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Urban Air Quality Model Inter-Comparison Study (UMICS) for Improvement of $PM_{2.5}$ Simulation in Greater Tokyo Area of Japan

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ABSTRACT

The urban model inter-comparison study (UMICS) was conducted in order to improve the performance of air quality models (AQMs) for simulating fine particulate matter ($PM_{2.5}$) in the Greater Tokyo Area of Japan. UMICS consists of three phases: the first phase focusing on elemental carbon (UMICS1), the second phase focusing on sulfate, nitrate and ammonium (UMICS2), and the third phase focusing on organic aerosol (OA) (UMICS 3). In UMICS2/3, all the participating AQMs were the Community Multiscale Air Quality modeling system (CMAQ) with different configurations, and they similarly overestimated $PM_{2.5}$ nitrate concentration and underestimated $PM_{2.5}$ OA concentration. Various sensitivity analyses on CMAQ configurations, emissions and boundary concentrations, and meteorological fields were conducted in order to seek pathways for improvement of $PM_{2.5}$ simulation. The sensitivity analyses revealed that $PM_{2.5}$ nitrate concentration was highly sensitive to emissions of ammonia (NH_3) and dry deposition of nitric acid (HNO_3) and NH_3, and $PM_{2.5}$ OA concentration was highly sensitive to emissions of condensable organic compounds (COC). It was found that $PM_{2.5}$ simulation was substantially improved by using modified monthly profile of NH_3 emissions, larger dry deposition velocities of HNO_3 and NH_3, and additionally estimated COC emissions. Moreover, variability in $PM_{2.5}$ simulation was estimated from the results of all the sensitivity analyses. The variabilities on CMAQ configurations, chemical inputs (emissions and boundary concentrations), and meteorological fields were 6.1-6.5, 9.7-10.9, and 10.3-12.3%, respectively.

Key words: Air quality simulation, Sensitivity analysis, Nitrate, Organic aerosol, Variability in simulation

1. INTRODUCTION

Particulate matter (PM) with an aerodynamic diameter of 2.5 μm or less ($PM_{2.5}$) is an atmospheric pollutant that consists of various chemical species, including the following five major components: sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), elemental carbon (EC) and organic aerosol (OA). Primary OA (POA) and EC are emitted directly through combustions of fossil fuel and biomass (Bond *et al*., 2004). Secondary OA (SOA) is formed from various volatile organic compounds (VOC) and the corresponding processes are highly complex and varied (Hallquist *et al*., 2009). $PM_{2.5}$ SO_4^{2-} and NO_3^- are typically secondary pollutants produced through oxidations of sulfur dioxide (SO_2) and nitrogen oxides (NO_X), respectively. $PM_{2.5}$ NH_4^+ is produced from ammonia (NH_3) and generally a counter ion of SO_4^{2-} and NO_3^-. $PM_{2.5}$ has been of increasing concern because of its adverse effects on human health (e.g., Goto *et al*., 2016; Nawahda *et al*., 2012). While air quality over Japan has been gradually improved, ambient $PM_{2.5}$ concentration still exceeds the environmental quality standard in Japan (Wakamatsu *et al*., 2013).

To design effective $PM_{2.5}$ control strategies, it is essential to use air quality models (AQMs) that represent detailed physical and chemical processes in the atmosphere. However, in the first model inter-comparison study in Japan (Morino *et al*., 2010a, b), participat-

ing AQMs were not able to adequately simulate mass concentrations of $PM_{2.5}$ and its major components in the Greater Tokyo Area, which includes one of the largest megacities in the world. In order to seek possible ways for improvement of the AQM performance for $PM_{2.5}$ simulation, the urban air quality model inter-comparison study in Japan (UMICS) was conducted (Chatani *et al*., 2014; Shimadera *et al*., 2014a). In UMICS, the major components of $PM_{2.5}$ in the Greater Tokyo Area of Japan were focused; common datasets, including meteorological, emission and boundary concentration data, were provided to evaluate uncertainties originating from AQMs; and participants conducted sensitivity analyses in their fields of expertise to identify factors influencing the AQM performance for $PM_{2.5}$ simulation. UMICS consists of three phases: the first phase focusing on EC in summer 2007 (UMICS1), the second phase focusing on SO_4^{2-}, NO_3^- and NH_4^+ in winter 2010 and summer 2011 (UMICS2), and the third phase focusing on OA in winter 2010 and summer 2011 (UMICS3).

In UMICS1 (Chatani *et al*., 2014), the Community Multiscale Air Quality modeling system (CMAQ) (Byun and Schere, 2006) with seven different configurations and the Regional Air Quality Model 2 (RAQM2) (Kajino *et al*., 2012) participated in the inter-comparison of simulated $PM_{2.5}$ EC concentration. All the eight participating AQMs underestimated $PM_{2.5}$ EC concentrations. Results of sensitivity analyses implied that local emissions, vertical diffusion and long-range transport were critical factors for the AQM performance. According to Shimadera *et al*. (2016, 2013), CMAQ well simulates the effect of long-range transport from the Asian Continent on $PM_{2.5}$ concentration in Japan. On the other hand, vertical diffusion process has not been sufficiently verified because of lack of observed vertical concentration profiles.

In UMICS2 (Shimadera *et al*., 2014a), CMAQ with five different configurations participated in the inter-comparison of simulated $PM_{2.5}$ SO_4^{2-}, NO_3^- and NH_4^+ concentrations. The different configurations approximately reproduced $PM_{2.5}$ SO_4^{2-} concentration, but consistently overestimated $PM_{2.5}$ NO_3^- concentration. Sensitivity analyses revealed that simulated $PM_{2.5}$ NO_3^- concentration was highly sensitive to NH_3 emissions and dry deposition of nitric acid (HNO_3) and NH_3. The importance of NH_3 emissions for $PM_{2.5}$ simulation has been also emphasized in other studies (e.g., Gonzalez-Abraham *et al*., 2015; Kim *et al*., 2014). The results indicate that larger dry deposition velocities of the precursor gases may improve the overestimation of $PM_{2.5}$ NO_3^- concentration, and the finding has been applied to some air quality modeling studies in Japan (Itahashi *et al*., 2017; Morino *et al*., 2015).

