LIDAR AEROSOL PROFILE CATEGORISATION IN SÃO PAULO, BRAZIL

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ABSTRACT

A backscattering LIDAR system, 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). The measurements have been carried out during the second half of the so-called Brazilian dry season, September and October 2001 and during the first half of the dry season in August and September 2002. The LIDAR data are presented and analysed in synergy with aerosol optical thickness (AOT) measurements in the visible region (532 nm) obtained by a CIMEL sun-tracking photometer (belonging to the AERONET network) for purposes of validation of the data. The LIDAR data were also used to retrieve the Planetary Boundary Layer (PBL) height, aerosol layering and the structure of the lower troposphere over the city of São Paulo, and allowed us to categorise the profiles according to three distinctive patterns followed by the meteorological conditions of each type of categorisation. Here we show these categorisations, how we defined them and their distribution over the period of measurements.

1. INTRODUCTION

Suspended aerosol particles play a significant role in Global Change issues, since they influence the earth's radiation balance and climate by scattering or absorbing both incoming and outgoing radiation and by acting as cloud condensation nuclei (CCN). Tropospheric aerosols arise from natural sources, such as airborne dust, sea-spray and volcanoes and also from anthropogenic sources, such as combustion of fossil fuels and biomass burning activities and from gas-to-particles conversion processes.

Air pollution in mega cities is one of the most important problems of our era. São Paulo is among the five largest metropolitan areas of the world, as well as one of the most populated with about 10 million inhabitants. Therefore, in all these mega-cities the human activities have an enormous impact on the air quality, 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 São Paulo metropolitan area, which carries more than 3.000 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, exceeding already 5 million vehicles.

Regarding its topography the city of São Paulo is located in a plateau at about 800 meters asl. And is surrounded by mountains of about 1200 m height. During the summer season the precipitation increases and many cold fronts generate meteorological instabilities, which indeed favour the pollution dispersion. These periods can extend over the autumn months of May and June, further on when the wintertime begins, a high-pressure semi-static regime over the São Paulo area is generally observed. This event becomes highly favourable to air pollutants accumulation, especially during episodes of intense temperature inversions, occurring typically at 1000 m asl. In this study in addition to local urban pollution, were able to find patterns in the aerosol profiles retrieved by a lidar system and thus propose a categorisation of such profiles which could be used as air-quality parameter for the local environmental monitoring agencies in addition to the instrumentation they used for air pollution control. Our measurements were mainy concentrated during the winter season in Brazil which shows two distinctive types of meteorological characteristics for the dry season, one whose days present synoptic systems of low pressure associated with high wind speed and occasional rain, which are favorable to pollution dispersion, against days which are characterized by synoptic systems of high pressure that brought mostly sunny weather, low wind speed and a low height thermal inversion layer, in these conditions the pollution dispersion is rather unfavorable. The sun photometer data are used to provide AOT values at selected wavelengths and thus to derive the Angstroem Exponent values over S. Paulo. The synergy of CIMEL and LIDAR measurements also acts in minimizing the uncertainties of the assumptions made, especially when inverting the LIDAR signal, using the Klett's technique [1].

Aerosol optical thickness measurements performed with the sunphotometer were taken under 'cloud free' conditions during the last period of the wintertime, September and October months in 2001, and in August and September 2002 [2].

