TAO, Vol.6, No.3, 403-408, September 1995

# High Concentration of SO<sub>2</sub> Observed Over the Sea of Japan

SHIRO HATAKEYAMA<sup>1</sup>, KENTARO MURANO<sup>2</sup>, HIROSHI BANDOW<sup>2</sup>, HITOSHI MUKAI<sup>3</sup> and HAJIME AKIMOTO<sup>3</sup>

(Manuscript received 7 July 1994, in final form 30 November 1994)

### ABSTRACT

An aircraft observation of atmospheric pollutants was made over the western part of the Sea of Japan based on the IGAC/APARE/PEACAMPOT Program on Nov. 11 and 12, 1992. High concentrations of  $SO_2$  and NOx were observed, and their distribution showed a clear dependence on wind direction. When wind blew from the west or north west, a very high concentration of  $SO_2$  (up to 10 ppb) was observed. The result of back trajectory analysis points to the contribution from industrialized areas of Korea.

(Key words: IGAC, The Sea of Japan, SO<sub>2</sub>, Back trajectory analysis, East Asia)

#### **1. INTRODUCTION**

The east Asian area is one of the largest regions for the anthropogenic emission of  $NO_x$ 

and SO<sub>2</sub> (Rodhe, 1989). In fact, because of population growth and the rapid development of industrial activity, it has been projected that this area will become the largest source of NO<sub>x</sub> and SO<sub>2</sub> in the world in the 21st century (Galloway, 1989). It is important to analyze the present status of air pollution over the East Asian region, to evaluate the amount of the anthropogenic emission, to accurately predict the future situation by the use of computer models and to apply these results to work out countermeasures. However, there is no intensive measurement of atmospheric pollutants such as SO<sub>2</sub>, NO<sub>x</sub>, and O<sub>3</sub> in this area. For the purpose of clarifying the present status of atmospheric pollution in the Pacific rim region of East Asia as well as of predicting the future situation in this area, the authors have been conducting intensive field study named the IGAC/APARE/PEACAMPOT Survey since 1991, and a detailed emission inventory of SO<sub>2</sub> and NO<sub>x</sub> has also been made (Kato and Akimoto, 1992; Akimoto and Narita, 1994). From November 8-12, 1992 the authors surveyed above the East China Sea as well as the Sea of Japan at 4 different altitudes. In this paper a high

#### <sup>3</sup> Research Center for Advanced Science and Technology, University of Tokyo, Japan



<sup>&</sup>lt;sup>1</sup> Global Environment Division, National Institute for Environmental Studies, P.O. Tsukuba, Ibaraki 305, Japan

<sup>&</sup>lt;sup>2</sup> Faculty of Engineering, University of Osaka Prefecture, Japan

#### **404**

#### TAO, Vol.6, No.3, September 1995

concentration of  $SO_2$  and  $NO_x$  observed over the Sea of Japan between Korea and the Oki Islands, Japan are reported.

#### 2. METHODS

Measurements of atmospheric pollutants were made from an aircraft on November 8, 10, 11, and 12, 1992. The aircraft employed was a turboprop twin-engined Fairchild Swearingen SA226-AT (Merlin-IV, owned by Showa Aviation Co., Ltd.). Flight areas were south of Cheju Island (Korea) over the East China Sea between 32°15'N, 127°E (Point C) and 31°5'N, 126°20'E (Point D) and west of the Oki Islands over the Sea of Japan between 35°20'N, 132°30'E (Point B) and 36°40'N, 132°30'E (Point A). Thirty-minute level flights were made at each altitude of about 10,000, 7,000, 3,500 and 1,500 feet in one cruise.

Ambient air was introduced into the airplane through a 3/8" Teflon tube which was connected to the air inlet settled on a window ahead of the air intake of the engine. The other end of the tube was connected to a glass manifold through which each instrument sampled air.

Ozone was monitored with a TECO Model 49 UV absorption ozone analyzer with a 4-sec switching of the light pass for its dual cell system. Pressure and temperature were automatically corrected. Calibration was made against a standard ozone supplier (TECO Model 49PS owned by the National Center of Atmospheric Research, U.S.A.).

