

IONIC CONSTITUENTS IN AEROSOL PARTICLES AT SYOWA STATION, EAST ANTARCTICA, DURING 1996

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Abstract: This preliminary study was conducted as part of an atmospheric aerosol monitoring program at Syowa Station, East Antarctica. Atmospheric aerosol particles were collected at Syowa Station from April 1996 to January 1997 using a cascade impactor. Although aerosols were collected on selected and limited days of the year, concentrations of CH_3SO_3^- and nssSO_4^{2-} showed distinct summer maxima. Concentration levels of sea-salt components were sporadically high in winter to spring. NO_3^- and $\text{C}_2\text{O}_4^{2-}$ concentrations were high in spring but long-term data are needed to obtain statistically significant seasonal variations.

Results of size-fractionated aerosol samples suggest that sea-salt particles in spring and early summer were more depleted for a coarse fraction ($>2 \mu\text{m}$ diameter) than in winter. CH_3SO_3^- and nssSO_4^{2-} were predominantly found in submicron aerosols whereas the average size fraction of NO_3^- through the observation period was 20% for $>2 \mu\text{m}$, 50% for 2 to $0.2 \mu\text{m}$ and 30% for $<0.2 \mu\text{m}$ diameter.

1. Introduction

Antarctica is far from areas where the air is polluted by heavy human activity. To study natural chemical cycles through the atmosphere, Antarctica is a crucial place as a vast sink area for most water soluble constituents. Major water soluble constituents in Antarctic atmospheric aerosol particles include Na^+ , Cl^- , SO_4^{2-} , NO_3^- , CH_3SO_3^- , and others. These components have also been found in Antarctic snow drift and ice cores (CLAUSEN and LANGWAY, 1989; LEGRAND, 1995; OSADA, 1994, 1996). Long-term monitoring of aerosol chemistry of Antarctic atmosphere has also been published (*e.g.*, TUNCEL *et al.*, 1989; PROSPERO *et al.*, 1991; SAVOIE *et al.*, 1992, 1993; WAGENBACH *et al.*, 1988; WAGENBACH, 1996; KANAMORI *et al.*, 1997) but details of their atmospheric sources, heterogeneous reactions and transport processes are not well known. To deduce these details of atmospheric chemical cycles via aerosols, year-long size-segregated chemistry of aerosols is essential information, yet few studies have been reported on Antarctic aerosols.

To obtain fundamental knowledge of size-segregated aerosol chemistry in coastal Antarctic areas, aerosol samples were collected by a cascade impactor as a part of an aerosol monitoring program at Syowa Station, east Antarctica. This monitoring program includes year-long observations for size-segregated aerosol chemistry (this study), number-size distribution of aerosol particles (IWASAKA *et al.*, in preparation) and Rn as a tracer of crustal material (UI *et al.*, in preparation). The prime objective of this

study was to obtain size distribution and ionic concentration range of aerosol particles through a year at Syowa Station.

2. Samples and Chemical Analysis

Atmospheric aerosol samples of surface air were obtained from April 6, 1996 to January 24, 1997 at an observatory called "Kansoku-tou" in Syowa Station (69°00' S, 39°35' E, 21 m a.s.l.) located in East Antarctica. Aerosol particles in the surface air were collected by a compact 2-stage cascade impactor, having 7 jet nozzles 1.6 mm in diameter for the first stage and 19 0.4 mm jet nozzles for the second stage, made from electrically conductive plastics with a backup filter (47 mm in diameter, teflon membrane filters, 0.5 μm nominal pore size, Advantech Toyo). Assuming density of particulate matter of 1 g/cm^3 , the estimated 50% cut-off aerodynamic diameters was 2 μm for the first stage and 0.2 μm for the second stage at a flow rate of 20 L/min (STP).

The cascade impactor and an air pump (MF-120, M & F Enterprise) were placed within the observatory. The surface air was taken from outside by using tetlon braid hose (3 m long, 9 mm i.d. and 15 mm o.d.) at a height of about 5 m above the ground. The typical sampling duration was 24 hours. Severe blizzard conditions were avoided for the aerosol sampling period. Sample air volumes were about 30 m^3 per sample. Discontinuous 32 sets of aerosol samples were obtained during the study period.

Commercially available polycarbonate membrane filters (manufactured by Costar) were used as a sample impaction sheet because of low background concentration levels of the filter for major ionic species measured. The impaction sheets sampled were placed into a polypropylene 15 ml centrifuge vial (Iwaki) with an air tight cap immediately after the sampling to reduce air contamination of ambient NH_3 in the laboratory, but some samples were accidentally not shut tight until after several hours. Therefore, NH_4^+ data of this study are not included here. The sample vials were packed in polyethylene bags and transported to STEL/Nagoya University, Japan. Before chemical analysis, 14 ml ultra-pure water (18M Ω , MilliPore) was added and treated by ultrasonic digestion for 5 min and immersed for about 12 hours.

