AEROSOL PROPERTIES DERIVED FROM LIDAR/SUNPHOTOMETRY, A FOUR-YEAR SYSTEMATICS STUDY OVER THE CITY OF SAO PAULO, BRAZIL

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ABSTRACT

A backscattering Lidar (light detection and ranging) system (Measures, 1984), the first of this kind in Brazil, has been set-up in a suburban area in the city of São Paulo (23°33' S, 46°44' W) to provide the vertical profile of the aerosol backscatter coefficient at 532 nm up to an altitude of 4-6 km above sea level (asl). This lidar system was developed in the Laboratory of Environmental Laser Applications at the Center for Laser and Applications (CLA) at the Instituto de Pesquisas Energéticas e Nucleares – IPEN and it is operational since September 2001.

The Lidar measurements have been complemented with aerosol optical thickness measurements obtained by a CIMEL sun-tracking photometer in the visible spectral region, not only to validate the lidar data, but also to provide an input value of the so-called extinction-to-backscatter ratio (Lidar Ratio - LR).

At ther present a four-year validation methodology is being carried out mainly with 2004 Lidar and will be added to those made in previous years, namely 2001, 2002 and 2003 Lidar data. A correlation between Lidar data and the data obtained by a CIMEL sun-tracking photometer (which belongs to the AERONET network) placed in São Paulo is also being made to set a temporal database with both datasets.

INTRODUCTION

Air pollution in mega cities is one of the most important problems of our era. The city of São Paulo is in the rank of the five largest metropolitan areas of the world, as well as one of the most populated areas having about 11 million inhabitants. Therefore, in all these mega-cities the human activities have an enormous impact on the local atmosphere, as well as on their population health. Concerning the atmospheric quality, we highlight the suspended aerosol particles as a subject of continuous interest due the on-going expansion of the metropolitan area, which carries over than 15,000 industries. Among them the main aerosol sources include heavy industries, such as iron and steel works, refineries, chemical manufacturing, cement, sulphuric acid, petrochemical plants, and the automotive fleet, the latter exceeding already 5 million vehicles.

In Brazil, a continental sized country, there are only two operating lidar systems; the first is devoted to stratospheric studies (Clemesha et al. 1971) and the second, an elastic backscatter lidar system devoted to tropospheric aerosol profiling for air pollution applications (Landulfo et al., 2004). This limited number of lidar systems is in contrast to the large networks now in operation over Europe and North America.

The synergy of ancillary meteorological measurements and simultaneous investigations of the optical properties of the suspended aerosols (by sun photometers or spectrophotometers) can provide additional information for reducing the lidar data retrieval errors (aerosol extinction and backscatter profiles). Especially, this synergy of measurements helps to minimize the uncertainties of the assumptions made, when inverting the lidar signals (Takamura et al. 1994) using the input value of the extinction-to-backscatter ratio (lidar ratio), since it is well known that the lidar ratio has a wide range of values, which depend on the relative humidity (RH) and on the origin of the air masses sampled (Anderson et al. 2000, Ackermann 1998).

EXPERIMENTAL SET-UP

The lidar system in São Paulo is a single-wavelength backscatter system pointing vertically to the zenith and operating in the coaxial mode. The light source is based on a commercial Nd:YAG laser (Brilliant by Quantel SA) operating at the second harmonic frequency (SHF), namely at 532 nm, with a fixed repetition rate of 20 Hz. The average emitted power can be selected up to values as high as 3.3 W. The emitted laser pulses have a divergence of less than 0.5 mrad. A 30 cm diameter telescope (focal length f=1.3 m) is used to collect the backscattered laser light. The telescope's field of view (FOV) is variable (0.5-5 mrad) by using a small diaphragm. The lidar is currently used with a fixed FOV of the order of 1 mrad, which according to geometrical calculations permits a full overlap between the telescope FOV and the laser beam at heights higher than 300 m above the lidar system. A

Raman channel is currently being added and is foreseen for the near future upgrade of our system. This will enable us to determine the aerosol extinction and the aerosol backscatter coefficients independently at 355 nm.

The backscattered laser radiation is then sent to a photomultiplier tube (PMT) coupled to a narrow band (1 nm FWHM) interference filter, to assure the reduction of the solar background during daytime operation and to improve the signal-to-noise ratio (SNR) at altitudes greater than 3 km. The PMT output signal is recorded by adual Analog - Photoncounting system. Data are averaged between 2 to 5 minutes and then summed up over a period of about one hour, with a typical spatial resolution of 15 m, which corresponds to a 100 ns temporal resolution.

