Model Evaluation based on a Relationship Analysis between the Emission 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 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, 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 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 [(NH$_4$)$_2$SO$_4$]” and “ammonium nitrate [NH$_4$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 PM$_{2.5}$ states that the daily and annual averaged concentrations should not exceed 35 $\mu$g m$^{-3}$ and 15 $\mu$g m$^{-3}$, respectively (established in 2009). The achievement rates for 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 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 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 PM$_{2.5}$ in the summer seasons.
previous studies have also reported overestimations of NO$_3^-$ in 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, 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$_3$.

### 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).

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**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; Skamarock 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 × 4,950 km² for Domain 1, 1,095 × 1,200 km² for Domain 2, and 335 × 335 km² 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). NOₓ emissions from vehicles were modified by reducing them by approximately 75% according to the reduction rate of the annual averaged concentration of NOₓ 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 SO₂ 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 NH₃ 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 NH₃ emission used in the simulation for Domain 3 (5-km grid resolution). The annual emission amounts of NH₃ in each grid were Shinjuku: 4.92 ton km⁻² year⁻¹, Komae: 4.52 ton km⁻² year⁻¹, Hino: 3.24...
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 NH$_3$ 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 NH$_3$ 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 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 NH$_3$ emissions for each season compared to the annual amount. In general, the emission strength of NH$_3$ 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 NH$_3$ 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 NH$_3$ 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 NH$_3$ 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%, respectively.

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

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

$n$: number of samples from April 2015 to March 2017
respectively. Regarding the simulation result for the Komae (rural) site, the observations and simulation suggest that NH₃ 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 higher despite the smaller emission amount of NH₃ in its grid. Fig. 5 indicates the ensemble mean of the diurnal variation in the simulated NH₃ 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 NH₃ increased in the daytime according to the temperature rise and human activities. Accordingly, the increase of NH₃ 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 NH₃ emission from vehicles during the morning rush hours.

Conversely, the daily maximum concentration of NH₃ 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 NH₃ 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 NH₃ 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

Table 2. Statistics between simulated and observed NH₃ for the annual, the warm season and the cold season during the period from April 2015 to March 2016.

| Site      | Season | Mean Sim. (ppbv) | Mean Obs. (ppbv) | NMB (%) | 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 ∑(Sim-Obs)/∑Obs
^4)r: correlation coefficient
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 from the Hino site.

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 suburbs and 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 PM₂.₅, further validation is required for the seasonal variation in the NH₃ emissions.

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REFERENCES

Byun, D.W., Ching, J.K.S. (1999) Science algorithms of the EPA Models-3 community multi-scale air quality (CMAQ) modeling system. EPA/600/R-99/030.

Carmichael, G.R., Sakurai, T., Streets, D., Hozumi, Y., Ueda, H., Park, S.U., Fung, C., Han, Z., Kajino, M., Engardt, M., Bennet, C., Hayami, H., Sartelet, K., Holloway, T., Wang, Z., Kamari, A., Fu, J., Matsuda, K., Thongbooncho, N., Amann, M. (2008) MICS-Asia II: The model intercomparison study for Asia Phase II methodology and overview of findings. Atmospheric Environment 42, 3468-3490.

Chatani, S. (2015) Contribution of Three-Dimensional Regional Air Quality Simulation for Mitigating Loads
on Atmospheric Environment. Journal of Japan Society for Atmospheric Environment 50(2), 76-84 (in Japanese).

Emmons, L.K., Walters, S., Hess, P.G., Lamarque, J.-F., Pfister, G.G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S.L., Kloster, S. (2010) Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4). Geoscientific Model Development 3, 43-67.

Fukui, T., Kokuryo, K., Baba, T., Kannari, A. (2014) Updating EAGrid2000-Japan emissions inventory based on the recent emission trends. Journal of Japan Society for Atmospheric Environment 49(2), 117-125 (in Japanese).

Hasegawa, S., Yonemochi, S., Yamada, D., Suzuki, Y., Ishii, K., Saito, S., Kamoshida, M., Kumagai, K., Jo, H. (2014) Analysis of the High Concentration of PM$_{2.5}$ Observed in the Kanto area in November 2011. Journal of Japan Society for Atmospheric Environment 49(6), 242-251 (in Japanese).

Hayami, H. (2013) Present Performance and improvement of Air Quality Models for PM$_{2.5}$. Journal of Japan Society for Safety Engineering 52(6), 383-387 (in Japanese).

Matsumoto, R., Yonemochi, S., Umezawa, N., Sakamoto, K. (2010) The influence of vehicles emission on air concentration of ammonia and nitrogen oxides at roadside. Chikyu Kankyo 15, 103-110 (in Japanese).

Ministry of Environment, Japan (2017) http://www.env.go.jp/press/103858.html (in Japanese).

Morino, Yu., Chatani, S., Hayami, H., Sasaki, K., Mori, Y., Morikawa, T., Ohara, T., Hasegawa, S., Kobayashi, S. (2010) Inter-comparison of Chemical Transport Models and Evaluation of Model Performance for O$_3$ and PM$_{2.5}$ Prediction - Case Study in the Kanto Area in Summer 2007. Journal of Japan Society for Atmospheric Environment 45(5), 212-226 (in Japanese).

Ocean Policy Research Foundation (2013) Report for comprehensive study for environmental impact lead by the establishment of emission control area in Japan. ISBN978-4-88404-299-8 (in Japanese).

Roadman, M.J., Scudlark, J.R., Meisinger, J.J., Ullman, W.J. (2003) Validation of Ogawa passive samplers for the determination of gaseous ammonia concentration in agricultural settings. Atmospheric Environment 37, 2317-2325.

Sakurai, T., Fujita, S., Hayami, H., Furushashi, N. (2003) A case study of high ammonia concentration in the nighttime by means of modeling analysis in the Kanto region of Japan. Atmospheric Environment 37, 4461-4465.

Sakurai, T., Satake, S., Matsuda, K. (2015) Measurement of the Inorganic Ions in PM$_{2.5}$ at Western Tokyo and the Evaluation for AQM Performance Based on the Measurement. Earozoru Kenkyu 30, 134-141 (in Japanese).

Shimadera, H., Hayami, H., Chatani, S., Morino, Y., Mori, Y., Morikawa, T., Yamaji, K., Ohara, T. (2014) Sensitivity analyses of factors influencing CMAQ performance for fine particulate nitrate. Journal of the Air & Waste Management Association 64(4), 374-387.

Skamarock, W.C., Klemp, J.B., Dudhia, J., Gill, D.O., Barker, D.M., Duda, M., Huang, X.-Y., Wang, W., Powers, J.G. (2008) A description of the advanced research WRF version 3. NCAR/TN-475+STR.

Ueno, H., Akiyama, K., Ishii, K., Miyoshi, T., Yokota, H., Nagoya, T. (2011) Relationship between PM$_{2.5}$, water-soluble organic carbon and oxidants in Tokyo during the summer. Journal of Japan Society for Atmospheric Environment 46(2), 124-130 (in Japanese).

van der Werf, G.R., Randerson, J.T., Giglio, L., Collatz, G.J., Mu, M., Kasibhatla, P.S., Morton, D.C., DeFries, R.S., Jin, Y., van Leeuwen, T.T. (2010) Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997-2009). Atmospheric Chemistry and Physics 10, 11707-11735.

Yonemochi, S., Umezawa, N. (2010) Parallel continuous observation of submicron particle (PM$_{1}$) and PM$_{2.5}$, and characterization of PM$_{1}$ in a suburban of Tokyo. Journal of Japan Society for Atmospheric Environment 45(6), 271-278 (in Japanese).

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