

## Technical Data

# Concentrations of Chemical Components, Including $^{210}\text{Pb}$ , Present in Aerosols Collected at Naha, Okinawa Prefecture, a Sub-tropical Region of Japan

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Aerosol samples were collected at Naha, Okinawa Prefecture, Japan during July 2001 to March 2005 to elucidate the concentrations of chemical components, including  $^{210}\text{Pb}$ , present in air for a sub-tropical region of Japan. The  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$  and  $\text{Ca}_2^+$  concentrations had clear seasonal variations and were high in winter to spring and low in summer.  $^{210}\text{Pb}$  concentrations ranged from 0.10 to 1.52  $\text{mBq m}^{-3}$  and were low in summer and high in the other three seasons. The source of sulfur compounds in the aerosol samples was clarified by combining the sulfur isotope ratio  $\delta^{34}\text{S}$  and  $^{210}\text{Pb}$  concentrations. The multi-isotope method using  $\delta^{34}\text{S}$  and  $^{210}\text{Pb}$  concentrations was judged to be a good tool for understanding the transported sulfur source.

**KEY WORDS:**  $^{210}\text{Pb}$ , aerosols, Okinawa, long-range transport, source of sulfur compounds.

## I INTRODUCTION

Long-range transported pollutants cause environmental problems worldwide.<sup>1,2)</sup> In the Far East Asian region, strong northwestern monsoons from the continental inland area blow to coast areas including the Japanese archipelago. These monsoons carry air masses that include many kinds of natural and anthropogenic materials like soil particles and pollutants.<sup>3)</sup> For example, long-range transported soil particles from the Asian continent are a well-known phenomenon called Asian dust or *Kosa*.<sup>4)</sup> These Asian dust events are mainly observed over Japan in the spring. They have been attracting attention in many fields because of their significant influences on both local and global environments.<sup>5,6)</sup> On the other hand, atmospheric pollutants, such as sulfur dioxide ( $\text{SO}_2$ ) that originates from fossil fuel combustion (coal and

petroleum), are one of the causes of acid rain in Far East Asia, and the amount of  $\text{SO}_2$  released into the atmosphere by fossil fuel combustion has generally increased due to economic growth.<sup>7)</sup> It is expected that atmospheric aerosols, brought by the western monsoons to the Japanese archipelago, contain  $\text{SO}_2$  coming from Far East Asian countries. It is important to understand the concentration levels and contributions of long-range transported components, such as sulfur compounds, in this area.

Both stable and radioactive isotopes have been used successfully in the past to attribute long-range transported materials in the atmosphere to their source regions. Sources of atmospheric sulfur compounds include sea spray  $\text{SO}_4^{2-}$ , anthropogenic sulfur such as  $\text{SO}_2$  by fossil fuel combustion, biogenic sulfur such as  $\text{H}_2\text{S}$  and dimethyl sulfide, and volcanic sulfur such as  $\text{H}_2\text{S}$  and  $\text{SO}_2$ , and the sulfur isotope ratio  $\delta^{34}\text{S}$  has been used as an indicator to determine the source and behavior of sulfur compounds in the environment.<sup>8)</sup> On the other hand,  $^{210}\text{Pb}$  is a natural radionuclide in the  $^{238}\text{U}$  decay series with a relatively long half-life ( $T_{1/2}$  of 22.3 y). Airborne  $^{210}\text{Pb}$  is produced from its precursor,  $^{222}\text{Rn}$  ( $T_{1/2}$  of 3.82 d), emanated from the ground; the produced  $^{210}\text{Pb}$  attaches to fine particles near the ground surface, and it is transported through the troposphere.  $^{210}\text{Pb}$  has proven to be a useful tracer for studying the source of air-mass and its transportation process in the atmosphere.<sup>9,10)</sup> In addition, KOTANI et al. have suggested that a multi-isotope method using  $\delta^{34}\text{S}$  of sulfate and  $^{210}\text{Pb}$  concentration is useful as an index for long-range transported aerosols travelling over the Japan Sea to northern Japan.<sup>11)</sup> However, reports including such data are limited.

