Complex assessment of the conditions of the air basin over Norilsk industrial region. Part 2. Admixture balance in Norilsk region

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Composition of gaseous and aerosol constituents arriving at and leaving from the air basin of the Norilsk industrial region in cold and warm seasons has been analyzed. Main constituents enriching the urban admixture column are revealed. Scattering properties of the atmosphere are estimated. Strong seasonal variations in composition of air, both arriving at and leaving from the territory of the region, are recorded.

Introduction

Amount of pollutants emitted by industrial plants and motor vehicles is calculated quite exactly from fuel and material balance. However, besides the big plants, with proper emission control, there are many small industrial enterprises and non-organized emission sources sometimes contributing appreciably to urban atmospheric and environmental pollution. In addition, the species emitted to the atmosphere may be transformed to other, more toxic compounds in the course of chemical and photochemical reactions and condensation. coagulation, and sedimentation processes. For instance, ozone is emitted to air by none of the enterprises, and is formed in the atmosphere from precursor gases. In parallel with the ozone, the formation of formaldehydes and other aldehydes also takes place in the atmosphere, well as of the ultrafine aerosol including peroxyacetylnitrates, etc. Therefore, it is necessary to have an independent estimate of the balance of pollutants in the region of a city or territory.

Foreign researchers use for this purpose the method of construction of vertical transects of the atmosphere on upwind and downwind sides.^{1,2} Thus obtained data are then used in calculations. Our experience of work in different regions has shown that the vertical transects are not always enough. The presence of local circulation may deflect air particle trajectories from direction of the main flow; therefore, we generally applied the "square" scheme,³ in which these deflections can be taken into account.

Location of Norilsk or, more precisely, the orographic obstacles around it, impeded implementation of the "square" scheme in these experiments. Instead, we had to use the variant applied by foreign scientists: construction of vertical transects on up- and downwind sides of the city. The planes of the transects were perpendicular to the main flow in accordance with the data of meteorological station and pressure topography maps. The scheme of the experiment is presented in Fig. 1.

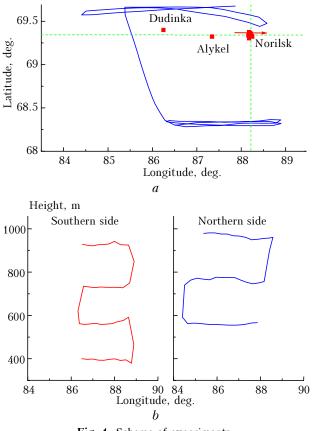


Fig. 1. Scheme of experiments.

On up- and down-wind sides of the city, flight legs were flown in a single vertical plane at heights 400, 600, 800, and 1000 m above sea level. Clearly,

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because of variations in relief, the flight altitude, though kept constant, nevertheless varied relative to the earth's surface.

Measurement results

We shall first consider upwind side of the city, using data for cold period (Figs. 2–6). These data must reflect the background concentrations of admixtures in air arriving at the city.

Figure 2 shows that, across the entire area of a vertical transect, the sulfurous anhydride concentrations are $30-40 \ \mu m/m^3$, typical for the background conditions of this region. Deviations are within standard measurement error. Therefore, no sulfurous anhydride arrives at the city from outside.

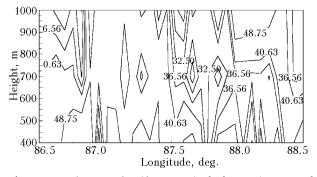


Fig. 2. Distribution of sulfurous anhydride on the upwind side, $\mu g/m^3$.

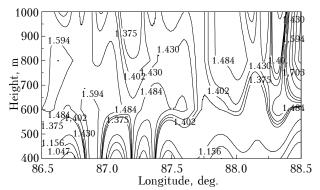


Fig. 3. Distribution of submicron fraction of aerosol $(d \ge 0.4 \ \mu\text{m})$ on the upwind side, cm⁻³.

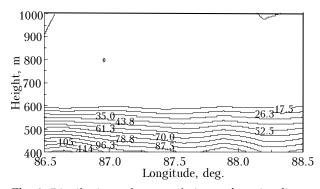


Fig. 4. Distribution of accumulation-mode microdisperse aerosol fraction on upwind side (d > 70 nm), cm⁻³.

