BIFREQUENCY LASER SOUNDING OF STRATOSPHERIC OZONE UNDER CONDITIONS OF HIGH DEGREE OF AEROSOL LOADING

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Received August 3, 1992

Laser sounding of stratospheric ozone under conditions of thick aerosol formations of volcanic origin is considered. The method is proposed of accounting for the aerosol contribution to the results of lidar sounding of ozone. The vertical ozone profiles reconstructed from the lidar data based on the proposed procedure are compared with those obtained from the weather–balloon measurements.

Increasing attention of specialists in ecology, radiation processes, climate formation, etc. has attracted the problem of stratospheric ozone depletion. This in combination with advances in laser techniques has stimulated widespread adoption of laser methods of studying ozone.[1] Among the laser methods, the differential absorption technique is of considerable current use. In this method sounding is accomplished simultaneously at two wavelengths one of which falls on the ozone absorption band (in sounding of the stratospheric ozone it is, as a rule, $\lambda_1 = 308$ nm emitted by an excimer XeCl laser) and the second wavelength is not absorbed by ozone. According to Ref. 1 the second wavelength λ_2 is most common equal to 353 nm and is obtained by SRS conversion of the radiation with the wavelength λ_1 in the cell filled with hydrogen. For these wavelengths the lidar equation is written down in the form[2]

$\lambda_1 = 308$ nm

$$N(H, \lambda_1) = C\beta_\pi(H, \lambda_1) \times$$

$$\times \exp\left\{-2\int_0^H \left[\alpha_m(Z, \lambda_1) + \alpha_a(Z, \lambda_1) + \sigma(\lambda_1)\, n(Z)\right] dZ\right\}, \quad (1)$$

$\lambda_2 = 353$ nm

$$N(H, \lambda_2) = C\beta_\pi(H, \lambda_2) \exp\left\{-2\int_0^H \left[\alpha_m(Z, \lambda_2) + \alpha_a(Z, \lambda_2)\right] dZ\right\},$$

where $N(H, \lambda_i)$ is the recorded lidar return, $\alpha_m(Z, \lambda_i)$ and $\alpha_a(Z, \lambda_i)$ are the coefficients of total molecular and aerosol extinctions, $\beta_\pi(H, \lambda_i)$ is the backscattering coefficient being equal to the sum of the coefficients of molecular backscattering $\beta_\pi^m(H, \lambda_i)$ and aerosol backscattering $\beta_\pi^a(H, \lambda_i)$, i.e., $\beta_\pi(H, \lambda_i) = \beta_\pi^m(H, \lambda_i) + \beta_\pi^a(H, \lambda_i)$ ($i = 1, 2$), C is the instrumental constant of the lidar, $\sigma(\lambda_1)$ is the absorption cross section of ozone molecule at $\lambda = 308$ nm being equal to $1.19 \cdot 10^{-14}$ cm^2, $n(Z)$ is the ozone concentration or the number of ozone molecules in cm^{-3}.

By solving the system of equations (1) for the ozone concentration $n(H)$, we derive the equation

$$n(H) = \frac{1}{2\sigma(\lambda_1)} \left\{ \underbrace{\frac{d}{dH}\left[\ln \frac{N(H, \lambda_1)}{N(H, \lambda_2)}\right]}_{(A)} - \underbrace{\frac{d}{dH}\left[\ln \frac{\beta_\pi(H, \lambda_1)}{\beta_\pi(H, \lambda_2)}\right]}_{(B)} - \right.$$

$$\left. - \underbrace{2\left[\alpha_m(H, \lambda_1) - \alpha_m(H, \lambda_2)\right]}_{(C)} - \underbrace{2\left[\alpha_a(H, \lambda_1) - \alpha_a(H, \lambda_2)\right]}_{(D)} \right\}. \quad (2)$$

It is easy to calculate the terms (A) and (C) of this equation, since (A) involves the recorded lidar returns and (C) is calculated based on the model representations or meteorological data. If need be, the molecular backscattering coefficient entering into $\beta_\pi(H, \lambda_i)$ is calculated analogously to the term (C). As to the terms (B) and (D), they can be neglected under conditions of background content of stratospheric aerosol when its backscattering is less than 10 % of the molecular one and the scattering properties of the adjacent high–altitude layers vary smoothly due to the proximity of the wavelengths λ_1 and λ_2.

