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A Technical Proposal for Active Sensing Monitoring Activities in the Area of Viggiano (South of Italy) and in Rural and Urban Areas: LIDAR/DIAL Techniques to Detect the Presence of Pollutants and to Control Biomass Combustion Products Like CO or PM_{10}

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Introduction

LIDAR (Light Detection And Ranging) and DIAL (Differential Absorption LIDAR) are techniques based on the principle of laser remote sensing that allow the atmosphere exploration at long distances for wide areas (till 10 Km^2)[1,2,3] in order to derive information, in real time, on the presence of anomalies in atmosphere; the type of pollutants and the concentrations are integrated over time or over space. The data acquired from these systems can also be used to develop and validate numerical models to predict the path of the contaminants and their dispersion and diffusion.

The Laboratory of Quantum Electronic and Plasma Physics, University of Rome Tor Vergata, has developed laser systems based on the LIDAR technique and the DIAL techniques. These systems are mobile, compact and transportable. The LIDAR system uses a Nd-YAG laser source[4] for the detection, and the DIAL system uses a TEA CO_2 laser source[5] for the identification.

These systems, based on the active laser sensing principles, have been applied to a wide range of remote sensing problems over the past 30 years:

1 Pollutants detection of minor compounds;[6,7,8,9,10,11]
2 Analysis of urban traffic emissions;[12,13] Detection of forest fires;[4,14] detection and identification of contaminants in open or closed environments to improve safety and security of public health worldwide.[15]

In order to monitor a very large area it is possible to deploy a low-cost detection system, composed of a LIDAR apparatus (already tested in an urban,[12] industrial and covered[14] areas) to detect pollutant and emission sources coupled with a DIAL based system, used as an identification system, able to investigate and to identify the nature of the pollutants emission.

In order to identify the substances in atmosphere it is fundamental that the transmission source is able to work in a large spectral range. A tuneable CO_2 laser is a good solution to identify the pollutants with man-made origin (VOCs-Volatile Organic Compounds; TICs-Toxic Industrial Compounds; TIMs: Toxic Industrial Materials). A lot of these pollutants have strong absorption bands in the "working" region of the CO_2 lasers, within the range 9-11 µm (Table 1).

In this spectral region, there is a wide atmospheric spectral window around 10 µm (Figure 1), which allows working without a consistent reduction of the signal due to the main components present in the atmosphere (CO_2; H_2O). Operation in the 10 µm region permits to operate in an eye safe region. An approach to the problem of environmental monitoring of accidental emissions of pollutants can be to detect before and to identify after the probable anomalies.

In the present paper, a review of the laser-based detection and identification will be presented and discussed together with a technical proposal to deploy these modern methodologies in the area of Viggiano or, in general, in urban and rural areas that can be affected by the diffusion of pollutants or by the dispersion of biomass combustion products like CO or PM_{10}.

The LIDAR System as Detection Technologies

The basic concept of the LIDAR has directly analogues with the more conventional and well known Radar. A pulse of energy is sent to the atmosphere and the backscattering light detected is related to some proprieties of the air volume involved.

LIDAR measurements have a broad application in the characterization of the atmosphere, ranging from the determination of properties of cloud particles[16] or aerosols[17] to the profiling of trace gas concentrations,[18] air temperature[19] and wind velocities.[20]

Two principal blocks characterize a LIDAR system: a transmitter and a receiver equipment.[1] The transmitter equipment is a laser source with high power pulses, whose characteristics are well known (high spatial/temporal coherence, low beam divergence, uniform polarization, brightness and mono-chromaticity of light, etc.).

The receiver is composed of a telescope with a specific detector placed at its focal plane. The backscattered radiation is collected by the telescope and converted by the detector to an electrical signal. An ADC (Analogic-Digital Convertor), connected to a computer, digitizes and stores the electrical signals as a function of space range and time of acquisition, in order to allow the computer to post-process and evaluate the elaborated data. The general scheme of a LIDAR system[21] is reported in Figure 2.

A technical improvement of the mini-LIDAR system was developed by Quantum Electronic and Plasma Physics research group (QEP). It is named "COLI" (COmpact LIDAR system).

