Long-term trends for nitrate and sulfate ions in snowcover on Hokkaido, northern Japan

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Abstract

On the island of Hokkaido in northern Japan, a survey of air pollutants in snowpack has been carried out every four years from 1988 to 2012. In this report Hokkaido was divided into four areas, and we determined the concentration and deposition of non-sea salt (nss) sulfate, ammonium and nitrate ions in each. Transported air pollution was found to have accumulated in the snowcover, especially on the Sea of Japan side (JS). The amount of nss-sulfate decreased continuously after 1988, but nitrate increased after 2000, especially in JS, from 3.7 meq m⁻² (2000) to 5.3 meq m⁻² (2012). Nitrogen deposition (nitrogen in ammonium and nitrate) was estimated at 8.9 meq m⁻² (1.2 kg N ha⁻¹) in JS in 2012. The nitrate/nss-sulfate concentration ratio shows a clear increasing trend. Previous studies have found that sulfur emission in China decreased after 2006. The present result appears to reflect the change in winter emissions in northern/northeastern China.

Key words: East Asia, Long range transported air pollution, Nitrogen deposition, NO₃⁻/nss-SO₄²⁻ ratio, Snowcover.

1. Introduction

Nitrate (NO₃⁻) and non-sea salt sulfate (nss-SO₄²⁻), derived respectively from nitrogen oxides (NO_x) and sulfur oxides (SO₂), are the major components of acid deposition. They have been studied as factors contributing to a regional-scale environmental problem since the 1970s. Recent studies have found that nitrogen deposition alters and often decreases the diversity of plant communities. (Bobbink *et al.*, 2010; Galloway *et al.*, 2008; Stevens *et al.*, 2004, 2011) The effect of nitrogen deposition is likely to be greater in mountainous areas because of their nutrient-poor soils and the shorter vegetation period. (Roth *et al.*, 2013)

The long-range transportation of air pollution in East Asia is an international problem based in rapid economic growth. In Japan, the effect of transported air pollution is especially seen in winter months. China's contribution of emissions to the concentration of sulfate particles in Japan is 50–70%, with a winter maximum of approximately 65–80% (Aikawa *et al.*, 2010), and the concentration in rainwater and wet deposition of air pollutants is higher in winter. (Tomoyose *et al.*, 2009) A chemical transport model study also found that the Chinese contribution has been increasing, from 40% in the 1980s to 50% in the early 2000s; this upward trend was greater on the Sea of Japan side. (Kuribayashi *et al.*, 2012)

The NO_x emission in China has been increasing rapidly, and in the absence of pollution control, it will continue to increase in conjunction with energy consumption (*e.g.*, Zhao *et al.*, 2013; Xing *et al.*, 2011). Emission of SO₂ decreased after 2006. (Lu *et al.*, 2011) However, NO_x emission has been increasing in the North China Plain and in Northeast China, and emissions are

greater from October to May than from June to September. (Gu *et al.*, 2013)

Long-term trends in air pollutant deposition in Japan have been previously studied. Calculated and observed NO₃⁻ deposition both show a slight increase from 1988 to 2002. (Noguchi *et al.*, 2007) The trend in the NO₃⁻/nss-SO₄²⁻ concentration ratio in precipitation from 1970 to 2011 reflects the changes in NO_x and SO₂ emissions in China. (Fujita, 2013) A similar study found that the increase in the NO₃⁻/nss-SO₄²⁻ concentration ratio in precipitation (namely 0.57%/year during 2000–2005 and 2.58%/year during 2006–2011) correspond to the changes in the NO_x/SO₂ emissions ratio in China. (Itahashi *et al.*, 2014) China's contribution to NO₃⁻ wet deposition in northern Japan was estimated to be 35–59% (Morino *et al.*, 2011); the contribution from the northern and northeastern regions in China to sulfur deposition in Hokkaido was significant. (Inoue *et al.*, 2005) These authors also reported that more air pollutants are transported from China in the winter.

In Hokkaido, which is located in the northern part of Japan, the land is covered by snow in the winter, for half of each year. Air pollutants that have accumulated in the snowcover are released during the short period of snowmelt. (Johannessen and Henriksen, 1978) This release has a significant effect on the ecosystem due to episodic acidification. (Lepori *et al.*, 2003) A previous study reports that 70% of hydrogen ions stored in Hokkaido's snowcover are released in a 20% snowmelt. (Sato *et al.*, 1999)

Particularly in winter, the dominant wind direction is westerly or northwesterly, so the deposition of air pollution in the snowcover is greatly affected by the Asian continent. Since 1988, the present authors have studied the spatial distribution and deposition of atmospheric pollutants in the snowcover. (Noguchi *et al.*, 2005, 2001; Yamaguchi *et al.*, 2009) Below, we examine long-term trends in the nitrate (NO₃⁻) and non-sea salt sulfate (nss-SO₄²⁻) concentration and deposition.

