C-1.2.1 International Cooperative Observations Using Aircrafts and Ships on the Balance of Materials Acidifying the Environment

Contact Person Shiro Hatakeyama
Section Chief
Atmospheric Environment Division
National Institute for Environmental Studies
Environment Agency
16-2, Onogawa, Tsukuba, Ibaraki 305-0053, Japan
Phone +81-298-50-2502, Fax +81-298-50-2579
E-mail hatashir@nies.go.jp

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Abstract International cooperative observations were agreed among Japan (NIES), Korea (NIER, KIST) and China (Peking Univ.). According to this agreement Aircraft observations and ground-based observations of atmospheric pollutants were performed over the East China Sea (FY1996: Jan. 6-25, 1997, FY1997: Dec. 2-19, 1997, and FY1998: Feb.2-6, 1999). In Jan. 1997 experiment, it was found that concentrations of SO_2 and trace acidic gases were high when northwest wind prevailed. In Dec., 1997 experiment, SO_2 was generally low. It may have been caused by clouds over observation area. Very high concentrations of pollutants were observed in Feb. 2, 1999 flight. SO_2 higher than 10 ppb was observed. It seemed that the cyclone, which had moved from Taiwan to northeast of Japan along the Honshu Island, brought about such high concentration of pollutants. The observation supported the results of model simulation well.

Key Words Cooperative observations, SO_2, NOy, Trace acidic gases, Northwest Pacific Ocean

1. Introduction

The East Asian area is one of the largest regions for the anthropogenic emission of NOx and SO_2.[1] 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 NOx and SO_2 in the world in the 21st century.[2] 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 study how to cope with the situation. In our previous project carried out from FY 1993 to FY1995 we performed aircraft observations over the seas between Japan and the Asian continent. We found that the vertical distribution of pollutants reflected characteristics intermediate between those of the continental polluted atmosphere and the marine background atmosphere. It was also found that air masses transported from the Asian continent to Japan can be divided into 3 types;[3] (a) China--Yellow Sea--Kyushu: low SO_2 and high SO_4^{2-}/SO_2 ratio, (b) Korea--western Japan: high SO_2 and low SO_4^{2-}/SO_2 ratio, and (c) North Asia--eastern Japan: low SO_2 and low SO_4^{2-}. In China 80% of energy is still produced by coal combustion. Thus, it may take some more time to reduce the emission of sulfur dioxide. Moreover, emission of NOx is growing rapidly. We need to pay attention to not only sulfur oxides but also nitrogen oxides.

2. Research Objectives

Model simulations studied also in the this project suggested that a large polluted air mass comes to Japan from central/southern China when a cyclone passes by south of Japanese main islands. It can be expected that the most typical pollution pattern transported from central/southern China appears in the East China Sea. Thus, the main objective of this three years project was established to see the status of atmospheric environment over the East Chinas Sea. For this purpose it is necessary to cooperate with scientists of Korea and China to cover wide area. Scientists in Kist (Korean Institute of Science and Technology) and Peking University agreed to the collaboration with us.

3. Research Method

Flight plans are shown in Fig. 1. Namely, between west of Fukue and south of Cheju. Nagasaki-Omura airport was the base for all the experiments for three years.