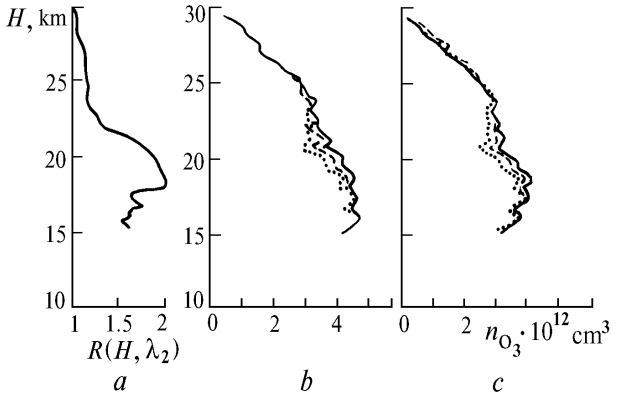

FIG. 1. Aerosol and ozone stratifications: a) vertical profile of the scattering ratio at $\lambda = 353$ nm; b) ozone profiles at the first iteration for $x = 0.5$ (solid curve), 1.25 (dashed curve), and 2 (dotted curve)); and, c) ozone profiles at the second iteration for $x = 0.5$ (solid curve), 1.25 (dashed curve), and 2 (dotted curve)).

0235-6880/92/10 681-03 $02.00

However, under conditions of high degree of aerosol loading of cloud type typical of the today's stratosphere after the Pinatubo volcano eruption[3] in which aerosol backscattering at $\lambda = 353$ nm is comparable with the molecular one (Fig. 1*a*), the 45–nm separation between the sounding wavelengths becomes of primary importance, and the neglect of the terms (*B*) and (*D*) in Eq. (2) results in essential distortions of the ozone profiles being obtained. The present paper deals with the estimate of the coefficients of aerosol backscattering and total scattering for the wavelengths of ozone sounding as well as with the correct account for their contribution to the reconstructed vertical profiles of the ozone concentration.

Taking into account such a parameter as the scattering ratio $R(H, \lambda_i)$, reconstructed by the method detailed in Ref. 4, which describes the backscattering properties of the atmosphere and has the form

$$R(H, \lambda_i) = \frac{\beta_\pi^m(H, \lambda_i) + \beta_\pi^a(H, \lambda_i)}{\beta_\pi^m(H, \lambda_i)}, \qquad (3)$$

the total backscattering coefficient $\beta_p(H, \lambda_i)$ can be represented in the form

$$\beta_\pi^m(H, \lambda_i) + \beta_\pi^a(H, \lambda_i) = R(H, \lambda_i)\, \beta_\pi^m(H, \lambda_i) . \qquad (4)$$

It should be noted that this equation is correct only for $\lambda_2 = 353$ nm, since the scattering properties of the atmosphere at $\lambda_1 = 308$ nm are distorted by ozone absorption and there is no way to obtain the accurate solution $R(H, \lambda_1)$.

However, if we make use of the following relations[2]:

$$\beta_\pi^m(H, \lambda_1)/\beta_\pi^m(H, \lambda_2) = (\lambda_1/\lambda_2)^4 \qquad (5a)$$

for the molecular (Rayleigh) scattering,

$$\beta_\pi^a(H, \lambda_1)/\beta_\pi^a(H, \lambda_2) = (\lambda_1/\lambda_2)^x \qquad (0.5 < x < 2) \qquad (5b)$$

for the aerosol (Mie) scattering,

$$\beta_\pi^m(H, \lambda_2) = \beta_\pi^a(H, \lambda_2)/[R(H, \lambda_2) - 1] \qquad (5c)$$

from Eq. (3), and

$$\beta_\pi^a(H, \lambda_i) = g(H, \lambda_i)\, \alpha_a(H, \lambda_i) , \qquad (5d)$$

where $g(H, \lambda_i)$ is the aerosol lidar ratio, and assume that $g(H, \lambda_i)$ is invariant ($g = 0.03$) with respect to the altitude and the wavelengths λ_1 and λ_2 (see Ref. 5), then the terms (*B*) and (*D*) in Eq. (2) can be represented as

$$\ln \frac{\beta_\pi(H, \lambda_1)}{\beta_\pi(H, \lambda_2)} = \ln \left\{ \frac{(\lambda_1/\lambda_2)^4 + (\lambda_1/\lambda_2)^x\, [R(H, \lambda_2) - 1]}{R(H, \lambda_2)} \right\},$$

$$\alpha_a(H, \lambda_1) - \alpha_a(H, \lambda_2) = \left[R(H, \lambda_2) - 1 \right] \times$$

$$\times \left[\left(\frac{\lambda_2}{\lambda_1} \right)^x - 1 \right] \beta_\pi^m(H, \lambda_2)/g . \qquad (6)$$

