MODELING STUDY ON LONG-RANGE TRANSBOUNDARY AIR POLLUTIONS IN NORTHEAST ASIA

Tatsuya SAKURAI

Japan NUS Co., Ltd. (JANUS), Tokyo, Japan

Abstract: Model validation and Source-Receptor relationship analysis for Sulfur compounds in Northeast Asia in March, July and December 2001 were conducted using RAQM (Regional Air Quality Model). RAQM is a three-dimensional off-line Eulerian model. In RAQM, 12 layers stretch vertically from surface to about 10km with a horizontal resolution of 0.5 degree. Emission inventories of SO_2, NOx, CO, NH_3, and VOCs are derived from emission inventories prepared in MICS-Asia Phase 2 (Model InterComparison Study in Asia). The MM5 is used to provide meteorological fields for RAQM. NCEP FNL reanalysis data with 1.0 degree resolution were used to provide initial and boundary conditions for meteorological fields. Four-dimensional data assimilation (nudging by three-dimensional reanalysis data) was utilized to improve the meteorology results, especially for wind field and temperature. The domain for MM5 is larger than that of RAQM, with the center located at 35N, 125E. There are 125x95 grid numbers in plane with 45 km grid resolution, and 23 sigma layers from surface to 100hPa. MM5 outputs are adequately interpolated from Lambert projection to spherical coordinate of RAQM. Experimental monitoring data from EANET (Acid Deposition Monitoring Network in East Asia) and their relevant information were made use of for the evaluation of model results.

Key words: Air Quality Modeling; Acid Deposition in East Asia; EANET; MICS-Asia; LTP

1. INTRODUCTION

Because of a rapid economic growth in East Asian region for the last several decades, some impacts for air quality and ecosystem are getting more and more obvious as well. Significant increases in emissions and changes in environment in East Asia also have important implications for atmospheric chemical cycling and climate change at both regional and global scales. Chemical transport models (CTMs) contribute for the regional assessment of the air quality and policy design by simulating the fate and transport of emissions.

In order to evaluate the air quality in East Asia, Model validation and Source-Receptor relationship analysis for Sulfur compounds in Northeast Asia in March, July and December 2001 were conducted using RAQM (Regional Air Quality Model). RAQM is a three-dimensional off-line Eulerian model (Han et al., 2006) developed by Acid Deposition and Oxidant Research Center (ADORC). Meteorological fields as input data for RAQM were calculated by MM5 ver. 3.7. Emission input was derived from emission inventories prepared in MICS-Asia Phase 2 project (Model InterComparison Study in Asia). Experimental monitoring data from the Acid Deposition Monitoring Network in East Asia (EANET) and their relevant information were made use of for the evaluation of model results.

2. METHODOLOGY

2.1 MODELING DESCRIPTION

In RAQM, 12 layers stretch vertically from surface to about 10km (50, 150, 300, 500, 750, 1500, 2500, 3500, 4500, 6000, 7500, 8950m) with a horizontal resolution of 0.5 degree. Initial and boundary conditions were taken as the lower end of available observations from recent studies for East Asia. Side boundary conditions were held fixed during simulation, whereas top boundary condition for O_3 was taken as a constant level with seasonal variation. Emission input of SO_2, NOx, CO, NH_3, and VOCs were derived from emission inventories prepared in MICS-Asia Phase 2. The emission inventories consider anthropogenic sources (area sources and SO_2 point sources), biomass burning, and volcano, on 0.5°x0.5° grid resolution basis.

MM5 was used to provide meteorological fields for RAQM (wind, temperature, water mixing ratio, precipitation and surface variables). NCEP FNL reanalysis data with 1.0 degree resolution were used to provide initial and boundary conditions for meteorological fields. Four-dimensional data assimilation (nudging by three-dimensional reanalysis data) was utilized to improve the meteorology results, especially for wind field and temperature. The domain for MM5 was larger than that of RAQM, with the center located at 35N, 125E. There were 125x95 grid numbers in plane with 45 km grid resolution, and 23 sigma layers from surface to 100hPa. MM5 outputs were adequately interpolated from Lambert projection to spherical coordinate of RAQM.

This study adopted the same simulation periods with MICS-Asia Phase2. The simulation was conducted for March, July and December in 2001. March 2001 was a period of high dust transport, high volcanic emissions, and elevated ozone. July and December 2001 provided examples of a summer periods with warm and high precipitation conditions, and a winter period with cold and low precipitation conditions, respectively.

