

RESULTS OF CLIMATIC–ECOLOGICAL MONITORING AT TOR STATION. III. ATMOSPHERIC AEROSOL

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The average annual and daily variations of aerosol have been analyzed based on the results of its 24-hour monitoring. Comparison with the data obtained for other regions have been performed as well as with aerosol total content in the vertical air column. Inhomogeneities of aerosol fields have been estimated.

There are some reasons for investigating the aerosol dynamics in the atmosphere. First, suspended particles, scattering and absorbing solar radiation incident on the Earth's surface, participate in the formation of the radiation balance and, hence, the climate of the planet. Second, aerosol particles, being active condensation nuclei, aid in formation of cloudiness that also affects the dynamics of the climate. Third, aerosol is one of the forms of air pollution of the atmosphere, which determines the ecological conditions of one or another region. Depending on the mechanism of origin of particles (terrestrial, *in situ*, or anthropogenic) and on their relative contribution in each specific region, its "aerosol weather" is created,¹ determining climatic and ecological regime of a given locality. Therefore, the study of spatiotemporal variability of atmospheric aerosol and its characteristics must be indispensable for long-term geophysical monitoring. This was embodied in the Program of Climatic–Ecological Monitoring of Siberia² (CEMS). This paper analyzes the aerosol dynamics in the atmospheric surface layer in the vicinity of Tomsk based on the data of measurements at the TOR station, operating in 24-hour and all-year-round regimes.³

As in the preceding papers of this series, we start our analysis with the annual variations of aerosol number density.

Figure 1 shows the annual variations of the number density of aerosol ($d > 0.4 \mu\text{m}$), obtained at the TOR station in 1993, and the annual variations of the integral aerosol content in a vertical air column of 1 cm^2 cross-section from the data of airborne sensing over Western Siberia averaged over a period of many years.⁴ The above variations are seen to differ significantly. In the surface layer, in addition to two secondary maxima in July and November, the maximum of the number density is observed in cold season and the minimum – in warm one. Such annual variations depend upon the fact that for constant generation of aerosol its number density in the surface layer will be determined by the height of the mixing layer, that is, by the air volume through which particles are distributed. The height of the mixing layer, derived from the data of airborne sensing over Western Siberia, averaged over a period of many years, exhibits the directly opposite behavior with the maximum in July and the minimum in December–January.⁵ Seasonal behavior of the total aerosol content in the vertical air column is determined by the superposition of a number of mechanisms.⁶ In this case, the key point is the *in situ*

photochemical formation of particles in spring by analogy with ozone. Secondary maxima in 1993 are due to the peculiarities of atmospheric circulation in this period.⁷

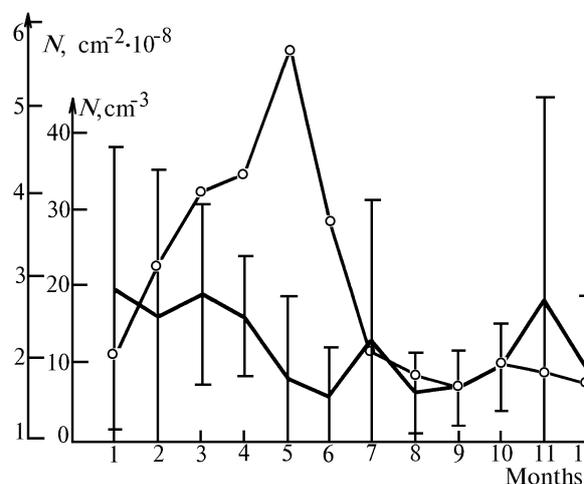


FIG. 1. Annual variations of the aerosol number density in the atmospheric surface layer in Tomsk (—) and the annual variations of the total aerosol content in the vertical air column over Western Siberia averaged over a period of many years (---). Vertical bars denote the standard deviation.

Such difference between annual variations of aerosol number density in the surface layer and total aerosol content in the vertical air column calls for not only a theoretical explanation but also an independent test. Almost the same study of aerosol was conducted in the vicinity of Moscow in 1991 (see Ref. 8). The aerosol mass concentration was monitored by means of an air-flow nephelometer during a year. For comparison of the data, the number density was converted to the mass concentration based on the particle size distribution and then normalized to its maximum. The same operation was performed with the data published in Ref. 8.

As is seen from Fig. 2, where the normalized values are shown, despite the different locations of points and periods of measurements, the common tendency of annual variations is retained, namely, the concentration peak is observed in cold season, and the concentration minimum – in warm season.

