Bulletin of Glaciological Research 24 (2007) *23–28*

Deposition analysis of non sea-salt sulfate and nitrate along to the northwest winter monsoon in Hokuriku district by a snow boring core and bulk samples

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(Received October 3, 2005; Revised manuscript accepted September 13, 2006)

Abstract

The depositions of non sea-salt sulfate and nitrate of both the Asian Continent and domestic origin were evaluated in the Hokuriku district, central Japan. Evaluation periods were from December 1, 1999 to March 17, 2000.

Observations were done by using bulk samplers in a plain area and by using a snow boring core on the high mountain area. Depositions of non sea-salt sulfate in the studying area (1000 km^2) were 2.743×10^6 kg and contributions of non sea-salt sulfate of the Asian Continent origin were 1.402×10^6 kg. Depositions of nitrate in the studying area (1000 km^2) were 1.204×10^6 kg and contributions of nitrate of the Asian Continent origin were 0.257×10^6 kg.

However, it was considered that acid rain constituents in precipitation at the high mountain area had a different origin from those in precipitation in the plain area, because the ratio of both non sea-salt sulfate and nitrate of the Asian Continent origin to sea-salt sulfate in precipitation at the high mountain was very different from those in precipitation at the plain area.

1. Introduction

Hokuriku district is located in the Sea of Japan side, central Japan. Winter precipitation in the Hokuriku district is mainly due to northwestern winter monsoon that blows from the Asian Continent to Japan. The precipitation in the plain area in Hokuriku district due to winter monsoon contains high concentrations of sea-salt constituents and non sea-salt sulfate (Honoki and Hayakawa, 2001). The amount of wet depositions of non sea-salt sulfate in winter is the largest in Hokuriku district compared to other districts in Japan (Fujita, 1996).

In Hokuriku district, the area affected by precipitation due to winter monsoon expands from the seacoast to about 100 km leeward. The behaviors of both non sea-salt sulfate and nitrate in this area are very important for revealing acid rain problems. The area affected by winter monsoon is not only the plain areas but also the high mountain areas, for example, Mountain Tateyama has maximum altitude of 3000 m.

Origins of non sea-salt sulfate in winter precipitation in the plain area in Hokuriku district were both the Asian Continent and domestic (Honoki and Hayakawa, 2001, Honoki *et al.,* 2001). Concentrations of both non sea-salt sulfate and nitrate originated from the Asian Continent in winter precipitation in plain part of Hokuriku district were reported in about 10 km steps from the seacoast to 55 km inland (Honoki and Hayakawa, 2001). However, no evaluations of those have not been performed on high mountain area on Mountain Tateyama, because it is difficult to take precipitation samples continuously on high mountain area especially during winter.

Snow boring core was also collected in spring at Murododaira (2450 m a.s.l) on Mountain Tateyama, and deposition data at Murododaira was combined with bulk precipitation data at plain area. Moreover, depositions of non sea-salt sulfate originated from both the Asian Continent and domestic have been evaluated in every 10 km to 20 km steps from the seacoast of the Sea of Japan to leeward of winter monsoon. Same evaluations of nitrate have also been performed.

In this paper, depositions of non sea-salt sulfate and nitrate originated from both the Asian Continent and domestic are evaluated in the 10 steps of 10 km square area from the seacoast of the Sea of Japan to leeward of winter monsoon.

2. Methods and Materials

2.1 Sampling

Sampling sites are shown in Fig. 1. Profile of altitude through a-a' line of Fig. 1 is shown in Fig. 2. Bulk precipitation samplers (open area of 224 cm^2) were set from station 1(abbreviated as St. 1) to St. 7, also at St. 10. St. 1 to St. 7 were located in the plain areas in Toyama Prefecture and Ishikawa Prefecture. St. 10 was located at east submontane of Ushiro Tateyama Mountain Range in Omachi Nagano Prefecture. Station 10 was located at the most leeward part of winter monsoon. All of these samples of St. 1 to St. 7 and St. 10 were collected on the same day. Usually, sampling was taken once per ten days (Honoki and Hayakawa, 2001). As St. 8 was set at the national acid rain observatory, acid rain data at St. 8 was obtained from Ministry of Environment (Ministry of Environment, 2004). St. T was Toyama Science Museum located at urban area of Toyama City. Evaluate period in this study was from December 1, 1999 to March 17, 2000.

