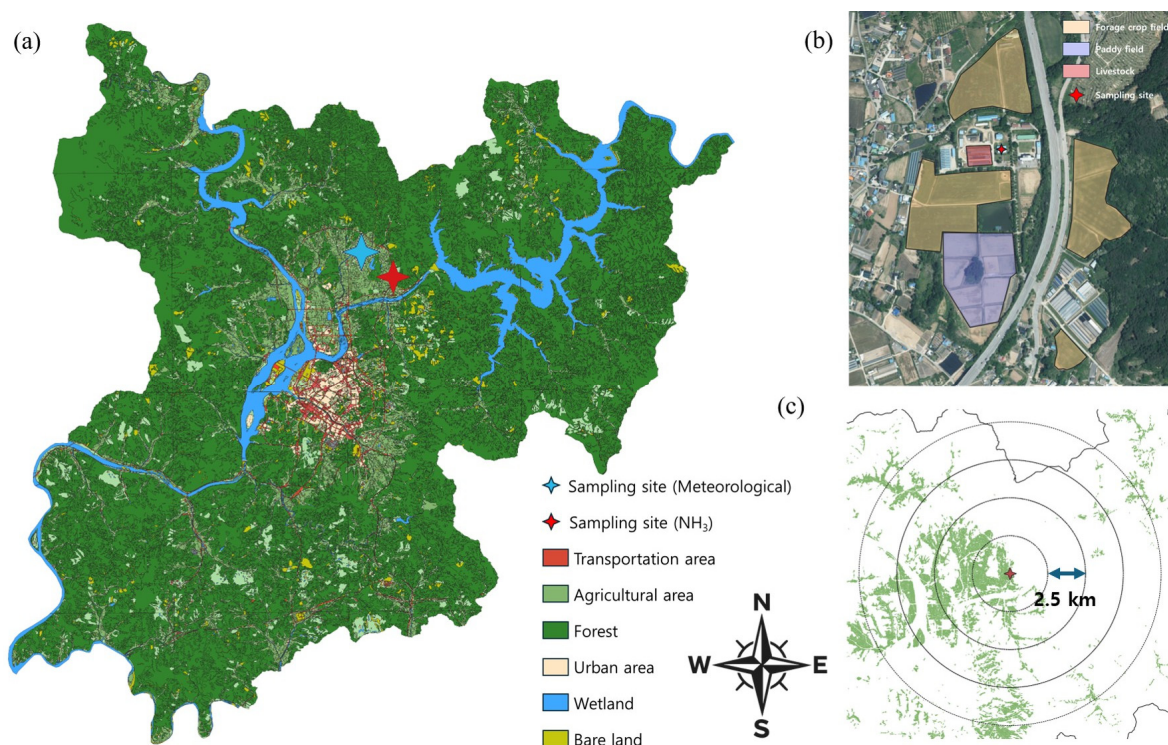


Figure 1. Study area and NH_3 sampling location. (a) Location of Chuncheon, South Korea (scale 1:260,000); (b) NH_3 measurement site and surrounding land-use features, including forage crop fields (yellow), livestock facilities (red), and paddy fields (blue) (scale 1:5000); and (c) Spatial distribution of agricultural areas around the NH_3 sampling site, with concentric buffers drawn at 2.5 km intervals (scale 1:130,000).

2.2. Ammonia Measurement

Atmospheric NH_3 concentrations were continuously monitored using a high-precision ammonia analyzer (LGR-ICOSTM GLA311-EAA, ABB Inc., Québec, QC, Canada). The instrument employs Off-Axis Integrated Cavity Output Spectroscopy (OA-ICOS), a cavity-enhanced absorption technique. In this method, a tunable diode laser is introduced off-axis into an optical cavity comprising two highly reflective mirrors. This configuration excites multiple transverse modes and significantly extends the effective optical path length (up to several kilometers), thereby enhancing detection sensitivity. The NH_3 concentration is retrieved from the steady-state light transmission through the cavity.

The NH_3 analyzer was calibrated annually by APM Engineering, the authorized representative company for the instrument manufacturer in Korea, as part of the regular maintenance and performance-check procedure. During the field campaigns, routine zero/span calibration using an external NH_3 standard gas was not conducted at the sampling site; however, instrument operation was checked using the internal diagnostic parameters, including sampled gas temperature, cell pressure, and laser offset.

The manufacturer-reported 1σ precision for NH_3 was 0.9, 0.3, and 0.1 ppb at averaging times of 1, 10, and 100 s, respectively. Assuming a detection limit of three times the 1σ noise

level, the corresponding estimated detection limits are 2.7, 0.9, and 0.3 ppb, respectively. The analyzer also had a manufacturer-reported maximum drift of less than 3 ppb for a 15 min average at STP over 24 h.

The sampling inlet was installed approximately 2 m above ground level, and the sampling line length was approximately 3 m. The inlet line was heated and maintained at approximately 40 °C during sampling to minimize NH₃ adsorption onto the sampling line and to prevent water condensation inside the line. Prior to sampling, the analyzer was warmed up and stabilized for at least 1 h. Data collection was started only after the sampled gas temperature and cell pressure had reached stable operating conditions, with the gas temperature maintained at 35–45 °C and the cell pressure at 75.00 Torr. The laser offset was also checked and adjusted, if necessary, so that the absorption peaks were properly centered on the instrument target lines.

Raw data were processed into 1 min averages. Data collected during the warm-up and stabilization period were excluded. Additional data were removed when the instrument diagnostics indicated unstable cell pressure, abnormal sampled gas temperature, or improper laser alignment. Periods potentially affected by inlet condensation or unstable instrument operation were also excluded from the final dataset.