This paper describes additional results of UMICS2 on $PM_{2.5}$ NO_3^- simulation, and results of UMICS3 on $PM_{2.5}$ OA simulation. In addition, variability in $PM_{2.5}$ simulation was estimated from results of all the sensitivity analyses on CMAQ configurations, chemical inputs (emissions and boundary concentrations), and meteorological fields. Findings obtained in UMICS will be useful for future air quality modeling studies including a new model inter-comparison project called Japan's study for reference air quality modeling (J-STREAM) (Chatani *et al*., 2018).

2. METHODOLOGY

2.1 Common Dataset

Common datasets, including meteorological, emission and boundary concentration data, were prepared for UMICS2/3. Fig. 1 shows common modeling domains and the target area for UMICS2/3. The computational domain consists of three nested horizontal domains from East Asia domain with 64-km grid cells (D1), East Japan domain with 16-km grid cells (D2), Kanto domain with 4-km grid cells (D3), and 30 vertical layers from the surface to 100 hPa with the middle height of the first layer being approximately 28 m. For comparisons between model results, the target area is defined as land areas with elevations lower than 200 m above sea level in D3. The target area includes the Greater Tokyo Area that is the largest urban areas in Japan, and therefore is characterized by huge population and air pollutant emissions. Field campaigns were conducted at Kisai, Komae, Maebashi, Saitama and Tsukuba in the Greater Tokyo Area (Fig. 1) to provide observation data for model validation (Ministry of the Environment, 2013).

Meteorological fields were produced using the Weather Research and Forecasting model (WRF) (Skamarock and Klemp, 2008) v3.2.1 for periods from November 15 to December 5, 2010 and July 11 to 31, 2011. Topography height and land use were obtained from the United States Geological Survey (USGS) data with 30-sec resolution. Initial and boundary conditions were derived from the final analysis data of the U.S. National Centers for Environmental Prediction (NCEP FNL) and the high-resolution, real-time, global sea surface temperature analysis (RTG_SST_HR) data of NCEP. WRF was configured with the cumulus parameterization of Kain (2004) (D1 and D2), the Asymmetrical Convective Model v2 (ACM2) (Pleim, 2007), the 5-class microphysics scheme of Hong *et al*. (2004), the Pleim-Xiu land surface model (PX LSM) (Xiu and Pleim, 2001), the longwave radiation scheme of Mlawer *et al*. (1997) and the shortwave radiation scheme of Dudhia (1989). Shimadera *et al*. (2014a) showed that the WRF simula-

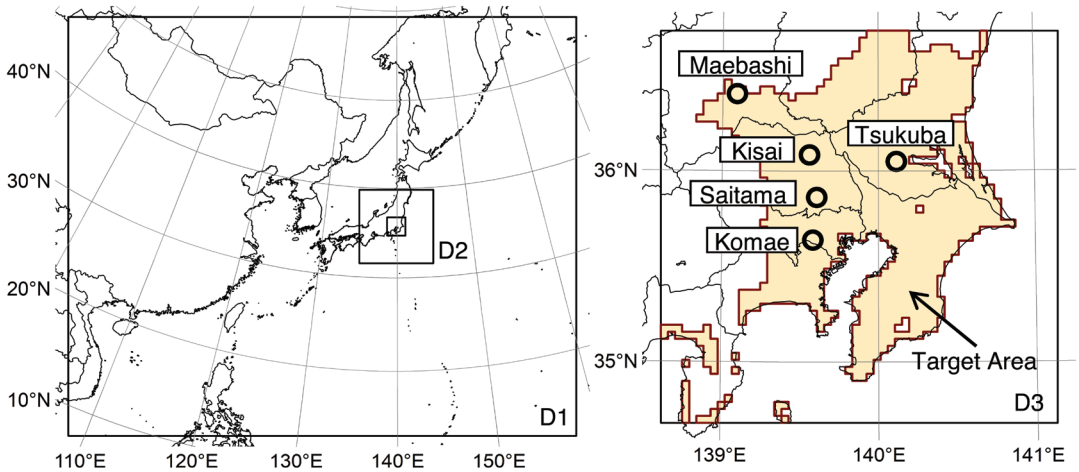

Fig. 1. Common modeling domains and the target area for UMICS2/3 with location of observation sites.

Table 1. Configurations of AQMs participating in UMICS2/3.

	M0	M1	M2	M3	M4	M5
AQM	CMAQ v4.7.1	CMAQ v4.7.1	CMAQ v4.6	CMAQ v4.7.1	CMAQ v5.0	CMAQ v5.0.1
Met.	MCIP v3.6	MCIP v3.6	MCIP v3.6	MCIP v3.6	MCIP v4.1	MCIP v3.6
Domain	D1, D2, D3	D3[a]	D3[a]	D1, D2, D3	D3[a]	D2[b], D3
Advection[c]	Yamartino/ Yamartino	Yamartino/ Yamartino	Yamartino/ Yamartino	PPM/ PPM[d]	Yamartino/ WRF	Yamartino/ WRF
Diffusion[c]	Multiscale/ ACM2	Multiscale/ ACM2	Multiscale/ ACM2	Multiscale/ ACM2	Multiscale/ ACM2	Multiscale/ ACM2
Photolysis	Lookup table	In-line calc[e]	Lookup table	In-line calc[e]	In-line calc[e]	In-line calc[e]
Chem.	SAPRC99 (EBI[f])	SAPRC99 (EBI[f])	SAPRC99 (ROS3[g])	SAPRC99 (EBI[f])	SAPRC99 (EBI[f])	SAPRC99 (EBI[f])
Aerosol	AERO5	AERO5	AERO4	AERO5	AERO5	AERO5
Cloud	RADM/ACM	RADM/ACM	RADM	RADM/ACM	RADM/ACM	RADM/ACM
Dry Dep.	Models-3	Models-3	Models-3	Models-3	Models-3	Models-3

[a]Boundary concentrations were derived from D2 of M0.
[b]Boundary concentrations were derived from D1 of M0.
[c]Horizontal/Vertical.
[d]Piecewise-Parabolic Method (Colella and Woodward, 1984).
[e]Binkowski *et al.* (2007).
[f]Euler Backward Iterative solver (Hertel *et al.*, 1993).
[g]Rosenbrock solver (Sandu *et al.*, 1997).

tions well agreed with the observed ground-level meteorology in D3.

