ON THE NATURE OF ANOMALOUS ABSORPTION OF SHORT-WAVE RADIATION BY THE ATMOSPHERE

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Continuous attenuation of radiation in the region 0.44–3.97 μm *having a positive linear relation to the absolute air humidity has been revealed based on the measurements of atmospheric spectral transmission along the extended horizontal near-ground paths. The comparison made among the coefficients of continuous attenuation at the near-ground and slant paths shows their satisfactory agreement. To elucidate the physical nature of the attenuation under discussion, the line-by-line absorption coefficients of the* m_2O–N_2 *mixture in the spectral region of* 9450–9475 cm^{-1} *were calculated using some empirical and theoretical expressions for the line shape. It is shown that the measured coefficients of the continuous attenuation being about* 0.02–0.03 $g^{-1}cm^2$ *significantly exceed the calculated level of the spectroscopic continuum. The form of their spectral dependence points to some connection between the atmospheric continuum and finely dispersed soot aerosol. It is expected that the presence of the finely dispersed soot aerosol in the atmosphere may be one of the important factors causing the anomalous absorption of optical radiation by clouds.*

1. INTRODUCTION

In recent years the problem of the anomalous (excess) absorption of the short-wave optical radiation by clouds has been widely debated when studying the Earth's radiation balance. As a matter of fact, the radiation absorption measured above and under the clouds exceeds the absorption calculated with the use of the radiation models allowing for a contribution of all the absorbing gases. It is obvious that the elucidation of the nature of the excess absorption is of great importance for creating climatic models which adequately take into account the main factors affecting the Earth's albedo and determining its radiation balance both in the cloudy and cloud free atmosphere.

Now there exist several hypotheses for explanation of the nature of the anomalous absorption. One of them assumes that the excess absorption observed in clouds is in reality apparent and related to the horizontal radiation transfer in the stochastically inhomogeneous clouds.[1] Doctor G.A. Titov also followed this hypothesis. He studied[2,3] the influence of the horizontal transfer on the radiation absorption in inhomogeneous stratocumulus clouds on the base of mathematical modeling. In the context of this hypothesis a discrepancy between the experimental and simulated data can be removed by spatial averaging of experimental data over the intervals of ~ 6 km and larger. Model estimates of the influence of the clouds' random geometry on the mean absorption of short-wave radiation in the atmosphere under broken cloud conditions[4] have shown that the random geometry of clouds does not affect essentially the absorption of the short-wave radiation in the atmosphere. This means that the excess absorption in the stochastically inhomogeneous clouds can not be explained only by the horizontal radiative transfer.

A number of authors attribute this effect to the presence, in the atmosphere, of really absorbing components. A finely dispersed aerosol fraction mainly consisting of the soot is treated in Refs. 5 and 6 as such a component. It is considered to be present in inter-drop space of a cloud and does not manifest itself in the scattering. The radiation absorption, according to this hypothesis, should be slightly selective and observable in the whole optical wavelength region. The

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absorption spectra of the finely dispersed aerosol presented in Ref. 7 for the region $\lambda = 0.25–0.8$ μm on the whole are consistent with the above assumption. It is shown in Ref. 8 on the base of the model of a multilayer concentrically inhomogeneous water drop that the anomalous absorption of the short-wave radiation in clouds may be related to non-additive light absorption by particles of atmospheric haze as a part of the liquid-drop clouds. It follows from the calculations that the aerosol particles concentrated at the center of drops absorb visible radiation more efficiently (by a factor of 1.5–3). It is supposed in Ref. 9 that besides the aerosol, a noticeable part in anomalous absorption of the short-wave radiation in clouds is played by an increase of the molecular scattering contribution due to an increase of the mean free path of photons at multiple scattering. This fact should manifest itself in an increase of the spectral dependence of the absorption coefficients in the visible region.

Finally, in Ref. 10 the excess radiation absorption by clouds is associated with the presence of a continuous absorption by water vapor in the short-wave spectral region. In this case the excess absorption can exist in the cloud free atmosphere as well, and the absorption coefficients must depend on the partial pressure of the water vapor. Weak water vapor absorption in the region of 0.648–3.97 μm was found experimentally in measurements of optical density of the cloud free atmosphere $\tau(\lambda)$ along slant paths[11] (a relative error of measurements was ~25 %). Absolute values of the spectral absorption coefficients $k(\lambda)$ were determined from the slope angle of the curve $\tau(\omega)$ (where ω is the precipitated layer of the water vapor) and ranged from 0.015 to 0.067 $g^{-1}\cdot cm^2$.

