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AEROSOL OBSERVATIONS IN THE SIBERIAN ARCTIC

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Abstract: Atmospheric concentrations of total particulate mass (TPM) and aerosol chemical species were measured at three sites in the Siberian Arctic; Tiksi, Norilsk and Yakutsk.

Continuous samplings of aerosols were carried out from August 1993 in Norilsk and Yakutsk, and from August 1994 in Tiksi. Tiksi is located on the coast of the Laptev Sea connecting to the Arctic Ocean. Norilsk is located at the base of the Taimyr Peninsula, which is the largest single sulfur dioxide emission source in the world. Yakutsk is situated on the middle reaches of the Lena river.

Measured concentrations of the main aerosol chemical species at Tiksi ranged from 0.06-0.3 μgm^{-3} for elemental carbon, 0.55-1.5 μgm^{-3} for organic carbon and 0.09-1.6 μgm^{-3} for SO_4^{2-}. The measured sulfate and elemental carbon concentrations showed remarkable seasonal variations with winter maxima and summer minima. The seasonal variations were consistent with other studies of Arctic haze in the Alaskan, Canadian and Norwegian Arctic.

There were no significant seasonal variations of pollutant concentrations at Norilsk and Yakutsk. The TPM ranged from 6 to 30 μgm^{-3} at Norilsk and from 7 to 39 μgm^{-3} at Yakutsk. Major aerosol composition was sulfate at Norilsk, and carbonaceous particles at Yakutsk.

We also performed trajectory analyses of air parcels to estimate the potential source regions which caused elevation of pollutant concentrations in winter at Tiksi. We calculated 10-day backward trajectories at Tiksi and forward trajectories at Norilsk in 1994. According to the calculations, the pollutants which arrived at Tiksi were frequently transported from Norilsk, the Urals and other source areas in winter. Further, it was suggested that Norilsk was one of the source areas for Arctic haze.

1. Introduction

The Arctic atmosphere has been considered to be one of the most pristine quality atmospheric regions in the world. However, in recent years, the Arctic region has been damaged by industrial pollutants. MITCHELL (1956) discovered the arctic haze in the 1950's as a condensed haze layer similar to urban smog. Recent studies have revealed that the haze mainly consists of sulfate and elemental carbon, and occurs over the entire Arctic region, exhibiting strong seasonal variation with winter and spring maxima and with a summer minimum (BARRIE, 1986).

There are some large emission sources within the Arctic, such as nickel-copper

smelting in Norilsk, which has been considered to be a major source of the Arctic sulfate aerosols (SHAW, 1982). RAHN *et al.* (1983), however, estimated that Norilsk was not a major contributor to the Arctic haze. In any case, not only the sources in the Arctic but also those in mid-latitudes around the Arctic cause Arctic pollution. The potential source areas are, for example, the northeastern U.S. and southeastern Canada, western and eastern Europe, western Russia, Ukraine, southern Urals and western Siberia (RAATZ, 1991).

Extensive research on Arctic haze has been conducted in the Alaskan, Canadian and Norwegian Arctic. It has not been done, however, in the Siberian Arctic. In this study we have researched pollution in the Siberian Arctic at 3 sites, Tiksi, Norilsk and Yakutsk.

The 10-day isentropic trajectory analyses of air parcels in 1994 were also carried out in order to estimate potential source areas for the pollution in Tiksi and the potential contribution of the Norilsk source area to Arctic haze.

2. Research Areas

2.1. Tiksi

Tiksi (72°N, 129°E), located on the coast of the Laptev Sea connecting to the Arctic ocean (Fig. 1), was chosen to observe the Arctic haze. There were no significant local pollutant sources. A sampling apparatus was installed in the building of the Cosmic Ray Observatory about 7 km to the south of Tiksi city. The sampling air was taken from about 5 m above the ground.

2.2. Norilsk

Norilsk (69°N, 89°E) has the largest nickel-copper smelting plant in Russia, which is the largest single SO_2 emission source in the world. The amount of emitted SO_2 is 2

