Statistical Estimation of the Atmospheric Aerosol Absorption Coefficient Based on the Data of Optical Measurements

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Introduction

The problem with the aerosol optical constants and, in particular, the imaginary part of the refractive index of particles in the visible and infrared (IR) wavelength ranges is important for the calculation of the atmospheric global albedo in climatic models. The available models of the aerosol optical constants obtained for the prescribed chemical composition of particles (Ivlev et al. 1973; Ivlev 1982; Volz 1972), are often far from real aerosol. In Krekov et al. 1982, model estimates of the atmospheric optical characteristics that depend on the accuracy of real and imaginary parts of the aerosol complex refractive index can differ by several hundred percent.

The aerosol extinction coefficient $\alpha(\lambda)$ obtained from measurements on a long horizontal path can be represented as $\alpha(\lambda)=\sigma(\lambda)+\beta(\lambda)$, where σ is the directed light scattering coefficient, and β is the aerosol absorption coefficient. The coefficient $\sigma(\lambda)$ is measured by a nephelometer. If the values $\alpha(\lambda)$ and $\sigma(\lambda)$ are determined, it is easy to determine the value $\beta(\lambda)$. However, it is almost impossible for a number of reasons. First, the real values $\alpha(\lambda)$ and $\sigma(\lambda)$ are close to each other, and the estimate of the parameter $\beta(\lambda)$ is concealed by the errors of measurements. Second, the aerosol optical characteristics on the long path and in the local volume of nephelometer can be different, which also leads to errors in estimating $\beta(\lambda)$. It is difficult to obtain spectral measurements of $\sigma(\lambda)$ in the IR wavelength range. Taking these circumstances into account we consider the statistical technique that makes it possible to estimate the absorption coefficient of real aerosol based on the analysis of simultaneous measurements of the spectral aerosol extinction coefficients $\alpha(\lambda)$, the directed scattering coefficient of dry aerosol $\sigma_0(0.55)$ and the mass concentration of aerosol containing black carbon (BC) Ms.

Characteristics of the Initial Data

Analysis was performed using data from round-the-clock measurements of the spectral aerosol extinction coefficients in the wavelength range 0.45 - 12 μm obtained on the 830-m long near-ground path with the meter (Pkhalagov et al. 1988) that determined the spectral aerosol extinction coefficients $\alpha(\lambda)$ using the Pkhalagov et al. 1992 technique. The scattering coefficients of dry aerosol $\sigma0(0.55)$ were measured in the local volume by the nephelometer FAN at a 45° angle in a heated chamber. The mass concentration of the aerosol containing BC Ms ($\mu g/m^3$) was measured by the aethalometer, the design and operation principle are described in Kozlov et al. 2002. Meteorological observations were carried out simultaneously with optical measurements, and the parameter of aerosol condensation activity γ was measured once a day. Let us note that the value σ_0 is related to the aerosol scattering coefficient σ by the known formula $\sigma=\sigma_0\,(1–RH/100)^{-\gamma}$, where RH is the relative humidity of air.

When processing, the entire array of the coefficient $\alpha(\lambda)$ was divided into two sub-arrays obtained at RH of below 60%, “dry” atmosphere (mainly daytime measurements), and above 60%, “wet” atmosphere (mainly nighttime measurements).

The mean values and rms deviations of the measured parameters (X) for two sub-arrays are presented in Table 1. The wide range of variability of these characteristics demonstrates how representative the obtained experimental data is.

Table 1. Mean values of the measured parameters, their rms deviations and the ranges of variability.

