Statistical estimation of light absorption by atmospheric aerosol using data of optical measurements

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A statistical technique is proposed for estimating absorption coefficient of atmospheric aerosol in a wide wavelength range based on data of simultaneous long-path measurements of the spectral coefficients of the radiation extinction by aerosol, scattering coefficients of the dry aerosol at $\lambda = 0.52$ μm, and mass concentration of the soot-containing aerosol M_{BC} in a local volume. The technique was tested on the experimental material obtained during complex studies of the atmospheric aerosol in June 2004 near Tomsk. We have revealed spectral dependence of the absorption coefficient of soot-containing aerosol. A well-pronounced decrease of the absorption occurs with the wavelength increasing from 0.45 to 4 μm. Such a wavelength dependence of the absorption coefficient, which can be approximately estimated as $1/\lambda$, evidences that the absorbing particles have very small size.

Introduction

In calculating the global albedo of the atmosphere for climate models it is very important to correctly select optical constants of the aerosol and, in particular, the imaginary part of the complex refractive index of the particulate matter in the visible and infrared (IR) spectral regions. The available models of the aerosol optical constants were obtained for a preset chemical composition of particles[1–3] and they do not always correspond to real aerosol. It is shown in Ref. 4 that model estimates of the optical characteristics of the atmosphere can differ by hundreds percent depending on how correct the real and imaginary parts of the aerosol refractive index were set.

It is known that the extinction coefficient of aerosol $\beta(\lambda)$ obtained from long-path near-ground measurements can be presented as $\beta(\lambda) = \beta^S(\lambda) + \beta^A(\lambda)$, where $\beta^S(\lambda)$ is the scattering coefficient, and $\beta^A(\lambda)$ is the aerosol absorption coefficient. It seems that, if the value $\beta^S(\lambda)$ is simultaneously measured in a closed volume by means of a nephelometer, one can easily determine the value $\beta^A(\lambda) = \beta(\lambda) - \beta^S(\lambda)$. However, it is practically impossible to do this for several reasons. First, the quantities $\beta(\lambda)$ and $\beta^S(\lambda)$ have very close values, and the estimate of the parameter $\beta^A(\lambda)$ will be noised by the measurement errors. Second, the aerosol optical characteristics on a long path and in a closed volume of a nephelometer at each moment in time can differ because of spatial inhomogeneity of the aerosol field, that always will lead to errors in determining $\beta^A(\lambda)$. Third, nephelometers with closed volume dry aerosol and actually measure the scattering coefficient of dry aerosol $\beta_0^S(\lambda)$, which noticeably differs from $\beta^S(\lambda)$ measured under conditions of enhanced air humidity. Besides, there are serious technical difficulties in conducting spectral measurements of $\beta^S(\lambda)$ in the IR wavelength range.

Taking into account these facts, a statistical technique is proposed in this paper, which makes it possible to estimate the absorption coefficient of real atmospheric aerosol in a wide wavelength range using data of simultaneous long-path measurements of the coefficients $\beta(\lambda)$, nephelometric measurements of the coefficients $\beta_0^S(\lambda)$ at the wavelength of $\lambda = 0.52$ μm and the mass concentration of aerosol containing black carbon M_{BC} measured in a local volume.

Characterization of the initial data

The data of round-the-clock measurements of the atmospheric aerosol characteristics near Tomsk obtained during a comprehensive radiative experiment in June 2004 were used in analysis. Measurements of the spectral transmission of the atmosphere in the wavelength range from 0.45 to 12 μm were carried out along a 830-m-long near-ground path every 2 hours with an automated instrumentation complex.[5] Transparency of the atmospheric layer $T(\lambda)$ and spectral coefficients of the total extinction $\varepsilon(\lambda)$ were determined using the technique proposed in Ref. 6. The extinction coefficients of aerosol $\beta(\lambda)$ were isolated from the obtained values of the coefficients $\varepsilon(\lambda)$ using the statistical technique[7] based on the procedure of multiple linear regression. The scattering coefficient of dry aerosol $\beta_0^S(0.52)$ was measured in a local volume by means of a FAN nephelometer at the scattering angle of 45° in a heated chamber,[8] and the