Table 1. Absorption coefficient for several pollutants gas.

SUBSTANCES	*Line (on)*	α [atm cm]$^{-1}$	*Line (off)*	α [atm cm]$^{-1}$	β [km]$^{-1}$	Δα	Sensibility for 5.4 km SNR = 1,5
Ammonia (NH_3)	9R8	25.8	9R10	0.031	0.2280	25.5	18 ppb
Ozone (O_3)	9P14	12.07	9P24	0.07	0.2375	12	39 ppb
Water vapour (H_2O)	10R20	8.8E-4	10R18	1.1E-4	1.013	7.7E-4	614 ppm
Sulphur dioxide (SO_2)	9R26	0.105	9R28	0.092	0.2131	0.013	36 ppm
Ethylene (C_2H_4)	10P14	35.1	10P16	5.8	0.2653	29.3	16 ppb
Freon 11 ($CFCl_3$)	9R22	29.2	9P18	0.1	0.2114	29.1	16 ppb
Freon 12 ($CFCl_2$)	10P32	35.7	10P12	0.08	0.2267	35.6	13 ppb
Hydrazine (N_2H_4)	10P22	4.77	10P28	2.06	0.307	2.71	174 ppb
Ethylmercaptan (C_2H_5SH)	10R26	0.56	10P20	0.18	0.2279	0.38	1218 ppb
Vinylchloride (C_2H_3Cl)	10P22	8.8	9R18	0.05	0.2516	8.75	53 ppb
Trichloroethylene (C_2HCl_3)	10P20	12.6	10R20	0.04	0.2388	12.56	37 ppb
Ethyl Cloride (C_2H_5Cl)	10R16	3.3	10P20	0.06	0.2962	3.24	142 ppb
Ethylene chloride ($C_2H_4Cl_2$)	10P20	0.52	10R16	0.01	0.2388	0.51	907 ppb
Chloroprene (C_4H_5Cl)	10R18	9.15	9P22	0.1	0.2453	9.05	51 ppb
Perchloroethylene (C_2Cl_4)	10P34	4.9	10R24	0.1	0.2076	4.8	96 ppb
UDMH ($(CH_3)_2N_2H_2$)	10P320	2.22	10R10	0.18	0.2372	2.04	226 ppb
MMH ($CH_3N_2H_3$)	10R30	1.69	10R18	0.1	0.2491	1.38	335 ppb

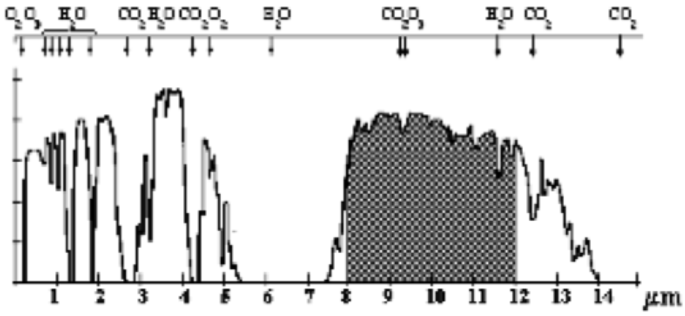

Figure 1. Atmospheric transmission.

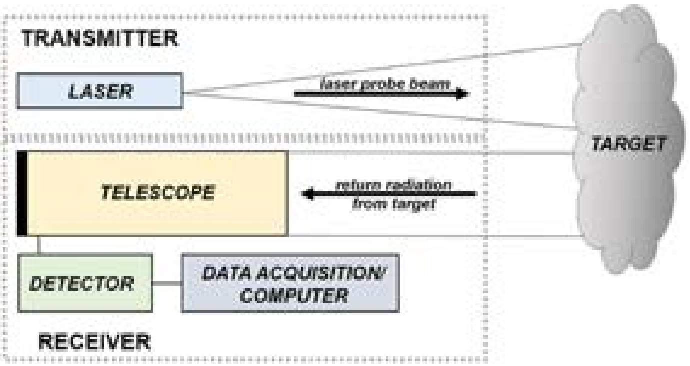

Figure 2. A conceptual scheme of a biaxial LIDAR system.

