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A. SZCZUREK*, A. SZYMAŃSKI**,
A. CZYŻEWSKI**, K. STELMASZCZYK**

APPLICATION OF OPTICAL REMOTE SENSING TECHNIQUES TO AIR QUALITY MONITORING

An analytical methods and techniques are essential for environment monitoring. The measurements do not improve directly environmental conditions, but they provide information about pollutants. This paper is a comprehensive and critical review of remote techniques applied to monitoring of air pollutants. In particular, DOAS and LIDAR methods are described and discussed.

1. INTRODUCTION

Air quality is mainly monitored by point measurements [1]–[4], which means that the concentrations of air pollutants are measured at a selected point in space. Though this method of monitoring offers some unique advantages, it still suffers from drawbacks. The point measurements of air pollutants are inadequate for identification and quantification of poorly mixed gases over large areas. Such conditions are characteristic of:

- plumes from smoke stacks and hazardous waste landfills,
- fugitive emissions from industrial plants or leakage emissions from chemical storage tanks,
- exhaust emissions from road traffic (especially during rush hours).

This method cannot be used for the measurements of trace species in remote points of atmosphere (troposphere and stratosphere) either. Optical remote sensing techniques are particularly suitable for the analysis of gaseous pollutants over the areas that are not easily accessible [5].

Generally, in the remote sensing techniques, the analyzing instruments physically removed from the air region under study are used. Therefore, the measuring procedure involving these techniques does not require a sample to be taken into the analyzer.

* Wrocław University of Technology, Institute of Environment Protection Engineering, Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland.

** Department of Physics, Optics Division, Warsaw University, Hoża 69, 00-681 Warsaw, Poland.

2. ADVANTAGES AND DISADVANTAGES OF OPTICAL REMOTE METHODS

There are a number of advantages of optical remote methods [6]. They may be itemized as follows:

remote sensing is particularly advantageous to monitor air pollutants in distinct, inaccessible, large areas,

it is possible to monitor a number of pollutants simultaneously,

remote sensing provides continuous, rapid, in near-real time data,

the optical remote methods are based on the direct, nonintrusive, contactless measurements.

Despite important advantages there are also some drawbacks associated with optical remote sensing of air pollutants:

- Major limitations are spectral interferences caused by atmospheric constituents other than the target gas. Water vapour, CO_2, O_3, hydrocarbons and many other compounds absorb electromagnetic radiation. The absorption bands of these species may preclude both qualitative and quantitative analyses of gases of interest.
- Optical measurements have to be restricted to wavelength ranges of atmospheric windows.
- Results of measurements depend also on atmospheric conditions. Fog and rain can preclude data collection. The data are also affected by extremes and fluctuations of temperature.
- An important drawback is the limitation of path length that can be monitored.
- The lowest detection limit of remote instruments may not be adequate for the desired application because the concentrations of trace species present in the atmosphere are so low that they range from 0.1 ppt to several ppb.
- Another serious problem is the calibration of the remote systems. Traditional calibration methods cannot be applied to remote optical systems because gaseous species of known concentrations cannot be put into a fixed path length gas cell [7].
- The remote, optical systems have to be supervised by highly qualified staff.
- They are very expensive.
- These instruments do not exist in portable forms. They are too heavy to be transported by hand. The optical remote systems are usually used in the stationary or transportable form.

3. PHYSICAL PRINCIPLES OF OPTICAL REMOTE METHODS

Identification and quantification of air pollutants by means of optical remote methods are based on interactions between radiation and matter. A number of complex, physical processes can occur as a result of an electromagnetic beam transmission across a volume of measured air. Scattering, fluorescence and absorption are considered to be such processes. Information about air pollutants can be obtained based on all these processes. However, detection of molecular species in the atmosphere is possible only as a result of Raman scattering, resonance fluorescence and absorption phenomena which are atomic or molecular specific. Additionally, the absorption cross-sections are larger than cross-sections for other optical interactions and are not affected by quenching. Therefore, in the most sensitive remote techniques we use absorption to detect and to determine the concentrations of air pollutants. Only homonuclear diatomic molecules such as N_2, O_2, H_2 and atoms such as He, Ar, Kr are not detectable by absorption measurements.

