ANALYTICAL FORMULAS FOR DETERMINING OPTICAL PARAMETERS OF A CLOUD LAYER BASED ON THE MEASURED CHARACTERISTICS OF THE SOLAR RADIATION FIELD. 2. EXPERIMENTAL RESULTS

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The method of determination of the optical parameters of a cloud is applied to aircraft spectral radiation measurements in cloud layers and the spectral dependencies of the scattering and absorption coefficients are calculated for four experiments. A strong spectral dependence of the scattering coefficient and a high value of the absorption coefficient are obtained. The explanation of these facts and empirical formulas for obtaining true values of optical parameters of clouds are proposed. The obtained values of true absorption can explain the so–called "anomalous short–wave absorption" by clouds.

INTRODUCTION

The analytical formulas which express the absorption and optical thickness of a cloud layer in terms of a scattered solar radiation are applied to spectral measurements of fluxes at the boundaries of a cloud layer. The spectral dependences of the volume scattering and absorption coefficients in stratus have been calculated based on the interpretation of the aircraft measurements conducted during the four experiments over different underlying surfaces and in different geographic regions.[1-4] Based on the obtained data the enhancement of contributions from the molecular seattering and aerosol absorption due to multiple scattering on cloud drops is estimated. It is shown that the increase of aerosol and molecular absorption due to multiple scattering can result in recently discovered[5,6] so–called "anomalous short–wave absorption" in clouds. The analytical relations for determining the scattering and absorption coefficients from measured hemispherical radiation fluxes outgoing from a cloud layer have been derived in Ref. 7.

EXPERIMENTAL RESULTS AND THEIR INTERPRETATION

As was mentioned above, the results of four experiments were used in this paper to determine the scattering and absorption coefficients. Hemispherical incident and upwelling fluxes of solar radiation were measured above and below the stratus within the spectral range 0.350–0.950 μm. The spectral resolution was 0.002 μm, the measurement time for a single spectrum was 10 s. The upwelling and downwelling fluxes were measured using both two and one instruments. The single–instrument measurements were conducted with the help of a periscopic system which provids for a possibility of alternate measurement of the upwelling and downwelling fluxes.

Shown in Fig. 1 are the measurement results in relative units which were conducted:

a) on April 10, 1971 over the Black Sea surface at altitudes of 1 and 0.2 km. The zenith angle of the Sun was 35.1°. Two instruments were used, the measurement error was about 6%. The experiment was conducted within the framework of the KENEKS program. The radiation was measured simultaneously with the cloud moisture content (W = 0.39 g/m3) and the extinction coefficient (ε = 50 km^{-1}) We estimated the mean droplet radius r_0 = 7.5 μm and the droplet number density N = 200 cm^{-3}, (see Refs. 1 and 2). The albedo of the underlying surface is constant over the spectrum (A = 0.05)

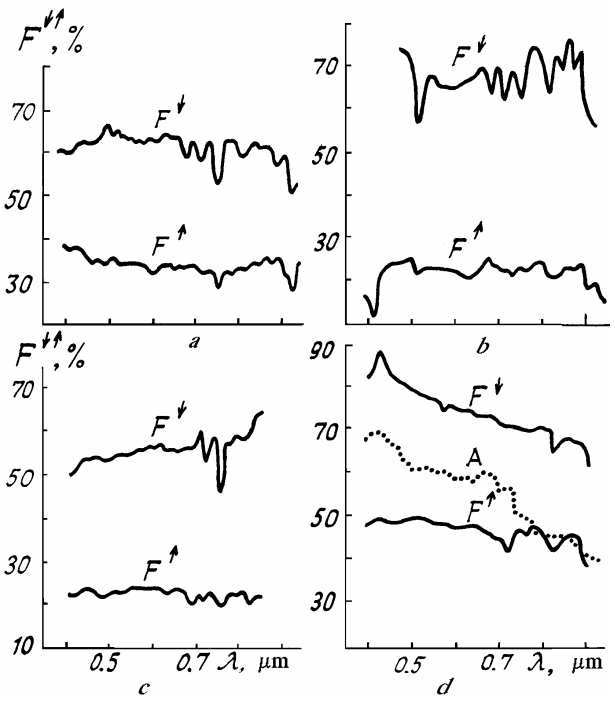

FIG. 1. Relative values of the solar radiation fluxes measured in the experiments (a), (b), (c), and (d) described in the text.