Aerosol samples were collected at Naha City, Okinawa

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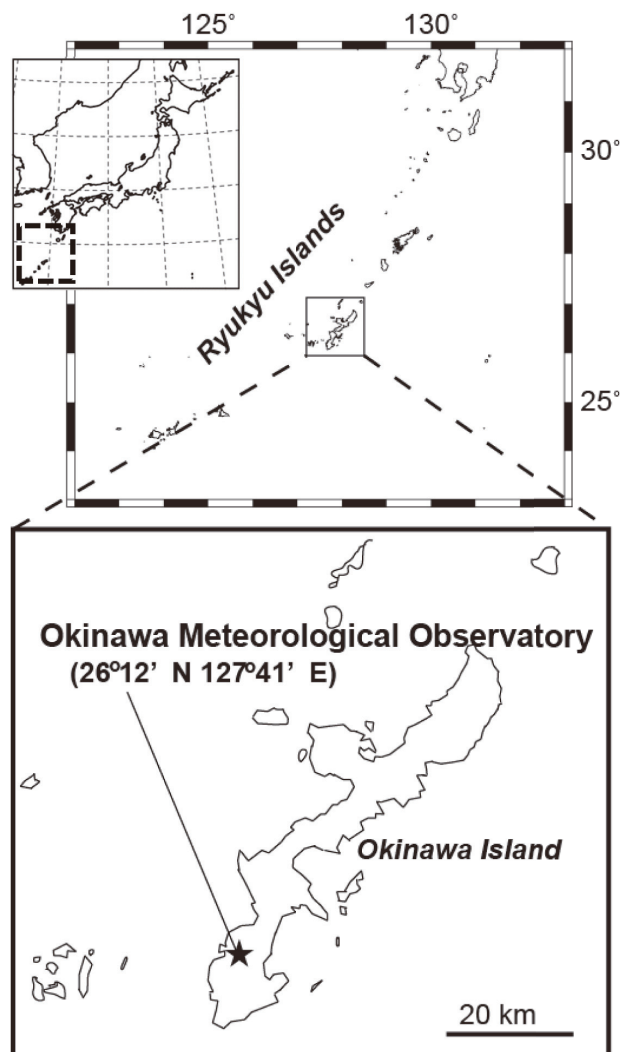
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Prefecture during 2001 to 2005 as part of the ADEC (Aeolian Dust Experiment on Climate impact) project,<sup>12)</sup> and the data on total suspended particle (TSP) and the sulfur isotope ratios of sulfate  $\delta^{34}\text{S}$  in them were reported.<sup>13, 14)</sup> In this paper, concentrations of chemical components, including  $^{210}\text{Pb}$ , in aerosol samples collected at Naha, Japan are reported. Furthermore, the relationship between  $\delta^{34}\text{S}$  and  $^{210}\text{Pb}$  concentrations that can be used to evaluate the long-range transported sulfur compounds in this region is presented.

## II MEASUREMENT METHOD

### 1. Overview of sample collection site

The sampling site was on a building roof of the Okinawa Regional Headquarters, Japan Meteorological Agency (JMA) at Naha City, Okinawa Prefecture ( $26^{\circ}12'\text{N}$ ,  $127^{\circ}41'\text{E}$ ) and approximately 40 m above sea level. **Figure 1** shows the sampling location. Naha City is located in the southern part of Okinawa Island which is the largest of the Ryukyu Islands. There are no high elevation mountains on this island. The



**Fig. 1** Location of the sampling site in Naha City, Okinawa, Japan.

island has a subtropical climate, and the annual average temperature is approximately  $23.1^{\circ}\text{C}$ .<sup>15)</sup> The rainy season starts early in May and continues until June, and typhoons occur in July to October. In late October, the frequency of winds blowing from the north increases.