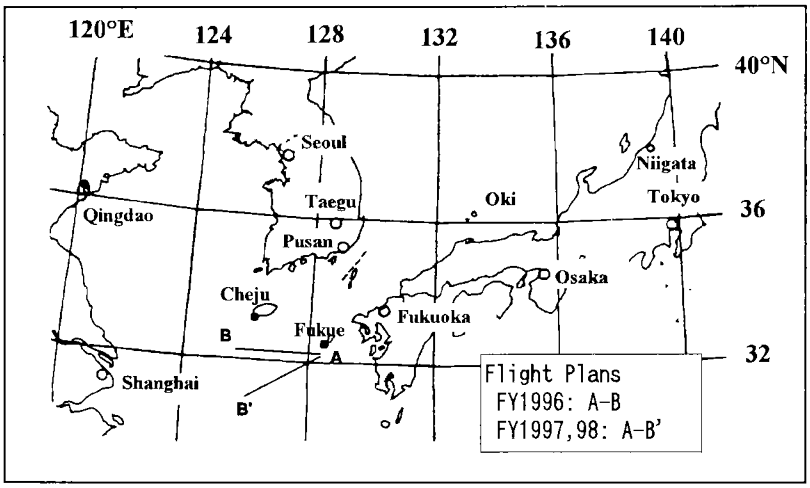

Fig. 1: Flight plans for aircraft observations in the project

(1) FY1996: Observation flights in FY1996 were made on January 11 and 13, 1997 between A and B in Fig 1. One hour level flights at 800-1000 m and ~300 m, respectively, were performed.

(2) FY1997: Observations were made on December 9, 12, and 13, 1997 between A and B' in Fig. 1. One hour level flights at ~2800 m and 500 m on Dec. 9, ~1000 m and 500 m on Dec. 12 and 13.

(3) FY1998: Observations were made on February 2, 4, and 6, 1999 between A and B' in Fig. 1. Flight altitude (mainly ~500 m and 2500 m) was variable on Feb. 2 because of very strong wind and the presence of clouds. On Feb 4 and 6, one hour level flights at ~2500 m

and 500 m were made.

Pollutants measured on board or collected are listed in Table 1. The aircraft employed was a reciprocal twin-engined airplane, Cessna 404, chartered from Showa Aviation Co., Ltd.,

Table 1: Pollutants measured on board and experimental techniques

pollutant	experimental technique
SO_2	pulse fluorescence
Ozone	UV absorption
NOx	Ozone chemiluminescence
PAN	Collection on a low temp. column
HC	Grab sampling
Aerosols	High volume sampler
Acidic gases	Diffusion scrubber

Japan. Outside air was introduced into the airplane through a 3/8" Teflon tube running from the nose of the airplane to a glass manifold located in the middle of the cabin. The other end of the tube was connected to a glass manifold through which each instrument sampled air. For the high volume sampler, a 1" stainless steel tube was also set on the co-pilot's window. Ozone was monitored with TECO Model 49 UV absorption ozone analyzer with 4 sec switching of the light pass for its dual cell system. Pressure and temperature were automatically corrected. SO_2 was measured with TECO Model 43S pulse fluorescence SO_2 monitor. Outer temperature and humidity were measured by use of a Vaisala HMP133Y thermometer-hygrometer. Measurement techniques of NOx and acidic gases will be described later.

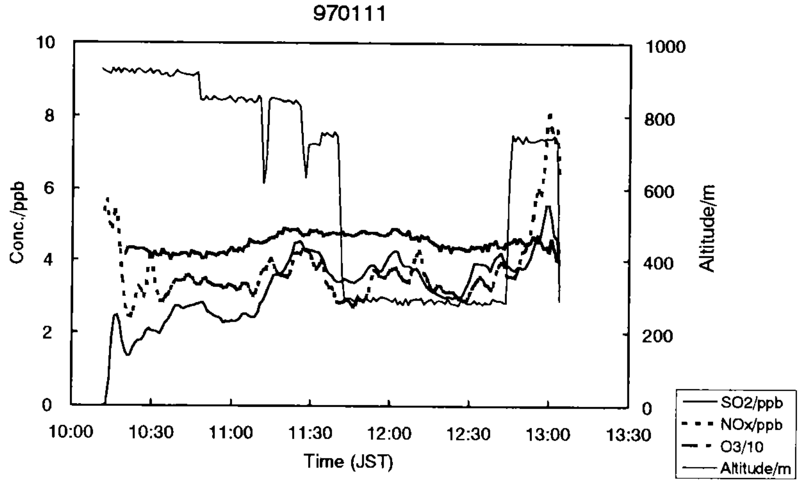

Fig. 2: Concentrations of SO_2, NOx and ozone (Jan. 11, 1997)

4. Results and Discussion

[1] Sulfur oxides and transport patterns

Concentrations of SO_2 showed very clear difference depending on the transport pattern. Fig. 2 shows the concentration profile of SO_2, NOx, and ozone observed on Jan. 11, 1997. SO_2 and NOx were generally high in the marine boundary layer. On the other hand, SO_2 concentration observed on Dec. 12, 1997 was generally low (less than 1 ppb everywhere) as shown in Fig. 3. In order to clarify the reason for those differences, back trajectories were calculated. The results are shown in Figs.