2. THE EXPERIMENTAL SET-UP AND METHODOLOGY OF ANALYSIS

The LIDAR system operated over the city of São Paulo is a coaxial mode single-wavelength backscatter system pointing vertically to the zenith. The light source is a commercial pulsed Nd:YAG laser operating at the second harmonic frequency (532 nm) with a fixed repetition rate of 20 Hz. The average power can be selected as high as 3.3 W and the emitted laser beam has 7 mm diameter and a divergence of 0.5 mrad. The laser beam is sent to the atmosphere through a Newtonian telescope, which is equipped with a 30 cm receiving mirror and has a 1.3 m focal length. The optics set-up is such that the maximum overlap is reached at about 350 m above the LIDAR system [3]. The backscattered laser radiation is detected by a S-20 photomultiplier (PMT) and a narrow band (1 nm FWHM) interference filter at 532 nm is used to select the desired wavelength and assure the efficient reduction of the background skylight during daytime operation, thus, improving the signal-to-noise ratio at 532 nm. The PMT output signal is recorded in the analog mode by a 1-GSa/s digitising oscilloscope (DSO), having 11-bit resolution of analog-to-digital conversion (ADC). Data are averaged every 2 to 5 minutes, with a typical spatial resolution of 15 to 30 m.

As stated before, co-located CIMEL aerosol measurements were performed to determine the AOT values at several wavelengths in the visible spectrum and thus to enable the assessment of the AE values at the same spectral region. The principle of operation of the CIMEL instrument is to acquire aureole and sky radiances measurements. The standard measurements are taken 15 minutes apart, in order to allow for cloud contamination checking. These measurements are taken in the whole spectral interval, and their number depends on the daytime duration. The instrument precision and accuracy follow the standard Langley plot method within the standard employed by the AERONET network [2].

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 The determination of the vertical profile of the aerosol backscatter coefficient relies on the LIDAR inversion technique following the Klett's algorithm [1]. One has however to bear in mind that this inversion technique is an ill-posed problem in the mathematical sense, leading to errors as large as 30% when applied. Therefore, in this region and above it, the LIDAR signal shows a decay, which follows the molecular contribution only.

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. Such a procedure is not novel [4]. Once the 'correct' values of the vertical profile of aerosol backscatter coefficient were derived (when the difference of the AOTs derived by CIMEL and LIDAR was less than 10%) 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. The LR obtained from the CIMEL database was used as well, according to the following expression [5]:

$$S_i = \frac{4\pi}{\omega_o P_i(180)} \qquad (1)$$

where ω_o is the Single Scattering Albedo and $P_i(180)$ is the backscaterring Phase Function.

The inversion of the solar radiances measured by the CIMEL sunphotometer to retrieve the aerosol optical thickness values, is based on the Beer-Lambert equation, assuming that the contribution of multiple scattering within the small field of view of the sunphotometer is negligible. The molecular (Rayleigh) scattering contribution is taken into account to retrieve the aerosol optical thickness values at 532 nm, determined by the relation:

$$\frac{\tau_{532}^{aer}}{\tau_{500}^{aer}} = \left(\frac{532}{500}\right)^{-\mathring{a}} \qquad (2)$$

Where, the Ångström exponent [6] å was derived from the measured optical thickness in the blue and red channels (440 nm and 670 nm):

$$\mathring{a} = -\frac{\log\left(\tau_{440}^{aer} / \tau_{670}^{aer}\right)}{\log\left(440/670\right)} \qquad (3)$$

3. CATEGORISARION OF THE EXPERIMENTAL DATA – DRY SEASON YEARS 2001 AND 2002

The first LIDAR measurements started over São Paulo in August 2001. For the so-called dry season we selected some specific cloud-free days in the years 2001 and 2002, where CIMEL, lidar and satellite date were available and were typical for the aerosol loadings over the city of São Paulo. Table 1 summarizes the dates of simultaneous CIMEL and LIDAR measurements during the dry season months of the years 2001 and 2002. Also for the sake of comparison Figures 1 and 2 show the lidar Ratio (LR) distribution along the years of 2001 and 2002. In those figures the 3 methods for retrieving the LR´s are also given. The first LR is the one retrieved directly from CIMEL, the second LR is the one used by LIDAR with 10% difference against the CIMEL and the last LR is the one applied to obtain a matching between the AOT's provided by CIMEL and LIDAR, therefore, the majority of the aerosol load is trapped inside the PBL, which means between ground level and the top of the mixing layer (ML), typically 1 km above the ground. These profiles are sorted in three categories according to some common features of the vertical distribution of the aerosol backscatter coefficient in which the following characteristics are taken into account:

(a) Number of layers at various heights

Category A shows only one aerosol layer and above 1.5-2.0 km height the atmosphere is considered to be *aerosol free* (b is less than 0.005 $km^{-1}sr^{-1}$), *Category B* shows more layers above 2.5-3.0 km, and *Category C* show layers above 3.0 km.

