

Seasonal variation of chemical composition of aerosols at Syowa Station, Antarctica in 2001

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Abstract: Antarctic aerosols collected at Syowa Station in 2001 were analyzed to investigate their seasonal variations and long-range transport of anthropogenic aerosols. The measured chemical species were elemental carbon (E.C.) and organic carbon (O.C.) using a combustion technique, SO_4^{2-} , NO_3^- , Cl^- , Na^+ , NH_4^+ , K^+ , Ca^{2+} and Mg^{2+} using ion chromatographs, and metals such as Al, V, Cr, Mn, Fe, Co, Ni, Zn and Pb using an inductively coupled plasma mass spectrometer. Total mass concentration of aerosols ranged from 0.366 to 2.72 $\mu\text{g}/\text{m}^3$ and increased from winter to spring. The NO_3^- concentration was lower than 0.01 $\mu\text{g}/\text{m}^3$ in March–July; in contrast, it was higher than 0.02 $\mu\text{g}/\text{m}^3$ in August–November. Concentration of elemental carbon was relatively low in April–June and was high in March, October and November. In October and November, NO_3^- concentration was also high. Therefore, the air mass was possibly affected by biomass burning. The concentration of SO_4^{2-} was low around 0.02 $\mu\text{g}/\text{m}^3$ in May–July, and it increased to higher than 0.1 $\mu\text{g}/\text{m}^3$ in August–December. The concentrations of Al, V, Co, Ni and Pb were sometimes lower than the detection limits. Nevertheless, spikes of the highest concentrations of V, Cr, Fe, Co, Ni, Zn and Pb were recorded in August–October, while V and Fe showed second spikes in March. The enrichment factors of Cr, Ni, Zn and Pb were high during 13–20 September and 12–22 October. The E.C. concentrations in these periods were also relatively higher than before and after the sampling periods. Moreover, blizzards occurred in these periods.

key words: Antarctic, aerosols, carbon, ion, metals

1. Introduction

Antarctic aerosols have only low concentration. The aerosols, however, play an important role in climate, physical and chemical processes in the atmosphere (Shaw, 1988). The aerosols scatter solar radiation and contribute to the formation of clouds. These properties are decided by shape, size, refractive index and chemical composition of aerosols. Because of a close relation to the refractive index and the chemical

composition it is important to measure chemical species of aerosols. On the other hand, since the Antarctic is far from important anthropogenic sources, it is a good site to measure the trends in background aerosol concentrations. Aerosol components and concentrations can be a source of information on the state of the atmospheric circulation.

Since there are no human activities in Antarctica, analyzing anthropogenic components of aerosols provides an estimate of the contribution of human activities to the atmosphere. Products of human activities include elemental carbon (E.C.) and metals from combustion of fossil fuels and of vegetation (biomass burning). These elements affect global climate (Haywood and Shine, 1995).

A five-year research project named "Atmospheric Circulation and Material Cycle in the Antarctic" was started by the 38th Japanese Antarctic Research Expedition (JARE-38) in 1997 (Yamanouchi *et al.*, 1999). The project includes studies of not only greenhouse gases but also atmospheric aerosols. As part of the JARE-42 activities, aerosols were sampled at Syowa Station (69°00'S, 39°35'E) in 2001. To estimate the optical and cloud nuclei properties of aerosols, water-soluble and carbonaceous component analysis were performed. In addition, to determine the origins of aerosols, trace elements were measured. In this paper, we report these measurements and discuss the contribution of human activities to the aerosols at Syowa Station.

2. Sample collection and analysis

Sampling systems were installed in the Observation Laboratory located southeast of Syowa Station. Since the direction of prevailing wind at Syowa Station is northeast, an air inlet for sampling aerosols was attached to a cable rack located northeast of the laboratory at a 4.75 m high position above the ground. Atmospheric aerosols were sampled on quartz fiber filters (Pallflex 2500QAT-UP) and Teflon filters (Sumitomo, Fluoropore FP-1000) 47 mm in diameter at flow rates of 80 and 55 L/min, respectively. For pretreatment, the quartz fiber filters were burned at 850°C for 2 hours. We sampled atmospheric aerosols until the sampling volume became about 300 m³. When the wind velocity was lower than 3 m/s, or the wind direction was not from the northeast, we stopped sampling in order to avoid contamination of the exhaust from the electric power generator. We also stopped when a snow vehicle was running on the windward side of the air inlet. For qualitative check of measurement, blank samples were taken from each sampling system periodically.

