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Evaluation of Ensemble Approach for O_3 and $PM_{2.5}$ Simulation

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ABSTRACT

Inter-comparison of chemical transport models (CTMs) was conducted among four modeling research groups. Model performance of the ensemble approach to O_3 and $PM_{2.5}$ simulation was evaluated by using observational data with a time resolution of 1 or 6 hours at four sites in the Kanto area, Japan, in summer 2007. All groups applied the Community Multiscale Air Quality model. The ensemble average of the four CTMs reproduced well the temporal variation of O_3 (r=0.65-0.85) and the daily maximum O_3 concentration within a factor of 1.3. By contrast, it underestimated $PM_{2.5}$ concentrations by a factor of 1.4-2, and did not reproduce the $PM_{2.5}$ temporal variation at two suburban sites (r=~0.2). The ensemble average improved the simulation of SO_4^{2-}, NO_3^-, and NH_4^+, whose production pathways are well known. In particular, the ensemble approach effectively simulated NO_3^-, despite the large variability among CTMs (up to a factor of 10). However, the ensemble average did not improve the simulation of organic aerosols (OAs), underestimating their concentrations by a factor of 5. The contribution of OAs to $PM_{2.5}$ (36-39%) was large, so improvement of the OA simulation model is essential to improve the $PM_{2.5}$ simulation.

Key words: Chemical transport model, Ensemble average, Ozone, $PM_{2.5}$, CMAQ

1. INTRODUCTION

Urban air pollution problems have not been resolved in Japan despite substantial efforts at controlling emissions during the last several decades. Recently, concentrations of primary aerosols (particles emitted directly into the atmosphere) have decreased in the Tokyo urban area because of a reduction in their emission, but concentrations of secondary aerosols (particles formed by gas-to-particle conversion in the atmosphere) have not been reduced as expected (Minoura *et al.*, 2006). In general, production pathways and atmospheric behavior of secondary pollutants are more complicated than those of primary pollutants, and concentrations of secondary pollutants do not decrease linearly with reductions in their precursors' emission rates. Furthermore, the contribution of trans-boundary transport to secondary pollutant concentrations in Japan is modest to large (e.g., Ohara *et al.*, 2008), so a multi-scale analysis is necessary to understand source-receptor relationships of secondary pollutants. To evaluate source-receptor relationships of O_3 and particulate material with diameter less than 2.5 μm ($PM_{2.5}$), chemical transport models (CTMs) are a useful tool, as they can explicitly simulate the physical and chemical processes that control their concentrations. However, CTM results typically include large uncertainties, because of problems with their input data (e.g., meteorological, boundary, and emissions data), the parameterization of each process, and missing science elements. Recently, it has been proposed that inter-comparison of CTMs can provide valuable information on the validity and variability of CTM results (e.g., Carmichael *et al.*, 2008; Hass *et al.*, 1997). Delle Monache and Stull (2003) have shown that the ensemble average of several CTMs may simulate O_3 concentrations better than the CTMs do individually. The ensemble approach has been applied to the evaluation of source-receptor relationships of O_3 and particulate species at hemispheric (Fiore *et al.*, 2009), regional (van Loon *et al.*, 2007), and local scales (Vautard *et al.*, 2007).

Table 1. Model settings of each individual modeling group.

	M1	M2	M3	M4
Meteorological model	WRF-ARW 2.2	MM5 v3.7	WRF-ARW v2.2*	MM5 v3.6*
CTM	CMAQ v4.6	CMAQ v4.5	CMAQ 4.6*	CMAQ 4.6*
Number of domains	3	3	3	2
Δx, Δy (km)	36/16/4	54/18/6	80/15/5	80/5

*RAMS v4.3 and CMAQ v4.4 were used for the coarse domain in the East Asia simulation.

We conducted an inter-comparison of CTMs for O_3 and $PM_{2.5}$ simulation for the Kanto area of Japan during summer 2007 and evaluated the CTM performance, focusing mainly on the model evaluation of secondary pollutants at urban scale in summer, when the level of photochemical air pollution is high in the Kanto area (e.g., Kondo *et al.*, 2010; Wakamatsu *et al.*, 1999). One advantage of this CTM inter-comparison is that the model performance for several $PM_{2.5}$ species was evaluated at multiple stations with high time resolution. In this paper, we evaluate the model performance of the ensemble approach to O_3 and $PM_{2.5}$ simulation. Morino *et al.* (2010) have reported the individual CTM results in detail.

