A new generation of mobile Raman lidar: Application to MEGAPOLI project

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ABSTRACT
The Commissariat à l'Energie Atomique (CEA) and the Centre National de la Recherche Scientifique (CNRS) have developed the Lidar pour l'Etude et le suivi des Aérosols Atmosphériques (LESAA) [1] and the Lidar Aérosols UltraViolet Aéroporté (LAUVA) [2]. The new version of these prototypes is now commercialized with success under license by the LEOSPHERE Company with the name EZ LIDAR®. This eyesafe instrument is based on a Nd:Yag laser tripled at the wavelength of 355 nm and has a high spatial resolution of 1.5 m along the line of sight. It allows performing measurement of both aerosols optical properties and atmospheric structure up to 14 km. This lidar has the advantage to be compact and light what makes it easily transportable (in a truck [1] [3] or onboard an UltraLight Aircraft (ULA) [2]).
The CEA and LEOSPHERE have modified the LESAA Rayleigh-Mie lidar into a two-wavelength nitrogen Raman lidar to build the LESAA-N2 lidar system. This prototype has recently been involved in the MEGAPOLI campaign "Megacities: Emissions, urban, regional and Global Atmospheric POLlution and climate effects, and integrated tools for assessment and mitigation" which took place at Paris in July 2009 (http://megapoli.dmi.dk/).
We present and analyze here daytime and nighttime observations obtained with this Raman lidar during the MEGAPOLI campaign at Saclay in the suburb of Paris.

1. INTRODUCTION

Thanks to technological progress Raman lidars have considerably spread during the past decade in atmospheric research for water vapor, ozone, temperature or aerosol profiling. Raman lidar 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 allow compensating the low Raman cross section. For many years, Raman lidar observations were only possible the night. With the development of high-power transmitters and narrow-band filters which permits to considerably reduce the daylight background daytime measurements are now available.
The lidar used here is an aerosol lidar measuring the backscattering signal by nitrogen molecules. It allows, by separating the molecular and particulate components, to determine without any assumptions the aerosol backscatter-to-extinction ratio [4] (BER) or the lidar ratio (LR) profile (contrarily to classic Rayleigh-Mie lidar which requires an a priori knowledge of the aerosol type or specific measurement strategy to inverse the signal). The BER is characteristic of the aerosol type and is directly related to single scattering albedo which permits to determine the aerosol radiative forcing.

In urban areas, the pollution from traffic or industries has become a subject of the greatest importance as it impacts the health of inhabitants and result in cardiovascular trouble [5]. The study of these polluted areas is indispensable in order to improve our understanding of the physical and chemical processes which play a key role on the pollution peaks. Such understanding will help improve chemistry-transport models and the forecasts of pollution events. Several urban regions have already been fully investigated during scientific campaigns (Mexico [6], the Po Valley [7], Hong Kong [8]). Pollution in the megacity of Paris, which comprises about 12M inhabitants, has been studied in the framework of 'Etude et Simulation de la QUalité de l'air en Ile-de-France' (ESQUIF, [9] [10]) and 'LIdar pour la Surveillance de l'AIR' (LISAIR, [11]) campaigns. Those campaigns allowed in particular retrieving aerosol complex refractive index from instrumental synergies of active and passive remote sensing and in situ measurements.

The first campaign of MEGAPOLI project took place around Paris in July 2009. Chemical analyzers, remote sensing and meteorological instruments from several European research groups have been deployed at ground level and in the air (in the ATR-42 French research aircraft).

© Proceedings of the 8th International Symposium on Tropospheric Profiling, ISBN 978-90-6960-233-2
Delft, The Netherlands, October 2009. Editors, A. Apituley, H.W.J. Russchenberg, W.A.A. Monna

The main objectives of MEGAPOLI campaign are:
1) To assess impacts of megacities and large air-pollution hot-spots on local, regional and global air quality and climate,
2) To quantify feedbacks between megacity emissions, air quality, local and regional climate, and global climate change,
3) To develop and implement improved, integrated tools to assess the impacts of air pollution from megacities on regional and global air quality and climate and to evaluate the effectiveness of mitigation option.

