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# Comparison between Atmospheric Chemistry Model and Observations Utilizing the RAQMS-CMAQ Linkage, Part II : Impact on PM2.5 Mass Concentrations Simulated

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**ABSTRACT**

In the companion paper (Lee *et al.*, 2012), it was showed that CMAQ simulation using a lateral boundary conditions (LBCs) derived from RAQMS-CMAQ linkage, compared to the CMAQ results with the default CMAQ LBCs, improved ozone simulations in the conterminous US domain. In the present paper, the study is extended to investigate the influence of LBCs on PM2.5 simulation. MM5-SMOKE-CMAQ modeling system was used for meteorological field generation, emissions preparation and air quality simulations, respectively. Realtime Air Quality Modeling System (RAQMS) model assimilated with satellite observations were used to generate the CMAQ-ready LBCs. CMAQ PM2.5 simulations with RAQMS LBCs and predefined LBCs were compared with U.S. EPA Air Quality System (AQS) measurements. Mean PM2.5 lateral boundary conditions taken from RAQMS outputs showed strong variations both in the horizontal grid and vertical layers in the northern and western boundaries and affected the results of CMAQ PM2.5 predictions. CMAQ with RAQMS LBCs could improve CMAQ PM2.5 predictions resulting in the improvement of index of agreement from 0.38 to 0.63.

**Key words:** CMAQ PM2.5 simulations, RAQMS-CMAQ linkage, Lateral boundary conditions, CMAQ, RAQMS

## 1. INTRODUCTION

In order to understand the complex physical and chemical processes in the atmosphere, a number of urban and regional scale air quality models have been developed and applied to address various air quality issues related to aerosols, regional haze, ozone and so on (Russell and Dennis, 2000; Seigneur *et al.*, 1999). The Community Multiscale Air Quality (CMAQ) model (Byun and Schere, 2006; Byun and Ching, 1999) is a state-of-the-art science atmospheric chemistry model extensively used for studying various air quality issues. The CMAQ contains numerous algorithms of physical and chemical processes to understand atmospheric trace gases and aerosol transformations. Despite this, predicting aerosol concentration is more difficult than predicting trace gas (for example, ozone) concentrations (Smyth *et al.*, 2006). This is partially due to the complexity and the limited knowledge on aerosol compositions, structure, phases, and formation mechanisms in the atmosphere. Some of the difficulties in the aerosol predictions are caused by the lack of episodic emissions inputs such as forest fires and lack of dynamic boundary conditions depicting the effects of long-range transport events (In *et al.*, 2007). Since the current CMAQ model utilizes fixed lateral boundary condition profiles of the pollutant species, they could not consider spatial and temporal changes at the boundaries. It is, therefore, essential to create and provide appropriate boundary conditions data for CMAQ simulations.

The key hypothesis of this study is that such limitations can be improved by the utilization of the NASA's substantial archives of earth science remote sensing and modeling data products. The Realtime Air Quality Modeling System (RAQMS) (Pierce *et al.*, 2009; Pierce *et al.*, 2007; Pierce *et al.*, 2003) model with satellite observations assimilated can provide such dynamic lateral boundary conditions (LBCs) for CMAQ. In the companion paper (Lee *et al.*, 2012), we showed that CMAQ simulation using a LBCs derived from RAQMS-CMAQ linkage, compared to the CMAQ results with the default CMAQ LBCs, improved ozone simulations in the conterminous US domain. In this paper, we investigated the impacts of the LBCs on CMAQ PM2.5 simulations and characterized the model results by comparing with measurement data. We discussed the improvement of CMAQ PM2.5 simulation by using the improved dynamic boundary conditions taken from RAQMS results.

## 2. METHODOLOGY

### 2. 1 MM5-SMOKE-CMAQ Modeling System

The air quality modeling system used in this study is the United States (U.S.) Environmental Protection Agency (EPA)'s Community Multiscale Air Quality (CMAQ) modeling system. The CMAQ version 4.6 was used to simulate regional air quality in this study. The key components of the CMAQ modeling system include: (1) A meteorological modeling system (for example, MM5 used in this study) for the description of meteorological phenomenon such as wind field, air temperature, cloudiness and so on; (2) Sparse Matrix Operating Kernel for Emissions (SMOKE) models for processing emissions that are injected into the atmosphere; and (3) CMAQ Chemistry Transport Modeling (CTM) system for the simulation of chemical transformations and fate of emissions.

