Analyses of Regional Air Pollution by Sulfur Oxides in East Asia Using A Long-Range Transport Model

Proceedings: International Symposium of the Kanazawa University 21st-Century COE Program Vol. 1 (2003)
Edited by Naoto KAMATA

Analyses of Regional Air Pollution by Sulfur Oxides in East Asia Using A Long-Range Transport Model

Takahisa MAEDA
National Institute of Advanced Industrial Science and Technology, Tsukuba, Ibaraki 305-8569, JAPAN

Abstract - A computer simulation model for long-range transport (LRT) of air pollutants in East Asia region developed and operated in AIST is introduced. A brief description of the model, some analyses of the atmospheric sulfur oxides for long term averaged distribution in China, and hourly basis analysis of the recent intensive observation campaign held around Amami-Oshima are indicated.

I. Introduction

Since 1980's, as the economy has been grown and the energy consumption has been increased, intensification of the air pollution in the continental scale has been apprehended in East Asia region. It firstly attracted attention as the 'acid rain' problem that caused by the 'trans-boundary' transport of acidic pollutants such as sulfur oxides and nitrogen oxides. Studies on the acidic deposition in East Asia have been carried out from many aspects, theoretical analyses, field monitoring, simulation models, impact assessment and so on. In the last decade, several simulation models dealing with transport of acidic substances in 1000km scale were developed to estimate the deposition amount in East Asia on monthly or annual basis (eg. [1], [2], [3] and so on).

In recent years, as the interest in the global climate change caused by the greenhouse gases has been heightened, impacts of the atmospheric particulate matters, those directly or indirectly affect the atmospheric radiation, onto the regional climate have been also a concern. Those particulate matters are originated from not only anthropogenic sources but also natural sources such as deserts supplying soil dusts or active volcanoes erupting huge amount of ash and gases. And research projects on the continental scale air pollution by aerosols in Asia region such as ACE-Asia (Asian Pacific Regional Aerosol Characterization Experiments) and APEX (Asia Atmospheric Particulate Environment Change Studies) those include field campaigns as their core activity have been organized. For analyses and predictions of the episodes those occur during such intensive observations, fine time resolution and capability of dealing with variety of chemical constituents have been required to models. And with the progress of the computer technology, long-range transport models mentioned above are being improved to equip more complicated chemical reaction modules and to have more fine resolution of time and space.

In this paper, a simulation model of long-range transport of air pollutant developed in AIST (National Institute of Advanced Industrial Science and Technology, Japan) is introduced with some results of analyses for atmospheric sulfur oxides, for both long-term average and fine time resolution.

II. Brief description of the model

The long-range transport model (LRT model, LRTM) being developed in AIST is a three-dimensional Eulerian transport model [4]. It can be categorized as an engineering model that is equipped with simplified chemical processes. The domain of the model includes most of the densely populated areas in East Asia region where large anthropogenic emission sources of air pollutants are located, and was divided by 1°x1° grids. The vertical coordinate is given with the sigma-coordinate system defined by $\sigma=p/p_s$, where p and p_s denote the pressure at arbitrary altitude and the surface pressure respectively, then 10 layers up to the level of σ=0.6 (approximately 4000m from the sea surface) are arranged.

The processes in each grid box are handled by:

$$\frac{\partial(\rho Q)}{\partial t}=-\frac{\partial(u\rho Q)}{a\cos\varphi\partial\lambda}-\frac{\partial(v\rho Q)}{a\partial\varphi}-\frac{\partial(\dot{\sigma}\rho Q)}{\partial\sigma}+\frac{1}{a^2\cos^2\varphi}\frac{\partial}{\partial\lambda}\left(K_h\frac{\partial(\rho Q)}{\partial\lambda}\right)+\frac{1}{a^2\cos\varphi}\frac{\partial}{\partial\varphi}\left(K_h\cos\varphi\frac{\partial(\rho Q)}{\partial\varphi}\right)+\left(\frac{g}{p_s}\right)^2\cdot\frac{\partial}{\partial\sigma}\left(\rho^2K_z\frac{\partial(\rho Q)}{\partial\sigma}\right)+R_i-D_{di}-D_{wi}+E_i \quad (1)$$