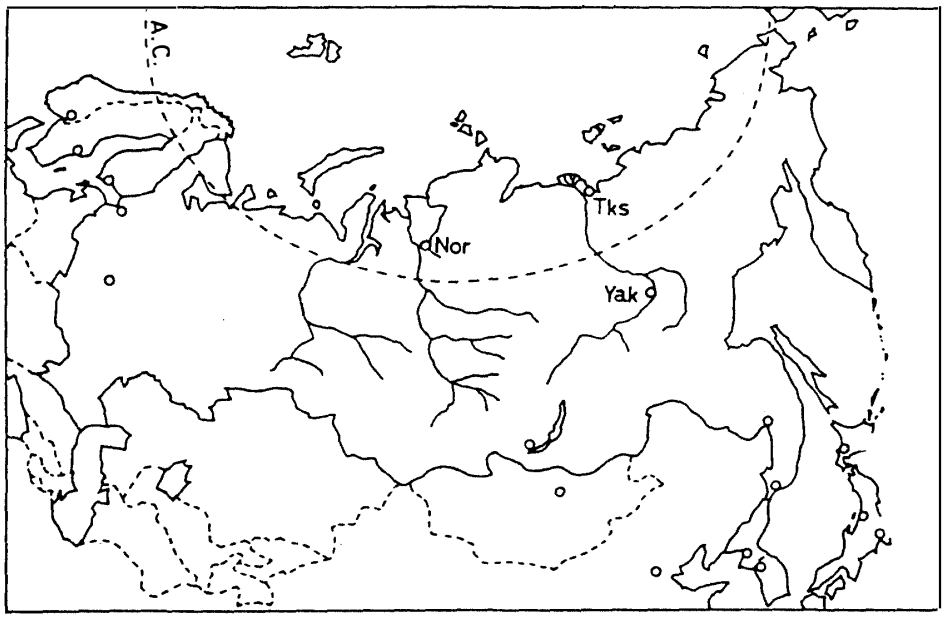

Fig. 1. Map of the sampling sites. Tks: Tiksi, Nor: Norilsk, Yak: Yakutsk, A.C.: Airctic Circle.

million tons per year. The sampling apparatus was installed on the 2nd floor of the Norilsk Experimental Center, which is surrounded by factories such as nickel and copper smelting plants.

2.3. Yakutsk

Yakutsk (62°N, 130°E) is the capital of the Republic of Sakha, located on the middle reaches of the Lena Liver. The Permafrost Institute is in the suburbs of Yakutsk city. The sampling apparatus was installed on the 6th floor of the institute, 25 m above the ground.

3. Experimental Procedure

3.1. Sampling of aerosols

The atmospheric aerosols were continuously collected once or twice a month by using filters. We used three kind of filters, such as Teflon filters (Sumitomo Fluoropore AF07P) for TPM and water-soluble matter, quartz fiber filters (Palluflex 2500QAST-UP) for carbonaceous particles and Nuclepore filters (Nuclepore co., pore-size 0.4 μm) for heavy metals. Atmospheric aerosols which had passed through an impactor whose 50% cut off diameter is 2 μm were collected on each filter with a pump at a flow rate of 5 L min^{-1}.

Table 1. Measured atmospheric concentrations of total particulate mass (TPM) and the chemical species. The unit is $\mu g/m^3$.
The blanks in the tables show that these components were not analyzed. Averaged concentrations were shown with the exception of those marked (*).

(a) Tiksi

	Month	TPM	E.C.	O.C.	SO_4^{2-}	Cl^-	NO_3^-	NH_4^-	Na^+	Ca^{2+}	Mg^{2+}	K^+
1994	AUG	0.90	0.075	0.55	0.17	0.018	0.015	0.023	0.032	0.0025	0.0028	0.010
	SEP	0.96	0.097	0.80	0.22	0.019	0.0085	0.040	0.029	0.0017	0.0029	0.0087
	OCT	0.65	0.073	1.1	0.19	0.031	0.012	0.050	0.031	0.0050	0.0031	0.010
	NOV	1.4	0.072	0.95	0.35	0.11	0.030	0.040	0.11	0.011	0.018	0.016
	DEC	2.1	0.22	1.1	1.0	0.044	0.018	0.16	0.11	0.014	0.026	0.021
1995	JAN	1.7	0.21	1.1	1.0	0.0023	0.030	0.10	0.049	0.0083	0.011	0.021
	FEB	2.6	0.31	1.5	1.5	0.0100	0.036	0.24	0.042	0.0093	0.0064	0.021
	MAR	3.0	0.19	1.2	1.6	0.038	0.026	0.16	0.12	0.021	0.050	0.026
	APR	1.4	0.20	1.1	0.78	0.016	0.031	0.15	0.047	0.0068	0.0090	0.010
	MAY	1.5	0.081	1.0	0.23	0.020	0.029	0.14	0.022	0.0064	0.0030	0.016
	JUN	1.0	0.15	1.1	0.33	0.010	0.019	0.21	0.0092	0.0017	0.00072	0.0087
	JUL	0.70	0.064	0.98	0.086	0.18	0.033	0.052	0.0074	0.0012	0.00051	0.0057
	max.	3.0	0.31	1.5	1.6	0.18	0.036	0.24	0.12	0.021	0.050	0.026
	min.	0.65	0.064	0.55	0.086	0.0023	0.0085	0.023	0.0074	0.0012	0.0005	0.0057
	mean	1.5	0.15	1.0	0.63	0.041	0.024	0.11	0.051	0.0073	0.011	0.014