### 2. Sample collection and analysis

The methods of aerosol sampling were the same as described in previous reports.<sup>13, 14)</sup> Briefly, aerosol samples were collected at intervals of approximately 10 to 20 days from July 2001 to March 2005 using a high-volume air sampler (HV-1000F, Sibata Co. Ltd., Japan) on a polyflon filter ( $25\text{ cm} \times 20\text{ cm}$ , PF040, ADVANTEC Co. Ltd., Japan). The airflow rate was approximately  $1,000\text{ L min}^{-1}$ .

A quarter section of each filter after the air-dried using desiccator was used in extraction of water-soluble components. The quarter section was cut into fine pieces and put into a Teflon™ beaker filled with 6 mL of 95% ethanol and 4 mL of distilled water. The water-soluble ionic species were extracted using an ultrasonic treatment for 5 minutes. After collecting the extraction solution into another container, the fine filter pieces were rinsed 3 times with 30 mL distilled water. All rinse solutions were collected and mixed with the extraction solution, and the mixed solution was filtered through a  $1.2\text{ }\mu\text{m}$  membrane filter (RAWP04700: Millipore, Japan). The concentrations of  $\text{Cl}^-$ ,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  were measured by ion chromatography (CompactIC 761; Metrohm, Switzerland). Concentrations of  $\text{Na}^+$ ,  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  were measured by inductively coupled plasma atomic emission spectroscopy (ICP-AES; SPS7000A; Seiko Instruments Inc., Japan). Concentration of  $\text{NH}_4^+$  was measured by indophenol blue absorptiometry (model no. 101; Hitachi, Japan).

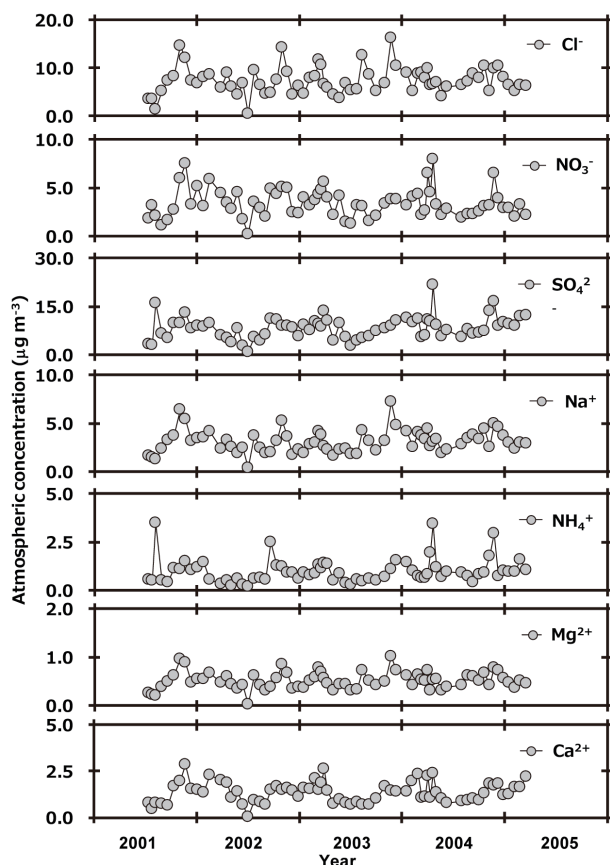
The remaining three-quarter section of each filter was air-dried using desiccator and cut into  $47\text{ mm}\phi$  that were placed into a plastic container (U8, AS ONE, Japan).  $^{210}\text{Pb}$  concentrations on the filters were measured by a high-purity Ge spectrometer (HPGe-GX-3018; Canberra, USA) with a multi-channel analyzer for 100,000 s to determine  $\gamma$ -rays of 46.5 keV. Counting uncertainty was less than 6% as relative standard deviation. Measured values were corrected for radioactive decay to the middle day of the sampling period.

