# Indirect aerosol hygroscopic growth observations with a backscattering lidar, part II: Five day breeze onset data analyses

Patricia Ferrini Rodrigues[a], Eduardo Landulfo[a], Adilson Wagner Gandu[b], Fabio Juliano da Silva Lopes[a], Renata Facundes da Costa[a], Walter Nakaema[a]

[a]IPEN-CNEN/SP, Av. Lineu Prestes 2242, São Paulo, Brazil.
[b]IAG/USP, Rua do Matão 1226, São Paulo, Brazil.

## ABSTRACT

Atmospheric aerosol particles have received much attention in recent years due to their importance in climate change. The influence of these particles on Earth's radiative budget depends on a number of factors, including their size distribution and chemical composition. This work addresses a particular property of aerosols, namely, the extent to which they have affinity for water vapor. The size increase of aerosol particles resulting from water vapor uptake has important implications for the direct scattering of radiation and cloud droplets formation. We used a single-wavelength backscatter LIDAR (532 nm), and relative humidity profiles obtained from radiosounding to assess the hygroscopic growing factor of aerosols over Sao Paulo metropolitan region, for five days altogether on March and September 2007 and August 2009. In these days we had a breeze onset over the metropolitan area, potentially bringing marine aerosols and humidity from the Atlantic Ocean. In this way we were able to detect a change in the boundary layer aerosol optical properties during these onsets using range corrected backscattering signal from LIDAR and a detailed analysis on the changes in backscattering coefficient profiles by a Klett analysis. In order to infer the hygroscopic growing factor, we developed a fitting model algorithm, proposed in the literature, calculating the backscattering coefficient at 532 nm for periods before and during the breeze and comparing the same profiles at various altitude levels with a reference profile at the lowest relative humidity level whithin the mixing layer. In addition, we performed a comparison between the thirty minutes backscattering profiles inside the breeze with a reference thirty minutes backscattering profile before the breeze.

**Keywords:** Hygroscopic Growth, Aerosols, LIDAR, Sea breeze

## 1. INTRODUCTION

The size increase of aerosol due the water uptake has important effects on direct radiation scattering (direct effect), but it has also influences on indirect effects, related to the capacity of this aerosol population to work as CCN (cloud condensation nucleus) - i.e. the ability of an aerosol particle to grow its liquid water content and form cloud droplets. As clouds contribute for the enhancement of the albedo from earth, the indirect effect leads to a radiative cooling of the global system. This feature depends on the chemical nature of aerosol population and it is known that working as a CCN is common in more hygroscopic aerosols. There are some particles, like black carbon, which work more as absorbers than CCN, leading to a decrease of the total albedo of the earth.[1]

At low relative humidities, aerosols particles containing salts remain solid. As the RH increases, the particles still remain solid until a threshold RH, characteristic of each aerosol, is reached. At this RH (known as deliquescence relative humidity, DHR), the solid particle spontaneously absorbs water, producing a saturated aqueous solution. If the RH increases, there is additional condensation into the salt, to maintain the thermodynamic equilibrium. If there is a decrease in RH, evaporation occurs, but in general there is no crystallization is the

Further author information: (Send correspondence to Patricia Ferrini Rodrigues)
E-mail: patricia.ferrini.rodrigues@usp.br

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solution when de DHR is reached. This behavior is known as hysteresis, it is, the point of deliquescence and crystallization are different.[2]

As the relative humidity (RH) of the environment increases, condensation of water vapor may occurs over aerosols, depending on their chemical properties. This phenomenon leads to an increase of the size of the particles (hygroscopic growth) and consequently causes changes in size and refractive index of the particles. Therefore, significant variations in the backscattering signal detected with a LIDAR are expected when changes in RH are observed.[3] This is particularly true for high RH levels, where hygroscopic growth of aerosols is more pronounced.[2] Generally, as the water uptake of the particle become more pronounced, the real and imaginary parts of the refractive index decrease. As the real and imaginary part of the refractive index of pure water is lower than the one of dry particles, and its imaginary part is zero, it would suggest a decrease in backscattering as RH increases. But as scattering is a function of size and refractive index, both effects need to be taken into account. In fact, as the cross section is dependent of the square of the particles radius, the decrease in refractive index is not large enough to counteract this effect. The size dependence dominates, then, leading to an increase in backscattering.[2]