Absorption measurements are based on the well-known Beer–Lambert extinction law according to which the transmitted radiation intensity $I(\lambda, L)$ at wavelength λ is related to the incident intensity $I_0(\lambda, 0)$:

$$I(\lambda, L) = I_0(\lambda, 0) \cdot \mathrm{e}^{-L\alpha(\lambda)}, \qquad \alpha = \alpha_{\text{molecular}} + \alpha_{\text{aerosol}} + c\sigma(\lambda)\,, \tag{1}$$

where $\alpha_{\text{molecular}}$ is related to the molecular (Rayleigh) scattering, α_{aerosol} describes scattering on aerosols particles (Mie scattering), $\sigma(\lambda)$ indicates the absorption cross-section, L is the length of optical path and c is gas concentration. Typical Raman scattering and fluorescence are not taken into account because they can be neglected in total extinction. The absorption cross-section is a characteristic property of any species and can be measured in laboratory. Having known the shape and value of $\sigma(\lambda)$ it is possible to determine the concentration of an unknown gas inside a cell or along an atmospheric optical path.

Various methods can be used to monitor pollutants remotely. DOAS and DIAL attract marked attention because they have some unique features.

4. DOAS METHOD

DOAS (Differential Optical Absorption Spectroscopy) method bases on the measurements of transmission in the function of wavelength along optical path [5]. The

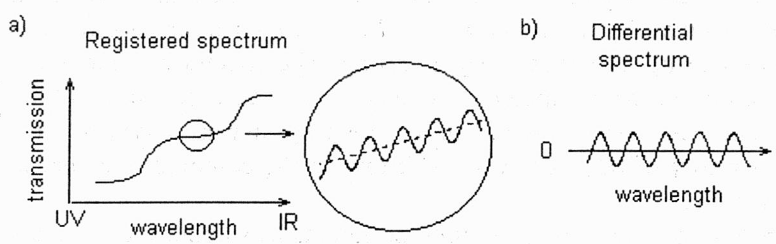

Fig. 1. Principles of DOAS method

transmission spectrum investigated over broad region (typically from near IR to UV) consists of two parts. One part varies slowly with wavelength, while the other, which is weaker, varies much faster (figure1a). Only absorption can cause fast changes. The slow changes are due to scattering (Mie and Rayleigh), transmission factor of the optics and unknown measurement errors. Therefore, the natural way to obtain absorption from such a spectrum is to subtract the slowly varying part. After this operation differential absorption spectrum can be obtained (figure1b).

The DOAS method is commercially available in the OPSIS system. OPSIS is a registered trade mark of Opsis Aktiebolog for the ABB systems. It is manufactured by OPSIS Inc. of Furulund, Sweden, and Old Greenwich, Connecticut [8]. The measurement principle of OPSIS is presented in figure 2.

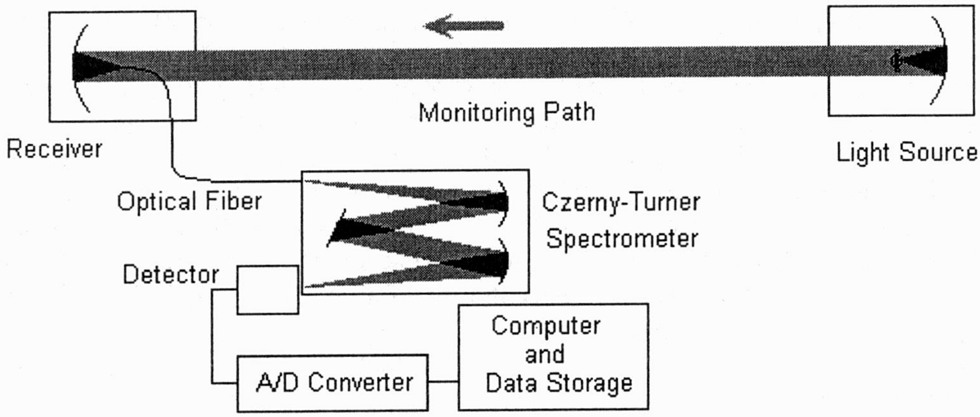

Fig. 2. OPSIS system

The transmitter with a high-pressure xenon lamp generates a concentrated beam of light that consists of wavelengths from short-wave UV to long-wave IR. The beam of light from the source is emitted along a chosen path. The signal collected at the receiver is sent over a fiber (optic cable) to the Czerny–Turner spectrometer in which the light is broken up into spectra by a grating. The spectra are converted to digital signals and stored in the computer's memory. The scan is repeated a hundred times per second. The calculations carried out by the computer are based on library spectra recorded under controlled laboratory conditions in the computer memory at 1000 different wavelengths.

The OPSIS system can monitore such pollutants as sulphur oxide, oxides of nitrogen, hydrochloric acid, formaldehyde, mercury vapours, ozone and several hydrocarbons including methane, styrene, benzene, toluene and xylene.

OPSIS system allows the distance between the transmitter and receiver to range from 200 meters up to a few kilometres. This is a standard system. The detection limits for most components are of the order of μg/m^3.