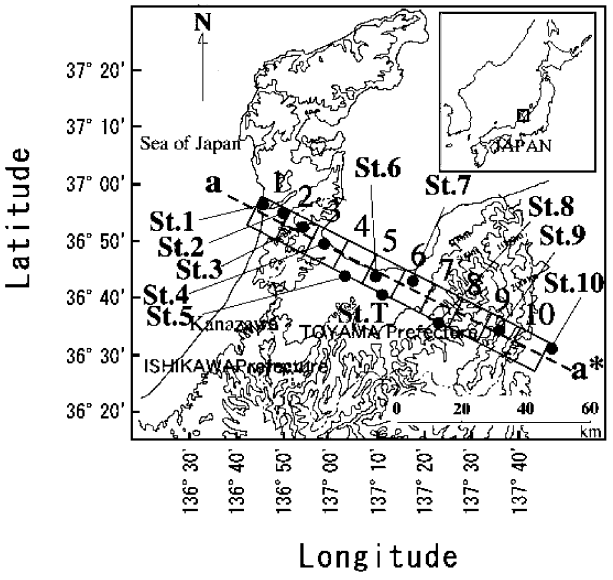

Fig. 1. Location of studying area, sampling stations and evaluation areas in Hokuriku district, Japan.

2.2 Snow boring core sampling and piece selection

Sample at St. 9 (Fig. 1) was snow boring core sample at Murododaira (2450 m a.s.l.), the western slope in Tateyama Mountain Range. At St. 9 (Murododaira), all the snows fallen were accumulated from November to next July. Snow boring core sample was collected on May 8 and 9, 2000. About 9 m depth of the core sample was divided into 50 pieces from the bottom. The snow of bottom layer fell in the mid November 1999, and this snow was granular snow. However, other pieces were compacted snows, and temperature of each piece was subfreezing. Fifty pieces of snow boring core collected at St. 9 were selected by using precipitation data at St. 8 for adjusting observation period, and deposition data of from 6 to 37 pieces of core from the bottom were used in this analysis (Fig 3).

2.3 Analysis

Samples were kept under 10℃ until processed in our analysis. In the analysis, snow samples melted at room temperature. The melted samples were filtrated by membrane filter (pore size of 0.45μm), pH and electric conductivity was analyzed. Anion and cation constituents were analyzed by an ion chromatography (Shimadzu Co., non-suppressor column type). The sea-salt sulfate ($ssSO_4^{2-}$) and non sea-salt sulfate ($nssSO_4^{2-}$) were calculated by eq. (1) and (2).

$$ssSO_4^{2-} = Na^+ \times (SO_4^{2-}/Na^+)_{seawater} \quad (1)$$

$$nssSO_4^{2-} = SO_4^{2-} - ssSO_4^{2-}. \quad (2)$$

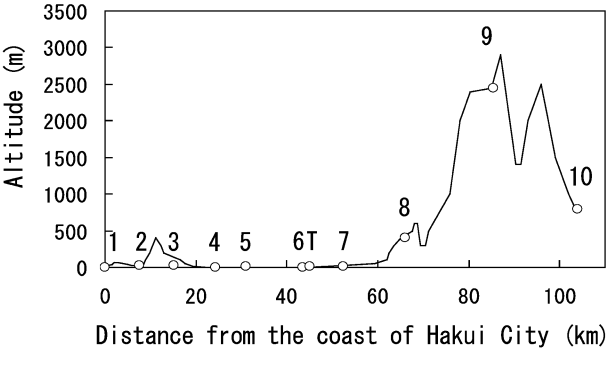

Fig. 2. Altitude profile along the a-a* line in Fig. 1. Location of each sampling station is also superimposed in Fig. 2.

