16th International Conference on
Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes
8-11 September 2014, Varna, Bulgaria

# EVALUATION OF AIR QUALITY MODEL PERFORMANCE FOR LONG-TERM $PM_{2.5}$ SIMULATION IN JAPAN

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**Abstract**: In order to evaluate the performance of air quality models for long-term simulations, the Community Multiscale Air Quality model (CMAQ) version 5.0.1 and the Comprehensive Air quality Model with extensions (CAMx) version 6.00 were driven with the Weather Research and Forecasting model (WRF) version 3.5.1 from April 2010 to March 2011 in the Kinki region of Japan. The two air quality models used common input meteorological fields, emissions and boundary concentrations considering the effect of long-range transport from the Asian continent. Although CAMx-simulated surface concentrations of air pollutants tended to be higher than CMAQ-simulated values except for $O_3$, which is strongly affected by titration with $NO_X$, temporal variation patterns simulated by the two models were quite similar to each other. As a result, statistical comparisons indicated that the overall long-term performances of CMAQ and CAMx were also similar to each other. The both model approximately captured the total $PM_{2.5}$ mass concentrations except for underestimates in summer. However, the models clearly underestimated OA, which was compensated by overestimates of dust transported from the Asian continent and anthropogenic unspeciated $PM_{2.5}$. Although CMAQ and CAMx similarly well simulated long-term day-to-day variations of $PM_{2.5}$ concentrations, they need to be revised for better representation of individual $PM_{2.5}$ components.

***Key words:*** *Fine particulate matter, Model performance, CMAQ, CAMx, Annual simulation*

## 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, nitrate, ammonium, elemental carbon (EC) and organic aerosol (OA). $PM_{2.5}$ has been of increasing concern because of its adverse effects on human health. 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 attained only at about 30% of ambient air quality monitoring stations. 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, current AQMs cannot adequately simulate $PM_{2.5}$ concentrations in Japan.

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., 2014a). In UMICS, the major components of $PM_{2.5}$ in the Greater Tokyo Area are focused; common datasets, including meteorological, emission and boundary data, are provided to participating models; participants conduct sensitivity runs in their fields of expertise. It is important to understand long-term performance of AQMs because the $PM_{2.5}$ AQS is evaluated on the basis of a single year. However, such performance was not evaluated in UMICS. In addition, almost all of AQMs participating in UMICS were the Community Multiscale Air Quality model (CMAQ) (Byun and Ching, 1999) with different configurations. Therefore, UMICS is an intra-comparison study for CMAQ rather than an inter-comparison study for AQMs.

This study conducted one-year air quality simulations in the Kinki region of Japan. In addition to CMAQ, the Comprehensive Air quality Model with extensions (CAMx) (ENVIRON, 2013) was used for the simulations. The CMAQ and CAMx performances for long-term $PM_{2.5}$ and other air pollutants were evaluated and difference and similarity between the two models were discussed.

