

Full Paper

Aerosol Chemical Species and Volatile Organic Compounds at Barrow, Alaska

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Abstract

This is a report of results of 2 years of measurements of aerosol chemical compositions and volatile organic compounds (VOCs) at Barrow, Alaska. The submicron aerosol consisted of 7 components (sea salt, non-sea-salt (nss) sulfate, nitrate, elemental carbon (EC), organic carbon, soil and NH_4^+). The total amount of chemical species increased in winter and spring (November-April) and decreased in summer and autumn (May-October). In particular, concentrations of EC and nss-sulfate showed seasonal variations with minimums in summer and autumn and maximums in winter and spring corresponding to the development and persistence of Arctic Haze. Heavy metals showed a similar seasonal variation.

Also reported are the seasonal variations of Volatile Organic Compounds (VOCs). Some components of VOCs (Xylene etc.) showed seasonal variations similar to that of EC, which suggests that the Arctic haze includes VOCs emitted from sources thousands of km away.

The sources and their contributions to the aerosols measured at Barrow were estimated by a new type of factor analysis, positive matrix factorization (PMF). The PMF analysis identified three factors. Based on the source compositions, a time series of the source contributions, and the results from trajectory analysis, the factors are considered to represent sea salt, Arctic haze and both soil and photochemically formed particles.

KEYWORDS: *atmospheric aerosols, Arctic haze, VOCs, PMF analysis*

1. Introduction

The Arctic region had been considered clean and free from anthropogenic pollutants. However, the results of recent studies have established that large quantities of pollutants including pesticides, hazardous chemicals and heavy metals pour into the Arctic, and their effects on ecosystems, human health, and climate are significant. Atmospheric long-range transport is a major pathway for the introduction of anthropogenic contaminants to the Arctic. Due to more frequent transport and the cold season meteorological conditions (i.e., weak wet and dry removal), aerosols accumulate during winter and spring to cause the so-called Arctic haze (Shaw, 1981; Iversen and Joranger, 1985). Arctic

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haze has been studied extensively for decades (e.g., Barrie, 1986; Shaw and Khalil, 1989). Shaw (1995) indicated that the major components of the aerosols of Arctic haze include nss-sulfate and carbonaceous compounds, and that pollutants emitted from sources thousands of km away contribute to the phenomena. Studies so far have emphasized the behavior of some important aerosol components in the Arctic haze such as soot and nss-sulfate, their sources and relative contributions. For example, at the Atmospheric Research Observatory at Barrow maintained by the National Oceanic and Atmospheric Administration (NOAA) Climate Monitoring and Diagnostics Laboratory (CMDL), real-time wind speed and direction are monitored and used to stop sampling to exclude contributions from the local air pollutions. This monitoring has been conducted mainly from an interest in establishing the impact of anthropogenic pollutants on the global environment, especially on global warming.

Besides the impact on local visibility and toxicities to humans and the Arctic ecosystem, the Arctic aerosols play an important role in both the Arctic and global climate. For example, soot particles strongly absorb solar radiation, which changes the thermal structure of the Arctic atmosphere. Deposited soot reduces the snow surface albedo, which perturbs the radiation budget in the Arctic (Blanchet, 1991; Aoki *et al.*, 1998]. The impacts on the climate system depend greatly on the long-term chemical composition including pollutants emitted from local sources and the vertical distribution of the aerosols. Total aerosol characterization including local air pollutants in the Arctic is thus important.

This study provides information critically needed to understand aerosol behavior in the Arctic and the impact of aerosols on the climate system. Moreover, the results of this study can serve as basic data to determine aerosol optical properties from satellite observations.

2. Methods

Sampling systems were installed at the Barrow Observatory, NOAA CMDL. This observatory was established in 1973, it is near sea level and 8 km east of Barrow, Alaska at 71°N (Fig.1). The aerosol sampling system has been used and described in detail in previous studies (e.g., Fujitani *et al.*, 2002; Ohta *et al.*, 1998, 1999). Sample air was drawn through an inlet stack 10 m above ground level. Fig.2 shows the sampling system for the aerosols (system A) and VOCs (system B).

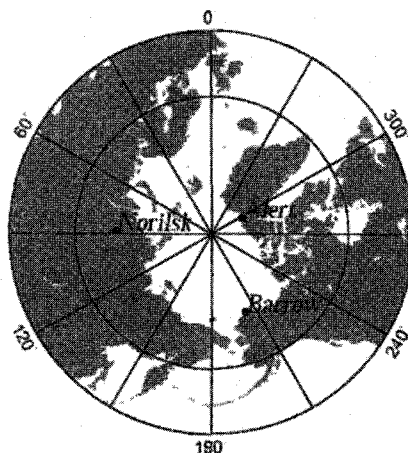


Fig.1 Map of Barrow and other sites.

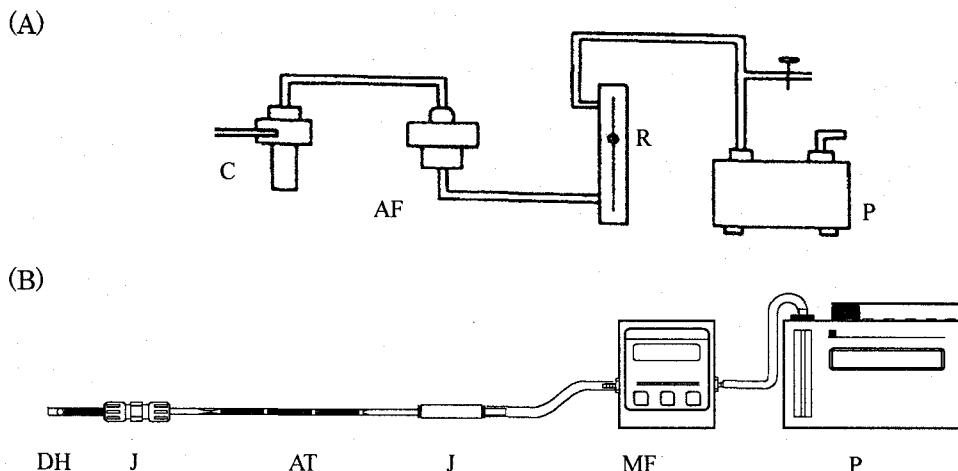


Fig.2 Sampling system of aerosols (A) and VOCs (B). C: Cyclone separator; AF: Aerosol collection filter; R: Rota meter; P: Pump; BP: Bypass; DH: Dehumidifying tube; J: Joint; AT: Absorption