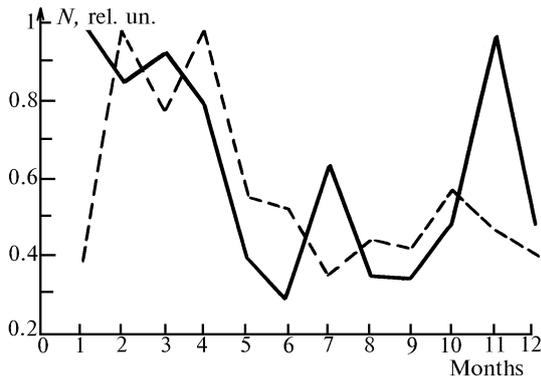


FIG. 2. The normalized annual variations of aerosol mass concentration in Tomsk (—) and in the vicinity of Moscow (---).

If the curve obtained in Tomsk is shifted to the left by a month, the similarity between the annual variations, especially in spring–summer period, will be more pronounced. It is possible that the products of aerosol photochemical formation, which takes place inside the European territory, arrive then in the territory of Western Siberia due to the west–eastern transport. However, for the final conclusion we need a longer series of measurements.

As is known, the aerosol field of the atmosphere is formed as a result of action of many sources, whose strength can change during a year. This can be manifested through the dispersed distribution of particles and their chemical composition. We analyze the dynamics of annual variations of the aerosol dispersed composition shown in Fig. 3.

As one can see from Fig. 3a, the submicron fraction has almost the same annual variations as the number density (see Fig. 2). It is not surprising since the finely dispersed fraction was not measured in the experiment and hence the value of the total number density was largely determined by the submicron one. We note only, that the curve behavior in Fig. 3a is smoother than that in Fig. 2.

Based on the established concepts, the minimum must be about 1 μm between the submicron and coarsely dispersed fractions of aerosol in the curve of the particle size distribution. Figure 3b shows the variations of number density of particles whose diameters are close to minimum. This figure shows almost steady decrease of aerosol number density from January to August–December. The exception is the May–June period, breaking this monotony.

The source of the coarsely dispersed fraction is, as a rule, the underlying surface and particles of photochemical origin partly "aged" in the atmosphere. From Figs. 3c and d, it follows that in the region of measurement the main source is the underlying surface since in winter months the number density of particles of these diameters is almost zero. In our opinion, this is due to the fact that the surface covered with snow is only a sink for coarsely dispersed fraction, and the surface itself does not produce this fraction. In summer, strengthening of wind and drying of the ground promote the processes of dispersion and particle rise. The turbulence more intense than in winter promotes the transport of coarsely dispersed particles upward.

It should be noted that there is close similarity of the behavior of curves in Fig. 3d and the curve of the total aerosol content in the vertical air column shown in Fig. 1. Their close similarity enables us to assume that a

certain contribution to the annual variations of coarsely dispersed fraction of aerosol in spring–summer period comes from particles of photochemical origin coarsened and aged in the higher atmospheric layers during the period of natural smog formation, and then settled on the ground.