3.2. *Analyses of aerosols and their chemical species*

First, the total particulate mass of collected aerosols on Teflon filters was measured. The collected aerosols were then extracted ultrasonically in distilled-deionized water. We analyzed the concentrations of sulfate (SO_4^{2-}), nitrate (NO_3^-) and chloride (Cl^-) by an ion chromatograph (Yokogawa Electric Works Inc., IC-100, column SAM3-125), and the concentrations of sodium (Na^+), calcium (Ca^{2+}), magnesium (Mg^{2+}) and potassium (K^+) by an atomic absorption spectrometer (Hitachi Inc., 170-30) in the extracted solution. The ammonium (NH_4^+) concentrations were measured colorimetrically by using the indophenol method (WEATHERBURN, 1967).

The carbonaceous components on the quartz filters were analyzed by a NC Analyzer (SUMIGRAPH NC-80) and GC-FID (HITACHI 164).

Table 1 (Continued).

(b) Norilsk

	Month	TPM	E.C.	O.C.	SO_4^{2-}	Cl^-	NO_3^-	NH_4^-	Na^+	Ca^{2+}	Mg^{2+}	K^+
1993	AUG.f	28	1.7	5.6	3.7	0.14	0.19	0.26	0.15	1.6	0.088	
	AUG.s	11	1.2	2.7	2.1	0.077	0.10	0.65	0.089	0.41	0.041	
	SEP.f	23	1.9	3.7	3.7	0.17	0.16	0.48	0.099	1.1	0.060	
	SEP.s	22	1.8	4.0	2.9	0.10	0.16	0.15	0.071	0.42	0.045	
	OCT.f	28	4.2	12	2.8	0.10	0.11	0.60	0.14	0.64	0.045	
	OCT.s	23	1.1	3.3	4.0	0.24	0.22	0.18	0.25	1.2	0.11	
	NOV.f	22	2.0	5.9	5.8	0.16	0.49	1.1	0.24	1.1	0.12	
	NOV.s	13	1.5	3.6	3.3	0.27	0.17	0.79	0.29	0.47	0.082	
	DEC.f	21	2.3	5.4	4.8	0.044	0.18	1.1	0.12	0.22	0.067	
	DEC.s	14	2.8	4.2	3.1	0.12	0.14	0.76	0.16	0.71	0.074	
1994	JAN.f	7.8	1.00	2.6	2.2	0.011	0.064	0.52	0.090	0.094	0.030	
	JAN.s	23	2.7	3.8	5.9	0.052	0.20	0.95	0.18	1.9	0.076	
	FEB.f	26	3.4	4.6	6.9	0.0043	0.13	1.1	0.17	0.65	0.071	
	FEB.s	18	2.7	4.0	5.0	0.015	0.27	1.2	0.16	0.64	0.061	
	MAR.f	16	2.0	3.4	6.8	0.046	0.10	1.5	0.17	0.85	0.085	0.24
	MAR.s	22	2.8	3.8	8.1	0.076	0.11	1.5	0.14	2.2	0.082	0.34
	APR.f	18	2.2	3.5	12	0.021	0.050	1.1	0.18	0.53	0.080	0.17
	APR.s	30	3.4	5.1	14	0.016	0.038	1.9	0.18	0.95	0.14	0.30
	MAY.f	26	1.5	2.9	16	0.049	0.022	1.3	0.20	0.62	0.12	0.48
	MAY.s	23	1.3	2.4	15	0.14	0.039	1.5	0.13	0.58	0.11	0.22
	JUN.f	14	0.93	2.9	8.6	0.019	0.045	1.8	0.074	0.25	0.028	0.097
	JUN.s	29	2.5	8.0	9.7	0.11	0.11	1.8	0.14	1.0	0.052	0.23
	JUL.f	27	0.11	1.9	12	0.075	0.094	0.24	0.12	1.3	0.074	0.28
	JUL.s	6.1	1.8	4.0	0.89	0.028	0.054	0.88	0.040	0.30	0.016	0.086
	max.	30	4.2	12	16	0.27	0.49	1.9	0.29	2.2	0.14	0.48
	min.	6.1	0.11	1.9	0.89	0.0043	0.022	0.15	0.040	0.094	0.016	0.086
	mean	20	2.0	4.3	6.6	0.087	0.14	0.97	0.15	0.83	0.073	0.24

3.3. Trajectory analyses

To estimate the potential source areas for the pollution in Tiksi and the potential contribution of the Norilsk source area to Arctic haze, we calculated 10-day backward trajectories of air parcels to Tiksi and forward trajectories from Norilsk in every month in 1994 by using an isentropic air trajectory model (NAKAYAMA *et al.*, 1994).

