

TIME VARIABILITY OF THE TROPOSPHERIC AEROSOL OVER WESTERN SIBERIA

B.D. Belan and G.N. Tolmachev

*Institute of Atmospheric Optics,
Siberian Branch of the Russian Academy of Sciences, Tomsk
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Time variability of integral and mean aerosol concentration in the atmospheric layer up to 5 km over Western Siberia is studied. Annual behavior of the concentration is shown to be asymmetrical due to an increase in the photochemical component in spring. The data processed reveal a long-term trend in aerosol concentration within this layer.

Aerosol plays an important role in many atmospheric processes such as cloud formation, extinction of solar radiation, generation of primary and secondary admixtures in air, that often are pollutants, etc. Contribution of aerosol into the atmospheric processes is mainly determined by its concentration in the lower troposphere, where the main portion of suspended substances is concentrated.

The data on spatial aerosol distribution obtained at the Institute of Atmospheric Optics during many years airborne sounding of the atmosphere¹ make it possible to partially fill this gap.

The aim of this paper is to summarize these data in order to reveal the temporal (long-term) variability of the aerosol content in the lower and middle troposphere. Long-term variability means here the variations of aerosol concentration starting from monthly mean. The measurement techniques and instrumentation are described in Refs. 2–5.

Let us note that individual and even monthly mean vertical profiles of the concentration strongly vary depending on the weather conditions, time, day, season, etc.⁶ These variations can be inclination of the curve of vertical distribution, increase or decrease in the total concentration or in separate layers, that makes their comparison and interpretation of the data very difficult. So it is expedient to consider the average or integral distribution in the whole layer.

We have selected Western Siberian region for these investigations because just for this region we have most complete set of measurement data. The vertical profiles obtained in the "caps" of cities were excluded from processing, i.e., only the data obtained under background conditions were analyzed.

In order to consider the annual behavior of aerosol concentration, we have calculated the integral number of particles in the vertical column of a unit cross section area:

$$\hat{N} = \int_0^H N(h) dh, \quad (1)$$

where $N(h)$ is the number density of aerosol at the height h . The value H was taken to be equal to 5 km that corresponds to the ceiling of the IL-14 airplane. The long-term average (1981–1988) annual behavior of aerosol calculated by Eq. (1) for Western Siberia is shown in Fig. 1. About 3000 profiles of the number density were used for its construction.

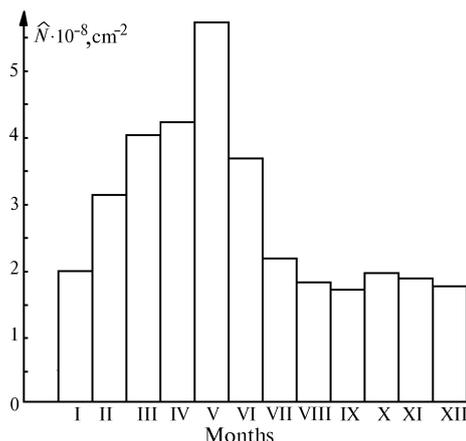


FIG. 1. Long-term average annual behavior of the aerosol concentration in the vertical column of air up to 5 km per 1 cm² of the surface over Western Siberia.

It is seen from Fig. 1 that the long-term average annual behavior of the integral aerosol content in the vertical column of air has an asymmetrical shape with a well pronounced maximum in May and a wide minimum since August till January. Such a temporal behavior of the aerosol number density is very similar to the annual behavior of the ozone column density⁷ and its near ground concentration measured in the same region.⁸ As is well known, ozone has only photochemical origin that can be an evidence of similarity in mechanisms of their production. The cause of the increase in aerosol concentration in the troposphere over Western Siberia is also the intensification of photochemical processes in spring.