LIDAR has several advantages over other methods on measuring hygroscopic growth. Foremost, the fact that this remote sensing system is able to measure changes in backscattering under unperturbed atmospheric conditions, besides the fact that the range of measurements can be extended to very close to saturation, as the traditional methods using nephelometers can not expose dry samples of particles to a relative humidity over 85%, the region where particles experience their most noticeable growth.[4]

In this work, we tried to evaluate the hygroscopic growth of aerosols over Sao Paulo, using data obtained from a single wavelength LIDAR operating at 532 nm. For this purpose, we selected backscattering data for five days of breeze onset over the metropolitan region in 2007 and 2009, aiming to evaluate relative differences in backscattering of aerosol population before and during the breeze, as it is expected to bring additional aerosol species and humidity from the sea. For this purpose, we used relative humidity profiles from radiosounding, and chose a reference backscattering half hour profile before the breeze onset to compare with half hour profiles during the passage of the breeze. Henceforth, we used a fitting model of aerosol hygroscopic growth factor described on the literature to compare the aerosol population inside the breeze subjected to different levels of relative humidity.

# 2. METODOLOGY

## 2.1 EQUIPMENTS

### 2.1.1 LIDAR

We have chosen 5 days (7, 10, 11 and 12 September 2007 and 27 August 2009), in which we could verify and characterize breeze onsets over the metropolitan region of São Paulo. These onsets could be verified using the LIDAR system located at the Nuclear and Energy Research Institute (IPEN), in São Paulo

The LIDAR system employed in this work 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(neodymium:yttium/aluminium/garnet) laser (Brillant by Quantel SA) operating at the second harmonic frequency (SHF), 532 nm, with a fixed repetition rate of 20 Hz. The emitted laser pulses have a divergence of less than 0.5 mrad after expansion. A 30 cm diameter telescope (Focal length =1.5m) is used to collect the backscattered laser light. The telescope's field of view (FOV) is variable (0.5 mrad) by using a small diaphragm. The system is currently used with a fixed FOV of 1mrad, which permits a full overlap between the telescope FOV and the laser beam at heights around 300 m above the ground level. This FOV value, in accordance with the detection electronics, permits the probing of the atmosphere up to the free troposphere (12-15 km).

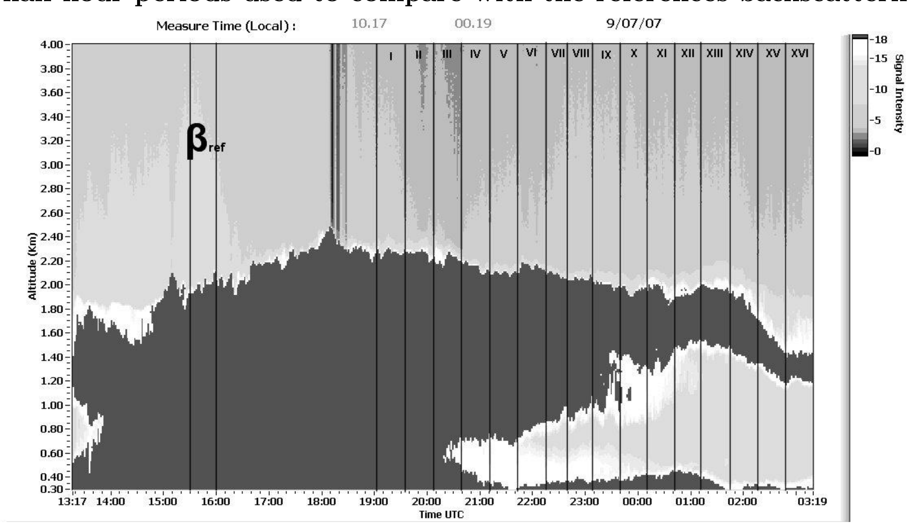
The backscattered laser radiation is then sent to a photomultiplier tube (PMT) coupled to a narrowband (1 nm FWHM) interference filter to assure the reduction of the solar background during daytime operation and to improve the signal-to-noise ratio (SNR) at altitudes greater than 3 km. The PMT output signal is recorded by a transient recorder in both analog and photoncounting mode. Data are averaged between 2 and 5 min and then summed up over a period of about 30 min, with a spatial resolution of 3.75 m for the day in 2009 and 15m for days in 2007.[5]

