Indirect Aerosol Hygroscopic Growth Observations with a Backscatter Lidar

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ABSTRACT

Aerosol Higroscopicity addresses a particular aspect of aerosol, namely, the extent to which they have an affinity for water vapor. The size increase of aerosol particles resulting from uptake of water vapor has important implications for the direct scattering of radiation and, under the right circumstances, to form cloud droplets. Ultimately this effect should have an important effect on the Earth's radiative budget and belongs to a category well known as aerosol indirect effect. Thus we performed an experiment using a single-wavelength backscatter LIDAR (532 nm), combined with a sea-breeze onset phenomenum and able to qualitatively describe a hygroscopic effect of aerosols over São Paulo metropolitan region (23^o S, 46^o W). To test this factor assessment we employed data obtained in a single day, namely on 11 September 2007, when a well characterized humidity intrusion is onset due the transport of water vapor by a sea-breeze phenomenon. For this data, we calculated the backscatter coefficient at 532 nm, and used this parameter to obtain the hygroscopic growing factor, assuming a well-mixed boundary layer where a cloud cap condition is present or a well defined and pronounced mixing layer boundary. These assumptions guarantee that any changes in the backscatter coefficient could be due to changes in relative humidity coupled to a mixing in aerosol size and type distribution. The results shown here should be regarded as a first step on an ongoing monitoring process of aerosol growth factor and will in the near future be merged with a Water Vapor Raman Lidar system in order to have a simultaneous water vapor mixing ratio profile together with the aerosol profile and it should be mainly used when clear, low-aerosol load conditions are available.

Keywords: LIDAR, aerosol, hygroscopic growth

1. INTRODUCTION

Aerosols and their relationships with clouds and rainfall regimes are one of the weakest understood aspects of current climate modeling.[1] Aerosols can affect climate directly by scattering and absorbing both solar radiation and that which is re-emitted from the surface of the Earth to the atmosphere. They also play a critical role in water vapor nucleation processes, working as cloud condensation nuclei. In this way, they can affect the concentration and size distribution of cloud droplets and thus affect nature and distribution of rainfall.One of the important issues that may be taken into account to understand the extension to which aerosols can affect rainfall and cloud regimes is the higroscopicity, the property of aerosols to condensate water vapor upon them (work as cloud condensation nuclei) and form cloud droplets. This property is function of the chemical nature of the particles and relative humidity. Although the term aerosol is applied to a large range of particles (with diameter ranging from tens of nanometers to hundreds of micrometers),[2] of different types and chemical natures, its well known that the hygroscopic nature of different aerosol populations has to be taken under consideration when understanding these climatic effects. Hänel[3] proposed simple models for the growth of hygroscopic aerosols as a function of relative humidity based on theoretical considerations and observational evidence. He has observed that the increase in particle size, as described by the ratio of the size of the particle over its dry size, is a complex function of relative humidity (RH), but can also be a function of the particle dry size. Apart from the change in size, hygroscopic aerosols experience a change in their refractive index as RH increases. Generally, as the water uptake by the particles gets more and more important, the real and imaginary parts of their refractive index tend to decrease, as the real part of the refractive index of pure water is lower than the one associated to dry

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particles and its imaginary part is zero. This would suggest a decrease in aerosol backscattering and absorption as RH increases. But as the scattering is a complex function of both refractive index and particle size, both effects need to be taken into account. In fact, variations in refractive index as RH increases are not large enough to counteract the variation of the particless cross-section due to size increase. So the size dependence dominates, leading to an increase in backscattering as RH increases. Junge[4] discussed hygroscopic properties of different types of aerosol populations and, for simplicity, divided these populations into two categories of hygroscopic particles: continental and maritime. The continental was assumed to be spherical particles of ammonium sulfate about a insoluble core of silicon dioxide, thus having a soluble mass fraction of about 0.25. The maritime aerosol was considered to be entirely formed of sodium chloride, having a soluble mass fraction of 1(thus, being more hygroscopic than urban aerosols).[5] Other differences between maritime and urban aerosol particles are the number, size and lognormal distribution.[2] These differences will be important when we discuss the observed results in this work. The ability to verify the hygroscopic growing factor with a single-wavelength LIDAR was described in literature in a set of initial conditions.[6–9] This ability is based on the fact that if a population of aerosols is changing its size distribution, then we expect to verify a change in measured values of backscattering and extinction coefficients.