2. METHODOLOGY

Four modeling research groups participated in this CTM inter-comparison. All four groups, the Central Research Institute of the Electric Power Industry (CRIEPI), the Japan Auto-Oil Program (JATOP), the Japan Weather Association (JWA), and the National Institute for Environmental Studies (NIES) groups, applied Community Multiscale Air Quality (CMAQ) CTMs. General information on the individual models is summarized in Table 1. Three groups used CMAQ v4.6, and the other group employed CMAQ v4.5. The smallest domain in all CTMs covered the Kanto area (~150×150 km^2) with a horizontal resolution of 4-6 km (Fig. 1). Two groups used the fifth-generation PSU/NCAR mesoscale meteorological model (MM5, Dudhia, 1993), and the other two groups used the Weather Research and Forecast model (WRF, Skamarock *et al.*, 2005). Emissions data were not unified; each group used a different data set. Details of the emissions data sets are reported by Morino *et al.* (2010). Hereafter, we designate the four CTMs as M1, M2, M3, and M4 in random order, because the objective of this CTM inter-comparison was not to rank individual models but to evaluate uncertainties in the CTM results. This is the same policy as was used in a previous CTM inter-comparison (Model Inter-Comparison Study Asia Phase II, Carmichael *et al.*, 2008). One CTM (M2) did not take into account emissions

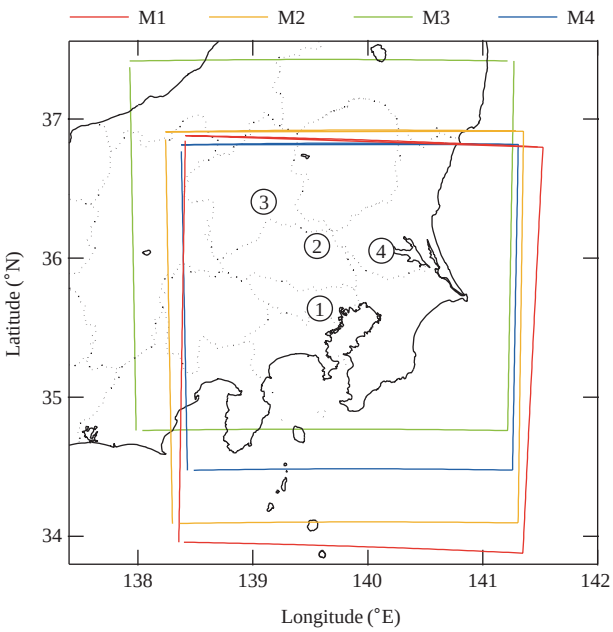

Fig. 1. Smallest domains of the individual CTMs (colored lines) and the measurement sites (numbered circles: 1, Komae; 2, Kisai; 3, Maebashi; 4, Tsukuba).

of elemental carbon (EC) or organic aerosols (OAs); thus, the M2 results were not used in the analysis of EC, OA, or total $PM_{2.5}$.

We evaluated model performance for O_3 and $PM_{2.5}$ simulation using observational data with a time resolution of 1 or 6 hours at four Kanto area sites in summer 2007 (Fig. 1). One site (Komae) is ~10 km southwest of the Tokyo urban center, and the other three sites (Kisai, Maebashi, and Tsukuba) are in suburban areas within 100 km of Tokyo. Details of the measurement techniques and sites are given by Hasegawa *et al.* (2008). To evaluate the $PM_{2.5}$ simulation, we analyzed the sum of the concentrations of five particulate species (EC, OA, SO_4^{2-}, NO_3^-, and NH_4^+; $\sum(PM_{2.5})$). The OA concentration was derived from that of organic carbon (OC) by assuming an organic mass-to-carbon ratio (OA/OC) of 1.6 (Turpin and Lim, 2001). As observed concentrations of NO_3^- and NH_4^+ at Tsukuba were not available (Morino *et al.*, 2010),