Table 1: **The IPEN LIDAR system summarized features**

| | |
|---|---|
| **Laser Energy/pulse** | Up to 130 mJ |
| **Telescope Config.** | Newtonian |
| **Telescope Diam.** | 300 mm |
| **Telescope F#** | 5 |
| **Detection Channel** | 532 nm |

Using this LIDAR system, a breeze onset was detected by the different feature on the curtain plot given at Figure 1, which refers to the days chosen. We divided each day into two groups of backscattering analyses, one of them using atmospheric data from the first radiosounding (1200 UTC) and the second using atmospheric data from the second radiosounding (0000 UTC of the next day). After that we divided the days of measurement is groups of half hour each, focusing on the profiles of the second group, which contain the breeze onset. These profiles were compared with one standard profile ($\beta_{ref}$) chosen in a moment before the breeze. To choose this profile, we considered the absence of clouds and proximity to period where the boundary layer was next to maximum altitude, to grant a better mixing of aerosols. Then, the profiles containing the breeze onset were compared with the reference profile calculating the displacement $1 - \frac{\beta_i}{\beta_{ref}}$, in order to verify the effects of the breeze onset over the urban aerosol population and its evolution in time. The results will be shown and discussed next section.

Figure 1: **Range corrected LIDAR signal for 07 September 2007. It is possible to see the breeze onset and the half hour periods used to compare with the references backscattering profile.**



### 2.1.2 RH PROFILES FROM RADIOSONDES

To obtain information about the relative humidity profiles for the chosen days, we used data from radiosounding. The soundings are launched twice a day, at 12OO UTC and 0000 UTC and are distant about 10km from the place where the LIDAR is located. It is possible to calculate relative humidity profiles from radisoundings using the temperature, the dew point temperature and Clausius Clapeyron equation (the values of RH obtained this way are provided as product of these mesurements). As the soundings provide information for five or six levels of altitude below the planetary boundary layer, we created an interpolation function of the values at those levels, to obtain information for all points in the same spacial resolution of the LIDAR (3.75 m for the day in 2009 and 15m for days in 2007).