Measurements of nitrogen oxides were made with an ozone-chemiluminescence NO- $NO_x$  analyzer (TECO, Model 42S). A commercial analyzer was modified for the aircraft measurement and for the improvement of its detection limit. Major modifications consisted of (1) the use of flow-controlled air sampling with a thermal mass-flow-meter system, which improved the pumping ability so as to reduce pressure in its chemiluminescence chamber, and (2) the use of pure oxygen to make ozone. In this analyzer, a molybdenum converter operating at 320°C was used to convert nitrogen oxides to NO. The conversion efficiency for nitric acid and some organic nitrates has not been established, but it may be less than unity. Thus, the nitrogen species detected as NO<sub>x</sub> with this analyzer include some part of NO<sub>y</sub>, which hereafter is dealt with as  $NO_x^*$ . In this measurement, a two-minute running mean (four intermittent measurements for one species) as an averaged value of NO and NO<sub>x</sub> \* was obtained. This gave a detection limit of 25 ppt with a S/N ratio of one. SO<sub>2</sub> was measured with a TECO Model 43S pulse fluorescence SO<sub>2</sub> monitor. The outer temperature and humidity were measured with a Vaisala HMP133Y thermometer-hygrometer. Isobaric back-trajectories were calculated for the 800, 850, 900, 950, and 1,000 hPa pressure levels, based on the program by Hayashida-Amano et al. (1991). The global wind field data compiled and analyzed by the Japan Meteorological Agency and available for 0000 and 1200 UT over a 1.825° mesh at each level were used as input.

#### **3. RESULTS AND DISCUSSION**

The weather was generally mild on both the 11th and 12th of November, 1992. The western part of the Sea of Japan was covered with high pressure originating from China. However, cloud was present between 3,500 and 7,000 feet on both days. Thus, it can be said that the upper layer ( $\geq$ 7,000 feet) was clearly separated from the lower layer ( $\leq$  3,500 feet)

## on those days.

#### Hatakeyama et al. 405

Figure 1 shows the flight courses of this mission. In this paper, only the results obtained on the course over the western part of the Sea of Japan is discussed, i.e., north of Honshu and west of the Oki Islands between Points B and A as depicted in Figure 1. Figures 2 and 3 show the concentration of  $SO_2$ ,  $NO_x$  and  $O_3$  along with flight altitude. Since the weather conditions were as mentioned above, the air covering this area seemed very uniform. The concentration of ozone did not show clear dependence on the altitude although the average concentration of the upper layer (average 46.9 and 45.2 ppb for Nov. 11 and 12, respectively) was, as expected, higher than that of the lower layer (average 43.5 and 43.3 ppb for Nov. 11 and 12, respectively). The profile of the temperature and humidity (Figures 4 and 5) also supports this uniformity.

On the other hand, the concentration profile of  $SO_2$  and  $NO_x$  showed a remarkable feature. At the higher altitudes of 7,000 and 10,000 feet,  $SO_2$  and  $NO_x$  were both very low. To illustrate,  $SO_2$  had an average of 0.24 and 0.18 ppb on Nov. 11 and 12, respectively, while  $NO_x$  had an average of 0.21 and 0.18 ppb, respectively. At the lower altitudes of 3,500 and 1,500 feet, average  $SO_2$  and  $NO_x$  concentrations were 1.78 and 1.82 ppb, respectively, on Nov. 11 and 2.62 and 2.71 ppb, respectively, on Nov. 12.