Carbon contents in the quartz fiber filters were determined by a combustion technique consisting of sample combustion at 850°C in an NC-analyzer (Sumitomo Chemical, Sumigraph NC-80) and analysis by a gas chromatograph (Shimadzu, GC-14 A) equipped with a nickel catalyst methanizer and a flame ionization detector (Ohta and Okita, 1984). The filters were cut in half. One half of the samples served to determine total carbon content (T.C.). Another half was heated in an electric furnace at 300°C in air for 30 min to remove organic carbon (O.C.), and then elemental carbon content (E.C.) was measured by the combustion technique. The difference between the total carbon and elemental carbon gave the amount of organic carbon.

Teflon filters were cut in half; one half served to determine the metal contents and

the other half served to determine the water-soluble contents. Collected aerosols on the half Teflon filters were extracted ultrasonically in a mixed solution of nitric acid (Ultrapur, Kanto Kagaku) hydrofluoric acid (Ultrapur, Kanto Kagaku), and ultra-pure water ($18.3 \text{ M}\Omega\text{cm}^{-1}$, Mili-Q water). The concentrations of metals in the solution were measured with an inductively coupled plasma mass spectrometer (ICP-MS; Yokogawa Analytical Systems, HP4500).

Collected aerosols on the other half Teflon filters were extracted with distilled deionized water ($16.7 \text{ M}\Omega\text{cm}^{-1}$). The concentrations of anions (SO_4^{2-} , NO_3^- , Cl^-), and cations (Na^+ , NH_4^+ , K^+ , Ca^{2+} , Mg^{2+}) in the extracted solution were measured with an ion chromatograph (Yokogawa Analytical Systems, IC7000) and another ion chromatograph (DIONEX, DX100), respectively.

3. Results and discussion

Figure 1 shows the seasonal variation of mass concentrations of water-soluble and carbonaceous aerosols at Syowa Station in the Antarctic. Letters in Fig. 1, such as “A”, “B” and “C”, denote the blizzard classes recorded by the JARE Meteorological Observation Team during each sampling period, *e.g.* “BC” means that blizzards of “B” and “C” classes were observed during that period. The criterion of the blizzard class is shown in Table 1. The measurements of water-soluble and carbonaceous elements of blank samples are shown in Table 2.

The blank values were different in each sampling period, because the sampling volumes were not constant in each sample. The averages and standard deviations of blank values in the case of minimum, average and maximum sampling volumes are shown in Table 2. The detection limit was twice the standard deviation of the blank value. In Fig. 1, the detection limits of NO_3^- , SO_4^{2-} and E.C. in the case of average sampling volume are shown; white bars mean that the measurements were under the detection limit. Total mass concentrations of aerosols ranged from 0.366 to $2.72 \mu\text{g}/\text{m}^3$. They decreased in October and increased from winter to spring (hereafter seasons are austral unless otherwise stated). Figure 1 further that the concentration became high during blizzards. In winter and spring, total mass concentrations of aerosols in sampling periods with blizzards were up to twice as high as in periods with no blizzards. The results suggest that snow blown by strong wind was collected on filters and/or low-pressure systems bring air masses containing rich aerosols to Antarctica. The maximum concentration of total aerosol was recorded during 30 September – 2 October; however, there was no official record of blizzards. From meteorological observation data, strong winds blew with average velocity over 20 m/s in this period, although this does not meet the condition of a “C” class blizzard.

Concentrations of each species of aerosols ranged as follows: elemental carbon (0.0026 – $0.215 \mu\text{g}/\text{m}^3$), organic carbon (0.18 – $2.90 \mu\text{g}/\text{m}^3$), SO_4^{2-} (0.0064 – $0.369 \mu\text{g}/\text{m}^3$), NO_3^- (0.0024 – $0.0538 \mu\text{g}/\text{m}^3$), Cl^- (0.047 – $1.31 \mu\text{g}/\text{m}^3$), Mg^{2+} (0.0017 – $0.0635 \mu\text{g}/\text{m}^3$), Ca^{2+} (0.0048 – $0.130 \mu\text{g}/\text{m}^3$), K^+ (0.0017 – $0.0393 \mu\text{g}/\text{m}^3$), NH_4^+ (0.0012 – $0.0127 \mu\text{g}/\text{m}^3$) and Na^+ (0.014 – $0.623 \mu\text{g}/\text{m}^3$). Concentrations of E.C. and O.C. at Syowa Station in December 1991 were 0.017 and $0.026 \mu\text{g}/\text{m}^3$, respectively (Nishikawa *et al.*, 1991). This E.C. concentration was in good agreement with our results, but this O.C. concen-

