ANNUAL BEHAVIOR OF THE CONTENT OF SUBMICRON AEROSOL IN THE TROPOSPHERE OVER WEST SIBERIA

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In this paper we present analysis of the annual behavior of the vertical profile of scattering coefficients of the dry matter of submicron aerosol particles. Experimental data analyzed have been obtained from nephelometric airborne sounding of the atmosphere over West Siberia in 1986–1988. Sounding was carried out at altitudes from 0 *to* 5 km. *It is shown in the paper that in spring time there was observed an enhanced atmospheric turbidity at altitudes above* 2 km, *that was caused by the long–range transport of aerosol produced in the western regions of Siberia after snow thawing. We also consider in this paper the variability of the vertical profile of aerosol content for four seasons. Based on data on mean temperature of the low atmospheric layers we propose an empirical formula for estimating the mixing layer height. Annual behavior of the mixing layer height is analyzed.*

The main purpose of the International Global Aerosol Program (IGAP) formed by scientific community is "improvement of understanding of the role of atmospheric aerosol in the mechanisms of the influence on the global climate change and geospheric and biospheric processes."[1]

The submicron fraction particles, that have the longest lifetime and participate almost in all atmospheric processes,[2,3,4] are of the special importance among the great variety of aerosol particles being present in the atmosphere. Submicron particles mainly determine the optical state of the atmosphere in the visible spectral range, the absorption of IR radiation[5,6] and, because they are the products and participants of all aerosol–to–gas transformations, accumulate a lot of various chemical compounds and transfer them to long distances.

Investigations of the processes of the spatiotemporal variability of aerosol particles for different climatic zones of the Earth make up the experimental basis for studying their effect on the climatically and ecologically significant factors and estimating the unfavorable tendencies. It is clear, taking into account the lifetime of submicron particles in the atmosphere, that their observations at the regional scale are most optimum.[7]

In this paper we consider, basing on the results of airborne nephelometric investigations of aerosol characteristics,[7,8] the annual behavior of variability of the vertical profile of scattering coefficients over West Siberia. Note that considered problems concern only the submicron aerosol because the nephelometric technique used in our investigations provides reliable data only on the particles of this size range.[7]

Detailed discussion of basic aspects of measurement technique and the substantiation of suitability of the formed bulk of observational data for the analysis of the processes at regional scale can be found in Ref. 7.

Describing the main factors of the aerosol variability on the basis of measured values of the scattering coefficient σ, we would like to remind that few parametric models available allow us to reconstruct a large number of optical characteristics in the wavelength range[9–11] of $\lambda \sim 0.5$–0.8 μm and some integral parameters of the size distribution, including the specific volume of particles V, from known value of σ.

Use of active nephelometric tools[12] allows one to separate the effect of relative humidity, that noticeably masks the action of geophysical processes of larger scale. Let us briefly explain the idea of such an approach using the Kasten––Hänel formula[13]

$$\sigma = \sigma_d (1 - f)^{-\gamma}, \qquad (1)$$

where σ is the *in situ* scattering coefficient, σ_d is the scattering coefficient of the dry matter of aerosol particles, γ is the parameter of condensation activity, and f is the relative humidity.

The content of submicron aerosol fraction (in our case $\sigma_d \sim V_d$, where V is the specific volume) depends on the whole of complex of the processes of different scales (from local to global). In theoretical sense σ_d is the function of the aerosol lifetime.[3,4]

The parameter of condensation activity γ depends on the particle substance, mainly on the ratio of soluble and insoluble species in the dry matter of aerosol particles,[14] therefore, in the general case it is governed by the processes determining σ_d.

Relative humidity f is not directly related to the content of aerosol particles in a particular air mass, and its variation[15] (as more noticeable in the near–ground layer) is controlled by relatively quick processes (diurnal variations of temperature, modulation of the radiation flux by clouds, the presence of local sources of higher moisture, and so on). Hence, in this approach one can relate the relative humidity to the inner scale weather factors, whose effect superimposes on the atmospheric processes of longer duration.

