INFLUENCE OF OPTICAL CHARACTERISTICS OF AEROSOLS ON THE RESULTS OF THE OZONE LIDAR SOUNDING DUE TO CORRECTION OF THE INITIAL DATA FOR AEROSOL

S.L. Bondarenko, A.V. El'nikov, and V.V. Zuev

Institute of Atmospheric Optics,
Siberian Branch of the Russian Academy of Sciences, Tomsk
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This paper concerns with the problem on laser sounding of the stratosphere under conditions of aerosol formations of volcanic origin erupted into the stratosphere. In particular, we study the adequacy of reconstructing the ozone vertical profile using correction for aerosol, with no data on optical characteristics of aerosol (backscattering and extinction coefficients, and so on). The estimation calculation we have done and comparison with the results of balloon borne measurements show that the lack of such data is of no principal importance.

Of the laser methods of remote measurements of gas the concentration, the most popular are DIAL techniques based on the use of sounding radiation at two wavelengths. One of the wavelengths, λ_{on}, which is within an absorption band of a gas under study is the basic one. A laser pulse at this wavelength during its propagation through the atmosphere is absorbed by a gas and scattered by molecules of air and aerosols thus bearing information about spatial distribution of the gas and scattering properties of air and aerosol. The information about the latter is not needed for in this method. Normalization of lidar returns obtained at the second (reference) wavelength λ_{off} which does not fall into the absorption band of the gas studied but is close to the basic one allows the removal of such an information. The proximity of λ_{off} to λ_{on} enables one to neglect spectral behaviors of the molecular and aerosol optical characteristics.

Necessity of considering the problem of laser sounding of ozone based on a DIAL technique in more detail is caused by the following. Absorption of light by molecular ozone occurs in a wide region of spectrum (200–300 nm) where the ozone absorption coefficient is a smooth function. This fact forces us to widely space the sounding wavelengths λ_{on} and λ_{off} for the optical depth

$$\tau(H) = \int_0^H \left\{\sigma(\lambda_{on}) - \sigma(\lambda_{off})\right\} n(H)\, dH\,, \qquad (1)$$

where $\sigma(\lambda_i)$ is the absorption cross section at the corresponding wavelength and $n(H)$ is the ozone concentration, to be sufficient for providing a reasonable accuracy when solving an inverse ill–posed problem, namely, the reconstruction of ozone concentration from lidar data[1]

$$n(H) = \frac{1}{2\left\{\sigma(\lambda_{on}) - \sigma(\lambda_{off})\right\}} \times$$

$$\times \left\{ \underbrace{\frac{d}{dH}\left[\ln \frac{N(H, \lambda_{on})}{N(H, \lambda_{off})}\right]}_{(a)} - \underbrace{\frac{d}{dH}\left[\ln \frac{\beta_\pi(H, \lambda_{on})}{\beta_\pi(H, \lambda_{off})}\right]}_{(b)} - \right.$$

$$\left. - \underbrace{2[\alpha_m(H, \lambda_{on}) - \alpha_m(H, \lambda_{off})]}_{(c)} - \underbrace{2[\alpha_a(H, \lambda_{on}) - \alpha_a(H, \lambda_{off})]}_{(d)} \right\}, \qquad (2)$$

where $N(H, \lambda_i)$ is the intensity of a lidar return recorded, $\alpha_m(H, \lambda_i)$ and $\alpha_a(H, \lambda_i)$ are the coefficients of molecular and aerosol extinction; $\beta_\pi(H, \lambda_i)$ is the backscattering coefficient equal to the sum of coefficients of molecular $\beta_\pi^m(H, \lambda_i)$ and aerosol $\beta_\pi^a(H, \lambda_i)$ backscattering, i.e.,

$$\beta_\pi(H, \lambda_i) = \beta_\pi^a(H, \lambda_i) + \beta_\pi^a(H, \lambda_i)\ (\lambda_i \div \lambda_{on}, \lambda_{off}).$$

