

Report

## Regional distribution of chemical tracers in snow cover along the route from S16 to Dome Fuji Station, east Dronning Maud Land, Antarctica

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**Abstract:** We show and discuss the regional distributions of  $\text{Cl}^-$ ,  $\text{Na}^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $^7\text{Be}$  concentrations in snow cover along the route from S16 to Dome Fuji Station in east Dronning Maud Land, Antarctica. The concentrations of  $\text{Cl}^-$  and  $\text{Na}^+$  at the coast exponentially decreased with distance from the coast up to 200 km inland. Between 200–1000 km inland, the concentrations of  $\text{Na}^+$  were nearly constant. On the other hand, the concentrations of  $\text{Cl}^-$  increased with distance beyond 750 km inland. Furthermore, the contribution factors between  $\text{Cl}^-$  and  $\text{Na}^+$  decreased and the  $\text{Cl}^-/\text{Na}^+$  ratio and the  $\text{nssCl}^-/\text{Cl}^-$  ratio increased toward the interior of the continent. These results indicate that sea salt aerosols are transported from the coastal region toward the inland region of the Antarctic continent, and that a source of  $\text{Cl}^-$  other than sea salt exists in the inland plateau of Antarctica. The concentrations of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $^7\text{Be}$  in snow were relatively high in the coastal region and the inland plateau region compared with those in the katabatic-wind region.

### 1. Introduction

Antarctic ice sheet records atmospheric deposition continuously in time and space. Because of the remoteness of Antarctica from major gases and aerosol sources, the information recovered from the ice sheet generally has a global significance. Therefore, vertical analyses of the ice sheet provide us with information about past climatic change (e.g. Delmas, 1992) and horizontal analyses of the ice sheet provide us with knowledge regarding long-range transport of airborne materials (e.g. Kamiyama *et al.*, 1989). The Japanese Antarctic Research Expeditions (JARE) have conducted many glaciological observations including the Dome Fuji Project (Dome-F Coring Group, 1998) on the ice sheet in east Dronning Maud Land during the past several decades. The study route to Dome Fuji Station, 1000 km inland, has been established through these observations (Ageta *et al.*, 1989; Kamiyama *et al.*, 1996). The 40th JARE (1998–2000) carried out three inland expeditions for glaciological investigation along this study route. In this report, we show and discuss the regional distributions of  $\text{Cl}^-$ ,  $\text{Na}^+$ ,

$\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $^7\text{Be}$  concentrations in snow cover along the route during the austral summer and winter seasons.

## 2. Methods

A map of the study route is shown in Fig. 1. The traverse from the coast, S16 ( $69^\circ 02' \text{S}$ ,  $40^\circ 04' \text{E}$ , 591 m a.s.l.), to Dome Fuji Station ( $77^\circ 19' \text{S}$ ,  $39^\circ 42' \text{E}$ , 3810 m a.s.l.) was carried out during the austral summer, from December 27, 1998 to February 15, 1999. From August 23 to September 13, 1999, the traverse in the winter season was performed from S16 to Mizuho Station ( $70^\circ 42' \text{S}$ ,  $44^\circ 20' \text{E}$ , 2230 m a.s.l.). Snow samples in this study were collected during these two inland expeditions. In the research area along the route, samples for ion analyses were taken from the snow cover at a depth of 0–2 cm by pushing a 100-ml pre-cleaned plastic bottle into the snow. The sample collection was performed every 10 km along the route. During the return traverse of the expedition, each sampling site was shifted 5 km further along the route than those of the outward traverse. On the traverse to Dome Fuji Station in the summer season, snow samples for  $^7\text{Be}$  measurement were also obtained at 50 km

