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# DETERMINATION OF ATMOSPHERIC TRACE GASES BY INFRARED DIFFERENTIAL ABSORPTION LIDAR *

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A differential absorption lidar system for detection of atmospheric pollutants based on the middle infrared tandem optical parametric oscillator is presented. The characteristics of the signal obtained by detecting radiation backscattered from different topographical objects were measured in the field test of the system. These characteristics were used to calculate the minimum detectable concentration of several atmospheric pollutants: ozone, methane, ammonia, etc. Most of the gases can be detected below typical background concentrations for the low-polluted atmosphere.

**Keywords:** differential absorption, lidar, atmospheric pollutants, laser spectroscopy

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## 1. Introduction

Increasing rates of technological progress and urbanization raise concerns about the ecological state of environment. Alongside with industrial and traffic pollution the threat of terrorist attacks should also be considered. These factors cause increased interest in the devices for control of atmospheric composition. Laser-based remote sensing methods are the most promising solutions for control of industrial and urban environments. An especially sensitive analysis method under troposphere conditions is a differential absorption lidar (DIAL) operating in the middle infrared spectral region. Though there were several attempts to apply this method to the detection of atmospheric pollutants, most of these systems were cumbersome [1–3].

$CO_2$ lasers have long been the primary choice for the laser source for middle IR DIAL systems. Their main drawback was a limited spectral region and discrete tuning. As an alternative to $CO_2$ lasers optical parametric oscillators (OPO) based on such crystals as ZnGaP, CdSe, and AgGaSe can be used. They promise continuous tuning in the middle IR region. However, there are also challenges to overcome, namely they can not be pumped by 1 $\mu$m radiation and the spectrum of generated radiation is much broader than that of the $CO_2$ laser. Both of these issues are addressed in this paper. In this work we describe an original compact DIAL system and universal solution for the early pollution warning system.

## 2. Experimental set-up

The mid-IR laser source was based on the tandem optical parametric oscillator. The pump source was a commercial Nd : YAG actively Q-switched nanosecond laser (NL303G, EKSPLA Ltd.) that produced 1.064 $\mu$m 3–6 ns pulses with the energy up to 500 mJ. The OPO consisted of two stages: the first one based on the KTP crystal and the second one on the AgGaSe crystal. A detailed description of the set-up was presented previously [4]. In brief, it produced up to 1 mJ of radiation tunable in the 5–12 $\mu$m region with a spectral bandwidth of less than 10 cm$^{-1}$ (typical values 7–8 cm$^{-1}$).

As the complete description of the DIAL set-up can be found in the previous paper [5], only a concise description will be provided. The optical set-up of the lidar is presented in Fig. 1. It is based on the coaxial transmitter-receiver system built around the main 250 mm diameter gold-coated parabolic mirror ($f$ = 1250 mm, surface quality $\lambda/8$ at visible wavelengths). In addition, a ZnSe lens ($f$ = 70 mm) and two gold-coated mirrors are used for transmitting

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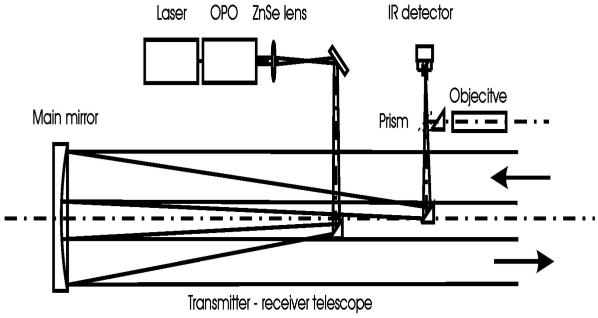


Fig. 1. Optical schema of the differential absorption lidar.