The CIMEL 318A spectral radiometer is a solar-powered weather hardy robotically pointed sun and sky instrument. This instrument is installed at the roof of the Physics Department at the University of São Paulo. The CIMEL photometer performs measurements of the aerosol optical thickness (AOT) at several wavelengths in the visible and the near infrared spectral region to enable the assessment also of the Ångström coefficient (Holben et al. 2001). For the relevant study, the channels used are centered at 440, 500 and 670 nm, with a 1.20 full angle field of view. The measurements are taken pointed directly to the sun or to the sky in nine different pre-programmed sequences (Holben et al. 1998). Three of them are dedicated to retrieve the AOT's, while the other six are taken to obtain the calibration parameters, the sky radiance, the aerosol particle size distribution, the refractive index (both real and imaginary components), the phase function, the total column abundance and the perceptible water content. The CIMEL sun photometer is calibrated periodically by a remote computer or locally under the supervision of the AERONET network. The calibration methodology assures a coefficient error between 1 and 3%, nonetheless various instrumental, calibrations, atmospheric, and methodological factors influence the precision and accuracy of the derived optical thickness and effectively the total uncertainty in the AOT is about 10%.

In the present stage, the retrieval of the aerosol optical properties is based on the measurements of the aerosol backscatter coefficient (βaer) at 532 nm, up to an altitude of 5-6 km asl. The determination of the vertical profile of the aerosol backscatter coefficient relies on the LIDAR inversion technique following the Klett's algorithm, as proposed by Klett (1985).

In this paper we did not consider multiple scattering effects, since no low-visibility conditions were taken into account. One can infer the Lidar equation presents two unknown parameters: β(λ,R) e α(λ,R). Therefore, is necessary to establish a relation between these two parameters to make the equation solvable. In order to achieve that, some considerations have to be made: a simple relation between α(λ,R) and β(λ,R), named as Lidar Ratio (LR) or extinction-to-backscatter ratio, is assumed.

However, it is known that the LR depends on several physical-chemical parameters inherent to the aerosols to be inspected, such as aerosol refractive index, and size and shape distribution of the aerosols particles. Besides, LR has a strong dependence on temperature profile and relative humidity that might cause variations on the aerosol optical parameters and on the presence of turbulence in the atmospheric volume being probed by the LIDAR beam as well.

To derive the appropriate 'correct' values of the vertical profile of aerosol backscatter coefficient in the lower troposphere we used an iterative inversion approach (by 'tuning' the LR values) based on the inter-comparison of the AOT values derived by LIDAR and CIMEL data, assuming the absence of stratospheric aerosols and that the PBL is homogeneously mixed between ground and 300 m height, where the lidar overlap factor is close to 1. Once the 'correct' values of the vertical profile of aerosol backscatter coefficient were derived we reapplied the Klett method, using the appropriate LR values, to retrieve the final values of the vertical profiles of the backscatter and extinction coefficient at 532 nm. This systematic error can be minimized using an iterative algorithm by direct measurements of the AOT from co-located CIMEL measurements. Using the information obtained by the CIMEL sunphotometer, one can derive the appropriate values of backscattering coefficient in the low troposphere and the Lidar AOT at 532 nm is determined by the following relation:

$$\tau_{532}^{aer} = \tau_{500}^{aer} \cdot \left(\frac{532}{500} \right)^{-a^\circ} \qquad (3)$$

Where the Ångström exponent , a° was derived from the measured optical thickness in the blue and red channels (440 nm and 670 nm respectively):

$$a^\circ = -\frac{\log\left(\tau_{440}^{aer} / \tau_{670}^{aer} \right)}{\log\left(440/670 \right)} \qquad (5)$$

The Ångström exponent is also an indirect mean to retrieve the particle size distribution and its possible composition (D'Almeida et al. 1991). Concerning the uncertainty, the major source of error would be in the calibration procedure, which is proportional to the associated uncertainty of the AOT at a given wavelength (Landulfo et al. 2003).

RESULTS

From the figure 1 below one can see the frequency of measurements taken during the 2001- 2004 period. Since we are mainly focused on the pollution events which are worse and more present during the dry season (June through September) one can see that the majority of measurements were taken in those corresponding months. Also the system has been fully operational by the end of the year 2002 which is realized by an increase in the number of measurements in the last years. In the years of 2003 and 2004 one can try to infere a seasonal signature in the data due the spreading of acquisition days throughout these years.

