



Model Evaluation based on a Relationship Analysis between the Emission and Concentration of Atmospheric Ammonia in the Kanto Region of Japan

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ABSTRACT

This study aims to evaluate the performance of the Air Quality Model (AQM) for the seasonal and spatial distribution of the NH_3 concentration in the atmosphere. To obtain observational data for the model validation, observations based on biweekly sampling have been conducted using passive samplers since April 2015 at multiple monitoring sites in the Tokyo metropolitan area. AQM, built based on WRF/CMAQ, was applied to predict the NH_3 concentration observed from April 2015 to March 2016. The simulation domain includes the Kanto region, which is the most densely populated area in Japan. Because the area also contains large amount of livestock, especially in its northern part, the density of the NH_3 emissions derived from human activities and agriculture there are estimated to be the highest in Japan. In the model validation, the model overestimated the observed NH_3 concentration in the summer season and underestimated it in the winter season. In particular, the overestimation in the summer was remarkable at a rural site (Komae) in Tokyo. It was found that the overestimation at Komae was caused by the transportation of NH_3 emitted in the northern part of the Kanto region during the night. It is suggested that the emission input used in this study overestimated the NH_3 emission from human sources around the Tokyo suburbs and agricultural sources in the northern part of the Kanto region in the summer season. In addition, the current emission inventories might overestimate the difference of the agricultural NH_3 emissions among seasons. Because the overestimation of NH_3 in the summer causes an overestimation of NO_3^- in $\text{PM}_{2.5}$ in the AQM simulation, further investigation is necessary for the seasonal variation in the NH_3 emissions.

Key words: Ammonia, Air quality model, Secondary inorganic aerosol, Emission sources, Passive sampler

1. INTRODUCTION

In practice, $\text{PM}_{2.5}$ is created by the accumulation and condensation of various substances such as Secondary Inorganic Aerosol (SIA), Elemental Carbon (EC), Organic Carbon (OC) and Metals. Especially, it is important to assess SIA, which cause severe $\text{PM}_{2.5}$ pollution in urban areas (e.g., Hasegawa *et al.*, 2014; Ueno *et al.*, 2011; Yonemochi and Umezawa, 2010). SIA can be generally divided into “ammonium sulfate $[(\text{NH}_4)_2\text{SO}_4]$ ” and “ammonium nitrate $[\text{NH}_4\text{NO}_3]$ ”. They are created as secondary particles in the atmosphere primarily by heterogeneous reactions between acid gases (SO_2 and NO_x) and alkaline gases (NH_3) on suspended particulate matter. In this context, NH_3 in the atmosphere has an important role for the creation of SIA.

In Japan, the environmental air quality standard for $\text{PM}_{2.5}$ states that the daily and annual averaged concentrations should not exceed $35 \mu\text{g m}^{-3}$ and $15 \mu\text{g m}^{-3}$, respectively (established in 2009). The achievement rates for $\text{PM}_{2.5}$ environmental standard in recent years were 2015: 74.5%, 2014: 37.8% and 2013: 16.1% at the Ambient Air Pollution Monitoring Stations (Ministry of Environment, Japan, 2017). Although the rate has been getting better, it is still important to make an effort to reduce the SIA concentration because it is one of the major components of $\text{PM}_{2.5}$ in urban areas. To evaluate the mechanism of increasing concentrations and to consider control measures for SIA, it is necessary to develop a numerical model, which can take the emissions of precursors and the physical/chemical processes in the atmosphere into account properly (e.g., Chatani 2015; Shimadera *et al.*, 2014; Morino *et al.*, 2010; Carmichael *et al.*, 2008). Sakurai *et al.* (2015) reported that the Air Quality Model (AQM) based on WRF/CMAQ could reproduce the weekly averaged concentration of SO_4^{2-} in $\text{PM}_{2.5}$ observed in western Tokyo from August 2009 to August 2011. However, it was also reported that the model overestimated the observed NO_3^- in $\text{PM}_{2.5}$ in the summer seasons. Other

