Difference method of determination of the optical depth due to aerosol scattering from data on sky brightness in the visible range: Part 1

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Based on solution of radiative transfer equation obtained, for the clear atmosphere, numerically by the Monte Carlo method we derived approximation formulas for determination of the atmospheric optical depth due to aerosol scattering from data on the sky brightness in the solar almucantar. It is demonstrated that they can be used over a wide range of atmospheric turbidity and different absorption and scattering properties of aerosol particles.

Aerosol particles, scattering and absorbing solar and thermal radiation in the atmosphere, influence directly the earth radiation balance. For better treatment of radiation effects of aerosol, it is necessary to improve our knowledge of optical (scattering phase function, optical depth, single scattering albedo) and/or microphysical (particle size distribution, complex refractive index) characteristics of aerosol.

A reliable source of information on the characteristics of aerosol, present in the entire atmospheric column, are the data of ground-based measurements of unscattered and diffuse radiation. This is because the variations of clear-sky brightness are determined mainly by variations of atmospheric aerosol constituent, characterized by strong scattering. The regularities, inferred from experimental observations, facilitated the development of different methods of solving inverse problems on retrieval of optical and microphysical properties of the aerosol from measurement data on scattered radiation in solar aureole, solar almucantar, and, to a lesser extent, in the plane of sun's vertical.[1–10] The development of global Aerosol Robastic Network (AERONET)[11] of automated scanning sun photometers in combination with modern computer technologies[12,13] makes it possible to obtain data on global distribution of microphysical and optical characteristics of aerosol particles in nearly real time.

The existing methods of retrieving the aerosol microphysical and optical properties can be conventionally divided into two groups. The first and most numerous group includes iterative algorithms, schematically performing the following operations[2,6,7,9,10,13]: in each step, by one or another method, they (1) solve the task of retrieval of aerosol microstructure (microphysics); (2) assuming particle sphericity, they use Mie theory to calculate aerosol optical properties, then being used as input data in making the the radiative transfer calculations; and (3) by comparing the calculated intensities with measured ones, they either decide to terminate or continue the iteration process. A characteristic feature of the second group of methods[1,3,4,8] is that they are not concerned directly with questions on estimating the aerosol microstructure, and, hence, they are less labor-intensive both in terms of time consumptions and required mathematical resources. Owing to these advantages, these simplified approaches can be used to derive simple and efficient techniques for estimating aerosol optical parameters and construct, on their basis, the radiation models of the atmosphere in different regions of the world.

One of the main optical characteristics is the optical depth of the atmosphere due to aerosol scattering (ASOD), or single scattering albedo, referenced to the entire atmospheric column. The goals of the present paper are:

- to develop the method of ASOD (τ_{as}) determination from sky brightness in solar almucantar in a wide range of atmospheric optical parameters, firstly in the visible spectral range, and
- to reduce this method to the level of simple engineering formulas.

The method (henceforth called the "difference" method) is based on the idea of the possibility of determining the scattering optical depth τ_{as} through the quantity τ^* (Ref. 1):

$$\tau^* = 2\pi \int_0^{\pi/2} f(\varphi)\sin\varphi\, d\varphi - 2\pi \int_{\pi/2}^{\pi} f(\varphi)\sin\varphi\, d\varphi, \quad (1)$$

where φ is the scattering angle. The absolute scattering phase function of brightness in the solar almucantar $f(\varphi)$ is related to the sky brightness $B(\varphi)$ by the formula

$$B(\varphi) = E_0 \exp(-\tau m) m f(\varphi),$$

where E_0 is the extraterrestrial solar constant, $\tau = \tau_a + \tau_m$ is the atmospheric optical depth as a sum of the aerosol and molecular components, and m is the atmospheric mass in the direction toward the sun.

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We represent $f(\varphi)$ (see, e.g., Ref. 8) in the form

$$f(\varphi) = f_{as}(\varphi) + f_{ms}(\varphi) + f_2(\varphi) + f_{sur}(\varphi). \qquad (2)$$

Here $f_{as}(\varphi)$ and $f_{ms}(\varphi)$ are the coefficients of directional aerosol and molecular single scattering

$$f_{as}(\varphi) = g_a(\varphi)\tau_{as}/2\pi, \quad f_{ms}(\varphi) = g_m(\varphi)\tau_{ms}/2\pi,$$

where $g_a(\varphi)$ and $g_m(\varphi)$ are the aerosol and molecular scattering phase functions, respectively; $f_2(\varphi)$ and $f_{sur}(\varphi)$ are the terms accounting for multiple scattering and surface effects.

