SENSITIVITY ANALYSIS OF SO_2 EMISSIONS TO AEROSOL SULFATE OVER EAST ASIA BY CMAQ-DDM

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1. INTRODUCTION

In East Asia, its air quality has been dramatically changing due to the new policy of Chinese government in air pollution regulation. In our previous study, we found the variability in the fine-mode (submicron) aerosol optical depth (AOD) over the oceans adjacent to East Asia increased (4-8 %/yr) to a peak around 2005-2006 and subsequently decreased (4-7 %/yr) during 2000-2010, as revealed based on MODIS aerosol sensor and CMAQ modeling analysis. Such fluctuations in AOD are thought to reflect the widespread installation of fuel-gas desulfurization (FGD) devices in coal-fired power plants in China because aerosol sulfate is a major determinant of the AOD in East Asia [Itahashi et al., 2011].

Under the above mentioned background, it is necessary to clarify the source-receptor relationships of emissions from each country to the impact of chemical concentrations over East Asia. In this work, we investigate the sensitivity of SO_2 emissions to aerosol sulfate based on the Decoupled Direct Method (DDM) implemented in CMAQ version 4.7.1.

2. METHODOLOGY

2.1 CMAQ-DDM

We used the Community Multi-scale Air Quality (CMAQ) modeling system version 4.7.1 released by the US EPA [Byun and Schere, 2006] equipped with DDM [Napelenok et al., 2008; Kim et al., 2009]. This model is driven by meteorological fields generated by the Weather Research and Forecasting (WRF; ver.3.1.1) model with initial and boundary conditions defined by NCEP/NCAR reanalysis data with 1° × 1° resolution, 6 hour interval. The horizontal grids comprise 98×78 grids, with a resolution of 80 km. The vertical grids extend from the surface to 50 hPa with 38 stretching grid layers on eta coordinate. Modeling domain is shown in Fig. 1. In this study, the Statewide Air Pollution Research Center version 99 (SAPRC-99) chemical mechanism was used for producing gas phase chemistry, and the AERO5 module was employed for aerosol calculation. An applications of CMAQ model simulation were reported by Uno et al. [2007] and Itahashi et al. [2010].

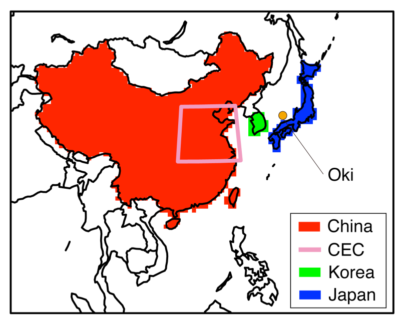

Figure 1. Modeling domain and the region settings as source regions (China, Central Eastern China (CEC), Korea, and Japan) and a receptor location (Oki).

DDM enables accurate and computationally efficient calculation of sensitivity coefficients for evaluating the impact of sensitivity parameter variations on chemical concentration. Sensitivity coefficients represent the response of chemical concentration, C, to the perturbations in a sensitivity parameter, p (e.g., emissions, initial condition, boundary condition, or reaction rate). An unperturbed (base case) sensitivity parameter, P, have the relation with p;

$$p = \varepsilon P = (1 + \Delta\varepsilon) P \qquad (1)$$

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where ε is a scaling factor , with a nominal value of 1. The semi-normalized first-order sensitivity coefficients, $S^{(1)}$, are defined as follows;

$$S^{(1)} = P\,(\partial C / \partial p) = \partial C / \partial \varepsilon \quad (2)$$

To project to any fractional perturbations away from the base case simulation, by incorporating Taylor series expansion, the corresponding concentration can be approximated by

$$C = C_0 + \Delta\varepsilon\, S^{(1)} + \cdots \quad (3)$$

where C_0 are the concentrations in the base case simulation.

The zero-out source contribution (ZOC) of an emission source is defined as the differences between C_0 and the concentration that would occur if the source did not exist. The first-order approximation of ZOC can be calculated from DDM results by setting $\Delta\varepsilon = -1$ in equation (3) and expressed;

$$ZOC \sim C_0 - C(p{=}0) = S^{(1)} \quad (4)$$

In this sense, the ZOC is equal to the semi-normalized first-order sensitivity itself.

2.2 Model Configuration

We set the four source regions (China, Central Eastern China (CEC), Korea, and Japan) for CMAQ-DDM (Fig. 1). The receptor site is set at Oki in Japan (Fig. 1), which located the down-wind region of China and Korea, and far from the Japanese industrial regions, considered as a remote site. In this study, to clarify the source-receptor relationships of sulfate which serve as the major component of aerosols over East Asia, we focused the sensitivity of anthropogenic SO_2 emissions to the sulfate concentration from four source regions. Anthropogenic emissions data were obtained from the Regional Emission Inventory in Asia (REAS) [*Ohara et al.*, 2007].