The application of the OPSIS systems is focused on:
measurements of urban pollution and road traffic pollutants,
monitoring of airport areas, industrial environments, dumping sites,
fence line monitoring.

Table

OPSIS systems in Poland

Measurement	City	Path length	Pollutants	Year
Traffic pollutants	Warsaw	328.8 m 144.2 m	NO_2, SO_2, O_3, formaldehyde, benzene, toluene, *p*-xylene NO	1992
Traffic pollutants	Poznań	328.0 m	NO_2, SO_2, O_3, formaldehyde, benzene, toluene, *p*-xylene	1992
Traffic pollutants	Toruń	426.0 m	NO_2, SO_2, O_3, formaldehyde, benzene, toluene, *p*-xylene	1994
Traffic pollutants	Tarnobrzeg	variable	NO_2, SO_2, O_3, CS_2, benzene, toluene, *p*-xylene	1994
Traffic pollutants	Bydgoszcz	593.3 m	NO_2, SO_2, O_3, formaldehyde, benzene, toluene, *p*-xylene, phenol	1994
Pollution of background in suburbs	Częstochowa	648.0 m	NO_2, SO_2, O_3, formaldehyde, benzene, toluene, *p*-xylene, phenol	1992
Pollution of background in suburbs	Lublin	280.0 m 142.0 m	NO_2, SO_2, O_3, formaldehyde, NO, NH_3	1994
Pollution of background in suburbs	Bielsko-Biała	505.0 m	NO_2, SO_2, O_3, benzene, toluene, phenol	1995
Pollution of background in urban agglomeration	Gdańsk	531.5 m	NO_2, SO_2, O_3, formaldehyde, benzene, toluene, *p*-xylene	1994
Pollution of background in urban agglomeration	Płock	340.0 m	NO_2, SO_2, benzene, toluene, phenol	1995
Pollution of background in urban agglomeration	Włocławek	384.5 m	NO_2, SO_2, O_3, formaldehyde, benzene, toluene, *p*-xylene	1995
Pollution of background in urban agglomeration	Czechowice-Dziedzice	486.0 m	NO_2,SO_2,O_3,formaldehyde, benzene, toluene, *p*-xylene, phenol	1996

The OPSIS systems have been applied to measurements of ambient air pollutants at 13 places in Poland [9]. They are used to conduct the following air quality monitorings:

monitoring of urban air quality at street level, monitoring of background levels in suburbs,

monitoring of urban air quality at roof-top level.

In the table, a list of the OPSIS systems working in Poland is presented.

The advantages of remote systems with the long-open path lie in the fact that they increase their sensitivity with the increase in range. However, they have also the disadvantages of being double-ended and lacking spatial resolution. The latter problem is partly solved by remote optical systems described later on.

5. LIDAR SYSTEMS

LIDAR is an acronym of Light Detection and Ranging. The LIDAR works similarly to RADAR (Radio Detection and Ranging) [10]. The transmitter of the LIDAR produces the pulses of light and sends them to the atmosphere (figure 3). When the pulse is travelling through the medium it is scattered and absorbed. The part of the radiation, which is scattered in backward direction, is collected and registered by receiver as a time-dependent signal.

Typically the transmitter is a high-power laser, which produces pulses of radiation of nanosecond duration time. The time width of the pulse is very important because it is related to spatial resolution of measurements. Spatial resolution is equal to half time of pulse duration. The receiver consists of an optical telescope, light detector, signal acquisition system and computer.

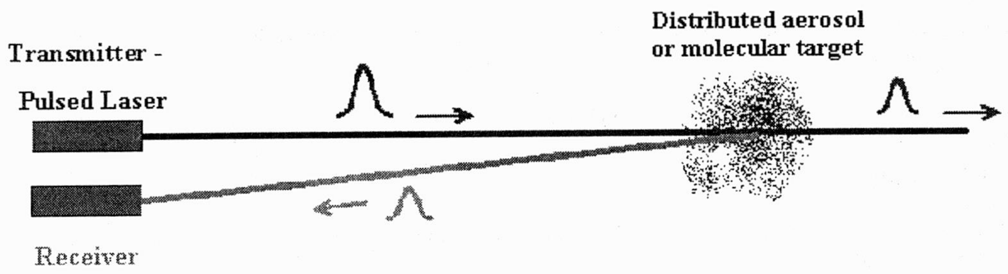

Fig. 3. Principles of the LIDAR

As it was mentioned earlier, light attenuation processes in the atmosphere are described by the extinction coefficient α (1). Since the part of extinction coefficient is absorption, which changes from one molecule to the other, it is possible to detect selectively the concentration of the chosen gas. LIDAR technique based on this idea of the measurements of gas concentration is called DIAL (Differential Absorption Lidar).

