# METHOD OF CORRECTING FOR AEROSOL WHEN SOUNDING THE TROPOSPHERIC OZONE WITH A MULTIFREQUENCY LIDAR

## S.L. Bondarenko, V.V. Zuev, and B.S. Kostin

*Institute of Atmospheric Optics,*
*Siberian Branch of the Russian Academy of Sciences, Tomsk*
*Received December 28, 1995*

*We propose here a technique for correcting the ozone profiles for the aerosol influence which assumes the use of multiwavelength sensing at 0.277, 0.313, 0.532, and 1.064 μm. The return signal of rotational Raman scattering has to be measured at the wavelength of 0.532 μm. Numerical experiments show this approach to be quite practical.*

The method is considered for aerosol correction in sounding the tropospheric ozone. The method assumes the use of a lidar operating at $\lambda = 0.277$, $0.313$, $0.532$, and $1.064$ μm. The signal due to rotational Raman scattering by oxygen and nitrogen molecules is used at the wavelength of $0.532$ μm.

The lidar equations for signals at $\lambda_i = 0.277$ and $0.313$ μm are used for determining the ozone concentration sought by the differential absorption method. Let us designate $\lambda_{on} = 0.277$ μm and $\lambda_{off} = 0.313$ μm and write the expression for the ozone concentration in the form[1]:

$$\rho(h_l') = \frac{1}{2\,[k(\lambda_{on}, h_l') - k(\lambda_{off}, h_i')]\,\Delta h} \times$$

$$\times \left\{ \ln \frac{P(\lambda_{on}, h_l)\,P(\lambda_{off}, h_{l+1})}{P(\lambda_{on}, h_{l+1})\,P(\lambda_{off}, h_l)} + B(h_l') + T(h_l') \right\}. \quad (1)$$

Here $k(\lambda_{on}, h_l)$ and $k(\lambda_{off}, h_l)$ are the absorption coefficients at $\lambda_{on}$ and $\lambda_{off}$, respectively; $P(\lambda_{on}, h_l)$ and $P(\lambda_{off}, h_l)$ are the lidar returns recorded at the same wavelengths from the height $h_l$ ($h_l = h_{l-1} + \Delta h$; $l = 1, 2, ...$); $B(h_l')$ and $T(h_l')$ are the correction coefficients determined by formulas:

$$B(h_l') = -\ln \frac{\beta_\pi(\lambda_{on}, h_l)\,\beta_\pi(\lambda_{off}, h_{l+1})}{\beta_\pi(\lambda_{on}, h_{l+1})\,\beta_\pi(\lambda_{off}, h_l)} \quad (2)$$

and

$$T(h_l') = -\{[\beta_{ex}(\lambda_{on}, h_l) + \beta_{ex}(\lambda_{on}, h_{l+1})] -$$

$$- [\beta_{ex}(\lambda_{off}, h_l) + \beta_{ex}(\lambda_{off}, h_{l+1})]\}\,\Delta h. \quad (3)$$

Here $\beta_\pi(\lambda_i, h_l) = \beta_\pi^a(\lambda_i, h_l) + \beta_\pi^m(\lambda_i, h_l)$ are the total (aerosol plus molecular) backscattering coefficients at the wavelengths $\lambda_{on}$ or $\lambda_{off}$; $\beta_{ex}(\lambda_i, h_l) = \beta_{ex}^a(\lambda_i, h_l) + \beta_{sc}^m(\lambda_i, h_l)$ are the total extinction coefficients at the same wavelengths. It is seen from Eqs. (1)−(3) that they contain six unknown values. In addition to the concentration profile and the profile of molecular backscatter that describes the behavior of the molecular

component, these equations contain two profiles of backscattering coefficients and two profiles of the extinction coefficients describing the aerosol component. Since one can determine two unknown values from two equations, then use the lidar equation for the signal at $0.313$ μm for determining the molecular component. Three lidar equations for wavelengths of $0.532$ and $1.064$ μm, including the equation for rotational Raman scattering by oxygen and nitrogen molecules at the wavelength of $0.532$ μm, are used for determining the aerosol component.

