

A NEW GENERATION OF MOBILE RAMAN LIDAR

Royer P.^{1,2}, Chazette P.¹, Lardier M.², Butter K.², Sauvage L.²

¹Laboratoire des Sciences du Climat et de l'Environnement, CEA-CNRS-UVSQ, 91191 Gif-sur-Yvette Cedex, France, philippe.royer@lsce.ipsl.fr, patrick.chazette@lsce.ipsl.fr,

²Leosphere, 76 rue Monceau, 75008, Paris, France. mlardier@leosphere.fr, kbutter@leosphere.fr, lsauvage@leosphere.fr

ABSTRACT

The Commissariat à l'Énergie Atomique (CEA) and the LEOSPHERE Company have recently developed a new generation of mobile Raman lidar. This eye-safe instrument is based on a 16 mJ Nd:Yag laser at 355 nm and is composed of three reception channels: two dedicated to the measure of the two cross-polarizations and one to the nitrogen Raman backscattered signal. This compact and light lidar is easily transportable in a small truck or a commercial vehicle and allows the retrieval of aerosol optical properties and atmospheric structure with a spatial sampling of 1.5 m along the line of sight. It is particularly well adapted to air quality and pollution study thanks to its full-overlap reached at ~150 m. This prototype has been recently tested in the suburb of Paris.

We will present and analyze here daytime and nighttime observations performed with this new Raman lidar at Saclay in the south-west suburb of Paris.

1. INTRODUCTION

Aerosols come from natural (for sea salt, desert dust, volcanic aerosols) or anthropogenic sources (pollution emitted by traffic, industries and biomass burning activities). The high heterogeneity of these sources leads to a high spatial and temporal variability of aerosol chemical, optical and physical properties which make their study and characterization difficult.

The impact of aerosols on climate still remains out-of-the-way as shown in the last report of the Intergovernmental Panel on Climate Change [1]. The direct effect of aerosols is due to absorption and scattering of solar and infrared telluric radiations [2]. The aerosol absorption induce a local heating (semi-direct effect) which can lead to modifications in atmospheric circulation and cloud burn-off [2]. The indirect effect affects the cloud formation by acting as cloud condensation nuclei what increases the number and decrease the size of droplets in the cloud [3]. A lot of studies have also shown that aerosols had an impact on human health especially for pollution and biomass

burning aerosols [4]. Nowadays this major problem impacts almost half of the world population living in urban areas. It is thus important to study, understand and quantify the impact of anthropogenic aerosols on the air quality in high population density areas (megalopolis) in order to improve chemistry-transport models and the forecasts of pollution events.

The aerosol Backscatter-to-Extinction ratio (BER) which is to the product of the single scattering albedo and the normalized backscattering phase function is a crucial parameter to assess the radiative forcing and the aerosol type. Different techniques have been developed to determine this parameter from lidar measurements. The BER profile can be retrieved with a Rayleigh-Mie lidar pointing at horizontal onboard an Ultralight Aircraft [5] or using multi-angular measurements [6]. Both approaches need to suppose the horizontal homogeneity of the atmosphere what is not necessarily the case. A more usual method used is the synergy with a passive instrument [7] such as a sun-photometer giving the total aerosol optical depth (AOD). The main inconvenient of this approach is that it only gives access to the mean aerosol BER in the atmosphere. High Spectral Resolution Lidar and Raman lidar are based on the use of two measured profiles instead of just one. HSRL [8] takes advantage of the Doppler frequency shifts due to the different velocities of molecules (~300 m.s⁻¹) and aerosols (~1-10 m.s⁻¹) whereas the Raman lidar technique [9] measures the weak inelastic signal backscattered by nitrogen or oxygen molecules.

For years, Raman lidar is widely used in atmospheric research for water vapor, ozone, temperature or aerosol profiling. Raman lidars are based on the weak inelastic scattering of atmospheric molecule. The frequency shift, which results of a change in the rotational and vibrational state, is characteristic of the interacting molecule. Molecules with a high atmospheric concentration (such as nitrogen, oxygen and water vapor) are generally studied because they compensate for the low Raman cross section. For many years, Raman lidar observations were only possible during the night. Thanks to the development of high-power transmitters and narrow-band filters daytime

measurements are now possible by considerably reducing the daylight background.

The first results obtained around Paris with this new Raman lidar will be here presented and analyzed in detail.