MICS-Asia project (*http://www.adorc.gr.jp/adorc/mics.html*) is an international intercomparison study of existing models to be applied to East Asia region with the purpose of creating a common understanding of model performance and uncertainties in Asia on long-range transport and deposition. Phase 2 activities had conducted during the period from 2003 to 2007 as a collaborative study between ADORC, which is the Network Center for EANET, and IIASA (International Institute for Applied System Analysis), with the participation of scientists from China, Korea, Japan, and other countries from Europe and North America (Carmichael et al., 2008).

2.2 SOURCE - RECEPTOR RELATIONSHIP ANALYSIS

Figure 1 shows the simulation domain. Source-Receptor relationship was analyzed based on the 5 regions, Region I-III for China, Region IV for Republic of Korea and Region V for Japan. Source-receptor relationship for total sulfur deposition was analyzed based on the "reverse method". The reverse method evaluates the contribution from i^{th} domain to j^{th} domain by a difference between a simulation result with all emissions and a simulation result with all emission except for i^{th} domain.

The domain division used in this study is originated in LTP project (Long-range Transboundary Air Pollutants in Northeast Asia). LTP project is an international project established in 1995 upon the initiative of the Korean government, with China, Japan and the Republic of Korea now as member countries (Park et al., 2005; Kim et al., 2010)

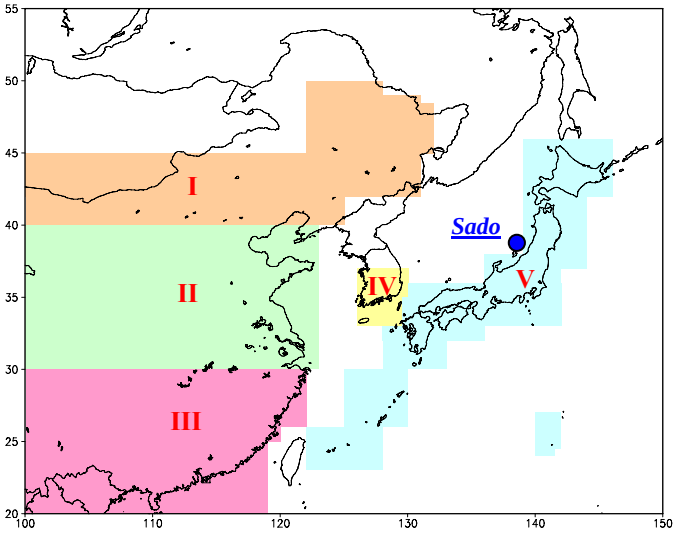

Figure 1 Study domain for simulation works and Source-Receptor relationship analysis

2.3 MONITORING DATA FOR MODEL VALIDATION

Observation data of EANET (http://www.eanet.cc/) were applied to model validation in this study. EANET was established in 1998 as regional cooperative initiative to promote efforts for environmental sustainability and protection of human health. Currently, EANET consists of 13 participating countries, and monitoring data of air quality and/or wet deposition are submitted from a total of 52 stations including 20 urban sites, 13 rural sites and 19 remote sites in 2008, with each country managing its own sites (EANET, 2009). This report introduces the model validation results using the EANET data observed at *Sado* station, which location is shown in Figure 1.

3 RESULTS AND DISCUSSIONS

3.1 MODEL VALIDATION USING EANET MONITORING RESULTS

Figure 2 indicates the comparison between simulated and observed SO_2, NOx and O_3 concentration at *Sado* station. Both of the simulated and observed concentrations are hourly averaged data.

As regarding SO_2, quite high concentration was observed in July and the concentration level became low in December. Although the simulation result misses several high concentration peaks, it is likely that the model shows the reasonable agreement on the level of SO_2 concentration in all months generally. The reason of high concentration observed in July is the volcanic emission. Mt. Oyama on Miyake-jima Island (Miyake-jima volcano: 139.53E, 34.08N, 813m MSL), located in the northwest Pacific Ocean, 180 km south of Tokyo metropolitan area, began to erupt on July 8, 2000, and has emitted huge amounts of SO_2 since then. SO_2 emission from the Miyake-jima volcano considered in the simulation is March: 27,350 ton day^{-1}, July: 14,611 ton day^{-1} and December: 12,960 ton day^{-1} in the target year of 2001. The Miyake-jima Volcano played a large role in SO_2 concentration in Japan because the prevailing seasonal winds from the southeast (Pacific Ocean) exists in summer season (Figure 3). In fact, peaks of high SO_2 concentrations disappear in the simulation without emission from the Region V (blue line). As regarding NOx, although the underestimation appears in December, the model predicted the observation results as well as SO_2. Since the high concentration peaks disappear on the blue line, these peaks likely originate in the local sources.