outgoing radiation. Incoming radiation is focused by the main mirror to the detector. One quarter of the main mirror area is used for transmitting outgoing radiation and other 3/4 for receiving incoming radiation. A thermoelectrically cooled mercury cadmium telluride (MCT) detector (PCI-2TE-12, Vigo System S.A., $D^* = 7.5 \cdot 10^8$ cm Hz$^{1/2}$ W$^{-1}$, time constant 5 ns) was used in this DIAL set-up. High sensitivity of this detector is useful for detection of the radiation backscattered from topographical objects. The short response time can help determine the distance to the object with high precision, thus minimizing the error in the optical path length determination (see Eq. (2)). For the pulse energy reference we used a small part of the light leaving the lidar transmitter. A delayed returning signal was registered using the same detector. Reliable detection of trace gases in the atmosphere requires a sufficient signal amplitude, which can be provided only by artificial back scatterers such as retroreflectors. The retroreflector consisting of three gold-coated mirrors was used in our field experiments. With the retroreflector in use and placed 2250 metres from the DIAL system, we measured the standard deviation of the signal to be 20%, without the use of the reference pulse. This value is used in minimal detectable concentration calculations.

## 3. Numerical calculations

The minimal concentration of the gas molecules that can be detected by the DIAL system can be evaluated from the characteristics of the DIAL signal and the absorption coefficient of gas. The minimal detectable change $y_{\min}$ can be expressed as follows:

$$y_{\min} = \frac{E_{\text{off}} - E_{\text{on}}}{E_{\text{off}}} , \qquad (1)$$

where $E_{\text{off}}$ is the signal measured at wavelength $\lambda_{\text{off}}$, which is just outside the absorption line of the gas under investigation, and $E_{\text{on}}$ is the signal at wavelength $\lambda_{\text{on}}$ – at the centre of the absorption line. The value of $y_{\min}$ is limited by the dominating source of noise. In our case it was caused by laser pulse instability and resulted in the 20% standard error. Assuming averaging over 100 shots, the signal to noise ratio being equal to 3, we estimated that the minimum detectable change in the signal due to absorption could be 7.8%. This result can be used to estimate the minimum detectable concentration of the gas in ppm [1]:

$$\langle C \rangle_{\min} = \frac{1}{2\alpha_{\text{on}} R} \ln \left( \frac{1}{1 - y_{\min}} \right) , \qquad (2)$$

where $\alpha_{\text{on}}$ is the absorption coefficient normalized to the concentration of 1 ppm, and $R$ is the length of optical path of the beam. It is assumed that the absorption coefficient and the optical path are known exactly. The characteristic absorption coefficient of ozone is presented in Fig. 2.

Exact information about concentration of molecules can be obtained from DIAL experiments only if the absorption coefficient at corresponding wavelengths is known. However, the laser source of the DIAL system reported in this paper has a broad spectrum. Its width was comparable to the widths of absorption lines of the species under investigation as shown in Fig. 2, where the ozone absorption linewidth is compared with the simulated laser spectral profile assuming Gaussian form and 10 cm$^{-1}$ linewidth. Additional procedures should be performed to take into account the finite spectral width of the source. One of the possibilities is using the effective absorption coefficient $\alpha_{\text{eff}}$. The method described below is very similar to the method of effective cross-section described in [6] and is adapted to the notion of the absorption coefficient instead of cross-section.

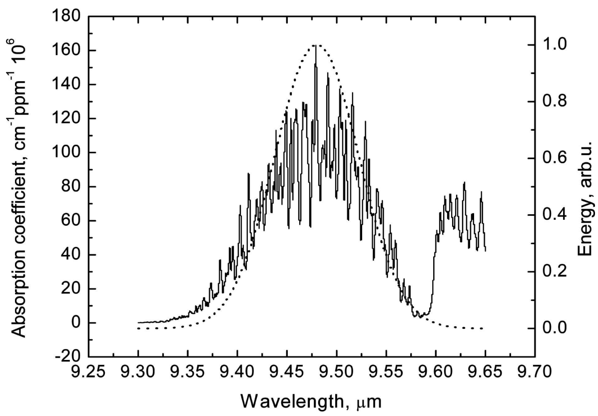


Fig. 2. Ozone absorption line profile and simulated laser line profile (Gaussian, spectral width 10 cm$^{-1}$) at 9.48 $\mu$m.