In the calculations, the ECMWF (the European Center for Medium Range Weather Forecast) data of geopotential, temperature, wind velocity and relative humidity at 7 pressure levels (1000, 925, 850, 700, 600, 500, 400 hPa) and latitude-longitude grid with 2.5 degree resolution in every six hours in 1994 were used.

We assigned the 850 hPa pressure level to the arrival or starting elevation, and then

Table 1 (Continued).

(c) Yakutsk

	Month	TPM	E.C.	O.C.	SO_4^{2-}	Cl^-	NO_3^-	NH_4^-	Na^+	Ca^{2+}	Mg^{2+}	K^+
1993	AUG.f	26	2.7	9.0	0.65	0.20	0.31	0.55	0.31	1.0	0.16	
	AUG.s	39	5.3	16	0.46	0.089	0.20	0.57	0.14	0.72	0.077	
	SEP.f	19	2.7	4.9	0.49	0.093	0.28	0.31	0.040	0.84	0.029	
	SEP.s	13	2.0	4.1	0.30	0.031	0.10	0.47	0.13	0.56	0.016	
	OCT.f	7.2	1.8	3.9	0.42	0.027	0.067	0.65	0.020	0.18	0.0013	
	OCT.s	11	3.6	5.4	0.76	0.037	0.11	1.1	0.015	0.0051	0.0024	
	NOV.f	18	7.5	5.5	1.3	0.051	0.29	2.0	0.10	0.066	0.019	
	NOV.s	18	4.6	10	0.99	0.079	0.29	1.5	0.14	0.18	0.018	
	DEC.f	14	3.6	7.1	0.83	0.20	0.17	1.0	0.28	0.12	0.068	
	DEC.s	12	4.2	10	0.45	0.038	0.12	1.2	0.032	0.060	0.0044	
1994	JAN.f	13	4.3	2.9	0.70	0.036	0.12	0.60	0.072	0.046	0.016	
	JAN.s	13	5.7	5.5	0.47	0.033	0.088	0.89	0.033	0.74	0.0070	
	FEB.f	11	4.1	5.6	0.89	0.052	0.18	1.4	0.047	0.015	0.0077	
	FEB.s	*42	12	11				1.6	0.040	0.083	0.014	
	MAR.f	24	7.8	7.5	3.1	0.10	0.49	1.4	0.084	0.16	0.023	
	MAR.s	17	5.6	6.1	1.4	0.060	0.25	0.86	0.052	0.14	0.015	
	APR.f	13	8.8	4.2	1.6	0.056	0.21	0.89	0.052	0.18	0.024	
	APR.s	7.7	3.2	2.2	1.2	0.051	0.26	0.54	0.056	0.36	0.026	
	MAY.f	13	2.6	3.3	0.64	0.028	0.13	0.24	0.041	0.28	0.020	
	MAY.s	16	3.9	6.6	1.1	0.12	0.19	0.50	0.065	0.40	0.035	
	JUN.f	*2.4	2.7	4.4	0.056	0.0079	0.032	0.22	0.014	0.073	0.0078	
	JUN.s	12	2.1	3.9	0.17	0.024	0.096	0.11	0.050	0.36	0.018	
	JUL.f	14	2.5	3.3	0.30	0.040	0.14	0.14	0.038	0.51	0.021	
	JUL.s	8.3	2.6	3.8	0.19	0.029	0.089	0.092		0.041	0.010	
	max.	42	12	16	3.1	0.20	0.49	2.0	0.31	1.0	0.16	
	min.	2.4	1.8	2.2	0.056	0.0079	0.032	0.092	0.014	0.0051	0.0013	
	mean	15	4.2	6.1	0.80	0.064	0.18	0.79	0.081	0.27	0.026	