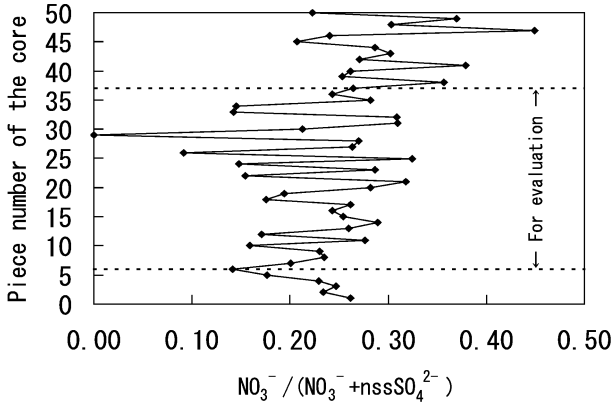

Fig. 3. Nitrate contribution ratio of each piece of snow boring core collected at St. 9.

Where, $(SO_4^{2-}/Na^+)_{seawater}$ is the ratio of SO_4^{2-} concentration to Na^+ concentration in the seawater.

2.4 Evaluation of the Asian Continent origin $nssSO_4^{2-}$ *and nitrate.*

Concentration of $nssSO_4^{2-}$ and NO_3^- of the Asian Continent origin from St. 1 to St. 7 was used from previous report (Honoki and Hayakawa, 2001). In this report, concentration of $nssSO_4^{2-}$ and NO_3^- of the Asian Continent origin at St. 1 was calculated by eq. (3) and (4), respectively (Honoki and Hayakawa, 2001).

$$nssSO_4^{2-}{}_{(Asian)} = \frac{Rc2(1-Rc1)(Ts+Tn)-(1-Rc1)Tn}{(Rc2-Rc1)} \quad (3)$$

$$NO_3^-{}_{(Asian)} = nssSO_4^{2-}{}_{(Asian)} \times \frac{Rc1}{(1-Rc1)}. \quad (4)$$

Here, $nssSO_4^{2-}{}_{(Asian)}$ is the $nssSO_4^{2-}$ concentration of the Asian Continent origin, $NO_3^-{}_{(Asian)}$ is the NO_3^- concentration of the Asian Continent origin, Rc1 is the nitrate contribution ratio $(NO_3^-/(NO_3^- + nssSO_4^{2-}))$ (in equivalent concentration) of acid rain constituents of the Asian Continent origin, Rc2 is the nitrate contribution ratio of those which have domestic origins, and Ts and Tn is the concentration of $nssSO_4^{2-}$ and NO_3^- in precipitation, respectively. As parameters for eq. (3) and (4), 0.14 and 0.59 were used for Rc1 and Rc2 at St. 1, respectively (Honoki and Hayakawa, 2001). The value of 0.14 for Rc1 was mean value (weighted mean by precipitation) of nitrate contribution ratio of new snow collected at St. 1 (Honoki *et al.,* 2001) and 0.59 for Rc2 was calculated from bulk precipitation observation from Jun 5 to November 9, 1998 at St. 1 (Honoki and Hayakawa, 2001), respectively.

Concentration of sea-salt constituents, for example $ssSO_4^{2-}$, decreased in accordance with the increase in the distance from the seacoast in the plain area (so-called inland effect), and regression equation (5) was calculated.

$$ssSO_4^{2-} = 51.2\exp(-0.022L). \quad (5)$$

Here, $ssSO_4^{2-}$ is the concentration of sea-salt sulfate (unit is μeq l^{-1}), 51.2 is the concentration of $ssSO_4^{2-}$ at St. 1 and L is the distance from the seacoast (km) and −0.022 is the decrease coefficient. The distance from the seacoast is the distance from each station to the seacoast in Hakui City parallel to the a-a* line in Fig. 1

Concentrations of both the $nssSO_4^{2-}{}_{(Asian)}$ and $NO_3^-{}_{(Asian)}$ also indicate inland effect. Moreover, decrease coefficients of the regression equations of those were the same as that of $ssSO_4^{2-}$ (Honoki *et al.,* 2001). From above results, ratio of $nssSO_4^{2-}{}_{(Asian)}$ to $ssSO_4^{2-}$ $(nssSO_4^{2-}{}_{(Asian)}/ssSO_4^{2-})$ and $NO_3^-{}_{(Asian)}$ to $ssSO_4^{2-}$ $(NO_3^-{}_{(Asian)}/ssSO_4^{2-})$ take constant value in plain area. The value of $nssSO_4^{2-}{}_{(Asian)}/ssSO_4^{2-}$ and $NO_3^-{}_{(Asian)}/ssSO_4^{2-}$ at St. 1 was 1.12 and 0.182, respectively (Table 1). Then, concentrations of $nssSO_4^{2-}{}_{(Asian)}$ and $NO_3^-{}_{(Asian)}$ from St. 2 to St. 7 were calculated by using eq. (6) and (7) (Honoki and Hayakawa, 2001).

