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Research Article

Evaluation of Air Quality Model Performance for Simulating Long-Range Transport and Local Pollution of $PM_{2.5}$ in Japan

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The Community Multiscale Air Quality Model (CMAQ) v5.0.2 was applied to $PM_{2.5}$ simulation in Japan, which is strongly affected by long-range transport (LRT) from anthropogenic sources in the Asian Continent, for one year from April 2010 to March 2011. The model performance for LRT and local pollution (LP) of $PM_{2.5}$ was evaluated to identify the model processes that need to be improved. CMAQ well simulated temporal and spatial variation patterns of $PM_{2.5}$ but underestimated the concentration level by 15% on average. The contribution of LRT was estimated from the difference between the baseline simulation case and a zero-emission case for anthropogenic emissions in the continent. The estimated LRT contribution to $PM_{2.5}$ was 50% on average and generally higher in the western areas of Japan (closer to the continent). Days that were dominantly affected by LRT or LP were determined based on the contribution of LRT to sulfate, which was fairly well simulated and strongly affected by LRT among major $PM_{2.5}$ components. The underestimation of $PM_{2.5}$ was larger in LP days (by 26% on average) than LRT days (by 10% on average). Therefore, it is essential to improve local emissions, formation, and loss processes of precursors and $PM_{2.5}$ in Japan.

1. Introduction

Particulate matter (PM) with aerodynamic diameter less than 2.5 μm ($PM_{2.5}$) is an atmospheric pollutant that mainly consists of several major components, such as organic aerosol (OA), elemental carbon (EC), sulfate, nitrate, and ammonium. Primary OA (POA) and EC are emitted directly through combustions of fossil fuel and biomass [1]. Secondary OA (SOA) is formed from various volatile organic compounds (VOC) and the corresponding processes are highly complex and varied [2]. Sulfate and nitrate are typically secondary pollutants produced through oxidations of sulfur dioxide (SO_2) and nitrogen oxides (NO_X), respectively. Ammonium is produced from ammonia (NH_3) and generally a counter ion of sulfate and nitrate.

The Ministry of the Environment of Japan (MOE) introduced an air quality standard (AQS) for $PM_{2.5}$ concentration (35 $\mu g\,m^{-3}$ for daily mean and 15 $\mu g\,m^{-3}$ for annual mean) in 2009. However, the AQS has not been attained in most urban areas in Japan. Although it is essential to use air quality models (AQMs) to design effective $PM_{2.5}$ control strategies, the performance of current AQMs for $PM_{2.5}$ simulation is not adequate for the purpose. Therefore, in order to improve AQM performance, a variety of studies have been conducted, including AQM intercomparisons, sensitivity analyses, and revisions of AQM subprocesses [3–5].

In East Asia, the rapid growth in economic activity and energy consumption in recent decades have resulted in increased anthropogenic emissions of air pollutants [6, 7]. Since air pollutants from the Asian Continent are efficiently transported to the Pacific region by eastward traveling high- and low-pressure systems because of the Westerlies in spring and autumn and by northwesterly monsoon in winter, long-range transport (LRT) of air pollutants from the continent in these seasons has been a field of scientific interests [8–10]. In addition, even in summer, LRT can strongly contribute to sulfate concentration in Japan [11]. Therefore, AQMs applied to air quality management in Japan have to properly simulate both local pollution (LP) and LRT.

In this study, one-year air quality simulations by the Community Multiscale Air Quality Model (CMAQ) [12] were conducted in regions from East Asia to Japan. The model

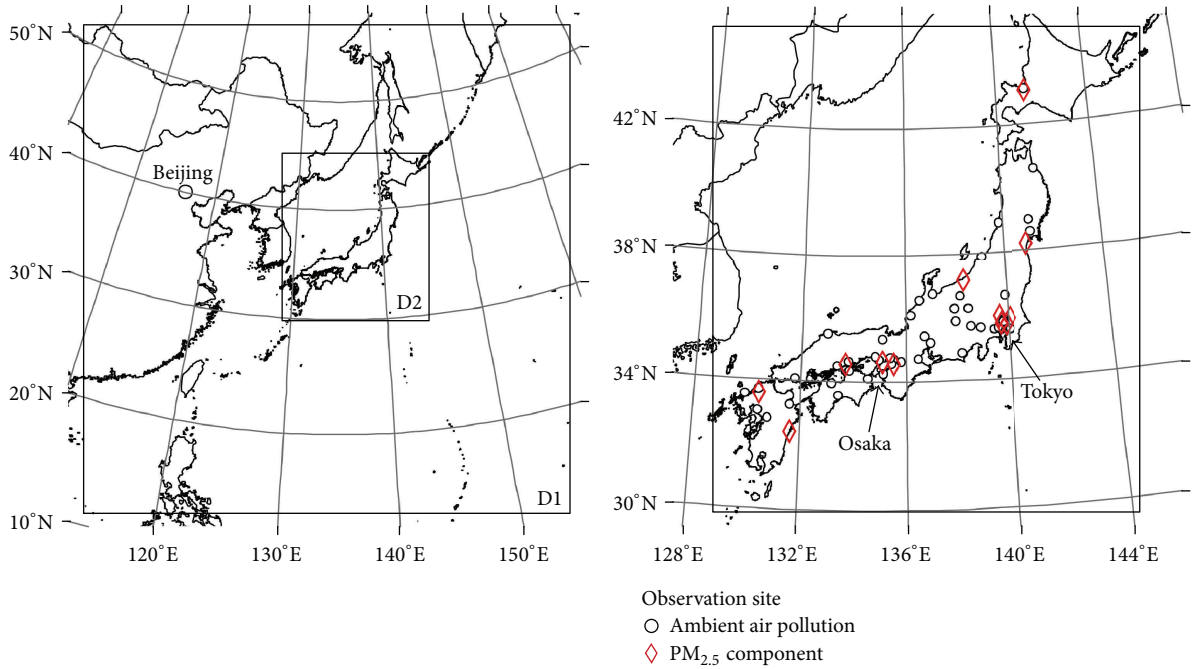


Figure 1: Modeling domains and location of observation sites.

performance for simulating LRT and LP of $PM_{2.5}$ in Japan was evaluated to identify aspects of AQM processes that need to be improved.

2. Materials and Methods

Air quality simulations were conducted by using CMAQ v5.0.2 from April 2010 to March 2011 (Japanese fiscal year 2010: JFY2010) with an initial spin-up period of 22–31 March 2010. The study period was selected because nation-wide $PM_{2.5}$ observations were launched in JFY2010 in Japan. Figure 1 shows the modeling domains for the CMAQ simulations including domain 1 (D1) covering a wide area of Northeast Asia and domain 2 (D2) covering almost the entire area of Japan. The horizontal resolutions and the number of grid cells are 64 km and 76×76 grid cells for D1 and 16 km and 92×104 grid cells for D2, respectively. The vertical layers consist of 30 sigma-pressure coordinate layers from the surface to 100 hPa with the middle height of the first layer being approximately 28 m.

Meteorological fields were produced by the Weather Research and Forecasting Model (WRF) [13] v3.5.1. The physics options and objective analysis data were the same as those used by Shimadera et al. [14]: the Yonsei University planetary boundary layer (PBL) scheme [15], the Kain-Fritsch cumulus parameterization [16], the WRF single-moment 6-class microphysics scheme [17], the Noah land surface model [18], the rapid radiative transfer model [19] for the long wave radiation, and the shortwave radiation scheme of Dudhia [20], the real time, global analysis, high-resolution sea surface temperature (RTG_SST_HR) data by the US National Centers for Environmental Prediction (NCEP), the mesoscale model grid point value (MSM GPV) data by the Japan Meteorological Agency (JMA), and the final analysis (FNL) data by NCEP. Grid nudging was applied to horizontal wind components, temperature, and humidity with a nudging coefficient of $3.0 \times 10^{-4}\ s^{-1}$ in the entire simulation domain and period. Shimadera et al. [14] showed that WRF successfully produced meteorological fields for air quality simulations in JFY2010 in Japan.

CMAQ was configured with the Carbon Bond mechanism developed in 2005 (CB05) [21] for the gas-phase chemistry, the sixth generation CMAQ aerosol module (AERO6) for the aerosol process. The hourly WRF results were processed using the Meteorology-Chemistry Interface Processor (MCIP) v4.2 for CMAQ. Initial and boundary concentrations in D1 were derived from the Model for Ozone and Related Chemical Tracers v4 (MOZART-4) [22].

