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DIAL Measurements of Vertical Distribution of Ozone at the Siberian Lidar Station in Tomsk

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***Abstract*—**The paper presents the results of DIAL measurements of the vertical ozone distribution. The ozone lidar operate as part of the measurement complex at Siberian Lidar Station (SLS) of V.E. Zuev Institute of Atmospheric Optics SB RAS, Tomsk (56.5ºN; 85.0ºE) and designed for study of the vertical ozone distribution in the upper troposphere–lower stratosphere. Most suitable wavelengths for measurements of ozone profiles are selected. We present an algorithm for retrieval of vertical distribution of ozone with temperature and aerosol correction during DIAL lidar sounding of the atmosphere. The temperature correction of ozone absorption coefficients is introduced in the software to reduce the retrieval errors. Results of lidar measurement at wavelengths of 299 and 341 nm agree with model estimates, which point to acceptable accuracy of ozone sounding in the 6–18 km altitude range.

***Keywords*—**Lidar, ozone distribution, atmosphere, DIAL.

I. INTRODUCTION

OZONE plays a key role as a shield against hard UV solar radiation for all living on our planet. Also, it is an important climate-forcing agent, which plays a significant role in thermal balance of the planet. Activation of the processes of destruction of stratospheric ozone layer, formation, and spread of ozone anomalies ("ozone holes") over Antarctica, Europe, and Siberia necessitated arrangement of planetary-scale monitoring. This has led to creation of network of ground-based ozonometer stations, which measure the total ozone (TO) content with the help of different spectrophotometers (foreign instruments such as Dobson, Junker, and Brewer spectrophotometers, and national instrument such as M – 124 ozonometer). In addition, information on TO is inferred from satellite (TOMS and other) measurements. Laser sensing of ozonosphere had been regular at a number of observatories since the second half of 1980s. It provides information on vertical ozone distribution (VOD), successfully complicating a similar information obtained *in situ* with the help of ozonesondes, as well through "onion-peeling" approach from satellites (SAGE-II, Terra-Aqua, etc.) [1]. The multiyear lidar observations of stratospheric ozone had made it possible to obtain information on climatology of ozonosphere, especially above 30 km, where ozonesonde data become unrepresentative. The lidar measurements of VOD are performed on the basis of method of differential absorption of backscattered energy of laser radiation in UV wavelength range of 240-360 nm (the so-called Hartley-Huggins band). As a rule, for sensing the stratospheric ozone, the fundamental frequency of excimer XeCl laser (308 nm) is used as λ_{on}, where absorption of sensing radiation by ozone is strong. At the same time, as a reference frequency (with weak absorption) λ_{off}, researchers use either the first component of its SRS conversion in hydrogen (353 nm), with efficiency of conversion reaching 40%, or the third harmonic of Nd:YAG laser (355 nm), the energy of which may reach more than 100 mJ.

Long-term period of lidar observations of stratospheric ozone (since 1989) at the measurement complex SLS, V.E. Zuev Institute of Atmospheric Optics, Siberian Branch, Russian Academy of Sciences (Tomsk: 56.5°N; 85.0°E) [2] showed that the part, most significant for studying the ozonosphere, is located in the lower stratosphere, where ozone is subject to the effect of dynamic factor. This part totally determines the TO variations in atmospheric column. Poorly studied scenarios of stratosphere-troposphere exchange, deformation of ozonosphere by jet streams, and formation of fine tongue-like structure of ozone layer also develop in this part. However, higher concentration sensitivity of lidar measurements is required to perform these studies in altitude ranges of upper troposphere – lower stratosphere, thus necessitating passage to shorter-wavelength region of UV spectrum, where ozone absorption cross section is greater.

II. SELECTION OF WAVELENGTHS

Excimer KrF laser (248 nm) or the fourth harmonic of Nd:YAG laser (266 nm) in combination with technique of SRS conversion in H_2, D_2, CO_2, and other gases [3]-[6] are usually used for tropospheric measurements of ozone. Hydrogen and deuterium are most widespread, in this regard. A possible set of wavelengths, corresponding to the 1st, 2nd, and 3rd Stokes (C) frequencies of SRS conversion in H_2, D_2, and CO_2, is presented in Table I.

TABLE I
SPECIFICATIONS OF LQ529B PUMPING LASER

Pumping radiation	Wavelengths (nm) corresponding to the Stokes frequencies (*C*) of the SRS conversion in H_2 C1 C2	In D_2 C1 C2 C3	in CO_2 C2 C3
Nd:YAG, 266 nm	299 341	289 316	287 299
KrF, 248 nm	277 313	268 291 319	

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Diverse wavelength combinations are used in different altitude ranges of the troposphere and lower stratosphere. For instance, the wavelength pairs (289, 316) and (287, 299) nm make it possible to obtain ozone profile up to the heights of about 10 km [3], [4]; the pair (292, 319) nm can be used up to the heights of 14-16 km [5]; and the pairs (277, 313) and (292, 313) nm can be used up the heights of 8-12 and 15 km, respectively [6].