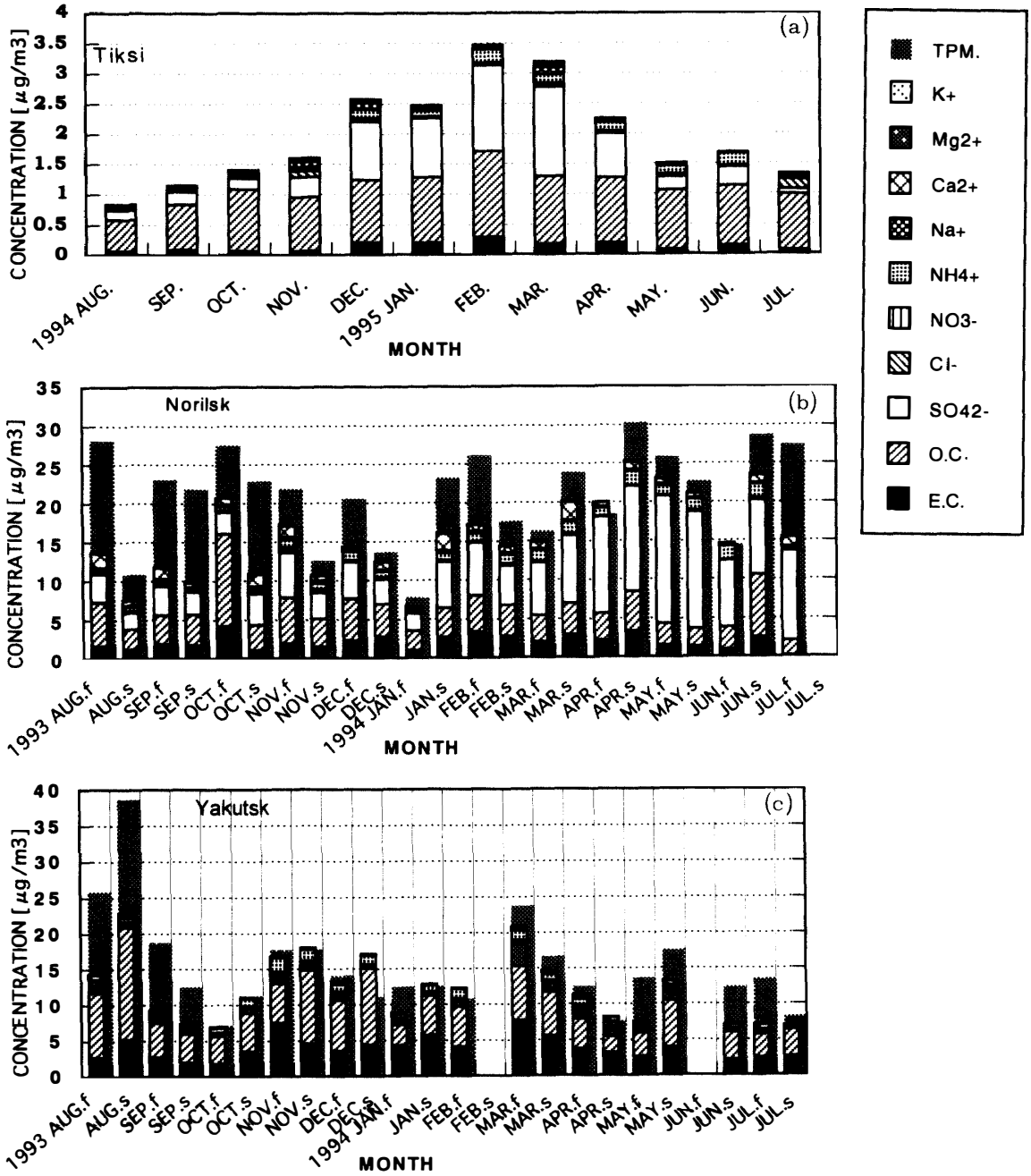

Fig. 2a–c. Monthly atmospheric concentrations of aerosol chemical species at Tiksi from August 1994 to July 1995 (a), TPM and aerosol chemical species at Norilsk (b) and Yakutsk (c) from August 1993 to July 1994.

calculated the trajectories. Trajectories arrived two times daily, at 00 and 12 UT. When the isentropic surface intersected the Earth's surface, we switched from the isentropic mode to the 3 dimensional advection mode, in which air parcels were advected by 3 dimensional wind until returning to the original isentropic surface.

4. Results and Discussion

4.1. Concentrations of atmospheric aerosols and their chemical species

Table 1 shows the results of the measured atmospheric concentrations of aerosol chemical species from August 1994 to July 1995 at Tiksi and from August 1993 to July 1994 at Norilsk and Yakutsk. TPM concentrations at Tiksi could not be measured exactly because of the low amounts of aerosols collected on each filter. In Tiksi the range and the mean of the mass concentrations of aerosol chemical species were elemental carbon, 0.06–0.31, 0.15; organic carbon, 0.55–1.5, 1.0; SO_4^{2-}, 0.09–1.6, 0.6; Cl^-, 0.002–0.18, 0.04; NO_3^-, 0.009–0.04, 0.02; NH_4^+, 0.02–0.24, 0.1; Na^+, 0.007–0.12, 0.05; Ca^{2+}, 0.001–0.021, 0.007; Mg^{2+}, 0.001–0.050, 0.01; K^+, 0.006–0.026, 0.01 μgm^{-3}. They were the same level as those measured at Ester Dome, Alaska by OHTA *et al.* (1995) as follows: TPM, 0.72–2.1; SO_4^{2-}, 0.2–1.3; organics, 0.3–0.8; elemental carbon, 0.07–0.2 μgm^{-3}.