Measured parameter X	$\bar{X}$	rms deviation	Xmax	Xmin
«dry» atmosphere (N = 141)				
$\alpha(0.55)$, km^{-1}	0.188	0.064	0.34	0.063
$\alpha(3.9)$, km^{-1}	0.147	0.044	0.271	0.064
σ_0, km^{-1}sr^{-1}	0.0081	0.0049	0.024	0.002
Ms, μg/m^3	1.062	0.652	4.77	0.302
t, °C	22.26	4.92	32.08	7.72
RH, %	43.9	9.3	59.9	22.1
γ	0.288	0.091	0.49	0.14
«wet» atmosphere (N = 97)				
$\alpha(0.55)$, km^{-1}	0.120	0.059	0.32	0.006
$\alpha(3.9)$, km^{-1}	0.076	0.033	0.160	0.014
σ_0, km^{-1}sr^{-1}	0.0099	0.0062	0.034	0
Ms, μg/m^3	1.095	0.612	3.88	0
t, °C	14.68	4.88	23.33	1.0
RH, %	77.7	10.2	94.2	60.1
γ	0.269	0.089	0.49	0.14

Technique for Statistical Comparison of the Aerosol Extinction and Scattering

Data were processed using the mathematical algorithm of multi-parameter linear regression, assuming that variations of the coefficient $\alpha(\lambda)$ are determined by coarse and submicron particle concentration changes. The measured value $\alpha(\lambda)$ can be presented in the form $\alpha(\lambda) = \alpha_{coarse}(\lambda) + \alpha_{subm}(\lambda)$. Then, the value $\alpha^*_{coarse}= [\alpha(2.2)+\alpha(3.9)]/2$ can be used as the parameter α_{coarse}, and the parameter $\alpha_{subm}(\lambda)$ can be used to represent a combination of the parameters measured in the local volume (σ_0, $\sigma_{RH} = \sigma - \sigma_0$ and Ms). The linear regression equation for $\alpha(\lambda)$ can be written in the form

$$\alpha(\lambda) = K_0(\lambda) + K_1(\lambda)\cdot\alpha^*_{coarse} + K_2(\lambda)\cdot\sigma_0 + K_3(\lambda)\cdot\sigma_{RH}+ K_4(\lambda)\cdot Ms+\delta(\lambda), \qquad (1)$$

where Ki are the spectral regression coefficients, α^*_{coarse}, σ_0, σ_{RH}, Ms are the input parameters, and $\delta(\lambda)$ is the error in reconstruction of $\alpha(\lambda)$. The coefficients $Ki(\lambda)$ were calculated by the method of least squares taking into account the mutual correlations of the input parameters of the equation in Table 2.

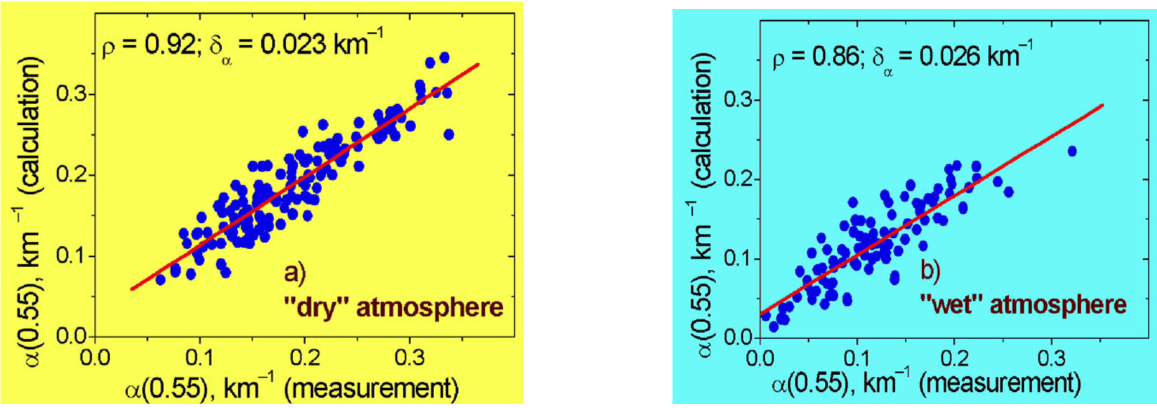
Table 2. Correlation coefficients of the input parameters of Eq. (1)

Input parameters	α^*_{coarse}	σ_0	σ_{RH}	Ms
«dry» atmosphere				
α^*_{coarse}	1.0	0.121	-0.148	-0.119
σ_0		1.0	0.696	0.573
σ_{RH}			1.0	0.542
Ms				1.0
«wet» atmosphere				
α^*_{coarse}	1.0	0.174	-0.058	0.188
σ_0		1.0	0.402	0.715
σ_{RH}			1.0	0.269
Ms				1.0

