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COMPREHENSIVE SENSITIVITY ANALYSES ON AIR QUALITY MODEL PERFORMANCE FOR $PM_{2.5}$ SIMULATION

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Abstract: The urban air quality model inter-comparison study in Japan (UMICS) was conducted in order to improve the Community Multiscale Air Quality model (CMAQ) performance of the for $PM_{2.5}$ simulation. UMICS consist of three phases including the first phase focusing on elemental carbon, the second phase focusing on major ionic components (sulfate, nitrate and ammonium) and the third phase focusing on organic aerosol (OA) (UMICS1, 2 and 3). The results of UMICS3 for improvement of substantial OA underestimation were described in this paper. Because primary OA accounted for most of OA in the participating models, changes in volatile organic compounds emissions caused only slight changes in OA concentrations. Meanwhile, additional primary OA emissions because of a large amount of semi-volatile organic compounds and condensable organic compounds emissions substantially increased OA concentrations. The results emphasized the importance of emission sources that were not considered in the existing emission data. In addition, sensitivity analyses on various processes including meteorology and emission were conducted in order to show errors of $PM_{2.5}$ simulation originating from model input data and configurations. The results indicated that the importance of model input data is comparable to, or greater than that of model configurations in improvement of model performance for $PM_{2.5}$ simulation.

Key words: *Fine particulate matter, Model sensitivity, CMAQ, Organic aerosol, Emission*

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 sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), elemental carbon (EC) and organic aerosol (OA). Because of increasing concern of adverse health effects caused by $PM_{2.5}$, the Ministry of the Environment of Japan (MOE) introduced an air quality standard (AQS) for $PM_{2.5}$ concentration (35 μg m^{-3} for daily mean and 15 μg m^{-3} for annual mean) in 2009. Although $PM_{2.5}$ concentrations have decreased in recent years in Japan, the $PM_{2.5}$ AQS is not attained in most urban areas. 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, the performance of current AQMs for $PM_{2.5}$ simulation is not adequate for the purpose.

The urban air quality model inter-comparison study in Japan (UMICS) was conducted in order to improve AQM performance (Chatani et al., 2014; Shimadera et al., 2014). In UMICS, the major components of $PM_{2.5}$ in the Greater Tokyo Area of Japan are focused; common datasets, including meteorological, emission and boundary data, are provided to participating AQMs; participants conduct sensitivity runs in their fields of expertise. UMICS consist of three phases including the first phase focusing on EC, the second phase focusing on SO_4^{2-}, NO_3^- and NH_4^+, and the third phase focusing on OA (UMICS1, 2 and 3). In UMICS1, all the participating AQMs underestimated EC concentrations in the Greater Tokyo Area in

summer 2007; process analyses revealed that local EC emission and vertical diffusion were dominant factors that controlled EC concentrations (Chatani et al., 2014). In UMICS2, the participating AQMs approximately reproduced SO_4^{2-} concentrations in winter 2010 and summer 2011, but clearly overestimated production of ammonium nitrate; sensitivity analyses revealed that NH_3 emission and dry deposition of HNO_3 and NH_3 are key factors for improvement of NO_3^- simulation (Shimadera et al., 2014).

This paper describes results of UMICS3, which focused on OA in winter 2010 and summer 2011, and sensitivity analyses on various processes including meteorology and emission, which were conducted in order to show errors of $PM_{2.5}$ simulation originating from AQM input data and configurations.

METHODOLOGY

Fig. 1 shows the common modeling domains from East Asia domain (D1), East Japan domain (D2) to Kanto domain (D3) that includes the Greater Tokyo Area. The target area for comparisons between model results is defined as the surface layer in land areas with elevations < 200 m in D3, which includes the largest urban and industrial areas in Japan. Table 1 summarizes common datasets for UMICS3. Common meteorological fields were produced using the Weather Research and Forecasting model (WRF) v3.2.1. The target area and the common datasets are identical to those for UMICS2 (Shimadera et al., 2014).