Very often, when sounding stratospheric ozone, the operating wavelengths are $\lambda_{on} = 308$ nm with the absorption cross section $\sigma(\lambda_{on}) = 1.19 \cdot 10^{-19}$ cm^{-2} (the XeCl excimer laser radiation wavelength) and $\lambda_{off} = 353$ nm with $\sigma(\lambda_{off}) = 0$ (the radiation wavelength of the first Stokes component of the stimulated Raman scattering cell filled with hydrogen and excited at $\lambda_{on} = 308$ nm). Under conditions of low aerosol loading (background conditions) such separation of wavelengths is not critical since the terms "b" and "d" in Eq. (2) are negligible. Under conditions of heavy aerosol loading (for stratosphere – after powerful eruption of volcanos when vertical stratification of aerosol has a complicated structure and the aerosol backscattering exceeds the molecular one by several times) the values of the terms "b" and "d" become comparable in magnitude with "a" and "c" terms and one cannot neglect them since this can result in substantial distortions in the ozone concentration profiles reconstructed. In such atmospheric situations the lidars for measuring ozone concentration can be used to correctly take into account the scattering and extinction properties of the atmosphere at sounding wavelengths. It is not difficult to do this at the reference wavelength λ_{off} provided that the profile of scattering ratio can be obtained.[2] The scattering characteristics at the base wavelength are estimated, as a rule,[3,4] by approximating the scattering characteristics from $\lambda = 353$ nm to the region at $\lambda = 308$ nm using certain relations.[1,5] Very often such an operation does not provide required accuracy of estimations of the aerosol scattering characteristics necessary to obtain correct results of ozone sounding.

This problem has been further developed in Ref. 6 the authors of which proposed to use the ozone profile obtained by the DIAL technique and the estimate of aerosol

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scattering properties at $\lambda = 308$ nm based on the results derived at $\lambda = 353$ nm to correct a lidar return at $\lambda = 308$ nm for the atmospheric transmission determined by ozone

$$T_{O_3}(H) = \left[- \int_0^H \sigma(\lambda_{on})\, n(H)\, dH \right]. \quad (3)$$

As a result it becomes possible to determine the scattering ratio at $\lambda = 308$ nm and to obtain a more correct profile of ozone by using this scattering ratio in Eq. (2).

However, it should be noted that each of the stages of aerosol correction needs for some assumptions and *a priori* values which can hardly be tested and estimated under conditions when volcanic or anthropogenic aerosols occur in the stratosphere. In this paper we concern with the problem on the effect of such uncertainties on the accuracy of the ozone profile reconstruction.

Aerosol lidar ratio. A possibility of reconstructing vertical aerosol stratification (scattering ratio $\tilde{R}$) is very important for facilitating the aerosol correction of the ozone profiles reconstructed from lidar data. However, for $\tilde{R}$ to be adequately reconstructed the knowledge of aerosol component of the atmospheric transmission is needed. It is possible to estimate this value from lidar data[5,7] based on the relation

$$\beta_\pi^a(H) = g_a(H)\, \alpha_a(H)\,, \quad (4)$$

where $b_p^a(H)$ is the aerosol backscattering coefficient, $\alpha_a(H)$ is the coefficient of aerosol extinction, $g_a(H)$ is the aerosol lidar ratio, and H is the height.

These estimates are usually made based on model values of $g_a(H)$ (see Ref. 7). However, these values can strongly differ from actual ones for volcanic aerosol. Let us consider the influence of the aerosol lidar ratio profile on the profiles of scattering ratio reconstructed from lidar data using different $g(H)$ values. Lidar data at different wavelengths obtained at nighttime during a 3 hrs period and reduced, by an approximation, to one instant of time were used in calculations. The sources of radiation at the wavelengths used were: an excimer laser with a SRS cell filled with hydrogen (308 and 353 nm), a solid–state ND:YAG laser (second harmonic at $\lambda = 532$ nm), and a gold–vapor laser (628 nm). The calculational results are presented in Fig. 1. It is seen that different values of the lidar ratio bring specific changes in the $\tilde{R}$ profile and the shorter the wavelength, the larger are variations in the $\tilde{R}$ profile. It should be noted that $\tilde{R}$at $\lambda = 308$ nm was obtained from the lidar return corrected for the atmospheric extinction due to radiation absorption by ozone.

