Complex assessment of the conditions of the air basin over Norilsk industrial region. Part I. Dimensions and dynamics of the impurity column

M.Yu. Arshinov, B.D. Belan, D.K. Davydov, G.A. Ivlev, V.A. Pirogov, D.V. Simonenkov, G.N. Tolmachev, and A.V. Fofonov

Institute of Atmospheric Optics, Siberian Branch of the Russian Academy of Sciences, Tomsk

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The measurements of air composition in the Norilsk industrial region have been carried out twice, in November 2002 and in August 2004. The measurements have been conducted using AN-30 "Optik-E" instrumented aircraft and a small ground-based complex mounted in an automobile van. In this paper we describe some results of the investigation of impurity dispersal in the city column. The city impurity column was found to retain its completeness both in cold and warm periods. In this case, its primary scale remains constant. This is determined probably by the city dimensions and orography. The concentration of SO_2, the basic compound of the city emissions, in summer reaches 1300 μg/m^3 that more than twice exceeds the value recorded in winter. The temperature superheating of the column in summer (0.8–1.0°C) is weaker than in winter. The observations of column spread in winter and summer along the horizontal has shown that at a distance of 50 km the column width increases twice during summer. This shows that in summer the dispersive capability of the atmosphere in a given region are much higher than in winter.

Introduction

At present there is no a continent on the planet, including the Antarctic, where the evidences of human activities were not observed. It seems quite reasonable that the anthropogenic influence in each region manifests itself in different ways that can be revealed in observing the state of the environment on the regional scale. One of such places is Norilsk industrial region, the enterprises of which make an up to 78% contribution to the emissions of Krasnoyarsk Territory or about 8% of gross industrial emission of the Russian Federation.[1]

Norilsk is a large industrial center in the Arctic. Because of great volumes of emissions observed by the end of 1980's the city atmosphere was characterized by the following parameters.[2] One-time concentrations of sulfur dioxide 40 times exceeded maximum permissible concentration (MPC) and the concentrations of nitric oxide exceeded it 36 MPC. The annual mean values of the same compounds were 3–4 MPC. Single MPC of chloride, phenol, hydrogen sulfide, carbon monoxide were exceeded by a factor of 4 to 6 and that of nitrogen peroxide – by a factor of 15.

Basic sources of air pollution are the enterprises of nonferrous metallurgy. The emissions of sulfur dioxide of these enterprises were the highest in the former Soviet Union and reached 10% of the total volume. Sulfur dioxide constituted 96%, the dust, carbon dioxide and nitrogen peroxides constituted 1–1.5%.

According to the data of the territorial committee on the environmental protection,[1] the Norilsk mining and metallurgical enterprises showed a decrease of the level of cleaning and utilization of sulfur dioxide from 20% in 1989 to 3.5% in 1994 that resulted in an additional amount of emissions (thousands of tons) to the atmosphere[3–6]:

Year	Dust	SO_2	NO_2	CO
1996	24.1	2014.4	13.7	53.1
1997	26.0	2015.0	12.8	32.0
1998	20.1	2065.0	12.8	25.5
1999	20.4	2104.1	12.3	23.6

Because there are no economically justified technologies available[7] for utilization of sulfuric anhydride one can hardly expect improvement of air quality in the city in the nearest future.

The situation is complicated by the unfortunate disposition of enterprises relative to housing areas in the city.[2] The enterprises are located to the north and south of the city. The repetition of north and northwestern wind directions is 19%, south and southeast – 28%. Thus, the city is alternately under the effect of emissions of either one or another enterprise of nonferrous metallurgy.

It is assumed that Norilsk region is characterized by a comparatively good dispersive power of the atmosphere.[2] Although, in the paper by E.Yu. Bezuglaya[8] this region is related to poorly self cleaning ones. It is evident that here the confusion of concepts took place. The atmospheric dispersive properties are really poor. However, the intensive winds, observed on the territory,[9] are favorable for further transport of impurities. This is confirmed by data on the state of vegetation and soil on the vast extensions.

