

EMPIRICAL MODEL OF THE AEROSOL OPTICAL PROPERTIES IN THE TROPOSPHERE OVER WEST SIBERIA

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This paper concludes a series of publications (see Refs. 1 to 5) that deal with the construction of an empirical model of the aerosol optical characteristics in the visible and near IR regions for the low troposphere. The model proposed provides for reconstruction of the aerosol scattering coefficient profile. In so doing one needs for, as input data, the scattering coefficient value of the dry aerosol substance at the near ground level measured at the wavelength of 0.52 μm , relative air humidity, aerosol optical thickness, and mean temperature of air in the low troposphere. To estimate the entire set of optical characteristics, we suggest to use, as the first order approximation, a single parameter model of the near ground aerosol with the relevant corrections for the vertical profile $\sigma(H)$.

INTRODUCTION

The empirical model of the aerosol optical properties proposed is based on the data of airborne sounding of the troposphere over West Siberia. It is known that the basic principles of any model are closely related to the model purpose and depend on the bulk and quality of information, experimental or theoretical, available.

Based on the practice of nowadays aerosol studies we isolate two basic approaches to the development of empirical aerosol models that may conditionally be called the optical and microphysical ones.⁶ Naturally each of the approaches has its own advantages and drawbacks. Thus, the microphysical approach provides for a possibility of calculating any of the optical parameter of aerosol using some theoretical grounds. However, in this case the question on possible errors in such estimations remains to be addressed. Actually, when setting empirically the size-distribution function of aerosol particles and optical constants of the particulate matter, what is a difficult task alone, it is hard to assess the contribution coming to the aerosol optical parameters to be calculated from the particles that have not been counted because of some instrumental limitations. Those particles are, as a rule, from the most fine aerosol (nucleation) mode as well as a significant part of the coarse aerosol fraction.

The optical approach is, in principle, free of these problems, but, at the same time, it is restricted only by the optical parameters measured and the spectral region used in the experiments. In the case of near-ground atmospheric hazes this type of limitations existing in the optical method has essentially been overcome by

using the method of microphysical extrapolation.⁷ That enables the extension of the applicability limits of this technique over the entire visible region.⁶

The development of few-parameter models of the near-ground atmospheric hazes and their microphysical extrapolation^{6,8} have closed a very important and fruitful stage in the studies of atmospheric aerosols. However, no proper attention has been paid to this approach in the practice of modern atmospheric modeling. For this reason this technique is not so widely spread in practice as it deserves to be. In our opinion this is most likely due to poor practicality of such models and insufficiently clear explanations the physical backgrounds of the models. May be a separate publication could help in resolving the latter circumstance.

Nevertheless, when planning a long-term and large-scale airborne experimental study of the tropospheric aerosol, we have incorporated the experience compiled from using a single-parameter model for interpreting the data on angular behavior of scattering properties of near-ground atmospheric hazes as well as of the variability regularities that follow from such a representation.^{6,8}

STRATEGY OF THE EXPERIMENT

Our approach is based on the following considerations.

1. Since the single-parameter model of the aerosol light scattering properties angular behavior has been constructed using the experimental data acquired at the wavelengths in the visible region, thus revealed regularities in the variability of aerosol properties are mainly caused by the *sub-micron aerosol particles*.

2. *The scattering coefficient* is one of the aerosol characteristics that is most *sensitive* to variations in microphysics of the sub-micron aerosol (that, according to G.V. Rosenberg⁹ are 'memorized' by the atmosphere).

3. The variations of the aerosol scattering coefficient in the atmosphere are governed by two basic processes: the first one is natural variability in the content of dry aerosol substance (or in other words by the processes of generation, aging, accumulation, and sinking of the particulate matter in the atmosphere), while the second one is the transformation of aerosol microphysical properties under the action of the air *relative humidity*. It is evident that those two types of processes are regulated by the geophysical factors of different spatiotemporal scales.

It follows from such an understanding of the aerosol-active atmospheric processes that in order to correctly assess the role of that or other atmospheric factor in the variability of aerosol properties and, as a consequence for a more efficient incorporation of those variations into the dynamic aerosol models one always must measure, in parallel, the characteristics of dry aerosol matter and its response to variations of the air relative humidity.