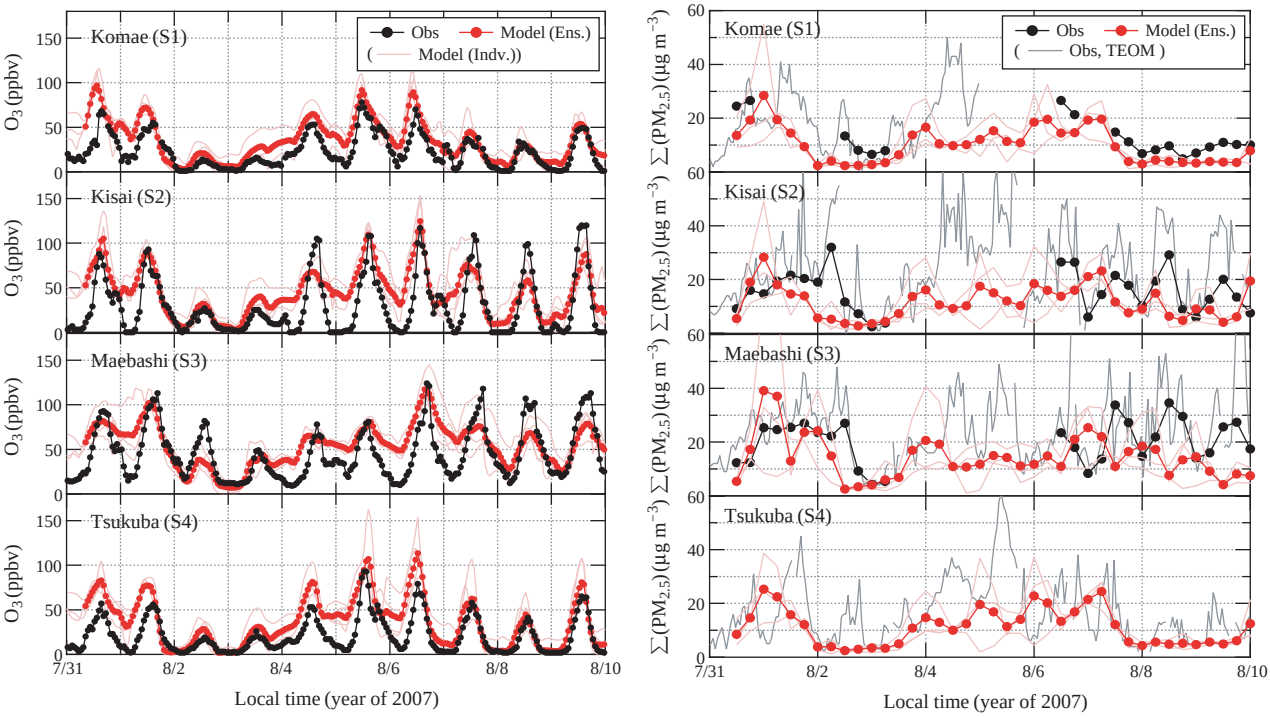

Fig. 2. Time series of O_3 (left) and $\sum(PM_{2.5})$ (right) concentrations from 31 July to 9 August 2007. Black circles indicate measurement results, and red circles indicate the ensemble average of the four CTMs. Pink lines indicate the individual CTM results. $PM_{2.5}$ concentrations measured by TEOM are also shown (gray lines).

we analyzed $\sum(PM_{2.5})$ at only three sites (Komae, Kisai, and Maebashi). The CTM inter-comparison was conducted for the period from 0900 Japan Standard Time (JST) on 31 July 2007 to 0900 JST on 10 August 2007.