Penn State-NCAR Fifth generation Mesoscale Model (MM5) (Grell *et al.*, 1994) is used for meteorological modeling to generate 2006 meteorological inputs to CMAQ. It is configured with multiscale domains: a $36\times36$ km grid domain ($157\times127$) which covers the Contiguous United States (CONUS) including southern Canada and northern Mexico and, a $12\times12$ km domain ($174\times135$), and a $4\times4$ km domain ($175\times136$). To investigate a LBCs impact on CMAQ PM2.5 results, only outputs from the coarse domain ($36\times36$ km grid) were utilized in this study. The initial and boundary conditions were provided by the National Centers for Environmental Prediction (NCEP) North American Mesoscale (NAM) model products. Physical options for the MM5 simulation include the Rapid Radiative Transfer Model (RRTM) radiation scheme, simple ice scheme for microphysics, Grell convection scheme, the Medium-Range Forecast (MRF) model boundary layer parameterization (Hong and Pan, 1996), and the Noah Land-surface model (LSM). To achieve better meteorological simulations, the assimilated MM5 inputs were utilized in this study for air quality simulations (detailed description of MM5 assimilation can be found in Ngan, 2008).

Since emissions determine the air pollutant concentration through chemical reactions, it is important to use high quality emissions inputs for air quality simulations. The 2002 U.S. EPA National Emission Inventory (NEI2002) was used to provide the anthropogenic emissions. The emissions inventories were processed using the SMOKE for providing emission inputs compatible with CMAQ.

### 2. 2 RAQMS-CMAQ Linking Tool

Boundary conditions are one of the most important inputs for the regional scale simulations of chemical transport models because they influence the temporal variations and spatial distributions of the pollutants. One of the most common methods to generate boundary condition inputs is utilization of global scale model outputs. A global chemistry model, the NOAA NESDIS/NASA LaRC/University of Wisconsin Real-time Air Quality Modeling System (RAQMS) (Pierce *et al.*, 2009, 2007, 2003), is used here to provide analyzed trace gas and aerosol boundary conditions to the CMAQ. The RAQMS analysis utilizes satellite trace gas measurements from the Ozone Monitoring Instrument (OMI), Tropospheric Emission Spectrometer (TES), and Microwave Limb Sounder on the NASA Aura satellite and satellite aerosol optical depth (AOD) measurements from the Moderate Resolution Imaging Spectroradiometer (MODIS) on the NASA Terra and Aqua satellites to constrain the analysis. It is run at the spatial resolution of 2 degrees longitude $\times2$ degrees latitude with 36 hybrid isentropic-sigma vertical layers in a standard global assimilation mode. RAQMS incorporate the Goddard Chemistry Aerosol Radiation and Transport (GOCART) aerosol modules (Chin *et al.*, 2003, 2002) which simulates major tropospheric aerosol components, including sulfate, dust, black carbon, organic carbon, and sea-salt aerosols. Chin *et al.* (2003) demonstrated that the GOCART aerosol modules, which are used in the RAQMS, can adequately simulate aerosols not only for the largescale intercontinental transport but also for the small-scale spatial and temporal variations. As mentioned, the RAQMS assimilates the MODIS AOD to adjust aerosol predictions and the benefits of the MODIS AOD assimilation are described in Kittaka *et al.* (2007).

RAQMS-CMAQ converter was updated from the previous version for this study. In the previous version (Ver. 1) (Song *et al.*, 2008), only Carbon Bond (CB4) gaseous species are taken into account to generate CMAQ-ready boundary conditions from RAQMS outputs. In the new version (Ver. 2), CMAQ aerosol modules (AE3 and AE4) are incorporated into the converter as well as additional gas phase species available from RAQMS. Several routines, such as an Meteorology-Chemistry Interface Processor (MCIP) data reading routine and input data error handling routines, were added and upgraded. Fig. 1 shows the schematic diagram of the RAQMS-CMAQ linking procedure. To match the grid system, units and species between the two models, three-dimensional (3-D) grid information, pressure, chemical, and aerosol concentration data are all extracted from RAQMS outputs and grid information, pressure, air density fields are also extracted from CMAQ. Nearest grid points between the two models are used for the horizontal grid point matching and