Figure 2a shows the variations of the mass concentrations of aerosol chemical species at Tiksi. The concentrations of aerosol chemical species, especially elemental carbon and sulfate, were remarkably higher from December to April than in summer. The maximum sulfate concentration at Tiksi was about seven times higher than that in summer. This suggests that Arctic haze occurs over Tiksi during winter and spring. The averaged concentrations of major aerosol chemical species at Tiksi in this period were SO_4^{2-}, 1.0; organic carbon, 1.1; elemental carbon, 0.19 μgm^{-3}. BARRIE (1986) reported a January–April mean SO_4^{2-} concentration of 1.5–3.9 μgm^{-3} in the Norwegian Arctic and 1.2–2.2 μgm^{-3} in the North American Arctic. RAHN and HEIDAM (1981) concluded that the average concentrations of anthoropogenic aerosols in the Arctic during winter and spring were 2 μgm^{-3} for SO_4^{2-}, 1 μgm^{-3} for organic compounds and 0.3–0.5 μgm^{-3} for elemental carbon. We had about the same level of major aerosol chemical species mentioned above.

On the other hand, the mean concentrations at Norilsk from August 1993 to July 1994 were TPM, 20; elemental carbon, 2.0; organic carbon, 4.3; SO_4^{2-}, 6.6; Cl^-, 0.09; NO_3^-, 0.14; NH_4^+, 1.0; Na^+, 0.15; Ca^{2+}, 0.83; Mg^{2+}, 0.07; K^+, 0.2 μgm^{-3}. There was no significant seasonal variation as shown in Fig. 2b, because the sampling site was surrounded by factories. The atmospheric aerosol at Norilsk had extremely high concentrations of sulfate. This suggests that the Norilsk Nickel complex emitted a large amount of SO_2 to the Arctic atmosphere.

At Yakutsk, the mean concentrations from August 1993 to July 1994 were TPM, 15; elemental carbon, 4.2; organic carbon, 6.1; SO_4^{2-}, 0.8; Cl^-, 0.06; NO_3^-, 0.18; NH_4^+, 0.79; Na^+, 0.08; Ca^{2+}, 0.27; Mg^{2+}, 0.03 μgm^{-3}. Figure 2c shows the monthly variations of measured TPM and the aerosol chemical species at Yakutsk. The aerosols at Yakutsk contained a large amount of carbonaceous matter, which might be emitted from forest or forest fires which frequently occur in the taiga.

The analyzed aerosol chemical species did not include soil components such as aluminum, which might cause disagreement between the TPM and the sum of the chemical species.

4.2. Potential transport pathways

4.2.1. Potential source areas for pollution in Tiksi

We calculated 10-day backward trajectories of air parcels to Tiksi. Figure 3 shows