It consists of a compact, robust, stable, rugged Nd-YAG laser based on a monostatic configuration (the laser and telescope are located in the same place[22] with a biaxial arrangement.[23] In **Figure 3** the final configuration scheme is reported.

In the COLI system a commercial standard pulsed Nd:YAG Laser operating at 1064 nm in Q-switching mode is used as the transmitter. Back-scattered radiation is collected by a commercial telescope in a Cassegrain configuration. In order to suppress the background in daytime operation a 2 nm wide interference filter (Omega Optical Inc., USA), with a centre wavelength of 1064 nm, is used. Finally, the backscattered signal focalized

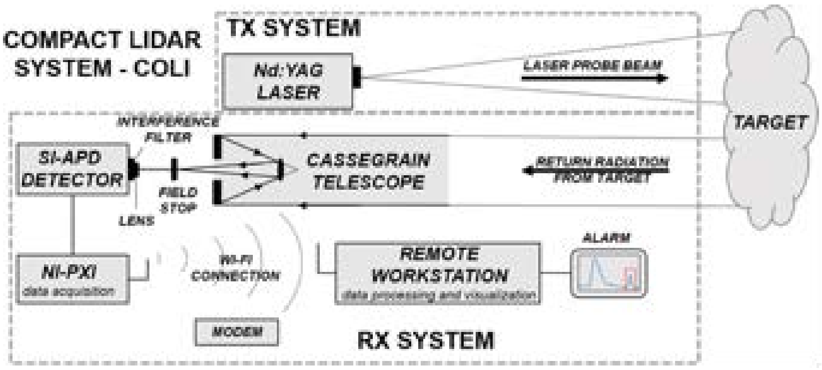

Figure 3. Technical scheme of COLI in its final configuration.

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by a telescope on the detector area of an IR-enhanced avalanche photodiode (APD) is acquired and stored by NI-PXI digitizer.

In Table 2 the principal characteristics of COLI system are reported.

The COLI system is able to scan the atmosphere in both vertical (elevation range from 0 to 90 degrees) and horizontal (azimuth range from 0 to 270 degrees) planes, using computer-controlled motors incorporated into the telescope mechanic. It is possible to design and implement a specific mount adapting it to particular applications. Therefore, once deployed in the field, COLI could be completely autonomous since it can be remotely controlled by our own software package written in LabVIEW and MATLAB, developed for mechanical handling of the laser-telescope block and of both the data acquisition and processing procedures.

Table 2. Specifications of COLI in its final configuration.

Laser – Big Sky CFR 400 by Quantel, France	
Pulse frequency	20 Hz
Energy pulse	400 mJ (max.)
Pulse time width	8 ns
Beam divergence	< 4.5 mrad (full angle)
Beam diameter	< 7 mm
Telescope – MAHK 130 by Ziel, Italy	
Nominal focal length	1300 mm
Primary mirror diameter	102 mm
f-number	f/12.7
Field of View	1.2 mrad
Detector – EG&G C30954/5E, URS Corporation, USA	
Responsivity	34 A/W (typ.)
Quantum efficiency	38% (typ.)
Diameter	1.5 mm
Response time	< 5 ns
Gain (M)	100 (typ.)
Digitizer – NI-PXI 5122, National Instruments, USA	
Resolution	14 bit
Sampling Rate	100 Ms/s
Bandwidth	100 Mhz

The pictures layout of the COLI system is reported in Figure 4.

The COLI system is able to acquire, store and elaborate the backscattered signals in order to evaluate particulate and aerosol in atmosphere.