(b) Potential Sources of Aerosol Particles

In the profiles shown under *Category A* the aerosol load has probably originated in the neighborhood region of the LIDAR site. In the profiles shown under Category B local air pollution sources play the main role. *Category C* has a distinctive pattern from category B since its shape up to 2.0 – 2.5 km is very similar to category A, which might be an indication the atmosphere above would be aerosol free, with the exception that a large layer in higher heights suggest that a long-range transportation might be occurring from remote areas outside the metropolitan area of São Paulo.

(c) Meteorology

The meteorology features for each category follows large scale parameters such as relative position of the anticyclone or cold front (Sánchez-Ccoyllo and Andrade, 2002):

Category A days are characterized as South Atlantic Subtropical High days which are sunny preceded by rainy days where a wash out of pollutants occurred, with very little wind and a high temperature amplitude and visibility conditions are extremely good (5-10 km). *Category B* days are sampled as pre-frontal days distinguished by a series of days without rain with some scattered clouds, and the presence of some wind at ground level and higher altitudes, concerning visibility they are hazy therefore the conditions are not so good as in category A. *Category C* days are tagged as post-frontal days which become very dry after sunrise and with very poor dispersion conditions, with some occurrences of thermal inversions, they are without clouds and with little wind as well at ground level.

4. PROFILES AND PARAMETERS FOR CATEGORY A , B AND C DAYS

The mean aerosol backscatter profiles obtained for year 2001 (Fig. 1) and year 2002 (Fig. 2) show that the majority of the aerosol load in category A is confined between ground level and 1250-1750 m height. In these days the mixing layer did not evolve to higher heights and thus an "practically" aerosol free atmosphere is found above 2000 m height, expressed as a value of $\beta_{aer} \leq 0.005\ km^{-1}.\ sr^{-1}$ or less. In this context the aerosol load observed inside the PBL should be due mainly to local urban activities, like car traffic, industrial emissions and other urban sources [7].

The AOT values obtained at 532 nm by the CIMEL and LIDAR during the years of 2001, the LR´s obtained through the three criteria and the Ångström exponent values calculated for each category are shown in table 1. The mean aerosol backscatter profiles representing category B in 2001 (Fig. 1) and in 2002 (Fig. 2) present a different pattern from category A, since one can observe higher and broader aerosol layers between 500 and 1500m (2001), 1500 and 2500 m (2001) and 2000 and 3500 m (2002) in altitude. Also at higher altitudes one can observe a discrete aerosol layer around 6000 m height. An aerosol "free" atmosphere is reached only above 4000 m. concerning the potential aerosol sources one can still assume originating from the nearby regions mainly due to the urban activity concerning the first 1000 m. The discrete aerosol layers found higher than 2500 m can be attributed to the residual layer from the previous day or most probably to particles originating from more remote areas.

The mean aerosol backscatter profiles belonging to category C in 2001 (Fig. 1) and in 2002 (Fig. 2) show a very peculiar pattern distinctive from categories A and B, since besides one the presence of a higher and broader aerosol layer above 1000 m up to about 2000 m, there are also the presence of much higher layers at 2500 to 4000 m (2001), and 4000 to 7500 m (2002), therefore one can not only have the influence of not only endogenous sources of aerosols but also from exogenous sources, as the rural areas of intensive harvesting in the State of São Paulo and with more remote sources where biomass burning activities take place during this period of the year as similar cases were observed in other parts of the globe [8].

