

## ON CONTINUOUS ATTENUATION OF OPTICAL RADIATION IN THE SHORT-WAVE SPECTRAL REGION

Yu.A. Pkhalagov, V.N. Uzhegov, and N.N. Shchelkanov

*Institute of Atmospheric Optics,  
Siberian Branch of the Russian Academy of Sciences, Tomsk  
Received December 16, 1997*

*Based on field measurement data on the atmospheric transmission spectra along the extended near-ground paths in an arid zone and in a region of the West Siberia, continuous absorption of radiation in the 0.44–3.97  $\mu\text{m}$  wavelength region has been revealed, being linearly dependent on the absolute air humidity. By the magnitude of the absorption coefficient (about 0.02–0.03  $\text{g}^{-1}\text{cm}^2$ ) it noticeably exceeds the continuous absorption by water vapor; and the character of its spectral dependence points to its relation to the finely dispersed soot aerosol. The absorption coefficients obtained on the near-ground and slant paths are being compared. The comparison made shows quite a satisfactory agreement between them. The presence of the finely dispersed soot aerosol in the atmosphere is assumed to be one of the important factors causing the anomalous absorption of optical radiation by clouds.*

1. In modern climatology there is an important problem to be addressed, namely, the nature of the anomalous (excess) absorption of short-wave optical radiation in clouds. The excess of absorption in clouds, as compared to that calculated using radiation models allowing for contributions from all gases, is often observed in field measurements of total radiation fluxes made above and under the clouds. It is evident that clear understanding of the nature of excess absorption is important when one needs for quantitative estimates of the main factors affecting the Earth's albedo and radiation balance under clear sky conditions and in a cloudy atmosphere.

At present there are several hypotheses to explain the nature of this phenomenon. Thus, in Ref. 1 the authors are reasoning that the excess absorption observed in clouds is a natural fact that follows from the horizontal radiation transfer in a stochastically inhomogeneous cloud. It is assumed that the discrepancy between the experiment and model calculations can be removed by averaging the experimental data over the paths about 6 km long. In some papers the excess absorption in clouds is related to some specific light-absorbing components that may present in the atmosphere. In particular in Refs. 2 and 3 the main cause of the anomalous absorption of short-wave radiation in clouds is assumed to be the fine aerosol fraction that may occur in the clouds between the droplets. The above aerosol fraction has a soot composition and does not manifest itself in scattering. According to this hypothesis the radiation absorption in clouds should be only weakly selective while noticeable throughout the optical range. This assumption agrees, on the whole, with the experimental data in Ref. 4 where

presented are spectral behaviors of the absorption coefficient of fine aerosol fraction, in the spectral range from 0.25 to 0.8  $\mu\text{m}$ , measured for aerosol samples taken in the mountains of Abastumani.

In Ref. 5, it is proposed that, apart from the absorbing aerosol, the anomalous absorption of short-wave radiation in clouds may also be explained by an increase in the contribution from molecular scattering because of the enhanced photon free path due to multiple scattering. In this case a stronger spectral dependence of the absorption coefficients should occur in the visible wavelength range. Finally, in Ref. 6 the excess radiation absorption in clouds is assumed to be due to continuous absorption of light by water vapor, in the short-wave spectral region. To estimate quantitatively this absorption we have used, in Ref. 6, an empirical formula proposed earlier in Ref. 7 for the spectral region 3.3 to 4.2  $\mu\text{m}$ .

The experimental evidences of the existence of weak absorption by water vapor in the wavelength region from 0.648 to 3.97  $\mu\text{m}$  have been obtained (see Ref. 8) under field conditions when measuring optical thickness of a cloudless atmosphere  $\tau(\lambda)$  along slant paths. The relative error of these measurements was about 25%. Absolute values of the absorption coefficient  $k(\lambda)$  were determined by the slope of the dependence  $\tau(\omega)$  (where  $\omega$  is the water vapor precipitated layer) and were from 0.015 to 0.067  $\text{g}^{-1}\text{cm}^2$ .