Such an understanding of the basic (temporal and spatial) scales of the aerosol properties variability makes it reasonable to study σ_d and γ parameters separately.

Then the further analysis will be performed for the scattering coefficients of the dry matter of aerosol particles σ_d (the study of the variability of the condensation activity parameter is a subject for a separate study, it will not be discussed in this paper).

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The sets of observations of σ_d available and the factors determining its variability are analyzed[17–20] for the near–ground atmospheric layer, but the data on the vertical profile of this parameter in the lower troposphere are practically absent.

Before the discussion let us define the terms more exactly. Let us understand stratification as a rate of the profile variation with height (for example, the normalized profile $\sigma(H)/\sigma(0)$). Let us call the absolute value of σ_d as the aerosol "content", because it is the parameter characterizing the total amount of particles in air mass and connected with its prehistory and with the nature and power of aerosol sources.

The average monthly profiles of aerosol scattering coefficient $\sigma_d(H)$ for the West Siberian region[7] are shown in Fig. 1. As is seen from this figure, both the profile stratification and the aerosol content strongly vary during a year. In order to estimate the impact of local sources on the formation of vertical profile, we have compared average monthly values of σ_d obtained at different heights separately over Tomsk and other sites of West Siberia. The results are presented in Figs. 2*a–e*. Noticeable differences are observed only in the near–ground atmospheric layer. Above this layer there are no any pronounced differences. This is indicative of the homogeneity of aerosol properties over all region and, in addition to Ref. 7, confirms the applicability of our bulk of data to the analysis of the factors of the regional scale. The average monthly values of the mass of submicron particles per 1 m^2 of the surface in the atmospheric column up to 5 km are shown in Fig. 2*f*. The calculations were done by the following formula:

$$M = C \int_0^{5\ \mathrm{km}} \sigma_d(H)\, dH\,, \qquad (2)$$

where $C = 0.45\ \mathrm{g/cm^2}$. The transition from σ_d to the specific volume has been done using the empirical relationship.[10,11] The density of aerosol substance taken was $\rho = 2\ \mathrm{g/cm^3}$.

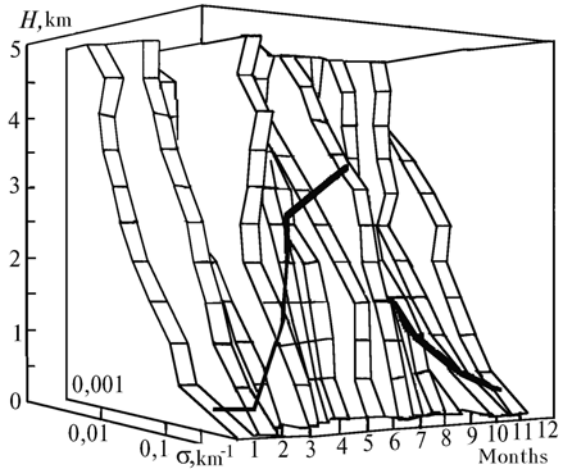

Fig. 1. Average monthly vertical profiles of the scattering coefficient of dry matter of aerosol particles. Annual behavior of the mixing layer height, as reconstructed from the bulk of data analyzed, is shown by solid line.