According to the physical meaning of the parameters of Eq. (1), the component $K_1(\lambda)\cdot\alpha^*_{coarse}$ determines the contribution of coarse aerosol into variability of the parameter $\alpha(\lambda)$. The components $K_2(\lambda)\cdot\sigma_0$ and $K_3(\lambda)\cdot\sigma_{RH}$, respectively, are related to variations of the dry matter and condensation part of submicron aerosol, and the component $K4(\lambda)$Ms determines the sought contribution of absorbing aerosol containing BC into variability of the parameter $\alpha(\lambda)$.

Results

Correlation of the aerosol extinction coefficients at the wavelength 0.55 µm, which were obtained from measurements on the horizontal path and calculated using Eq. (1), is shown in Figures 1 a and b for two data arrays. A high level of correlation (0.92 and 0.88 for "dry" and "wet" atmosphere, respectively) is evidence of good applicability of Eq. (1) to divide the total aerosol extinction into components.



(a) (b)

Figure 1a-b. The correlation of the aerosol extinction coefficients at the wavelength 0.55 µm, which were obtained from measurements on the horizontal path and calculated using Eq. (1) for two data arrays.

Figures 2 - 5 show the effect of changing the input parameters of Eq. (1) on the spectral structure of the aerosol extinction coefficients α(λ) dry (Figures 2a - 5a) and wet (Figures 2b - 5b) atmospheric conditions. Three spectral dependences of the coefficients α(λ) are shown in each figure. Curve 1 corresponds to the mean values of all input parameters. Vertical lines show the error in reconstruction of the coefficients α(λ) at each wavelength. Curve 2 in each figure shows maximum, and Curve 3 shows the minimum values of one of the input parameters of Eq. (1) provided that other input parameters take their mean values.

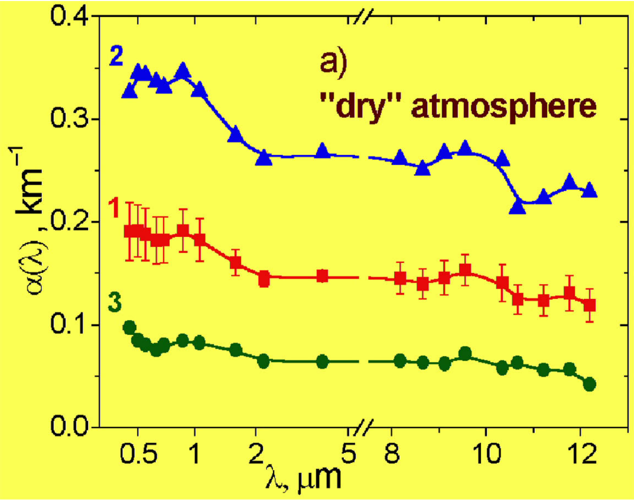


Figure 2a. The effect of changing the input parameters on Eq. (1) on the spectral structure of the aerosol extinction coefficients ά(γ) are shown in dry atmospheric conditions. Transformation is considered part of the spectrum of the coefficients ά(γ) due to the coarse aerosol concentration change.

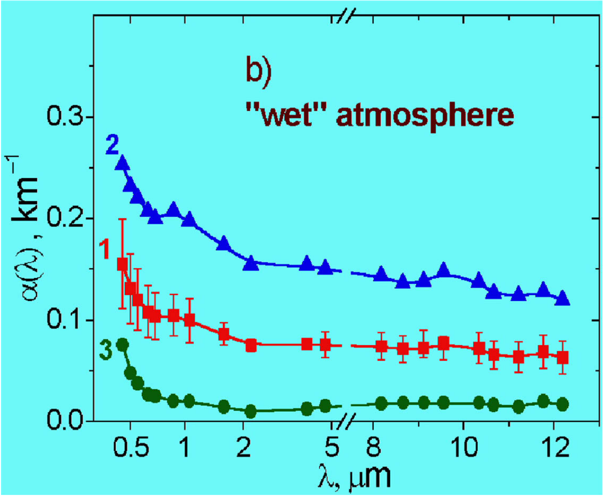


Figure 2b. The effect of changing the input parameters on Eq. (1) on the spectral structure of the aerosol extinction coefficients ά(γ) are shown in wet atmospheric conditions. Transformation is considered part of the spectrum of the coefficients ά(γ) due to the change in the coarse aerosol concentration.

