Lidar/photometry studies at São Paulo in the 2003-2005 period, Brazil

Eduardo Landulfo[a],Patricia Sawamura[a], Sandro T. Uehara[a], Walter M. Nakaema[a], Ani S. Torres[a], Fábio J. S. Lopes[a], Caio A. Matos[a], and Wellington C. de Jesus[a]

[a]Instituto de Pesquisas Energéticas e Nucleares, Av. Prof. Lineu Prestes 2242 CEP 05508-000, São Paulo, Brazil

ABSTRACT

A lidar system has been operational at São Paulo, Brazil (23^o S, 46^o W) since 2001 and colocated is a sunphotometer belonging to AERONET . During this last years aerosol properties has been extracted from both systems and seasonal trends have been observed specially when long range transport takes place bringing plumes with biomass burning aerosol which can distinctively be extracted from a heavy loaded atmosphere as São Paulo. These events trigger poor air quality conditions which can be easily correlated. The parameters for studying these patterns are Aerosol Optical Depth, Angström Exponent and Lidar Ratio. We show here some case studies belonging to years 2003, 2004 and 2005.

Keywords: Aerosols, LIDAR, Sunphotometry

1. INTRODUCTION

Suspended aerosol particles play a significant role in Global Change issues, since they influence the earths 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, seaspray and volcanoes and also from anthropogenic sources, such as combustion of fossil fuels and biomass burning activities and from gas-toparticles conversion processes.[1] 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 millionvehicles. Regarding its topography the city of São Paulo is located in a plateau at about 850 meters asl. And is surrounded by mountains of about 1200m 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 1000m above sea level. The study shown here is part of a long term aerosol observation which started in 2001.[2] We will show that the LIDAR generated data in coordination with those data acquired by a sunphotometer can help in the study of the aerosol vertical distribution over the metropolitan area of São Paulo. During the so-called dry season spanning from late June until early November there are some very poor air quality episodes over the city than can not only understood from local conditions but also from external sources. In this direction we performed some studies the aerosol optical properties namely *Aerosol Optical Thickness*, *Ångström exponent* and *Lidar Ratio* (extinction-to-backscattering ratio) over the period of 2001-2005 giving a more climatological aspect to our study. In this analysis we perfomed some statistical studies to check seasonal signature patterns between the

Further author information: (Send correspondence to E. Landulfo)
E. Landulfo: E-mail: elandulf@ipen.br, Telephone: 55 11 3816-9314

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dry and wet seasons. We should present also some case study days when very high AOT(532 nm) were observed in order to show the type of analysis we conducted and to see if any specific features could be realized from this type of analyses. A more intensive study is currently under process to be submitted in the near future.

2. EXPERIMENTAL SETUP

2.1 Backscattering Lidar

The LIDAR system operated over the city of Sãao 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 1.3m focal length. The PMT output signal is recorded by a dual Analog - Photoncounting system.The optics set-up is such that the maximum overlap is reached at about 350m above the LIDAR system. Data are averaged every 2 to 5 min, with a typical spatial resolution of 15 to 30 m.

Table 1. Lidar System Summarized Features

LASER	3.3 W, 20 Hz, 532 nm
TELESCOPE	30 cm primary mirror, focal length=1.3 m
FILTER	Interference 1 nm *FWHM*
DATA DIGITAL PROCESSING	Analog 20 MHz and Photocounting 250 MHz, dual acqusition

2.2 Sunphotometer

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 Angström coefficient. 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.[3] Three of them are dedicated to retrieve the AOTs, 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%.

3. METHODOLOGY

In the present stage, the retrieval of the aerosol optical properties is based on the measurements of the aerosol backscatter coefficient (β_{aer}) 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 Kletts 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: $\beta(\lambda, R)$ e $\alpha(\lambda, R)$. 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 $\beta(\lambda, R)$ e $\alpha(\lambda, R)$), named as Lidar Ratio (LR) or extinction-to-backscatter ratio, is assumed:

$$LR = \frac{\alpha(\lambda, R)}{\beta(\lambda, R)} \tag{1}$$

However, it is known that the LR depends on several physical-chemical parameters inherent to the aerosols to be inspected,[4] 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 relation:

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

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

$$\mathring{a} = -\frac{\log(\frac{\tau_{440}^{aer}}{\tau_{670}^{aer}})}{\log(\frac{440}{670})} \tag{3}$$

The Ångström exponent is also an indirect mean to retrieve the particle size distribution and its possible composition. 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.