2. LIDAR DESCRIPTION

The technical characteristics of the LESAA-N2 system are summarized in Table 1. It is a compact (~100x50x20cm) and light (~60kg for optical and electronic system) lidar. A Nd:Yag Brilliant laser manufactured by QUANTEL gives pulses at 20Hz with a mean pulse energy of 65 mJ at 355 nm. The beam diameter is about 40 mm and its divergence <0.2mrad. The lidar resolution along the line of sight is 1.5m in analog mode and 15 m in photon counting. The full overlap is obtained from 200 m.
The lidar is composed of two independent 15 cm-diameter reception channels with a field-of-view of about ± 3 mrad: the elastic channel at 355 nm and the raman-N_2 channel at 387 nm. The detection is realized with photomultiplier tubes and narrow-band filters (0.3 nm) which allow only selecting the pure rotational part of the raman-N_2 spectrum.
The lidar operated onboard the ground based mobile experimental station MAS (Mobile Aerosol Station).

Table 1 : *Lidar characteristics*

Lidar head size (cm)	~ 100 x 50 x 20
Lidar head weight (kg)	~ 20
Electronic weight (kg)	~ 40
Electric tension (V)	220
Power (W)	< 1200
Laser	Nd:Yag Brilliant (Quantel)
Laser wavelength (nm)	355
Mean pulse energy (mJ)	65
Pulse repetition rate (Hz)	20
Pulse length (ns)	5
Bean diameter (mm)	~ 20mm
Beam divergence (mrad)	< 0.2
Detector	Photomultiplier
Detection mode	analog photon counting
Vertical resolution (m)	1.5 (analog) 15 (photon counting)
Reception channels (nm)	355 (elastic) 387 (raman N_2)
Reception diameters (mm)	150
Fields of view (mrad)	± 3
Filter bandwidths (nm)	0.3
Full overlap (m)	~ 200

3. OBSERVATIONS AND ANALYZE

The Figure 1 shows the different results obtained with the lidar during the preparation of MEGAPOLI campaign from 19:30 (local time) on 4 June 2009 to 9:30 on 5 June 2009 in Saclay. Measurements with the Raman lidar in photon counting mode have only been realized between 23:50 and 1:00 (see black rectangle on figure 1c). The mean aerosol backscatter coefficient (β_a) profile (blue solid line) and its variability (shading area) over this period are shown on figure 1a. This profile presents a slight increase of β_a from 2.5 to 3.5 $sr^{-1}.km^{-1}$ between 0.25 and 1 km and then regularly decreases to 0 $sr^{-1}.km^{-1}$ over the top of the planetary boundary layer (PBL) at about 1.7 km.

