

Practical solutions for calibration of DIAL system

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We propose and describe two simple methods which can be applied for calibration of DIAL systems. One of them is related to SO₂ measurements. It is based on the on-line investigation of the fluorescence signal coming from the cell containing SO₂ gas and allows us to control the laser wavelength. The other solution which we call the autocalibration of the DIAL system is due to the simultaneously performed DOAS measurements.

1. Introduction

Monitoring of air pollution can be performed with various techniques and methods [1]. In order to get reliable and reproducible results, accurate calibration of measurement systems is required [2]–[6]. If we want to compare two different systems we should perform measurements at the same time and cover the same area. It is of particular importance in the case of measurements related to such a dynamic medium as the atmosphere. It is not easy to satisfy the requirements mentioned above for systems which differ from each other in their principles of operation as much as lidars, DOAS (Differential Optical Absorption Spectroscopy) instruments and chemical analysers.

The DIAL (Different Absorption Lidar) system [1], [7] is able to detect selectively specific pollutants. The basic idea of its operation consists of simultaneous use of two laser beams which are sent to the atmosphere and give rise to the return signals due to backscattering processes occurring along their path. The first wavelength λ_{on} is tuned to an absorption line of the desired pollutant. The second wavelength λ_{off} is much less absorbed and it is used as a reference. The difference between both echo signals reflects the concentration of the pollutant and may provide its range resolved information.

In DIAL measurements one of the main problems is how to control precisely the wavelength of the laser light. Optogalvanic techniques [8] frequently used for calibration of lidar systems are in many cases not sufficiently precise. For gas species which have their spectrum consisting of narrow absorption lines even small changes of the wavelengths may cause substantial error in the measured value of differential absorption. The direct consequence is incorrect determination of concentration values.

The DOAS technique [9] is based on measurements of light absorption along a chosen path, for various wavelengths. As a result one can get information on

concentration of various pollutants, but integrated over the optical path of the light beam. DIAL and DOAS are both purely optical methods.

Chemical methods are based on chemical reactions and/or chemiluminescence. Air samples can be analysed either *in situ* or transported to the lab. In both cases the information is limited only to the point (local) concentration. Typical volume of the sample for *in situ* measurements is about few litres per minute. In a similar lidar measurement the analysed air volume corresponds to a cylinder of the length of a few kilometres and of the diameter of a few centimetres. A single measurement with lidar at a distance of 1 km takes only about 6 μs , *i.e.*, the time resulting from the value of the light velocity. Since measurements are typically averaged over 1000 laser pulses we get 6 ms as their effective time. For laser repetition rate of 10 Hz the real time of measurements is 100 s, which gives the value of the so-called "duty time" as low as 6 ms/100 s = 0.00006. For chemical methods and for DOAS technique the value is practically equal to 1.

It is worthwhile to note that each of the methods mentioned above determines slightly different parameters. The chemical methods give the point values of a pollutant concentration, the DIAL measurements can give information on its spatial distribution, while the DOAS technique provides the concentration value averaged over a certain distance. This is one of the reasons for which the comparison of results obtained with the use of different methods requires careful analysis. More information about the features of these techniques can be found elsewhere [2].

The calibration of measurement systems is still one of very important problems in lidar techniques. Any progress in this direction can significantly improve their reliability and precision. In this paper, we propose two practical solutions which can be applied to DIAL system. One of them is related to the control of laser wavelength and it is based on the on-line investigation of the fluorescence signal coming from the cell containing SO_2 gas. This gives the calibration precision better than 0.1 nm. The other solution is the autocalibration of the DIAL system based on the simultaneously performed DOAS measurements. The latter can be applied if the laser light is scattered on a macroscopic object situated in the background.

2. Wavelength calibration for SO_2 measurements

— theoretical aspects

The SO_2 spectrum in the near UV has very well pronounced and narrow maxima [10]. As a consequence, very precise tuning for both λ_{on} and λ_{off} wavelengths is required. For this reason the calibration can be based on the absorption of laser light by SO_2 and the observation of fluorescence light. In our case, the laser beam is sent into the absorption cell of length l which contains SO_2 gas. The fluorescence signal is measured at a distance x from the entrance window. According to Lambert–Beer formula, the transmitted light intensity at a distance x from the entrance window is equal to

$$I(x) = I_0 e^{-\sigma N x}, \quad (1)$$

where $\sigma = \sigma(\lambda)$ is the absorption cross-section and N is the concentration of SO_2 molecules.