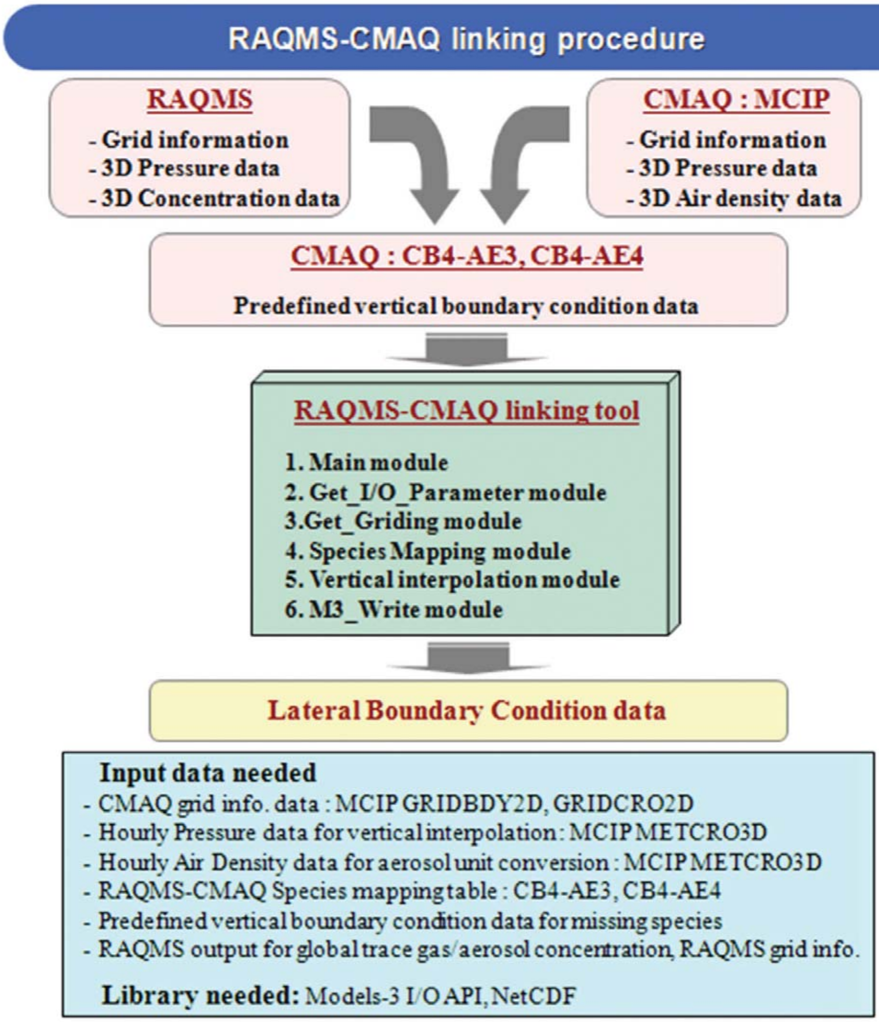


**Fig. 1.** Schematic diagram of RAQMS-CMAQ linking procedure.

the two model's 3-D pressure fields are utilized for the vertical interpolation of gas phase and aerosol species concentration. A species mapping table for the CB4-AE3 and CB4-AE4 mechanisms is used to convert RAQMS species into CMAQ species. The detailed RAQMS-CMAQ aerosol species mapping table can be found in the Appendix 3 of the companion paper (Lee *et al.*, 2012). For this study, hourly LBCs were prepared by RAQMS-CMAQ linking tool and applied during the CMAQ simulations.

## 3. RESULTS AND DISCUSSION

### 3. 1 Episode, Data and Analysis Regions

In order to demonstrate the boundary conditions' impacts on CMAQ PM2.5 predictions, MM5-SMOKE-CMAQ were run from August 30 to September 1 in 2006 with three different spatial resolutions (36, 12, and 4 km). The dynamic boundary conditions were provided by the 36 km resolution CONUS domain CMAQ simulations only from the RAQMS results. CMAQ was run for this period with two different lateral boundary conditions: (1) Predefined constant LBCs (simulation number denoted as 'cc_09'), and (2) LBCs from RAQMS outputs (simulation number denoted as 'cc_16').

U.S. EPA Air Quality System (AQS) 2006 ambient monitoring data were used for the comparison of CMAQ PM2.5 simulations. AQS PM2.5 data are available at http://epa.gov/ttn/airs/airsaqs/. For the purpose of model performance evaluation, CMAQ CONUS domain were evenly divided into three regions (western, central, and eastern region) as shown in Fig. 2.