Of course, it would be an ideal situation if one can measure angular behavior of the scattering phase matrix elements in different spectral regions, but unfortunately we had no sufficient funding and equipment to design such an instrument that could, in addition, withstand the long-term exploitation onboard an aircraft. Therefore, in our studies of the aerosol scattering coefficient by the nephelometric method, we have used an airborne instrument equipped with an attachment capable of artificially regulating the relative humidity of air samples and heating the aerosol particles (for details see Ref. 1).

BRIEF ANALYSIS OF THE EXPERIMENTAL DATA SET

When flying over West Siberia, we have compiled an array of 602 vertical profiles acquired during measurement campaigns in different seasons and under different meteorological and synoptic situations. The data acquired over cities and in the regions nearby big industrialized centers have been excluded from thus formed data array. Then the data array formed has been divided into seasonal sub-arrays according to climatic criteria of seasons¹⁰ characteristic of the region under study. For each season we have analyzed the frequency of occurrence of different situations depending on type of air mass and pressure system. As analysis has shown, the frequencies of occurrence obtained well agree with the data of many-year observations. In a similar way we have also compared the vertical profiles of the meteorological quantities measured with the data on many-year mean profiles measured at the aerological network.¹

The agreement of synoptic and meteorological characteristics of the atmosphere during the observation period with the climatic mean ones enables us to consider the array of data on aerosol properties compiled to be representative of this geographical region as well.

SELECTION OF THE INPUT PARAMETERS

It is evident that the models (that are inherently statistical) describe only most common properties of the aerosol particles as well as of their variability under the action of different atmospheric factors while washing out those that manifest themselves in every particular case. As a result two basic aspects of using such models in practice become quite clear. The first one is that those models can not be used when solving the problems that require highly accurate description of the aerosol optical characteristics in any particular atmospheric situation. Second, these models may only be few-parameter ones since the variability of the aerosol optical characteristics that is memorized by a model is capable of representing only the most frequent situations that occur under the action of the variety of external aerosol and weather factors which, in their turn, are interrelated in a complex way.

Apparently, the construction of a versatile, independent of seasons, model of optical properties of aerosol is unrealistic because it is an *a priori* understandable fact that the state of the underlying surface changes during a year as well as the sources of aerosol particles and their power also vary together with the capacity of sinks for aerosol particles.

Based on these considerations we took as the first and the basic one the natural parameterization principle of dividing the data arrays into seasonal sub-arrays. Further analysis of the data available showed that no additional subdivision, according to calendar principle, into monthly sub-arrays is useful. The matter is that even similar synoptic and weather situations in the region under study undergo significant variations from year to year.¹⁰ It is worth mentioning in this connection that the time of covering the underlying surface with snow and rivers with ice as well as the reverse processes occur in different calendar times every year.

In figure 1 we present, for illustration, vertical profiles of the scattering coefficient values of the dry aerosol matter acquired in two seasons. It is explicitly seen from this figure that variations of this quantity are very wide even during same season.

So, one may readily conclude that even a season mean model can hardly provide for any acceptable for practice accuracy of estimations.

As analysis of the within the season factors causing the variability of the scattering coefficient of the dry aerosol matter showed, for the submicron aerosol fraction, the account for types of air masses and meteorological parameters enables one use these factors for the model parameterization.²⁻⁴