4 and 5. Differences are obvious. The air mass caught on Jan. 11 had started near Shanghai, China three days before. It stayed for one day near Shanghai and came across the East China Sea. Starting altitude was low (less than 2000 m). Highly polluted air must have been included in the air mass. On the other hand, the air mass caught on Dec. 12 had started Siberia three days before and had come down from high altitude (5000 m). It could not contain air pollution, since there was no big emission source in the originating area.

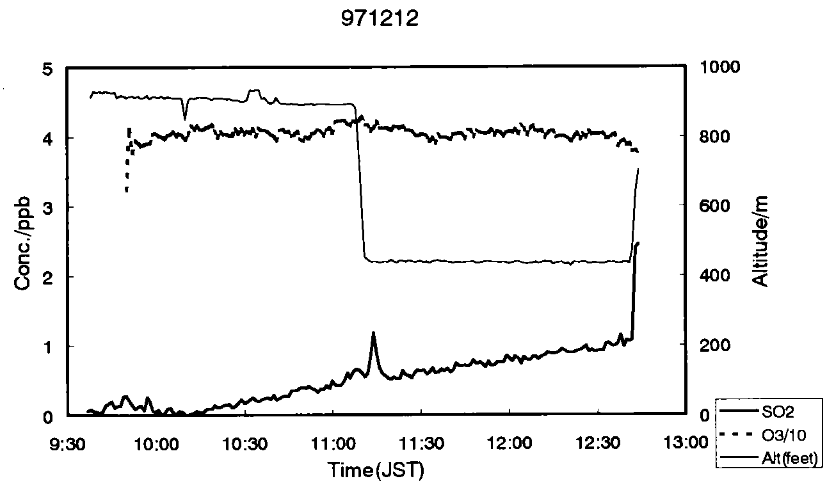

Fig. 3: Ozone and SO_2 observed on Dec. 12, 1997

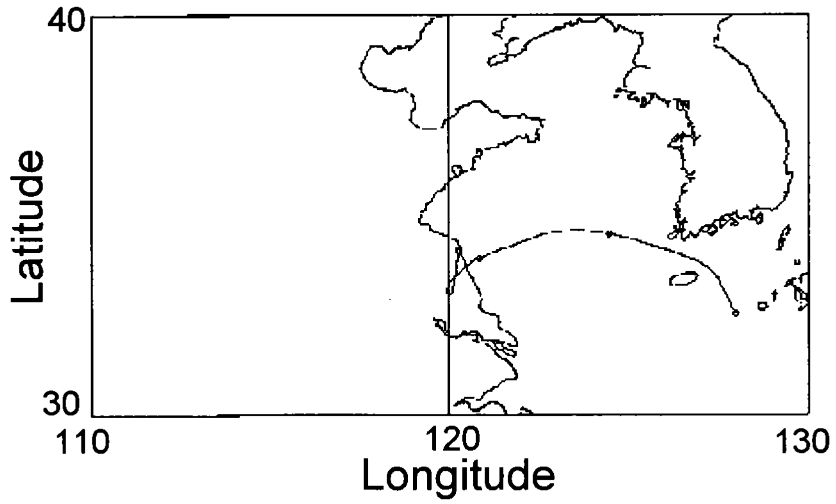

Fig. 4: Back trajectory of the air mass caught near Fukue on Jan. 11, 1997

On the basis of above analysis it became clear that there is another transport route from the Asian continent to Japan in addition to the three routes mentioned in Introduction section; a path from central/southern China to north Kyushu across the northern East China Sea.