Anthropogenic emissions in D1 were derived from an emission inventory by Zhang *et al.* (2009) except NH_3 and from an emission inventory by Ohara *et al.* (2007) for NH_3. Anthropogenic emissions in D2 and D3 were estimated with the Japan Auto-Oil Program (JATOP) vehicle emission estimate model (Chatani *et al.*, 2011) and the Georeference-Based Emission Activity Modeling System (Nansai *et al.*, 2004). Biogenic VOC (BVOC) emissions were estimated by using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther *et al.*, 2006) v2.04 with the common meteorological fields processed by the Meteorology Chemistry Interface Processor (MCIP). Common initial and boundary concentrations for D1 were obtained from the Model for Ozone and Related Chemical Tracers v4 (Emmons *et al.*, 2010).

2.2 Participating AQM

Table 1 summarizes configurations of AQMs participating in UMICS2/3. All the six participating AQMs (M0 to M4 in Shimadera *et al.* (2014a) and M5) are CMAQ with different configurations. M0 is the baseline

simulation of UMICS2/3. The Statewide Air Pollution Research Center v99 (SAPRC99) gas-phase chemistry (Carter, 2000) was used with the fourth or fifth generation CMAQ aerosol module (AERO4 or 5). Although emissions of sea salt particles were computed with the aerosol modules, the emission enhancement in the coastal surf zone (Kelly *et al*., 2010) was not considered in M0-M5 runs. The aqueous chemistry and wet deposition process were based on the Regional Acid Deposition Model (RADM) (Chang *et al*., 1987) in all the configurations. The sub-grid cloud process was based on RADM in M2 and ACM (Pleim and Chang, 1992) in the others. All the models used ACM2 for the vertical diffusion and the Models-3 routine (Pleim *et al*., 2001) for the dry deposition process.

M0 was driven by the common datasets in order to produce common boundary concentrations for the inner domains, which were used for M1, M2, M4, M5, and sensitivity runs. CMAQ runs were conducted for periods from November 15 to December 5, 2010 and July 11 to 31, 2011. The first seven days of each period are initial spin-up periods, and the next 14 days of each period are the target periods for comparisons between model results.

2. 3 Sensitivity Analysis

Table 2 summarizes all the sensitivity analysis cases in this study. The cases are categorized into CMAQ configuration (CC), emission and boundary concentration (EB) as chemical inputs, and meteorological model (MM). There are 8 cases for CC (CC01-08), 11 cases for EB (EB01-11) and 4 cases for MM (MM01-04). Each individual case focuses on a specific target and has 1-5 runs with different settings. There are 27, 24 and 11 runs in total for CC, EB and MM cases, respectively. The sensitivity was evaluated by the difference of mean $PM_{2.5}$ concentrations between the baseline and sensitivity runs in the target area (Fig. 1) during the target periods in November 22 to December 5, 2010 (winter 2010) and July 18 to 31, 2011 (summer 2011).

For CC cases, there are a comparison of participating CMAQ in UMICS2/3 (CC01) and vertical transport processes associated with the difference between CMAQ v4 and v5 (CC02), sensitivity analyses on factors influencing $PM_{2.5}$ NO_3^- simulation in UMICS2 (CC03/04/05/06), sensitivity analyses using different advection schemes (CC07) and the sixth generation CMAQ aerosol module (AERO6) (CC08). In CC02, the impact of reduction in the minimum eddy diffusivity introduced in CMAQ v5.0 (from 2.0 to 1.0 m^2 s^{-1} for urban areas and from 0.5 to 0.01 m^2 s^{-1} for the other areas) (Maker *et al*., 2014), changes in the vertical advection scheme or the treatment of non-precipitating cloud convection associated with the CMAQ and MCIP versions were investigated to complement the analysis in CC01. For UMICS2, CC03/04/05 were described in Shimadera *et al*. (2014a), and photolysis rates relevant to HNO_3 formation were reduced by 20% in CC06.

EB cases include sensitivity analyses on factors influencing $PM_{2.5}$ NO_3^- simulation in UMICS2 (EB01/03/07/08), factors influencing $PM_{2.5}$ OA simulation in UMICS3 (EB02/04/05/06), different emission inventories in Japan (EB09) and different boundary concentrations (EB10/11). For UMICS2, EB01/03 were described in Shimadera *et al*. (2014a). Emissions of nitrous acid (HONO), which is an important source of hydroxyl radical, from diesel-powered vehicles were estimated based on the concentration ratio of HONO to nitrogen dioxide (NO_2) measured by chassis dynamometer experiments (Ministry of the Environment, 2013) in EB07. The impact of sea salt emissions to $PM_{2.5}$ NO_3^- was investigated by increasing the emissions uniformly and by taking into account enhanced emissions from the coastal surf zone (Kelly *et al*., 2010) in EB08. For UMICS3, VOC emissions were uniformly changed in EB02. Anthropogenic VOC (AVOC) emissions from evaporative sources were respectively increased and decreased based on upper and lower limits of 95% confidence interval of emission factors (European Environment Agency, 2009) in EB04. The upper and lower limits of AVOC emissions were respectively larger and smaller by about 80% and 50% than those in the common emission data. BVOC emissions were estimated by MEGAN v2.04 and v2.1 (Guenther *et al*., 2012) with land cover data obtained from the Japan Integrated Biodiversity Information System (J-IBIS) in EB05 (Chatani *et al*., 2015). Emissions of semi-volatile organic compounds (SVOC) were estimated based on the Pollutant Release and Transfer Register (PRTR) system of the Ministry of Economy, Trade and Industry in Japan (2011) and emissions of condensable organic compounds (COC) from large point sources were estimated based on a survey on emission sources by Tokyo Metropolitan Government (2011) in EB06. SVOC with boiling point of 240 to 400°C (SVE400), SVOC with boiling point less than 240°C and not included as VOC in the common emission data (SVE240), or COC (COCE) were assumed to be emitted as non-volatile POA in the case. The total POA emissions in SVE400, SVE200 and COCE were respectively larger by 3, 277, and 597% than that of the common emission data. In EB09, anthropogenic emissions in Japan were replaced by the JATOP emission inventory database (JEI-DB) (Japan Petroleum Energy Center, 2012a, b) (EJ_JEI) or an emission inventory called EAGrid2000-JAPAN (Kannari *et al*., 2007) (EJ_EAG). The amounts of anthropogenic NO_X, SO_2 and $PM_{2.5}$ emissions changed from the common

Table 2. Sensitivity analysis cases in UMICS2/3.

