

# DISTRIBUTIONS OF SULFUR POLLUTANTS IN EAST ASIA

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**ABSTRACT:** A Eulerian sulfur deposition model is used to study the distributions of sulfur pollutants in East Asia, in which important physical and chemical processes such as transport, diffusion, dry and wet deposition, vertical transportation in cumulus cloud, gas phase chemistry, aqueous phase chemistry and heterogeneous aerosol chemistry are included. Two cases, the periods of 1987-07-02 GMT00- 1987-07-03 GMT00(case 1) and 1989-04-20 GMT00- 1989-04-21 GMT00(case 2), are studied and the distributions of SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> in East Asia are shown. It is revealed that the patterns of distributions of SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> are different under different meteorological conditions. In the upper layer, the concentrations of SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> are 0.4- 1.0 μg/m<sup>3</sup> and 0.2- 0.5 μg/m<sup>3</sup>, 4.0- 10.0 μg/m<sup>3</sup> and 2.0- 6.0 μg/m<sup>3</sup> in the middle layer. In the surface layer, the levels of SO<sub>2</sub> range from 4.0 to 30.0 μg/m<sup>3</sup> and from 2.0 to 8.0 μg/m<sup>3</sup> for SO<sub>4</sub><sup>2-</sup>. Distributions of SO<sub>x</sub> are controlled by emissions in the surface layer, while in the upper layer it is mainly affected by precipitation. The distributions of SO<sub>x</sub> show the interaction of emissions, winds and precipitation in the middle layer.

**KEY WORDS:** sulfur pollutants; deposition model; East Asia, sulfur distribution

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## 1 INTRODUCTION

It is very important to realize the distributions of pollutants before the pollution can be controlled successfully. Owing to the lack of systematical observations in a broad area, numerical simulation has become a powerful tool to understand the distributions of pollutants and their features. CARMICHAEL and PETERS (1984) have studied the transportation of SO<sub>x</sub> in the east part of America by a Surfer Transport Eulerian Model (STEM). The 24h-averaged concentrations of SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> were shown. WANG Zi-fa *et al.* (1996) has given the year-averaged distributions in 1989 of sulfur pollutants in East Asia using a sulfur deposition model. Few attempts have been made to study the day-averaged spatial distributions of sulfur pollutants. In this paper, an Eulerian sulfur

deposition model (GAO *et al.*, 1997a) is used to study two cases and realize the day-averaged distributions of SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup>.

## 2 MODEL DISCRIPOTION

The regional sulfur deposition model used for the present study has been described in detail in our previous paper (GAO *et al.*, 1997b). A brief description is shown here. Like other Eulerian models, this model is based on the solution of the mass conservation equation for a pollutant. The important physical and chemical processes included are transport, diffusion, dry and wet deposition, vertical transportation in cumulus cloud, gas phase chemistry, aqueous phase chemistry and heterogeneous aerosol chemistry. Transport and diffusion are calculated using temporally

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and spatially varying meteorology provided by the LASG-REM (State Key Laboratory of Numerical Modeling for Atmospheric Sciences and Geophysical Fluid Dynamics-Regional Eta( $\eta$ )-coordinate Model) (YU, 1989) meso-scale meteorological model developed in IAP (Institute of Atmospheric Physics, the Chinese Academy of Sciences). The necessities for the origins of LASG-REM are obtained from the datasets provided by ECMWF (Europe Center for Middle-range Weather Forecasts) by appropriate interpolation. A gas-phase chemical model including 31 species and 52 reactions is used to simulate chemical oxidation rate of  $\text{SO}_2$  (GAO, 1997b). The aqueous-phase chemical process includes sulfur oxidation by  $\text{H}_2\text{O}_2$  and  $\text{O}_3$ . Dry depositions of  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  are computed using a resistance model. Pollutants scavenging by clouds and rain are also included. A heterogeneous chemical model (GAO *et al.*, 1997b) is to simulate the chemical process on aerosol surface. The role of cumulus cloud in the vertical redistribution of species is described by parameterization. The spatial resolution in the horizontal is about 80 km. The vertical region from the Earth's surface to 100 hPa is divided into 8 sigma( $\sigma$ ) levels (0.0, 0.2, 0.4, 0.6, 0.78, 0.89, 0.96, 0.99) and the time step is 300 s.

