C-1.1.1 Studies on the Behavior of Acidic and Oxidative Component in East Asia by Ground Based Observation

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Abstract In order to clarify the behavior of atmospheric pollutants, observation of atmospheric pollutants had been carried out at several places where the air pollutants from Asian continent may reach. In Okinawa Is., maximum ozone level of 91ppb was observed in May. The lowest monthly average ozone concentration of 11ppb was recorded in July. Back trajectory analysis of air mass from Oki Is. showed that the contribution from Asian continent were 70-80% in winter, while it was 50-60% and 30-40% in the spring and summer, respectively. The long term record of particulate non-sea-salt sulfate concentrations showed nearly 5% increase. The main contributor of inorganic ion of particulate matter on Happo-one is sulfate. The deposition of non-sea-salt sulfate on Happo-one is twice as much of that in urban Nagano city. In the western part of Aomori Pref., the concentration of non-sea-salt sulfate was low in the summer and high in the winter when the northwest wind prevailed. The sampling point on the coast of Sea of Japan exhibited a similar level of the concentration of non-sea-salt sulfate compared to those of inland areas, so long range transport of air pollutants were significantly indicated.

Key Words East Asia, Long Range Transport, Ozone, Back Trajectory

1. Introduction

A large amount of SO_2 and NOx 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_2 and NOx may increase in the near future by the development of industry and increment of population. These precursor were converted to sulfate and nitrate by oxidative reaction. 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.

From the air pollutant monitoring at less polluted mountainous area and remote islands or the sampling site where the air pollutants from the Asian continent may be transported, we can learn the behavior of air pollutants and can detect the yearly variation of acidic deposition in Japan.

2. Research Objectives

In order to clarify the behavior of atmospheric pollutants, observation of atmospheric pollutants had been carried out at several places where the air pollutants from Asian continent may reach. The observation was conducted in Okinawa Is. where the sea and continent air mass dominate depending on the season. Oki Is. is situated in Sea of Japan and separated 60-

80 km from the main land of Japan and there are no significant emission of air pollutant in the island. It is a good sampling point to detect the contribution of air pollutant from the Asian continent and main land of Japan.

3. Research Method, Results and Discussion

[1] Okinawa Island (Hedo)

Ozone concentration was monitored by Dasibi ozone monitor. From the data obtained from February to November 1992, ozone concentration ranged from 0 to 91ppb and its average is 29.1ppb(Fig. 1). The characteristics of concentration variation is as follows; in autumn to

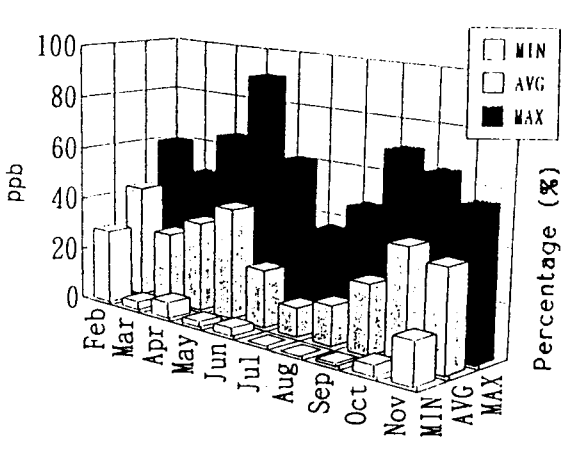

Fig. 1 Monthly averaged ozone concentration

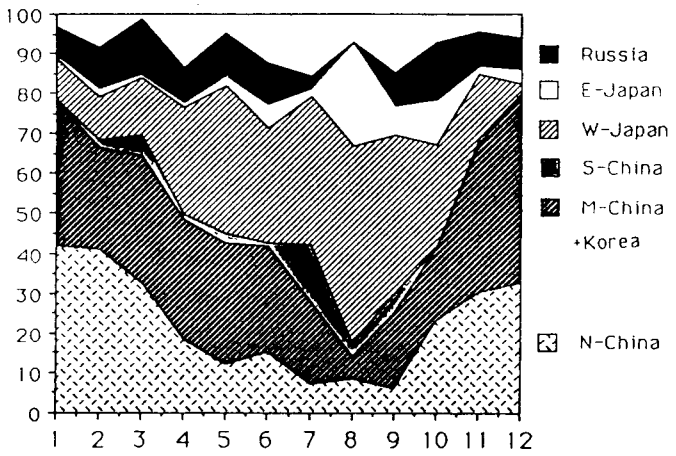

Fig. 2 Contribution of air mass from each sector

winter the concentration is relatively high, in March temporally it decreased a little bit, early summer in May the maximum concentration of 91ppb was observed and then it gradually decreased to show the minimum value in July to August, because the sea air mass prevails. Especially in July the concentration of ozone ranged from 0 to 37ppb, and the lowest monthly average concentration of 11ppb was observed.