The existence of such transport route for polluted air mass was predicted by the model developed in this institute based on C-1.1.1 sub-theme within this project. The model suggested that highly polluted air mass accompanied a cyclone that moved from central/southern China or Taiwan to north west Pacific along the south coast of Japan. The data supporting this prediction was obtained in the experiment in FY1998. As shown in Fig 6. more than 10 ppb of SO_2 was observed not only in boundary layer but also in free troposphere. It shows that the air was well mixed among low and high altitude (<3000 m).

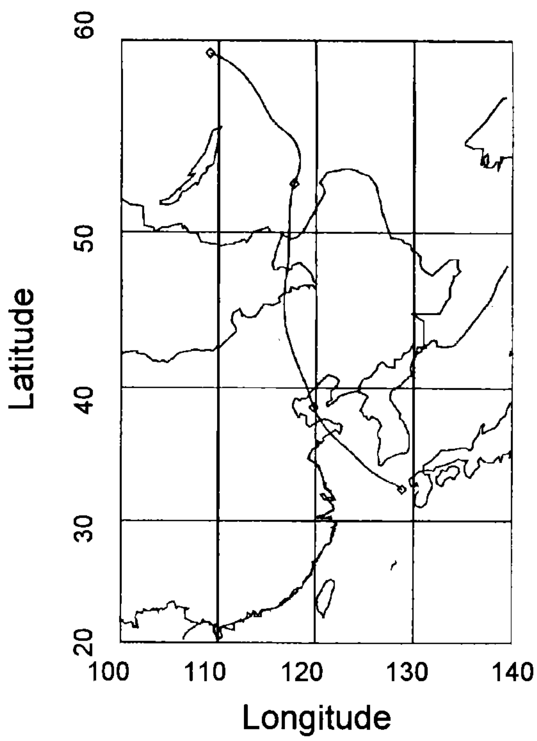

Fig. 5: Back trajectory of the air mass caught near Fukue on Dec. 12, 1997

A cyclone moved quickly leaving around Taiwan on Feb. 1, 1999 and reached northwest Pacific on Feb. 3. Very strong west wind brew on Feb. 2. In the experiments carried out on Feb. 4 and 6, such high concentrations of SO_2 were not observed, although the pressure pattern was west-high and east-low on both days.

[2] Nitrogen oxides

Measurements of nitrogen oxides were made with an ozone-chemiluminescence NO-NOx analyzer (TECO, Model 42S). A commercial analyzer was modified for the aircraft measurement and for improvement of its detection limit.[4] In FY1998 experiments NOx analyzer was again modified to be used as an NOy analyzer (HNO_3 conversion was improved from ~10 % for previous system to 92 ± 5 % for new system).

In the FY1998 experiments NOy was analyzed with the NOy analyzer described above and PAN was collected by use of a low temperature column cooled at -78 ° C.