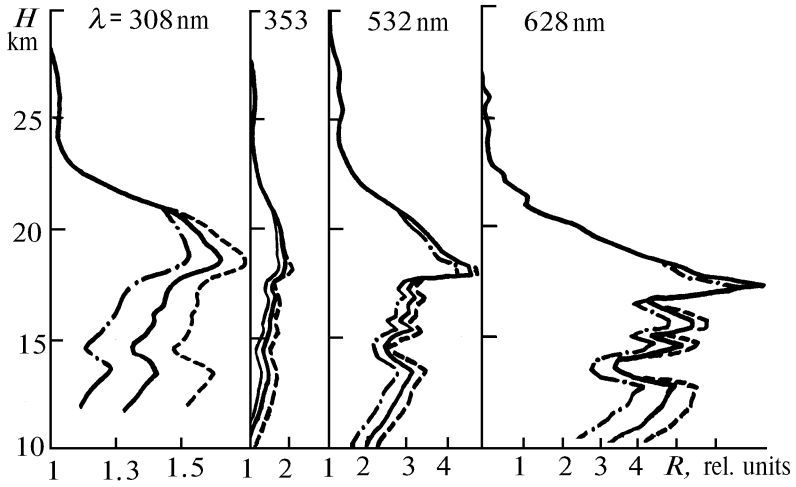

FIG. 1. Profiles of scattering ratio for corresponding wavelengths for different $g_a(H)$: $g_a(H) = 0.5 \cdot g_{mod}(H)$ (dot–dashed line), $g_a(H) = g_{mod}(H)$ (solid line), $g_a(H) = 2 \cdot g_{mod}(H)$ (dashed line), and $g_{mod}(H)$ (model from Ref. 7).

However, calculations of the ozone profile from lidar data much stronger depend not on the profile of aerosol scattering ratio itself, but on the derivative of the ratio of scattering ratios profiles at two wavelengths, λ_{on} an λ_{off} (see term "b" in Eq. (2)). Figure 2 depicts a vertical profile of the term "b" in units of ozone concentration for the pair of wavelengths, 308 and 353 nm. As is seen from Fig. 2 *b*, when variations in the aerosol lidar ratio at one wavelength are accompanied by variations in $g_a(H)$ at the other wavelength but in the opposite direction, there is a significant difference in the values of the term "b" calculated using different combinations of $g_a(H)$. This can cause an essential systematic error in calculations of ozone concentration, if the aerosol lidar ratio has been chosen improperly.