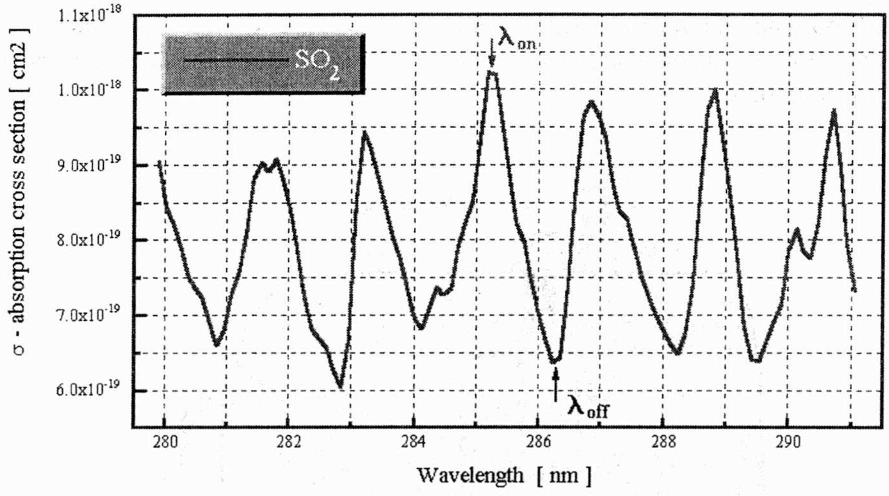

Fig. 4. Absorption spectrum of SO_2. DIAL 'on' and 'off ' wavelength are marked

In order to get information about molecular absorption only from extinction coefficient, in the DIAL system the pulses of light at two different wavelengths are used. One of the wavelengths is tuned in to the absorption line of the investigated species (the so-called λ_{on}), while the other (λ_{off}) is slightly detuned (figure 4). For small difference between the two wavelengths (within a few nanometers) the coefficients $\alpha_{molecular}$ and $\alpha_{aerosol}$ for the respective 'on' and 'off ' pulses are almost equal. Therefore the difference between 'on' and 'off ' signals is only due to the difference in the well-known absorption cross-sections σ_{abs} for λ_{on} and λ_{off} multiplied by a concentration for which we are looking:

$$\alpha(\lambda_{on}) - \alpha(\lambda_{off}) = c[\sigma_{abs}(\lambda_{on}) - \sigma_{abs}(\lambda_{off})]. \qquad (2)$$

It's clear from the above that the idea of DIAL is very similar to DOAS except that the measurement is performed for two chosen wavelengths, and the results are distance-dependent.

Under real conditions, because of very small ratio of signal to noise for a single 'on','off ' measurement, return signal is averaged over a few hundred laser pulses. Then the signal is processed with a proper inversion algorithm by a computer in order to get the concentration as a function of distance from the instrument.

The DIAL technique allows us to measure the concentrations of ozone (O_3) [11]–[13], [34], sulphur dioxide (SO_2) [11], [12], [14], [34], nitrogen monoxide (NO) [15], [16], nitrogen dioxide (NO_2) [15], [17]–[19], [34], methane (CH_4) [20], carbon dioxide (CO_2)

[20], water vapour (H_2O) [21]–[26], mercury (Hg) [27],[28], hydrogen chloride (HCl) [29],[30], chloride (Cl_2) [31], ethylene (C_2H_4) [32], toluene ($C_6H_5CH_3$) [33], [34], benzene (C_6H_6) [34], and other VOC's [34]. The detection limit is within the range of few ppb (hundreds ppb) and the measurement distance varies from a few hundred meters up to 40 kilometers. Both parameters strongly depend on the kind of investigated species and atmospheric conditions.

