Aerosol absorption of single- and multiple-scattered light in the cloudless atmosphere

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Received March 7, 2007

The dependence of the multiple-scattered light brightness in the absorbing atmosphere on the absorption optical depth and the solar zenith angle is studied based on numerical results of the transfer equation solution for the case of the cloudless atmosphere with a reflecting bottom. Vital differences from the case of single scattering are ascertained.

Solution of a wide range of problems on retrieving atmospheric aerosol parameters from the observed spectral brightness $B(\Psi, Z_0, Z)$ of the cloudless sky, it is often necessary to divide it into the components of single $B_1(\Psi, Z_0, Z)$ and multiple $B_{2,q}(\Psi, Z_0, Z)$ scattering. The latter usually includes the adding, caused by the light reflection from the underlying surface:

$$B(\Psi,Z_0,Z) = B_1(\Psi,Z_0,Z) + B_{2,q}(\Psi,Z_0,Z), \quad (1)$$

where Ψ is the azimuth count off from the solar vertical plane; Z_0 and Z are the zenith angles of the Sun and the observed sky point; q is the spectral albedo of the underlying surface. If $Z_0 = Z$, i.e., for points lying in solar almucantar, the azimuth Ψ relates to the angle of scattering φ as

$$\cos\varphi = \cos^2 Z_0 + \sin^2 Z_0 \cos\Psi. \quad (2)$$

Light reflection from the underlying surface is usually considered as orthotropic. When atmospheric turbidity is uniformly horizontally distributed, the sky brightness also depends on the wavelength λ, molecular $f_{\mathrm{m}}(\varphi)$ and aerosol $f_{\mathrm{a}}(\varphi)$ scattering indicatrices, and molecular τ_{m} and aerosol τ_{a} optical depths. The latter are divided into absorption depths $\tau_{\mathrm{m.a}}$ and $\tau_{\mathrm{a.a}}$ and scattering $\tau_{\mathrm{m.s}}$ and $\tau_{\mathrm{a.s}}$ depths. The division of $B(\Psi, Z_0, Z)$ into $B_1(\Psi, Z_0, Z)$ and $B_{2,q}(\Psi, Z_0, Z)$ is quite difficult without a preliminary detailed analysis of these quantities on the brightness, especially if it is based on approximate equations and does not use directly the radiation transfer equation.

According to the rigorous single scattering theory,[1] the sky brightness in solar almucantar for the plane-parallel model of atmosphere is connected with the absorption depth in the same manner as the direct solar radiation intensity:

$$B_1(\Psi,Z_0,\tau_{\mathrm{m.s}},\tau_{\mathrm{a.s}},\tau_{\mathrm{m.a}},\tau_{\mathrm{a.a}}) = \\ = B_1(\Psi,Z_0,\tau_{\text{м.р}},\tau_{\text{а.р}})\exp\left[-\sec Z_0(\tau_{\text{м.п}} + \tau_{\text{а.п}})\right]. \quad (3)$$

Here $B_1(\Psi, Z_0, \tau_{\mathrm{m.s}}, \tau_{\mathrm{a.s}})$ is the brightness of single-scatter light in the non-absorbing atmosphere. The absolute phase function of single scattering

$$f_1(\varphi) = f_{\mathrm{a}}(\varphi) + f_{\mathrm{m}}(\varphi) \quad (4)$$

is supposed to be the same in calculations of $B_1(\Psi, Z_0, \tau_{\mathrm{m.s}}, \tau_{\mathrm{a.s}})$ and $B_1(\Psi, Z_0, \tau_{\mathrm{m.s}}, \tau_{\mathrm{a.s}}, \tau_{\mathrm{m.a}}, \tau_{\mathrm{a.a}})$.

It is necessary to specify the normalization conditions for absolute indicatrices of molecular $f_{\mathrm{m}}(\varphi)$ and aerosol $f_{\mathrm{a}}(\varphi)$ scattering, entering into Eq. (4):

$$\tau_{\mathrm{m.s}} = 2\pi\int_0^\pi f_{\mathrm{m}}(\varphi)\sin\varphi\,d\varphi, \quad (5)$$

$$\tau_{\mathrm{a.s}} = 2\pi\int_0^\pi f_{\mathrm{a}}(\varphi)\sin\varphi\,d\varphi. \quad (6)$$

In this case, the stratification of molecular and aerosol absorption coefficients is not important: whether absorbing components are over the light-scattering layer, under it, or somehow distributed with height inside it.