previous studies have also reported overestimations of NO_3^- in $\text{PM}_{2.5}$ (e.g., Shimadera *et al.*, 2014; Hayami, 2013; Morino *et al.*, 2010), and it has been pointed out that (i) uncertainties could exist in the seasonal fluctuation of the NH_3 emission as input data for the model, (ii) uncertainties could exist in the model performance regarding the concentrations and the dry deposition process for the precursors (HNO_3 and NH_3), (iii) the model reproduced higher HNO_3 concentrations under the condition of overestimated O_3 , and (iv) there could be an artifact in the observational data based on the volatilization of ammonium nitrate. An artifact due to volatilization would mean that the ammonium nitrate (particulate) volatilizes into nitric acid gas and ammonia gas on the particle collection filter in the official method, so that the actual particle concentration is underestimated.

As for the uncertainties regarding ammonium nitrate, this study focuses on the performance of the AQM for the seasonal and spatial distribution of the NH_3 concentration in the atmosphere. Because NH_3 is not regarded as an air pollutant, monitoring networks and official observation methods for NH_3 have not been established in Japan. Because information for the seasonal and spatial distribution of the NH_3 concentration is limited in Japan, in this study, observations of the atmospheric NH_3 concentration were conducted at multiple sites in the Tokyo metropolitan area using passive samplers. In addition, a simulation based on WRF/CMAQ was applied to predict the NH_3 concentration observed during the period from April 2015 to March 2016.

2. METHODOLOGY

2.1 Observation of Atmospheric Ammonia

In Japan, a national standard has been established for acid gases (SO_2 and NO_2) and their concentrations have been monitored by a national monitoring network that consists of approximately 1500 sites throughout Japan (Ministry of Environment, Japan, 2017). Conversely, because NH_3 is not regarded as an air pollutant, as mentioned above, monitoring networks and official observation methods for NH_3 have not yet been established in Japan. In this context, for this study, observations of the NH_3 concentration have been conducted at multiple sites in the Tokyo metropolitan area since April 2015. This manuscript introduces the observational results from April 2015 to March 2017 at the five sites shown in Fig. 1. The sites were located in urban (Shinjuku), rural (Hino and Komae), remote (Tsukui), and agricultural areas (Hiratsuka).

In this study, a passive sampler, manufactured by Ogawa Shokai Co., Ltd., was adopted as the observational method for the NH_3 concentration. The Ogawa passive sampler has been used for NH_3 sampling inside and outside of Japan (e.g., Matsumoto *et al.*, 2010; Roadman *et al.*, 2003). The sampling was carried out every two weeks basically. Captured NH_3 on the sampling filter was extracted into pure water (5 mL), and the amount was detected using the Flow Injection Analysis method. The concentration was derived by deducting a blank value on unused filter from the collected NH_3 .