b) on May 10, 1972 over the Azov Sea surface at the altitudes 1.2 and 0.2 km, (see Refs. 1 and 2). The zenith angle of the Sun was 52°. Two spectrometers were used, the measurement error was ~ 6%. The cloudiness boundaries were at altitudes of 0.85 and 0.3 km. The extinction coefficient ε = 45 km^{-1}, and A = 0.06;

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c) on December 5, 1972 over the dry land near Rustavi at altitudes of 3.30 and 1.85 km, (see Ref. 3). The zenith angle of the Sun was 64°. Two spectrometers were used, the measurement error was 4%, $A = 0.06$; and,

d) on April 20, 1985 over Ladozhskoe Lake surface covered with ice and snow at altitudes of 1.7 and 0.2 km, (see Ref. 4). The cloudiness boundaries were at altitudes of 1.7 and 0.2 km, the measurement error was ~ 3%, the surface albedo varied over the spectrum, the zenith angle of the Sun was 49.7°.

The volume absorption κ and scattering σ coefficients were calculated using the formulas derived in Ref. 7. Thusly obtained spectral dependences $\kappa(\lambda)$ and $\sigma(\lambda)$ for four experiments are shown in Figs. 2 and 3.

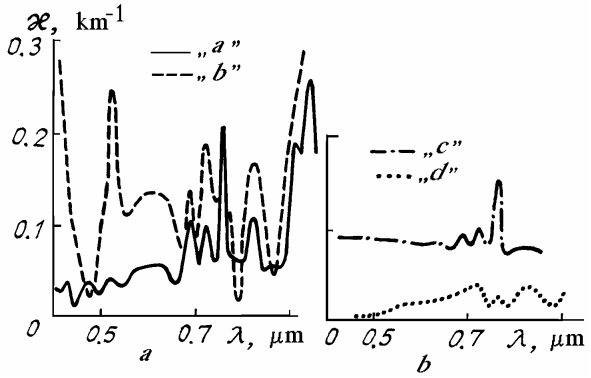

FIG. 2. Spectral dependences of the effective values of the absorption coefficient in cloud layers based on the results of the experiments (See (a), (b), (c),and (d)).

It should be noted that in determining the scattering coefficient the spectral behavior of the scattering phase function parameter g was taken into account according to Ref. 8. In the cases (*a*) and (*b*) when the measurements were accompanied by the measurements of the extinction coefficient it was possible to determine the quantity g. Therefore we have chosen the reference values $g = 0.87$ (*a*) and $g = 0.86$ (*b*) at the wavelength $\lambda = 0.55$ μm with the spectral behaviour $g(\lambda)$ taken from Ref. 8.

In the experiments (*a*), (*b*), and (*c*) the measurement errors are gross. Just these values determine the measurement errors in the volume coefficient. On the other hand, the absorption in the cloud layers is sufficiently strong too and the values of the influx are 0.15, 0.05, and 0.20, respectively. Calculations of the error using the formulas from Ref. 9 given $\sigma\kappa \sim 6\%$ and $\delta\sigma \sim 10\%$ for a) and b) and $\delta\kappa \sim 4\%$ and $\delta\sigma \sim 5\%$ for c). In more accurate measurements (*d*) the effect of the narrow range of the asymptotics applicability on the accuracy in determining the coefficients κ and σ is stronger $\delta\kappa \sim 2\%$ and $\delta\sigma \sim 4\%$. Here are presented the spectrum–averaged errors, the errors in determining the scattering coefficient markedly increase with the inrease of actual absorption and attain 12% in the regions of absorption bands.

