

## **C-1.2.2 Budget Study of Environment Acidifying Components with International Ground-Based Observation**

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**Total Budget for FY1996-FY1998** 34,841,000 Yen (FY1998; 11,472,000 Yen)

**Abstract** A large amount of SO<sub>2</sub> and NO<sub>x</sub> which were the precursor of acid rain is emitted in East Asia followed by European continent and Northern America continent. Transboundary air pollution is one of the most important environmental issues in East Asia. In order to clarify the transboundary air pollution to Japan from Asian continent, we conducted an intensive field survey of air pollutants at remote island in East China Sea and semi-urban sampling point in winter season, January and December 1997. Occasional high concentration of nss-SO<sub>4</sub><sup>2-</sup> were observed in both sampling sites indicating the air pollutants covering larger than 200 km, for the two sampling points are separated around 190 km. From the ion balance of chemical component of collected aerosol, under transboundary air pollution, unneutralized sulfate contributed to the acidity of aerosol.

**Key Words** East Asia, Budget Study, Ground-Based Observation, Remote Island

### 1. Introduction

A large amount of SO<sub>2</sub> and NO<sub>x</sub> which were the precursor of acid rain is emitted in East Asia followed by European continent and Northern America continent. It was forecasted that the emission of SO<sub>2</sub> and NO<sub>x</sub> may increase in the near future by the development of industry and increment of population. Therefore, the deposition of acidic component will increase in East Asia. It is anxious that the damage by acid rain expand in East Asia including Japan. In order to understand the transboundary air pollution to Japan from Asian continent, we conducted an intensive field survey of air pollutants at remote island in East China Sea and semi-urban sampling point in winter season. Northwestern air flow prevails in the winter time to increase the possibility of transboundary air pollution at both sampling sites. We observed transboundary air pollution at both sampling sites, with air pollutants observation, the construction of air flow fields and isentropic backward trajectory analysis.

### 2. Research Objectives

To clarify the transboundary air pollution from the Asian continent to Japan, we conducted ground-based observations. From the air pollutant monitoring at less polluted remote island(Goto Is.: G) and semi-urban point(Fukuoka Prefecture, Dazaifu city: F), we can learn the behavior of air pollutants and can detect the transboundary air pollution from Asian continent to Japan.

### 3. Research Method

#### (1) Sampling sites

Two field sampling sites were surveyed intensively, on the roof of the building of the Fukuoka Institute of Health and Environmental Sciences in Fukuoka Prefecture (hereafter termed F), which is located in a suburban area and a low-pollution site (National Acid Rain Monitoring Station) established on Goto Island (hereafter termed G), a remote island in the East China Sea. The sampling sites are about 190 km apart. The filter-pack method was used to collect gas and aerosol samples. The sampling interval was 6 h at G and 12 h at F, from 7 to 26 January 1997 (Phase I) and 6h at G and F from 2 to 19 December 1997(Phase II). The sampling sites are illustrated in Fig. 1.

#### (2) Gas and Aerosol Sampling and Chemical analysis

In order to monitor the variation in concentration of atmospheric pollutants, a filter-pack method was applied. Ambient aerosols were collected on a polytetrafluoroethylene (PTFE) filter. After removal of particulate matter, SO<sub>2</sub> and NH<sub>3</sub> were trapped on a cellulose filter (TOYO No. 51A) impregnated with 6% potassium carbonate solution in water, and a glass-fiber filter impregnated with 5% phosphoric acid in ethanol solution, respectively. Total nitrate (gaseous + particulate = t-NO<sub>3</sub>) was collected on a nylon filter (Pall, pore size 0.45 μm). All the filters used were 47 mm in diameter. The collected particulate matter and gases were extracted with solvent and analysed

#### (3) Wind field and backward trajectory analysis

Wind fields were modeled from ECMWF data with the Support System for Evaluation of Troposphere Monitoring Data of CGER(NIES). 3-Day isentropic backward trajectory analysis at 850 hPa was calculated with the method developed by Hayashida et al.(S. Hayashida-Amano et al., 1991).

#### (4) Data treatment

SO<sub>2</sub> and NH<sub>3</sub> concentration divided by 32 and 17, respectively, were considered to be equivalent concentrations.

### 4. Results and Discussion

Average concentrations of nss-SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>(p) and related chemical components at F and G, as well as the ratio of concentrations at F and G and the correlation coefficients of chemical component concentrations at the two sampling sites during the intensive field surveys are given in Table 1.