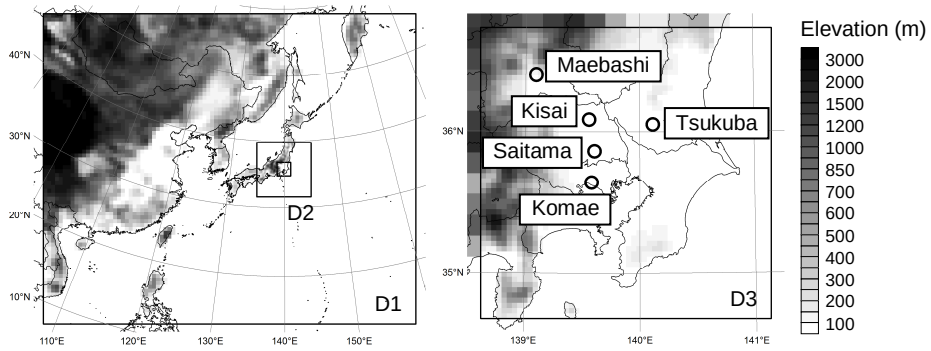

Figure 1. Modeling domains and locations of observation sites for $PM_{2.5}$ and its components

Table 1. Common datasets for UMICS3

	Original data
Meteorology (WRF v3.2.1)	Geography data: USGS 30-sec topography and land-use Analysis data: NCEP FNL and RTG_SST_HR Physics option: Kain-Fritsch, ACM2, WSM5, PX LSM and RRTM/Dudhia
Emission	Anthropogenic in land area in D1: INTEX-B v1.2 and REAS v1.11 Anthropogenic in land area in D2/D3: JATOP vehicle, G-BEAMS Ship: inventories by NMRI and by OPRF Natural sources: MEGAN v2.04 with default PFT, volcanic activity report by JMA
Boundary concentration of D1	MOZART-4

Table 2. Configurations of AQMs participating in UMICS3

	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
Domain	D1, D2, D3	D3	D3	D1, D2, D3	D3	D2, D3
H adv	Yamartino	Yamartino	Yamartino	PPM	Yamartino	Yamartino
V adv	Yamartino	Yamartino	Yamartino	PPM	WRF	WRF
H diff	Multiscale	Multiscale	Multiscale	Multiscale	Multiscale	Multiscale
V diff	ACM2	ACM2	ACM2	ACM2	ACM2	ACM2
Photolysis	Lookup table	In-line calc	Lookup table	In-line calc	In-line calc	In-line calc
Chem (Solver)	SAPRC99 (EBI)	SAPRC99 (EBI)	SAPRC99 (ROS3)	SAPRC99 (EBI)	SAPRC99 (EBI)	SAPRC99 (EBI)
Aerosol	AERO5	AERO5	AERO4	AERO5	AERO5	AERO5
Cloud	ACM	ACM	RADM	ACM	ACM	ACM
Dry dep	Models-3	Models-3	Models-3	Models-3	Models-3	Models-3

Table 3. List of sensitivity analysis cases

ID	Target	Base	Modified configurations
MM01	WRF version	M0	WRF v3.4.1 (M0W341)
MM02	PBL scheme	M0	YSU; MYJ; MYNN2.5 in D2/D3
MM03	Analysis data	M0	ACM2 (MACM2); MYJ (MMYJ) with JMA MANAL in D2/D3
MM04	Land surface	M0	MYJ/Noah with USGS land-use (UMN); ACM2/Noah with USGS (UAN); ACM2/PX with USGS (twofold Z0) (UAPz0x2); ACM2/PX with J-IBIS (JAP); MYJ/Noah with J-IBIS (JMN) 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) (NH3Emod)
EB04	AVOC emission	M0	Estimated evaporative AVOC emission 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)
EB06	SVOC emission	M5	Estimated SVOC emission with boiling point 240 to 400 °C (SVE400); < 240 °C (SVE240); (model input as primary OA emission)
EB07	COC emission	M5	Estimated COC emission (model input as primary OA emission) (COCE)
EB08	HONO emission	M5	Estimated HONO emission as 1% (HONO1); 5% (HONO5) against NO_2 from diesel-powered vehicles
EB09	Sea salt emission	M5	Considering surf zone (SSEsurf); uniform 30% increase (SSE+30) in D2/D3
EB10	Emission inventory in Japan	M0	JATOP emission inventory in D1/D2/D3 (JEI); EAGrid2000-JAPAN in D2/D3 (EAGJ)
EB11	Boundary concentration	M0	D1 boundary concentration derived from CMAQ default profile (D1BDP)
EB12	Boundary concentration	M5	D3 boundary concentration derived from M0 (D3BM0)
AQM01	CMAQ configuration	M0	M1, M2, M3, M4, M5
AQM02	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
AQM03	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
AQM04	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
AQM05	Photolysis	M1	Reduction of photolysis rate of NO_2 (jNO2); O3 → O1D2 (jO3O1D); O3→ O3P (jO3O3P); HNO3 (jHNO3) by 20%
AQM06	Advection scheme	M3	Eddy (M3eddy); Yamartino (M3yamo)
AQM07	MCIP version	M5	MCIP v4.1 (M5MC41) (Baseline case: MCIP v3.6)
AQM08	Vertical transport	M1	M1 with minimum eddy diffusivity of CMAQ v5 (M1Kmod); M4 with Yamartino (M4yamo) in D3
AQM09	Chemical scheme	M5	SAPRC99&AERO6 (S99A6); SAPRC07tc&AERO6 (S07A6)