3. RESULTS AND DISCUSSION

We compared time series of O_3 and $PM_{2.5}$ concentrations among observations, the ensemble average of the four CTMs, and the individual CTM results (Fig. 2). The observed O_3 concentration sometimes exceeded 100 parts per billion by volume (ppbv) at Kisai and Maebashi, which are downwind from Tokyo during the daytime, whereas it was lower than 100 ppbv at Komae and Tsukuba throughout the period. On 6-9 August 2007, a Pacific anticyclone was located over Honshu Island, Japan, and sea-land breeze circulation developed locally in the Kanto area. Under these meteorological conditions, the O_3 concentration usually becomes higher downwind of Tokyo than in the Tokyo urban area itself (e.g., Chang *et al.*, 1989). All four CTMs reproduced well the diurnal and interdiurnal variation in the O_3 concentration at Komae, though they sometimes overestimated the O_3 concentration itself. At Kisai and Maebashi (downwind of Tokyo), the O_3 concentration was underestimated by some CTMs during the daytime on 7-9 August, suggesting that CTM simulations may have problems simulating the O_3 production rate during downwind air mass transport from an urban area or the transport pathways of urban air masses to the surrounding area. Most CTMs reproduced well the diurnal and interdiurnal variation in the O_3 concentration at Tsukuba, though one CTM sometimes greatly overestimated the O_3 concentration.

The average model O_3 and $PM_{2.5}$ simulation performance during the analytical period is shown in Fig. 3. Three CTMs predicted well the average O_3 concentration, and in most cases, the correlation coefficient (r) between the observations and each model was higher than 0.6. All CTMs overestimated the average O_3 concentration at all four sites. One CTM (M1) greatly overestimated the O_3 concentrations, but the ensemble average of CTMs effectively reduced the effect of these outlying data. The correlation coefficients of the ensemble average of O_3 with observations were higher (0.65-0.85) than those of most individual CTMs at all four sites.

During nighttime, most CTMs overestimated the O_3 concentration at all four sites, which is one reason