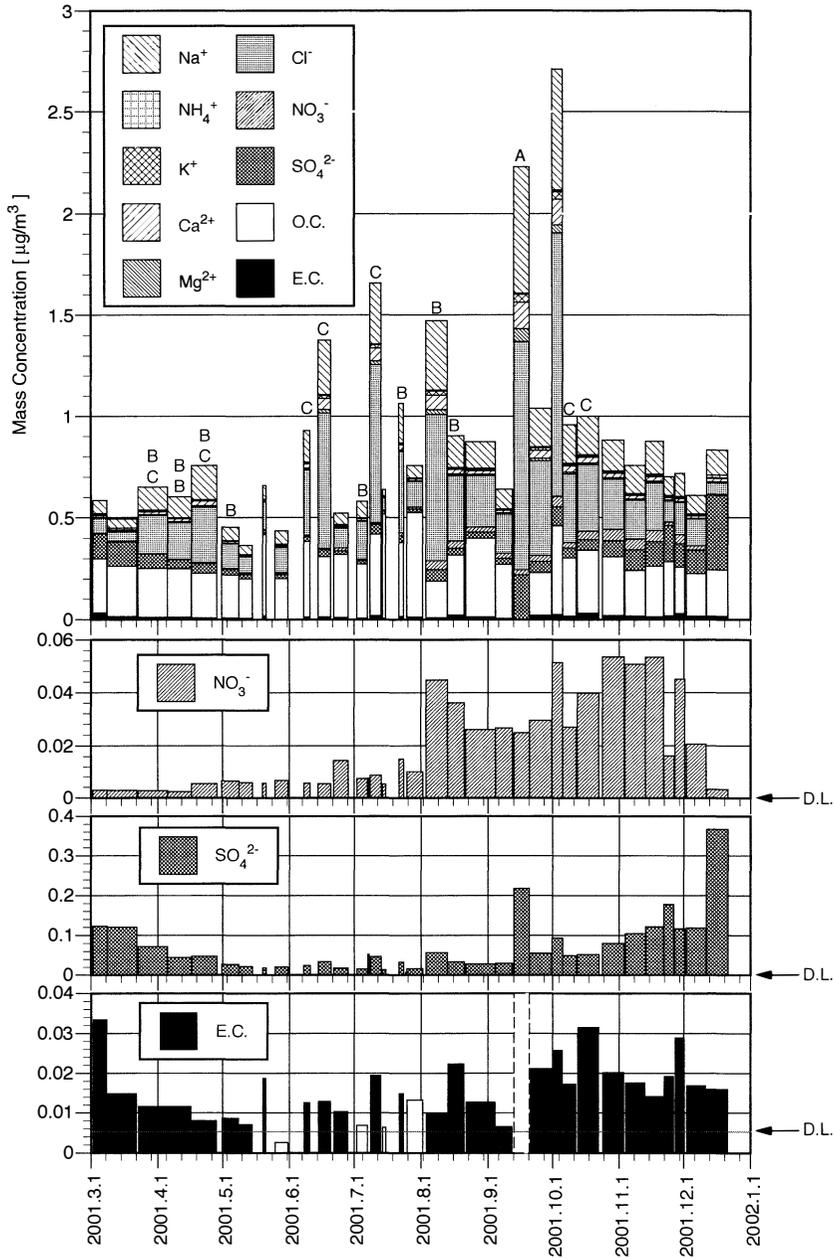


Fig. 1. Seasonal variation of mass concentration of water-soluble and carbonaceous aerosols at Syowa Station in the Antarctic. Letters, such as "A", "B" and "C", denote the blizzard classes recorded by the JARE Meteorological Observation Team during each sampling period. Expanded seasonal patterns of NO_3^- , SO_4^{2-} and elemental carbon (E.C.) are shown below. The bar with broken line denotes no data of E.C. White bars below the figure indicate that the concentrations were lower than the detection limits. Dotted lines show the detection limits (D.L.) in the case of average sampling volume.

Table 1. Criteria for blizzard classes recorded by the JARE Meteorological Observation Team.