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In the annual variation, the most unfavorable meteorological conditions, favorable for accumulation of impurities in the atmosphere, are observed in winter,[2] when the recurrence of air stagnations reaches 9–19%, and of fogs – 8–17%. At high sulfur dioxide content, its precipitation on the fog drops takes place and sulfuric acid is formed. The formation of smog situations is possible of the famous London smog type.

The measurements of air pollution level are performed in Norilsk at 4 ground-based stations located in the city.[3–6] Of course, these measurement data cannot present the total information on the state of air basin of industrial center and the impact of industrial emissions on the environment. However, many evidences exist of that these emissions are transported to distances of thousands of kilometers and can reach Alaska and Canada. It is unclear how in this case the impurities are dispersed and transformed. All the above-mentioned served a basis for making a complex evaluation of the air basin state in the city of Norilsk. Investigations of regularities of the formation and transformation of the air pollution field in big industrial centers have been progressing rapidly during the past 10–15 years. These results radically alter foregone concepts in this research area and are not fully described yet in the currently available textbooks and monographs. Therefore, let us briefly consider the present-day understanding of regularities of the formation and transformation of the fields of air pollution in big industrial centers.

First, we shall remind the essence of traditional ideas. Historically circumstances turned out so that any big industrial center, while developing, had been built during several stages. In any city one can recognize the historical part, along the perimeter of which, in due time, industrial objects were built. Thereafter, as the city enlarged, residential areas were beyond the limits of historical part and the manufactures occurred inside the city. In some cities, such process was realized repeatedly. For example, in Tomsk the electric power plant GRES-2 was built in an outskirts, and now GRES-2 is located in the center of the city. More complicated variant is observed in Novosibirsk.

Over a long time, it was assumed that during ordinary days with moderate wind, the wind blew freely through the city and the enhanced background of air pollution was observed only near industrial enterprises or according to the plume of the impurity transport. To plan the location of future plants, the climatic wind rose was used, that is, new plants and factories were located outside the city, in its lee side. It was assumed that in this case the impact of industrial emissions on the city population would be least harmful. And only when two conditions: low inversion and calm coincide, the industrial emissions remain reside long over the city territory. Present-day theoretical and experimental data on the aeration characteristics of big cities indicate that this is a dangerous illusion.

If we observe any city from an aircraft, even in the presence of strong and moderate wind, one can easily see that the industrial emissions from elevated sources (stacks) do move along the wind and are swept away beyond the limits of the city. At the same time, the city is blanketed by a haze that is indicative of the accumulation, first, of aerosol impurities within the city limits. Consequently, no direct removal of impurities out of the city limits takes place, but there is a certain mechanism favorable for aerosol impurity accumulation.

In this case, if we observe the city atmosphere during several days without precipitation, for example, under conditions of anticyclone, we can see that the city haze, during this period, modifies its color from whitish, to dirty grey. What does this evidence of? This points to the fact that the industrial emissions occurring in the city are not totally removed from the city but, in addition, undergo transformations in the course of chemical, photochemical, condensation and coagulation processes. As measurement data show, in the course of such processes the materials are formed, which are more toxic and those are absent in the direct emissions. The airborne sensing data of the city atmosphere have shown that the impurity accumulation is observed not only in the atmospheric boundary layer, but in the upper layers, encompassing in some periods the entire atmospheric boundary layer.

From the data of numerous researches performed under the city conditions, it follows that the most effective mechanism of cleaning air of impurities are frontal divisions and rains that accompany them. In the front rear, as a rule, the pollution concentration decreases almost by one order of magnitude and the balance formed between the volume of emissions, transfer, transformation, and sink of different pollutants is disturbed. This process enables one to assess the rate of reconstruction of this balance.

Thus it is clear that, based on the traditional concepts on the aeration of industrial center, it is impossible to explain the presence of pollution cap over the center.

Congestion of industrial objects on a limited territory, which is a modern city, results in that at this point a great number of pollutants is emitted to the environment, which were not observed under natural conditions. These are chemical materials in different states and of different nature, additional heat energy, electromagnetic radiation, and so on. Because of this the city becomes, as known, an heat island. Besides, the city, as a rule, is constructed at a high place on a bank of a large reservoir, therefore orography provides the contrast of temperatures between dry land and water surface.