The backscattered signals are described by the elastic LIDAR equation[1] here reported (0):

$$P_r(\lambda, R) = O(\lambda, R)\frac{A_r}{R^2}P_0(\lambda)\frac{c\tau}{2}\beta(\lambda, R)\exp -2\int_0^R \alpha(\lambda, R')\,dR' \quad (0)$$

where:

- $P_r(\lambda,R)$ is the backscattered power received at the specific laser operative wavelength λ from distance R,
- $O(\lambda,R)$ is the overlap and transmission factor determined by the geometric configurations of the receiver optics between the overlap of the emitted laser beam with the field of view of the receiver and the transmitter and receiver spectral efficiencies;
- A_r/R^2 is the acceptance solid angle of the receiver optics with collecting area A_r;
- $P_0(\lambda)$ is the output power of the laser pulse,
- c is the speed of light;
- τ is the laser pulse duration;

The last two terms describe the optical parameters of the laser atmosphere interactions.

The first backscattering coefficient – $\beta(\lambda,R)$ – is a measure of the scattering (in $m^{-1}sr^{-1}$) in the backwards direction (i.e. towards the incident direction, at a scattering angle of 180°) of the one of laser-atmosphere interaction with constituents as aerosol, particles and molecules.

The second one is the extinction coefficient – $\alpha(\lambda,R)$ – that consists of a measure of the attenuation of the laser pulse (in m^{-1}) propagating through the atmosphere, due to the scattering and absorption by aerosol particles and molecules present in the atmosphere.

An example from a measurement campaign with COLI is shown below. The measurement of aerosol and particulate dispersed in a suburban area of Crotone in the south of Italy[12] vs time is reported in Figure 5. During this campaign, a backscattering value related to urban traffic was measured, in order to measure particulate increments due to vehicles. A strong correlation of PM (Particulate

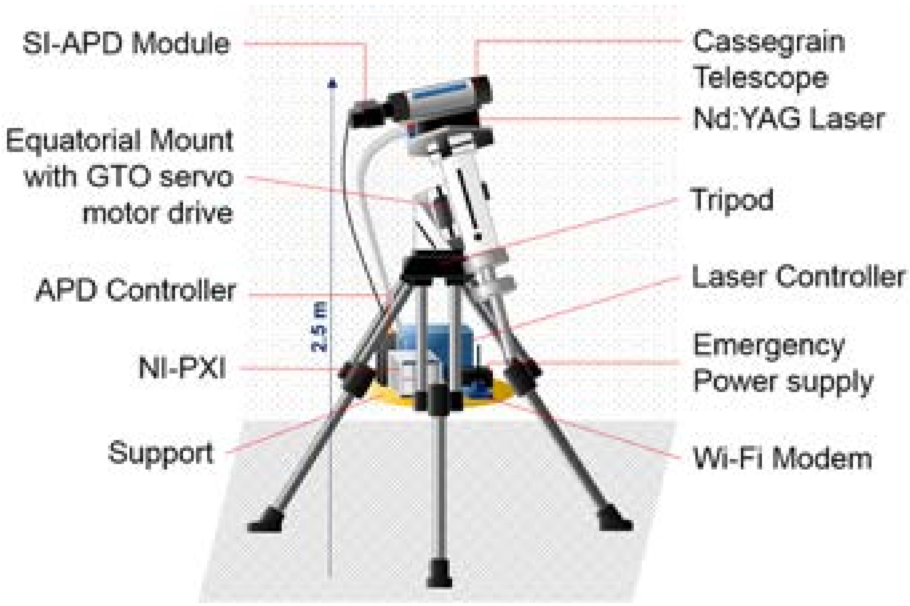

Figure 4. Picture of COLI system.