We have tried to elucidate the physical nature of the absorption observed, Ref. 8, as well as to calculate the contribution to the total absorption coming from the water-vapor continuous absorption component in this wavelength region, Ref. 9. In so doing we have assumed the continuous absorption in

the visible and near IR region, as well as in the region from 8 to 12  $\mu\text{m}$ , to occur mainly due to the absorption within the water vapor line wings. To estimate quantitatively continuous absorption we used the Lorentzian contour for a line wing at spectral distance of 500  $\text{cm}^{-1}$ .

Thus calculated values may be considered as the upper limit for the continuous absorption coefficient. These calculations have shown that, though overestimated, the values of the continuous absorption coefficients calculated are well below, by one or two orders of magnitude, the measured ones, Ref. 8. By this we mean that in a clear atmosphere and, consequently, in clouds the main contribution into the absorption of short-wave radiation comes not from the continuous absorption by water vapor but from some other substance absorbing that absorbs light.

2. It is normally expected that this substance is evenly mixed in the atmosphere, due to turbulent diffusion. So, in this paper we present an attempt made to identify the continuous absorption of short-wave radiation in the atmosphere from measurements only along a near-ground path where higher measurement precision may be achieved. It should be noted that in the general case isolation of continuous absorption in the short-wave spectral region, when measuring the atmospheric transmission  $T(\lambda)$  spectrum under field conditions, is too a complicated task. The matter is that it is very low and in the majority of cases completely masked by aerosol scattering of radiation on submicron particles. To minimize this factor, we need the data on the atmospheric transmission to be obtained in a clear atmosphere along an extended path. This is a necessary condition to increase the accuracy of  $T(\lambda)$  measurements.

3. Taking this into account we have compiled, to solve the above-stated problem, a spectral array of data on the coefficients of total extinction of radiation,  $\epsilon(\lambda)$ , in the wavelength region from 0.44 to 3.97  $\mu\text{m}$ . The data have been collected during the field campaigns to measure the atmospheric transmission near Balkhash lake, characterized by a very good visibility in warm seasons,<sup>10</sup> and near Tomsk city (Western Siberia) under anticyclonic conditions when the visual range exceeded 40 km.<sup>11</sup> The measurements of  $T(\lambda)$  were made using the instrumentation described in Refs. 12 and 13. To isolate necessary wavelengths, we have used either interference ( $\lambda = 0.44\text{--}2.2 \mu\text{m}$ ) filters or a combination of bandpass filters at  $\lambda > 3 \mu\text{m}$ . The filter halfwidth was about 0.010  $\mu\text{m}$  in the visible region, 0.015–0.020  $\mu\text{m}$  at  $\lambda$  from 0.8 to 2.2  $\mu\text{m}$  and 0.15  $\mu\text{m}$  at  $\lambda = 3.97 \mu\text{m}$ . The rms measurement error in  $T_\lambda$  (at  $T < 0.8$ ) did not exceed 2% in the spectral region from 0.44 to 1.06  $\mu\text{m}$  and 3% in the region from 1.06 to 3.97  $\mu\text{m}$ . Data acquired in different seasons (spring, summer, and fall) have also been included into the above data array to extend the range of variation of the absolute air humidity.

Thus compiled data array includes 35 series of measurement data on  $\epsilon(\lambda)$  and the corresponding data on the absolute air humidity,  $a$ . The latter was measured as an integral quantity along the path using a long-path absorption technique within the water vapor absorption band at  $\lambda = 0.94 \mu\text{m}$ . The integration over the sounding path provides for a higher measurement accuracy. The relative air humidity during the measurement period varied from 40 to 93%, the air temperature varied from  $-10^\circ\text{C}$  to  $+32^\circ\text{C}$ , the absolute air humidity was from 1.23 to 18.7  $\text{g}/\text{m}^3$ , and the wind velocity varied from 1.6 to 8.8  $\text{m}/\text{s}$ .