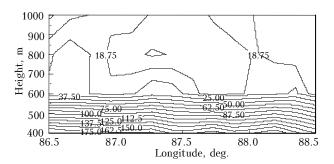


Fig. 5. Distribution of nucleation-mode microdisperse aerosol fraction (d = 3-70 nm) on the upwind side, cm⁻³.

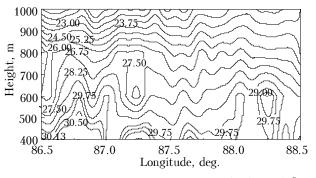


Fig. 6. Distribution of ozone on the upwind side, $\mu g/m^3$.

Number concentration of submicron aerosol fraction $(1-1.5 \text{ cm}^{-3})$ turned out to be close to the background value (see Fig. 3).

In analysis, we divide the microdisperse fraction into two (accumulation and nucleation) modes, which are usually observed in the atmosphere.^{4,5} The accumulation mode typically consists of ready particles formed from the gas-phase material; they participate only in the coagulation process and subsequently grow to submicron fraction.⁶ The nucleation mode constitutes the particles just (minutes to hours ago) formed from gas-phase material.

We shall first consider the distribution of accumulation-mode aerosol (see Fig. 4). As seen, in the subinversion layer, up to the height of 600 m, there is small amount $(17-100 \text{ cm}^{-3})$ of particles, unable to effectively coagulate because of small collision probability.⁶

The distribution of nucleation-mode aerosol suggests (Fig. 5) that in the arriving air, especially in the subinversion layer, there takes place particle generation from gas-phase substance, though not very intense. Here, the concentration is 25 to 175 cm⁻³. In the case of intense generation, it reaches 2000–10000 cm⁻³ (Ref. 13).

At last, we shall present data on ozone which, as was already mentioned above, is formed directly in the atmosphere (see Fig. 6). As seen, its distribution over transect plane is quite uniform and ranges from $30 \ \mu\text{g/m}^3$ in the subinversion layer, to which ozone-forming gases are lifted from the underlying surface, to $23 \ \mu\text{g/m}^3$ in the above-inversion layer.

Thus, sensing from upwind side of the city in winter period has shown that the city receives air in which admixture concentration is close to the background level.

We shall analyze, which compounds are blown off the city (Figs. 7-11).

From Fig. 7 it is seen that sulfurous anhydride concentration increased in the upper part of the transect, i.e., over stable layer. Most likely, this was due to the spread of admixture on the boundary of involvement in urban column. Whereas the background values at city entry were $30-40 \ \mu\text{g/m}^3$, here they increased to $160 \ \mu\text{g/m}^3$.

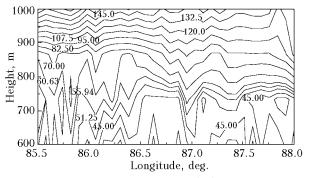


Fig. 7. Distribution of sulfurous anhydride concentration on the downwind side, $\mu g/m^3$.

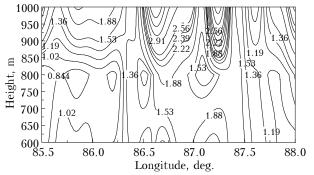


Fig. 8. Distribution of submicron-mode aerosol on the downwind side, cm^{-3} .

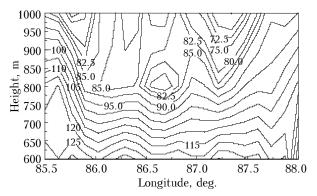


Fig. 9. Distribution of accumulation-mode aerosol on the downwind side, $\rm cm^{-3}.$

In the lower part of the transect, the submicronmode aerosol $(d = 0.4-2.0 \ \mu\text{m})$ also had almost background concentration (see Fig. 8). In the upper part, we can isolate two regions of enhanced concentrations. However, they do not correspond to plume-level values. Rather, these are particles spread off the urban column.

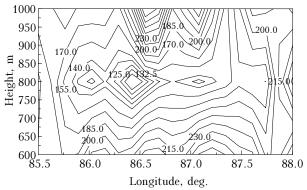


Fig. 10. Distribution of nucleation-mode aerosol on the downward side, $\rm cm^{-3}.$

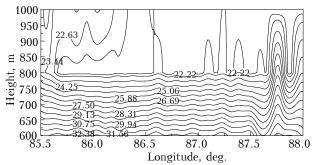


Fig. 11. Distribution of ozone on downward side, $\mu g/m^3$.