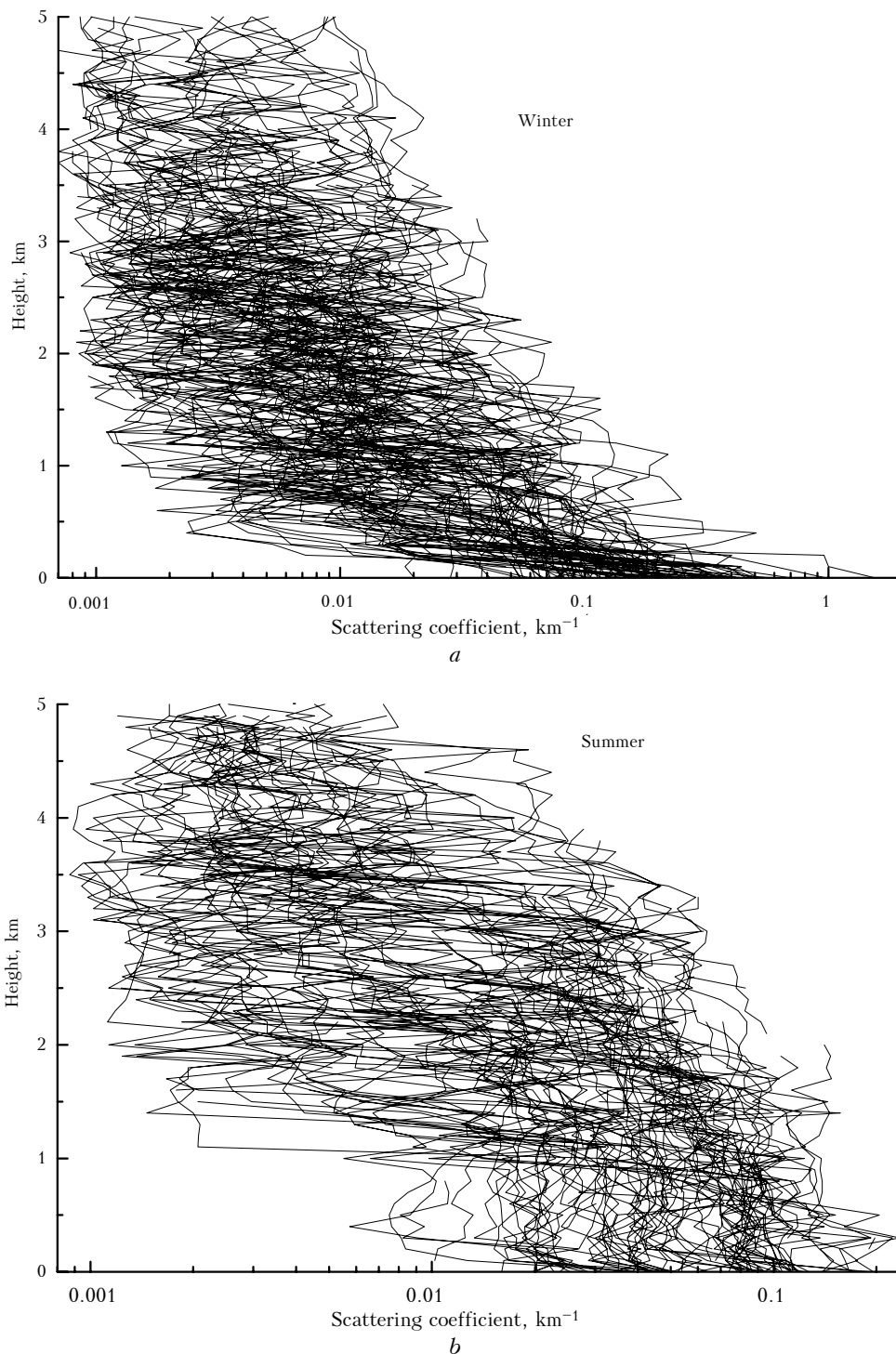


FIG. 1. Examples of vertical profiles of the "dry" scattering coefficient of aerosol.

However, we should like to underline once more that the experience of aerosol studies compiled up to now (see, for example, Ref. 11) shows that the attempts to construct dynamic models of the aerosol optical or microphysical parameters that use, as the input parameters, only synoptic or meteorological quantities are yet too problematic.

Actually, the 'aerosol' weather at a concrete point of observations and moment is determined

not only by synoptic and meteorological factors, but also by the whole complex of global and local, *inherently aerosol*, factors of natural and anthropogenic origin.¹¹

The development of an aerosol model that could provide for the account and forecasting of the whole variety of factors, though being much promising, would require many-year experiments and a wide measurement network for monitoring a large number of atmospheric

parameters including the aerosol ones as well as of advanced climate and weather models.

From this point of view, it seems to be reasonable that the development of an aerosol model uses, at the first stages, as most important input parameters, those aerosol parameters that are measured *in situ* and which bear qualitative and quantitative information on the state of a particular aerosol.

In principle, among the great variety of optical or microphysical parameters of aerosol that may be measured or calculated, as in the case of climatic modeling, and then used as the input data one can conditionally isolate two groups of characteristics. One group could involve the data on aerosol in the near-ground atmospheric layer while the other one those acquired with the ground-based systems on vertical profiles or on the total optical thickness. This question has been addressed in Ref. 5 in a more detail, so in this paper we only note that at this stage of the model approbation we have chosen only those aerosol parameters that are easy to measure.

The parameters from the first “near-groundB group are the scattering coefficients (“moistB that means measured *in situ* or “dryB or reduced to zero relative humidity), temperature, and relative humidity of the air.

