Structure of many-year aerosol trend near Tomsk

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The many-year trend of aerosol number density, earlier revealed by the authors, was shown to be mainly due to variations of the particle submicron fraction. The particle number density of the coarse-dispersed fraction varies randomly near some mean value. As a result, the total amount of the aerosol matter in the atmosphere varies within much more narrow limits than the number density. Analysis of the ion and element composition of aerosol has shown that the aerosol number density decreases due to compounds of natural origin.

Aerosol particles play a leading role in a number of atmospheric processes. Scattering and absorbing solar radiation, they cause a cooling of the atmospheric boundary layer if they reside in the stratosphere, or its heating if they are in the troposphere. As active participants of condensation processes, they contribute to formation of clouds and fogs. Being formed during chemical and photochemical processes, aerosol particles have in their composition heavy metals and other carcinogenic admixtures affecting adversely on the ecological situation. Such a role of the atmospheric aerosol requires constant monitoring of its concentration in the air to reveal the reasons of its variability, as well as development of methods of its short-term and long-term prediction.

We began the monitoring of the aerosol number density in 1983, first by means of the aircraft-laboratories IL-14 and AN-30 "Optik-E"[1] and then at the TOR station.[2] The results of the long-term measurements are summarized in Refs. 3 and 4. The aerosol number density was found to have long-term variability, at which its magnitude has been changed by a factor of 4.7 in the 1980s and by a factor of 13 in the 1990s. At the same time, the aerosol number density behavior followed the solar activity behavior with a lag of 2–3 years. Similar behavior was also described in Refs. 5 and 6 for the atmospheric aerosol optical thickness.

In spite of good agreement between the results on trends in variation of the aerosol concentration,[3–6] other data obtained during the same periods differ markedly. Thus, the monitoring data, obtained in the 1990s at the Institute of Atmospheric Physics RAS with a nephelometer, which measured the aerosol scattering coefficient at an angle of 45° and $\lambda = 0.55$ μm,[7] as well as at the Institute of Atmospheric Optics SB RAS with a photoelectric aerosol nephelometer (PAN) measuring the same characteristic at an angle of 45° and $\lambda = 0.52$ μm,[8] have not revealed such a significant its variation as reported in Refs. 3 and 4. This is well seen in Fig. 1, where the long-term variability of the aerosol number density and volume concentration is given according to the TOR-station data together with the aerosol mass concentration measured at Zvenigorod (Moscow Region) and Tomsk. The obtained nephelometric data are recalculated from the aerosol scattering coefficient to the mass concentration. Figure 1 shows that the mass concentration by the nephelometric data in the 1990s changed by a factor of 1.5 at most, while the number density – by a factor of 13, and the volume concentration – by a factor of 7. Probably, such divergences are due to the transformation of the particle size distribution, whereas the volume of aerosol matter varied insignificantly. A check of this hypothesis is one of the goals of this paper.

To analyze variations of the dispersion composition, let us examine Fig. 2. Recall that at the TOR station the 12-channel measurements of particles with $d = 0.4$–10 μm are conducted. It is seen that the submicron fraction ($d = 0.4$–1.5 μm), which mainly determines the aerosol number density ($d > 0.4$ μm), varies in the same way as described in Refs. 3 and 4. Its maximum was observed in 1994.

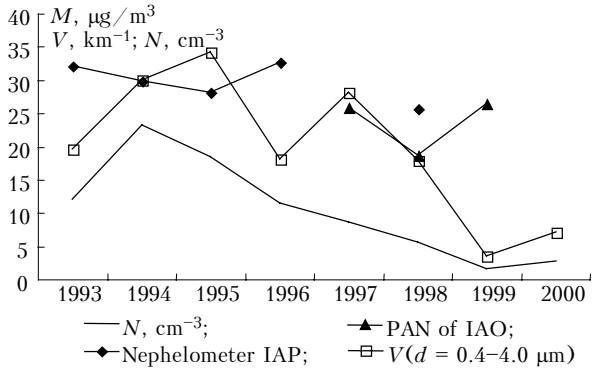

Fig. 1. Annual mean number density N (—) and volume aerosol concentration V (□) measured at the TOR station; mass concentration (M) measured at Zvenigorod (◆—◆) and at Tomsk (▲—▲), μg/m^3.

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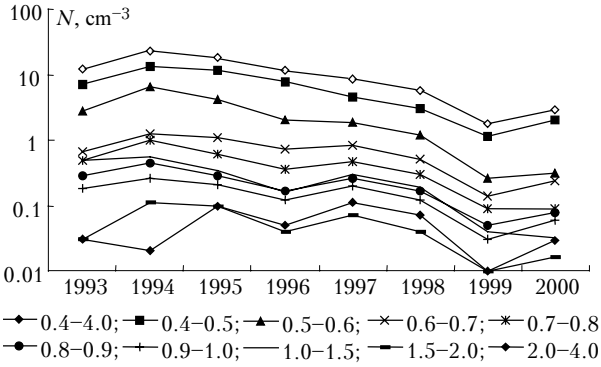

Fig. 2. Variation of aerosol disperse composition near Tomsk over a period of 1993–2000.