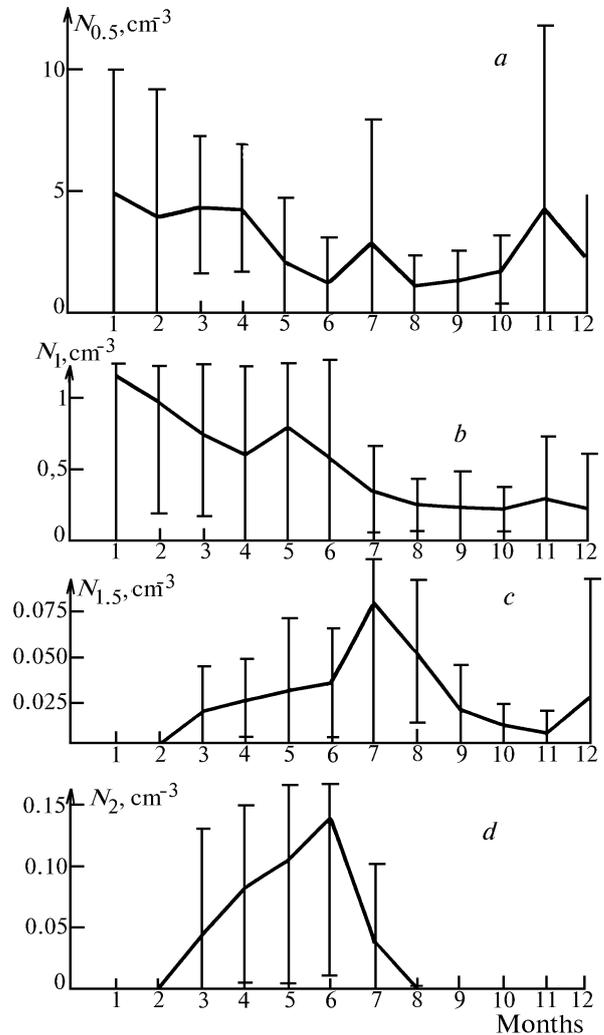


FIG. 3. Variations of the number density of individual fractions of aerosol in 1993 in the vicinity of Tomsk: the particle diameters $d = 0.5\text{--}0.6$ (a), $1\text{--}1.5$ (b), $1.5\text{--}2$ (c), and $2\text{--}4$ μm (d). Vertical bars denote the standard deviation.

Thus, the annual variations of dispersed composition of particles show the peculiarities of action of different sources in different seasons.

Without presenting the diagrams of daily variations, we note that, in analogy with Ref. 8, the daily variations of aerosol are more pronounced during summer months than during winter ones. In this case, two maxima are typical, namely, during morning hours (9:00–11:00, LT) and during the sunset (21:00–24:00, LT). During the winter period, the daily variations are less pronounced. The morning maximum remains but is shifted toward later time (10:00–12:00, local time) evidently, due to later sunrise. The evening maximum disappears but the night maximum appears from 22:00 till 2:00, then the decrease of the aerosol number density is observed.

The lack of strong anthropogenic aerosol sources close to the point of measurements hinders thus far the interpretation of such daily variations, namely, whether they are due to the effect of natural processes or of Tomsk located in the vicinity of measurement point. This problem will be discussed in detail in the next paper of this series.

The test of chemical composition is of paramount importance for aerosol monitoring. This test makes it possible to identify the sources of aerosol emission in the atmosphere as well as to define the optical constants necessary for calculation for the radiative models. However, at present there are no effective methods of solving this problem. In modern practice, we use preliminary taken samples with their concentration and subsequent analysis in the laboratory. This procedure is laborious and costly. Therefore, in 1993 measurements of this kind were performed only occasionally, in April and July.

Sampling and concentration of particles were performed using the filters of AEA-BP-20 type. The duration of sampling was 24 hours. Then the filters were processed in the Analytical Laboratory of the Tomsk State University by group of scientists headed by Dr. Z.I. Otmakhova. The employed methods and thresholds of detection were described in detail in Ref. 9. The average monthly characteristics of chemical composition of particles are shown in Fig. 4, where all detected ions and elements are conventionally divided into three groups: upper figure shows the elements contained in particles of natural origin, usually called terrigenous; intermediate figure shows the elements entering the particles as a result of anthropogenic activity or photochemical processes; lower figure shows the microelements that may be both of terrigenous and of anthropogenic origin. The relative contribution of the first and second groups to the concentration is almost