2.1 Aerosols

Atmospheric fine particles were simultaneously collected on Teflon filters (Sumitomo Electric Industry Inc., FP-1000, 47 mm) and quartz fiber filters (Pallflex 2500 QAT-UP, 47 mm) by drawing outdoor air through a cyclone separator with a 50% cut-off diameter of 2- μm at a flow rate of 20 $\text{dm}^3 \text{min}^{-1}$. Two-week samplings were continuously conducted from August 1999 through June 2001. Ionic aerosol components collected on the Teflon filters were extracted ultrasonically with distilled-deionized water and analyzed with an ion chromatograph (IC, Yokogawa Analytical Systems) for anions (model IC 7000, column Excelpack ICS0-A23) and cations (ICS-C25). Metal components of the aerosols on the Teflon filters were extracted with a mixed solution of nitric acid and hydrofluoric acid and analyzed with an inductively coupled plasma mass spectrometer (ICP-MS, Yokogawa Analytical Systems, HP 4500). The carbonaceous components of aerosols (elemental and organic carbon) on the quartz fiber filters were determined by combustion in a condensation nuclei (CN) coder at 850°C coupled with a flame ionization detector (FID) and a nickel catalyst methanizer (Ohta and Okita, 1984). The OC concentrations here may have been overestimated because of the absorption of gaseous organics in the sampled air.

2.2 Volatile organic compounds (VOCs)

The VOCs were sampled once a month from March 2000 to August 2001. Air was drawn into an absorption tube (Carbotrap 317, SUPELCO) at a flow rate of 100 ml min^{-1} for 2 hours. To eliminate contamination from local pollutants, samples were taken when the wind direction was not from local pollution sources as well as when the wind speed was greater than a threshold value (0.5-1 m s^{-1}). Two successive samplings were made to confirm the reproducibility and a blank tube was used to check contamination during the shipment of tubes to our laboratory. Analyses were carried out within a week or two of the samplings. The VOCs were analyzed by a thermal desorber attached to a gas

chromatograph (HP 5890 series II) and mass selective detector (HP-5971A). Chromatographic separation was achieved using a HP-1 capillary column (60m, 0.53mm id., 5mm film thickness).

2.3 Positive matrix factorization (PMF) analysis

Positive matrix factorization (PMF) is a factor analysis algorithm developed by Paatero and Tapper (1994) and has been used in aerosol characterization studies (e.g., Poirot *et al.*, 2001; Polissar *et al.*, 2001). PMF analyzes a matrix containing the measured aerosol components and computes a small number of source compositions and their contributions to each measured components. Since PMF requires non-negativity for the contributions and the source compositions, the results are simpler to interpret than conventional factor analysis or principal component analysis. The model is given by

$$\mathbf{X} = \mathbf{GF} + \mathbf{E}$$

where \mathbf{X} is a matrix of measured aerosol components (mg m^{-3}), \mathbf{F} is an unknown source composition (g g^{-1}), \mathbf{G} is an unknown source contribution to each measured data value (mg m^{-3}) and \mathbf{E} is a matrix of residuals. The PMF computes the solution by minimizing the least squares error of the fit ($|\mathbf{E}|^2$).

The 5-day backward isentropic trajectories arriving at Barrow at a height of 925 hPa were constructed using meteorological data supplied by the Japan Meteorological Agency. As has been pointed out elsewhere (Kahl *et al.*, 1989), the calculated trajectories in the Arctic are highly uncertain because of the sparseness of meteorological observatories in the Arctic. Moreover, the trajectories in the lowest 1-2 km of the atmosphere are not always accurate (Murao *et al.* 1997). Thus the use of a single trajectory to interpret chemical measurements is problematic. In this study, the results from the PMF analysis were used to select sampling periods in which a certain source factor dominated the measured aerosol fine mass. Trajectories were then constructed for each period and the group characteristics are discussed.

3. Results and Discussions

3.1 Aerosol chemical component

Two-week average concentrations of chemical species in fine particles in the atmosphere collected at CMDL, Barrow, from August 1999 to June 2001 are presented in Fig.3. The concentration range of species were as follows: elemental carbon (EC), 0.001-0.32 $\mu\text{g C m}^{-3}$; organic carbon (OC), 0.20-0.69 $\mu\text{g C m}^{-3}$; nss-sulfate (SO_4^{2-}), 0.09-1.7 $\mu\text{g m}^{-3}$; nitrate (NO_3^-), 0.00-0.15 $\mu\text{g m}^{-3}$; chloride (Cl^-), 0.001-1.81 $\mu\text{g m}^{-3}$; ammonium (NH_4^+), 0.00-0.14 μgm^{-3} ; seasalt cations (S.S.C.), 0.007-1.8 $\mu\text{g m}^{-3}$; and soil particles (Soil), 0.003-0.25 $\mu\text{g m}^{-3}$. The concentrations of S.S.C. were obtained assuming that Na^+ derived from sea water with standard composition only (Mason, 1966), i.e.,

