

# SUMMER CONCENTRATIONS OF ATMOSPHERIC POLLUTANTS IN URBAN AND RURAL AREAS OF SIBERIA

Sachio Ohta<sup>1</sup>  
Tatsuya Fukasawa<sup>2</sup>  
Naoto Murao<sup>3</sup>  
Vladimir N. Makarov<sup>4</sup>

## Abstract

Atmospheric concentrations of aerosol chemical species, SO<sub>2</sub> and HNO<sub>3</sub> were measured at a radio relay station 20 km west of Yakutsk city, Siberia, during July 23–31, 1992. The mean concentrations of the aerosol species were SO<sub>4</sub><sup>2-</sup>, 0.38; NO<sub>3</sub><sup>-</sup>, 0.04; Cl<sup>-</sup>, 0.02; NH<sub>4</sub><sup>+</sup>, 0.13; Na<sup>+</sup>, 0.04; Ca<sup>2+</sup>, 0.56; Mg<sup>2+</sup>, 0.02 μgm<sup>-3</sup>. The mean concentrations of SO<sub>2</sub> and HNO<sub>3</sub> were 0.04 ppbv and 0.006 ppbv, respectively. These concentrations were similar to those recorded at a rural site near Fairbanks, Alaska. Atmospheric concentrations of NO<sub>2</sub> and SO<sub>2</sub> in urban areas of Siberia were also measured in the summer of 1993. The NO<sub>2</sub> concentrations at Khabarovsk, Yakutsk and Norilsk were 5–11, 2–10 and 2–6 ppbv, respectively. They were lower than those in Sapporo in summer. The SO<sub>2</sub> concentrations in Norilsk were high due to poor emission control from copper and nickel smelting.

**KEYWORDS:** *atmospheric pollutants, Siberia, aerosol chemical species, SO<sub>2</sub>, HNO<sub>3</sub>, NO<sub>2</sub>*

## 1. Introduction

Atmospheric aerosols affect the global climate by changing the planetary albedo (reflectivity of short wavelength solar radiation at the top of the atmosphere) through the scattering and absorption of solar radiation (Potter and Cess, 1984; Charlson *et al.*, 1992). Furthermore, atmospheric aerosols, in particular sulfate aerosols (SO<sub>4</sub><sup>2-</sup>) which are mainly converted from sulfur dioxide (SO<sub>2</sub>) in the atmosphere, may also change the global climate due to modification of cloud albedo (shortwave cloud reflectivity) through the increase of cloud droplet concentration (Twomey *et al.*, 1984; Kaufman and Chou, 1993).

In order to evaluate the abovementioned effects on the climate, we need to determine the global distribution of aerosol chemical species. There are, however, only sparse background data on aerosol concentrations of different species, in particular in central Eurasia. In this study, therefore, we measured the concentrations of atmospheric aerosol species in a rural area of Yakutsk, Siberia, Russia. We also measured concentrations of atmospheric SO<sub>2</sub> and nitric

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1 D. Sci., Prof., Dept. of Sanitary and Environ. Engineering, Hokkaido Univ., Sapporo 060, Japan.