From comparison of Figs. 7 and 8, we see another one characteristic feature, i.e., the regions of increased concentrations of sulfurous anhydride and aerosol do not coincide, for the first time in our practice. Seemingly, the sources of these ingredients differ.

The behavior of microdisperse fraction is drastically different on the downwind side. Its generation is observed both in the upper and lower parts of the transect; whereas on the upwind side, it was mainly formed in the subinversion layer.

Turning to accumulation mode (see Fig. 9), we see that it remained almost unchanged in the lower part of the transect and became detectable in the upper part. Its concentration is still insufficient for intense coagulation processes.

The nucleation-mode aerosol (see Fig. 10) became detectable throughout in the entire altitude range, again confirming the hypothesis on the possibility of photochemical processes in the atmosphere of Norilsk. Also favoring this hypothesis is the comparison of concentrations in Figs. 9 and 10. The accumulation-mode concentrations are much less than nucleation-mode level, paving grounds to believe that the nucleation-mode particles are newer, formed less than an hour ago.^{5,6}

Ozone concentration somewhat increased in the lower part of transect and decreased in its upper part (see Fig. 11). This may evidence of two reverse processes in the atmosphere of Norilsk. The first process is that ozone is indicator of photochemical processes, so its increase means that they are present in the lower part.⁷ The second process essentially consists in that, being active oxidant, ozone starts interacting with aerosol, and destructs it stronger in the upper part of the transect where aerosol concentration is higher.^{8,9}

In addition to gaseous components and aerosol disperse composition, at each altitude we took samples for analysis of aerosol chemical composition. The analysis itself was performed at analytical laboratory of Tomsk State University, certified by Federal Bureau of Standards. Figure 12 presents mean relative aerosol composition ($d \ge 0.1 \mu$ m) on up- and downwind sides of Norilsk.

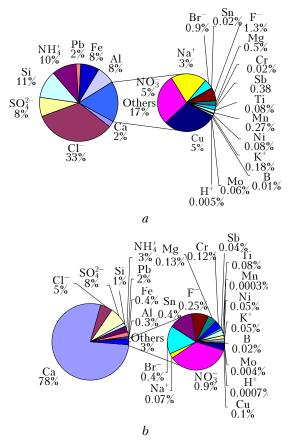


Fig. 12. Relative chemical composition of atmospheric aerosol on the upwind (*a*) and downwind (*b*) sides.

Data for each plot were obtained using a combination of instrumental and arithmetic averaging. The instrumental averaging was performed during air sampling along the entire flight track, at each altitude. The arithmetic mean was obtained through averaging over all altitudes.

From Fig. 12a it follows that the city received aerosol with chemical composition characteristic of background conditions.¹⁰

At the exit from the city (Fig. 12b) the pattern is substantially different. The dominating element in aerosol composition is now calcium whose contribution is over 75%. It should be stressed that, over 20-year period of atmospheric aerosol study, we encountered no such situations, neither in the background¹¹ nor in urban environment, $^{12-14}$ though in Ref. 14 we indicated that a certain single element dominates in each region. Possibly, coal used in Norilsk is enriched with limestone, explaining that high calcium content in coal-burning aerosol. As is evident from Fig. 12,*a*, calcium content at the city entry did not exceed a few percent.

Bezuglaya et al.¹⁵ reported on high nitrogen monoxide concentrations in the atmosphere of Norilsk. Our experiments did not confirm that. Judging from Fig. 12, nitrate-anion is present in the aerosol also in the background amounts.

Measurement data obtained in the vertical planes on up- and downwind sides of Norilsk were used to compile Table 1, which presents average concentrations and their differences.

Transport is calculated based on the speed of motion of air mass, determined using map AT-850, rather than wind. Transport per year is case specific because it is determined based on the data obtained during a single flight. It is given here only to assess the scale of the phenomenon. Concentrations with minus sign pertain to cases of deposition of these constituents on the territory of the city. Positive differences signify outflow of these constituents off the city.

From Table 1 it is seen that in measurement period, constituents, mainly transported off the city, were calcium and sulfurous anhydride, with the transport rates of 42.5 and 51.4 g \cdot s⁻¹, respectively.

It is also seen that constituents arriving at the territory were aluminium, iron, silicon, and ammonium, whose concentration was higher on upwind than downwind side. Usually, these elements compose coarse-mode particles,^{16,17} which have high sedimentation rate. It is quite probable, hence, that they could have deposited on the earth's surface.