$$[\text{S.S.C.}] = 1.194 \times [\text{Na}^+].$$

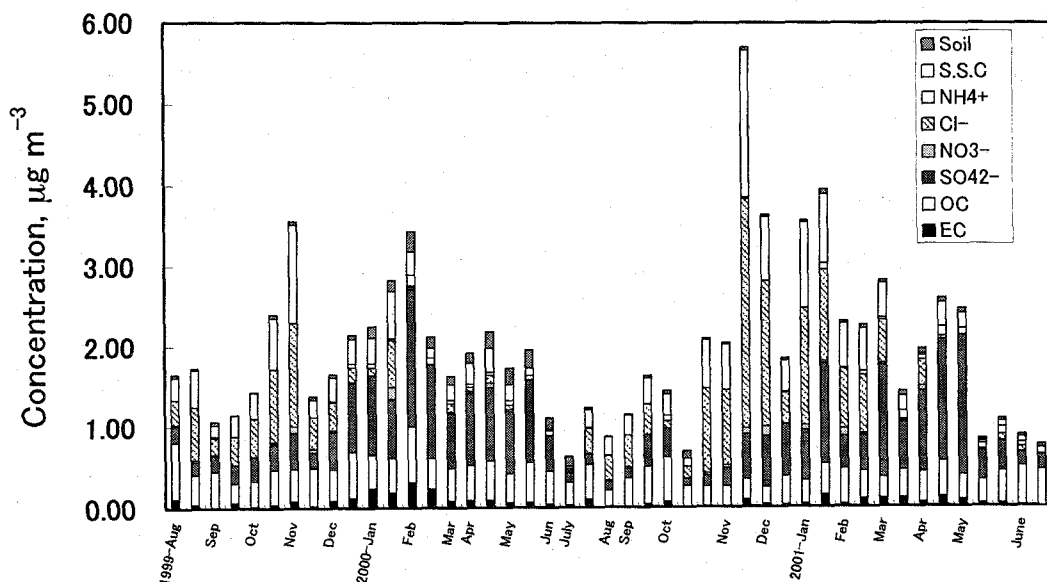


Fig.3 Two-week average concentrations of chemical species in fine particles at Barrow

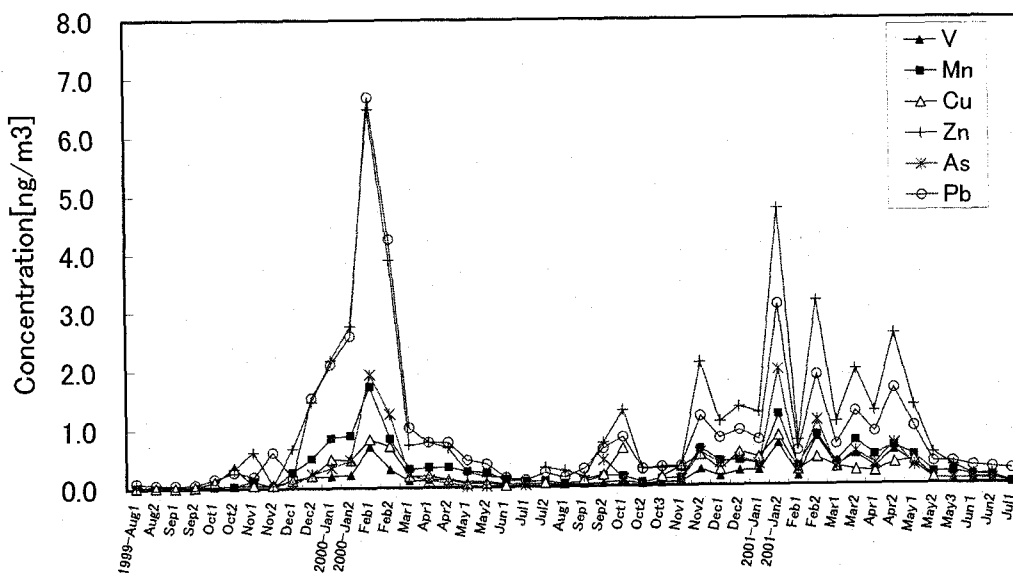


Fig.4 Two-week average concentrations of metal components of fine particles at Barrow

To estimate the concentrations of soil, Al was selected as the soil tracer, and assigned 8.1% to the mass fraction in the standard soil (Mason, 1966),

$$[\text{Soil}] = 12.35 \times [\text{Al}].$$

The total amount of the chemical species increased in winter and spring (November-April) and decreased in summer and autumn (May-October). In particular, the concentrations of EC and nss-SO_4^{2-} increased in winter and spring, in concert with the occurrence of Arctic haze. The seasonal OC variation was less pronounced. Fig.4 shows 2-week average concentrations of the metal components of fine particles in the atmosphere at Barrow. The concentration (ng m^{-3}) range of species were as follows: vanadium (V), 0.02-0.82; manganese (Mn), 0.00-1.73; copper (Cu), 0.00-0.83; zinc (Zn), 0.00-6.48; arsenic (As), 0.00-1.97; and lead (Pb), 0.06-6.68. These concentrations also increased in winter and spring (November-April) and decreased in summer and autumn (May-October), again together with the development and persistence of Arctic haze. Concentrations of Zn and Pb, increased in January and February 2000 and January to May 2001; Zn and Pb are emitted mainly from solid waste incineration and automobile exhaust, respectively.

3.2 VOC components

Table 1 shows the atmospheric concentrations of VOCs from March 2000 to August 2001 at Barrow. The concentration (ppbv) ranges were as follows: chloroform (0.005-0.014); 1, 2-dichloroethane (0.00-0.012); carbon tetrachloride (0.00-0.11); ethylbenzene (0.008-0.11); m,p-xylene (0.01-0.15); styrene (0.006-0.092); o-xylene (0.007-0.096); 1-ethyl-3-methyl-benzene (0.002-0.04); 1, 3, 5-trimethylbenzene (0.002-0.11); 1, 2, 4-trimethylbenzene (0.008-0.50); and 1, 4-dichlorobenzene (0.001-0.057). Except for chloroform, 1, 2-dichloroethane, and carbon tetrachloride, these concentrations showed a seasonal variation with spring maxima. The seasonal variations of four of the compounds listed in Table 1 are plotted in Fig.5. Ethylbenzene is used as a solvent, and xylene, styrene, and other benzenes are emitted from automobile exhaust, suggesting