2 M. Eng., Dept. of Sanitary and Environ. Engineering, Hokkaido Univ.

3 D. Eng., Associate Prof., Dept. of Sanitary and Environ. Engineering, Hokkaido Univ.

4 D. Sci., Head of Geochem. Lab., Permafrost Inst., Yakutsk, Russia.

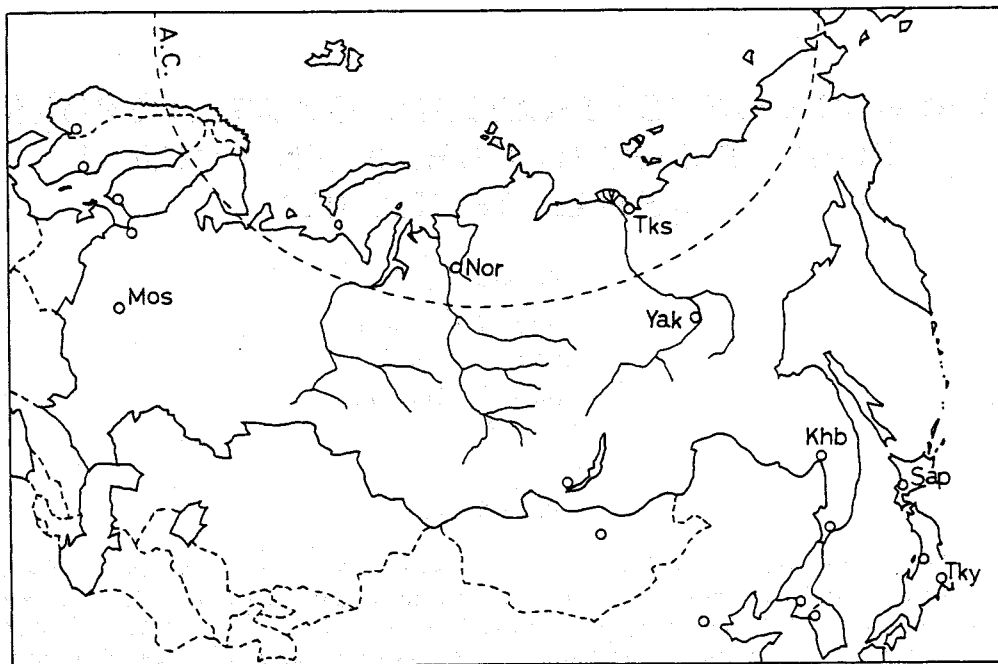


Figure 1. Map of Russia and Japan. A.C.: Arctic Circle, Mos: Moscow, Nor: Norilsk, Tks: Tiksi, Yak: Yakutsk, Khb: Khabarovsk, Sap: Sapporo, Tky: Tokyo.

acid ( $\text{HNO}_3$ ) since they are precursors which are converted into sulfate and nitrate aerosol, respectively, in the atmosphere. Urban environmental degradation has been reported recently in Russia (Green, 1993), but, we have no available data on the air pollution, in particular in Siberia. Thus, plans were made to measure the concentrations of atmospheric nitrogen dioxide ( $\text{NO}_2$ ) in Khabarovsk, Yakutsk and Norilsk, and of  $\text{SO}_2$  in Norilsk. Nitrogen dioxide and sulfur dioxide are deleterious compounds of air pollution. They affect human health by causing respiratory diseases such as bronchitis and asthma. They also cause photochemical smog and acid rain problems. Monitoring of the atmospheric concentration is, thus, important to make plans for the protection of the human health and the environment.

Figure 1 shows a map of Russia and Japan. In this research project we visited Khabarovsk and Yakutsk in the summer of 1992, and Khabarovsk, Yakutsk and Norilsk in the summer of 1993.

Khabarovsk is the capital of the Khabarovsk Region, Russia, and is an industrial city on the banks of the Amur river, with a population of 600,000. Yakutsk is the capital of the Republic of Sakha, and is an agricultural city on the banks of the Lena river, with a population of 200,000. Norilsk is a mining city, the leading copper and nickel producer in Russia, situated 80 km east of the Yenisei river, with a population of 285,000.

## 2. Experimental Procedures

### 2.1 Sampling of atmospheric aerosols, $\text{SO}_2$ and $\text{HNO}_3$

We collected atmospheric aerosols and atmospheric  $\text{SO}_2$  and  $\text{HNO}_3$  every 24 or 48 hours during July 23–31, 1992, at a radio relay station 20 km west of Yakutsk city shown in Figure

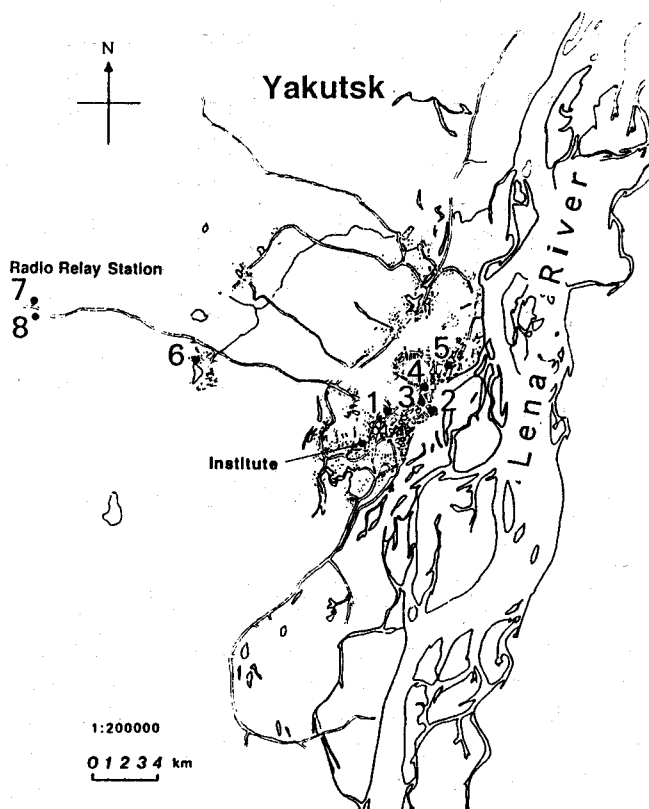


Figure 2. Map of Yakutsk.