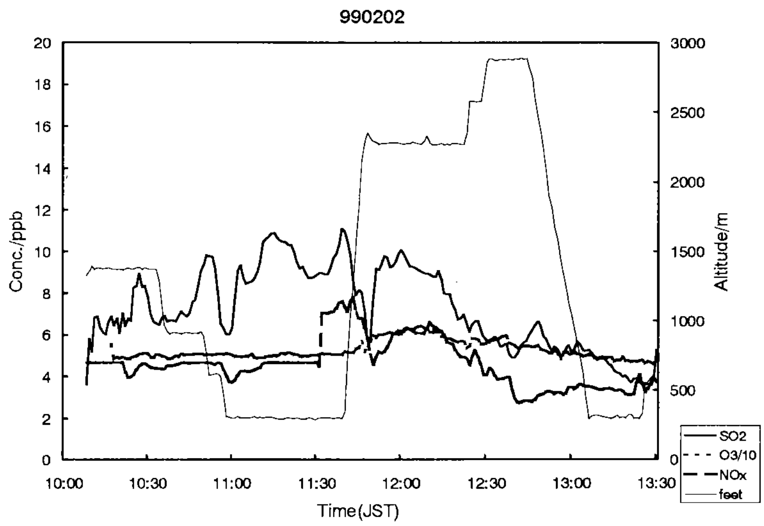

Fig. 6: Concentrations of ozone, SO_2 and NOx measured on Feb. 2, 1997

In the experiment on Feb. 2, 1999 the highest concentration of NOy so far was recorded. In the previous experiments the highest concentration of NOx was 4 ppb in the boundary layer and the concentration of NOx was always less than 1 ppb in the free troposphere. However, NOy was higher than 3 ppb during the flight on Feb. 2 and 8 ppb was recorded in the boundary layer. It is not due to the change of the analyzing system but to transport pattern of pollutants as discussed in the previous section.

[3] Trace acidic gases and aldehydes

In order to monitor trace acidic gases such as HCl, HNO_3, and SO_2 or aldehydes such as HCHO and CH_3CHO, we developed an automated continuous analysis system usable on an small airplane. It utilizes diffusion scrubber systems as collecting devise for gaseous