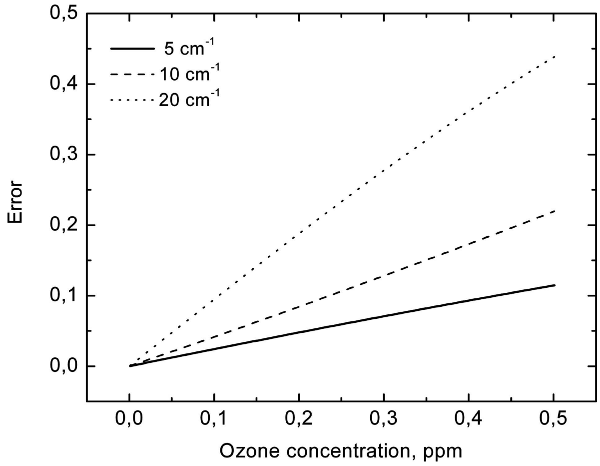


Fig. 3. Error in the 0.5 ppm ozone concentration detection caused by different spectral linewidth of the laser line: 5, 10, and 20 $\mathrm{cm}^{-1}$ (optical path 4500 m).

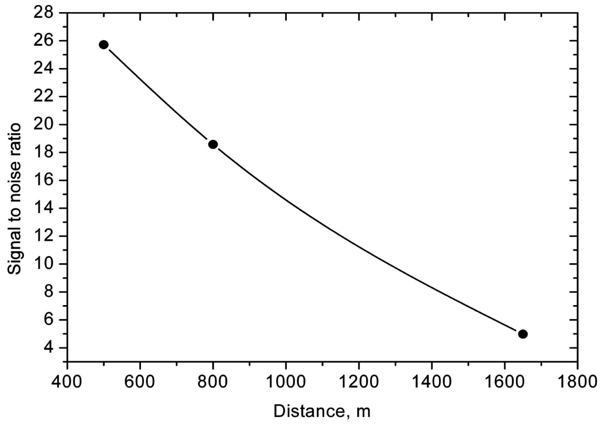


Fig. 4. Dependence of the backscattered signal/noise ratio on the distance from the object (road sign).

The effective absorption coefficient of gas molecules is calculated as follows:

$$\alpha_{\mathrm{eff}} = \frac{\int L(\nu, r)\alpha(\nu, r)\,\mathrm{d}\nu}{\int L(\nu, r)\,\mathrm{d}\nu}, \quad (3)$$

where $L(\nu, r)$ is the spectral profile of the laser source and $\alpha(\nu, r)$ is the profile of the absorption line. When the laser light propagates through absorbing media, its spectral profile changes. The value of $\alpha_{\mathrm{eff}}$ also changes, as one can see from Eq. (3). At some distance $R$, the value of $\alpha_{\mathrm{eff}}$ will be

$$\alpha_{\mathrm{eff}}(R) = \frac{\int L(\nu, r)\alpha(\nu, r)\mathrm{e}^{-\alpha R}\mathrm{d}\nu}{\int L(\nu, r)\mathrm{e}^{-\alpha R}\,\mathrm{d}\nu}\,. \quad (4)$$

The change can be negligible if absorption is weak. We estimated the error produced by assuming a constant value $\alpha_{\mathrm{eff}}$ through all the path of the light propagation:

Table 1. Signal detected by the DIAL detector from various topographical objects.

| Object | Signal, V | Distance to the object, m | Signal/noise |
|---|---|---|---|
| Brick wall | 0.11 | 146 | 13.5 |
| Cardboard sheet | 0.09 | 146 | 11.3 |
| Tree leaves | 0.11 | 105 | 14.3 |
| Concrete post | 0.44 | 34.5 | 55 |
| Tree | 0.92 | 26.3 | 115 |

$$\frac{\Delta\alpha_{\mathrm{eff}}}{\alpha_{\mathrm{eff}}} = 1 - \alpha_{\mathrm{eff}}(R)\,\frac{\int L(\nu, r)\,\mathrm{d}\nu}{\int L(\nu, r)\alpha(\nu, r)\,\mathrm{d}\nu}\,. \quad (5)$$

The relative error as a function of the concentration in the case of ozone absorption at 9.48 $\mu$m and the path of 4500 m is presented in Fig. 3.