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mass concentration of aerosol containing black carbon M_{BC} ($\mu g/m^3$) was determined by means of the aethalometer, the design and operational principle of which can be found in Ref. 9. Meteorological observations were carried out simultaneously with the optical measurements. The parameter of the condensation activity of submicron aerosol γ was measured once a day. It allows one to relate the scattering coefficients of dry, $\beta_0^S(0.52)$, and wet, $\beta^S(0.52)$, aerosol using the relationship

$$\beta^S(0.52) = \beta_0^S(9.52)(1 - RH/100)^{-\gamma}, \quad (1)$$

where RH is the relative humidity of air. Let us note that $\beta^S(0.52)$ was measured at the Aerosol Monitoring Station of the Institute of Atmospheric Optics every hour. To determine the elements of the $\{\beta^S(0.52)\}$ array corresponding to the *i*th realization of $\beta(\lambda)$ measured at different time, the values γ were calculated by linear approximation between two points obtained from measurements on the nearest days.

In data processing, all arrays were divided into two subarrays obtained at RH < 60% ("dry" atmosphere, mainly daytime measurements) and RH > 60% ("wet" atmosphere, mainly nighttime measurements).

Mean values and rms deviations of the measured parameters (X) for the two subarrays are presented in Table 1. The quite wide variability of these parameters is evidence of the representativeness of the experimental data obtained.

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

Measured parameter X	$\bar{X}$	rms	X_{max}	X_{min}
"dry" atmosphere (N = 141)				
$\beta(0.55)$, km^{-1}	0.188	0.064	0.34	0.063
$\beta(3.9)$, km^{-1}	0.147	0.044	0.271	0.064
$\beta_0^S(0.52)$, km$^{-1}\cdot$ sr^{-1}	0.0081	0.0049	0.024	0.002
M_{BC}, $\mu g/m^3$	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)				
$\beta(0.55)$, km^{-1}	0.120	0.059	0.32	0.006
$\beta(3.9)$, km^{-1}	0.076	0.033	0.160	0.014
$\beta_0^S(0.52)$, km$^{-1}\cdot$ sr^{-1}	0.0099	0.0062	0.034	0
M_{BC}, $\mu g/m^3$	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 estimation of the aerosol absorption

The data were processed using the mathematical technique of linear multi-parameter regression. Assuming that, in the general case, variations of the $\beta(\lambda)$ coefficient are determined by variation of the concentrations of coarse and submicron particles, the measured $\beta(\lambda)$ value can be presented as $\beta(\lambda) = \beta_c(\lambda) + \beta_{sbm}(\lambda)$. Then, if one considers the value $\beta_c^* = [\beta(2.2) + \beta(3.9)]/2$ as the parameter β_c, and present the parameter $\beta_{sbm}(\lambda)$ in the form of a combination of the parameters β_0^S, β_{RH}^S, and M_{BC}, one can write the equation of linear regression for $\beta(\lambda)$ in the form

$$\beta(\lambda) = K_0(\lambda) + K_1(\lambda)\beta_c^* + K_2(\lambda)\beta_0^S + K_3(\lambda)\beta_{RH}^S + K_4(\lambda)M_{BC} + \delta_\beta(\lambda), \quad (2)$$

where $K_i(\lambda)$ are the spectral regression coefficients, β_c, β_0^S, and $\beta_{RH}^S = \beta^S - \beta_0^S$ are the input parameters, $\delta_\beta(\lambda)$ is the error in reconstruction of $\beta(\lambda)$. Let us remind that the parameter β^S was calculated from Eq. (1) using the set values of β_0^S, RH, and γ. The coefficients $K_i(\lambda)$ were calculated by the least squares method taking into account correlations between the input parameters of the equation (Table 2).