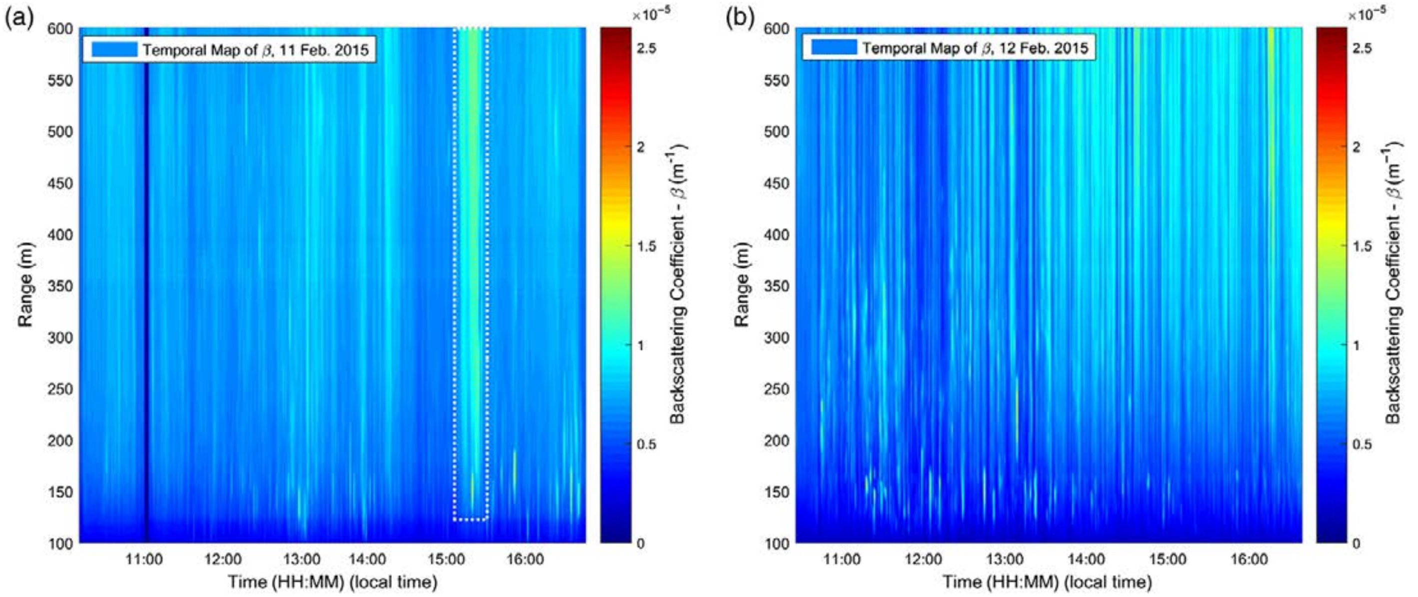

Figure 5. In the plot is shown the temporal maps of backscattering coefficients. An increment of pollutants source is strictly related to human activity in the relation to closure of offices or schools (Source[12]).

Materials) concentration levels between LIDAR measurements and a conventional point measurement station was observed. In the same work[12] the daily variation of the particulate in urban areas is shown by means of temporal maps; these measurements made in real time represents a strong innovation if compared with the classical measuring techniques (based on punctual measurement systems), able to monitor only a small volume of few cubic centimeter around the detector. The analysis of backscattered signals versus time was performed in order to investigate and detect potential aerosol sources due to daily variation of vehicle emissions in high-speed congested roads. With this aim it is possible to observe that an interesting increase of particulate concentration amount is strongly correlated to the human activity (**Figure 5**).

Finally, the experimental campaign has demonstrated the capability of LIDAR systems and in particular of the mini-LIDAR COLI, to produce real measurements of particle diffusion in the atmosphere.

The DIAL System as Identification Technologies

The theoretical aspects of laser remote sensing capabilities have been extensively investigated in the literature. It is well known that the DIAL technique[24] consists mainly of sending through the atmosphere two different laser pulses to analyse the backscattered signals. The selected wavelengths, λ_{on} and λ_{off} that are respectively "resonance" and "off resonance", which allow an absorption transition of molecular species that are measured with this technique. The concentration of the absorbing species is obtained from the differential absorption of the backscattered laser radiation at λ_{on} compared to the one at $\lambda_{off.}$[25] λ_{on} is strongly absorbed from molecules under study, while λ_{off} is not absorbed. It is possible to write the LIDAR equation for each wavelength used and, performing the ratio of the two equations, it is possible to obtain a concentration profile N(R) of molecules investigated:

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

where P is the power received by detector, $\Delta\sigma(\lambda_{off} - \lambda_{on})$ the differential absorption coefficient, $\beta(\lambda_{on/off}, R)$ the backscattering coefficient and $k_0(\lambda_{on/off}, R)$ the total atmospheric extinction exclusive of the quality relating to the absorption due to the substance that is being studied. Both backscattering and extinction coefficients depend on the wavelength of the laser radiation and, if the difference between λ_{on} and λ_{off} is small, they can be considered the same; in this case the concentration of the investigated substances can be represented as a first order function of the range. The DIAL equation, under the previous conditions, becomes:

$$N(R) = \frac{1}{2\ \sigma\left(\lambda_{on} - \lambda_{off}\right)} \cdot \frac{d}{dr} \ln \frac{P\left(\lambda_{off}, R\right)}{P\left(\lambda_{on}, R\right)} \qquad (2)$$

where N(R) is the concentration profile at first order approximation.