Concentration levels of anions and cations are measured by an ion chromatograph (Dionex, model DX-300) equipped with AS11A analytical and AG11 guard columns for anion separation, CS12 analytical and CG12 guard columns for cation separation. An automated sample injector was used for simultaneous analysis of anions and cations in the sample water. Using a 500 μl injection loop for each analytical flow system, procedural blank concentration levels of the polycarbonate and teflon filters used in the field were below or comparable to detection limits for some of ionic species under the present analytical conditions: 0.5 to a few ng/g for anions and cations, but CH_3COO^- , HCOO^- , NO_3^- , Na^+ , NH_4^+ and Ca^{2+} were detectable in procedural blank samples treated the same way as actual aerosol samples. All concentration levels in this study are corrected for the procedural blank values obtained.

3. Results and Discussion

Figure 1 shows stacked-bar plots of atmospheric concentration levels (ng/m^3) in aerosol particles as a function of time from April 1996 to January 1997. CH_3SO_3^- and nssSO_4^{2-} were found mainly in fine particle sizes less than $2 \mu\text{m}$ in diameter. Total concentration levels of CH_3SO_3^- and nssSO_4^{2-} in aerosols show maxima in the austral summer and minima in winter. This is consistent with the results obtained from

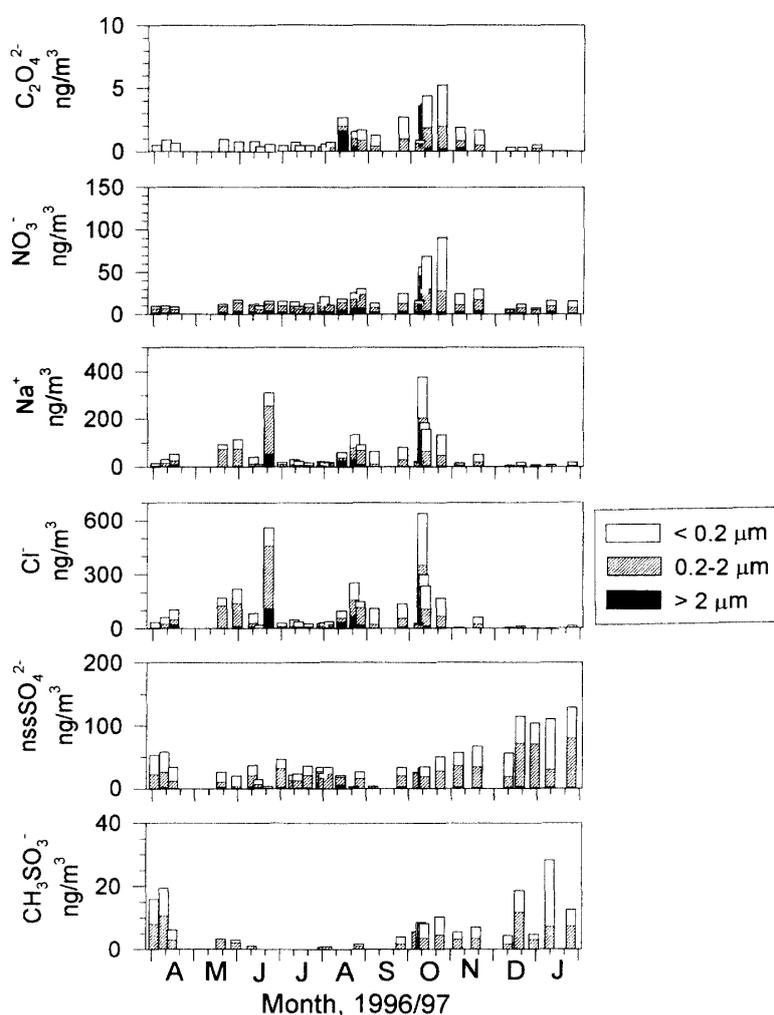


Fig. 1. Stacked-bar plots of concentration levels (ng/m^3) in aerosol particles at Syowa Station as a function of time from April 1996 to January 1997. The top height of each bar represents the total amount of a constituent, and patterns in the bar represent the partition of size distribution. Filled bars represent concentration levels in aerosols larger than $2 \mu\text{m}$ in diameter; hatched bars represent the fraction between 2 and $0.2 \mu\text{m}$ and open bars indicate concentration levels found in the backup filters. Non-sea-salt SO_4^{2-} (nssSO_4^{2-}) concentration is the non-sea-salt originated part of the total SO_4^{2-} content. The nssSO_4^{2-} fraction is calculated from the measured SO_4^{2-} and Na^+ content in the samples as compared with the $\text{SO}_4^{2-}/\text{Na}^+$ weight ratio in sea water (0.25).