2. METHODOLOGY

The aerosol backscattering profile could be retrieved by solving the Lidar equation using a Klett like algorithm[11] .

$$P(\lambda, R) = P_L\left(\frac{c\tau}{2}\right)\frac{\beta(\lambda,R)\; A_0\; \xi(\lambda)\; \zeta(R)}{R^2}\; exp\left[-2\int_0^R \alpha(\lambda,r)r\right] \tag{1}$$

where, P(λ,R) is the lidar signal received from a distance R at the wavelength λ, P_L is the emitted laser power, A_0 is the telescope receiving area, $\xi(\lambda)$ is the receiver's spectral transmission factor, $\beta(\lambda, R)$ is the atmospheric volume backscatter coefficient, $\zeta(R)$ is the overlap factor between the FOV of the telescope and the laser beam, $\alpha(\lambda, R)$ is the extinction coefficient, c is the light speed and τ is the laser pulse length. In this calculation one has to assume the ratio $\frac{\alpha(\lambda,r)}{\beta(\lambda,r)}$. To observe the influence of the humidity over the aerosol backscattering coefficient we looked up for a day when a sea breeze onset was observed which should bring and rise the humidity level in the lower atmosphere. To give a complementary overview of this event we used a BRAMS simulation which describes synoptically the sea breeze as a result of the pressure gradient between the ocean and the continent due the different heating processes on each environment. The sea breeze acts as a pollutant dispersion drive the same way a frontal system does.[12] The range corrected LIDAR signal should describe when this event occurs and at wich levels in the atmosphere and coupled to the weather service radiosoundings which are launched twice every day allowing to verify the humidity values within the boundary layer.

3. RESULTS

In this study we used the data obtained at IPEN/SP[10] LIDAR system on September, 11 2007. These present a interesting feature by early night when we had a sea breeze coming from Atlantic Ocean over the city of São Paulo, starting at 19:00 UTC. From the LIDAR data we obtained fourteen backscatter coefficients profiles as for every thirty minutes, between 15:00 and 21:00 UTC. The eight backscatter profiles obtained after the breeze (henceforth β_I to β_{VIII}) were compared with one chosen standard profile before the breeze (taken as β_o) in order to evaluate the extent of which this breeze could affect the aerosol characteristics over the metropolitan region. As seen in Figure 1, the signal returned by the LIDAR dramatically changes after 19:00 UTC. This observation fits with our perception of the sea breeze onset and described by the BRAMS simulation below in Figure2. In figures 03 through 05 we can see how the breeze alters the backscattering coefficient profile.

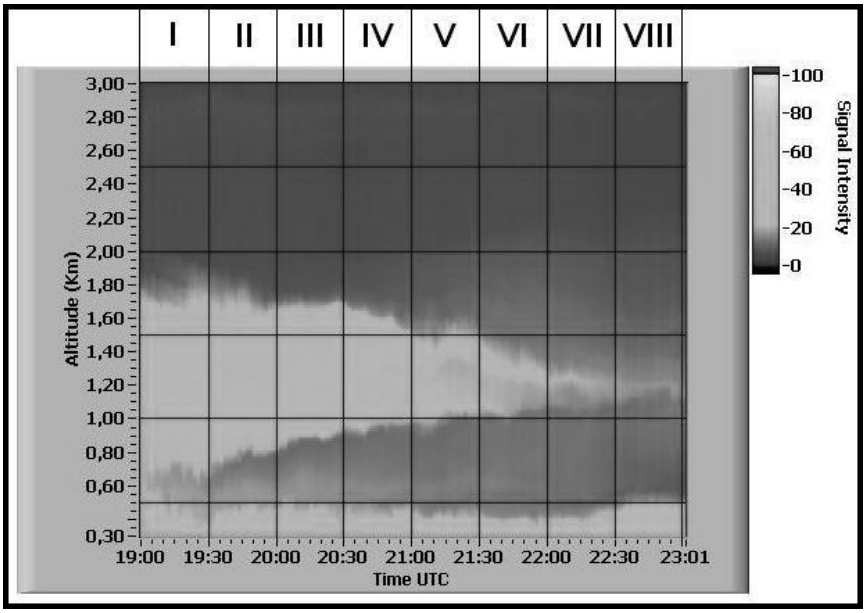

Figure 1. Range Corrected LIDAR plot showing the sea-breeze onset. The regions I-VIII refer to the different time slices taken for the analysis.