Some data on laboratory measurements with the absorption cells are also available from which a weak absorption of the short-wave radiation has been revealed. Thus, in Ref. 12 the value of the absorption coefficient $k = 6\cdot10^{-10}$ cm^{-1} at the frequency $\nu = 9466$ cm^{-1} is given which is obtained with the use of an interferometric calorimeter under partial pressure of water vapor $P_{H_2O} = 16.5$ Torr, the pressure $P_{N_2} = 1$ atm of nitrogen as a broadening gas and the temperature $t = 30°$q. In Ref. 13 the air transmission was measured by methods of laser spectrophotometry and laser opto-acoustic spectroscopy in the spectral region of 0.5–1.35 μm at the partial pressure of water vapor from 5 to 15 Torr and the upper boundary of the water vapor absorption coefficient of 10^{-7} cm^{-1}.

Starting from 1996 the cycle of investigations has been pioneered by G.A. Titov at the Institute of Atmospheric Optics into the problem under discussion. The objective was to estimate a magnitude of the continuous attenuation of the short-wave radiation in the real cloud free atmosphere (the atmospheric continuum) based on the data from Ref. 11 and our experimental data and thus to answer the question if the excess absorption can be explained by the continuous absorption by water vapor (the spectroscopic continuum) as well as to draw some inference about its physical nature. These investigations were based, on the one hand, on the long-term study of the aerosol attenuation of optical radiation in the near-ground atmosphere[14,15] and, on the other hand, on the theoretical treatment of the spectral line shape behavior in far wings (see, for example, Refs. 16 and 17). The results obtained may be found in Refs. 18–21 and are analyzed in the present paper.

2. RESULTS OF FIELD EXPERIMENT ON SEPARATING OUT THE ATMOSPHERIC CONTINUUM

The data from Ref. 11 were obtained under cloudless atmospheric conditions. Therefore, it may be considered with a high degree of confidence that in this case the continuous attenuation of short-wave radiation is produced by some absorbing substance. Assuming that this substance is uniformly mixed in the atmosphere due to the turbulent diffusion we undertook an attempt to separate a continuous absorption of the short-wave radiation out of the data on spectral atmospheric transmission $T(\lambda)$ along the ground path, where more accurate measurements can be performed.

It should be noted that under field conditions the separation of a continuous absorption in the short-wave region of the spectrum is a difficult problem because it is very weak, and in the majority of cases it is masked by light scattering from submicron aerosol particles. To minimize this factor, the data on atmospheric attenuation are required, which were obtained under very clean conditions on a sufficiently long path. The latter condition is necessary to increase the accuracy of the $T(\lambda)$ determination.

To solve the problem stated, a set of spectral coefficients $\varepsilon(\lambda)$ of the radiation attenuation in the region of $\lambda = 0.44–3.97$ μm was formed. We have obtained the coefficients from atmospheric transmission measurements under conditions of very high meteorological visibility range in the coastal zone of the Balkhash lake[14] and in Tomsk region.[15] The measurements of $T(\lambda)$ were carried out in the atmospheric transmission windows using the instrumental systems described in Refs. 22 and 23, respectively. To fix the necessary wavelength, the interference ($\lambda = 0.44–2.2$ μm) and combined ($\lambda > 3$ μm) filters were used. The filters' half-width was about 0.010 μm in the visible region, 0.015–0.020 μm in the region of $\lambda = 0.8–2.2$ μm, and 0.15 μm in the region of $\lambda = 3.97$ μm. The root-mean-square error of measurements T_λ (at $T < 0.8$) did not exceed 2% in the region of $\lambda = 0.44–1.06$ μm and 3% in the region of

$\lambda = 1.06–3.97$ μm. To extend the limits of the absolute air humidity variation, the realizations obtained in Ref. 14 for different seasons (spring, summer, and fall) were included into the set under consideration.

Thus formed set involved 35 realizations of $\varepsilon(\lambda)$ and the corresponding values of the absolute air humidity a. To improve the accuracy, the absolute humidity was measured along the whole path by optical method within the absorption band of water vapor in the region of $\lambda = 0.94$ μm. The relative air humidity during the measurements varied from 40 to 93%, the ranges of the temperature, absolute humidity, and wind velocity variation were from 10 to +32°C, from 1.23 to 18.7 g/m^3, and from 1.6 to 8.8 m/s, respectively.

