ON THE CONTRIBUTIONS OF DISPERSE FRACTIONS OF THE NEAR GROUND HAZE TO THE EXTINCTION OF VISIBLE AND IR RADIATION

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Using several arrays of the aerosol extinction coefficients $\alpha(\lambda)$ as an example, it is shown that the coefficients $\alpha(\lambda)$ can be statistically separated into components caused by particles of different size in a wide spectral range. For separation, the apparatus of linear multivariate regression analysis is applied. It is shown, in particular, that the component of $\alpha(\lambda)$ caused by particles of the fine-disperse fraction not only contributes in the visible spectral range (due to scattering), but also shows itself in the infrared spectral range (due to absorption). This approach allows statistical detection of the weak atmospheric absorption in the spectral range $\lambda > 9$ μm in the real atmosphere. The character of the spectral dependence suggests that it is caused by radiation absorption by small water particles, which are optically inactive in the visible spectral range.

1. When studying regularities in the behavior of spectral coefficients of aerosol extinction $\alpha(\lambda)$ by atmospheric hazes in the visible and IR spectral ranges is of primary importance to provide for adequate physical interpretation of the $\alpha(\lambda)$ dynamics of both the spectral structure and magnitude under varying environmental conditions. Such an interpretation is very complicated, because the aerosol extinction coefficients measured in the atmosphere are caused by joint effect of particles of different size, different composition, that differently respond to changes in the environmental conditions. Consequently, to solve this problem, it is necessary to estimate, in some way, separate contributions from particles of different size into the total aerosol extinction in the wide spectral range. Having this problem solved, we could study the optical response of a separate fraction of particles to changes in the meteorological conditions or some other factors.

To calculate the components of $\alpha(\lambda)$ caused by particles of different size, representative information is normally needed about the particle size distribution function of aerosols, as well as an appropriate model of optical constants of aerosol in a given spectral range. The particle size distribution function, in its turn, can be found directly from microphysical measurements or from solution of an inverse problem based on measured aerosol extinction coefficients in the visible and IR spectral ranges. The general drawback of this approach is a subjective choice of the optical constants of aerosol, as well as errors inevitably arising in the particle size distribution function obtained from both the microphysical measurements and from a solution of the inverse problem based on optical data.

In this paper we consider the possibility of applying statistical approach to estimation of separate contributions from particles of different size into the total aerosol extinction of optical radiation. This approach is based on application of the apparatus of multivariate linear regression to the array of $\alpha(\lambda)$ coefficients measured in a wide spectral range. The possibility of applying this approach in the 0.5–4 μm wavelength range has been proved in Ref. 1. Figure 1 demonstrates the result of separation of the total aerosol extinction coefficient of a coastal haze (curve *1*) into the submicron (curves *2* and *2′*) and coarse-disperse (curves *3* and *3′*) components. The separation was performed using different methods. Curves *2* and *3* were calculated by statistical method, while curves *2′* and *3′* were calculated using Mie formulas based on the microphysical data obtained from inversion of curve *1*. Good agreement between these curves allows the conclusion to be drawn that the statistical approach is applicable to separation of the coefficient $\alpha(\lambda)$ into the above-indicated components.

2. Let us consider the potentialities of the discussed method in a wider spectral range (λ = 0.44–12 μm) using, as an example, the array of the aerosol extinction coefficients measured in the atmospheric hazes of the Crimean Coast of the Black Sea.[2] The corresponding spectral measurements of the atmospheric transmittance were conducted along a 3800-m long path above the sea surface. A total of 168 spectra of the coefficients $\alpha(\lambda)$ has been recorded. The average values of atmospheric meteorological parameters (with their rms deviations) were the following: the air temperature $\bar{t} = (+20 \pm 3.3)°$q, the partial pressure of

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water vapor $\overline{e} = (17.4 \pm 4.1)$ mbar, the relative air humidity $\overline{f} = (73.6 \pm 12.4)\%$, the aerosol extinction coefficient in the visible spectral range $\overline{\alpha}(0.55) = (0.235 \pm 0.152)$ km^{-1}. The average spectral dependence of the coefficients $\alpha(\lambda)$ for this array of data is shown in Fig. 2 (curve *1*). As is seen, it monotonically decreases with increasing wavelength in the range $\lambda = 0.48 - 3.9$ μm, then it is almost constant while exhibiting a slight increase at $\lambda > 10$ μm.