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Atmospheric aerosols were collected on Teflon filters (Sumitomo Fluoropore AF07P) with a high-volume air sampler (Shibata Scientific Technology Ltd., HVS-500) at a flow rate of 500 liter per minute equipped with an impactor whose 50 % cut-off diameter that was  $5\ \mu\text{m}$ .

The collected samples were extracted ultrasonically with distilled-deionized water. The concentrations of sulfate ( $\text{SO}_4^{2-}$ ), nitrate ( $\text{NO}_3^-$ ) and chloride ( $\text{Cl}^-$ ) in the extracted solution were determined by using an ion chromatograph (Yokogawa Electric Works Inc., IC-100, column SAM3-125). The concentrations of sodium ( $\text{Na}^+$ ), calcium ( $\text{Ca}^{2+}$ ) and magnesium ( $\text{Mg}^{2+}$ ) were determined with an atomic absorption spectrometer (Hitachi Inc. 170-30). The ammonium ( $\text{NH}_4^+$ ) concentration was measured colorimetrically by using the indophenol method (Weatherburn, 1967).

Atmospheric  $\text{SO}_2$  and  $\text{HNO}_3$  were collected with reagent-soaked paper filters at a flow rate of 25 liter per minute. We used filters soaked with  $\text{Na}_2\text{CO}_3$  and  $\text{NaCl}$ , respectively, for sampling  $\text{SO}_2$  and  $\text{HNO}_3$ . Figure 3 shows the sampling system for collecting atmospheric  $\text{SO}_2$  or  $\text{HNO}_3$ . We removed atmospheric aerosol by using Teflon filter AT, and collected atmospheric  $\text{SO}_2$  or  $\text{HNO}_3$  on reagent-soaked paper filters G1 and G2.

The absorbed  $\text{SO}_2$  on the reagent-soaked paper filter was extracted with distilled-deionized water and oxidized to sulfate by dropping  $\text{H}_2\text{O}_2$ . The sulfate concentrations were determined with the ion chromatograph. The absorbed  $\text{HNO}_3$  was measured colorimetrically using the

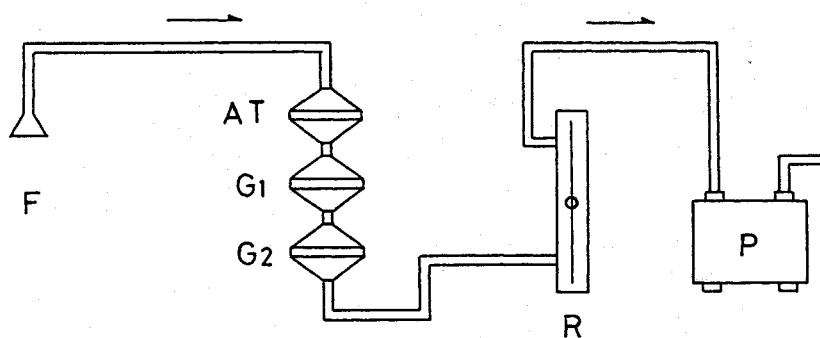


Figure 3. Sampling system of atmospheric  $\text{SO}_2$  or  $\text{HNO}_3$ . F: Funnel, AT: Teflon filter, G1, G2: Gas sampling filter, R: Rota meter, P: Air pump.

hydrazine reduction GR reagent method (Ohta *et al.*, 1970) after extraction with distilled-deionized water.

## 2.2 Diffusion sampling of atmospheric $\text{NO}_2$ and $\text{SO}_2$

In the summer of 1993 we measured atmospheric concentrations of  $\text{NO}_2$  in Khabarovsk, Yakutsk and Norilsk, and of  $\text{SO}_2$  in Norilsk. In the measurements we used diffusion samplers of the type shown in Figure 4. The sampler was made of a plastic pot 36 mm in diameter and 33 mm in depth, which was packed with aluminum foil, polyethylene sheet, reagent-soaked paper, Polyflon filter (TOYO PF040), and an O-ring. The reagent-soaked paper was made by dipping

