

Evaluation of Ensemble Approach for O₃ 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 O₃ 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 O₃ ($r=0.65-0.85$) and the daily maximum O₃ 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 ($r\sim 0.2$). The ensemble average improved the simulation of SO₄²⁻, NO₃⁻, and NH₄⁺, whose production pathways are well known. In particular, the ensemble approach effectively simulated NO₃⁻, 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 O₃ 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 O₃ concentrations better than the CTMs do individually. The ensemble approach has been applied to the evaluation of source-receptor relationships of O₃ 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
$\Delta x, \Delta y$ (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 O₃ 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 O₃ 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 ($\sim 150 \times 150$ km²) 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

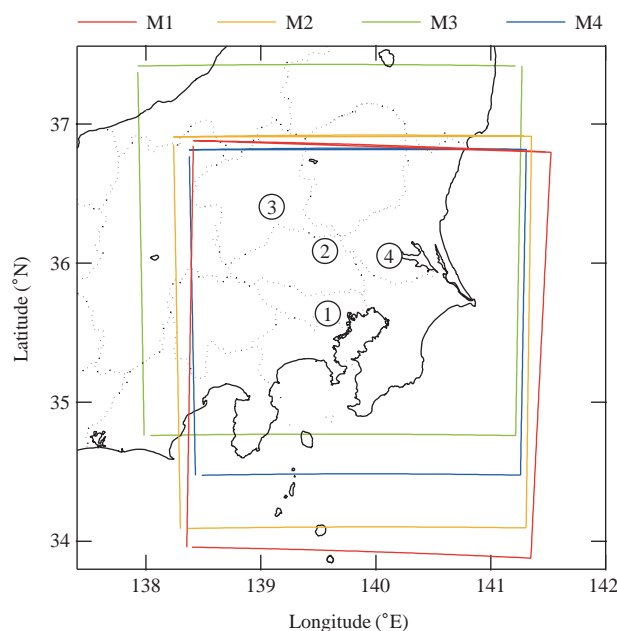


Fig. 1. Smallest domains of the individual CTMs (colored lines) and the measurement sites (numbered circles: 1, Komae; 2, Kisai; 3, Maebashi; 4, Tsukuba).

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 O₃ 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, SO₄²⁻, NO₃⁻, and NH₄⁺; $\Sigma(\text{PM}_{2.5})$). 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 NO₃⁻ and NH₄⁺ at Tsukuba were not available (Morino *et al.*, 2010),

