SINGLE–FREQUENCY LASER SOUNDING OF THE STRATOSPHERIC OZONE LAYER

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The stratospheric ozone measurements were carried out within the 20–30 km *altitude ranges using a single–frequency lidar. The error in determining the ozone concentrations was at best , 5–10 %. Therefore a single–frequency lidar can be used for evaluation of anthropogenic effects on the ozone layer.*

1. As is well known, the lidar sensing equation in the far–diffraction zone in the single scattering approximation is given in the form

$$N(H) = N_0\eta K_0 S\Delta H H^{-2}[\sigma_{\pi a}(H) + \sigma_{\pi m}(H)]\, q^2(H)\ ;$$

$$q^2(H) = \exp\left\{ -2\int\limits_{H_0}^{H}[\sigma_a(h) + \sigma_m(h) + \alpha_{O_3}(h)]\,dh\right\}. \qquad (1)$$

Employing the models of the atmosphere developed in Ref. 1, the values of $\sigma_{\pi a}$, σ_m, $\sigma_{\pi m}$, σ_a, α_{O_3}, and q were calculated at $\lambda = 308$ nm. (The analysis made in Ref. 2 showed that absorption due to such gases as SO_2, NO_2, HNO_3, H_2O_2, and N_2O_5 near 300 nm can be ignored in this altitude range.) The data on molecular scattering and absorption cross sections of ozone were borrowed from Ref. 2: $\sigma^0_m = 5.59\cdot10^{-26}$ cm^2, $\sigma^0_{\pi m} = 6.71\cdot10^{-27}$ cm$^2\cdot$str^{-1}, $\sigma^0_{O_3} = 1.17\cdot10^{-19}$ cm^2.

TABLE. Cross sections of absorption and molecular scattering.

λ, nm	σ_m, cm^2	$\sigma^0_{\pi m}$, cm$^2\cdot$str^{-1}	$\sigma^0_{O_3}$, cm^2
308	5.59 (–26)	6.71 (–27)	1.17 (–19)

For the lower stratosphere (10–30 km) $\sigma_m > \sigma_a$ and $\sigma_{\pi m} > \sigma_{\pi a}$. This condition is also satisfied for the altitudes $H > 30$ km.

Let the backscattered signals be received from the two altitudes H_1 and H_2 separated by the gating interval ΔH. Then, neglecting the values σ_a and $\sigma_{\pi a}$ and taking into account that $\sigma_m = \sigma^0_m\rho$ and $\sigma_{\pi m} = \sigma^0_{\pi m}\rho$, we obtain

$$\frac{N(H_2)}{N(H_1)} = \frac{\rho(H_2)H_1^2}{\rho(H_1)H_2^2}\exp\left\{-2\int\limits_{H_1}^{H_2}[\sigma^0_m\,\rho(h) + \sigma^0_{O_3}[O_3(h)]]\,dh\right\} \qquad (2)$$

In formulas (1) and (2) N is the number of recorded photoelectrons, N_0 is the number of emitted photons, η is the quantum efficiency of the photomultiplier, K_0 is the total efficiency of the optical train, S is the area of the receiving antenna, ΔH is the spatial resolution (gating interval), H is the altitude of the sounded layer of the atmosphere, $\sigma_{\pi a}$, $\sigma_{\pi m}$, σ_a, and σ_m are the coefficients of backscattering and total aerosol and molecular scattering, respectively, α is the gas absorption coefficient, q^2 is the laser radiation transmission along the path lidar–sounded layer–lidar, and ρ is the mass concentration of atoms and molecules in the atmosphere (subsequently referred to as the density of the atmosphere, for brevity).

Taking the logarithm of Eq. (2) we obtain

$$[O_3(\Delta H)] = \frac{1}{2\Delta H s^0_{O_3}}\ln\frac{N_1\rho_2H_1^2}{N_2\rho_1H_2^2} - \frac{\sigma^0_m}{\sigma^0_{O_3}}\rho(\Delta H)\ , \qquad (3)$$

where $[O_3(\Delta H)]$ and $\rho(\Delta H)$ are the corresponding values averaged over the layer ΔH, $N_1 = N(H_1)$, $\rho_1 = \rho(H_1)$, etc.