Let us start the analysis of the results with Fig. 2 in which the spectral dependences of the volume absorption coefficients are shown. The O_2 and water vapor absorption bands can be clearly seen in all of the curves. In curves (*a*) and (*b*) there is a Chappeau band at the wavelength 0.6 μm. An approximate elimination of the gas absorption from the plots makes it possible to extract the absorption of light by an atmospheric aerosol (see Fig. 4). The strongest aerosol absorption caused by severe industrial emissions is observed in cases (*a*) and (*b*). The absence of spectral dependence (cases (*a*) and (*b*)) is indicative of the fact that the pollutant is carbon black. In the case (*d*) an insignificant increase in χ_a tindicates the presence of exhaust products of organic fuel combustion.[10]

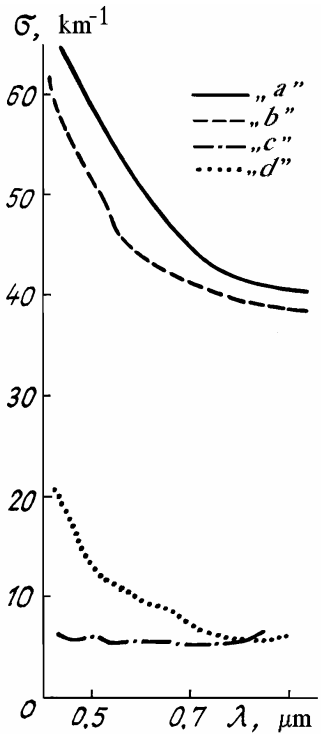

FIG. 3. Spectral dependences of the values of the scattering coefficient in cloud layers based on the results of the experiments (a), (b), (c), and (d).

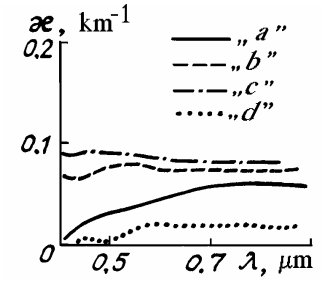

FIG. 4. The effective values of the aerosol absorption coefficients in clouds based on the experiments (a), (b), (c), and (d).

Let us now consider the volume scattering coefficient $\sigma(\lambda)$ shown in Fig. 3. The value of the scattering coefficient in the measurements conducted over the sea surface (cases (*a*) and (*b*)) is approximately twice as large as that measured over the ice in a slightly polluted atmosphere (case (*d*)) and about 8 times larger than that in the case (*c*) which is related to the measurements over a dry land in a strongly polluted atmosphere.

As is well known in cloud layers the scattering coefficient is related to the total water vapor content of the cloud W and can be represented by an empirical relation $\sigma \sim W(dr_0)^{-1}$ (Refs. 1 and 11), where d is the water density and r_0 is the mean size of droplets. As was mentioned above, in the case (*a*) the measurements gave

$W = 0.39$ g·m^{-1}, $r_0 = 7.5$ μm, and $\varepsilon = 50$ km^{-1}. In addition, in the cases (*a*) and (*b*) $g = 0.87$ and 0.86 that indicates that r_0 values in these cases are close. Therefore we may conclude that in the case (*b*) the total water vapor content is the same as that in the case (*a*): ($W \sim 40$ g/m^3). The lower values of σ in the cases (*c*) and (*d*) can be obviously related to the lower moisture content of the clouds over the land than of the clouds over the water surface.

SPECTRAL BEHAVIOR OF THE VOLUME SCATTERING COEFFICIENT

As can be seen from Fig. 3, the spectral behavior of the scattering coefficient is missing only for a strongly absorbing and weakly scattering cloud (*c*). In the case of the weak absorption (*d*) the spectral behavior is very strong and very close to λ^{-4}. At the same time, the numerical simulations made by many authors[12,13] do not show any wavelength dependence of the optical thickness of a cloud layer τ_0 (and related to it scattering coefficient) or reveal a slight increase of τ_0 with λ.

Let us now explain the aforementioned effect. In calculations of the radiation field in a cloud and in description of a multiple scattering process a cloud layer is considered as additively superimposed on the molecular atmosphere. Normally the molecular scattering is not taken into account since the molecular (Rayleigh) scattering coefficient is approximately two orders of magnitude smaller than the coefficient of scattering by droplets. But because of the multiple scattering in cloud with large optical thickness the mean number of collisions of a photon passing through a purely scattering layer is proportional to τ_0^2. Thus the photon travel path in a cloud is markedly enhanced in comparison with a clear atmosphere and the number of collisions with molecules of the air increases. The contribution from molecular scattering in a cloud increases as in a cell with reflecting walls.