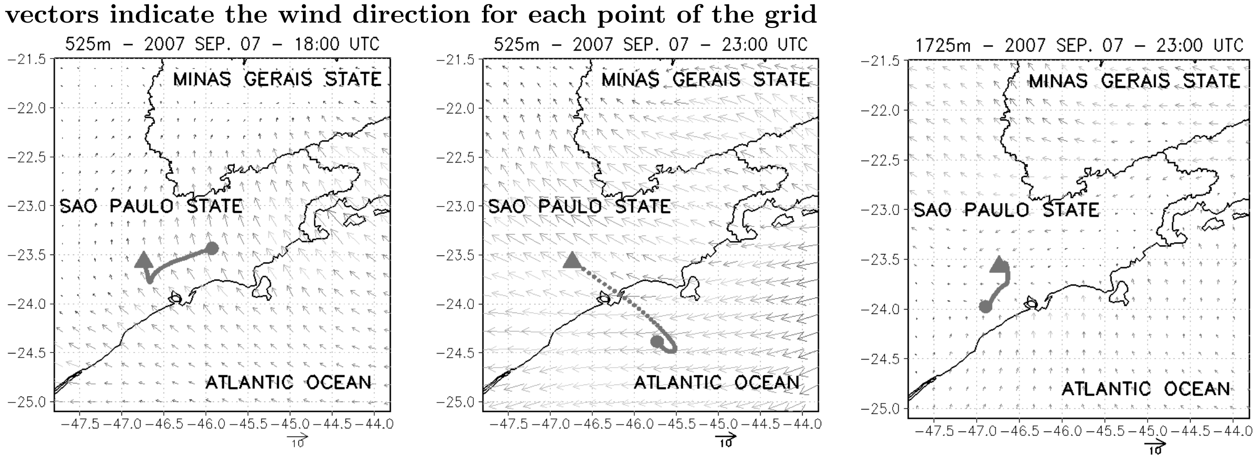
## 2.2 DATA ANALYSIS

### 2.2.1 BRAMS

BRAMS (Brazilian developments on the Regional Atmospheric Modelling System) is the brazilian adaptation for the version 5.04 of RAMS (Regional Atmospheric Modelling System), a numerical prediction model that simulates atmospheric circulations, being most frequently used to simulate atmospheric phenomena on the mesoscale (horizontal scales from 2 km to 2000 km) for applications from operational weather forecasting to air quality regulatory uses.[6]

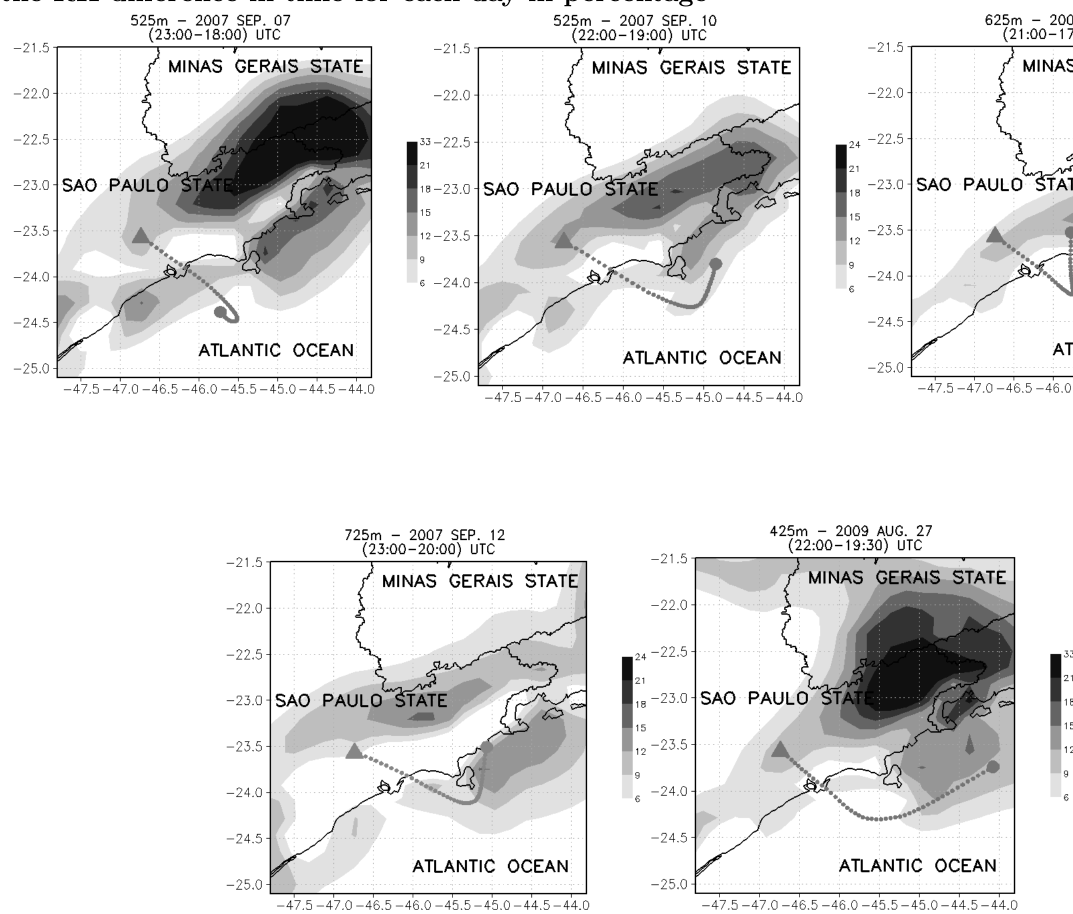
We ran the model for the four days in 2007 and for the day in 2009 using a horizontal grid of 25 km, aiming to model characteristics of the sea breeze onset over the metropolitan region during these day. Figure 2 show the origins of the air parcels over the metropolitan region ($23° 32' 51'' S$ and $46° 39' 10'' W$) for 07 september 2007 for tree moments of interest, with the circle indicating the origin and the triangle indicating the destiny. The first for each day is at the same altitude of the breeze, but before the onset. The second and the third for each day is at a moment where the onset was already detected by the LIDAR, at the altitude of the breeze and above it, respectively. These BRAMS analyses were performed to bring information about the origin of the breeze detected by the LIDAR and consequently of the aerosol population and characteristics of relative humidity. The analyses were consistent with the perception of a marine origin of the air parcels during the breeze. Before and above the onset is possible to see that the origin of the air parcel is continental. All the other days showed the same agreement (data not shown).