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2. Methods

2.1 Sampling periods and sites

Snow sampling was conducted every four years (1988, 1992, 1996, 2000, 2004, 2008, and 2012), mainly in February, which is the time of greatest snowcover. (Yamaguchi *et al.*, 2009) To reduce contributions from local emission sources, the sample sites were selected in rural areas, away from main roads carrying traffic and artificial disturbances.

2.2 Sampling method

Snow samples were collected from all depths using a stainless steel pipe sampler (diameter 5.0 or 5.5 cm) at 80~62 sites and packed in plastic bags, which had been washed with deionized water. The number of sites varied from year to year due to traffic or weather conditions. The depth of the snowcover at the site was recorded and was used to calculate the snow volume and density. The samples were melted at room temperature and weighed. After being passed through a membrane filter (pore size: 0.2μm), the electrical conductivity and pH were promptly measured using a conductivity meter and a pH meter. The samples were then stored in a refrigerator. The major inorganic ions, sulfate (SO_4^{2-}), nitrate (NO_3^-), chloride (Cl^-), ammonium (NH_4^+), calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+) and potassium (K^+) were measured by ion chromatography or automated colorimetry or by

atomic absorption spectrography. Non-sea salt sulfate and calcium (nss-SO_4^{2-} and nss-Ca^{2+}) were calculated using Na^+ as a sea salt index, as follows:

$$\text{nss-SO}_4^{2-} (\text{mol L}^{-1}) = \text{SO}_4^{2-} (\text{mol L}^{-1}) - 0.06028 \times \text{Na}^+ (\text{mol L}^{-1}) \quad (1)$$

2.3 Spatial analysis

The amount of precipitation was calculated from precipitation data taken from AMeDAS (Automated Meteorological Data Acquisition System, Japan Meteorological Agency) during the snowcover period. The snowcover period was defined as falling between the date when the seven-day moving average temperature fell below 0°C and the date when the seven-day moving average temperature rose above 0°C . Spatial distribution maps of the amount of accumulated precipitation were drawn using Kriging interpolation. (Oliver and Webster, 1990) The maps of ion concentrations were also interpolated using the Kriging method. The accumulated ion components deposited in the snowcover was calculated by multiplying the grid data for the amount of precipitation and the concentrations of each ion.

For regional evaluation, we divided Hokkaido into four areas (JS: Sea of Japan, OH: Sea of Okhotsk, PO: Pacific Ocean, and IL: Inland) and calculated the mean concentrations and the deposition of ions in each area, as shown in Fig. 1. Table 1 shows the areas, the mean snow depth as precipitation and the total amount of precipitation in each region. Adding the calculations for the

Table 1. Area of each region, mean snow depth and amount of snow in 2012.

Region	Area (km ²)	Mean snow depth in 2012 (mm)	Snow amount in 2012 (Mm ³)
IL	16390	298	4880
JS	18690	326	6090
OH	14100	177	2500
PO	28570	188	5370
Total	78440	242	19020

Fig. 1. Sampling sites in the 2012 survey and partition into regions.

Fig. 2. Temporal trend in the amount of snow as precipitation and its spatial distribution in 2012.

areas in Table 1 does not result in the total due to rounding.

3. Results

3.1 Amount of precipitation

Figure 2 shows the change in the amount of accumulated precipitation. The amount of precipitation was approximately 250 mm, with a minimum of 180 mm occurring in 2008. In 2012 the amount of precipitation was 242 mm. Region by region, the amount of precipitation was 177 mm in OH, 188 mm in PO, 326 mm in JS and 298 mm in IL. The amount in the JS and IL regions was usually high (Table 1, Fig. 2).

3.2 Concentration and accumulated deposition of ion components

The temporal trend of the regional mean concentration ($\mu\text{eq L}^{-1}$) and the deposition (meq m^{-2}) of each ion in the snowcover are shown in Table 2 and Fig. 3. As an example, Fig. 4 shows the spatial distribution of the concentration and the deposition of each ion in 2012.

3.2.1 Concentration

The nss-SO_4^{2-} concentration was high in the JS region and the NO_3^- concentration was high in JS and IL. The NH_4^+ concentration did not vary from region to region (Fig. 3, Table 2). The NO_3^- concentration held steady at about $10 \mu\text{eq L}^{-1}$ from 1988 to 2000, and then increased, especially in IL and JS, to 12–21 $\mu\text{eq L}^{-1}$.

