MEASUREMENT OF OZONE CONCENTRATION IN THE LOWER STRATOSPHERE – UPPER TROPOSPHERE

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ABSTRACT

We describe an ozone lidar and consider an algorithm for retrieving the ozone concentration, taking into consideration the aerosol correction. Results of lidar measurements at wavelengths 299 and 341 nm well agree with model estimates, indicating that ozone is sensed with acceptable accuracies in the altitude range of about 6-18 km. It should be noted that the retrieved profiles of altitude distribution of ozone concentration more closely resemble those from satellite data than according to Krueger model.

A lidar is developed and put into operation at Siberian Lidar Station (SLS) to measure the vertical ozone distribution (VOD) in the upper troposphere-lower stratosphere. Sensing is performed according to the method of differential absorption and scattering at wavelength pair 299/341 nm, which are respectively the first and second Stokes components of stimulated Raman scattering (SRS) conversion of the fourth harmonic of Nd:YAG laser (266 nm) in hydrogen.

Figure 1 presents a block-diagram of lidar. As a source of laser radiation, we use fourth harmonic (266 nm) of the principal frequency of radiation of Nd:YAG laser (LS-2134UT model, LOTIS TII firm, Minsk) with its subsequent SRS conversion in hydrogen into first (299 nm) and second (341 nm) Stokes components.

We will consider an algorithm for calculating the vertical ozone distribution, taking into consideration the aerosol correction. The formula for determining the ozone concentration in lidar sensing of the atmosphere by the differential absorption method has the form:

$$n(H) = \frac{1}{\underbrace{2\{k_{on}(H,T) - k_{off}(H,T)\}}_{A}} \cdot$$

$$\cdot\left\{\underbrace{\frac{d}{dH}\left[\ln\frac{N_{off}(H)}{N_{on}(H)}\right]}_{B} - \underbrace{\frac{d}{dH}\ln\left[\frac{\beta^a_{off}(H)+\beta^m_{off}(H)}{\beta^a_{on}(H)+\beta^m_{on}(H)}\right]}_{C} - \underbrace{2\cdot\left[\alpha^a_{off}(H)-\alpha^a_{on}(H)\right]}_{D} - \underbrace{2\cdot\left[\alpha^m_{off}(H)-\alpha^m_{on}(H)\right]}_{F}\right\}$$

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where $N(H)$ is the recorded return signal; α^{α} is the extinction coefficient of aerosol scattering; β^{α} is the aerosol backscattering coefficient; α^{m} is the extinction coefficient of molecular scattering; k is the absorption coefficient; and $n(H)$ is the ozone concentration.

The absorption coefficients $k_{on}(H)$ and $k_{off}(H)$ are used in term A. According to work [1], due to the neglect of temperature dependence of $k(T)$, the uncertainty in ozone concentration may increase by as much as 9%. The ozone absorption coefficients as functions of temperature for different wavelengths are presented in Table 1 [2, 3].

Table 1 – Ozone absorption cross sections (cm^2) for the range of 218 – 295 K at ozone sensing wavelengths.

Wavelength, nm	Temperature, K				
	218	228	243	273	295
On line					
299	$4.1\ 10^{-19}$	$4.1\ 10^{-19}$	$4.25\ 10^{-19}$	$4.3\ 10^{-19}$	$4.6\ 10^{-19}$
Off line					
341	$6\ 10^{-22}$	$6\ 10^{-22}$	$6\ 10^{-22}$	$6\ 10^{-22}$	$1.2\ 10^{-21}$

Aerosol backscattering is several-fold larger than molecular backscattering when aerosol loading of the atmosphere is large, which introduces substantial distortions in retrieved ozone profile when scattering and attenuating properties of the atmosphere at sensing wavelengths are disregarded [4, 5].