2.3. Meteorological Data

Synchronized meteorological parameters—including air temperature, relative humidity, wind speed, wind direction, and solar radiation—were recorded at 6 s intervals during the April and May campaigns using an onsite weather station (SENSEKA, Remscheid, Germany). For the December campaign, due to onsite equipment maintenance, meteorological data were obtained from the Korea Meteorological Administration (KMA) at the Bukchuncheon station (37.947° N, 127.754° E), located approximately 2.5 km from the sampling site (Figure 1). Both locations are situated within the same relatively flat agricultural area and have similar surrounding land-use characteristics; therefore, the meteorological data were considered broadly representative of the local conditions at the sampling site, although small-scale differences between the two locations cannot be completely excluded.

2.4. Conditional Probability Function (CPF) Analysis

To identify the directional dependence of NH₃ sources and the influence of transport conditions, a Conditional Probability Function (CPF) analysis was performed using the openair package in R. CPF accounts for both wind direction and wind speed bins to estimate the probability of observing elevated concentrations. The CPF for a given bin ($\Delta\theta, \Delta u$) is defined as follows (Equation (1)).

$$CPF(\Delta\theta, \Delta u) = \frac{m(\Delta\theta, \Delta u)}{n(\Delta\theta, \Delta u)} \quad (1)$$

where $n(\Delta\theta, \Delta u)$ is the total number of observations within a specific wind direction sector $\Delta\theta$ and wind speed interval Δu , and $m(\Delta\theta, \Delta u)$ is the number of those observations that exceed a predefined high-concentration threshold.

CPF analyses were conducted using campaign-specific NH₃ concentration thresholds. The 75th percentile within each campaign was used as the main threshold to characterize elevated NH₃ conditions. To evaluate the robustness of the directional patterns, sensitivity analyses were additionally performed using the 65th and 85th percentile thresholds. Observation frequencies in each wind direction-wind speed bin were examined together with CPF values, and high-probability regions were interpreted cautiously when sample counts were low.

2.5. Leaf-Surface Moisture Inference: The Penman-Monteith (P-M) Equation

To infer the potential formation and removal of leaf-surface moisture, latent heat flux (LE) at the vegetation surface was estimated using the Penman-Monteith (P-M) equation, following the approach of Sentelhas and Gillespie [14]. In this study, LE was not directly measured by micrometeorological techniques; instead, it was used as a diagnostic indicator of the surface moisture exchange state. The governing equation is expressed as:

$$LE = - \frac{\left\{ sRn + \left[\frac{1200(es_{Ta} - ea)}{r_b} \right] \right\}}{(s + \gamma^*)} \quad (2)$$

where LE is the latent heat flux at the leaf surface, 1200 represents the approximate volumetric heat capacity of air ($\rho_a C_p$), obtained from an air density of approximately 1.2 kg m^{-3} and specific heat capacity of approximately $1000 \text{ J kg}^{-1} \text{ K}^{-1}$, s is the slope of the saturation vapor pressure curve ($\text{hPa } ^\circ\text{C}^{-1}$), Rn is the leaf net radiation, es_{Ta} is the saturated vapor pressure at the weather station air temperature (hPa), ea is the actual air vapor pressure (hPa), r_b is the boundary-layer resistance for heat transfer, and γ^* is the modified psychrometric constant. The term $(es_{Ta} - ea)$ represents the vapor pressure deficit (VPD), indicating the atmospheric drying capacity.

The slope of the saturation vapor pressure curve (s) was calculated from air temperature using a Tetens-type equation [15]:

$$s = \frac{4098 \times \left[0.6108 \times \exp\left(\frac{17.27 \times T}{T + 273.3}\right) \right]}{(T + 273.3)^2} \quad (3)$$

In Equation (3), the numerical constants are empirical coefficients in the saturation vapor pressure formulation and its temperature derivative, where T is air temperature in $^\circ\text{C}$. All vapor pressure terms used to calculate VPD, including saturation vapor pressure and actual vapor pressure, were calculated using consistent pressure units.

Because leaf net radiation was not directly measured, Rn was estimated from the measured solar radiation (Rg) using the empirical relationship proposed by Iziomon et al. [16], which was also evaluated by Sentelhas and Gillespie [14] for leaf wetness applications:

$$Rn = 0.837 \times (Rg \times 0.77) - 9.9275 \times \epsilon \left[5.67 \times 10^{-8} \times (273 + T)^4 \right] - 37.7 \quad (4)$$

where ϵ is the leaf emissivity. A value of $\epsilon = 0.98$ was adopted for the Italian ryegrass canopy, which was the dominant forage crop surrounding the sampling site. This value was selected to reflect the relatively dense and tall canopy structure of Italian ryegrass compared with general grassland surfaces. Sentelhas and Gillespie [14] previously validated the accuracy of this estimation method, reporting excellent agreement between measured and estimated net radiation with a confidence index of 0.9773 and a coefficient of determination (R^2) of 0.9794. In Equation (4), 0.77 represents the assumed shortwave absorptivity ($1 - \alpha$), 5.67×10^{-8} is the Stefan-Boltzmann constant, and 273 is used to convert air temperature from $^\circ\text{C}$ to K. The coefficients 0.837, 9.9275, and 37.7 are empirical coefficients of the net-radiation estimation model.

The boundary-layer resistance for heat transfer, r_b , was calculated as:

$$r_b = 307(d/U)^{1/2} \quad (5)$$

where d is the effective characteristic dimension of the vegetation and U is wind speed. Because Italian ryegrass at the study site reached approximately 94 cm in height, d was set to 0.94 m to represent the canopy-scale characteristic dimension. In Equation (5), 307 is the

coefficient used in the boundary-layer resistance formulation adopted from Monteith and Unsworth. For calm conditions, a minimum wind speed of 0.1 m s^{-1} was applied to avoid computational divergence.