### 3. Backward air-mass trajectory analysis

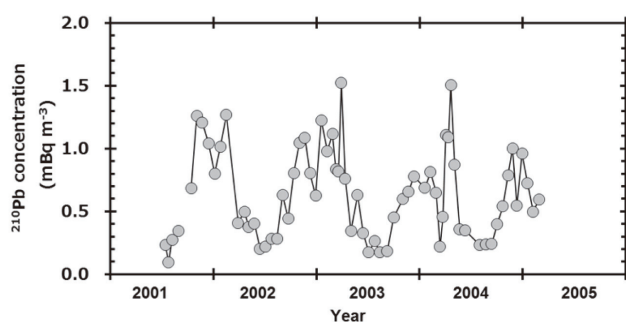
The air-mass trajectory analysis was carried out in order to investigate the air-mass transportation course to the sampling site in Naha City. The three-dimensional backward air-mass trajectories for 96 hours starting at 100 m altitude were calculated by METEX.<sup>16)</sup> The grid point values (GPV) data sets produced by the National Centers for Environmental Prediction, USA were used as weather data.<sup>17)</sup> The trajectories were calculated for every 6 hours from every 09 UTC during the sampling period.

## III RESULTS AND DISCUSSION

Atmospheric concentrations of anions and cations are shown in **Fig. 2**. Atmospheric concentration ranges of the anions were:  $\text{Cl}^-$ , 0.47 to  $16.30\text{ }\mu\text{g m}^{-3}$ ;  $\text{NO}_3^-$ , 0.18 to  $7.92$



**Fig. 2** Variation of ionic species in aerosol samples collected at Naha City.



**Fig. 3** Variation of  $^{210}\text{Pb}$  concentration in aerosols collected at Naha City.

$\mu\text{g m}^{-3}$ ; and  $\text{SO}_4^{2-}$ , 0.94 to  $21.66 \mu\text{g m}^{-3}$ . The ranges of the cations were:  $\text{Na}^+$ , 0.34 to  $7.28 \mu\text{g m}^{-3}$ ;  $\text{NH}_4^+$ , 0.18 to  $3.47 \mu\text{g m}^{-3}$ ;  $\text{Mg}^{2+}$ , 0.02 to  $1.01 \mu\text{g m}^{-3}$ ; and  $\text{Ca}^{2+}$ , 0.04 to  $2.86 \mu\text{g m}^{-3}$ . Atmospheric concentrations of  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$  and  $\text{Ca}^{2+}$  were high in spring and fall, but on the other hand,  $\text{Cl}^-$ ,  $\text{Na}^+$  and  $\text{Mg}^{2+}$  had no clear seasonal variation. In summer (May to September), all ion concentrations were low. KANAI et al.<sup>13</sup> reported that the total suspended particle (TSP) concentration at Naha City was high in fall to spring and low in summer. This area has heavy rainfalls in summer from plume rain

**Table 1** Correlation coefficients of ion species in aerosol samples collected at Naha City, Japan.

	$\text{Cl}^-$	$\text{NO}_3^-$	$\text{SO}_4^{2-}$	$\text{SO}_4^{2-}$	$\text{Na}^+$	$\text{NH}_4^+$	$\text{Mg}^{2+}$	$\text{Ca}^{2+}$	$\text{Ca}^{2+}$
TSP	0.42	0.54	0.32	0.28	0.47	0.16	0.52	0.60	0.58
$\text{Cl}^-$		0.45	0.17	0.09	0.95	0.03	0.95	0.39	0.32
$\text{NO}_3^-$			0.67	0.64	0.51	0.50	0.58	0.77	0.75
$\text{SO}_4^{2-}$				1.00	0.32	0.85	0.36	0.69	0.69
$\text{SO}_4^{2-}$					0.24	0.86	0.28	0.66	0.67
$\text{Na}^+$						0.15	0.96	0.46	0.39
$\text{NH}_4^+$							0.15	0.35	0.35
$\text{Mg}^{2+}$								0.55	0.48
$\text{Ca}^{2+}$									1.00