Table 2 summarizes configurations of AQMs (M0 to M5) participating in UMICS3. All the models are the Community Multiscale Air Quality model (CMAQ) with different configurations, and M0 to M4 are the same as UMICS2. M0 results were used to produce common boundary concentrations for the inner domains, which were used for M1, M2, M3, 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. Table 3 summarizes the sensitivity analysis cases in this study. The cases are identified by sequentially-numbered categories: meteorological model (MM), emission/boundary concentration (EB) and AQM. The participating AQMs clearly overestimated NO_3^- and underestimated OA. Therefore, EB01, EB03, AQM02, AQM03 and AQM05 were conducted in UMICS2 (Shimadera et al., 2014), and EB02, EB04, EB05, EB06 and EB07 were conducted in UMICS3 to improve the model performance. The cases in UMICS3 focused on emissions of anthropogenic and

biogenic volatile organic compounds (AVOC and BVOC), semi-volatile organic compounds (SVOC), and condensable organic compounds (COC). SVOC and COC emissions were not considered in the common emission data. In addition, sensitivity analyses on various processes including meteorology and emission were conducted to show errors of $PM_{2.5}$ simulation originating from AQM input data and configurations.

RESULTS AND DISCUSSION

Figure 2 shows comparisons of the results of AQM01/07/08. Although the differences between the results of M0 and those of M1 and M3, which are CMAQ v4.7.1, are 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 are CMAQ v5, predicted higher ground-level concentrations of species that were strongly affected by ground-level emissions. The difference between M1Kmod and M1 indicates that higher concentrations in M4 and M5 are mainly attributed to smaller minimum diffusivity introduced in CMAQ v5.0. The difference between M5MC41 and M5 indicates that the difference between M4 and M5 in summer is attributed to the change in convective cloud parameterization by Meteorology Chemistry Interface Processor (MCIP) versions.

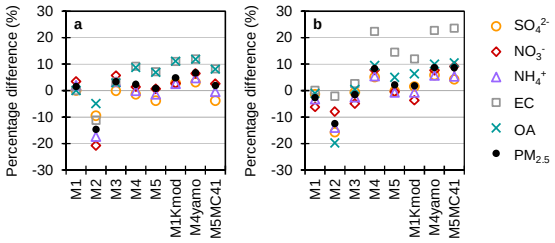

Figure 2. Percentage differences between M0 and AQM01/07/08 cases for mean $PM_{2.5}$ concentrations in the target area during the target periods in winter 2010 (a) and summer 2011 (b)

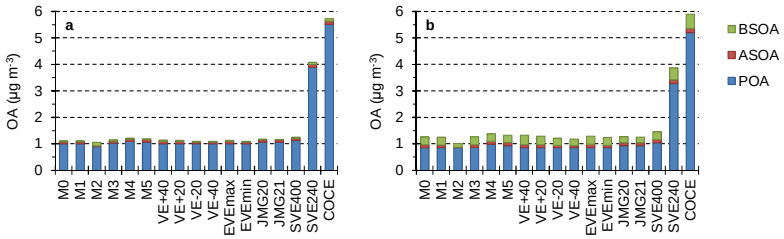

Figure 3. Simulated mean OA concentrations in AQM01, EB02/04/05/06/07 in the target area during the target periods in winter 2010 (a) and summer 2011 (b)

