

LIDAR MEASUREMENTS OF OZONE IN THE UPPER TROPOSPHERE – LOWER STRATOSPHERE AT SIBERIAN LIDAR STATION IN TOMSK

O. A. Romanovskii^{1,2*}, S.I. Dolgii¹, V. D. Burlakov¹, A.A. Nevzorov¹ and A. V. Nevzorov¹

¹ V.E. Zuev Institute of Atmospheric Optics SB RAS, 1 Zuev Square, Tomsk 634021, Russia, roa@iao.ru

² National Research Tomsk State University, 36 Lenina Pr., Tomsk 634050, Russia

ABSTRACT

The paper presents the results of DIAL measurements of the vertical ozone distribution at the Siberian lidar station. 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 vertical ozone distribution in altitude range of 6-16 km. The temperature correction of ozone absorption coefficients is introduced in the software to reduce the retrieval errors.

1. 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 Siberian Lidar Station (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.

2. 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 H₂, D₂, CO₂, 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 H₂, D₂, and CO₂, is presented in Table.

Pumping radiation	Wavelengths (nm) corresponding to the Stokes frequencies (C) of the SRS conversion					
	in H ₂		In D ₂			in CO ₂
	C1	C2	C1	C2	C3	C2 C3
Nd:YAG, 266 nm	299	341	289	316		287 299
KrF, 248 nm	277	313	268	291	319	

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 $\lambda_{on}=299$ nm lies in the region of ozone absorption band with absorption cross section $\sigma_{299}=4.4 \cdot 10^{-19}$ cm², a factor of 3 larger than the absorption cross section at the wavelength of 308 nm: $\sigma_{308}=1.4 \cdot 10^{-19}$ cm². 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, $\lambda_{on}=299$ 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 the vertical ozone distribution 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 vertical ozone distribution in altitude range of 6-16 km.

3. LIDAR FOR SENSING THE VERTICAL OZONE DISTRIBUTION IN THE UPPER TROPOSPHERE – LOWER STRATOSPHERE

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

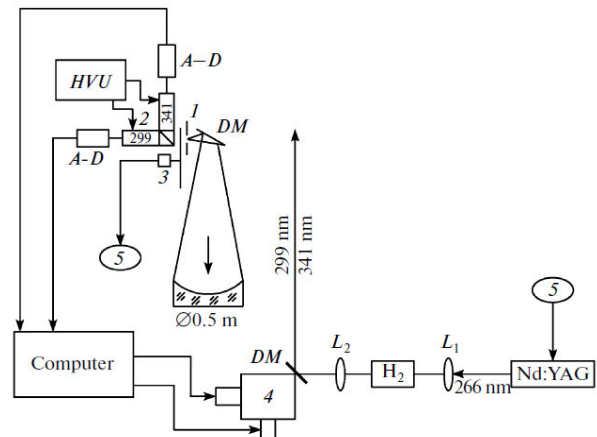


Fig. 1. Block-diagram of the lidar: (1) field diaphragm, (2) cell for spectral selection with a PMT, (3) mechanical shutter, (4) automated adjustment unit of the rotating output mirror, (5) system for synchronizing the shutter operation time and the moment of emission of laser pulses, (DM) deflecting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

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.

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.

4. RESULTS OF MEASUREMENTS

Examples of retrieved profiles of vertical ozone distribution are presented in Figure 2 (a - November 2014, b - January-February 2015).

Figure 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.