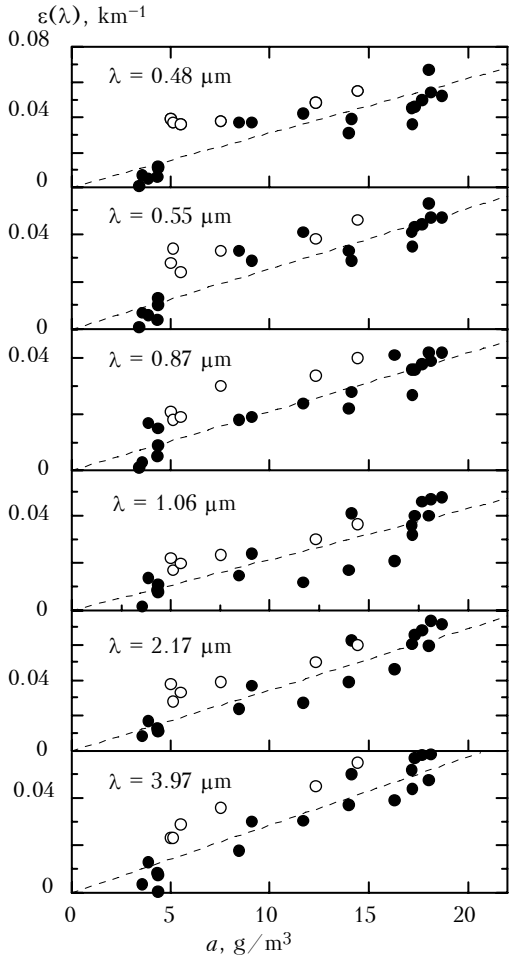

FIG. 1. The attenuation coefficients vs. the absolute air humidity at the six wavelengths measured near the Balkhash Lake (•) and in Tomsk (○). The direct regressions are calculated by the least-square method with extrapolation into the zero point.

Using the obtained data, the dependence of the attenuation coefficients $\varepsilon(\lambda)$ on the absolute humidity was constructed for the wavelengths of 0.44; 0.48; 0.55; 0.69; 0.87; 1.06; 1.22; 1.60; 2.17; and 3.97 μm. The results of these investigations were presented in Ref. 21. As an illustration, the functions $\varepsilon_\lambda(a)$ for some wavelengths are presented in Fig. 1 using the Balkhash (•) and Tomsk (○) data. It can be seen, that, as in Ref. 11, within the limits of the points' scatter the coefficients $\varepsilon(\lambda)$ have positive relation to the absolute air humidity. Statistical analysis has shown a significant correlation between these parameters (the correlation coefficients were equal to 0.88–0.96). Spectral coefficients $k(\lambda)$ of the total absorption related to the water vapor were found by the least squares method using linear approximation.

The selective water vapor absorption was then subtracted from the absorption coefficients at the given values of air humidity and temperature obtained above. The calculation was made[20] using the spectroscopic database[24] and the Lorentzian line profile cut at the distance $\Delta\nu = 20$ cm^{-1} from the line center. As a result the coefficients of the continuous attenuation $k_{\text{cont}}(\lambda)$ were derived which present solely the atmospheric continuum.

Absolute values of the spectral coefficients of the total and continuous attenuation from Refs. 11 and 21 are listed in the Table I and shown in Fig. 2 (curves *1* and *2*). It can be seen that the continuous attenuation coefficients found from the measurements along the near-ground and slant paths correlate quite well (except for some points on the borders of the region). This fact shows a good spatial mixing of the absorbing substance.