As a result, summed actions of these factors, in the city environs a local circulation occurs. The peculiarity of it is that in the city shade the airflows occur, which are opposite to the direction of the main airflow observed at the windward side. This circulation “blocks” the industrial emissions in the atmospheric boundary layer. Over this layer the haze with admixtures occurs consisting of gaseous and aerosol substances, which received the name the pollution cap. One more typical peculiarity of circulation is that it resides over the city not only at gentle wind but at moderate wind as well, disappearing for a short time at passage of atmospheric fronts. After the passage of the fronts, the circulation is rapidly reconstructed.

Methods and models developed by V.V. Penenko[10–12] and then the calculations, made for specific regions, have shown that the city is not only the heat island but also pollution island because of peculiarities of local air circulation appeared in the city outskirts. The data presented by V.V. Penenko not only change considerably the modern understanding of city aeration, but also allow one to explain a series experimentally observed facts that cannot be explained based on the traditional concepts. This, in particular, refers to rapid reconstruction (1–2 days) of photochemical equilibrium in the air of industrial center after the passage across the city of atmospheric front, clearing the city from pollutants.

The local circulation area depends on the scale of the city, the number and power of industrial centers. Industrial emissions are accumulated in this area, but not infinitely. Because the air temperature inside the column is higher, the air flux begins to ascend. Based on data from Ref. 13 first the column with impurities takes a vertical shape, and then under the action of main flux begins to tilt. It takes the horizontal shape at a significant distance from the city and travels close to the upper boundary of the atmospheric boundary layer. The height of this layer depends on season. This layer is separated from the free atmosphere by a impeding layer.[14]

The experience accumulated at IAO SB RAS on ecological sensing of air basin of a series of cities[15] has made it possible, on the one hand, to check theoretical findings, and on the other hand, to assess the actual state of air basin. For this purpose an AN-30 "Optik-E" instrumented aircraft is used. This aircraft-laboratory is fully described in the literature,[16] and the original methodology is given in detail in Ref. 17.

The measurements of air composition in Norilsk industrial region have been carried out twice, in November 2002 and in August 2004. For measurements the AN-30 "Optik-E" aircraft-laboratory and a small ground-based complex were used. The complex was mounted in an automobile van and moved on the territory surveyed.

Previously we described some results of atmospheric sensing in Norilsk industrial region.[18] In the present series of papers a complete characteristics of the air basin state in the region is described.

Scheme of the research flights

It was noted in the introduction that near the city a particular air circulation occurs, which "blocks" the emitted pollutants in its territory. Because of relative overheating, the polluted urban air first ascends as a vertical column, and then it moves in the horizontal plane along the key airflow as a whole plume. According to calculations,[10–12] depending on the city area, the power and number of enterprises, the scale of such a column can vary from several hundreds of meters up to tens kilometers.

To determine the existence of such a column in Norilsk industrial region, the flights were carried out using the "cross" scheme at altitudes of 600 and 800 m above sea level during cold and warm seasons. Taking into account the complex orography of Norilsk, the height of a relative terrain was variable and at some places the height did not exceed 100 m. The "cross" beam directions were chosen before the flight using the data of aviation meteorological station: perpendicular and parallel to the wind direction. The flight diagram is shown in Fig. 1.