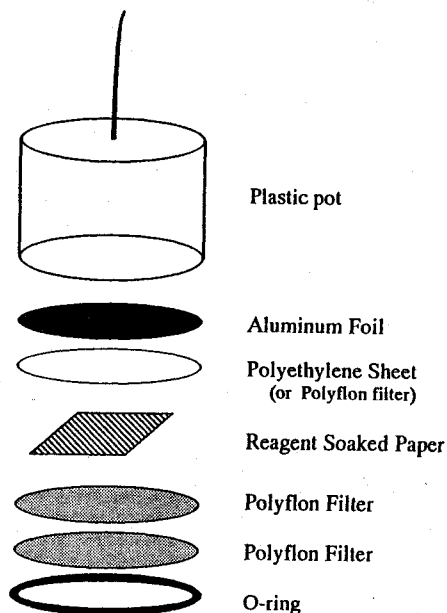


Figure 4. Diffusion sampler.

chromatography paper (TOYO No. 51A) with a size of  $2 \times 2.5$  cm into aqueous triethanolamine solution and aqueous  $\text{Na}_2\text{CO}_3$  solution, respectively, for  $\text{NO}_2$  and  $\text{SO}_2$  sampling. We used the soaked paper packed in the pot in dry condition for a sampling period longer than two weeks, and in wet condition for shorter sampling periods.

After exposure of the samplers in the atmosphere, we took out the reagent-soaked papers and extracted nitrite or sulfite, derived from absorbed  $\text{NO}_2$  or  $\text{SO}_2$ , respectively, with distilled-deionized water. The sulfite was oxidized to sulfate by dropping  $\text{H}_2\text{O}_2$ , and the sulfate concentrations were determined by using the ion chromatograph. The nitrite concentrations were analyzed by the Saltzman method (Saltzman, 1954).

We exposed the samplers at 6 sites in Khabarovsk for 21 days (July 21–August 11), 8 sites in Yakutsk for 14 days (July 25–August 8), and 9 sites in Norilsk for 6 days (July 29–August 4).

Because the reagent-soaked paper absorbs sample gas in proportion to the atmospheric concentration, we must determine a conversion coefficient that expresses the amount of absorbed gas to the atmospheric concentration. This coefficient was obtained in Sapporo in the summer of 1993. We put the diffusion samplers close to the sampling nozzles of the automatic  $\text{NO}_2$  or  $\text{SO}_2$  meter at eight monitoring sites in Sapporo. At each site we put three  $\text{NO}_2$  diffusion samplers and three  $\text{SO}_2$  diffusion samplers.

Figure 5 shows the correlation between the amount of  $\text{NO}_2$  absorbed in the soaked paper in dry condition for 30 days exposure and the mean atmospheric concentration of  $\text{NO}_2$  measured with the automatic  $\text{NO}_2$  monitor for the 30 days. Figure 6 shows the corresponding correlation for the  $\text{SO}_2$  sampling with the reagent-soaked paper in wet condition. From these figures we determined the conversion coefficients.

For the  $\text{NO}_2$  sampling we obtained values on the correlation for atmospheric concentrations up to 30 ppbv. For the  $\text{SO}_2$  sampling, however, only concentrations between 4 and 6 ppbv were available. Thus, we cannot place high confidence in  $\text{SO}_2$  concentrations above 10 ppbv.

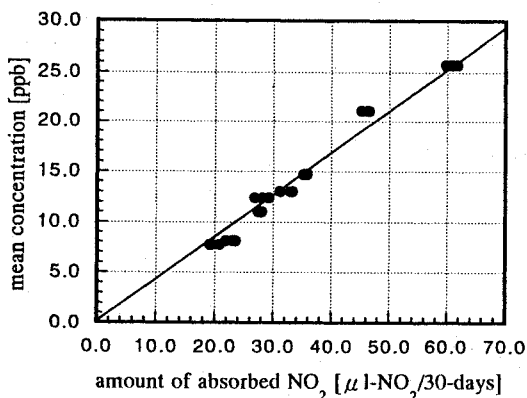


Figure 5. Correlation between the amount of  $\text{NO}_2$  absorbed in the reagent-soaked paper in dry condition for 30 days exposure and the mean atmospheric  $\text{NO}_2$  concentration measured by automatic  $\text{NO}_2$  monitor for the 30 days.

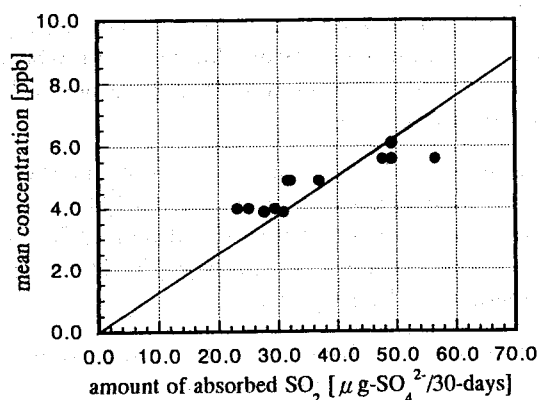


Figure 6. Correlation between the amount of SO<sub>2</sub> absorbed in the reagent-soaked paper in wet condition for 30 days exposure and the mean atmospheric SO<sub>2</sub> concentration measured by automatic SO<sub>2</sub> monitor for the 30 days.