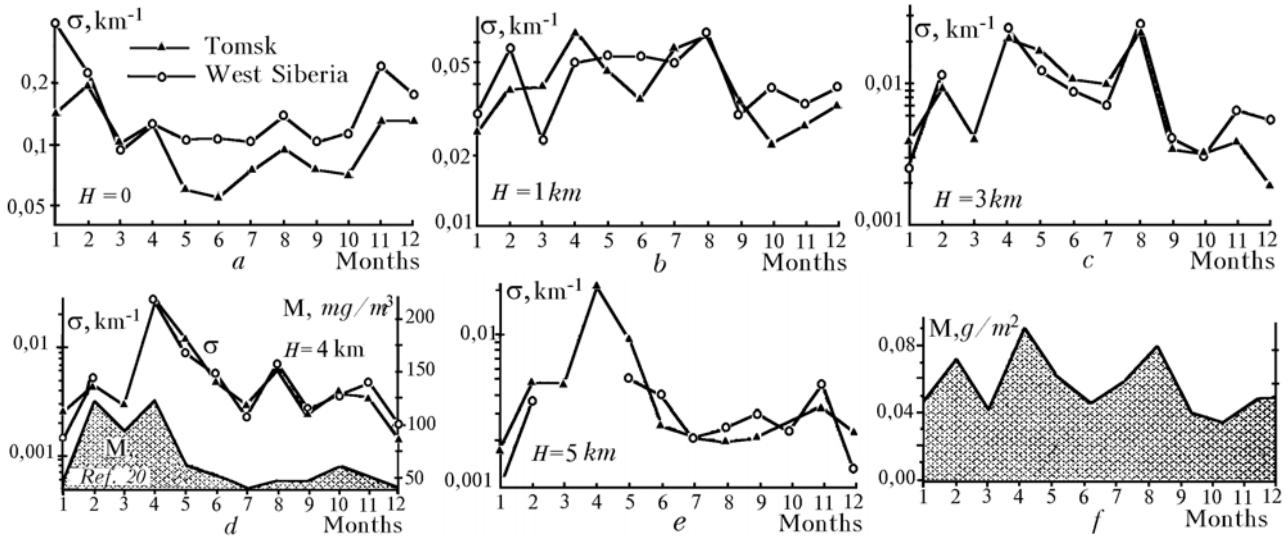

Fig. 2. Average monthly values of aerosol scattering coefficient at different heights over Tomsk and other West Siberian sites (a–e) and the annual behavior of mass of submicron aerosol particles in the atmospheric column up to 5 km *with the column base area of* 1 m^2 *(f).*

One can explain high near–ground values in winter by the intensification of anthropogenic aerosol sources (during cold period) and by the fact that the inverse temperature profile in the near–ground atmospheric layer[21] prevents penetration of aerosol emissions into upper layers of the atmosphere.

High values of the aerosol content that are distinctly seen in the second half of summer both in the vertical profiles at all heights and in the total content can be explained by the following factors. The atmosphere is most strongly heated in this season. As a result, the emission of aerosol particles from surface reaches high altitudes and, therefore, their lifetime in the atmosphere increases. Then the insolation and long daytime determine the intensity and duration of photochemical processes. The next factor is the prevalence of weakly mobile low–rate pressure fields[21] that favor the accumulation of tropospheric aerosol. The maximum aerosol content is observed in August instead of July, though the values of the above–listed factors are the largest in July.

Evidently, this is connected with a smaller amount of precipitation[21] in August. The additional aerosol sources can be forest fires that arise, as a rule, in the region under investigation during this season.

A decrease of the total aerosol content is observed during the fall. Evidently, it happens due to a weaker role of factors that are important in summer. The next cause are more frequent, compared to other seasons, fogs, drizzle, and steady rain[21] that favor the clearing of the atmosphere and switch off the action of the underlying surface.

Thus, for three seasons the main processes determining the variability and the effect of aerosol sources are similar for large areas (at least, for midlatitudes of the northern hemisphere) at the regional scale. But the situations observed in spring require special consideration.

Let us pay attention to the well pronounced maximum of the scattering coefficient observed in April at the heights above 2 km (Figs. 2*c*, *d*, and *e*). We have revealed this fact (the contribution of aerosol at these heights is also revealed in the total aerosol content in the atmospheric layer up to 5 km, Fig. 2*f*).

One can try to understand this phenomenon based on the following considerations. Quick variation of the Sun elevation angle and the great contrast between albedo and the thermal regime of the underlying surface, connected with the motion of the snow thawing boundary in spring, initiate enhanced dynamics of the atmospheric circulation.[22] For these reasons a significant spatial contrast of the state and power between the sources of particles and aerosol producing vapors should take place.