ID	Target	Baseline	Modified setting
CC01	CMAQ participating in UMICS2/3	M0	M1, M2, M3, M4, M5
CC02	CMAQ v4 and v5	M0	M1 with minimum eddy diffusivity of CMAQ v5 (M1Kmod); M4 with Yamartino (M4yamo); M5 with MCIP v4.1 (M5MC41) in D3
CC03	Gas/particle equilibrium	M0	Uniform change of temperature by ±2 K (T+2; T-2); relative humidity by ±10% (RH+10; RH-10) in aerosol module in D3
CC04	Dry deposition	M1	Uniform multiplication of HNO_3 and NH_3 dry deposition velocities by 5 (Vd5); 2 (Vd2); 0.5 (Vd0.5); 0.2 (Vd0.2) in D3
CC05	Heterogeneous reaction	M0	N_2O_5 reaction probability set to 0 (Γ0); 0.1 (Γ0.1); and calculated by the method in AERO3 (Γae3); AERO4 (Γae4) in D3
CC06	Photolysis	M1	Reduction of photolysis rate of HNO_3 (jHNO3); NO_2 (jNO2); $O_3 \rightarrow O$ (1D) (jO3O1D); $O_3 \rightarrow O$ (3P) (jO3O3P) by 20% in D3
CC07	Advection scheme	M3	Eddy (M3eddy); Yamartino (M3yamo) in D1/D2/D3
CC08	Aerosol scheme	M5	AERO6 (M5ae6) in D2/D3
EB01	NO_X emission	M1	Uniform change by from −40% to +40% (NE-40; NE-20; NE+20; NE+40) in D3
EB02	VOC emission	M1	Uniform change by from −40% to +40% (VE-40; VE-20; VE+20; VE+40) in D3
EB03	NH_3 emission	M0	Monthly profile (larger in winter and smaller in summer) (NH_3Emod) in D2/D3
EB04	AVOC emission	M0	Evaporative AVOC with maximum (EVEmax); minimum (EVEmin) in 95% confidence interval of emission factor in D3
EB05	BVOC emission	M5	MEGAN v2.04 with J-IBIS (JMG20); MEGAN v2.1 with J-IBIS (JMG21) in D2/D3
EB06	SVOC and COC emissions	M5	Additional emission of SVOC (as POA) with boiling point 240 to 400°C (SVE400); <240°C (SVE240); and additional emission of COC (as POA) (COCE) in D2/D3
EB07	HONO emission	M5	HONO from diesel-powered vehicles with HONO/NO_2 ratio of 1% (HONO1); 5% (HONO5) in D2/D3
EB08	Sea salt emission	M5	Considering surf zone (SSEsurf); uniform 30% increase (SSE+30) in D2/D3
EB09	Emission inventory in Japan	M0	JATOP emission inventory in D1/D2/D3 (EJ_JEI); EAGrid2000-JAPAN in D2/D3 (EJ_EAG)
EB10	Boundary concentration	M0	D1 boundary concentration derived from CMAQ default profile (D1BDP)
EB11	Boundary concentration	M5	D3 boundary concentration derived from M0 (D3BM0)
MM01	WRF version	M0	WRF v3.4.1 (W341) in D1/D2/D3
MM02	PBL scheme	M0	YSU; MYJ; MYNN in D2/D3
MM03	Analysis data	M0	ACM2 (MACM2); MYJ (MMYJ) with JMA MANAL in D2/D3
MM04	Land surface	M0	ACM2/Noah with USGS (UAN); ACM2/PX with USGS (twofold roughness length) (UAPz0x2); ACM2/PX with JIBIS (JAP); ACM2/Noah with J-IBIS (JAN); MYJ/Noah with J-IBIS (JMN) in D2/D3

emission data by −1, +2 and −7% in EJ_JEI, and −8, −39 and +6% in EJ_EAG in the target area, respectively.

For MM cases, WRF v3.4.1 was utilized with the identical configurations described in subsection 2.1 in MM01. Three different PBL parameterizations, namely the Yonsei University (YSU) scheme (Hong *et al*., 2006), the Mellor-Yamada-Janjic (MYJ) scheme (Janjic, 1994) and the Mellor-Yamada Level-2.5 model proposed by Nakanishi and Niino (2006) (MYNN), with the Noah LSM (Chen and Dudhia, 2001) were compared with the ACM2 scheme with PX LSM in MM02. Initial and boundary conditions for WRF were obtained from the mesoscale objective analysis data of the Japan Meteorological Agency (JMA MANAL) with higher spatial and temporal resolutions than those of NCEP FNL in MM03. The impacts of land surface data and model on $PM_{2.5}$ simulations were investigated in MM04. The land use data obtained from J-IBIS were characterized by more widely distributed urban areas than the USGS data used in the WRF simulation to produce the common meteorological fields.

