Optica Applicata, Vol. XXIX, No. 4, 1999

Monitoring of an industrial area in southern Italy by using a multiwavelength lidar

P.F. Ambrico*, A. Amodeo**, S., Amoruso*, M. Armenante***, A. Boselli**, M. Pandolfi**, G. Pappalardo**, N. Spinelli****, R. Velotta***

Istituto Nazionale per la Fisica della Materia (INFM), Unità di Napoli, Pad. 20 Mostra d'Oltremare I-80125 Napoli, Italy, and

*Dipartimento di Ingegneria e Fisica dell'Ambiente (DIFA), Università della Basilicata, via della Tecnica 3, I-85100 Potenza, Italy.

**Istituto di Metodologie Avanzate di Analisi Ambientale (IMAAA), CNR-Area della Ricerca di Potenza — Tito Scalo, I-85050, Italy.

***Istituto Nazionale di Fisica Nucleare (INFN), Sezione di Napoli, Complesso Universitario di Monte S. Angelo, via Cintia, I-80126 Napoli, Italy.

****Dipartimento di Scienze Fisiche (DSF), Università di Napoli, Complesso Universitario di Monte S. Angelo, via Cinita, I-80126 Napoli, Italy.

A multiwavelength lidar system has been developed for air monitoring of an industrial area in southern Italy. The system based on two dye lasers pumped by a Nd:YAG allows us to perform differential absorption lidar measurements in the UV spectral region together with measurements of water vapour, atmospheric transmissivity and aerosol load. The system has been designed to deliver simultaneously in the atmosphere the Nd:YAG fundamental and its harmonics (II and III) as well as the output beams of two dye lasers. The return signals, collected by means of two Newtonian telescopes, make it possible to retrieve information on atmospheric aerosols, water vapour and pollutans concentration, density and transmissivity profiles. In this paper, we report the preliminary results obtained in a measurement campaign performed in the periods of November–December'98, and June–July'99.

1. Introduction

Among optical remote sensing techniques, the lidar technique represents the most powerful tool for studying processes occurring in the atmosphere, because of its capability to provide information on several atmospheric parameters with very high spatial and temporal resolution. Lidar techniques have been widely used to monitor stratospheric and topospheric aerosols [1], [2], water vapour [3] and other main pollutants concentration [4], atmospheric transmissivity [5], density and temperature [6], [7]. In particular, elastic (Rayleigh and Mie) and Raman backscattering in the visible and ultraviolet (UV) regions allow a comprehensive investigation of aerosol profiles. Rayleigh and N_2 Raman scattering are used to obtain information on the atmospheric number density profile and a subsequent application of the hydrostatic equation allows the determination of atmospheric temperature profiles.

Though a large number of atmospheric parameters can be currently monitored separately, correlation between parameters is still a complex task. For example, the correlation between atmospheric temperature, water vapour content and pollutants density appears important in order to monitor the evolution of reacting chemical species. Therefore, it is of fundamental importance to perform simultaneous measurements of several atmospheric parameters with high spatial and time resolution.

In this paper, we report some of the results obtained during a field campaign in two different industrial areas located in southern Italy: San Nicola di Melfi (41°N, 15°39′E – 200 m a.s.l.) and Tito Scalo (40°36′N, 15°44′E – 820 m a.s.l.). The former has undergone a large industrialisation in the last decade. Moreover, since an incinerator has been installed in this area, an investigation of the air quality before it becomes operative is of particular interest. The latter has been selected as a test site to obtain some reference results and is characterised by a low industrial density. Measurements of aerosol, water vapour, transmissivity profiles, and major pollutants (O_3, NO_2, SO_2) have been performed in daytime and night-time conditions.

2. Experimental set-up

The experimental apparatus is shown in Fig. 1. It is based on a Nd:YAG laser delivering simultaneously the fundamental (1064 nm), the second (532 nm) and the third (355 nm) harmonic, with a maximum repetition rate of 20 Hz. Part of the second and third harmonics ($E_{355} \leqslant 10\%$; $E_{532} \leqslant 10\%$), taken by means of beam splitters, is mixed with the fundamental and the three laser beams are delivered along the same optical path. In order to avoid damage to the delivering mirrors and to reduce the beam divergence, the laser beams are expanded by a factor of 5 and deflected into the atmosphere coaxially with respect to the telescope. The ramaining part of the second and third harmonics is used to pump two dye lasers.

The first dye laser is pumped by Nd:YAG third harmonic in order to emit in the 400 nm spectral region, for NO_2 DIAL measurements. This dye laser is equipped with a dual wavelengths option which allows the laser to alternatively oscillate at two close wavelength (446.8 nm and 448.1 nm). In such a way DIAL on-line (λ_{ON}) and off-line (λ_{OFF}) beams are alternatively sent into the atmosphere at a repetition rate of 10 Hz. The second dye laser is equipped with a second harmonic generation (SHG) crystal which covers the UV region for the monitoring of SO_2 and O_3. The laser is alternatively tuned to two close wavelengths corresponding to 282.4 nm and 286.3 nm for O_3 and to 286.9 nm and 286.3 nm for SO_2 concentration measurements, respectively.