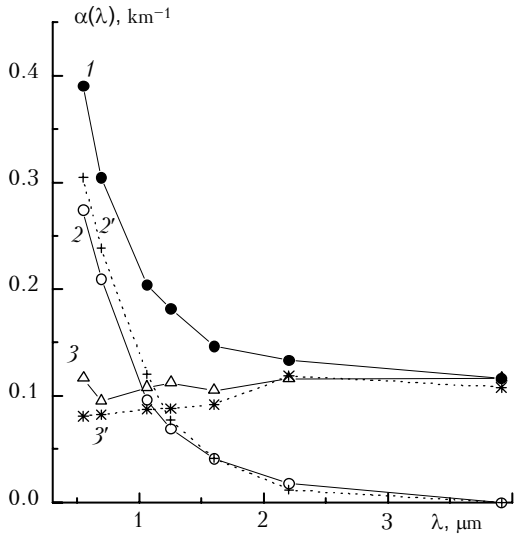

FIG. 1. Comparison of two methods for separation of the spectral coefficients of the total aerosol extinction in the atmospheric haze (curve 1) into the submicron (curves 2 and 2′) and coarse-disperse components (curves 3 and 3′). Curves 2 and 3 are obtained by use of statistical method; curves 2′ and 3′ are calculated by the Mie formulas based on the microphysical data obtained from inversion of curve 1 (Ref. 1).

Such a spectral behavior of the coefficients $\alpha(\lambda)$ suggests that the total aerosol extinction in this case is the combination of two main components: one of them determines the spectral dependence in the short-wave part of the spectrum, while another is responsible for the wavelength-independent level of the aerosol background. It is natural to suppose that the former is determined by radiation absorption and scattering on submicron aerosol particles. The parameter $\Delta\alpha = [\alpha(0.48) - \alpha(3.9)]$, in km^{-1}, can be chosen as its numerical measure. The latter is determined by either radiation scattering on giant particles or absorption by fine-disperse soot aerosol, which does not manifest itself in scattering. The coefficient of aerosol extinction $\alpha(3.9)$, in km^{-1}, at the wavelength of 3.9 μm can serve as a numerical measure for this component. With the values of $\Delta\alpha$ and $\alpha(3.9)$ as input parameters of the regression equation, this equation can be presented as

$$\alpha(\lambda) = K_0(\lambda) + K_1(\lambda)\,\Delta\alpha + K_2(\lambda)\,\alpha(3.9)\,, \qquad (1)$$

where $K_0(\lambda)$, $K_1(\lambda)$, and $K_2(\lambda)$ are the spectral coefficients of regression, which are calculated with known variances of the parameters $\alpha(\lambda)$, $\Delta\alpha$, and $\alpha(3.9)$ and the coefficients of correlation between them $\rho_{\alpha_\lambda,\Delta\alpha}$, $\rho_{\alpha_\lambda,\alpha_{3,9}}$, and $\rho_{\Delta\alpha,\alpha_{3,9}}$. The obtained values of the coefficients $K_0(\lambda)$, $K_1(\lambda)$, and $K_2(\lambda)$ are presented in Table I. Note that Eq. (1) and the Table I comprise the empirical model. This model enables one, first, to reconstruct the whole spectrum of the coefficients $\alpha(\lambda)$ from only two measured aerosol extinction coefficients $\alpha(0.48)$ and $\alpha(3.9)$. Second, using this model and having measured the coefficients $\alpha(\lambda)$ in a wide wavelength range, one can separate any of $\alpha_i(\lambda)$ from this array into components.

TABLE I. Spectral coefficients of regression for Eq. (1).

λ, μm	$K_0(\lambda)$, km^{-1}	$K_1(\lambda)$	$K_2(\lambda)$
0.44	0.001	0.973	0.94
0.48	0	1	1
0.55	0	0.842	0.885
0.69	-0.001	0.585	0.781
0.84	−0.003	0.371	0.745
1.06	−0.005	0.249	0.909
1.25	−0.005	0.193	0.933
1.6	−0.003	0.107	0.931
2.2	−0.001	0.05	0.97
3.9	0	0	1
4.6	0	0.044	0.961
8.1	0.003	0.029	0.912
8.3	0.002	0.036	0.871
8.6	0	0.041	0.909
9.15	0	0.063	0.909
10.2	0.005	0.051	0.87
10.7	0.008	0.069	0.857
11.1	0.012	0.08	0.805
11.6	0.02	0.074	0.906
11.9	0.023	0.123	0.816

With such input parameters, the component $K_1(\lambda)\Delta\alpha$ determines the contribution of the submicron fraction into the aerosol extinction of radiation, and $K_2(\lambda)\alpha(3.9)$ determines the value of the aerosol background. Note that the component $K_0(\lambda)$ includes, by definition, a part of $\alpha(\lambda)$ which does not correlate with the input parameters of Eq. (1). The calculated results for these components are also presented in Fig. 2 (curves *2*, *3*, and *4*, respectively).