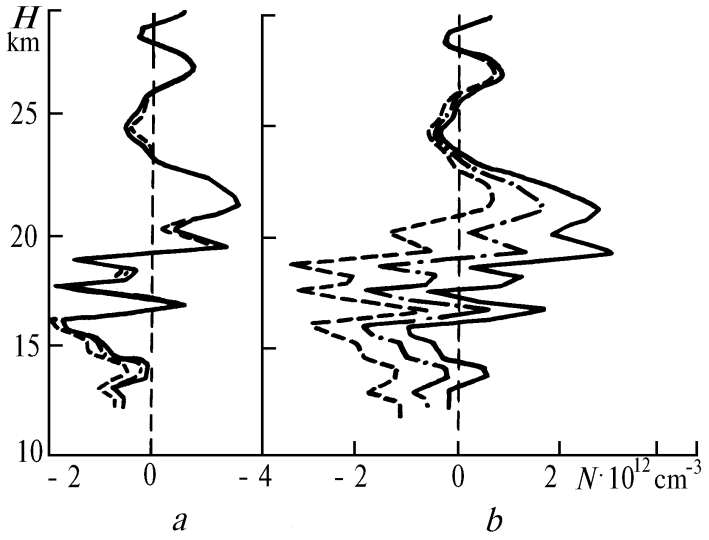

FIG. 2. Altitude variations of "b" from Eq. (2) at $\lambda_{on} = 308$ *and* $\lambda_{off} = 353$ nm: *a)* $g_a^{on}(H) = g_{mod}^{on}(H)$ *and* $g_a^{off}(H) = g_{mod}^{off}(H)$ *(dot–dashed line),* $g_a^{on}(H) = 2\,g_{mod}^{on}(H)$ *and* $g_a^{off}(H) = 2\,g_{mod}^{off}(H)$ *(solid line), b)* $g_a^{on}(H) = g_{mod}^{on}(H)$ *and* $g_a^{off}(H) = g_{mod}^{off}(H)$ *(dot–dashed line),* $g_a^{on}(H) = 2\,g_{mod}^{on}(H)$ *and* $g_a^{off}(H) = g_{mod}^{off}(H)/2$ *(solid line),* $g_a^{on}(H) = g_{mod}^{on}(H)/2$ *and* $g_a^{off}(H) = 2\,g_{mod}^{off}(H)$ *(dashed line).*

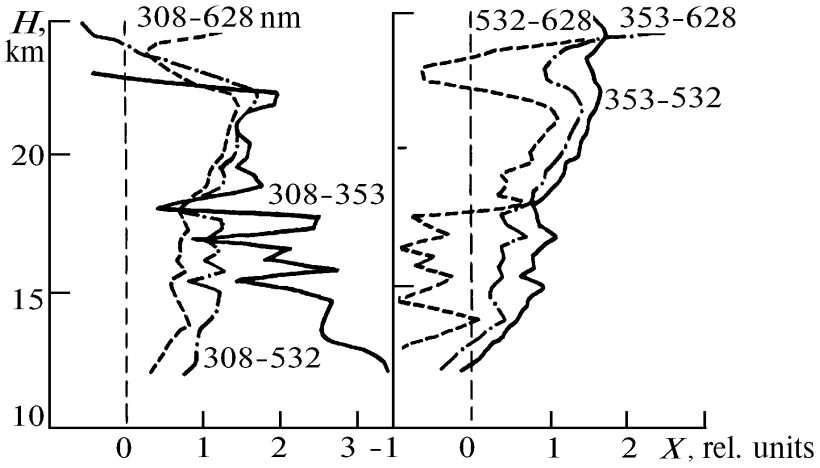

FIG. 3. Vertical profile of the X parameter for corresponding wavelength combinations.

When a microphysical structure of aerosol changes resulting either in the decrease or in the increase of the aerosol lidar ratio at both wavelengths, the derivative of the ratio $\tilde{R}(\lambda = 308)/\tilde{R}(\lambda = 353)$ is in fact invariable for any value of $g_a(H)$ (Fig. 2 *a*).

If we neglect the resonance effects which do not allow one to obtain correct results from lidar sounding of ozone, then the most probable situation is presented by the ozone profile in Fig. 2 *a*. This situation can occur only if similar variations of scattering and extinction properties of the aerosol at both these wavelengths take place. However, if ever possible, the situation with variations of $g_a(H)$ depicted in Fig. 2 *b* can occur with its much smaller variations and, hence, smaller variations of "b" (Fig. 2 *b* represents variations of $g_a(H)$ at sounding wavelengths of about 400%), since the 45–nm spectral range is too small for a substantial change of optical characteristics.

Thus the value of the aerosol lidar ratio only slightly affects the results of reconstruction of ozone concentration.