Moreover, a very clear difference in the profile of SO<sub>2</sub> concentration was observed depending on wind direction, particularly at the lower altitudes. On Nov. 11, a south wind blew. A high concentration of SO<sub>2</sub> up to 6 ppb was found near Point B at lower altitudes (3,500 and 1,500 feet) as shown in Figure 2. A clear symmetrical pattern was observed in the concentration profile of SO<sub>2</sub> and NO<sub>x</sub>, having A as a mirror surface. Since the peaks of SO<sub>2</sub> in Figure 2 are very narrow, this can be considered to be the result of local emission in Japan. In contrast, a very high and broad peak of SO<sub>2</sub> was observed on Nov. 12 as shown in Figure 3 around Point A with a maximum concentration of 9.61 ppb. Here the peak was also symmetrical. There is no local source around Point A. Wind blew from west or northwest clearly indicating the transport of pollutants from the East Asian continent including the Versen Depincula

Korean Peninsula.



Fig. 1. Flight courses of the survey.

**406** 

#### TAO, Vol.6, No.3, September 1995



Fig. 2. Concentration profiles of  $SO_2$ ,  $NO_x$  and ozone on Nov. 11, 1992. A and B indicate Point A and Point B in Figure 1.

![](_page_3_Figure_4.jpeg)

# Fig. 3. Concentration profiles of $SO_2$ , $NO_x$ and ozone on Nov. 12, 1992. A and B indicate Point A and Point B in Figure 1.

This contention was supported by the calculation of back trajectory. Figure 6 shows the back trajectory of the air mass reaching Point A on Nov. 12. The starting point of the calculation was Nov. 9. Trajectories calculated for all the altitudes pass through China and/or Korea. The concentration of  $SO_2$  at higher altitudes was very low, and the high concentration of  $SO_2$  was only observed at lower altitudes. This suggests that the impact of the emission in Korea was larger in this case. Highly industrialized areas in Korea are in the upstream of

![](_page_3_Figure_7.jpeg)

![](_page_4_Figure_0.jpeg)

Fig. 4. Profiles of temperature and relative humidity on Nov. 11, 1992.

1-20

![](_page_4_Figure_2.jpeg)

![](_page_4_Figure_3.jpeg)

Fig. 5. Profiles of temperature and relative humidity on Nov. 12, 1992.

should also suggest a short distance transport from Korea since the lifetime of  $NO_x$  is short. However, the  $NO_x/SO_2$  ratio at Point A and that at Point B were very different as can be clearly seen in Figure 3, indicating that the origins of the air mass around both points were

different.

**408** 

TAO, Vol.6, No.3, September 1995

![](_page_5_Figure_2.jpeg)

Fig. 6 Back trajectory of air mass reaching Point A at 11:10 am on Nov. 12, 1992. (1) 800 hPa, (2) 850 hPa, (3) 900 hPa, (4) 950 hPa and (5) 1,000 hPa.

Acknowledgments The authors are grateful to Drs. I. Watanabe of the National Institute of Public Health, S. Tanaka of Keio University and M. Yamato of Gunma University who participated in this survey mission. S.H. is also grateful to Drs. K. Takeuchi and N. Kaneyasu of the National Institute of Resources and Environment for their help in calibrating the  $SO_2$  analyzer.

This study was performed based on the Global Environment Research Program Budget of the Environment Agency of Japan.

#### REFERENCES

- Akimoto, H., and H. Narita, 1994: Distribution of  $SO_2$ ,  $NO_x$  and  $CO_2$  emissions from fuel combustion and industrial activities in Asia with  $1^{\circ} \times 1^{\circ}$  resolution. Atmos. Environ., **28A**, 213-225.
- Galloway, J. N., 1989: Atmospheric acidification: projections for the future. Ambio, 18, 161-166.
- Hayashida-Amano, S., Y. Sasano, and Y. Iikura, 1991: Volcanic disturbance in the stratospheric aerosol layer over Tsukuba, Japan, observed by the National Institute for Environmental Studies of Lidar from 1982 through 1986. J. Geophys. Res., 96, 15,469-15,478.
- Kato, N, and H. Akimoto, 1992: Anthropogenic emissions of  $SO_2$  and  $NO_x$  in Asia: emission inventories. Atmos. Environ., 26A, 2297-3017.
- Rohde, H., 1989: Acidification in a global perspective. Ambio, 18, 155-160.