Consider the systematic errors in determining the ozone concentration. They are essentially summed over the errors due to fluctuations in the number of recorded photoelectrons and uncertain knowledge of atmospheric density at the sounding altitudes and of the distance to the corresponding layers.

Using the well–known relation

$$\Delta\,[O_3(\Delta H)] = \sqrt{\left\{\frac{\delta\,|\,O_3(\Delta H)\,|}{\delta H_1}\,\Delta H_1\right\}^2 + \dots}\ , \qquad (4)$$

where $[O_3(\Delta H)]$ is the ozone concentration in the layer ΔH and assuming that $\rho(\Delta H) \simeq \dfrac{\rho_1 + \rho_2}{2}$, $\Delta H = H_1 - H_2$, and $\Delta N = \sqrt{N}$ (the Poisson distribution), after differentiation of the quantity $[O_3(\Delta H)]$ with respect to N_1, N_2, H_1, H_2, ρ_1, and ρ_2, we obtain

$$\left\{\frac{\partial[O_3(\Delta H)]}{\partial N_1}\Delta N_1\right\}^2 + \left\{\frac{\partial[O_3(\Delta H)]}{\partial N_2}\Delta N_2\right\}^2 =$$

$$= \frac{1}{(2\Delta H\sigma^0_{O_3})^2}\left(\frac{1}{N_1} + \frac{1}{N_2}\right); \qquad (5)$$

$$\left\{\frac{\partial[O_3(\Delta H)]}{\partial\rho_1}\Delta\rho_1\right\}^2 + \left\{\frac{\partial[O_3(\Delta H)]}{\partial\rho_2}\Delta\rho_2\right\}^2 \simeq$$

$$\simeq 2(\Delta\rho)^2\left[\left(\frac{1}{2\Delta H\sigma^0_{O_3}\rho}\right)^2 + \left(\frac{\sigma^0_m}{2\sigma^0_{O_3}}\right)^2\right]. \qquad (6)$$

Formula (6) is written down on the assumption that $\rho_1 \simeq \rho_2 = \rho$ and $\Delta\rho_1 \simeq \Delta\rho_2 = \Delta\rho$. And, finally,

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$$\left\{\frac{\partial[O_3(\Delta H)]}{\partial H_1}\,\Delta H_1^*\right\}^2 + \left\{\frac{\partial[O_3(\Delta H)]}{\partial H_2}\,\Delta H_1^*\right\}^2 \simeq$$

$$\simeq 2(\Delta H^*)^2\left(\frac{1}{2\sigma^0_{O_3}\,(\Delta H)^2}\ln\frac{N_1\rho_2H_1^2}{N_2\rho_1H_2^2} - \frac{1}{2\sigma^0_{O_3}\,\Delta H}\,\frac{1}{H}\right)^2 . \qquad (7)$$

Formula (7) is written down on the assumption that $\Delta H_1^* \simeq \Delta H_2^* = \Delta H^*$ and $\frac{1}{H_1} \simeq \frac{1}{H_2} = \frac{1}{H}$, where ΔH_1^* and ΔH_2^* are the errors in determining the altitudes in the atmosphere. Thus, the resultant error in determining the ozone concentration from the data of single–frequency sounding is specified by the three terms given by Eqs. (5)–(7).

For the subsequent estimates we choose the parameters of a concrete lidar, e.g., close to the parameters of one of the modern lidar ozonometers. The energy of radiation at a wavelength of 308 nm is 200 mJ, $\eta = 0.2$, $K_0 = 1.6\cdot10^{-1}$, $S \simeq 1.96\cdot10^3$ cm^2 (D = 50 cm), and $\Delta H = 2\cdot10^5$ cm.

The calculation showed that the ozone measurement error is, in fact, determined by Eqs. (5) and (6). The resultant error, at best, for $\Delta\rho = 1$ % and $\Delta H^* = 10$ m attains several per cents or several tens of per cents increasing by several times for $\Delta\rho = 3$ % and $\Delta H^* = 100$ m. Shown in Fig. 1 are the calculated results.