3. RESULTS AND DISCUSSION

A base CMAQ simulation was performed prior to DDM sensitivity simulation. In summer, AOD marked its peak through the entire year, therefore this base simulation was conducted for July, 2005 with 10 days spin-up. This base simulation results are compared with the EANET (Acid Deposition Monitoring Network in East Asia) observation data at Oki and shown in Fig. 2.

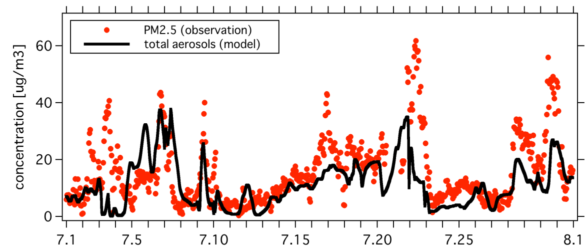

Figure 2. Time-series of observed $PM_{2.5}$ concentration and modeled total aerosol concentration at Oki.

CMAQ underestimate the peak concentration of observed $PM_{2.5}$, however, captures the general temporal variation. Observed $PM_{2.5}$ was 15.5 μg/m^3, in monthly averaged value, and highest in 22nd, July, 2005 with around 60 μg/m^3. From the CMAQ modeling results, monthly mean total aerosols was 11.3 μg/m^3 and sulfate was 8.0 μg/m^3 (over 70%). We targeted this highest peak event, and analyzed its source-receptor relationships by CMAQ-DDM for the period of 20-22 July (3 days) in 2005 with 5 days spin-up.

In Fig. 3, the spatial distribution of sulfate concentration and sulfate contribution by ZOC (see, eq. (4)) for domain-wide SO_2 emissions averaged in 22nd, July, 2005 are shown.

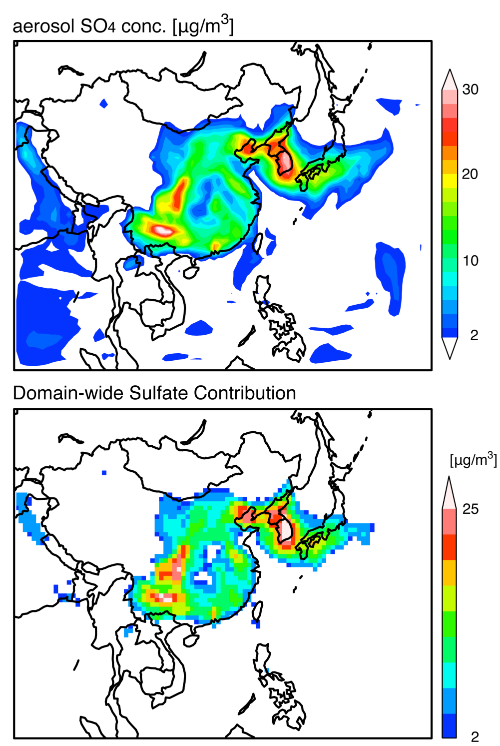

Figure 3. Modeled sulfate concentration (top) and sulfate contribution by ZOC in domain-wide SO_2 emissions (bottom) averaged in 22nd, July, 2005.

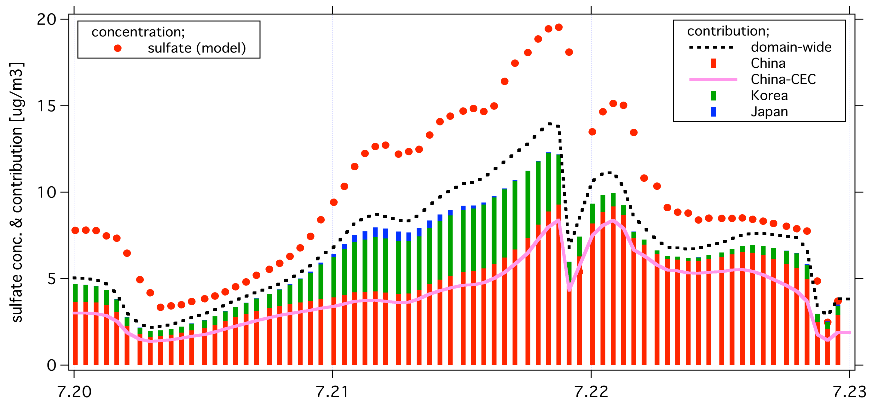

Figure 4. Time-series of sulfate concentration (red circle) and sulfate contribution by ZOC (black dashed line; domain-wide, red bar; China, pink line, CEC, green bar; Korea, blue bar; Japan) at Oki site.

We found that the sulfate contribution are well correspond to the sulfate concentration. For comparison of DDM results, we also perform the sensitivity analysis via brute-force method (BFM) by comparing the base case and 20% perturbated case. DDM reproduce the BFM results well and its correlation coefficient was 0.97.

The time-series of sulfate concentration and sulfate contribution from four source regions computed by ZOC are shown in Fig.4, and its spatial distribution are illustrated in Fig. 5. The daily averaged contribution from each regions are summarized in Table 1.