Simulated O_3 shows almost the same trends in March and December between red and blue lines. In July, high concentration observed on 10^{th} July is successfully predicted by the model, but the peak disappears on the blue line. As a result, it is suggested that the O_3 observed at Sado are generated from the sources in Region V in July and are transported from the outside of Region V in March and December.

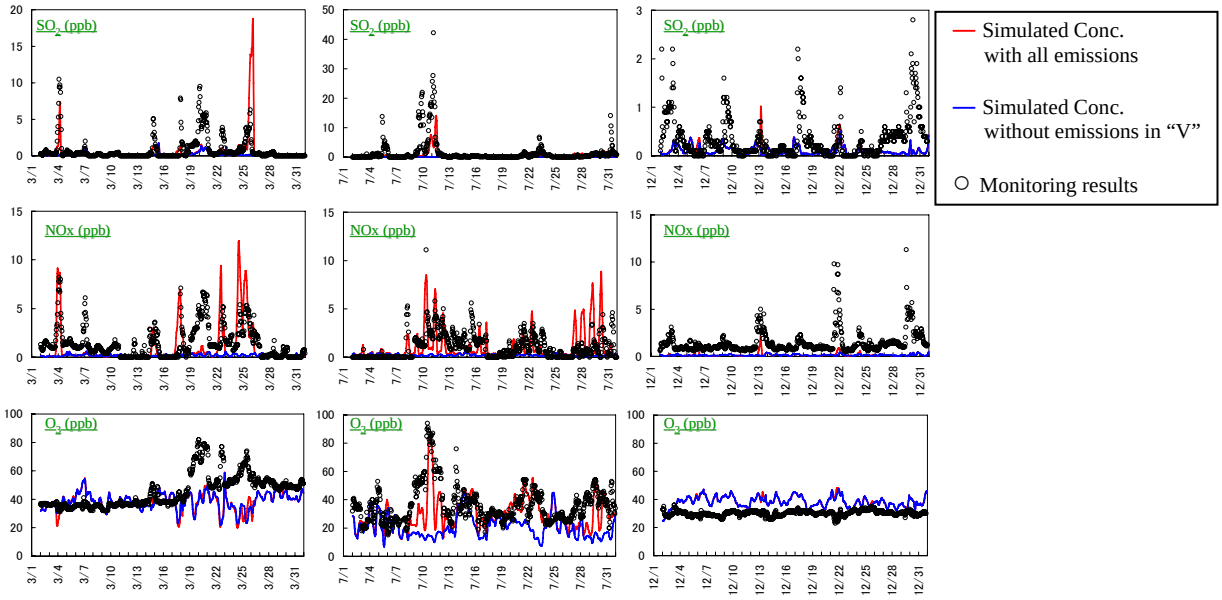

Figure 2 Comparison between simulated and observed SO_2, NOx and O_3 concentrations at Sado station on hourly basis. Red and blue lines indicate the simulated results with all emissions or without emissions from Region V, respectively.

3.2 SENSITIVITY ANALYSIS OF CONCENTRATIONS RELATED TO THE EMISSION CONTROL

Table 1 summarizes the variation of the monthly averaged simulated concentrations at *Sado* station related to the emission reduction for Region I to V. "without ALL" in Table 1 includes the contribution from boundary/initial conditions. Region I and II have relatively large Impacts to SO_2 and NOx concentration at *Sado* in December. Especially the contribution of SO_2 from Region II reaches almost 40%, although the concentration level is low. On the other hand, those impacts are little in July. Instead, the concentration tends to increase with emission reduction, which is likely caused by the atmospheric chemical reaction. As mentioned above, Region V has a large contribution to O_3 concentration at *Sado* in July. Since the contribution from Region I to V are little in March and December, it seems that O_3 concentration at *Sado* is mainly occupied by the background concentration from the boundary conditions in those months.