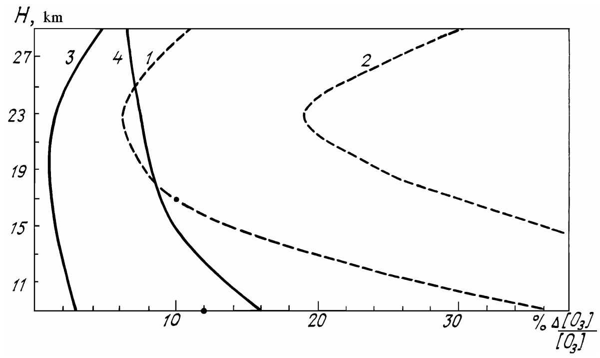

FIG 1. Error in determining the ozone concentration from the data of at single–frequency (dashed curves) and bifrequency (solid curves) sounding. The lidar parameters are indicated in the text. The number of sounding pulses is 10^4. 1 *and* 3) $\frac{\Delta\rho}{\rho} = 1$ % *and* $\Delta H^* = 10$ m *and* 2 *and* 4) $\frac{\Delta\rho}{\rho} = 3$ % *and* $\Delta H^* = 100$ m.

For comparison, the same figure shows the calculational errors in bifrequency sounding ($\lambda = 353$ nm, $E_0 = 50$ mJ, and the number of sounding pulses is 10^4).

It is quite obvious that for determining low–intensity long–term trends, the ozone measurement accuracy should be of the order of 1–2 %, which can be obtained only in the bifrequency sounding. In this case the increase in the lidar potential (increase in the radiant energy, area of the receiving antenna, and pulse repetition rate) does not lead to higher accuracy in determining the ozone concentration at altitudes of from 10 to 20 km. The errors due to the prescribed atmospheric density profile in bifrequency sounding are much smaller than those caused by the altitude measurements. If we assume that the minimum value $\Delta H^* = 10$ m then the maximum error in determining $\frac{\Delta[O_3]}{O_3}$ is ~ 1 % at H ~ 20–25 km.

Quite different is the case of single–frequency sounding. The error due to the use of the quantity ρ in the calculations of the distribution plays a decisive role here. Therefore the stratospheric ozone concentration can be derived, at best, with an accuracy of several or several tens of per cents from the data of single–frequency sounding. Such a method of sounding is applicable for solving only some specific problems associated, e.g., with very significant local variations of the ozone concentration. In this case, one can make use of the temperature profile derived from the data of radiosonde observations.

Using the well–known formulas $\rho = \frac{P}{kT}$ and $P = P_0\exp\left(-\frac{gM}{RT}H\right)$, where P_0 is the ground pressure and T is the temperature, and the remaining symbols are generally acceptable, we obtain

$$\rho_2 = \frac{P_0\exp\left(-\frac{gM}{RT_2}H_2\right)}{kT_2}\ ;\ \rho_1 = \frac{P_0\exp\left(-\frac{gM}{RT_1}H_1\right)}{kT_1}\ ;$$

$$\frac{\rho_2}{\rho_1} = \frac{T_1}{T_2}\exp\left[-\frac{gM}{R}\left(\frac{H_2}{T_2} - \frac{H_1}{T_1}\right)\right] . \qquad (8)$$

From Eqs. (8) and (2) we have

$$\frac{N_2H_2^2T_2}{N_1H_1^2T_1}\exp\left[\frac{gM}{R}\left(\frac{H_2}{T_2} - \frac{H_1}{T_1}\right)\right] = q^2(H_1, H_2) . \qquad (9)$$

It follows from this that at λ = 308 nm

$$q^2(H_1, H_2) \simeq q_m^2(H_1, H_2)\, q_{O_3}^2(H_1, H_2) =$$

$$= \exp\left\{-2\int_{H_1}^{H_2}[\sigma_m^0\,\rho(h) + \sigma^2_{O_3}[O_3(h)]\right\}dh\ ; \qquad (10)$$

$$[O_3(\Delta H)] \simeq \frac{[O_3(H_1)] + [O_3(H_2)]}{2} =$$

$$= \frac{1}{2\Delta Hs^0_{O_3}}\left[\ln\frac{N_1H_1^2T_1}{N_2H_2^2T_2} - \frac{gM}{R}\left(\frac{H_2}{T_2} - \frac{H_1}{T_1}\right) - \sigma_m^0\,\Delta H\times\right.$$

$$\left.\times\frac{P_0}{\kappa}\left[\frac{1}{T_2}\exp\left(-\frac{gM}{RT_2}H_2\right) + \frac{1}{T_1}\exp\left(-\frac{gM}{RT_1}H_1\right)\right]\right] \qquad (11)$$