Various datasets were used to produce emission data for the CMAQ simulations. Anthropogenic emissions in Japan were derived from the Japan Auto-Oil Program (JATOP) Emission Inventory-Data Base (JEI-DB) in the year 2010 developed by Japan Petroleum Energy Center (JPEC) [23] for vehicles, an emission inventory in the year 2005 developed by the Ocean Policy Research Foundation (OPRF) [24] for ships, and an emissions inventory in the year 2010 called EAGrid2010-JAPAN [25] for the other sectors. Anthropogenic emissions in East Asia except Japan were derived from an emission inventory for Asia in the year 2006 to support the Intercontinental Chemical Transport Experiment phase B (INTEX-B) [7] v1.2. Because of a lack in the INTEX-B emission data, NH_3 emissions were derived from the Regional Emission inventory in Asia (REAS) [6] v1.11. Daily emissions from open biomass burning were derived from

a fire inventory of Wiedinmyer et al. [26]. Biogenic emissions were estimated with the Model of Emissions of Gases and Aerosols from Nature (MEGAN) [27] v2.04. Volcanic SO_2 emissions were derived from the Aerosol Comparisons between Observations and Models (AEROCOM) data [28].

The CMAQ simulations were conducted in the following two cases: a baseline simulation case with all the emission data (EBase) and a zero-emission case for anthropogenic emissions outside Japan (EJapan). Results in EJapan roughly indicate the contribution of LP in Japan and differences between results of EBase and EJapan indicate the contribution of LRT from anthropogenic sources in the Asian Continent to Japan. Note that emission data in EJapan includes the entire natural emissions such as biomass burning and biogenic and volcanic emissions.

Figure 2 shows spatial distributions of mean SO_2, NO_X, NH_3, and primary $PM_{2.5}$ emissions. Major anthropogenic emission sources of SO_2 are large point sources, industrial areas, and ships. Major sources of NO_X and primary $PM_{2.5}$ are vehicles in addition to those of SO_2. Major sources of NH_3 are agricultural sources such as volatilization from livestock wastes and fertilized fields. In China, these emissions were particularly large in and around Beijing, and, in Japan, emissions were particularly large in and around Tokyo, followed by Osaka. The total SO_2, NO_X, NH_3, and primary $PM_{2.5}$ emissions in D1 were 37.5, 23.3, 15.7, and 11.6 Tg yr^{-1} in EBase and 8.2, 3.3, 0.6, and 1.6 Tg yr^{-1} in EJapan, respectively.

Results of the CMAQ simulation in EBase were compared with ground-level concentration data at observation sites shown in Figure 1. The observation sites include ambient air pollution monitoring stations conducting $PM_{2.5}$ observations in JFY2010 and national monitoring stations in Japan (49 stations in total) [29], the US embassy in Beijing for $PM_{2.5}$ [30], and 24 h filter sampling sites for $PM_{2.5}$ components (Fukuoka City in Fukuoka Prefecture, Hyuga in Miyazaki, Kurashiki in Okayama, Kobe in Hyogo, Sakai in Osaka, Joetsu in Niigata, Ichikawa in Chiba, Hasuda in Saitama, Itabashi in Tokyo, Toride in Ibaraki, Sendai in Miyagi, and Sapporo in Hokkaido in order from west to east) [31]. At the 12 sampling sites, concentration data of the five major $PM_{2.5}$ components (OA, EC, sulfate, nitrate, and ammonium) are available for two weeks in each season in JFY2010 (spring: May 14–27, 2010, summer: July 26-August 11, 2010, autumn: November 5–18, 2010, and winter: January 26-February 10, 2011). The model performance was evaluated using statistical measures including Pearson's correlation coefficient (r), the mean bias error (MBE), the root mean square error (RMSE), MBE normalized by the mean observed value (NMB), RMSE normalized by the mean observed value (NRMSE), the proportion of simulated values within a factor 2 of observation (PF2), and the index of agreement (IA).

3. Results and Discussion

3.1. Model Performance and LRT for SO_2, NO_X, and $PM_{2.5}$. Table 1 shows statistical comparisons of the observations and the CMAQ simulation in EBase for daily mean SO_2, nitrogen dioxide (NO_2), and $PM_{2.5}$ concentrations at the ambient air pollution monitoring stations in JFY2010. For SO_2, while CMAQ approximately captured the mean concentration level, relatively small r, PF2, and IA and large NRMSE indicate that the model performance was inferior to that for NO_2 and $PM_{2.5}$. For NO_2, although the IA value indicates better agreement between the observation and simulation compared to SO_2, the model underestimated the mean concentration level. For $PM_{2.5}$, the model better simulated daily mean concentrations than those of its precursors, SO_2 and NO_2. Most of the simulated daily $PM_{2.5}$ concentrations were within a factor 2 of the observed values in both Japan and Beijing. In addition, the values of r and IA were the highest among the three pollutants. Therefore, CMAQ well simulated their temporal and spatial variation patterns of $PM_{2.5}$. However, the model moderately underestimated the mean $PM_{2.5}$ concentration in Japan. The CMAQ performance is further discussed through the following comparisons on temporal and spatial variations.

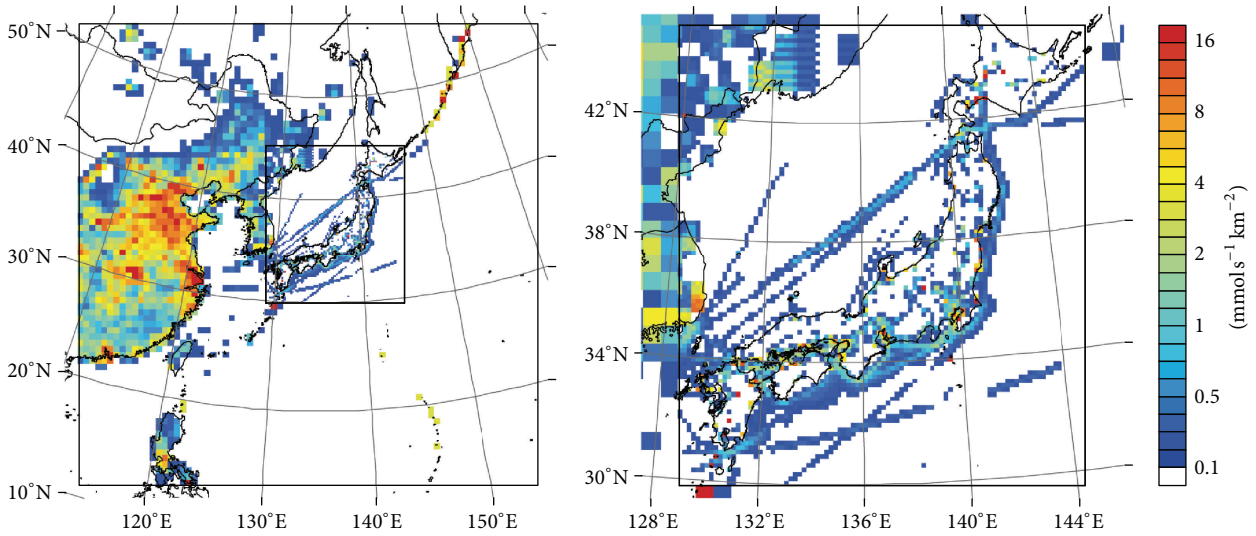
Table 1: Statistical comparisons between observed and simulated (EBase) daily mean SO_2, NO_2, and $PM_{2.5}$ concentrations at the ambient air pollution monitoring stations in JFY2010.