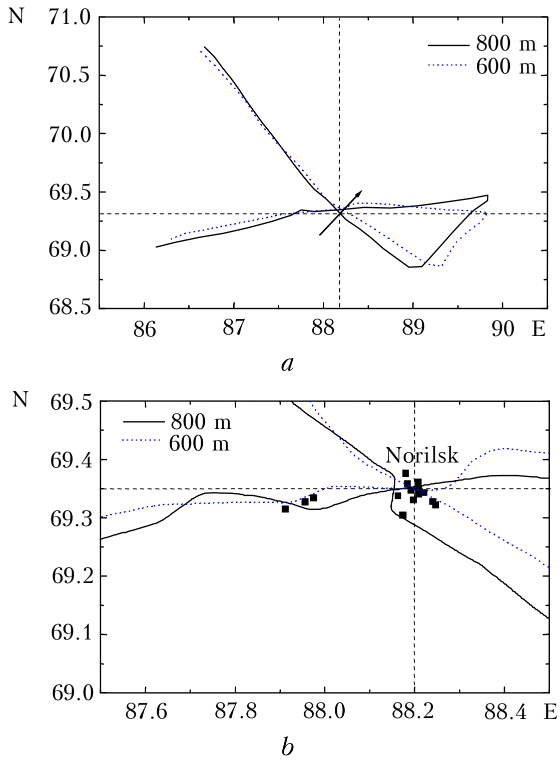

Fig. 1. Scheme of the experiment: (*a*) general, (*b*) in the environs.

Because the air inside the column is warmer than in the surrounding space, the condensation process occur in the column less intensely. Therefore, the column, as a rule, contains an increased number of particles of the ultrafine aerosol fraction. Coarse-fraction aerosol particles precipitate rapidly from the plume. As a result, the column cannot be seen visually.

Measurement results

The measurement data for winter are shown in Fig. 2. Figure 2 shows that, depending on the parameter being studied, the size of the city column can be estimated from 20 to 40 km.

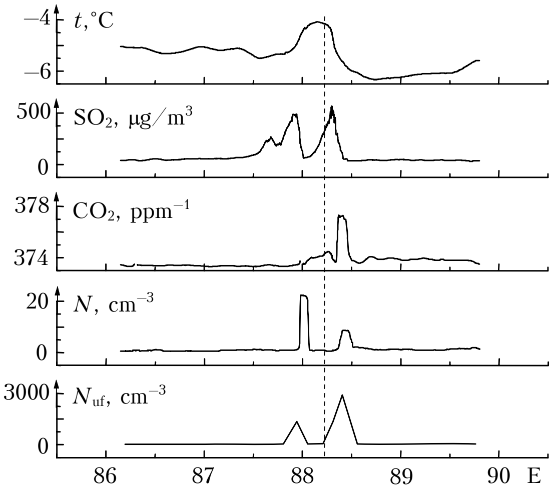

Fig. 2. Distribution of air temperature, concentration of sulfurous anhydride, carbon dioxide, number density of aerosol ($d \geq 0.4$ μm), and ultrafine aerosol ($d \leq 0.2$ μm) at 600 m height on November 5, 2002 in the west–eastern direction.

Such a spread in data can be understood because in the plume the transition processes take place. We noticed earlier that the air inside the column is warmer than the surrounding air by more than 1°C. At the same time, as the plume moves far from the city, this difference increases up to 2°C and even 3°C. If the background SO_2 concentration was at this height 40–50 μg/m^3, then at intersecting the column the SO_2 concentration increased up to 500–550 μg/m^3, i.e., by a factor of 10. In the distribution of sulfurous anhydride 3 peaks can be identified. This is, evidently, the reflection of industrial emissions of three main enterprises of Norilsk. It is obvious that in the initial part of the urban plume the plumes of industrial emissions have no time to mix. Strange as it may seem, but in the plumes no detectable increase of CO_2 concentration is observed.

An aerosol component both in submicron and ultrafine fractions manifested itself by two maxima. The submicron fraction gave the concentration increase from 2–3 cm^{-3} to 20–25 cm^{-3}, i.e., by a factor of 10, as SO_2. The ultrafine fraction, showing the transition of gas to particle, indicates that in the column quite intense heterogeneous condensation processes proceed. The concentration of aerosol particles' embryos in the plume is higher by a factor of 50–100 than outside the column. These particles cannot be recognized visually so that this does not contradict the above-mentioned facts in the introduction to this section. Figure 3 shows the disperse composition of aerosol submicron fraction in two peaks of city column detected at 600 m height and outside it. These data point to the particle size, which are rapidly removed from the atmosphere[19] and in which usually heavy metals are contained.[20] This figure shows that the particle concentration in the plumes is by 1 to 2 orders of magnitude higher than outside the plums.