5. DISCUSSION AND CONCLUDING REMARKS

In this paper we presented synergetic measurements of aerosols over the São Paulo area, using lidar, sunphotometer. The aerosol-backscattered profiles at 532nm.

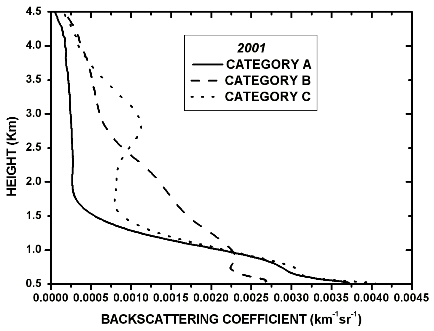

Fig. 1. Categories for comparison in the shape profile in year 2001 (raw range-resolution 15 m).

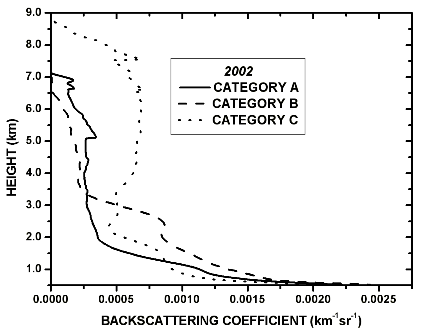

Fig. 2. Categories for comparison in the shape profile in year 2002 (raw range-resolution 30 m).

Table 1. List of selected days measured in the dry seasons of years 2001 and 2002. The corresponding AOT, LR and Ångström Exponent values retrieved by sunphotometer (CIMEL) data are also shown.

Categ.	# of times	LR averaged	AOT averaged	Angs.
A (2001)	4	60 ± 6	0.20 ± 0.03	1.8
A (2002)	6	51 ± 5	0.13 ± 0.02	1.7
B (2001)	13	36 ± 4	0.17 ± 0.02	1.6
B (2002)	7	60 ± 7	0.23 ± 0.04	1.8
C (2001)	6	52 ± 5	0.26 ± 0.04	1.6
C (2002)	11	55 ± 6	0.27 ± 0.04	1.7

The aerosol-backscattered profiles at 532 nm were obtained during the dry season of the years 2001 and 2002. The Klett inversion technique was applied in an iterative mode, with the synergy of the CIMEL data. From the profiles we could sort out three distinctively categories and characterized their features according to their morphology, potential aerosol sources and meteorological conditions.

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REFERENCES

1. Klett, J., Lidar inversion with variable backscatter/extinction ratios, *Applied Optics*, Vol. 24, 1638-1643, 1985.

2. Holben, B.N., et al. Aeronet – A Federal Instrument Network and Data Archive for Aerosol Characterization, *Remote Sensing and Environment*, 66, 1-16, 1998.

3. Chourdakis, G., et al. Analysis of the receiver response for a non-coaxial LIDAR system with fiber-optic output, *Applied Optics*, Vol. 41, 2715-2723, 2002.

4. Chazette, P., et al. Comparative Lidar Study of the optical geometrical, and dynamical properties of stratospheric post-volcanic aerosol, following the eruptions of El-Chinchon and Mount-Pinatubo, *Journal of Geophysical Research A*, Vol. 100, D11, 23195-23207, 1995.

5. Welton, E.J., et al., Measurements of Aerosol vertical profiles and optical properties during INDOEX 1999 using micropulse lidars, *Journal of Geophysical Research*, Vol. 107, D19, 8019, 2002

6. Angström, A., The Parameters of Atmospheric Turbidity. *Tellus*, Vol. 16, 64-75, 1964.

7. Landulfo, E., et al. 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.

8. Wandinger, U., et al., Optical and physical characterization of biomass burning and industrial pollution aerosols from multiwavelength lidar and aircraft measurements, *Journal of Geophysical Research*, Vol. 107, DOI 10.1029/2000, ID000202, 2002.