Differential absorption lidar for ozone sensing in the upper troposphere – lower stratosphere

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A lidar for measurements of ozone distribution in the upper troposphere – lower stratosphere for studying the ozone dynamics in the tropopause region and, in particular, the processes of stratospheric – tropospheric exchange was developed at Siberian Lidar Station of the Institute of Atmospheric Optics SB RAS in Tomsk (56.5°N, 85.0°E). The sensing is performed using the method of differential absorption and scattering at a pair of wavelengths (299 and 341 nm), which are the first and second Stokes components of radiation transformation of the Nd:YAG laser pumping line at 266 nm, based on stimulated Raman effect in hydrogen. Using lidar with the receiving mirror of 0.5 m in diameter, we realized the ozone sensing in altitude range 5–18 km.

The sensing channel of the vertical ozone distribution (VOD) in the stratosphere by the method of differential absorption (DA) is based on the use of an excimer Xe–Cl laser (308 nm) in combination with the first Stokes component of its transformation based on the effect of the simulated Raman scattering (SRS) in hydrogen (353 nm) operates at the Siberian Lidar Station (SLS) of the Institute of Atmospheric Optics SB RAS. The obtainable lidar returns at wavelengths of 308 (on) and 353 nm (off) provide for the concentration sensitivity of the DA-method, sufficient for determining the stratospheric ozone concentration at altitudes of 13–35 km, i.e., in the layer with maximal ozone content, where the ozone concentration is $(2–6)\cdot 10^{12}$ mol/cm^3.

The lidar observational data on the stratospheric ozone obtained during an extended period since 1989 have shown that the region of ozonosphere, significant for investigation, is located in the low stratosphere, where the ozone is subjected to the dynamic factors. This region determines fully the variability of the total ozone content (TOC) in the atmospheric column. In the same area we can observe the poorly known phenomena of stratospheric – tropospheric exchange, deformation of the ozonosphere by jet flow, the formation of the fine "tongue" structure of the ozone layer. However, for such investigations in the altitude ranges of the upper troposphere – lower stratosphere a large concentration sensitivity of lidar measurements is required, i.e., it is necessary to use a shorter-wave range of UV spectrum, where the ozone absorption cross section is greater.

1. Estimation of the possibility of ozone sensing in the troposphere – stratosphere at 299/341 nm wavelengths

For measurements of ozone in troposphere we traditionally used an excimer KrF laser (248 nm) or 4th harmonics of Nd:YAG laser (266 nm) in combination with the technique of SRS transformation in H_2, D_2, CO_2, and other gases.[1–4] The most widespread gases are hydrogen and deuterium. A possible set of wavelengths corresponding to the 1st, 2nd, and 3rd Stokes (S) frequencies of SRS transformation in H_2, D_2, and CO_2 is given in Table.

Table. Wavelengths corresponding to Stokes frequencies of SRS transformation of radiation at 266 and 248 nm in hydrogen, deuterium, and carbon dioxide

Pumping radiation	Wavelengths (nm) corresponding to Stokes frequencies of SRS transformation						
	in hydrogen		in deuterium			in carbon dioxide	
	C1	C2	C1	C2	C3	C2	C3
Nd:YAG, 266 nm	299	341	289	316	–	287	299
KrF, 248 nm	277	313	268	292	319	–	–

In different altitude ranges of troposphere and lower stratosphere different combinations of wavelengths are used. Thus, wavelengths of 289/316 and 287/299 nm allow us to obtain the ozone profile up to an altitude of 10 km [Refs. 1 and 2]; the wavelengths 292 /319 nm – up to 14–16 km [Ref. 3]; the wavelengths 277/313 and 292/313 nm – up to 8–12 km and 15 km, respectively.[4]