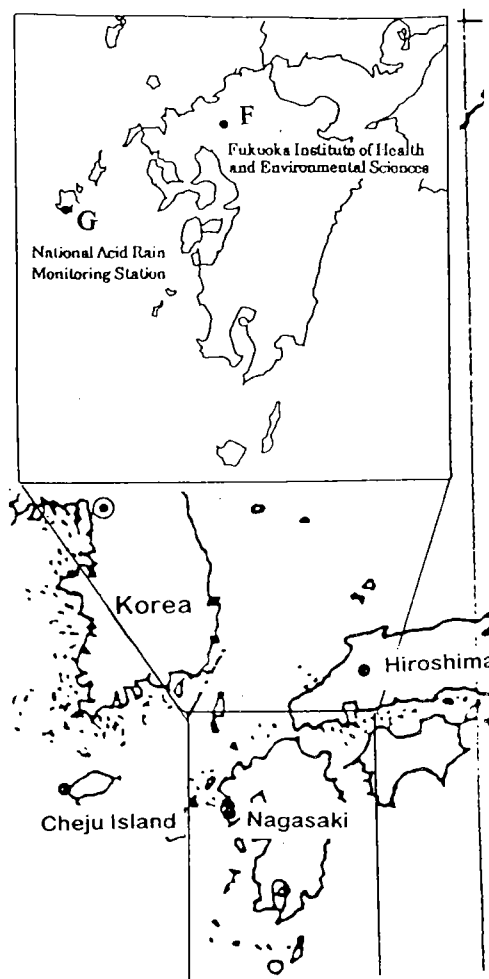


Fig. 1 Sampling sites for intensive field survey

Table 1 Average concentrations of nss-SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup>(p) at F and G, the ratio of concentrations at F and G, and correlation coefficients of chemical component concentrations at the two sampling sites during the intensive field surveys

	Concentration(neq m <sup>-3</sup> )		Ratio <sup>a</sup>	Correlation <sup>b</sup> coefficient
	Fukuoka	Goto		
nss-SO <sub>4</sub> <sup>2-</sup>	133	86.9	1.53	0.63
NO <sub>3</sub> <sup>-</sup>	96.5	26.5	3.65	0.30
NH <sub>4</sub> <sup>+</sup>	253	71.1	3.55	0.48
SO <sub>2</sub>	619	129	4.78	-0.07
HNO <sub>3</sub>	4.1	26.2	0.16	0.16
NH <sub>3</sub>	211	18.8 <sup>c</sup>	-	-

<sup>a</sup>Concentrations ratios at Fukuoka to those at Goto.

<sup>b</sup>Correlation coefficient of the concentrations between at Fukuoka and Goto.

<sup>c</sup>Measured in December 1997.

Variations of nss-SO<sub>4</sub><sup>2-</sup> concentration in aerosols at F and G during 7 and 26 January are shown in Fig. 2. The concentrations at the two sites were well correlated (correlation coefficient, 0.63). However, the average concentration at F was approximately 1.5 times higher than that at G, indicating that some urban air pollution reached F. Severe air pollution episodes were observed on 11, 14, 18, and 20-21 January. The wind field in the East Asian region was governed by a high pressure system on the Asian continent, and strong northwesterly air flow was observed on 11 January as shown in Fig. 3. On 22 and 23 January, in the absence of air pollution, the concentrations were similar to those of obtained from remote island sampling sites scattered over the Pacific Ocean( D. L. Savoie and J. M. Prospero, 1989).

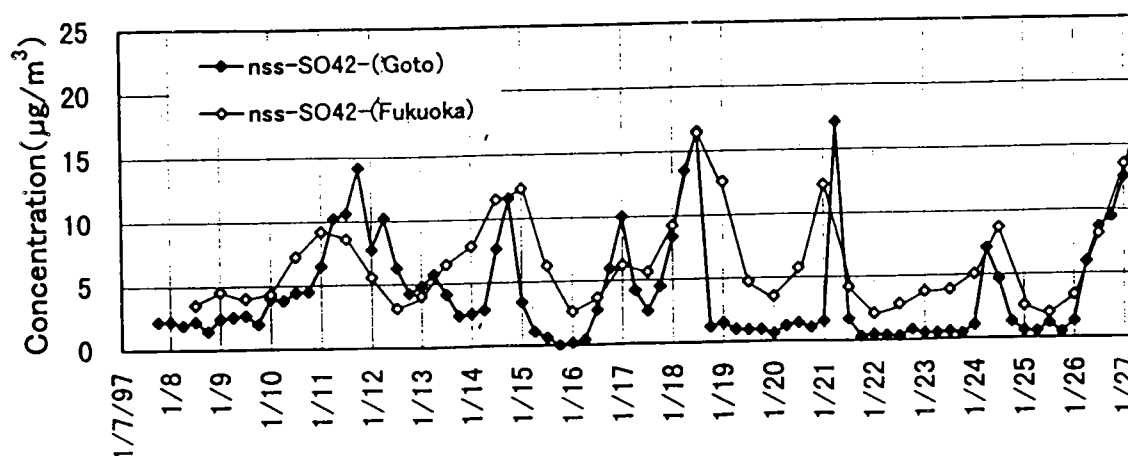


Fig. 2 Daily variations in nss-SO<sub>4</sub><sup>2-</sup> concentrations at G and F