Since the westward transfer is characteristic of midlatitudes, one can suppose that the aerosol particles produced from various chemical compounds, that have been accumulated in winter and emitted into the atmosphere because of snow melting in west regions, come into the atmosphere of West Siberia. Possibly, the products of the aroused vegetation make some contribution to aerosol generation (by April the majority of conifers are free of snow and can emit the aerosol producing vapors under the action of solar radiation[23]).

Getting ahead, let us note that the Arctic air masses coming to West Siberia in winter and spring contain higher amount of aerosol at these heights, the maximum is precisely in spring. (This question will be discussed in detail in the next paper.) Of course, one cannot exclude other mechanisms, though unknown yet, that could be the reasons of this maximum. However, since the largest filling is observed above 2 km height, one can suppose that during this period the major portion of aerosol particles comes into the atmosphere over West Siberia from remote sources. Evidently, this conclusion is also confirmed by the fact that the annual behavior of submicron particles content in the near–ground atmospheric layer observed in the vicinity of Moscow [20] is similar to our data obtained at the heights above 2 km (the data from Ref. 20 are shown in Fig. 2*d*).

Let us consider now the questions concerning the annual behavior of the aerosol vertical stratification. To reveal most characteristic features of formation of the vertical profile, it is appropriate to consider its variability as assessed from seasonal ensembles. As is shown in Ref. 7, the meteorological and synoptic parameters of these ensembles are close to the average climatic data for the region under investigation. The histograms of the scattering coefficient of dry matter of aerosol particles are shown in Fig. 3 for four seasons. The average values are shown by solid lines. As is seen, the variations of the scattering coefficient within the whole of height range under investigation are more then one order of magnitude. In the majority of events the vertical profile $\sigma_d(H)$ has a layered structure. It has been observed by many authors.[23–25] As a rule, the layers are near the levels where the alternation of stable and unstable microregions is observed in the temperature profile (ΔH = 100–200 m), and their height and contrast vary occasionally. The average seasonal profiles $\sigma_d(H)$ are quite smooth, that is indicative of the absence of the heights of preferrable layer position.

The vertical stratification vary noticeably from season to season. It is seen more distinctly in Fig. 4, where the average seasonal profiles normalized to the near–ground value $\sigma_d(0)$ are shown.

It is clear that the formation of the vertical profile is mainly caused by thermal regime of the atmosphere. Actually, the maximum differences in stratification of optical characteristics are observed for the most contrast seasons, winter and summer.

As a rule, in winter the inverse temperature profile is observed over West Siberia[21] and, hence, the main amount of aerosol particles produced by near–ground sources is concentrated in the low 200–500 m layer. In this layer the fastest decrease of aerosol content with height is observed. The exponential decrease of the vertical profile of the scattering coefficient continues up to the height of 2.5 km. Almost constant value characteristic of the free atmosphere[25] is observed above this height.

In summer the high temperature of the underlying surface and low atmospheric layers provides good mixing of air and aerosol particles along the vertical direction. After small decrease in the near–ground layer (~100 m) the values of the aerosol scattering coefficient decrease insignificantly up to 3 km, then quickly decrease at the distance of 300–400 m, and then the regime of the free atmosphere is observed. Vertical profiles $\sigma_d(H)$ for spring and fall seasons are close to each other up to 2.5 km and intermediate between winter and summer profiles (as it should be expected from the temperature stratification). Above 2.5 km in spring the enhanced filling of the atmosphere with aerosol is observed, as it is mentioned above. It is caused by the action of remote sources instead of temperature stratification.

As is seen, the principal features of the variability of $\sigma_d(H)$ in season ensembles well agree with the three–layer concept of the aerosol height distribution.[26–27] Within the framework of the approach proposed in Ref. 24 and according to the terms accepted in meteorology, one can select the following height ranges: up to H_1 is the ground layer, H_2 is the height of the boundary layer or the layer of intense turbulent exchange, above H_2 is the layer of free atmosphere.