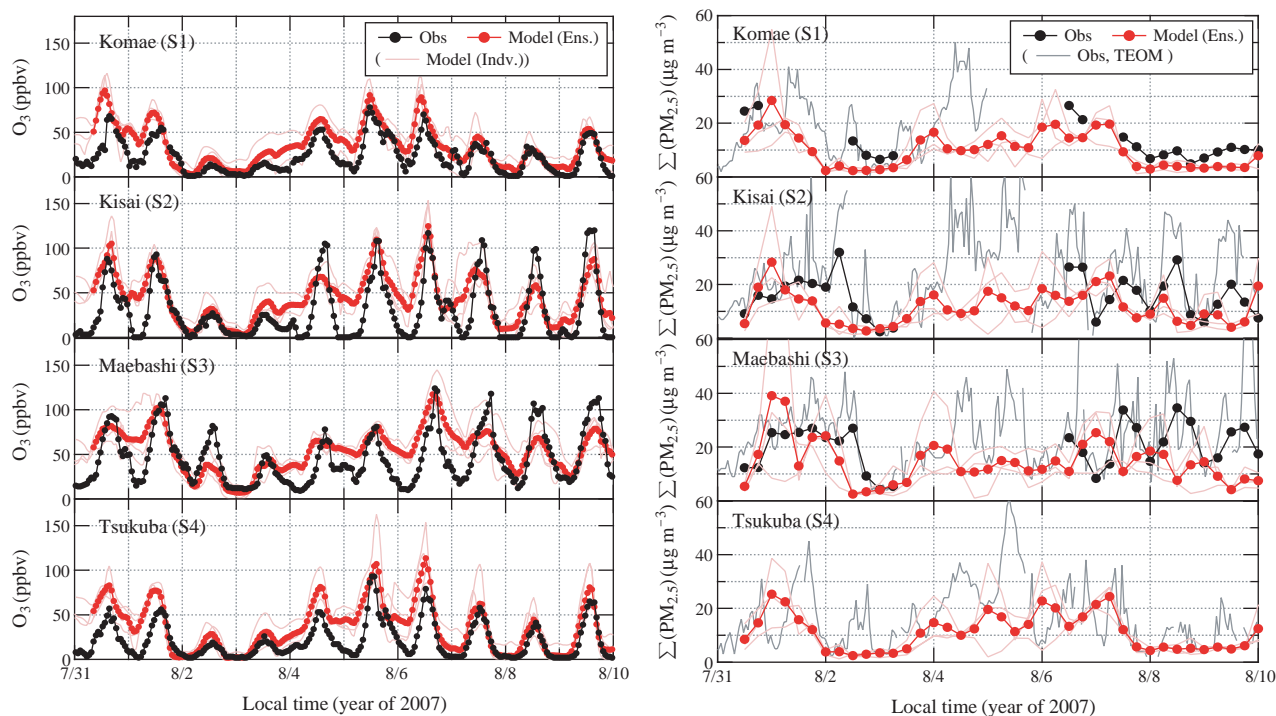


Fig. 2. Time series of O_3 (left) and $\Sigma(PM_{2.5})$ (right) concentrations from 31 July to 9 August 2007. Black circles indicate measurement results, and red circles indicate the ensemble average of the four CTMs. Pink lines indicate the individual CTM results. $PM_{2.5}$ concentrations measured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

we analyzed $\Sigma(PM_{2.5})$ 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 inter-diurnal variation in the O_3 concentration at Komae, though they sometimes overestimated the O_3 concen-

tration 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 inter-diurnal 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

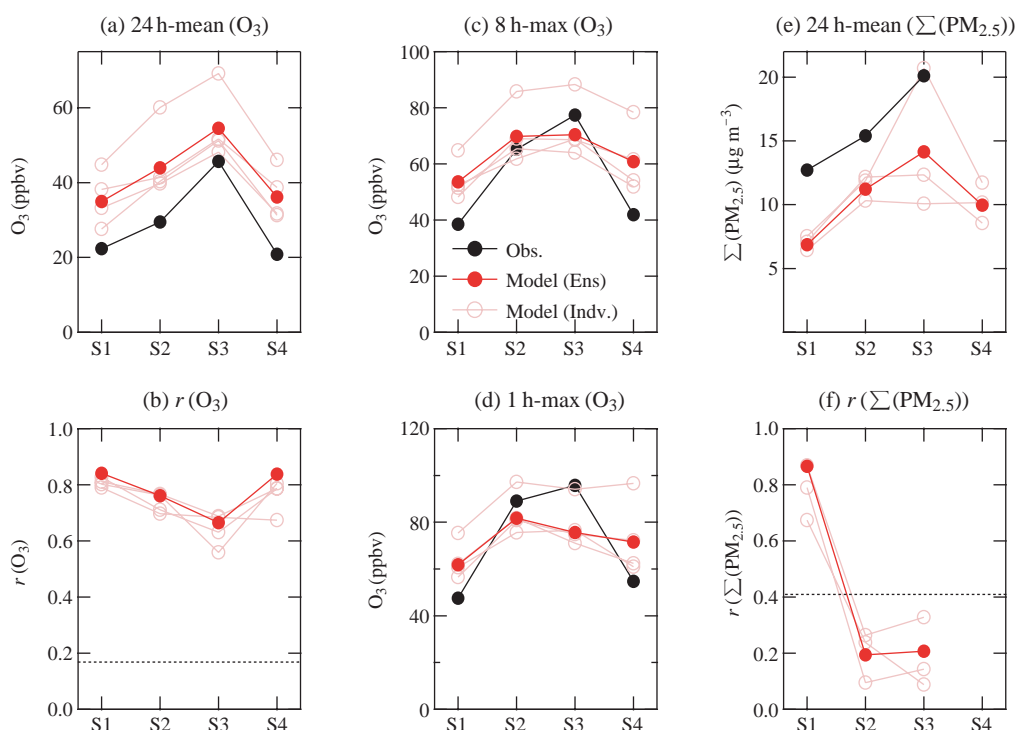


Fig. 3. Average model performance at the four measurement sites (S1, Komae; S2, Kisai; S3, Maebashi; S4, Tsukuba) during the analytical period (from 31 July 2007 to 10 August 2007). Fig. 3(a) and 3(e) show the average concentrations of O₃ and $\Sigma(\text{PM}_{2.5})$, respectively. Fig. 3(b) and 3(f) 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 O₃ 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 O₃ concentrations (Fig. 3(a)). This overestimation can be attributed to a poor model representation of O₃ titration by NO during nighttime (Morino *et al.*, 2010). For the evaluation of the model performance for daytime O₃ concentrations, we also compared daily 8- and 1-hour maximum O₃ 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 O₃ concentrations, whereas air quality standards in the United States have been established for daily 8-hour maximum O₃ 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 O₃ 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 O₃ concentrations better than the individual CTMs, it reduced the effect of outlying data (the large overestimation of

O₃ by one CTM in this case) and effectively increased the reliability of the O₃ simulation by the CTMs.