Mean NH<sub>4</sub><sup>+</sup>(p) concentrations were 4.6 and 1.3 µg m<sup>-3</sup> at F and G, respectively. High concentration peaks were well correlated with nss-SO<sub>4</sub><sup>2-</sup> concentration variations at F and G, although the NH<sub>4</sub><sup>+</sup>(p) concentrations were very low at G. The average NH<sub>4</sub><sup>+</sup>(p) concentration of 1.28 µg m<sup>-3</sup> during the research period was similar to the mean NH<sub>4</sub><sup>+</sup>(p) concentration of approximately 1 µg m<sup>-3</sup> (January, 1993, 1994 and 1995) obtained at Kosan on Cheju Island (Li-Ling Chen et al., 1997), which is about 50 km north-northwest of G. The highest concentration was observed on 14 January at F, 12 µg m<sup>-3</sup>. From the variations of NH<sub>3</sub> concentration at F and G in phase II of the field survey, at G, there was no clear pattern, because the concentrations were very low. G is surrounded by the sea; thus the total NH<sub>4</sub><sup>+</sup>

ECMWF(2.5) Wind Field  
 Geopotential Height  
 Date : 97.01.11.12 Pressure : 850.0hPa

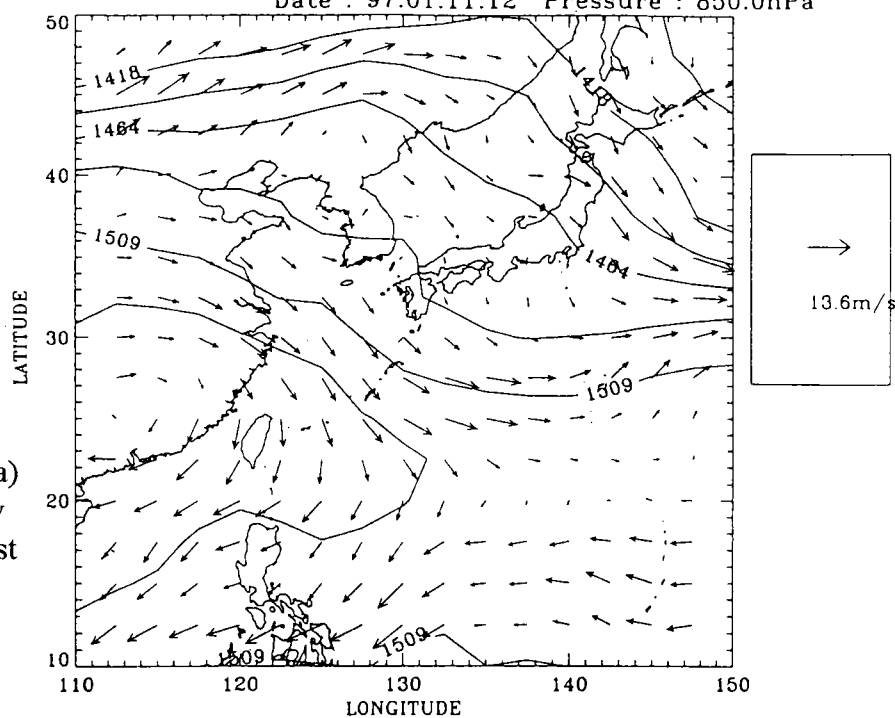
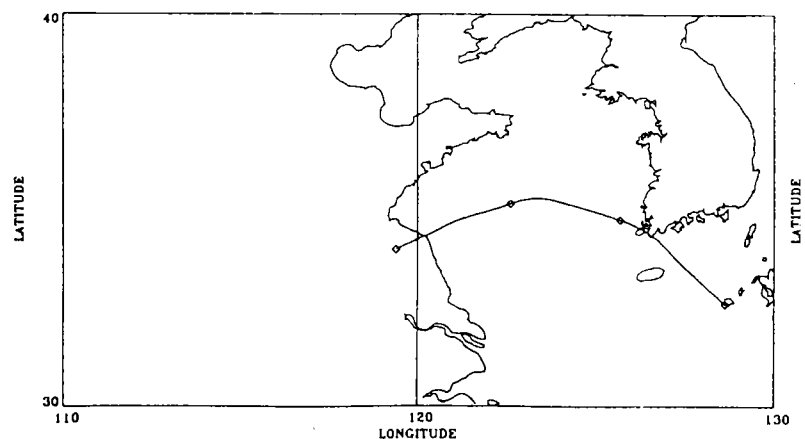


