DETERMINATION OF MICROPHYSICAL PARAMETERS OF A STRATIFIED CLOUD FROM THE DATA OF AIRBORNE RADIATIVE MEASUREMENTS

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Simple formulas that relate the microphysical parameters (the mean radius and the imaginary part of the complex refractive index) of a large particle to its scattering and absorption coefficients have been derived in the approximation of the Van de Hulst anomalous diffraction. The accuracy of these formulas and their applicability limits have been estimated. The obtained formulas have been applied to process the data on spectral dependence of the optical parameters of a stratified cloud obtained previously in four runs of airborne radiative measurements. The mean radius of stratified cloud droplets has been determined for each run of measurements as well as the spectral dependence of the imaginary part of the complex refractive index.

INTRODUCTION

The microphysical parameters of a real stratified cloud are important for solving the problems associated with the study of the cloud structure, dynamics of its evolution, cloud modeling, and ecological monitoring of atmospheric pollution. The main parameters of cloud particles from the viewpoint of radiative problems are the effective cloud particle radius r averaged over their geometric cross sections (or the mean particle radius for a given cloud particle size distribution) and the imaginary part $\varkappa$ of the complex refractive index. Feasibility of determination of the mean radius of cloud particles from the data on airborne measurements of the intensity of solar radiation reflected from a cloud layer in the near-IR range was analyzed in Refs. 1 and 2 with the use of statistical methods of inverse problem solution.

Here, simple formulas for r and $\varkappa$ are derived from the approximate relations of the light scattering theory suggested in Ref. 3 for the volume scattering and extinction coefficients. The problem of determining the microphysical parameters from the data of radiative measurements is solved in two stages. The first stage – the derivation of the optical characteristics of a cloud layer (its volume scattering and absorption coefficients) – was considered in Refs.4 and 5, where rigorous formulas were derived that related the scattering and absorption coefficients to the solar radiation fluxes into the hemisphere measured at the boundaries of the cloud layer.

The accuracy of the method was also estimated, the range of its applicability was defined, and the optical parameters of stratified cloudiness were obtained for four measurement runs. The second stage – the determination of the effective cloud particle radius averaged over their cross sections and of the imaginary part of the complex refractive index – is considered below.

STARTING FORMULAS

Simple but sufficiently exact formulas for the extinction (σ^{ext}), scattering (σ^{sc}), and absorption (σ^{abs}) coefficients and for the single scattering albedo $\omega_0 = \sigma^{\text{sc}}/\sigma^{\text{ext}}$ of polydispersed media were derived in Ref. 3 in the approximation of the Van de Hulst anomalous diffraction in case of the gamma distribution of particles over their size. The accuracy of these formulas was also estimated in Ref. 3 by way of their comparison with results of calculations by the Mie theory. Here, these formulas are the starting ones for further consideration. They are:

$$\left.\begin{aligned}\sigma^{\text{sc}} &= S\,[1 + C\,(1 + B)^{-\,(p+3)}]\\ \sigma^{\text{abs}} &= S\,[1 - (1 + B)^{-\,(p+3)}]\\ \sigma^{\text{abs}} &= 2\,S\,(1 + C/2)\end{aligned}\right\}, \tag{1}$$

where S is the effective geometric cross section of particles per unit volume,

$$S = \pi\, r^2\, N\,\frac{p+2}{p+1}\,,\quad B = \frac{8\pi\,\varkappa\, r}{\lambda(p+1)}\,,$$

$$C = \frac{(p+1)\,\lambda^2}{(p+2)\,\pi^2\, r^2}\;\frac{(m-1)^2 - \varkappa^2}{[(m-1)^2 + \varkappa^2]^2}\,, \tag{2}$$

m is the real part of the refractive index, p is the parameter of the gamma distribution (for narrow particle size distribution, typical of low clouds above the

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continent, $p = 6$; for wide particle size distribution, more typical of frontal and maritime clouds, $p = 2$), λ is the incident radiation wavelength, and N is the number density of particles in the cloud. According to Ref. 6, $N > 200$ for continental air masses and $N \approx 50$ for maritime ones. Considering that in the expression for C $\varkappa^2 \sim 10^{-12}$ and $\varkappa^4 \sim 10^{-24}$ for the visible range, it can be shown that

$$C = \frac{p+1}{p+2}\,\frac{\lambda^2}{4\pi^2\, r^2\, (m-1)^2}\,. \tag{3}$$

Then the formulas that relate the effective mean radius r and the imaginary part of the refractive index $\varkappa$ to the volume scattering and absorption coefficients

$$r = \left(\frac{\sigma^{\mathrm{ext}} - N\lambda^2/[4\pi\,(m-1)^2]}{2\pi\, N}\,\frac{p+1}{p+2}\right)^{1/2}, \tag{4}$$

$$\varkappa = \frac{\lambda\,[(1-\sigma^{\mathrm{abs}}/S)^{-1/(p+3)} - 1]}{8\pi\, r}\,(p+1) \tag{5}$$

can be easy derived from the above expressions.