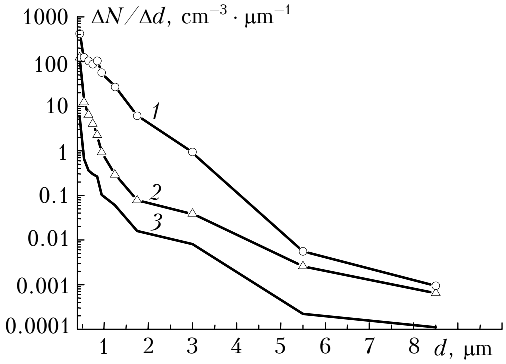

Fig. 3. Disperse composition of aerosol on November 5, 2002 in the city column (*1*, *2*) and outside the column (*3*).

Thus, using the data of winter measurements we notice that according to the present-day concepts on parameters of city plumes, it can be observed in Norilsk region too. Typical dimensions of the plume for the period of the experiment are 40 km. The column of impurities has inhomogeneous structure. In the column 2 or 3 high-power plumes can be observed, which were emitted from basic industrial enterprises of Norilsk.

Our subsequent experiments in 15 cities of the Siberian region have revealed that during winter period such columns can be revealed even over small populated areas.[21] In large populated areas (Novosibirsk, Krasnoyarsk, Tomsk) the city column can be observed even during warm season. Therefore, taking into account the power of Norilsk industry, it was advantageous to reveal the city column during warm season. This problem was solved using the first flight of the aircraft-laboratory in August 2004. The flight was also performed using the "cross" scheme at heights of 600 and 800 m above sea level.

The weather during the day of flight was determined by a specific cyclone, formed in the saddle between the anticyclone crest and the old cyclone. The cloudiness was variable. The wind in the atmospheric boundary layer was northwestern. In the free atmosphere the wind direction varied and became western. The wind speed in lower layers was 6–7 m/s, in the upper layers the wind speed was decreased to 3–4 m/s. No intense turbulence was observed though in some sectors of the flight the aircraft slightly rocked.

First we consider the vertical profiles perpendicular to the wind direction. They begin outside the city, cross the forest fire plume, and then the city column, including the plume of Norilsk Mining and Metallurgical Company (NMMC).

Traditionally the city column is identified by the temperature field. From Fig. 4 it follows that the forest fire and city emissions affect the temperature field. This value is 0.8–1.0°C. The layer from 700 to 900 m is a transition between the atmospheric boundary layer and free atmosphere. Figure 4 shows that there exist perturbations in the free atmosphere.

Because the city column during the day of the experiment was located close to the mountains surrounding the city, it was impossible to intersect the column fully, and the aircraft should turn the other way. If we consider it to be symmetric, and this is admissible, following data in Fig. 4, then the horizontal size of the column is 40 km, i.e., close to that obtained in the winter experiment.

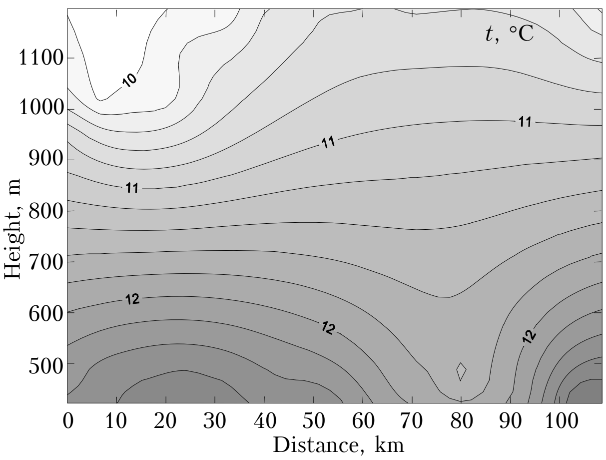

Fig. 4. Distribution of air temperature (°C) along the vertical plane passing through Norilsk, perpendicular to the wind direction.

It was revealed in the previous experiment, that the key elements of the city industrial emissions are aerosol and sulfurous anhydride. Data on their distribution are given in the next figures.

Figure 5*a* shows that the forest fire practically did not contribute to SO_2 field over the city. The main source is emission from NMMC plants. In this case, the concentration of sulfurous anhydride in the column reaches 900 μg/m^3 that exceeds more than twice the values detected in the winter experiment.