Then it smoothly decreases up to 1999, when a minimum was determined. The coarse fraction (d = 2–4 μm) behaved differently. Early in the 1990s when the submicron fraction concentration increased, the content of the coarse fraction varied randomly. In 1997 concentration of the coarse fraction reached its maximum while that of the submicron fraction decreased by a factor of 2 or 3. Over a period from 1998 to 2000 the both fractions varied simultaneously.

The number density of aerosol particles is not always adequate to mass and volume content of aerosol matter in the atmosphere, and some scientists prefer not to use this characteristic. Figure 3 presents data on the volume concentration of particles of different sizes.

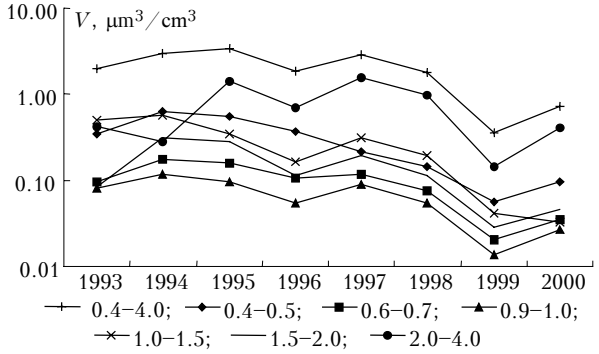

Fig. 3. Aerosol volume concentration V near Tomsk over a period of 1993–2000.

Figure 3 demonstrates the same trend as Fig. 2. From 1994 the concentration of submicron fraction decreases. The concentration of the coarse fraction at first somewhat falls and then increases in 1995. Over a period from 1995 to 1998 it fluctuates about some mean value and falls only in 1999. The total volume of the aerosol matter V (d = 0.4–4.0 μm) varies insignificantly during the period from 1993 to 1998. We can see the interannual variations which, like the nephelometric data, do not exceed 2 times. In 1999 the volume concentration of particles of all fractions simultaneously decreases that is reflected in the total volume of the aerosol matter.

A comparison between Figs. 2 and 3 shows that the volume concentration varies slightly as compared to the number density during the most part of the period under analysis (1993–1997). This is indicative of the fact that in these years the multiple decrease of the aerosol matter in the atmosphere was absent, and only a qualitative change of the particle composition took place; the concentration of the submicron fraction decreased and that of the coarse fraction practically remained constant.

Taking into account the regularities of aerosol generation and transformation in the atmosphere[10,11] we may suppose that over the period from 1993 to 1999 for some reasons the power of the sources of the aerosol-producing vapor or the intensity of processes of the atmospheric purification gradually decreased. Therefore, either submicron fraction had no time to be replenished by new particles from microdisperse fraction, or the partial pressure of the aerosol-producing vapor was deficient for heterogeneous condensation on particles of microdispersed fraction. At the same time, the coarse aerosol particles arrived by the common laws. Since the mass of aerosol in the atmosphere is largely determined by the coarse fraction, it remained practically unchanged, what is reflected in the above-mentioned nephelometric measurements and the total aerosol volume. Probably, the post-volcanic purification of the atmosphere plays some role as well.[8] But our earlier study of the direct post-volcanic impact has led us to a negative result.[12] We may assume that this mechanism, if it exists, is realized through some intermediate processes, for example, photochemical ones.

The second question, to be considered in this paper, is the following. By the available estimates,[9] at present the content of aerosol of anthropogenic origin in the atmosphere averages 10%. According to the data from Ref. 3, the aerosol number density in the 1990s decreased by a factor of 10–11. This raises the question: where does the anthropogenic aerosol vanish from the atmosphere provided its content there is not overestimated?

During the 90s, at the TOR station the aerosol samples were casually analyzed for their chemical composition. The obtained data allowed us to see changes not only in disperse composition but also in chemical composition, which may reflect the nature of the aerosol-generating compounds. The results of determination of the ion-element composition in 1993, 1997, and 1998 are given in Fig. 4.

Figure 4 shows that the concentration of ions and elements of natural origin (top histogram) during the period from 1993 to 1998, as a rule, decreases. The exceptions are Mg, K^+, NO_3^-, Cl^-, Al, which have a variable concentration. The concentration of ions and elements of anthropogenic origin (bottom histogram), on the contrary, relatively grows either in 1997 or in 1998, although, as compared with natural ones, it is by two orders of magnitude less. This is due to the fact that the aerosol trend, reported in Refs. 3 and 4, is mainly due to natural processes and the concentration decrease was at the cost of ions and elements of natural origin. The growth of concentration of anthropogenic compounds

shows that this share of the aerosol matter was not decreased in the period of analysis. Thus, Fig. 4 shows that contribution of anthropogenic component to the total aerosol mass in the vicinity of Tomsk is much less than on average around the world, where its contribution is 10% (Ref. 9); and at a period, when contribution of the natural component of aerosol particles decreases, relative contribution of the anthropogenic component increases.