### 3. 2 Lateral Boundary Conditions for PM2.5

Mean PM2.5 lateral boundary conditions taken from RAQMS outputs for the August 30-September 1, 2006 period are shown in Fig. 3. Strong variations both in the horizontal grid and vertical layers can be found in the northern and western boundaries. Maximum PM2.5 values of 50-60 μg/m$^3$ are present at the height of 2-4.5 km (14-17 layer) in the northern boundary, representing the aerosols transported from Canada possibly due to wild fires or so. High PM2.5 values of 10-40 μg/m$^3$ are found near the surface layers (1-10 layer, surface-800 m) in the northern and western boundaries.

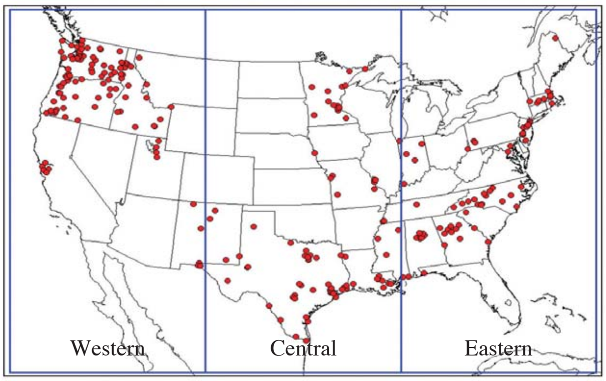


**Fig. 2.** CMAQ 36-km domain and three analysis regions (western, central, and eastern region). Red circles indicate EPA AQS monitoring stations.

Because the western boundary adjoins to the Pacific Ocean, it seems that the high values in the western boundary are associated with air mass recirculation. Aerosols from local emissions and transported from Canada go out and return back through the western boundary, resulting in high values in the western boundary. PM2.5 values in the southern and eastern boundaries are much lower than those in northern and western boundaries. Since CMAQ provide predefined PM2.5 boundary conditions for sulfate only and their values are less than 1 μg/m$^3$ (Byun and Ching, 1999), PM2.5 LBCs from RAQMS will affect the CMAQ PM2.5 predictions by adding aerosols through the boundaries.

### 3. 3 Comparisons between CMAQ with AQS Hourly PM2.5

The CMAQ predicted PM2.5 concentrations are compared with AQS hourly PM2.5 measurements as presented in Fig. 4. There are little changes (less than 1 μg/m$^3$) of PM2.5 concentration in the eastern region due to the addition of aerosols from the northeastern boundary. Slight changes (~1 μg/m$^3$) in PM2.5 concentration are found in the central region due to the increased aerosols from the northern boundary. As expected by PM2.5 boundary conditions in Fig. 3, PM2.5 concentration predicted with RAQMS LBCs are significantly enhanced in the western region. High PM2.5 boundary conditions from northern and western boundaries increased PM2.5 concentration by 2-4 μg/m$^3$ in the western region, which resulted in better agreement