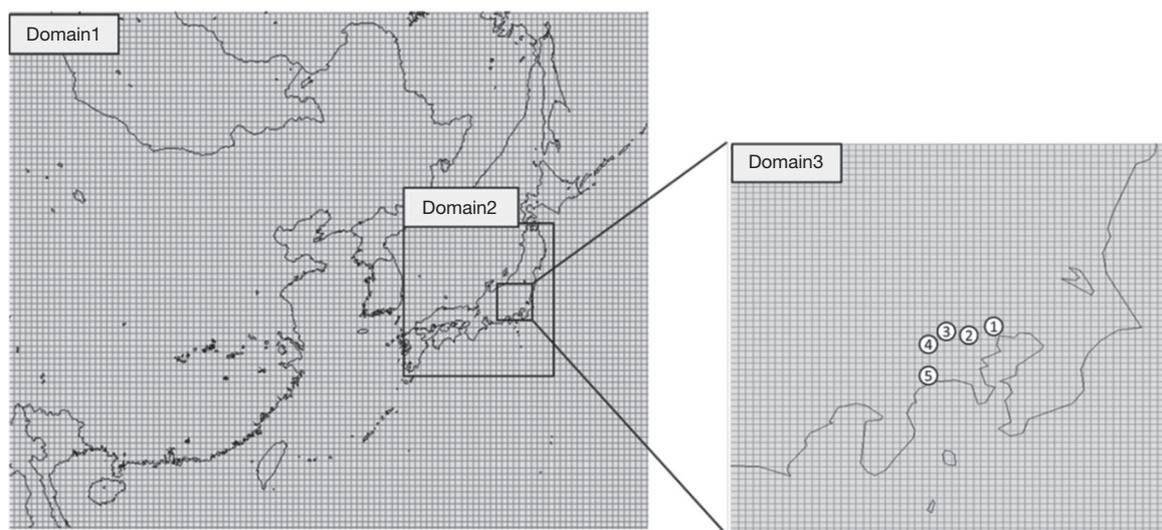


Fig. 1. Model domains and locations of the five monitoring sites for atmospheric NH_3 to be compared with the calculated concentrations (1: Shinjuku [urban], 2: Komae [rural], 3: Hino [rural], 4: Tsukui [remote], and 5: Hiratsuka [agricultural]).

2.2 Model Description

In this study, a modeling analysis was performed using CMAQ (Community Multiscale Air Quality; Byun and Ching, 1999) version 4.7.1. The selected chemical reaction scheme was the same as that of Sakurai *et al.* (2015). The meteorological data in the three-dimensional space was calculated using WRF (Weather Research Forecast model; Skamorock *et al.*, 2008) version 3.7.1. The global objective analysis data (FNL) of the National Centers for Environmental Prediction (NCEP) were used in the WRF simulation as the initial and boundary conditions. In addition, RGT_SST of NCEP was used for the sea surface temperature. The simulation period was from March 2015 to March 2016. As shown in Fig. 1, the modeling domain of WRF had a nesting system with grid resolutions of 45 km (East Asia as Domain 1), 15 km (Japan as Domain 2), and 5 km (the Kanto region as Domain 3). The domain sizes were $3,825 \times 4,950 \text{ km}^2$ for Domain 1, $1,095 \times 1,200 \text{ km}^2$ for Domain 2, and $335 \times 335 \text{ km}^2$ for Domain 3 with the domain center at 36° N and 140° E . The vertical layers consisted of 30 sigma-pressure layers from the surface to 100 hPa with the top height of the lowest layer being approximately 22 m.

Air quality simulations based on CMAQ were conducted only in Domains 2 and 3 due to the limited information regarding the continental emission inventories during the recent years. Inland anthropogenic emissions in Domains 2 and 3 were derived from EAGrid-Japan 2010 (Fukui *et al.*, 2014). NO_x emissions from vehicles were modified by reducing them by approximately 75% according to the reduction rate of the annual averaged concentration of NO_x observed at all Motor Vehicle Exhaust Monitoring Stations in Japan from 2010 (416 sites) to 2015 (413 sites) (Ministry of

Environment, Japan, 2017). Ship emissions were derived from the emission inventories developed by the Ocean Policy Research Foundation (2013). In addition, GEIA (Global Emission Initiative database) and GFED Ver. 3.1 (van der Werf *et al.*, 2010) were applied for vegetable origin VOCs and biomass burning origins, respectively. The volcanic origin SO_2 emissions were also taken into account in the same way as in Sakurai *et al.* (2015). The boundary concentration of air pollutants in Domain 2 was derived from MOZART-4 (Model for Ozone and Related chemical Tracers version 4) (Emons *et al.*, 2010).

3. RESULTS AND DISCUSSION

3.1 Observation Results for Atmospheric Ammonia Concentration

Fig. 2 introduces the seasonal variation in the NH_3 concentrations observed from April 2015 to March 2017 at the sites indicated in Fig. 1. Because the observations indicated that the level and seasonal variation in the observed concentrations at the Hino (rural), Komae (rural), and Tsukui (remote) sites were nearly the same, the observations at the Komae (rural) and Tsukui (remote) sites were terminated in April 2016.