The well-known aphorism by G.V. Rozenberg is "aerosol is a process" (Ref. 9) or, in other words, that

means that the aerosol particles are produced directly in the atmosphere from the aerosol producing gases. The principal mechanism of production and growth of these particles is heterogenetic condensation of the aerosol producing vapors, to which the chemical capture of admixture gas precursors joins.¹⁰

Based on the above said, one can suppose that the geographical position of the Western Siberia causes (enhances) the annual behavior of \hat{N} revealed. On the one hand, a significant part of its territory is covered with forests that generate a lot of terpenes and isoprenes in spring,¹¹ which are material for aerosol particles.¹² As known, it often leads to generation of natural photochemical smog.¹³ On the other hand, a lot of anthropogenic products are transported to this territory⁵ (or produced over it), that favors the growth of aerosol particles due to the capture of the admixture gases. Joint action of these two mechanisms under conditions of high humidity characteristic of the region¹¹ results in this effect.

According to Ref. 14, 35–85 % of the mass of natural aerosol is water soluble fraction including primarily the ions SO_4^{2-} , NO_3^- , K^+ , Na^+ , and Cl^- that play the main role in the process of growth and transformation of particles. Then the validity of all the aforementioned statements should also be confirmed by the annual behavior of the aerosol chemical composition.

Let us look at Fig. 2 to test such a conclusion. It shows the long-term average chemical composition of aerosol in May and December (maximum and minimum in the annual behavior of concentration). For its construction we used 165 aerosol samples collected in May and 167 samples collected in December in the 0.5 to 5 km height range since 1983 till 1987.

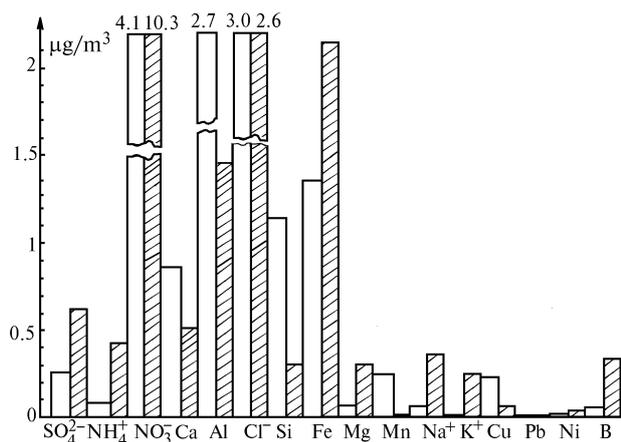


FIG. 2. Long-term average chemical composition of aerosol over Western Siberia in May (hatched areas) and December (light areas).

It is seen from Fig. 2 that the May increase in the aerosol concentration occurs mostly due to its water soluble fraction. The content of ions SO_4^{2-} , NH_4^+ ,

NO_3^- , Na^+ and K^+ in the composition of particles increases since December till May by 2–4 times. Contrary, the mineral component of aerosol, that, as a rule, is produced by dispersing,¹⁵ has higher concentration in December. This applies to such elements as Ca, Al, Si, Mn, and Cu. Thus, it follows from data shown Fig. 2, that photochemical generation of aerosol particles in the atmosphere is essentially weaker in December than in May. It is caused by a sharp decrease in the amount of the aerosol producing substances in the lower atmosphere during the cold season because of termination of the vegetation activity and a decrease in evaporation from the underlying surface covered with snow.

The absence of data on annual behavior of the aerosol concentration for other regions does not allow us to compare them. Nevertheless, one can suppose that the data presented are interesting for the specialists who study the climate formation processes.

The long-term airborne sounding of the atmosphere makes it possible to estimate the inter-annual variability of the aerosol concentration.

To do this, let us consider first Fig. 3 which shows the dynamics of aerosol number density averaged over the layer up to 3 km in May and December, as calculated by the formula:

$$\bar{N} = \frac{1}{H} \int_0^H N(h) dh. \tag{2}$$

May and December were chosen for the above reasons, as well as due to the fact that a good data sets are available for these months.

It is seen from Fig. 3 that in the period from 1983 till 1987 there was observed a distinct tendency in the aerosol number density over Western Siberia to decrease at a rate of 4.5 in December and 5.5 in May.