TSP concentration was the reported the total suspended particle concentration ( $\mu\text{g m}^{-3}$ ).<sup>13)</sup>

and typhoons. It seemed that the low ion concentrations in summer were affected by a washout process. The relationship between  $\text{Na}^+$  and  $\text{Cl}^-$  concentrations was found to have a strong correlation ( $r = 0.95$ ,  $p < 0.01$ ) and the  $\text{Na}^+/\text{Cl}^-$  was similar to the sea water ratio. It was suggested that  $\text{Na}^+$  and  $\text{Cl}^-$  originated from seawater. SHIGETOMI et al.<sup>18)</sup> reported that atmospheric pollutant concentrations at Cape Hedo, Okinawa Prefecture demonstrated a clear seasonal variation: they were high in winter to spring and low in summer. The present results for  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$  and  $\text{Ca}^{2+}$  concentrations had a similar trend to the reported one. The correlation coefficients between ion species concentrations are shown in **Table 1**. Non-sea salt sulfate ( $\text{SO}_4^{2-}$ ) and non-sea salt calcium ( $\text{Ca}^{2+}$ ) were calculated from the conservative assumption that  $\text{Na}^+$  is a sea-salt tracer. The calculation equations are as follows:

$$\text{SO}_4^{2-} = \text{SO}_4^{2-} - (\text{SO}_4^{2-}/\text{Na}^+)_{\text{sea}} \times \text{Na}^+ \quad (1)$$

$$\text{Ca}^{2+} = \text{Ca}^{2+} - (\text{Ca}^{2+}/\text{Na}^+)_{\text{sea}} \times \text{Na}^+ \quad (2)$$

where  $(\text{SO}_4^{2-}/\text{Na}^+)_{\text{sea}}$  and  $(\text{Ca}^{2+}/\text{Na}^+)_{\text{sea}}$  are the concentration ratio in sea water.  $\text{Na}^+$  also showed a good correlation with  $\text{Mg}^{2+}$  ( $r = 0.96$ ,  $p < 0.01$ ) which had the same source (sea spray). Both  $\text{SO}_4^{2-}$  and  $\text{SO}_4^{2-}$  had a strong correlation with  $\text{NH}_4^+$  ( $\text{SO}_4^{2-}$ ,  $r = 0.85$ ,  $p < 0.01$ ;  $\text{SO}_4^{2-}$ ,  $r = 0.86$ ,  $p < 0.01$ ) which suggested that they were present as  $(\text{NH}_4)_2\text{SO}_4$ .  $\text{Ca}^{2+}$  was significantly correlated with  $\text{NO}_3^-$  ( $r = 0.77$ ,  $p < 0.01$ ) and  $\text{SO}_4^{2-}$  ( $r = 0.69$ ,  $p < 0.01$ ). It is well known that the source of  $\text{Ca}^{2+}$  is mainly soil particles. The present result indicated that  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  were transported to Naha with soil particles.

