CORRECTION OF ATMOSPHERIC OZONE SOUNDING DATA OBTAINED BY THE DIFFERENTIAL ABSORPTION METHOD FOR THE AEROSOL CONTRIBUTION

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The aerosol particle size spectrum is measured with a multifrequency lidar. The measured spectrum is then used for calculating the aerosol optical characteristics (to correct for the aerosol contribution) at the ozone sounding wavelength. Numerical experiments verify the practical applicability of the proposed approach.

The atmospheric aerosol makes the problem of interpretation of the data of laser ozone sounding very difficult. When sounding the atmosphere by the differential absorption method, the reliability of determination of the ozone concentration is primarily caused by an irregularity in the aerosol behavior. The problem is especially urgent for sounding of stratospheric ozone. The stratospheric aerosol can have the narrow particle size distribution. This fact suggests that the behavior of the aerosol optical characteristics in the ozone sounding wavelength range may be irregular.[1] New layers of different structures are formed in the stratosphere in addition to the Junge layer after volcanic eruptions. The effect of such layers on the reliability of retrieval of the ozone concentration from the laser sounding data can be significant. Results of numerical experiment on determination of the ozone concentration in the stratosphere are shown in Fig. 1 as an illustration. They were obtained by two different techniques from the same laser sounding data. The aerosol component was not taken into account in this series of experiments.

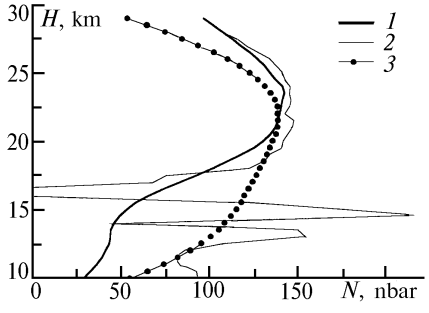

FIG. 1. Ozone concentration profiles obtained without their correction for aerosol contribution at λ_{on} = 0.308 μm and λ_{off} = 0.353 μm: *1) exact profile, 2) profile obtained by differential scheme, and 3) profile obtained by the algorithm of differentiation of empirical dependence.*

The experimental results shown in Fig. 1 are for the situation shown in Fig. 2 where the profile of the scattering ratio at a wavelength of 0.532 μm is drawn. The profile is synthesized from the results of real measurements carried out from December 1991 to March 1993 after Mt. Pinatubo eruption. As is seen from the figure, there is the aerosol layer at altitudes of from 12 to 18 km. Let us consider now the data shown in Fig. 1. Curve *2* in this figure was obtained by the differential scheme. One can see from the comparison of Figs. 1 and 2 that the greatest deviation and even appearance of negative components in the sought after ozone profile is due to the aerosol layer. Curve *3* in Fig. 1 was obtained by the regularization algorithm of differentiation of empirical functions.[2] Although the curve has only positive values of deviation due to the specific feature of the algorithm, its deviation from the exact profile (curve *1*) is quite noticeable. Thus, it follows from Fig. 1 that none of the methods will allow one to determine the ozone concentration with sufficient accuracy, unless the behavior of the aerosol component is considered.

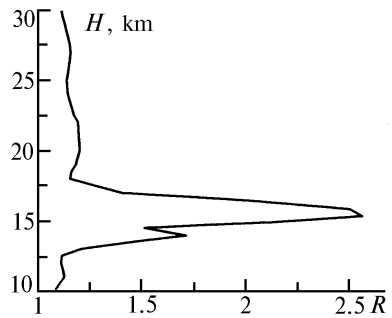

FIG. 2. Profile of the scattering ratio at a wavelength of 0.532 μm *obtained when sounding the stratosphere.*

So, when sounding the atmosphere by the differential absorption method, one use the modified

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expression in the form[3]:

$$\rho(h_l') = \frac{1}{2[k(\lambda_{on}, h_l') - k(\lambda_{off}, h_l')]\Delta h}\,[S(h_l') + B(h_l') + T(h_l')], \quad (1)$$

where

$$S(h_l') = \ln\frac{N(\lambda_{on}, h_l)N(\lambda_{off}, h_{l+1})}{N(\lambda_{on}, h_{l+1})N(\lambda_{off}, h_l)}.$$

Here $\rho(h_l')$ is the sought – after ozone concentration at the height h_l'; $h_l' = (h_{l+1} + h_l)$, $h_{l+1} = h_l + \Delta h$; $k(\lambda_{on}, h_l')$ and $k(\lambda_{off}, h_l')$ are the absorption coefficients of an ozone molecule at the height h_l' at wavelengths λ_{on} and λ_{off}, respectively; $N(\lambda_{on}, h_l)$ and $N(\lambda_{off}, h_l)$ are the signals measured at these wavelengths; $B(h_l')$ and $T(h_l')$ are the correction factors determined by the formulas