For $R = 8.31\cdot10^3$ J/kmol·K, $M = 28.966$ kg/kmol, $g = 9.81$ m/s^2, $gM/R = 3.42\cdot10^{-2}$ K/m, $\sigma_M^0(308\text{ nm})/\kappa = 3.78\cdot10^{-7}$ s^{2}k/kg and $\sigma^0_{O_3}$ (308 nm) $= 1.17\cdot10^{-23}$ m^2, we obtain the calculational formula

$$[O_3(\Delta H)] = 4.27\cdot10^{22}\,\frac{1}{\Delta H}\left[\ln\frac{N_1H_1^2T_1}{N_2H_2^2T_2} - \right.$$

$$- 3.42\cdot10^{-2}\left(\frac{H_2}{T_2} - \frac{H_1}{T_1}\right) - 3.78\cdot10^{-7}\,P_0\Delta H\times$$

$$\left.\times\left[\frac{1}{T_2}\exp\left(-3.42\cdot10^{-2}\frac{H_2}{T_2}\right) + \frac{1}{T_1}\exp\left(-3.42\cdot10^{-2}\frac{H_1}{T_1}\right)\right]\right] \qquad (12)$$

where $\Delta H = H_2 - H_1$, $N(H_1) = N_1$, etc., all of the quantities are in M.K.S.

The last term in the right side of Eq. (12) is smaller than the second term. At an altitude of 10 km their values equal 0.01 and 0.31, respectively, at an altitude of 18 km they are $5{\cdot}10^{-3}$ and 0.316, and then (with increase of altitude) their difference grows.

In the calculations we can therefore employ the formula

$$[O_3(\Delta H)] \simeq 4.27{\cdot}10^{22}\,\frac{1}{\Delta H}\ln\frac{N_1H_1^2T_1}{N_2H_2^2T_2} - \frac{1.46{\cdot}10^{21}}{\Delta H}\left(\frac{H_2}{T_2} - \frac{H_1}{T_1}\right). \quad (13)$$

Let us estimate the error in the quantity $[O_3(\Delta H)]$ found from formula (13).

Omitting the intermediate calculations we obtain

$$\Delta[O_3(\Delta H)] \simeq \sqrt{2\left\{\Delta H^*\left[4.27{\cdot}10^{22}\left(\frac{1}{(\Delta H)^2}\ln\frac{N_1H_1^2}{N_2H_2^2} + \frac{2}{\Delta HH}\right) + \frac{1.46{\cdot}10^{21}}{T\Delta H}\right]\right\} + 1.83{\cdot}10^{45}\left(\frac{1}{N_1} + \frac{1}{N_2}\right)\frac{1}{(\Delta H)^2} + 2\left[\Delta T\left(4.27{\cdot}10^{22}\,\frac{1}{\Delta HT} + \frac{1.46{\cdot}10^{21}\,H}{\Delta HT^2}\right)\right]^2}. \quad (14)$$

In formula (14) $H = \dfrac{(H_1 + H_2)}{2}$ and $T = \dfrac{(T_1 + T_2)}{2}$. The first term under the root of Eq. (14) characterizes the error due to the altitude determination, the second term is determined by the error in recording of the signals, and the third term – by the error in the temperature measurement.

With the accuracy of the radiosonde temperature measurement $\Delta T = 2K$ and the above–described lidar parameters we obtain, e.g., at an altitude of 18 km $\Delta[O_3(\Delta H)] = 3.8{\cdot}10^{11}$ cm^{-3}, i.e., $\dfrac{\Delta[O_3(\Delta H)]}{[O_3(\Delta H)]} \simeq 9$ %. The principal error in this case is caused by the last term in the right side of Eq. (14), which equals $1.2{\cdot}10^{35}$ cm^{-6}. For comparison, the first term in the right side of Eq. (14) is $2.7{\cdot}10^{34}$ cm^{-6} and the second term is $1.7{\cdot}10^{32}$ cm^{-6}.