On the other hand, the absorption removes a portion of photons from this process and attenuates the effect of molecular scattering amplification. Therefore it is possible to state that the cloud layer is not merely superimposed on the molecular atmosphere but affects the process of light scattering by molecules through additional contribution to its intensity due to multiple scattering. The same considerations are valid in the case of dry aerosol particles.

It is clear that the theory of multiple light scattering and the radiation transfer equation take into account all scattering and absorption processes but only if they are allowed for in the model of a scattering medium. Since the asymptotic formulas for the radiation fluxes are a solution of the radiation transfer equation for a medium in which molecular scattering and aerosol absorption are neglected they describe the relations between the fluxes (intensities) of radiation and the parameters of the medium which consists only of a single component, i.e., cloud droplets (of course, the apsorption of light by droplets is accounted for). By inverting the asymptotic formulas we obtain the corresponding relations for the medium parameters (scattering and absorption) in terms of characteristics of the radiation field with the same drawbacks, and by substituting the values of fluxes (intensities) into them we obtain not the scattering and absorption coefficients describing a real unit volume but some " effective" values .

Let us now consider a possibility of inverting the "effective" values of σ and κ measured experimentally on actual values which could be obtained if we were primarily based on the transfer equation describing a multicomponent medium. The scattering (absorption) coefficient is usually written as a sum of scattering (absorption) coefficients of corresponding components. For example, in the visible spectral region, neglecting the aforementioned effect, we have

$$\sigma = \sigma_m + \sigma_a + \sigma_d\ ;\quad \kappa = \kappa_m + \kappa_a\ . \tag{1}$$

Since we have a combined effect of scattering and absorption by different components it is possible to write the empirical expressions relations

$$\sigma = (\sigma_R + \sigma_a)\,\tau_d^p\,\Lambda^q + \sigma_d;\quad k = (k_m + k_a)\,\tau_d^p\,\Lambda^q\ . \tag{2}$$

In these relations there is no any factor at the coefficient of scattering by droplets because the effect of multiple scattering on droplets has already been taken into account in the initial asymptotic formulas for σ_d. The products $\sigma_R\tau_d^p\Lambda^q$, $\sigma_a\tau_d^p\Lambda^q$, $\kappa_m\tau_d^p\Lambda^q$ and $\kappa_a\tau_d^p\Lambda^q$ can be called the effective coefficients of scattering and absorption. The term $\kappa_m\tau_d^p\Lambda^q$ in the second relation of Eqs. (2) differs from 0 in the region of molecular absorption bands, σ_R is the coefficient of Rayleigh scattering at the corresponding wavelength and altitude in the atmosphere. It should be noted that all above considerations have been made for the case of large optical depth $\tau_0 \gg 1$.

Let us estimate the power factor *p*. In the case (*d*) for $\lambda = 0.42$ μm $\kappa = 0$ (Fig. 3). The values σ in a cloud are 18.5 km^{-1} and $\sigma_R = 0.035$ km^{-1} at an altitude of 0.6 km from the surface. At the wavelength $\lambda = 0.8$ μm $\sigma_R = 0.002$ km^{-1} and $\sigma = 9.3$ km^{-1}, i.e., this σ value can be entirely referred to scattering by dry aerosol by particles and cloud droplets. We then have $\tau_d = 14$, $\sigma_M(\lambda) = 9.2$ km^{-1}. Based on relations (2) we can write

$$p = \ln(\sigma_m(\lambda)/\sigma_R)/\ln\tau_d = 2.1 \tag{3}$$

which is in a satisfactory agreement with the above–mentioned fact that the mean number of photon collisions in a cloud is $\sim\tau_d^2$.