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

$$T(h_l') = -2[\beta_{ex}(\lambda_{on}, h_l') - \beta_{ex}(\lambda_{off}, h_l')]\Delta h, \quad (3)$$

where $\beta_{ex}(\lambda_i, h_l') = [\beta_{ex}(\lambda_i, h_l) + \beta_{ex}(\lambda_i, h_{l+1})]/2$ and $\lambda_i = \lambda_{on}$ or λ_{off}. In Eqs. (2) and (3), the following designations are used for the correction factors: $\beta_\pi(\lambda_i, h_l) = \beta_\pi^a(\lambda_i, h_l) + \beta_\pi^m(\lambda_i, h_l)$ are the total (aerosol plus molecular) backscattering coefficients, $\beta_{ex}(\lambda_i, h_l) = \beta_{ex}^a(\lambda_i, h_l) + \beta_{ex}^m(\lambda_i, h_l)$ are the total extinction coefficients for two sounding wavelengths, λ_{on} and λ_{off}.

It is seen from the modified Eq. (1) that it contains more unknown parameters than we can retrieve from measurements at two wavelengths. Here the aerosol and molecular scattering coefficients are the unknown parameters.

To determine the above-indicated optical characteristics, let us consider the method based on application of a multifrequency lidar. Let us suppose that the lidar has not two but n working wavelengths. Let us select the ozone channel and consider the remaining wavelengths. The problem in determination of the optical characteristics (interpretation of measurements) reduces to a solution of the system of laser sounding equations

$$F(\lambda_i, h_l) = \left[\beta_\pi^a(\lambda_i, h_l) + \beta_\pi^m(\lambda_i, h_l)\right]T_a^2(\lambda_i, h_l - h_0) \times T_m^2(\lambda_i, h_l - h_0), \quad (4)$$

$$F(\lambda_i, h_l) = \frac{N(\lambda_i, h_l)h_l^2}{b(\lambda_i)T_a^2(\lambda_i, h_0)T_m^2(\lambda_i, h_0)},$$

$$i = 1, 2, \ldots, n-1. \quad (5)$$

The parameter h_0 in Eq. (1) is the altitude of the lidar calibration; $T_a^2(\lambda_i, h_l - h_0)$ and $T_m^2(\lambda_i, h_l - h_0)$ are the variations of the squares of aerosol and molecular transparency, respectively, in comparison with the altitude h_0. In Eq. (5), $b(\lambda_i)$ are the calibration constants; $T_a^2(\lambda_i, h_0)$ and $T_m^2(\lambda_i, h_0)$ are the squares of aerosol and molecular transparency at the altitude h_0. We note that the values of the parameters included in Eq. (5) are determined when calibrating the lidar.[4]

It is well known that the molecular optical characteristics are related by the expressions:

$$\beta_\pi^m(\lambda_i, h_l) = \frac{3}{8\pi}\,\beta_{sc}^m(\lambda_i, h_l), \quad (6)$$

$$\beta_\pi^m(\lambda_i, h_l) = p_i\,\beta_\pi^m(\lambda_1, h_l),$$

$$\beta_{sc}^m(\lambda_i, h_l) = p_i\,\beta_{sc}^m(\lambda_1, h_l), \quad (7)$$

where p_i is the well-known analytical expression specified in the molecular scattering theory. If we take into account the dispersion properties of air, then

$$p_i = \{[(m_i - 1)/\lambda_i^2][\lambda_1^2/(m_1 - 1)]\}^2. \quad (8)$$

The value of the refractive index of air in Eq. (8) is determined by the Edlen formula

$$(m_i - 1)10^6 = 64.328 + 29498.1/(146 - 1/\lambda_i^2) + 255.4/(41 - 1/\lambda_i^2). \quad (9)$$

Formula (9) is satisfied at a temperature of 288.15 K and a pressure of 760 mm Hg.

Expressions (6)–(8) mean that the totality of the molecular optical characteristics included in the laser sounding equation is determined by a single parameter, namely, $\beta_\pi^m(\lambda_1, h_l)$. So when solving Eq. (4), one wavelength is selected for determining the molecular component.

The set of $(n - 2)$ wavelengths remains for determining the aerosol component. We note that in general the number of working wavelengths of the lidar should be sufficient for correct inversion of the aerosol particle size spectrum. However, in the case under consideration we are confronted with another problem. It is the conversion of the aerosol optical characteristics of one kind (backscattering coefficients) to the others (extinction coefficients). In this case, the particle size spectrum plays an auxiliary role. The above-indicated aerosol optical characteristics form a system of the interrelated parameters. This means that we can do with the minimum number of lidar wavelengths for determining the aerosol component. If we use the parametric form of the lidar data inversion scheme, two wavelengths will be necessary in the case under consideration.[2]

For the method being considered the algorithm for signal processing and determining the ozone concentration is constructed in two stages. The system of equations (4) is solved at the first stage.