The two dye laser beams are sent in the atmosphere by mirrors located on the secondary mirror of the second telescope in a monostatic configuration. The tuning of dye lasers is controlled by a wavelength meter (Burleigh WA-4500, wavelength accuracy ± 0.02 cm^{-1}, spectral resolution 0.05 cm^{-1}). This is sensitive to radiation in the wavelength range 400 – 1100 nm, thus for the dye laser emitting in the UV the dye fundamental output wavelengths have been measured.

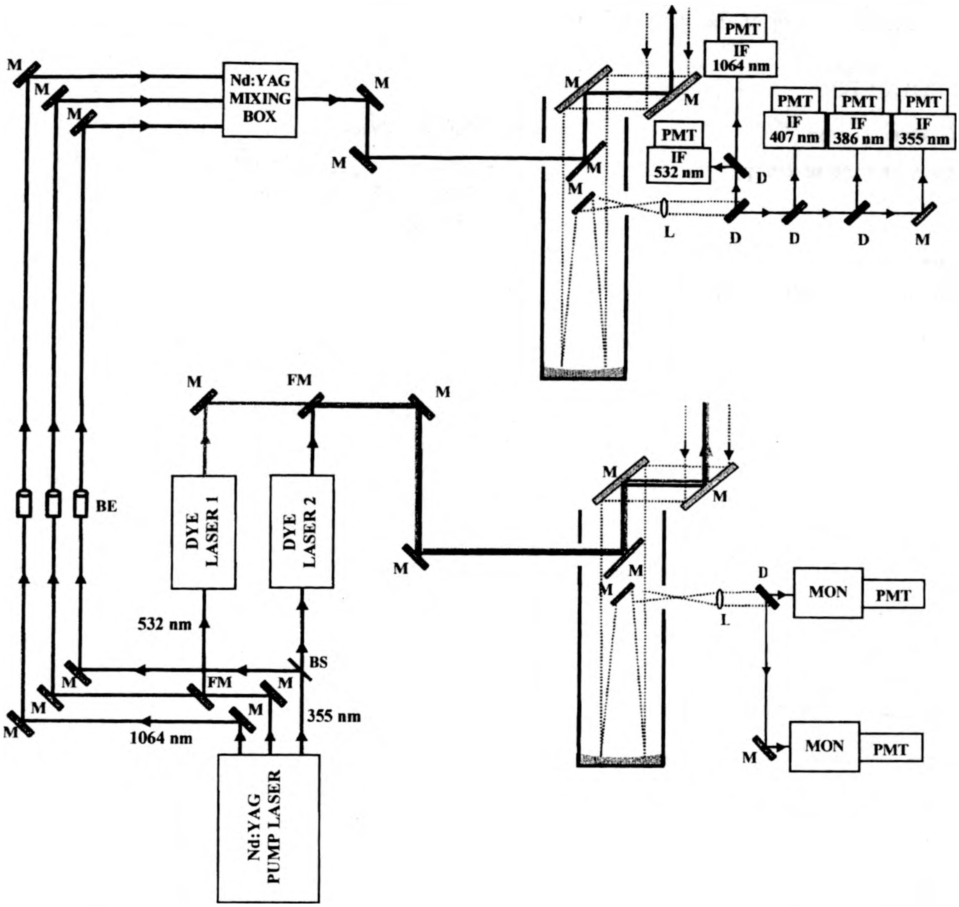

Fig. 1. Experimental setup. BE – beam expander, D – dichroic mirror, FM – flipper mirror, IF – interferential filter, MON – monochromator, M – mirror, BS – beam splitter, L – lens.

The receivers consist of two vertically pointing telescopes in Newtonian configuration, with a 0.3 m diameter primary mirror and a focal length of 1.2 m. The telescopes are both equipped with periscopes enabling a 3D monitoring of the atmosphere. The field of view of each telescope is selected by means of irises. One telescope is used to collect the elastically backscattered radiation at 1064, 532 and 355 nm, as well as the Raman return radiation of the water vapour (407 nm) and nitrogen (386.6 nm) excited at 355 nm. The other telescope is intended for the DIAL measurements.

The spectral selection is accomplished through interference filters and monochromators. The interference filters are used to select the backscattered radiation of elastic and Raman shifted signals, whereas the monochromators are used to select the λ_{ON} and λ_{OFF} in the DIAL measurements.

Photomultiplier tubes are used as detectors. All detectors are cooled down to -30 °C in order to reduce dark current. Signals from photomultipliers are sampled

Table 1. Characteristics of the lidar system.