Total fluorescence intensity at point x can be written as

$$F(\lambda, x) = K(\lambda) N^*(x, \lambda) \quad (2)$$

where $K(\lambda)$ is the coefficient of the fluorescence efficiency and $N^*(x, \lambda)$ is the concentration of SO_2 molecules at distance x , excited by light at wavelength λ . The latter can be expressed as

$$N^*(\lambda, x) = P(\lambda) I(x) = P(\lambda) I_0 e^{-\sigma N x}, \quad (3)$$

where $P(\lambda)$ is a function describing excitation efficiency.

Combining formulae (1), (2) and (3) one obtains

$$F(\lambda, x) = K(\lambda) P(\lambda) I_0 e^{-\sigma N x}. \quad (4)$$

We define a function Φ which is the ratio of total fluorescence intensity $F(x, \lambda)$ excited at λ to the intensity of transmitted light $I(l)$

$$\Phi(\lambda, x) = \frac{F(\lambda, x)}{I(l)}, \quad (5)$$

which, using Eqs. (1) and (4), gives

$$\Phi(\lambda, x) = K(\lambda) P(\lambda) e^{\sigma N(l-x)}. \quad (6)$$

In the case of DIAL measurements we use two wavelengths: λ_{on} (absorbed by SO_2 gas) and λ_{off} (reference wavelength). So, the functions Φ can be written for both wavelengths and their ratio can be expressed in the following form:

$$\frac{\Phi_{\text{on}}(\lambda, x)}{\Phi_{\text{off}}(\lambda, x)} = \frac{K(\lambda_{\text{on}}) P(\lambda_{\text{on}})}{K(\lambda_{\text{off}}) P(\lambda_{\text{off}})} e^{(\sigma_{\text{on}} - \sigma_{\text{off}}) N(l-x)}. \quad (7)$$

3. Experimental set-up and results

The scheme of experimental set-up is presented in Figure 1. The light from the lidar Ti:Sa laser system, passing through the nonlinear crystals is transformed into the UV region. The 3rd harmonics pulses having energy of about 0.5 mJ are directed towards the quartz cell of the length l (e.g., 20 cm) filled with SO_2 gas. The fluorescence signal F from the SO_2 cell is collected by a fiber placed at a distance x (e.g., 1 cm) from the entrance window, and measured by a photomultiplier. The transmitted intensity of the 3rd harmonic $I(l)$ is measured by UV detector placed behind the cell. Both signals are displayed on digital oscilloscope.

For DIAL measurements of SO_2 concentrations we chose $\lambda_{\text{on}} = 286.9$ nm and $\lambda_{\text{off}} = 286.3$ nm, for which $\sigma_{\text{on}} = 1.09 e^{-18} [\text{cm}^2]$ and $\sigma_{\text{off}} = 6.45 e^{-19} [\text{cm}^2]$. In order to determine the best parameters of the calibration cell, the SO_2 fluorescence was investigated for three different pressures: 0.05 Tr, 0.5 Tr and 1.5 Tr. The lowest

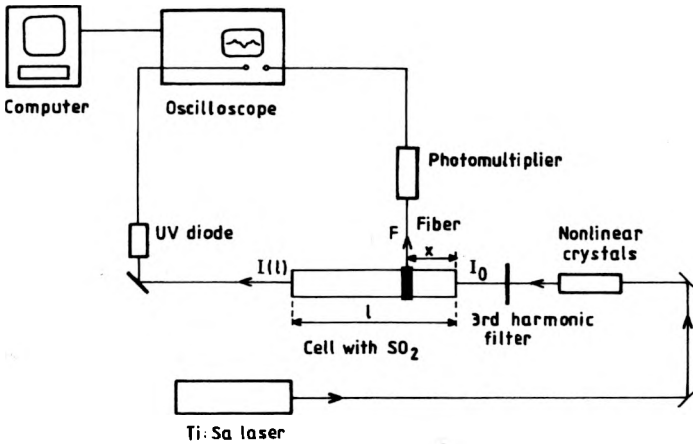


Fig. 1. Scheme of experimental set-up for fluorescence investigations.

pressure was chosen for calibration purposes because of the longest fluorescence decay time. It was equal to $2.2 \mu\text{s}$, while for the other two pressures the measured values of decay times were much shorter because of the collisional fluorescence quenching. They were equal to about $0.9 \mu\text{s}$ and $0.18 \mu\text{s}$, respectively. The difference between transmitted signals at λ_{on} and λ_{off} by SO_2 gas under pressure 0.05 Tr in the cell of 20 cm in length is only about 0.015 . For this reason the calibration based on absorption measurements only would not be possible. The ratio of functions Φ under identical conditions is about 2 and is greater than the ratio of the respective cross-sections. It is related to the K and P wavelength dependence.