The winter data show that SO_2 emissions are accompanied by intense gas-to-particle conversions. As a result the ultrafine aerosol fraction is formed ($d = 3...200$ nm). Figure 5*b* demonstrates that such processes take place in warm period. And their intensity is much higher than in cold period.

If in winter the concentration of ultrafine fraction was 200–400 cm^{-3} (Ref. 18), in summer it was 1100–1600 cm^{-3}. Figure 5*b* shows that the nucleation processes in the forest fire increased twice compared to that in the industrial plume. It is probable that here the effect of organic matter is observed, which is much larger in the forest fire plume, and it is shown in Ref. 22 that in the laboratory experiment organic gases greatly enhance the nucleation processes.

Submicron fraction (particle diameter of 0.1 to 1.0 μm), the most long-lived component of aerosol,[23] although shows the presence of plumes, but not to such an obvious degree as the ultrafine fraction (Fig. 5*c*). One should keep in mind that in order that ultrafine fraction increased to submicron one, the period of 5–6 days is necessary.[24]

Figure 5*c* shows that as compared with the background, the concentration of submicron fraction grows in the plumes, both of forest fire and in the plumes of industrial emissions by a factor of 2 to 3. In the left part of Fig. 5*c* special attention must be given to high values of the submicron fraction concentration over areas, in which there are no sources of impurities. The checking indicated that this is not the experimental error. This situation cannot be explained so far.

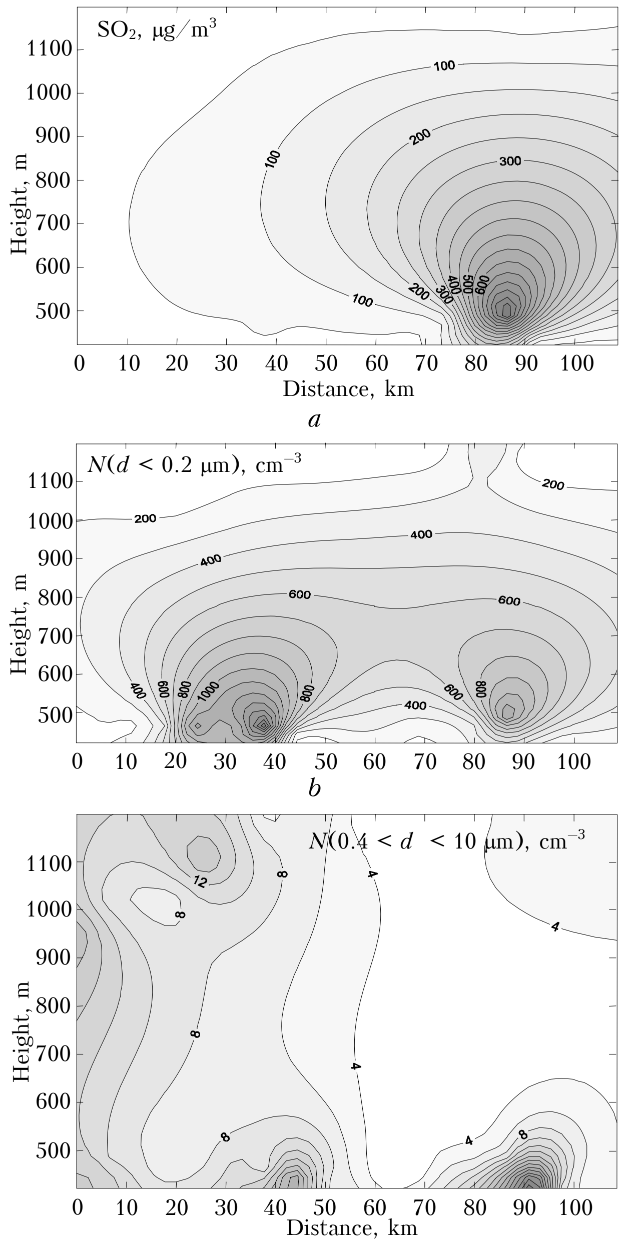

Fig. 5. Distribution of sulfurous anhydride (*a*), ultrafine (*b*) and submicron (*c*) aerosol fractions over the vertical plane passing through Norilsk, perpendicular to the wind direction.