The wavelength λ_{on}=299 nm lies in the region of ozone absorption band with absorption cross section $\sigma_{299}=4.4\cdot10^{-19}$ cm^2, a factor of 3 larger than the absorption cross section at the wavelength of 308 nm: $\sigma_{308}=1.4\cdot10^{-19}$ cm^2. The maximal height of sensing is determined primarily by the range from which the signal at λ_{on} is recorded, which is always shorter than the range from which the signal at λ_{off} is recorded, due to stronger absorption by ozone. From this viewpoint, λ_{on}=299 nm is more preferable than 277 or 292 nm. Wavelengths 299 and 341 are implemented in one sensing beam (in one SRS cell), in contrast to, e.g., pair (292, 313) nm. It is noteworthy that system on the basis of hydrogen-filled SRS cell is cheaper than deuterium-filled cell. It is also important to remember about technical feasibility of spectral separation, during reception, of signals at closely lying wavelengths.

To study VOD in the upper troposphere – lower stratosphere, we developed and put into measurement mode a lidar, as part of SLS, for measuring the ozone concentration in the upper troposphere – lower stratosphere [7]. Sensing is performed according to the method of differential absorption and scattering at wavelength pair of 299/341 nm, which are, respectively, the first and second Stokes components of SRS conversion of 4th harmonic of Nd:YAG laser (266 nm) in hydrogen. Lidar with receiving mirror 0.5 m in diameter is used to implement sensing of VOD in altitude range of 6-16 km.

III. ALGORITHM FOR CALCULATING THE VOD

We will consider an algorithm for calculating VOD, 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 following form:

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

$$\cdot\left\{\begin{array}{l}\underbrace{\frac{d}{dH}\ln\left[\frac{N_{off}(H)}{N_{on}(H)}\right]}_{B}-\underbrace{\frac{d}{dH}\ln\left[\frac{\beta^{a}_{off}(H)+\beta^{m}_{off}(H)}{\beta^{a}_{on}(H)+\beta^{m}_{on}(H)}\right]}_{C}-\\ \underbrace{2\cdot\left[\alpha^{a}_{off}(H)-\alpha^{a}_{on}(H)\right]}_{D}-\underbrace{2\cdot\left[\alpha^{m}_{off}(H)-\alpha^{m}_{on}(H)\right]}_{F}\end{array}\right\}$$

where $N(H)$ is the recorded return signal; α_{α} is the extinction coefficient of aerosol scattering; β_{α} is the aerosol backscattering coefficient; αm 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. 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 II.

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.

TABLE II
OZONE ABSORPTION COEFFICIENTS AS FUNCTIONS OF TEMPERATURE FOR DIFFERENT 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	$6\ 10^{-22}$	$6\ 10^{-22}$	$6\ 10^{-22}$	$6\ 10^{-22}$	$1.2\ 10^{-21}$

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:

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

$$D=2\cdot0.04\cdot\left\{\beta^{a}_{off}(H)\cdot\left[1-\left(\frac{\lambda_{off}}{\lambda_{on}}\right)^{x}\right]\right\}=2\cdot0.04\cdot\left\{\left[R_{off}(H)-1\right]\cdot\beta^{m}_{off}(H)\cdot\left[1-\left(\frac{\lambda_{off}}{\lambda_{on}}\right)^{x}\right]\right\}$$

$$F=2\cdot0.119\cdot\beta^{m}_{off}(H)\cdot\left[1-\left(\frac{\lambda_{off}}{\lambda_{on}}\right)^{4}\right],$$

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

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.

IV. LIDAR FOR VOD MEASUREMENTS

As the source of laser radiation, we use the 4th harmonic (266 nm) of fundamental frequency of Nd:YAG laser (model LS-2134UT, LOTIS TII firm, Minsk) with its subsequent SRS conversion in hydrogen to the first (299 nm) and second (341 nm) Stokes components. The vertical ozone profiles are retrieved from lidar signals with the help of universal software [8], which allows the altitude profiles of ozone concentration to be calculated according to the method of differential absorption and scattering for three wavelength pairs (272, 289) nm, (299, 341) nm, and (308, 353) nm.

The software applies linear smoothing for both input lidar data and for retrieval results. Linear smoothing (smoothing by sliding average) is a well-known procedure, widely used for processing the experimental data in different natural science regions. Linear smoothing is a particular case of digital filtering, possessing random signal error, filter with rectangular window, and unit weighting coefficients.

Fig. 1 presents the block-diagram of the developed lidar.