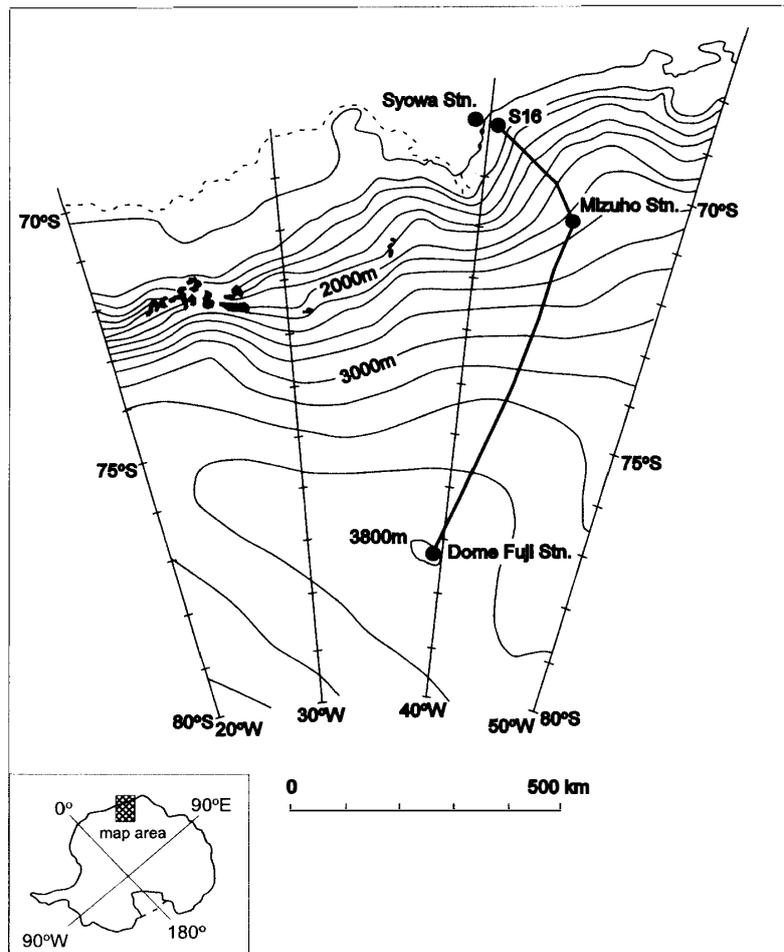


Fig. 1. Map showing the study route in east Dronning Maud Land, Antarctica.

intervals. Samples for  $^7\text{Be}$  analyses were taken from the snow cover at a depth of 0–2 cm by using a stainless shovel to fill a 4-l pre-cleaned plastic bottle. Samples were analyzed at Syowa Station ( $69^\circ 00'S$ ,  $39^\circ 35'E$ , 29 m a.s.l.) immediately after the inland expeditions. The concentrations of ionic species and the activities of  $^7\text{Be}$  were measured by ion chromatography and  $\gamma$ -spectrometry, respectively. Beryllium-7, with a half-life of 53 days, is a naturally occurring cosmogenic radionuclide, which is produced by nuclear spallation reactions between high energy cosmic-ray and atmospheric nuclei. The production rate of  $^7\text{Be}$  increases with altitude and reaches a maximum at 15 km at higher latitudes, and approximately 70% of the production of  $^7\text{Be}$  occurs in the stratosphere (Lal *et al.*, 1958, 1960).

### 3. Results and discussion

The concentrations of  $\text{Cl}^-$  and  $\text{Na}^+$  in snow cover along the study route are shown in Fig. 2. Variation of the altitude is also indicated in the figures. There were no significant differences between the results of the summer and winter expeditions, and between the summer results of the outward and return traverses to and from Dome Fuji Station, respectively. The concentrations of  $\text{Cl}^-$  and  $\text{Na}^+$  at the coast were approximately  $0.6 \text{ mg kg}^{-1}$  and  $0.3 \text{ mg kg}^{-1}$ , respectively. These values decreased one order of magnitude until about 200 km inland, 2.0 km a.s.l., from S16. This result indicates that sea salt aerosols are transported from the coastal region toward the inland region of the Antarctic continent. The concentrations of  $\text{Na}^+$  were nearly constant between 200–1000 km inland. On the other hand, the concentrations of  $\text{Cl}^-$  increased with distance beyond 750 km inland, 3.6 km a.s.l. We divided the study route into three sections, 0–