The function Φ plotted against wavelength in the spectral range $283\text{--}290 \text{ nm}$ is shown in Fig. 2b. As one can see it strongly resembles the absorption spectrum of SO_2 shown in Fig. 2a. Only the positions of maxima are shifted by 0.2 nm with respect to those shown on SO_2 absorption spectrum. It shows that the calibration based on the opto-galvanic effect is not sufficient for DIAL measurements. It is also clear that such a small displacement can be easily detected by observation of the function Φ .

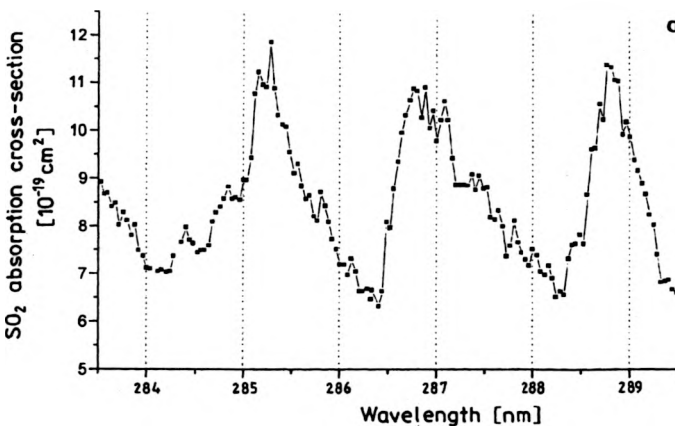


Fig. 2a

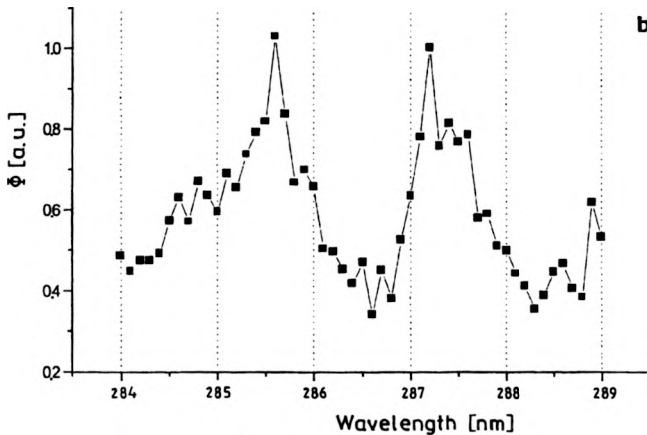


Fig. 2b

Fig. 2. SO₂ absorption cross-section (a), function Φ (b). Displacement of function Φ against absorption spectrum is well pronounced (about 0.2 nm).

The method can be easily applied to any DIAL system by adding an SO₂ cell and using the fluorescence signal and subsequently the function Φ as an indicator for wavelength setting. In the case of SO₂ DIAL measurements, investigation of function Φ can be limited only to finding its local maximum or minimum. It defines the proper wavelengths for atmospheric DIAL measurements. Such a method will be cheaper than any commercial wavemeter for UV and much more reliable in field conditions. It is also important that the function Φ does not depend on fluctuations of laser energy. It should be noticed that this method works only with another calibration system, which allows a rough tuning of the laser to the investigated spectral region with an accuracy of about 1 nm.

4. Autocalibration of DIAL system with DOAS technique

In order to use the DOAS signal for DIAL system calibration the laser pulses sent to the atmosphere have to be scattered on a macroscopic object. In this way we simulate one-ended DOAS system working at two different wavelengths. Normally, the white light is used for DOAS. Two wavelength system is a modification of traditional DOAS though its principle of operation is the same. Comparing intensities of the DOAS signals on both wavelengths, λ_{on} (strong absorption) and λ_{off} (weak absorption), one can determine the concentration of the chosen gas averaged over the distance between the lidar and the scattering object.

In the experiment we performed during the ozone campaign at the Kamięńczyk Fall (near Szklarska Poręba, Poland) the quarry wall (about 1.5 km distant from lidar system) turned out to be a very convenient object to generate the DOAS signal and to use it for autocalibration of the DIAL system.