Figure 2: **BRAMS analysis for 07 September 2007. "10" indicates the wind speed (m/s) and the vectors indicate the wind direction for each point of the grid**



Also, using BRAMS, we calculated the differences in RH levels before and during the breeze onset over the metropolitan region, as the breeze is expected to bring humidity from the ocean. Figure 3 shows the differences in percentage, for a specific moment before and during the breeze for each day. There is also an air parcel trajectory within these two moments, with the circle indicating the origin and the triangle indicating the destiny. For all the days is possible to see an increasing in RH of about 12 percent.

Figure 3: **Differences in RH before and after the breeze onset. The figures show the altitude and the RH difference in time for each day in percentage**




### 2.2.2 THE HYGROSCOPIC GROWTH ASSESSMENT

To minimize errors due to the distance between the LIDAR and the radisounding, we have chosen five thirty minutes backscattering profiles during the breeze and close to the time the radiosounding data were available (00:00 UTC).

After that, we selected the minimum and the maximum altitude of the breeze for each day using the LIDAR range corrected backscattering signal, and the respective values of RH and backscattering for the altitude range inside the breeze. Then we selected the lowest RH and its respective backscattering value and then compared all the other backscattering values with this reference one, aplying in sequence the fitting model described in Tardif[2] and Im,[7] shown in sequence.

$$\frac{\beta_a}{\beta_0} = a(1 - (\frac{RH}{100}))^{-b} \tag{1}$$

where $\beta_a$ is the backscattering value for aerosol subjected to a RH value higher than the reference one, and $\beta_0$ is a reference backscattering value for a chosen level of RH. In this work we used a particular value of $\beta_0$ for each day. The fitting parameter **b** can be used to efficiently describe the variability of the data set when its mean value and standard deviation are known.[8,9] The parameter **a** is representative of the total scattering of aerosol population. The values will be shown and discussed in sequence.