The model area is from  $100^\circ\text{E}$  to  $145^\circ\text{E}$  in the longitude, and from  $16^\circ\text{N}$  to  $53^\circ\text{N}$  in the latitude, including most of China, Korea Peninsula, Japan, Hong Kong and part of Russian and Mongolia. The emission inventory for  $\text{SO}_2$  used in the regional sulfur deposition model is shown in Fig. 1 (KATO, 1992) and the emissions of  $\text{SO}_4^{2-}$  is assumed to be equal to 5% of  $\text{SO}_2$ .

### 3 ANALYSIS OF THE METEOROLOGICAL BACKGROUND OF CASES

In order to indicate the distributions of sulfur pollutants and their features in East Asia, two cases, the periods of 1987-07-02 GMT00 - 1987-07-03 GMT00 (case 1) and 1989-04-20 GMT00 - 1989-04-21 GMT00 (case 2), are selected to show the different patterns in different dates. The two cases represent the

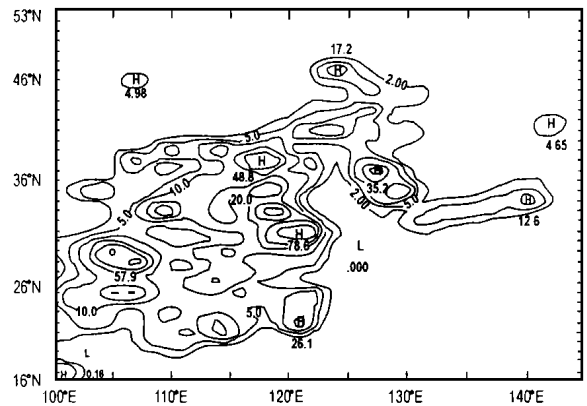


Fig. 1 Emission intensity of  $\text{SO}_2$  in East Asia ( $10^4 \mu\text{gm}^{-3} \text{s}^{-1}$ )

typical weather in summer and spring respectively.

On July 2, 1987, in the 500 hPa weather map, there is a trough located along the latitude of  $45^\circ\text{N}$  with a low center near  $120^\circ\text{E}$ . Contrast to it there is a cold front located in the northeast and north of China on the ground with precipitation maintained. Small waves come into being and move eastward in this layer, while in the 700 hPa and 850 hPa weather map, high-pressure centers stand in the area of the Qinghai-Xizang Plateau. From Jinan to Chengdu in the 700 hPa weather map and from Bengbu to Guiyang in the 850 hPa weather map, shear lines are located with short time precipitation maintained. From bottom layer to upper layer, the winds in the simulated areas are mainly westerly. In the south boundary, winds blow northward from the outside of model area. 24 h (from July 2, 1987, GMT00 to July 2, 1987, GMT00) integrated maximum precipitation cell is located at  $122^\circ\text{E}$ ,  $50^\circ\text{N}$  with the value of 25.7 mm. The precipitation areas include the northeast part of China, Korea and the north part of Japan. Another west-east rain band stretches from the South of China to the south of Japan, with the rainfall of 1.0-12.0 mm.

On April 20, 1989, it was a typical weather in spring with a little rain. The pattern of one trough and one ridge stand in the 500 hPa weather map in the Eurasian continent with a relatively stable weather trend. Small waves appear and move eastward in

the area of mid-latitude. In the 700 hPa and 850 hPa weather map, from the Qinghai-Xizang Plateau to Kinawa Island of Japan, there is a strong shear line. Therefore the winds blow from the Bohai sea and the Yellow Sea to the continent in the 700 hPa and 850 hPa weather map. Three 24 h (from April 20, 1989, GMT00 to April 21, 1989, GMT00) integrated maximum precipitation cells are located in northwest, north of China and the middle part of Japan with the values of 23.7, 6.5 and 3.1 mm.