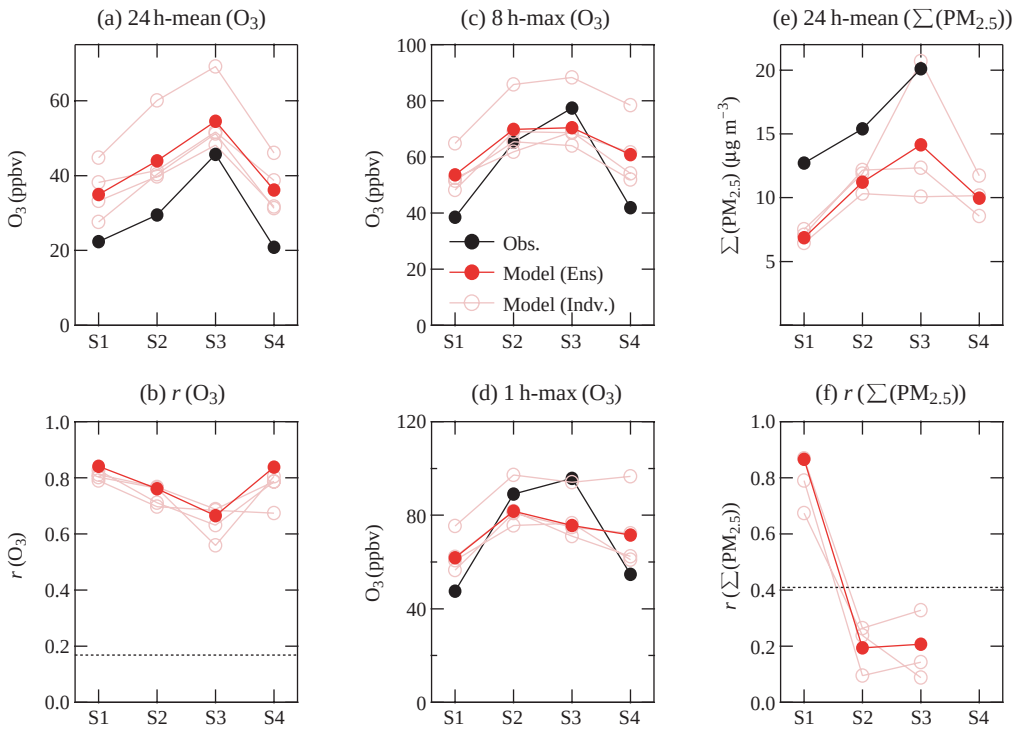

Fig. 3. Average model performance at the four measurement sites (S1, Komae; S2, Kisai; S3, Maebashi; S4, Tsukuba) during the analytical period (from 31 July 2007 to 10 August 2007). Fig. 3(a) and 3(e) show the average concentrations of O_3 and $\sum(PM_{2.5})$, respectively. Fig. 3(b) and 3(f) show the correlation coefficients between the observations and the CTM results. Fig. 3(c) and 3(d) show daily 8-h and 1-h maximum O_3 concentration, respectively. Red circles indicate the ensemble average of the CTMs, and pink circles indicate the individual CTM results. Dotted lines indicate the level of statistical significance ($P < 0.01$).

for the overestimation of the 24-hour average O_3 concentrations (Fig. 3(a)). This overestimation can be attributed to a poor model representation of O_3 titration by NO during nighttime (Morino *et al.*, 2010). For the evaluation of the model performance for daytime O_3 concentrations, we also compared daily 8- and 1-hour maximum O_3 concentrations between observations and CTM results (Fig. 3(c) and 3(d)), as recommended by the US Environmental Protection Agency (2007). The Japanese air quality standards are for hourly O_3 concentrations, whereas air quality standards in the United States have been established for daily 8-hour maximum O_3 concentrations as well as for hourly concentrations.

In general, the ratio of CTM results to observations decreased from the 24-hour average to the daily 8-hour and 1-hour maxima. Average O_3 concentrations for the 24-hour average, the daily 8-hour maximum, and the daily 1-hour maximum were reproduced within 77%, 45%, and 31%, respectively, by the ensemble average at all four sites. Although the ensemble average did not necessarily reproduce the O_3 concentrations better than the individual CTMs, it reduced the effect of outlying data (the large overestimation of O_3 by one CTM in this case) and effectively increased the reliability of the O_3 simulation by the CTMs.

$\sum(PM_{2.5})$ represented 60-80% of the total $PM_{2.5}$ concentration measured by tapered element oscillating microbalance (TEOM) (Fig. 2). The diurnal variation of $\sum(PM_{2.5})$ was smaller at Komae than at the other two sites where $\sum(PM_{2.5})$ was analyzed, whereas the inter-diurnal variation of $\sum(PM_{2.5})$ at Komae was roughly reproduced by all three CTMs. Observed $\sum(PM_{2.5})$ was distinctly higher during daytime than during nighttime at Kisai and Maebashi on 6-9 August. This behavior was not reproduced, however, by all three CTMs, and, accordingly, the ensemble average of the CTMs did not reproduce this daytime increase either. This underestimation is largely explained by the underestimation of the OA concentration, which increased during daytime (Morino *et al.*, 2010). All CTMs underestimated the average $\sum(PM_{2.5})$ concentration, and, consequently, the ensemble average underestimated the average $\sum(PM_{2.5})$ concentration by a factor of 1.4-2 (Fig. 3(e)). $\sum(PM_{2.5})$ was highest at Maebashi, and this behavior was reproduced by most CTMs. The correlation coefficient between observations and the ensemble average of $\sum(PM_{2.5})$

Table 2. Model:observation ratios of the ensemble average (Ens.) and the four individual CTMs (M1-M4) for $\sum(PM_{2.5})$, EC, OA, SO_4^{2-}, NO_3^-, and NH_4^+. Observed concentrations (Obs.) of these species are also given. The numbers in bold indicate the CTM results that are closest to the observation.