We made preliminary numerical estimates of potentialities of vertical ozone distribution sensing in the upper troposphere – lower stratosphere using the pair 299/341 nm. We used λ_{on} = 299 nm for the region of the ozone absorption band with the absorption cross section $\sigma_{299} = 4.4\cdot 10^{-19}$ cm^2, which is by three times higher than the absorption cross section at λ = 308 nm ($\sigma_{308} = 1.4\cdot 10^{-19}$ cm^2). The maximal sensing altitude was first determined by the distance of signal recording at λ_{on}, which is always less than the signal recording distance at λ_{off} due a greater ozone absorption. From this standpoint

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λ_{on} = 299 nm is more preferable, than 277 or 292 nm. The wavelengths 299 and 341 nm are realized in one sensing beam (in one SRS cell) as differentiated from, for example, 292/313 nm. In this case, the system based on the SRS cell with hydrogen is cheaper than that with deuterium. It is necessary to take into account the technical possibility of spectral separation, when receiving signals at close wavelengths.

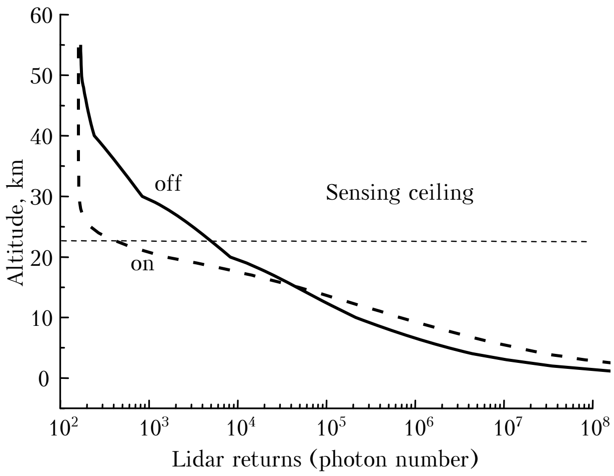

Fig. 1. Calculated lidar returns accumulated during 1.5 h at wavelengths of 299 (on) and 341 nm (off).

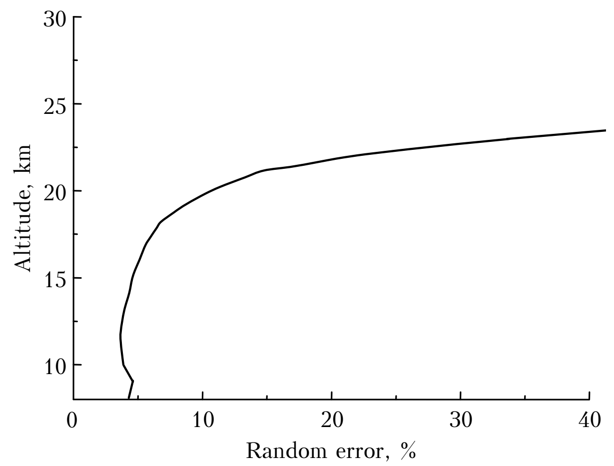

Fig. 2. The error of ozone concentration determination from lidar returns in Fig. 1.

The following actual lidar parameters were used in calculations: a radiation energy of 20 mJ at both wavelengths; a laser pulse repetition rate of 15 Hz; a receiving mirror diameter of 0.5 m; a time of signal accumulation of 1.5 hours. To determine the transceiver efficiency, we used the actual values of transmission of optical elements of spectral selection and the photomultiplier efficiency, the noise was taken from real measurements by UV lidar based on the Xe–Cl laser. Figures 1 and 2 show the calculated lidar returns (on and off), as well as errors in determination of ozone concentrations, respectively.

The calculations have shown that the use of λ_{on} = 299 nm, at which σ is greater than at λ_{on} = 308 nm, affects the decrease of the sensing ceiling height approximately up to 22 km (the ozone maximum in Tomsk is located in altitude range 19–21 km), but there appeared a possibility to measure ozone in troposphere. An error of determination of ozone concentration is within 4–10% up to altitudes of about 20 km.

2. Technical description of lidar

Figure 3 shows the block-diagram of the lidar. As a source of laser radiation, the fourth harmonics (266 nm) of the basic frequency of Nd:YAG laser radiation is used (model LS-2134UT (LOTTS TII, Minsk) with its subsequent SRS-transformation in hydrogen to the first (299 nm) and second (341 nm) Stokes components.