LASER SOURCES	
Nd:YAG laser punmp	**Continuum NY60**
Wavelength and pulse energy	$\lambda = 1064$ nm, $E_{max} = 600$ mJ $\lambda = 532$ nm, $E_{max} = 300$ mJ $\lambda = 355$ nm, $E_{max} = 170$ mJ
Pulse duration	5–7 ns
Maximum pulse repetition rate	20 Hz
Beam divergence	<0.5 mrad
Dye lasers	
	Lambda Physik LPD6000
Dye	Rhodamine 6G
Line width	$\leqslant 0.08$ cm^{-1}
Divergence	$\leqslant 0.5$ mrad
Pulse width	<pump laser
	Continuum ND60
Dye	Coumarin 120
Line width	$\leqslant 0.08$ cm^{-1}
Divergence	$\leqslant 0.5$ mrad
Pulse width	<pump laser
Equipped with a dual wavelength device	
RECEIVER	
Newtonian telescope	
Diameter of primary mirror	0.3 m
Focal length	1.2 m
Field of view	0.2–1 mrad
Interference filters	
Bandwidth	2.0 nm
Out-of-band rejection	10^{-8}
Single-grating monochromator	
Bandwidth	4.0 nm
Grating efficiency	30%
Out-of-band rejection	10^{-6}
Used in the DIAL channel	
Detectors	
Photomultipliers enclosed in cooled housing (−30 °C)	

both in analog and photon counting mode. Analog signals are acquired by means of a digital oscilloscope (HP54502A, 400 MHz) or transient digitizer (IMTEC, T3012, 60 MHz, 12 bit). The photon counting chain is based on fast discriminators (300 MHz) and Multi-channel Scaler boards (EG&G TurboMCS, dwell time – 20 ns). Characteristics of the lidar system are presented in Tab. 1.

3. Data analysis and results

In this section, we report the preliminary results obtained during a field campaign performed in the industrial area of San Nicola di Melfi in the periods of November–December '98 and June–July '99. Moreover, some results obtained in the low industrial density area of Tito Scalo are reported as reference.

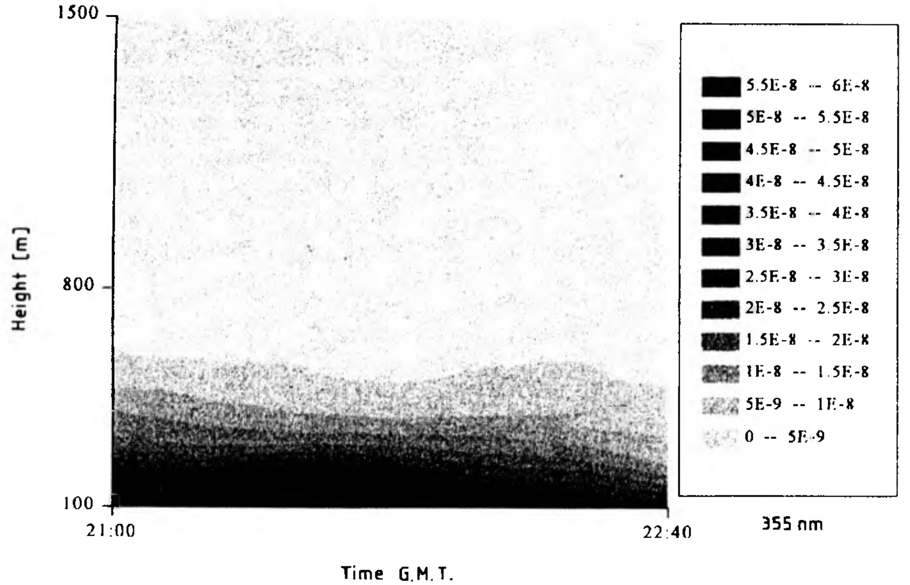

Fig. 2. Temporal evolution of the backscattering coefficient at 355 nm (Tito Scalo, February 11, 1998).

The map presented in Fig. 2 shows the temporal evolution of the backscattering coefficient at 355 nm, on February 11, 1998 in Tito Scalo. The backscattering coefficient has been obtained by means of an iterative procedure, as described in [8]. The backscattering coefficient gives information on the density, mass and dimension of the aerosols that can be retrieved by means of inversion techniques [9]. In the map, 50 different aerosol backscattering coefficient profiles are reported, each one obtained through integration over 6000 laser shots. The measurements show a stratified structure around 650 m above the lidar station and a clear air above this height. The maximum value of the backscattering coefficient is around $6\times10^{-6}\,m^{-1}\,sr^{-1}$.

In previous works we showed that Raman and DIAL lidar techniques are almost equivalent for water vapour measurements in the night-time conditions [10], [11] observing quite a good agreement between them and with balloon-borne radiosondes. In the measurement campaign performed in the industrial area of San Nicola di Melfi, the water vapour content has been monitored by using the Raman technique. In particular, in such a campaign, the water vapour vertical profiles have been collected by means of the simultaneous collection of the nitrogen and H_2O Raman signals. Moreover, this allows us to obtain also information on the atmospheric transmissivity [5], a parameter that gives an indication on the overall quality of the atmosphere.