The modified psychrometer constant, γ^* , was assigned according to the assumed direction of leaf-surface moisture exchange, following Sentelhas and Gillespie [14]. Values of 6.4 and 12.8 hPa K^{-1} were used for dew formation and evaporation, respectively, reflecting differences in heat and vapor transfer: dew formation assumes heat and moisture exchange on both sides of the leaf, whereas evaporation is assumed to occur from one side only. Under the sign convention adopted in this study, positive LE values ($LE > 0$) indicate condensation or wetness formation, in which atmospheric water vapor condenses onto the leaf surface. In contrast, negative LE values ($LE < 0$) indicate evaporative or drying conditions, in which liquid water on the leaf surface is removed through evaporation. This sign convention differs from the conventional micrometeorological convention, in which positive latent heat flux generally represents upward evaporation. Therefore, the estimated LE values were interpreted only within this diagnostic framework and were used as an auxiliary indicator of relative moisture-accumulating or evaporative conditions. They were not used as a direct measure of NH_3 emissions, manure-surface wetness, soil moisture, or fertilizer-related volatilization. The LE results were used to provide complementary context for interpreting short-term atmospheric NH_3 variability together with temperature, wind conditions, and CPF-based source-directional patterns.

3. Result and Discussion

3.1. Meteorological Factors Associated with Atmospheric NH_3 Variability

The mean NH_3 concentrations measured at the sampling site were $200.8 (\pm 91.1)$ (arithmetic mean $\pm$ standard deviation) ppb during the April campaign and $83.0 (\pm 34.4)$ ppb during the May campaign (Table 1), which were considerably higher than those reported for urban areas [17–19]. The mean concentration during the December campaign was much lower than those during the April and May campaigns, with a value of $27.9 (\pm 16.2)$ ppb. This contrast among the selected short-term observation periods suggests that NH_3 volatilization potential was substantially reduced under cold-season, non-fertilization conditions, rather than indicating a climatologically representative seasonal pattern [20]. The box-plot distributions of NH_3 and meteorological variables further illustrate the differences among the three short-term campaigns, showing higher NH_3 levels in April and May than in December, together with distinct differences in air temperature and solar radiation among the observation periods (Figure 2).

Table 1. Arithmetic mean ($\pm$ standard deviation) of NH_3 concentration, air temperature and relative humidity during the measurement periods.

Avg (± 1 SD)	4/11~4/12	5/23~5/24	12/10~12/11
NH_3 (ppb)	200.76 ± 91.07	82.95 ± 34.36	27.87 ± 16.24
Temp ($^{\circ}\text{C}$)	13.18 ± 5.65	21.22 ± 4.68	-0.79 ± 3.28
RH (%)	73.16 ± 20.69	64.82 ± 21.64	64.21 ± 17.88
Solar radiation (W m^{-2})	243.45 ± 321.07	239.81 ± 328.86	93.29 ± 155.27
Wind speed (m s^{-1})	0.75 ± 0.56	1.60 ± 1.64	0.46 ± 0.38

Because correlations calculated using pooled data from all campaigns may primarily reflect differences among observation periods rather than within-campaign micrometeorological relationships, pooled correlation results were not used for interpretation. Campaign-specific correlation analyses were conducted to examine the short-term relationships between NH_3 and micrometeorological variables during each observation period. To reduce short-term temporal dependence, the original 1 min data were first averaged to

10 min resolution. In the conventional Spearman correlation analysis, several micrometeorological variables showed statistically significant correlations with NH_3 during the April and May campaigns. However, high-time-resolution atmospheric observations commonly exhibit temporal autocorrelation, and p -values derived from conventional correlation tests may overestimate statistical significance when individual observations are treated as independent. To address this issue, we applied a moving block bootstrap approach to evaluate the robustness of the Spearman correlation coefficient. Unlike conventional bootstrap resampling of individual observations, the moving block bootstrap resamples consecutive time blocks and therefore preserves part of the temporal dependence structure within each block. In this study, a block size of 18 observations, corresponding to 3 h for the 10 min averaged data, was used, and 5000 bootstrap samples were generated. For each bootstrap sample, the Spearman correlation coefficient was recalculated, and the 95% confidence interval was estimated from the 2.5th and 97.5th percentiles of the bootstrap correlation distribution. Correlations were interpreted as robust descriptive associations, rather than independent causal effects, when the 95% confidence interval did not include zero [21,22].