### 3. Results and Discussion

#### 3.1 Atmospheric concentrations of aerosol chemical species, SO<sub>2</sub> and HNO<sub>3</sub> in the rural area of Yakutsk

Table 1 shows results of the measured atmospheric concentrations of aerosol chemical species, SO<sub>2</sub> and HNO<sub>3</sub> at the radio relay station (r.r.s.) 20 km west of Yakutsk city during July 23–31, 1992. The mean concentrations of the aerosol chemical species were SO<sub>4</sub><sup>2-</sup>, 0.38; NO<sub>3</sub><sup>-</sup>, 0.04; Cl<sup>-</sup>, 0.02; NH<sub>4</sub><sup>+</sup>, 0.13; Na<sup>+</sup>, 0.04; Ca<sup>2+</sup>, 0.56; Mg<sup>2+</sup>, 0.02 μgm<sup>-3</sup>. The mean concentrations of SO<sub>2</sub> and HNO<sub>3</sub> were 0.04 ppbv and 0.006 ppbv, respectively.

In Table 2 we show for comparison the concentrations of atmospheric aerosol chemical

Date	Aerosol (μg/m <sup>3</sup> )							Gas (ppbv)	
	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	Na <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	SO <sub>2</sub>	HNO <sub>3</sub>
July 23-24	0.53	0.03	0.01	0.19	0.05	0.57	0.02	0.03	0.001
24-25	0.48	0.05	0.02	0.16	0.05	0.57	0.02	0.08	0.008
25-26	0.21	0.03	0.02	0.08	0.03	0.47	0.02	0.03	0.004
26-27	0.11	0.01	0.00	0.04	0.02	0.27	0.01	0.01	0.000
27-29	0.35	0.06	0.01	0.11	0.04	0.80	0.03	0.05	0.005
29-30	0.75	0.10	0.02	0.23	0.06	1.16	0.04	0.11	0.020
30-31	0.24	0.01	0.03	0.09	0.03	0.05	0.01	0.00	0.001
Mean	0.38	0.04	0.02	0.13	0.04	0.56	0.02	0.04	0.006

Table 1. Atmospheric concentrations of aerosol chemical species, SO<sub>2</sub> and HNO<sub>3</sub> at the radio relay station 20 km west of Yakutsk city, Siberia, during July 23–31, 1992.

	Ester Dome		Wolkersdorf		Bermuda
	July	Aug.	July	Aug.	Aug.
SO <sub>4</sub> <sup>2-</sup>	0.20	0.20	8.3	9.5	1.76
NO <sub>3</sub> <sup>-</sup>	0.001	0.001	1.6	2.4	0.20
Cl <sup>-</sup>	0.000	0.000	0.02	0.01	0.51
NH <sub>4</sub> <sup>+</sup>	0.067	0.069	3.0	4.6	0.34
Na <sup>+</sup>	0.002	0.007	0.17	0.15	1.90
Ca <sup>2+</sup>	0.002	0.004			0.13
Mg <sup>2+</sup>	0.001	0.001			
SO <sub>2</sub>	0.045	0.111	2.45	2.90	0.32
HNO <sub>3</sub>	0.014	0.014	0.43	0.50	0.10

Table 2. Summer concentrations of aerosol chemical species ( $\mu\text{gm}^{-3}$ ), SO<sub>2</sub> and HNO<sub>3</sub> (ppbv) in the rural areas of Ester Dome, Alaska (Ohta *et al.*, 1995), Wolkersdorf in northeastern Austria (Puxbaum *et al.*, 1993), and the southwest coast of Bermuda (Wolff *et al.*, 1986).

species ( $\mu\text{gm}^{-3}$ ) and atmospheric SO<sub>2</sub> and HNO<sub>3</sub> (ppbv) recorded in other rural areas. Ohta *et al.* (1995) measured the concentrations at Ester Dome (at a height of 720 m), 12 km west of Fairbanks, Alaska, from August 1991 to August 1992. Puxbaum *et al.* (1993) measured the concentrations from November 1990 to October 1991 at the air quality measurement site Wolkersdorf (16° 31' E, 48° 23' N), Lower Austria, located in a hilly region 40 km south of the Czech and 70 km northwest of the Hungarian border. Wolff *et al.* (1986) measured the concentrations at the southwest coast of Bermuda Island during August 1982, and January and December 1983.