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

Input parameters	β_c^*	β_0^S	β_{RH}^S	M_{BC}	β_c^*	β_0^S	β_{RH}^S	M_{BC}
	«dry» atmosphere				«wet» atmosphere			
β_c^*	1.0	0.12	–0.15	–0.12	1.0	0.17	–0.06	0.19
β_0^S		1.0	0.70	0.57		1.0	0.40	0.72
β_{RH}^S			1.0	0.54			1.0	0.27
M_{BC}				1.0				1.0

According to physical meaning of the input parameters of Eq. (2), the component $K_1(\lambda)\beta_c^*$ determines the contribution of the coarse aerosol to variability of the parameter $\beta(\lambda)$ in the entire wavelength range, the components $K_2(\lambda)\beta_0^S$ and $K_3(\lambda)\beta_{RH}^S$ determine the contributions of dry and wet submicron aerosol, respectively, and the component $K_4(\lambda)M_{BC}$ determines the contribution of the absorbing aerosol containing black carbon. Let us note that introducing two parameters, β_0^S and β_{RH}^S, responsible for the scattering properties of submicron aerosol seems, at first sight, to be odd, and, one would think, it is sufficient to introduce the only parameter $\beta^S = \beta_0^S + \beta_{RH}^S$. However, the estimates of the error in reconstruction of the value $\beta(\lambda)$ using the model (2) show that such a replacement is admissible only under conditions of low humidity, but at high humidity introducing two parameters, β_0^S and β_{RH}^S, makes it possible to noticeably decrease the error in reconstruction of $\beta(\lambda)$.

Results of investigations

Correlation of the aerosol extinction coefficients at the wavelength of 0.55 μm obtained from measurements along the horizontal path and calculated by Eq. (2) for two data arrays is shown in Fig. 1.

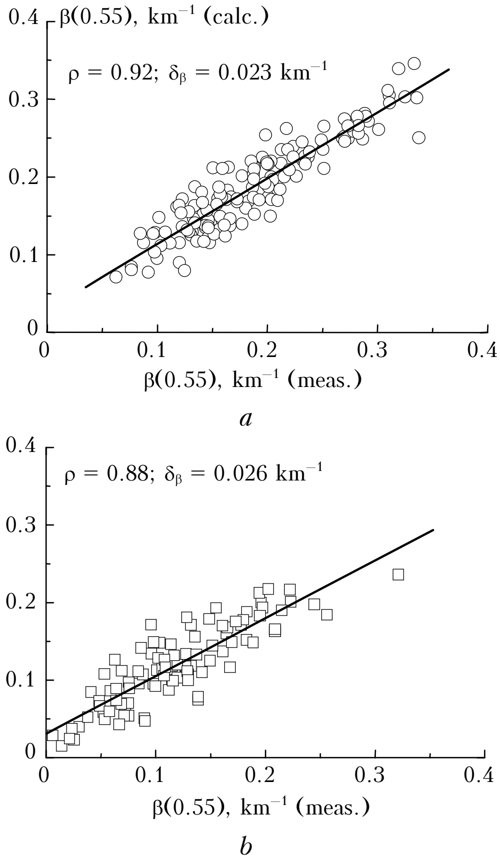

Fig. 1. Comparison of the measured and calculated, by model (2), aerosol extinction coefficients at the wavelength of 0.55 μm in "dry" (*a*) and "wet" (*b*) atmosphere.

High correlation (0.92 and 0.88 for "dry" and "wet" atmosphere, respectively) is an evidence of the correctness of the choice of input parameters of Eq. (2) and makes it possible to study the role of each predictor in formation of the spectra of the aerosol extinction coefficients. Variations of the spectra of the aerosol extinction coefficients $\beta(\lambda)$ are shown in Figs. 2–5 as functions of the input parameters for "dry" and "wet" atmosphere. In particular, transformation of the spectra of $\beta(\lambda)$ coefficients due to the change of the contribution of coarse aerosol (see Fig. 2), dry matter (Fig. 3), and submicron aerosol fraction caused by condensation of water vapor (Fig. 4), submicron aerosol and black carbon (Fig. 5) is considered. Curve *1* in Figs. 2 to 5 corresponds to the mean values of all input parameters, curve *2* is for maximum, and curve *3* is for minimum under condition that other input parameters take their mean values. Vertical bars on the curve *1* in Fig. 2 show the error in reconstruction of the coefficients $\beta(\lambda)$ at each wavelength. Let us note that the spectra of $\beta(\lambda)$ coefficients shown in Figs. 2 to 5 are drawn relative to that at the wavelength of 3.9 μm that is caused by the choice of input parameter in Eq. (2) responsible for variations of coarse aerosol.