## 4. Results

For practical applications of the DIAL system, it is very important to know the backscattering coefficients of various topographical objects. We performed a series of measurements where the signal backscattered from several objects as well as the signal / noise ratio were detected. The results are presented in Table 1. The values at distance $R$ used in these experiments were small because one of the possible applications of this system could be monitoring of some industrial area where the open view path does not exceed several hundred metres. Much longer distances were used in the measurement of backscattering by road signs to simulate possible monitoring of the traffic pollution. The results of measurements are presented in Fig. 4. One can see that distances up to 1.5 km can be successfully monitored by the presented system.

The most important characteristic of the pollution detection system is its detection threshold for various gases. We performed estimation of minimal detectable concentrations for several gases. The absorption coefficient data were taken from the spectral database [7], which is based on the HITRAN [8] and GEISA [9] databases. The finite spectral width of the laser source was taken into account by calculating the effective absorption coefficient according to Eq. (3). We used the characteristics of the signal that were recorded in the field tests and described before. A typical optical path in these field tests was 4500 m. The possible interference from other species was ignored. The results of the calculations are presented in Table 2. Typical

Table 2. Calculated minimal detection limit of several gases.

| Gas | $\lambda_{on}$, $\mu$m | $\lambda_{off}$, $\mu$m | $\alpha_{on}R$, ppm$^{-1}$ | $C_{min}$, ppb | $C_{back}$, ppb [10] |
|---|---|---|---|---|---|
| $O_3$ | 9.48 | 9.22 | 2.0 | 21 | 55 |
| $SF_6$ | 10.6 | 10.4 | 30 | 1.4 | 0.042 |
| $NH_3$ | 10.3 | 10.2 | 2.1 | 20 | 6–20 |
| $N_2O$ | 7.84 | 8.2 | 4.33 | 43 | 250 |
| $CH_4$ | 7.66 | 7.1 | 2.15 | 93 | 1500 |

background concentrations $C_{back}$ for the low-polluted atmosphere are presented in Table 2. One can see that most of the gases can be detected at their background concentrations and only ammonia and sulfur hexafluoride can be detected when they are exhausted.

The gases were chosen only to demonstrate the feasibility of the method. This method can be easily applied to other more complex pollutants, for example chemical warfare agents.

## 5. Conclusion

The calculations based on characteristics of the presented middle infrared DIAL system show that most of the pollutants can be detected at ppm and ppb level. As a result, this DIAL system can serve as a part of the early warning system that can warn of elevated pollution levels until they reach concentrations harmful for human health.

## Acknowledgement

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# DUJŲ PĖDSAKŲ NUSTATYMAS ATMOSFEROJE INFRARAUDONOSIOS SKIRTUMINĖS SUGERTIES LIDARU

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**Santrauka**

Pateikti skirtuminės sugerties lidarinės sistemos, skirtos atmosferos teršalams aptikti, eksperimentiniai rezultatai. Sistemos infraraudonosios spinduliuotės šaltinis buvo sudarytas iš dviejų pakopų parametrinio šviesos generatoriaus. Lauko bandymų metu buvo nustatytos lidarinės sistemos registruojamų signalų charakteristikos, pagal jas buvo įvertintos kai kurių atmosferos komponenčių (ozono, metano, amoniako ir kt.) minimalios aptinkamos koncentracijos. Skaičiavimai rodo, kad daugelis šių medžiagų gali būti aptinkamos dar koncentracijoms nepasiekus žalingo žmonių sveikatai lygio arba net žemesnės nei tipinės foninės koncentracijos mažai užterštoje atmosferoje. Buvo įvertinta lazerinio šaltinio spektro pločio įtaka kiekybinės informacijos patikimumui.