4. RESULTS

From the figure 1 below one can see the frequency of measurements taken during the 2001- 2005 period. Since we are mainly focused on the extreme 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 year of 2005 the CIMEL sunphotometer went through its routine maintenance and was kept away for more than 3 months which generated a gap in our time series. At present we are filling these missing data with those taken by a Multi-Filter Rotating Shadow Band Radiometer(MFSRS)[5] which provides values of AOT as well. In the years of 2003, 2004 and 2005 one can try to infere a seasonal signature in the data due the spreading of acquisition days throughout these years. This can be seen through figures 3 and 4 where one can see the mean LR values for the months corresponding to the wet season (March - June) and the dry season (July - November). In those plots we could fit to our data two bimodal gaussian curves. In both seasons the mean peaks can be found at 48 sr and 69 sr, respectively. This second peak can be not only understood as a change of local meteorological conditions but also as result of aerosol transport from remote areas,[2,6] which is more pronunced during the dry seasons. From the AERONET data the AOT yearly distribution at figure 2) so the variability of this quantity can be checked during the wet/dry season months when some extreme pollution events can be observed as the AOT(532 nm) can reach fairly high values.[6] From these days we selected some days as case studies in which three key parameters: *Ångström Exponent*, *Lidar Ratio* and *Aerosol Optical Thickness* were verified. Those days, *August 13th, 2003*, *August 19th, 2004*, *September 23th, 2004* and *August 30th, 2005* have in common poor air dispersion conditions and the sypnotic features favored the transport of air masses loaded from biomass burning aerosol sources.[2,7–9] On Table 2 we show the three optical parameters obtained for each of the selected days and their variation during those days that can be observed also in figures 5, 6, 9 and 10. In that table we show also if a significative change in the values of Lidar Ratio/Ångström exponent occurred then we correlated that with a change in the type of aerosol. However one should not discard changes in the meteorological conditions such as relative humidity variation which can also change drastically the aerosol optical properties.[10,11] One can see that in some cases a direct correlation with a change in LR/AE is not observed in accordance with a high value of AOT. This can be understood as the

Table 2. *Ångström Exponent*, *Lidar Ratio* and *Aerosol Optical Thickness* Summarized Features for the selected days

Day	AE Range	LR Range	AOT range	Aerosol Change due LR	Aerosol Change due AE
08/13/2003	1.6 - 1.8	18 - 23	0.15 - 0.30	-	-
08/18/2004	1.6 - 1.7	40 - 70	0.30 - 0.45	√	-
09/23/2004	1.3 - 1.4	22 - 30	0.70 - 0.80	-	-
08/30/2005	1.5 - 1.8	40 - 60	0.30 - 1.70	√	√

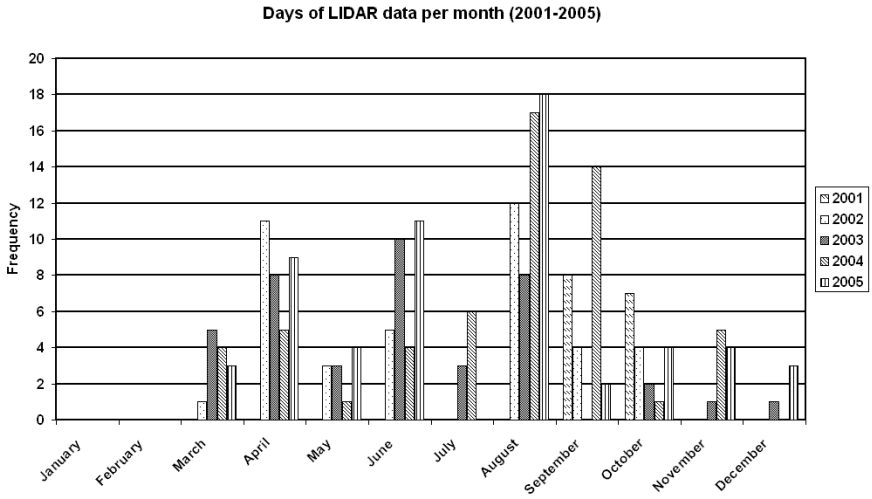

Figure 1. Histogram of Lidar Data acquisition since 2001.

systems were taking data in the exact time as the incoming air masses appeared as it is the case on August 30, 2005. This has to be inspect more in depth in order to draw further conclusions.