Table 1 Variation of the monthly averaged simulated concentrations related to the emission reduction

		Simulated Conc. (ppb)	without I	without II	without III	without IV	without V	without ALL
SO_2	Mar	0.64	-6.7%	-12.0%	-0.2%	-5.0%	-69.1%	-91.4%
	Jul	0.65	0.4%	0.1%	0.6%	-2.8%	-83.8%	-88.4%
	Dec	0.12	-21.1%	-39.6%	-1.3%	-5.6%	-11.3%	-68.4%
NOx	Mar	0.99	-3.7%	-0.5%	0.1%	-1.5%	-80.4%	-88.6%
	Jul	1.03	0.8%	0.9%	0.8%	0.6%	-78.9%	-82.4%
	Dec	0.17	-24.2%	-8.5%	-1.9%	-4.4%	-21.4%	-62.1%
O_3	Mar	38.69	-2.9%	-2.3%	-0.1%	-0.9%	-0.3%	-7.6%
	Jul	29.99	-1.6%	-4.5%	-2.6%	-7.6%	-31.5%	-50.5%
	Dec	38.44	-1.7%	-1.6%	-0.2%	-0.1%	-0.3%	-4.3%

* "without ALL" includes the contribution from boundary/initial conditions.

3.3 SOURCE - RECEPTOR RELATIONSHIP ANALYSIS FOR SULFUR DEPOSITION

Figure 3 indicates the simulated monthly mean wind fields and the amount of "SO_2 dry deposition", "Sulfate dry deposition" and "SO_4^{2-} wet deposition". Predicted wind conditions around Japan on March, July and December are Westerly, Southerly and Northwesterly, respectively. Northwestern area of the study area does not have the deposition in March and December due to rainless and no large emission sources around the area. On the other hand, large deposition is predicted in the area from Eastern China to Korean Peninsula to Japan on all of the months. In July, relatively large deposition is concentrated in or in the vicinity of major source areas, such as Beijing and Tianjing, Chongqing, Yangtze Delta, Shandong province, Seoul of Republic of Korea and Miyakejima volcano of Japan, with maximum amounts exceeding 2,400 mg m^{-2} around Beijing and Chongqing.

Table 2 summarizes the estimated contribution of source to receptor for the the amount of "SO_2 dry deposition", "Sulfate dry deposition" and "SO_4^{2-} wet deposition" on March, July and December 2001. "Others" in Table 2 is calculated by the difference between 100% and the sum of the contributions from all regions, which includes the contribution from boundary/initial conditions and nonlinear increase or decrease produced by the atmospheric chemical reaction. "S" and "R" indicate the source region and the receptor region, respectively.

Generally, Region V is the downwind region in March and December. Moreover, Region V covers a large area of ocean. Thus, the contributions to the total deposition amount from outside become larger and its ratio is March: 47.8% and December: 64.2%. In July, the outside contribution becomes smaller, with the ratio of 21.6%. This is mainly because the prevailing winds from Pacific Ocean which is caused by the existence of high atmospheric pressure around region V in summer season. Moreover, it is suggested that there are the apparent increases in precipitation and consequent more scavenging in the other regions, which can also be demonstrated by the increases in wet depositions in all of the regions.

The estimated self contribution of Region V was March: 52.2%, July: 78.4% and December: 35.8%. However, the self contribution rate can be divided into the volcanic emissions and the other emission sources, with the ratio of 43.8% and 8.4% on March, 58.7% and 19.7% on July and 25.0% and 10.8%, respectively. In fact, the volcanic emissions have a large influenced on the Sulfur deposition in Region V as in the SO_2 concentration observed at *Sado*.