where i denotes the chemical species, i.e. sulfur dioxide

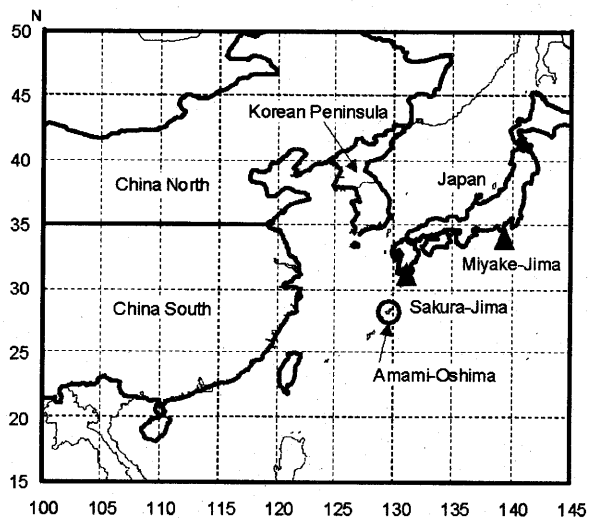

Fig. 1. The domain of the LRT model. Thick lines indicate the boundaries of areas of anthropogenic SOx emission used for source analysis at Amami-Oshima described in section IV [5]. Triangles are the major volcanoes those can influence the SOx concentration around Amami-Oshima.

(SO_2) or sulfate (SO_4^{2-}), Q_i is mixing ratio, t is time, a is the radius of earth, ϕ and λ are longitude and latitude, u and v are horizontal winds, $\dot{\sigma}$ is vertical wind, K_h and K_z are horizontal and vertical diffusion coefficients, ρ is air density, and g is the gravity acceleration. The terms R_i, D_{di} D_{wi} and E_i are reaction, deposition and emission rate respectively. The advection term is calculated by the method with the conservation of the second-order moments. The vertical diffusion term is solved by Crank-Nicholson scheme using the vertical diffusion coefficients with diurnal variation. The chemical reaction term of SO_2 is processed by the simple first order conversion to SO_4^{2-}. The dry deposition term, D_{di}, is calculated with deposition velocities for various land usage and vegetation types in Japan applied to the lowest layer of each grid as $D_{di}=k_{di}(\rho Q_i)$ and $k_{di}=v_{di}/\triangle z$, where v_{di} and $\triangle z$ denote the dry deposition velocity and the depth of the grid box respectively. The wet deposition term D_{wi} is processed as $D_{wi}=k_{wi}(\rho Q_i)$ with deposition coefficient given by $k_{wi}=a_i r^{b_i}$, where r is precipitation rate and a_i and b_i are empirical constants. For the constants a and b, $a_{so2}=2.0\times10^{-5}$ and $b_{so2}=1$, $a_{so4}=2.0\times10^{-4}$, $b_{so4}=0.7$ are applied as the average of several literary values. For the emission, a part of Global Emissions Inventory Activity (GEIA) Global SO_X Inventory Version 1B, that has seasonal variation of point sources and area sources, is used. Since the GEIA data is an estimation for the year 1985, the emission rate is adjusted in proportion to the increase of national coal energy consumption in China [6] to apply to the LRTM. For the case study of the year 1992 described in section III, national coal energy consumption in China increased 1.4 times as that in 1985. And in 2000 described in section IV, the factor of increase is 1.7.

III. A long-term average analysis in China in 1992

Using the LRTM described above, an analysis for long-term average distribution of sulfur oxides was carried out to investigate the acidic deposition in China [4]. The objectively analyzed meteorological data (ECMWF/WCRP Level III-A data set) of European Centre for Medium Range Forecast was applied to the LRTM. And precipitation data from the world meteorological dataset compiled by the Japan Meteorological Agency was used to estimate wet deposition.

Fig. 2 shows the model-calculated distributions of the monthly total wet deposition in July, September, November,