The nss-SO_4^{2-} concentration fell from 1988 to 2000 and rose temporarily in 2008. The amount of precipitation affects the concentrations of atmospheric pollutants. (Barrie, 1985) The high concentration of air pollutants in 2008 is therefore believed to be due to the small amount of precipitation in that year.

3.2.2 Deposition

The deposition of NO_3^- was steady below 4.0 meq m^{-2} in IL and JS in the 1990s but has increased since 2000. In 2012, the deposition of NO_3^- was 4.8 meq m^{-2} ($0.67 \text{ kg N ha}^{-1}$) in IL and 5.3 meq m^{-2} ($0.74 \text{ kg N ha}^{-1}$) in JS (Table 2, Fig. 3b).

Deposition of NO_3^- in PO and OH ($1.3\text{--}2.8 \text{ meq m}^{-2}$) was about half of that in IL and JS, although they also showed an increasing trend after 2000. The fall in PO in 2008 was due to the small amount of precipitation that year. Ammonium deposition did not show any clear temporal trend. It fell until 1996, then increased, and decreased slightly in 2012. Nitrogen deposition ($\text{NH}_4^+\text{-N} + \text{NO}_3^-\text{-N}$) in 2012 ranged from 3.0 meq m^{-2} (OH, 0.4 kg N ha^{-1}) to 8.9 meq m^{-2} (JS, 1.2 kg N ha^{-1}); see Table 2.

Deposition of nss-SO_4^{2-} was greatest in all regions in 1988. Since then it has been falling continuously. For example, it was 9.4 meq m^{-2} (3.0 kg S ha^{-1}) in JS and had fallen steadily to half that value by 2012.

3.2.3 $\text{NO}_3^-/\text{nss-SO}_4^{2-}$ concentration ratio

The temporal trend in the $\text{NO}_3^-/\text{nss-SO}_4^{2-}$ concentration ratio in

Table 2. Regional mean concentration ($\mu\text{eq L}^{-1}$) and deposition (meq m^{-2}) of each ion in snow cover from 1988 to 2012.

		NH ₄ ⁺								NO ₃ ⁻								nss-SO ₄ ²⁻							
		1988	1992	1996	2000	2004	2008	2012	1988	1992	1996	2000	2004	2008	2012	1988	1992	1996	2000	2004	2008	2012			
IL	(μeq L ⁻¹)	8.8	12.8	8.2	9.1	10.5	14.7	9.7	10.3	12.2	9.5	11.1	13.8	21.7	16.2	24.0	23.7	18.9	14.5	16.1	20.6	12.7			
	(meq m ⁻²)	3.1	3.6	2.4	3.3	3.8	3.7	2.9	3.6	3.7	2.9	3.8	4.9	5.6	4.8	8.2	6.6	5.5	5.2	5.8	5.2	3.8			
JS	(μeq L ⁻¹)	14.5	13.6	8.9	11.0	12.4	15.5	11.2	9.2	11.1	10.1	10.1	12.4	19.9	16.7	25.5	29.4	25.9	19.6	21.1	22.7	16.9			
	(meq m ⁻²)	5.3	3.9	2.6	4.0	4.1	4.0	3.6	3.4	3.3	2.9	3.7	4.1	5.2	5.3	9.4	8.4	7.5	7.2	6.9	5.9	5.5			
OH	(μeq L ⁻¹)	12.8	11.4	6.1	7.9	7.3	13.4	8.0	9.0	7.2	6.6	5.7	6.5	14.3	9.0	27.7	19.9	16.4	10.6	12.6	20.3	11.7			
	(meq m ⁻²)	2.12	1.58	1.02	1.81	2.19	1.86	1.43	1.51	1.36	1.33	1.38	1.97	2.12	1.56	4.6	2.8	2.7	2.4	3.8	2.8	2.0			
PO	(μeq L ⁻¹)	15.0	11.9	7.1	7.7	8.0	14.9	11.6	11.7	10.7	9.2	7.6	8.9	15.6	14.7	32.5	23.2	19.1	10.8	13.2	23.3	15.7			
	(meq m ⁻²)	2.4	1.6	1.2	1.9	2.1	1.5	2.2	1.9	1.4	1.4	2.0	2.4	1.6	2.8	5.1	3.2	3.3	2.7	3.5	2.2	2.9

Fig. 3. (a) Mean concentration ($\mu\text{eq L}^{-1}$) and (b) deposition (meq m^{-2}) of each ion in snow cover at each region from 1988 to 2012.