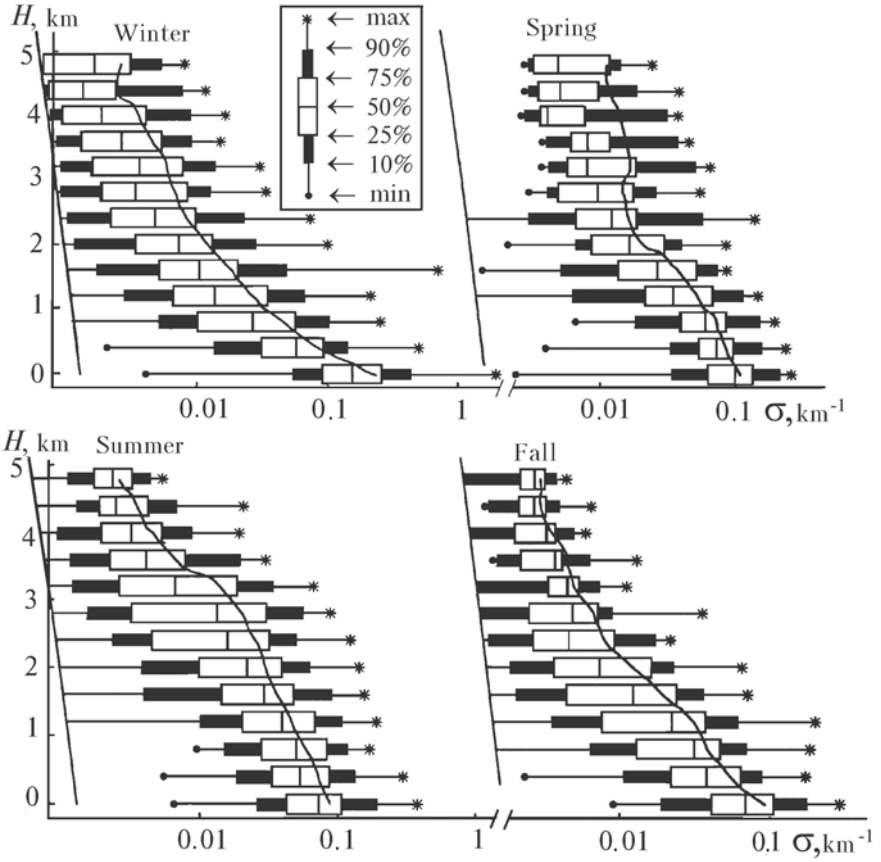

Fig. 3. Histograms of aerosol scattering coefficient at different heights.

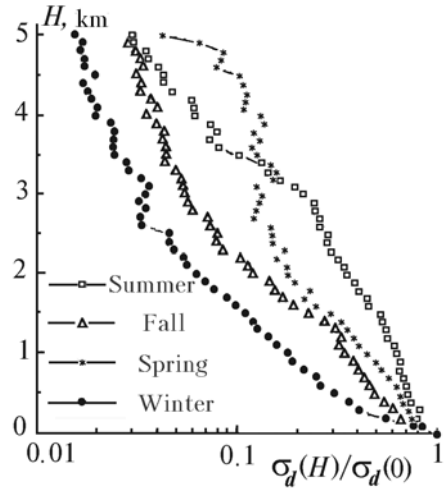

Fig. 4. Normalized vertical profiles of aerosol scattering coefficients.

It follows from Fig. 4 that the major dynamics of annual variability of the stratification $\sigma_d(H)$ occurs in the layer up to H_2 (except the spring season, when the effect of the long–distance transport of admixtures essentially changes not only the filling of the atmosphere by aerosol substance but also the vertical stratification within the height range above 2.5 km).

To estimate the height H_2, often called the mixing layer height, different techniques are used in meteorology, and this problem has been discussed in papers devoted to the aerosol processes many times. However, when estimating it from the intensity of the turbulent mixing (that is absolutely right from the standpoint of the principal mechanism), a number of problems arise because the diurnal variations of temperature and underlying surface strongly affect the turbulent exchange processes. The next reason is the gradual heating (or cooling) of the lower troposphere day by day. Aerosol processes have greater time lag, and the height H_2 is caused by a relatively prolonged mixing of particles along the vertical direction[24] that occurs during the lifetime of an air mass.