	Japan			Beijing
	SO_2	NO_2	$PM_{2.5}$	$PM_{2.5}$
Sample number	15053	16133	15315	323
Mean obs.	2.6 (2.1)	13.3 (8.1)	15.0 (10.2)	102.2 (85.3)
Mean sim.	2.2 (2.0)	9.7 (8.4)	12.7 (9.3)	110.4 (65.7)
r	0.40	0.71	0.75	0.83
MBE	−0.4	−3.7	−2.2	8.3
RMSE	2.3	7.3	7.3	48.8
NMB	−14%	−27%	−15%	8%
NRMSE	88%	55%	49%	48%
PF2	55%	63%	81%	83%
IA	0.62	0.80	0.85	0.89

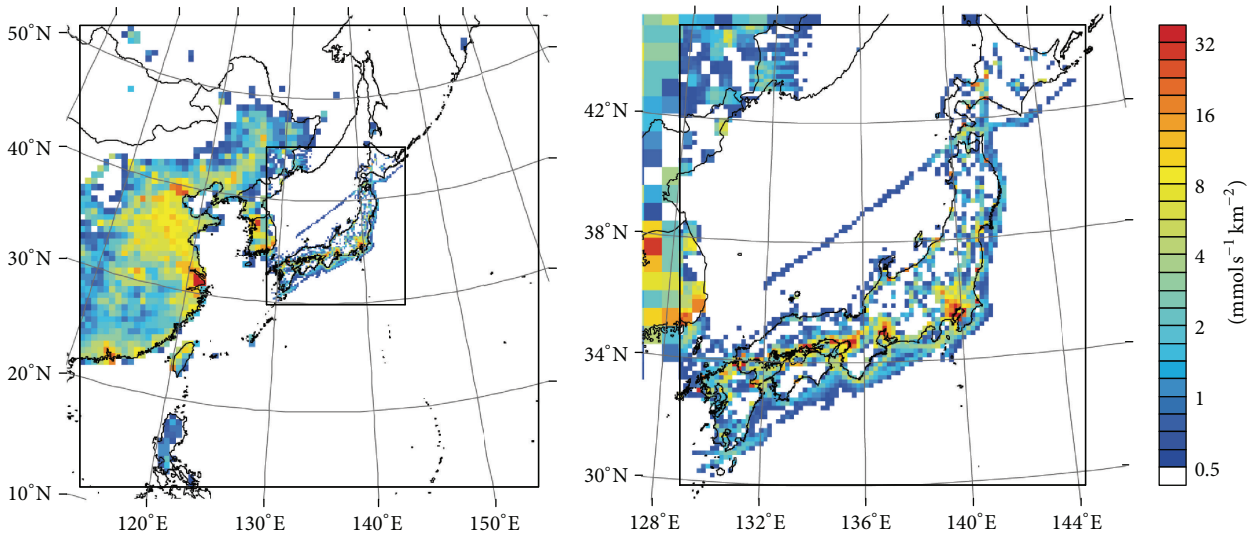
Note: parenthetical values show standard deviations. Units of mean, MBE, and RMSE values are ppbv for SO_2 and NO_2 and $\mu g\,m^{-3}$ for $PM_{2.5}$.

Figure 3 shows time series of daily mean observed and simulated SO_2, NO_2, and $PM_{2.5}$ concentrations averaged for all the ambient air pollution monitoring stations in D2 in JFY2010. For SO_2, CMAQ well captured day-to-day variation averaged for the monitoring stations in Japan. For NO_2, while both the observed and simulated concentrations tended to be high in the cool season from November 2010 to March 2011, relatively large underestimations frequently occurred in the period. For $PM_{2.5}$, the model fairly well simulated day-to-day variation of $PM_{2.5}$ concentration including occurrence of several high concentration peaks. The temporal variation pattern was similar to that of SO_2, but the variation range was larger than that of SO_2.

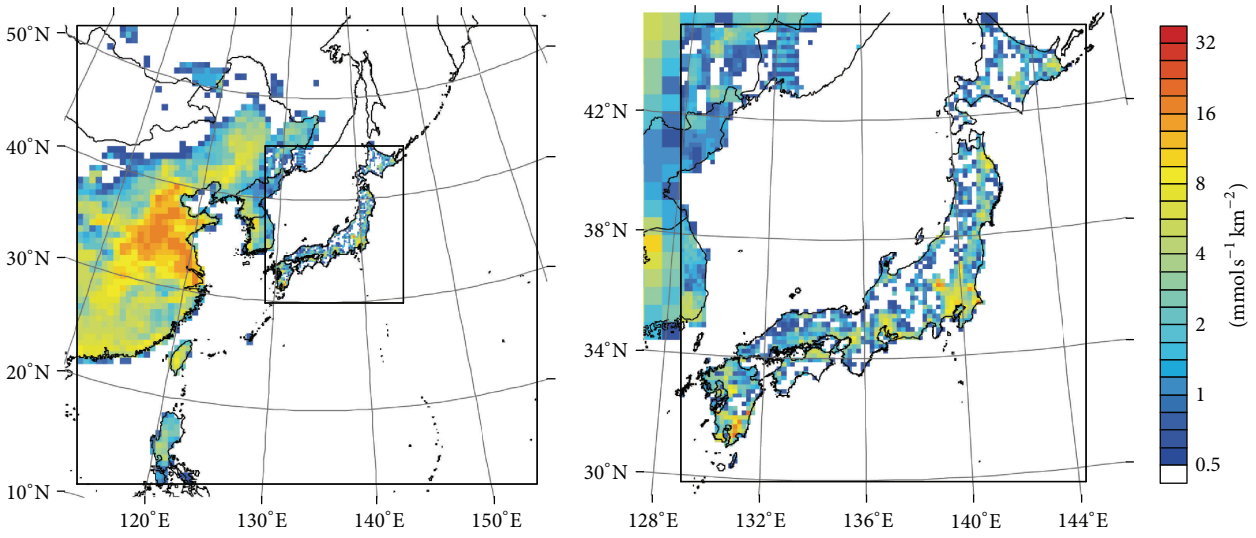
Figure 4 shows spatial distributions of simulated annual mean ground-level SO_2, NO_2, and $PM_{2.5}$ concentrations in JFY2010. Figure 5 shows observed and simulated annual mean SO_2, NO_2, and $PM_{2.5}$ concentrations at the ambient air pollution monitoring stations in D2. For SO_2, the simulated concentration in China was much higher than that in Japan. The bias between the observation and simulation considerably varied with the monitoring stations from large



(a) SO_2 emission

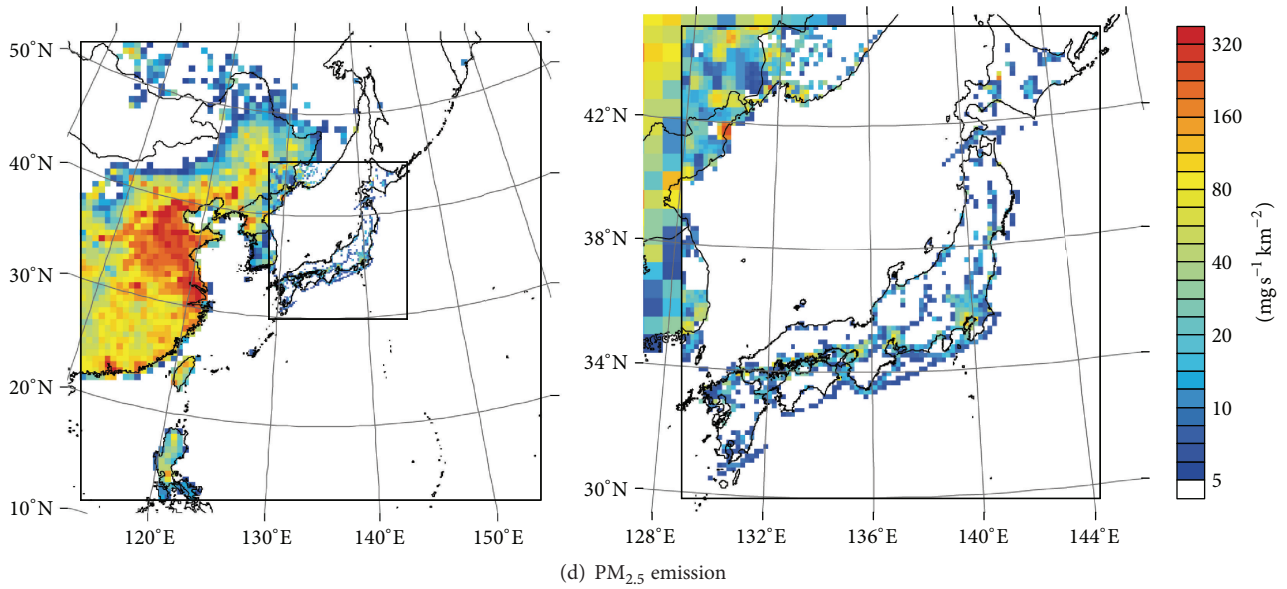


(b) NO_X emission



(c) NH_3 emission

Figure 2: Continued.



(d) $PM_{2.5}$ emission

FIGURE 2: Spatial distributions of mean SO_2, NO_X, NH_3, and $PM_{2.5}$ emissions in D1 (left) and D2 (right).