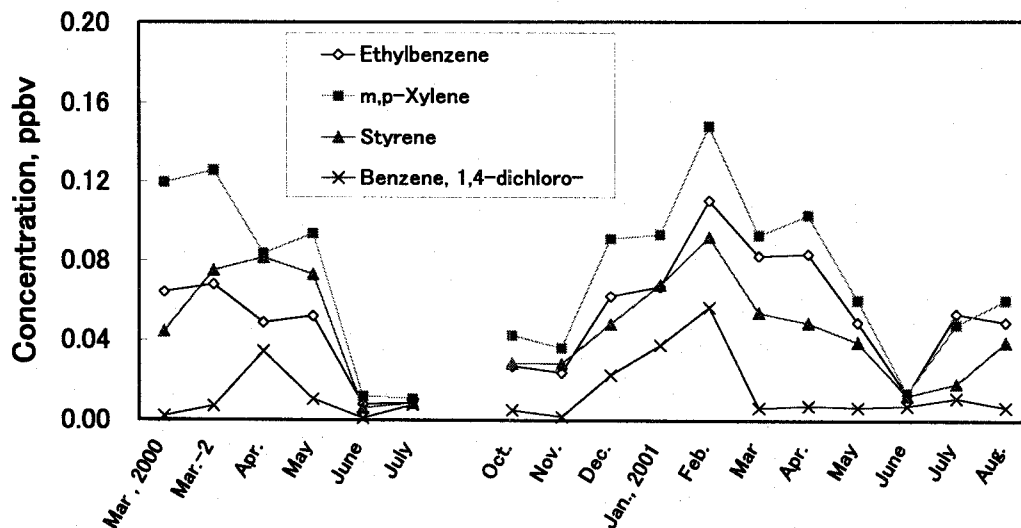


Fig.5 Concentrations of VOCs at Barrow.

Table 1. Concentrations of Volatile Organic Compounds (VOCs) in the Atmosphere at Barrow.

Compound	Concentration (ppbv)								
	March-December 2000								
	Mar. 9	Mar. 30	Apr. 27	May. 26	Jun. 26	Jul. 26	Oct. 26	Nov. 15	Dec. 21
Chloroform	0.012	0.012	0.013	0.011	0.013	0.013	0.011	0.014	0.010
Ethane, 1,2-dichloro-	0.007	0.007	0.009	0.006	0.004	0.006	0.005	0.005	0.007
Carbon Tetrachloride	0.060	0.064	0.083	0.075	0.051	0.088	0.073	0.065	0.092
Ethylbenzene	0.064	0.068	0.049	0.052	0.008	0.009	0.027	0.024	0.062
m,p-Xylene	0.120	0.126	0.084	0.094	0.012	0.011	0.043	0.036	0.091
Styrene	0.044	0.075	0.081	0.073	0.006	0.009	0.029	0.028	0.048
o-Xylene	0.090	0.096	0.054	0.059	0.009	0.007	0.026	0.021	0.064
Benzene, 1-ethyl-3-methyl-	0.018	0.021	0.010	0.010	0.002	0.003	0.005	0.003	0.012
Benzene, 1,3,5-trimethyl-	0.023	0.026	0.015	0.015	0.002	0.003	0.007	0.004	0.016
Benzene, 1,2,4-trimethyl-	0.098	0.110	0.058	0.060	0.008	0.011	0.027	0.017	0.061
Benzene, 1,4-dichloro-	0.002	0.007	0.035	0.011	0.001	0.008	0.005	0.002	0.023
	January-August 2001								
	Jan. 18	Feb. 26	Mar. 30	Apr. 25	May. 18	Jun. 15	Jul. 18	Aug. 25	
Chloroform	0.011	0.013	0.011	0.008	0.008	0.005	0.008	0.008	
Ethane, 1,2-dichloro-	0.012	0.012	0.011	0.008		0.005			
Carbon Tetrachloride	0.106	0.101	0.103	0.100	0.099		0.084	0.099	
Ethylbenzene	0.067	0.111	0.082	0.083	0.049	0.013	0.053	0.049	
m,p-Xylene	0.093	0.148	0.093	0.103	0.060	0.014	0.048	0.060	
Styrene	0.068	0.092	0.054	0.049	0.039	0.013	0.019	0.039	
o-Xylene	0.066	0.089	0.057	0.077	0.038	0.011	0.028	0.038	
Benzene, 1-ethyl-3-methyl-	0.040	0.023	0.010	0.022	0.010	0.004	0.005	0.010	
Benzene, 1,3,5-trimethyl-	0.111	0.034	0.015	0.046	0.015	0.006	0.006	0.015	
Benzene, 1,2,4-trimethyl-	0.498	0.127	0.058	0.210	0.058	0.025	0.023	0.058	
Benzene, 1,4-dichloro-	0.038	0.057	0.006	0.007	0.007	0.008	0.011	0.007	

that these gases have been transported from far away sources. Since the VOCs were sampled only 2 hours on each sample day, the measured VOC concentrations depend on the weather conditions and the transport history. Thus more extensive measurements are needed to verify the variations and sources.

3.3 PMF Analysis

The PMF model identified three sources, and the results are shown in Fig.6. The left side of Fig.6 shows the source compositions (**F**), i.e., the chemical loading of each factor (g g^{-1}). Displayed on the right side are relative contributions to the species by the factors. The summation of the relative contributions across the 3 factors (plus residual) is one. The source names shown in Fig.6 are not produced by the model. They were added based on the source compositions, time series of the source contributions, and the results from trajectory analysis detailed below.

A comparison of the reconstructed mass contributions from the 3 factors with the measured total mass is shown in Fig.7. The PMF model explained well the variation of total fine mass (Squared correlation coefficient (R^2) 0.92). The factor contributions (**G**) to the measured total mass for each sampling period and season are shown in Figs.8 and 9.