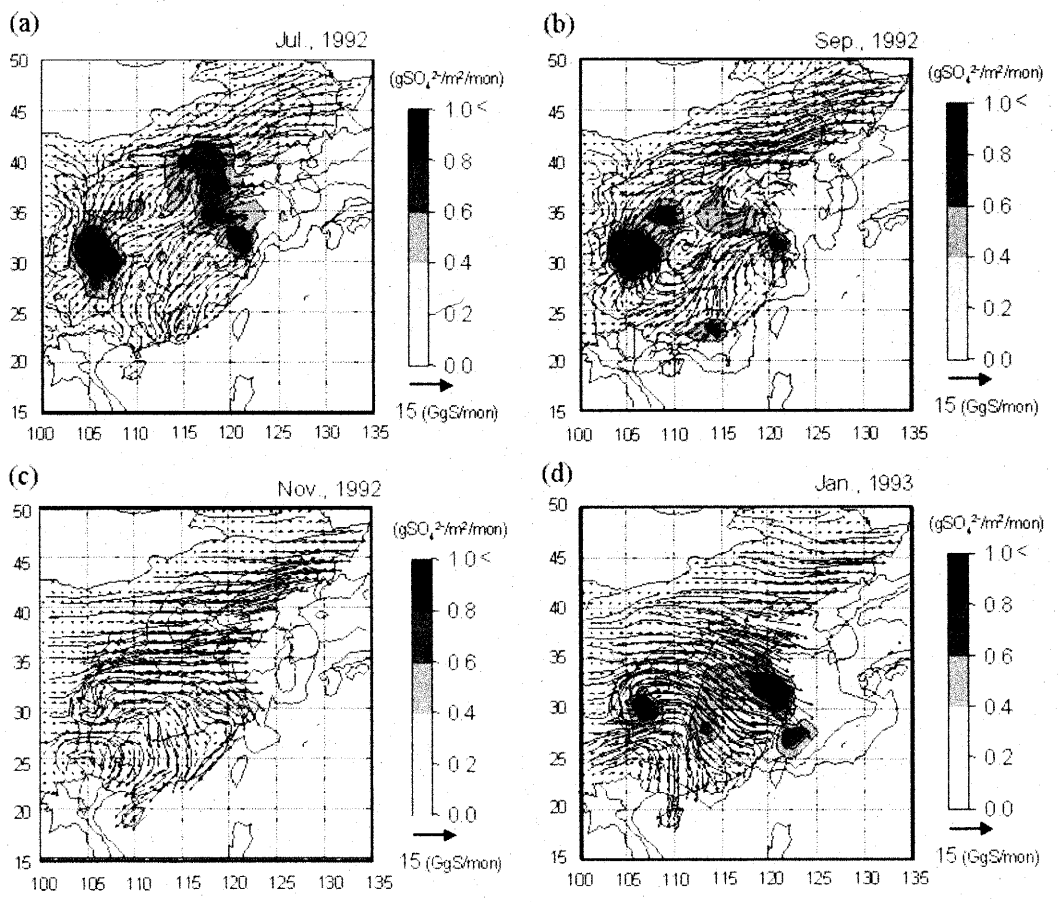

Fig. 2. Distribution of wet deposition of sulfur oxides in (a) July, (b) September, (c) November 1992, (d) January 1993. Arrows are monthly averaged flow of sulfur oxides [4].

and January of 1992. The calculation results were compared with the monthly mean concentration of sulfuric acid in rainwater measured at 69 locations those widely distributed in China [7]. The comparison showed that the calculated concentrations were in the rage from 1/2 to 2 times (figure not shown) as those measured.

Taking an overview, it was found that the wet deposition covered the most of populated areas in China distributing similarly to the emission sources in July. The reason seems to be that the amount and frequency of precipitation were large in summer and the sulfur oxides were supposed to deposit shortly after being emitted from the source, before being transported long distance. While the precipitation decreased as the season proceeds, the wet deposition amount also decreased especially in the areas north from 35°N. The arrows in Fig.2 indicate the monthly averaged direction and the monthly total amount of sulfur oxides passed through the grid box. The figures indicate that there are large differences in seasonal change of the transport pathways between the north side and the south side of 35°N. In the north side, transport northeastward or eastward from the inland areas to the Sea of Japan through northeast China was always dominant. In the south side from Shandong peninsula, the seasonal variation of the transport paths was considerable.

IV. Hourly basis analyses around Amami-Ohshima in Dec. 2000 and Apr. 2001

In December 2000 and April 2001, the field campaigns of APEX project were conducted around Amami-Oshima (28°N, 129°E, shown in Fig. 1). In order to support the observations at the ground surface of Amami-Oshima, analyses of regional scale distribution and transport paths of