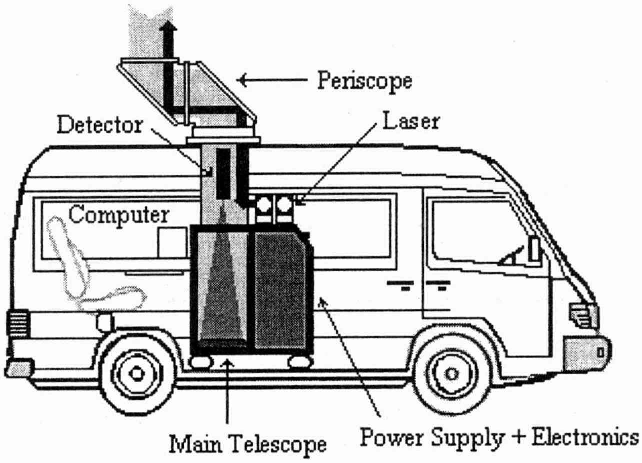

Fig. 5. Mobile DIAL system used by IFD UW Lidar Group

In Poland, a mobile DIAL system has been used by the group from the Institute of Experimental Physics of Warsaw University. The group developed in last years an advanced system constructed by Elight Laser System GmbH, Germany. This is one of few such systems operating in Europe. Figure 5 presents general view of the setup, and figure 6 shows block scheme which is also typical of other DIAL systems. Their basic parts are as follows:

transmitter containing tuned Ti:Saphire laser and accompanying optics with a rotating periscope,

receiver with a 40 cm diameter mirror collecting backscattered radiation, detector which is photomultiplier (PMT) used as a detector, digital oscilloscope for signal acquisition and a computer for data evaluation and storage.

More detailed description of the setup can be found elsewhere [35]. Using this system it is possible to investigate such species as ozone, nitrogen dioxide, sulphur dioxide, benzene and toluene with detection limit of about 5–10 ppb in the range up to 2.5 km. The time of measurement in one direction is 0.5–3 minutes. The periscope allows us to perform measurements in different directions. The time necessary for two-dimensional scan (10 directions) varies from 5 to 30 minutes, depending on the species being investigated and atmospheric conditions.