The minimum of $(n-1)$ wavelengths destined for separating the scattering components is selected for determining the molecular component. The inverse problem of aerosol light scattering is solved for the rest of wavelengths and the aerosol optical characteristics are determined. At the first stage the algorithm is constructed by the iteration method. We omit the description of the first stage, because it is beyond the scope of this paper. We note that thesolution of the inverse problems of aerosol light scattering was considered in detail in Refs. 2 and 5.

The peculiarities of the algorithm for separation of the scattering components were described in Ref. 6. The sought-after profile of the ozone concentration is calculated at the second stage according to Eqs. (1)–(3). It should be noted that it is indifferent for the approach being considered which of $(n-1)$ wavelengths, used for solving system (4), is selected as λ_{off}.

Let us consider a stratospheric lidar with working wavelengths of 0.308, 0.339, 0.353, 0.532, and 0.683 μm. Let us assume that it has been calibrated. Remind that when sounding the stratosphere, the lidar is calibrated by the atmospheric layer at the altitude[4] $h \approx 30$ km. Then the interpretation of signals is performed in descending order, from the altitude of the lidar calibration. To check the algorithm, a numerical experiment was carried out. The molecular component was calculated for the winter and summer models of thermodynamic parameters at mid-latitudes.[7] The altitude range changed from 30 down to 10 km. When calculating the aerosol component, the vertical profile of the aerosol optical characteristics was specified by the scattering ratio at wavelength of 0.532 μm (Fig. 2). The spectral behavior of the aerosol optical characteristics was specified for specially selected models of aerosol microstructure. The Deirmendjian's model H (Ref. 8) was selected as a background model of microstructure. It serves for describing the stratospheric aerosol size distribution. As is seen from Fig. 2, there is an aerosol layer at altitudes from 12 to 18 km. Microstructure of this layer was described by the lognormal distribution.[9] Spectral dependence of the refractive index of aerosol particles was selected according to the recommendations of Ref. 10. It was assumed that the refractive index remained unchanged with altitude. The error, distributed according to the Poisson law and equal to 5% of the signal value, was introduced to the signal profiles calculated in such a way. The scattering components were separated and the aerosol optical characteristics were determined at four wavelengths 0.339, 0.353, 0.532, and 0.683 μm. Since we considered four wavelengths, the Tikhonov regularization method was chosen to invert the aerosol characteristics. The result of numerical experiment for summer at mid-latitudes is shown in Fig. 3. This result was obtained for $\lambda_{on} = 0.308$ μm and $\lambda_{off} = 0.353$ μm.

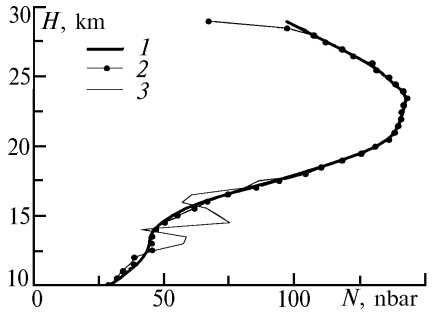

FIG. 3. Profiles of the stratospheric ozone concentration corrected for aerosol contribution and derived from the data of multifrequency lidar at $\lambda_{on} = 0.308$ μm *and* $\lambda_{off} = 0.353$ μm: *1) exact profile, 2) profile obtained by the differentiation algorithm and 3) profile obtained by the differential scheme.*

We note that any wavelength of four wavelengths used for separating the scattering components can be selected as λ_{off}. Curve *1* in the figure shows the exact distribution, curve 2 is obtained by the regularization algorithm of differentiation, and curve *3* is obtained by the differential scheme. It follows from the figure that both results are satisfactory, though the result obtained by the differentiation algorithm is preferable.

When sounding the troposphere with a ground-based lidar, the algorithm of calculation remains the same. Only the calibration of the lidar is changed. The tropospheric lidar was calibrated on a horizontal path with a screen with the known reflection coefficient placed at the end of the path. When calibrating the lidar, the parameters $b(\lambda_i)$ were determined as well as the optical characteristics of the atmosphere. The values of the optical characteristics determined on the horizontal path were taken as initial values in processing of the tropospheric sounding data. Let us describe the numerical experiment on sounding of the tropospheric ozone with a lidar operating at wavelengths of 0.271, 0.289, 0.510, 0.578, 0.628, and 0.726 μm.