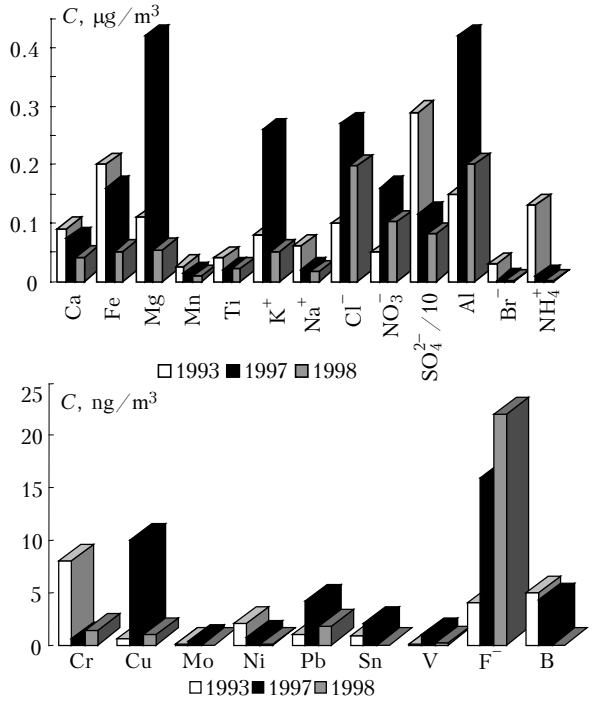

Fig. 4. Aerosol chemical composition at Tomsk in 1993, 1997, and 1998.

The measurements at the TOR station are carried out in the vicinity of the city, therefore, naturally, urban sources somehow contribute to the results. In 1993 and 1997 the measurements of chemical composition were performed simultaneously at 2 points: at the TOR station and in the settlement of Kireevsk, Tomsk Region. There are no industrial objects in the neighborhood of Kireevsk, therefore this region can be considered as a background one. In Ref. 13 the classification of elements and compounds is given according to their contribution to the origin of aerosol particles. The results are demonstrated in the Table. They allow one to compare the Tomsk and background data.

It follows from Table that the elements and ions of anthropogenic origin (> 60%) increased their contribution to aerosol both at Tomsk and Kireevsk. Elements and ions, whose sources are natural processes (< 40%), on the contrary, decreased markedly their contribution at the both points. Therefore the results obtained are characteristic not only of Tomsk but the region as a whole.

Comparing the aerosol behavior and solar activity,[3,4] we have predicted the beginning of growth of the aerosol number density in 2000 and its peak in 2003. The data of 2000 and 2001, presented in Fig. 5, testify that the first part of our prediction works well. Further measurements must show a feasibility of the prediction as a whole. If our assumption is true, then, based on the prediction of solar activity, we will be able to make the long-term prediction of the aerosol number density in the atmosphere.

Table. Relation between concentrations of chemical elements in aerosol in 1997 and 1993

Anthropogenic contribution, %	Element	Tomsk	Kireevsk
	Anthropogenic contribution exceeds 60%*		
88	Cu	17	3.5
81	V	11	6
94	Pb	4.4	5.8
83	Mo	4	2.5
93	Cl^-	27	34.4
89	F^-	4.5	
80	Si	15.8	3.8
79	Sn	2.1	11.8
	Anthropogenic contribution is below 40%*		
18	Ti	0.5	
10	Ca	0.8	0.6
25	Fe	0.8	0.4
23	Mn	0.5	1
39	Na^+	0.3	0.2
7	Br^-	0.1	0.3
40	SO_4^{2-}	0.4	0.4
38	NH_4^+	0.1	0.03
	Anthropogenic contribution is 40–60%*		
51	Al	2.9	2.9
45	Cr	0.1	0.8
50	B	0.9	1.3

* By S.V. Savenko.

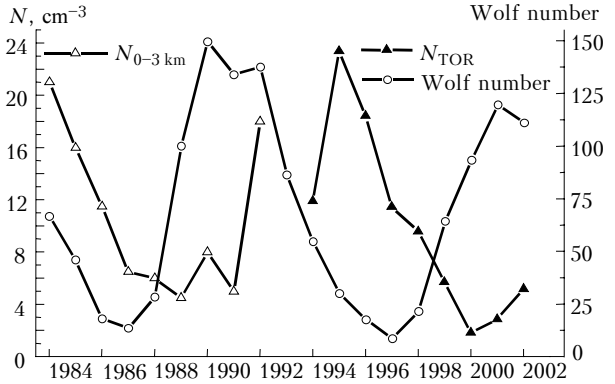

Fig. 5. Long-term behavior of aerosol number density and the Wolf numbers. $N_{0-3\ km}$ are airborne data, N_{TOR} are data of TOR station.

From the above it follows that the trend, determined for the total number density, has a complex structure. It is dictated by transformation of disperse and chemical composition of aerosol and, most probably, has a natural ground, namely, many-year cyclicity of atmospheric processes.

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