The algorithm for VOD calculation accounts for the aerosol correction in terms C and D through introduction of real distribution of scattering ratio $R_{off}(H)$; while VOD calculations in the usual “non-disturbed” atmosphere can be performed taking $R_{off}(H)=1$. Mathematically rearranged C, D, and F terms look:

$$C=\frac{d}{dH}\left\{\ln\left[\left(\frac{\lambda_{off}}{\lambda_{on}}\right)^{x}\cdot\left[1-\frac{1}{R_{off}(H)}\right]+\frac{1}{R_{off}(H)}\cdot\left(\frac{\lambda_{off}}{\lambda_{on}}\right)^{4}\right]\right\}$$

$$D=2\cdot 0.04\cdot\left\{\beta_{off}^{a}(H)\cdot\left[1-\left(\frac{\lambda_{off}}{\lambda_{on}}\right)^{x}\right]\right\}=2\cdot 0.04\cdot\left\{\left[R_{off}(H)-1\right]\cdot\beta_{off}^{m}(H)\cdot\left[1-\left(\frac{\lambda_{off}}{\lambda_{on}}\right)^{x}\right]\right\}$$

$$F=2\cdot 0.119\cdot\beta_{off}^{m}(H)\cdot\left[1-\left(\frac{\lambda_{off}}{\lambda_{on}}\right)^{4}\right],$$

where λ is the wavelength of radiation *on* and *off* absorption line, and x is the parameter which characterizes the size of aerosol particles.