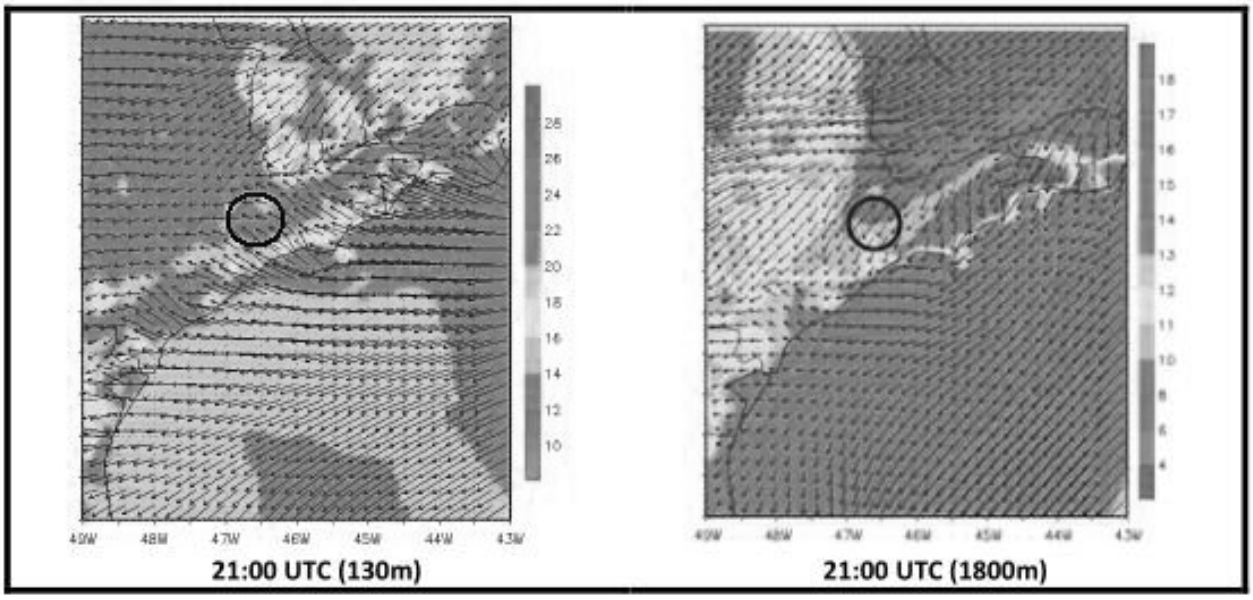

Figure 2. BRAMS generated plot to show the wind pattern at different levels, 130 m and 1800 m. The greylevel bar refers to the temperature.

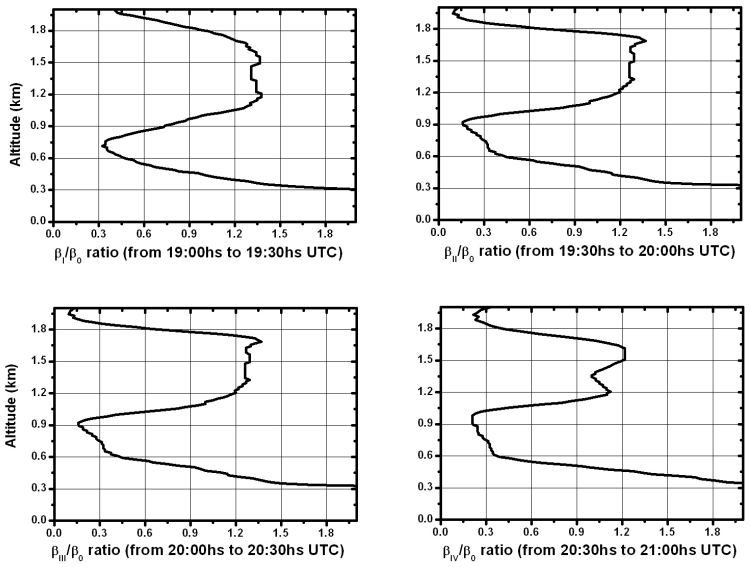

Figure 3. $\frac{\beta_{(I-IV)}}{\beta_o}$ ratios. β_o corresponds to a backscattering profile at a given time frame when the sea breeze was absent during an earlier time in this measurement.