The relationship between the aerosol backscattering coefficients at different wavelengths. In the scheme for calculating the ozone concentration[6] the aerosol backscattering coefficients are recalculated from $\lambda = 353$ to $\lambda = 308$ nm using the relation

$$\beta_\pi^a(H, \lambda_{308}) \;/\; \beta_\pi^a(H, \lambda_{353}) = (353/308)^X, \qquad (5)$$

where X for different types of aerosol takes the values from the range between 0.5 and 2, see Ref. 5. The ozone concentration profiles given in Ref. 6 and calculated for different X values are indicative of the specific role of X in the formation of the reconstructed ozone profiles. Hence, to obtain adequate results we must know the value of X. The value X can be estimated from lidar returns by the formula

$$X = \ln\left\{\frac{R(H, \lambda_1) - 1}{R(H, \lambda_2) - 1}\right\} \Big/ \ln\left(\frac{\lambda_1}{\lambda_2}\right), \quad (6)$$

obtained from Eq. (5) by simple mathematical transform with the account of

$$\beta_\pi^a(H, \lambda_i) = [\tilde{R}(H, \lambda_i - 1]\, \beta_\pi^m(H, \lambda_i),$$
$$\beta_\pi^m(H, \lambda_1) / \beta_\pi^m(H, \lambda_2) = (\lambda_2 / \lambda_1)^4, \quad (7)$$

where $\tilde{R}(H, \lambda_i)$ is the profile of the scattering ratio at the corresponding wavelength λ_i.

The results of reconstructing the vertical profile of X from lidar returns obtained during the complex multiwavelength (λ_i = 308, 353, 532, and 628 nm) experiment on aerosol sounding for different combinations of wavelengths are represented in Fig. 3. The effect of ozone absorption at the wavelength 308 nm was taken into account following Eq. (3). This figure depicts a sufficiently large difference between the values of X depending not only on the height, which can be explained by different microphysical and optical properties of the aerosol, but also on different combinations of wavelengths. As to the high stratification of the vertical profiles of $X(H)$, it can be explained both by the structure of aerosol layers and by the measurement errors in the values of the parameters used in the $X(H)$ reconstruction. The latter can be related, to a greater extent, to $X_{308-353}$, through the general tendency of the vertical profile of $X_{308-353}$ can be well seen in this figure. The largest difference is observed between the vertical profiles of $X_{308-353}$ and $X_{532-628}$ as well as for other combinations of wavelengths. The vertical profile $X_{308-353}$ has larger and $X_{532-628}$ smaller values as compared to the remaining X_{i-j} combinations. This can be accounted for by the fact that $X_{308-353}$ and $X_{532-628}$ are related to the edge regions of the spectral range (308–628 nm) and their behaviors are determined by different size fractions of the aerosol. A submicron fraction is the most active in the ultraviolet region while the coarse one in the red region. Widening of the range of the wavelength separation used to find $X(H)$ widens an interval of aerosol particle size which determines the scattering and results in corresponding transformation of $X(H)$.

Thus, the X value cannot be considered fixed and its value must be estimated from the lidar returns when reconstructing the ozone concentration profiles. If necessary, a preliminary smoothing should be made.

Aerosol extinction. When reconstructing the ozone profiles, by Eq. (2), the effect of aerosol extinction is determined by the term D. As a rule, its value is found from:

$$\alpha_a(H, \lambda_{on}) - \alpha_a(H, \lambda_{off}) = g_a(H)\, \beta_\pi^a(H, \lambda_{off}) \left[\left(\frac{\lambda_{off}}{\lambda_{on}}\right)^y - 1\right], \quad (8)$$

where y is, for the aerosol extinctions similar to X for the backscattering. Assuming that the scattering ratio at the wavelengths λ_{on} and λ_{off} is known as well as the equality of aerosol lidar ratios at these wavelengths $g_a(H, \lambda_{on}) = g_a(H, \lambda_{off})$, the term D can be written as

$$\alpha_a(H, \lambda_{on}) - \alpha_a(H, \lambda_{off}) = g_a(H)\, \beta_\pi^m(H, \lambda_{off}) \times$$
$$\times \left[\tilde{R}(H, \lambda_{on}) \left(\frac{\lambda_{off}}{\lambda_{on}}\right)^4 - \tilde{R}(H, \lambda_{off}) + 1 - \left(\frac{\lambda_{off}}{\lambda_{on}}\right)^4\right]. \quad (9)$$

Equation (9) should be preferred to Eq. (8) since it contains only $g_a(H)$ as an unknown.