TABLE I. Coefficients of the continuous attenuation $k_{\text{cont}}(\lambda)$, g^{-1} · cm^2 *(from Ref. 11 and 21).*

Wavelength λ, μm	k_{cont} (λ) (Ref. 21)	$k_{\text{cont}}(\lambda)$ (Ref. 11)
0.44	0.028	–
0.48	0.028	–
0.55	0.025	–
0.65	–	0.040
0.69	0.024	0.028
0.87	0.0205	0.018
1.06	0.021	0.018
1.22	0.022	0.023
1.60	0.0204	0.016
2.17	0.021	0.018
3.97	0.021	0.083

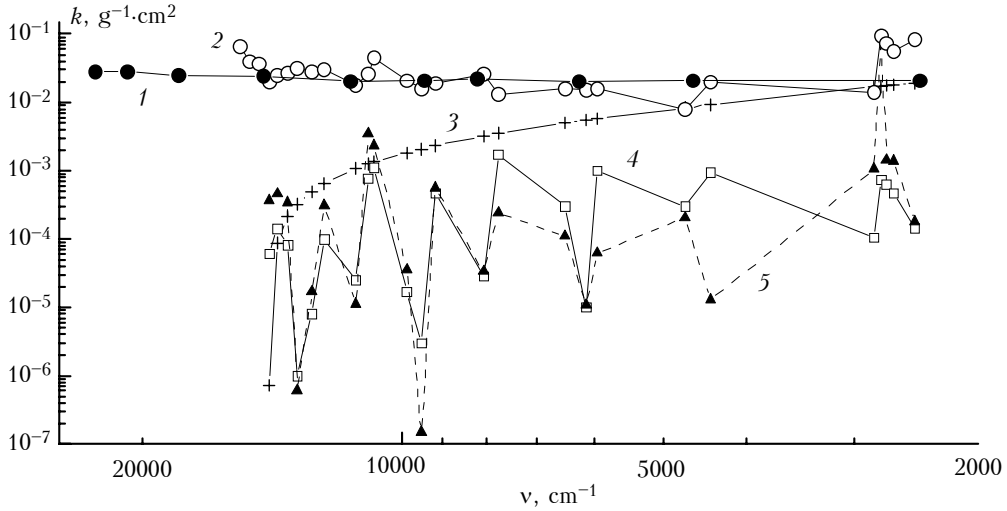

FIG. 2. Special coefficients of continuous attenuation: field experiments (1, 2) along near-ground[21] and slant[11] paths, respectively; continuous absorption calculated by the technique from Ref. 9 (3); the Lorentzian spectroscopic continuum calculated up to 500 cm^{-1} *from the line center[18,20] (4); the spectroscopic continuum calculated using the Clough profile[25,26] (5).*

3. CALCULATION OF THE MOLECULAR ABSORPTION

The available experimental data are indicative of an essential contribution of water vapor into the anomalous absorption. Therefore it was necessary to treat first of all the versions of the phenomenon explanation on the base of molecular absorption. The continuum determined by the molecular absorption (in the case under consideration it is the absorption in line wings) can be referred to as "spectroscopic" continuum.

It is well known that the contour of the vibrational-rotational lines has a non-Lorentzian shape at a distance far from their centers and at large distances it is characterized by an exponential decay. Detailed calculations of the water vapor line shape for the bands localized in the visible and near IR regions of the spectrum are rather cumbersome. Therefore it was worthwhile to make some preliminary estimates. For this purpose the water vapor absorption coefficient k_{Lor} was calculated[18,20] for the case of broadening by the nitrogen with the Lorentzian line shape cut at the distance $\Delta\nu \cong 500$ cm^{-1} from the line center. Since the variations from the Lorentzian contour to the exponential decay were observed at the frequency detuning less than 500 cm^{-1}, the calculated values k_{Lor} can be considered as the upper boundary of the absorption coefficient. The results of the k_{Lor} calculations are shown in Fig. 2 (curve *4*). Note, that the points, for which the calculations were performed, are connected with the straight lines only for the sake of visualization. It can be seen that the calculation even with the Lorentzian line wings results in significantly lower absorption coefficients than the experimental data.[11,21] The calculations by the procedure from Ref. 9 (curve *3*) and the calculations using the Clough line shape[25,26] (curve *5*) also give strongly underestimated values of the continuous absorption coefficients. Hence, it follows from this that the absorption in the far wings of the water vapor absorption lines in the visible and near IR regions is too low to explain the observed absorption.

The change of the line shape should somehow change the calculated values. The influence of the line shape on the value of the absorption coefficient was studied in Ref. 19. Note that the line shape obtained for the 8–12 μm region is used most frequently in calculations of the water vapor absorption in the visible and near IR spectral regions using both theoretical line shapes and empirical relationships.