[2] Oki Island

Back trajectory analysis at Oki Is. was conducted from March 1988 to March 1991. The sector is divided as follows, (1) Northern part of China, (2) Korea and Middle part of China, (3) Southern part of China, (4) Western part of Japan, (5) Eastern part of Japan, (6) Russia, (7) Impossible to classify. The contribution of each sector is shown in Fig. 2. From the Asian continent including Russia air mass was transported 80-90%, 50-60% and 30-40% in winter, spring and summer, respectively. The frequency of air mass from Russia stayed a constant level of 10% in the whole year. As for Japan, the contribution from the western part is significant and the contribution of eastern part is the biggest(20%) in August. Detailed analysis showed that the contribution of Mt. Sakurajima which is the most active volcano in Japan is occasionally detected. As is shown in Fig. 3, the long term record of particulate non-sea-salt sulfate (nss-sulfate) concentrations showed nearly 5% increase. Some deviation is observed because the meteorological parameter as if rain fall amount is different. It is clear that the air pollution in the Sea of Japan area is progressing apparently.

[3] Happo-one

Monthly average value of ozone concentration is high in spring(April to June) and reached above 50ppb and is low in summer(August to September). High concentration of ozone

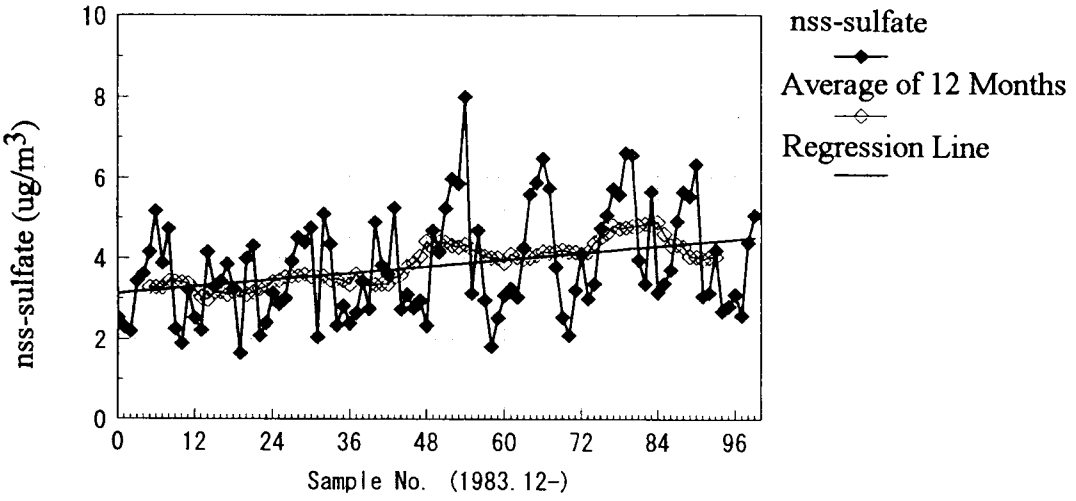

Fig. 3 Trend of nss-sulfate concentration

appeared in interval of a few days in spring and in some year it exceeded 100ppb. The high concentration of ozone is attributed to the intrusion of stratospheric ozone and photochemical production.

Monthly variations of chemical component of particulate matter collected on Happo-one were exhibited in Fig. 4. It had a tendency that the chemical component increase in spring to summer. Particulate sulfate is the main contributor in every month and it comprise about 50% of the total inorganic ion. The concentration of particulate sulfate is high in spring to summer and low in autumn to winter. This seasonal variation coincide with the variation of total nitrate and Pb, so it is suggested that anthropogenic SO_2 are converted by photochemical reaction to sulfate. Calcium and aluminum concentration increased in spring(March to May) and also Fe, Ti and Mn exhibited nearly same variation. These component are involved in yellow sand, so the transport of yellow sand from the Asian continent is expected.

A large amount of sulfuric acid mist may exist in the atmosphere on mountainous area. Nitrate and chloride content on the quartz filter is low because the sulfuric acid mist evaporate the collected particulate nitrate and chloride. If we assume that all the nitrate and chloride on the second stage alkaline reagent impregnated filter are evaporated by sulfuric acid mist, it comprised about 40% of the total sulfate. A large amount of sulfuric acid mist are thought to be transported as acid.

Monthly variation of rain fall amount, pH and deposition amount of chemical component of total deposition on Happo-one are shown in Fig. 5. The value of pH ranged from 4.61 to 5.87 and average is 5.1. As seen characteristically in spring of 1992, pH value increased in spring. It is because a large amount of Ca contained in yellow sand may neutralize the acidity of deposition. The Ca concentration of deposition increased in spring. The contribution of each chemical component in the season of yellow sand transport(March and April) and in Summer(May to September) in Fig. 6. In the spring about 50% of anthropogenically originated nitrate and sulfate were neutralized by Ca. This percentage is high compared to the summer time value of 15 %, and the difference of 35 % may be attributed to the Ca in yellow sand. Yellow sand is a important material to consider the neutralization of acid rain in the spring time.