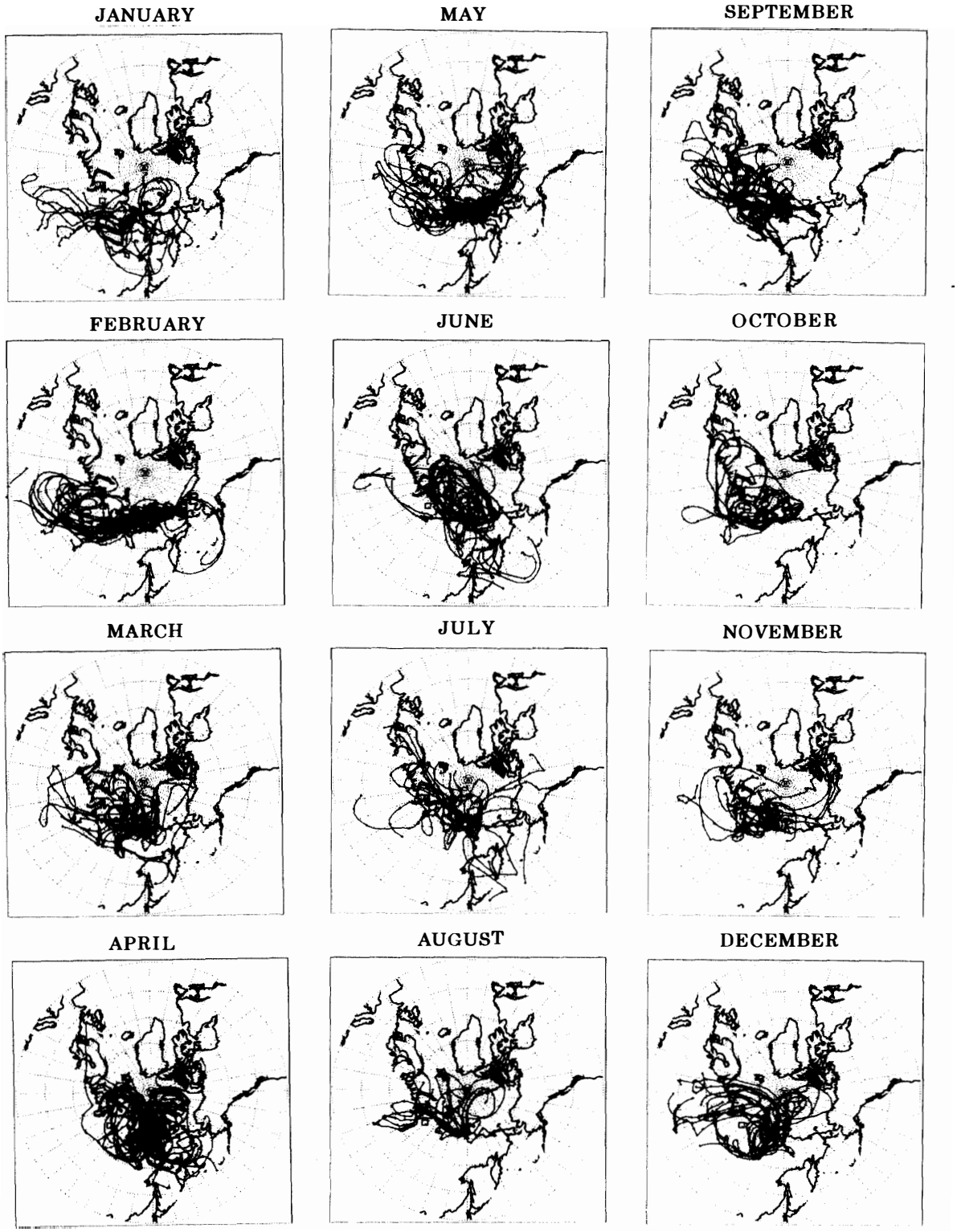

Fig. 3. Ten-day isentropic backward trajectories of air parcels arriving at the 850 hPa pressure height over Tiksi in each month in 1994.