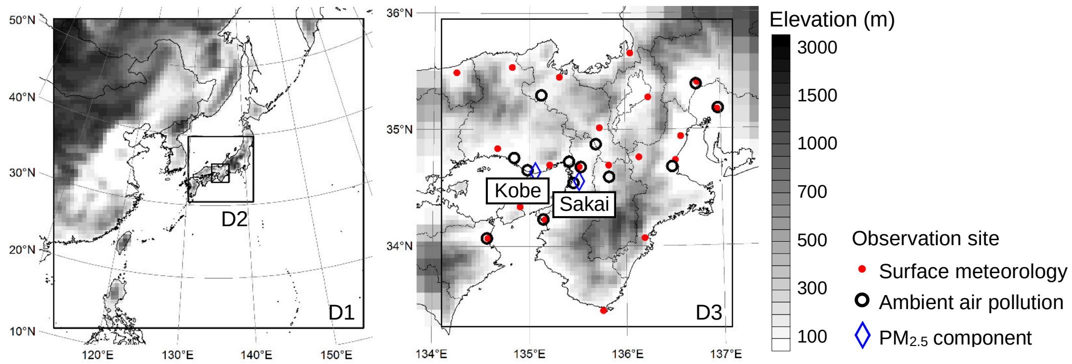


**Figure 1.** Modeling domains for air quality simulations and locations of observation sites

## METHODOLOGY

This study utilized the Weather Research and Forecasting model (WRF) (Skamarock et al., 2009) version 3.5.1 to produce meteorological fields, and CMAQ version 5.0.1 and CAMx version 6.00 for air quality simulations. The numerical models were run for April 2010 to March 2011 (Japanese fiscal year 2010: JFY2010) with an initial spin-up period of 22-31 March 2010. Figure 1 shows modeling domains for air quality simulations and locations of observation sites used for model evaluations. The horizontal domains consist of three domains: domain 1 (D1) covering a wide area of Northeast Asia, domain 2 (D2) covering the main land of Japan, domain 3 (D3) covering of the Kinki region, in which there are some megacities such as Osaka, Kyoto and Kobe. The horizontal resolutions and the number of grid cells are 64, 16 and 4 km, and 76 × 76, 64 × 64 and 68 × 72 for D1, D2 and D3, respectively. The vertical layers consist of 30 sigma-pressure coordinated layers from the surface to 100 hPa with the middle height of the first layer being approximately 28 m. The WRF performance was evaluated with observation data at meteorological observatories in D3 by the Japan Meteorological Agency (JMA). The CMAQ and CAMx performances were evaluated with concentration data observed at ambient air pollution monitoring stations conducting $PM_{2.5}$ observations in JFY2010 and national monitoring stations in D3, which were derived from the Environmental Numerical Databases by the National Institute for Environmental Studies of Japan. The performances for major $PM_{2.5}$ components were evaluated with concentration data obtained from 24-h filter sampling at Sakai and Kobe sites by MOE.

Meteorological fields were produced using WRF configured with the same physics options as those used by Shimadera et al. (2014b). Sea surface temperature was derived from the high-resolution, real time, global analysis data developed at the U.S. National Centers for Environmental Prediction (NCEP). Initial and lateral boundary conditions for WRF were derived from the mesoscale model grid point value data by JMA and the final analysis data by NCEP. Grid nudging using these analysis data was applied to horizontal wind components, temperature and humidity in D1 and D2 with nudging coefficient of $3.0 \times 10^{-4}$ $s^{-1}$ and horizontal wind components in D3 with nudging coefficient of $7.5 \times 10^{-5}$ $s^{-1}$ for the entire simulation period. The WRF simulation was conducted with on-line one-way nesting in the three domains.

Emission data for the air quality simulations were produced in a similar way to Shimadera et al. (2014b) with the following differences. Anthropogenic emissions in Japan other than from vehicles were derived from EAGrid2000-JAPAN. Ship emissions were derived from an emission inventory developed by the Ocean Policy Research Foundation. Emissions from open biomass burning were derived from the fire inventory from the U.S. National Center for Atmospheric Research version 1.0. Initial and boundary concentrations for D1 were obtained from the Model for Ozone and Related Chemical Tracers version 4.

Table 1 summarizes CMAQ and CAMx configurations. The CMAQ simulation was conducted with off-line one-way nesting in the three domains. The CAMx simulation in D3 was conducted with boundary concentrations derived from results of the CMAQ simulation in D2. Fine particles are represented by two lognormal distributions called the Aitken and accumulation modes in CMAQ and by a static fine mode in CAMx. The total mass of particles except water in the two modes in CMAQ and that in the fine mode in CAMx were used as approximations of $PM_{2.5}$. The total mass except water in all modes was used as approximations of suspended particulate matter (SPM; approximately equivalent to $PM_7$).

**Table 1.** Configurations of CMAQ and CAMx

| | CMAQ v5.0.1 | CAMx v6.00 |
|---|---|---|
| Meteorology-chemistry interface | MCIP v4.1 | WRFCAMx v4.0 |
| Domain | D1, D2, D3 | D3 |
| Horizontal/vertical advection | Yamartino/ WRF-based scheme | PPM/PPM |
| Horizontal/vertical diffusion | Multiscale/ ACM2 | Smagorinsky/ACM2 |
| Photolysis rate | on-line photolysis method | TUV v4.8 |
| Gas phase chemistry (solver) | SAPRC99 (EBI) | SAPRC99 (EBI) |
| Aerosol process (size distribution) | AERO5 (two fine/one coarse modes) | CF (static fine/coarse modes) |
| SIA partitioning | ISORROPIA | ISORROPIA |
| SOA partitioning | SORGAM | SOAP |
| Aqueous process | RADM/ACM convective cloud | RADM/Seinfeld and Pandis |
| Dry deposition | M3Dry Pleim model | Wesely |