4. Based on the data obtained in our study and data from Ref. 8, the atmospheric extinction coefficient  $\epsilon(\omega)$  has been calculated for the wavelength  $\lambda = 0.44; 0.48; 0.55; 0.69; 0.87; 1.06; 1.22; 1.60; 2.17$  and  $3.97 \mu\text{m}$  as functions of the absolute air humidity. As an example, six of these dependences for Balkhash ( $\bullet$ ) and Tomsk ( $\circ$ ) are depicted in Fig. 1. It is evident that within the limits of the existing point spread the coefficients  $\epsilon(\omega)$  have, as in Ref. 8, positive correlation with the absolute air humidity. Statistical analysis made shows a significant correlation between these values (with the correlation coefficients of 0.88 to 0.96).

Within the limits of linear approximation and using the least squares method we have related the spectral coefficients of the total absorption  $k(\lambda)$  to water vapor content. Since measurements of the atmospheric transmission had been made with a finite spectral resolution, the calculated absorption coefficients were corrected for the selective absorption by water vapor. The calculations presented in Ref. 9 were made using the database from Ref. 14. As a result, the coefficients of continuous absorption  $k_{\text{cont}}(\lambda)$  were obtained, which represent the "atmospheric continuumB (according to terminology from Ref. 9).

The spectral values of the absorption coefficients and corresponding coefficients of the continuous absorption are given in the Table I. The values  $k_{\text{cont}}(\lambda)$  calculated for close wavelengths using data from Ref. 8 are also given in this table for a comparison. The comparison between these data shows that the absolute values of the continuous absorption coefficients are in a good agreement with the data from Ref. 8. The exception is only observed at several points near the ends of the range considered that may be due to the lack of data available in Ref. 8 for the proper account of the selective transmission component in these spectral regions. In this case it is of certain interest to note the closeness of the absorption coefficients values obtained on the near-surface and slant paths that is indicative of a good vertical mixing of the absorbing substance. However, the question on the most probable nature of this substance is still to be addressed.<sup>8</sup>

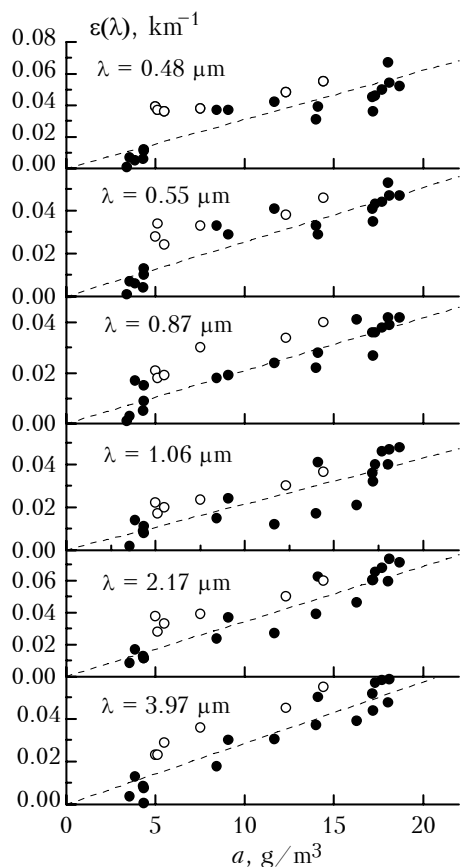


FIG. 1. The dependence of the total extinction coefficients on the absolute air humidity at six wavelengths according to data obtained near lake Balkhash (●) and in Tomsk (○). The direct regressions are obtained using the method of least squares with the extrapolation to “0B

TABLE I. The values of the total absorption coefficient  $k(\lambda)$  and the corresponding coefficients of continuous absorption  $k_{cont}(\lambda)$  [ $g^{-1} \cdot cm^2$ ] obtained in this paper and in Ref. 8.

