Detection and monitoring of pollutant sources with Lidar/Dial techniques

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Abstract. It's well known that air pollution due to anthropogenic sources can have adverse effects on humans and the ecosystem. Therefore, in the last years, surveying large regions of the atmosphere in an automatic way has become a strategic objective of various public health organizations for early detection of pollutant sources in urban and industrial areas.

The Lidar and Dial techniques have become well established laser based methods for the remote sensing of the atmosphere. They are often implemented to probe almost any level of the atmosphere and to acquire information to validate theoretical models about different topics of atmospheric physics. They can also be used for environment surveying by monitoring particles, aerosols and molecules.

The aim of the present work is to demonstrate the potential of these methods to detect pollutants emitted from local sources (such as particulate and/or chemical compounds) and to evaluate their concentration. This is exemplified with the help of experimental data acquired in an industrial area in the south of Italy by mean of experimental campaign by use of pollutants simulated source. For this purpose, two mobile systems Lidar and Dial have been developed by the authors. In this paper there will be presented the operating principles of the system and the results of the experimental campaign.

Keywords - Lidar; Dial; pollutants; water vapour

1. Introduction

Particulate matter (PM), harmful gases/aerosols or other pollutants, emitted by vehicles in urban traffic and industrial activities, can greatly affect environment air quality and have direct implications on human health. Air pollution is a huge problem and not just for people living in smog-choked cities; the consequences for society are relevant through such things as global warming and damage to the ozone layer, it has the potential to affect us all. In fact, air pollution and urban air quality are listed as two of the world's worst toxic pollution problems in the 2008 Blacksmith Institute World's Worst Polluted Places report [1]. According to the 2014 WHO report, air pollution in 2012 caused the deaths of around 7 million people worldwide [2].

Limits and thresholds of pollutants are typically regulated by government agencies. In Europe, emissions of many air pollutants have decreased substantially over the past decades, resulting in improved air quality across the region. However, air pollutant concentrations are still too high, and air quality problems persist. A significant proportion of Europe's population live in areas, especially cities, where exceedances of air quality standards occur [3, 4]. Therefore, monitoring the quality of the

atmosphere in such urban areas is particularly relevant and is one of the key safety aspect of the 21st century.

Lidar and Dial techniques are two examples of laser remote sensing methodologies which allow the exploration of the atmosphere. Traditionally they have been used to acquire information necessary to create or validate several models about different topics of atmospheric physics [5-9]. On the other hand, if properly automated, they have the potential of being deployed in environmental surveying to monitor particulate and gas. A typical example of the use of Lidar in the detection of backscattered peaks is the detection of forest fires [10-12]. They can also be deployed for environment surveying of particulate, aerosol and molecule concentrations (e.g. in urban areas) [13-17].

In order to understand the causes of air pollution, several divisions can be made. Primarily air pollutants can be caused by primary sources or secondary sources. The pollutants that are a direct result of the process can be called primary pollutants. A classic example of a primary pollutant would be the sulfur-dioxide emitted from factories. Secondary pollutants are the ones that are caused by the inter mingling and reactions of primary pollutants. Smog created by the interactions of several primary pollutants is known to be as secondary pollutant [3].