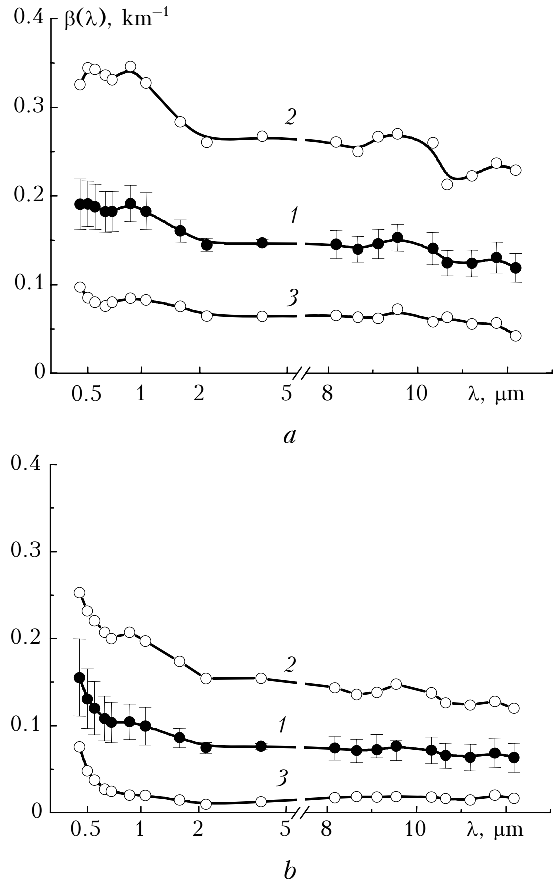

Fig. 2. Transformation of the spectra of the aerosol extinction coefficients at variations of the concentration of the coarse aerosol in "dry" (*a*) and "wet" (*b*) atmosphere.

It follows from comparison of these data that the change of the radiation extinction by aerosol related to the coarse aerosol makes the greatest contribution to variability of $\beta(\lambda)$ coefficients in the entire wavelength range. Variability of $\beta(\lambda)$ in "dry" atmosphere due to this factor is a little bit stronger than in "wet" atmosphere. Obviously, this is related to the fact that the conditions of "dry" atmosphere (RH < 60%) are realized mainly in daylight, when, in summer, convective and turbulent fluxes are well developed, which favor emission of large particles of soil into the atmosphere. The estimates show that introduction of the parameter β_c^* into the model (2) leads to dramatic decrease of the error in reconstruction of the coefficient $\beta(\lambda)$ in both "dry" and "wet" atmosphere.

It is interesting to note that the variations of the spectral structure of $\beta(\lambda)$ with the change of β_c^* in "dry" and "wet" atmosphere are significantly different. Variations of the parameter β_0^S (see Fig. 3) noticeably affect the spectrum of $\beta(\lambda)$ only in the shortwave range ($\lambda < 0.6$ μm) and especially in "dry" atmosphere. The spectral dependence under conditions of "wet" atmosphere practically is not changed. As to the parameter β^{S}_{RH}, its variations in "dry" atmosphere weakly change the spectrum β(λ) (see Fig. 4*a*), while variations of β^{S}_{RH} in "wet" atmosphere very strongly affect the spectrum of the coefficient β(λ) in the entire wavelength range (see Fig. 4*b*). As the air humidity increases, and, hence, increases the parameter β^{S}_{RH}, the well-pronounced increase of the coefficients β(λ) in the shortwave range is observed, and the spectrum becomes smoother, without any extremes.