Determination of the aerosol component should be discussed in more detail. It is known[2] that there is a relation between the spectral behavior of two optical characteristics entering into the lidar equation. Let us write the aerosol backscattering and extinction coefficients in the form:

$$\beta_\pi^a(\lambda_i, h_l) = \int_{R_1}^{R_2} K_\pi(r, m(\lambda_i, h_l), \lambda_i)\,\pi r^2\,n(r, h_l)\mathrm{d}r; \quad (4)$$

$$\beta_{ex}^a(\lambda_i, h_l) = \int_{R_1}^{R_2} K_{ex}(r, m(\lambda_i, h_l), \lambda_i)\,\pi r^2\,n(r, h_l)\mathrm{d}r. \quad (5)$$

Let us remind that the equations for optical characteristics are written in the approximation for spherical homogeneous particles. Here $s(r, h_l) = \pi r^2 n(r, h_l)$ is the particle size spectrum; $m(\lambda_i, h_l) = \bar{m}(\lambda_i, h_l) - i\kappa(\lambda_i, h_l)$ is the complex aerosol refractive index that is related to the whole spectrum and characterizes the spectral dependence $m(\lambda) = \bar{m}(\lambda) - i\kappa(\lambda)$. Let us assume that there are particles with the size distribution (spectrum) $s(r, h_l)$ at the height $h_l$. The spectral dependence of the refractive index $m(\lambda)$ relates to all particles from this distribution. It is obvious that these two aerosol microphysical parameters determine both the value and the spectral behavior of the optical characteristics. One should pay due attention to the fact that Eqs. (4) and (5) are the linear integral equations with respect to the

particle size spectrum $s(r)$. The refractive index of aerosol particles makes a nonlinear term in the expressions for the efficiency factors. Analysis of optical characteristics calculated for different refractive indices shows the nonlinear dependence.

The peculiarity of the problem under consideration is that the particle size spectrum is determined from the aerosol backscattering coefficient, and then the extinction coefficient is calculated. That means that the operator $W$ considered in detail in Ref. 2 is constructed, which does not depend on the size spectrum. To construct the operator $W$, the assumption is necessary about the sphericity of scattering particles, as well as on the refractive index value and the size spectrum limits. Since the spectrum plays an auxiliary role and is necessary for estimation of the size limits, then to solve the problem considered one may use the algorithms that require minimum number of wavelengths. In particular, such algorithms are the algorithms of optimum filtration (method of model estimates), which in practice allow one to use two wavelengths.[2] They are $\lambda = 0.532$ and $1.064$ μm, which serve for estimating the particle size spectrum.

It is obvious that the particle size spectrum obtained in such a way depends on the value of the refractive index of particles, so its estimation is necessary. To solve this problem, the signal of rotational Raman scattering at the wavelength of $0.532$ μm is used. The peculiarity of measurements at this wavelength is that the rotational Raman backscattering coefficient is equal to the molecular backscattering coefficient to a high accuracy.[3] Omitting the argument $\lambda$, let us write the equation for rotational Raman scattering signals

$$P^R(h_l) = P_0^R \, h_l^{-2} \, b \, \Delta h \, \beta_\pi^m(h_l) \, T_a^2(h_l) \, T_m^2(h_l);$$

$$T_a^2(h_l) = \exp\left\{-2 \int_0^{h_l} \beta_{ex}^a(z) \, dz\right\};$$

$$T_m^2(h_l) = \exp\left\{-2 \int_0^{h_l} \beta_{sc}^m(z) \, dz\right\}, \qquad (6)$$

where $P^R(h_l)$ are the rotational Raman scattering signals recorded; $b$ is the calibration constant; $\beta_\pi^m(h_l)$ is the molecular backscattering coefficient; $T_a(h_l)$ and $T_m(h_l)$ are the atmospheric transmissions due to the aerosol and molecular extinction of light.

As follows from Eq. (6), in the signal recorded only the aerosol extinction coefficient in the expression for the aerosol component of the transmission depends on the refractive index of aerosol particles. It is necessary to note that the dependence of the efficiency factors in Eqs. (4) and (5) on the refractive index of aerosol particles is unknown. The dependence $m(\lambda)$ related to all spectrum is also unknown. So let us use the following procedure for determining the refractive index. Let us set *a priori* some range of variations of

the real $[\bar{m}_1, \bar{m}_2]$ and imaginary $[\kappa_1, \kappa_2]$ parts of the refractive index of aerosol particles. For each complex value $m = \bar{m} - i\kappa$ formed by arbitrarily selected values of the real part from the range ($\bar{m} \in [\bar{m}_1, \bar{m}_2]$) and the imaginary part ($\kappa \in [\kappa_1, \kappa_2]$), the scattering components are separated, i.e. the aerosol and molecular scattering coefficients are determined.[4] The rotational Raman scattering signals at the wavelength of $0.532$ μm are calculated according to Eq. (6).