Considering that the function $f_{ms}(\varphi)$ is symmetrical about the point $\varphi = \pi/2$, and assuming that

$$\int_0^{\pi/2} f_{sur}(\varphi)\,d\varphi \approx \int_{\pi/2}^{\pi} f_{sur}(\varphi)\,d\varphi,$$

it can be expected that τ^*, given by formula (1), is quite informative with regards to the atmospheric aerosol constituent, and that some functional dependence will exist between the difference τ^* and the optical depth due to aerosol scattering τ_{as}. In constructing such a function, we will take into account that

- $f(\varphi)$ depends weakly on vertical stratification of atmospheric optical characteristics[14,15] (at least within the limits of applicability of the plane-parallel model). Therefore, to calculate sky brightness in the solar almucantar, the model of vertically homogeneous atmosphere can be used;
- the difference τ^*, defined by formula (1), generally is rather insensitive to variations of the surface albedo A_s. This is important if considering the spotty behavior of the surface reflectivity such as during partial snow cover.

The attempts to construct a function, expressing τ_{as} in terms of τ^*, on the basis of numerical simulation results are not new (see, e.g., Refs. 4 and 8). For instance, Muldashev et al.[4] obtained the formulas, which allowed them to determine ASOD in the visible spectral range for different optical depths due to molecular and aerosol scattering (with the latter not exceeding 0.3) and different solar zenith distances Z_0. The phase functions of brightness $f(\varphi)$, required to construct these formulas, were calculated by the spherical harmonics method. The results by Muldashev et al.[4] are important in that they demonstrated that, in the calculations of τ_{as} via τ^*, both the spectral variations of τ_{as} and τ^* and the value of Z_0 should be taken into account.

Based on numerical solution of radiative transfer equation by the Monte Carlo method, Smerkalov extended the difference method to a wider range of turbidity in the real atmosphere. Smerkalov asserts[8] that his formula for estimation of τ_{as} holds over ASOD variability range $\tau_{as} \le 0.6$–0.7. As will be clear from the discussion below, a serious drawback of the formula is the absence of dependence on solar zenith distances Z_0 in it. It should be stressed that the formula is valid for $\sec Z_0 \approx 5$.

In deriving the formulas in Refs. 4 and 8, it was assumed that aerosol is purely scattering, i.e., that the single scattering albedo of aerosol particles ω_a is unity. However, for more adequate treatment of the optical properties of aerosol, the optical depth due to aerosol absorption must be introduced in the τ^* calculations, precisely what is to be done in the present paper.

As in conservative scattering case, for $\omega_a < 1$ the method of τ_{as} determination from $B(\varphi)$ observations in the solar almucantar is based on preliminary calculations of the scattering phase function of brightness $f(\varphi)$ over a wide range of atmospheric turbidity for different scattering and absorbing properties of aerosol particles and solar zenith distances. To solve the radiative transfer equation in scalar form, the Monte Carlo method (method of adjoint walks, see, e.g., Refs. 16 and 17) was used. Use of a large number ($\approx 5 \cdot 10^5 - 10^6$) of photon trajectories enabled us to calculate $f(\varphi)$ with the accuracy of 1% for all scattering angles over the entire range of input model parameters. Calculations were made for two wavelengths: $\lambda = 439.4$ and $\lambda = 675$ nm, corresponding to maxima of light filter passbands of AERONET scanning photometers. The absorption by air molecules except for ozone was assumed negligible in these wavelength regions. Since most of the ozone mass is located much higher than the bulk of the scattering medium the aerosol absorptivity, if necessary, can readily be taken into account through introduction of the so-called "sub-ozone solar constant."