Class	Visibility	Wind velocity	Duration time
A	< 100 m	> 25 m/s	> 6 hour
B	< 1 km	> 15 m/s	> 12 hour
C	< 1 km	> 10 m/s	> 6 hour

Table 2. Blank values of water-soluble and carbonaceous elements.

tration was not. Huebert and Charlson (2000) reported that the data on organic aerosols have uncertainties caused by positive and/or negative sampling artifacts. Kim *et al.* (2001) indicated that 30% of measured PM_{2.5} organic carbon concentration is positive artifacts. The discrepancy was caused by the positive artifact with absorption of vapors of organic constituents on a quartz fiber filter. The abundance of NO_3^- was lower than $0.01\mu\text{g}/\text{m}^3$ in March–July. In contrast, it increased in late winter to spring; it was higher than $0.04\mu\text{g}/\text{m}^3$ in August and October–November. The high values of NO_3^- in spring suggest that the NO_3^- origin might be PSCs in the stratosphere. In Antarctica, it is generally known that Polar Stratospheric Clouds (PSCs) appear in winter. Particles of PSCs contain HNO_3 and a possible intrusion process of PSC particles from stratosphere to troposphere has been indicated by Wagenbach *et al.* (1998). Shibata *et al.* (2003) reported a case of PSCs observed on 30 June 2001 by a micro-pulse lidar at Syowa Station. Several successive PSC events were observed over Syowa Station until mid-August. Therefore, it is suggested that NO_3^- around Syowa Station might come from the stratosphere into the troposphere in spring. The concen-

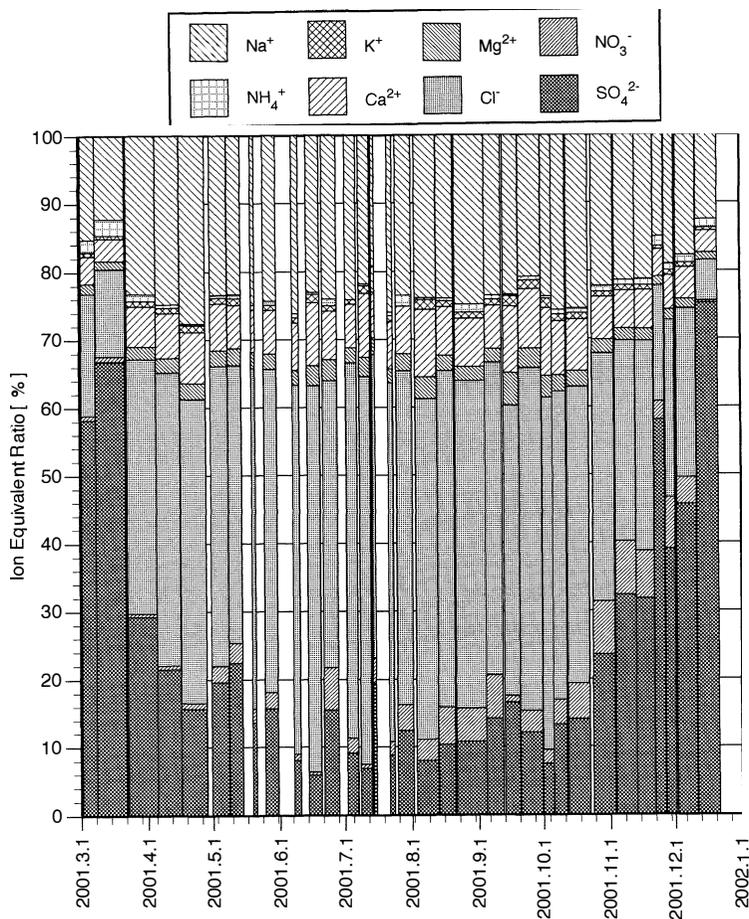


Fig. 2. Seasonal variation of ion equivalent ratios of the water-soluble components.

Table 3. Seasonal variations of metal concentrations at Syowa Station, Antarctica in 2001.