Search of simple dependences of the component $B_{2,q}(\Psi, Z_0, Z)$ on $\tau_{\mathrm{m.a}}$ and $\tau_{\mathrm{a.a}}$ are of interest when solving intricate radiation problems like in Ref. 2. It is reasonable that the absorption coefficient in this case should be somehow distributed inside a scattering medium. The aim of this work is just in in-depth study of the dependence of sky brightness, stipulated by the multiple light scattering and reflection from an underlying surface, on the optical absorption depth at uniform mixing of the scattering and absorbing components. The case $Z = Z_0$ is analyzed for $B_{2,q}(\Psi, Z_0, Z)$ in the visible spectrum range.

This problem is to be considered on the base of numerical solution results of the fundamental equation of the radiation energy transfer in atmosphere. The Monte Carlo method was used for this purpose. The corresponding effective algorithm for brightness computation was developed by T.B. Zhuravleva[3] and kindly put at our disposal.

0235-6880/08/02 107-04 $02.00
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The aerosol model of atmosphere included three fractions of spherical particles: ultra-fine, fine, and coarse.[4] Their size distribution in each mode was considered as logarithmic. The distribution parameters are the following: $\sigma^2 = 0.3$ and $a = -0.1$ (Aitken nuclei), $\sigma^2 = 0.4$ and $a = 0.4$ (submicron particles), and $\sigma^2 = 0.5$ and $a = 0.8$ (coarse particles). Here σ is the dispersion of radius logarithms; $a = \ln\rho_0$; $\rho_0 = 2\pi r_0$; r_0 is the mean geometrical radius of particles.

Note, that if the contributions of these fractions in aerosol optical scattering depth are in the ratio 15, 60, and 25%, the Mie-calculated[5] total aerosol scattering phase function for spherical particles within the 0.55 μm spectral range for the scattering angles $2° \le \varphi \le 160°$ closely approximates the average scattering phase function $f_a(\varphi)$, determined from long series of sky brightness observations in the South-East Kazakhstan.[6] The real part of the refraction index equals to 1.5. Asymmetry coefficients of scattered light fluxes

$$\Gamma_a = \frac{\int_0^{\pi/2} f_a(\varphi)\sin\varphi\, d\varphi}{\int_{\pi/2}^{\pi} f_a(\varphi)\sin\varphi\, d\varphi}, \qquad (7)$$

calculated for these aerosol phase functions and related to the center of the visible spectral range, equal to 4.3, 10.3, and 15.6, respectively.

The radiation transfer equation was solved to the approximation of the plane-parallel atmosphere model. It is applicable to calculations of sky brightness in solar almucantar for zenith angles $Z_0 \le 75°$. The results of $B(\Psi, Z_0, \tau_{m.s}, \tau_{a.s}, \tau_{m.a}, \tau_{a.a})$ calculation, analyzed below, have been obtained for wavelengths of 0.651, 0.546, and 0.416 μm. The molecular optical scattering depths $\tau_{m.s}$, equal to 0.05, 0.10, and 0.30, answer them at standard atmosphere pressure.

The optical depths of the molecular absorption $\tau_{m.a}$ are considered negligible in the chosen spectral regions, i.e., the aerosol was supposed to be the absorbing component. In fact, for resulting brightness calculations, it does not matter which component ($\tau_{m.a}$ or $\tau_{a.a}$) really absorbs the light, since the transfer equation includes the combined quantum survival probability

$$\Lambda = (\tau_{m.s} + \tau_{a.s})/(\tau_{m.s} + \tau_{a.s} + \tau_{m.a} + \tau_{a.a}).$$

If the absorbing effect is attributed to aerosol particles, then the corresponding model aerosol quantum survival probabilities $\Lambda_a = \tau_{a.s}/(\tau_{a.s} + \tau_{a.a})$ varied between 1 and 0.79; while the aerosol optical scattering depths – between 0.1 and 0.6. These values cover the majority of situations in the cloudless atmosphere.[7–9]

The underlying surface albedo was equal to 0.15 (in summer) and 0.8 (in winter). Note, that spectral behavior of the summer albedo in the visible range is of specific character in contrast to the snow one, that is connected with the type of soil or vegetation.[7,10]

However, to study the absorption effect on the brightness component, caused by only multiple scattering at different wavelengths, i.e., at different values of molecular depth $\tau_{m.s}$, q should be fixed through the use, e.g., its spectrum-averaged value. The same relates to the shape of the model aerosol scattering phase function $f_a(\varphi)$, calculated, e.g., for the green spectral region and considered as independent of λ throughout the visible range. Besides, it is considered as independent of the quantum survival probability Λ_a for aerosol particles.