2. LIDAR CHARACTERISTICS

Table 1 summarizes the main technical characteristics of the Raman lidar. It is a compact (~70x45x18 cm) and light (< 50 kg for optical and electronic system) lidar which operates onboard the ground based mobile experimental station MAS (Mobile Aerosol Station) in a small truck. A Nd:Yag Ultra laser manufactured by QUANTEL produces pulses at 20 Hz with a mean pulse energy of 16 mJ at 355 nm. The lidar sampling along the line of sight is 1.5 m in analog mode and 15 m in photon counting.

Laser	Nd:Yag Ultra (Quantel)
Energy (mJ)	16 at 355 nm
Frequency (Hz)	20
Reception channels	Elastic // 355 nm Elastic \perp 355 nm Raman-N ₂ 387 nm
Reception diameters (cm)	15
Field of view (mrad)	4
Full overlap (m)	~150
Detector	Photomultiplier tubes
Filter bandwidth (nm)	0.3
Vertical resolution (m)	1.5 (analog) 15 (photon counting)
Acquisition system	PXi technology at 100 MHz
Lidar head size (cm)	~ 70x45x18
Lidar head and electronic weight (kg)	< 50

Table 1. Lidar technical characteristics

The lidar is composed of three 15 cm-diameter reception channels with a field-of-view of about ~4 mrad: two dedicated to the measurement of the two elastic cross-polarizations and one to the inelastic nitrogen Raman backscattered signal at 387 nm. The signal detection is realized with photomultiplier tubes and narrow-band filters (0.3 nm) which allow only selecting the pure rotational part of the raman-N₂ spectrum.

It enables to retrieve aerosol optical properties (extinction, backscatter coefficient and depolarization ratio) and atmospheric structures (planetary boundary layer (PBL) heights, aerosol layers and clouds). It is particularly well adapted to air quality and pollution study thanks to its full-overlap reached at ~150 m.

3. RESULTS

The first measurements have been performed during the first half of March 2010 at the CEA site of Saclay about 20 km in the south-east suburb of Paris. This cold period is characterized by anti-cyclonic conditions and relatively low levels of pollution with mean daily PM10 concentrations between 20 and 40 $\mu\text{g}\cdot\text{m}^{-3}$ and aerosol optical depths between 0.05 and 0.2 at 355 nm. These values are close to those generally observed around Paris area.

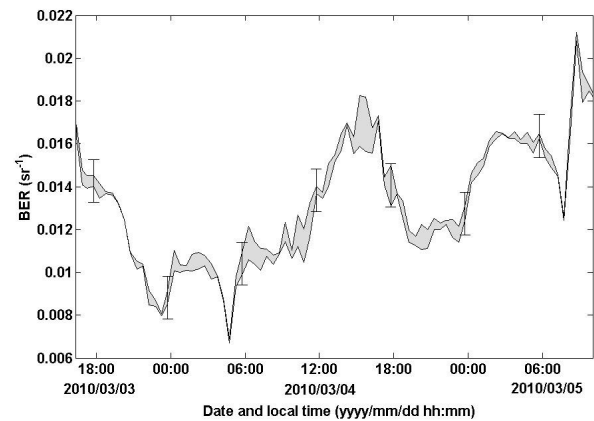


Figure 1. Temporal evolution of the mean aerosol BER at 355 nm between 0.2 km and 0.6 km retrieved at the CEA site of Saclay between 16h LT on March 3rd 2010 and 9h LT on March 5th 2010. The error on the BER values assessed to 0.002 sr^{-1} is represented by the errorbars on the figure.

Figure 1 shows the temporal evolution from March 3rd 2010 16h local time (LT) and March 5th 2010 9h LT of the aerosol BER at 355 nm. The BER values have been determined with a Klett algorithm [10] and a

dichotomous approach [7] converging on the aerosol optical depth determined with the N2-Raman channel between 0.2 and 0.6 km. The error on the BER has been assessed to 0.002 sr^{-1} considering the uncertainty on the aerosol optical depth values, which have been derived from the Raman-N2 channel.

A significant diurnal cycle is clearly visible with higher values between 0.013 and 0.018 sr^{-1} during the day from 12h to 18h LT and lower values close to $0.010 - 0.012 \text{ sr}^{-1}$ during the night. These night-time BER values are consistent with those retrieved during both the 'Etude et Simulation de la Qualit e de l'air en Ile-de-France' (ESQUIF, [11]) and 'Lidar pour la Surveillance de l'AIR' (LISAIR, [12]) campaigns.