According to the data, obtained for conservative scattering case, and analyzed jointly in Ref. 8, the ASOD value also depends on the asymmetry factor of aerosol scattering phase function in addition to all the other parameters

$$\Gamma_a = \int_0^{\pi/2} g_a(\varphi)\,d\varphi \Big/ \int_{\pi/2}^{\pi} g_a(\varphi)\,d\varphi.$$

This means that the development of τ_{as} retrieval method involves specification of atmospheric aerosol model, in some or another way. Naturally, such a model must fit, as close as possible, most frequent real situations.

Our choice of aerosol models was based on the data array obtained in 1999–2000 with a specialized sun photometer that is operated in Barnaul as a part of the AERONET system.[18] Most frequently, three aerosol size distribution modes, namely ultrafine (Aitken nuclei), submicron, and coarse fractions, are distinguished.[19] By varying the contribution of each mode to the total extinction of light, it is possible to tune the spectral behavior of the total aerosol scattering coefficient and the shape of the scattering phase function. (Usually, the contribution is adjusted to fit observations of aerosol optical depth and angular distribution of sky brightness.) In accordance with the aforesaid we have constructed, for each wavelength, three aerosol optical models, characteristic of the urban atmosphere. The asymmetry factors Γ_a are presented in Table 1.

Table 1. Asymmetry factors of aerosol scattering phase functions for three different aerosol models

λ, nm	Model 1	Model 2	Model 3
439	7.03	8.77	10.2
675	7.03	9.66	11.55

Remaining input parameters, required to calculate the absolute phase function of the brightness $f(\varphi)$, are presented in Table 2. We recall that the difference τ^* depends little on the surface albedo A_s (we chose A_s in accordance with the recommendations in Refs. 20 and 21).

Table 2. Input parameters used for calculation of the phase function of brightness $f(\varphi)$

Input parameters	λ, nm	
	439	675
$\sec Z_0$	2, 3, 4, 5	
τ_a	0.1; 0.3; 0.5; 0.7; 0.9	
ω_a	0.7; 0.8; 0.9; 1.0	
τ_{ms}	0.239	0.043
A_s (summer)	0.06	0.15
A_s (winter)	0.5	

The phase functions of brightness $f(\varphi)$ were calculated for scattering angles φ from 1 to 6° (with a 1°-step), 6 to 10° (2°-step), 10 to 30° (5°-step), and to $\varphi_{max} = 2Z_0$ (10°-step). The variable step is used because $f(\varphi)$ strongly varies as a function of scattering angle, so that its integration might lead to marked errors. Measurements of the phase function of brightness are constrained by some scattering angle $\varphi_{max} < \pi$; whereas in τ^* calculations from formula (1) it is required to evaluate $\int_{\pi/2}^{\pi} f(\varphi)\sin\varphi\, d\varphi$. This necessitates extrapolation of the product $f(\varphi)\sin\varphi$ to the interval $\varphi \in [\varphi_{max}, \pi]$. The function $f(\varphi)\sin\varphi$ can be extrapolated by different methods, such as using third order polynomials. (Since $f(\varphi)\sin\varphi$ is a smooth function, the uncertainty of selection of extrapolation method introduced only little error in evaluation of integrals over the backward hemisphere.[4]) We note that, for a reliable extrapolation, the solar zenith distance Z_0 must be no less than 60° what exactly explains why the smallest $\sec Z_0$ value was set to 2 here. The largest $\sec Z_0$ value used for calculations equaled 5, providing the possibility of using brightness, calculated within plane-parallel atmospheric model,[17] for analysis of experimental data. Thus, we can state that the difference between the integrals τ^* for $\sec Z_0 = 2$ was calculated accurate to 2–3%.

Figure 1 presents τ^* values, calculated for different aerosol optical depths and single scattering albedo ω_a at $\lambda = 439$ nm. Similar dependence is also observed for $\lambda = 675$ nm. It is clearly seen from Fig. 1 that τ^* values, calculated for a given solar zenith distance Z_0, all fall (with a little spread) on the same curve as ω_a varies in the range $0.7 \le \omega_a \le 1$. This means that, for each Z_0 we can find approximation formulas, fitting these curves with reasonable accuracy and, thereby, determine the sought functions relating τ^* and τ_{as}. Also it is clear that, whereas at a low atmospheric turbidity $\tau_{as} < 0.2$, τ^* changes little with varying $\sec Z_0$; for $\tau_{as} > 0.2$ its dependence on $\sec Z_0$ becomes evident. This strongly suggests that the formula proposed by Smerkalov[8] has limited application: with no dependence on solar zenith distance, it is only valid assuming conditions accepted at its derivation, i.e., for low sun elevation angles ($\sec Z_0 \sim 5$).