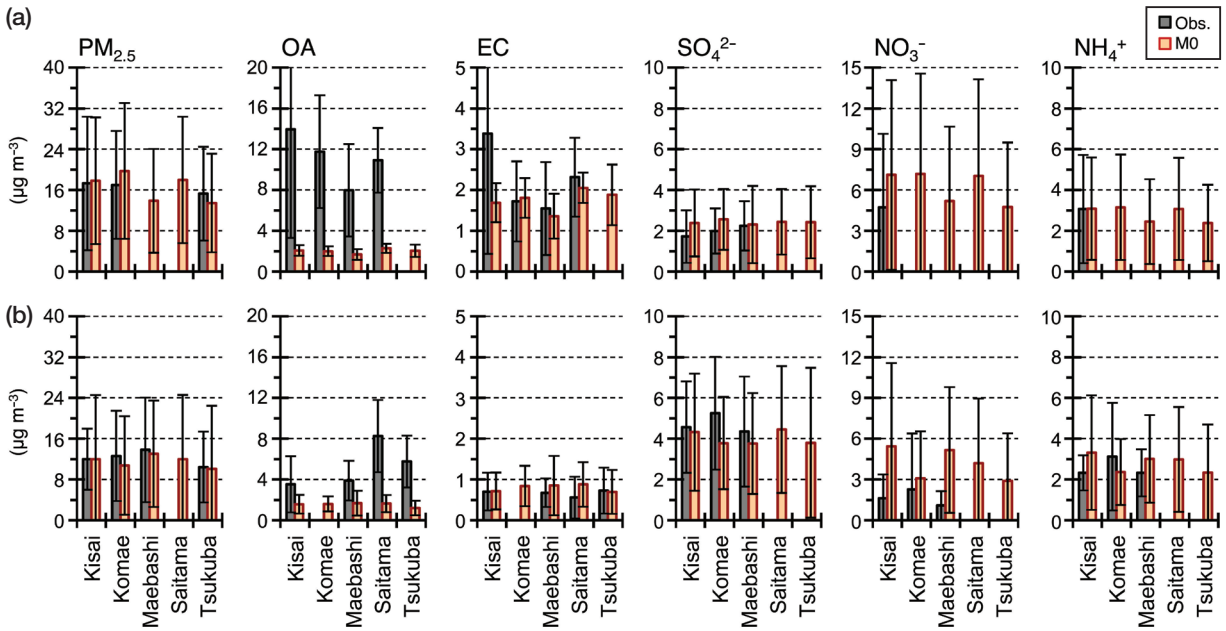

Fig. 2. Comparisons of observed and M0-simulated mean concentrations of $PM_{2.5}$ and its major components in (a) winter 2010 and (b) summer 2011. Error bars represent standard deviation.

3. RESULTS AND DISCUSSION

3.1 Comparison with Observed $PM_{2.5}$

Fig. 2 shows comparisons of observed and M0-simulated mean concentrations of $PM_{2.5}$ and the five major components in winter 2010 and summer 2011. The M0-simulated mean concentrations of $PM_{2.5}$, and $PM_{2.5}$ EC and SO_4^{2-} approximately agreed with the observations. However, the model overestimated $PM_{2.5}$ NO_3^- concentration and substantially underestimated $PM_{2.5}$ OA concentration at the observation sites. The model also overestimated total (gaseous + $PM_{2.5}$) nitrate and ammonium, and the overestimation of $PM_{2.5}$ NO_3^- likely attributed to overestimation of particulate NH_4NO_3 (Shimadera *et al.*, 2014a). The agreement of mean $PM_{2.5}$ concentrations between the observation and simulation does not indicate a good model performance because it is due to compensation of the overestimations and underestimations of $PM_{2.5}$ components. Sensitivity analyses to identify influencing factors on the overestimation of $PM_{2.5}$ NO_3^- and the underestimation of $PM_{2.5}$ OA were conducted in UMICS2 and UMICS3, respectively.

3.2 Comparison of Participating AQM

Fig. 3 shows percentage differences between M0 and the other participating AQMs, all of which are CMAQ with different configurations, in UMICS2/3 for mean concentrations of $PM_{2.5}$ and its major components in the target area during the target periods in winter 2010 and summer 2011. While the differences between the results of M0 and those of M1 and M3, which are CMAQ v4.7.1, were relatively small, M2 showed relatively large differences compared to the others because of various different configurations, such as CMAQ version, gas-phase chemistry solver, aerosol and aqueous modules. M4 and M5, which were CMAQ v5.0, tended to predict higher ground-level concentrations for primary pollutants such as $PM_{2.5}$ EC that was mainly emitted from ground-level emission sources.

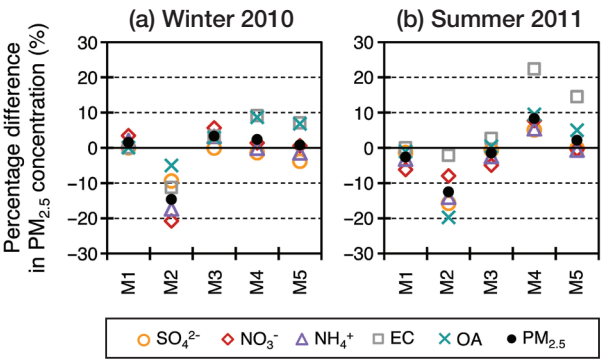

Fig. 3. Percentage differences between M0 and the other participating AQMs in UMICS2/3 for mean concentrations of $PM_{2.5}$ and its major components in the target area during the target periods in (a) winter 2010 and (b) summer 2011.

The differences in simulated $PM_{2.5}$ concentrations among the participating CMAQ with different configu-

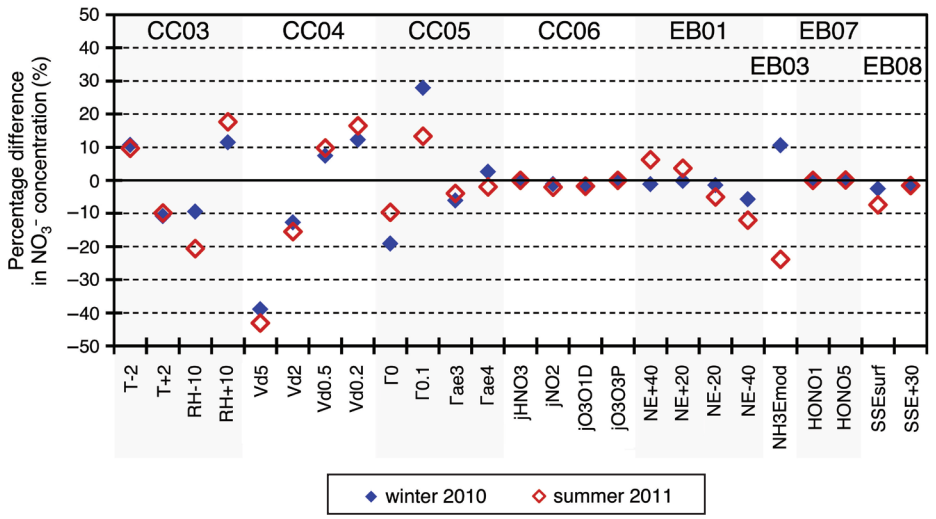

Fig. 4. Percentage differences between the baseline and sensitivity runs in CC03/04/05/06 and EB01/03/07/08 for mean $PM_{2.5}$ NO_3^- concentration in the target area during the target periods in winter 2010 and summer 2011.

rations typically ranged within 20%, which was smaller than the magnitudes of the overestimation of $PM_{2.5}$ NO_3^- and the underestimation of $PM_{2.5}$ OA by M0 (Fig. 2). Therefore, all the participating CMAQ shared common problems in $PM_{2.5}$ simulation.