As to the group of data that may conditionally be called “soundingB data group we have considered the possibility of including there the data on vertical profiles of meteorological quantities and data acquired with sun photometers on the aerosol optical thickness.

ANALYSIS OF THE RECONSTRUCTION ERRORS

In Ref. 5 one can find a detailed description of the calculation scheme for reconstructing vertical profiles of the aerosol scattering coefficient. Therefore we present here only some concrete empirical parameters of the model that we have used when reconstructing the profiles and illustrate the practicability of the model.

So, if we have data on the near-ground scattering coefficient value σ , measured *in situ*, or on its value σ_d , reduced to zero relative humidity of the air, then the reconstruction of the profile starts with the value $\sigma_d(0)$. In the case when only σ values are available we first have to calculate $\sigma_d(0)$ values using Kasten-Haenel formula

$$\sigma = \sigma_d (1 - R)^{-\gamma}, \tag{1}$$

where R is the relative humidity of air and γ is the parameter of condensation activity.

Next, the profile $\sigma_d(H)$ is being reconstructed using the empirical linear equations of the following form:

$$\sigma_d(H) = K(H) \sigma_d(0) + C(H), \tag{2}$$

where $K(H)$ and $C(H)$ are empirical coefficients for the relevant, according to external factors, data array.

Table I gives the values of the empirical coefficients of the regression equation (2) for different seasons.

If, additionally, the data on temperature profile of air are also available, then we calculate the height of the mixing layer by formula proposed in Ref. 2, and use it to correct the coefficients $K(H)$ and $C(H)$ in the empirical equation (2) and, finally, reconstruct the profile $\sigma_d(H)$ using already thus corrected equation.

TA” LE I. Coefficients of the regression equation.

Winter			Spring			Summer			Fall		
H, km	K(H)	C(H)	H, km	K(H)	C(H)	H, km	K(H)	C(H)	H, km	K(H)	C(H)
0	1	0	0	1	0	0	1	0	0	1	0
0.4	0	0.072	0.4	0.57	0.024	0.6	0.58	0.011	0.4	0.32	0.016
1.2	0	0.028	1.4	0.23	0.014	2.8	0.30	-0.0054	1.2	0.10	0.022
2.6	0	0.0077	2.4	-0.06	0.026	3.4	0.081	0.003	2.4	0	0.0065
5	0	0.004	5	-0.094	0.024	5	0	0.0026	5	0	0.0028

Then the scattering coefficient values calculated for the dry aerosol base are reduced to relative humidity values at the corresponding height by formula (1). There are three options to do this. If no information on $R(H)$ profiles is available one may use the corresponding season mean profile. In the case when a researcher has at his disposal only the data on near-ground values of the relative air humidity then it is possible to reconstruct, using some empirical

relations like equation (2), the $R(H)$ profile by making use of the correlation between $R(H)$ and $R(0)$. And, finally, if data on the $R(H)$ profile are available, for instance as measured with a radiosonde, one uses the true profile of the relative air humidity.

At the final stage the profile of scattering coefficient is to be corrected using the optical thickness value according to the technique proposed in Ref. 5.