It appears that the observed concentrations increased toward the warm seasons, and relatively higher concentrations were observed at the Shinjuku (urban) and Hiratsuka (agricultural) sites throughout the study period. Fig. 3 shows the horizontal distribution of the annual NH_3 emission used in the simulation for Domain 3 (5-km grid resolution). The annual emission amounts of NH_3 in each grid were Shinjuku: $4.92 \text{ ton km}^{-2} \text{ year}^{-1}$, Komae: $4.52 \text{ ton km}^{-2} \text{ year}^{-1}$, Hino: 3.24

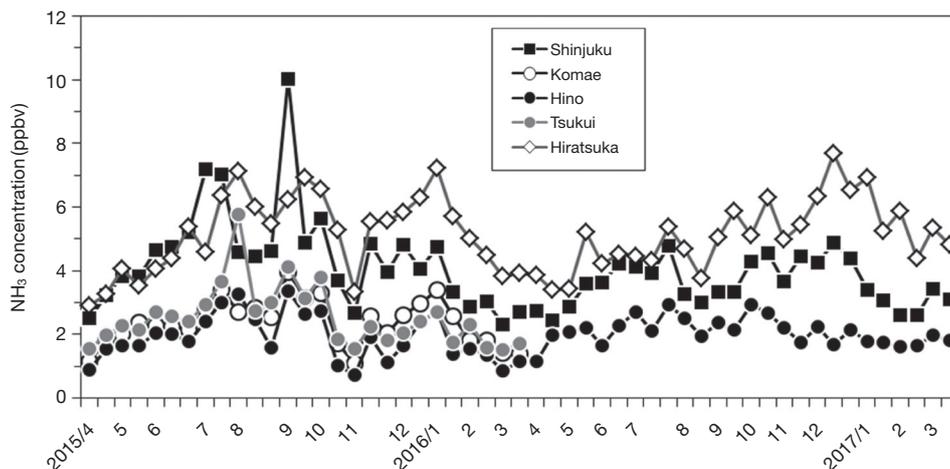


Fig. 2. Seasonal variation in the NH_3 concentrations observed from April 2015 to April 2017 at the sites indicated in Fig. 1.

ton km⁻² year⁻¹, Tsukui: 1.36 ton km⁻² year⁻¹, and Hiratsuka: 4.60 ton km⁻² year⁻¹. Fukui *et al.* (2014) estimated that the annual emission amount of NH₃ in Japan was 404,393 ton year⁻¹ in 2010 and that the agricultural and human sources contributed 66% and 18% of the total amount, respectively. The large emission amounts at the Shinjuku (urban) and Hiratsuka (agricultural) sites originate from human and agricultural emission sources, respectively. Therefore, for these two sites, the relationship between the emission amount and the observed concentrations was consistent. Conversely, even though the emission amount at the Komae (rural) site, which originated primarily from human sources, was nearly as large as that at the Shinjuku (urban) and Hiratsuka (agricultural) sites, a relatively lower concentration of NH₃ was observed there. The Komae (rural) site is located in a suburb of Tokyo, and the human activities there are substantially different from those near the Shinjuku (urban) site. Therefore, the inconsistency between the emission amount and the concentration at the Komae (rural) site suggests that the estimated emissions around the Tokyo suburb might

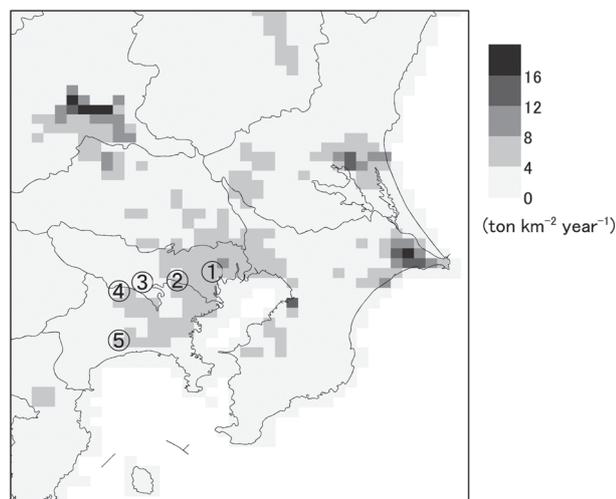


Fig. 3. Horizontal distribution of the annual NH₃ emissions (ton km⁻² year⁻¹) used in the simulation for Domain 3 (5-km grid resolution).

be overestimated compared to the actual situation.