3.3 Sensitivity Analysis on $PM_{2.5}$ NO_3^-

Fig. 4 shows results of UMICS2 by percentage differences between the baseline and sensitivity runs in CC03/04/05/06 and EB01/03/07/08 for mean $PM_{2.5}$ NO_3^- concentration in the target area during the target periods in winter 2010 and summer 2011. As described in Shimadera *et al.* (2014a), there were large sensitivities of $PM_{2.5}$ NO_3^- concentration to dry deposition velocities of precursor gases (CC04) and NH_3 emissions (EB03). Compared to these factors, the reduction in photolysis rates (CC06) and the estimated HONO emissions (EB07) showed quite small impacts on $PM_{2.5}$ NO_3^- simulation. In EB08, the increase in sea salt emissions slightly decreased $PM_{2.5}$ NO_3^- concentration, which was due to enhancement of the reaction between HNO_3 and sea salt aerosols to produce coarse particulate NO_3^-. Although the sensitivity of $PM_{2.5}$ NO_3^- was smaller in EB08 than CC04 and EB03, the impacts of ocean-related phenomena including sea salt emissions should be further investigated given the regional characteristics of Japan surrounded by ocean.

The finding that larger dry deposition velocities of precursor gases may improve the overestimation of $PM_{2.5}$ NO_3^- has been applied to some air quality modeling studies in Japan (Itahashi *et al.*, 2017; Morino *et al.*, 2015). However, corresponding field measurements have to be performed in order to verify the finding. While several factors were investigated in UMICS, there are still other unconsidered factors relevant to the AQM performance for $PM_{2.5}$ NO_3^- simulation (e.g., a heterogeneous reaction of nitric acid to NOx (Li *et al.*, 2015)). In addition, although the CMAQ performance for $PM_{2.5}$ SO_4^{2-} simulation was better than that for $PM_{2.5}$ NO_3^-, some studies have reported that AQMs tend to underestimate $PM_{2.5}$ SO_4^{2-} concentration particularly in winter (e.g., Zheng *et al.*, 2015; Shimadera *et al.*, 2014b). Some of the remaining challenges in UMICS will be tackled in J-STREAM (Chatani *et al.*, 2018).

3.4 Sensitivity Analysis on $PM_{2.5}$ OA

Fig. 5 shows results of UMICS3 by simulated mean concentrations of mean POA, anthropogenic and biogenic SOA (ASOA and BSOA) in CC01 and EB02/04/05/06 in the target area during the target periods in winter 2010 and summer 2011. Although the fraction of SOA was higher in summer than in winter, POA accounted for most of $PM_{2.5}$ OA in the participating CMAQ in UMICS2/3 in the both seasons. Therefore, there were quite small sensitivities of $PM_{2.5}$ OA concentrations in EB02/04/05, in which VOC emissions were modified. Meanwhile, $PM_{2.5}$ OA concentrations substantially increased in SVE240 and COCE of EB06, in which POA emissions were increased based on the estimated SVOC and COC emissions, respectively. These results indicate the importance of estimates of additional emission sources that are not considered in the existing emission data for improvement of the AQM performance for $PM_{2.5}$ OA simulation.

It must be noted that the contributions of SVOC and

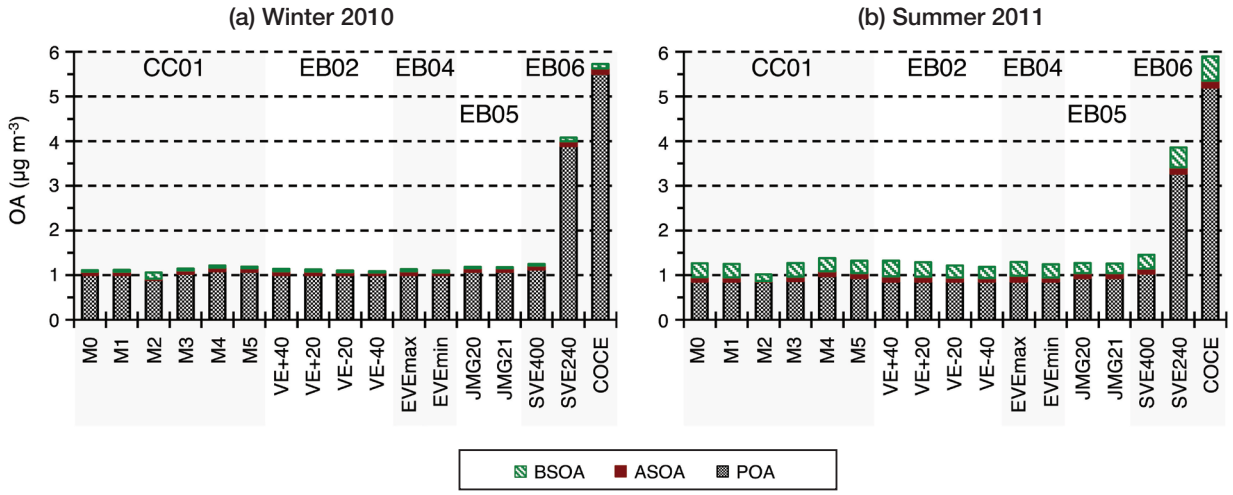

Fig. 5. Simulated mean $PM_{2.5}$ OA concentrations in CC01 and EB02/04/05/06 in the target area during the target periods in (a) winter 2010 and (b) summer 2011.

COC were likely overestimated in this study because these semi-volatile compounds were assumed to be non-volatile POA in EB06. AQMs incorporating the volatility basis set (VBS) approach (Robinson *et al*., 2007; Donahue *et al*., 2006), which can deal with direct emissions and gas-particle partitioning of these semi-volatile compounds as well as corresponding SOA formations, may provide more realistic results. According to Morino *et al*. (2015), the VBS approach improves OA simulation in warm season in Japan.

The fraction of oxygenated OA, which is a proxy of SOA, to the total OA concentration was estimated to be 44-55% at Kisai, Maebashi and Saitama (Fig. 1) according to the measurement and receptor modeling based approach by Hagino *et al*. (2012) in summer 2011. Therefore, the participating CMAQ in UMICS2/3 probably underestimated the formation of SOA and the sensitivity of $PM_{2.5}$ OA concentration to VOC emissions. Updated SOA formation processes, such as epoxide pathways of isoprene (Pye *et al*., 2013), have to be employed for further discussions.