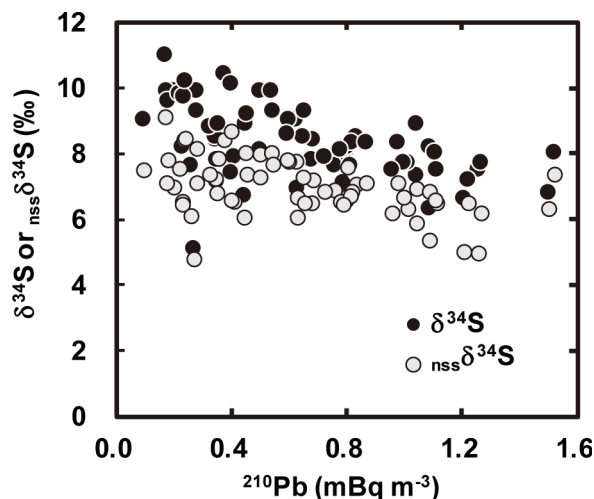
Atmospheric concentrations of  $^{210}\text{Pb}$  are shown in **Fig. 3**. They ranged from 0.10 to  $1.52 \text{ mBq m}^{-3}$  with the mean value ( $\pm$  S.D.) of  $0.64 \pm 0.36 \text{ mBq m}^{-3}$ , and there was a clear seasonal variation which was high in fall to spring and low in summer. TANAHARA et al.<sup>19)</sup> reported that atmospheric  $^{210}\text{Pb}$  concentration on Okinawa Island during 2004 to 2011 ranged from 0.06 to  $1.98 \text{ mBq m}^{-3}$  with the mean value of  $0.80 \text{ mBq m}^{-3}$ . The present results were comparable to the reported values. **Table 2** shows the correlation coefficients between  $^{210}\text{Pb}$  concentration and ion concentrations. There were good correlations between  $^{210}\text{Pb}$  and  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{Ca}^{2+}$  concentrations (correlation coefficients  $> 0.7$ ,  $p < 0.01$ ).

**Table 2** Correlation coefficients between  $^{210}\text{Pb}$  and ion concentrations in aerosol samples collected at Naha City, Japan.

	Correlation Coefficient
TSP	0.51
$\text{Cl}^-$	0.34
$\text{NO}_3^-$	0.75
$\text{SO}_4^{2-}$	0.70
$_{\text{nss}}\text{SO}_4^{2-}$	0.68
$\text{Na}^+$	0.40
$\text{NH}_4^+$	0.45
$\text{Mg}^{2+}$	0.45
$\text{Ca}^{2+}$	0.77
$_{\text{nss}}\text{Ca}^{2+}$	0.76

The air-mass origin for Okinawa Island in summer is mainly the Pacific Ocean,<sup>19)</sup> and heavy rainfalls by plume rain and typhoons are observed. On the other hand, the northwestern monsoons blow to Okinawa Island in fall to spring. The source of  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{Ca}^{2+}$  species is mainly the Asian continent (exhaust gas from automobiles and soil particles from semi-arid and arid regions).<sup>20)</sup> It seemed that the seasonal variation of  $^{210}\text{Pb}$  concentrations depended on the regional meteorological conditions.

It is known that in all the emission inventory the two largest fluxes to the atmosphere are those from fossil fuel combustion and from oceanic biological activity, excluding the direct input of sea-salt sulfate.<sup>21)</sup> TADANO et al.<sup>14)</sup> reported that  $\delta^{34}\text{S}$  and  $_{\text{nss}}\delta^{34}\text{S}$  (sulfur isotope ratio of non-sea salt sulfate) ranged from 5.1 to 11.0‰ and 4.8 to 9.1‰, respectively, and seasonal variation was low in summer and high in fall and spring. AKATA et al.<sup>22)</sup> reported that the  $_{\text{nss}}\delta^{34}\text{S}$  values were to around 5‰ with increasing  $_{\text{nss}}\text{SO}_4^{2-}$  concentration in aerosols collected at Tsuruoka, northern Japan. They suggested that the recent  $\delta^{34}\text{S}$  value of sulfur compounds from the Asian continent with the source of fossil fuel combustion was around 5‰. And there are some reports that biogenic sulfur sources like dimethyl sulfide have a high  $\delta^{34}\text{S}$  composition.<sup>23, 24)</sup> TADANO et al.<sup>14)</sup> suggested that volatile organic sulfur oxides from the sea which has high  $\delta^{34}\text{S}$  values contributed to the atmospheric  $_{\text{nss}}\text{SO}_4^{2-}$  concentration in summer, while on the other hand, continental anthropogenic sulfur compounds which has slightly lower  $\delta^{34}\text{S}$  values than those of volatile organic sulfur oxides contributed to it in winter. **Figure 4** shows the relationships between  $^{210}\text{Pb}$  concentration and  $\delta^{34}\text{S}$  or  $_{\text{nss}}\delta^{34}\text{S}$ .<sup>14)</sup> As shown there,  $\delta^{34}\text{S}$  and  $_{\text{nss}}\delta^{34}\text{S}$  decreased with increasing  $^{210}\text{Pb}$  concentration and the correlation coefficients of  $^{210}\text{Pb}$  with  $\delta^{34}\text{S}$  and  $_{\text{nss}}\delta^{34}\text{S}$  were  $-0.53$  and  $-0.47$ , respectively. High  $^{210}\text{Pb}$  concentration and  $_{\text{nss}}\delta^{34}\text{S}$  of 5 to 6‰ were observed in spring (March to April), and low  $^{210}\text{Pb}$  concentration and  $_{\text{nss}}\delta^{34}\text{S}$  of 7 to 8‰ were observed in summer (July to August). **Figure 5** shows the back trajectories for 96 hours starting at 100 m above the sampling site; high  $^{210}\text{Pb}$  concentrations were observed in spring (a, b), and the air-mass from the Asian continent was included. On the other hand, back trajectories in summer were transported from the ocean (c, d). The obtained