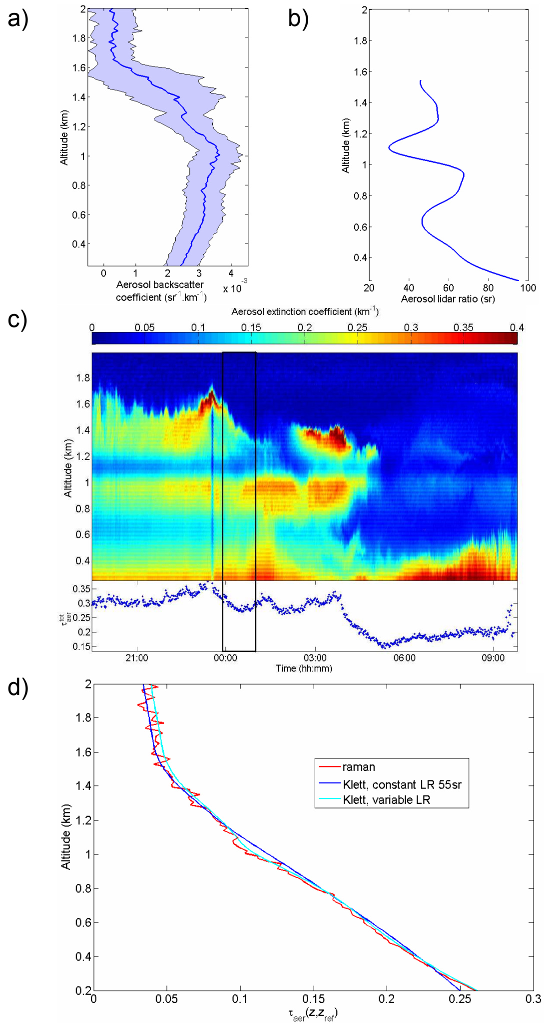

Figure 1: Nighttime lidar measurements realized in Saclay between 19:30 (local time) on 4 June 2009 and 9:30 (local time) on 5 June 2009. Results are shown for (a) the mean aerosol backscatter coefficient profile (blue solid line) and its variability (shading area), (b) the mean aerosol lidar ratio profile, (c) the temporal evolution of the aerosol extinction coefficient (top) and the total aerosol optical depth (bottom) at 355nm and (d) the cumulated aerosol optical depth determined with raman-N_2 channel (red solid line), Klett [12] inversion with a constant aerosol BER (in blue) and Klett inversion with the aerosol BER profile of figure 1b (in sky blue).

The mean profile of aerosol lidar ratio (see figure 1b) has been inversed with the Tikhonov regularization algorithm [13]. The lidar ratio is not constant through the PBL and different layers can clearly be identified. Between 0.4 and 1.7 km the lidar ratio oscillates around 55 sr (i.e. a BER of 0.018 sr^{-1}). Those weak values are generally associated to dust aerosols. The strong depolarization rate observed with another lidar at the same time (not shown here) let suppose that a dust event occurred during the night. Between 0.4 and 0.2 km the lidar ratio profile increases until 90 sr. Those larger values are typical of urban pollution aerosols emitted at the surface by traffic and industries [11].

This lidar ratio profile has then been used to inverse lidar data with a Klett inversion algorithm [12] and thus to obtain the temporal evolution of the aerosol extinction coefficient profile during the night (Figure 1c). Values of the extinction coefficient are significantly higher near the surface. Pollutants emitted by human activities are trapped either in PBL or in the nocturnal inversion layer (NIL). The top of the latter is very close to the surface. The PBL grows slightly after 06:00 (local time) with the increase of human activities. During the night important advection of pollutant aerosol is observed just above NIL and in the residual layer between 0.6 and 1 km. Another layer could be observed between 1.2 and 1.8 km corresponding to the advection of dust aerosols from the Saharan regions.

We also computed the cumulated aerosol optical depth $\tau_a(z,z_{ref})$ (see Figure 1d) between an altitude z and the reference altitude z_{ref}:

$$\tau_a(z,z_{ref}) = \int_{z}^{z_{ref}} \alpha_a(z')dz' \qquad (1)$$

where α_a is the aerosol extinction coefficient. The reference altitude is taken where there is no more aerosols, i.e. above 1.8 km. This cumulated aerosol optical depth can be directly obtained from elastic and Raman channels (red curve) or indirectly with a Klett inversion method supposing a constant lidar ratio profile (blue solid line) or variable (sky blue curve). The agreement between cumulated aerosol optical depth derived from Raman channel and the variable lidar ratio is quite good whereas there is some slight differences by considering a constant lidar ratio (especially below 0.4 km where the lidar ratio increases).