On September 23, 2004 and on August 30, 2005 one can observe through "the curtain plots" available in figures 7 and 8 the presence of layers above the Planetary Boundary Layer (PBL) around 3-4 km suggesting a long range transport.[2] From the AERONET and LIDAR optical parameters in figures 9 and 10 one observes that a large aerosol concentration peaks up when AOT values over 1.4 and 1.8 are observed, respectively. One might also infere for these days that the presence of biomass burning transported from remote areas since the LIDAR data shows layers at heights above the Mixed Layer. It is expected the occurrence of strong absorbing particles transported over a large distance one expects increases in size as result of coagulation, condensation and gas-to-particle conversion[12] therefore size distribution can present both coarse and fine modes in equal amounts.

4.1 HYSPLIT backtrajectories calculations

The HYSPLIT4 (Hybrid Single-Particle Lagrangian Integrated Trajectory) Model[13] developed by NOAA/Air Resources Laboratory can be utilized to analyze particle transport over the areas of interest to verify the influence of external aerosol sources incoming in the area where our experiment took place. NCEP/NCAR reanalysis data on a 2.5 grid and with a temporal resolution of 6-h are used as input meteorological data into the HYSPLIT4 model in this study. Figures 11 and 12 present a transport map of particles calculated with the HYSPLIT4 model to investigate the origin of air masses incoming in São Paulo. In general in these days due a temperature gradient there is a high pressure system that resides over the South-Southeast Brazilian states drawing air inland from the Atlantic Ocean and from the North-Central States. This air circulation can bring a great amount of aerosols into the city of São Paulo specially that the cropping cycle is over and a great deal of biomass burning activities take place during this period. As mentioned from the lidar profiles taken during this period one could

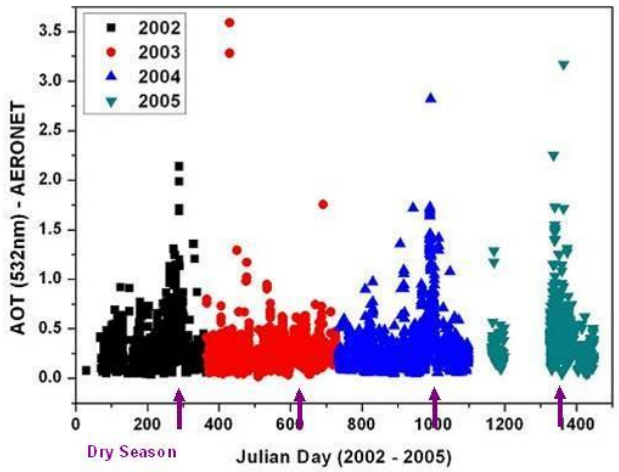

Figure 2. Scattergram of AERONET's retrieved AOT's during the period 2002-2005. Day One is considered as 01/01/2002. The Gap in the year of 2005 is due a recalibration process at the manufacture's site and filters exchange.

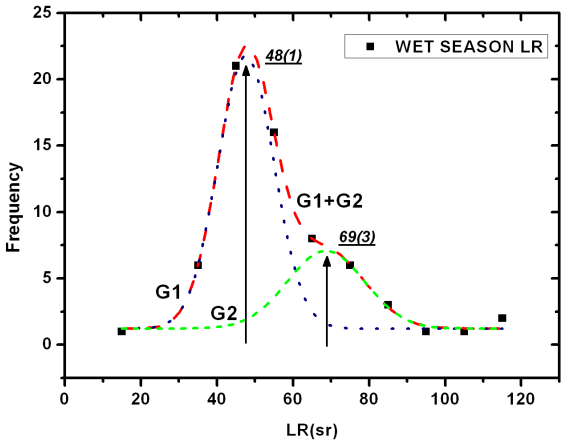

Figure 3. Lidar Ratio seasonal signature during the 2001-2005 wet season period. The two gaussian peaks could be related with two different kinds of aerosol. The first peak with a LR around 48 sr and a second one at 69 sr.