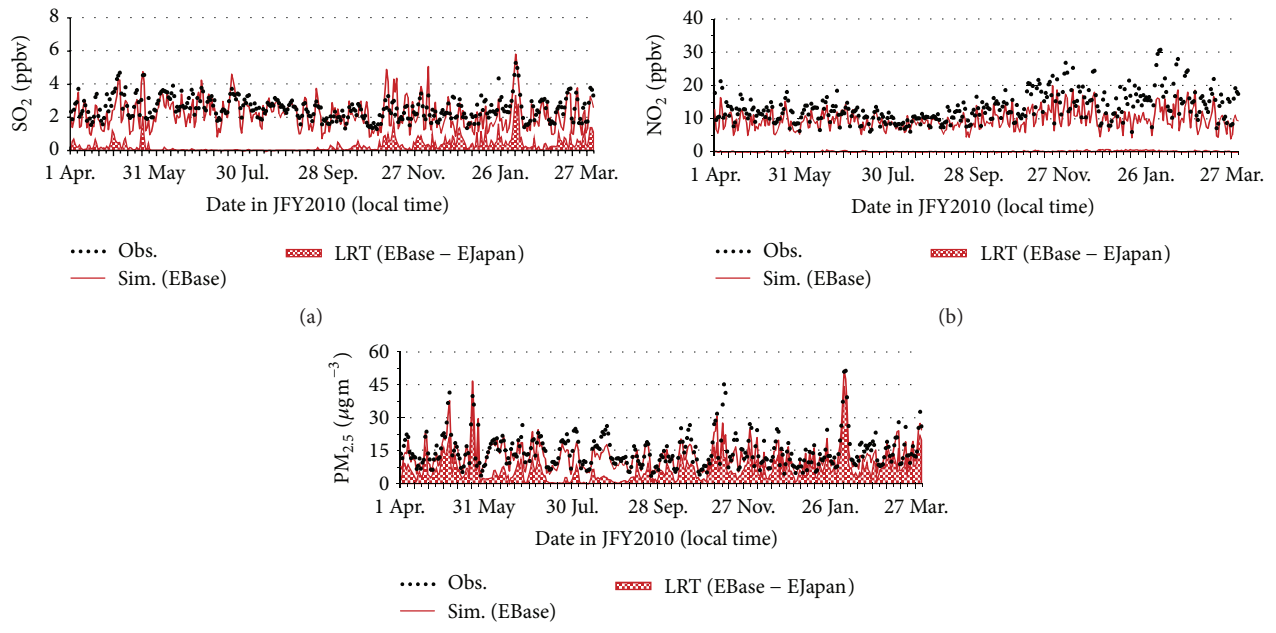
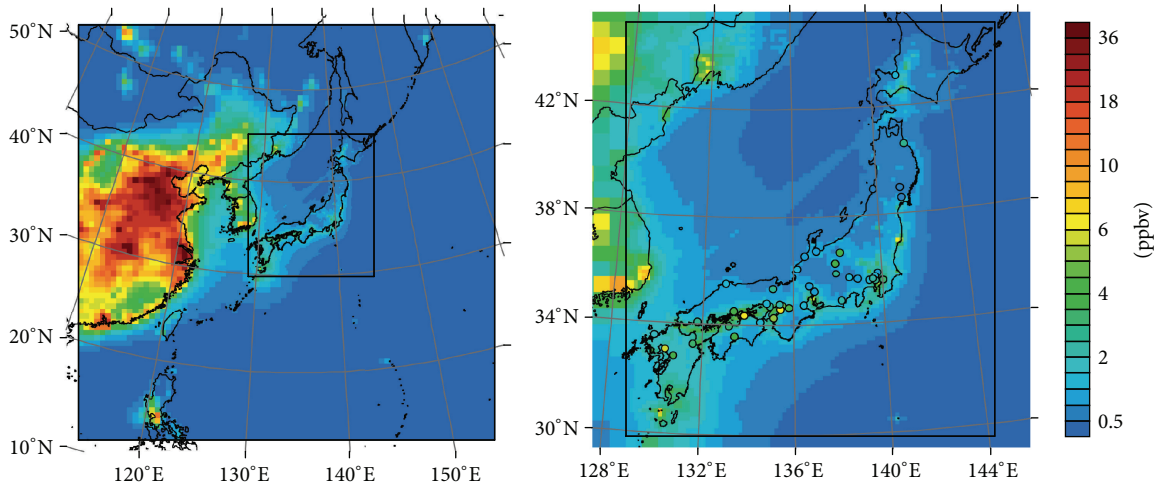
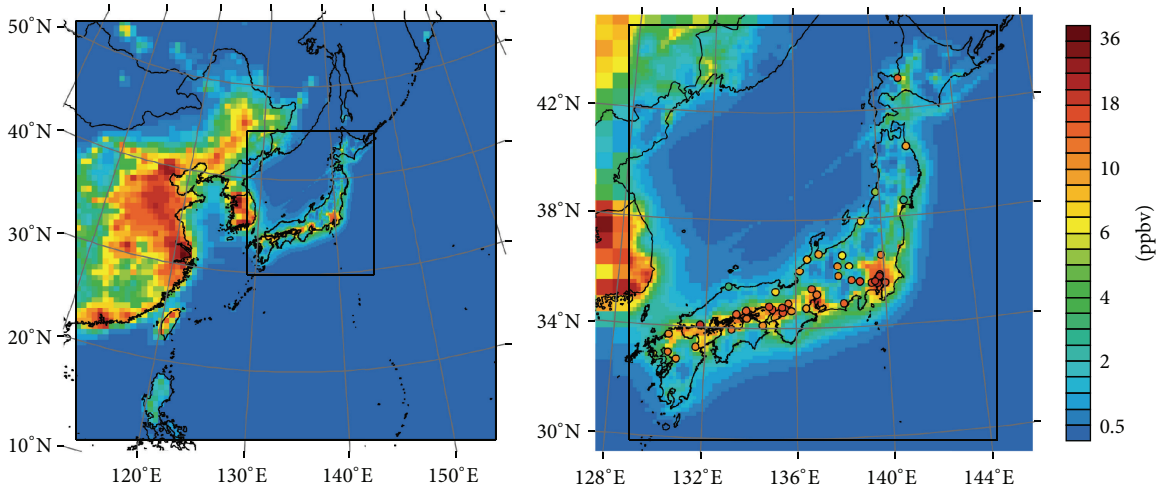


FIGURE 3: Time series of observed and simulated daily mean SO_2, NO_2, and $PM_{2.5}$ concentrations and corresponding contributions of LRT averaged for all the ambient air pollution monitoring stations in D2 in JFY2010.

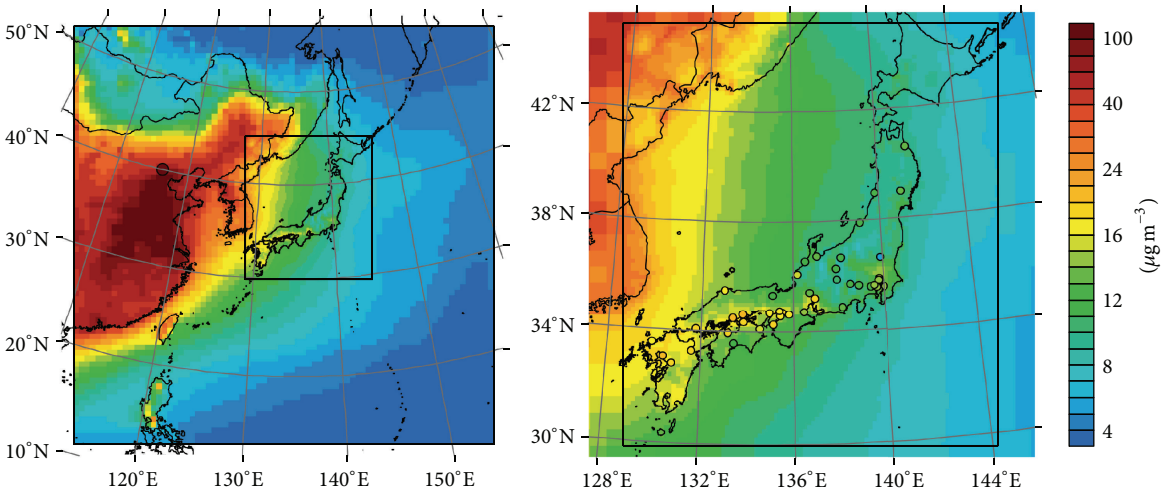
positive to large negative. Because CMAQ captured the temporal variation of SO_2 averaged for the monitoring stations (Figure 3(a)), the inferior model performance (Table 1) was attributed to the difficulty in reproducing the spatial variability. For NO_2, the simulated concentration in large metropolitan areas in Japan, such as Tokyo and Osaka, was comparable to that in Beijing. While CMAQ well captured the NO_2 concentration level in such metropolitan areas and coastal industrial areas with large emissions (Figure 2(b)), the model underestimated it at the monitoring stations in small cities, which resulted in the underestimation of the mean concentration (Table 1). For $PM_{2.5}$, the spatial gradient of the simulated concentration from the Asian Continent to Japan indicates a large contribution of LRT to Japan. Although there was a decreasing trend from west to east in both the observed and simulated concentrations in D2, CMAQ tended to underestimate the concentration at most of the monitoring stations.