$\Sigma(\text{PM}_{2.5})$ represented 60-80% of the total PM_{2.5} concentration measured by tapered element oscillating microbalance (TEOM) (Fig. 2). The diurnal variation of $\Sigma(\text{PM}_{2.5})$ was smaller at Komae than at the other two sites where $\Sigma(\text{PM}_{2.5})$ was analyzed, whereas the inter-diurnal variation of $\Sigma(\text{PM}_{2.5})$ at Komae was roughly reproduced by all three CTMs. Observed $\Sigma(\text{PM}_{2.5})$ 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 $\Sigma(\text{PM}_{2.5})$ concentration, and, consequently, the ensemble average underestimated the average $\Sigma(\text{PM}_{2.5})$ concentration by a factor of 1.4-2 (Fig. 3(e)). $\Sigma(\text{PM}_{2.5})$ was highest at Maebashi, and this behavior was reproduced by most CTMs. The correlation coefficient between observations and the ensemble average of $\Sigma(\text{PM}_{2.5})$

Table 2. Model:observation ratios of the ensemble average (Ens.) and the four individual CTMs (M1-M4) for $\Sigma(\text{PM}_{2.5})$, 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)
$\Sigma(\text{PM}_{2.5})$	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 $\mu\text{g}/\text{m}^3$	15.40 $\mu\text{g}/\text{m}^3$	20.10 $\mu\text{g}/\text{m}^3$	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 $\mu\text{g}/\text{m}^3$	2.22 $\mu\text{g}/\text{m}^3$	2.45 $\mu\text{g}/\text{m}^3$	1.57 $\mu\text{g}/\text{m}^3$
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 $\mu\text{g}/\text{m}^3$	5.85 $\mu\text{g}/\text{m}^3$	6.94 $\mu\text{g}/\text{m}^3$	4.30 $\mu\text{g}/\text{m}^3$
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 $\mu\text{g}/\text{m}^3$	3.58 $\mu\text{g}/\text{m}^3$	4.16 $\mu\text{g}/\text{m}^3$	5.37 $\mu\text{g}/\text{m}^3$
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 $\mu\text{g}/\text{m}^3$	1.24 $\mu\text{g}/\text{m}^3$	3.07 $\mu\text{g}/\text{m}^3$	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 $\mu\text{g}/\text{m}^3$	1.94 $\mu\text{g}/\text{m}^3$	2.90 $\mu\text{g}/\text{m}^3$	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 $\Sigma(\text{PM}_{2.5})$ at those sites.

Model performance for $\text{PM}_{2.5}$ 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 $\Sigma(\text{PM}_{2.5})$ was large (36-39%); thus, underestimation of the OA concentration caused the poor model performance for $\text{PM}_{2.5}$, 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 pro-

duction pathways of secondary organic aerosols (SOAs) are mostly unknown. Improvement of the SOA simulation model is necessary to improve model performance for $\text{PM}_{2.5}$ simulation. The contribution of SO_4^{2-} to $\Sigma(\text{PM}_{2.5})$ 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

NO₃⁻ are inhomogeneous (e.g., Eder and Yu, 2006). The ensemble average reproduced the NO₃⁻ concentration within a factor of 2 at all three sites where it was measured, whereas none of the four CTMs individually reproduced the NO₃⁻ concentration within a factor of 2 at every site. These results suggest that the ensemble approach could effectively simulate NO₃⁻. The model performance for NH₄⁺ was roughly determined by the model performance for SO₄²⁻ and NO₃⁻ simulation (not shown).

In this study, the ensemble approach improved the model performance for SO₄²⁻ and NO₃⁻ simulation. This result suggests that current CTMs capture the factors controlling SO₄²⁻ and NO₃⁻ concentrations relatively well. The ensemble approach effectively reduced the uncertainties in CTM results for SO₄²⁻ and NO₃⁻, 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 O₃ 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 O₃ ($r=0.65-0.85$) than the individual CTMs. Also, the ensemble average could effectively remove the effect of outlying data and reproduced the daily maximum O₃ concentration within 31% at all four sites. By contrast, the ensemble average did not reproduce the $\sum(\text{PM}_{2.5})$ temporal variation at two suburban sites ($r\sim 0.2$), and underestimated $\sum(\text{PM}_{2.5})$ by a factor of 1.4-2. The ensemble average improved the model performance for SO₄²⁻ and NO₃⁻, whose production pathways are relatively well known, suggesting that the ensemble approach is suitable for SO₄²⁻ and NO₃⁻ 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 improv-

ed 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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