The calculated signals should depend on the refractive index of aerosol particles due to the peculiarities in the algorithm of separation of the scattering components.[4] Therefore, the difference between the measured signal $P_{obs}^R$ of the rotational Raman scattering at the wavelength of $0.532$ μm, which weakly depends on the refractive index of aerosol particles, and the same signals $P^R$, but calculated for different values $\bar{m} \in [\bar{m}_1, \bar{m}_2]$ and $\kappa \in [\kappa_1, \kappa_2]$, characterizes the value of the refractive index at each height $h_l$. The value of the refractive index can be determined from the condition of the minimum sought in the ratio

$$(|P^R - P_{obs}^R|)/P_{obs}^R = \min_{\bar{m}, \kappa}.$$

Let us note some principle peculiarities of the algorithm while omitting its detailed description. The first peculiarity is connected with the fact that the wavelength $0.313$ μm used for determining the molecular component is in the ozone absorption band. So the algorithm is constructed by the method of iteration, and $\rho_1(h_l) \equiv 0$ is taken as the first approximation. The algorithm converges quite quickly, because the number of iterations did not exceed five. The second peculiarity is connected with the lidar calibration. The multifrequency lidar is calibrated on the horizontal path by means of the screen with known reflection coefficient. Aerosol backscattering and absorption coefficients are determined simultaneously with the calibration constants. The refractive indices, that were taken as the initial conditions for interpretation of the signals, were determined from these values of aerosol coefficients by using special computer program.

The method proposed was used in the numerical experiments for different models of the molecular atmosphere.[5] Aerosol optical characteristics in these experiments were calculated for Deirmendjian continental L model.[6] Different models of the refractive index of aerosol particles[7] were used in calculations. The error distributed according to Poisson law and equal to 5% of the calculated signal values was added to the profiles of signals calculated.

The results of the experiments showed that the ozone concentration profile sought is calculated quite accurately in all cases when no significant variations of the refractive index of particles occur along the sounding path. In the cases when the variation of the

refractive index of aerosol particles is great, the deviation of the reconstructed values of the ozone concentration from the exact values is also quite significant.
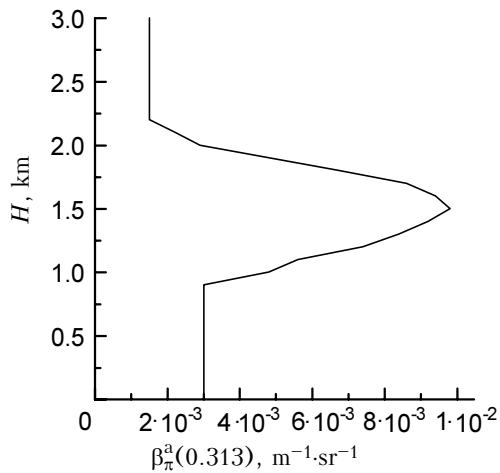


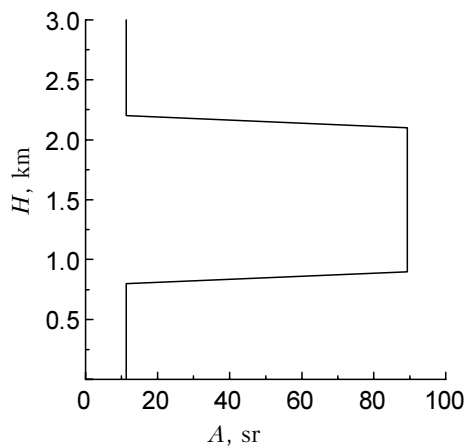FIG. 1. *Profile of the backscattering coefficient at the wavelength of* 0.313 *μm.*



FIG. 2. *Profile of the value reciprocal of the lidar ratio at the wavelength of* 0.532 *μm.*

Let us consider these numerical experiments in detail. The example of significant variations of the refractive index of aerosol particles is shown in Figs. 1 and 2. The profile of the backscattering coefficient at the wavelength of 0.313 μm is shown in Fig. 1. The profile of the value reciprocal of the lidar ratio is shown in Fig. 2. To satisfy the requirements presented in the figures, the aerosol optical characteristics at the heights below 0.9 km and above 2.2 km were calculated for the L model and the refractive index $m = 1.470 - i0.001$. This values of the refractive index is most close to sea salt particles. The spectral dependence of the refractive index of sea salt particles was taken constant, according to the data from Ref. 7. Aerosol optical characteristics at the heights between 0.9 and 2.2 km were calculated for L model and the refractive index $m = 1.470 - i0.022$. It was taken the same for all

wavelengths, because no spectral model for this refractive index is available.