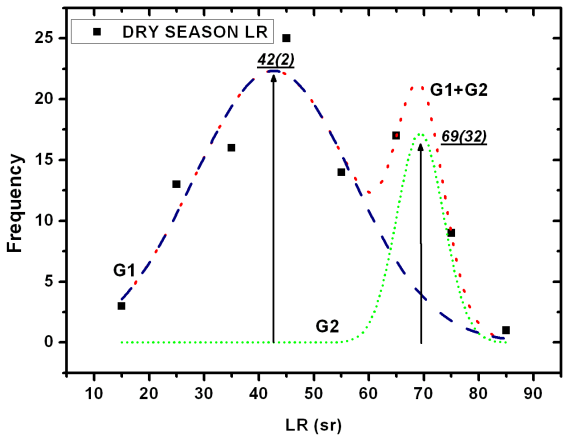

Figure 4. Lidar Ratio seasonal signature during the 2001-2005 dry season period. Again the two gaussian peaks are present, however the second at around 69 sr is more pronounced than that one present in the wet season.

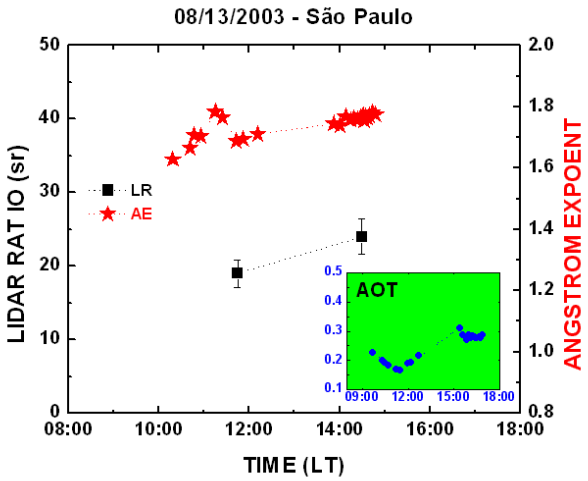

Figure 5. Lidar Ratio, Ångström exponent and AOT(532nm) variation on August 13, 2003. More details are covered on Table 2

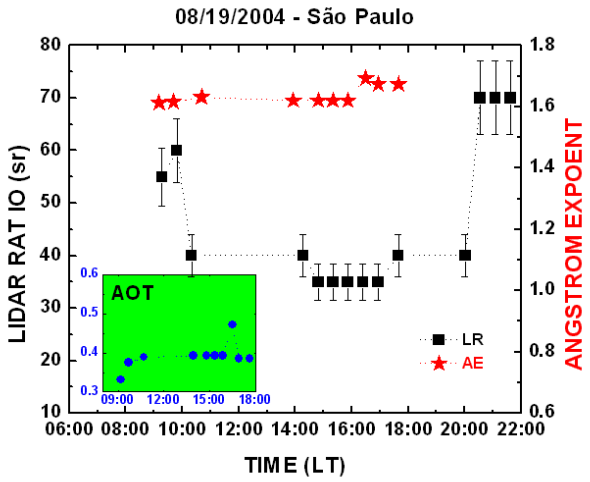

Figure 6. Lidar Ratio, Ångström exponent and AOT(532nm) variation on August 19, 2004. More details are covered on Table 2