From Table 1 it also follows that the ratio of the transferred mass of SO_4^{2-} ion and gaseous SO_2 is approximately 1:16. Seemingly, industrially emitted SO_2 has little time to condense or react immediately in the atmosphere of the city and is transported to the environment in the initial (i.e., gaseous) phase.

Approximate estimate of the annual transport for all measured parameters for Norilsk yields over 3 million tons. At the same time, it should be stressed that the vertical transects contain no urban plume aerosols.

On a day of a similar experiment in summer, there was northeasterly wind with the speed ranging from 3-4 m/s in the lower layers and to 5-7 m/s at the upper levels. Therefore, the vertical planes for sensing were constructed on northeastern (upwind) and southwestern (downwind) sides.

From Fig. 13 it is seen that the air arriving at the city contains SO_2 with the concentration 70–80 µg/m³. The distribution of the species over transect plane is quite uniform, with some increase in

Chemical		Concentration	m 41	The man and the /							
constituent	Upwind side	Downwind side	Difference	Transport, g·s	Transport, t/yr,						
ng/m ³											
Mo	6.9	7.6	0.7	0.84	26						
В	1.3	10.3	9.1	10.90	344						
\mathbf{K}^+	20.3	21.0	0.7	0.90	28						
Ni	8.8	23.3	14.5	1750	552						
Mn	30.2	32.1	1.9	2.34	74						
Ti	8.8	37.3	28.5	34.30	1082						
Sb	42.5	46.7	4.2	5.04	159						
Cr	2.4	55.6	53.2	63.91	2015						
Mg	52.1	56.9	4.8	5.83	184						
Sn	2.0	160.0	158.0	189.60	5979						
Br^-	97.5	191.7	94.2	113	3564						
$\mu g/m^3$											
F^-	0.143	0.165	0.022	26.1	823						
Na^+	0.381	0.389	0.008	9.3	293						
NO_3^-	0.553	0.567	0.015	17.4	549						
Cu	0.558	0.560	0.003	3.0	95						
Al	0.868	0.156	-0.711	-639.9	-20180						
Fe	0.878	0.175	-0.703	-632.7	-19953						
Pb	0.234	0.938	0.704	844.4	26629						
NH_4^+	1.185	1.159	-0.026	-23.4	-738						
Si	1.293	0.515	-0.778	-700.2	-22082						
SO_4^{2-}	0.910	3.520	2.610	3132.0	98771						
Cl	3.763	4.150	0.388	465.0	14664						
Ca	0.273	35.653	35.381	42457.0	1338924						
Aerosol			39.404	47398.4	1494754						
SO_{2} , $\mu g/m^3$	38.4	81.2	42.80	51360	1619689						
				Total	3114443						

 Table 1. Concentration of polluting species on up- and downwind sides of Norilsk and their transport across vertical planes

the upper part of transect. This background concentration is somewhat higher than the concentration observed in winter. Back trajectory analysis showed that the air mass has arrived from Arctic Ocean. In this case, the source of elevated sulfur dioxide concentration may have been seaderived dimethylsulfide which, as shown in Ref. 18, is subsequently converted in the atmosphere into sulfur dioxide. Precisely this mechanism was used by Tu et al.^{19,20} to explain the elevated background SO₂ concentration over Pacific Ocean, revealed in the course of airborne experiments.

Also favoring this mechanism are the data on distribution of microdisperse aerosol fraction presented in Fig. 14.

In winter, its generation on the background side took place only in the boundary layer. Above the boundary layer, the content of nanoparticles has decreased almost to zero. In summer, the concentration of microdisperse fraction increased with height.

American scientists studied formation of microdisperse fraction of aerosol in the atmosphere over oceans and have shown^{21–23} that it is formed from SO_2 and its concentration in the free atmosphere may exceed concentration of this aerosol in the boundary layer. Obviously, similar pattern is observed in Fig. 14.

In contrast to sulfur dioxide and microdisperse aerosol fraction, the background concentrations of ozone were found to be almost identical to those measured in winter; therefore, its vertical transects will not be presented here.