Fig. 3 Wind field (850 hPa) and backward trajectory analysis (from G) in East Asia: Every dot means 24 hrs interval



concentration is very low and most of the total  $\text{NH}_4^+$  is used to neutralize  $\text{H}_2\text{SO}_4$  as well as  $\text{HNO}_3$ . As a result,  $\text{NH}_4^+(\text{p})$  concentrations were higher than those of  $\text{NH}_3$ . If we consider the situation from 19 to 26 January, the concentrations of  $\text{nss-SO}_4^{2-}$  and  $\text{NH}_4^+(\text{p})$  were normally very low. However, there were occasional peaks indicating long-range transport of air pollutants from the Asian continent, based on data on the wind field in East Asian region and backward trajectory analysis (Fig. 3). The air mass originated at 1200 h on 8 January on the seashore area of China, traveled over the Yellow Sea, and reached G at 1200 h on 11 January.

The relative distribution of ionic species in aerosols during the research period is shown in Fig. 4.  $\text{NH}_4^+(\text{p})$  was the major cation contributor at both sampling sites. The contributions

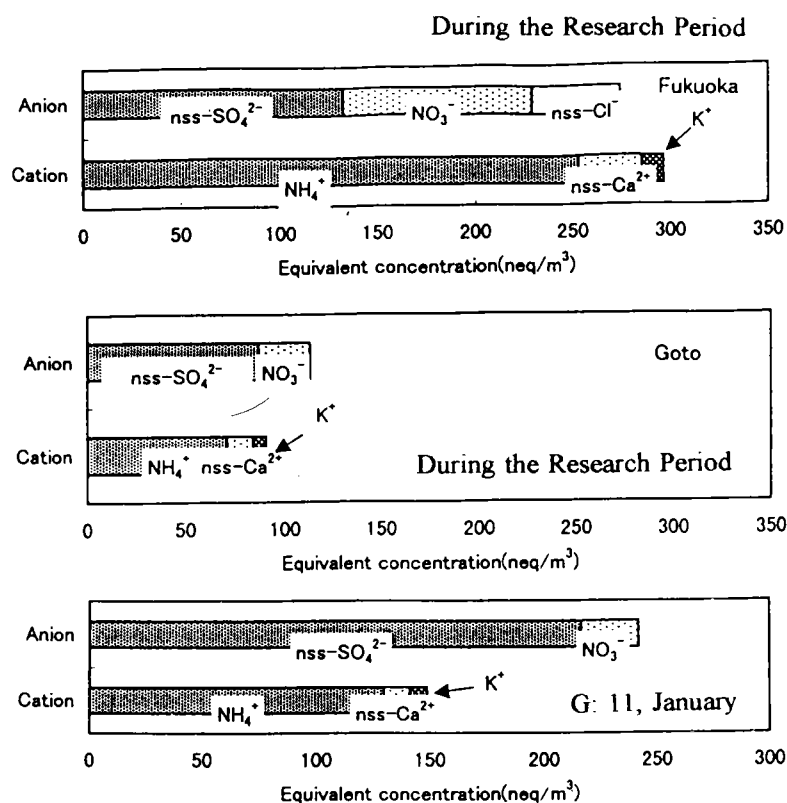


Fig. 4 Averaged relative distribution of each ionic component in aerosols at F and G during the research period, and relative distribution of each ionic component in the aerosol at G, 11 January 1997

of anions differed significantly between the two sites. In G, the contribution of  $\text{nss-SO}_4^{2-}$  was very large compared to that of other anion species. However, in F, the contribution of  $\text{NO}_3^-$  was about equal to that of  $\text{nss-SO}_4^{2-}$  and there was a significant contribution of  $\text{Cl}^-$ . F is located in a semi-urban area and emission of HCl from incinerators is a source of  $\text{Cl}^-$ .

On 11 January, we observed a high concentration of air pollutants at both sampling sites. The relative distribution of ionic species on that date is shown in Fig. 4. The major inorganic ions in the aerosol were  $\text{nss-SO}_4^{2-}$  and  $\text{NH}_4^+(\text{p})$ . However, the imbalance between cations and anions (anion rich) implied the existence of  $\text{H}^+$ , such as in  $\text{NH}_4\text{HSO}_4$ . As Fig. 3 shows, long-range transport of air pollutants was expected on 11 January, 1997. The outflow from the Asian continent to the Pacific Ocean consists mainly of  $\text{nss-SO}_4^{2-}$ ,  $\text{NH}_4^+(\text{p})$  and  $\text{H}^+$ .

## 5. Acknowledgement

Winds fields were calculated with the Support System of CGER(NIES).

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