Figure 2 shows the Lidar Ratio extracted from the CIMEL data (Level 2.0, quality assured) on the days which the sunphotometer and the lidar took data together. One can observe an increase in the LR´s indicating a substantial change in the aerosol properties in the year 2004, in terms of absolute values one can see LR´s over 200, which is an unusual value at this wavelength (Anderson et al., 200, Liu et al. 2002, Cattral et al. 2005), if this value is due some miscalculation or calibration problem in the AERONET data is yet to be better explored, however the same pattern repeats in the Lidar extracted LR. This can be seen in figure 3 which represents the ratio of LR extracted from the CIMEL by the one from the LIDAR. There we can see that by the early stage of 2004 an increase in the LR value is observed.

We believe this high value of LR is a strong indication of a major aerosol presence from biomass burning sources and a work to identify its origin has to be pursued.

In figure 4 we compare the seasonal signature in the LR data, this procedure shows very useful in understanding aerosol properties (Mona et al., 2004), one can realize that the higher values of LR are given during the wet season (March – June) while lower values are in the dry season. This behaviour is somehow puzzling as one would expect that due the presence of transport in biomass burning more likely in the dry season and therefore a higher value in the LR, again this is a point to be further explored after the full analysis for year 2004.

For purpose of extending our analysis we also provide the 532 nm aerosol average optical thickness for the whole period as seen in figure 5. As we can see there is a similar trend between the LR plots and the AOT ones.

For future analysis we plan also to run a complete aerosol backscattering coefficient analysis in order to get a categorisation for 2004 – 2005 period like done before (Landulfo et al. 2004).

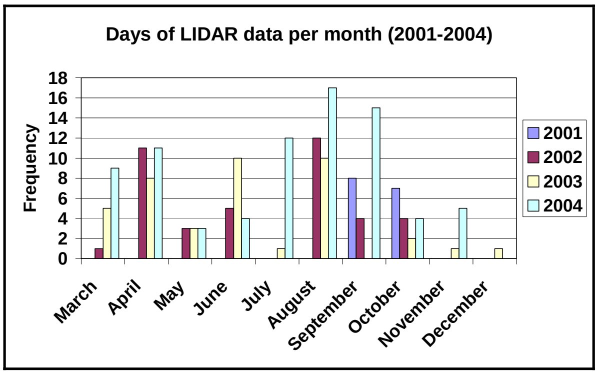

Figure 1 – Lidar Data Measurements Frequency in the 2001 – 2004.

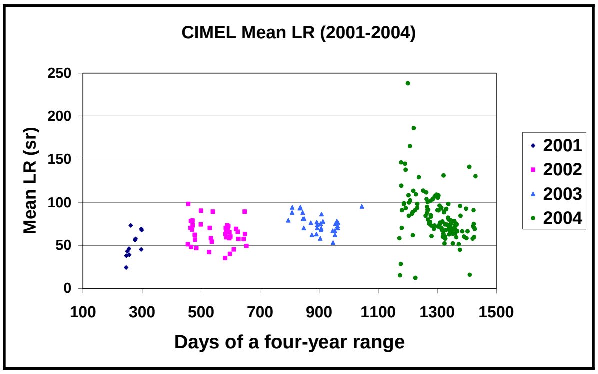

Figure 2 – CIMEL Lidar ratio during the 2001 – 2004 period; the indicated days are those which both CIMEL and LIDAR measured together.- Day 1 = 01/01/2001

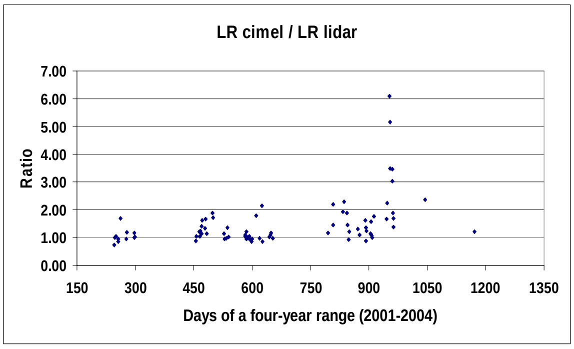

Figure 3 –Ratio between LR extracted from CIMEL and from LIDAR - Day 1 = 01/01/2001