The second part of this flight was devoted to sensing of the impurity column leaving the city. We have managed to intersect the trace of the city plume at a distance of 50 km from the city. In Fig. 6 the sections are presented obtained in the horizontal plane constructed based on data obtained at 1200 m height.

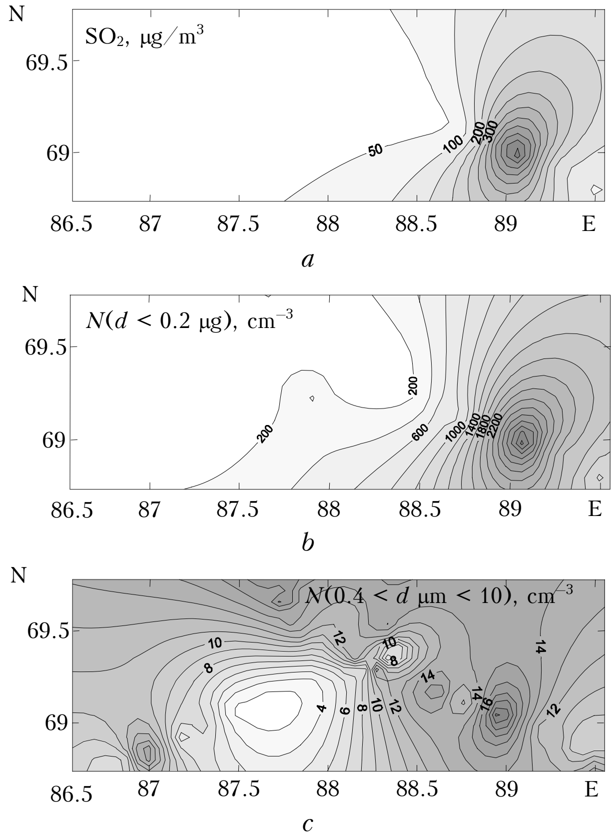

Fig. 6. Horizontal section of the sulfurous anhydride distribution (*a*), ultrafine (*b*) and submicron (*c*) aerosol fraction over the horizontal flight leg at 1200 m height.

Figure 6*a* shows that the concentration of sulfurous anhydride, as the column moves and expands, decreased by almost 2 times and is equal to 600 μg/m^3. The column itself is extended in the plane up to 80 km. This is the first evidence (during both periods of measurements) that in the Norilsk area the dispersive atmospheric properties can be high, at least, in the horizontal plane. Because of the orography of the locality the vertical profile along the route was not constructed.

From Fig. 6*b* an important conclusion can be drawn that the decrease of SO_2 concentration occurred not only due to the dispersal but also due to the gas-to-particle conversions. It is evident that the number density of nucleation mode increased up to 3600 cm^{-3} in the plume. In this case, we can observe the sink of sulfurous anhydride from the atmosphere precisely through an aerosol channel because outside the plume the particle concentration of this fraction does not exceed 200 particles per cubic centimeter.

At the same time, low concentrations of submicron fraction (Fig. 6*c*) evidence that the deposition of sulfates from the plume has not yet started. It should be noted that in winter this process began at a distance of 60 km and lasts till the distance of 100 km is reached. If this supposition is correct, in summer the area of the action of industrial emissions on the environment is expanded considerably. However, for checking this proposition it is necessary to make a separate experiment using special methods.

Conclusion

The column of industrial impurities maintains its condition of being intact in winter and summer. In this case, its original scale remains stable. Evidently, this is determined by the size of the city and by its orography. The concentration of basic emitted compound, SO_2, in summer reaches 900 μg/m^3 that more than twice exceeds the value detected in winter. The temperature overheating of the column in summer (0.8–1.0°C) is less than in winter.

The observation of the column transport in winter and summer along the horizontal path has revealed that in summer the column width increases twice at a distance of 50 km. The reason is that in summer the dispersive properties of the atmosphere in this region are much higher than in winter.

Acknowledgments

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