The Monte Carlo computation accuracy of the scattered light brightness depends on the number of photon trajectories in a dispersion medium.[11] Hence, it was necessary to choose the computation time interval Δt. Computations of sky brightness in the units of spectral solar constant (intensity) $I = B/S$ has shown that the error $\delta I \le 1\%$ is reached for $\Delta t \le 1$ min in the majority of tasks computed with a 900 MHz computer. The time increased in case that the optical scattering depths $\tau_s = \tau_{m.s} + \tau_{a.s}$ and zenith angles of the Sun Z_0 were large, and the maximal phase function elongation was considered. For example, an error of 1% was reached for 10 min at $\lambda = 0.416$ μm, $\tau_{m.s} = 0.3$, $\tau_{a.s} = 0.4$, $\sec Z_0 = 5$, $\tau_{m.a} = \tau_{a.a} = 0$, $q = 0.15$, and $\Gamma_a = 15.6$.

To exclude maximally the influence of aerosol scattering phase function shape on the obtained result, we considered the calculation results of sky brightness in nephelometric scattering angles φ_0 satisfying the following condition:

$$B(\varphi_0) = \frac{1}{2}\int_0^{\pi} B(\varphi)\sin\varphi\, d\varphi. \qquad (8)$$

The integrals were calculated numerically. The average value of the angle $\varphi_0 \pm \Delta\varphi_0$ turned out to be $(65.8 \pm 0.7)°$ for the atmosphere model containing only Aitken nuclei, $(57.9 \pm 0.4)°$ for the submicron fraction, and $(52.5 \pm 0.5)°$ for the coarse fraction. RMS deviations $\Delta\varphi_0$ were resulted from variations of aerosol scattering optical depths and zenith angles of the Sun. Note, that cases with only Aitken nuclei in the real atmosphere have not been recorded in practice, while two last φ_0 values are in good agreement with observation data.[13,14]

The optical absorption depths $\tau_{a.a}$ (on the assumption that $\tau_{m.a} = 0$) and the secants of zenith angle of the Sun $\sec Z_0$ enter into the exponent in Eq. (3) as prime factors. The product $\tau_{a.a} \cdot \sec Z_0$ is the optical depth of aerosol absorption towards the Sun.

Earlier G.V. Rosenberg assumed that $\exp(-\tau_{a.a} \cdot \sec Z_0)$ could be used not only in single-scattered light calculations, but in approximate estimations of multiple scattering effects[1] not only in plane parallel atmosphere but in spherical one by substituting $\sec Z_0$ to the Bemporad function.

Such an approach naturally simplifies the calculation pattern, especially for the intensity I in twilight, however, the competence of the approach needs in direct proof. In this work, the brightness dependence on $\tau_{a.a}$ and $\sec Z_0$ is considered separately for each parameter.

To study the dependence of multiple scattered radiation $B_{2,q}(\varphi_0, \sec Z_0, \tau_{a.a})$ on aerosol absorption depth, $B_{2,q}(\varphi_0)$ was calculated for three above mentioned wavelengths λ, two values of albedo of underlying surface, and five values of $\sec Z_0$ (1.5, 2, 3, 4, and 5). The values of aerosol optical depths of scattering $\tau_{a.s}$ and absorption $\tau_{a.a}$ were set according to the table below.

Table. Model optical absorption and scattering depths accepted in calculations

$\tau_{a.s}$	0.1	0.2	0.4	0.6
$\tau_{a.a}$	0	0	0	0
	0.005	0.01	0.02	0.02
	0.01	0.02	0.04	0.04
	0.015	0.03	0.06	0.06
	0.02	0.04	0.08	0.08
	0.025	0.05	0.1	0.1
				0.12
				0.14
				0.16

The $\tau_{a.a}$ values were selected so that the quantum survival probability for aerosol particles varied from 1 to 0.79. The last number approximately corresponds to Λ_a for urban aerosol.[15,16]

By analogy with commonly used techniques for processing of solar radiation spectral fluxes, used, e.g., in ozonometry, calculate $B_{2,q}(\varphi_0, \sec Z_0, \tau_{a.a})/B_{2,q}(\varphi_0, \sec Z_0, \tau_{a.a} = 0)$. As it follows from the analysis, these ratios within limits of 2–3% are independent of wavelengths, i.e., of $\tau_{m.s}$, optical depths of aerosol scattering $\tau_{a.a}$, and particle size distribution, i.e., elongation of the scattering phase function Γ_a, which allows their averaging over the listed parameters. Figure 1 shows the logarithms of the ratio as functions of $\tau_{a.a}$ for two values of $\sec Z_0$ (2 and 5) and calculation results for the case of single scattering.