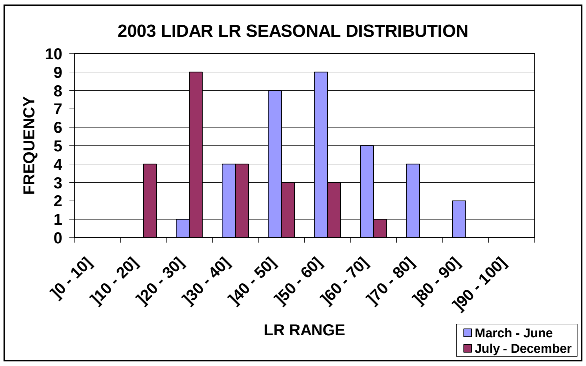

Figure 4 – A seasonal LR Lidar distribution for 2003

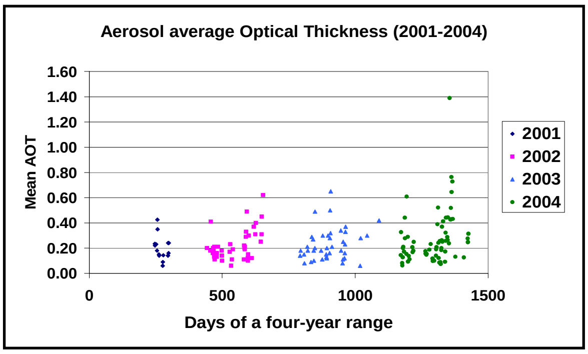

Figure 5 - 532 nm aerosol average optical thickness for the whole period

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REFERENCES

Ackerman, J. The *Extinction-to-Backscatter Ratio of Tropospheric Aerosol: A Numerical Study*, *Journal of Atmospheric and Oceanic Technology:* Vol. 15, No. 4, pp. 1043–1050, 1998

Anderson, T., Masonis, S.J., Covert, D.S., Charlson. R.J., Rood, M.J., *In situ measurement of the aerosol extinction-to-backscatter ratio at a polluted continental site*, , Journal of Geophysical Research 105, D22, 26,907 – 26,915, 2000.

Cattral, C., Reagan, J., Kurt, T., Dubovik, O. *Variability of aerosol and spectral lidar and backscatter and extinction ratios of key aerosol types derived from selected Aerosol Robotic Network locations*, Journal of Geophysical Research 110, D10S11, doi: 10.1029/2004JD005124, 2005.

Clemesha, B.R. and Rodrigues, S.N., *Stratospheric scattering profile at 23 degrees south.* Journal of Atmospheric and Terrestrial Physics, 33, pp. 1119–1125, 1971.

D'Almeida, G. A. et al: *Atmospheric Aerosols, Global Climatology and Radiative Characteristics*. Hampton, Virginia; Deepak Publishers, 1991.

Holben, B.N. et al*: AERONET - A federated instrument network and data archive for aerosol characterization*. Remote Sensing of Environment, 66: 1-16, 1998.

Klett, J. *Lidar inversion with variable backscatter/extinction ratios*, Applied Optics, 24, 11, 1638 – 1643, 1985.

Landulfo, E., Papayannis, A., Artaxo, P., Castanho, A. D. A., De Freitas, A. Z., Souza, R. F., Vieira Junior, N.D., Jorge, M. P. M. P., Sanchez-Ccoyllo, O.R., Moreira, D.S. *Synergetic Measurements of Aerosols over São Paulo, Brazil using LIDAR, Sunphotometer and Satellite Data During the Dry Season*, Atmospheric Chemistry and Physics, Vol. 3, 1523 –1539, 2003.

Landulfo, E. ; Papayannis, A. ; Souza, R. F. ; Freitas, A.Z. - *Lidar Aerosol Profile Categorisation in São Paulo, Brazil*. In: 22nd INTERNATIONAL LASER RADAR CONFERENCE, 2004, Matera. Reviewed and Revised Papers Presented at the 22nd International Laser Radar Conference (ILRC2004). Noordwijk, Holanda : ESA Publications Division. v. 1. p. 499-502, 2004.

Liu, Z., Sugimoto, N., Murayama, T., *Extinction-to-backscatter ratio of Asian dust observed with high-spectral-resolution lidar and Raman lidar*, Applied Optics, 41, 15, 2761 – 2767, 2002.

Mona, L., Amodeo, A., Pandolfi, M., Pappalardo, G., *Three years of Saharan dust observations over Potenza in the framework of Earlinet.* Reviewed and Revised Papers Presented at the 22nd International Laser Radar Conference (ILRC2004). Noordwijk, Holanda: ESA Publications Division. v. 2. p. 849-852, 2004.

Takamura, T., Sasano, Y. and Hayasaka, T., *Tropospheric aerosol optical properties derived from lidar, sun photometer, and optical particle counter measurements*. Applied Optics, 33, pp. 7132–7140, 1994.