When the liquid water content of the cloud

$$q = 4/3\; N\,\pi\, r^3\,\rho\,(p+2)\,(p+3)/(p+1)^2, \tag{6}$$

is used instead of the particle number density, where ρ is the density of the material of the particles (for water, $\rho = 1$ g/cm^3), the formula for the mean particle radius takes the form

$$r = \frac{1.5\, q}{\sigma^{\mathrm{ext}}\,\rho}\,\frac{p+1}{p+3}\,. \tag{7}$$

In the derivation of formula (7), we neglected the term comprising $\lambda^2/r^2 < 0.01$ for the visible incident radiation and particle size $r > 4$ μm.

Now we consider the ratio σ^{abs}/S in formula (5). Taking into account the expression for σ^{ext} we derive $\sigma^{\mathrm{abs}}/S = 2(1-\omega_0)$ with the error no more than 0.3%. Then the formula for $\varkappa$ can be written as

$$\varkappa = \frac{\lambda\,[(2\,\omega_0 - 1)^{-1/(p+3)} - 1]}{8\pi\, r}\,(p+1)\,. \tag{8}$$

ERRORS IN RECONSTRUCTING THE MICROPHYSICAL PARAMETERS

The above technique can be considered as a solution of the inverse problem whose errors are determined by the speed of variations of functions $\sigma^{\mathrm{ext}}(r)$ and $\omega_0(\varkappa)$. Therefore, the formulas for the relative errors in reconstructing the parameters r and $\varkappa$ in terms of the errors in measuring or *a priori* assignment of the input parameters can be obtained by differentiation of the main formula. The error in determining the effective mean particle radius is calculated from the formula

$$2\,\frac{\Delta r}{r} \le \frac{\Delta\sigma}{\sigma} + \frac{\Delta N}{N} + \frac{\Delta p}{p+1}\,, \tag{9}$$

or with the use of the liquid water content

$$\frac{\Delta r}{r} \le \frac{\Delta\sigma}{\sigma} + \frac{\Delta q}{q} + \frac{\Delta p}{p+1}\,. \tag{10}$$

Let us analyze the relative contribution of individual terms.

1) $\Delta\sigma/\sigma$ denotes the relative error in determining the volume extinction coefficient. It was estimated in Refs. 4 and 5 and was within 10%.

2) The uncertainty in *a priori* assignment of the particle number density N and the particle size distribution, manifested through the uncertainty in the parameter p, is given by the second and third terms of formula (9). The contribution of the term $\Delta p/(p+1)$ is ≤15%. The error in assignment of N may reach 200% and its contribution to the error in determining the mean radius will be decisive.

3) The contribution of $\Delta q/q$ at an altitude of ~1 km (typical of stratified clouds) does not exceed 20%, as follows from the data presented in Ref. 6. Therefore, it is desirable to use formula (7) for determining the mean radius. In this case, $\Delta r/r < 45\%$.

The errors in determining the imaginary part of the refractive index are calculated from the following formulas:

$$\frac{\Delta\varkappa}{\varkappa} \le \frac{\Delta r}{r} + \frac{\Delta\lambda}{\lambda} + \frac{\Delta p}{p+1} +$$

$$+ \frac{\Delta\omega_0}{\omega_0}\,\frac{2}{(p+3)\,[(2\,\omega_0 - 1)^{-1/(p+3)} - 1]}\,, \tag{11}$$

$$\frac{\Delta\varkappa}{\varkappa} \le \frac{\Delta r}{r} + \frac{\Delta\lambda}{\lambda} + \frac{\Delta p}{p+1} +$$

$$+ \frac{\Delta\sigma^{\mathrm{abs}} + \sigma^{\mathrm{abs}}/S\,[2\Delta r/r + \Delta N/N + 2\Delta p/(p+1)]}{S\,(p+3)\,(1-\sigma^{\mathrm{abs}}/S)\,[(1-\sigma^{\mathrm{abs}}/S)^{-1/(p+3)} - 1]}\,. \tag{12}$$

An analysis of the errors in this case shows that:

1) the error in determining the imaginary part of the refractive index is primarily determined by the error in determining the mean radius of particles;

2) the contribution from the term $\Delta\lambda/\lambda$ is rather small in case of radiative measurements with wavelength resolution of ~0.002 μm;

3) the term comprising the error in assignment of the single scattering albedo ω_0 involves a small value ($\sim 5\cdot 10^{-4}$) as a denominator; however, $\Delta\omega_0$ itself is very small ($\sim 2\cdot 10^{-6}$) and does not introduce large error in determining $\varkappa$. The term involving $\Delta\sigma^{\mathrm{abs}}$ contributes nearly 50%. For this reason, formula (8) can be recommended to determine $\varkappa$.