In particular, transformation is considered part of the spectrum of the coefficients $\alpha(\lambda)$ due to the change of the coarse aerosol concentration (Figure 2), the dry matter of submicron aerosol concentration (Figure 3), the condensation portion of submicron aerosol (Figure 4), and aerosol containing BC (Figure 5). It follows from the data comparison that the coarse aerosol concentration change makes the greatest contribution to the variability of the coefficients $\alpha(\lambda)$ in the entire wavelength range. The variability of $\alpha(\lambda)$ caused by this factor is maximized in the dry atmosphere. The change of the parameter σ_0 noticeably changes the structure of the spectrum $\alpha(\lambda)$ in the shortwave range and only at maximum values σ_0 in the "dry" and "wet" atmosphere (Figure 3). As for the parameter σ_{RH}, its

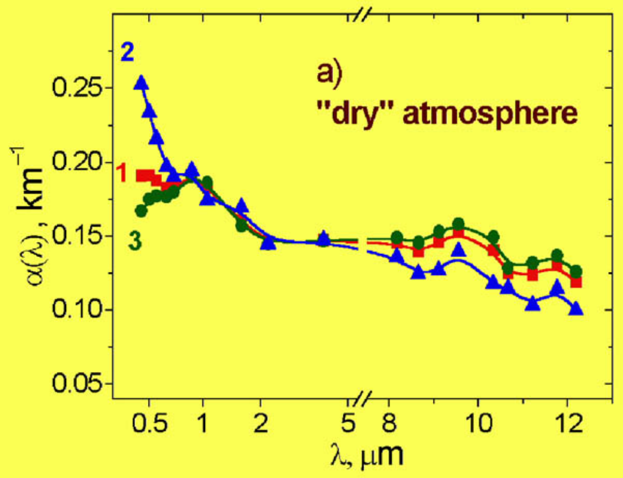


Figure 3a. The effect of changing the input parameters on Eq. (1) on the spectral structure of the aerosol extinction coefficients ά(γ) are shown in dry atmospheric conditions. Transformation is considered part of the spectrum of the coefficients ά(γ) due to the change in the dry matter concentration of submicron aerosol.

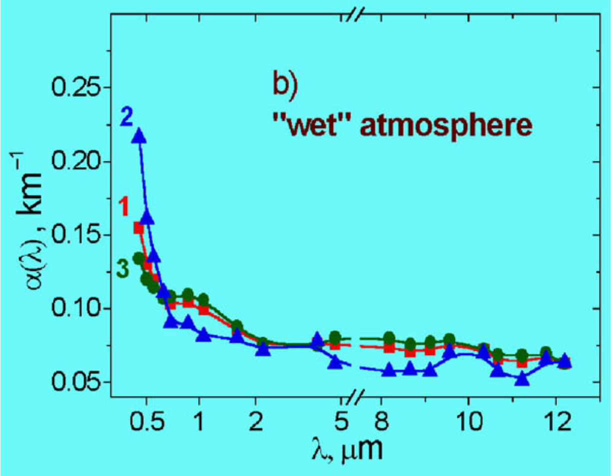


Figure 3b. The effect of changing the input parameters on Eq. (1) on the spectral structure of the aerosol extinction coefficients ά(γ) are shown in wet atmospheric conditions. Transformation is considered part of the spectrum of the coefficients ά(γ) due to the change in the dry matter concentration of submicron aerosol.

variations in the dry atmosphere hardly affect the spectrum $\alpha(\lambda)$ (Figure 4a), while variations of σ_{RH} in the wet atmosphere dramatically change the spectral structure of the aerosol extinction coefficients in the entire wavelength range (Figure 4b). On the contrary, the content change of the absorbing aerosol is better pronounced in the dry atmosphere and hardly has an affect under wet conditions (Figures 5a and 5b).