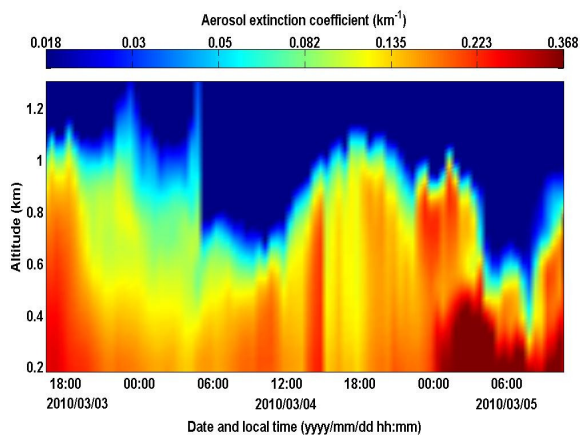


Figure 2. Temporal evolution of the aerosol extinction coefficient profile at 355 nm between 16h (LT) on March 3rd 2010 and 9h (LT) on March 5th 2010 at Saclay. The inversion has been realized using a Klett algorithm with the BER values determined in figure 1.

The BER values have been used to compute the temporal evolution of the aerosol extinction coefficient profile shown on Figure 2. We can notice the diurnal cycle of the Planetary Boundary Layer Height (PBLH) with values close to 0.4-0.5 km for the night and an increase during the day until about 1 km at 18h LT. The aerosol extinction coefficient values at 355 nm are comprised between 0.1 and 0.3 km^{-1} in the PBL. The higher values observed the 5th of March 2010 are due to hazy conditions as revealed by relative humidity measurements higher than 70%. Note that the presence of haze induced a significant increase of the BER from 0.011 to 0.016 sr^{-1} corresponding to a decrease of lidar ratio from 91 to 63 sr, respectively.

Figure 3 shows the mean profiles of aerosol backscatter, BER and extinction coefficient measured the 4th of March 2010 between 19h45 and 22h45 LT with the Raman lidar. The inversion has been realized

using a numerical differentiation (blue curve) or the Tikhonov regularization method (red curve) [13]. The BER profile looks almost constant between 0.5 and 0.8 km with a mean value close to 0.013 sr^{-1} and progressively decreases to reach 0.008 sr^{-1} at ground level. The mean BER between 0.2 and 0.6 km is close to 0.012 sr^{-1} which is in good agreement with the values retrieved using the dichotomous approach and shown in Figure 1. The aerosol extinction coefficient is close to 0.15 km^{-1} between 0.5 and 0.8 km and increases until 0.25 km^{-1} at 0.2 km due to pollution emission near the surface. This type of behavior is in agreement with previous results which highlighted significant differences between ground level observations and the same parameters within the mixed layer [14]. This may show that constrain air quality models by the only ground-level measurements is not enough to improve the air quality prediction.

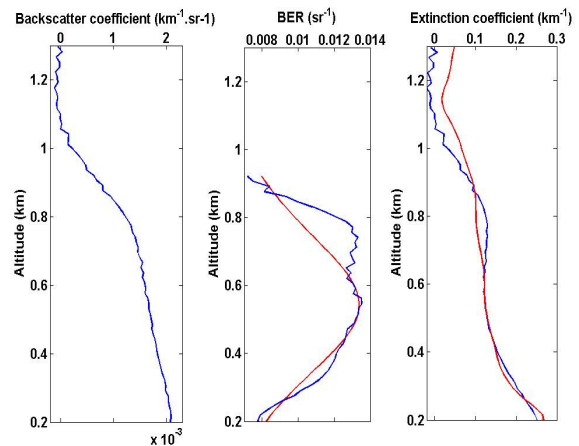


Figure 3. Mean profiles of aerosol backscatter coefficient (left), BER (center) and extinction coefficient (right) between 19:45 (LT) and 22:45 (LT) the 4th of March 2010 at Saclay. The BER profile has been retrieved with a numerical differentiation (blue curve) and with the Tikhonov regularization method (red curve).

4. CONCLUSION

These first results are encouraging and show that a low energy Raman lidar can be used to retrieve a mean BER value in the PBL until 0.7 km with a temporal resolution of 30 minutes during daytime. Mobile measurements are planned with this compact and light Raman lidar. Numerical simulations with a direct-inverse model are in progress in order to assess the night-time performances in photon counting mode of this lidar with a H₂O-Raman channel at $\sim 407 \text{ nm}$ to measure the profile of water vapor mixing ratio in the PBL.