Begin date (UT)	End date (UT)	Sampling Vol. [m ³]	Na	Mg	Al	Ca
2001/3/1 19:16	3/8 10:54	445.6	63	8.0	3.48	1.9
3/8 11:20	3/22 5:22	492.4	36	4.1	0.35	0.8
3/22 19:26	4/5 11:00	411.0	117	12.6	0.23	2.0
4/5 11:38	4/16 11:45	388.8	97	11.0	0.19	2.0
4/16 13:20	4/28 8:07	376.8	198	22.5	0.25	3.7
4/30 14:03	5/8 11:54	388.8	70	8.8	-	1.4
5/8 13:12	5/14 19:21	371.5	47	6.0	1.34	1.2
5/19 11:47	5/21 4:39	130.7	68	7.6	-	1.1
5/25 6:46	5/31 7:11	393.7	82	9.4	0.21	1.4
6/7 15:01	6/10 14:22	167.8	181	21.4	-	3.0
6/14 6:49	6/20 4:49	224.7	356	39.6	-	6.1
6/21 12:16	6/28 5:53	165.8	67	9.2	-	1.0
7/2 5:54	7/7 5:32	210.1	92	10.8	-	1.4
7/8 10:27	7/13 13:16	164.7	472	57.3	-	8.5
7/14 4:34	7/15 16:06	121.9	41	5.4	-	0.7
7/21 19:04	7/23 23:17	173.1	174	19.8	-	2.9
7/25 11:31	8/1 18:51	100.7	62	8.2	-	0.8
8/3 7:29	8/13 5:42	339.1	556	61.5	-	9.5
8/13 7:35	8/21 0:55	200.6	170	22.0	-	3.5
8/21 17:22	9/4 11:54	213.8	165	20.3	-	2.9
9/4 13:30	9/12 8:24	273.8	98	11.7	-	1.6
9/13 3:05	9/20 5:37	326.0	O.R.	58.0	0.60	12.0
9/20 8:10	9/30 14:47	283.0	281	33.6	-	5.2
9/30 18:44	10/5 17:28	161.6	872	101.5	-	15.4
10/5 18:39	10/12 0:32	271.2	190	24.9	-	3.5
10/12 13:34	10/22 13:09	219.6	152	17.7	0.32	3.8
10/24 5:15	11/3 6:00	237.0	163	18.6	-	3.9
11/3 17:50	11/13 5:30	405.7	149	14.9	1.06	3.4
11/13 6:18	11/21 15:43	295.1	159	16.5	-	3.5
11/21 19:44	11/26 12:05	294.8	107	11.0	-	2.4
11/26 22:20	12/1 10:29	262.3	164	17.6	-	3.6
12/2 4:25	12/11 8:42	325.0	87	9.2	-	1.9
12/11 13:47	12/21 11:09	405.5	135	13.1	-	2.8
			Na	Mg	Al	Ca
Average of blank	Minimum		1.26	0.375	0.941	-0.501
(Standard deviation of blank)	100.7		(0.91)	(0.159)	(0.327)	(0.134)
[ng/m ³]	Average		0.45	0.135	0.338	-0.180
	280.1		(0.33)	(0.057)	(0.118)	(0.048)
	Maximum		0.26	0.077	0.193	-0.103
	492.4		(0.19)	(0.032)	(0.067)	(0.027)

- indicates lower than detection limit.

O.R. denotes over range of instrument.

Table 3 (Continued).

Mass concentration [ng/m ³]							
V	Cr	Mn	Fe	Co	Ni	Zn	Pb
0.0107	0.96	0.141	5.76	0.0058	0.41	0.450	0.0101
-	-	0.012	0.60	-	-	0.038	0.0040
-	0.20	0.020	0.95	-	-	0.085	-
0.0033	0.93	0.088	3.10	0.0055	0.36	0.172	0.0049
-	0.26	0.028	0.99	0.0038	-	0.107	-
0.0020	0.58	0.062	1.94	0.0041	0.23	0.096	0.0153
-	0.18	0.033	0.67	-	-	0.041	0.0046
-	-	-	-	-	-	0.054	-
-	-	-	-	-	-	0.018	-
-	0.25	-	0.73	-	-	0.007	-
-	0.48	0.028	1.79	0.0065	-	0.080	0.0047
-	-	-	-	-	-	-	-
-	0.37	0.026	0.88	-	0.33	0.010	-
-	-	-	-	-	-	-	-
-	-	-	-	-	-	-	-
-	-	-	0.77	-	-	0.016	-
-	-	-	-	-	-	-	-
0.0028	0.46	0.048	1.39	0.0081	-	0.073	-
0.0070	2.08	0.275	6.50	0.0145	0.89	0.306	0.0222
-	-	-	-	-	-	0.009	-
-	-	-	-	-	-	-	-
0.0054	0.92	2.248	3.36	0.0142	0.53	2.772	0.0117
-	-	-	-	-	-	0.030	-
-	-	-	-	0.0088	-	0.192	-
-	-	-	-	-	-	0.014	-
-	-	-	1.05	-	-	0.106	0.0313
-	-	-	0.89	-	-	0.103	-
-	-	-	0.69	-	-	0.059	0.0032
-	-	-	0.58	-	-	0.005	0.0044
-	-	-	0.74	-	-	0.205	-
-	-	-	0.55	-	-	0.023	-
-	-	-	-	-	-	0.054	-
-	-	-	-	-	-	0.061	-
V	Cr	Mn	Fe	Co	Ni	Zn	Pb
0.0164	-0.061	0.0273	0.890	0.0065	0.175	0.1665	0.0122
(0.0038)	(0.184)	(0.0196)	(0.571)	(0.0049)	(0.313)	(0.0041)	(0.0051)
0.0059	-0.022	0.0098	0.320	0.0024	0.063	0.0599	0.0044
(0.0014)	(0.066)	(0.0070)	(0.206)	(0.0018)	(0.112)	(0.0015)	(0.0018)
0.0034	-0.012	0.0056	0.182	0.0013	0.036	0.0341	0.0025
(0.0008)	(0.038)	(0.0040)	(0.117)	(0.0010)	(0.064)	(0.0008)	(0.0010)