The summer SO<sub>4</sub><sup>2-</sup> concentrations at r.r.s. of Yakutsk in Table 1 were similar to those at Ester Dome in Table 2. The SO<sub>4</sub><sup>2-</sup> concentrations at Bermuda were higher than those in Yakutsk, because in coastal areas a considerable amount of sulfate is formed from DMS (dimethylsulfide) which is released by planktonic algae at the ocean surface (Andreae, 1986).

The concentrations of NO<sub>3</sub><sup>-</sup> at r.r.s. in Yakutsk were a little higher than those at Ester Dome, but much lower than those at Wolkersdorf and Bermuda. The nitrate aerosol is formed through nucleation from HNO<sub>3</sub> gas in the atmosphere. However, the concentrations of HNO<sub>3</sub> at r.r.s. in Yakutsk were also much lower than those at Wolkersdorf and Bermuda, because less HNO<sub>3</sub> was produced at Yakutsk or Ester Dome due to less NO<sub>x</sub> and lower hydrocarbon concentrations, in addition to a lower air temperature than at Wolkersdorf or Bermuda.

At Wolkersdorf the concentrations of SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> were much higher than at the other locations. This was due to transport of large amounts of SO<sub>x</sub>, NO<sub>x</sub> and hydrocarbons discharged in Eastern Europe, where coal is mainly combusted for power generation and domestic heating. In contrast, in Yakutsk natural gas is combusted, which discharges little pollutants.

The concentrations of NH<sub>4</sub><sup>+</sup> at r.r.s. in Yakutsk were similar to those at Bermuda. The concentrations at Wolkersdorf were again much higher than at the other locations. The concentrations of Cl<sup>-</sup> and Na<sup>+</sup> at r.r.s. in Yakutsk were a little higher than those at Ester Dome, but much lower than those at Bermuda where they were derived from sea salts.

The concentrations of  $\text{SO}_2$  at r.r.s. in Yakutsk were similar to those at Ester Dome, but about one tenth of those at Bermuda, where  $\text{SO}_2$  was formed due to conversion from DMS in the atmosphere. The high  $\text{SO}_2$  concentrations at Wolkersdorf were also due to the transport of large amounts of  $\text{SO}_2$  discharged in Eastern Europe. The calcium concentrations ( $\text{Ca}^{2+}$ ) at r.r.s. in Yakutsk were higher than those in the other areas. Because Yakutsk is located on the riverbed of the Lena river, the ground surface is covered with fine sand rich in calcium. In summer strong solar radiation dries up the surface soil and fine sand particles blow up and fly over r.r.s.

	Sampling site		NO <sub>2</sub> concentration (ppbv)			
	No					Mean
Khabarovsk (July 21- August 11)	1	Downtown	6.4	6.2	6.6	6
	2	Downtown	10.6	9.5	8.7	10
	3	Downtown	11.2	10.5	11.9	11
	4	Downtown	8.7	8.0	9.1	9
	5	the suburbs	6.0	5.5	5.6	6
	6	the suburbs	4.7	5.1	4.6	5
Yakutsk (July 25- August 8)	1	Downtown	5.6	6.2	6.0	6
	2	Downtown	3.8	3.9	3.8	4
	3	Downtown	5.5	5.9	6.1	6
	4	Downtown	10.2	10.5	10.0	10
	5	Environmental center	6.2	6.0	6.1	6
	6	the suburbs	1.5	1.5	1.4	2
	7	Radio relay station	0.6	0.7	0.6	< 1
	8	Forest	0.3	0.3	0.2	< 1
Norilsk (July 29- August 4)	1	Norilsk city	6.3	3.6	3.9	5
	2	Norilsk city	4.9	6.0	5.5	6
	3	Norilsk city	4.9	4.5	4.4	5
	4	Norilsk city	5.5	5.6	4.9	5
	5	Norilsk city	4.9	4.8	4.7	5
	6	Experimental center	6.3	5.7	5.2	6
	7	the suburbs	2.2	2.2	1.9	2
	8	Oganer	2.5	3.2	2.3	3
	9	Tarnax	3.1	3.3	2.5	3

Table 3. Atmospheric concentrations of NO<sub>2</sub> in Khabarovsk, Yakutsk and Norilsk in the summer of 1993



### 3.2 Summer concentrations of atmospheric $\text{NO}_2$ and $\text{SO}_2$ in urban areas of Siberia

Table 3 shows the measurement results for the atmospheric  $\text{NO}_2$  concentration in Khabarovsk, Yakutsk and Norilsk in the summer of 1993. We exposed a set of three diffusion samplers at every sampling site, and averaged the concentrations for the three samplers.