Seasonal averaged concentrations over the two years at the Shinjuku (urban), Hino (remote), and Hiratsuka (agricultural) sites are summarized in Table 1. The table also shows the proportion of NH₃ emissions for each season compared to the annual amount. In general, the emission strength of NH₃ based on volatilization increases as the temperature rises. Accordingly, the emission ratio reached its maximum in the summer season (from July to September) at all sites. Even though the seasonal averaged concentrations at the Shinjuku (urban) and Hino (rural) sites reached their maximums in the summer season, the concentration at the Hiratsuka (agricultural) site became high not only in the summer but also in the winter (from January to March) and the autumn (from October to December). The inconsistency between the emission and the observed concentration at the Hiratsuka (agricultural) site suggests that there might be uncertainties in the seasonal variation in the NH₃ emitted from agricultural sources.

3.2 Model Validation based on the Observations

To examine the model performance for atmospheric ammonia, comparisons between the observed and simulated NH₃ were conducted at the five sites, as shown in Fig. 4, during the period from April 2015 to March 2016. In addition, Table 2 shows statistics of mean value of simulated concentration (mean Sim.), mean value of observed concentration (mean Obs.), normalized mean bias (NMB) and correlation coefficient (*r*) between the simulation and observation during the period from April 2015 to March 2016. NMB indicates a level of model overestimation (plus value) and underestimation (minus value), and the calculated NMB in this study were ranged from -56% (at Hiratsuka in the cold season) to 170% (at Hino in the warm season).

It was found that the model overestimated the observed NH₃ concentration in the warm season and underestimated the concentration in the cold season. In particular, the values of NMB in the warm season at Hino (rural) and Komae (rural) sites reached 170% and 70%,

Table 1. Averaged concentrations (ppbv) and the ratio (%) of emission in each season at the Shinjuku, Hino, and Hiratsuka sites.

	Shinjuku		Hino		Hiratsuka	
	Conc. (ppbv)	Emis. (%)	Conc. (ppbv)	Emis. (%)	Conc. (ppbv)	Emis. (%)
Jan-Mar as winter (n = 12)	3.10	14	1.63	11	5.23	10
Apr-Jun as spring (n = 13)	3.65	27	1.76	28	4.01	28
Jul-Sep as summer (n = 14)	4.90	39	2.53	43	5.44	45
Oct-Dec as autumn (n = 14)	4.30	20	1.94	18	5.77	18