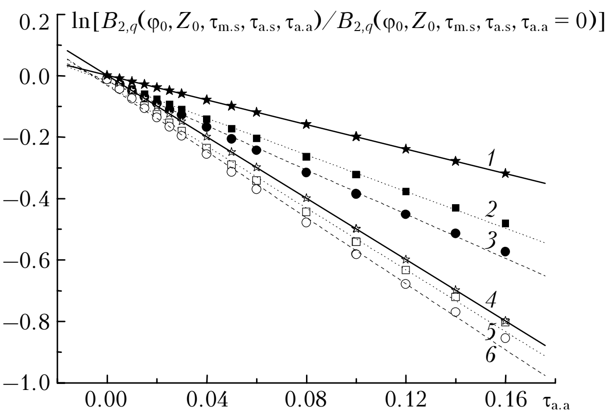

Fig. 1. The logarithm of brightness ratio as a function of the optical depth $\tau_{a.a}$ at $\sec Z_0 = 2$ (*1–3*) and 5 (*4–6*) for single- (*1* and *4*) and multiple-scattered light (*2*, *3*, *5*, and *6*) at the albedo of underlying surface $q = 0.15$ (*2* and *5*) and 0.8 (*3* and *6*).

The following conclusion can be drawn: the dependence of $\ln[B_{2,q}(\varphi_0, Z_0, \tau_{m.s}, \tau_{a.s}, \tau_{a.a})/B_{2,q}(\varphi_0, Z_0, \tau_{m.s}, \tau_{a.s}, \tau_{a.a} = 0)]$ on $\tau_{a.a}$ within the above variations of atmospheric parameters is linear. The slopes of these straight lines relative to the x-axis are evidence of a more rapid decrease of the brightness component of multiple-scattered radiation with an increase of optical depths as compared to those of single-scattered light. The rate of the decrease falls with augmenting zenith angle of the Sun and albedo of underlying surface.

The dependence of multiple-scattered light brightness on the secant of solar coaltitude is of more complicated character. Calculate the values $B_{2,q}(\varphi_0, \tau_{m.s}, \tau_{a.s}, \tau_{a.a}, \sec Z_0)/B_{2,q}(\varphi_0, \tau_{m.s}, \tau_{a.s}, \tau_{a.a}, \sec Z_0 = 1.5)$ and plot the logarithms of the ratios as functions of $\sec Z_0$ similar to Fig. 1. As in the previous case, the calculation results weakly depend on the optical scattering depths τ_s (hence, on λ), which allows corresponding averaging. Averaged data for two fixed absorption depths 0.04 and 0.16 are shown in Fig. 2, as well as single scattering straight lines.

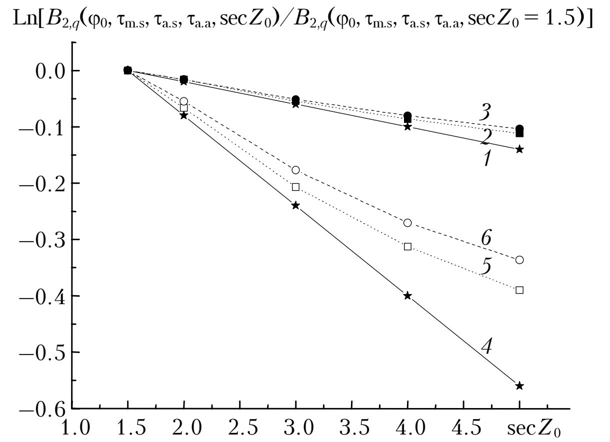

Fig. 2. The logarithm of brightness ratio as a function of $\sec Z_0$ at $\tau_{a.a} = 0.04$ (*1–3*) and 0.16 (*4–6*) for single- (*1* and *4*) and multiple-scattered light (*2*, *3*, *5*, and *6*) at the albedo of underlying surface $q = 0.15$ (*2* and *5*) and 0.8 (*3* and *6*).