Figure 7 shows a map of the sampling sites in Khabarovsk. Atmospheric concentrations of  $\text{NO}_2$  ranged from 5 to 11 ppbv in Khabarovsk. The concentrations in the downtown area (site No. 1-4) were a little higher than those in the suburbs (No. 5 and 6).

The sampling sites in Yakutsk are shown in Figure 2. Sites No. 1 to 5 were in the downtown area and the measured concentrations ranged from 4 to 10 ppbv. The concentration at site No. 6 was 2 ppbv, which was obtained at a villa 10 km west from the center of Yakutsk city. Site No. 7 was the radio relay station 20 km west of Yakutsk city and site No. 8 was in the forest nearby. The concentrations at both sites were less than 1 ppbv. In Khabarovsk and Yakutsk,  $\text{NO}_2$  concentrations in urban areas were a little higher than those in rural areas due to emission of  $\text{NO}_x$  from vehicles. For comparison, we cite the concentrations in Sapporo. In July and August 1992 atmospheric concentrations of  $\text{NO}_2$  were, respectively, 28 and 25 ppbv in downtown Sapporo, and 10 and 9 ppbv in the suburbs of Sapporo. In Khabarovsk and Yakutsk, thus, the summer  $\text{NO}_2$  concentrations were much lower than those in Sapporo.

Figures 8 and 9 show maps of the Norilsk area and Norilsk city, respectively. The Norilsk area consists of four residential sections. The resident population in each of these sections

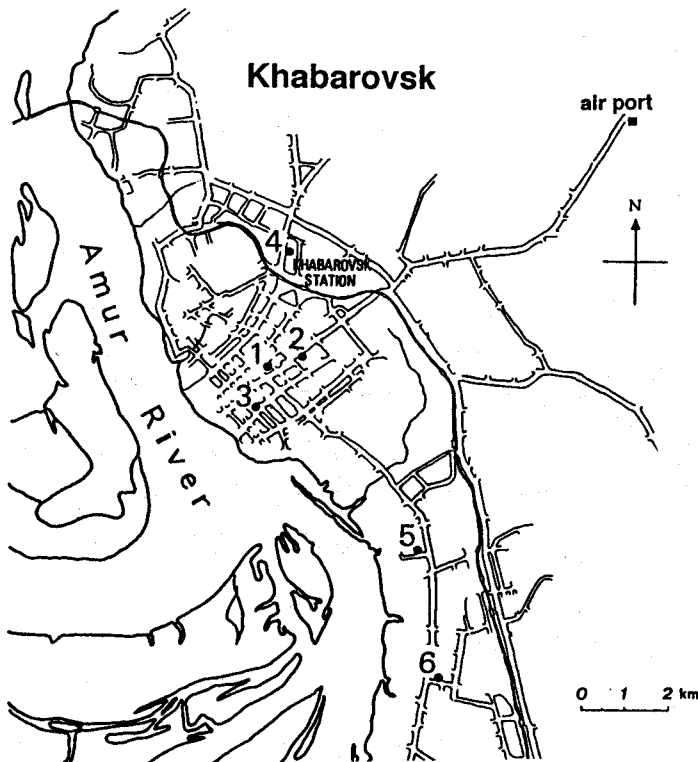


Figure 7. Map of Khabarovsk.