The deposition amount of acidic component are shown in Fig. 7. The deposition amount of nss-sulfate on Happo-one is twice that of Nagano city(urban area), and also the deposition of nitrate is large. It was clarified that a large amount of acidic component deposited on

mountainous area.

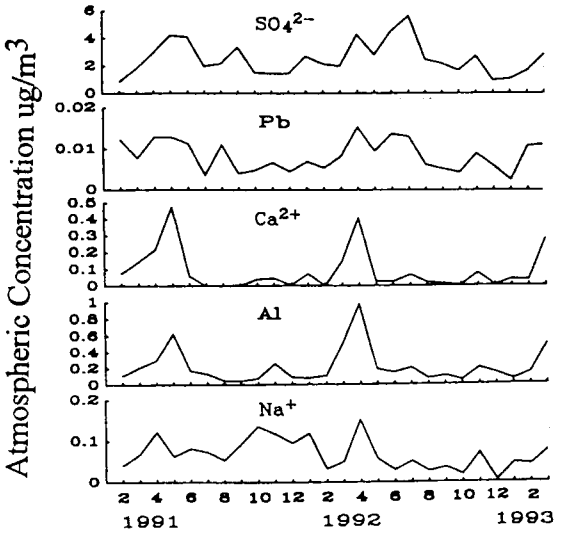

Fig. 4 Monthly variation of ionic species of particulate matter

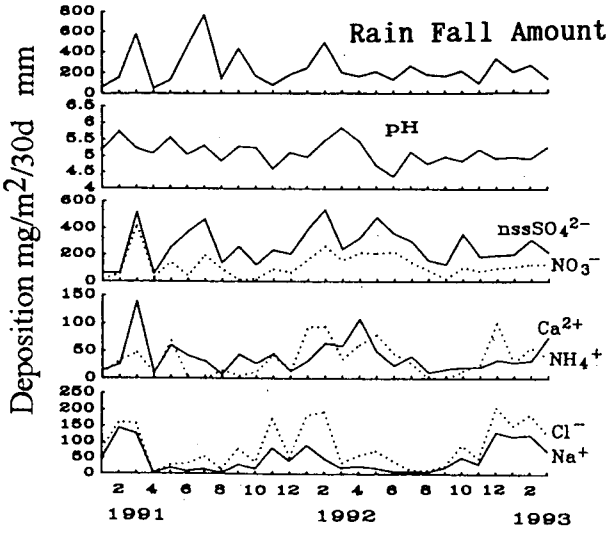

Fig. 5 Monthly variation of deposition

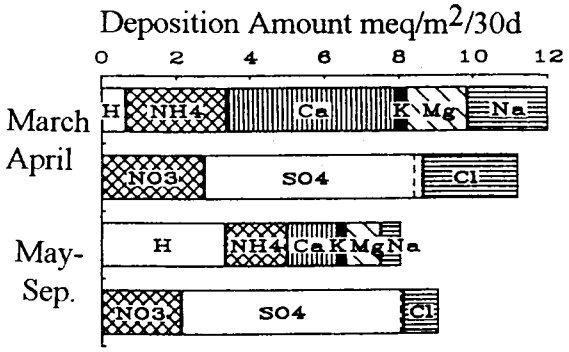

Fig. 6 Contribution of each chemical component

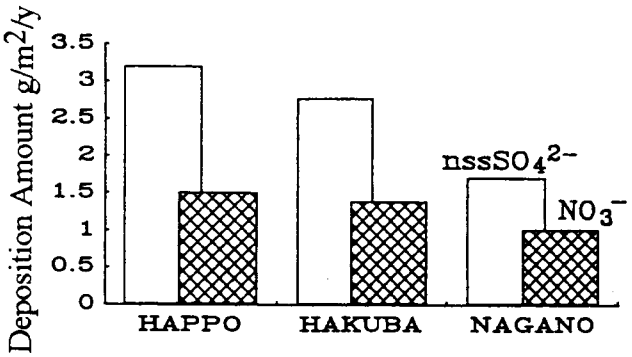

Fig. 7 Deposition of nss-sulfate and nitrate

[4] Hakkoda(Aomori)

Total deposition was collected in 4 sampling sites(Ajigasawa: Western shore, Mt. Hakkoda: Altitude 1300 m, Center: Aomori city, Rokkasyo village: Eastern part of Prefecture) in Aomori Prefecture in 1992. Yearly average of pH of each sampling site were 4.75, 4.96, 4.89 and 4.65, respectively. Yearly variation of pH showed maximum value in March and it was well correlated to the variation of Ca, and is high in urban area, so it was attributed to the road dust. The variation of monthly averaged concentration of nss-sulfate is correlated well in three sampling site which are all low pollution site except for the Center.

Publication

T. Okita, K. Murano, M. Matsumoto, T. Totsuka, Determination of Dry Deposition Velocities to Forest Canopy from Measurements of Through Fall, Stem Flow and the Vertical Distribution of Aerosol and Gaseous Species, *Environ. Sci.*, **2**, 103-111, 1993.