n: number of samples from April 2015 to March 2017

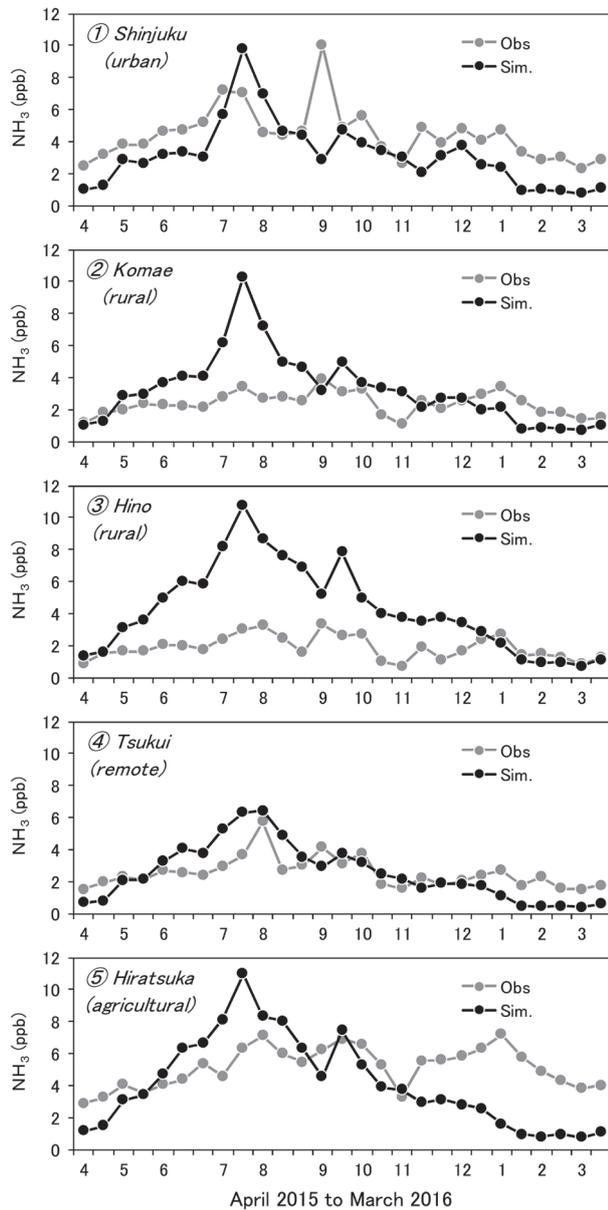


Fig. 4. Comparison between the observed and simulated NH_3 concentrations during the period from April 2015 to March 2016.

respectively. Regarding the simulation result for the Komae (rural) site, the observations and simulation suggest that NH_3 emissions from human sources around the Tokyo suburb might be larger than in reality, especially in the warm season.

As for the Hino (rural) site, there is a consistency in the relationship between the emissions and the observed concentrations, as mentioned in the previous section. However, the simulated concentrations at the Hino (rural) site in the warm season became unexpectedly

Table 2. Statistics between simulated and observed NH_3 for the annual, the warm season and the cold season during the period from April 2015 to March 2016.

		Mean Sim. (ppbv)	Mean Obs. (ppbv)	NMB ³⁾ (%)	r ⁴⁾
Shinjuku	Annual	3.16	4.43	-29	0.56
	Warm ¹⁾	4.04	5.05	-20	0.41
	Cold ²⁾	2.23	3.76	-41	0.68
Komae	Annual	3.14	2.38	32	0.49
	Warm	4.31	2.53	70	0.61
	Cold	1.89	2.22	-15	0.24
Hino	Annual	4.25	1.88	126	0.65
	Warm	5.83	2.16	170	0.73
	Cold	2.55	1.59	61	0.27
Tsukui	Annual	2.53	2.53	0	0.75
	Warm	3.57	2.93	22	0.72
	Cold	1.41	2.10	-33	0.53
Hiratsuka	Annual	4.11	5.13	-20	0.43
	Warm	5.76	5.02	15	0.79
	Cold	2.33	5.26	-56	0.30

¹⁾warm: April to September 2015 (n = 14)

²⁾cold: December 2015 to March 2016 (n = 13)

³⁾NMB: Normalized Mean Bias calculated by $\sum(\text{Sim}-\text{Obs})/\sum\text{Obs}$

⁴⁾r: correlation coefficient

higher despite the smaller emission amount of NH_3 in its grid. Fig. 5 indicates the ensemble mean of the diurnal variation in the simulated NH_3 in the sampling period from July 21 to August 3, 2015, when the largest overestimations were simulated at all five sites. The simulation was configured so that the emission strength of NH_3 increased in the daytime according to the temperature rise and human activities. Accordingly, the increase of NH_3 concentration was simulated in the late morning at the Shinjuku (urban), Komae (rural), and Tsukui (remote) sites. The simulated concentration at Shinjuku (urban) increased earlier than those at Komae (rural) and Tsukui (remote), and it seems to be derived from the NH_3 emission from vehicles during the morning rush hours.

Conversely, the daily maximum concentration of NH_3 was simulated in the early morning at the Hino (rural) and Hiratsuka (agricultural) sites. The simulated high concentration in the early morning was inconsistent with the diurnal variation in the NH_3 emissions. To evaluate why high concentrations appeared in the early morning during the summer season at these two sites, ensemble mean of the spatial distributions of the NH_3 concentration at 5 AM (Japan Standard Time) in the sampling period from July 21 to August 3, 2015, simulated by WRF/CMAQ, are illustrated in Fig. 6. In addition, Fig. 7 indicates a comparison between the observed and simulated wind direction in the same