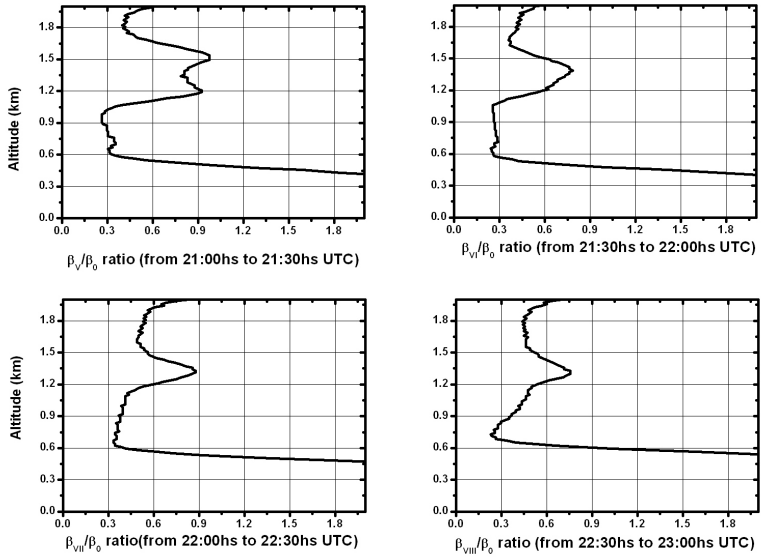

Figure 4. $\frac{\beta_{(VI-VIII)}}{\beta_o}$ ratios. β_o corresponds to a backscattering profile at a given time frame when the sea breeze was absent during an earlier time in this measurement.

Figures 3 and 4 present the $\frac{\beta_i}{\beta_o}$ for each period I through VIII. β_o corresponds to the backscattering coefficient at an earlier time previous to the sea-breeze onset and works as a reference.

Figure 5 shows the (1 - $\frac{\beta_i}{\beta_o}$) quantity indicating the trend of the sea breeze entrance on the aerosol backscattering coefficient between 1.0 and 1.8 km over the I-VIII period. It is noticeable that between period I and III this difference is negative when it switches to positive number and increases up to ≈ 0.5.

The data obtained from radiosonde taken from the weather service were used to have a relative humidity

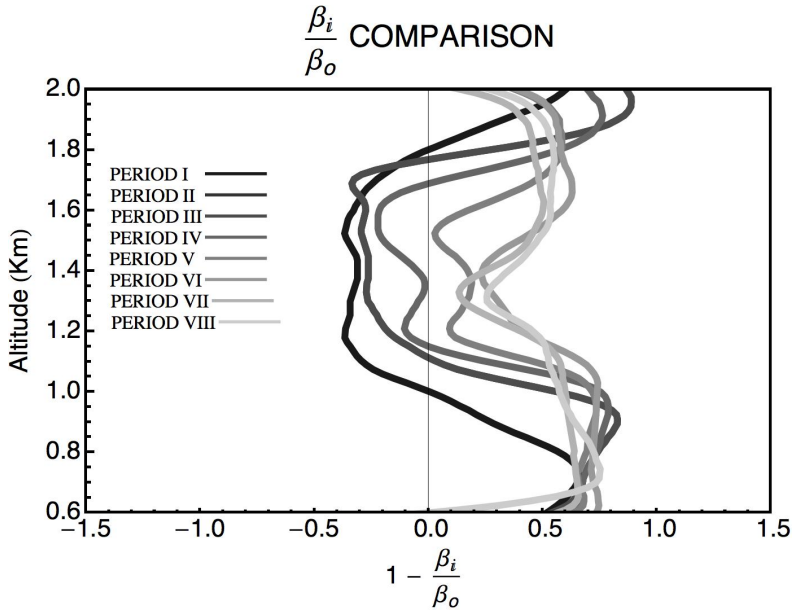

Figure 5. 1 - $\frac{\beta_i}{\beta_o}$ quantity indicating the trend of the sea breeze entrance on the aerosol backscattering coefficient between 1.0 and 1.8 km over the I-VIII period.