It is clear that there is a possibility of using the integral characteristics of temperature in the low tropospheric layer as a parameter that is responsive to the intensity and duration of heating or cooling.

The next problem, that must be solved for parametrization of the relationship between the temperature regime of the atmosphere and the stratification of aerosol characteristics, is connected with the objective character of estimating the height of the mixing layer. The criterion for estimating H_2 based on finding the height of the break of correlation is proposed in Ref. 28. However, this approach has some disadvantages, since there is some arbitrariness in selecting the degree of the break of correlation; such a problem is related to the class of ill–posed ones.[29]

To improve this approach, let us analyze the matrices for season ensembles (Fig. 5). Let us take the height where the correlation coefficient $r(\sigma_0, \sigma_H)$ reaches the value of 0.5 as a criterion for the estimate of the sought height and designate it H_c.

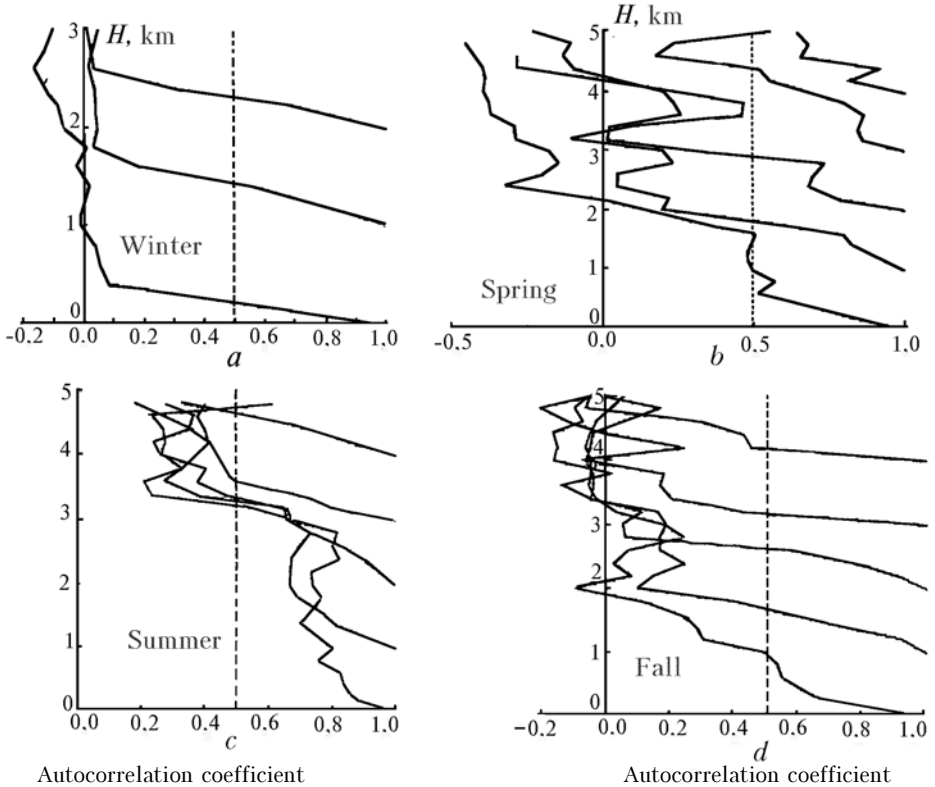

FIG. 5.

As is seen from Fig. 5 (it is especially well seen in the correlation matrix for the summer season), near the height where $r(\sigma_0, \sigma_H)$ reaches the value of 0.5 the closeness of lines is observed, i.e., within this layer the values $\sigma_d(H)$ are correlated with each other at all heights. Whichever the initial height we take, $r(\sigma_0, \sigma_H)$ reaches the value of 0.5 at the same level. Above this layer a sharp decrease of the correlation coefficient is observed ($r(\sigma_0, \sigma_H)$ decreases down to 0.5 in 200–500 m). The spring events continue to be the exception. Here one can select two regions of high correlation. They are up to ~ 1.5 km and above 3 km. This confirms the effect of remote sources on the formation of the aerosol vertical profile above the mixing layer in spring.