snow drift (year of 1986) at Mizuho Station (OSADA, 1996), aerosols collected at Syowa Station by other investigators (years from 1988 to 1990 by KANAMORI *et al.*, 1997; year of 1993 by KOGA *et al.*, 1995), and other coastal stations in Antarctica (Mawson Station, SAVOIE *et al.*, 1993; Neumayer Station, WAGENBACH, 1996). The differences in amplitudes of the total concentration levels for summer to winter averages are about one to two orders of magnitude for CH_3SO_3^- (about 4 to 28 ng/m^3 for summer and mostly below 1 ng/m^3 for winter) and about 5 times for nssSO_4^{2-} (about 30 to 130 ng/m^3 for summer and nearly zero to about 30 ng/g for winter). The peak values in summer for CH_3SO_3^- and nssSO_4^{2-} in this study are slightly lower than the earlier results at Syowa Station (KANAMORI *et al.*, 1997) but are about half to 1/3 of those at Mawson Station (SAVOIE *et al.*, 1993).

Concentration levels of Cl^- and Na^+ are high in winter to spring with large variability and always low in summer. The ranges and seasonal variations of Cl^- and Na^+ concentrations are nearly comparable with the earlier results at Syowa Station (KANAMORI *et al.*, 1997). As will be discussed later in Fig. 2b, both ions are presumably derived from sea-salt particles. Both species show similar size distributions through the year but the distributions are different from those in the usual fresh marine atmosphere; the fraction of coarse particles ($>2 \mu\text{m}$) is less than 50% of the total at Syowa, compared to more than 50% in fresh marine air masses (see for example: HARA *et al.*, 1997). In some samples, especially in summer, $>50\%$ of the total amount is found in particles below $0.2 \mu\text{m}$. Although the sampling artifact due to the long intake line may give underestimations for coarse fractions, differences of dominant size fractions among samples are noticeable. For example, the highest concentration ($377 \text{ ng}/\text{m}^3$) of total Na^+ was obtained on October 10, 1996. The size fractions in the total Na^+ concentration for this sample were 4% for $>2 \mu\text{m}$, 50% for 2 to $0.2 \mu\text{m}$ and 46% for $<0.2 \mu\text{m}$ size ranges. On the other hand, for the second highest concentration ($309 \text{ ng}/\text{m}^3$) of total Na^+ on June 24, 1996, the size fractions were 17%, 65% and 18%, respectively.

Although Syowa Station is located on a coastal island, the distance from sea-salt sources may be quite long, especially in springtime, because sea ice extends about one thousand kilometers to the north in September (PARKINSON and GLOERSEN, 1993). This will provide more favorable conditions to preferentially transport a fine fraction of sea-salt particles during long-range transport to Syowa Station due to differences in deposition velocity with particle size. Although a very limited number of samples were studied, similar depletion of coarse particles has also been reported in KOIDE *et al.* (1981) and KANAMORI *et al.* (1997).

The highest sea-salt concentration with depleted coarse fraction was associated with abnormally low surface O_3 concentration, starting on October 10, for several days at Syowa Station (AOKI *et al.* personal communication). During the low O_3 concentration episode, no significant changes in sulfur species were observed, but relatively higher levels of NO_3^- and $\text{C}_2\text{O}_4^{2-}$ concentration were found in the aerosol samples. Significant changes in chemical (this study) and number-size distributions of aerosol particles (IWASAKA *et al.*, in preparation) during the low O_3 episode at Syowa and comparison with the Arctic cases (STAEBLER *et al.*, 1994; BARRIE *et al.*, 1994) may imply a relationship between aerosol particles and the low O_3 episode in Antarctica.

Seasonal variations of NO_3^- and $\text{C}_2\text{O}_4^{2-}$ concentrations were not clear but maximum total concentrations (91 ng/m^3 for NO_3^- and 5 ng/m^3 for $\text{C}_2\text{O}_4^{2-}$) were obtained for the same sample on October 23, 1996. Concentrations were slightly higher from August to November with peaks in October for both species. Similar broad maxima in spring for aerosol NO_3^- with approximate peak values of about 60 to 90 ng/m^3 were reported at Mawson Station (SAVOIE *et al.*, 1993) and Neumayer Station (WAGENBACH, 1996) but their results showed higher concentrations which extended to midsummer. In the earlier results at Syowa Station (KANAMORI *et al.*, 1997), they reported no distinctive seasonal variation with large year-to-year variations for aerosol NO_3^- but their range of atmospheric concentration was nearly the same as in this study.