Table 2: **Parameters for the five days chosen**

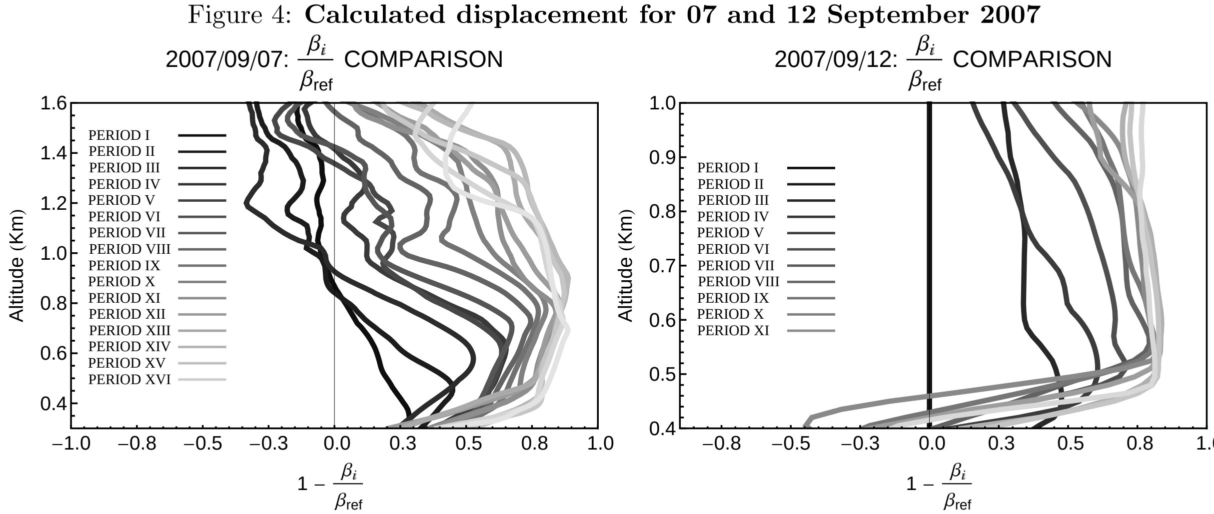
| Date | Half Hour Periods used in the fitting | Half Hour Periods Used as $\beta_{ref}$ | Lowest RH value (LRH) | Altitude of LRH | Altitude of breeze |
|---|---|---|---|---|---|
| 09/07/2007 | 22h16min - 00h42min | 18h34min - 19h04min | 40% | 421 m | 300m to 1600m |
| 09/10/2007 | 20h54min - 23h16min | 19h20min - 19h49min | 48% | 843 m | 350m to 1100m |
| 09/11/2007 | 20h30min - 23h00min | 17h00min - 17h30min | 24% | 736 m | 400m to 1000m |
| 09/12/2007 | 22h41min - 01h21min | 18h59min - 19h29min | 73% | 617 m | 450m to 1000m |
| 08/27/2009 | 20h46min - 23h06min | 19h44min - 20h14min | 18% | 788 m | 300m to 900m |

# 3. RESULTS

## 3.1 Comparisons with a reference profile before the breeze

Figure 4 shows the calculated $1-\frac{\beta_i}{\beta_{ref}}$ for 07 September 2007 and 12 September 2007, evidencing the temporal evolution of the effects of the sea breeze onset over the aerosol backscattering coefficients. For those days was possible to see a pattern of displacement to most positive values, indicating that there is a progressive decrease of value of backscattering inside the breeze (as $\beta_{ref}$ remais constant, a decrease in $\beta_i$ would result in lower values for the fraction). This patters could be observed in all days analysed (data not shown). For 07 September 2007 there is also a shifting inside the breeze, with a displacemente to most negative values (compared with lower altitudes inside the breeze), indicating an increase of backscattering. This findings could be explained by the two effects acting over the aerosol population, wich are the effect of size and the effect of changing the aerosol population (see duscission next section).It is also possible too see the results and discussion for 11 September 2007 in Rodrigues at all.[10]

Figure 4: **Calculated displacement for 07 and 12 September 2007**



### 3.2 Hygroscopic growth factor modeling

The results of the fitting adjusted according to the equation 1 are show in Figure 5 for 07 September 2007 and 12 September 2007 and in Table 3 for all the days analysed. It is also possible too see the results and discussion for 11 September 2007 in Rodrigues at all.[10] We can see here a big variation between the parameter of the regression (**a** and **b**), presented in the form: mean value for the half hour profiles evaluated and their respective mean standard deviation (Table 3). We also performed the error propagation for each day to evaluate the quality of the fittings (Table 4). As the parameter **b** is a measurement of the variability of the data, it is possible to see the higher dispersion of data for higher values of this parameter. As **a** is a property of the aerosol population, the variability between the values encountered is lower.