However, if (d) in Eq. (2) has the value comparable to the remaining terms, the possibility of obtaining correct profiles of ozone concentration from lidar data becomes too problematic, since the values $g_a(H)$ and y in Eq. (8) strongly depend on the aerosol microphysics and it is impossible to estimate them from the lidar return signals. The estimate of (d) made based on the model values of $g_a(H)$ and y lying between 0.5 and 2, Ref. 5, at λ = 308 and 353 nm shows that it is smaller by an order of magnitude than (c), to say nothing about the terms (a) and (b).

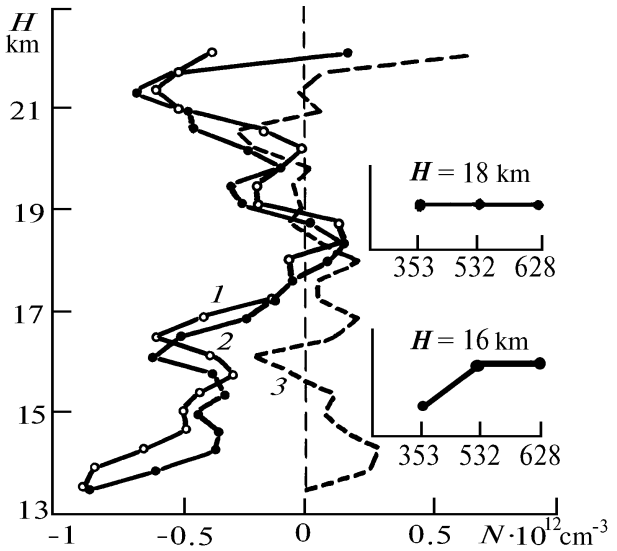

FIG. 4. Vertical profile of difference of extinction coefficients: $\Delta\lambda_{353-532}$ (curve 1), $\Delta\lambda_{358-628}$ (curve 2), and $\Delta\lambda_{532-628}$ (curve 3).

Based on the lidar returns obtained during the complex multifrequency experiment on sounding the aerosol using the DIAL technique we have estimated the difference of the extinction coefficients:

$$\Delta\alpha_{i-j} = \alpha_a(H, \lambda_i) - \alpha_a(H, \lambda_j), \quad (10)$$

where λ takes the values 353, 532, and 628 nm. It is not possible to derive the aerosol extinction coefficients from the system of equations (10), since it is degenerate. The calculational results for $\Delta\alpha_{i-j}$ presented in the form of vertical profiles are depicted in Fig. 4. The analysis of relative behavior of $\Delta\alpha_{i-j}$ provides a qualitative representation of the aerosol extinction coefficient. Its spectral behavior for two altitude ranges (16 and 18 km) is schematically shown in the right part of Fig. 4. It is seen that the extinction properties of aerosol undergoes some altitude transformations and these properties cannot be extrapolated to λ = 308 nm from the results obtained at 353, 532, and 628 nm. However, $\Delta\alpha_{i-j}$ have small values since in this case the difference between the operating

wavelengths is only 45 nm while for $\Delta\alpha_{i-j}$ presented in Fig. 4 the wavelength difference is 93, 179, and 275 nm, the values $\Delta\alpha_{308-353}$ could be much smaller.

Thus the above considerations concerning the value of $\Delta\alpha_{308-353}$ and the estimates made allow us to assume that the term (*d*) in Eq. (2) can be neglected.