From the water vapour and nitrogen Raman backscattered signals the water vapour mixing ratio w as a function of height is directly obtained by the following relation [12]:

$$w = k \frac{P_{H_2O}(\lambda, z)}{P_{N_2}(\lambda, z)} \tag{1}$$

where: k is the ratio between the Raman scattering cross-sections of H_2O and N_2, and $P_{H_2O}(\lambda, z)$ and $P_{N_2}(\lambda, z)$ are the corresponding intensities of signals. Moreover, the transmissivity profile is evaluated by using the nitrogen Raman measurements following the procedure described in [5].

As an example, in Fig. 3, a typical water vapour profile obtained in San Nicola di Melfi around 20:25 local time, on October 10, 1998, is shown. The vertical profile has been acquired with a vertical resolution of 3 m, that has been reduced to 30 m in the analysis in order to reduce the statistical fluctuations. Signals have been averaged over 6000 laser shots at a pulse repetition rate of 20 Hz, corresponding to a time resolution of 5 minutes. Moreover, a ground humidity sonde has been acquired in order to calibrate the measurement, and the corresponding data is shown as a full square.

From the nitrogen Raman measurements the atmospheric transmissivity profiles have been obtained. The data show a systematic lower value of the transmissivity in the industrial area of San Nicola di Melfi with respect to the low density indus-

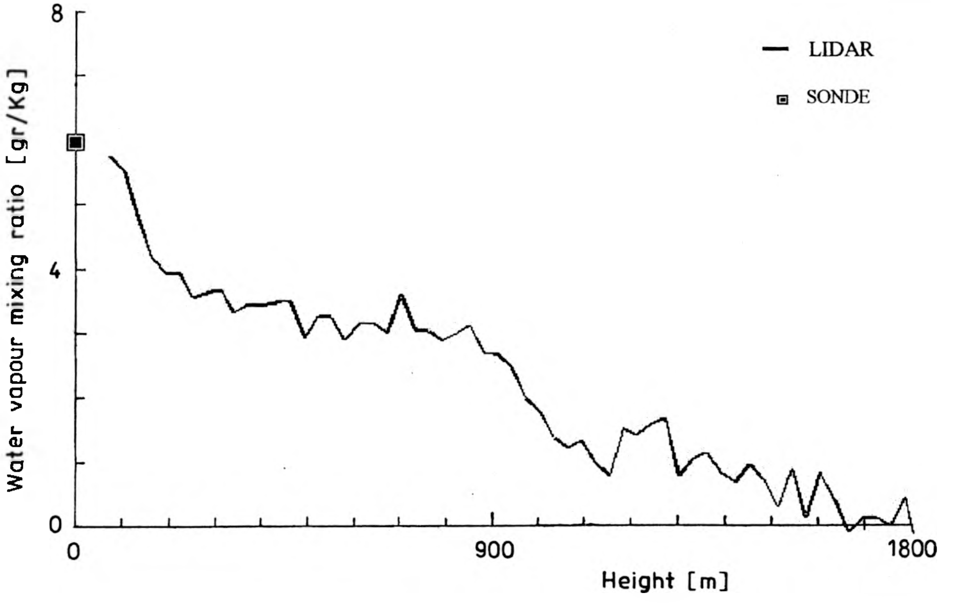

Fig. 3. Water vapour profile (San Nicola di Melfi, 20:25 local time, October 10, 1998).

trial area of San Nicola di Melfi with respect to the low density industrial area of Tito Scalo. This suggests that a much higher dust content is present in the atmosphere of the industrial area of San Nicola di Melfi with respect to Tito Scalo.

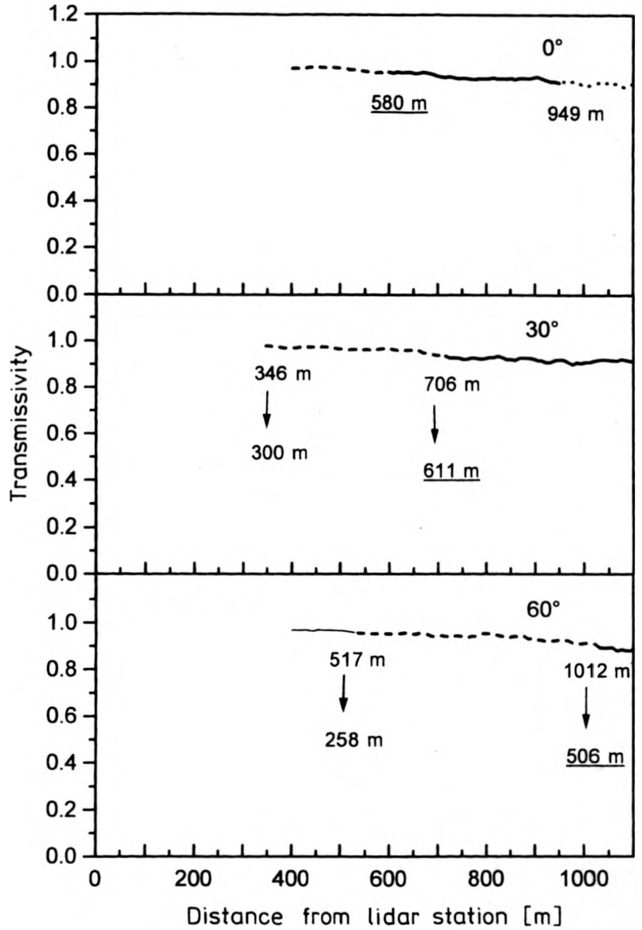

Fig. 4. Transmissivity profiles at three different zenithal angles. The same range in the vertical direction has been indicated by the same line type. The arrows indicate the corresponding height along the vertical direction.