The shapes of registered signals are shown in Fig. 3. Each of them is averaged over 1000 laser pulses. The lidar is situated in the origin of the coordinate sys-

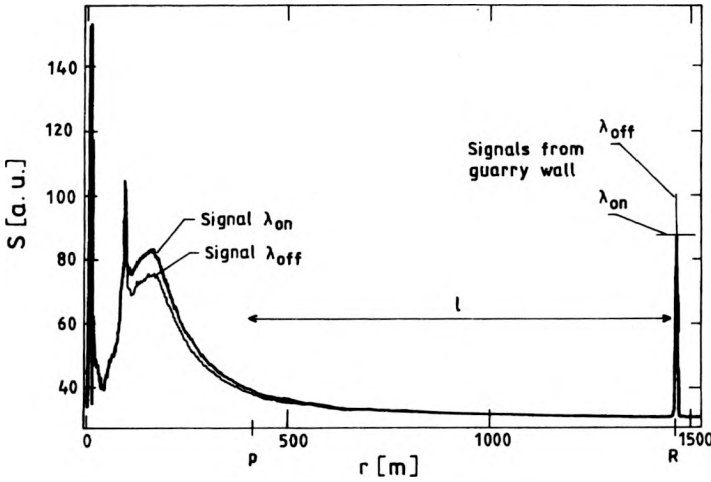


Fig. 3. Ozone lidar signals registered for λ_{on} and λ_{off} at the Kamięńczyk Fall.

tem (point 0). The distance L corresponds to the DIAL measurement range. Two strong pulses observed at the end of this range are signals related to the scattering of laser light on quarry wall and are used for DOAS measurements. The difference in their intensity is due to stronger O_3 absorption of the λ_{on} wavelength.

5. Theoretical aspects of autocalibration method

Signal S_R arising due to the scattering on the quarry wall situated at a distance R from the lidar system may be written as

$$S_R = DBI(t) \frac{A}{R^2} \exp \left[-2\sigma \int_0^R c(r) dr \right] \tag{8}$$

where: A is the telescope area, D – coefficient describing the lidar geometry and the sensitivity of its detection system, B – backscattering coefficient for the quarry wall, $I(t)$ – intensity distribution of the laser pulse emitted by the system (at point $r = 0$), σ – cross-section for absorption, $c(r)$ – concentration of the gaseous pollutant (O_3 in our case). Only two of the above parameters (I and σ) are different for λ_{on} and λ_{off} laser beams. We assume that all the others are identical for both wavelengths and the difference between the two DOAS signals is due to the absorption only. We also assume that the scattering process does not change the shape of the laser pulse.

Integrating Equation (8) over the time we get

$$\int_{-\infty}^{\infty} S_R dt = \hat{S} = DBE \frac{A}{R^2} \exp \left[-2\sigma \int_0^R c(r) dr \right] \tag{9}$$

where $E = \int_{-\infty}^{\infty} I(t) dt$ is the laser pulse energy.

The ratio of signals \hat{S} corresponding to λ_{off} and λ_{on} is equal to

$$\frac{\hat{S}_{\text{off}}}{\hat{S}_{\text{on}}} = \frac{E_{\text{off}}}{E_{\text{on}}} \exp \left[2\Delta\sigma \int_0^R c(r) dr \right] \tag{10}$$

where $\Delta\sigma = \sigma_{\text{on}} - \sigma_{\text{off}}$.

Taking the logarithm of both sides we have

$$\ln \frac{\hat{S}_{\text{off}}}{\hat{S}_{\text{on}}} = \ln \frac{E_{\text{off}}}{E_{\text{on}}} + 2\Delta\sigma \int_0^R c(r) dr. \tag{11}$$

Then replacing $\int_0^R c(r) dr$ by $\bar{c} \cdot R$, where \bar{c} is an average concentration on the OR length, we get the following formula for \bar{c}

$$\bar{c} = \frac{\ln \frac{\hat{S}_{\text{off}}}{\hat{S}_{\text{on}}} - \ln \frac{E_{\text{off}}}{E_{\text{on}}}}{2\Delta\sigma R}. \tag{12}$$

As one can see, by measuring both the energy E of laser pulses emitted at lidar output and the integrated signals \hat{S} coming from the scattering on the quarry wall we can determine the average ozone concentration between the lidar system and the quarry wall. The scattered signal is strong enough to allow measurements with precision of about 1%.