Figure 2 shows results of daytime measurements on 1 July 2009 between 15:45 and 18:45 LT during the MEGAPOLI campaign. It was a sunny day with high temperature (30 °C), weak wind (< 5 ms^{-1}) and high level of pollution in Paris area. Mean profiles of β_a (see figure 2a) and lidar ratio (2c) have been obtained by averaging the 3 hours of data. The sky background being too high during the day, this lidar measures have been acquired in analog mode. The signal-to-noise ratio (SNR) is clearly lower than in the previous example despite the fact that we have averaged much more profiles. Due to strong convection the PBL is fully developed and keep a constant β_a value between 1.6 and 1.8 km height (see Figure 2c). The aerosol backscatter profile present higher values at the top of the PBL which are certainly due to the presence of water vapor. Indeed, polluted aerosols emitted from Paris traffic have been shown to be very hydroscopic [14].
The lidar ratio is about 50-55 sr from 0.8 km to the top of the PBL and then regularly decreases until 35 sr at 0.35 km.
The total aerosol optical depth derived from lidar measurements is comprised between 0.5 and 1 at 355 nm (see Figure 3c and 3d). Those high values are in agreement with those from the sunphotometer-derived aerosol optical thickness over Palaiseau (http://aeronet.gsfc.nasa.gov/). Such a value is significantly higher than the mean value of 0.28 observed over Paris area [10].

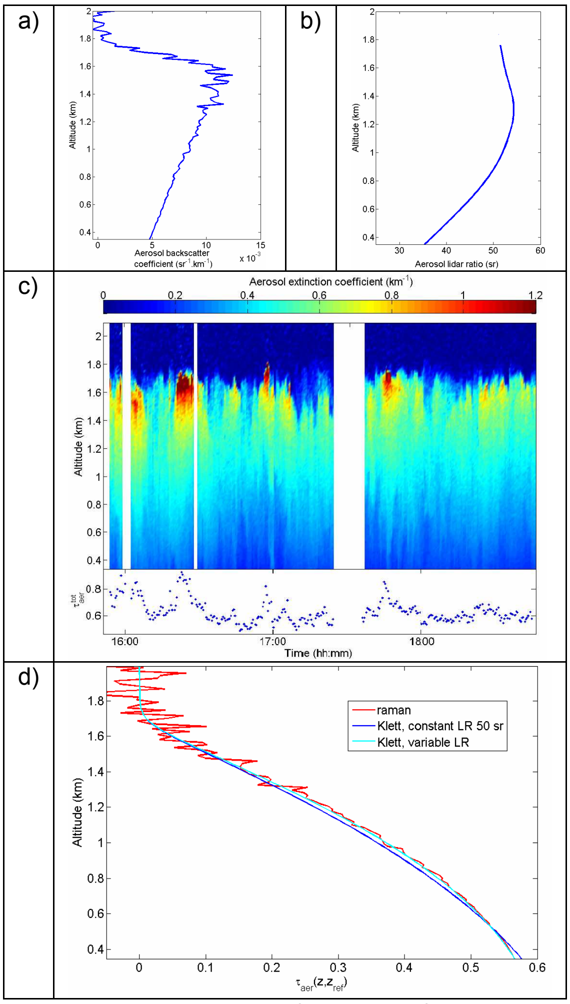

Figure 2: *similar as figure 1 for daytime observations during the MEGAPOLI campaign on 1 July 2009 between 15:51 and 18:44 (local time).*

3. CONCLUSION AND PERSPECTIVES

Simultaneous measurements with the Raman lidar and a vertical scanning Rayleigh-Mie lidar are envisaged in order to compare the lidar ratio profiles derived from the two methods (see [15]) and thus to

validate the Raman lidar results. Monte Carlo simulations are also in progress to assess the lidar ratio errors with the Tikhonov regularization algorithm with different lidar signal SNR.
Nevertheless, lidar observations performed during the MEGAPOLI project presented here show the ability to derive aerosol optical properties from Raman lidar LESAA-N2 during both daytime and nighttime measurements. This lidar will be soon modified to allow measuring the backscatter signal from water vapor molecules at 408 nm and thus to determine profiles of water vapor mixing ratio. It will probably be involved in other future campaigns such as the ADM-AEOLUS satellite validation (http://www.esa.int/esaLP/LPadmaeolus.html).
Eventually an airborne version of this compact and light lidar is envisaged.

4. REFERENCES

[1] Chazette P., P. Couvert, H. Randriamiarisoa, J. Sanak, B. Bonsang, P. Moral, S. Berthier, S. Salanave, and F. Toussaint, 2005: Three dimensional survey of pollution during winter in French Alps valleys, *Atmosph. Env*., **39**, 1345-1047,.