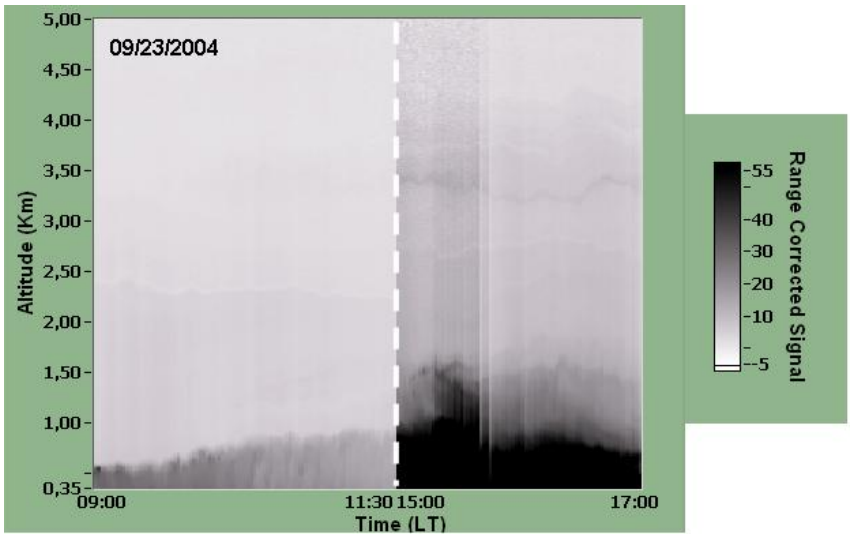

Figure 7. September 23rd, 2004 Curtain Plot. In this kind of plot one has the continuous range corrected signal displayed and it is possible to notice the layering structure in the atmosphere both above nd below the PBL. In the 15:00 - 17:00 LT period one can extract some layering between 2.5 and 4 km which is an indication of long range transport over the city of São Paulo and was present in a previous period at lower altitudes ($\approx$ 2.5 km).

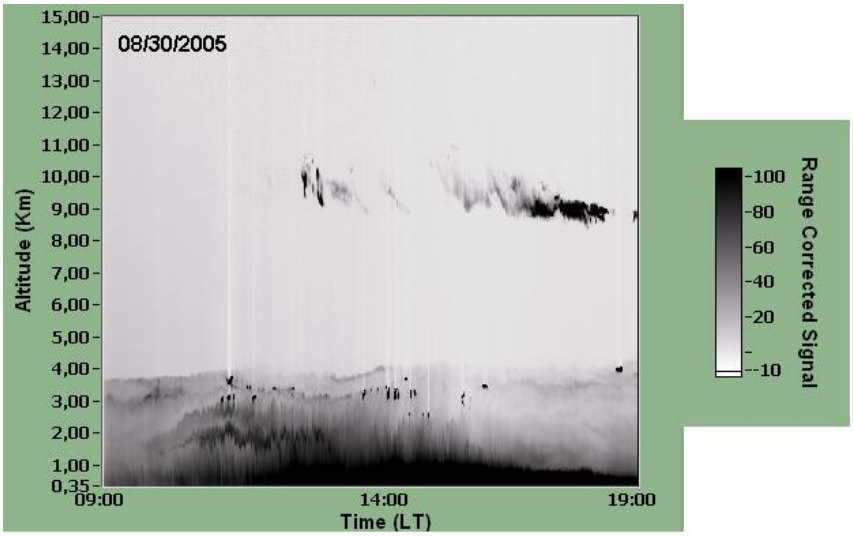

Figure 8. August 30th, 2005 Curtain Plot.This day is a very interesting one as both systems could catch the incoming air mass in the city. The cirrus clouds present in this plot suggest the air mass entrance time. Again plumes can be observed above the PBL indicating a transport of aerosols.

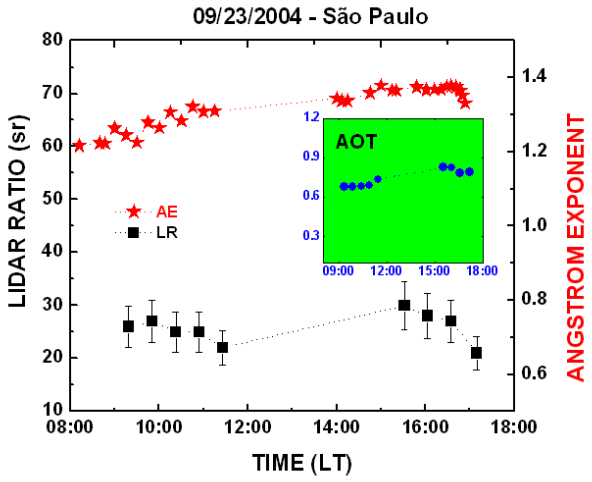

Figure 9. Lidar Ratio, Ångström exponent and AOT(532nm) variation on September 23, 2004. More details are covered on Table 2