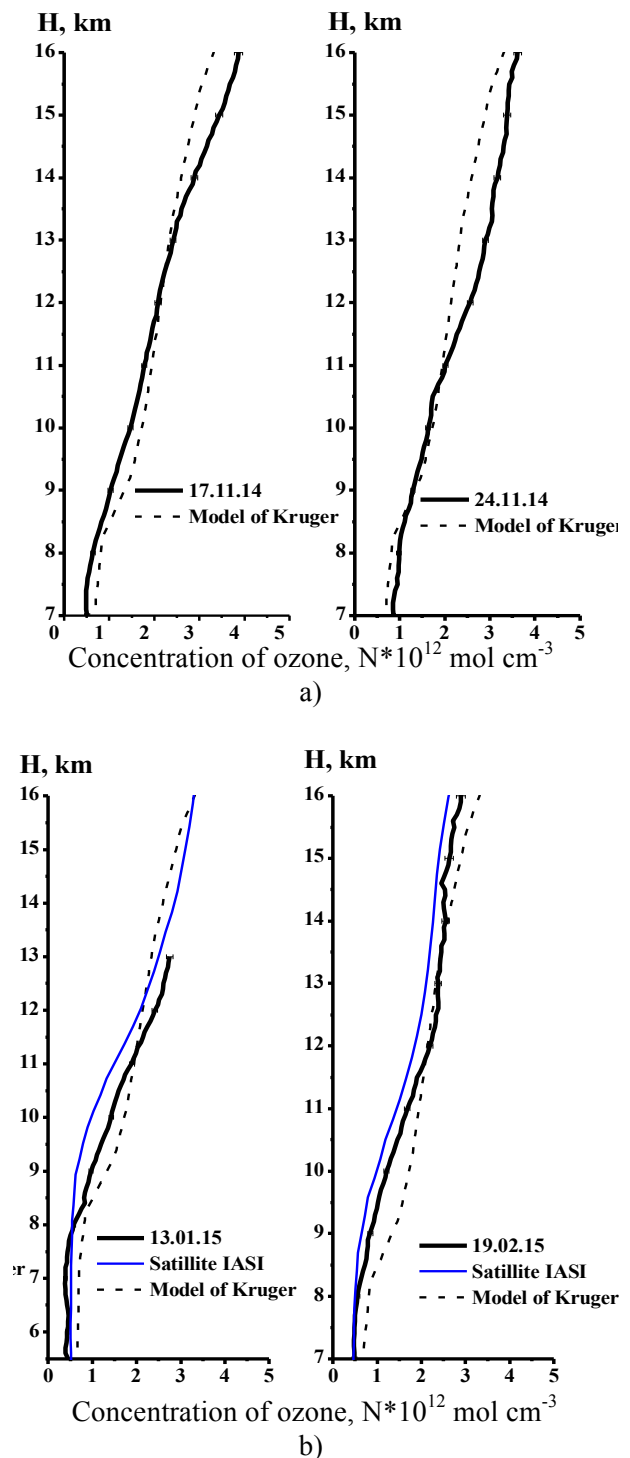


Figure 2 – Retrieved vertical profiles of ozone concentration at wavelengths 299/341 nm (a - November 2014, b - January-February 2015) compares them with Krueger midlatitude model and IASI satellite data.

5. CONCLUSIONS

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.

ACKNOWLEDGEMENTS

This work was supported in part by the Ministry of Science and Education of the Russian Federation (Agreement No. 14.604.21.0100 (RFMEFI60414X0100)), the Russian Science Foundation (Agreement no. 14-27-00022), The President of the Russian Federation (Grant NSh-4714.2014.5).

REFERENCES

- [1] <http://sage.nasa.gov/SAGE2/>
- [2] Burlakov, V.D., Dolgii, S.I., Nevzorov, A.V., 2004, Modification of the measuring complex at the Siberian Lidar Station. *Atmospheric and oceanic optics*. **17** (10), 756-762.
- [3] Galani, E., Balis, D., Zanis, P., Zerefos, C., Papayannis, A., Wemli, H., and Gerasopoulos, E., 2003, Observations of stratosphere-to-troposphere transport events over the eastern Mediterranean using a ground-based lidar system. *J. Geophys. Res.* **108**, D12. P.STA12/1-STA12/10.
- [4] Masahisa Nakazato, Tomohiro Nagai, Tetsu Sakai, and Yasuo Hirose, 2007, Tropospheric ozone differential-absorption lidar using stimulated Raman scattering in carbon dioxide. *Appl. Opt.* **46** (12), 2269-2279.
- [5] Bukreev, V.S., Vartapetov, S.K., Veselovskii, I.A., Galustov, A.S., Kovalev, Yu.M., Prokhorov, A.M., Svetogorov, E.S., Khmelevtsov, S.S., Li, Ch.Kh., 1994, Lidar system for sensing the stratospheric and tropospheric ozone on the basis of excimer lasers. *Quantum Electronics*. **21** (6), 591- 596.
- [6] Eisele, H., H.E. Scheel, R. Sladkovic, and T. Trickl, 1999, High resolution lidar measurements of stratosphere-troposphere exchange. *J. Atmos. Sci.*, **56** (3), 319-330.
- [7] Burlakov, V.D., Dolgii, S.I., Makeev, A.P., Nevzorov, A.V., Romanovskii, O.A., Kharchenko, O.V., 2010, A differential-absorption lidar for ozone sensing in the upper atmosphere-lower stratosphere. *Instruments and Experimental Techniques*. 2010. **53** (6). 886-889.
- [8] Nevzorov, A.V., Nevzorov, A.A., Romanovskii, O.A., 2014, Software for retrieving the ozone altitude profiles from data of atmospheric laser sensing. *Proc. SPIE*. **9292**, 92923L. doi:10.1117/12.2074718.
- [9] Krueger A.J. , Minzner R.A. , 1976, A mid-latitude ozone model for the 1976 U.S. standard atmosphere. *J. Geophys. Res.*, **81** (D24) 4477-4481.