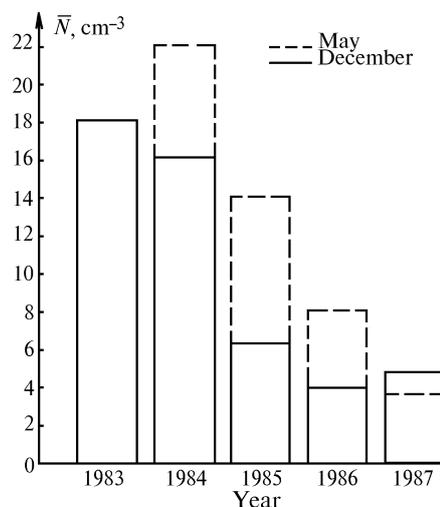


FIG. 3. Long-term average aerosol number density in the layer up to 3 km in May (dashed line) and December (solid line).

Although the sounding has been carried out outside the industrial centers, the result obtained seems to be unexpected against the background of the increasing anthropogenic pollution of the atmosphere. So let us consider other data. Since there was no regular vertical sounding of aerosol over the former USSR, we must make estimation based on the data of the ground-based measurements carried out at the network of air pollution monitoring.

The total emission of suspended substances over USA and former USSR as well as the variation of aerosol concentration over some cities are shown in Figs. 4 and 5 from the data published in Ref. 16.

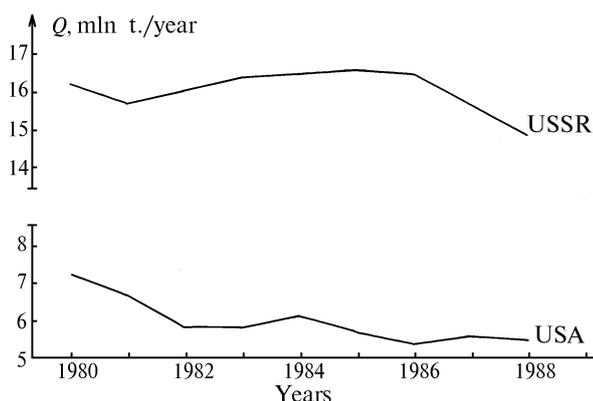


Fig. 4. Total emission of suspended substances in the USSR and USA.

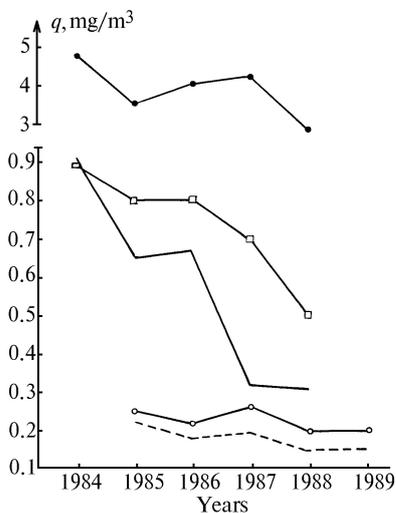


Fig. 5. The concentration of suspended substances as given in Ref. 16: Karaganda (---), Novosibirsk (—○—), Omsk (●—●), Kaliningrad (□—□), Tol'yatti (○—○).

It is seen from Fig. 4 that since 1983 till 1988 the amount of emission over USSR decreased by 9 %, and over USA by 4 %. Taking into account that the contribution of these two states is a significant part of the total emission in the world, and that the decrease of concentration revealed in Fig. 3 is manifold, one can

suppose that the data on Fig. 4 exclude the idea on the anthropogenic nature of the trend revealed.

In addition, the data on Fig. 5 show that there is a tendency in the aerosol concentration over some cities of the former USSR to decrease. It is confirmed by the data presented in Fig. 3. The tendency of decreasing the concentration of suspended substances is also revealed in the air over some foreign cities,¹⁷ such as Calcutta, Athens, Madrid, and Milano during ten years since 1975 till 1985.