The first "Sea Salt" factor had high loadings of Na^+ and Cl^- . As shown in Fig.7, this factor explained the aerosol fine mass variation well ($R^2=0.67$). The time series for the factor shown in

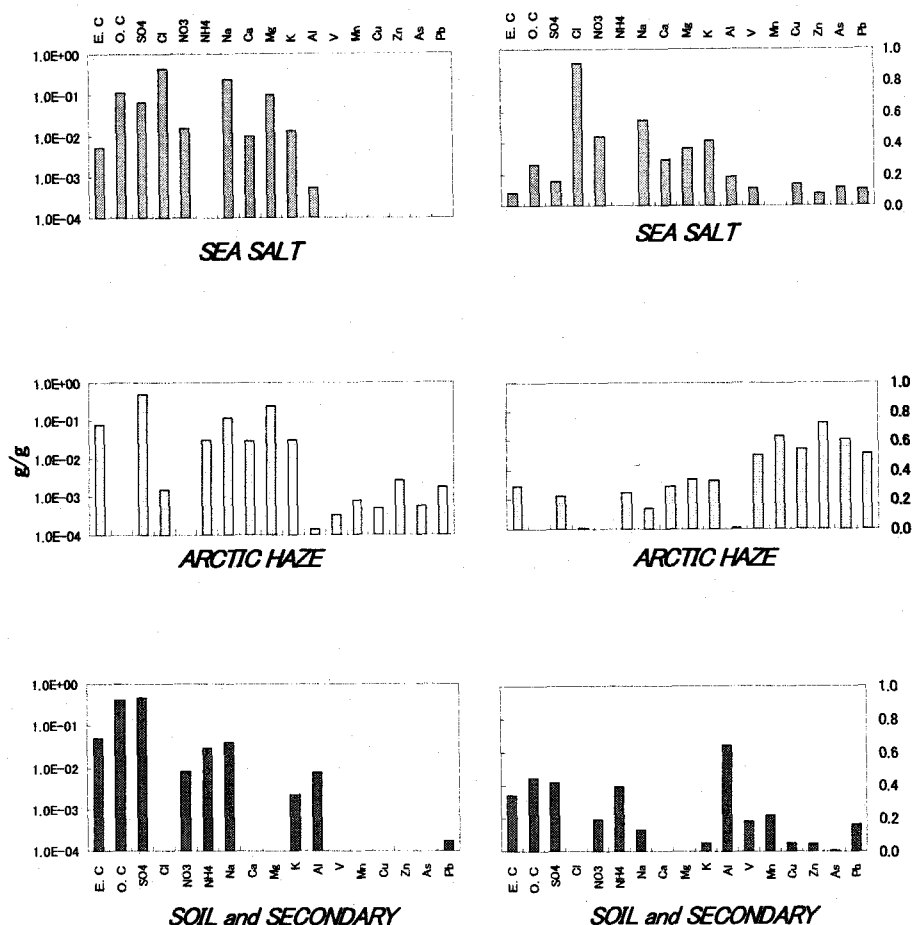


Fig.6 Source compositions of the factors identified by the PMF model, left: absolute composition of each factor, g g^{-1} , right: relative contributions to each component

Fig.9 had a maximum in winter. From November to December 2000, the factor contributed more than 80 % of the fine mass (Fig.8). The result of the trajectory analysis for the period from 9 to 24 Nov. 2000 (Fig.10) indicated that the sea salt particles were likely transported from the northern Pacific Ocean. This factor had smaller contributions in summer and the seasonal difference is in agreement with observations of submicron particles at Barrow Observatory (Quinn *et al.*, 2002). Quinn *et al.* (2002) also showed that super-micron sea salt peaks in summer corresponding to a decrease in the extent of sea ice. The "Sea Salt" factor also had small but nonzero EC, OC, and nss-SO_4^{-2} values. These anthropogenic components are probably incorporated during the long-range transport.

Two other factors with high loadings of nss-SO_4^{-2} (i.e., "Arctic Haze" and "Soil and Secondary")

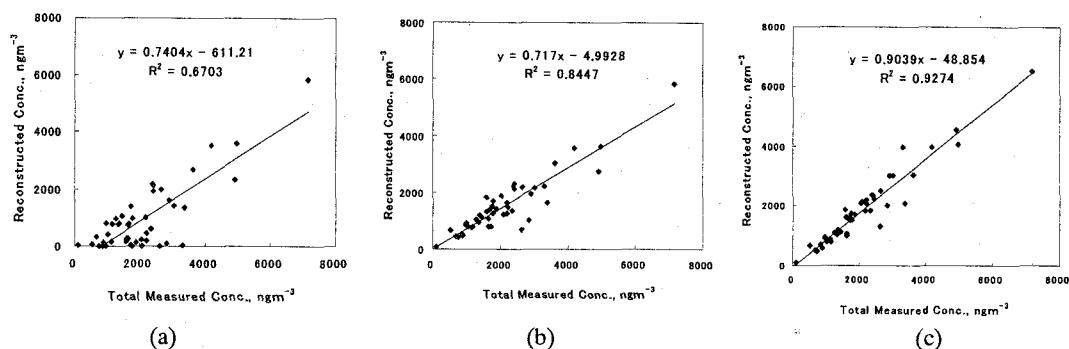


Fig.7 Observed total fine mass vs reconstructed mass from (a) "Sea salt" factor, (B) "Sea salt" and "Soil and Secondary" factors and (C) all three factors.

were obtained. These factors had small loadings of heavy metals, because the concentrations of heavy metals were low compared with the ionic components. However, as shown in the right side of Fig.6, the factors explained the variations of anthropogenic components including heavy metals, nss-SO_4^{2-} , EC and OC.