tration of SO_4^{2-} was low, around $0.02 \mu\text{g}/\text{m}^3$, in May–July, and it increased to higher than $0.1 \mu\text{g}/\text{m}^3$ in early summer. Minikin *et al.* (1998) reported that SO_4^{2-} origin of the boundary layer at high southern latitudes is mainly marine biogenic. Thus, the seasonal variation of SO_4^{2-} concentration in Antarctica relates to biotic activity;

increasing chlorophyll concentration called blooming occurs in the Antarctic coastal area in summer and raises the abundances of Dimethylsulfide (DMS) and Dimethylsulfoxide (DMSO), which are precursors of sulfuric acid. The DMS and DMSO oxidized sulfuric acid and then converted to particulate sulfur oxide in a gas-to-particle conversion process. For these reasons, the SO_4^{2-} concentration at Syowa Station was high in summer and low in winter. The concentration of elemental carbon (E.C.) was relatively low in April to June and was high in March, October and November. Aerosols containing elemental carbon are generally affected by human activity (Wolff and Cachier, 1998). In October and November, NO_3^- concentration was also high. Elemental carbon and NO_y , which is a precursor of NO_3^- , are emissions of biomass burning (Andreae and Merlet, 2001). Therefore, it is possible that the air mass might be affected by biomass burning.

During the 13–20 September sampling period, we have no data on E.C. or O.C. In this period, an A-class blizzard was observed at Syowa Station. Since the quartz filter was broken at the end of the sampling period, we could not analyze this filter. On the other hand, although the Teflon filter was torn, we measured the water-soluble species. Therefore, the real value might be higher, because some aerosols passed through the crack.

Figure 2 shows the seasonal variations of the ion equivalent ratios of the water-soluble constituents. The ratio of SO_4^{2-} was high, around 60–70%, in summer; on the other hand, it decreased to 10% in winter. This result is consistent with a previous study by Osada *et al.* (1998). In contrast, the ratio of Cl^- was around 10% in summer; it increased to 60% in winter.

Seasonal variations of metal concentrations are shown in Table 3 and Fig. 3. The measurements of blank samples are also shown in Table 3. Since the blank values were different in each sampling period, similar to the measurements of water-soluble and carbonaceous elements, the averages and standard deviations of blank values in the cases of minimum, average and maximum sampling volumes are shown in Table 3. The detection limit was twice the standard deviation of the blank value. Measurements lower than the detection limit are shown by white bars. The concentrations of Al, V, Co, Ni and Pb were sometimes lower than the detection limits. The results showed no apparent seasonal variations of each metal. These variations were different for each component. But spikes of the highest concentrations of V, Cr, Fe, Co, Ni, Zn and Pb were recorded in August–October and these of V and Fe were recorded in March. Although the amount of Al was sometimes high, 1.0 ng/m^3 or more, it was usually lower than 0.2 ng/m^3 . As the winter average concentration of Al at the South Pole was lower than 0.3 ng/m^3 (Cunningham and Zoller, 1981), the concentration at Syowa was as low as that at the South Pole. When the concentration is high, it indicates that the aerosol includes soil particles because Al is a major constituent of the Earth's crust. We compared our results with average elemental concentrations at the Brazilian Antarctic Station, on King George Island, Antarctic Peninsula (Artaxo *et al.*, 1992) that is located in the coastal area like our sampling site, and found that the Al concentration at Syowa Station was lower than that at the Brazilian station. This shows that the contribution of continental soil to aerosols at Syowa Station may not be more than that at the Brazilian Station on the Antarctic Peninsula.