Generally speaking, the spectral line shape changes from one band to another and depends on the type of a broadening gas. This fact is beyond the question if we deal with the CO_2 spectrum in the IR region. In the case of the water vapor absorption, the change of the line shape from one band to another only begins to be recognized. We know only one paper that applies different line shapes in different absorption bands of the water vapor. It is the paper by Thomas and Nordstrom[23] who obtained their empirical contours for the rotational, ν_2-, and $\nu_1\,\nu_3$ bands. Their results show that the deviations from the Lorentzian line shape can increase with the wavelength decrease.

The calculation was performed using the line-by-line method and a number of empirical and theoretical line shapes from the 9450–9475 cm^{-1} region where the laboratory and field data were available. The results of calculation of the H_2O absorption coefficients with N_2 as a broadening gas in this spectral region at the temperature T = 296 K and pressures P_{H_2O} = 0.02 atm, $P_{H_2O + N_2}$ = 1 atm are depicted in Fig. 3. The absorption coefficients are presented for the cases of the Lorentzian line shape

cut at 20 and 1000 cm^{-1}, the Thomas-Nordstrom line shapes[27] obtained for three water vapor bands, the total Clough contour[25,26] and the line shape following from the line wing theory.[16,28] As to the latter one, this line shape was used previously as derived to describe the water vapor absorption at the N_2 broadening in the transmission window of 8–12 μm. The value of the absorption coefficient obtained with that line shape at the frequency of 9466 cm^{-1} is close to that estimated by Ma and Tipping for the sum of absorption coefficients due to the self- and the N_2-broadening.[29,30] When calculating each curve in Fig. 3, the line shape chosen was used for all lines from the observed region.

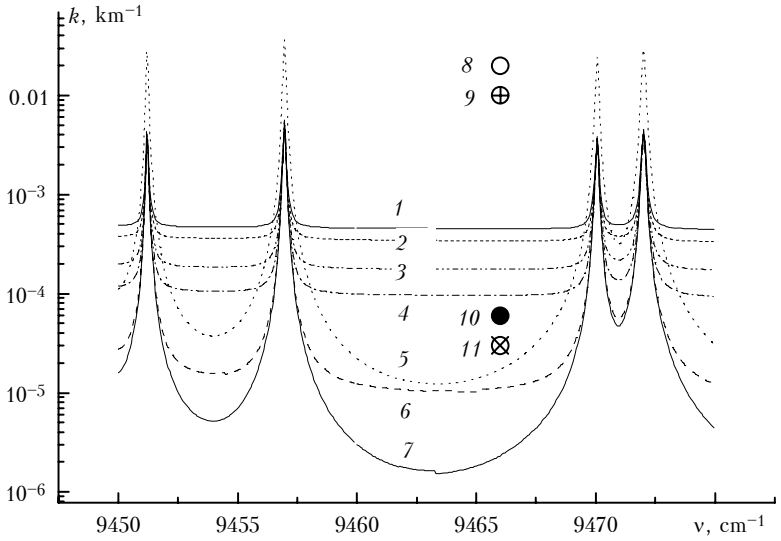

FIG. 3. The absorption coefficients in the H_2O–N_2 *mixture near* 9466 cm^{-1} *calculated using the following line profiles: the Tomas–Nodstrom line profile fitted to the* H_2O *absorption band (1); the Tomas–Nodstrom profile fitted to the* ν_1, ν_3 *band of* H_2O *(2); The Lorentzian profile cut at a distance of* 1000 cm^{-1} *from the line center (3); the Tomas–Nodstrom profile fitted to the* ν_2 *band of* H_2O *(4); the Clough profile (5); the profile following from the line wing theory (6); the Lorentzian profile cut at a distance of* 20 cm^{-1} *from the line center (7); the Ma and Tipping calculation[30] (11). The experimental data: field measurements[11,12] (8); measurements reported in Ref. 13 (9); laboratory measurements[12] (10).*

We see from Fig. 3, that the absorption measured in Ref. 12 can be well interpreted with the help of molecular absorption though for more confident conclusions it would be desirable to have additional experimental data on the transmission windows in the near IR region. However, the absorption retrieved from the field measurements[11,21] may be hardly described if changing a spectral line shape only. All the values calculated are distinctly lower than the absorption coefficients obtained from the field measurements.

Regular disagreement between the measured and calculated data makes us to assume the presence of an unknown factor causing the absorption of radiation under field conditions. Note, that the presence of this factor effects only the absorption in the transmission windows whereas the continuum even being equal to the experimental one makes a negligible contribution within the absorption bands.