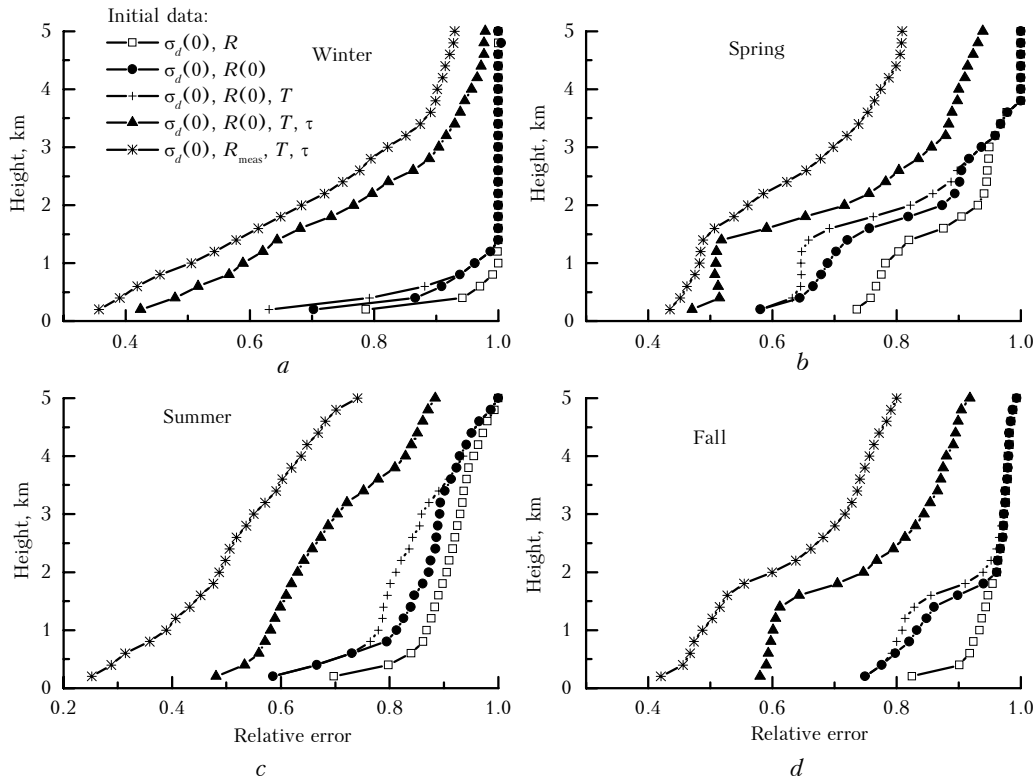


FIG. 2. Ratio of the rms error of reconstruction to the rms deviation of the initial data set.

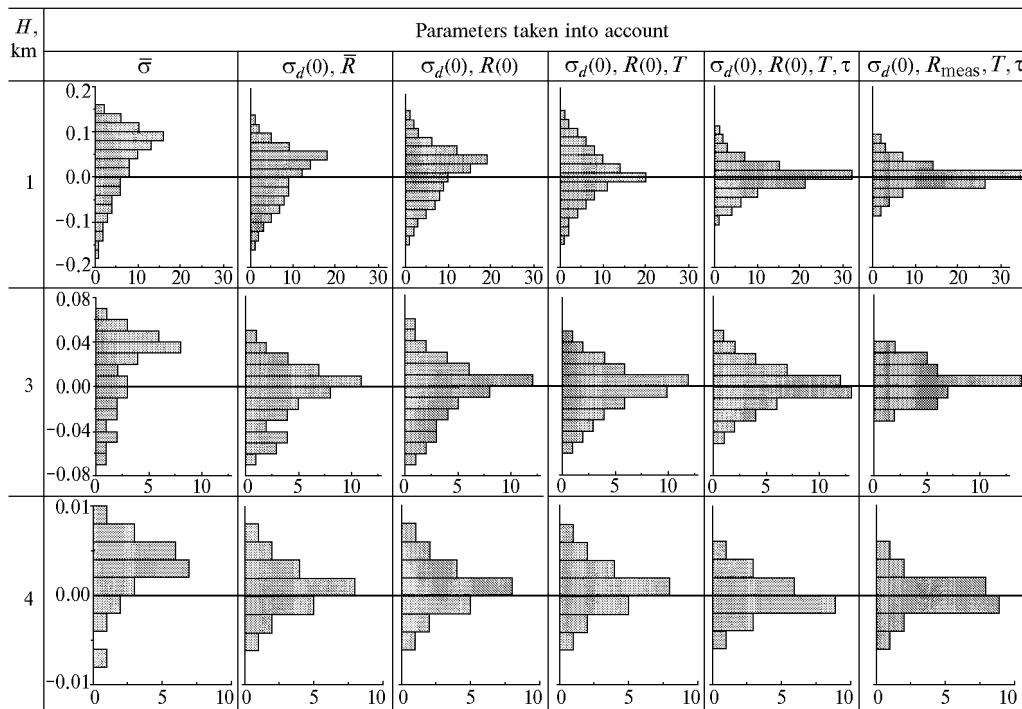


FIG. 3. Histograms of the reconstruction errors.

Figure 2 presents the ratio profiles between the rms error of $\sigma(H)$ reconstruction and the rms deviation of the initial data array for each season.