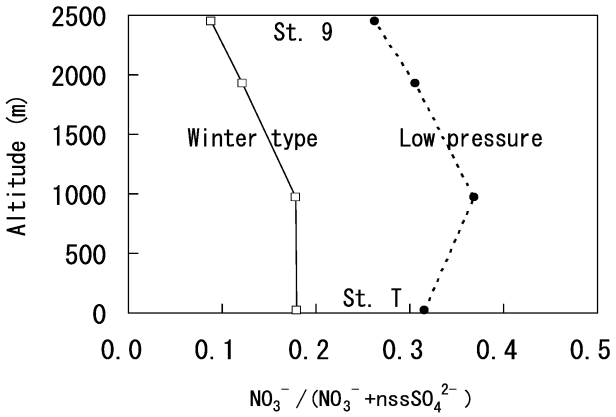

Fig. 4. Changes of nitrate contribution ratio of bulk samples collected between St. T and St. 9. Open box and solid line indicates precipitation by winter type. Solid circle and broken line indicates by low pressure. Plot of altitude at both 970 m and 1930 m is located between St. 8 and St. 9.

$$nssSO_4^{2-}{}_{(Asian)} = ssSO_4^{2-} \times (nssSO_4^{2-}{}_{(Asian)}/ssSO_4^{2-})_{St.1} \quad (6)$$

$$NO_3^-{}_{(Asian)} = ssSO_4^{2-} \times (NO_3^-{}_{(Asian)}/ssSO_4^{2-})_{St.1}. \quad (7)$$

Mean concentrations of $nssSO_4^{2-}{}_{(Asian)}$ and $NO_3^-{}_{(Asian)}$ at St. 8 were calculated by eq. (6) and (7), because $ssSO_4^{2-}$ concentration at St. 8 was the same as the calculated concentration by regression equation (5) (Fig. 4). Mean concentrations of $nssSO_4^{2-}{}_{(Asian)}$ and $NO_3^-{}_{(Asian)}$ at St. 9 and St. 10 were calculated by eq. (3) and (4), because $ssSO_4^{2-}$ concentration at St. 9 and 10 were very different from regression equation (5) (Fig. 4). As a parameter for eq. (3) and (4), Rc1 at St. 9 and St. 10 was 0.10. It was determined by mean of 14 samples data of new snow collected at St. 9. The value of Rc2 at St. 9 and St. 10 was 0.35. This value was calculated from acid rain data in summer (from April 4 to October 2, 2000) at St. T. The ratio of $nssSO_4^{2-}{}_{(Asian)}/ssSO_4^{2-}$ and $NO_3^-{}_{(Asian)}/ssSO_4^{2-}$ at St. T was also calculated by using eq. (3) and (4) to compare these values to those at St. 1 (Table 1). As a parameter for eq. (3) and (4) at St. T, the value of Rc1 and Rc2 was 0.14 and 0.35, respectively.

2.5 Calculation of Deposition in each mesh box

Ten mesh boxes of 10 km squares each were set from the seacoast to 100 km leeward of the northwest winter monsoon (Fig. 1). Deposition of mesh box 1 was calculated by the mean deposition between St. 1 and St. 2 data. Depositions of mesh boxes from 2 to 7 were calculated by the deposition from St. 3 to St. 8, respectively. Deposition of mesh box 8 was calculated by the mean deposition between St. 8 and St. 9.

Table 1. Acid rain observation data at each station (from December 1, 1999 to March 17, 2000).