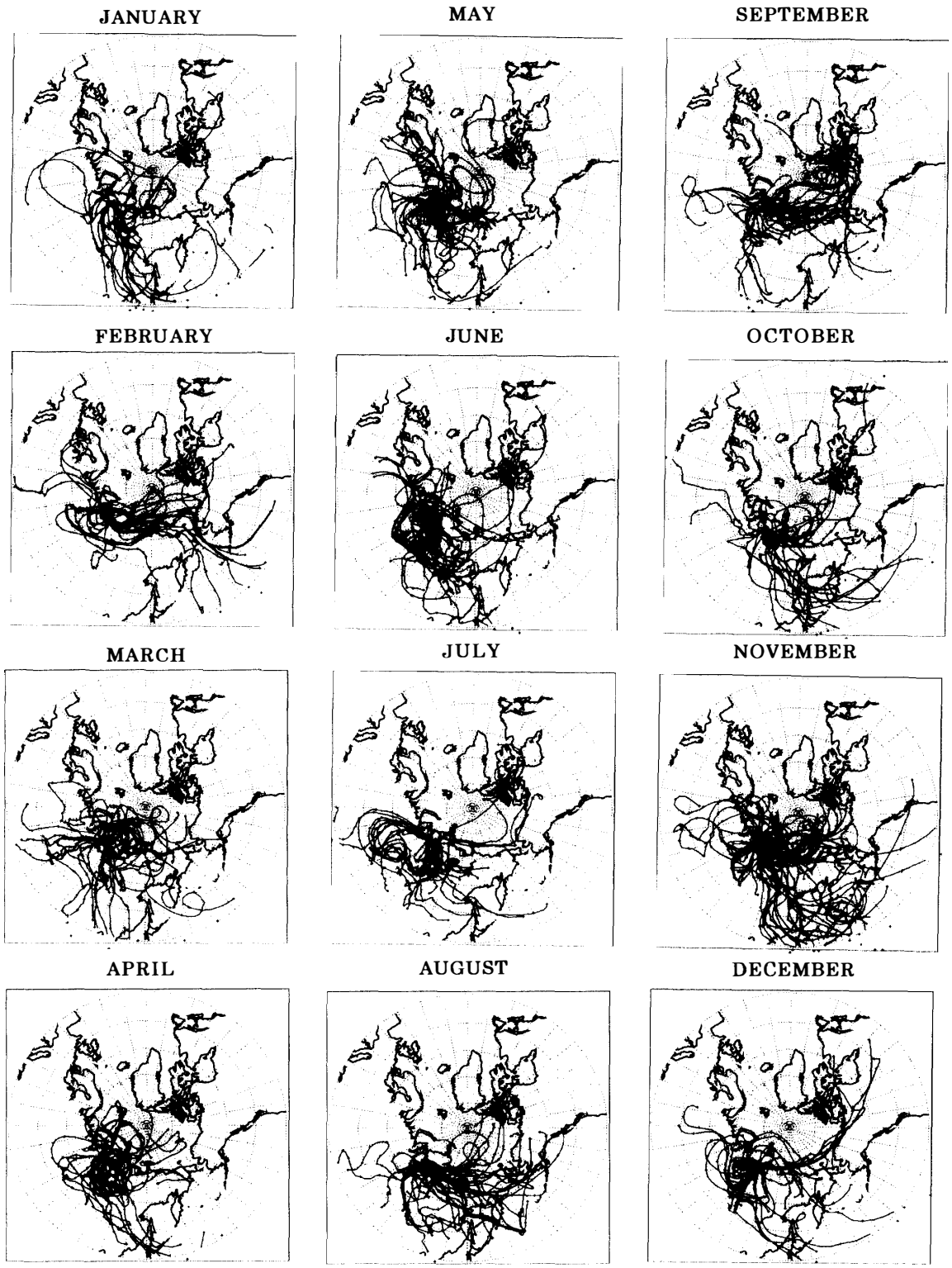

Fig. 4. Ten-day isentropic forward trajectories of air parcels starting at the 850 hPa pressure height over Tiksi in each month in 1994.)

the monthly isentropic trajectories arriving at the 850 hPa pressure level over Tiksi.

Most trajectories arriving at Tiksi from June to August in 1994 showed that frequent transport pathways usually came from the Arctic Ocean so that there were almost no anthoropogenic emission sources. Further, there are more clouds and precipitation in summer than in other seasons in the Arctic (HUSCHKE, 1969). It has been hypothesized that the seasonal variation of arctic air polluiton could be controlled by the seasonal variation of clouds and precipitation due to scavenging by increased dry and wet depostion (RAATZ, 1991). We, thus, supposed that this situation produced low concentrations of pollutants at Tiksi during the summer. The trajectories from September to October passed over the Norilsk area, Ural area and other source areas around the Arctic Ocean. However there will be also the scavenging processes mentioned above during the transport so that pollutants might not be transported to Tiksi. In winter, the trajectories showed the following three major transport pathways to Tiksi: (i) transport directly from the Urals and/or Norilsk, (ii) transport from the Urals and other source areas as the Kola Peninsula and the Pechora basin and so on through the Arctic Ocean and (iii) transport from southern Yakutia. The former two transport pathways affect the atmospheric pollution in Tiksi. In spring, the air mass tended to be transported from the Arctic Ocean.

4.2.2. Potential transport pathways for pollutants from Norilsk

Figure 4 shows 10-day forward trajectories of air parcels from Norilsk in each month. According to the trajectories, air masses from Norilsk were frequently transported to Tiksi and/or the Arctic Ocean in February and from August to December. However, except in winter, the pollutants are scavenged due to the above-mentioned processes. In fact, when we visited Norilsk, this region was usually covered by persistent and extensive layers of cloud. In March and April, the air masses tended to drift around the Norilsk area, and then were transported to the Arctic Ocean and other areas. Thus the pollutant emission from Norilsk are one of the sources of Arctic haze as suggested in SHAW (1982).

This also implies that the emitted SO_2 gas affects the forest in the taiga zone because the air masses from Norilsk were transported to the southern taiga area through the year.

5. Conclusions

Atmospheric aerosols and the chemical species at three sites in the Siberian Arctic were continuously measured near the ground. The TPM concentrations ranged from 6–30 μgm^{-3} at Norilsk and 7–39 μgm^{-3} at Yakutsk during August 1993–July 1994.

The aerosol chemical species concentrations at Tiksi showed significant seasonal variation mainly derived from that of sulfate. On the other hand, there were no seasonal variations in Yakutsk or Norilsk. Major aerosol constituents were sulfate at Norilsk, and carbonaceous particles at Yakutsk.

According to the trajectory analyses, the atmospheric pollutants at Tiksi during winter/spring would be mainly transported from Norilsk,Ural and other source areas. And pollutant emissions from Norilsk are one of the sources of Arctic haze

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