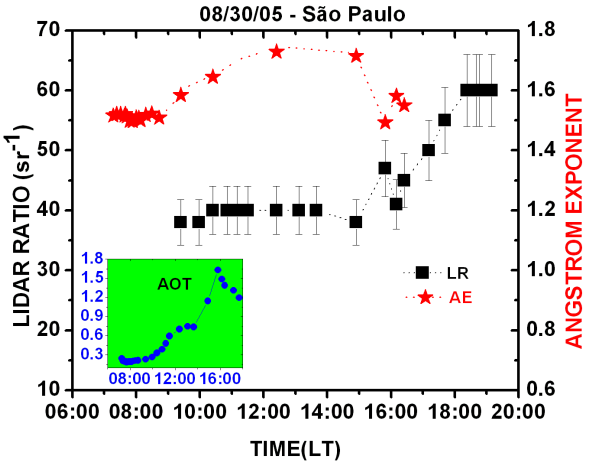

Figure 10. Lidar Ratio, Ångström exponent and AOT(532nm) variation on August 30, 2005. More details are covered on Table 2

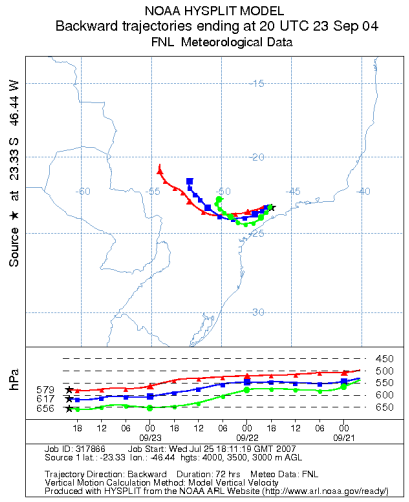

Figure 11. HYSPLIT4 simulated air mass backtrajectory with 72 hour integration time before 09/24/2004.

detect that the incoming air parcels bringing aerosols were about 3-4 km discarding the possibility the aerosol load was only from local sources.

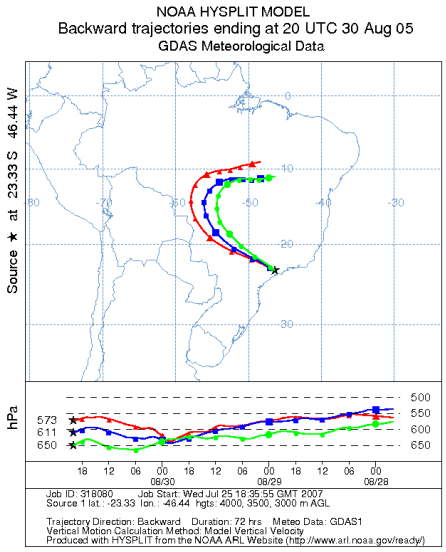

Figure 12. HYSPLIT4 simulated air mass backtrajectory with 72 hour integration time before 08/30/2005.

5. CONCLUSIONS

We showed here the study of aerosols over the city of São Paulo with sunphotometers and a backscattering lidar. The period covered was 2003-2005. We could pinpoint some seasonal signature through the statistical study of the Lidar Ration occurence during the months correspondig to the dry and wet seasons, respectively. In both seasons a bomidal gaussian could be fitted with central peaks at 48 sr and 69 sr, the second peak was more pronunced in the day season period; suggesting the influence of a optically diverse type of aerosol related to long range transport into the metropolitan area of São Paulo. Also we showed some selected days when high values of AOT were observed and checked the behavior of other optical parameters: *Ångström Exponent* and *Lidar Ratio* for these days. It is important we could realize that the change on these parameters and the incidence of a high AOT were more "drastic" when coincident with the incoming of an air mass with a great amount of biomass burning originated in more remote areas in Central-NW Brazil. In the future we are planning to extend our results by the inclusion of the data taken in 2006-2007 thus we will be able to improve our statistical analysis and have more cases of long range transport of aerosols in São Paulo.

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