Most pollutants are present in the atmosphere in a gaseous state in concentrations between 0.01 and 10 ppm for molecules and between 0.01 and 10 ppb for metal vapours. Generally, in order to detect such substances and determine quantitatively their concentration, point monitoring stations are employed. These fixed monitoring stations implement different measurement techniques with integration times varying from one minute to a few hours. Unfortunately they can typically evaluate the concentration of the substance, for which they have been calibrated, only in a limited area close to the detector. So, to cover large areas, these fixed monitoring stations have to be deployed in large numbers. They have also to be connected in suitable networks. In addition to the practical difficulties and the economic implications, it is to be emphasized that no sound methodology exists to determine the optimal number of measurement points and their localization. Therefore, all these reasons motivate the development of a sensitive, flexible and fast method with the capability of measuring concentrations of atmospheric gaseous pollutants over a wide areas. With the development of reliable lasers, emitting in the appropriate range of wavelengths, Lidar and Dial systems have become suitable and competitive techniques. In fact, they are widely recognized as a cost-effective alternative to monitor large region of atmosphere [18]. These techniques are successfully used to measure stratospheric parameters in order to obtain experimental data to benchmark various atmospheric models. Other authors are using the Lidar technique in the field of environmental monitoring. Most of these applications work in the UV or visible region [11, 12]. On the other hand, it is worth trying to extend the range of operation to the infrared spectral region, where there are many molecular absorption bands. Indeed, the vast majority of the substances to be detected (SO_x, ammonia, ozone etc.) emit and absorb in the spectral range between 9 and 11 μm. Furthermore a wide atmospheric spectral window is located around the wavelength of 10 μm. This characteristic of the atmosphere allows working without significant attenuation by the main components of the atmosphere (CO_2, H_2O). This is especially true in the low troposphere, where more strict should be the monitoring of the pollution levels.

Taking into account all these aspects, it has been decided to develop an integrated monitoring system using a Lidar and Dial station combining the properties of a Nd:YAG laser for the Lidar system and a TEA CO_2 laser for the Dial one.

In this work, attention is focused on surveying particulate/aerosol generated by combustion of vegetable matter in order to simulate the presence of pollutants sources produced in urban and industrial areas. Therefore, the main operational approach envisages the continuous monitoring of the area to be surveyed with the Nd:YAG laser in order to evaluate any aerosol variation. When a significant backscattered signal is detected the Dial system is activated with the purpose of determining whether the increase of the aerosol level is associated to an increase in water vapour. This approach has been tested, up to the detection of water vapour, in an extensive experimental campaign which has been carried out in Calabria, in the south of Italy.

In the present paper the authors, after a technical description of the system used, will analyse and discuss the results of the experimental campaign carried out using two different laser systems.

2. Material and methods

The measurements described in the paper have been performed with the Lidar system developed by the Quantum Electronic Plasma Physics and Materials research group (QEPM) at the University of Rome Tor Vergata. Furthermore, a ground-based Dial station has been built and continuously upgraded at the CRATI s.c.r.l. c/o University of Calabria [19-22]. The physical and constructive parameters of this mobile station, used to obtain the results described in this paper, are reported in Table 1 and 2 [23, 24].

Table 1. Parameters of CO_2 Dial system.

Transmitter TEA CO_2 laser	
Output power	10^9 W
Beam divergence	0.77 mrad
Spectral range	9:11 μm
Receiver	
Primary ROC	2400 mm
Primary diameter	400 mm
ZnSe focal length	100 mm
Total Focal length	576.6 mm
F.O.V.	0.88 mrad
Detector type	HgCdTe
Detector Sensitivity D*	$3.38 \cdot 10^{10}$ cmHZ$^{1/2}$/W
Detector size	1 mm^2

Table 2. Parameters of Nd:YAG Lidar system.

Transmitter Q-Switch Nd:YAG laser	
Energy pulse at 1064 nm	360 mJ
Pulse time wdith	5 ns
Divergence angle	0.5 mrad
Pulse frequency	10 Hz
Receiver	
Telescope type	Newtonian
Nominal focal length	1030 mm
Primary mirror diameter	210 mm
Detector	Photomultiplier Tube - PMT
Photocatode sensibility	72 mA/W
Response time	30 ns

Table 1 summarizes the main characteristics of the CO_2 laser and Table 2 is an overview of the Nd:Yag laser [25, 26]. The Dial laser is a homemade TE (Transversal-Excited) CO_2 source. The laser is tunable over about 60 laser lines in the spectral range between 9 and 11 μm. The telescope is based on the Newtonian configuration. The primary mirror has a diameter of 400 mm and the system presents a total focal length of 576.6 mm. The Lidar measurements are performed with a commercial Nd:Yag laser; the main characteristics of the whole system developed are reported in Table 2.