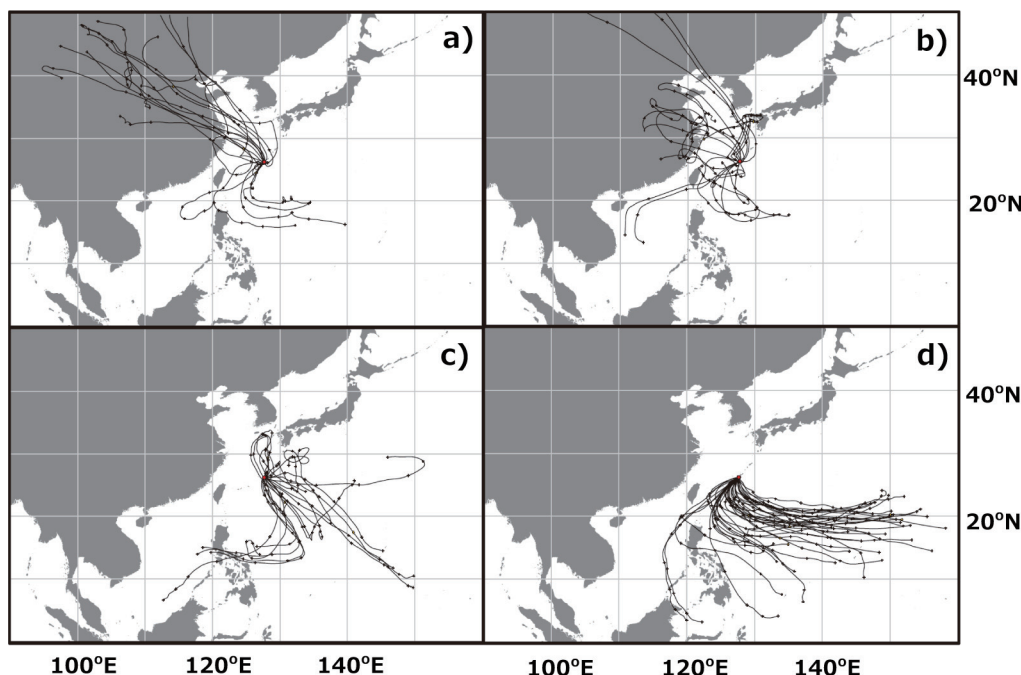
**Fig. 4** Relationship between atmospheric  $^{210}\text{Pb}$  concentration and sulfur isotope ratio of sulfate ( $\delta^{34}\text{S}$ ) or sulfur isotope ratio of non-sea salt sulfate ( $_{\text{nss}}\delta^{34}\text{S}$ ) at Naha City.