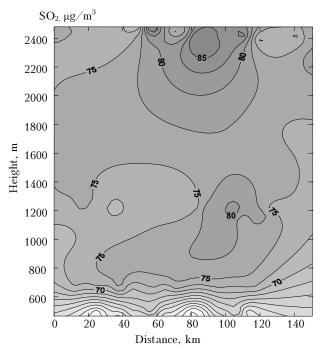


Fig. 13. Distribution of sulfurous anhydride on the upwind side of Norilsk in summer.

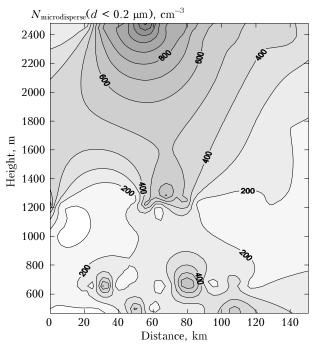


Fig. 14. Distribution of microdisperse aerosol fraction on the upwind side of Norilsk in summer period.

Let us now consider the vertical transect passing through the urban atmosphere.

Since this transect passed over a hill the lower height of sensing was 1200 m. To increase the areal coverage, the flight track was extended, and a few legs were flown at heights up to 4 km in a step of 500 m.

From Fig. 15 it is seen that in the main core of the propagating urban column, the concentration of sulfurous anhydride reaches 500 μ g/m³. At height 2500 m, there appears the secondary core with the concentration of 100 μ g/m³. Seemingly, some. overheated part of the plume penetrates through the stable stratification layer. If the urban column is to be defined by 50 μ g/m³ contour line, its width will be larger than 175 km. That is, the scales of horizontal dispersal are much larger than corresponding vertical scales. The concentration of the background SO₂ (beyond the borders of the urban column) substantially decreases as compared with its value on the upwind side. Possibly, appearance of additional condensation nuclei in the urban atmosphere accelerated the processes of condensation of sulfur oxide and the gas-to-aerosol conversion.

This conjecture is supported by data of Fig. 16, which shows distribution of microdisperse aerosol fraction. In the vertical transect, we see two cores with increased content of nanoparticles. Aloft, at the height of 4 500 m, their number concentration reaches 10 000 cm⁻³; therefore, it can be suggested that in this region the particles formed from the background SO₂ are collected. Below, near 1200-m altitude, there is urban column in which the nucleation processes are quite intense. The concentration of microdisperse particles in the column reaches 40 000 cm⁻³.

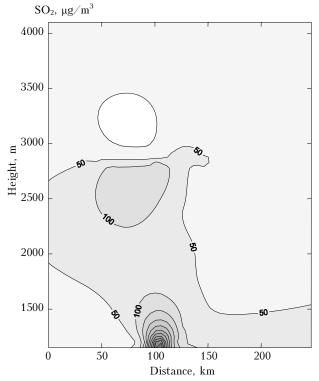


Fig. 15. Distribution of sulfur dioxide on the downwind side of Norilsk in summer period.

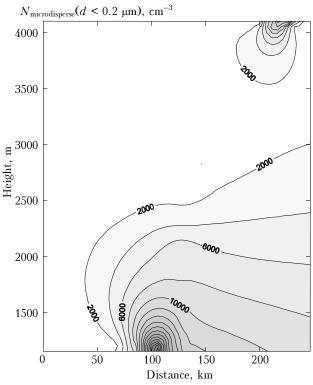


Fig. 16. Distribution of microdisperse aerosol fraction on the downwind side of Norilsk in summer.

That high particle content per unit volume must lead to intense coagulation processes and precipitate particle growth to submicron fraction.²⁴ Therefore,

this will manifest itself in distribution of submicron fraction.

From Fig. 17 it is seen that, when compared with sulfur dioxide and microdisperse fraction, the submicron fraction in the vertical transect occurs where the former two are absent. Such a behavior of submicron fraction suggests that it is formed on the periphery of zones of increased content of ozone and microdisperse fraction. In the plume, possibly, it rapidly exceeds 1- μ m size and is deposited onto the earth's surface.

$N(0.4 < d < 10 \ \mu m), \ cm^{-3}$

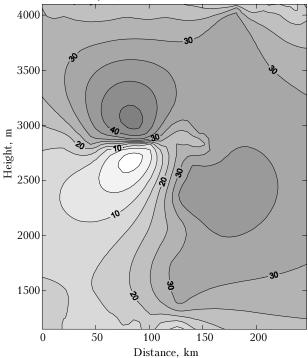


Fig. 17. Distribution of submicron aerosol fraction on the downwind side of Norilsk in summer period.