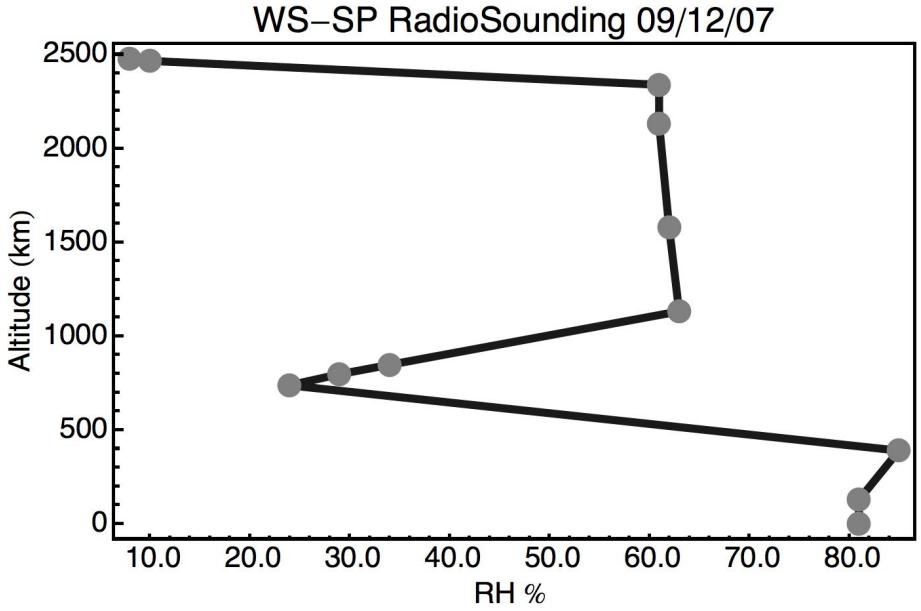

Figure 6. Relative humidity profile taken with a radiosonde as from the weather service taken at 0 UTC on September 12.

profile as depicted in Figure 6.

4. DISCUSSION

The characteristics of urban aerosols, due to their size and number distribution and chemical composition, are quite different from characteristics of marine aerosols. As seen, urban aerosols are smaller in size distributions but higher in number distributions and concentrations when compared to marine ones. Marine aerosols are expected to be more hygroscopic, due to the fraction of soluble ions, like sodium and chloride.These differences

could possible explain the effects observed on backscattering patterns. Because of the various routes by which particles can be formed, the atmospheric aerosol physical-chemical features are extremely complex and dynamic. Furthermore, individual aerosol particles rarely exist as a pure type (e.g. a pure sea-salt droplet, a specific sulphate compound). Rather, most particles are comprised of a wide range of compounds with properties and atmospheric lifetimes that differ from those of their individual components. As a result, atmospheric particles often display a wide range of chemical and physical properties. If we compare urban to marine aerosols, for example, its possible to identify a lot of differences. As seen, urban aerosols are smaller in size distributions (the particles radii are 10 to 20 times smaller and depending on Lognormal summation taken over the different types of aerosols[2]), but higher in number distribution and concentration (thousands of times per cm^3), when compared to marine ones. Marine aerosols are expected to be more hygroscopic, due to the fraction of soluble ions, like sodium and chloride.

Based on our observations of the profiles plotted on figure 02 through 05, is possible to see that for the first six analyzed periods, there is a decrease of the backscattering ratio. And for the last two periods, an increase. This may possible be explained by the changes in the aerosol population, since the breeze is expect to bring marine aerosols. Although the marine particles are bigger (and one would expect a higher backscattering, since the cross section is a size dependent function), the concentration of urban aerosol particles overcomes this difference in 2-3 orders of magnitude, and we expect to have a low backscattering when comparing marine with urban aerosol population, whereas the backscattering is strongly dependent of particle concentration (the cross-section is dependent of the number density).

From the last two profiles (VII and VIII), one can realize that the relative increase in the $\frac{\beta_i}{\beta_o}$ ratio (when compared to the first ones), could possibly be explained by the mix of urban and marine aerosol populations, or due to hygroscopic growth. The relative humidity profile suggests that there is not enough humidity to verify this phenomenon, but we have to remind that this profile was obtained from a radiosonde, not from the LIDAR. The relative humidity profile for 11 september 2007 shows an almost constant value of 63% between 1100m and 2300m. In this altitude, we have the perception, based on figure 05, that the backscattering ratios from I to VIII periods were in fact decreasing their absolute values. These results lead us to realize that the sea-breeze onset met two different types of aerosols which due the increase and presence of humidity displayed a different pattern when we observed the $\frac{\beta_i}{\beta_o}$. We should extend this procedure to other days when the same pattern was observed and also should start some simultation through Mie calculations. So far yet this method is simple for a backscattering LIDAR system it covers different aspects of aerosol-humidity interaction which can by all means be further explored with a intesive radiosounding campaign coupled to a water vapor Raman LIDAR.

5. CONCLUSIONS

We conducted an experiment using a single-wavelength backscatter LIDAR (532 nm), combined with a sea-breeze onset phenomenum and able to qualitatively describe a hygroscopiceffect of aerosols over São Paulo metropolitan area. To test this factor assessment we employed data obtained in a single day, namely on 11 September 2007, when a well characterized humidity intrusion is onset due the transport of water vapor by a sea-breeze phenomenon. For this data, we calculated the backscatter coefficient at 532 nm, and used this parameter to observe possible hygroscopic aerosol swallowing, assuming a well-mixed boundary layer where a cloud cap condition is present or a well defined and pronounced mixing layer boundary. These assumptions guarantee that any changes in the backscatter coefficient could be due to changes in relative humidity coupled to a mixing in aerosol size and type distribution.

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