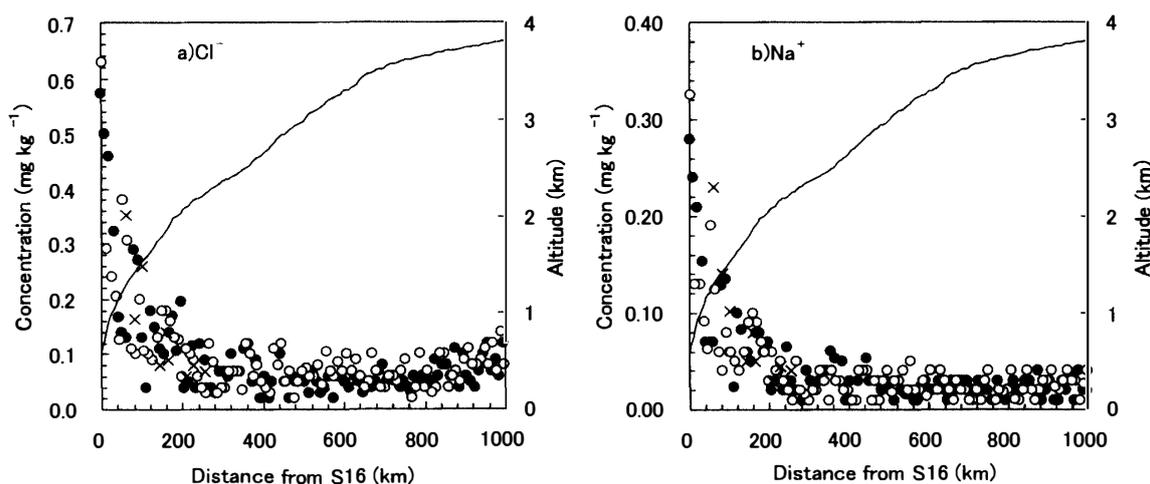


Fig. 2. The distributions of the concentrations of a)  $\text{Cl}^-$  and b)  $\text{Na}^+$  in snow cover along the study route. Solid circles and open circles express the data obtained in outward and return traverses during the summer expedition performed from December 27, 1998 to February 15, 1999, respectively. The crosses represent data obtained in the winter expedition performed from August 23 to September 13, 1999. Variation of the altitude is also indicated by the solid line.

Table 1. The contribution factors between  $\text{Cl}^-$  and  $\text{Na}^+$ , the  $\text{Cl}^-/\text{Na}^+$  and the  $\text{nssCl}^-/\text{Cl}^-$  ratios in three sections of the study route.

Distance from S16 (km)	c.f. <sup>a</sup> (%)	$\text{Cl}^-/\text{Na}^+$ <sup>b</sup>	$\text{nssCl}^-/\text{Cl}^-$ <sup>b</sup>
0–200	97	$2.0 \pm 0.3$	$0.09 \pm 0.13$
200–750	61	$2.6 \pm 1.0$	$0.22 \pm 0.24$
750–1000	33	$3.4 \pm 1.5$	$0.38 \pm 0.24$

<sup>a</sup>Contribution factors between  $\text{Cl}^-$  and  $\text{Na}^+$ .

<sup>b</sup>Errors are  $\pm 1$  standard deviation.

200 km (0.6–2.0 km a.s.l.), 200–750 km (2.0–3.6 km a.s.l.) and 750–1000 km (3.6–3.8 km a.s.l.). This division corresponds to the definition based on the characteristics of snow surface features: a coastal region, characterized by a high frequency of small sastrugi and low frequency of dunes; a katabatic-wind region characterized by the coexistence of small and large sastrugi, dunes and a glazed surface; and an inland plateau region characterized by low frequencies of small sastrugi and dunes (Furukawa *et al.*, 1996). The contribution factors ( $r^2 \times 100$ ) between  $\text{Cl}^-$  and  $\text{Na}^+$ , the average  $\text{Cl}^-/\text{Na}^+$  ratios and the average  $\text{nssCl}^-/\text{Cl}^-$  ratios in each section are calculated and summarized in Table 1. The concentrations of  $\text{nssCl}^-$  (non-sea-salt  $\text{Cl}^-$ ) were obtained as follows:

$$[\text{nssCl}^-] = [\text{Cl}^-] - 1.8[\text{Na}^+], \quad (1)$$

where 1.8 is the  $\text{Cl}^-/\text{Na}^+$  ratio in bulk sea water (Broecker and Peng, 1982), considering that  $\text{Na}^+$  is the best reference element for sea salt. The contribution factors between  $\text{Cl}^-$  and  $\text{Na}^+$  were 97% in the coastal region, 61% in the katabatic-wind region and 33% in the inland plateau, respectively. The decrease in the contribution factor indicates that the correlation between the concentration of  $\text{Cl}^-$  and  $\text{Na}^+$  is weakened toward the inland region. In the coastal region, the average value of the  $\text{Cl}^-/\text{Na}^+$  ratio,  $2.0 \pm 0.3$ , was close to the ratio in bulk sea water, 1.8, and the proportion of  $\text{nssCl}^-$  to total  $\text{Cl}^-$  was less than 10%. On the other hand, the  $\text{Cl}^-/\text{Na}^+$  ratio in the inland plateau region,  $3.4 \pm 1.5$ , was approximately twice the ratio in sea water, and roughly 40% of  $\text{Cl}^-$  in this section was the non-sea-salt fraction. Not only the average values of  $\text{Cl}^-/\text{Na}^+$  and  $\text{nssCl}^-/\text{Cl}^-$ , but also their standard deviations, increased toward the interior of the continent. These results suggest that a source of  $\text{Cl}^-$  other than sea salt exists in the inland plateau of Antarctica. The increase in the concentration of  $\text{Cl}^-$  in the snow inland may be due to an inflow of  $\text{nssCl}^-$  into the Antarctic boundary layer from the stratosphere and the upper troposphere along with atmospheric convergent flow in the interior of the Antarctic continent (e.g. Parish and Bromwich, 1991). Although we cannot identify the origin of  $\text{nssCl}^-$  in this study, the anthropogenic  $\text{Cl}^-$  which is injected into the upper atmosphere in the mid-latitude region or the naturally occurring  $\text{Cl}^-$  which is produced by the photochemical reaction in the Antarctic atmosphere are possibilities.

In Fig. 3, the concentrations of  $\text{Na}^+$  in snow are plotted against a) the distance from S16 up to 300 km and b) the altitude up to 2.5 km. Regression analyses using an exponential formula were performed on these data sets based on the assumption that sea salt aerosols in the atmosphere are removed to the ice sheet in proportion to its

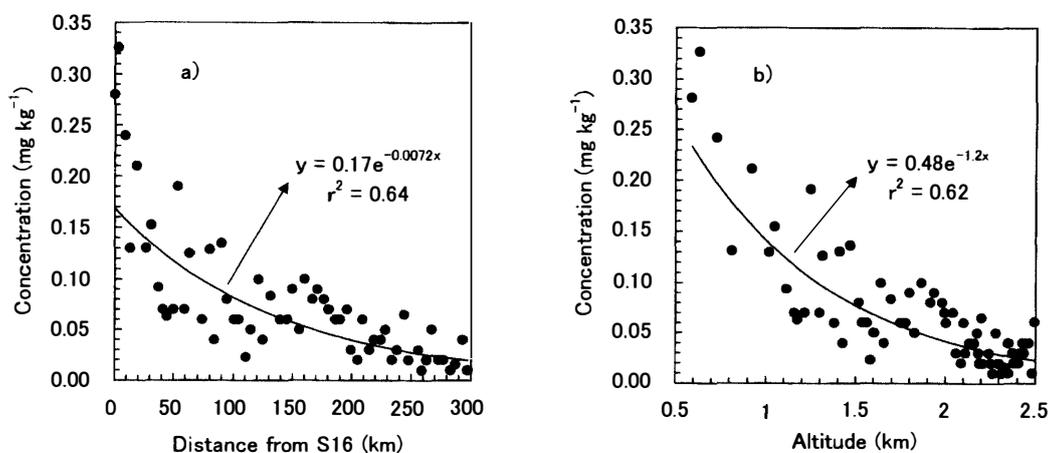


Fig. 3. The concentrations of  $\text{Na}^+$  in snow obtained in the summer expedition plotted against a) the distance from S16 and b) the altitude. The curves in the figures are obtained by the least squares method.