relationship between  $^{210}\text{Pb}$  concentration and  $_{\text{nss}}\delta^{34}\text{S}$ , and its relationship to air-mass transportation courses supported the idea that the source of sulfur to Okinawa Island in summer was mainly volatile organic sulfur oxides from the sea and in spring, it was continental anthropogenic sulfur. The source identification of sulfur compounds in the atmosphere could be clarified using the multi-isotope method combined with  $\delta^{34}\text{S}$  and  $^{210}\text{Pb}$  concentrations, although the source of atmospheric sulfur compounds could not be clarified using only  $\delta^{34}\text{S}$ . Thus, the multi-isotope method using  $\delta^{34}\text{S}$  and  $^{210}\text{Pb}$  concentrations was a good tool to understand the transported sulfur source to Okinawa Island.

#### IV CONCLUSIONS

Aerosol samples were collected at Naha City, Okinawa Prefecture, Japan and concentrations of the ionic species and  $^{210}\text{Pb}$  were measured. Atmospheric concentration ranges of the anions were:  $\text{Cl}^-$ , 0.47 to 16.30  $\mu\text{g m}^{-3}$ ;  $\text{NO}_3^-$ , 0.18 to 7.92  $\mu\text{g m}^{-3}$ ; and  $\text{SO}_4^{2-}$ , 0.94 to 21.66  $\mu\text{g m}^{-3}$ . The ranges of the cations were:  $\text{Na}^+$ , 0.34 to 7.28  $\mu\text{g m}^{-3}$ ;  $\text{NH}_4^+$ , 0.18 to 3.47  $\mu\text{g m}^{-3}$ ;  $\text{Mg}^{2+}$ , 0.02 to 1.01  $\mu\text{g m}^{-3}$ ; and  $\text{Ca}^{2+}$ , 0.04 to 2.86  $\mu\text{g m}^{-3}$ . Furthermore, atmospheric concentrations of  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$  and  $\text{Ca}^{2+}$  were high in spring and fall, but,  $\text{Cl}^-$ ,  $\text{Na}^+$  and  $\text{Mg}^{2+}$  had no clear seasonal variation.  $^{210}\text{Pb}$  concentrations ranged from 0.10 to 1.52  $\text{mBq m}^{-3}$  and were high in fall to spring and low in summer. As a result of back trajectory analysis, it was concluded the air-mass from the Asian continent blew into Okinawa in the spring when  $^{210}\text{Pb}$  concentration was high. On the other hand, the air-mass was transported from the ocean in summer when  $^{210}\text{Pb}$  concentration was low. It seemed that the source of atmospheric sulfur in Okinawa in summer was mainly volatile organic sulfur compounds from the ocean, while in spring the main source was continental anthropogenic sulfur.





**Fig. 5** Back trajectories for 96 hours starting at 100 m above the sampling site.  
 a) 25 March 2002–5 April 2002;  $^{210}\text{Pb}$  concentration,  $1.52 \text{ mBq m}^{-3}$ ;  $\delta^{34}\text{S}_{\text{nss}}$ , 7.4‰.  
 b) 16 April 2002–26 April 2002;  $^{210}\text{Pb}$  concentration,  $1.50 \text{ mBq m}^{-3}$ ;  $\delta^{34}\text{S}_{\text{nss}}$ , 6.3‰.  
 c) 20 July 2004–30 July 2004;  $^{210}\text{Pb}$  concentration,  $0.10 \text{ mBq m}^{-3}$ ;  $\delta^{34}\text{S}_{\text{nss}}$ , 7.5‰.  
 d) 25 June 2003–15 July 2003;  $^{210}\text{Pb}$  concentration,  $0.17 \text{ mBq m}^{-3}$ ;  $\delta^{34}\text{S}_{\text{nss}}$ , 9.1‰.

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The authors indicated no conflicts of interest.

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