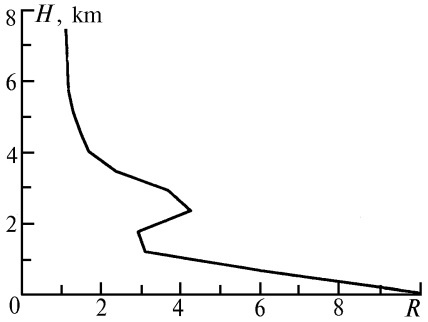

FIG. 4. Profile of the scattering ratio obtained when sounding the troposphere at a wavelength of 0.532 μm.

As before, the molecular component was calculated for the winter and summer models of thermodynamic parameters[7] at mid-latitudes at altitudes up to 7 km. Spectral dependence of the aerosol optical characteristics was specified for two Deirmendjian's distribution functions.[8] Optical characteristics of the rural aerosol were calculated for the model *L* that characterizes the continental aerosol size distribution. The vertical profile of the optical characteristics was specified by the scattering ratio at a wavelength of 0.532 μm shown in Fig. 4.

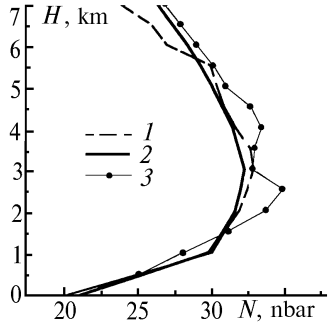

FIG. 5. Profiles of the tropospheric ozone concentration corrected for the aerosol contribution and derived from the data of multifrequency lidar at λ_{on} = 0.271 μm *and* λ_{off} = 0.289 μm: *1) profile obtained by the differentiation algorithm, 2) exact profile, and 3) profile obtained by the differential scheme.*

As is seen from the figure, there is the aerosol layer at altitudes of from 1 to 3 km. Spectral dependence of the aerosol optical characteristics in the layer was for the model *M* that describes the marine aerosol size distribution. As before, the spectral dependence of the refractive index was specified according to recommendations of Ref. 10. It was also assumed that the refractive index remained unchanged with altitude. The scattering components were separated and the aerosol optical characteristics were determined at wavelengths of 0.510, 0.578, 0.628, and 0.726 μm. The regularization method was applied as a method for inverting the aerosol optical characteristics. The result of the numerical experiment for summer at mid-latitudes is shown in Fig. 5 for λ_{on} = 0.271 μm and λ_{off} = 0.289 μm. Curve *1* was obtained by the regularization algorithm for differentiation. Curve *2* shows the exact profile of the ozone concentration in the troposphere. Curve *3* was obtained by the differential scheme. As follows from the figure, the result of determination of the ozone concentration is satisfactory in both cases.

To summarize, we note the following. First, as the numerical experiments have shown, the aerosol correction method based on the use of a multifrequency lidar makes it possible to determine the ozone concentration with satisfactory accuracy. Second, the regularization algorithm for differentiation of empirical dependence gives better results, except of the end points, in comparison with the differential scheme.

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REFERENCES

1. I.E. Naats, in: *Remote Methods for the Study of the Atmosphere* (Nauka, Novosibirsk, 1980), pp. 41–89.
2. V.E. Zuev and I.E. Naats, *Inverse Problems of Laser Sounding of the Atmosphere* (Nauka, Novosibirsk, 1982), 241 pp.
3. E.D. Hinkley, ed., *Laser Monitoring of the Atmosphere* [Russian translation] (Mir, Moscow, 1979), 416 pp.
4. V.E. Zuev, V.V. Zuev, and B.S. Kostin, Atmos. Oceanic Opt. **5**, No. 6, 415–416 (1992).
5. V.E. Zuev and I.E. Naats, *Inverse Problems of Atmospheric Optics* (Gidrometeoizdat, Leningrad, 1990), 286 pp.
6. V.E. Zuev, V.V. Zuev, and B.S. Kostin, Atmos. Oceanic Opt. **5**, No. 10, 698–700 (1992).
7. I.I. Ippolitov, V.S. Komarov, and A.A. Mitsel, in: *Spectroscopic Methods for Sounding of the Atmosphere* (Nauka, Novosibirsk, 1985), pp. 4–44.
8. D. Deirmendjian, *Electromagnetic Scattering on Spherical Polydispersions* (Elsevier, New York, 1969).
9. B. Halperin and D. G. Murcray, Appl. Opt. **26**, No. 11, 2222–2235 (1987).
10. P.B. Russel, T.J. Swisser, M.P. McCormic et. al., Atmos. Sci. **38**, 1279–1294 (1981).