   The two apparatuses are installed on a mobile system and therefore constitute a fully mobile station, which can be used for in field campaigns in any location.

   The experimental campaign, described in this paper, has taken place in the industrial area of Lamezia Terme, in the Calabria region in the south of Italy (see Figure 1), in the springtime. The Dial laser has been tuned on the wavelengths 10R20 and 10R18, corresponding to the absorbing and non-absorbing wavelengths of the water molecule [26-29]. The wavelength 1064 nm is used for the Lidar system. A controlled fire, produced by the combustion of a known quantity of vegetables (2 Kg of vegetable matter every 30 minutes), has been lighted into a chimney at a distance of about 400 m from the mobile station. An effort has been made to keep the quantity of smoke constant by periodically adding a suitable amount of vegetable material. This trick has already been successfully used to simulate the presence of air pollution in several experimental campaigns [18, 19, 23, 25, 30].

   The backscattered signals, in the presence of smoke, have been acquired along system pointing direction with a Line of Sight – LOS of 270 degrees to the North (see Figure 1). In fact, this LOS corresponds to the position of the chimney; nonetheless, the system is able to rotate its pointing angle with a mechanical support.
The signals at both wavelengths have been averaged over 100 individual laser pulses.

Figure 1. Map of the site of the experimental campaign. The red arrow indicates the Line of Sight of the system.

The entire apparatus is controlled by a software package, written in Labview and Matlab, explicitly developed for this application. The laser activation and the wavelength selection, together with the rotation of the telescope, are controlled by an hardware system working with a Labview program developed by QEPM. The data acquisition, the signal processing algorithms and the graphic presentation have been implemented using Matlab language. The Matlab scripts elaborate raw data acquired by the system and calculate the distance of smoke from the station. This last piece of information is of course an essential input to give a warning when something strange is found in atmosphere and for the coordination of timely remedial action. Then, in case of particulate significant

peaks, they also evaluate the water vapour concentration. The QEPM has also developed a new method to detect the LIDAR peaks [31].

3. Campaign Results

The main purpose of the monitoring campaign was to track the pollutants sources emitted by the urban traffic or factories and to give a warning of their presence in atmosphere. For this aim, our system can be operated in both Lidar and Dial configurations in sequence.

In the Dial method, Lidar measurements are performed at two wavelengths to deduce the concentration of a trace gas of interest. By properly selecting the Lidar operating wavelengths, it is possible to minimize the effect of unwanted trace gases in the atmosphere. The two wavelengths of the Dial system are chosen such that at the first wavelength (λ_{on}) the gas of interest absorbs strongly (on-absorption band) and at the second wavelengths (λ_{off}) the gas absorbs weakly (off-absorption band).