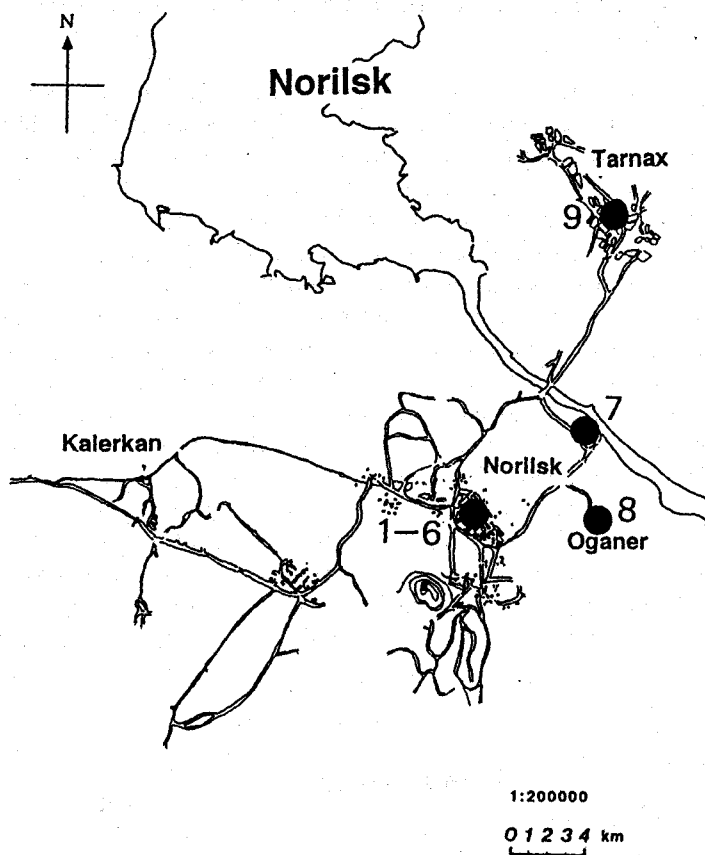


Figure 8. Map of the Norilsk area.

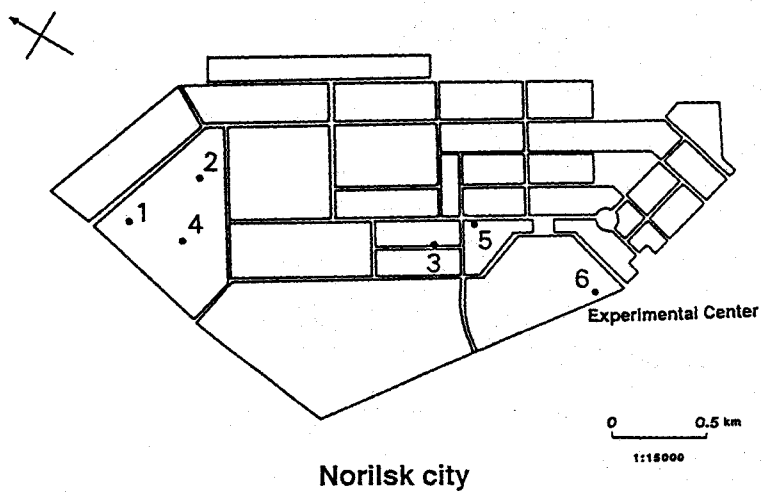


Figure 9. Map of Norilsk city.

is 180,000 in Norilsk city, 70,000 in Tarnax, 30,000 in Kaierkan, and 5,000 in Oganer. The total population in Norilsk area is 285,000. In Norilsk, copper and nickel mining, ore dressing and smelting are in operation. The smelting discharges large amounts of  $\text{SO}_2$  and particulate  $\text{SO}_4^{2-}$ .

Summer concentrations of  $\text{NO}_2$  in Norilsk shown in Table 3 ranged from 2 to 6 ppbv, which are similar to the concentrations in Khabarovsk and Yakutsk.

Table 4 shows atmospheric concentrations of  $\text{SO}_2$  in the summer in the Norilsk area. The concentrations ranged from 11 to 44 ppbv. Because the smelteries are located in the northern

Sampling site		$\text{SO}_2$ concentration (ppbv)			
No.					Mean
1	Norilsk city	45	44	42	44
2	Norilsk city	16	15	19	17
3	Norilsk city	22	17	18	19
4	Norilsk city	22	19	22	21
5	Norilsk city	12	10	10	11
6	Experimental center	21	18	21	20
7	the suburbs	27	28	24	26
8	Oganer	19	23	20	21
9	Tarnax	16	16	19	18

Table 4. Atmospheric concentrations of  $\text{SO}_2$  in Norilsk during July 29 – August 4, 1993

part of Norilsk city, the concentration at site No. 1 was the highest of all. Although, as shown in Figure 6, we cannot place high confidence in the concentration values because of the extrapolation regarding the correlation between the amounts of  $\text{SO}_2$  absorbed in the reagent-soaked paper and the mean atmospheric concentrations of  $\text{SO}_2$ , we may estimate that in the Norilsk area the  $\text{SO}_2$  concentrations were around the 20 ppbv level.

In Japan, the  $\text{SO}_2$  concentrations at monitoring stations in typical industrial cities in July and August 1992 were, respectively, 10 and 5 ppbv in Tokyo, 15 and 9 ppbv in Kawasaki, and 11 and 6 ppbv in Osaka. Thus, the  $\text{SO}_2$  concentrations in Norilsk were high due to poor emission control of  $\text{SO}_2$  and particulate  $\text{SO}_4^{2-}$  from the smelteries. It is an important issue to control the emissions of the pollutants in Norilsk.

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