Average size fractions of aerosol NO_3^- were 20% for $>2 \mu\text{m}$, 50% for 2 to $0.2 \mu\text{m}$ and 30% for $<0.2 \mu\text{m}$ size ranges. Coarse fractions of NO_3^- were about 10 to 30% of the total concentrations. Deposition of gaseous precursors onto coarse sea-salt particles, which survived after long-range transport, is presumed to be a source of NO_3^- in coarse particles found at Syowa Station. Such gaseous precursors may include HNO_3 and N_2O_5 in the atmosphere (VOGT *et al.*, 1996). As mentioned earlier for the Na^+ and Cl^- , size fraction of sea-salt particles in this study dominates frequently in fine particles. Not only coarse sea-salt particles survived to reach Syowa Station, fine sea-salt particles may also be a good candidate for a carrier of nitrate in fine particles after deposition of gaseous precursors.

For impactor samples except for backup filters, most of the $\text{C}_2\text{O}_4^{2-}$ concentrations were below our detection limit until August and after December. In summer samples, concentration levels of $\text{C}_2\text{O}_4^{2-}$ were lower than those in spring. This is similar to the temporal pattern of NO_3^- data. KAWAMURA *et al.* (1996) reported that $\text{C}_2\text{O}_4^{2-}$ concentration in submicron aerosols ($<0.7 \mu\text{m}$) at Syowa Station in 1991 was higher (10.3 ng/m^3 , a sample taken from November 28 to December 31) in summer than that (3.3 ng/m^3 , August 29 to October 29) in spring. Based on detailed analysis of dicarboxylic acids in aerosols at Syowa Station, they suggested, especially for summer aerosols, that the Antarctic organic aerosols originate from marine-derived lipids and are transformed largely by photochemical oxidations. For autumn to spring, they also suggested that oxalic acid may be produced from chemical transformation during long-range atmospheric transport of aerosols from mid latitudes to the southern Antarctic region.

Figure 2 presents scatter plots for 3 groups having similar seasonal variations: (a) for CH_3SO_3^- and nssSO_4^{2-} , (b) for Cl^- and Na^+ and (c) for NO_3^- and $\text{C}_2\text{O}_4^{2-}$ concentration (nmol/m^3) in aerosols with size information. No clear tendency is seen in differences with sizes for the 3 groups. Figure 2a shows no strong relationship between CH_3SO_3^- and nssSO_4^{2-} regardless of size fractions.

In the coastal areas of Antarctica, marine biogenic dimethylsulfide (DMS) has been considered as the dominant source of submicron nssSO_4^{2-} in aerosols (IWAI *et al.*, 1979; PROSPERO *et al.*, 1991; SAVOIE *et al.*, 1992, 1993). On the other hand, atmospheric oxidation of DMS also produces CH_3SO_3^- (HATAKEYAMA *et al.*, 1982). Assuming that all nssSO_4^{2-} in aerosols are solely originated from DMS and that the atmospheric oxidation with transformation pathways from DMS to submicron aerosol particles results in a relatively constant ratio between CH_3SO_3^- and nssSO_4^{2-} , a pos-