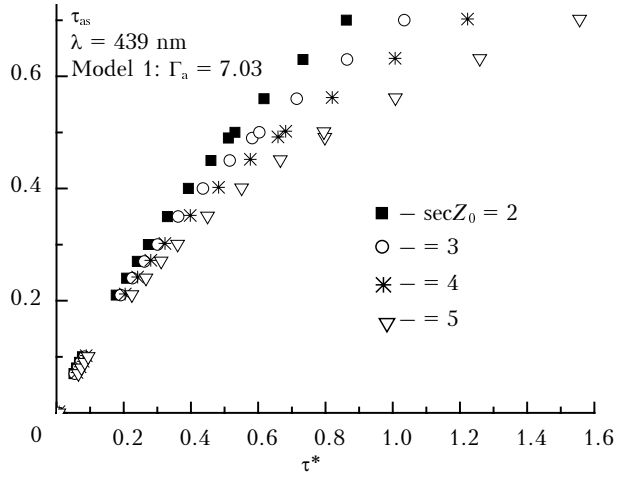

Fig. 1. Aerosol scattering optical depth versus difference τ^* for different solar zenith angles Z_0 (aerosol single scattering albedo $0.7 \le \omega_a \le 1$, aerosol depth $0.1 \le \tau_a \le 0.9$).

Analysis of calculations, performed over wide range of input parameters (Table 2), has shown that, in the general case, τ_{as} can be expressed in terms of τ^* by the following fit:

$$\tau_{as} = K_2(\tau^*)^2 + K_1\tau^* + K_0. \qquad (3)$$

For higher accuracy, it is advisable to divide the entire range of τ^* variability into two intervals[22]: $0 \le \tau^* \le 0.4$ and $0.24 \le \tau^* \le 1.5$ (blue region of the spectrum); $0 \le \tau^* \le 0.45$ and $0.24 \le \tau^* \le 1.36$ (red region of the spectrum). We propose that in each of the spectral intervals, the coefficients K_i, $i = 0, 1, 2$ as functions of $\sec Z_0$ (or the atmospheric mass m that weakly differs from $\sec Z_0$ at $Z_0 \le 78°$) for different values of the asymmetry factor of the aerosol scattering phase function Γ_a (models 1, 2, and 3) be described by the following function:

$$K_i = P_{i,1}\, m + P_{i,0},\ i = 0, 1, 2.$$

The values of $P_{i,j}$ are given in Table 3 (for $\lambda = 439$ nm) and Table 4 (for $\lambda = 675$ nm).

Table 3. Coefficients K_i, $i = 0, 1, 2$, entering into formula (3) for $\lambda = 439$ nm

Coefficient	Model		
	1	2	3
	$\tau^* \le 0.4$		
K_0	0	0	0
K_1	$1.44 - 0.04m$	$1.42 - 0.06\,m$	$1.37 - 0.06m$
K_2	−1.04	−0.99	−0.93
	$0.24 \le \tau^* \le 1.5$		
K_0	$-0.004 + 0.018m$	$0.022m$	$-0.02 + 0.028m$
K_1	$1.31 - 0.12m$	$1.27 - 0.15m$	$1.29 - 0.16m$
K_2	$-0.44 + 0.05m$	$-0.46 + 0.07m$	$-0.49 + 0.08m$

Table 4. Coefficients K_i, $i = 0, 1, 2$, entering into formula (3) for $\lambda = 675$ nm

Coefficient	Model 1	Model 2	Model 3
	$\tau^* \le 0.45$		
K_0	0	0	0
K_1	$1.39 - 0.0374m$	$1.326 - 0.045m$	$1.34 - 0.069m$
K_2	-1	-0.9	-0.84
	$0.24 \le \tau^* \le 1.36$		
K_0	$-0.002 + 0.015m$	$-0.002 + 0.019m$	$0.0025 + 0.022m$
K_1	$1.265 - 0.106m$	$1.183 - 0.1165m$	$1.142 - 0.139m$
K_2	$-0.441 + 0.044m$	$-0.396 + 0.048m$	$-0.369 + 0.0556m$