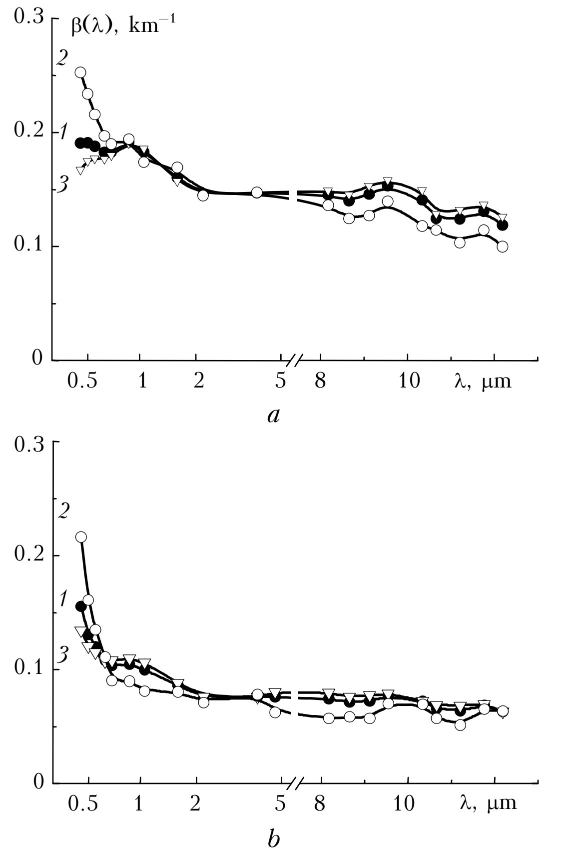

Fig. 3. Transformation of the spectra of the aerosol extinction coefficients at variations of the parameter β^{S}_{0}, related to the "dry" matter of submicron aerosol in "dry" (*a*) and "wet" (*b*) atmosphere.

Changes in the spectrum of the β(λ) coefficient at variations of the concentration of aerosol containing black carbon (M_{BC}) are shown in Fig. 5 for three values of the content of absorbing substance in aerosol particles: 0, 1, and 3 μg/m^3 (curves *3*, *1*, and *2*, respectively). It is seen from Fig. 5 that β(λ) spectrum noticeably differs from the mean one only at the high content of black carbon (M_{BC} = 3 μg/m^3, curve *2*).

The analysis performed makes it possible to estimate variations of the spectra of both the coefficient of total aerosol extinction and its components included in Eq. (2) depending on any external parameters under condition that correlations of these parameters with the predictors (2) have been revealed.

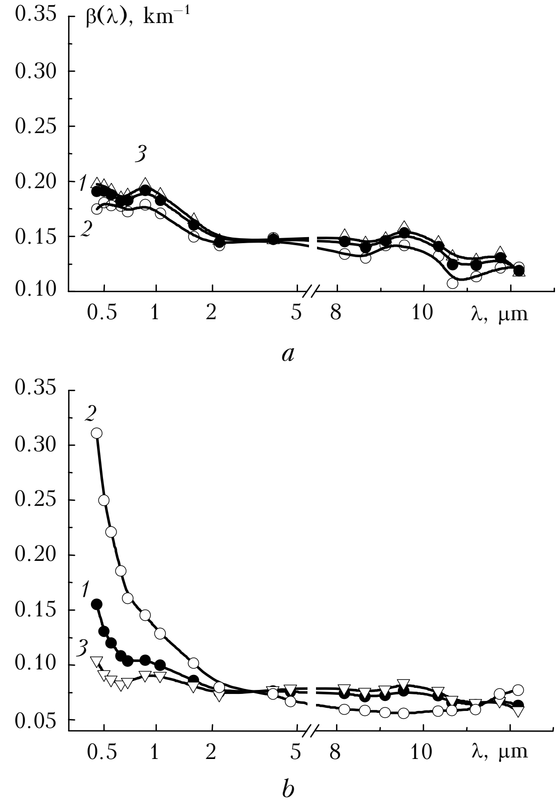

Fig. 4. Transformation of the spectra of aerosol extinction coefficients at variations of the condensation fraction of submicron aerosol in "dry" (*a*) and "wet" (*b*) atmosphere.

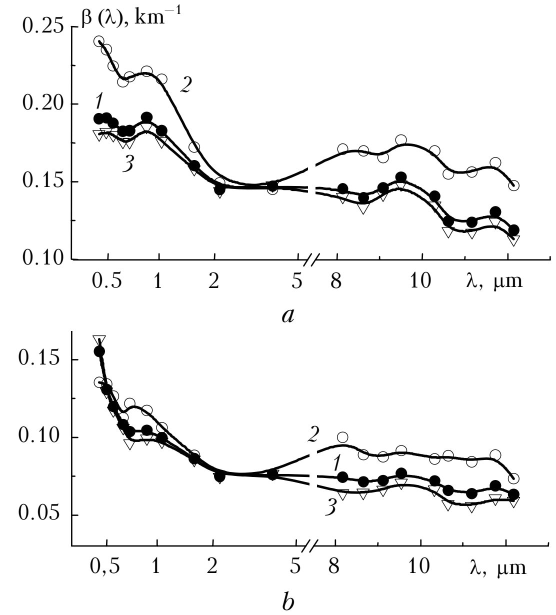

Fig. 5. Transformation of the spectra of aerosol extinction coefficients at variations of the content of black carbon in aerosol particles in "dry" (*a*) and "wet" (*b*) atmosphere.