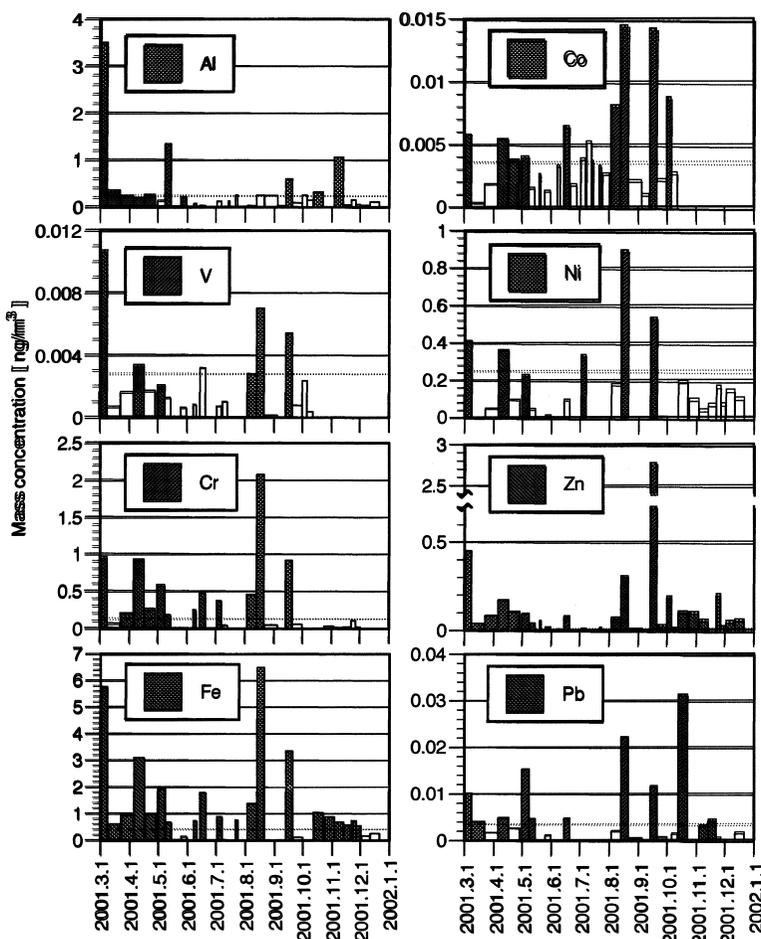


Fig. 3. Seasonal pattern of metal concentrations. White bars indicate that the concentrations were lower than the detection limits. Broken lines show the detection limits in the case of average sampling volume.

To evaluate the effects of human activities, enrichment factors (EF) were calculated. The factors are ratios of observed elemental concentrations to the calculated crustal contributions for those elements. Aluminum has been chosen here as a reference element. The *EF* is defined as:

$$EF = (X/Al)_{\text{aerosol}} / (X/Al)_{\text{crust}},$$

where X and Al refer to the concentrations of the metals of interest and of Al, respectively. Crustal values are taken from Bowen (1966). The Al concentrations of about half of the samples were lower than the detection limit; therefore, the enrichment factors were calculated only for samples having meaningful measurements of Al (Fig. 4). The EF values of Cr, Ni, Zn and Pb were high, especially in April, September and October. The species Cr, Pb, and Zn within these metals are attributed to emissions of heavy metals to the atmosphere from human activities in South America, southern