The comparison of the results presented leads to the following conclusion. Obviously, the decrease in aerosol concentration noted in Fig. 3 has not the anthropogenic origin, because no worldwide tendency to decreasing aerosol content has been observed.¹⁶ At the same time, the tendency of decreasing the aerosol concentration in the near ground atmospheric layer is revealed in some sites throughout the world.

If one takes into account that the aerosol concentration in urban air is a sum of the natural background and the aerosol formed by spreading the local emissions, one can assume that the background component caused by the natural processes essentially decreases. Hence, the result we obtained is quite reasonable and reflects the long-term dynamics of the atmospheric aerosol.

In order to extend the period under consideration, let us consider the annual concentration, instead of the monthly mean ones. The annually mean values of the aerosol number density calculated by Eq. (2) in the layer up to 3 km are shown in Fig. 6.

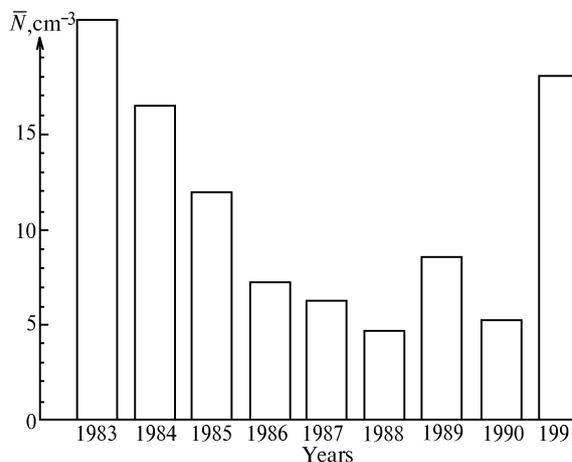


Fig. 6. Annual average aerosol number density in the layer up to 3 km over Western Siberia.

It is seen from Fig. 6 that the annually mean aerosol number densities decrease since 1983 till 1988, as the May and December monthly mean values do. Certain increase in the aerosol content can be seen in 1989. The obvious increase of the concentration is observed in 1991 up to the values obtained in 1983. Thus, during 9 years, since 1983 till 1991 a wave-like variation of the aerosol concentration was observed in the lower atmosphere over Western Siberia.

A number of oscillations of meteorological parameters with the periods longer than 1 year are known in the climate theory.^{18, 19} They are 26-months, 11-years, 22-years cycles, etc. The shape of the interannual behavior shown in Fig. 6 is most likely similar to the curve that is characteristic of the 11-years cycle.²⁰ So one can suppose that in this case we have observed a part of a 11-year cycle of the natural variability of atmospheric aerosol.

Analysis of the variability of the aerosol chemical composition, the samples for which were collected simultaneously with the measurements of the number density, show that during the period under consideration mainly the mineral fraction decreased in its composition. The most strong decrease is observed for Al, Ca, and Fe. Concentration of the SO_4^{2-} ions is practically the same. The components of the water soluble fraction that are usually related to aerosol of marine origin,²¹ including the ions NH_4^+ , Na^+ , and K^+ , increase their contribution to the composition of particles. In our opinion, this fact indicates that the long-term variations of the aerosol concentration are accompanied by variations in the regime of air circulation over the region. In particular, the increase of concentration of the ions NH_4^+ , Na^+ , and K^+ can be connected with the increased number of events when the marine air masses come to Western Siberia, that also indicates the possible relation between the aerosol variations with 11-year cycle of atmospheric processes. It is well known that circulation mechanisms also undergo this evolution.^{22, 23}

For summing up the results of the paper, let us note the principal results. The study of the annual behavior of the aerosol concentration in the lower atmosphere has revealed its asymmetric shape caused by the increase of the aerosol mass from winter to spring, the primary cause of which is the emission of aerosol producing substances by natural ecological systems (forests). The interannual variations of the aerosol concentration, revealing the long-term tendencies to decrease and growth, are connected with the dynamics of the mineral component, and most likely, reflect the natural cycles occurring in the atmosphere.

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