#### 4 DAY-AVERAGED DISTRIBUTIONS OF $\text{SO}_2$ AND $\text{SO}_4^{2-}$

The two cases are simulated for 24 h respectively and the day-averaged concentrations of  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  are obtained by average the outputs of each hour from the sulfur deposition model. The second, fifth and eighth layers from the top are considered to be the upper, middle and surface layer respectively. Due to the  $\delta$  coordinate system the same layer is located on the different heights in different places. In plain areas, the average height of the three layers is about 30 m, 1000 m and 6000 m respectively.

Fig. 2 shows the distributions of  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  in the surface layer, middle layer and upper layer. In

the surface layer, the levels of  $\text{SO}_2$  are about 5.0–140.0  $\mu\text{g}/\text{m}^3$  with the maximum of 141.0  $\mu\text{g}/\text{m}^3$  near Beijing City. The north part of the east China, including Shanghai, and most part of the east China, including Beijing connect together show the big values of  $\text{SO}_2$ . A big value area lies in Sichuan basin with the biggest level of 47.3  $\mu\text{g}/\text{m}^3$  near Chongqing. Another several local big value areas are located in Taiwan, South Korea and middle part of Japan. There exists a big value center of  $\text{SO}_2$  with 9.28  $\mu\text{g}/\text{m}^3$  in size, corresponding to a big cell of emission at  $107^\circ\text{E}$ ,  $46^\circ\text{N}$ . Concentrations of  $\text{SO}_2$  range from 1.0  $\mu\text{g}/\text{m}^3$  to 17.8  $\mu\text{g}/\text{m}^3$  in the middle layer. There are five big level centers located in Changchun, Beijing, Xi'an, Taiwan and south Korea with the maximum value of 17.8  $\mu\text{g}/\text{m}^3$  near Changchun. A low-level area stretches from Hunan of China to Nakasaki of Japan. The values of  $\text{SO}_2$  in the upper layer are 0.1–0.9  $\mu\text{g}/\text{m}^3$ . There are two low-level areas. One is a southwest–northeast band with the minimum value of 0.155  $\mu\text{g}/\text{m}^3$  located at the same place with that in the middle layer, the other lies in the northeast part of China with a lowest level of 0.176  $\mu\text{g}/\text{m}^3$ . This kind of distributions is affected by wet scavenging of cloud and rain.

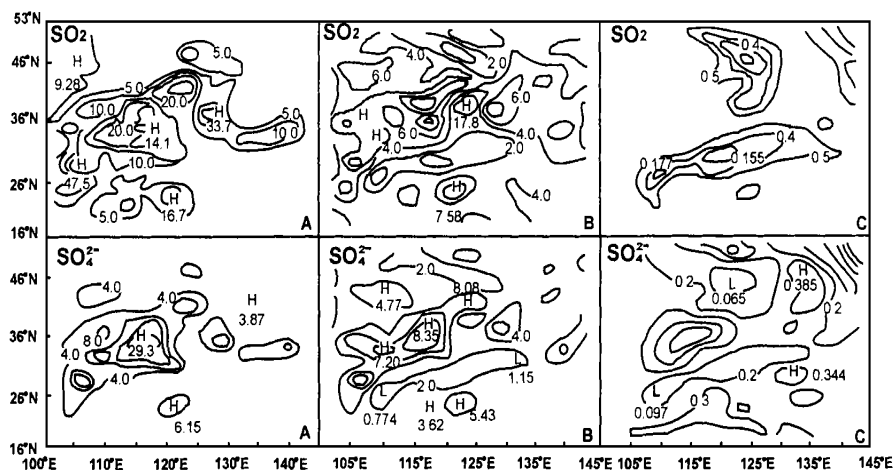


Fig. 2 Distributions of daily averaged  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  concentrations ( $\mu\text{g}/\text{m}^3$ ) (case 1)

A. Surface layer B. Middle layer C. Upper layer

In the surface layer, the levels of  $\text{SO}_4^{2-}$  are 1.0–29.0  $\mu\text{g}/\text{m}^3$ . The pattern of distribution is similar with that of  $\text{SO}_2$  with a maximum value of 29.3  $\mu\text{g}/\text{m}^3$  near Beijing. The distribution of  $\text{SO}_4^{2-}$  is similar with  $\text{SO}_2$  in the middle layer as well with the values of 1.0–8.0  $\mu\text{g}/\text{m}^3$ . The main high-level area and low-level area are nearly parallel like a band from southwest to northeast with the low-level area stretches from Hunan Province to Nakasaki of Japan and the high-level area from Chongqing to northeast part of China. In the upper layer the values of  $\text{SO}_4^{2-}$  range from 0.1  $\mu\text{g}/\text{m}^3$  to 0.5  $\text{g}/\text{m}^3$  and the pattern is a high-level area standing in the middle and two low-level areas lying in each side of north and south. The low-level area in the south is from Hunan Province to Nakasaki of Japan and the low-level area in the north lies in the northeast part of China. The high-level area stands in the north and northwest parts of China.