Wavelength $\lambda, \mu m$	$k(\lambda)$	$k_{cont}(\lambda)$ (this paper)	$k_{cont}(\lambda)$ (Ref. 8)
0.44	0.028	0.028	—
0.48	0.028	0.028	—
0.55	0.025	0.025	—
0.65	—	—	0.040
0.69	0.028	0.024	0.028
0.87	0.0205	0.0205	0.018
1.06	0.021	0.021	0.018
1.22	0.032	0.022	0.023
1.60	0.021	0.0204	0.016
2.17	0.0345	0.021	0.018
3.97	0.029	0.021	0.083

As was already mentioned, the calculations of continuous absorption by water vapor in the short-wave spectral region give the values, which are two orders of

magnitude smaller than the measured values of  $k_{cont}(\lambda)$ . Therefore, to make physical reasoning concerning the nature of the atmospheric continuum some other information is needed. In this connection, it is interesting to analyze the spectral dependence of the coefficients of continuous absorption that is depicted in Fig. 2 (curve 1). Figure 2 shows that the atmospheric continuum decreases markedly with increasing wavelength in the visible spectral region while being practically unchanged in the infrared one. From the data for the visible region,<sup>4</sup> one can see that the absorption spectra of the finely dispersed soot aerosol, which does not manifest itself in light scattering, are similar. One of these spectra, obtained for the wavelength region from 0.37 to 0.63  $\mu m$ , is shown in Fig. 2 (curve 2) in relative values. It is evident that in the overlapping spectral interval (0.44 to 0.63  $\mu m$ ) curves 1 and 2 have qualitatively same spectral dependence.

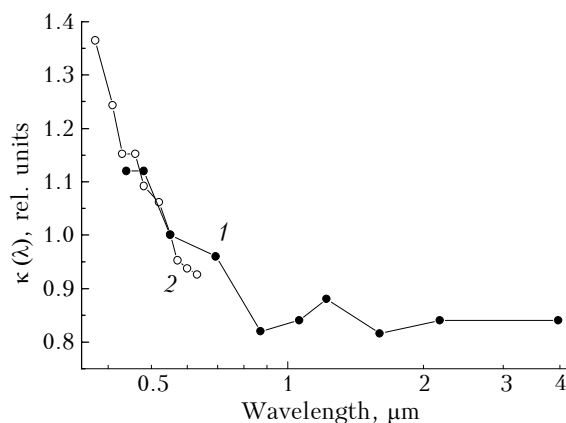


FIG. 2. The spectral dependence of the coefficient of continuous attenuation in the short-wave region for the surface atmosphere (curve 1) and finely dispersed aerosol containing the soot component<sup>4</sup> (curve 2).

Taking into account the fact that in the IR region the absorption by fine aerosol fraction is practically neutral in the entire spectral range (except for the absorption bands), it is supposed that the soot aerosol plays the main role in formation of the atmospheric continuum. Thus the aerosol concentration is established to be related to the absolute air humidity.

This interpretation agrees with the hypothesis proposed in Refs. 2 and 3, according to which the soot component of finely dispersed aerosol is an important radiation factor in the short-wave spectral region. Besides, the assumption made in Ref. 3 that the finely dispersed aerosol is accumulated on water clusters that are stable, in the thermodynamic sense, can explain the connection between the continuous extinction and the absolute air humidity. The conclusions, drawn in Ref. 15, show a strong interest in this interpretation. The latter paper suggests that there exists in the atmosphere a “grey” nonselectively absorbing substance. This absorber is a very important climatic

factor under clear sky conditions. A major contribution of absorbing tropospheric aerosol to the decrease of albedo of the "atmosphere – underlying surface" system was also mentioned in Ref. 16.

One more, though very close, physical mechanism of forming the atmospheric continuum can be the absorption of short-wave radiation by fractal structures consisting of water clusters and crystal carbon (soot). This assumption is based on the data from Ref. 17 where the authors consider the relative spectral behavior of the absorption coefficient in the short-wave region, in such a system, to be in a good agreement with the spectral behavior of the coefficient of continuous extinction depicted in Fig. 2.