2. The distribution of the ozone concentration was obtained using a specially designed lidar ozonometer.

A receiving system of the lidar was a Newton telescope. A spherical mirror 70 cm in diameter (235 cm in focal length) was finely adjusted in a horizontal frame rigidly fixed with the lidar base.

An excimer laser (model 1701) emitted radiation at a wavelength of 308 nm. Because of an unstable resonator of the emitter, the radiation divergence did not exceed 0.5 mrad which allowed one to do without a collimating system. The laser pulse energy was about 70 mJ, the maximum pulse repetition rate was 50 Hz. The laser was fixed on the lidar base in a horizontal position, an output beam was deflected in the upward direction with the help of the adjustable mirror with high reflectance at an operating wavelength.

To record a backscattered lidar return, a single–channel photoblock was used. It incorporated a changeable diaphragm forming a telescope field of view (varying from 4 to 11 min), a cutoff unit in the form of a mechanical obturator which covered a high–power flux of radiation scattered in the near–diffraction zone, two holders with neutral filters used for attenuating the light flux by factors of 3.5, 10, and 30 and with interference filters used for selection of two or three wavelengths, a photomultiplier operating in the photon counting mode, a preamplifier and a discriminator–shaper which reduced the level of noise and formed single–electron pulses fed into a counter of the recording system.

A FEU–140 FOTON photomultiplier was also used which possessed high sensitivity in the UV ($\eta \approx 0.2$). Short duration of a single–electron pulse of FEU–140 (5–7 ns) and low level of dark current allowed the signal to be recorded over a wide dynamic range in the photon counting mode.

Electron blocks from the previously developed lidars MAKET were employed as an information–measuring system of this lidar. The operation was controlled by the ELEKTRONIKA–60 computer which allowed one to integrate the backscattered signal over $3{\cdot}10^4$ radiation pulses. A control block of the emitter and the near–zone diffraction–cutoff unit limited the maximum pulse repetition rate by 20 Hz.

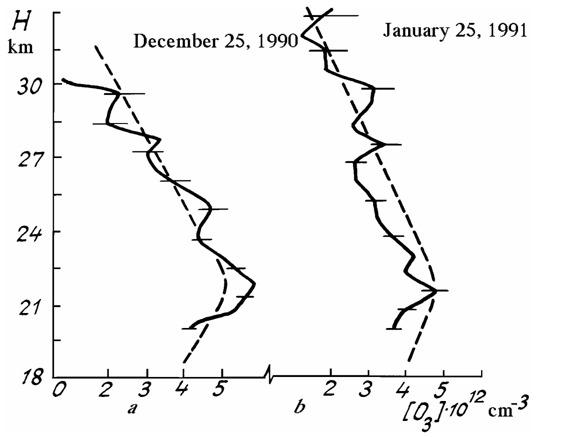

FIG. 2. Measurement results of ozone concentration in Obninsk.

The automated program of the lidar enabled one, in the interactive mode, to put the required data into the system: the distance of the near–diffraction zone (up to 30 km), the length of strobes (75–1500 m), the maximum measurement height (up to 100 km), pulse repetition rate, and the number of pulses. After this, the measurements proceeded automatically. The data were stored on magnetic disks in the form of files which contained the measurement data and comments.

The laser was fixed under the telescope and did not extend above the overall dimensions of the base frame with power supply and a pump for water cooling of the laser arranged inside. Such an arrangment of the lidar occupied a minimum area with the length of communications being substantially shortened.

The measurements of vertical profiles for ozone concentration were started in Obninsk in October, 1990 in a single–frequency mode.

Shown in Fig. 2 are the measurements of ozone concentration profiles in comparison with the model distribution of ozone taken from Ref. 1.

The further experiments are being performed in a bifrequency mode.

REFERENCES

1. I.I. Ippolitov et al., in: *Spectroscopic Methods for Sounding of the Atmosphere* (Nauka, Novosibirsk, 1985), pp. 4–44.
2. V.M. Zakharov, O.K. Kostko, and S.S. Khmelevtsov, *Lidars and Climate Research* (Gidrometeoizdat, Leningrad, 1990), 320 pp.
3. H. Claude and K. Wege, in: *Abstracts of Reports at the 14th International Laser Radar Conference Holy* (1988), pp. 392–395.