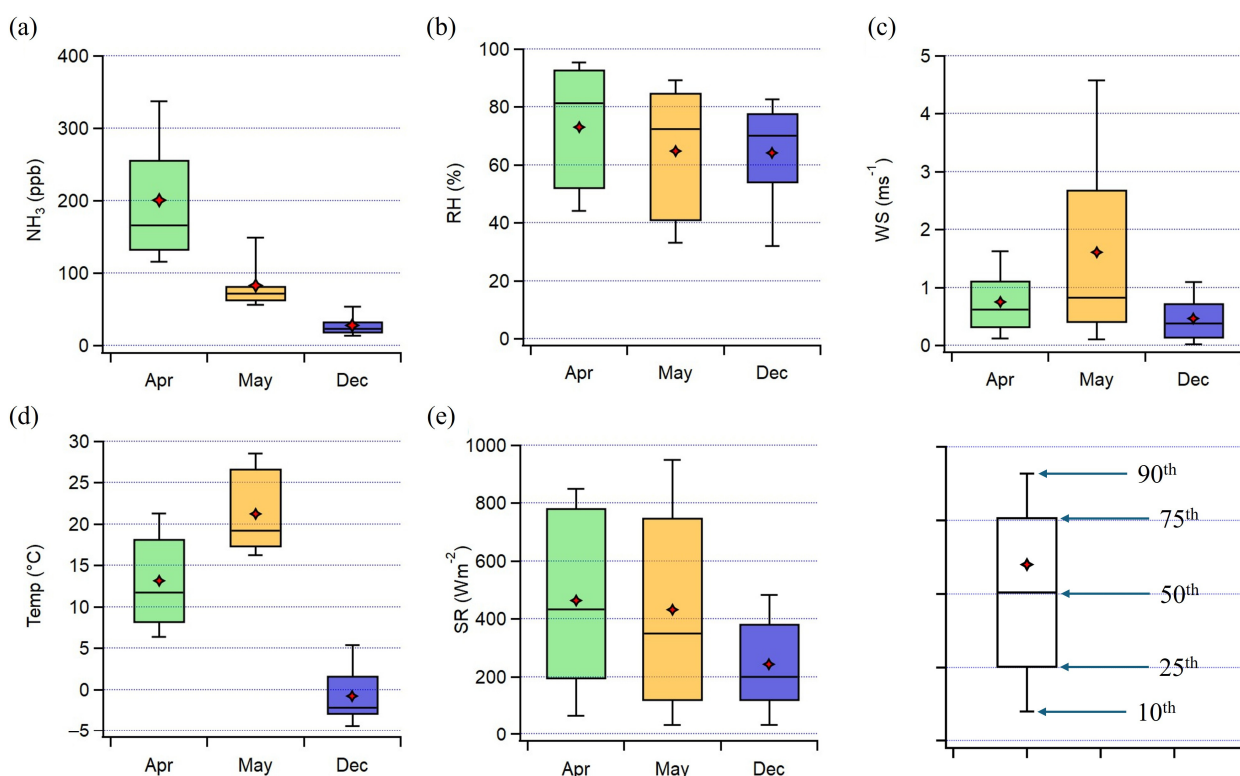


Figure 2. Box plot of NH_3 concentrations and meteorological variables during the April, May, and December measurement periods: (a) NH_3 , (b) relative humidity, (c) wind speed, (d) temperature and (e) solar radiation. Solar radiation was analyzed using daytime observations only. Red dots indicate arithmetic means.

Campaign-specific correlation results showed that the relationships between NH_3 and meteorological variables differed among the observation periods (Table 2). In April, NH_3 showed positive correlations with air temperature, wind speed, and solar radiation, with Spearman's rho values of 0.39, 0.46, and 0.60, respectively, and a negative correlation with relative humidity ($\text{rho} = -0.29$). However, after accounting for temporal dependence using moving block bootstrap confidence intervals, only wind speed and solar radiation retained robust positive associations with NH_3 . In May, NH_3 showed stronger positive correlations with air temperature and wind speed, with rho values of 0.68 and 0.65, respectively, and a strong negative correlation with relative humidity ($\text{rho} = -0.70$). These three relationships

remained robust after the moving block bootstrap analysis. In contrast, during the December campaign, NH₃ did not show any robust association with the micrometeorological variables examined.

Table 2. Campaign-specific Spearman's rank correlation coefficients between NH₃ and meteorological variables based on 10 min averaged data. Bootstrap confidence intervals were estimated using a moving block bootstrap approach.

Campaign	Temp					RH				
	Spearman's rho	p-value	CI _{low}	CI _{high}	Robust association based on bootstrap CI	Spearman's rho	p-value	CI _{low}	CI _{high}	Robust association based on bootstrap CI
Apr	0.39	<0.001	−0.07	0.75	No	−0.29	<0.001	−0.69	0.17	No
May	0.68	<0.001	0.26	0.94	Yes	−0.70	<0.001	−0.93	−0.31	Yes
Dec	0.07	0.417	−0.33	0.51	No	−0.01	0.937	−0.47	0.35	No

Campaign	WS					SR				
	Spearman's rho	p-value	CI _{low}	CI _{high}	Robust association based on bootstrap CI	Spearman's rho	p-value	CI _{low}	CI _{high}	Robust association based on bootstrap CI
Apr	0.46	<0.001	0.07	0.70	Yes	0.60	<0.001	0.18	0.81	Yes
May	0.65	<0.001	0.30	0.86	Yes	0.20	0.019	−0.66	0.81	No
Dec	−0.07	0.383	−0.27	0.22	No	0.02	0.791	−0.33	0.45	No

The higher NH₃ concentrations observed during the April and May campaigns than during the December campaign are consistent with the well-known temperature dependence of NH₃ volatilization. The mean air temperatures during the April, May, and December campaigns were 13.2 ± 5.7 °C, 21.2 ± 4.7 °C, and -0.8 ± 3.3 °C, respectively. In general, increasing temperature shifts the partitioning of ammoniacal nitrogen toward gaseous NH₃ and enhances NH₃ volatilization from manure, soils, and fertilized surfaces [23,24]. Therefore, the substantially lower NH₃ concentration observed in December is consistent with suppressed volatilization under cold conditions.

However, the comparison between April and May indicates that atmospheric NH₃ variability during the spring campaigns cannot be explained by temperature alone. Although the mean air temperature was lower in April than in May, NH₃ concentrations were higher in April. This difference may partly reflect campaign-specific source conditions, as the April campaign was conducted during the active fertilization period, whereas the May campaign was conducted approximately one month later. This suggests that, once thermal conditions are sufficient for NH₃ volatilization, atmospheric NH₃ concentrations may become more strongly influenced by other factors, such as emission strength, substrate availability, surface moisture conditions, source direction, and wind-dependent transport, dilution, or local accumulation. For example, Pedersen et al. [20] reported in a chamber study that NH₃ emissions following slurry-based fertilizer application increased markedly within the temperature range of 5–14 °C, whereas further warming above this range did not result in a clear additional increase in emissions. This suggests that the temperature response of NH₃ volatilization may become less pronounced beyond a certain temperature range, particularly when other limiting factors, such as total ammoniacal nitrogen (TAN; the sum of dissolved NH₃ and NH₄⁺), pH, exposed manure or soil surface area, and transfer resistance, become more important. This result refines previous understanding by showing that the well-known temperature dependence of NH₃ volatilization may be modified over short timescales by campaign-specific source conditions, substrate availability, wind-directional transport, and surface moisture-exchange conditions. Thus, in peri-urban environments where livestock and agricultural sources coexist in close proximity, the meteorological variables associated with NH₃ variability may differ among short observation periods rather than following a single generalized temperature–NH₃ relationship.