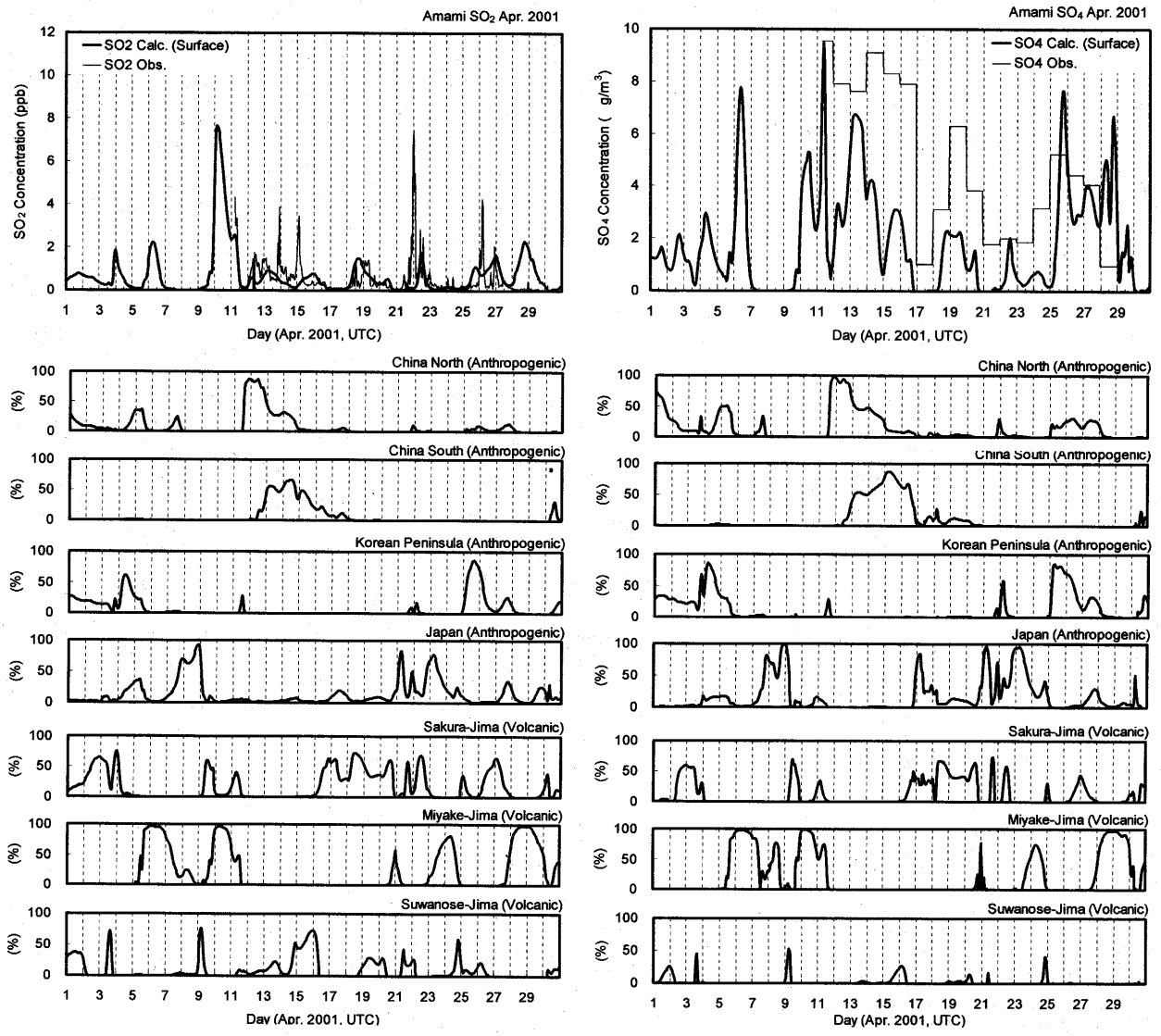

Fig. 3. Time variation of surface concentration at Amami-Oshima (upper chart) and relative contribution from each of seven sources described in section IV (lower seven charts) of SO_2 in April 2001 (left) and nss-SO_4^{2-} in April 2001 (right) [5]. In the upper charts, thick lines are model-calculated concentration, and thin lines indicate observed value. Lower seven rows are relative contribution from anthropogenic sources in northern part of China, southern part of China, Korean peninsula, Japan, and volcanic sources of Sakura-Jima, Miyake-Jima, Suwanose-Jima, respectively from the top.

sulfur oxides employing the LRTM were performed [5].

The meteorological data applied to the LRTM for the analyses was obtained from the output of PSU/NCAR MM5 (Pennsylvania State University / National Center for Atmospheric Research Mesoscale Model 5) that was run with NCEP (National Centers for Environmental Prediction) Reanalysis global data set. Precipitation rate was also estimated by MM5, because observed rainfall is unavailable over the sea areas.

The upper charts of Fig. 3 show the time variation of calculated and observed [8] concentration of SO_2 and nss-SO_4^{2-} (non-sea-salt sulfate) at Amami-Oshima site in April of 2001 including the period of the intensive observation of APEX-E2. The LRTM reproduced the trend of the observed concentrations reasonably to reveal the episodes those influenced the time variation of sulfur oxides concentrations, although the calculated nss-SO_4^{2-} concentration was underestimated in some extent. The analyses for other observation sites such as Cheju in Korea [9] suggested that the underestimation could be because of emission sources in the sea areas not considered in the calculation, especially the exhaust from numerous fishing boats working in Yellow Sea and East China Sea.