The profiles of the ratio $|P^R - P_{obs}^R|/P_{obs}^R$ calculated at the wavelength of 0.532 μm as a function of real and imaginary parts of the refractive index of aerosol particles at two points of the sounding path are shown in Fig. 3. The result shown in Fig. 3a is related to the point of the sounding path at 0.9 km. It is seen from the figure that there is a minimum at $m = 1.470 - i0.020$. This value of the refractive index is quite close to the exact value. The result related to the point of the sounding path at 2.2 km is shown in Fig. 3b. It is seen from the figure that there is a minimum at $m = 1.470 - i0.0$. This values of the refractive index is also close to its exact value.
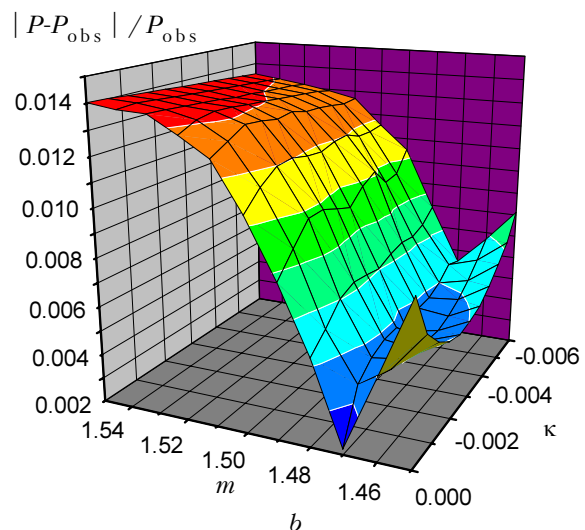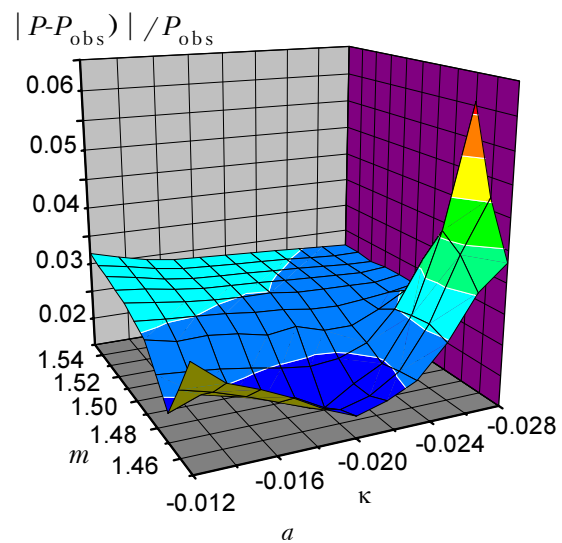


FIG. 3. *Ratio of the Raman scattering signals at the wavelength of* 0.532 *μm as a function of real and imaginary parts of the refractive index of aerosol particles at two points of the sounding path a)* 0.9 *km; b)* 2.2 *km.*
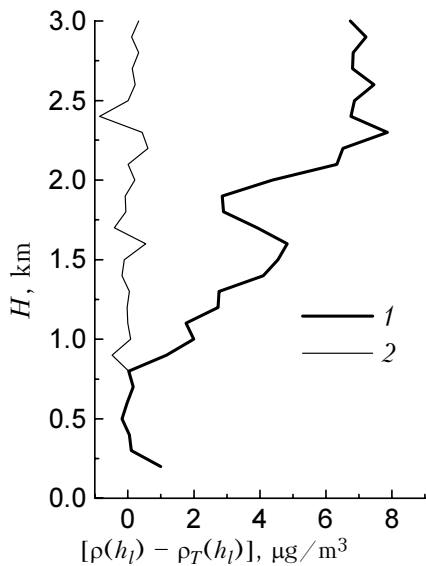
*FIG. 4. Profiles of the deviations of the reconstructed ozone concentrations from the exact values.*

It is also seen from Fig. 3*b* that the ranges of variations of real and imaginary parts of the refractive index are equal to $\bar{m} \in [1.540;\ 1.460]$ and $\kappa \in [0;\ 0.006]$, respectively. If the refractive index $m = 1.450 - i0.0$ is taken, the ratio becomes too great, that is indicative of divergence of the solution. Similar situation occurs at the refractive index $\bar{m} = 1.470 - i0.007$.

When increasing the real part of the refractive index, the ratio increases and tends to some value independent of the value of the imaginary part. The same is equally related to the results shown in Fig. 3*a*.

The ratio becomes too large at the refractive index $m = 1.470 - i0.029$ as well as at $m = 1.450 - i0.010$. Minimum is less pronounced in this case. When increasing the real part, the ratio tends to the value weakly decreasing as the imaginary part increases.

For conclusion let us consider the behavior of the reconstructed values of ozone concentration. Deviations of reconstructed values from the exact ones are shown in Fig. 4. Curve *1* shows the deviation at the refractive index determined at the calibration of the lidar and constant along the sounding path. The deviation, satisfactory at the beginning of the path, increases starting with the height of 0.9 km and reaches its maximum at the height of 2.2 km. Curve *2* is obtained for the refractive index of aerosol particles determined by the method proposed.

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