## RESULTS AND DISCUSSION

### Model performance for meteorology

Figure 2 shows observed and WRF-simulated monthly meteorological variables at the meteorological observatories in D3 in JFY2010. For temperature, WRF-simulated values fairly well agreed with observed values, including diurnal and day-to-day variations, at every meteorological observatory in D3. For wind speed, WRF well simulated day-to-day variation patterns, but tended to overestimate the strength. Because the overestimate was remarkable for strong wind at observatories along coastline or in small basin, the model may underestimate the effect of surface drag in such regions. For precipitation, WRF approximately captured seasonal and spatial variations except that the model tended to underestimate the amount in rainy season caused by a persistent stationary front over the study region in June to early July. Overall, the results indicate that the meteorological fields produced by WRF generally captured synoptic weather patterns that control $PM_{2.5}$ behaviours in the atmosphere.

### Model performance for ambient air pollution

Table 2 summarizes statistical values for the CMAQ and CAMx performances of daily concentrations of $O_3$, $SO_2$, $NO_2$, NO, CO, non-methane hydrocarbon (NMHC), SPM and $PM_{2.5}$ at the ambient air pollution monitoring stations in D3. The statistical measures include the Pearson's correlation coefficient (*r*), the mean absolute error (MAE), the root mean square error (RMSE), and the index of agreement (IA). The *r* values for $O_3$, $NO_2$, CO and $PM_{2.5}$ were relatively high in the both models, indicating that the models well simulated temporal and spatial variation patterns of these pollutants. The CAMx-simulated surface concentrations were generally higher than the CMAQ-simulated values except for $O_3$, which is strongly affected by titration with $NO_X$. As a result, the CAMx-simulated mean concentrations of the pollutants except for $SO_2$ were closer to the observed values than the CMAQ-simulated values while the both models clearly underestimated CO and NMHC. The minimum vertical eddy diffusivity in CMAQ that is higher in urban areas than other areas is partly responsible for the difference between the results of the two models. In spite of the differences between the two models, their overall performances were similar.

Figure 3 shows observed, CMAQ- and CAMx-simulated monthly concentrations at the ambient air pollution monitoring stations in D3 in JFY2010. The two model simulated quite similar seasonal variation patterns. While seasonal variation patterns of $NO_2$ and CO were well simulated, overestimates of $O_3$ in summer and $SO_2$ in winter, consistent underestimate of CO, and underestimates of NMHC and SPM in summer caused discrepancies between the observed and simulated mean concentrations. The $O_3$ overestimate and NMHC underestimate may indicate overestimate of photochemical activity in summer.

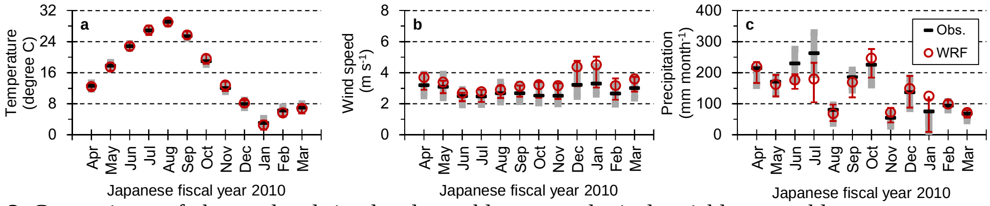


**Figure 2.** Comparisons of observed and simulated monthly meteorological variables: monthly mean temperature (a) and wind speed (b), and monthly precipitation (c). Mean value of all the meteorological observatories in D3 and range of 25-75th percentile rank of monthly values of individual observatories are provided