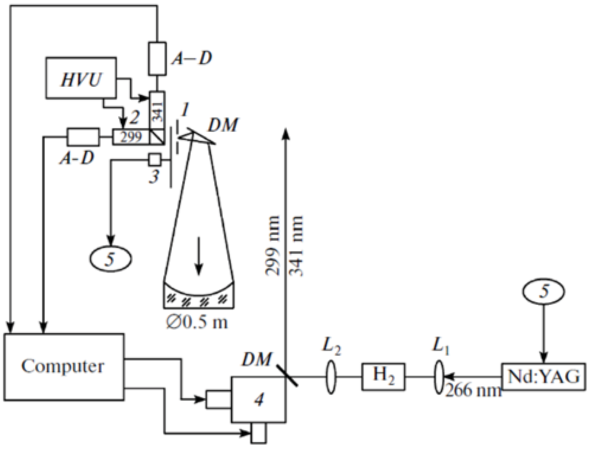

Fig. 1 Block-diagram of the lidar: (*1*) field diaphragm, (*2*) cell for spectral selection with a PMT, (*3*) mechanical shutter, (*4*) automated adjustment unit of the rotating output mirror, (*5*) system for synchronizing the shutter operation time and the moment of emission of laser pulses, (*DM*) deflecting mirrors, (Nd:YAG) solid-state laser, (H_2) SRS conversion cell with H_2, (*A–D*) amplifiers– discriminators, (*HVU*) high-voltage power supply units for the PMT, and (L_1, L_2) lenses

The software permits us: to read off lidar data and save the retrieval results in ASCII format; smooth the lidar signals and retrieval results using sliding mean. The software incorporates temperature correction of ozone absorption coefficients to reduce the retrieval errors. When lidar signals are retrieved at sensing wavelengths (272, 289) nm and (299, 341) nm, large aerosol concentrations in the altitude range of 0-20 km should be taken into consideration; therefore, aerosol correction is accounted for in the software.

Seasonal model values of altitude distribution of temperature and molecular backscattering coefficient for midlatitude winter and summer are introduced in the software for the calculations.

V. RESULTS OF MEASUREMENTS

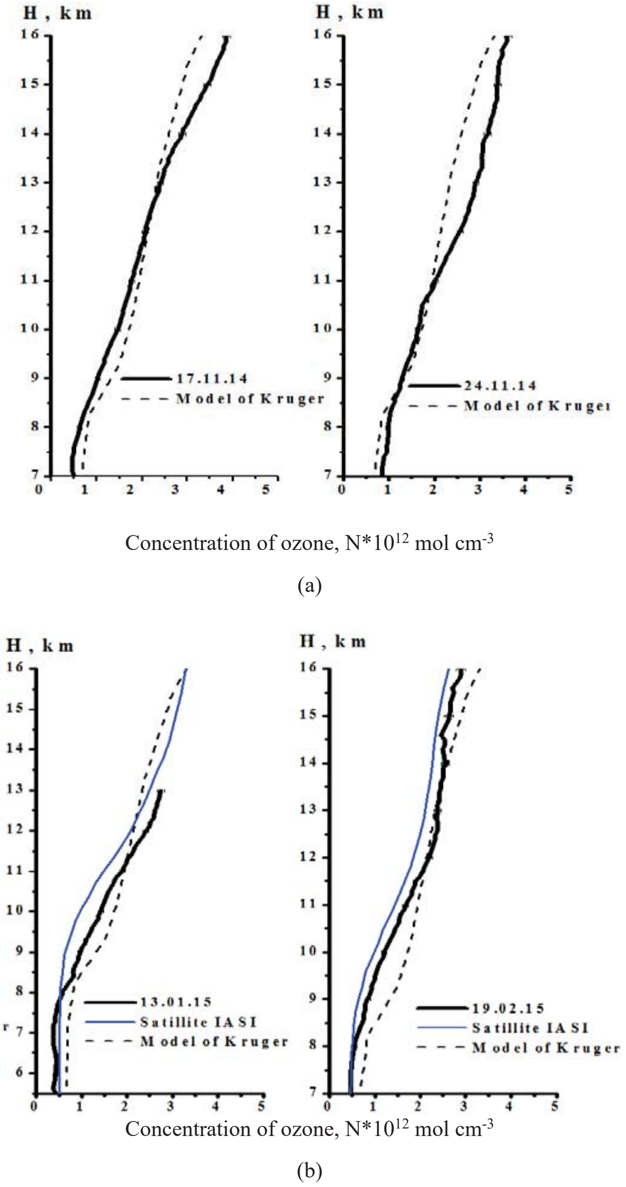

Fig. 2 Retrieved vertical profiles of ozone concentration at wavelengths (299, 341) nm (a) November 2014, (b) January-February 2015) compares them with Krueger midlatitude model and IASI satellite data

Examples of retrieved profiles of VOD are presented in Fig. 2 (a) November 2014 and (b) January-February 2015.

Fig. 2 presents the retrieved profiles of ozone concentration for November 2014 and January-February 2015 and compares them with Krueger midlatitude model [9] and IASI satellite data.

The retrieved and model profiles quite well coincide, while deviations from IASI satellite data are quite natural for ozone dynamics in separate observation days.

VI. CONCLUSION

Lidar measurements at wavelengths 299 and 341 nm agree with model estimates and satellite (IASI) observations, which indicate acceptable accuracies of ozone sensing in the altitude range of about 6-16 km. We can also note that, for ozone sensing in the altitude range of 5-20 km, lidar on the basis of Nd:YAG laser is more preferable than lidar on the basis of excimer KrF laser which is more costly, more complex in exploitation, needs especially pure gases for working mixture, frequent purification or replacement of resonator optics.

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