(a) SO_2 concentration



(b) NO_2 concentration



(c) $PM_{2.5}$ concentration

Figure 4: Spatial distributions of simulated annual mean ground-level SO_2, NO_2, and $PM_{2.5}$ concentrations in D1 (left) and D2 (right) in JFY2010. Corresponding observed concentrations are shown at the ambient air pollution monitoring stations.

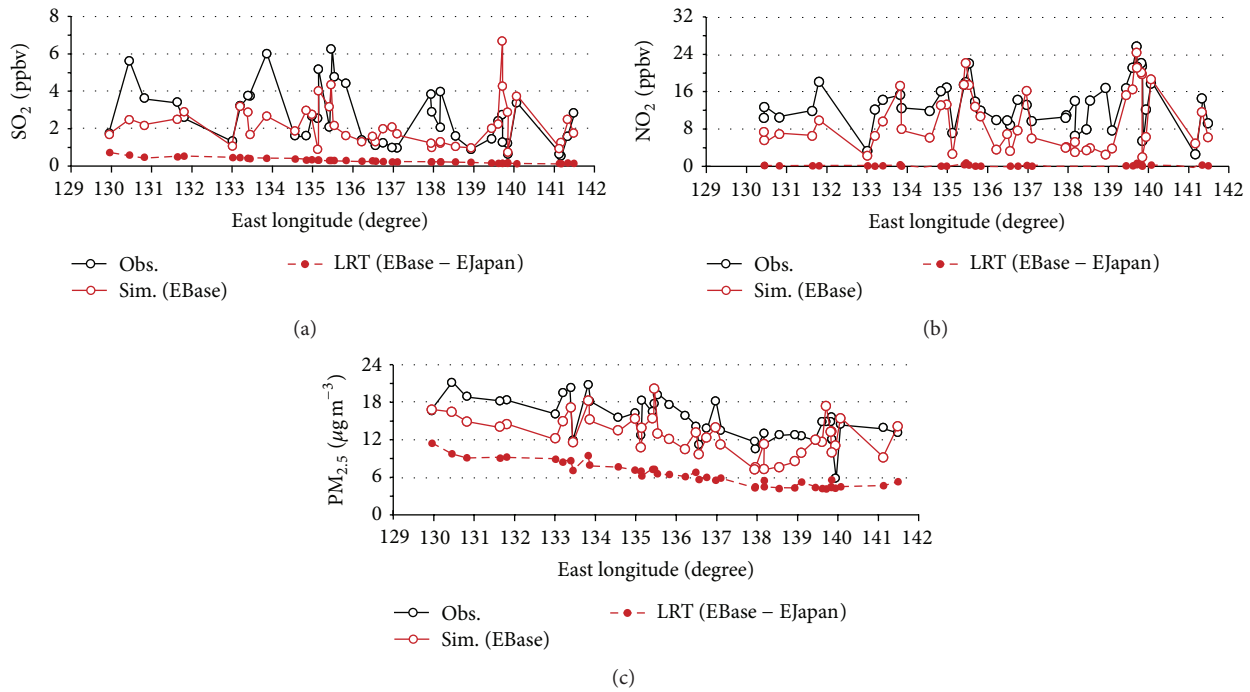


FIGURE 5: Observed and simulated annual mean SO_2, NO_2, and $PM_{2.5}$ concentrations and corresponding contributions of LRT in JFY2010 at the ambient air pollution monitoring stations in D2.

As shown in Figures 3 and 5, the contribution of LRT from anthropogenic emissions in the Asian Continent to Japan was substantial for $PM_{2.5}$ concentration and negligible for NO_2 concentration. The high concentration peaks of SO_2 and $PM_{2.5}$ concentrations were generally affected by LRT (Figures 3(a) and 3(c)), particularly for $PM_{2.5}$ in spring and winter. Annual mean estimated contribution ratios of LRT to NO_2, SO_2, and $PM_{2.5}$ concentrations averaged for the monitoring stations were 1, 13, and 50%, respectively. The contribution ratios were generally higher in the western areas of Japan, which are closer to the Asian Continent and have smaller emissions compared to the metropolitan areas. The contribution ratios to NO_2, SO_2, and $PM_{2.5}$ concentrations for the individual stations were up to 5, 43, and 73%, respectively. The contribution of LRT explains the decreasing trend from west to east in $PM_{2.5}$ concentration (Figure 5(c)). The higher LRT contribution ratio to $PM_{2.5}$ compared to SO_2 and NO_2 indicate that these precursors reacted to form sulfate and nitrate during the LRT process before arriving at Japan, and LP controlled the concentration level of these precursors in Japan.

Considering the findings described above, the discrepancies between the observation and simulation for the spatial variabilities of SO_2 and NO_2 concentrations (Figures 4 and 5) were mainly attributed to deficiencies in simulating LP in Japan. Therefore, local emission inventories still need to be revised.

In addition, a coarse horizontal grid resolution may be partly responsible for the limitation of LP simulation in Japan as pointed out by Morino et al. [5]. For example, in an air quality simulation using 16 km grid cells like this study, LP can be overestimated at a monitoring station located at 15 km from a large point source and can be underestimated at a station located at 1 km from a major arterial road, particularly if it is a dominant emission source within a grid cell.

3.2. Model Performance and LRT for Major $PM_{2.5}$ Components. Table 2 shows statistical comparisons of the observations and the CMAQ simulation in EBase for daily mean concentrations of $PM_{2.5}$ and its major components at the observation sites for $PM_{2.5}$ components for the four seasons (May 14–27, 2010, July 26-August 11, 2010, November 5–18, 2010, and January 26-February 10, 2011) in JFY2010. Figure 6 shows scatter plots for observed and simulated seasonal mean concentrations of $PM_{2.5}$ and its major components at the observation sites for each season. Because high $PM_{2.5}$ concentration peaks were observed in the reference period (Figure 3(c)), the mean $PM_{2.5}$ concentration of the four seasons was higher than that of the entire simulation period (Table 1). For EC, sulfate, and ammonium, the values of r and IA were particularly high, and most of the simulated daily concentrations were within a factor 2 of the observed values. These results indicate that CMAQ successfully captured the temporal and spatial variation patterns of these three components. For OA, the model clearly and consistently underestimated the concentration. Because CMAQ-simulated OA is dominated by POA even in summer [4], the underestimation of OA in warm season is likely caused by underestimation of SOA. The volatility basis set (VBS) approach may improve SOA formations in warm season [5]. Among the four seasons, the simulated OA concentration was the closest to the observed value in winter, in which POA