The power factor *q* in formulas (2) can be estimated from the curve (*c*) in Fig. 3. In the spectral region $\lambda = 0.5$–0.7 μm the scattering and absorption coefficients are constant $\sigma = 6.0$ km^{-1}, $\kappa = 0.09$ km^{-1}, at $z = 1.45$ km, $\tau_0 = \tau_d = 8.41$. The coefficient of Rayleigh scattering at $\lambda = 0.5$ μm at an altitude of 2 km $\sigma_R = 0.013$ km^{-1}. Since in this case the scattering coefficient is independent of the wavelength it is possible to assume that the product $\sigma_R\tau_d^2\Lambda^q$ does not exceed the error of determination of the scattering coefficient σ which attains 5% or 0.3, $\sigma_R\tau_d^p\Lambda^q$, from which we have

$$q \le \ln(0.3\sigma_R^{-1}\,\tau_0^{-2})/\ln\Lambda\ . \tag{4}$$

Since $\Lambda = 1 - s^2\cdot(3 - x_1) = 0.9846$, it is easy to calculate that $q \le 72.1$.

On the other hand, from the physical meaning of the value Λ is the probability of a photon servival at a single scattering act, and taking into account the fact that the probability of a group of independent events is the product of the probabilities we obtain $q = 70.7$ which is close to the estimate obtained using the experimental data.

However, Eqs. (2) also involve the term which takes the scattering by dry aerosol particles into account.

Assuming that $q = \tau_d^2$ one can estimate the contribution coming from dry aerosol to the scattering using the formula $\sigma_a \le 0.3\ \tau_d^2\ \Lambda_0^S - \sigma_R = 0.012\ \text{km}^{-1}$.

Thus dry aerosol in a polluted atmosphere gives the effect which can be compared with the molecular scattering as noted previously.[1]

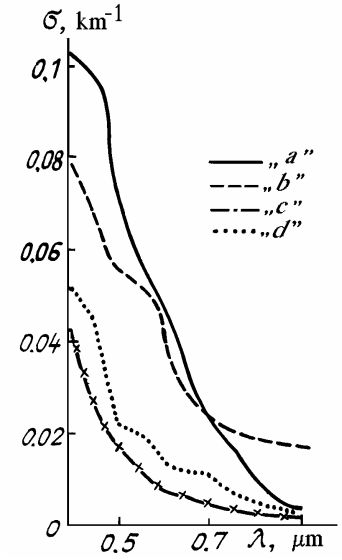

FIG. 5. True values of the coefficients of molecular and aerosol scattering obtained using formulas (2). The curve denoted by R corresponds to the Rayleigh scattering at the altitude 600 m.

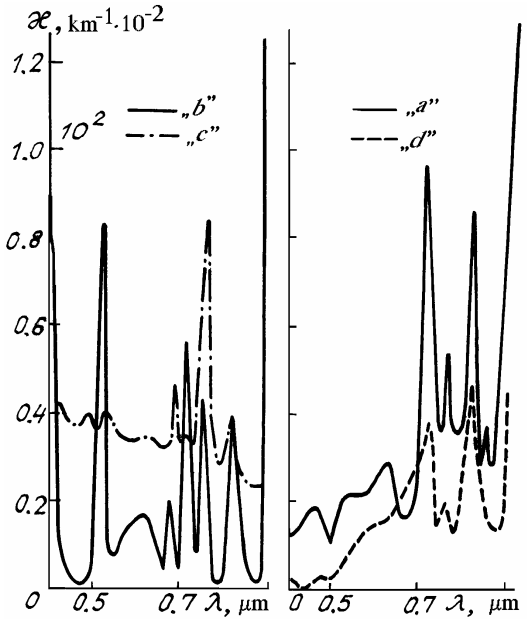

FIG. 6. True values of the absorption coefficients derived based on formulas (2) for the experiments (a), (b), (c), and (d).

By transforming the effective scattering and absorption coefficients based on the above–empirical formulas we obtain the quantities for a unit volume of an actual medium (Figs. 5 and 6). As can be seen the obtained scattering and absorption coefficients are close to the values calculated using the Mie theory[8,12,13] and Rayleigh formula.

The results obtained here make it possible to arrive at a conclusion that in the cases under consideration the absorption of light in a cloud is caused only by dry aerosol particles, otherwise the "effective" absorption coefficient should coincide with the model values.[12,13] Moreover, speaking more generally the presence of the "anomalous short wave absorption in a cloud" is indicative of the presence of a dry aerosol in a cloud.