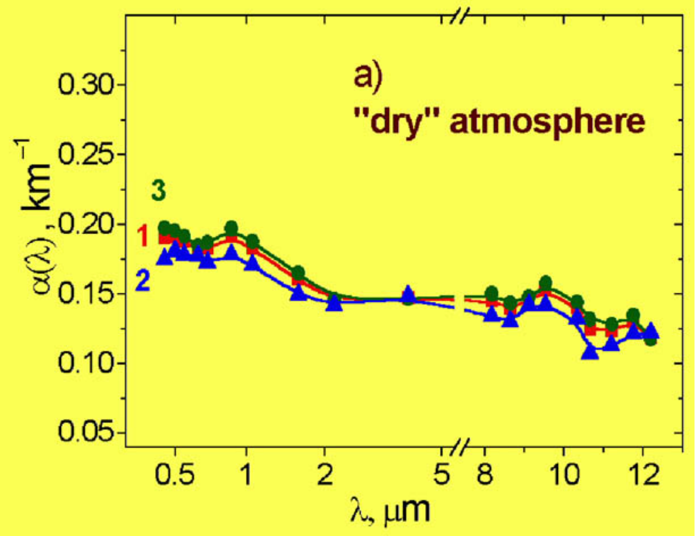


Figure 4a. The effect of the changing the input parameters on Eq. (1) on the spectral structure of the aerosol extinction coefficients ά(γ) are shown in dry atmospheric conditions. Transformation is considered part of the spectrum of the coefficients ά(γ) due to the change in the condensation portion of the submicron aerosol.

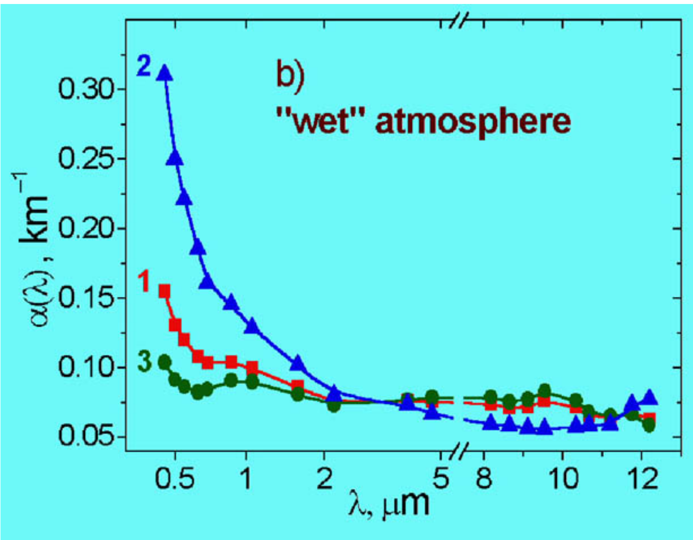


Figure 4b. The effect of changing the input parameters on Eq. (1) on the spectral structure of the aerosol extinction coefficients ά(γ) are shown in wet atmospheric conditions. Transformation is considered part of the spectrum of the coefficients ά(γ) due to the change in the condensation portion of the submicron aerosol.