		Komae (S1)	Kisai (S2)	Maebashi (S3)	Tsukuba (S4)
$\sum(PM_{2.5})$	Ens.	0.54	0.73	0.70	NA*
	M1	**0.59**	0.77	**1.03**	NA*
	M2	NA*	NA*	NA*	NA*
	M3	0.56	**0.79**	0.61	NA*
	M4	0.51	0.67	0.50	NA*
	Obs.	12.72 μg/m^3	15.40 μg/m^3	20.10 μg/m^3	NA*
EC	Ens.	**0.78**	0.42	0.31	0.46
	M1	0.61	0.28	0.32	0.46
	M2	NA*	NA*	NA*	NA*
	M3	1.29	**0.69**	**0.44**	**0.69**
	M4	0.45	0.28	0.18	0.24
	Obs.	1.00 μg/m^3	2.22 μg/m^3	2.45 μg/m^3	1.57 μg/m^3
OA	Ens.	0.15	0.18	0.18	0.19
	M1	**0.19**	0.20	**0.26**	**0.26**
	M2	NA*	NA*	NA*	NA*
	M3	0.17	**0.21**	0.20	0.18
	M4	0.12	0.17	0.12	0.15
	Obs.	5.16 μg/m^3	5.85 μg/m^3	6.94 μg/m^3	4.30 μg/m^3
SO_4^{2-}	Ens.	0.77	1.11	**1.01**	**0.78**
	M1	0.75	**0.99**	**1.01**	0.77
	M2	0.61	0.92	0.88	0.49
	M3	0.71	1.07	0.94	0.70
	M4	**1.05**	1.57	1.30	1.23
	Obs.	4.65 μg/m^3	3.58 μg/m^3	4.16 μg/m^3	5.37 μg/m^3
NO_3^-	Ens.	**1.30**	1.80	1.37	NA*
	M1	3.13	3.25	3.11	NA*
	M2	0.40	**0.79**	**0.91**	NA*
	M3	1.58	2.44	1.10	NA*
	M4	0.21	0.68	0.33	NA*
	Obs.	0.97 μg/m^3	1.24 μg/m^3	3.07 μg/m^3	NA*
NH_4^+	Ens.	0.81	**1.06**	**0.96**	NA*
	M1	**1.07**	1.27	1.50	NA*
	M2	0.57	0.78	0.75	NA*
	M3	0.83	1.19	0.84	NA*
	M4	0.83	1.10	0.79	NA*
	Obs.	2.03 μg/m^3	1.94 μg/m^3	2.90 μg/m^3	NA*

was high (~0.9) at Komae, and low (~0.2) at Kisai and Maebashi. This poor correlation at Kisai and Maebashi reflected the large underestimation of daytime $\sum(PM_{2.5})$ at those sites.

Model performance for $PM_{2.5}$ species is summarized in Table 2. Concentrations of EC and OA were underestimated by factors of 1.2-3 and ~5, respectively, by the ensemble average. The contribution of OAs to $\sum(PM_{2.5})$ was large (36-39%); thus, underestimation of the OA concentration caused the poor model performance for $PM_{2.5}$, particularly at Kisai and Maebashi. Underestimation of the OA concentration has been reported by several previous studies (e.g., Volkamer *et al.*, 2006). In particular, the production pathways of secondary organic aerosols (SOAs) are mostly unknown. Improvement of the SOA simulation model is necessary to improve model performance for $PM_{2.5}$ simulation. The contribution of SO_4^{2-} to $\sum(PM_{2.5})$ is also large (21-34%). The SO_4^{2-} concentration was generally well reproduced by all four CTMs (within 60%), and it was reproduced within 23% by the ensemble average. This value (23%) is smaller than any of those of the individual CTMs, suggesting that the ensemble average effectively simulated the SO_4^{2-} concentration. Predicted concentrations of NO_3^- showed large variability among the four CTMs (up to a factor of 10), partly because the temporal and spatial distributions of

NO_3^- are inhomogeneous (e.g., Eder and Yu, 2006). The ensemble average reproduced the NO_3^- concentration within a factor of 2 at all three sites where it was measured, whereas none of the four CTMs individually reproduced the NO_3^- concentration within a factor of 2 at every site. These results suggest that the ensemble approach could effectively simulate NO_3^-. The model performance for NH_4^+ was roughly determined by the model performance for SO_4^{2-} and NO_3^- simulation (not shown).

In this study, the ensemble approach improved the model performance for SO_4^{2-} and NO_3^- simulation. This result suggests that current CTMs capture the factors controlling SO_4^{2-} and NO_3^- concentrations relatively well. The ensemble approach effectively reduced the uncertainties in CTM results for SO_4^{2-} and NO_3^-, which were partly caused by uncertainties in the CTM input parameters (e.g., emission inventories and boundary conditions). By contrast, the ensemble approach did not improve the model performance for OAs, reflecting that production pathways of OAs are mostly unknown. Improvement of the OA simulation model is essential. Also, the ensemble approach had problems simulating EC concentrations. Concentrations of primary aerosols, including EC, are generally sensitive to local meteorology and EC emission data. These aspects should also be addressed by future study.