Using this model calculation, the origins of sulfur oxides reached around Amami area were analyzed on hourly basis. For the analysis, the major emission sources were divided into the four areas and the three volcanoes indicated in Fig. 1. (Suwanose-Jima, located about 200km north from Amami-Oshima, and erupted in Dec., 2000, is not indicated in Fig. 1.) Then the LRTM was run with the emission from each area and volcano eliminated. The relative contribution from the eliminated source was estimated by calculating the difference of the concentration at Amami-Oshima site from that calculated considering all emission sources in the domain of the model.

The results of analysis of origins on hourly basis are shown in the lower part of Fig. 3. According to the analysis, sulfur oxides reached around Amami-Oshima site from a variety of origins including the anthropogenic sources and the volcanoes located in the surrounding areas. Some of the sulfur oxides came even from Miyakejima that is located about 1000 km away from Amami-Oshima and has been hugely erupting since the summer of 2000. In December 2000, the dominant origin of the sulfur oxides around Amami-Oshima changed everyday and transport from the same origin repeated periodically with the interval of about 7 days (figure not shown). It seemed to be due to the periodical sequence of the formation and deformation of the 'winter weather pattern' in East Asia. As shown in Fig. 3, this periodical change of the dominant origin was also occurred in April 2001 while the interval was about 10 days, a little longer than that in December 2000, and was caused by the 'spring migratory anticyclone and cyclone with front' passed over East China Sea. In the middle of April 2001, a characteristic episode occurred that the origin changed rapidly from Miyakejima, east side of Amami, to the northern area of China, northwest side of Amami area, then changed to the southern area of China keeping high concentration. This episode also included a Kosa (yellow sand) event in Amami area.

V. Summary

The overview of the long-range transport model of air pollutants being developed in AIST was demonstrated with some case studies on analyses of atmospheric sulfur oxides in East Asia region for long-term average and hourly analysis of episodes. For both of the analyses, as mentioned in section III and IV, the LRTM reproduced the transport of sulfur oxides reasonably.

The major characteristics of the transport pathways of sulfur oxides were as shown in the figures and explained in section III and IV. Especially in the analysis of the intensive observation of APEX, the LRTM revealed the origins of the sulfur oxides reached around Amami-Oshima on hourly basis reasonably to explain the observed concentrations. More detailed analyses for the episodes using these results will be continued.

Furthermore, the source-receptor analysis and the transport pathway analysis mentioned here are available operationally in quasi-real time by running the LRTM with meteorological data distributed online by several meteorological organizations. The results of daily calculation during the field campaigns and the current results of continuing operation of the LRTM (currently only concentration distributions are available) are exhibited at http://staff.aist.go.jp/takahisa.maeda/LRTM/ .

References

[1] R. L. Arndt, G. R. Carmichael, D. G. Streets, N. Bhatti, "Sulfur dioxide emissions and sectorial contributions to sulfur deposition in Asia", *Atmospheric Environment* Vol. 31, pp. 1553-1572, 1997.

[2] M. Huang, Z. Wang, D. He, H. Xu, L. Zhou, "Modeling Studies on Sulfur Deposition and Transport in East Asia", *Water, Air and Soil Pollution* Vol. 85, pp. 1921-1926, 1995.

[3] Y. Ikeda and H. Higashino, "The Estimation of Acid Deposition in East Asia – The model development and validation –", *Journal of Japan Society for Atmospheric Environment* Vol. 32, pp. 116-135, 1997 (in Japanese).

[4] T. Maeda, M. Hayashi, R. Imasu, Z. Wang, "A model analysis of transport of sulfur oxides in china and outflow to the sea", *Water Air and Soil Pollution* Vol. 130, pp. 355-360, 2001.

[5] T. Maeda, R. Imasu, H. Tsuruta, S.Yonemura, "Model analyses of distribution and transport paths of atmospheric sulfur oxides in East Asia during APEX-E1/E2 periods", unpublished.

[6] China Statistical Press, *China Statistical Yearbook 1999*, 2000 (in Chinese).

[7] W. Wang, *The source, deposition, transport, effects on ecology of acid species and its control in China, Report for the Eighth Five Year National Scientific and Technological*

Project, 1995 (in Chinese).
[8] H. Tsuruta, S. Yonemura, S. Sudo, N. Kaneyasu, T. Maeda, Y. Shirasuna, K. Hirano, K. Sera, S. Futatsugawa, "Characterization of atmospheric aerosols measured at Amami-Oshima, southwest of Japan, in April 2001 in the APEX field experiment", unpublished.
[9] J. S. Han, unpublished.