Thus, the following conclusions may be drawn from what just have been discussed above. The measurements of the atmospheric transmission spectra along a near-ground path enabled us to prove the existence of the atmospheric absorption continuum in the visible and near IR regions. As to the value of the extinction coefficient, being equal to 0.02 to 0.03 g<sup>-1</sup> cm<sup>2</sup>, its magnitude for the atmospheric continuum is about two orders higher than that due to water vapor continuous absorption. The nature of the spectral structure of the atmospheric continuum shows that it is due to absorption by the finely dispersed soot aerosol. A good quantitative agreement of the absorption coefficients obtained in the surface layer (the present paper) with those obtained along a slant path, Ref. 8, points to a homogeneous distribution of the absorbing substance through the entire troposphere that indirectly supports the assumption that the radiation is mainly absorbed by the finely dispersed soot aerosol. In the case when the finely dispersed absorbing aerosol is in between the cloud droplets the absorbing aerosol can be one of the main factors of anomalous absorption of optical radiation in clouds due to the multiple scattering process that increases the photon free path.

#### ACKNOWLEDGMENT

This work has been financially supported by the Russian Foundation for Basic Researches (grant 97-05-65994).

#### REFERENCES

1. G.A. Titov, *Atmos. Oceanic Opt.* **9**, No. 10, 833–838 (1996).
2. G.V. Rozenberg, *Fizika Atmosfery i Okeana* **9**, No. 5, 460–470 (1973).
3. G.V. Rozenberg, *Fizika Atmosfery i Okeana* **18**, No. 11, 1192–1198 (1982).
4. Yu.S. Lyubovtseva and L.G. Yaskovich, *Fizika Atmosfery i Okeana* **18**, No. 9, 922–932 (1982).
5. K.Ya. Kondratiev, V.I. Binenko and I.N. Melnikova, *Meteorol. Gidrol.*, No. 2, 14–23 (1996).
6. G.L. Stephens and Si-Chee Tsay, *Quart. J. Roy. Meteorol. Soc.* **116**, 671–704 (1990).
7. M.E. Thomas and C.I. Delaye, in: *Proc. of the 14-th Annual Review Conference on Atmospheric Transmission Model* (1991), pp. 342–349.
8. C. Tomasi, R. Guzzi, and O. Vittori, *J. Atmos. Sci.* **31**, 255–260 (1974).
9. L.I. Nesmelova, O.B. Rodimova, and S.D. Tvorogov, *Atmos Oceanic Opt.* **10**, No. 2, 81–83 (1997).
10. Yu.A. Pkhalagov, V.U. Uzhegov, and N.N. Shchelkanov, *Atmos. Oceanic Opt.* **7**, No. 10, 714–720 (1994).
11. Yu.A. Pkhalagov, V.N. Uzhegov, and N.U. Shchelkanov, *Atmos. Oceanic Opt.* **9**, No. 6, 455–458 (1996).
12. A.N. Volkov, Yu.A. Pkhalagov, V.N. Uzhegov, and N.N. Shchelkanov, in: *Proceedings of the 8-th All-Union Symposium on Laser Radiation Propagation in the Atmosphere*, (Tomsk, 1986), pp. 228–231.
13. Ya.A. Pkhalagov, V.N. Uzhegov, and N.N. Shchelkanov, *Atmos. Oceanic Opt.* **5**, No. 6, 423–426 (1992).
14. L.S. Rothman, R.R. Gamache, R.H. Tipping, et al., *J. Quant. Spectrosc. Radiat. Transfer* **48**, 469–507 (1992).
15. R. Eiden and G. Eschelbach, *Z. Geophys.* **39**, 189–228 (1973).
16. T.A. Tarasova and E.M. Feigelson, *Atmospheric and Oceanic Physics*, **18**, No. 11, 1199–1206 (1982).
17. S.D. Andreev and E.F. Mikhailov, *Atmospheric and Oceanic Physics* **32**, No. 6, 808–817 (1996).