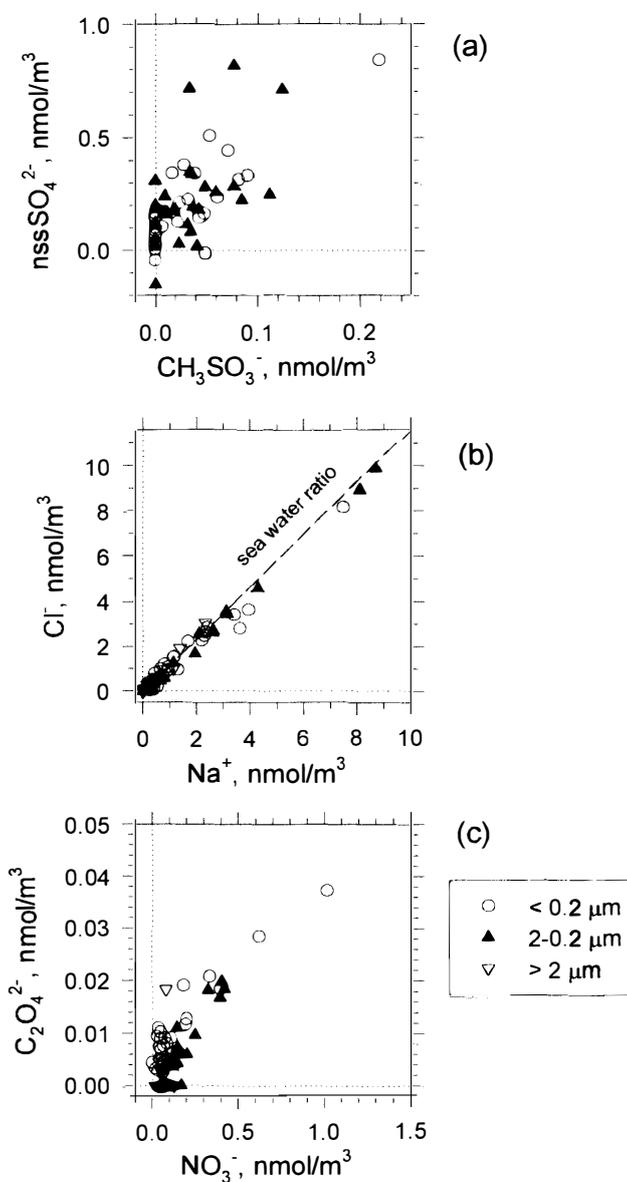


Fig. 2. Scatter plots for 3 groups: (a) for CH_3SO_3^- and nssSO_4^{2-} , (b) for Cl^- and Na^+ and (c) for NO_3^- and $\text{C}_2\text{O}_4^{2-}$ concentration (nmol/m^3) in aerosols with size information.

itive linear relationship should be expected in the scatter plot, but it is not that simple in our case. Possible explanations for this may include an additional contribution of nssSO_4^{2-} from (1) non-DMS sources of continental origin (SAVOIE *et al.*, 1993) in conjunction with vertical entrainment of air from higher free-tropospheric altitude (AYERS *et al.*, 1997; JEFFERSON *et al.*, 1998) and (2) oxidation of volatile sulfur compounds other than DMS such as COS and CS_2 , etc. (ITO, 1993).

Concentration ratios between Cl^- and Na^+ are mostly consistent with the ratio in sea water as seen in Fig. 2b. This suggests that both ionic species are derived from

sea-salt particles. However, ratios for the samples during October and November deviate negatively from the sea water ratio up to 2 nmol/m³ (depleted Cl⁻) on October 23. Similar seasonal trends between 1988 and 1990 have also been reported in KANAMORI *et al.* (1997).

Figure 2c shows an enigmatic relationship between NO₃⁻ and C₂O₄²⁻ concentration. It seems that there is a relationship between these two components during September to November when both concentrations are high. The dominant source of NO₃⁻ in Antarctica has been a matter of controversy (see for examples in HERRON, 1982; LEGRAND and KIRCHNER, 1990; WAGENBACH, 1996; OSADA *et al.*, 1996). SAVOIE *et al.* (1992, 1993) suggested that the primary source of NO₃⁻ in aerosols at Mawson is continental due to a similar temporal pattern of the mean seasonal variations between NO₃⁻ and ²¹⁰Pb. However, the strong relationship between NO₃⁻ and ²¹⁰Pb was not found at Neumayer Station (WAGENBACH, 1996).

As KAWAMURA *et al.* (1996) suggested for autumn to spring samples for submicron aerosols at Syowa Station in 1991, if oxalate in aerosols in this season is to be produced from chemical transformation during long-range atmospheric transport from mid-latitudes to Antarctica, the increase of NO₃⁻ in this period may also be related to the transport process from mid-latitudes, although primary source of NO₃⁻ or its precursors and transport pathways are not well understood. Simultaneous measurements of continental tracer species such as Al and Rn with aerosol NO₃⁻ are needed at Syowa Station to reveal the major sources and transport pathways of aerosol NO₃⁻.

4. Concluding Remarks

Although our sampling frequency and species measured are limited, ionic chemical analysis of 32 size-segregated sample sets obtained from April 1996 to January 1997 provide preliminary results on seasonal variations of concentration levels in atmospheric aerosols at Syowa Station. Results of size-segregated aerosol samples suggest that sea-salt particles in spring and early summer were more depleted for a coarse fraction than in winter. Further study, including comparisons with tracer radioisotopes, elements and gaseous compounds, is needed to study sources, long-range transport and transformation processes during transport toward Syowa Station. Complete year-long continuous data sets for size-segregated aerosols and precursor gaseous species at a contamination-free site near Syowa Station are desired to determine the atmospheric chemical cycles via aerosols for the coastal Antarctic atmosphere.

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