The concentration of water vapour, in the atmosphere, is highly variable (as low as few millibars and as high as few tens of millibars). A significant variation of water vapour is also a typical emission in the early phase of vegetable combustion process, such as a forest fire [10-12, 30]. For the calculation of this concentration, the chosen on and off lines are, respectively, 10R20 and 10R18 in the branch at 10 μm. The 10R20 corresponds to 10.591 μm and the 10R18 to 10.571 μm. At these wavelengths the difference in the absorption coefficients of the water molecule ($\Delta\sigma$) is equal to $7.7 \cdot 10^{-4} \mathrm{cm}^{-1}\mathrm{atm}^{-1}$ and the value of attenuation coefficient (α) is 0.05 Km^{-1} [32].
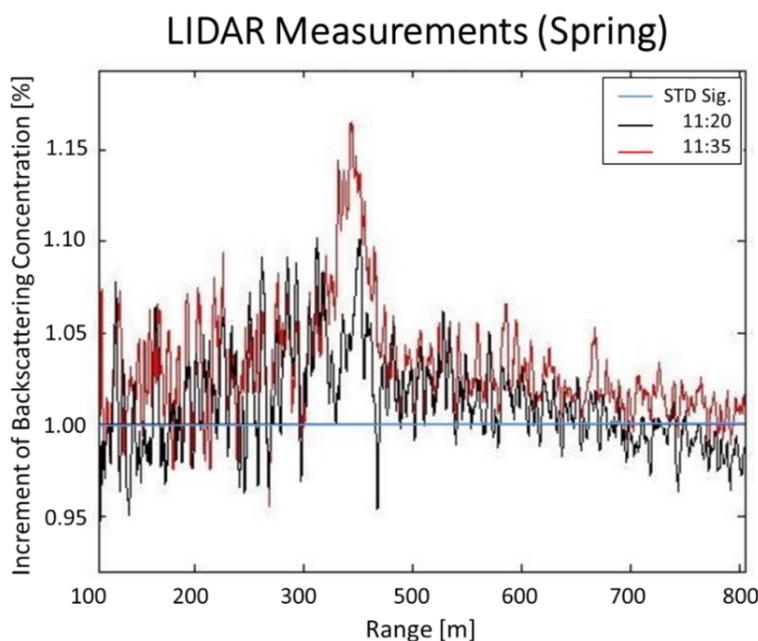
Figure 2. Backscattered Lidar signal in presence of smoke plume.

The first warning measurements are performed by first scanning the region to be surveyed with the 10R18 non absorption line.

As we can see in the Figure 2, there is an increment of aerosol backscattering concentration in proximity to the chimney, in particular during the time slot in which the fire is lighted.

When one or more peaks are detected in the backscattered signal (see Figure 2), a measurement is taken in the same direction using the second absorption line 10R20. The backscattered signal detected

at this second wavelength (see Figure 3) can then be used to determine the water vapour concentration. Assuming that the atmosphere is homogeneous along the path length, the path average concentration of the absorption molecule n can be calculated by [32]:

$$n(R) = \frac{1}{2\cdot(\sigma_{on}-\sigma_{off})} \cdot \left[\frac{d}{dR}\left\{ \ln\left[\frac{P(\lambda_{off},R)}{P(\lambda_{on},R)}\right] - \ln\left[\frac{\beta(\lambda_{off},R)}{\beta(\lambda_{on},R)}\right] \right\} + k_0(\lambda_{off},R) - k_0(\lambda_{on},R) \right] \qquad (1)$$

where the index *on* or *off* indicate the value referred to the absorption band or to detuned wavelength, *n(R)* is the concentration as a function of the range, P is the power received by the detector, σ the differential absorption cross-section, β the backscattered coefficient and k_0 the total atmospheric extinction exclusive of the quantity relating to the absorption due to the substance that is being studied.

Assuming that the backscattered coefficients and the total atmospheric extinction value are almost identical at the two wavelengths, the equation 1 can be re-written as:

$$n(R) = \frac{1}{2\cdot(\sigma_{on}-\sigma_{off})} \cdot \left[\frac{d}{dR}\left\{ \ln\left[\frac{P(\lambda_{off},R)}{P(\lambda_{on},R)}\right] \right\} \right] \qquad (2)$$
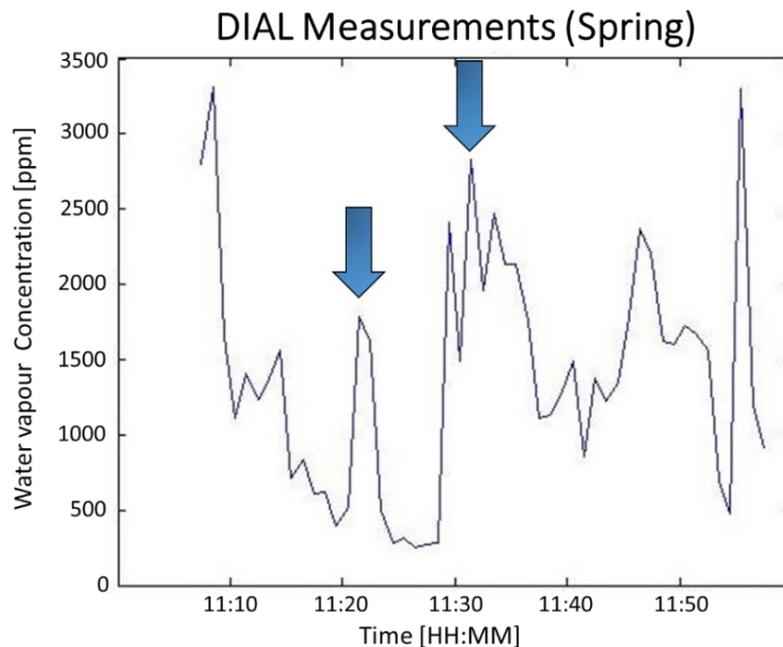
Figure 3. Dial signal detected in presence of fire: water vapour profile. The two peaks are quite small because a minimum amount of vegetable material has been burned.

As we can see in Figure 3, the highlighted peaks, that represent the increment of water vapour concentration, have been detected in the same position and time slot of Figure 2. Comparing the two figures, in fact, the increment of backscattering concentration is the result of strong presence of particulate/aerosol in atmosphere. This has been observed in a chimney located at a distance of about 400 m from the mobile station during a controlled fire lighted between 11:20 and 11:35. Therefore, the

experimental results indicate that the sensitivity of the Lidar/Dial measurements is quite good and confirm the effectiveness of the system to detect the emission in the atmosphere of (small amounts) of pollutants, evaluating their concentration.

4. Conclusions

The Lidar/Dial techniques are well established methods to explore the atmosphere and they are also widely recognized as a cost-effective alternative to monitor large regions of the atmosphere.

The aim of the present work is to demonstrate the capabilities of our mobile system to make a detection and a preliminary identification of the presence of pollutant sources in the atmosphere. For this purpose, our system can be operated in both Lidar and Dial configurations in sequence.

The first Lidar measurement is performed to evaluate the presence of aerosol/particulate into the atmosphere, using a non absorption water wavelength 10R18 (10.571 μm). If the returned signal reports a backscattering peak, the presence of a smoke plume is probable. To confirm this hypothesis, a second Dial measurement is carried out to reveal a second component emitted during the combustion process. The second component chosen is water vapour, which is largely produced during the first combustion stage. Moreover, thanks to a "fingerprint" database it will be possible to recognise the composition of plume, discriminating between industrial pollutant sources and forest fires. Indeed the advances in CO_2 laser technology permit to determine the water concentration, allowing to detect and\or exclude the presence of vegetable fires (depending on the application) [26, 30].

The volume backscattering coefficients of atmospheric aerosol for two wavelengths (10R20 and 10R18) in a Dial methodology were previously measured with cell tests [33]. As shown in the previous section, this information can be used to determine changes in the water concentration with respect to the background humidity level of the atmosphere.

The experimental results confirm the sensitivity and effectiveness of the Lidar/Dial measurements. In fact the proposed system detects even the first combustion stage, detecting the emission in the atmosphere of particulate/aerosol produced by vegetable fires and can be used to determine the concentrations of pollutants generated by urban traffic or industrial activities. Moreover the same system is expected to have the capability to give a warning in case of significant presence of pollutants in atmosphere and to reduce the false alarms [30].

A three stages approach for the surveying of large areas has been proposed: a) detection of backscattered radiation with Lidar measurements; b) determination whether a vegetable fire is present searching for water vapour; c) determination of the concentration of candidate pollutants with the Dial technique. These steps have have been successfully tested in field, during the experimental campaign performed near the CRATI s.c.r.l c/o University of Calabria.

Acknowledgements

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