The profiles of this ratio are shown in a succession of the increasing number of input

parameters. We consider the following versions of the reconstruction scheme. 1) The first one is in reconstructing the profile of dry aerosol base using a near-ground value $\sigma_d(0)$ with the following ‘moistening’ using a season mean profile of relative

humidity $\bar{R}(H)$. 2) The second way assumes reconstructing 'dry' aerosol profiles and the profile of relative humidity via the respective near-ground values. 3) The third option assumes, in addition to the procedure under previous point, the account for the mixing layer height based on mean temperature of the layer from the ground and up 3 km height. 4) In the fourth version the aerosol optical thickness is being taken into account, besides the factors mentioned in the third version. 5) The reconstruction scheme in this version is the same as the previous one except that 'moistening' of the "dry" scattering coefficients is performed using measured profile of the relative humidity, $R_{\text{meas}}(H)$.

To illustrate the distribution of the reconstruction errors, we show in Fig. 3 the histograms of these errors that are characteristic of summer season. For this illustration we have chosen the atmospheric layers at three specific heights.

The layer at 1 km height is taken because it is within the mixing layer, the layer at 3 km is at the height of season mean upper boundary of the mixing layer, and the layer at 5 km height is already in the free atmosphere. Histograms that are shown in the first column represent the errors when no measurement data are used and the season mean profile $\sigma(H)$ is taken as the reconstructed one. Other columns show the error histograms for the $\sigma(H)$ reconstruction versions used to obtain data shown in Fig. 2. It is seen from Fig. 3 that at all heights an increase in the number of input parameters used in reconstruction results in a narrower distribution of the reconstruction errors that becomes more close to the normal one. If reconstruction of the profile is being performed using only one input parameter, namely the near-ground value of the scattering coefficient, we obtain a very asymmetric histogram of errors at the height within the mixing layer. Moreover, at the height near the mixing layer top the error distribution may even become a bimodal one. By introducing into the reconstruction scheme the mean temperature of the low atmospheric layers or the temperature of the near-ground layer, one may calculate the mixing layer height for each individual profile. As a result, the rms error of reconstruction not only falls off but, in addition, becomes closer to the normal view. An increase in the accuracy of reconstructing the aerosol scattering coefficient at the heights in the free atmosphere can only be achieved when taking into account the aerosol optical thickness. Similar behavior of the reconstruction error with the increasing number of input parameters may be demonstrated for other seasons.

Of course, it is quite clear that the errors of reconstruction are too high and, as a consequence, thus reconstructed aerosol characteristics can hardly be used for some operative and accurate assessments. Moreover, the absence of knowledge of the condensation activity of the aerosol at different heights and at any concrete time is one more source of errors that may be essential in magnitude. In this study we have used only its

average value $\gamma = 0.5$. As our earlier airborne studies have shown, the value γ may vary from case to case having also certain seasonal peculiarities and some vertical behavior.¹² However, the bulk of experimental material compiled up to now on the behavior of this parameter is yet insufficient for reliably parameterizing it and thus we could not involve it into our scheme of reconstruction.

At the same time, it is clearly seen that even at this stage of the model development this approach enables reconstructing the aerosol scattering coefficient in the height region from 0 to 5 km with the accuracy that can hardly be achieved, at the present time, by other models currently in use. Thus, for summer conditions the use of a scheme that accounts for the near-ground values $\sigma_d(0)$, temperature profile $T(H)$, and the aerosol optical thickness τ allows the uncertainty in $\sigma(H)$ estimates to be decreased by two to three times, as compared to the case of using the season mean value $\bar{\sigma}(H)$.

POSSIBILITY OF RECONSTRUCTING SPECTRAL AND ANGULAR BEHAVIOR OF SCATTERED RADIATION

As has already been mentioned, the scattering coefficient is the aerosol characteristic that is most sensitive to variations in the microphysical properties of the sub-micron aerosol. For this reason it is often used as an input parameter in single-parameter models of the angular characteristics of scattered radiation and of spectral behavior of the extinction coefficients of the near-ground hazes.^{6,8} One can not directly make use of data on the aerosol scattering coefficient for reconstructing vertical profiles of other optical characteristics of aerosol. The first reason is that the amount of aerosol particles decreases, as a rule, with increasing height what requires that the coefficients entering the empirical relations of the scattering coefficient to optical parameters derived based on the near-ground measurements ought to be corrected in some way. This particular problem is quite simply resolved in our model since one of its stages of the profile reconstruction assumes reconstruction of this profile for 'dry' aerosol that exactly corresponds to the fall off of the amount of particles with height.