Fig. 3 shows the day-averaged concentrations of  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  of case 2. In the surface layer, the pattern of  $\text{SO}_2$  distribution shows several big value areas lying in southwest, east of China, south Korea and Japan with the maximum level of 64.2  $\text{g}/\text{m}^3$  in south Korea. The distribution of  $\text{SO}_2$  in the surface layer is very similar with that of emissions and the big value centers are located exactly at the places of big emission centers because of little influence of rain. Shanghai has the biggest level of emission and Chongqing follows. The levels of emission in the northeast of China and south Korea are not so big as those in Shanghai and Chongqing, but with the maximum concentrations of  $\text{SO}_2$  in the surface layer, which is probably because of the influences of wet scavenging and transport. In 500 hPa and 700 hPa weather map, there is a high-pressure system in the northeast part of China and south Korea. The pollutants can be transported to the northeast part of

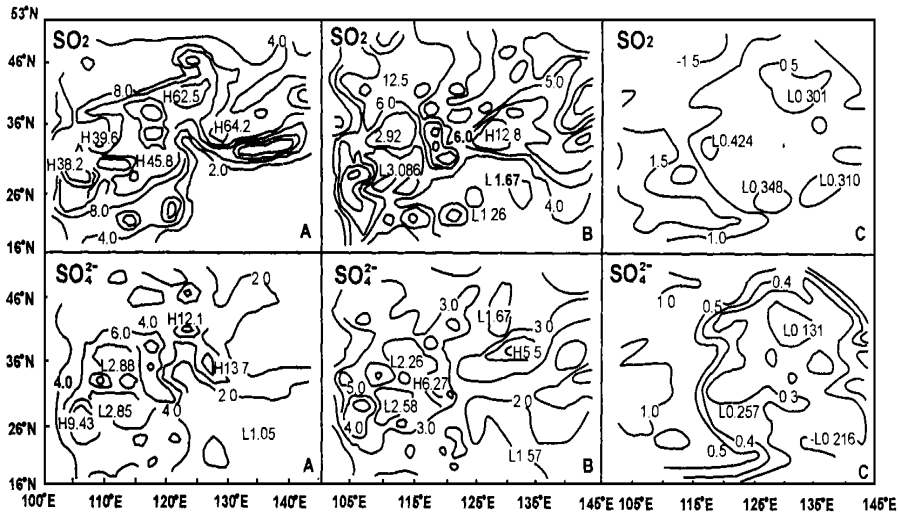


Fig. 3 Distribution of daily averaged  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  concentrations ( $\mu\text{g}/\text{m}^3$ ) (case 2)

A. Surface layer B. Middle layer C. Upper layer

China and south Korea along the north edge of the high-pressure system, which causes the high level of pollutants. The levels of  $\text{SO}_2$  are 1.0–12.5  $\mu\text{g}/\text{m}^3$  in the middle layer with the similar pattern in the surface layer. The concentrations are relatively big in most part of China with the biggest value of 12.8  $\mu\text{g}/\text{m}^3$  in

south Korea. In the upper layer, the levels of  $\text{SO}_2$  are 0.1–1.5  $\mu\text{g}/\text{m}^3$  with the biggest value of 1.92  $\mu\text{g}/\text{m}^3$  in the southwest and south parts of China. In this case, the values of  $\text{SO}_2$  are bigger than those in case 1, because of much less rain and cloud with less scavenging of pollutants.