Being one of the most active species, ozone behaves like submicron aerosol. It justifiably decreases inside the plumes where the particle concentration is high, and is generated on their periphery.⁸ At the same time, the measurement data showed that on the downwind side the ozone concentration is almost twice as high as that on the upwind side. Hence, the plumes of emissions contain considerable amount of ozone-forming species.^{26,27}

As an intermediate conclusion, we note that industrial emissions of Norilsk are not only of considerable amount, but also they qualitatively modify the processes taking place in the atmosphere.

Let us analyze composition of aerosol, arriving in the atmosphere of Norilsk, after being emitted by enterprises of the city in summer.

If we compare the aerosol chemical composition on up- and downwind sides of the city (Fig. 18), we shall not see large differences. As in winter, content of calcium increases in aerosol composition. The arriving aerosol is close in composition to the maritime aerosol. $^{10}\,$

Comparison of chemical composition of aerosol leaving the city in summer and winter periods reveals substantially different situations (Fig. 19), unexplainable just by seasonal factor. Possible extra cause may be technological change at the plants of Norilsk. At least, the authors only know of reconstruction at copper factory of the plant in summer of year 2004.

Identifications of non-natural changes in the aerosol composition frequently use the method of estimate of enrichment of components with respect to reference species (predominately of natural origin in aerosol composition) compared with this same characteristic of the earth crust or salt residuals in sea water.^{28–30} However, in our situation it is difficult to choose a reference constituent, usable for both seasons and for all constituents and introducing no distortion to the estimate of their origin; therefore, we consider simplified enrichment estimate based on interrelation of constituents in aerosol composition on the upwind and downwind sides on days of measurements inside the "squares" in each season (November 8, 2002 and August 10, 2004).

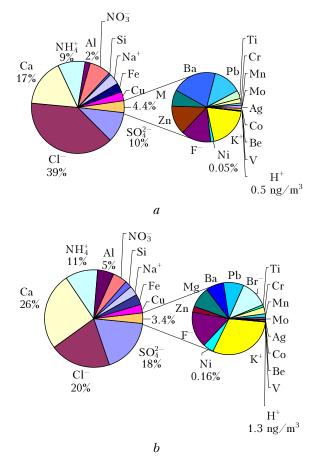


Fig. 18. Aerosol composition in Norilsk in summer: the upwind side (*a*), downwind side (*b*).

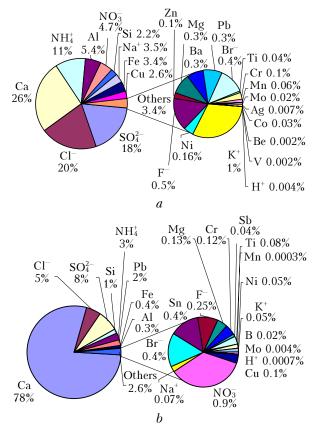


Fig. 19. Aerosol composition on the downwind side of Norilsk: summer (a), winter (b).

From Fig. 20 it follows that, in winter, when the underlying surface over large distances away from the city is covered with snow, the terrigenous elements Si, Al, Fe are diluted. Considerable enrichment is observed for Ca, Cr, Sn, Ba, and Ti in cold period. For the sum of all species, sum of all elements, and sulfate anion, the enrichment factor does not change from one season to another. In summer, all terrigenous elements and ions are enriched, as are ions and elements found in the composition of sea water. At the same time, it is still unclear why fluorine and titanium ions are not enriched in the summer season. Thus, in summer season, a marked contribution to aerosol composition comes from the underlying surface.

For summer season, the mean concentrations in the vertical planes on the upwind and downwind sides of Norilsk, their differences, and transport calculations are presented in Table 2. From Table 2 it is seen that, in the measurement period, calcium, chlorine, sulfates, and sulfurous anhydride mainly left the city. Quite noticeable became the inflow of aluminum, copper, iron, sodium, ammonium, nitrates, and silicon.

As compared with the winter period, there is now a considerable (orders of magnitude) increase of outflow of potassium, manganese, magnesium, chlorine, copper, sodium, ammonium, and nitrates from the city. At the same time outflow of chromium, bromine, lead, calcium, and total aerosol decreased.