4. TO AN INTERPRETATION OF THE ANOMALOUS ABSORPTION

As indicated earlier, the calculations of the water vapor continuous absorption in the short-wave spectral region result in the values that are two orders of magnitude lower than the measured values of $k_{cont}(\lambda)$. Therefore, it is necessary to consider other factors for physical explanation of the atmospheric continuum nature. In this connection it is of certain interest to analyze the spectral dependence of the above continuous absorption coefficients, which are shown in Fig. 4 in relative units (curve *1*).

It is seen from the figure that the atmospheric continuum noticeably decreases with increasing wavelength in the visible spectral region and is virtually unchanged in the IR range. According to the data from Ref. 7, the absorption spectra of finely dispersed aerosol including the soot component show a similar behavior in the visible range. For a comparison, one of these spectra in the wavelength range of 0.37–0.63 μm is presented (in relative units) in Fig. 4 (curve *2*). Note, that in the overlapping spectral ranges (0.44–0.63 μm) the curves *1* and *2* have qualitatively similar spectral behavior. Taking into account that the absorption by finely dispersed aerosol in the IR range is virtually independent of the wavelength (except within the absorption bands), it can be assumed, that the finely dispersed soot aerosol, the concentration of which is somehow related to the absolute air humidity, plays an important part in formation of the atmospheric absorption continuum. Similar interpretation is the

closest to the hypothesis proposed in Refs. 5 and 6 according to which it is just the soot component of the finely dispersed aerosol that is an important radiation factor in the short-wave spectral range. If the assumption made in Ref. 6 is adopted, that the accumulation of the finely dispersed aerosol occurs on the thermodynamically stable water clusters, then the connection between the continual absorption under discussion and the absolute air humidity becomes understandable. This interpretation is also confirmed by the conclusions drawn in Ref. 31, in which the belief is stated that there exists a non-selective "grayB absorber in the atmosphere which appears to be a climatic factor of great importance under clear sky conditions. A substantial contribution of the absorbing tropospheric aerosol to the decrease of the albedo of the "Earth – underlying surfaceB system is also noted in Ref. 32.

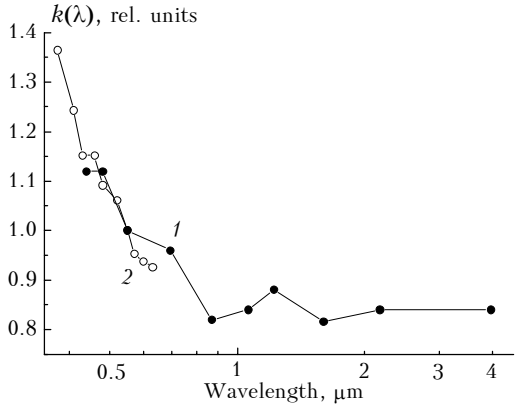

FIG. 4. Spectral dependence of the coefficient of continuous attenuation of the short-wave optical radiation in the near-ground atmosphere (curve 1) and the absorption spectrum of the fine-disperse aerosol containing the soot component[7] (curve 2).

Another physical way of formation of the atmospheric continuum is sufficiently close to the previous one and consists in absorbing the short-wave radiation by fractal structures including water clusters and crystal carbon (soot). This conclusion is based on the data from Ref. 33, where a relative spectral dependence of the absorption coefficient of the short wave radiation in such a system is presented. It agrees quite well with the spectral behavior of the continuous attenuation coefficient shown in Fig. 4.

5. CONCLUSION

The data, obtained from the measurements of the spectral atmospheric transmission along some near-ground path, show that the atmospheric continuum exists in the visible and near IR spectral ranges. It is characterized by the attenuation coefficient as large as 0.02–0.03 $g^{-1}\cdot cm^2$, being two orders of magnitude higher than the continuous absorption by water vapor. In its spectral structure it mostly corresponds to the radiation absorption by finely dispersed soot aerosol. Quite good quantitative agreement of the absorption coefficients measured in the near-ground layer (the present work) and along slant paths[11] points to the fact, that the absorbing substance is uniformly distributed in the troposphere, what indirectly supports the assumption of its finely dispersed structure. If the finely dispersed absorbing aerosol is distributed in the inter-droplet space of a cloud, then it can be one of the important factors of the anomalous absorption of the optical radiation in clouds at multiple scattering.

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