The formulas used in this paper were derived for asymptotic cases of the light scattering theory for large particles and for asymptotic cases of the radiative transfer theory for large optical thickness and weak absorption. Thus, their range of applicability is limited, and beyond this range methodological errors strongly increase. Measurements of solar radiation fluxes were numerically modeled to analyze the errors of the obtained formulas and the sensitivity of the method to particle size, value of the imaginary part of the refractive coefficient, and form of the particle size distribution function.

The following set of parameters was considered:

1) $r = 3–10$ μm, $\varkappa = 5{\cdot}10^{-8}$ and $5{\cdot}10^{-5}$ in case of a monodispersed medium;

2) for $r = 10$ μm, two gamma distributions of particle size were analyzed (with $p = 2$ and 6) and two distributions of the form[7]

$$f(r) = ar^{-p}\, e^{-p\, r_0/r}, \quad \text{where} \quad r_0 = r_{\max}. \tag{13}$$

For the above-indicated cases, we first calculated the optical parameters $\sigma^{\rm scat}$ and $\sigma^{\rm abs}$ by the Mie formulas. Second these parameters were used as model ones in calculations of the solar radiation fluxes into the hemisphere. And then we solved the inverse problem to derive the mean effective particle radius r and the imaginary part of the refractive index.

In case of monodispersed medium, the error in reconstructing the mean radius was $\Delta r/r \sim 8\%$ for $r = 3$ μm and 4% for $r = 10$ μm; $\Delta\varkappa/\varkappa \sim 4\%$ at $\lambda = 0.55$ μm and ~25% at $\lambda = 1$ μm.

For polydispersed medium, the minimum error (~4%) in determining the mean radius was obtained with the use of the formula for monodispersed medium without the factor $(p + 1)(p + 3)$ for all considered particle size distribution functions. This circumstance is very important, because we do not know the particle size distribution function in a cloud when we process the real experimental data.

The error in reconstructing $\varkappa$ varied from 3% for the gamma distribution with the parameter $p = 6$ to 25% for the distribution function[7] given by Eq. (16) with $p = 6$. It should be noted that the particle number density N was assumed to be known and did not introduce the error in the result.

DETERMINATION OF MICROPHYSICAL PARAMETERS FOR REAL STRATIFIED CLOUDS FROM THE DATA ON THE MEASURED SOLAR RADIATION FLUXES

As mentioned above, the spectral dependence of the absorption and scattering coefficients was measured in four measurement runs in Refs. 4 and 5.

The first two measurement runs were performed above the Azov and Black Seas, the third measurement run was performed above the Rustavi industrial center, and the fourth measurement run was performed above the surface of Lake Ladoga covered with ice.

The salient features of the results of these measurements are the strong wavelength dependence of the scattering coefficient and large values of the absorption coefficient. This can be explained by intensification of the Rayleigh scattering by air molecules and absorption of radiation by aerosol particles concentrated outside the water droplets in the process of multiple scattering of solar radiation within the cloud. To determine the mean particle radius, the scattering coefficient should be considered in the near-IR range, where the wavelength dependence disappears. For this reason, we take $\sigma = 40$, 39, 6.5, and 24 km^{-1} to determine the effective mean particle radius, Nakajima and King[1] also pointed out that radiative measurements in the IR range are best suited for determination of the mean particle radius in a cloud.

Considering the altitudes of the lower cloud boundary and the temperature at this altitudes, we chose $q = 0.25$, 0.25, 0.2, and 0.5 g/m^3 as the starting values of the liquid water content for the above-considered runs of measurements, in accordance with Ref. 6. Thus, using formula (7) we obtained $r = 7.5$, 7.3, 8.8, and 12.2 μm, respectively, for four measurement runs. In the first two measurement runs, the mean particle radius was also determined from the data of nephelometric measurements performed simultaneously with the radiative measurements. In both cases, it was 6.5 μm, that is, differed from our data by ~15%.