**Table 2.** Statistical comparisons between observed and simulated daily concentrations at ambient air pollution monitoring stations in D3 in Japanese fiscal year 2010

| | | **$O_3$ (ppb)** | **$SO_2$ (ppb)** | **$NO_2$ (ppb)** | **NO (ppb)** | **CO (ppb)** | **NMHC (ppbC)** | **SPM (μg $m^{-3}$)** | **$PM_{2.5}$ (μg $m^{-3}$)** |
|---|---|---|---|---|---|---|---|---|---|
| | *n* | 4306 | 4314 | 4297 | 4297 | 1454 | 2819 | 4648 | 3820 |
| Obs. | Mean | 43.7 | 2.9 | 14.4 | 4.2 | 369 | 159 | 21.8 | 16.4 |
| CMAQ | Mean | 51.2 | 3.5 | 12.0 | 2.8 | 220 | 96 | 17.2 | 13.5 |
| | *r* | 0.77 | 0.44 | 0.82 | 0.59 | 0.79 | 0.60 | 0.60 | 0.76 |
| | MB | 7.5 | 0.6 | -2.3 | -1.4 | -149 | -64 | -4.5 | -2.9 |
| | RMSE | 13.4 | 3.0 | 5.6 | 5.0 | 169 | 106 | 11.8 | 7.6 |
| | IA | 0.83 | 0.64 | 0.88 | 0.74 | 0.63 | 0.70 | 0.75 | 0.85 |
| CAMx | Mean | 49.2 | 3.7 | 13.2 | 4.1 | 248 | 118 | 19.2 | 16.4 |
| | *r* | 0.74 | 0.43 | 0.83 | 0.56 | 0.75 | 0.58 | 0.57 | 0.76 |
| | MB | 5.5 | 0.8 | -1.2 | -0.1 | -120 | -41 | -2.6 | 0.0 |
| | RMSE | 12.6 | 3.0 | 5.4 | 7.5 | 149 | 122 | 11.9 | 7.7 |
| | IA | 0.83 | 0.63 | 0.89 | 0.66 | 0.72 | 0.69 | 0.75 | 0.87 |

Note: Daily concentrations are daily mean values except for $O_3$ being daily maximum 8-h value.

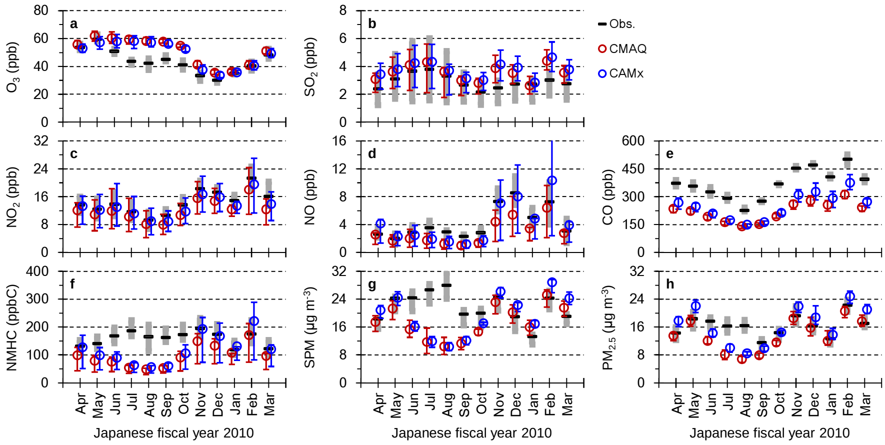


**Figure 3.** Comparisons of observed and simulated monthly concentrations of air pollutants: monthly mean daily maximum 8-h concentration of $O_3$ (a), and monthly mean concentrations of $SO_2$ (b), $NO_2$ (c), NO (d), CO (e), NMHC (f), SPM (g) and $PM_{2.5}$ (h). Mean value of all the ambient air pollution monitoring stations in D3 and range of 25-75th percentile rank of monthly values of individual stations are provided

## Model performance for $PM_{2.5}$

Figure 4 shows observed and simulated seasonal mean concentrations of $PM_{2.5}$ components at Sakai and Kobe sites in JFY2010. While the CAMx-simulated values were higher by 10-20% than the CMAQ-simulated values, component ratios by the two models were quite similar to each other at the both sites. The models approximately reproduced the total $PM_{2.5}$ mass concentrations except for underestimates in summer. However, the models clearly and consistently underestimated OA, which was compensated by overestimates of components other than the five major components. The other $PM_{2.5}$ is dominated by dust transported from the Asian continent and anthropogenic primary unspeciated $PM_{2.5}$ emissions. Therefore, the contribution of dust from the continent was possibly overestimated. In addition to underestimates of secondary OA productions, uncertainties in speciation profiles of $PM_{2.5}$ emissions may be partly responsible for the OA underestimates. The models tended to overestimate nitrate, which may be associated with an artefact in the observed data because of the volatilisation. The model tended to underestimate sulfate, particularly in winter. This underestimate and the overestimate of $SO_2$ in winter may indicate that the models underestimate the oxidation of $SO_2$ under cold condition.