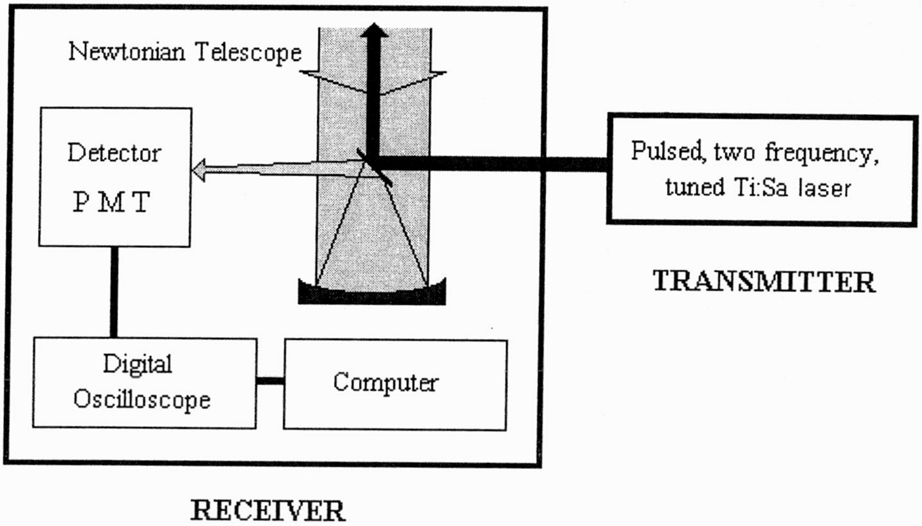

Fig. 6. Scheme of DIAL system

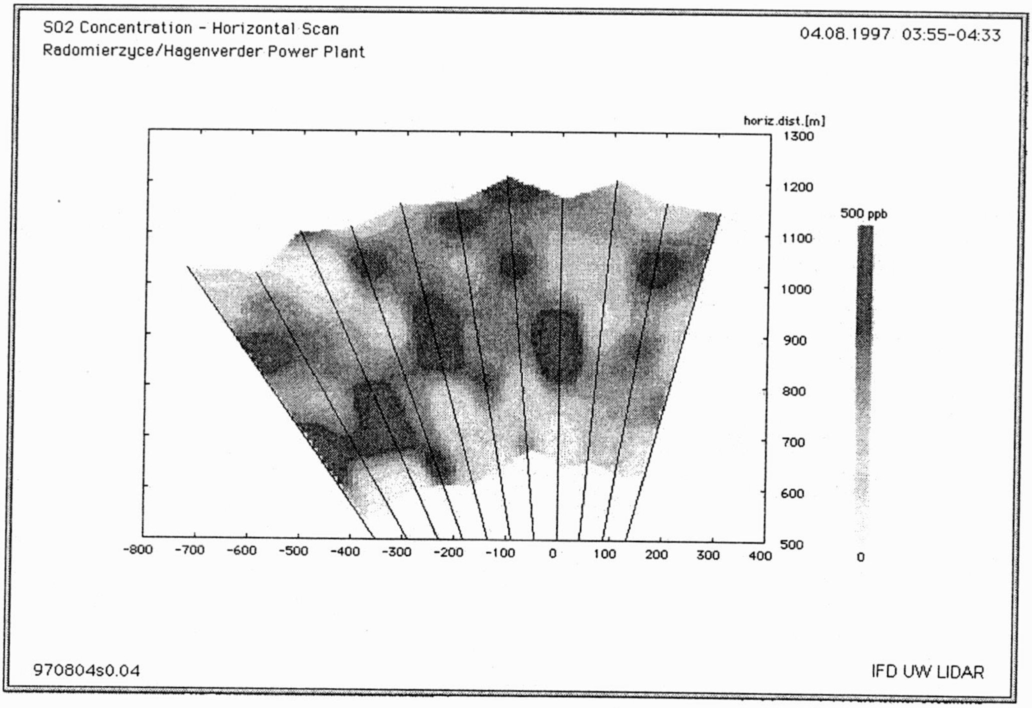

Fig. 7. SO_2 concentration over Hagenverder Power Plant, Germany. Measurement performed from Radomierzyce village, Poland