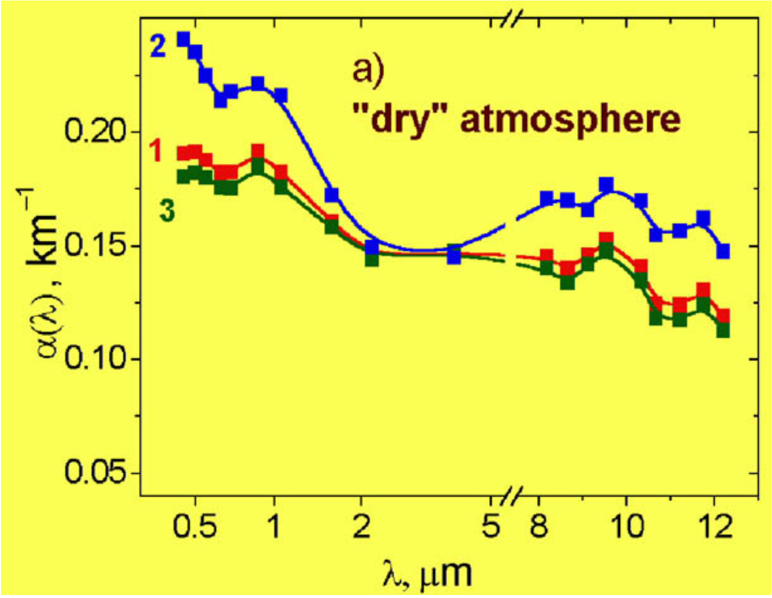


Figure 5a. The effect fo changing the input parameters on Eq. (1) on the spectral structure of the aerosol extinction coefficients ά(γ) are shown in dry atmospheric conditions. Thransformation is cnosidred part of the spectrum of the coefficients ά(γ) due to the change in aerosol containing BC.

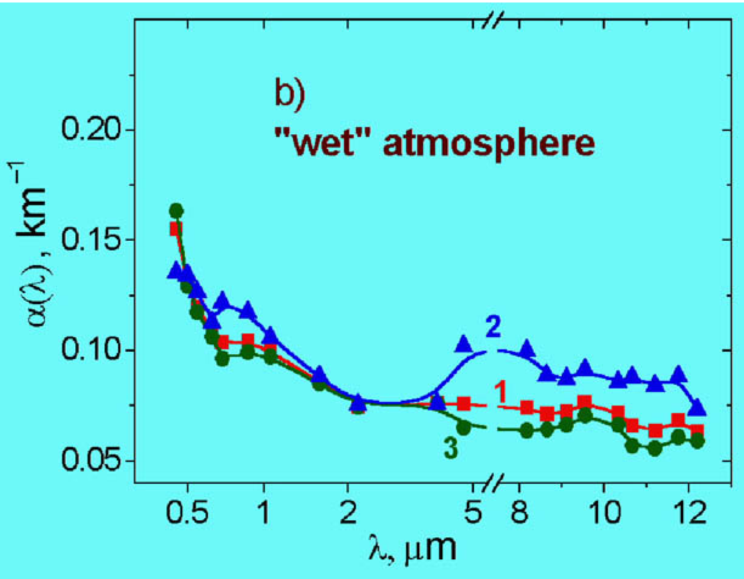


Figure 5b. The effect of changing the input parameters on Eq. (1) on the spectral structure of the aerosol extinction coefficients ά(γ) are shown in wet atmospheric conditions. Transformation is considered part of the spectrum of the coefficients ά(γ) due to the change in aerosol containing BC.

The calculated spectral dependences of the component $\alpha_{sbm}(\lambda) = K_2(\lambda)\cdot\sigma_0 + K_3(\lambda)\cdot\sigma_{RH} + K_4(\lambda)\cdot Ms$ in two RH ranges are shown in Figure 6. As seen, the spectral structure of the submicron component of extinction behaves in an expectant manner – it quickly decreases with wavelength, and its contribution in the range $\lambda \geq 2$ μm becomes practically equal to zero.