The role of relative humidity also differed among the campaigns. In May, NH₃ showed a strong negative correlation with relative humidity ($\rho = -0.70$), and this relationship

remained robust after the moving block bootstrap analysis. This suggests that NH_3 concentration tended to increase under drier conditions during the May campaign. Lower relative humidity can promote surface drying and evaporation of thin water films, which may shift the dissolved $\text{NH}_4^+/\text{NH}_3$ equilibrium toward gaseous NH_3 and enhance re-emission to the atmosphere. Conversely, under humid or dew-forming conditions, gaseous NH_3 can be absorbed into surface moisture films and temporarily stored as dissolved $\text{NH}_3/\text{NH}_4^+$. Subsequent drying of these films can then promote the release of stored NH_3 back to the atmosphere [10,13,25]. Therefore, the negative NH_3 –RH relationship observed in May likely reflects the influence of surface moisture exchange and drying processes on NH_3 volatilization and re-emission, although RH itself should not be regarded as a direct measure of surface wetness.

In April, NH_3 also showed a negative correlation with relative humidity ($\rho = -0.29$), but this relationship was not robust after accounting for temporal dependence. This suggests that RH alone did not consistently explain NH_3 variability during the April campaign. Instead, the robust positive associations with wind speed and solar radiation suggest that atmospheric NH_3 variability shortly after fertilization was linked to the combined influence of wind-related transport or mixing processes and solar-radiation-related surface heating. In December, no robust relationship between NH_3 and RH was observed, indicating that RH-related surface moisture processes were not a dominant driver of atmospheric NH_3 variability under cold-season conditions. Overall, RH should not be interpreted as a direct controlling factor by itself, but rather as an indicator of surface moisture conditions that interact with temperature, solar radiation, wind speed, and surface–atmosphere exchange.

Accordingly, the following sections further examine atmospheric NH_3 variability from two complementary perspectives: first, because the effect of wind speed may reflect dilution, local accumulation, or directional transport depending on wind direction and source location, the wind-directional characteristics of elevated- NH_3 events are examined using CPF analysis. Second, the role of latent heat flux is evaluated as an auxiliary indicator of meteorological conditions potentially favorable for surface moisture exchange and NH_3 volatilization, rather than as a direct measure of NH_3 emissions.

3.2. Wind-Directional Characteristics of NH_3 Variability

To identify the wind-direction and wind-speed conditions associated with elevated NH_3 concentrations, CPF analysis was conducted for each measurement period (Figure 3). Because wind speed can have competing effects on atmospheric NH_3 concentrations, it was not interpreted as an independent meteorological control. Low wind speeds may favor local accumulation near the sampling site, whereas higher wind speeds may either enhance dilution or transport NH_3 from upwind livestock and agricultural sources when wind direction is aligned with those sources. Therefore, wind-speed effects were interpreted together with wind direction and the spatial distribution of potential source areas. To assess the robustness of the CPF patterns, sensitivity analyses were performed using campaign-specific NH_3 thresholds defined at the 65th, 75th, and 85th percentiles, and the observation frequency in each wind-direction–wind-speed bin was also examined (Figure 3). Across the different threshold settings, the main high-CPF sectors remained generally consistent within each campaign, indicating that the identified directional patterns were not strongly dependent on threshold selection.

During the April and May campaigns, high CPF values were mainly observed under southwesterly to southerly winds, particularly at moderate to relatively high wind speeds (Figure 3a,b). Because livestock facilities, manure-related activity areas, and agricultural fields are located southwest to south of the sampling inlet, these wind conditions were

consistent with transport from livestock- and agricultural-influenced sectors toward the measurement site. As shown in Figure 1c, agricultural areas are widely distributed within a 10 km radius of the sampling site, particularly to the southwest. In addition, the April and May campaigns showed high CPF values over a broader range of wind direction–wind speed conditions than the December campaign. These results may be consistent with contributions from multiple agricultural source areas distributed southwest of the site, rather than with the influence of a single dominant source. Nearby forage crop fields and other agricultural areas may also have been associated with the elevated NH_3 concentrations observed during the selected spring campaigns. However, the observation frequency in some wind direction–wind speed bins with high CPF values was low during April, and the corresponding results should therefore be interpreted with caution. Nevertheless, high CPF values repeatedly appeared across adjacent wind direction–wind speed bins, and the main directional pattern was maintained under the different thresholds corresponding to the upper 35%, 25%, and 15% of NH_3 concentrations. These patterns suggest that the elevated NH_3 concentrations observed during the April campaign were more consistent with short-term influence from source areas southwest of the site during the active fertilization period than with an artifact arising solely from low sample counts in individual bins.