REFERENCES

- [1] Intergovernmental Panel on Climate Control (IPCC), 2007: Climate Change 2007, the Fourth Assessment Report of the IPCC, *Cambridge Univ. Press*, New York.
- [2] Ramanathan, V., Crutzen, P.J., Lelieveld, J., Althausen, D., Anderson, J., Andreae, M.O., Cantrell, W., Cass, G., Chung, C.E., Clarke, A.D., Collins, W.D., Coakley, J.A., Dulac, F., Heintzenberg, J., Heymsfield, A.J., Holben, B., Hudson, J., Jayaraman, A., Kiehl, T., Krishnamurthi, T.N., Lubin, D., Mitra, A.P., McFarquhar, G., Novakov, T., Ogren, J.A., Podgorny, I.A., Prather, K., Prospero, J.M., Priestley, K., Quinn, P.K., Rajeev, K., Rasch, P., Rupert, S., Sadourny, R., Satheesh, S.K., Sheridan, P., Shaw, G.E., and Valero, F.P.J., 2001: Indian Ocean experiment: An integrated analysis of the climate forcing and effects of the great Indo-Asian haze, *J. Geophys. Res.*, **106**(D22), pp. 28371–28398.
- [3] Twomey, S., 1974: Pollution and the planetary albedo, *Atmos. Environ.*, **8**, pp. 1251-1256.
- [4] Dockery D. and A. Pope, 1996: Epidemiology of acute health effects: summary of time-series, in: *Particles in Our Air: Concentration and Health Effects*, edited by: Wilson, R. and Spengler, J. D., *Harvard University Press*, Cambridge, MA, USA, pp. 123–147.
- [5] Chazette, P., Sanak, J., and Dulac, F., 2007: New Approach for Aerosol Profiling with a Lidar Onboard an Ultralight Aircraft: Application to the African Monsoon Multidisciplinary Analysis, *Environ. Sci. Technol.*, **41** (24), pp. 8335–8341, doi: 10.1021/es070343y.
- [6] Sicard, M., Chazette, P., Pelon, J., Won, J. G., and Yoon, S.-C., 2002: Variational method for the retrieval of the optical thickness and the backscatter coefficient from multiangle lidar profiles, *Appl. Opt.*, **41**, pp. 493-502.
- [7] Chazette, P., 2003: The monsoon aerosol extinction properties at Goa during INDOEX as measured with lidar, *J. Geophys. Res.*, **108** (D6), pp. 4187, doi: 10.1029/2002JD002074.
- [8] Shipley, S.T., Tracy, D.H., Eloranta, E.W., Trauger, J.T., Sroga, J.T., Roesler, F.L. and Weinman, J.A., 1983: High spectral resolution lidar to measure optical scattering properties of atmospheric aerosol. 1: Theory and instrumentation, *Appl. Opt.*, **22**, pp. 3716-3724.
- [9] Ansmann, A., Wandinger, U., Riebesell, M., Weitkamp, C., and Michaelis, W. , 1992: Independent measurement of extinction and backscatter profiles in cirrus clouds by using a combined Raman elastic-backscatter lidar, *Appl. Opt.*, **31**, pp. 7113-7113.
- [10] Klett, J. D., 1981: Stable analytical inversion solution for processing lidar returns, *Appl. Opt.*, **20**, pp. 211–220.
- [11] Chazette P., H. Randriamiarisoa, J. Sanak, P. Couvert, and C. Flamant, 2005: Optical properties of urban aerosol from airborne and ground-based in situ measurements performed during the ESQUIF program, *J. Geophys. Res.*, **Vol. 110**, No. D2, D0220610.1029/2004JD004810.
- [12] Raut, J.-C. and Chazette, P., 2007: Retrieval of aerosol complex refractive index from a synergy between lidar, sunphotometer and in situ measurements during LISAIR experiment, *Atmos. Chem. Phys.*, **7**, pp. 2797-2815.
- [13] Tikhonov., A. E. and Arsenin, V. Y., 1977: *Solutions of Ill-posed Problems*, *Wiley*.
- [14] Raut, J.-C. and Chazette, P., 2009: Assessment of vertically-resolved PM10 from mobile lidar observations, *Atmos. Chem. Phys.*, **9**, pp. 8617-8638.