Transmissivity measurements at different zenithal angles (see Fig. 4) have also been performed, showing the existence of a thicker layer of air between 600 and 950 m above the lidar station. The changes in the slope of the transmissivity profile in the ranges equivalent to the height of 600 – 950 m have been observed due to the presence of regions with different aerosol load. Measurements of the aerosol backscattering coefficient, reported below, seem to validate this interpretation of the transmissivity results.

Aerosol backscattering measurements have been performed in order to follow the time evolution of the aerosol load. Moreover, to obtain information about the dimension of the aerosol, two simultaneous measurements of wavelength backscattering coefficients have been performed at 350 nm and 532 nm. In particular, Fig. 5 shows the temporal evolution of the aerosol backscattering coefficient around sunset on November 10, 1998. The temporal resolution is 2 min 30 s, each profile is averaged over 3000 laser shots, at a repetition rate of 20 Hz. The aerosol backscattering coefficients at the two different wavelengths show the same temporal evolution, with maximum values of $6\times10^{-6}\,m^{-1}\,sr^{-1}$ at 355 nm, and $2\times10^{-6}\,m^{-1}\,sr^{-1}$ at 532 nm between 600 and 900 m after sunset.

It is clear that, due to the atmospheric turbulence, the atmospheric aerosol load seems to be mixed before sunset, and after sunset the aerosol load seems to increase between 600 and 900 m. Though the values at 355 nm are in agreement with the ones measured in the test site of Tito Scalo, it is evident that there is a different distribution of the aerosol load that seems to be stratified in San Nicola di Melfi between 600 and 900 m of height.

From the measurement of the backscattering coefficient at two different wavelengths, space and time resolved information on the aerosol size can be obtained [13] if a power law in the dependence of β on λ is assumed as $\beta_p \propto \lambda^{-\delta}$. In this hypothesis the Angström coefficient δ can be calculated by means of the following relation:

$$\delta(z) = \frac{\ln(\beta_p(\lambda_1,z)/\beta_p(\lambda_2,z))}{\ln(\lambda_2/\lambda_1)} \tag{2}$$