The "Arctic Haze" factor explained more than 50% of the concentrations of V, As, Cu, Pb, and Mn. Since these elements are emitted by oil combustion, coal combustion, solid waste incineration, and automobile exhaust, this factor is strongly related to primary pollutants. The "Arctic Haze" factor did not explain the OC variations. It is not clear whether the results indicate that secondary OC dominates in the Arctic or that they merely arose from measurement errors due to the absorption of

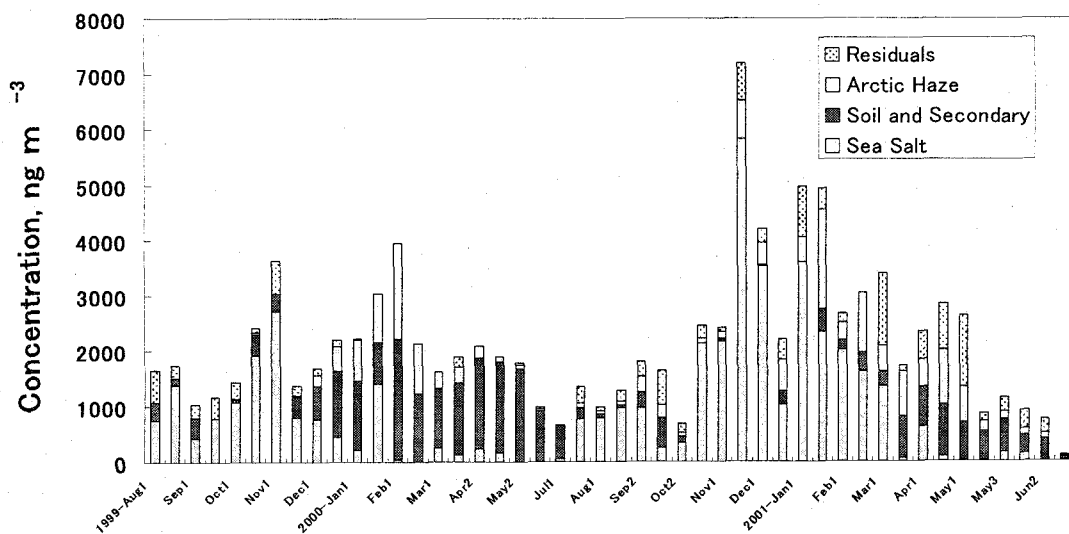


Fig.8 Time series of the contribution of each factor to the observed aerosol mass.

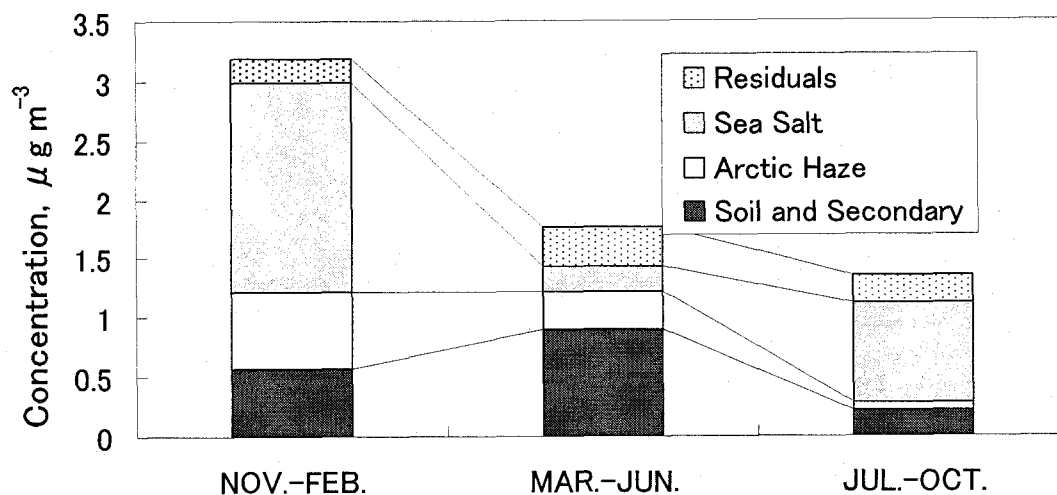


Fig.9 Seasonal average contributions of each factor to the observed aerosol mass.

organic gases. The time series for the factor (Figs.8 and 9) also had a maximum in winter. The contribution of this factor was the largest in February 2001. The trajectories for the period from 5 to 17 Feb. 2001 are shown in Fig.11. The result suggests that there was occasional long-range transport from north central Russia during this period. These results are in agreement with the climatic flow pattern analysis for Barrow (Harris and Kahl, 1994), which indicates transport of pollution from central Russian occurs near the surface during the Arctic haze season. The high contributions to the Cu and Zn variations could also be explained by the long-range transport from north central Russia, because there are a number of strong pollution sources, such as the Norilsk copper-nickel smelter on the Taimyr Peninsula.

Compared with the "Sea salt" factor and the "Arctic Haze" factor, the "Soil and Secondary" factor is difficult to interpret. As shown in Fig.6, the "Soil and Secondary" factor had high loadings of nss-SO_4^{2-} and OC and explained the variation of EC, OC, nss-SO_4^{2-} , NH_4^+ , and Al. The large contribution to Al indicates that the variation in soil particles was explained by this factor. The time series of for the factor (Fig.9) had a maximum in spring. The large contributions to OC and nss-SO_4^{2-} seem to be due to enhanced photochemical activity at that time of the year. The NH_4^+ was probably produced by the reaction of gaseous NH_3 with nss-SO_4^{2-} . As shown in Fig.8, the contribution of this factor was the largest in May 2001. The results of the trajectory analysis for the period from 13 to 26 May 2001 (Fig.12) did not show long-range transport to Barrow. These results indicate that this factor can be seen to represent both soil particles and secondary aerosols, although the source area responsible for the enhanced nss-SO_4^{2-} formation was not identified. A separation of the two kinds of particles may need a higher time resolution in the measurements. The results here are similar to those in the other studies applying statistical analysis to Arctic aerosol compositions (Pokissar *et al.*, 1998, 1999; Xie *et al.*, 1998). For example, Xie *et al.* (1998), applied the PMF model to the aerosol data from Alert, Arctic Canada, found three factors with high sulfate concentrations: an anthropogenic factor including elements like Zn, Pb, and As with a broad peak from January to March; a