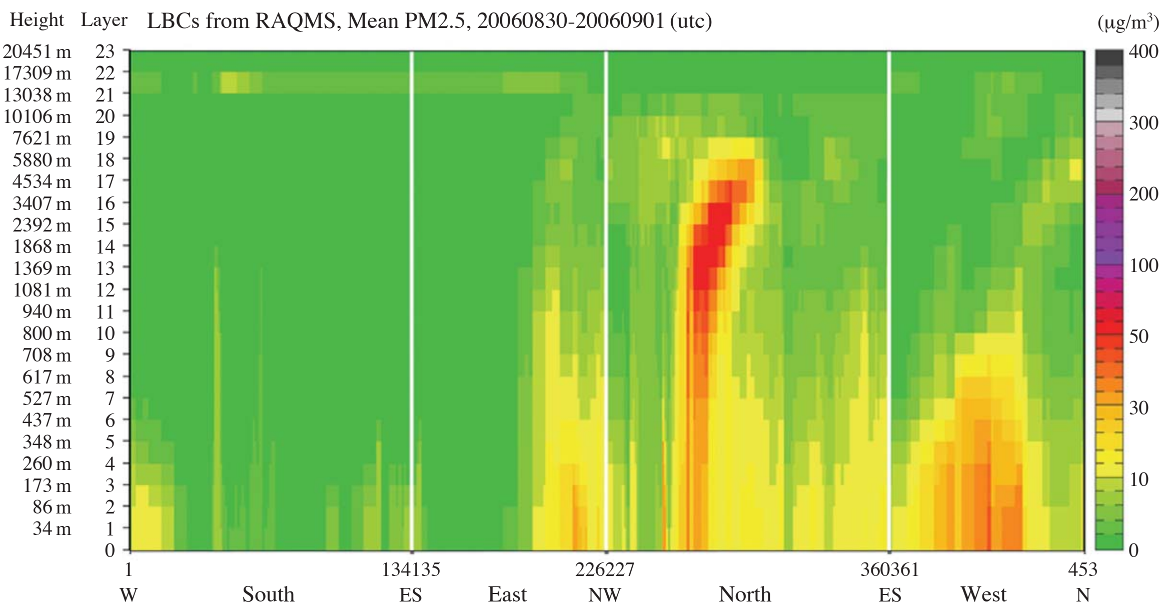


**Fig. 3.** Mean PM2.5 lateral boundary conditions along the boundaries of CMAQ 36 km domain. Mean LBCs values are generated from RAQMS outputs. The values are listed from west to east for southern and northern boundaries, and from south to north for eastern and western boundaries.

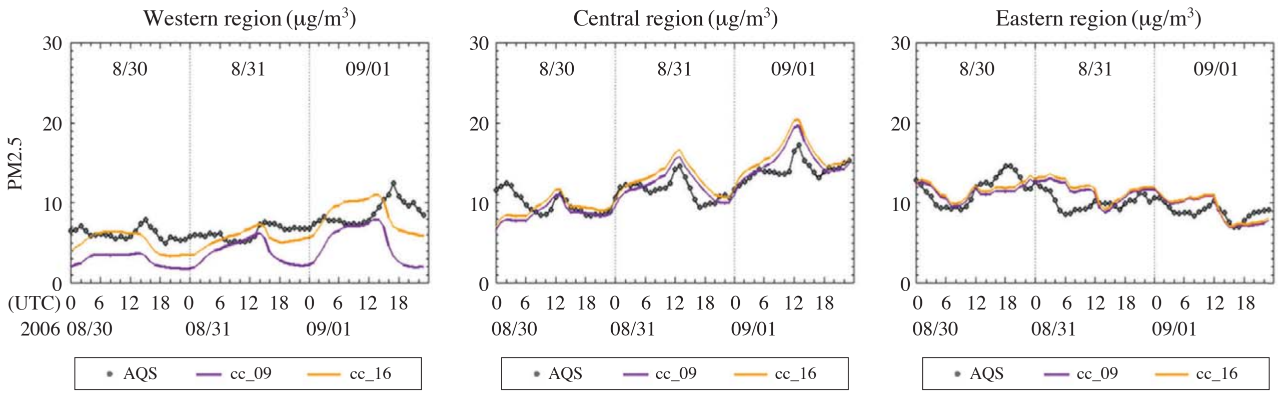


**Fig. 4.** Time series of CMAQ regional average PM2.5 concentration for three regions (western: left, central: middle, eastern USA: right), compared to AQS hourly PM2.5 measurements. Purple and orange lines are the PM2.5 concentration from CMAQ with predefined and RAQMS LBCs, respectively. Black circle lines are the observed PM2.5.