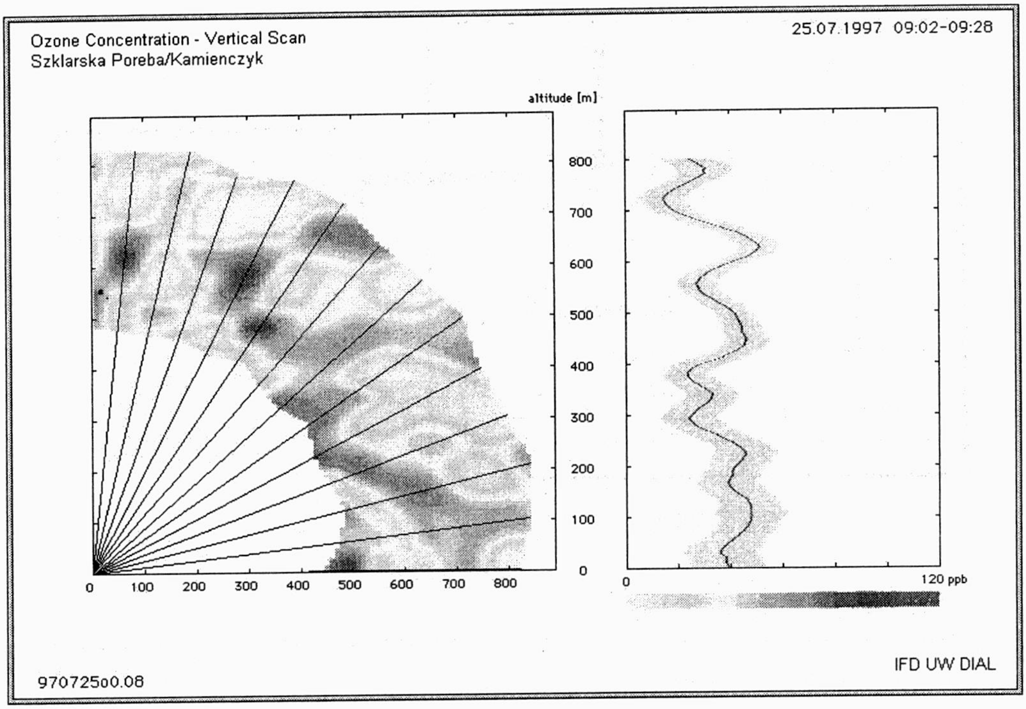

Fig. 8. Ozone concentration over Szklarska Poręba

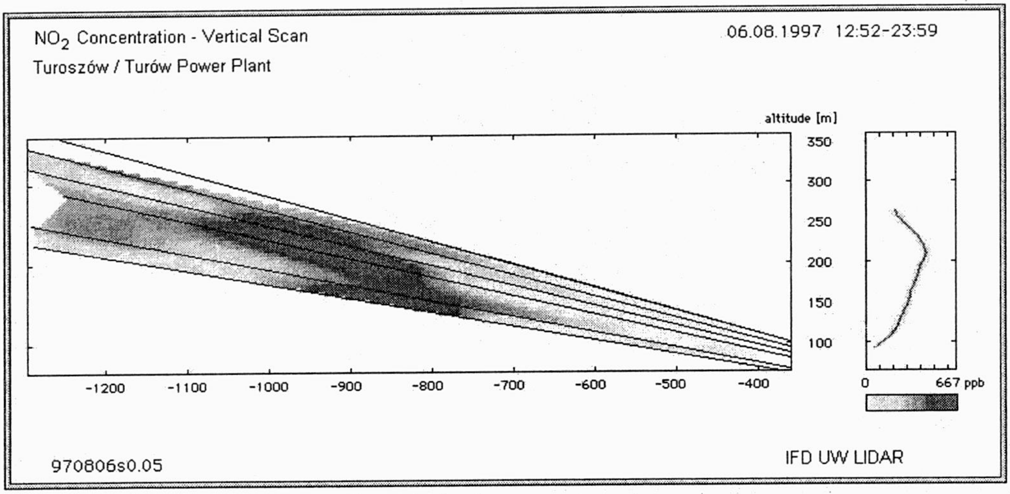

Fig. 9. Map of NO_2 concentration in the smoke stack cross-section. Data collected over Turów Power Plant in Turoszów

Figures 7, 8, 9 show some selected results of measurements performed by the system described above. The solid black lines in the pictures indicate directions of measurements. The curves on the right-hand side in figures 8 and 9 are vertical profiles of concentrations obtained by averaging values from a map along the horizontal direction. The shadows around curves show measurement error.

Figure 7 illustrates an obvious advantage of LIDAR system over other remote systems. The map shows concentration of sulphur dioxide in horizontal cross-section over Hagenverder Power Plant, Germany. The measurements were done from Radomierzyce, Poland, in September 1997. The strongest plums in the center of the picture are plums from a cooling towers which act as sources of SO_2. Another example is the imission measurement of ozone (figure 8).

The map presents vertical distribution of ozone concentrations over Szklarska Poręba town in the Karkonosze Mountains. Measurements were also done in July 1997. The average concentration approaches 25 ppb. Figure 9 shows the emission measurement again. In this case, the smoke plum containing nitrogen dioxide from one of the chimneys of Turów Power Plant in Turoszów was investigated. It is worth noticing that the rate of emission from such measurements can easily be calculated.

6. CONCLUSIONS

The optical remote methods such as DOAS and DIAL have proved to be very successful during recent years. A commercial DOAS instrument manufactured by OPSIS Inc. has been accepted in Europe for urban pollution measurements. DIAL method is now in operation for many pollutants, and several mobile system using this method has been developed.

REFERENCES

[1] VESILIND P.A., PEIRCE J.J., *Environmental Pollution and Control*, Boston, London, Sydney, Wellington, Durban, Toronto, Butterwoth Publishers, 1983.

[2] STERN A.C., *Air Pollution*, third edition, Vol. 3, *Measuring, Monitoring and Surveillance of Air Pollution*, New York, San Francisco, London, Academic Press Inc., 1976.

[3] STERN A.C., BOUBE R.W., TURNER D.B., FOX D.L., *Fundamentals of Air Pollution*, second edition, Orlando, Florida, Academic Press, 1984.

[4] LODGE J.P., *Methods of Air Sampling and Analysis*, third edition, Lewis Publishers Inc., 1989.

[5] SIGRIST M.W., *Air Monitoring by Spectroscopic Techniques*, New York, Chichester, Brisbane, Toronto, Singapore, A Wiley- Interscience Publication, 1994.

[6] GRANT W.B., KAGANN R.H., MCCLENNY W.A., *Optical Remote Measurement of Toxic Gases*, J. Air Waste Manage. Assoc., 1992, Vol. 42, 1, 18.

[7] PERRY R., YOUNG R., *Handbook of Air Pollution Analysis*, London, Chapman and Hall Ltd., 1977.