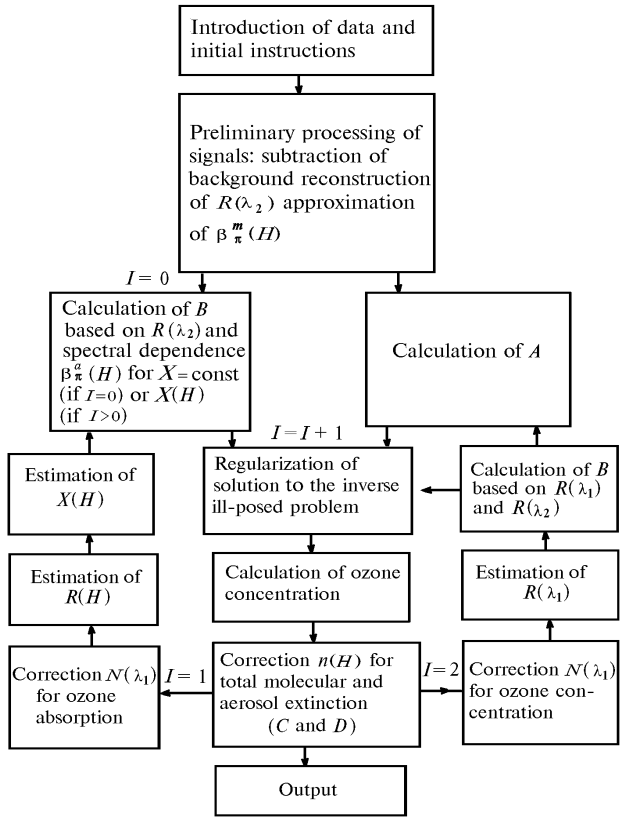

FIG. 5. Block–diagram of a software package for reconstructing the ozone concentration (I is a number of passages).

A block–diagram and methods of calculation. A software package was developed to realize calculations of ozone profiles using the algorithm[6] whose correctness was analyzed in this paper. The programs were written using the "TURBO PASCAL" language in the operational system "MS–DOS". A block–diagram of the program for reconstructing the ozone concentration from lidar returns realized in this software package is represented in Fig. 5. This package was constructed so that a block for a regularization of the solution to an inverse ill–posed problem can include different methods such as spline–functions[8] or Tikhonov's regularization.[9] In particular, in the present paper the calculations were made using the linear[10] and exponential[11] smoothings, and the derivative [Eq. (2)] was calculated using the numerical methods.[12]

The results of calculations of ozone profiles using this block–diagram are depicted in Fig. 6 with the model[13] (dashed lines) and balloonborne measured (circles) profiles of vertical ozone concentration. It is seen that at every stage of calculations the ozone profile approaches the sounding balloon profile.

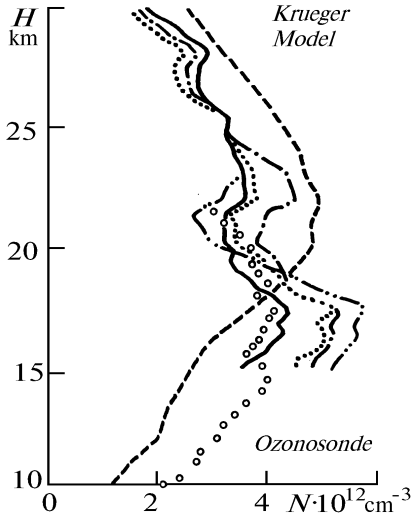

FIG. 6. Ozone profiles obtained from lidar returns: without aerosol correction (– · · –), for X = 1.75 (a dot–dashed line), for X obtained from lidar data (circles), and completely corrected profiles (solid curve).

The more accurate experimental test of adequacy of the obtained values of $n(H)$ to the actual ones cannot be realized.

Thus the analysis made shows that the calculational scheme[6] is useful and provides for obtaining reasonable results. However it requires further investigation based on numerical simulations since its experimental test is impossible.

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