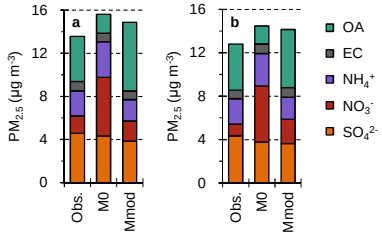

Figure 4. Mean observed, M0- and Mmod-simulated concentrations of major $PM_{2.5}$ components at Kisai (a) and Maebashi (b) in 24-29 July 2011

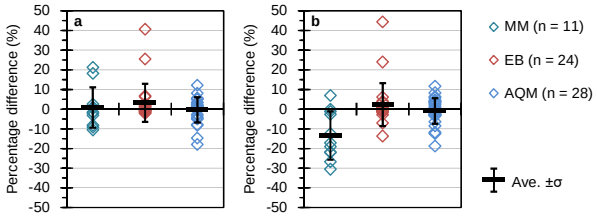

Figure 5. Percentage differences between all the sensitivity analyses and their baseline cases for mean $PM_{2.5}$ concentration in the target area during the target periods in winter 2010 (a) and summer 2011 (b)

Figure 3 shows simulated mean primary OA (POA), anthropogenic and biogenic secondary OA (ASOA and BSOA) concentrations in the sensitivity analysis cases of UMICS3. Although contribution of SOA is higher in summer than in winter, POA accounted for most of OA in the participating AQMs in UMICS3 in both season. Therefore, in EB02/04/05 in which VOC emissions were modified, there were only slight changes in OA concentrations. In EB06/07, the total POA emission of SVE400, SVE240 and COCE in D3 respectively larger by 3, 277 and 597% than that of the common emission data. Consequently, OA concentrations in SVE240 and COCE substantially higher than those in other cases. The results emphasize the importance of emission sources that are not considered in the existing emission data. It must be noted that the contribution of SVOC and COC should be overestimated in EB06/07 because SVOC and COC were assumed to be POA that was non-volatile component in CMAQ.

Another sensitivity analysis (Mmod), in which modifications of multiple factors that could improve NO_3^- overestimation (Shimadera et al., 2014) and OA underestimation were applied to M0, was conducted in D1 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 AERO3, and COC emissions as POA. Figure 4 shows mean concentrations of major $PM_{2.5}$ components at Kisai and Maebashi (Figure 1) in 24-29 July 2011, in which the simultaneous observation data of the five major components were only available. While there were only slight changes in EC and SO_4^{2-} concentrations between M0 and Mmod, NO_3^- and OA concentrations substantially decreased and increased in Mmod compared to M0, respectively. As a result, ratio of the five major components in Mmod-simulated $PM_{2.5}$ concentration was much closer to the observation.

Various sensitivity analyses summarized in Table 3 were conducted in order to show errors of $PM_{2.5}$ simulation originating from AQM input data and configurations. Figure 5 shows percentage differences between all the sensitivity analyses and their baseline cases for mean $PM_{2.5}$ concentration in the target area during the target periods. In 11 cases of MM, 24 cases of EB and 28 cases of AQM sensitivity runs, standard deviations of the percentage differences were 10.3, 9.7 and 6.4% in winter 2010 and 12.3, 10.9 and 6.5% in summer 2011. Therefore, AQM input data are as important as, or more important than AQM configurations for improvement of AQM performance for $PM_{2.5}$ simulation.

CONCLUSION

This paper described the results of UMICS3 for improvement of OA simulation. The sensitivity analyses in UMICS3 focused on emissions of AVOC, BVOC, SVOC and COC. Because POA accounted for most of OA in the participating AQMs in UMICS3, changes in VOC emissions caused only slight changes in OA concentrations. Meanwhile, a large amount of SVOC and COC emissions as additional POA emissions substantially increased OA concentrations. The results emphasize the importance of emission sources that are not considered in the existing emission data. Based on the results of UMICS2/3, modifications of multiple factors that could improve NO_3^- overestimation and OA underestimation were applied to M0. As a result, ratio of the five major components in the simulated $PM_{2.5}$ concentration became much closer to the observation. In addition, sensitivity analyses on various processes including meteorology and emission were conducted in order to show errors of $PM_{2.5}$ simulation originating from AQM input data and configurations. The results indicate that AQM input data are as important as, or more important than AQM configurations for improvement of AQM performance for $PM_{2.5}$ simulation.

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