As seen, the component of $\alpha(\lambda)$, due to fine-disperse aerosol component, dominant in the visible spectral range, rapidly decreases with the increasing wavelength and reaches minimum ~0.01 km^{-1} near $\lambda = 2.2$ μm. Then it is almost unchanged up to $\lambda = 8.6$ μm. Such a level of extinction and its spectral independence agree well with the conclusions drawn in Refs. 3–5, in which it is shown that the extinction is likely to depend on the radiation absorption by the soot aerosol. In the range of $\lambda = 8.6 - 12$ μm, the contribution of the fine-disperse fraction increases

somewhat. Note that in this spectral range, curve *2* has two weakly pronounced maxima (insertion in Fig. 2). Since the fine-disperse aerosol practically does not scatter radiation in this wavelength range, the detected spectral structure should be interpreted as a manifestation of the absorption bands of a substance of small particles. Thus, for example, the maximum near $\lambda = 9.2$ μm most probably corresponds to the well-known absorption band of sulfates, while the maximum near $\lambda = 11.1$ μm corresponds to the absorption band of aluminum oxides or carbonates.[6] The general growth of the coefficients near $\lambda > 10$ μm agrees well with the spectral behavior of light absorption in liquid water.[7] The close correspondence of the absorption bands of the aerosol substance for a coastal haze[2] and continental haze[6] confirms the thesis about global abundance of the fine-disperse aerosol.

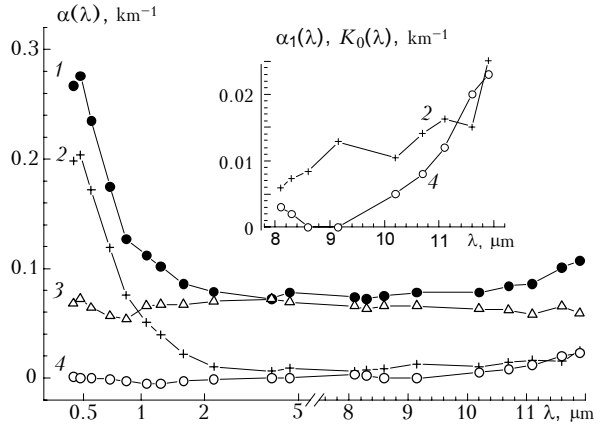

FIG. 2. Mean spectral dependence of the total aerosol extinction coefficients for coastal hazes over Black Sea (curve 1), its component due to submicron aerosol (curve 2) and the coarse-disperse aerosol (curve 3) calculated by the two-parametric model given by Eq. (1). Curve 4 is for the component K_0, which does not correlate with the input parameters of the model.

The spectral dependence of the component associated with the aerosol background (curve *3*) proved to be practically neutral over the entire wavelength range. Its contribution dominates in the region from 2 to 12 μm, being about 0.06 – 0.07 km^{-1} in the absolute value. Such a high level of the aerosol background and the absence of pronounced absorption bands suggest that in this case it is caused mainly by scattering of radiation on the coarse-disperse aerosol, rather than by absorption by particles of the fine-disperse fraction. This result agrees well with numerous data of investigations into the particle size distributions in the marine atmosphere, which is characterized by the enhanced concentration of the coarse-disperse aerosol.

As to the component $K_0(\lambda)$ (curve *4*), we expected that it will be close to zero in the entire wavelength range. However, the calculations have shown that it is non-zero, and its contribution is significant in the spectral range $\lambda > 9$ μm (see curve *4* in the insertion in Fig. 2). Judging from the character of the spectral dependence of this component, which coincides qualitatively with that of the absorption coefficient of liquid water, we can assume that it is caused by absorption of radiation by very small water particles, which are optically inactive in the visible spectral range.

In the cases, when spectral dependence of the aerosol extinction coefficients $\alpha(\lambda)$ is more complicated, the regression equation with a large number of input parameters should be used for separation.