	(Unit)	St. 1	St. 2	St. 3	St. 4	St. 5	St. 6	St. T	St. 7	St. 8	St. 9	St. 10
Distance from the seacoast*	(km)	0.0	7.6	15.2	24.4	31.0	43.6	45.0	52.4	66.0	85.5	109.8
Amount of precipitation	(mm)	646	768	928	826	770	717	625	730	974	3259	330
$ssSO_4^{2-}$	(μeq l^{-1})	51.2	42.1	33.6	34.1	28.9	18.4	18.0	16.0	10.7	3.2	2.5
$nssSO_4^{2-}$	(μeq l^{-1})	72.8	85.1	62.4	66.7	66.6	65.9	72.1	61.0	51.0	27.1	47.1
$nssSO_4^{2-}{}_{(Asian)}$	(μeq l^{-1})	57.1	47.0	37.5	38.0	32.2	20.6	23.0	17.8	12.0	18.0	13.0
$nssSO_4^{2-}{}_{(Domestic)}$**	(μeq l^{-1})	15.7	38.1	24.9	28.6	34.4	45.4	49.1	43.2	39.1	9.2	34.1
NO_3^-	(μeq l^{-1})	31.9	27.9	23.8	24.8	25.6	22.9	30.2	23.9	19.5	6.9	20.5
$NO_3^-{}_{(Asian)}$	(μeq l^{-1})	9.3	7.6	6.1	6.2	5.2	3.4	3.7	2.9	1.9	2.0	2.1
$NO_3^-{}_{(Domestic)}$***	(μeq l^{-1})	22.6	20.2	17.7	18.6	20.3	19.6	26.5	21.0	17.6	4.9	18.4
$NO_3^-/(NO_3^- + nssSO_4^{2-})$		0.30	0.25	0.28	0.27	0.28	0.26	0.30	0.28	0.28	0.20	0.30
$nssSO_4^{2-}{}_{(Asian)}/ssSO_4^{2-}$		1.12						1.28			5.57	5.30
$NO_3^-{}_{(Asian)}/ssSO_4^{2-}$		0.182						0.208			0.619	0.863

* Distance from the seacoast is the distance from each station to the seacoast in Hakui City parallel to the a-a* line in Fig. 1

** $nssSO_4^{2-}{}_{(Domestic)} = nssSO_4^{2-} - nssSO_4^{2-}{}_{(Asian)}$

*** $NO_3^-{}_{(Domestic)} = NO_3^- - NO_3^-{}_{(Asian)}$

Deposition of Mesh box 9 was calculated by the deposition at St. 9. Deposition of mesh box 10 was calculated by the mean deposition between at St. 9 and St. 10.

3. Results and Discussions

3.1 Nitrate contribution ratios in the pieces of snow boring core

Nitrate contribution ratio (Honoki *et al.,* 2001) in each piece of snow boring core collected at St. 9 is shown in Fig. 3. The x-axis shows nitrate contribution ratio and y-axis shows each piece number of snow boring core. The piece number 1 means the bottom sample and number 50 means the surface sample. Values of the nitrate contribution ratio ranges from 0.00 to 0.32 in the evaluation period.

Figure 4 shows examples of relationship between altitude and nitrate contribution ratios by bulk sample at St. T and St. 9 in October 2003. Cases of precipitation due to low pressure and winter monsoon were superimposed in Fig. 4. Nitrate contribution ratio which indicates high value in the precipitation is due to low pressure, and the ratio which indicates low value in the precipitation is due to winter monsoon at St. 9. Low values of nitrate contribution ratio means that origin of acidic constituents ($nssSO_4^{2-}$ and NO_3^-) are mainly derived from the Asian Continent and high values of the ratio means that origin of acid rain constituents are domestic (Watanabe *et al.,* 2001). Moreover, medium values of the nitrate contribution ratio suggested that both the Asian continent and domestic origin acid rain constituents were mixed in snow. It is suggested from Fig. 3 that precipitation due to both low pressure and winter monsoon fell in winter at St. 9.