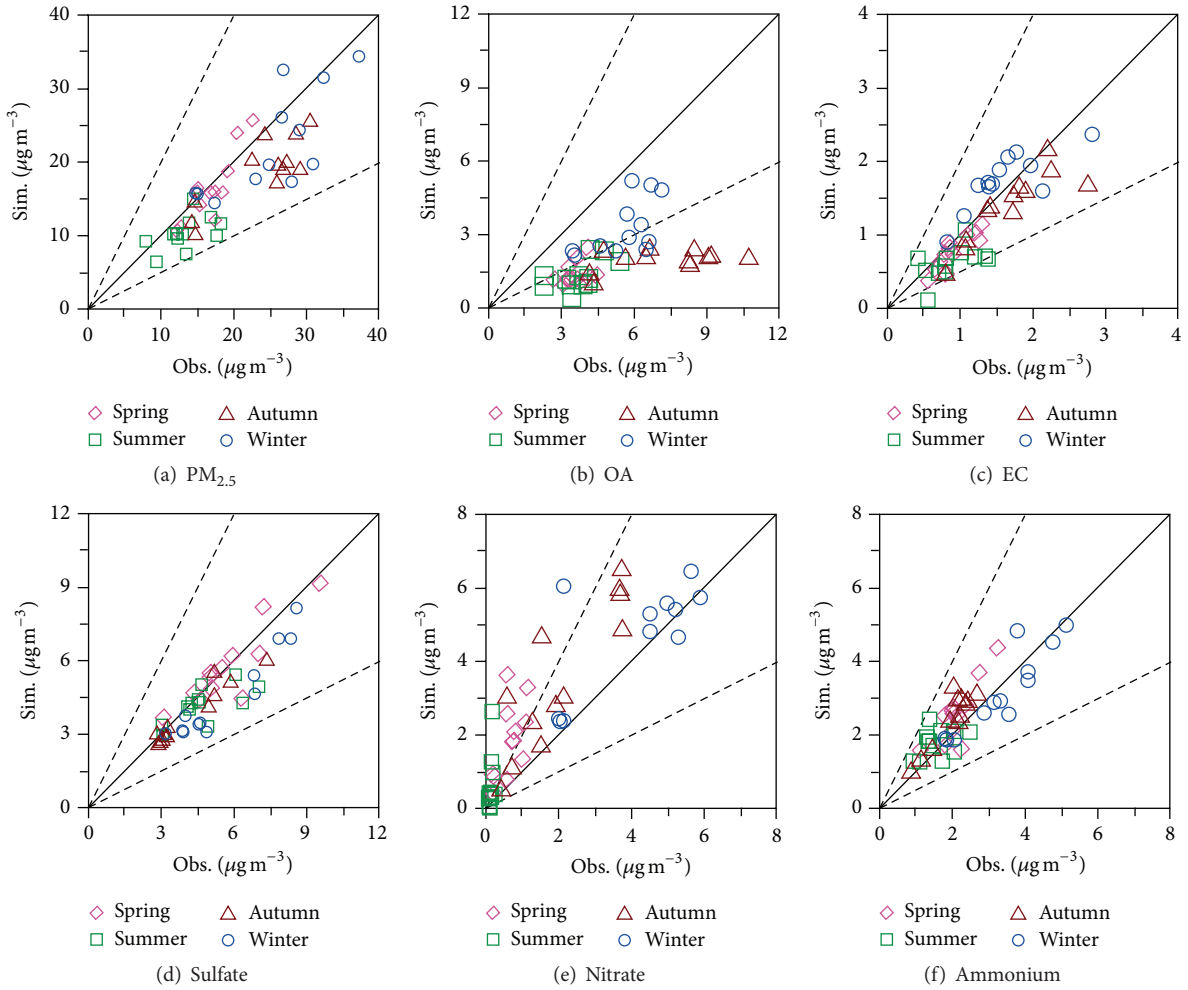


FIGURE 6: Scatter plots for observed and simulated seasonal mean concentrations of $PM_{2.5}$ and its major components at the observation sites for $PM_{2.5}$ components for each season in JFY2010. Reference lines for 1 : 1, 1 : 2, and 2 : 1 are provided.

was assumed to be dominant in the observation as well as the simulation. The observed OA concentration and the magnitude of underestimation were the largest in autumn that was harvest season in Japan. Therefore, it is essential to better estimate emissions from irregular open field burning after harvest. In addition, local emissions of condensable organic compounds (COC) that are not considered in the existing emission data can be a key factor for mitigating the underestimation [4]. For nitrate, the model clearly overestimated the concentration, particularly in warm season. Note that the overestimation in warm season may be partly attributed to a negative artifact in the observed data because of the volatilization from filter during 24 h sampling. As fine particulate nitrate is dominated by ammonium nitrate and the formation is highly sensitive to NH_3 [32], emission and deposition processes of NH_3 can be key factors for improving the model performance [3, 33]. In addition, the underestimation of NO_2 and the overestimation of nitrate may be improved by incorporating a heterogeneous reaction of nitric acid to NO_X [34].

Figure 7 shows observed and simulated mean concentrations of $PM_{2.5}$ and its major components and corresponding contributions of LRT at the observation sites for $PM_{2.5}$ components for the four seasons in JFY2010. For sulfate and ammonium, there was a clear decreasing trend from west to east in both the observed and simulated concentrations, which indicated a strong influence of LRT. Meanwhile, for OA, EC, and nitrate, the observed concentrations in and around Tokyo were comparable to or higher than those in the western areas of Japan that was more affected by LRT. Therefore, LP has a large impact on OA, EC, and nitrate in Japan. Mean estimated contribution ratios of LRT averaged for the observation sites for the four seasons were 57% for $PM_{2.5}$ concentration, 54% for OA, 29% for EC, 58% for sulfate, 55% for nitrate, and 58% for ammonium. There are large uncertainties in the estimated LRT contribution for OA and nitrate because the model performance for these two components are inferior to that for the other three components (Table 2), and, in addition, nitrate formation is strongly affected by nonlinear processes such as the formation though reactions of transported precursors and local precursors [35].

3.3. Model Performance for LP and LRT of $PM_{2.5}$. In order to evaluate the model performance for LRT and LP of $PM_{2.5}$,

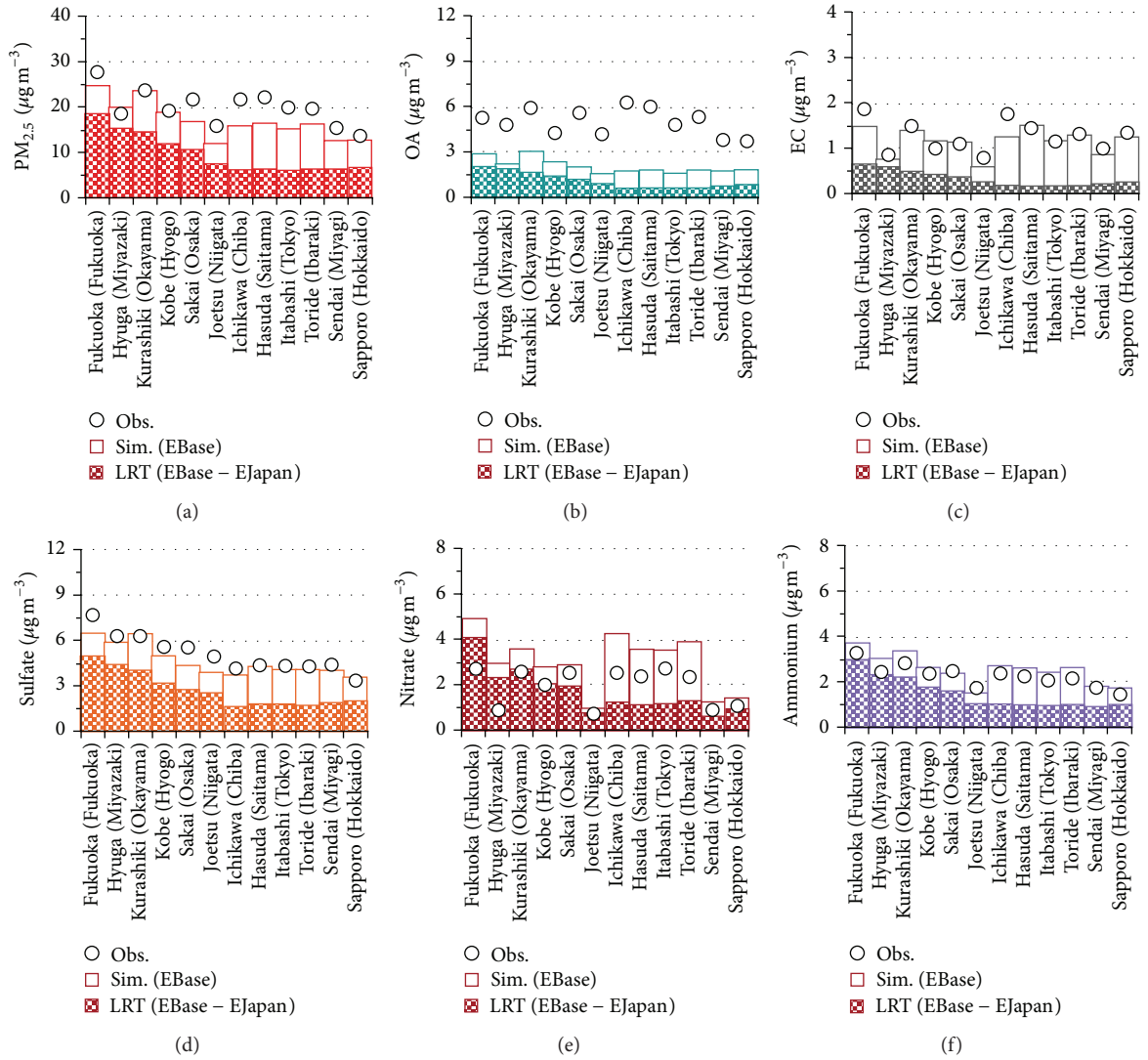


Figure 7: Observed and simulated mean concentrations of $PM_{2.5}$ and its major components and corresponding contributions of LRT at the observation sites for $PM_{2.5}$ components for four seasons in JFY2010. The observation sites are listed in order from west to east.