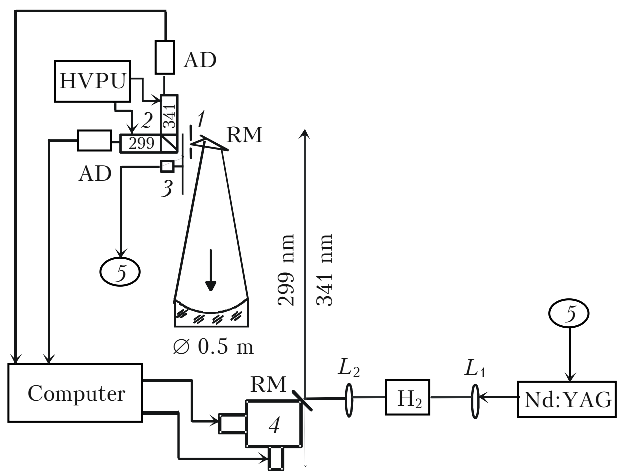

Fig. 3. Block-diagram of lidar: the field pin-hole (*1*); the cell of spectral selection with a photomultiplier (*2*); the mechanical obturator; the rotational mirrors RM (*3*); automated aligning block of the output rotational mirror; Nd:YAG laser; H_2 is the cell of SRS transformation with hydrogen; AD is amplifier-discriminator; HVPU is high-voltage power unit; L_1 and L_2 are lenses (*4*); the system of time synchronization of the obturator acting and laser pulse sending (*5*).

The receiving telescope is designed by the Newton scheme based on the basic mirror with a diameter of 0.5 m and a focal distance of 1.5 m. The recording channel of lidar includes photomultipliers (R7207-1) and HAMAMATSU amplifiers-discriminators (C3866). Recording of lidar returns is conducted in the count mode of photocurrent pulses.

The mechanical obturator provides for the linear regimes of photomultiplier operation, which cuts a high-power optical signal from the near sensing zone. The automated aligning block of the output rotating mirror is designed on the basis of the computer-controlled step engines.

The SRS-cell is made of a stainless steel tube with an inside diameter of 3 cm and a length of 1 m. Input and output windows are made of KU quartz. The pumping pulse energy at λ = 266 nm is 60 mJ.

The pumping power density, necessary for obtaining SRS transformation, is provided for by the lens L_1 with a focal length of 1 m, which is mounted before the SRS-cell and focuses the radiation to its centre. A collimating lens L_2 is mounted after the cell confocally to the focusing lens.

The efficiency of SRS transformation was determined depending on the hydrogen pressure in the cell, which varied from 1 to 9 atm. The dependence of relative intensities of pumping radiation (266 nm) of the first (299 nm) and second (341 nm) Stokes components of SRS transformation on the hydrogen pressure, obtained at the output from SRS-cell, is given in Fig. 4.

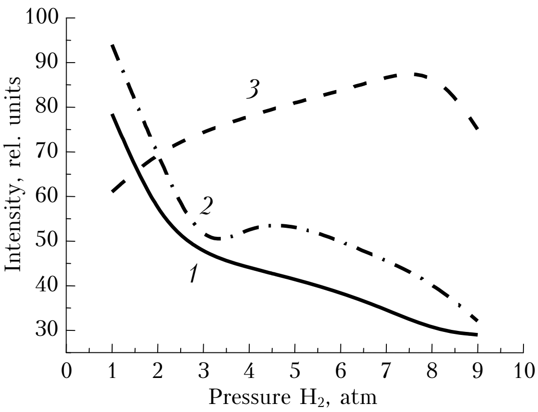

Fig. 4. The dependence of relative pumping intensities (*1* – 266 nm), of the first (*2* – 299 nm) and second (*3* – 341 nm) Stokes components of SRS transformation on the hydrogen pressure.