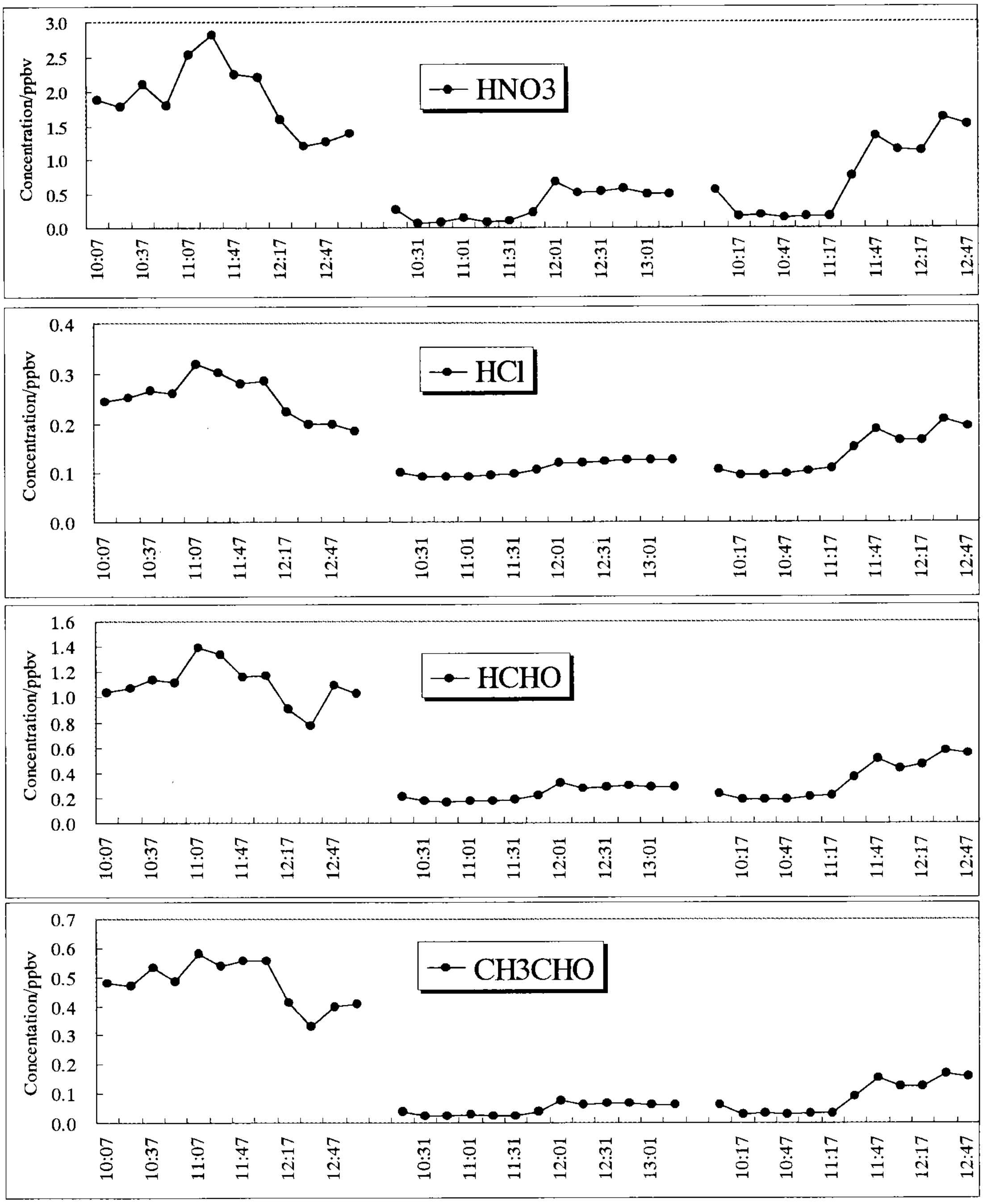

Fig. 7: Concentrations of acidic gases and aldehydes observed in Feb. 1999

compounds in sample air. Details of the diffusion scrubber have been reported already.[5)] A portable ion chromatograph for acidic gases and a portable HPLC for aldehydes were installed on the automated analysis system. A sequencer controlled pumps for air sampling and suppliers of adsorbents as well as automatic valves. Thus, it became possible to monitor the concentrations of HCl, HNO_3, SO_2, HCHO, and CH_3CHO at 15 min interval. Fig 7 shows the results of experiments in FY1998 (Feb., 1999).

All the gaseous pollutants existed in very high concentration on Feb. 2. The distribution of gases along the flight track is very similar one another. It supports the contention described above. Very polluted air mass was transported from central/southern China on that day. All the components showed the highest concentration so far, too.

5. Conclusion

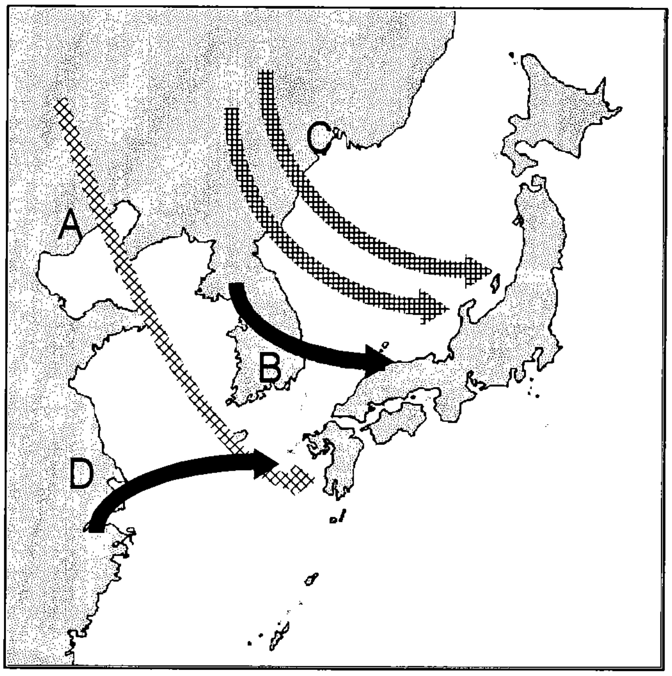

Fig. 8: The routes of transport of air masses from the Asian continent

It became evident that there is a route for transport of pollutants from central/southern China as depicted as D in Fig. 8. The model developed in the sub-theme C-1.1.1 in this project suggested the existence of such a transport route when a cyclone passes by south of Japanese main islands. It was expected that the most typical pollution pattern transported from central/southern China appears in the East China Sea. Just as our expectation, very polluted air mass was observed in the experiment on Feb. 2, 1999. The field observation data supported the model simulation results well.

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