The contribution from China is dominant through this period, and Korea also dominate the sulfate contribution at Oki in 21st. During this period, the contribution from Japan is quite low, because the wind from north-west and north are prevailing in 21st and 22nd, respectively (Fig. 5). We found that the sensitivity of SO_2 emissions from CEC region play an important role over the oceans adjacent to East Asia. From the REAS emissions inventory, SO_2 emissions in CEC region cover 44 % emissions in China, however, its contribution to sulfate sensitivity at Oki is above 85 % (Table 1). Due to the transport pattern of Asian monsoon in summer, CEC is considered as the most impacting region in China for down-wind regions.

Table 1. Summary of the daily averaged contribution by ZOC from each region to Oki site at 21st and 22nd, July, 2005.

	China	CEC	Korea	Japan
21st	5.50	4.84 (88%)	3.14	0.25
22nd	6.17	5.30 (86%)	0.49	0.01

unit: μg/m^3

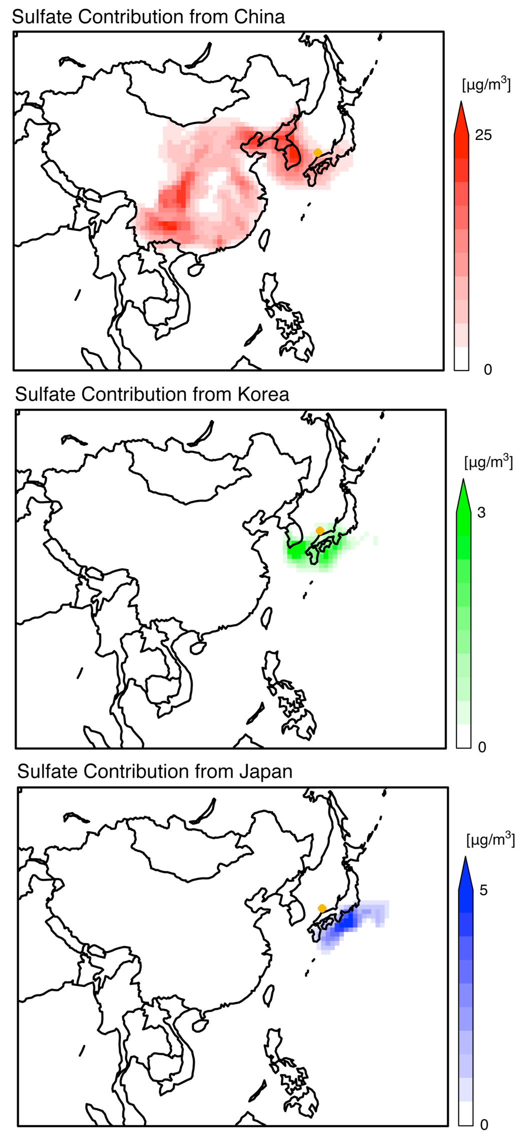

Figure 5. Spatial distribution of sulfate contribution by ZOC in China (top; red), Korea (middle; green) and Japan (bottom; blue) averaged in 22nd, July, 2005. Orange circle indicate the location of Oki site.

From the unit-based method, it is reported that CEC region became widespread of FGD equipment in the late 2000s. For instance, percentage of unit installed with FGD in Beijing region are under 20% in 2005 and expected to reach 60% in 2010 [Zhao et al., 2008]. By the results of DDM, in the down-wind region of China, CEC is the most dominant region of its SO_2 emissions. The widespread FGD penetration leads to the declining of SO_2 emissions, and could serve as a declining trend of sulfate, eventually fine-mode AOD above East Asian down-wind regions.

4. CONCLUSIONS

In our previous study, we clarified the variability in the fine-mode AOD over the oceans adjacent to East Asia show increasing (4-8 %/yr) to a peak around 2005-2006 and subsequently decreasing (4-7 %/yr) trends during 2000-2010 based on space-borne MODIS aerosol sensor and CMAQ modeling analysis. Here, in this work, we evaluated the sensitivity of SO_2 emissions to sulfate concentration by using the CMAQ-DDM to elucidate the source-receptor relationships over East Asia.

In July 2005, CMAQ model capture the observed temporal variation of $PM_{2.5}$ at Oki, which located in the down-wind region in China and Korea. We focused the highest episode in 20-22 July, and applied CMAQ-DDM by setting the four source regions. China was the most dominant contributor to sulfate contribution through this period. We also found the contribution from Korea in 21st. Because of the prevailing wind from north-east or north directions, the contribution from Japan was quite low. We found that the CEC is the most impacting region in China to the down-wind region. SO_2 emissions from CEC region was only 44% of the total Chinese emissions, however, its contribution rate at Oki was above 85% within total Chinese contribution of sulfate along of the Asian monsoon transport pattern in summer time. The fast-growing equipment of FGD during the late 2000s in CEC region is expected to link with the decrease of SO_2 emissions, and could serve as the decreasing trend of sulfate concentration, furthermore of fine-mode AOD observed by the satellite measurement.

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