At a hydrogen pressure of 2 atm the line intensities at 299 and 341 nm are comparable, which provides for a possibility of ozone sensing at equal radiation powers at these wavelengths. However to increase the sensing ceiling height, the pressure of 1 atm is more efficient, because in this case the power is redistributed in favor of 299 nm line, which is stronger absorbed by ozone than 341 nm line.

3. The first measurement results

Figure 5 shows the type of lidar returns and the reconstructed profile of ozone concentration, respectively. The lidar returns were recorded in the photocurrent pulse count mode over 25 000 shots (the accumulation time was about 30 minutes).

The reached ceiling height of the lidar return recording at 299 nm exceeded 20 km. The VOD profile (Fig. 5*b*) is given in comparison with the middle-latitude winter AFGL model.[5] In the altitude range between 5 and 14 km the reconstructed profile and model curve well agree. At altitudes higher than 14 km the measured profile differs from the model one, that is natural for ozone dynamics for individual observation days.

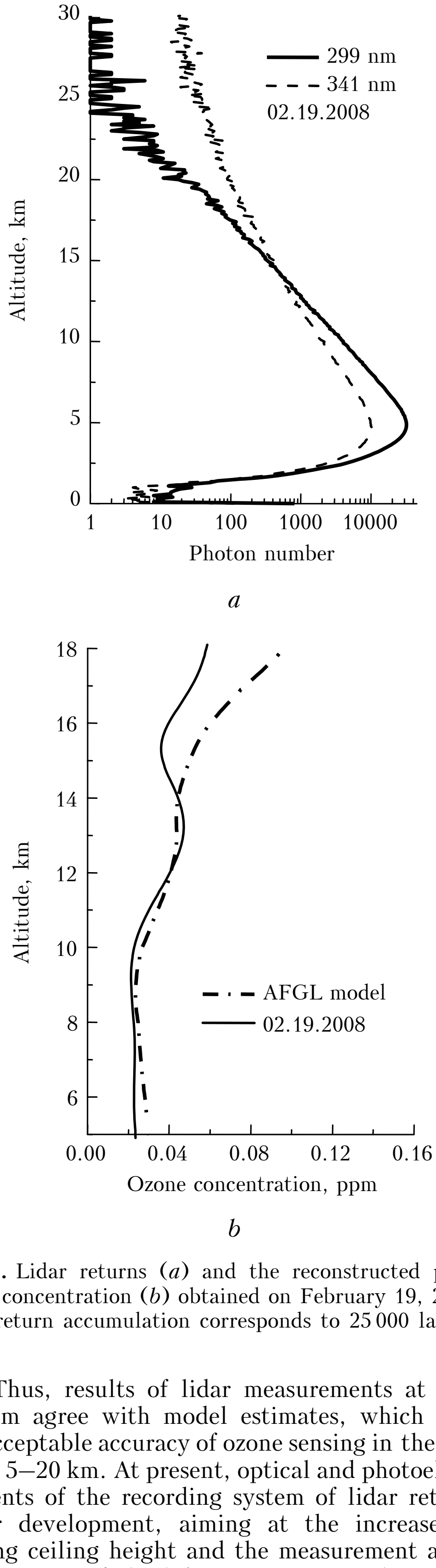

Fig. 5. Lidar returns (*a*) and the reconstructed profile of ozone concentration (*b*) obtained on February 19, 2008. The lidar return accumulation corresponds to 25 000 laser shots.

Thus, results of lidar measurements at 299 and 341 nm agree with model estimates, which point to the acceptable accuracy of ozone sensing in the altitude range 5–20 km. At present, optical and photoelectronic elements of the recording system of lidar returns are under development, aiming at the increase of the sensing ceiling height and the measurement accuracy. The increase of the lidar return accumulation time is also possible.

Note in conclusion that for ozone sensing at 5–20 km altitude range the lidar based on ND:YAG laser is preferable as compared to the lidar based on the excimer KrF laser, which is more expensive, more complicated in operation, requires very pure gases of working mixtures, and frequent cleaning or replacement of resonator optics.

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