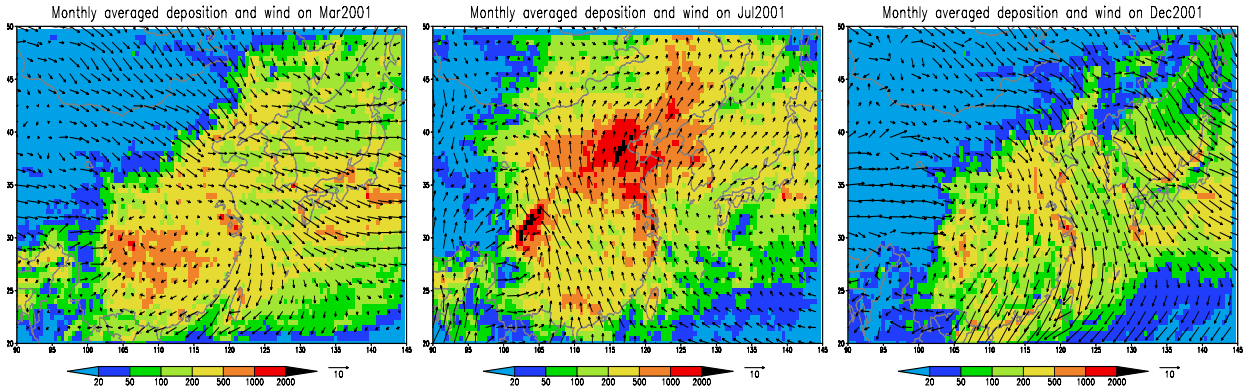

Figure 3 Predicted monthly mean wind fields (m s^{-1}) and the amount of "SO_2 dry deposition", "Sulfate dry deposition" and "SO_4^{2-} wet deposition" (mg m^{-2})

Table 2 Estimated contribution of source to receptor for Sulfur deposition on March, July and December 2001

2001	S \ R	I	II	III	IV	V
March	I	56.6%	1.4%	0.1%	4.6%	1.7%
	II	17.8%	86.9%	11.1%	22.3%	16.3%
	III	0.1%	6.1%	74.8%	2.6%	10.8%
	IV	0.6%	0.0%	0.0%	56.1%	1.6%
	V	0.6%	0.0%	0.0%	1.6%	52.2%
	Others	24.3%	5.6%	14.0%	12.8%	17.4%
July	I	31.9%	0.6%	0.0%	0.7%	1.1%
	II	50.4%	75.8%	4.8%	15.5%	5.9%
	III	1.0%	18.3%	81.8%	7.7%	2.2%
	IV	2.2%	0.2%	0.0%	61.7%	3.1%
	V	0.2%	0.1%	0.2%	6.0%	78.4%
	Others	14.3%	4.9%	13.2%	8.3%	9.3%
December	I	51.2%	2.1%	0.2%	5.4%	3.5%
	II	22.5%	83.3%	17.6%	23.3%	21.3%
	III	0.1%	7.0%	72.2%	1.8%	7.9%
	IV	0.1%	0.0%	0.1%	47.7%	3.1%
	V	0.0%	0.0%	0.3%	1.8%	35.8%
	Others	26.2%	7.6%	9.6%	20.0%	28.3%

* "Others" is calculated by the difference between 100% and the sum of the contributions from all regions, which includes the contribution from boundary/initial conditions and nonlinear increase or decrease produced by the atmospheric chemical reaction.
* "S" and "R" indicate the source region and the receptor region, respectively.

4 CONCLUSIONS

In order to evaluate the air quality in East Asia, Model validation and Source-Receptor relationship analysis for Sulfur compounds in Northeast Asia in March, July and December 2001 were conducted using RAQM (Regional Air Quality Model). Meteorological fields as input data for RAQM were calculated by MM5 ver. 3.7. Emission input was derived from emission inventories prepared in MICS-Asia Phase 2 project (Model InterComparison Study in Asia). Experimental monitoring data from the Acid Deposition Monitoring Network in East Asia (EANET) and their relevant information were made use of for the evaluation of model results.

It is likely that the model shows the reasonable agreement on the level of SO_2, NOx and O_3 concentration monitored at Sado station in all months generally. The volcanic emission played a large role in SO_2 concentration in Japan on July because the prevailing seasonal winds from the southeast (Pacific Ocean) exists in summer season. In fact, peaks of high SO_2 concentrations disappear in the simulation without emission from Japan (Region V). As regarding O_3, it is suggested that the O_3 observed at Sado are generated from the sources in Region V in July and are transported from the outside of Region V in March and December.

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As in the SO_2 concentration observed at Sado, the volcanic emissions have a large influenced on the Sulfur deposition in Region V. The estimated self contribution of Region V was March: 52.2%, July: 78.4% and December: 35.8%. However, the self contribution rate can be divided into the volcanic emissions and the other emission sources with the ratio of 43.8% and 8.4% on March, 58.7% and 19.7% on July and 25.0% and 10.8%, respectively.

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