Based on the technique for determining H_c and on the character of correlation within this layer one should call H_c as the height of correlation layer.

To create the parametrization of the relationship between H_c and the integral heating of lower atmospheric layers we used the ensembles of observational data formed for seasons, months, and different air masses (24 ensembles on the whole). The autocorrelation matrices $r(\sigma_0, \sigma_H)$ were calculated for them as well as the average temperature $\overline{T}_i$ of the layers of 1, 2, 3, 4, and 5 km high.

The relationship between H_c and $\overline{T}_i$ for the layers of different height was determined in the form

$$\ln(H_c) = A_i \overline{T}_i + B_i \,. \qquad (3)$$

The dependence of the empirical coefficients A_i and B_i on the height of the layer of temperature averaging occurs to be close to a linear one, that allowed us to propose the following general dependence for estimating H_c:

$$H_c = 0.75 \exp\left[(4.5 \cdot 10^{-3}\, \tilde{H} + 6.5 \cdot 10^{-2})\, \overline{T}_{\tilde{H}} + 0.2\, \tilde{H}\right], \qquad (4)$$

where $\overline{T}_{\tilde{H}}$ is the average temperature of the atmospheric layer of the height $\tilde{H}$. The rms error of determining H_c by this formula occurs to be ~50 m (that gives the error ~ 20% for winter and ~ 5% for summer).

Using Eq. (4) we estimated the annual behavior of the height of correlation layer for the region under investigation (the annual behavior constructed using our bulk of data is shown in Fig. 1). To calculate H_c we used the data on temperature obtained from aerological sounding of the atmosphere at a station in West Siberia.[30] The height H_c was determined from the average temperature $\overline{T}_3$ of the lower 3–km atmospheric layer (up to the pressure surface $P = 700$ mbar). Annual behavior of the correlation layer height for the region under investigation is shown in Fig. 6*a*.

The rms deviation of the values $\overline{T}_3$ caused by the spatial variability between different stations does not exceed 2°C for each month. To analyze the interannual temperature variations, the data on average monthly temperature in Tomsk during the period 1972–1992 were used (the data were kindly presented by A.I. Kuskov). The interannual scatter of $\overline{T}_3$ occurs to be ~ 5°C in winter and ~ 2°C in summer. Summing the variances of the spatial and interannual variations of $\overline{T}_3$ and adding the error of estimating by the approximation formula (4) we have estimated the variations of the annual behavior of the correlation layer for West Siberia region. The rms deviation ΔH_c is about 40% for winter and 20–25% for summer.

To analyze the role of the mixing layer (correlation layer in our case) in forming the vertical aerosol profile, the annual behavior of the relative content of submicron particles within and above this layer is shown in Fig. 6*b*. These curves are descriptive of the possibility of using the data on aerosol characteristics in the near–ground layer for retrieving their vertical profile using the developed empirical models.

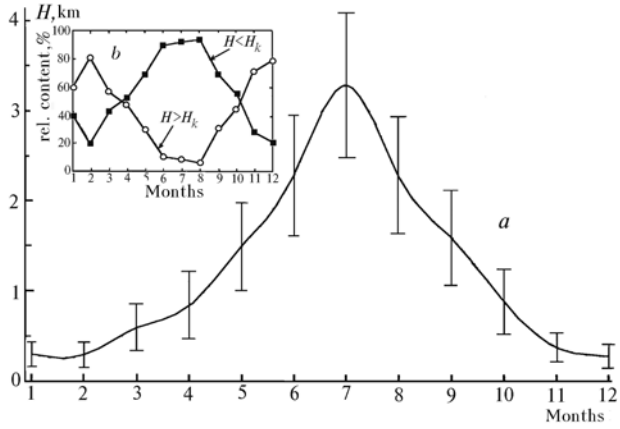

FIG. 6.

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