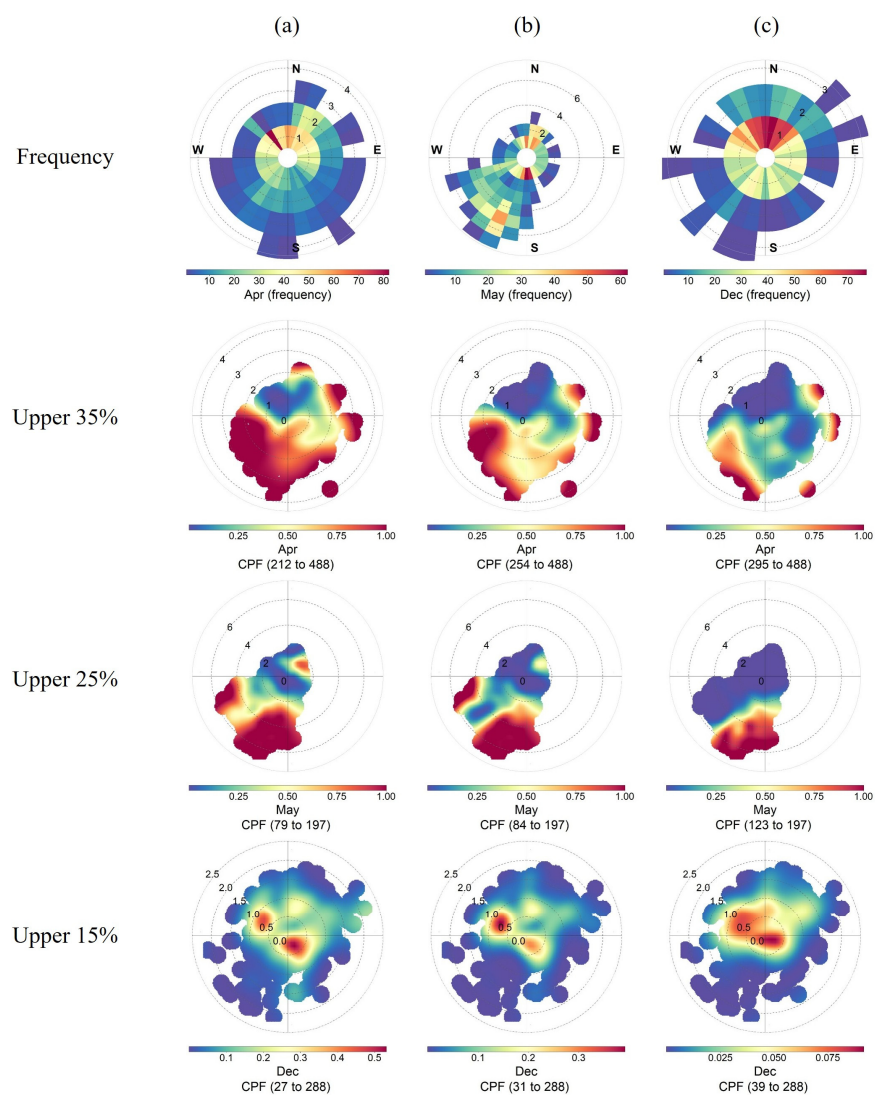


Figure 3. Wind-direction and wind-speed dependence of atmospheric NH_3 concentrations during the selected observation periods. Columns correspond to (a) 11–12 April, (b) 23–24 May, and (c) 10–11 December. The top row shows the observation frequency in each wind direction–wind

speed bin. The lower rows present conditional probability function (CPF) results calculated using campaign-specific NH_3 concentration thresholds corresponding to the upper 35%, 25%, and 15% of each campaign dataset (i.e., the 65th, 75th, and 85th percentiles, respectively). The corresponding threshold concentrations were 211.8, 253.6, and 295.3 ppb for April; 78.8, 83.7, and 123.1 ppb for May; and 27.4, 31.2, and 38.9 ppb for December. The numbers of observations exceeding each threshold were 505, 361, and 217 for April; 484, 346, and 208 for May; and 505, 361, and 217 for December. The identical sample counts for April and December reflect the fact that both campaigns covered a full 24 h period, whereas the May campaign lasted 23 h.

In contrast, the CPF pattern in the December campaign showed generally lower values than those in the April and May campaigns, with high CPF values confined mainly to a limited number of wind direction-wind speed bins under low-wind or near-calm conditions (Figure 3c). Although some high-CPF regions corresponded to the direction of nearby livestock facilities, the increased probability of elevated NH_3 under low-wind conditions suggests that local NH_3 accumulation due to limited atmospheric dispersion may have acted together with the direct influence of local emissions. In particular, when the threshold was increased to the upper 15%, the overall CPF values decreased to below 0.1 and the high-CPF regions became more dispersed rather than forming a stable directional sector. This suggests that the highest NH_3 concentrations in the December campaign may have been associated with the combined effects of intermittent local emissions and stagnant conditions, rather than with persistent transport from a specific direction.

Overall, the CPF analysis indicates that the wind direction and wind speed conditions associated with elevated NH_3 , as well as the patterns of source influence, differed among the campaigns. During the April and May campaigns, the observed directional patterns were consistent with contributions from livestock- and agricultural source areas located southwest to south of the site. Thus, the positive association between wind speed and NH_3 during these campaigns should be interpreted as evidence of wind-assisted directional transport from nearby upwind source areas, rather than as a simple wind-speed effect. In contrast, during the December campaign, nearby local sources and accumulation under weak-wind or stagnant conditions may have acted together.

However, this CPF-based source interpretation is subject to an important limitation. Detailed field-activity records, including fertilizer application, manure spreading, livestock housing operation, ventilation, cleaning events, crop type, and soil conditions, were not available during the observation periods. Therefore, although the CPF patterns were consistent with transport from nearby livestock and agricultural source areas, the specific activities responsible for individual atmospheric NH_3 enhancement events could not be directly identified.