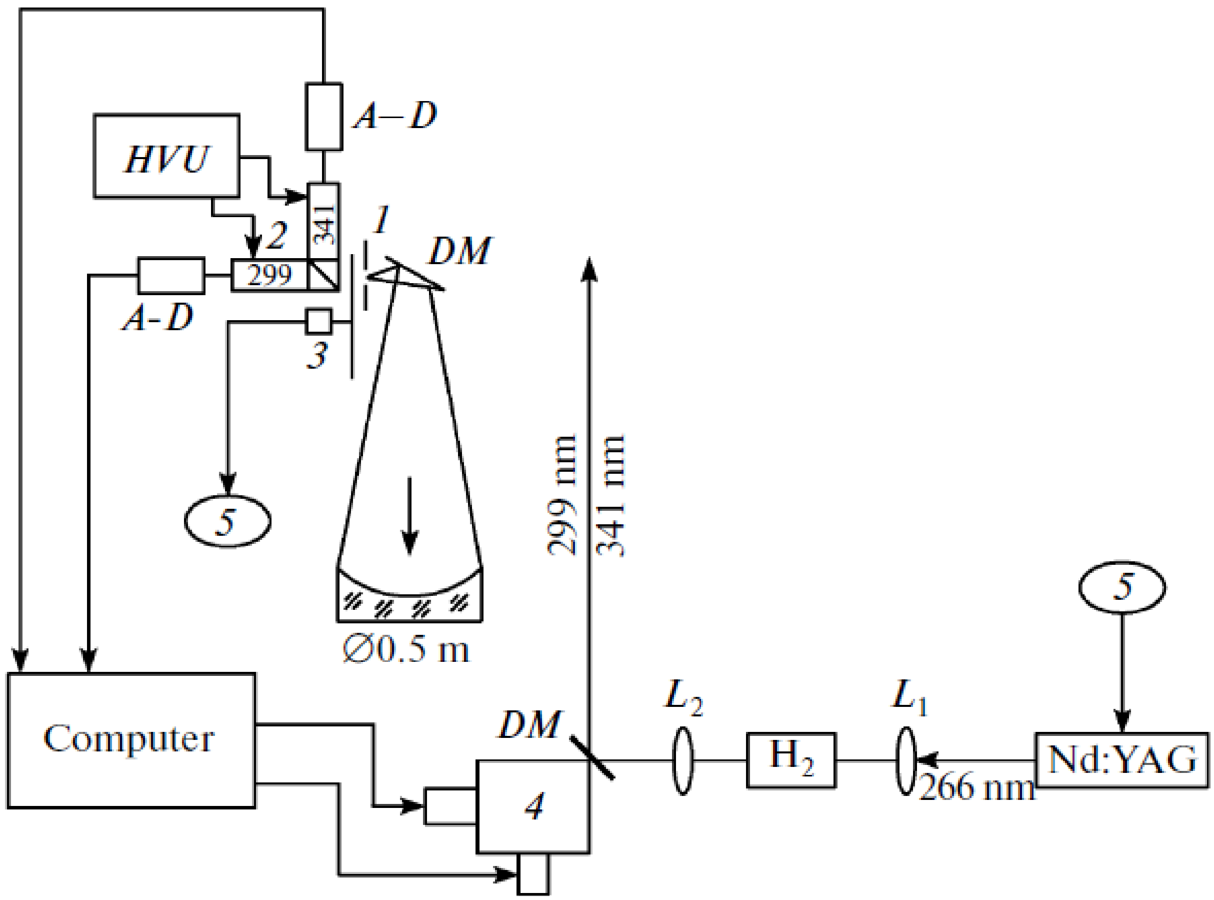

Figure 1. Block-diagram of ozone lidar

(1) field diaphragm, (2) cell for spectral selection with a PMT, (3) mechanical shutter, (4) automated adjustment unit of the rotating output mirror, (5) system for synchronizing the shutter operation time and the moment of emission of laser pulses, (DM) deflecting mirrors, (Nd:YAG) solid-state laser, (H2) SRS conversion cell with H2, (A–D) amplifiers– discriminators, (HVU) high-voltage power supply units for the PMT, and (L1, L2) lenses.

When lidar signals are retrieved at sensing wavelengths 299/341 nm, large aerosol concentration in the altitude range of 0-20 km should be taken into consideration. For calculation, latitudinally average seasonal model values of altitudinal distribution of temperature and molecular backscattering coefficient for winter and summer were incorporated in the algorithm. Examples of retrieved ozone profiles with aerosol correction over 2014 and 2015 are presented in Fig. 2.

The results of lidar measurements at wavelengths of 299 and 341 nm well agree with model estimates, indicating that ozone sensing in altitude range of about 6-18 km is performed with a reasonable accuracy. It should be noted that the retrieved profiles of altitude distribution of ozone concentration closer resemble those from satellite data than according to Krueger model.

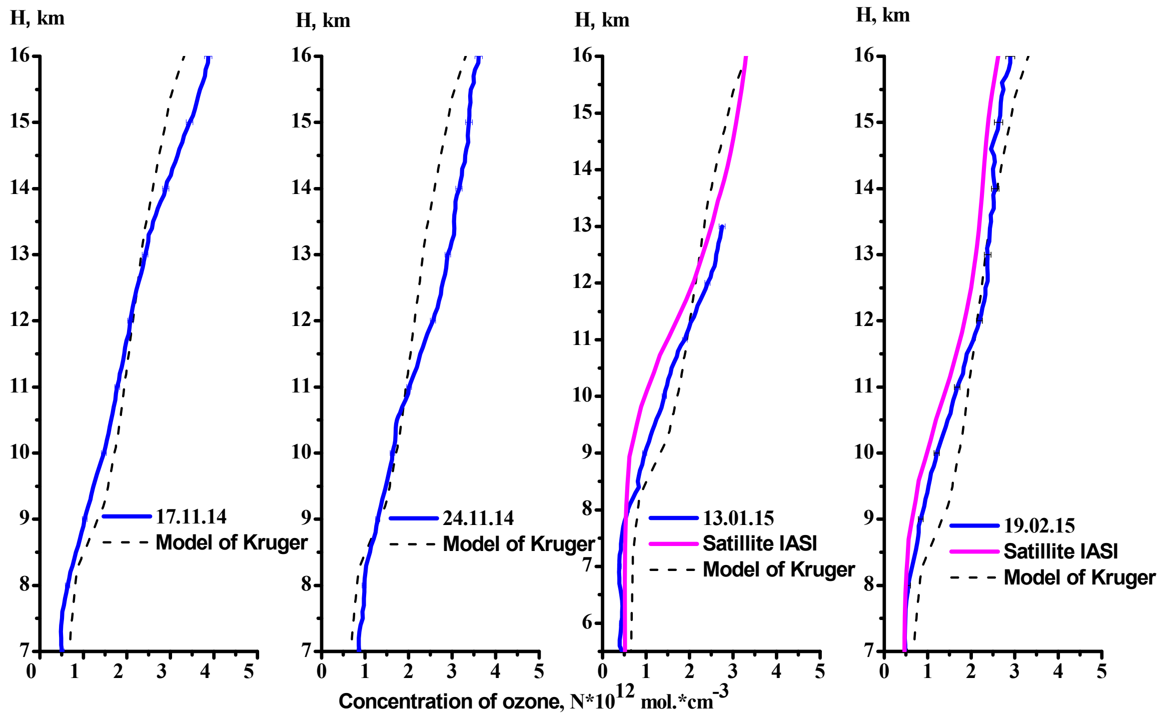

Figure 2. – Retrieved profiles of vertical ozone distribution over 2014 and 2015 at wavelength pair 299/341 nm

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