The calculated spectra for the submicron component

$$\beta_{sbm}(\lambda) = K_2(\lambda)\beta_0^S + K_3(\lambda)\beta_{RH}^S + K_4(\lambda)M_{BC}$$

in two ranges of relative humidity of air are shown in Fig. 6. As is seen, the spectra of extinction by the submicron component have usual shape: it quickly decreases with the increasing wavelength, and its contribution becomes practically equal to zero in the range of $\lambda \geq 2$ μm.

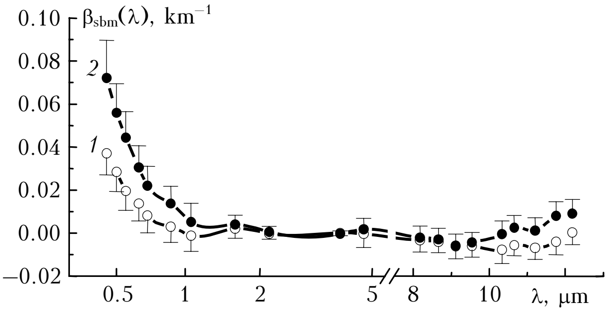

Fig. 6. Spectra of aerosol extinction by the submicron component under conditions of "dry" (*1*) and "wet" (*2*) atmosphere.

The spectra of the component $\beta_a(\lambda) = K_4(\lambda)M_{BC}$ caused by absorption of radiation by aerosol containing black carbon, calculated from Eq. (2), are shown in Fig. 7.

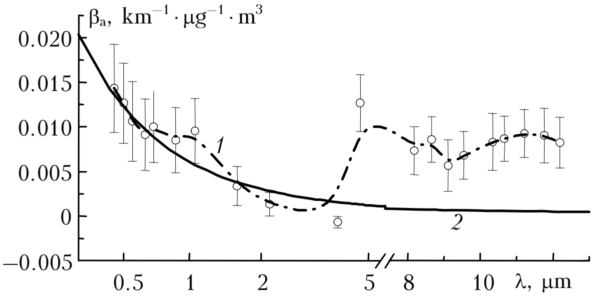

Fig. 7. Mean spectrum of the absorption coefficient of aerosol containing black carbon under conditions of "dry" atmosphere (curve *1*) and approximation of the dependence by $1/\lambda$ (curve *2*).

Here we consider only conditions of low air humidity, when this dependence has been better pronounced. It is seen that the well-pronounced decrease of the absorption coefficient with the wavelength is observed in the wavelength range from 0.45 to 4 μm. Such a shape of the $\beta_a(\lambda)$ curve, which can be approximately estimated as $1/\lambda$ (see curve *2*) is an evidence of the fact that this absorption is caused by particles of very small size.

The obtained result is qualitatively agrees with the conclusion,[10] where the spectrally continuous absorption of radiation by aerosol in the range from 0.44 to 4 μm was revealed by another technique based on the data of field measurements in the atmosphere of an arid zone (i.e., also under conditions of very low humidity).

Later, the fact that aerosol absorbs in the visible and near IR ranges was reliably revealed by means of optoacoustic spectrometry,[11] where the specific values of the absorption coefficients in the range from 0.53 to 1.06 μm are presented. Comparison of the absorption coefficients obtained in this paper with the data from Ref. 11 shows their good agreement. Then one can suppose that the statistical approach used in this paper can be applied to determination of the absorption coefficients of atmospheric aerosol in the wavelength range from ~0.45 to 4 μm under conditions of low air humidity.

As to the wavelength range from 8 to 12 μm, where noticeable increase of $\beta(\lambda)$ coefficient has been observed, additional investigations are necessary for clarification of this fact.

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