A preliminary field test of a DIAL system can be prepared using an integrated technique that guarantees a high sensitivity even in presence of substances at low concentrations. This technique uses the same method of DIAL but a topographic target has been employed to scatter the radiation in the backward direction. It allows an improvement of the performance in term of the sensibility in the detection. The backscattered signal is much larger than the atmospheric backscattered one even if this is jeopardized the spatial resolution. It is possible, starting from eq. 2, if obtain the average concentration as:

$$\overline{N}(R) = \frac{1}{R} \int_{0}^{R_t} N(R')\, dR' \qquad (3)$$

then:

$$N(R) = \frac{1}{R} \ln \frac{P_{off}}{P_{on}} \qquad (4)$$

where ΔR is the distance between the target and the DIAL station and σ_o is the differential absorption cross-section while P_{on}

and P_{off} are the power received at the two different wavelengths at target distances.

The QEP group is working to design, develop and improve DIAL systems based on a CO$_2$ laser for application in environmental monitoring, monitor of diffusion and emission sources from industrial area and also for safety and security applications.[21,28] All these systems are able to emit about 60 laser lines in the spectral range between 9÷11µm with a typical giant pulse width of the order of 100 ns that permit to obtain, at a first approximation, a spatial resolution of about 15 m. In fact, the CO$_2$ laser pulse, is constituted from a giant pulse, to some 100 ns in duration, following by a long tail (3 – 4 µs). Studies are in progress to clip the long tail.[26,27]

Table 3. Principal characteristics of mini-DIAL system.

Transmitter TEA CO$_2$ laser	Output energy	50 mJ
	Pulse width	100 nsec
	Beam divergence	1 mrad
	Spectral range	9 ÷ 11 mm
Receiver	Telescope type	Newtonian
	Primary diameter	200 mm
	ZnSe lens focal length	7 mm
	Focal length	1000 mm
	F.O.V.	8,7 mrad
Kolmar KMPV11 model	Detector type	HgCdTe
	Detector sensitivity	D* 1,9 (1010) [ones]
	Detector size	1 mm2

The last important goal was the realization of a compact DIAL system. The mini TEA CO$_2$ laser distributed by Edinburg Instruments is used for the development of a mini-DIAL based on a CO$_2$ laser (**Figure 6**). The energy of the transmitter system is maximum 50 mJ. The receiver system instead is based on a Newtonian telescope that focalised the backscattered radiation of atmosphere on the detector area. The detector is an HgCdTe type with 8 ns of rise time. The transmitter and receiver systems can be located on the mechanical support in order to allow the scanning of an entirely hemisphere. The principal characteristic of a mini-DIAL system developed is reported in **Table 3**. The characteristics of transmitter and receiver systems guarantee the capability of the mini DIAL system to measure concentration profiles up to a maximum range of 1.5 km covering a maximum area of about 10 km^2.

In case of chemical release in atmosphere it is essential to deploy also systems able to identify the chemical substances released. In case of organic compounds unfortunately the absorption spectra show different overlaps causing interference phenomena that makes difficult the exact identification of substances using only two wavelengths.[1,29] The DIAL multi-wavelengths approach can solve this problem for organic compounds, limiting the false alarms. The preliminary tests, on the classification of several absorption spectra of some organic compounds, give interesting results.[28] The sensitivity of DIAL methodologies is one of the most interesting information and it is expressed as the minimum concentration of the atmospheric trace gases that can be distinguished from the natural background or noise.[29] The minimum detectable concentration depends on the range and spatial resolutions and can be defined according to two different limits:[25,30]

1. A limitation is essentially due to the noise of the detector at long distances. In fact, the measurements error depends on the SNR ratio of the LIDAR backscatter that is a function of the range;[25]
2. A limitation may occur due to the inability of the DIAL system to distinguish, at short distances, between the fractional change in the LIDAR return signal ($\Delta P/P$) where ΔP is the minimum variation between P_{on} and P_{off} backscattering signals.