4. CONCLUSIONS

In an inter-comparison of CTMs, we evaluated the model performance of the CTM ensemble average for O_3 and $PM_{2.5}$ simulation in the Kanto area in summer 2007. Three modeling groups used CMAQ v4.6 and the other employed CMAQ v4.5. Each group used a different emissions data set.

The ensemble average of the four CTMs better reproduced the temporal variation of O_3 (r=0.65-0.85) than the individual CTMs. Also, the ensemble average could effectively remove the effect of outlying data and reproduced the daily maximum O_3 concentration within 31% at all four sites. By contrast, the ensemble average did not reproduce the $\sum(PM_{2.5})$ temporal variation at two suburban sites (r=~0.2), and underestimated $\sum(PM_{2.5})$ by a factor of 1.4-2. The ensemble average improved the model performance for SO_4^{2-} and NO_3^-, whose production pathways are relatively well known, suggesting that the ensemble approach is suitable for SO_4^{2-} and NO_3^- simulation. However, the ensemble approach did not improve the model performance for OA or EC concentrations. As the contribution of OAs to the $PM_{2.5}$ concentration is large, the model performance for $PM_{2.5}$ was not greatly improved by the ensemble average. Improvement of the OA simulation model is essential for better $PM_{2.5}$ simulation.

All groups that we studied used CMAQ; thus, we did not evaluate CTM variability itself. Also, this was a case study during 10 days in summer. Further studies in other areas or in other seasons would help us better understand CTM performance.

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REFERENCES

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., Kannari, A., Fu, J., Matsuda, K., Thongboocho, 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.

Chang, Y.S., Carmichael, G.R., Kurita, H., Ueda, H. (1989) The transport and formation of photochemical oxidants in central Japan. Atmospheric Environment 23, 363-393.

Delle Monache, L., Stull, R.B. (2003) An ensemble air-quality forecast over western Europe during an ozone episode. Atmospheric Environment 37, 3469-3474.

Dudhia, J. (1993) A nonhydrostatic version of the Penn State/NCAR mesoscale model: Validation tests and simulation of an Atlantic cyclone and cold front. Monthly Weather Review 121, 1493-1513.

Eder, B., Yu, S. (2006) A performance evaluation of the 2004 release of Models-3 CMAQ. Atmospheric Environment 40, 4811-4824.

Fiore, A.M., Dentener, F.J., Wild, O., Cuvelier, C., Schultz, M.G., Hess, P., Textor, C., Schulz, M., Doherty, R.M., Horowitz, L.W., MacKenzie, I.A., Sanderson, M.G., Shindell, D.T., Stevenson, D.S., Szopa, S., Van Dingenen, R., Zeng, G., Atherton, C., Bergmann, D., Bey, I., Carmichael, G., Collins, W.J., Duncan, B.N., Faluvegi, G., Folberth, G., Gauss, M., Gong, S., Hauglustaine, D., Holloway, T., Isaksen, I.S.A., Jacob, D.J., Jonson, J.E., Kaminski, J.W., Keating, T.J., Lupu, A., Marmer, E., Montanaro, V., Park, R.J., Pitari, G., Pringle, K.J., Pyle, J.A., Schroeder, S., Vivanco, M.G., Wind, P., Wojcik, G., Wu, S., Zuber, A. (2009) Multimodel estimates of intercontinental source-receptor re-

lationships for ozone pollution. Journal of Geophysical Research 114, D04301, doi:10.1029/2008JD010816.

Hasegawa, S., Kobayashi, S., Ohara, T., Tanabe, K., Hayami, H., Yomemochi, S., Umezawa, N., Iijima, A., Kumagai, K. (2008) Fine Aerosol Measurement and Modeling in Kanto Area (1), Overview of Measurement. Proceedings of the 49th Annual Meeting of the Japan Society for Atmospheric Environment, 377 (in Japanese).