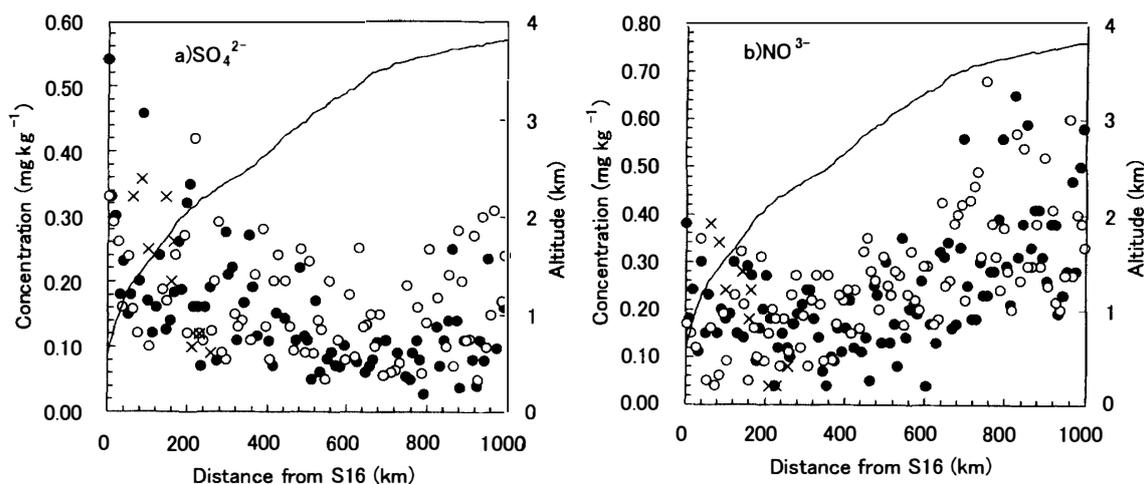


Fig. 4. The distribution of the concentrations of a)  $\text{SO}_4^{2-}$  and b)  $\text{NO}_3^-$  in snow cover along the study route. Meanings of symbols in the figures are same as those of Fig. 2.

concentration in the atmosphere, *i.e.* a first-order removal process. The half-decrease distance of sea salt in the ice sheet expected from the exponential term of the equation in Fig. 3a is approximately 100 km. In the same way, from Fig. 3b, we can recognize that the concentration of sea salt in the ice sheet decreases to half with increasing altitude every 0.6 km. Delmas (1992) suggested that sea salt deposition in polar snow is very high in coastal areas but decreases rapidly inland as a function of elevation rather than of distance from the sea. Observations of aerosol and snow at sites from the same elevation but different distance from the sea are needed to clarify which is the major factor controlling the attenuation of sea salt in the ice sheet.

The distribution of the concentrations of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  in snow along the study route are shown in Fig. 4. For both ions, relatively high concentrations were observed

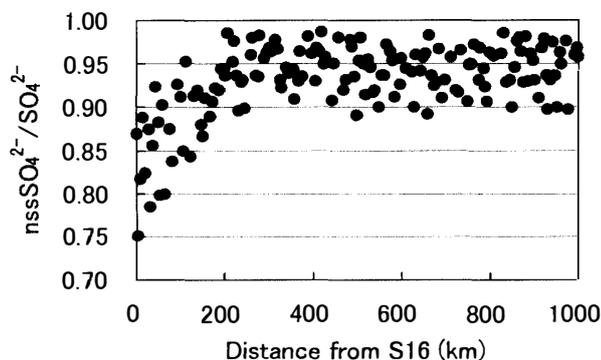


Fig. 5. The regional distribution of the concentration ratios of  $\text{nssSO}_4^{2-}$  to total  $\text{SO}_4^{2-}$ .

in the coastal region and the inland plateau compared with the katabatic-wind region. Although this trend roughly resembles the result for  $\text{Cl}^-$ , the fluctuations in the concentrations of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  were large compared with that of  $\text{Cl}^-$ . We cannot wholly explain the cause of this fluctuation. The fluctuation, if not due to analytical errors, may be due to spatial and/or temporal variability of the atmospheric fluxes of these species. In addition, the concentration of the volatile species  $\text{NO}_3^-$  may be affected by post-depositional migration and re-deposition of the gaseous species (Wagon *et al.*, 1999). The regional distribution of the ratio of concentration of  $\text{nssSO}_4^{2-}$  to that of total  $\text{SO}_4^{2-}$  is shown in Fig. 5. The concentrations of  $\text{nssSO}_4^{2-}$  (non-sea-salt  $\text{SO}_4^{2-}$ ) were obtained as follows:

$$[\text{nssSO}_4^{2-}] = [\text{SO}_4^{2-}] - 0.25[\text{Na}^+], \quad (2)$$

where 0.25 is the  $\text{SO}_4^{2-}/\text{Na}^+$  ratio in bulk sea water (Broecker and Peng, 1982). The proportion of the  $\text{nssSO}_4^{2-}$  to  $\text{SO}_4^{2-}$  was larger than 75% in all samples. This indicates that almost all the  $\text{SO}_4^{2-}$  in the snow cover along the study route originated from a source other than sea salt. The result also indicates that the contribution of sea salt aerosols is relatively high in the coastal region.