3.3. Surface-Energy and Moisture Exchange Conditions Inferred from Latent Heat Flux

Latent heat flux was estimated using the Penman–Monteith equation to examine whether surface-energy and moisture-exchange conditions were related to short-term NH_3 variability (Figure 4). In this study, the Penman–Monteith-derived latent heat flux was not interpreted as a direct measure of NH_3 volatilization from manure, nor as a flux representing only leaf evaporation or transpiration. Because the surrounding area includes heterogeneous surfaces, such as vegetation, bare soil, livestock facilities, manure-related surfaces, and agricultural fields, the estimated latent heat flux should not be interpreted as a footprint-integrated flux representative of all surrounding surface types. Instead, it was used as a vegetation-based diagnostic indicator of relative evaporative and moisture-accumulating conditions during each observation period. Leaf-surface moisture exchange, soil moisture, manure-surface wetness, and fertilizer-related volatilization are related but distinct processes; therefore, the latent heat flux analysis was used only to provide comple-

mentary meteorological context for interpreting atmospheric NH_3 variability together with temperature, wind conditions, and CPF-based source-directional patterns.

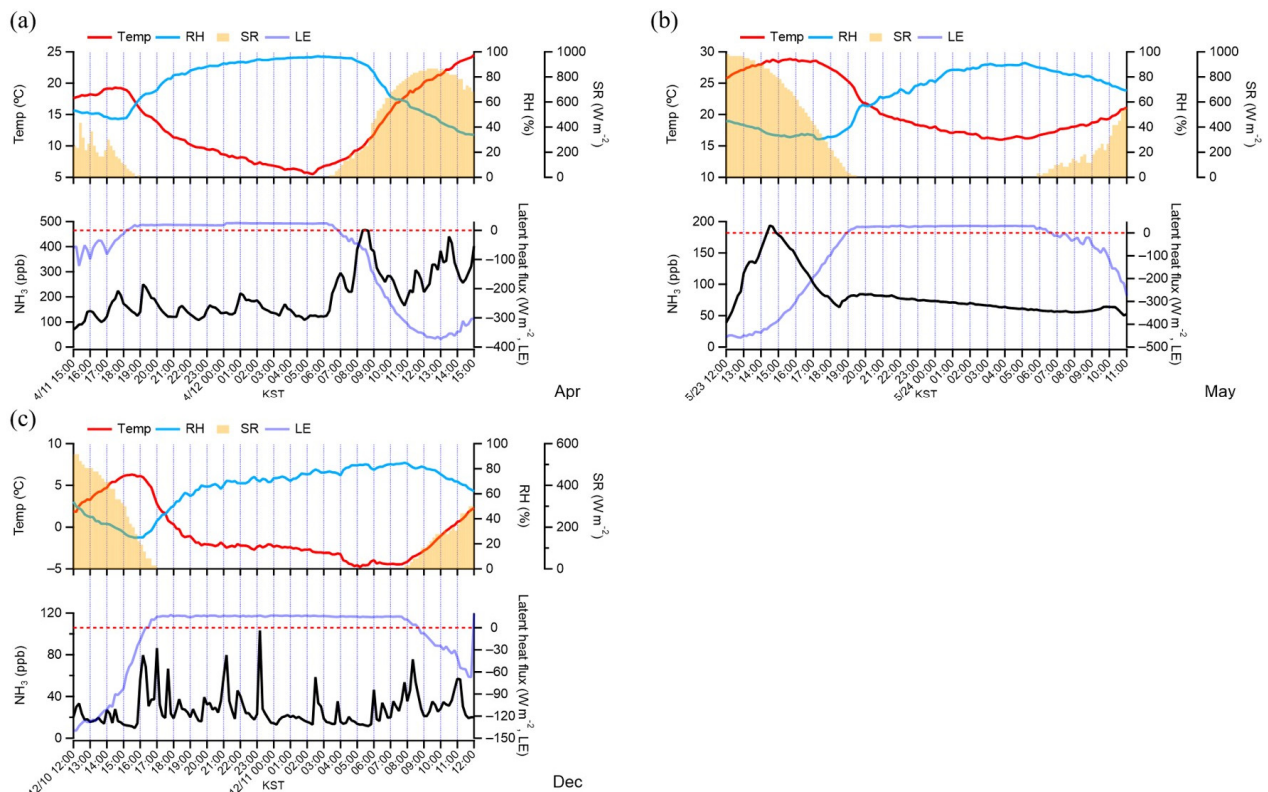


Figure 4. Time series of meteorological parameters and NH_3 concentrations during the measurement periods. (a) 11–12 April, (b) 23–24 May, and (c) 10–11 December. Upper panels show air temperature (Temp), relative humidity (RH), and solar radiation (SR), while lower panels show NH_3 concentration, latent heat flux (LE). The dashed line in the LE plots indicates the evaporation–condensation boundary. All times are given in Korea Standard Time (KST).

Under the sign convention adopted in the Penman–Monteith leaf-wetness framework used here, positive latent heat flux indicates condensation or moisture accumulation, whereas negative latent heat flux indicates evaporative conditions and surface moisture loss. This sign convention differs from the conventional micrometeorological convention, in which positive latent heat flux generally represents upward evaporation. Therefore, the LE values in this study should be interpreted only within this diagnostic framework. Strongly negative latent heat flux values do not directly quantify NH_3 emissions, manure-surface wetness, soil moisture, or fertilizer-related volatilization. Rather, they indicate periods when meteorological conditions favored evaporation and surface drying in the Penman–Monteith framework. These conditions may be broadly consistent with an environment favorable for NH_3 volatilization from wet or NH_3 -rich surfaces, but the specific contributions of leaf surfaces, soils, manure-related surfaces, and fertilized fields cannot be separated using this approach [11,12].