Secondly, and it is most important, one ought to be sure that no significant variations in the aerosol microphysical composition occur with the increasing height. Otherwise the empirical relations obtained based on a single-parameter model may not be used for making any estimations. To check up the possibility of using a single-parameter model of the near-ground aerosol through the whole height region from 0 to 5 km we have undertaken statistical analysis of experimentally measured particle size-distribution functions. Those size distributions were measured, with a photoelectric particle counter, in parallel with the measurements of the aerosol scattering coefficient

during an airborne experiment. Measurements of the aerosol microstructure have been carried out by the research group headed by B.D. Belan and kindly presented to authors for making analysis. We have

analyzed the profiles of 12 aerosol fractions with the particle diameters varying from 0.6 to 10 μm . Altogether we have analyzed more than 5000 aerosol size spectra acquired over West Siberia.

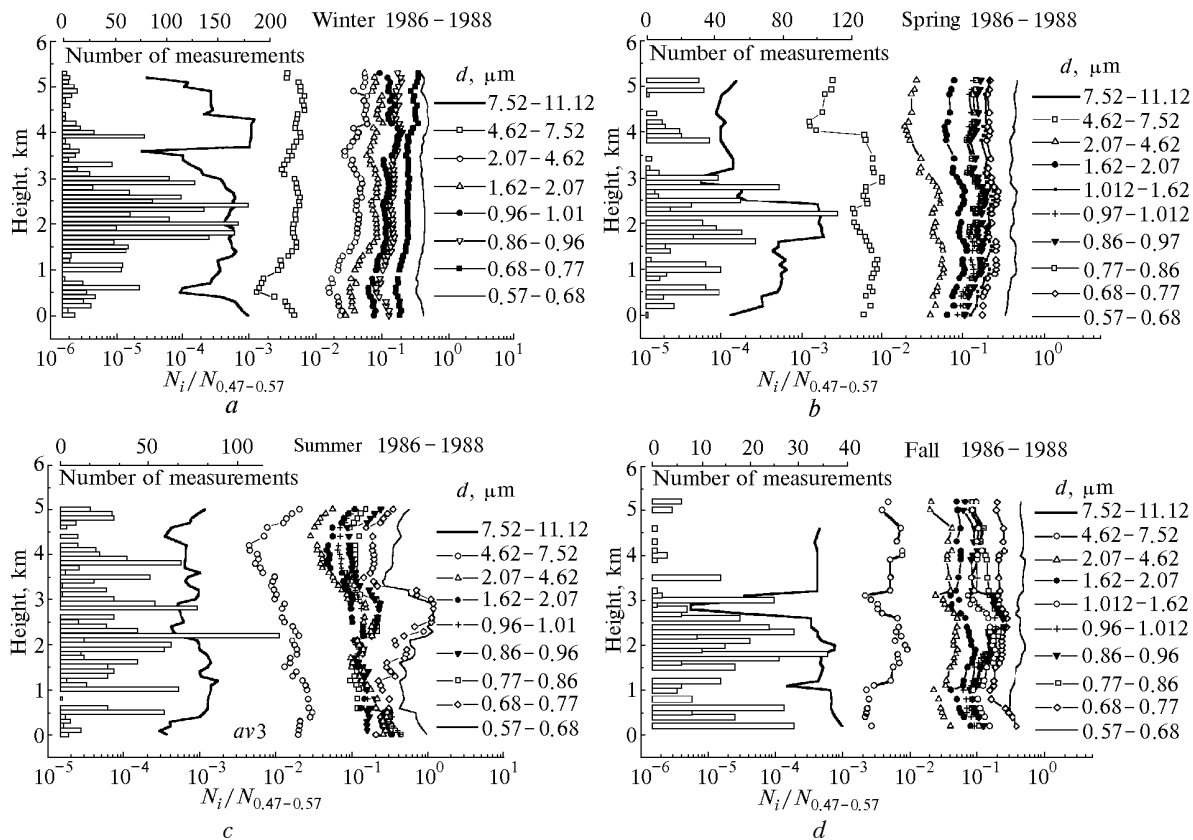


FIG. 4. Relative concentrations of isolated aerosol fractions.

Set out in Fig. 4 are the concentrations of different fractions normalized to the concentration of particles counted with the counter in its first measurement channel. The data presented in this figure are grouped from the data sets acquired in 1986-1988 according to seasons. The curves presented in the figure have been obtained by sliding average over three points. The size ranges shown in the figure have been calculated with the account for the refractive index of calibration particles and of those sampled in the atmosphere.¹³ The columnar histograms in the left parts of figures show the number of measurements made at each height.