The concentrations of  $^7\text{Be}$  in the snow cover along the study route are shown in Fig. 6. The regional variation of the concentration was also high in the coastal region and the inland plateau region. The high concentration of  $^7\text{Be}$  observed in the coastal region cannot be explained by the mechanism whereby the  $^7\text{Be}$  descends from the upper atmosphere inside of a polar vortex and is transported along the continental slope by the katabatic-wind. If  $^7\text{Be}$  is deposited onto the sea surface and then re-suspended into the atmosphere together with sea salt aerosols, the high concentration of  $^7\text{Be}$  in snow at the coastal region would be expected. Let us calculate the concentration of  $^7\text{Be}$  in sea salt aerosols. The concentration of  $\text{Na}^+$  in the coastal region obtained in this study is approximately  $0.2 \text{ mg kg}^{-1}$  (Fig. 2b). This value can be converted to  $18 \text{ mg-sea salt kg}^{-1}$  because the concentration of  $\text{Na}^+$  in seawater is  $11000 \text{ mg kg}^{-1}$  (Broecker and Peng, 1982). The concentration of  $^7\text{Be}$  in the surface water of the South Pacific is  $\sim 10^{-8} \text{ Bq mg}^{-1}$  (Young and Silker, 1980). Consequently, the concentration of  $^7\text{Be}$  in snow originating from sea salt is calculated to be  $\sim 1.8 \times 10^{-7} \text{ Bq kg}^{-1}$ ; but this value does not substantially contribute to the total concentration of  $^7\text{Be}$  in snow of the coastal region. Radioactive decay is another possible cause to explain the regional distribution

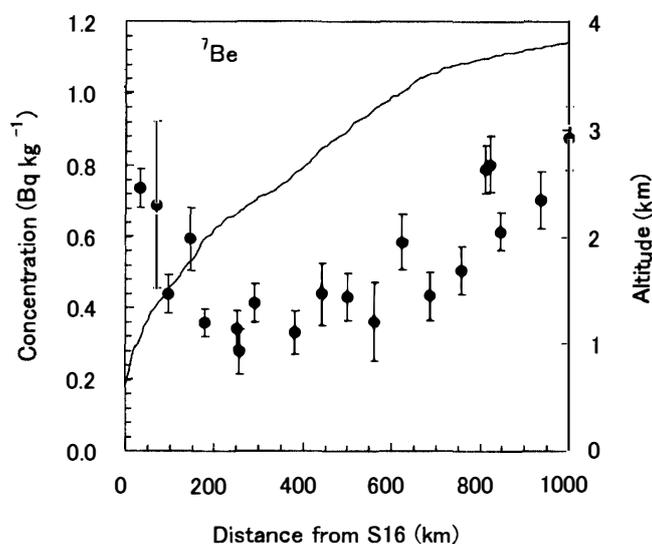


Fig. 6. The distribution of the concentrations of  $^7\text{Be}$  in snow cover along the study route. Error bars are given for each point.

of  $^7\text{Be}$ . The activity of  $^7\text{Be}$ , with a half-life of 53 days, in the katabatic-wind region should be lower than that in the other regions because the accumulation of fresh snow is disturbed by strong winds. At the present time, we do not have any supporting data to explain the regional distribution of  $^7\text{Be}$ . Measurement of  $^{10}\text{Be}$ , with a half-life of  $1.6 \times 10^6$  years, may provide us with some knowledge that may help us to interpret the distribution.

#### 4. Concluding remarks

We have shown the regional distributions of  $\text{Cl}^-$ ,  $\text{Na}^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $^7\text{Be}$  in snow cover along the study route from S16 to Dome Fuji Station in east Dronning Maud Land, Antarctica, and obtained the following results.