We have tested the multiparameter approach using the array of the total aerosol extinction coefficients $\alpha(\lambda)$ obtained for hazes of the arid zone near the Lake Balkhash in spring and summer seasons. The average values and rms deviations of the meteorological parameters in spring were the following: the air temperature $\bar{t} = (+6.2 \pm 4.34)°$q, the partial pressure of water vapor $\bar{e} = (6.91 \pm 2.29)$ mbar, the relative air humidity $\bar{f} = (72.1 \pm 18.0)\%$, and the coefficient of aerosol extinction in the visible spectral range $\bar{\alpha}(0.55) = (0.101 \pm 0.047)$ km^{-1}. The measurements were conducted in the wavelength range from 0.44 to 11.5 μm along a near-ground path of 4630 m length. The total of 230 spectra of the coefficients $\alpha(\lambda)$ were obtained. The average spectral dependence of the coefficients $\alpha(\lambda)$ for the spring array is shown in Fig. 3*a* (curve *1*). It is seen that the spectral dependence of the coefficients $\alpha(\lambda)$ in the ranges 0.44 – 0.87, 0.87 – 2.2, and 2.2 – 11.5 μm is characterized by different curve slopes. This means that the main contribution into the aerosol extinction comes from three different factors. In this connection, to separate out the components of coefficients $\alpha(\lambda)$, we have applied a three-parameter regression equation

$$\alpha(\lambda) = K_0(\lambda) + K_1(\lambda)\,\Delta\alpha_1 + K_2(\lambda)\,\Delta\alpha_2 + K_3(\lambda)\,\alpha(3.97), \quad (2)$$

where $\Delta\alpha_1 = [\alpha(0.44) - \alpha(0.87)]$; $\Delta\alpha_2 = [\alpha(0.87) - \alpha(3.97)]$; $K_0(\lambda)$, $K_1(\lambda)$, $K_2(\lambda)$, and $K_3(\lambda)$ are the spectral coefficients of regression with values not presented here.

With the given input parameters, the component $K_1(\lambda)\,\Delta\alpha_1$ corresponds to the contribution coming into $\alpha(\lambda)$ from the finest-disperse fraction of particles with the modal radius smaller than 0.4 μm. At the same time, the components $K_2(\lambda)\,\Delta\alpha_2$ and $K_3(\lambda)\,\alpha(3.97)$ determine the contributions into $\alpha(\lambda)$ from the medium-disperse fraction (the modal radius of particles from 0.4 to 0.8 μm) and the coarse-disperse fraction of aerosol, respectively. The component $K_0(\lambda)$, as in Eq. (1), is responsible for a part of aerosol extinction, which does not correlate with the input parameters. The results of calculation of the spectral components $K_1(\lambda)\,\Delta\alpha_1$, $K_2(\lambda)\,\Delta\alpha_2$, and $K_3(\lambda)\,\alpha(3.97)$ at average values of the input parameters for the spring season are shown in Fig. 3 by curves *2*, *3*, and *4*, respectively.

The component $K_0(\lambda)$ proved to be negligibly small in this case, so it is not shown in the figure.

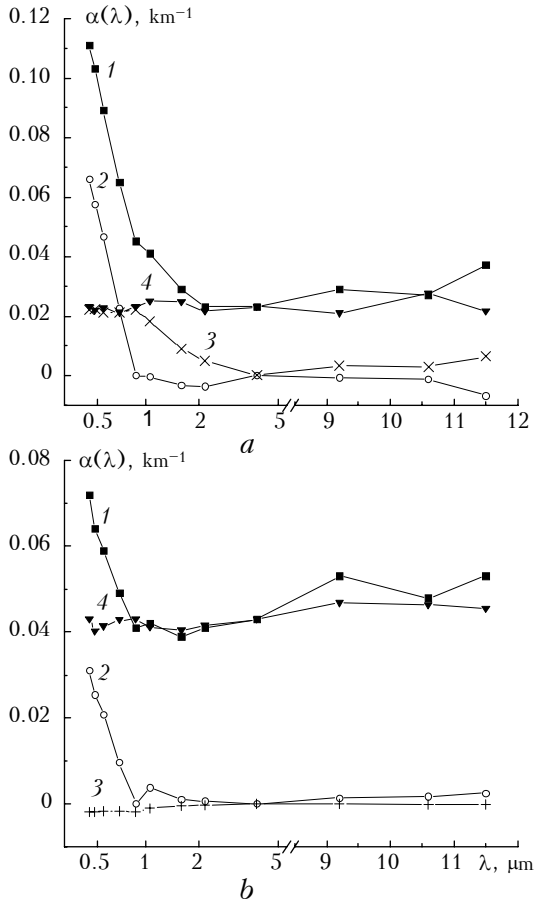

FIG. 3. Mean spectral dependence of the coefficients α(λ) *for the spring (a) and fall (b) arrays measured in the arid zone of Kazakhstan: the total aerosol extinction (curves 1), the aerosol extinction by particles of the fine-disperse, medium-disperse, and coarse-disperse fractions (curves 2–4).*