Table 2: Statistical comparisons between observed and simulated (EBase) daily mean concentrations of $PM_{2.5}$ and its major components at the observation sites for $PM_{2.5}$ components for four seasons in JFY2010.

	$PM_{2.5}$	OA	EC	Sulfate	Nitrate	Ammonium
Sample number	674	674	674	674	601	671
Mean obs.	19.9 (15.0)	5.0 (3.3)	1.3 (0.9)	5.1 (4.7)	2.0 (3.0)	2.3 (2.2)
Mean sim.	17.1 (14.8)	2.0 (2.2)	1.2 (0.9)	4.6 (4.5)	3.1 (4.0)	2.5 (2.5)
r	0.86	0.63	0.82	0.87	0.79	0.88
MBE	−2.8	−3.0	−0.1	−0.5	1.1	0.3
RMSE	8.4	3.9	0.6	2.4	2.7	1.2
NMB	−14%	−59%	−8%	−9%	53%	12%
NRMSE	42%	79%	44%	47%	133%	55%
PF2	86%	24%	86%	87%	50%	80%
IA	0.92	0.63	0.90	0.93	0.85	0.93

Note: parenthetical values show standard deviations. Units of mean, MBE, and RMSE values are $\mu g\,m^{-3}$.

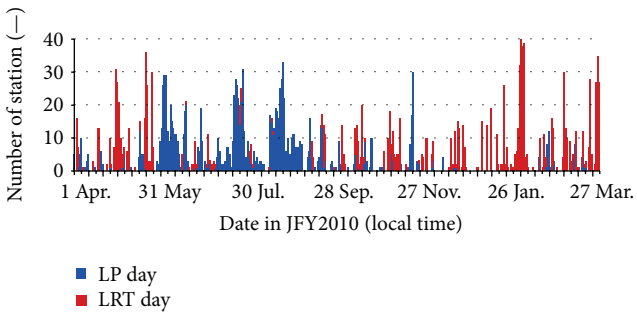


FIGURE 8: Time series of station numbers counted as LP and LRT days in JFY2010 among the ambient air pollution monitoring stations in D2.

the simulation period was classified into LP and LRT days. Because CMAQ fairly well simulated the temporal and spatial variations of sulfate that was strongly affected by LRT, the contribution ratio of LRT to sulfate was used as the index for LRT:

$$\text{LRT index} = \frac{[\text{sulfate}]_{\text{EBase}} - [\text{sulfate}]_{\text{EJapan}}}{[\text{sulfate}]_{\text{EBase}}}. \quad (1)$$

Among days with the observed $PM_{2.5}$ concentration >15 μg m^{-3} (AQS for annual mean $PM_{2.5}$), days with the daily LRT index being top and bottom 30 values were, respectively, classified into LRT and LP days at each of the ambient air pollution monitoring stations. Figure 8 shows time series of station numbers counted as LP and LRT days in JFY2010 among the ambient air pollution monitoring stations in D2. LP days were frequent in summer and LRT days were frequent in spring and winter. Most of the monitoring stations were counted as LRT days during the period of the highest $PM_{2.5}$ peak in February 2011 (Figure 3(c)).

The LP and LRT days classified with the LTR index were qualitatively verified through comparisons with backward trajectory analyses, which were conducted with the same method as that used by Shimadera et al. [11]. Figure 9 shows spatial distributions of simulated ground-level $PM_{2.5}$ concentrations with horizontal wind vectors and backward trajectories arriving at 300 m above Kokusetsu-Osaka station (135.535°E, 34.680°N) on July 24, 2010, and February 6, 2011, which are, respectively, typical LP and LRT days (Figure 8). The LP days in the latter part of July were characterized by prevailing southerly wind and local circulations under the pacific high-pressure system. The LRT days in the early part of February were caused by eastward traveling high-pressure systems that transported a large amount of $PM_{2.5}$ from the Asian Continent over a few days. The combination of the LRT index and the backward trajectory analysis enables detailed analysis of the atmospheric transport.

Figure 10 shows comparisons of observed and simulated $PM_{2.5}$ concentrations averaged for LP and LRT days in JFY2010 at the ambient air pollution monitoring stations in D2. While the observed concentrations averaged for LP days were almost spatially uniform, the observed and simulated concentrations averaged for LRT days similarly decreased from west to east. The $PM_{2.5}$ concentrations averaged for LRT days were generally higher than those for LP days, particularly in the western areas. The underestimation of $PM_{2.5}$ was larger in LP days (MBE = −5.5 ± 5.1 μg m^{-3} and NMB = −26 ± 24%) than LRT days (MBE = −2.8 ± 3.1 μg m^{-3} and NMB = −10 ± 12%). The results indicate that it is still essential to improve local emissions of precursors and primary $PM_{2.5}$ and local AQM subprocesses including formation and loss processes of precursors and $PM_{2.5}$ in Japan. In addition, because of the larger underestimation of $PM_{2.5}$ concentration in LP days than LRT days, the estimated contribution ratio of LRT to $PM_{2.5}$ (50%) was likely somewhat overestimated in this study.

4. Conclusion

CMAQ v5.0.2 driven with WRF v3.5.1 was applied to air quality simulations in Japan, which is strongly affected by LRT from anthropogenic sources in the Asian Continent, for one year from April 2010 to March 2011. The CMAQ performance for simulating LRT and LP of $PM_{2.5}$ in Japan was evaluated to provide information for future improvement of AQM performance.

The CMAQ performance for simulating $PM_{2.5}$ was better than that of its precursors, SO_2 and NO_2. The model well captured temporal and spatial variation patterns of $PM_{2.5}$ concentration, including the occurrence of several high concentration peaks and a decreasing trend from west (closer to the continent) to east in Japan. However, the model tended to underestimate the concentration level (by 15% on average). For major $PM_{2.5}$ components, the model successfully simulated EC, sulfate, and ammonium but underestimated OA by 59% and overestimated nitrate by 53%. The AQM processes that should be preferentially improved include SOA formations in warm season, POA emissions in cool season, and the atmospheric behavior and emissions of NH_3.

The contribution of LRT was estimated from the difference between the results of the EBase and EJapan cases. Annual mean estimated contribution ratios of LRT to NO_2, SO_2, and $PM_{2.5}$ concentrations in Japan were, respectively, 1, 13, and 50%, with the contribution ratios being generally higher in the western areas of Japan. The contribution of LRT explained most of high $PM_{2.5}$ concentration peaks and the decreasing trend from west to east. The higher LRT contribution ratio to $PM_{2.5}$ compared to SO_2 and NO_2 indicated that these precursors reacted to form sulfate and nitrate during the LRT process before arriving at Japan, and LP controlled the concentration level of these precursors in Japan.