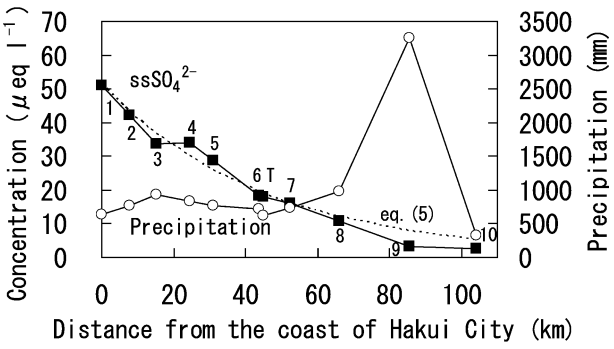

Fig. 5. Amount of precipitation and concentration of $ssSO_4^{2-}$ at each sampling station. The x-axis indicates the distance from each station to the seacoast in Hakui City parallel to the a -a* line in Fig. 1. Solid box and solid line indicates concentration of $ssSO_4^{2-}$. Open circle and solid line indicates amount of precipitation. Broken line indicates regression equation (5) of concentration of the $ssSO_4^{2-}$.

3.2 Precipitation and $ssSO_4^{2-}$ *concentration*

Amount of precipitation and $ssSO_4^{2-}$ concentrations (weighted mean by precipitation) at sampling stations in evaluation period are shown in Fig. 5 and Table 1. The x-axis indicates the distance from each station to the seacoast in Hakui City parallel to the a-a * line in Fig. 1. Amount of precipitation from St. 1 to St. 8 during the sampling period ranged from 646 mm to 974 mm. Amount of precipitation at St. 9 and St. 10 was 3259 mm and 330 mm, respectively. The concentrations of $ssSO_4^{2-}$ at St. 2 to St. 8 decreased with exponential function due to the inland effect. Equation (5) in Fig. 5 indicated regression function of $ssSO_4^{2-}$ concentration in plain area, which was calculated based on the data from St. 2 to St. 7 (Honoki and

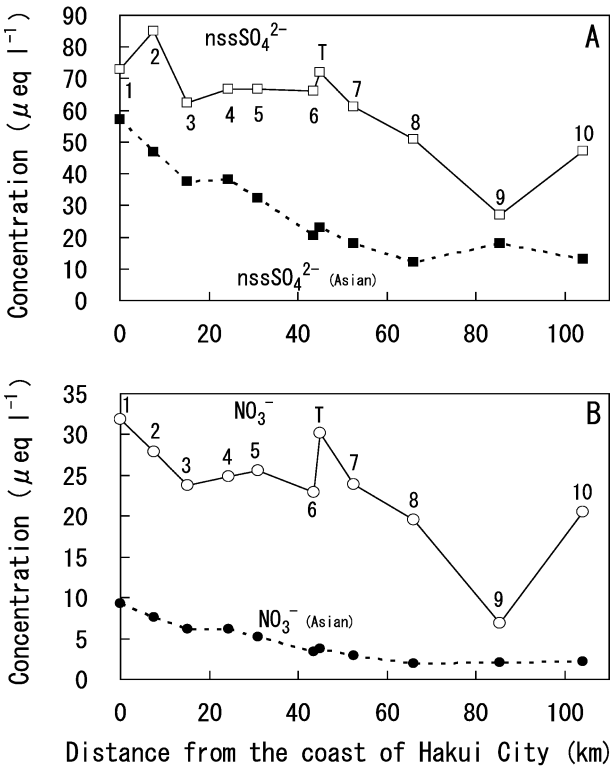

Fig. 6. Concentration of $nssSO_4^{2-}$ and NO_3^- at each sampling station. Plate A and B show concentrations of $nssSO_4^{2-}$ (open box and solid line) and NO_3^- (open circle and solid line) at each sampling station, respectively. Concentrations of $nssSO_4^{2-}{}_{(Asian)}$ (solid box and broken line) and $NO_3^-{}_{(Asian)}$ (solid circle and broken line) at each sampling station are also superimposed in plate A and B, respectively.