Hass, H., Builtjes, P.J.H., Simpson, D., Stern, R. (1997) Comparison of model results obtained with several European regional air quality models. Atmospheric Environment 31, 3259-3279.

Kondo, Y., Takegawa, N., Miyakawa, T., Koike, M., Miyazaki, Y., Kanaya, Y., Mochida, M., Kuwata, M., Morino, Y., Shiraiwa, M. (2010) Formation and transport of aerosols in Tokyo in relation to their physical and chemical properties: a review. Journal of the Meteorological Society of Japan 88, 597-624.

Minoura, H., Takahashi, K., Chow, J.C., Watson, J.G. (2006) Multi-year trend in fine and coarse particle mass, carbon, and ions in downtown Tokyo, Japan. Atmospheric Environment 40, 2478-2487.

Morino, Y., Chatani, S., Hayami, H., Sasaki, K., Mori, Y., Morikawa, T., Ohara, T., Hasegawa, S., Kobayashi, S. (2010) Intercomparison of Chemical Transport Model for Evaluation of Model Performance on O_3 and $PM_{2.5}$ Prediction-Case Study on Kanto Area in Summer 2007. Journal of Japan Society for Atmospheric Environment 45, 212-226 (in Japanese with English abstract).

Ohara, T., Yamaji, K., Uno, I., Tanimoto, H., Sugata, S., Nagashima, T., Kurokawa, J., Horii, N., Akimoto, H. (2008) Longterm simulations of surface ozone in East Asia during 1980-2020 with CMAQ and REAS. In NATO Science for peace and security series-C: Environmental Security, Air Pollution Modeling and its Application XIX (Borrego, C. and Miranda, A.I., Eds), Springer, Netherlands, pp. 136-144.

Skamarock, W.C., Klemp, J.B., Dudhia, J., Gill, D.O., Barker, D.M., Wang, W., Powers, J.G. (2005) A description of the Advanced Research WRF Version 2. NCAR Technical note, NCAR/TN-468+STR, Boulder, 99p.

Turpin, B.J., Lim, H.J. (2001) Species contributions to $PM_{2.5}$ mass concentrations: Revisiting common assumptions for estimating organic mass. Aerosol Science and Technology 35, 602-610.

US Environmental Protection Agency (2007) Guidance on the Use of Models and Other Analyses for Demonstrating Attainment of Air Quality Goals for Ozone, $PM_{2.5}$, and Regional Haze, Rep. USA EPA-454/B-07-002, US Environmental Protection Agency, Washington, DC.

van Loon, M., Vautard, R., Schaap, M., Bergstrom, R., Bessagnet, B., Brandt, J., Builtjes, P.J.H., Christensen, J.H., Cuvelier, C., Graff, A., Jonson, J.E., Krol, M., Langner, J., Roberts, P., Rouil, L., Stern, R., Tarrason, L., Thunis, P., Vignati, E., White, L., Wind, P. (2007) Evaluation of long-term ozone simulations from seven regional air quality models and their ensemble. Atmospheric Environment 41, 2083-2097.

Vautard, R., Builtjes, P.H.J., Thunis, P., Cuvelier, C., Bedogni, M., Bessagnet, B., Honore, C., Moussiopoulos, N., Pirovano, G., Schaap, M., Stern, R., Tarrason, L., Wind, P. (2007) Evaluation and intercomparison of ozone and PM10 simulations by several chemistry transport models over 4 European cities within the CityDelta project. Atmospheric Environment 41, 173-188.

Volkamer, R., Jimenez, J.L., San Martini, F., Dzepina, K., Zhang, Q., Salcedo, D., Molina, L.T., Worsnop, D.R., Molina, M.J. (2006) Secondary organic aerosol formation from anthropogenic air pollution: Rapid and higher than expected. Geophysical Research Letter 33, L17811, doi:10.1029/2006GL026899.

Wakamatsu, S., Uno, I., Ohara, T., Schere, K.L. (1999) A study of the relationship between photochemical ozone and its precursor emissions of nitrogen oxides and hydrocarbons in Tokyo and surrounding areas, Atmospheric Environment 19, 3097-3108.

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