From Tables 3 and 4 it follows that in both spectral intervals considered here, the coefficients K_i, $i = 0, 1, 2$ in formula (3) depend on the asymmetry factor of the aerosol scattering phase function Γ_a. Therefore, the ASOD retrieval with approach discussed here is obscured by the absence of *a priori* information on Γ_a. We can only believe that the asymmetry factor of aerosol scattering phase function, assumed during the measurements, largely remains within the interval we have chosen: $7.03 \le \Gamma_a \le 10.2$ for $\lambda = 439$ nm and $7.03 \le \Gamma_a \le 11.6$ for $\lambda = 675$ nm. (Recall that the Γ_a variability range is chosen to fit most of the real urban situations).

To demonstrate the capabilities of the proposed method, we have performed numerical experiments for moderate aerosol optical depths for Barnaul: mean aerosol optical depth is $\bar{\tau}_a \approx 0.3$ for $\lambda = 439$ nm and $\bar{\tau}_a \approx 0.2$ for $\lambda = 675$ nm.[18] The other input parameters are presented in Tables 5 and 6.

Table 5. Results of numerical experiment on ASOD calculation for $\lambda = 439$ nm: $Z_0 = 73.4°$ ($\sec Z_0 = 3.5$); $\Gamma_a = 9.5$; $A_s = 0.06$. (For the $\omega_a = 0.9$ case, the τ_{as} calculations from formulas are presented for different intervals: $\tau^* \le 0.4$ and $0.24 \le \tau^* \le 1.5$)

Input parameters	τ^*	Model 1 ($\Gamma_a = 7.03$)	Model 2 ($\Gamma_a = 8.77$)	Model 3 ($\Gamma_a = 10.2$)
$\tau_a = 0.3$ $\omega_a = 0.75$ $\tau_{as} = 0.225$	0.237	$\hat{\tau}_{as}^{(1)} = 0.25$	$\hat{\tau}_{as}^{(2)} = 0.231$	$\hat{\tau}_{as}^{(3)} = 0.223$
$\tau_a = 0.3$ $\omega_a = 0.9$ $\tau_{as} = 0.27$	0.303	$\hat{\tau}_{as}^{(1)} = 0.298$ $\hat{\tau}_{as}^{(1)} = 0.304$	$\hat{\tau}_{as}^{(2)} = 0.276$ $\hat{\tau}_{as}^{(2)} = 0.283$	$\hat{\tau}_{as}^{(3)} = 0.266$ $\hat{\tau}_{as}^{(3)} = 0.280$

Table 6. Results of numerical experiment on ASOD calculation for $\lambda = 675$ nm: $Z_0 = 77.17°$ ($\sec Z_0 = 4.5$); $\Gamma_a = 8.2$; $A_s = 0.15$

Input parameters	τ^*	Model 1 ($\Gamma_a = 7.03$)	Model 2 ($\Gamma_a = 9.66$)	Model 3 ($\Gamma_a = 11.55$)
$\tau_a = 0.2$ $\omega_a = 0.75$ $\tau_{as} = 0.15$	0.146	$\hat{\tau}_{as}^{(1)} = 0.157$	$\hat{\tau}_{as}^{(2)} = 0.145$	$\hat{\tau}_{as}^{(3)} = 0.133$
$\tau_a = 0.2$ $\omega_a = 0.9$ $\tau_{as} = 0.18$	0.185	$\hat{\tau}_{as}^{(1)} = 0.192$	$\hat{\tau}_{as}^{(2)} = 0.177$	$\hat{\tau}_{as}^{(3)} = 0.166$

From the ASOD calculations presented in Fig. 2 it follows that, for a fixed difference τ^*, the aerosol scattering optical depth is a monotonic function of Γ_a for both wavelengths. We will consider calculations for $\lambda = 439$ nm (Table 5) in a more detail.