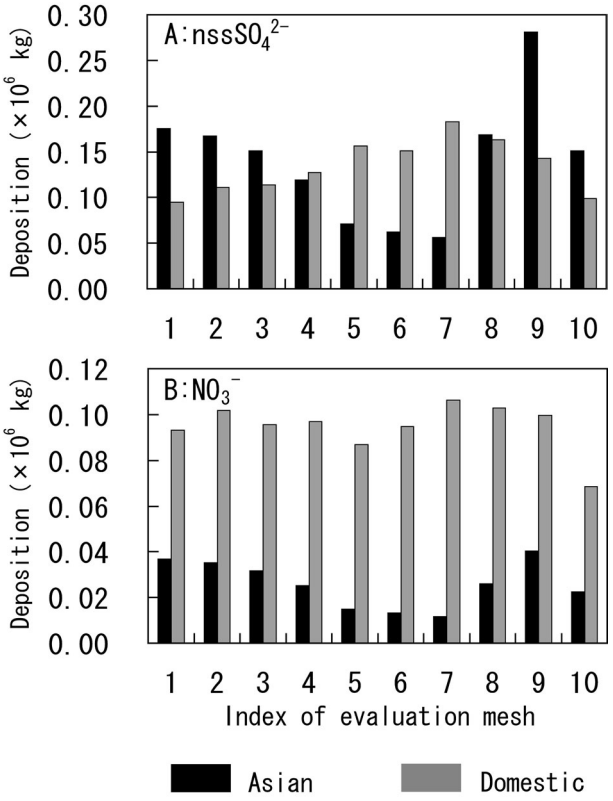

Fig. 7. Depositions of $nssSO_4^{2-}$ and NO_3^- of both the Asian Continent and domestic origins in each mesh box. Plate A and B show depositions of $nssSO_4^{2-}$ and NO_3^-, respectively. Solid bar and half tone bar indicate depositions of the Asian Continent origin and domestic origin in each mash (each area is 100 km^2), respectively.

Hayakawa, 2001). Concentration of $ssSO_4^{2-}$ at St. 9 was lower in observation than that in calculation by regression equation (Fig. 5). This result suggested that low concentration of $ssSO_4^{2-}$ at St. 9 was caused by the altitude effect. Sea salt sulfate concentration at St. 10 was the lowest compared to the other stations.

3.3 Concentrations of $nssSO_4^{2-}$ *and* NO_3^-*, and ratios of* $nssSO_4^{2-}{}_{(Asian)}/ssSO_4^{2-}$ *and* $NO_3^-{}_{(Asian)}/ssSO_4^{2-}$

Concentrations of $nssSO_4^{2-}$ and $nssSO_4^{2-}{}_{(Asian)}$ are shown in Fig. 6A and Table 1. Concentrations of NO_3^- and $NO_3^-{}_{(Asian)}$ are shown in Fig. 6B and Table 1. In Fig. 6A and 6B, the x-axis indicates the distance from each station to the seacoast in Hakui City parallel to the a-a* line in Fig. 1. The concentrations of $nssSO_4^{2-}$ and NO_3^- at St. 9 were the lowest compared to the other stations (Fig. 6A, 6B and Table 1). However, the concentration of $nssSO_4^{2-}{}_{(Asian)}$ was higher at St. 9 than at St. 8, while $NO_3^-{}_{(Asian)}$ was lower at St. 9 than those at St. 8 (Fig. 6A, 6B and Table 1).

Ratio of $nssSO_4^{2-}{}_{(Asian)}/ssSO_4^{2-}$ at St. 1, St. T and St. 9 was 1.12, 1.28 and 5.57, respectively (Table 1). Ratio of $NO_3^-{}_{(Asian)}/ssSO_4^{2-}$ at St. 1, St. T and St. 9 was 0.182, 0.208 and 0.619, respectively (Table 1). These results suggested that acid rain constituents in precipitation at the high mountain area had a different origin from those in precipitation in plain area.