In order to evaluate the CMAQ performance for LRT and LP of $PM_{2.5}$, days that were dominantly affected by LRT or by LP were determined based on the contribution of LRT to sulfate, which was fairly well simulated and strongly affected by LRT. LP days were frequent in summer and LRT days were frequent in spring and winter. The underestimation of $PM_{2.5}$ was larger in LP days (by 26% on average) than LRT days (by 10% on average). Therefore, it is essential to improve local emissions of precursors and primary $PM_{2.5}$ and local AQM subprocesses including formation and loss processes

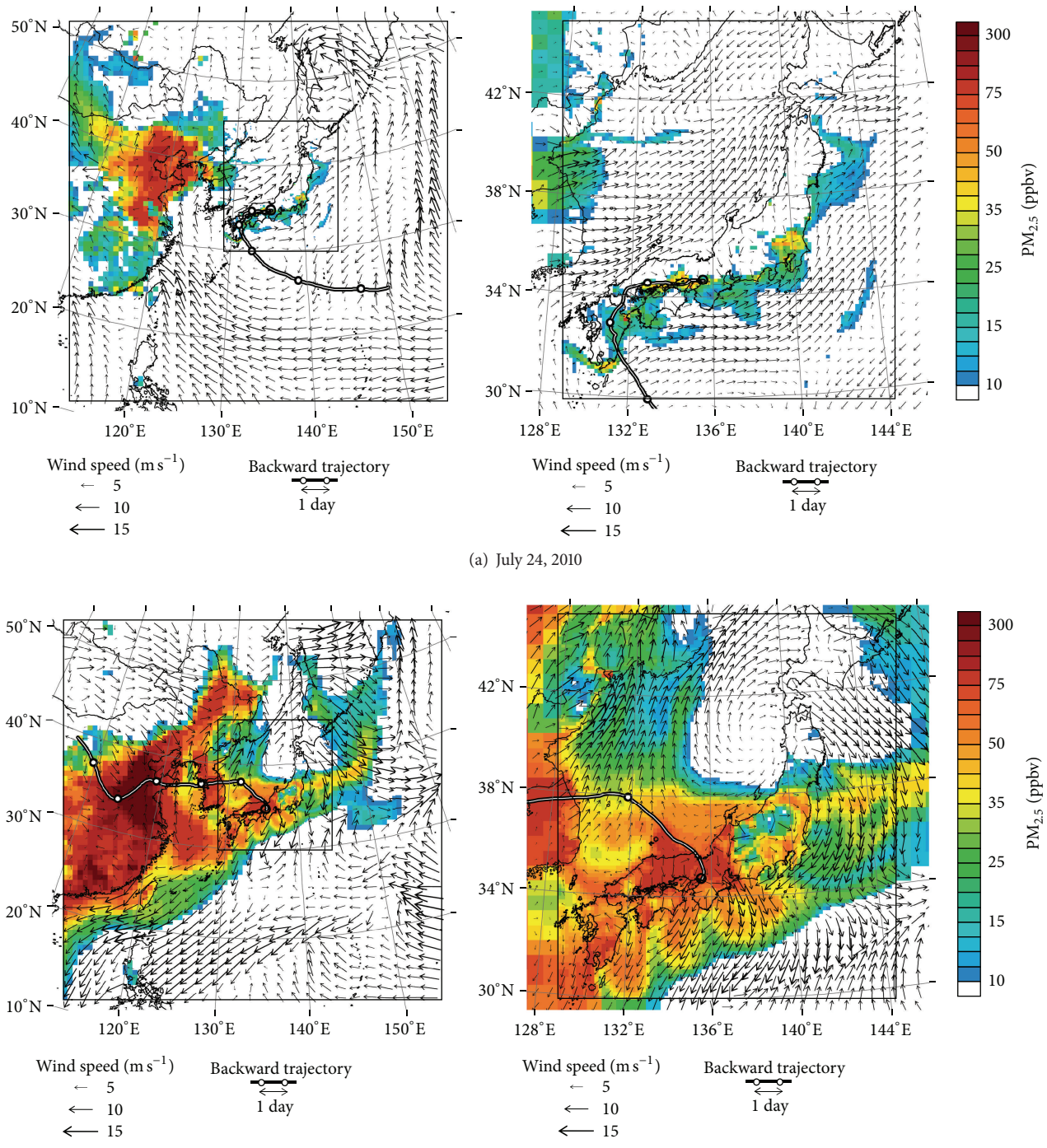


FIGURE 9: Spatial distributions of simulated ground-level $PM_{2.5}$ concentrations in D1 (left) and D2 (right) with horizontal wind vectors and backward trajectories arriving at 300 m above Kokusetsu-Osaka station at 0000 (local time) on July 24, 2010, and February 6, 2011.

of precursors and $PM_{2.5}$ in Japan. Because of the larger underestimation of LP in Japan, the estimated contribution ratio of LRT to $PM_{2.5}$ was likely somewhat overestimated in this study.

Competing Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

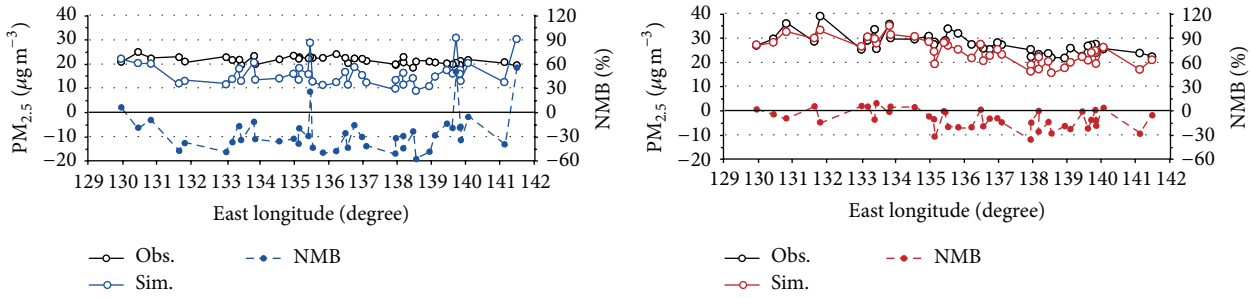


FIGURE 10: Comparisons of observed and simulated $PM_{2.5}$ concentrations averaged for LP and LRT days in JFY2010 at the ambient air pollution monitoring stations in D2.

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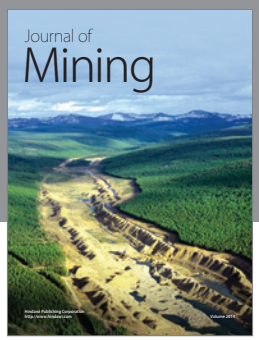
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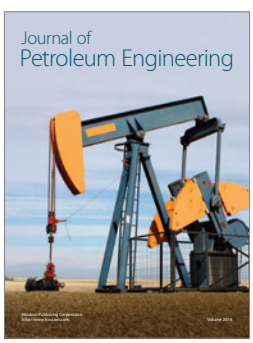
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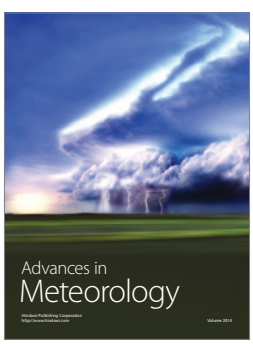
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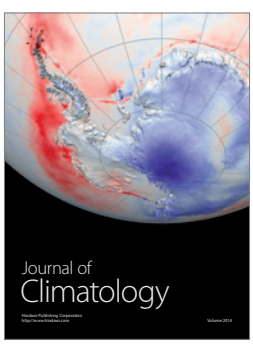
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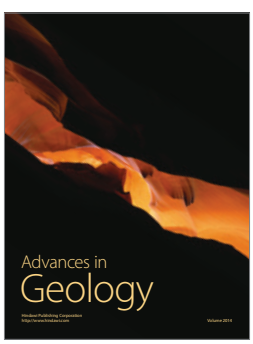
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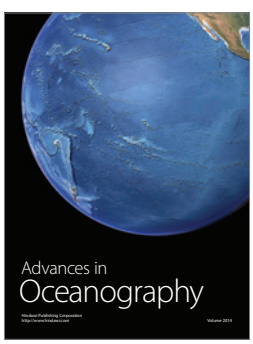
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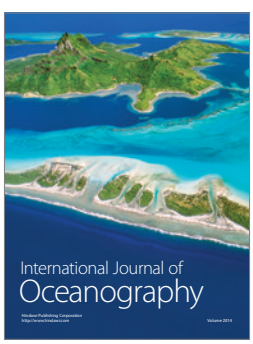
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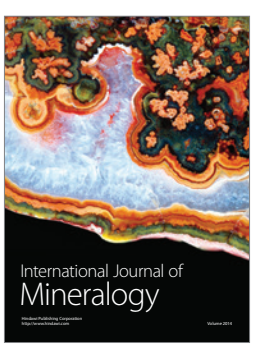
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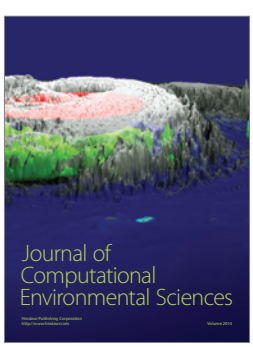
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