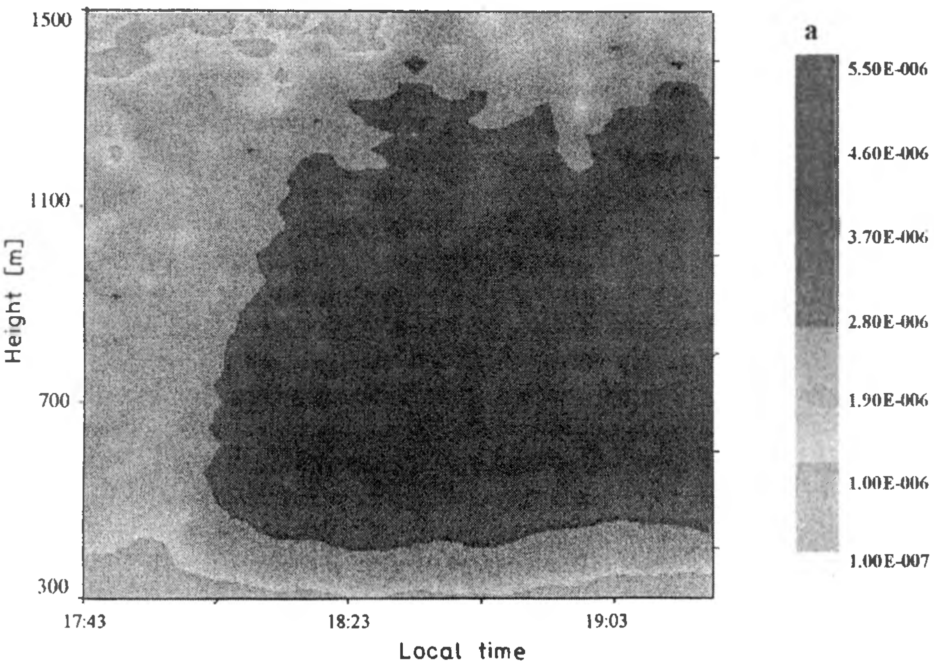

Fig. 5a

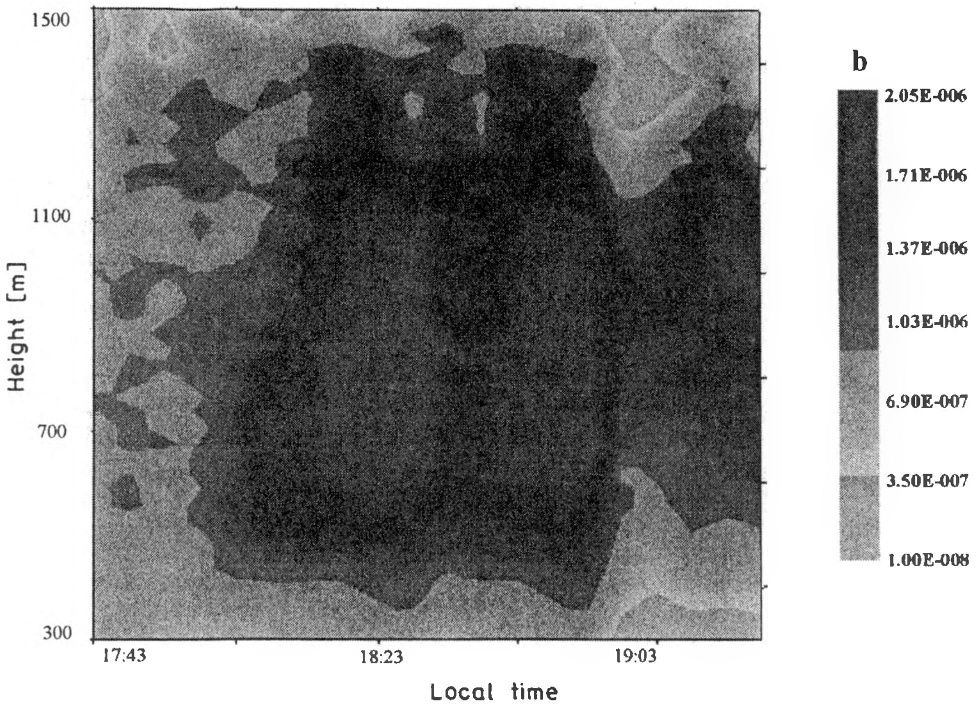

Fig. 5 b

Fig. 5. Time evolution of the aerosol backscattering coefficient obtained in San Nicola di Melfi, on November 10, 1998 at: 355 nm (a) and 532 nm (b).

where $\beta_p(\lambda_2, z)$ and $\beta_p(\lambda_1, z)$ are the aerosol backscattering coefficients at λ_2 and λ_1, respectively, with $\lambda_2 < \lambda_1$. Figure 6 shows the time evolution of the Angström coefficient, which reveals the presence of sub-micrometric particulate after 19:00 local time.

In order to verify that the aerosol detected in the atmosphere around the industrial plant is somehow due to the industrial activity, measurements of the stack emission plume have been performed at three different wavelengths (355 nm, 532 nm, 1064 nm), as shown in Fig. 7. Each profile has been acquired over 3000 laser shots and with spatial resolution of 3 m. The optical depth (τ_A) and the integrated backscattered coefficient of the plume has been evaluated, and the results are reported in Tab. 2. The wavelength dependence shows that sub-micrometric particles are emitted from the stack.

In the industrial area of San Nicola di Melfi a measurement campaign, settled at the end of July 1999, has also been performed to monitor NO_2, SO_2 and O_3 concentrations. Since the measurements are very recent, data analysis is currently in progress. In Figure 8, we give a preliminary result relative to O_3 concentration, as measured in the late morning (11:10 and 11:15 local time) of July 16, 1999. The profiles have been averaged over 6400 laser shots for each wavelength and acquired with a repetition rate of 20 Hz and at a sampling time of 40 ns, corresponding