During April and May, NH_3 concentrations showed recurrent daytime enhancement, particularly from morning to afternoon (Figure 4a,b). These increases coincided with increasing solar radiation, higher air temperature, and pronounced negative latent heat flux values. This pattern suggests that NH_3 concentrations tended to increase under meteorological conditions potentially favorable for evaporation, surface drying, and surface-atmosphere exchange. Importantly, this interpretation is consistent with the CPF results: high NH_3 concentrations in April and May were preferentially associated with southwest-

erly winds, indicating the potential influence of nearby livestock and agricultural source areas (Figure 3a,b). Therefore, the combined CPF and latent heat flux results suggest that the NH_3 enhancements observed during the April and May campaigns were associated with the co-occurrence of source-directional influence from livestock- and agriculture-influenced sectors and meteorological conditions potentially favorable for volatilization and surface-atmosphere exchange. However, this co-occurrence should not be interpreted as direct evidence that the Penman–Monteith-derived latent heat flux controlled or quantified NH_3 emissions.

The December results differed substantially from those in April and May (Figure 4c). NH_3 concentrations were much lower, daytime enhancement was weaker and less recurrent, and the CPF pattern did not show a strong southwesterly source signal (Figure 3c). In addition, negative latent heat flux values were much smaller in magnitude than those observed in April and May (Figure 4). The minimum latent heat flux values were -373 W m^{-2} in April and -459 W m^{-2} in May, whereas the minimum value in December was only -139 W m^{-2} . This suggests that the estimated evaporative surface-exchange signal was much weaker in December.

The weaker latent heat flux signal observed in December indicates that surface energy and moisture exchange conditions were less active than in April and May. Such conditions may have been less favorable for volatilization from manure and other NH_3 -rich wet surfaces and may therefore have been consistent with the lower NH_3 concentrations observed during the December campaign. However, because manure surface temperature, soil moisture, surface wetness, and NH_3 flux were not directly measured, the relative contribution of reduced volatilization cannot be quantified, and other processes such as reduced source activity, deposition, and atmospheric dilution may also have contributed.

These results indicate that the Penman–Monteith-derived latent heat flux provides process-oriented information on evaporative surface-exchange conditions but should not be interpreted as a direct measure of NH_3 emissions. In April and May, daytime NH_3 enhancement coincided with evaporative conditions and southwesterly source-directional patterns, whereas December showed weaker evaporative conditions and a less distinct source-directional pattern. Thus, the LE results provide complementary context for interpreting campaign-specific atmospheric NH_3 variability, although the direct contribution of volatilization to the observed concentrations cannot be quantified. Because a full sensitivity analysis of Penman–Monteith parameters was not conducted, the LE results were used only qualitatively, and conclusions were based primarily on the co-occurrence of evaporative conditions, CPF source-directional patterns, and observed NH_3 variations rather than on the absolute magnitude of LE.

4. Conclusions

This study investigated atmospheric NH_3 variability at a peri-urban site influenced by nearby livestock and agricultural activities, focusing on differences among selected short-term observation periods, source-directional characteristics, and micrometeorological factors. The mean NH_3 concentrations were 200.8 ± 91.1 ppb in April, 83.0 ± 34.4 ppb in May, and 27.9 ± 16.2 ppb in December. The much lower concentration during the December campaign suggests reduced NH_3 volatilization potential under low-temperature, non-fertilization conditions.

Campaign-specific correlation analyses showed that the relationships between atmospheric NH_3 concentrations and micrometeorological variables differed among the observation periods, with robust associations observed in April and May but not in December. Moreover, NH_3 concentrations were higher in April than in May despite the lower mean temperature, indicating that short-term atmospheric NH_3 variability at this site

could not be explained by temperature alone. Instead, local emission intensity, fertilization or manure-management activities, source direction, surface–atmosphere exchange, and wind-dependent transport, dilution, or local accumulation likely acted together.

CPF analysis showed that high NH_3 concentrations in April and May were preferentially associated with southwesterly winds, consistent with the influence of nearby livestock and agricultural source areas. This directional pattern was much weaker in December, consistent with reduced volatilization potential and possible intermittent local accumulation under low-wind conditions.

The Penman–Monteith-derived latent heat flux provided complementary information on relative evaporative and moisture-accumulating conditions, rather than a direct measure of NH_3 emissions. In April and May, elevated NH_3 concentrations coincided with evaporative conditions and southwesterly source-directional patterns, whereas December showed weaker evaporative conditions and a less distinct CPF pattern. These differences provide process-oriented context for the campaign-specific atmospheric NH_3 variability but do not quantify the direct contribution of volatilization to the observed concentrations.

Overall, atmospheric NH_3 variability during the selected campaigns appeared to be influenced by combined effects of source location, emission activity, temperature-dependent volatilization, wind-directional transport or local accumulation, and evaporative surface-exchange conditions. RH alone was insufficient to explain atmospheric NH_3 variability because surface moisture can mediate both uptake and re-emission depending on drying and evaporation conditions. Given that NH_3 is an important precursor of secondary inorganic aerosols, the elevated NH_3 concentrations observed during the spring campaigns may have potential implications for $\text{PM}_{2.5}$ formation. However, because particulate NH_4^+ , NO_3^- , SO_4^{2-} , acidic gases, and aerosol acidity were not measured in this study, the role of NH_3 in secondary aerosol formation could not be directly evaluated. Future studies should incorporate simultaneous gas- and particle-phase measurements to assess how livestock- and agriculture-related NH_3 contributes to secondary inorganic aerosol formation.

An important limitation of this study is the lack of detailed field-activity records during the observation periods, including fertilization, manure spreading, manure storage, livestock housing operations, ventilation, cleaning events, crop type, and soil conditions. Accordingly, although the observed NH_3 variability was associated with meteorological conditions, boundary layer development, and source-direction patterns, the specific agricultural or livestock activities responsible for individual NH_3 enhancement events could not be directly identified. Interpretations regarding source processes and campaign-specific emission conditions should therefore be viewed as inferential rather than definitive. Future studies should combine detailed activity records with longer-term NH_3 measurements and direct flux observations to more clearly separate source-specific emissions from micrometeorological influences.

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