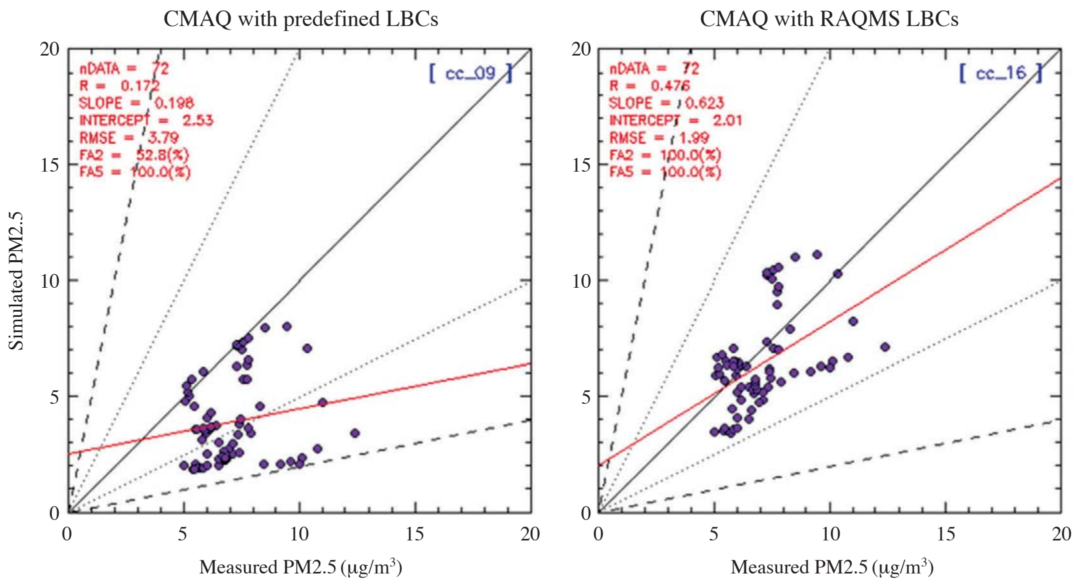


**Fig. 5.** Scatter diagram of regional average PM2.5 concentration for western region for August 30-September 1, 2006. CMAQ with predefined (left) and RAQMS (right) LBCs are compared with the EPA AQS PM2.5 data.

with observations. As presented in the Fig. 5, CMAQ with predefined LBCs seriously under-predicted when the observed level was over 8 μg/m$^3$, while CMAQ with RAQMS LBCs was able to predict observed high PM 2.5 in the western region. As shown in the Table 1, statistics for the eastern and central regions are almost same in the two CMAQ simulations, but the CMAQ with RAQMS LBCs case for the western region showed better PM2.5 prediction by improving the correlation coefficient from 0.17 to 0.48 as well as index of agreement from 0.38 to 0.63. These results clearly suggest that the CMAQ PM2.5 predictions can be improved by providing appropriate boundary conditions inher-

**Table 1.** Statistics between AQS observations and two CMAQ results for August 30-Semtember 1, 2006 (CMAQ results with predefined ('cc_09'), and RAQMS ('cc_16') LBCs; MM=model mean; OM=observation mean; R=correlation coefficient; IOA=index of agreement).

| Region | Case | MM | OM | R | IOA |
|---|---|---|---|---|---|
| Western | cc_09 | 3.93 | 7.04 | 0.17 | 0.38 |
| | cc_16 | 6.39 | | 0.48 | 0.63 |
| Central | cc_09 | 12.06 | 11.84 | 0.83 | 0.88 |
| | cc_16 | 12.81 | | 0.83 | 0.84 |
| Eastern | cc_09 | 10.70 | 10.25 | 0.62 | 0.78 |
| | cc_16 | 11.02 | | 0.63 | 0.76 |

ited from RAQMS outputs, demonstrating the importance of reliable PM2.5 boundary conditions in the CMAQ predictions.

## 4. CONCLUSIONS

In order to demonstrate the boundary conditions' impacts on CMAQ aerosol predictions, CMAQ results for August 30-September 1, 2006 were compared with AQS PM2.5 measurements. Mean PM2.5 lateral boundary conditions taken from RAQMS outputs showed strong variations both in the horizontal grid and vertical layers in the northern and western boundaries while mean PM2.5 values in the southern and eastern boundaries were much lower than those in northern and western boundaries.

CMAQ PM2.5 predictions with RAQMS LBCs were compared to CMAQ results with predefined LBCs for the three regions. CMAQ results in the eastern and central regions, there were little and slight changes of PM2.5 concentrations by adding aerosols from the northeastern northern boundaries, respectively. However, in the western region, PM2.5 predicted with RAQMS LBCs were significantly enhanced due to high PM2.5 boundary conditions from the northern and western boundaries resulting in increase of PM2.5 by 2-4 $\mu g/m^3$. CMAQ with predefined LBCs seriously under-predicted when observation was over 8 $\mu g/m^3$, while CMAQ with RAQMS LBCs was able to simulate observed high PM2.5 in the western region, which resulted in improvements of index of agreement from 0.38 to 0.63. These results obviously suggest that the CMAQ PM2.5 predictions can be improved by providing appropriate boundary conditions inherited from RAQMS outputs, demonstrating the importance of reliable aerosol boundary conditions in the CMAQ aerosol predictions.

We verified and characterized the CMAQ results by comparing with measurements data and showed the benefits of utilizing the fully assimilated RAQMS LBCs to improve regional air quality predictions of PM2.5. These CMAQ 36 km domain results can be nested down to 12-4 km domain simulations to generate high spatial resolution atmospheric chemistry model data.

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