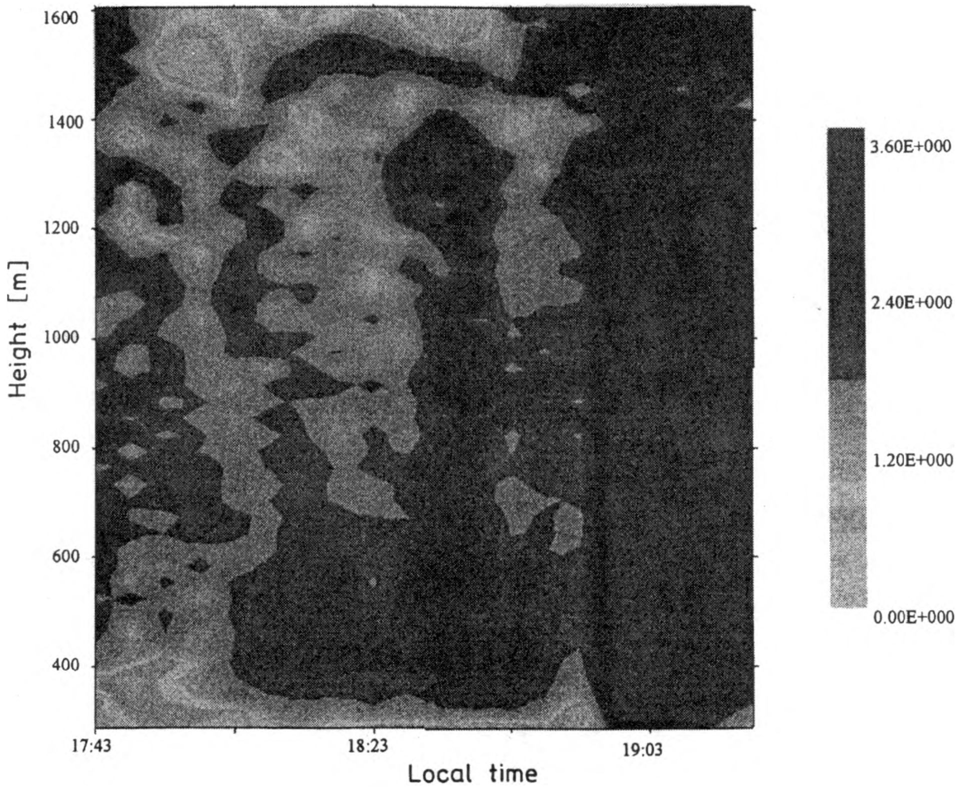

Fig. 6. Time evolution of the Angström coefficient (San Nicola di Melfi, November 10, 1998).

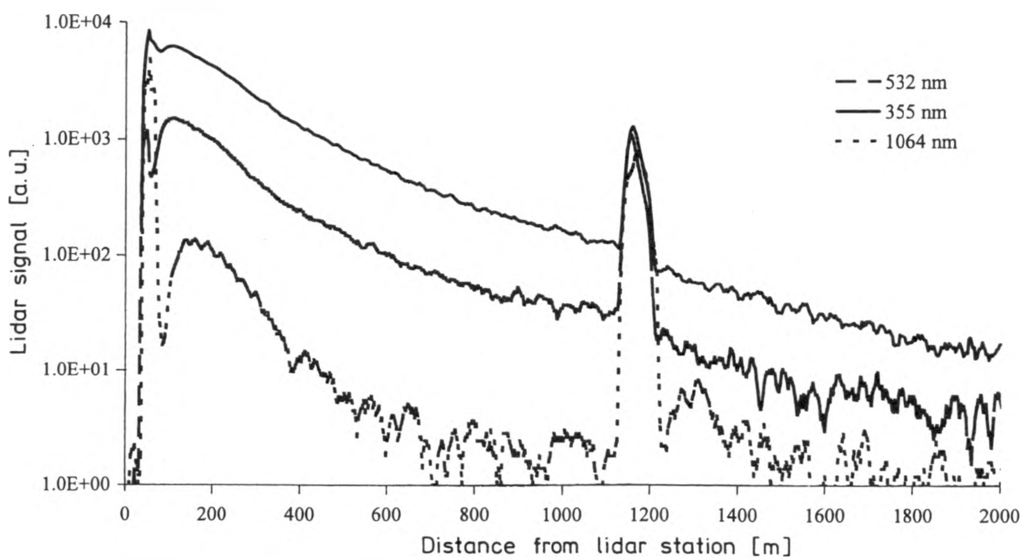

Fig. 7. Lidar echoes from the stack emission plume at three different laser wavelengths.

Table 2. Plume optical depth τ_A and integrated backscattering coefficient *BI*.

Wavelength [nm]	τ_A	BI [sr^{-1}]
355	0.22	3.64×10^{-3}
532	0.18	2.7×10^{-3}
1064	0.11	2.52×10^{-3}

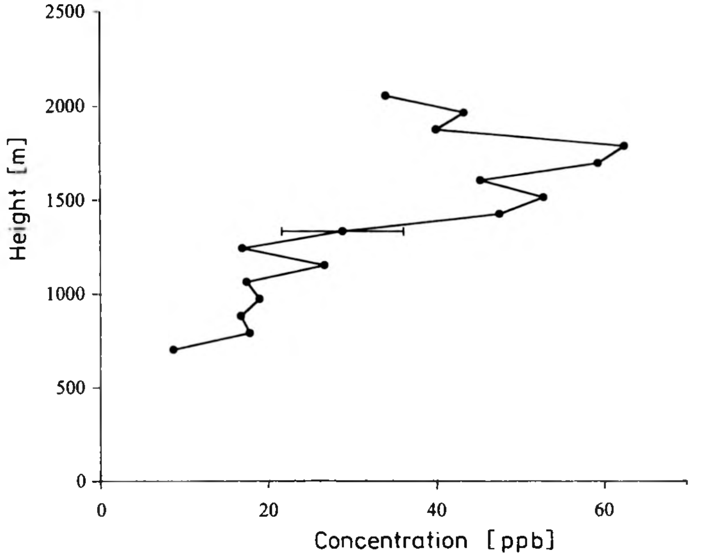

Fig. 8. Ozone vertical profile (San Nicola di Melfi, July 16, 1999).

to a spatial resolution of 6 m. Moreover, measurements of the background have also been performed. The background has been subtracted from the signal and the resulting waveform has been averaged to reduce the statistical fluctuations of the signal, obtaining a spectral resolution of 30 m. Then a smoothing procedure has been applied, as proposed by CHUDZYŃSKI *et al.* [14], obtaining a final spatial resolution of 90 m. The ratio of the on- and off-line background free signals has again undergone a smoothing procedure performed with current average technique. The concentration of O_3 is almost constant below 1.3 km and increases between 1.3 km and 1.7 km; it shows a maximum value of 70 ppb around 1.7 km of height.