Fig. 4. Estimated spatial distribution of (a) concentrations ($\mu\text{eq L}^{-1}$) and (b) deposition (meq m^{-2}) of nss-SO_4^{2-} , NO_3^- and NH_4^+ in snowcover in 2012.

each region is shown in Fig. 5. After 1996 all the regions showed clear increases, which were largest in IL (1.28 in 2012). The increase was smallest in OH, at 0.76 in 2012. The annual mean rate of increase was 4.6% in IL and 3.8% in JS. Values were 1.6% in OH and 1.8% in PO. In PO it rose in 2000, to 0.75, and the rate of increase thereafter was small.

4. Discussion

4.1 Concentration and $\text{NO}_3^-/\text{nss-SO}_4^{2-}$ concentration ratio

We observed an increase in the NO_3^- concentration and a de-

crease in the nss-SO_4^{2-} concentration. The $\text{NO}_3^-/\text{nss-SO}_4^{2-}$ ratio has therefore been increasing throughout the study period; the increase we found is greater than in a previous study. (1.4%, Fujita, 2013) One previous study found that the $\text{NO}_3^-/\text{nss-SO}_4^{2-}$ ratio in Japan increased after 2006 (Itahashi *et al.*, 2014) in a manner consistent with the change in the SO_2 emission in China (Lu *et al.*, 2011). The differing values found for nss-SO_4^{2-} change in the present study could be due to the geographical location of Hokkaido relative to China and seasonal differences. The contribution of north and northeast China to sulfur deposition in Hok-

Fig. 5. Long-term trend in the $\text{NO}_3^-/\text{nss-SO}_4^{2-}$ concentration ratio in snow cover from 1988 to 2012.

kaido was estimated to be 41%; these are the areas that contribute most. (Inoue *et al.*, 2005) The recent increasing NO_x trend in northern and northeastern China in the colder seasons was also larger than in other areas in China from 2005–2010. (Gu *et al.*, 2013) Observations in Japan also found that air pollution transported from the Asian continent was significant during winter. (Aikawa *et al.*, 2010; Tomoyose *et al.*, 2009; Var *et al.*, 2000) The present study is limited to the winter period, and our results could reflect changes in SO_2 and NO_x emission in the northern/northeastern part of China in winter.

The $\text{NO}_3^-/\text{nss-SO}_4^{2-}$ ratio has been above 1.0 in IL and is also close to 1.0 in JS and PO. We deduce that the dominant component of acidification in winter is changing from SO_2 to NO_x .

4.2 Deposition

Nitrogen deposition in 2012 was greatest in JS (8.9 meq m^{-2} (1.2 kg N ha^{-1})); see Table 2. The observed annual wet nitrogen deposition at Rishiri Island, located in JS, was $29 \text{ mmol m}^{-2} \text{ y}^{-1}$ ($4.0 \text{ kg N ha}^{-1} \text{ y}^{-1}$) in 2012 (Network Center for EANET, 2013). Nitrogen deposition in the snowcover in JS was close to one third of the annual wet nitrogen deposition.

The snowmelt period in Hokkaido lasts about two months, from mid-March to mid-May. Nitrogen stored in the snowcover will be eluted during this period. Furthermore, the chemicals in the snow are concentrated in the early snowmelt water (Johannessen and Henriksen, 1978; Satoh *et al.*, 1999; Brooks and Williams, 1999) and affect the ecosystem via episodic acidification. (Lepori *et al.*, 2003) If there continues to be an absence of air pollution regulations in China, NO_x emission will continue to increase as well. (Zhao *et al.*, 2013; Xing *et al.*, 2011)

The concentration of atmospheric pollution was large in the cloud/fog and rime, and deposition in correspondence with these was also significant in mountainous areas. (Igawa *et al.*, 2002; Nagafuchi *et al.*, 2001; Yamaguchi *et al.*, 2015) We excluded the mountainous area from our study because of the difficulty of sampling. The deposition of atmospheric pollution might therefore be larger than that found in this study. Furthermore, sensitive plants or microorganisms in high areas are strongly influenced by the increase in nitrogen deposition (e.g. Fenn *et al.*, 2003; Williams *et al.*, 1996). The snowcover in the mountainous areas should be studied further.

5. Conclusion

Deposition of air pollutants in the snowcover was significant on the Sea of Japan side of Hokkaido island. Nitrate deposition increased after 2000, whereas nss-SO_4^{2-} deposition has been decreasing throughout the study period. Nitrogen deposition on the Sea of Japan side was estimated to be 8.9 meq m^{-2} (1.2 kg N ha^{-1}), corresponding to one third of the annual nitrogen wet deposition. The $\text{NO}_3^-/\text{nss-SO}_4^{2-}$ concentration ratio increased continually throughout the study period, from 1988 to 2012. A possible explanation is that the changes are due to increased regional emission in the northern and northeastern China. The increase in nitrogen deposition is believed to affect the ecosystem, especially in the mountainous areas on the Sea of Japan side of Hokkaido island.

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