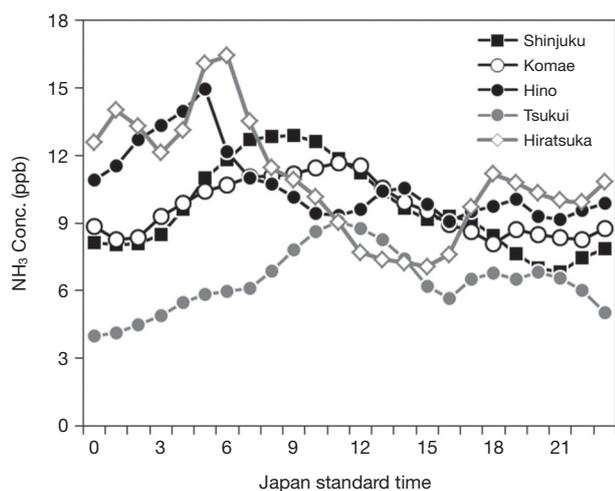


Fig. 5. Ensemble mean of the diurnal variation in the simulated NH_3 in the sampling period from July 21 to August 3, 2015, when the largest overestimations were simulated at each site.

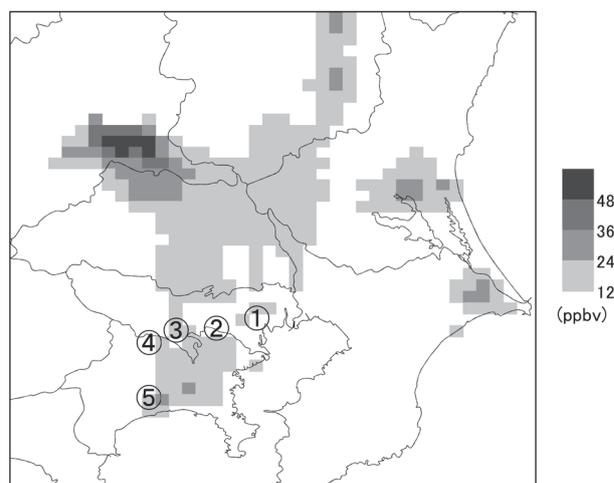


Fig. 6. Ensemble mean of the spatial distributions of the NH_3 concentration at 5 AM (Japan Standard Time) in the sampling period from July 21 to August 3, 2015.

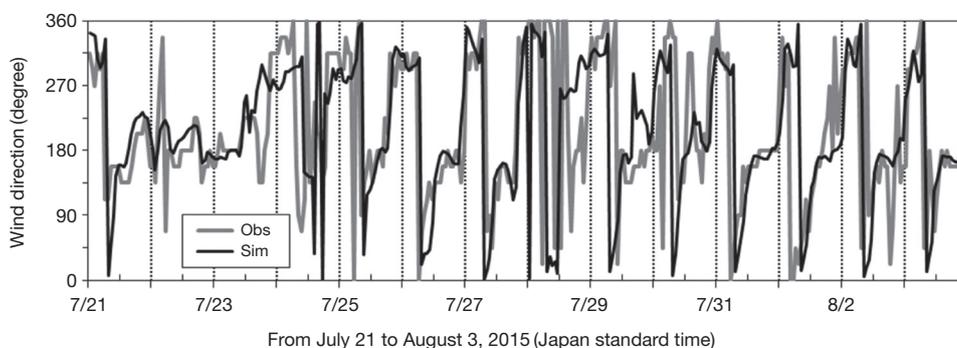


Fig. 7. Comparison between the observed (Obs) and simulated (Sim) wind direction from July 21 to August 3, 2015 (JST) at the nearest AMeDAS station to the Hino (rural) site, which is located in Hachioji city and is approximately 5 km west-northwest from the Hino site.

period at the nearest AMeDAS (Automated Meteorological Data Acquisition System) station to the Hino (rural) site, which is located in Hachioji city and is approximately 5 km west-northwest of the Hino site. The simulated wind direction at Hachioji showed reasonable agreement with the observations and it was found that a land breeze (north wind) generally prevailed in the middle of the night.