After the power x in the system of equations (5) is assigned a fixed value (the effect of arbitrary choice of x on the calculated results is discussed below), the ozone concentration can be calculated from Eq. (2). As a consequence of the accepted assumptions the obtained profile $n(H)$ will be distorted in a definite way and is only an estimate. This profile is called "the profile at the zeroth iteration" in the procedure of ozone calculation under consideration. However, its accuracy is sufficient for the estimate of such an integral characteristic of the atmosphere as the square transmission determined by the ozone absorption

$$T_{O_3}(H) = \exp\left\{ -2 \int_0^H [\sigma\, n(Z)]\, dZ \right\}, \qquad (7)$$

which affects the signal recorded at $\lambda = 308$ nm and makes it difficult to calculate the scattering ratio. If the return signal is corrected for the square transmission

$$\tilde{N}(H, \lambda_1) = N(H, \lambda_1)/T_{O_3}(H) , \qquad (8)$$

the possibility exists to determine the scattering ratio at $\lambda = 308$ nm and, hence, based on it and on $R(H, \lambda_2)$ to estimate more accurately the terms (*B*) and (*D*) in Eq.(2):

$$\ln \frac{\beta_\pi(H, \lambda_1)}{\beta_\pi(H, \lambda_2)} = \ln \left[\frac{R(H, \lambda_1)}{R(H, \lambda_2)} \left(\frac{\lambda_1}{\lambda_2} \right)^4 \right],$$

$$\alpha_a(H, \lambda_1) - \alpha_a(H, \lambda_2) = \left[\left(R(H, \lambda_1) - 1 \right) \left(\frac{\lambda_2}{\lambda_1} \right)^4 - \right.$$

$$\left. - \left(R(H, \lambda_2) - 1 \right) \right] \frac{\beta_\pi^m(H, \lambda_2)}{g} . \qquad (9)$$

This provides more correct results when calculating the ozone concentration from formula (2) which represent the profile at the first iteration.

This procedure for calculating the vertical profile of ozone concentration from the lidar return may be continued using the profile at the first iteration to find T_{O_3} (which enables one to refine the scattering ratio profile at $\lambda = 308$ nm) and by substituting it into Eq. (9) to calculate the profile at the second iteration from formula (2).

Depicted in Figs. 1*b* and 1*c* are the results of ozone profile reconstruction from the lidar data obtained on April 27, 1992 using the aforementioned procedure for different x. It can be seen that the arbitrary choice of x affects the calculated vertical ozone distribution (VOD) within the 18–23 km altitude range. This altitude range, in terms of aerosol, is characterized by the decrease of maximum scattering ratio due to the presence of volcanic aerosol, down to the background level R. The displacement of the ozone profile occurs both at the first and second iterations. Therefore, to obtain the most adequate solution in reconstructing $n(H)$ it is desirable to have the *a priori* determined x since its calculation from the lidar returns at λ_1 and λ_2 using Eqs. (5*b*) and (5*c*) and the estimates of the scattering ratio at $\lambda = 308$ nm according to the formula

$$x = \ln \left\{ \frac{\left(R(H, \lambda_1) - 1 \right)}{\left(R(H, \lambda_2) - 1 \right)} \right\} \Big/ \ln \left(\frac{\lambda_1}{\lambda_2} \right) \qquad (10)$$

does not provide the required solution. This is due to the fact that the determination of x based on Eq. (10) is the ill–posed inverse problem complicated by the logarithm and the absence of accurate solution $R(H, \lambda_1)$.

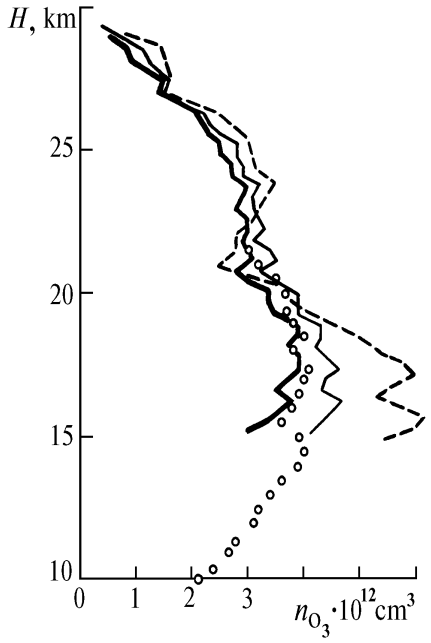

FIG. 2. Vertical profiles of ozone concentration obtained from lidar data without considering the terms (B) and (D) in Eq. (2) (dashed line) at the first iteration for $x = 1.25$ *(solid curve), at the second iteration for* $x = 1.25$ *(heavy curve), and from the ozone sonde data (dotted curve).*

However, the reconstruction of ozone concentration $n(H)$ using the proposed procedure both after first and second iterations is closer to the altitude behaviour of the VOD obtained with the ozone sonde compared to the ozone profile calculated without regard for the terms (*B*) and (*D*) in Eq. (2) (Fig. 2). As for the accuracy of the iterations in comparison with the real profile it calls for special consideration since the concept of iteration used in this paper does not fit the classical concept of iteration procedure in which the increased number of calculational runs brings the solution closer to the real values. It should be noted that the VOD measurements with the lidar and those with the ozone sonde shown in Fig. 2 were conducted at different instants and the observed differences in the ozone stratification are determined by its dynamics.

Thus the described procedure for reconstructing the vertical profiles of the stratospheric ozone concentration is efficient and can be employed to investigate the ozonosphere with lasers under conditions of high degree of aerosol loading. However, it must be further studied and tested in numerical experiments.

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