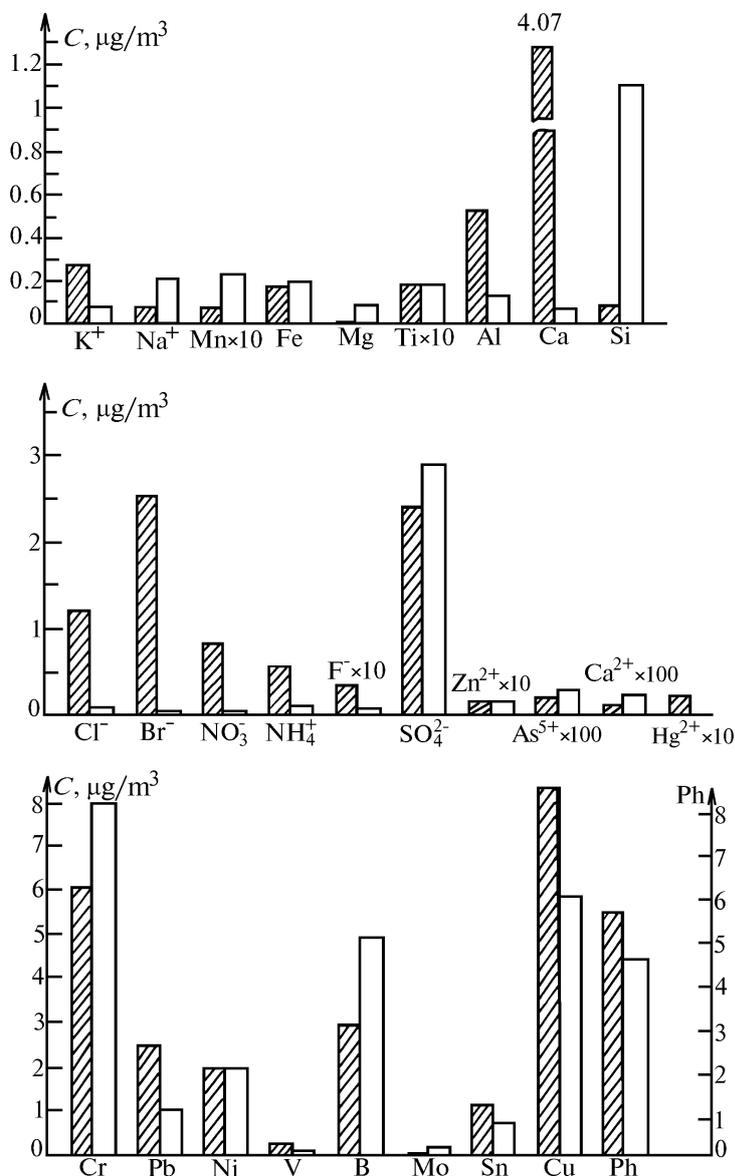


FIG. 4. Aerosol chemical composition in the vicinity of Tomsk: shaded regions are for April of 1993 and empty regions are for July of 1993.

the same for the territory of Western Siberia, the total contribution of microelements is several percent.¹⁰

Figure 4 shows that when going from spring to summer, the composition of terrigenous component changed. The Na⁺ concentration increased by a factor of two that points to the increased recurrence of marine air masses in the measurement region,⁷ the Al concentration decreased significantly (by a factor of three), and the Ca concentration decreased almost by a factor of 40, but the Si concentration increased (~10 times) as well as the Mg concentration. The last also points to the fact that in July the arrival of air masses to the territory of the region differed from that in April.

The contribution of anthropogenic component of Cl⁻, Br⁻, NO₃⁻, NH₄⁺, and F⁻ ions, except for the SO₄²⁻ ion, significantly decreased with the exception of the Zn²⁺, As⁵⁺, Cd²⁺, and Hg²⁺ ions, whose concentration in the region was from 10 to 100 times less than the concentration of the other ions. This is likely due to intensification of scattering of anthropogenic emissions in the summer period.

The concentration of microelements, as a whole, also decreased from April to July. The exception is provided by Cr, B, and Mo.

Comparison of the results obtained at the TOR station with the data obtained for the other regions, performed in Ref. 1, indicates that they differ considerably, and first of all, in rather high relative contribution of photochemical component.

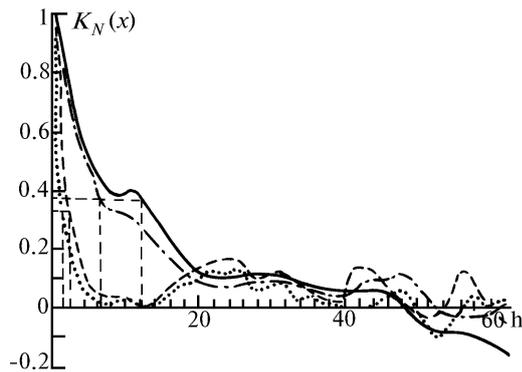


FIG. 5. Normalized autocorrelation functions of aerosol number density for $d > 0.4 \mu\text{m}$: solid curve is for January, and dashed curve is for July; $d = 1.5-2 \mu\text{m}$: dot-dashed curve is for January, and dotted curve is for July.

And finally, we estimate the scales of aerosol fields by the autocorrelation functions. For this purpose, we use the same methodology as for estimating gaseous components.¹⁰ The corresponding curves are shown in Fig. 5, where the horizontal lines show the 0.9995 confidence levels for the correlation coefficients. Table I gives the spatial scales of aerosol fields, calculated by the obtained correlation lengths.

TABLE I. Spatial scales (km) of aerosol fields in the vicinity of Tomsk in 1993.

Month	Diameter of particles	
	$d \geq 0.4 \mu\text{m}$	$d = 1.5-2.0 \mu\text{m}$
January	161	31
July	56	14

As follows from the data in Table I, the mean scales of aerosol fields for the number density are 161 and 56 km in January and July, respectively. For the coarsely dispersed fraction, these values are considerably less, namely, 31 km in January and 14 km in July. Such differences are quite natural since the coarsely dispersed fraction is poorly entrained by the air and is more variable in space.

At the same time, the mean scales of aerosol fields are considerably less than for gases considered in Ref. 10, especially during winter period that once more emphasizes the need for regular monitoring of air aerosol composition.

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