Sakurai *et al.* (2003) also reported that higher concentrations of NH_3 were observed at night in Tokyo in the summer of 2002. As shown in Fig. 3, a large emission area of NH_3 exists in the northern part of the Kanto region originating from agricultural sources. Because a land breeze (north wind) generally prevails at night in the Kanto region, Sakurai *et al.* (2003) concluded from the simulation analysis that the higher concentration of NH_3 observed in Tokyo at night was caused by the

transportation of NH_3 emitted in the northern part of the Kanto region. Accordingly, Fig. 6 shows that the NH_3 emitted in the northern part of the Kanto region was transported south by the land breeze and that the concentrations at Hino (rural) and Hiratsuka (agricultural) sites were affected by the transported NH_3 . In addition, vertical turbulence is generally reduced due to the cooling of the land surface at night. This likely leads to a low planetary boundary layer as well as a reduction in the vertical mixing. Therefore, it is also suggested that the higher concentration of NH_3 in the early morning at the Hino (rural) and Hiratsuka (agricultural) sites occurred due to the reduced dilution of the local NH_3 emission and the transported NH_3 in the simulated lower planetary boundary layer in the early morning.

Hayami (2013) reported the seasonal variation in the

NH₃ emission strength from agricultural sources, which was modified considering the fertilization time in Japan. In its estimation, the emission amounts in the warm and cold seasons were decreased and increased, respectively. According to the model overestimation at the Hino (rural) and Hiratsuka sites in the summer as shown in Fig. 4, it is suggested that the agricultural NH₃ emission in the northern part of the Kanto region might be larger than the actual emission in the summer season. Moreover, the simulated concentration underestimated the observations in the cold season (October 2015 to March 2016) at the Hiratsuka (agricultural) site (NMB = -56%). As a result, it is suggested that there might not actually be a large difference in the agricultural NH₃ emissions between seasons. The discussion here supports the seasonal variation in the NH₃ emission estimated by Hayami (2013).

4. CONCLUSIONS

Because atmospheric ammonia plays an important role in the creation of SIA, this study aims to evaluate the performance of the AQM for the seasonal and spatial distribution of the NH₃ concentration in the atmosphere. To obtain observational data for the model validation, observations based on biweekly sampling have been conducted using passive samplers since April 2015 at multiple monitoring sites in the Tokyo metropolitan area.

In the comparison analysis between the observed concentration and the estimated NH₃ emission at each site, it was seen that there was a consistency in the relationship between the annual emission amount and the concentration levels at the Shinjuku (urban) and Hiratsuka (agricultural) sites. Conversely, even though the emission amount at the Komae (rural) site was nearly as large as that at the Shinjuku (urban) and Hiratsuka (agricultural) sites, a relatively lower concentration was observed. Because the Komae (rural) site is located in a suburb of Tokyo and the human activities there are substantially different from those near the Shinjuku (urban) site, it is suggested that the estimated emission around the Tokyo suburb might be overestimated compared to the actual situation.

In the model validation, the model unexpectedly overestimated the observed concentration at the Hino (rural) site in the summer season despite the smaller emission amount of NH₃ in its grid. In addition, the NH₃ concentration was simulated to increase in the early morning in the summer at the Hino (rural) and Hiratsuka (agricultural) sites. The simulated spatial distribution suggests that this overestimation was caused by the transport of NH₃ emitted in the northern

part of the Kanto region, which originated from agricultural sources. The NH₃ was simulated as being transported from north to south in the Kanto region by a land breeze during the middle of the night during the summer season, and the concentrations at Hino (rural) and Hiratsuka (agricultural) sites were affected by the transported NH₃. Moreover, the model underestimated the NH₃ concentration observed at the Hiratsuka (agricultural) site in the autumn (from October to December) and winter (from January to March) seasons.

As a result, it is suggested that the emission input used in this study overestimated the NH₃ emission from human sources around the Tokyo suburbs and agricultural sources in the northern part of the Kanto region in the summer season. Moreover, because the simulated concentration underestimated the observations in the cold season at the Hiratsuka (agricultural) site, there might not actually be a large difference in the agricultural NH₃ emissions between seasons, unlike that in the emission input used in this study. Because the overestimation of NH₃ in the summer causes an overestimation of the NO₃⁻ in PM_{2.5} in the AQM simulation, further validation is required for the seasonal variation in the NH₃ emissions.

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