[8] VISWANATHAN S., SANDS M.D., *Monitoring Ambient and Source Pollutants*, Environ. Sci. Technol., 1992, Vol. 26, 4, 650.
[9] ŻEGLIN M., BOROWSKA M., *Application of Telemetric Technique DOAS to Air Pollution Measurements by OPSIS System in Poland*, Third International Symposium and Exhibition on Environmental Contamination in Central and Eastern Europe, Warsaw, 1996, 144.
[10] MEASURES R.M., *Laser Remote Sensing. Fundamentals and Applications*, J. Wiley & Sons, New York, 1984.
[11] HAWLEY J.G., *Dual-Wavelength Laser Radar Probes for Air Pollutants*, Laser Focus, 1981, Mar., 60–62.
[12] BROWELL E.V., Opt. Eng., 1982, 21, 128.
[13] WERNER J., ROTHE K.W., WALTHER H., Appl. Phys. B, 1983, 32, 113.
[14] UCHINO O., TOKUNAGA M., MAEDA M., MIYOZOE Y., Opt. Lett., 1983, 8, 347.
[15] KÖLSCH H. J., RAIROUX P., WOLF J.P., WÖSTE L., *Simultaneous NO and NO_2 DIAL Measurements Using BBO Crystals*, Appl. Optics, 1989, 28, 2052–2056.
[16] ALDÉN H., EDNER H., SVANBERG S., *Laser monitoring of atmospheric NO using ultraviolet differential-absorption techniques*, Opt. Letters, 1982, 7, 543–545.
[17] KILLINGER D.K., MOORADIAN A. (Eds.), *Optical and laser remote sensing*, Springer-Verlag, Berlin, 1983.
[18] ROTHE K.W., BRINKMAN U., WALTHER H., Appl. Phys., 1974, 4, 181.
[19] FREDRIKSSON K., GALLE B., NYSTROM K., SVANBERG S., *Mobile Lidar System for Environmental Probing*, Appl. Optics, 1981, 20, 4181–4189.
[20] UCHIUMI M., CHEE O., MUROAKA K., MAEDA M., UCHINO O., *DIAL measurements of CH_4, CO_2, N_2O using tunable IR source based on the Titan-saphire laser*, 17th IRLC Proceedings, 31–12, Sendai, Japan, 1984.
[21] SCHOTLAND R.M., Proceedings, *Third Symposium on Remote Sensing of the Environment*, Environmental Research Institute of Michigan, Ann Arbor, 1964, 215–224.
[22] SCHOTLAND R.M., Proceedings, *Fourteenth Weather Radar Conference*, U. Arizona, Tucson, 1971.
[23] SCHOTLAND R.M., J. Appl. Meteorol., 1974, 13, 71.
[24] BROWELL E.V., WILKERSON T.D., MCILRATH T.J., Appl. Optics, 1979, 18, 3474.
[25] CAHEN C., PELON J., FLAMANT P., MÉGIE G., C.R. Acad. Sci. Ser. II, 1981, 25, 292.
[26] WERNER C., HERRMANN H., J. Appl. Meteorol., 1981, 20, 476.
[27] ALDÉN H., EDNER H., SVANBERG S., *Remote measurements of Atmospheric Mercury Using Differential Absorption Lidar,* Opt. Lett., 1982, 7, 221.
[28] EDNER H., FARIS G.W., SUNESSON A., SVANBERG S., *Atmospheric Atomic Mercury Monitoring Using Differential Absorption Lidar Technique*, Appl. Optics, 1989, 28, 921–930.
[29] WEITKAMP C., HEINRICH H.J., HERRMANN W., MICHAELIS W., LENHARD V., SCHINDLER R.N., *Measurements of Hydrogen Chloride in the Plume of Incineration Ships*, 5th International Clean Air Congress, 20–26 Oct. 1980, Buenos Aires, Argentina; also in GKSS 80/E/55.
[30] WEITKAMP C., *The Distribution of Hydrogen Chloride in the Plume of Incineration Ships: Development of New Measurements Systems*, Wastes in the Ocean, 1981, Vol. 3, Wiley; also GKSS 81/E/57.
[31] EDNER H., FREDRIKSSON K., SUNESSON A., WENDT W., *Monitoring Cl_2 Using a Differential Absorption Lidar System*, Appl. Optics, 1987, 26, 3183–3185.
[32] ROTHE K.W., Radio Electron. Eng., 1980, 50, 567.
[33] MILTON M.,WOODS P.T., JOLLIFE B., SVANN N., MCMELLVEN, *Measurements of Toluene and other Aromatic Hydrocarbons by Differential-Absorption LIDAR in the Near-Ultraviolet*, Applied Physics B, 1992, 55, pp. 41–45.
[34] WEITKAMP C., GOERS U.-B. et al., *Laser Remote Sensing of Sulphur Dioxide, Nitrogen Dioxide, Toluene, Ozone and Dust in the Industrial Area of Cubatao (Brazil)*, 18th ILRC Berlin, Germany, 22–26 July 1996.
[35] CHUDZYŃSKI S., ERNST K., STACEWICZ T., SZYMAŃSKI A., *Mobile Lidar laboratory*; Proceedings of SPIE, 1997, Vol. 3188, 180.

ZASTOSOWANIE ZDALNYCH OPTYCZNYCH TECHNIK POMIAROWYCH
DO KONTROLI JAKOŚCI POWIETRZA

Metody i techniki analityczne są istotne w monitorowaniu środowiska. Pomiary nie polepszają bezpośrednio stanu środowiska, lecz dostarczają informacji o jego zanieczyszczeniu. W artykule przedstawiono obszerny i krytyczny przegląd zdalnych metod optycznych stosowanych do pomiaru zanieczyszczeń powietrza. W szczególności opisano metody DOAS i LIDAR.