In the surface layer, the levels of  $\text{SO}_4^{2-}$  are 1.0 – 13.0  $\mu\text{g}/\text{m}^3$ . The big value areas are located in southwest, middle, east, north of China and Korea Peninsula with the four biggest value centers lying in Chongqing, Shanghai, Changchun of China and south Korea. The maximum value is 13.7  $\mu\text{g}/\text{m}^3$  in south Korea and the second biggest value is 12.1  $\mu\text{g}/\text{m}^3$  near Changchun. In the south part of China, the concentrations are relatively low with 2.0– 4.0  $\mu\text{g}/\text{m}^3$  in values. The values of  $\text{SO}_4^{2-}$  range from 1.0 to 6.0  $\mu\text{g}/\text{m}^3$  in the middle layer. Comparing with those in the surface layer,  $\text{SO}_4^{2-}$  is distributed in a broad area with several big value centers located in Chongqing, Shanghai and over the sea to the east of Korea. The levels of  $\text{SO}_4^{2-}$  over the south China and its adjacent sea are relatively low. In the upper layer, the concentrations are 0.1– 1.0  $\mu\text{g}/\text{m}^3$  with the big value area located in the west part of model area and

the low-level areas in the middle and east parts. The levels of  $\text{SO}_4^{2-}$  are bigger than those in case 1 in the upper layer, because of the less wet scavenging.

## 5 SUMMARY AND DISCUSSION

It is indicated (Table 1) that the big value centers of  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  are located very near in the surface layers, while in the middle and upper layers, the locations are much difference. In different cases, the ratios of  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  concentrations show much difference in different layers with the ratio getting bigger down to the low layers. It is revealed that emission controls the levels of pollutants in the surface layer and in the middle and upper layer, the concentration of  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  lie on the interactions of emission, meteorological conditions, all kinds of chemical processes and scavenging.

Table 1 Characteristics of  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  distributions under different meteorological conditions

|        | Surface layer                             |                                  | Middle layer                              |                                  | Upper layer                               |                                  |             |
|--------|---|----------------------------------|---|----------------------------------|---|----------------------------------|-------------|
|        | Maximum value( $\mu\text{g}/\text{m}^3$ ) | Location of maximum value center | Maximum value( $\mu\text{g}/\text{m}^3$ ) | Location of maximum value center | Maximum value( $\mu\text{g}/\text{m}^3$ ) | Location of maximum value center |             |
| Case 1 | $\text{SO}_2$                             | 141.0                            | 118°E, 37°N                               | 17.8                             | 117°E, 43°N                               | 0.91                             | 134°E, 43°N |
|        | $\text{SO}_4^{2-}$                        | 29.3                             | 117°E, 37°N                               | 8.35                             | 118°E, 37°N                               | 0.5                              | 115°E, 37°N |
|        | $\text{SO}_2/\text{SO}_4^{2-}$            | 4.81                             |   | 2.13                             |   | 1.82                             |             |
| Case 2 | $\text{SO}_2$                             | 64.2                             | 129°E, 37°N                               | 12.8                             | 127°E, 39°N                               | 1.92                             | 117°E, 26°N |
|        | $\text{SO}_4^{2-}$                        | 13.7                             | 130°E, 37°N                               | 6.27                             | 114°E, 35°N                               | 1.0                              |             |
|        | $\text{SO}_2/\text{SO}_4^{2-}$            | 4.69                             |   | 2.04                             |   | 1.92                             |             |

In the two cases, the patterns of distributions of  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  in the surface layer are not very similar and the locations of maximum value centers of the two pollutants are not exactly the same too. Comparing with the pattern of emission, the big value centers are located at the big level centers of emission or nearby, which infers that the important effect of emission on the concentrations of  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  in the surface further. The differences of the locations of maximum value centers in the two cases show that meteorological condition is another important factor to affect the distribution pattern of pollutants. The meteorological influences on the distribution of pollutants, including

scavenging by rain, transport by wind, dry deposition and chemical processes, are very complicated. So it is not easy to give a simple explanation on the different distributions of  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  in the surface layer. The distributions of  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  in the upper layer are strongly affected by scavenging process. The areas with low pollutant level are not exactly located at the same place of the precipitation areas but nearby, which is because the scavenging of pollutants is not only controlled by precipitation but affected by cloud as well. In the middle layer, the distributions of  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  are the results of the interactions of emission, wind in this layer and wet scavenging.

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