3. 5 Improvement of $PM_{2.5}$ Simulation

Another sensitivity analysis (Mmod), in which modifications of multiple factors that could improve the $PM_{2.5}$ NO_3^- overestimation (Shimadera *et al*., 2014a) and the $PM_{2.5}$ OA underestimation were applied to M0, was conducted in D2 to D3. Mmod is different from M0 in using in-line photolysis rate calculation, modified monthly profile of NH_3 emission, fivefold dry deposition velocities of HNO_3 and NH_3, N_2O_5 parameterization in the third generation CMAQ aerosol module (AERO3), and COC emissions as POA.

Fig. 6 shows mean observed, M0- and Mmod-simulated concentrations of major $PM_{2.5}$ components at Kisai and Maebashi in summer 2011. While there were only slight changes in $PM_{2.5}$ EC and SO_4^{2-} concentrations between M0 and Mmod, $PM_{2.5}$ NO_3^- and OA concentrations substantially decreased and increased in Mmod compared to M0, respectively. As a result, the ratio of the five major components in the Mmod-simulation was much closer to that in the observation. This result indicates a possibility of improvement of the AQM performance for $PM_{2.5}$ simulation.

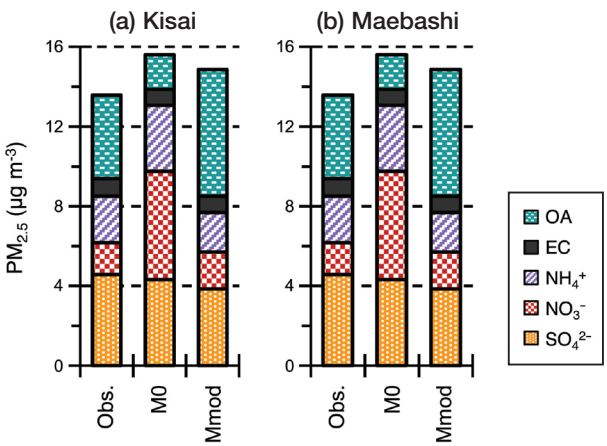

Fig. 6. Mean observed, M0- and Mmod-simulated concentrations of major $PM_{2.5}$ components at (a) Kisai and (b) Maebashi in summer 2011.

3. 6 Variability in $PM_{2.5}$ Simulation

Fig. 7 shows results of all the sensitivity analyses by percentage differences between the baseline and sensi-

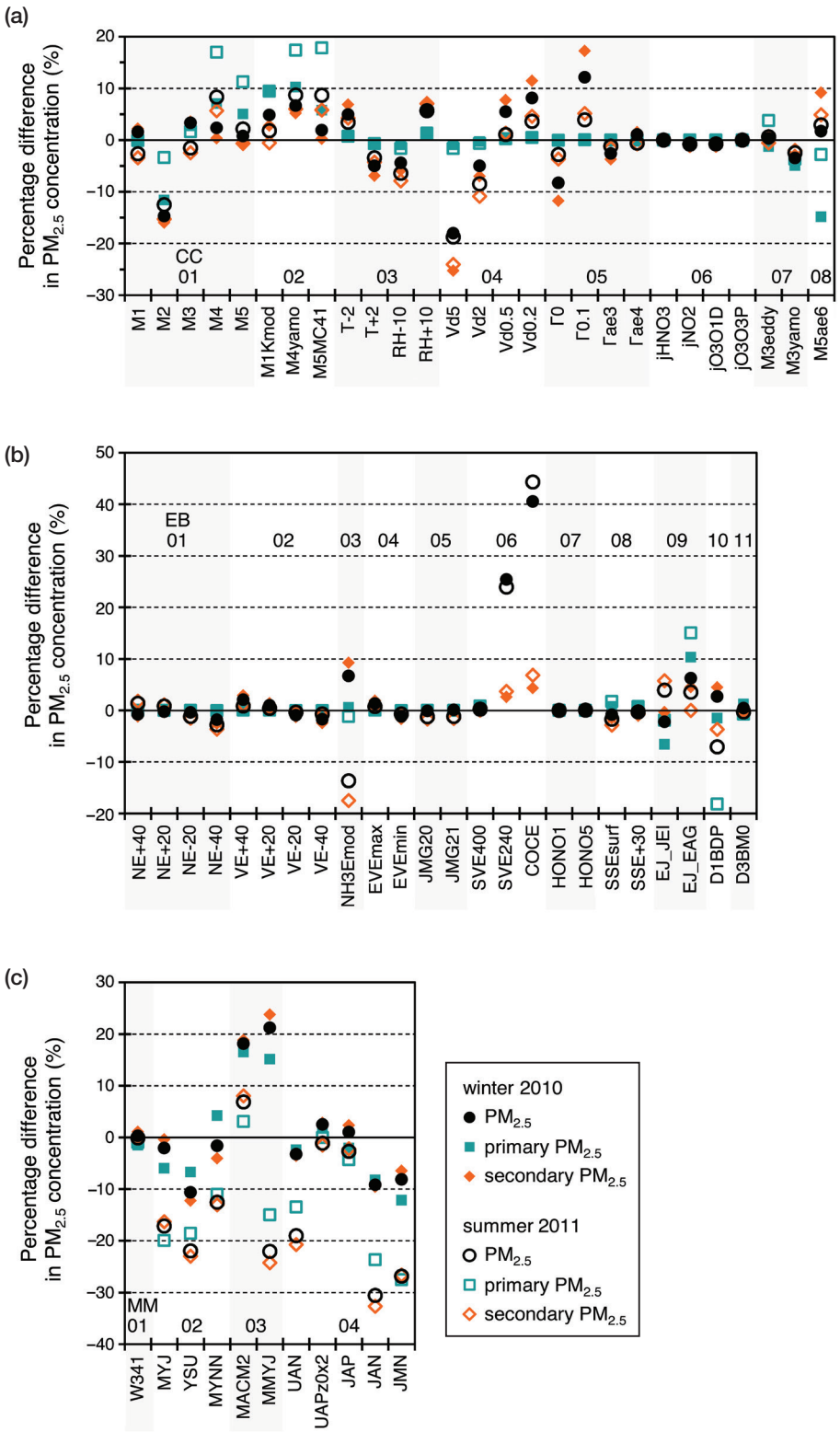

Fig. 7. Percentage differences between the baseline and sensitivity runs in (a) CC, (b) EB and (c) MM cases for mean concentrations of $PM_{2.5}$, primary $PM_{2.5}$ ($PM_{2.5}$-secondary $PM_{2.5}$) and secondary $PM_{2.5}$ ($SO_4^{2-} + NO_3^- + NH_4^+ + SOA$) in the target area during the target periods in winter 2010 and summer 2011.

tivity runs in CC, EB and MM cases for mean $PM_{2.5}$ concentrations in the target area during the target periods in winter 2010 and summer 2011. Secondary $PM_{2.5}$ concentration was assumed to be the total concentration of $PM_{2.5}$ SO_4^{2-}, NO_3^-, NH_4^+ and SOA.