The evaluation can be done according with these last two limitations evaluating the following the expression (4)[25] able to describe these limits, as function of the systems characteristics and absorption coefficients of the substances, that must to be identified.[25] The evaluation of sensitivity of our mini-DIAL system for the measurement of Benzene and Sulphur Dioxide is shown

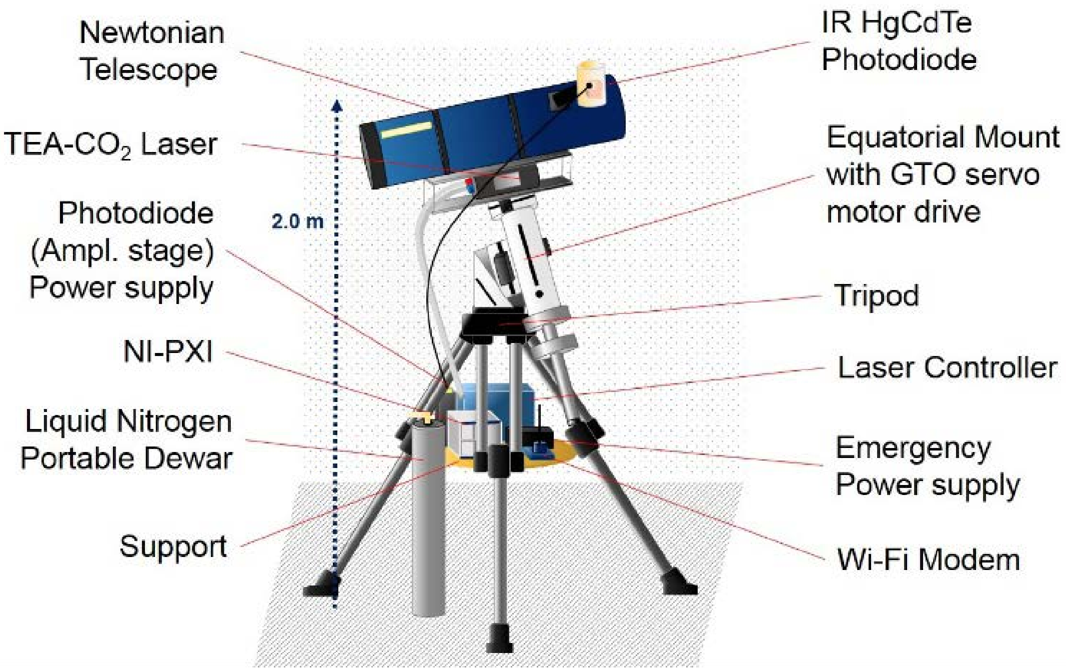

Figure 6. Picture layout of mini-DIAL system developed at University of Rome Tor Vergata.

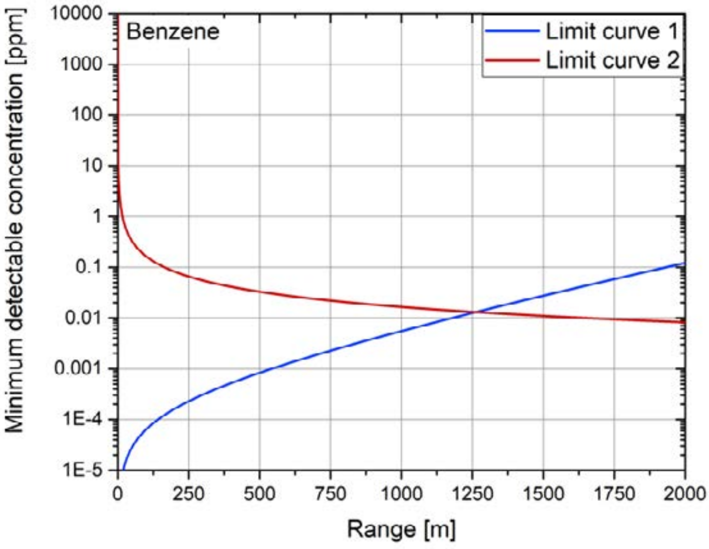

Figure 7. Benzene concentration limits vs range.