The dependences of $\ln[B_{2,q}(\varphi_0, \tau_{m.s}, \tau_{a.s}, \tau_{a.a}, \sec Z_0)/B_{2,q}(\varphi_0, \tau_{m.s}, \tau_{a.s}, \tau_{a.a}, \sec Z_0 = 1.5)]$ on $\sec Z_0$ are evidently nonlinear and less steep in comparison with single-scattered light. Curves for different albedos interchange in comparison with Fig. 1.

The described results show that approximately similar character of the dependences of brightness components of single B_1 and multiple $B_{2,q}$ scattering in absorbing atmosphere on $\tau_{a.a}$ and $\sec Z_0$, used by G.V. Rosenberg in twilight calculations, is quite conditional. Due to small Z_0 values, this concerns our techniques for determining quantum survival probability for aerosol particles[3,17] to a lesser extent.

In general, the techniques, where the results of radiation transfer equation solution can be presented as approximation formulas, should be considered as the first approximation in the iterative scheme for aerosol optical depth division into the scattering and absorption components.

An issue of the day is the development of second approximation in the techniques, similar to those, described in Ref. 17, more accurately taking into account the role of the components, caused by multiple scattering and light reflection from the underlying surface.

References

1. G.Sh. Livshits, *Light Scattering in the Atmosphere*. Part 1 (Nauka, Alma-Ata, 1965), 177 pp.
2. V.E. Pavlov, V.V. Pashnev, A.S. Shestukhin, and T.B. Zhuravleva, Vychisl. Tekhnol. Vestnik KazGU **7**, No. 4 (32), 34–41 (2002).
3. T.B. Zhuravleva, I.M. Nasrtdinov, and S.M. Sakerin, Atmos. Oceanic Opt. **16**, Nos. 5–6, 496–504 (2003).
4. G.V. Rosenberg, G.I. Gorchakov, Yu.S. Georgievskii, and Yu.S. Lyubovtseva, in: *Atmospheric Physics and Climate Problems* (Nauka, Moscow, 1980), pp. 216–257.
5. E.G. Yanovitskii and Z.O. Dumanskii, *Tables for Light Scattering by the Polydispersed System of Spherical Particles* (Naukova Dumka, Kiev, 1972), 123 pp.
6. T.Z. Muldashev, V.E. Pavlov, and Ya.A. Teifel', Atmos. Oceanic Opt. **2**, No. 11, 953–958 (1989).
7. V.A. Smerkalov, *Applied Atmospheric Optics* (Gidrometeoizdat, Leningrad, 1997), 334 pp.
8. http://aeronet.gsfc.nasa.gov
9. S.M. Sakerin, *"Passive sensing of optical atmospheric parameters over oceans and continents,"* Doct. Phys.-Math. Sci. Dissert., Tomsk (1999).
10. V.K. Kogan and K.Ya. Kondratyev, *Foundations of the Information Atmospheric Visibility Theory* (Gidrometeoizdat, Leningrad, 1968), 168 pp.
11. G.I. Marchuk, ed., *The Monte Carlo Method in Atmospheric Optics* (Nauka, Novosibirsk, 1976), 284 pp.
12. G.V. Rosenberg, *Twilight* (Fizmatgiz, Moscow, 1963), 380 pp.
13. E.V. Pyaskovskaya-Fesenkova, *Study of Light Scattering in the Earth's Atmosphere* (AS USSR Publishing House, Moscow, 1957), 219 pp.
14. V.N. Korovchenko, V.K. Oshlakov, V.E. Pavlov, and A.S. Shestukhin, Atmos. Oceanic Opt. **14**, No. 8, 605–606 (2001).
15. World Climatic Program. World Meteorological Organization. A preliminary cloudless standard atmosphere for radiation calculations. (Boulder, Colorado, USA, 1986), 112 pp.
16. A.I. Ivanov, L.M. Karimova, and V.N. Korovchenko, Izv. AN SSSR. Fiz. Atmos. i Okeana **16**, No. 8, 869–871 (1980).
17. T.B. Zhuravleva, V.E. Pavlov, V.V. Pashnev, and A.S. Shestukhin, J. Quant. Spectrosc. and Radiat. Transfer **88**, 191–209 (2004).