In CC01/02, the difference between M1Kmod and M1 indicates that the smaller minimum diffusivity introduced in CMAQ v5.0 increased primary $PM_{2.5}$ concentration by about 10%. The difference between M5MC41 and M5 indicates that the difference between M4 and M5 in summer was attributed to the treatment of non-precipitating cloud convection associated with the MCIP versions. In CC07, the sensitivities of $PM_{2.5}$ concentration to the advection schemes were ranged within 5%. In CC08, the difference was mainly due to the treatment of aging of OA introduced in AERO6. Although UMICS2/3 compared results of $PM_{2.5}$ simulations by CMAQ with various different configurations, there was no comparison between AQMs with different model frameworks. Therefore, UMICS2/3 was an intra-comparison of CMAQ rather than an inter-comparison of AQMs. In addition, the effect of chemical scheme on $PM_{2.5}$ simulation was not sufficiently evaluated in UMI CS2/3 because all the CMAQ runs used the SAPRC99 gas-phase chemistry mechanism. In J-STREAM, inter-comparisons between different AQMs with different chemical schemes will be conducted for further comprehensive study (Chatani *et al*., 2018).

In EB09, the usage of different emission inventories in Japan caused larger differences in $PM_{2.5}$ simulation than in the sensitivity runs in EB01-08 except NH_3 Emod, SVOC240 and COCE runs. EB10 showed that the impacts of the change in boundary concentrations of D1 on air quality in the Greater Tokyo Area was comparable to those of the change in CMAQ configurations although air quality in the area was dominantly controlled by pollutants emitted within D1.

For MM cases, the change in WRF version had negligible impacts on $PM_{2.5}$ simulation in MM01. Meanwhile, the impacts of the changes in PBL scheme, analysis data and LSM in MM02-04 were larger than those in most sensitivity runs in CC and EB cases. The ACM2 scheme tended to predict higher ground-level $PM_{2.5}$ concentration because of lower PBL height compared to the other PBL schemes, particularly in summer 2011. The increase in urban areas in J-IBIS typically decreased ground-level $PM_{2.5}$ concentration because of enhanced vertical diffusion associated with higher ground-level air temperature and larger minimum eddy diffusivity in urban areas. The PX LSM, in which land use parameters in a grid cell are derived from fractional land use coverages in the grid cell, was less sensitive to the change in land use data than the Noah LSM, in which land use parameters in a grid cell are determined by a dominant land use category in the grid cell.

Variability in $PM_{2.5}$ simulation was estimated from the results of 27 runs for CC cases, 24 runs for EB cases and 11 runs for MM cases. Standard deviations of the percentage differences in CC, EB and MM cases were 6.5, 9.7 and 10.3% in winter 2010 and 6.1, 10.9 and 12.3% in summer 2011, respectively. Therefore, AQM input data are as important as, or more important than AQM configurations for improvement of the AQM performance for $PM_{2.5}$ simulation.

4. CONCLUSIONS

The model inter-comparison study called UMICS was conducted in order to improve the AQM performance for $PM_{2.5}$ simulation in the Greater Tokyo Area of Japan. In UMICS, there were three phases: UMICS1 focusing on EC (Chatani *et al*., 2014), UMICS2 focusing on SO_4^{2-}, NO_3^- and NH_4^+ (Shimadera *et al*., 2014a), and UMICS3 focusing on OA. This paper described the results of UMICS3 with the additional results of UM ICS2. In UMICS2/3, all the participating AQMs, which were six CMAQ with different configurations, similarly overestimated $PM_{2.5}$ NO_3^- and underestimated $PM_{2.5}$ OA. According to the additional sensitivity analyses in UMICS2, simulated $PM_{2.5}$ NO_3^- concentration was less sensitive to photolysis rates, HONO emissions and sea salt emissions than the two important factors for improvement of $PM_{2.5}$ NO_3^- simulation, namely NH_3 emissions and dry deposition velocities of NNO_3 and NH_3 that were identified in the previous study (Shimadera *et al*., 2014a). The sensitivity analyses in UMI CS3 focused on emissions of AVOC, BVOC, SVOC and COC. Because POA accounted for most of $PM_{2.5}$ OA in all the participating CMAQ, there were quite small sensitivities of $PM_{2.5}$ OA concentration to AVOC and BVOC emissions. Meanwhile, the increase in POA emissions based on the estimated SVOC and COC emissions substantially increased $PM_{2.5}$ OA concentration. Therefore, to estimate additional emission sources that are not considered in the existing emission data is one of the most important factors for improvement of the $PM_{2.5}$ OA underestimation. Based on the results of UMICS2/3, multiple factors, such as the modified NH_3 emissions, the increased HNO_3 and NH_3 dry deposition velocities and the estimated COC emissions, were applied to the baseline simulation case to improve the $PM_{2.5}$ NO_3^- overestimation and the $PM_{2.5}$ OA underestimation. As a result, the ratio of the five major components in the simulated $PM_{2.5}$ concentration approximately agreed with the observation. Variability in $PM_{2.5}$ simulation was estimated from all the sensitivity runs for CMAQ configurations, emission and boundary concen-

trations, and meteorological fields. The variability on input data was comparable to, or larger than that on CMAQ configurations, indicating the importance of the input data for improvement of the AQM performance. Although several important findings were obtained in UMICS, there are a lot of remaining challenges (e.g., corresponding measurements to verify the findings, inter-comparisons between different AQMs with different chemical schemes; improvement of SO_4^{2-} simulation, SOA formation from directly emitted semi-volatile organic species), some of which will be tackled in a new model inter-comparison project called J-STREAM (Chatani *et al*., 2018).

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