As is seen from the figure, relative contributions of particles, that are measurable with the particle counter, to the resulting size spectrum only weakly, in the first approximation, varies with height. It is only in summer when one can reliably discern an enhanced contribution to the size spectrum of aerosol that comes from particles of 0.6 to 0.8 μm at the heights about the mixing layer top (from 1.5 to 3 km).

As the Mie calculations of the angular behavior of the scattering phase matrix elements showed, one can neglect, in the first approximation, the influence of variations in the size spectra on the aerosol scattering

properties at least for scattering angles outside the aureole. It is just the range of scattering angles for which the single-parameter model of near-ground hazes has been developed.⁶ Hence, if a moderate accuracy of reconstruction may be accepted one certainly can use the single-parameter model of aerosol while having in mind only visible wavelengths. It is especially true if one reminds that the accuracy of reconstructing the input parameter, in this case it is the scattering coefficient, is not very high.

At the same time we consider it to be too problematic to extend the applicability limits of this approach to a wider spectral range.

As the calculations made have shown, no correct extrapolation of the particle size-distribution functions outside the measurement range is possible even if the array of measurement data on aerosol microstructure acquired with a photoelectric particle counter is statistically full. The variations in the size spectra of particles with radii over several microns are especially large that makes the estimation of aerosol optical properties at the wavelengths over 1 micron practically impossible. As a result, the model we have proposed may only be used in the visible and near IR regions.

More detailed analysis of the aerosol microstructure peculiarities that may occur at the heights from 0 to 5 km in the atmosphere is out of the scope of this paper and may be a subject for a separate discussion. In this particular presentation we restrict ourselves by a conclusion that fortunately the absence of sharp changes in the size spectra of aerosol particles with height enables us to rely on the possibility of using the single-parameter representations for making, though rough, estimates of the angular behavior of radiation scattered by aerosol as well as of spectral features in the extinction coefficient values in the visible wavelength region.

So, according to the approach discussed, making corrections to the single-parameter model of the near-ground hazes is based on the assumption that only the content of dry aerosol substance varies with height while the size-distribution function and physicochemical properties of the particulate matter keep approximately unchanged. From this conclusion it naturally follows that the variability of normalized optical characteristics of aerosol is mainly determined by the ratio between the volumes of dry substance and water in the content of aerosol particles. In this case the entrance to a single-parameter model may be performed through an empirical scheme. Such a scheme assumes that first one reconstructs the value of "dry scattering coefficient at a given height. Then, using a particular value of the relative air humidity one calculates the *in situ* value of the scattering coefficient and after that determines the dry substance-to-water ratio. Using thus found ratio between the volumes of dry substance and water one may reconstruct based on the single-parameter model of near-ground atmospheric hazes all other optical characteristics needed as normalized to the scattering coefficient. Then one relates the absolute values to those of the scattering coefficient reconstructed for the given height.

It is possible, in this case, to enter the single-parameter model through the following empirical relation¹⁴:

$$\log \sigma_{\text{sing}} = 2.8 \gamma (1 - R) - 1.61. \quad (3)$$

The value σ_{sing} in this formula is to be considered as the input parameter to the single-parameter model; γ is the parameter of condensation activity occurring at the moment when a particular $\sigma(H)$ profile is to be reconstructed; R is the relative air humidity.

The process of entering the single-parameter model is as follows. Having reconstructed the value σ_d at some height H one finds, after moistening by formula (1), the profile $\sigma(H)$ and then determines using expression (3) the parameter σ_{sing} .

Having thus found the value σ_{sing} , one reconstructs the components of the reduced scattering phase matrix and scattering phase function $f(\varphi)$ (the values of the empirical coefficients of the single-parameter model may be found in Ref. 6). The absolute values of the angular characteristics of light scattering are then calculated via the scattering coefficient reconstructed for the given altitude, that is $\sigma(H)$.

CONCLUSION

Based on data of airborne sounding of the atmosphere we have developed an empirical model of optical properties of aerosol in the low troposphere over West Siberia. Since in so doing we have revealed certain leading factors that cause the variability of aerosol particles content along vertical direction, the applicability of this approach most likely is not bounded by the West Siberian region only. In our opinion the basic empirical relationships proposed in this paper for estimating the optical characteristics of aerosol can be used to interpret data acquired over similar geographic regions.

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