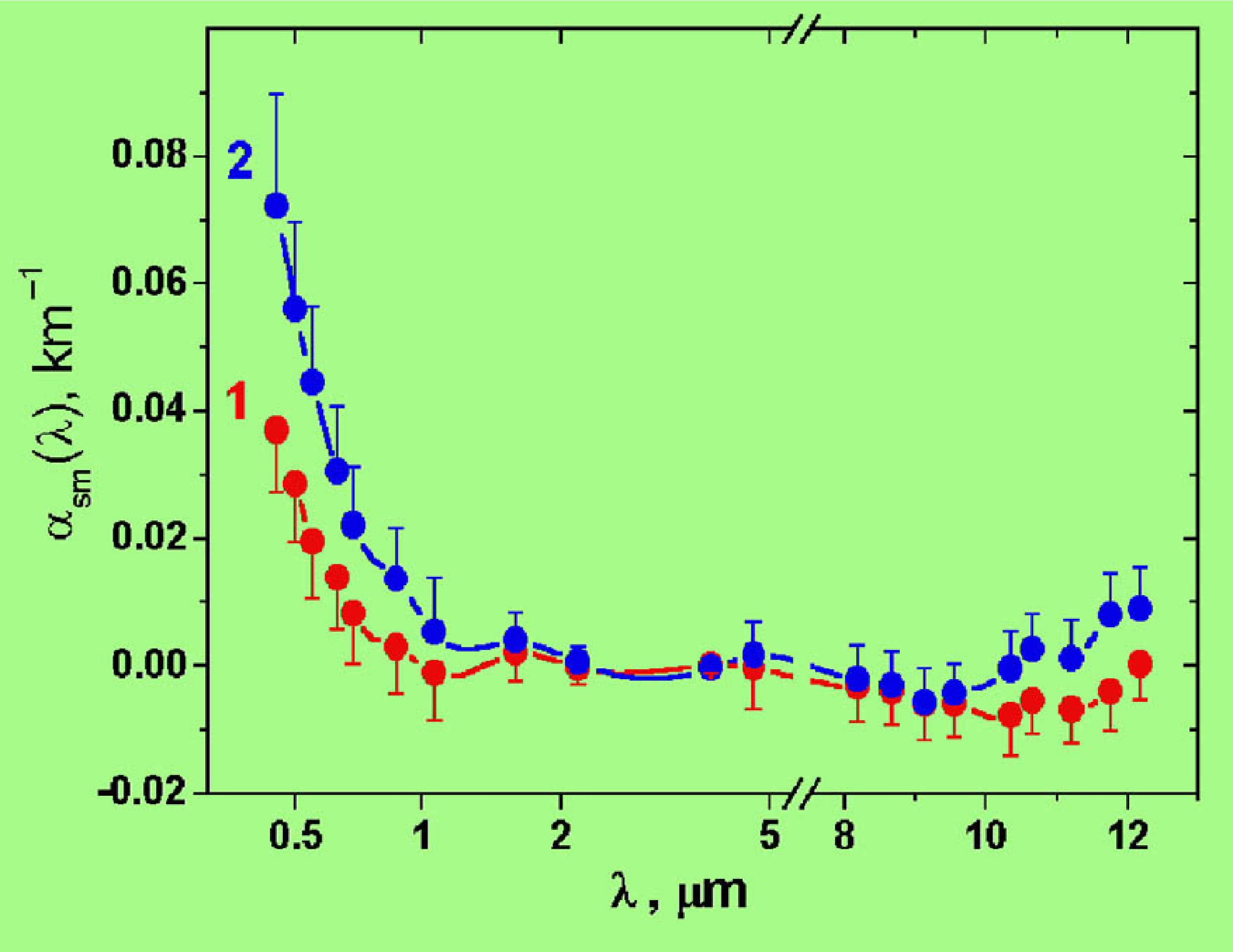


Figure 6. The calculated spectral dependences of the component (see first sentence p. 6 for equation) in two RH ranges.

The spectral dependence of the component β (λ) = $K_4(\lambda)\cdot Ms$ calculated by Eq. (1) is shown in Figure 7. It is caused by absorption of radiation by BC aerosol. Here, we consider only conditions of low RH, where this dependence is the strongest. It is seen that the well-pronounced decrease of the absorption coefficient with wavelength is observed in the range of 0.44 - 4 µm. Such a dependence of β on wavelength, which can be estimated as 1/λ (see Curve 2), is evidence that absorption is related to very small particles. The result is in qualitative agreement with the conclusions of Pkhalagov et al. 1998, where the continuous aerosol absorption of radiation in the range 0.44 - 4 µm was revealed by a distinctly different technique based on field measurements in the arid zone of the atmosphere.