In the absence of information about asymmetry factor of the aerosol scattering phase function, for the $\omega_a = 0.75$ case, the retrieved counterpart, $\hat{\tau}_{as}$, falls within the interval $0.223 \le \hat{\tau}_{as} \le 0.25$ when the "true" value is $\tau_{as} = 0.225$. If anything is known about Γ_a, such as that the "real" asymmetry parameter of the aerosol scattering phase function lies in the range $8.77 \le \Gamma_a \le 10.2$ ($\Gamma_a = 9.5$, in this experiment), the range of possible ASOD values reduces to the range $0.223 \le \hat{\tau}_{as} \le 0.231$.

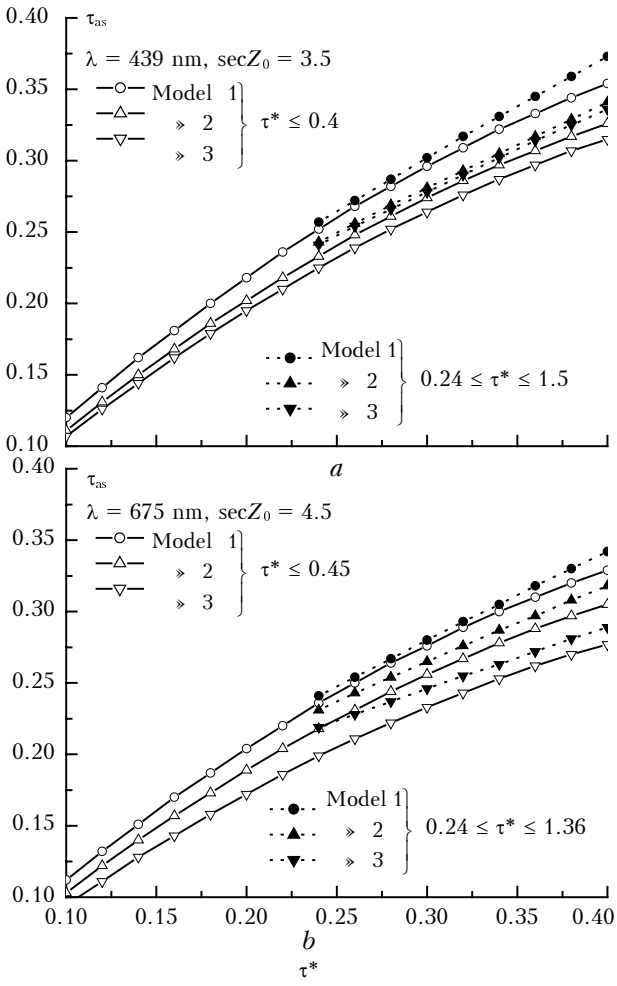

Fig. 2. Optical depth due to aerosol scattering, calculated from approximation formula (3) for different aerosol models in the range $0.1 \le \tau^* \le 0.4$.

For ASOD retrieval in the $\omega_a = 0.9$ case ("true" value $\tau_{as} = 0.27$), we used two sets of coefficients K_i, $i = 0, 1, 2$, as given in Table 3. (As the calculated results show, for $\tau^* \le 0.4$ the first set of K_i values is preferable.) It is noteworthy, because of the additional information about Γ_a available in ASOD determination, the $\hat{\tau}_{as}$ variability range

$0.266 \le \hat{\tau}_{as} \le 0.298$ has narrowed to $0.266 \le \hat{\tau}_{as} \le 0.276$. Similar situation takes place in the experiments with $\lambda = 675$ nm (Table 6).

The results of numerical simulation show that the uncertainty of ASOD calculations, made in the absence of *a priori* information on Γ_a, is ≈ 0.02. This is critically important for the problem posed here, because NASA photometers measure aerosol optical depth $\Delta\tau_a$ based on Bouguer law with the absolute error of 0.01.

Thus, analysis of numerical solution of equation of radiative transfer in the atmosphere by the Monte Carlo method for typical aerosol models allowed us to accomplish first step in development of difference method, namely, to obtain simple "engineering" formulas for the determination of aerosol scattering optical depth. Obviously, the accuracy of τ_{as} determination will be better if data on asymmetry factor of the aerosol scattering phase function Γ_a (or related observation parameters) are available. Hopefully, this additional information can be inferred from a more thorough analysis of measurements of the brightness of scattered radiation, and these results are prepared for publication in the second part of this paper.

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