3.4 Depositions of $nssSO_4^{2-}$ *and* NO_3^- *of both the Asian Continent and domestic origin*

Figure 7A and Table 2 shows depositions of $nssSO_4^{2-}$ of both the Asian Continent and domestic origin in the each mesh (10 km square, Fig. 1A). Depositions of $nssSO_4^{2-}{}_{(Asian)}$ in each mesh decreased with increase in the distance from seacoast, from mesh 1 to 7. However, depositions of $nssSO_4^{2-}{}_{(Asian)}$ was the largest in mesh 9 (in high mountain area) compared to those in the other meshes. Depositions of $nssSO_4^{2-}$ and that of the Asian Continent origin in the all mesh boxes ($10\times100\,km^2=1000\,km^2$) were 2.743×10^6 kg and 1.402×10^6 kg, respectively (Table 2). Depositions of $nssSO_4^{2-}$ of the domestic origin in the all mesh boxes were 1.341×10^6 kg. Sulfur dioxide emission in China is estimated about 20×10^9 to 25×10^9 kg (Streets *et al.*, 2001). These amounts are about 20 to 25 fold of those in Japan. However, it is revealed from our results that deposition ratio of $nssSO_4^{2-}$ between the Asian

Table 2. Depositions of $nssSO_4^{2-}$ and NO_3^- of both the Asian Continent and domestic origin in each mesh.

	$nssSO_4^{2-}$			NO_3^-		
evaluation mesh No.*	Observed deposition ($\times 10^6$ kg)	Asian Continent ($\times 10^6$ kg)	Domestic origin ($\times 10^6$ kg)	Observed deposition ($\times 10^6$ kg)	Asian Continent ($\times 10^6$ kg)	Domestic origin ($\times 10^6$ kg)
1	0.270	0.175	0.094	0.130	0.037	0.093
2	0.278	0.167	0.111	0.137	0.035	0.102
3	0.264	0.151	0.114	0.127	0.032	0.096
4	0.246	0.119	0.127	0.122	0.025	0.097
5	0.227	0.071	0.156	0.102	0.015	0.087
6	0.214	0.063	0.151	0.108	0.013	0.095
7	0.239	0.056	0.183	0.118	0.012	0.106
8	0.332	0.169	0.163	0.129	0.026	0.103
9	0.424	0.281	0.143	0.140	0.040	0.100
10	0.250	0.151	0.099	0.091	0.022	0.069
Sum from 1 to 10**	2.743	1.402	1.341	1.204	0.257	0.947

*Each mesh area is 100 km^2.

**Sum of areas from mesh 1 to mesh 10 are $10 \times 100\ km^2 = 1000\ km^2$.

Continent origin and domestic origin was almost 1: 1.

Figure 7B and Table 2 shows depositions of NO_3^- of both the Asian Continent and domestic origin in each mesh (10 km square, Fig 1A). Total depositions of NO_3^- and those of the Asian Continent origin in the all mesh boxes were 1.204×10^6kg and 0.257×10^6kg, respectively. Depositions of NO_3^- of domestic origin in the all mesh boxes were 0.947×10^6 kg (Table 2).

4. Conclusion

The depositions of $nssSO_4^{2-}$ and NO_3^- that are originated both in the Asian Continent and domestic regions like Hokuriku district, central Japan were evaluated in 10 mesh boxes of 10 km square from the seacoast to the leeward of northwest winter monsoon. Depositions of $nssSO_4^{2-}$ and those of the Asian Continent origin in the all mesh boxes (1000 km^2) from December 1, 1999 to March 17, 2000 were 2.743×10^6 kg and 1.402×10^6 kg, respectively. Depositions of NO_3^- and those of the Asian Continent origin in the all mesh boxes were 1.204×10^6 kg and 0.257×10^6 kg, respectively. Deposition ratio of $nssSO_4^{2-}$ of the Asian Continent origin to those of domestic origin in studying area was almost 1: 1, though sulfur oxide emission in China was 20 to 25 fold of those in Japan. Moreover, it was considered that acid rain constituents in precipitation at high mountain area had a different origin from precipitation in plain area.

Acknowledgments

A part of this study was funded by the Ministry of Education, Science, Sports and Culture and Toyama Prefecture Museum association. The authors thank to Toyama Prefecture agency, Toyama Forest administration office and Tateyama Kurobe Kanko Co. for admittance of investigation on Mt. Tateyama. The authors thank to Pension Beach Noto, Hotel Green Wood Noto, Buddhist temple Gyoninji, Himi Seaside Botanical Garden, Toyama city Hachiman Branch office, Toyama city Sango Branch office, Murodo-Sannso and Omachi Energy Museum for collecting samples.

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