It is seen that the contribution of the fine-disperse fraction into the total aerosol extinction (curve *2*) prevails in the visible spectral range, and it is practically not pronounced in the IR range. The contribution of the medium-disperse fraction (curve *3*) is clearly pronounced in the visible and near IR spectral ranges. It is 0.022 km^{-1} in the spectral range of 0.44 – 1.06 μm. Then it reduces to zero and again becomes pronounced in the wavelength range of 9.2 and 11.5 μm. As was noted above, the absorption bands of sulfates, aluminum oxides, and carbonates lie just in this wavelength range. This circumstance allows us to suppose that the maxima of aerosol extinction coefficients found at these wavelengths in the coastal haze are likely to be caused also by the particles of medium-disperse fraction.

The contribution of the coarse-disperse fraction into the total aerosol extinction of optical radiation (curve *4*) is characterized by the spectral behavior that is very close to neutral. It determines the level of α(λ) in the wavelength range λ > 1.6 μm, to be about 0.02 km−1. It is almost three times smaller than in the coastal zone. Note that in the summer period, when powerful upward flows carry the coarse-disperse soil aerosol into the ground atmospheric layer, the contribution from this fraction increases up to 0.05 – 0.07 km−1 (Ref. 8).

Similar calculations of the spectral components of the total aerosol extinction coefficient have been performed for the array of the data obtained in the arid zone during the fall season. The calculated results, as well as the averaged spectrum of the total aerosol extinction coefficients for the fall season are shown in Fig. 3b. It is seen that, in contrast to the spring season, the average values of the coefficients α(λ) (curve 1) decrease monotonically in the spectral range from 0.44 to 0.87 μm, and then they are practically unchanged. The spectral dependence of the coefficients α can have such a form only with no particles of medium-disperse fraction in the atmosphere. This supposition is confirmed by calculated results on the components K1(λ) Δα1, K2(λ) Δα2 , and K3(λ) α(3.97) shown in Fig. 3. As is seen from this figure, curve 3, that corresponds to the contribution of medium-disperse particles into the total aerosol extinction, turns out to be close to zero in the entire wavelength range.

As a summary, we would like to note that the apparatus of the linear multivariate regression analysis proves to be quite efficient as applied to the problem of separation of the total aerosol extinction coefficients into components in a wide spectral range. It allows statistical estimation of separate contributions from particles of different size into the total aerosol extinction in a wide spectral range. Qualitative (and, in some cases, even quantitative) physical interpretation of the spectral dependence of α(λ) components indicates that weak absorption of radiation by aerosol can be revealed in the real atmosphere in both visible and IR ranges. The latter conclusion is important for climatic problems. It should also be noted that the use of the developed regression models represented by Eqs. (1) and (2) provides for an interesting possibility separating out a needed component in each realization of the coefficient α(λ), to create the array for the component needed, and to study its variability depending on different factors.

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REFERENCES

1. V.V. Veretennikov, Yu.A. Pkhalagov, and V.N. Uzhegov, in: *Optics of the Sea and Atmosphere, Abstracts of Reports at the X Plenum of the Workgroup on the Optics of the Ocean,* October of 1988, Rostov-na-Donu (State Optical Ins. Press, 1988), pp. 332–333.

2. M.V. Kabanov, M.V. Panchenko, Yu.A. Pkhalagov, et al., *Optical Properties of Coastal Hazes* (Nauka, Novosibirsk, 1988), 201 pp.
3. G.V. Rozenberg, Fiz. Atmos. Okeana **18**, No. 11, 1192–1198 (1982).
4. Yu.S. Lyubovtseva and L.G. Yaskovich, Fiz. Atmos. Okeana **18**, No. 9, 922–932 (1982).
5. Yu.A. Pkhalagov, V.N. Uzhegov, and N.N. Shchelkanov, Atmos. Oceanic Opt. **11**, No. 4, 272–275 (1998).
6. S.D. Andreev, V.E. Zuev, L.S. Ivlev, M.V. Kabanov, and Yu.A. Pkhalagov, Izv. Akad. Nauk SSSR, Fiz. Atmos. Okeana **8**, No. 12, 1262–1267 (1972).
7. V.E. Zuev, *Propagation of Visible and Infrared Waves in the Atmosphere* (Sov. Radio, Moscow, 1970), 496 pp.
8. Yu.A. Pkhalagov, V.N. Uzhegov, and N.N. Shchelkanov, Atmos. Oceanic Opt. **7**, No. 10, 714–720 (1994).