4. Conclusions

A multiparametric lidar able to perform simultaneously DIAL, Raman and elastic measurements in the lower troposphere (up to 2 km of height) has been developed. The laser sources and the employment of two telescopes enable the system to measure simultaneously different atmospheric parameters. The system has been used to perform field campaigns in two different industrial areas located in southern Italy.

Results of the campaigns, in terms of water vapour, aerosol load an atmospheric transmissivity profiles have been reported. The measurements performed in the industrial area of San Nicola di Melfi have evidenced the presence of an aerosol layer between 550 and 900 m of height. The presence of the layer has been confirmed by aerosol backscattering coefficient measurements. A clear correlation with the industrial plant emission has been observed during measurements of the integrated backscattering coefficient of the plume emitted by a stack of the industrial plant. From these measurements, information about the aerosol dimensions has also been obtained. Moreover, the lower values of the atmospheric transmissivity observed in San Nicola di Melfi evidence a much higher dust content with respect to Tito Scalo, probably due to its larger industrial density.

Finally, measurements of the concentrations of atmospheric pollutants, such as SO_2, NO_2 and O_3, were performed in July 1999. Since these measurements are very recent, only preliminary results relative to the ozone concentration in daytime conditions have been reported.

Acknowledgements — This work has been carried out within an agreement between the Istituto Nazionale per la Fisica della Materia (INFM) and the Amministrazione della Provincia di Potenza. The partial financial support from the Amministrazione della Provincia di Potenza is acknowledged.

References

[1] DI GIROLAMO P., PAPPALARDO G., SPINELLI N., BERARDI V., VELOTTA R., J. Geophys. Res. **101** (1996), 18765.

[2] ANSMANN A., RIEBESELL M., WANDINGER U., WEITKAMP C., VOSS E., LAHMANN W., MICHAELIS W., Appl. Phys. **B 55** (1992), 18.

[3] WHITEMAN D.N., MELFI S.H., FERRARE R.A., Appl. Opt. **31** (1992), 3068.

[4] SVANBERG S., *Atmospheric pollution monitoring using laser lidars*, [In] *Optoelectronics for Environmental Science*, Plenum Press, New York 1990, p. 3–13.

[5] DI GIROLAMO P., CUOMO V., BOSELLI A., GAGLIARDI R.V., PAPPALARDO G., VELOTTA R., BARTOLI B., BERARDI V., SPINELLI N., Nuovo Cim. **18** (1995), 209.

[6] DI GIROLAMO P., CUOMO V., PAPPALARDO G., VELOTTA R., BERARDI V., Proc. SPIE **2310** (1994), 71.

[7] HAUCHECORNE A., CHANIN M.L., KECKUT P., NEDELJKOVIC D., Appl. Phys. **B 55** (1992), 29.

[8] DI GIROLAMO P., AMBRICO P.F., AMODEO A., BOSELLI A., PAPPALARDO G., SPINELLI N., Appl. Opt. (in press).

[9] MEASURES R.M., *Laser Remote Sensing: Fundamentals and Applications*, Wiley, New York 1984.

[10] DI GIROLAMO P., AMBRICO P. F., AMODEO A., BOSELLI A., CAPPOBIANCO R., PAPPALARDO G., SPINELLI N., *Water vapour and aerosol observation by lidar in the nocturnal boundary layer*, 19th ILRC, Annapolis, Maryland, USA, July 6–10, 1998.

[11] PAPPALARDO G., AMBRICO P., AMODEO A., BERARDI V., BOSELLI A., CAPOBIANCO R., DI GIROLAMO P., SPINELLI N., VELOTTA R., Proc. SPIE **3104** (1997), 158.

[12] MELFI S.H., WHITEMAN D., FERRARE R., J. Appl. Meteo. **28** (1989), 789.

[13] COLLIS R.T.H., RUSSEL P.B., *Lidar measurements of particles and gases by elastic backscattering and differential absorption*, [In] *Laser Monitoring of the Atmosphere*, [Ed.] E.D. Hinkley, Springer Verlag, Berlin 1976.

[14] CHUDZYŃSKI K., ERNST K., STACEWICZ T., STELMASZCZYK K., SZYMAŃSKI A., *A method of elaboration of a lidar signal*, EGAS'97, Berlin.

Received September 10, 1999