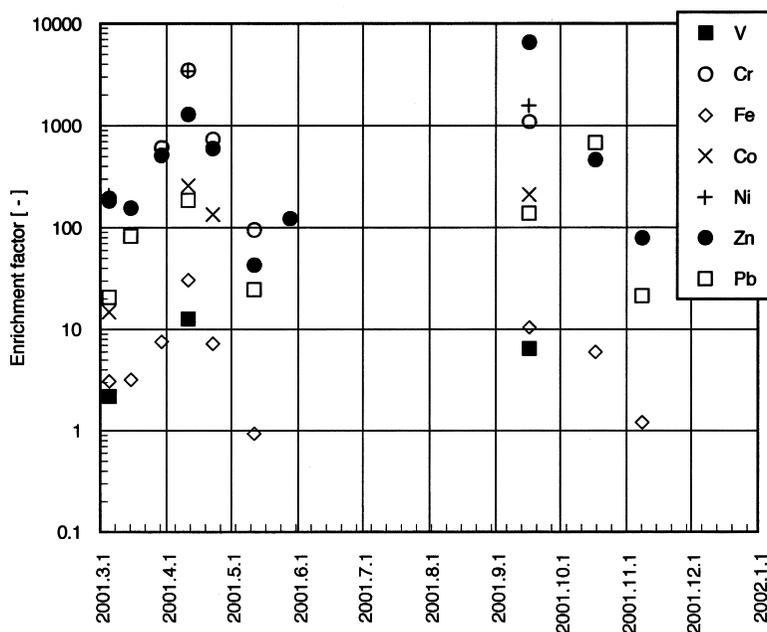


Fig. 4. Seasonal variation of enrichment factor (EF) values. Samples in which the concentrations of metals were lower than the detection limits are not shown.

Africa and Australia, particularly non-ferrous metal mining and smelting in Chile, Peru, Zaire, Zambia and Australia (Planchon *et al.*, 2002). During 13–21 August, high concentrations of Cr, Fe, Co, Ni, Zn and Pb were observed. However, Fig. 4 shows no data in this period because the Al concentration was lower than the detection limit. This indicates that the origins of these metals in this period were not only from the Earth's crust but also from human activities. The E.C. concentrations during 13–21 August and 12–22 October, when the EF values were high, were also higher than before and after the sampling periods. During 13–20 September, although the EF values were high, we have no data on the E.C. concentration due to damage to the quartz fiber filter. In these periods, blizzards were recorded at Syowa Station. It is possible that the air masses in these periods were affected by human activities and low-pressure systems brought air masses containing anthropogenic aerosols to Antarctica.

4. Conclusions

We measured atmospheric mass concentrations of aerosols at Syowa Station, Antarctica in 2001. Total mass concentrations of aerosols ranged from 0.366 to 2.72 $\mu\text{g}/\text{m}^3$. Concentrations decreased in autumn and increased in spring. In winter and spring, total mass concentrations in sampling periods when blizzards were recorded were sometimes twice as high as in periods with no records of blizzards. The results suggest that snow blown by strong wind was collected on filters and/or low-pressure systems brought air masses containing rich aerosols to Antarctica.

The concentration of NO_3^- was lower than $0.01\mu\text{g}/\text{m}^3$ in March–July. In contrast, it increased in August–November. High values of NO_3^- in spring mean that the origin might be PSCs in the stratosphere. The concentration of SO_4^{2-} was low, around $0.02\mu\text{g}/\text{m}^3$, in May–July; it increased to higher than $0.1\mu\text{g}/\text{m}^3$ in early summer. The seasonal variation of SO_4^{2-} concentration in Antarctica relates to such biotic activity as blooming. The elemental carbon concentration was relatively low in April–June and was high in March, October and November. In October and November, NO_3^- concentration was also high. Elemental carbon and NO_y , which is precursor of NO_3^- , are emissions of biomass burning. Therefore, air masses were possibly affected by biomass burning.

Metals concentrations showed no apparent seasonal variations. In addition, these variations were different in each component. But spikes of the highest concentrations of V, Cr, Fe, Co, Ni, Zn and Pb were recorded in August–October, and these of V and Fe were recorded in March. To evaluate effects of human activities, enrichment factors (EF) were calculated. EF values for Cr, Ni, Zn and Pb were high, especially in April, September and October. The species Cr, Pb, and Zn within these metals are attributed to emissions of heavy metals to the atmosphere from human activities in South America, southern Africa and Australia. The E.C. concentrations when the EF values were high were also relatively higher than before and after sampling periods. In these periods, blizzards were recorded at Syowa Station. It is possible that the air mass in these periods was affected by human activities and low-pressure systems brought air masses containing anthropogenic aerosols to Antarctica.

To learn more about the origins of air masses coming to Antarctica, we plan to combine measurements of seasonal variations of chemical compositions of aerosols, trajectory analysis and satellite remote sensing.

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