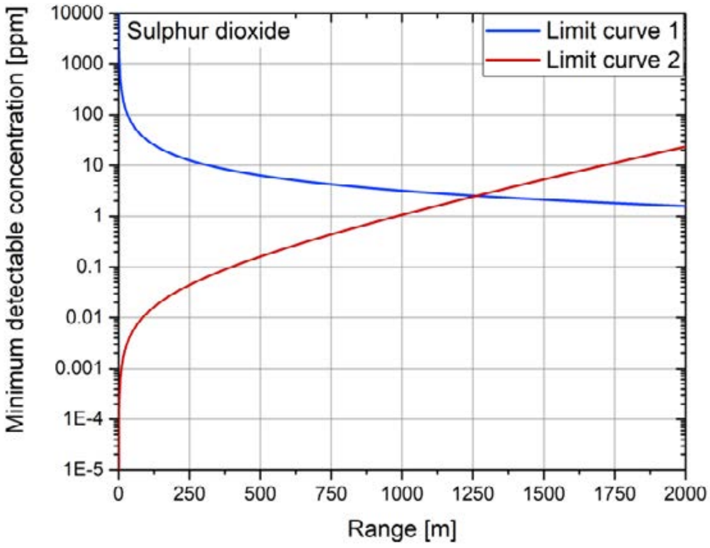

Figure 8. Sulphur dioxide concentration limits vs range.

in Figures 7 and 8. First of all, it is necessary to identify the *on* and *off* wavelengths for the two substances and their respectively absorption coefficients. An evaluation of the mini-DIAL system sensitivity for the Sulphur dioxide and Benzene measurements can be calculated starting from the absorption coefficients and laser wavelengths that can be found in literature or alternatively with measurements done using absorption cells. In particular, using the data of Sulphur dioxide[31] and Benzene obtained from experimental measurements using an absorption cell,[28] it is possible to evaluate the capability of detection versus the range of our system.

Conclusion

The sensitivity of the measurements of described systems, their capabilities to be transported and to monitor large areas (till 10 km^2), the possibility to detect anomalies giving a first alarm and to identify the substances giving map of concentration in space and time make the LIDAR and DIAL system developed by the University of Rome Tor Vergata (https://www.qepresearch.it/research-activities/laser-systems/) an adequate option to monitor area like Viggiano or, in general, urban and rural areas that can be affected by the diffusion of pollutants or by the dispersion of biomass combustion products like CO or PM$_{10}$. The future development can be a network of LIDAR and DIAL systems to cover hundreds of square kilometres of territory in real time.

Glossary

- *APD: Avalanche Photodiode*
- *CFR laser: Compact Pulsed Nd:YAG Lasers*
- *CO: Carbon monoxide*
- *COLI: Compact LIDAR*
- *DIAL: Differential Absorption LIDAR*
- *F.O.V: Field of View*
- *IR: Infra Red*
- *LabVIEW: Laboratory Virtual Instrumentation Engineering Workbench*
- *LIDAR: Light Detection and Ranging*
- *MATLAB: Matrix Laboratory*
- *Nd-YAG: Neodymium-Doped Yttrium Aluminum Garnet*
- *NI-PXI: National Instruments (NI) PCI eXtensions for Instrumentation (PXI)*
- *PM$_{10}$: Particulate Matter with a Diameter Equal or Inferior to 10 µm*
- *TEA CO2 laser: Transversely-Excited Atmospheric-Pressure (TEA) CO2 (Carbon dioxide) Laser*
- *TICs: Toxic Industrial Compounds*
- *TIMs: Toxic Industrial Materials*
- *VOCs: Volatile Organic Compounds*

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