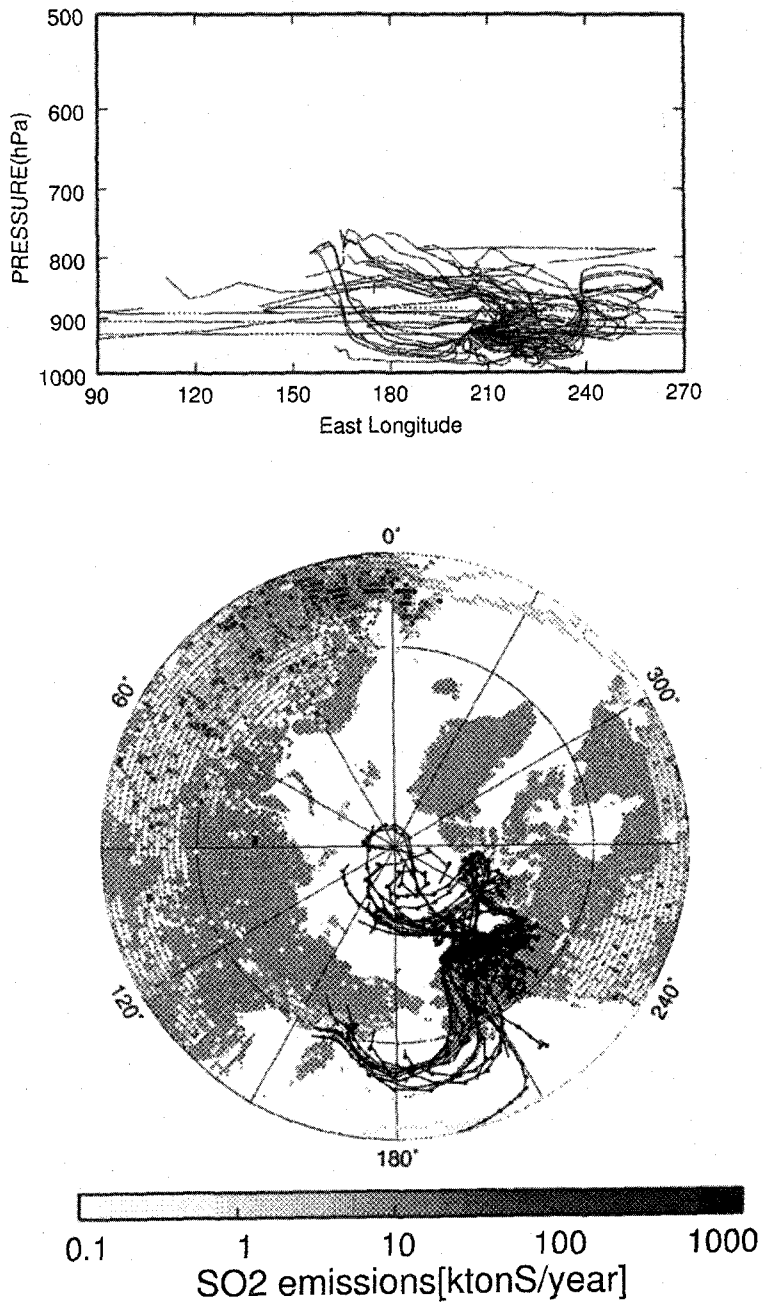


Fig.10 Trajectories for the period from 9 to 24 Nov. 2000. The source distribution was obtained from Olivier *et al.* (1996) and Ryaboshapko *et al.* (1996)