THE VOLUME ABSORPTION COEFFICIENT

It should be noted, first of all, that the spectral resolution of the instrument used in experiments is insufficient for the radiation absorption in complex bands to be discussed and the author understands that the use of the term absorption coefficient in the region of such bands is ill–founded. Therefore when the absorption coefficient is considered in the region of complex absorption bands of an atmospheric gas we deal with the mean values of the absorption coefficient in the spectral interval of 0.004 μm.

In the previous section we proposed a formula which takes into account the effect of multiple scattering on the volume absorption coefficient. Let us now analyze the curves (*a*) and (*b*) in Fig. 2. Near the wavelength of 0.6 μm there is an absorption band of O_3 (Chappean band). By subtracting the aerosol absorption we obtain: (*a*) $\kappa = 0.0258\ \text{km}^{-1}$, in this case $\tau_d = 22.0$ and $\Lambda = 0.9988$ and (*b*) $\kappa = 0.0604\ \text{km}^{-1}$, $\tau_d = 18.0$, $\Lambda = 0.9966$. Then in the absence of a cloud the coefficients of ozone absorption should be: (*a*) $\kappa = 0.95 \cdot 10^{-4}\ \text{km}^{-1}$ and (*b*) $\kappa = 5.62 \cdot 10^{-4}\ \text{km}^{-1}$. The absorption cross section of O_3 in the maximum of the Chappean band is known: $5 \cdot 10^{-21}\ \text{cm}^2$ (Ref. 14) and it is possible to estimate the ozone concentration at altitudes of (*a*) 0.6 km and (*b*) 0.9 km;

a) $n_{O_3} = 1.9 \cdot 10^{11}\ \text{cm}^{-3}$ or $\rho_{O_3} = 15.1\ \mu\text{g} \cdot \text{m}^{-3}$;

b) $n_{O_3} = 1.1 \cdot 10^{12}\ \text{m}^{-3}$ or $\rho_{O_3} = 84.5\ \mu\text{g} \cdot \text{m}^{-3}$.

Thusly obtained values of O_3 concentration are quite realistic even near the earth's surface to say nothing about an altitude of 1 km (Ref. 14). Similar effect of increasing absorption in the oxygen band at 0.76 μm in clouds as compared with the cloudless atmosphere was found from experiments.[15]

An approximate estimation of the total absorption of solar radiation in clouds in the spectral range 0.4–0.9 μm made for a mean value of the aerosol absorption coefficient of $0.08\ \text{km}^{-1}$, scattering coefficient of a cloud about $30\ \text{km}^{-1}$, and geometrical thickness of 1 km shows that the absorption in a cloud increases by 15% only due to absorption by dry aerosol. The increase of absorption in the ozone absorption band for a cloud is about 6–10% and in the oxygen band O_2 at 0.76 μm it is 10 to 12%. For heavier clouds the effect of the absorption increase due to multiple scattering can be stronger and this can explain the so–called "anomalous short wave absorption in clouds".

CONCLUSION

The measurement data used in this work were obtained with different accuracy, and not always sufficiently high, therefore the results presented here should be considered as an example of applying the proposed method to interpretation of the experiments carried out in clouds. It is quite clear that experimental data of higher quality would provide for more accurate estimates and more interesting results as regards the

properties of real clouds. Nevertheless, in spite of an insufficient accuracy of experimental data some new facts have been obtained which are important for adequate description of cloud layers.

As to the contribution of multiple light scattering to the increase of contributions coming from the Rayleigh and aerosol scattering and absorption to the radiation transfer process, similar result can obviously be obtained from rigorous theoretical calculations if one writes the transfer equation for each component in the atmosphere and solves the general system of these equations jointly or if in the Monte Carlo method one uses a model which takes into account all even weakly absorbing and scattering components.

If calculations of the radiation field characteristics (fluxes, intensities, and heat influxes) use radiation transfer equation written for a single component as it is usually the case, then it is advisable to use in the model the effective values of the absorption and scattering coefficients estimated by empirical formulas (2).

In conclusion the author would like to acknowledge his colleagues from the atmospheric physics department of the Leningrad State University for their fruitful discussions of the results obtained as well as E.A. Bezrukova and A.I. Shul'ts for their help.

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