Thus, in going from winter to summer, the chemical aerosol composition qualitatively changed, but the total outflow remained almost the same. Moreover, because we know the fuel balance of industrial plants and housing and municipal economy of the city, we can suggest several hypotheses. First, it is of great importance the above-mentioned change of technology or waste treatment at the plants. Second, the underlying surface is qualitatively different in the periods of experiments. In winter, the land is covered with snow, and nearby ocean with ice, both serving a kind of screen for aerosol replenishment. The situation is reverse in summer,

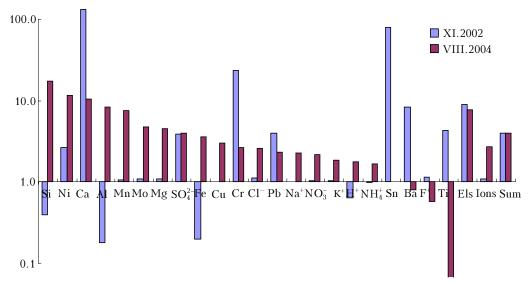


Fig. 20. Enrichment of ions, elements, and their sums (Ions, Els, Sum) in the atmosphere of Norilsk.

Element/ion	Concentration			Transport	Transport rate,	Transport rate,	summer				
	TT · 1 · 1	D · 1 · 1	D:0	rate,	$t \cdot yr^{-1}$	$t \cdot yr^{-1}$	$K \frac{\text{summer}}{\text{winter}}$				
	Upwind side	Downwind side	Difference	$g \cdot s^{-1}$	August 2004	November 2002	winter				
ng/m ³											
Mo	1.1	5.4	4.3	6.0	190	26	7.3				
K^+	62.7	115.1	52.4	73.4	2313	28	82.6				
Ni	5.7	65.2	59.5	83.3	2627	552	4.8				
Mn	5.1	38.5	33.4	46.8	1475	74	19.9				
Ti	21.9	0.5	-21.4	-30.0	-945	1082	-0.9				
Cr	14.4	38.3	23.9	33.5	1055	2015	0.5				
Mg	39.6	177.1	137.5	192.5	6071	184	33.0				
Br^{-}	<	0.2	0.2	0.3	9	3564	0.003				
Со	5.5	8.8	3.3	4.6	146	_	_				
F^{-}	46.6	27.0	-19.6	-27.4	-865	823	-1.1				
V	0.9	0.1	-0.8	-1.1	-35	_	_				
Ba	131.6	104.3	-27.3	-38.2	-1025	_	_				
Zn	7.9	31.6	23.7	33.2	1046	_	_				
Be	0.7	0.6	-0.1	-0.1	-4	_	-				
Pb	8.4	193.5	185.1	259.1	8172	26629	0.3				
Ag	2.6	8.0	5.4	7.6	238	-	-				
			µg∕⊥	m ³							
Al	0.24	1.94	1.70	2380	75056	-20180	-3.7				
Ca	0.96	9.89	8.93	12502	394263	1338924	0.3				
Cl ⁻	3.97	10.29	6.32	8848	279030	14664	19.0				
Cu	0.38	1.16	0.78	1092	34437	95	362.5				
Fe	0.54	1.94	1.40	1960	61811	-19953	-3.1				
Na^+	0.38	0.86	0.48	672	21192	293	72.3				
NH_4^+	0.74	1.23	0.49	686	21633	-738	-29.3				
NO_3^-	0.73	1.59	0.86	1204	37969	549	69.2				
Si	0.08	1.47	1.39	1946	61369	-22082	-2.8				
SO_4^{2-}	1.41	5.67	4.26	5964	188081	98771	1.9				
Aerosol			27.07	37897	1195129	1494754	0.8				
SO_2	75.3	121.5	46.2	64680	2039748	1619689	1.3				
Total					3234877	3114443	1.04				

Table 2. Balance of admixtures in the region of Norilsk in August 2004 and November 2002

Note. Concentration differences marked with minus sign correspond to deposition of these constituents on the territory of the city. Positive differences indicate outflow of these constituents off the city.

when both wind erosion and evaporation of aerosolforming species may take place. Third, in general emission flux of pollutants, there is a marked contribution of housing and municipal economy and, possibly, private sector, both using fuels with different chemical composition. Then, weaker activity in this area during summer period may lead to such cardinal changes.

An approximate estimate of annual transport for all measured species for Norilsk exceeds 3 million tons, much more than officially reported.²⁵

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