Using formula (8) and the measured wavelength dependence of the single scattering albedo, we obtained the wavelength dependence of the imaginary part of the refractive index shown in Fig. 1. It should be noted that gaseous absorption was not eliminated from the measured volume absorption coefficient; therefore, the results shown in Fig. 1 concern a small volume of the scattering media rather than a cloud droplet. This small volume comprises droplets, atmospheric gases, and dry aerosols.

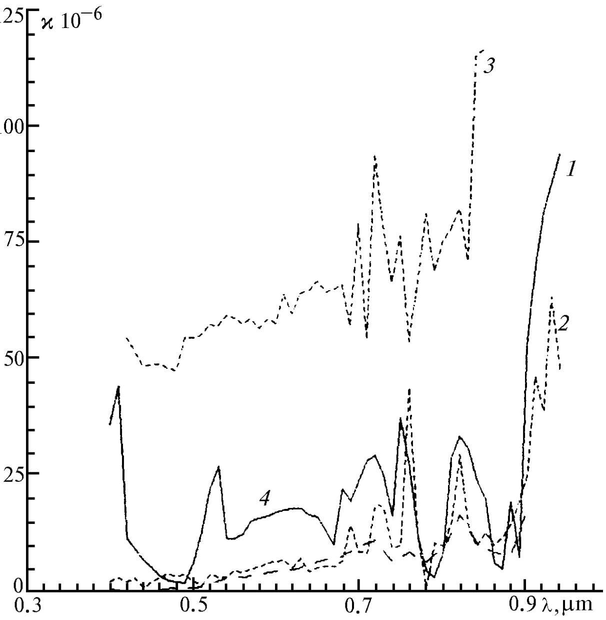

FIG. 1. Spectral dependence of the imaginary part of the refractive index.

Absorption bands of atmospheric gases are clearly seen in the figure, but some more salient features of the absorption spectra are also seen that differ for different clouds.

Rather similar features in the wavelength behavior of $\varkappa(\lambda)$ are noteworthy, in spite of essential differences in air masses and microstructural characteristics of clouds, likely caused by the optical constants of the material of cloud cells and the air outside of droplets. At $\lambda > 0.7$ μm, the absorption lines of oxygen are pronounced, and at $\lambda > 0.9$ μm the absorption lines of H_2O can be seen. At shorter wavelengths, the absorption bands may be caused by gaseous components presented in the atmosphere, such as ozone and nitrogen oxides. Selective character of aerosol absorption at $\lambda < 0.7$ μm is likely due to ferric and titanium oxides presented in aerosol substance. Sharp absorption bands in the shortwave region of the spectrum in the case of maritime clouds are likely due to the effect of the F-centers in alkali and alkali-earth metal compounds, especially in NaCl and KCl entering the atmosphere from sea splashes.

We note that the value of $\varkappa$ obtained by us is two orders of magnitude higher than that used for stratified cloud model. As indicated above, this may be explained by large values of the absorption coefficient[4] (resulting in the corresponding increase of the imaginary part of the refractive index) due to the effect of multiply scattered light within the cloud. Empirical formulas considering this effect were suggested in Ref. 4. If we take the value of the absorption coefficient corrected for the contribution of multiple scattering of light according to Ref. 4, the obtained values of $\varkappa$ will be of the same order as commonly accepted ones. This is one more indirect evidence that the assumption about the effect of multiple scattering made in Ref. 4 is valid.

CONCLUSION

It is well known that direct measurements of microphysical characteristics of cloudiness especially of the imaginary part of the refractive index of the material of particles $\varkappa$ and particle dispersity (r) present serious technical and methodical difficulties, in particular, due to their continuous variability within the cloud and in the course of measurements. The errors of direct measurements of the parameter r are no less than 20%, and those of $\varkappa$ are 30–40%. The errors in determining these characteristics by the procedure described above are less and vary from 4% for both parameters to 18% for r and 25% for $\varkappa$ for different model cases. Moreover, in this case the uncontrollable effect of an experimenter on the dispersed medium is absent, and the desired characteristics can be obtained immediately in the course of experiments.

The application of the method of determining r and $\varkappa$ to the data of real optical experiments yielded the results that agreed with the simultaneously measured mean particle radius as well as with the data of direct measurements in typical stratified clouds.[6] The wavelength dependence $\varkappa(\lambda)$ for the visible and near-IR ranges that was unknown previously was first obtained. Detailed study and interpretation of the measured dependence $\varkappa(\lambda)$ under different experimental conditions is of special interest for the authors and will be continued in future publications.

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