Figure 5 shows observed and simulated daily mean $PM_{2.5}$ concentrations at ambient air pollution monitoring stations neighboring Sakai and Kobe sites. The difference in *r* values between the two sites is mainly due to lack of observation data in winter at the station neighboring Kobe rather than difference of model performance at the two sites. Overall, CMAQ and CAMx similarly well simulated long-term day-to-day variations of $PM_{2.5}$ concentrations.

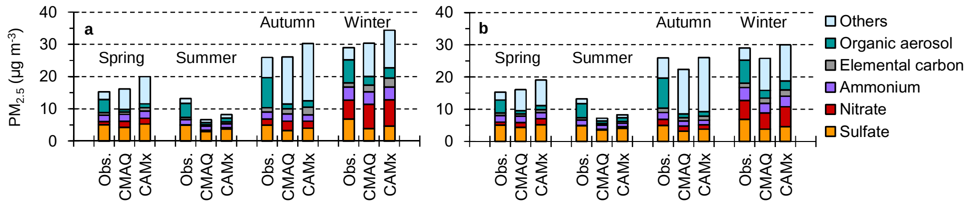


**Figure 4.** Comparisons of observed and simulated seasonal mean concentrations of major $PM_{2.5}$ components at Sakai (a) and Kobe (b) sites. Spring: 14-27 May 2010, Summer: 26 July-11 August 2010, Autumn: 5-18 November 2010, Winter: 26 January-10 February 2011

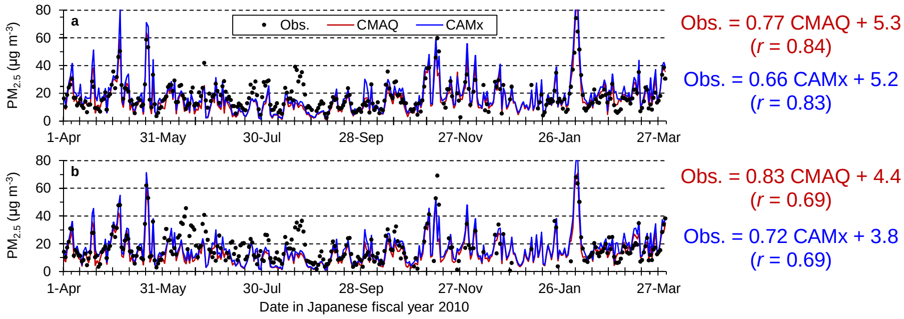


**Figure 5.** Time series comparisons of observed and simulated daily mean $PM_{2.5}$ concentrations at ambient air pollution monitoring stations neighboring Sakai (a) and Kobe (b) sites

## CONCLUSION

In order to evaluate the performance of CMAQ and CAMx for long-term simulations, the two models were driven in JFY2010 with common input meteorological fields, emissions and boundary concentrations considering the effect of long-range transport. Although CAMx-simulated surface concentrations of air pollutants tended to be higher than CMAQ-simulated values except for $O_3$, which is strongly affected by titration with $NO_X$, the overall long-term performances of the two models were similar to each other. While the both model approximately captured the total $PM_{2.5}$ mass concentrations, the simulated component ratios did not agreed with the observations. This is due to moderate underestimate of sulfate, moderate overestimate of nitrate, substantial underestimate of OA and substantial overestimate of components other than the major components. Overall, although CMAQ and CAMx similarly well simulated long-term day-to-day variations of $PM_{2.5}$ concentrations, they need to be revised for better representation of individual $PM_{2.5}$ components.

## ACKNOWLEDGEMENTS


This research was supported by JSPS KAKENHI Grant Number 26740038.


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