1) The concentrations of  $\text{Cl}^-$  and  $\text{Na}^+$  at the coast exponentially decreased with distance from the coast up to 200 km inland. This result may be due to the fact that sea salt aerosols are transported from the coast toward the interior of the continent and are removed onto the Antarctic ice sheet.

2) Between 200–1000 km inland, the concentrations of  $\text{Na}^+$  were nearly constant. On the other hand, the concentrations of  $\text{Cl}^-$  increased with distance beyond 750 km inland. Furthermore, the contribution factors between  $\text{Cl}^-$  and  $\text{Na}^+$  decreased and the  $\text{Cl}^-/\text{Na}^+$  ratio and the  $\text{nssCl}^-/\text{Cl}^-$  ratio increased toward the interior of the continent. These results indicate that a source of  $\text{Cl}^-$  other than sea salt exists in the inland plateau of Antarctica.

3) The concentrations of  $\text{Na}^+$  in snow cover in the ice sheet within the coastal region decreased to half with every 100 km of distance from the coast and 0.6 km of the altitude.

4) Although the concentrations of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  in snow were relatively high in the coastal region and the inland plateau, the fluctuations in the concentrations were

large compared with those of  $\text{Cl}^-$  and  $\text{Na}^+$ .

5) The concentrations of  $^7\text{Be}$  in snow were also high in the coastal region and the inland plateau region. Although we cannot explain its distribution clearly, the effect of re-suspension of  $^7\text{Be}$  from the sea surface does not significantly affect the concentration at the Antarctic coast.

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### References

- Ageta, Y., Kamiyama, K., Okuhira F. and Fujii, Y. (1989): Geomorphological and glaciological aspects around the highest dome in Queen Maud Land, East Antarctica. *Proc. NIPR Symp. Polar Meteorol. Glaciol.*, **2**, 88-96.
- Broecker, W.S. and Peng, T.-H. (1982): *Tracers in the Sea*. Eldigio Press, 26 p.
- Delmas, R.J. (1992): Environmental information from ice core. *Rev. Geophys.*, **30**, 1-21.
- Dome-F Coring Group (1998): Deep ice-core drilling at Dome Fuji and glaciological studies in east Dronning Maud Land, Antarctica. *Ann. Glaciol.*, **27**, 333-337.
- Furukawa, T., Kamiyama, K. and Maeno, H. (1996): Snow surface features along the traverse route from the coast to Dome Fuji Station, Queen Maud Land, Antarctica. *Proc. NIPR Symp. Polar Meteorol. Glaciol.*, **10**, 13-24.
- Kamiyama, K., Ageta, Y. and Fujii, Y. (1989): Atmospheric and depositional environments traced from unique chemical compositions of the snow over an inland high plateau, Antarctica. *J. Geophys. Res.*, **94**, 18515-18519.
- Kamiyama, K., Motoyama, H., Fujii, Y. and Watanabe, O. (1996): Distribution of hydrogen peroxide in surface snow over Antarctic ice sheet. *Atmos. Environ.*, **30**, 967-972.
- Lal, D., Malhorta, P.K. and Peters, B. (1958): On the production of radioisotopes in the atmosphere by cosmic radiation and their application to meteorology. *J. Atmos. Terre. Phys.*, **12**, 306-328.
- Lal, D., Arnold, J.R. and Honda, M. (1960): Cosmic ray produced rates of  $\text{Be}^7$  in oxygen, and  $\text{P}^{32}$ ,  $\text{P}^{33}$ ,  $\text{S}^{35}$  in argon at mountain altitudes. *Phys. Rev.*, **118**, 1626-1632.
- Parish, T.H. and Bromwich, D.H. (1991): Continental-scale simulation of the Antarctic katabatic wind regime. *J. Climate*, **4**, 135-146.
- Wagnon, P., Delmas, R.J. and Legrand, M. (1999): Loss of volatile acid species from upper firn layers at Vostok, Antarctica. *J. Geophys. Res.*, **104**, 3423-3431.
- Young, J.A. and Silker, W.B. (1980): Aerosol deposition velocities on the Pacific and Atlantic oceans calculated from  $^7\text{Be}$  measurements. *Earth Planet. Sci. Lett.*, **50**, 92-104.

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