[2] Chazette, P., J. Sanak., and F. Dulac, 2007: New Approach for Aerosol Profiling with a Lidar Onboard an Ultralight Aircraft: Application to the African Monsoon Multidisciplinary Analysis, Environ. Sci. Technol., **41**, 8335–8341.

[3] Raut,J-C. and P. Chazette, 2009: Assessment of vertically-resolved PM10 from mobile lidar observations, Atmos. Chem. Phys. Discuss., **9**, 13475-13521.

[4] Ansmann, A., U. Wandinger, M. Riebesell, C. Weitkamp, and W. Michaelis, 1992: Independent measurement of the extinction and backscatter profiles in cirrus clouds by using a combined Raman elastic-backscatter lidar, *Appl. Opt.,* **31**, 7113–7131.

[5] Dockery, D., A. Pope, 1996: Epidemiology of acute health effects: summary of time-series. In: Wilson, R., Spengler, J.D. (Eds.), Particules in Our Air Conecentration and Health Effects, *Harvard University Press*, MA, USA, 123-147.

[6] Doran, J.C., X. Bian, S. de Wekker, S. Edgerton, J. Fast, J.M. Hubbe, W. Shaw, C.D. Whiteman, S. Abbott, C. King, J. Leach, M. Mulhearn, C. Russell, B. Templeman, D. Wolfe, J. Archuleta, S. Elliott, A. Fernandez, D. Langley, J. Lee, W. Porch, L. Tellier, J. Chow, J. Watson, R. Coulter, T. Martin, J. Shannon, R. White, D. Martinez, J. Martinez, V. Mora, G. Sosa, G. Mercado, J. Pena, R. Salas, and R. Petty, 1998: The IMADA-AVER Boundary Layer Experiment in the Mexico City Area. *Bull. Amer. Meteor. Soc.*, **79**, 2497–2508.

[7] Highwood, E.J., J.M. Haywood, H. Coe, , J. Cook, S. Osborne, P. Williams, J. Crosier, P. Formenti, J. McQuaid, B. Brooks, G. Thomas, R. Grainger, F. Barnaba, G.P. Gobbi, G. De Leeuw, 2007: Aerosol Direct Radiative Impact Experiment (ADRIEX) overview. Q. J. R. Meteorol. Soc. **133(S1)**, 3–15.

[8] Kok, G. L., J. A. Lind, and M. Fang, 1997: An airborne study of air quality around the Hong Kong Territory, *J. Geophys. Res.*, **102(D15)**, 19,043–19,057.

[9] Menut, L., R. Vautard, C. Flamant, al., 2000: Measurements and modelling of atmospheric pollution over the Paris area: En overview of the ESQUIF project, *Ann. Geophys.,* **18(11)**, 1467-1481.

[10] 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.

[11] Raut, J.-C. and P. Chazette, 2007: Retrieval of aerosol complex refractive index from a synergy between lidar, sunphotometer and in situ measurements during LISAIR experiment, *Atmos. Chem. Phys.,* ***7**, 2797-2815.*

[12] Klett, J. D., 1981: Stable analytical inversion solution for processing lidar returns, *Appl. Opt.*, **20**, 211–220.

[13] Tikhonov., A. E. and V. Y. Arsenin 1977: Solutions of Ill-posed Problems, *Wiley.*

[14] Randriamiarisoa H., P. Chazette, P. Couvert, J. Sanak, and G. Mégie, 2006: Relative humidity impact on aerosol parameters in a Paris suburban area, 1389-1407, SRef-ID: 1680-7324/acp/2006-6-1389.

[15] Sicard, M., P. Chazette, J. Pelon, J.G. Won, and S.-C. Yoon, 2002: Variational Method for the Retrieval of Optical Thickness ans the Backscatter Coefficient from Multiangle Lidar Profile, *Appl. Opt.*, **41**, 493-502.