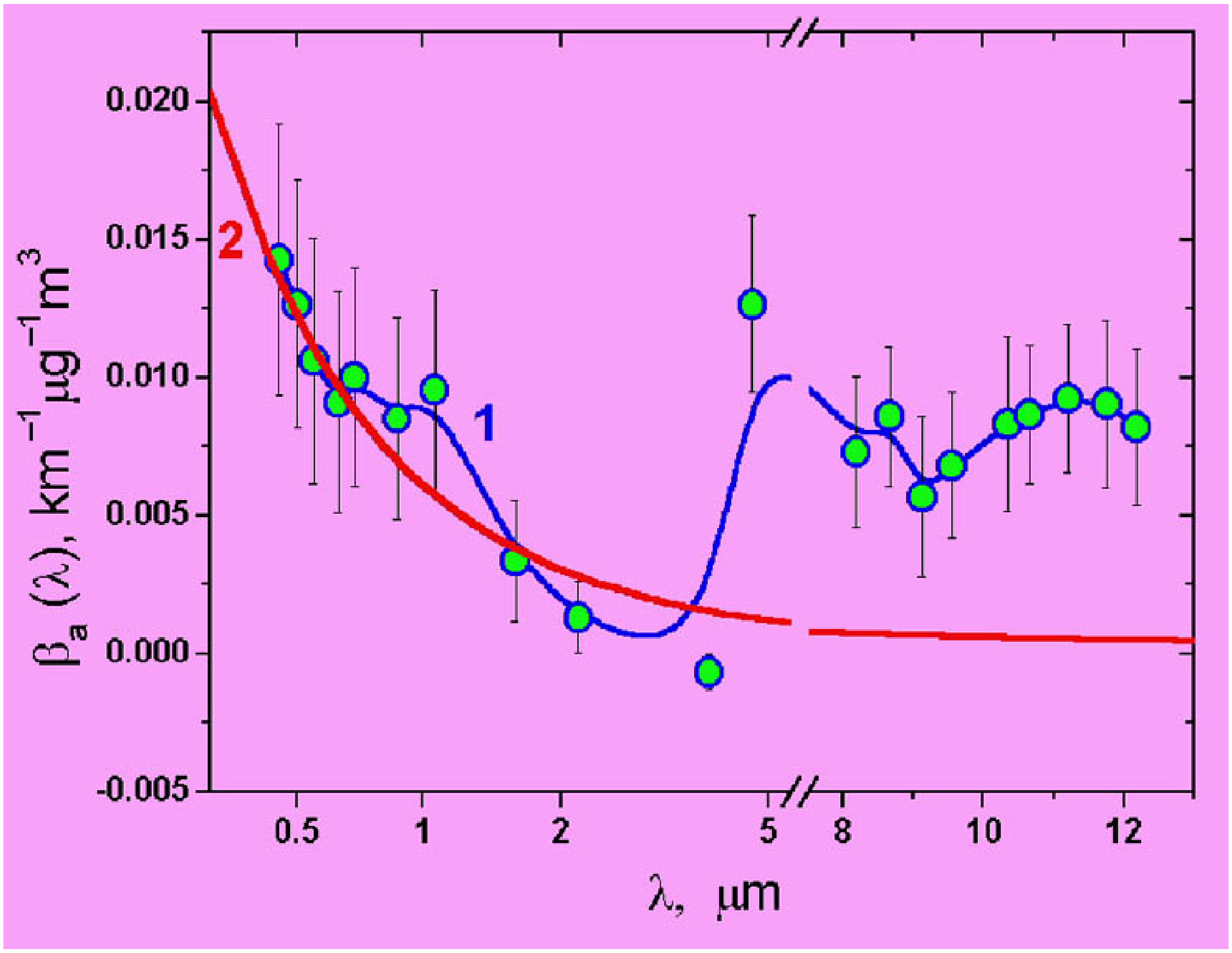


Figure 7. The spectral dependence of the component (see second paragraph, first sentence p. 6 for equation) calculated by Eq. 1.

Later, the presence of aerosol absorption in visible and near IR ranges was reliably revealed by optical-acoustic spectrometry (Kozlov et al. 2002). The specific values of the absorption coefficients in the range 0.53 - 1.06 µm are also presented there. Comparison of the absorption coefficients obtained in the present paper with the data Kozlov et al. 2002 shows solid agreement. It allows us to determine that the statistical approach used in this paper can be applied to atmospheric aerosol absorption coefficients in the wavelength range 0.4 - 4 µm under low RH conditions. As for the range 8 - 12 µm, where a noticeable increase of the coefficient β(λ) is observed, additional investigations are necessary for clarification.

Acknowledgements

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