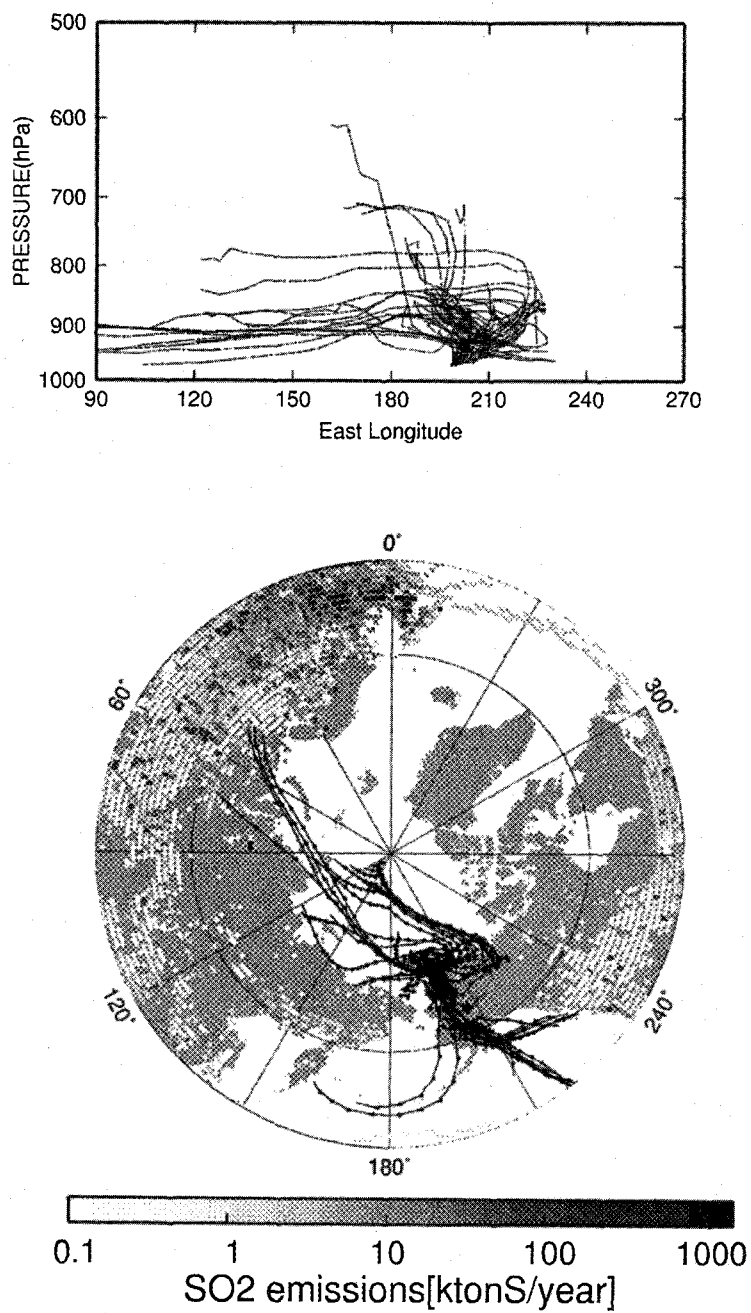


Fig.11 Same as Fig.10, but for the period from 5 to 17 Feb. 200

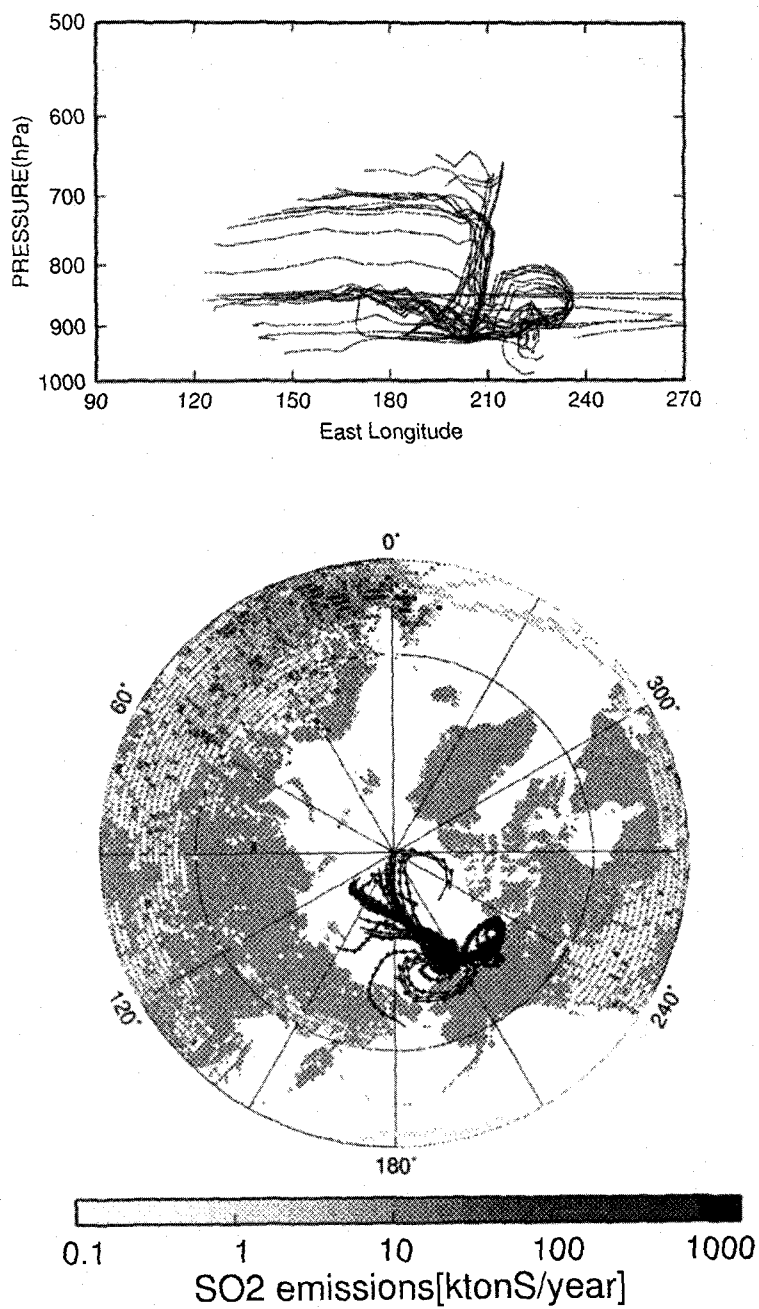


Fig.12 Same as Fig.10, but for the period from 13 to 267 May. 2001.

photochemical factor with a spring peak; and a biogenic sulfur factor including methane sulfonate (MSA) with peaks in the late spring and summer.

4. Conclusion

Atmospheric concentrations of mass and chemical species of aerosols, and volatile organic compounds (VOCs) at Barrow, Alaska were measured from August 1999 to July 2001. The submicron aerosols consisted of elemental carbon, organics, sulfate, nitrate, chloride, ammonium, sea-salt cations, and soil components. The total amount of the chemical species increased in winter and spring (November-April) and decreased in summer and autumn (May-October). The concentrations of EC and nss-SO_4^{2-} also increased in winter and spring, in concert with the occurrence of Arctic haze. Heavy metals also showed a minimum in summer and autumn and a maximum in winter and spring corresponding to the development and persistence of Arctic Haze. Some VOC components showed seasonal variations with a maximum in spring. The results suggest that the Arctic haze contains VOCs emitted from sources thousands of km away. The PMF analysis identified three factors, and based on the source compositions, the time series of the source contributions, and the results from trajectory analysis, the factors are named as "Sea Salt", "Arctic Haze" and "Soil and Secondary". They are considered to represent the sea salt, Arctic haze component, and the soil particles and photochemically formed particles. The "Arctic haze" factor dominated the aerosol mass during winter and explained almost all of the variation in heavy metals. The trajectory analysis showed that some transport from the north central Russia occurred in winter. The "soil and secondary" factor which dominated during spring had high loadings of organic carbon and nss-sulfate and explained the variation of Al. The results obtained in this study will serve as basic data to evaluate effects of pollutants on climate, human health, and ecosystems in the Alaskan Arctic.

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