Equation (12) shows how to simulate DOAS operation by means of a DIAL system. There are, however, several limitations. As one can see in Fig. 3, the signal corresponding to short measurement range (0–400 m) cannot be described by lidar equation because of fluctuations [11] which are mainly due to electric disturbances, non-linear response of photodetectors and optical limitations of the system. In order to avoid this problem we can choose for DIAL analysis only the path of the length L starting at point p (Fig. 3). To obtain the average concentration \bar{c} of the absorber we have to modify slightly Eq. (12). The distance R has to be replaced by $(R-p)$, and the ratio of output energies by the ratio of lidar signals $S_{\text{off}}/S_{\text{on}}$ corresponding to the point p . Such substitutions lead to the formula

$$\bar{c} = \frac{\ln \frac{\hat{S}_{\text{off}}}{\hat{S}_{\text{on}}} - \ln \frac{S_{\text{off}}(p)}{S_{\text{on}}(p)}}{2\Delta\sigma(R-p)}. \tag{13}$$

In this way we do not have to measure the energies of laser pulses at the output of the system. The above formula allows us to determine the average concentration of chosen pollutant using data obtained from lidar measurements. The point p should be located not too far away from the lidar system in order to get considerably good

signal/noise ratio and to determine the \bar{c} value as averaged over the long path. In our experiment we chose the point p at a distance of 400 m from the lidar.

The same concentration we can find also from the DIAL analysis. In order to get it we have to average the spatial distribution of the pollutant concentration. The value obtained in this way is compared with that obtained from DOAS technique. It is very important that the proposed autocalibration method allows us to compare data from the same measurements.

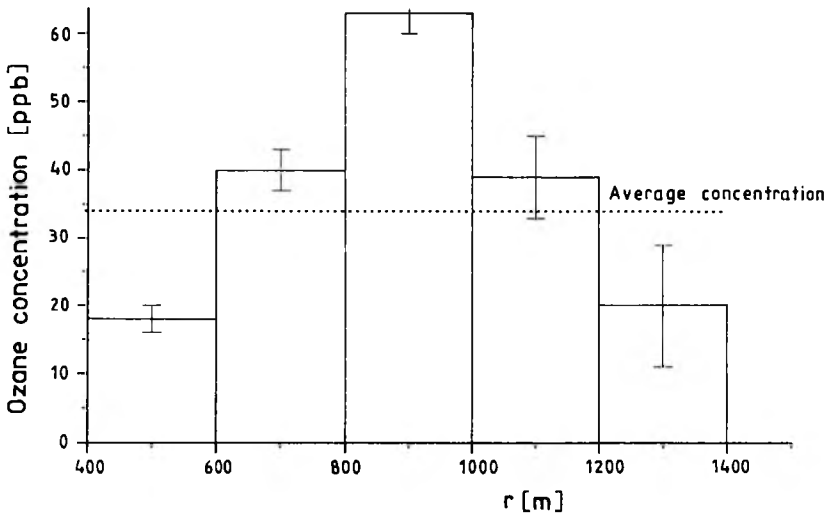


Fig. 4. Distribution of ozone concentration along the path between the lidar and the quarry wall obtained from 1000 pairs of laser shots. Dashed line denotes the average concentration while vertical lines denote uncertainties.

Figure 4 presents DIAL results of the ozone concentration distribution along the path between the lidar and the quarry wall. The data obtained from 1000 pairs of laser shots were smoothed according to the procedure described in [11]. Concentration values derived by the slope method [11] over 200 m sections as well as the mean (weighted) value over the whole distance pR are shown. The latter is equal to 34 ± 2 ppb while the DOAS analysis gives as the average concentration 32 ± 3 ppb. Such a good agreement is an indication of the correct lidar operation.

6. Conclusions

Using the function Φ for wavelength calibration of a DIAL system we can get precision better than 0.1 nm. The calibration method is particularly convenient for SO_2 measurements because of the character of its absorption spectrum. This simple and cheap method can be particularly useful during field campaigns.

The main advantage of the method of autocalibration which combines DIAL and DOAS techniques is that it satisfies a very important condition of the place and time conformity. The analysis of the experimental signals obtained during the ozone

campaign at the Kamieńczyk Fall has shown that the discrepancy between both techniques can be smaller than 10%.

Acknowledgements – The authors would like to thank D. Weidauer from ELIGHT Laser Systems GmbH for lending quartz spectroscopic cells, K. Kołacz and G. Flisiak from Physics Department of Warsaw University for their help with the measurements and the data processing.

This work was supported by the Polish Committee for Scientific Research (KBN) under the project No. 6 P04G 027 17.

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Received October 10, 1999