Figure 5: **Fitted model for 07 and 12 September 2007**

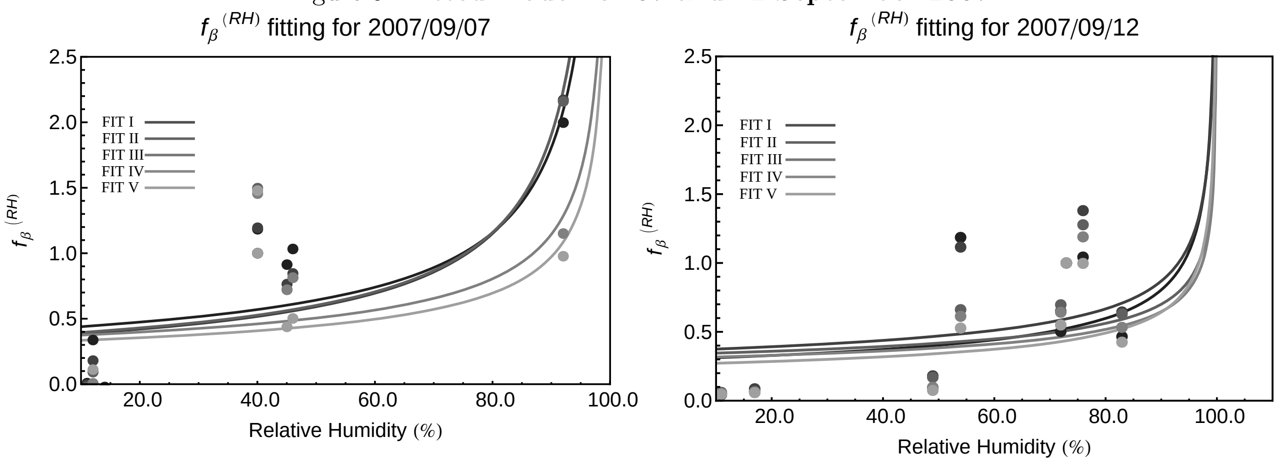



Table 3: Mean values of **a** and **b** for each day and the respectives mean deviations

| Date | a values | b values |
|---|---|---|
| 09/07/2007 | 0.361±0.021 | 0.616±0.095 |
| 09/10/2007 | 0.231±0.015 | 1.440±0.186 |
| 09/11/2007 | 0.593±0.155 | 0.671±0.300 |
| 09/12/2007 | 0.312±0.028 | 0.370±0.037 |
| 08/27/2009 | 0.256±0.077 | 1.480±0.255 |

## 4. DISCUSSION AND CONCLUSION

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). 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.[11]

The characteristics of urban aerosols, due to their size and number distribution and chemical composition, are quite different from characteristics of marine aerosols. Urban aerosols are smaller in size distributions but higher in number distributions and concentrations when compared to marine ones.[12] Urban aerosols are smaller

Table 4: Mean values of **a** and **b** for each day and the respectives fitting errors

| Date | a values | a error | b values | b error |
|---|---|---|---|---|
| 09/07/2007 | 0.361 | 0.028 | 0.616 | 0.034 |
| 09/10/2007 | 0.231 | 0.042 | 1.440 | 0.177 |
| 09/11/2007 | 0.593 | 0.045 | 0.671 | 0.143 |
| 09/12/2007 | 0.312 | 0.018 | 0.370 | 0.009 |
| 08/27/2009 | 0.256 | 0.032 | 1.480 | 0.500 |

in size distributions (the particles radii are 10 to 20 times smaller and depending on Lognormal summation taken over the different types of aerosols), but higher in number distribution and concentration (thousands of times per $cm^3$), when compared to marine ones.[13] Although the marine particles are bigger (and one would expect a higher backscattering, since the cross section for particles light scattering 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). These differences in size and concentration could possible explain the effects observed on backscattering patterns, which enables the LIDAR system to detect the breeze onset.

On the other hand, marine aerosols are expected to be more hygroscopic, due to the fraction of soluble ions, like sodium and chloride.[13] As the breeze evolves, is expected to observe a hygroscopic growth of marine aerosols, and the fitted model is an indicative of this fenomenum. But, as we can see in the results of the comparisons between backscattering before and during the breeze, there is a positive displacement, indicating the decrease of values of backscattering in the temporal evolution of the breeze. This could indicate that the predominant effect is of the aerosol concentration, not the hygroscopic effect, when we think about the mixing between urban and marine aerosols. Wulfmeyer and Feingold[14] found that aerosols with large mass fractions of soluble material exhibit a steeper increase in backscatter when compared to partially soluble particles.

Additionally, this fitting model has been found to adequately describe the light scattering hygroscopic growth for some nondeliquescent aerosols.[9] But in a study done by these same authors with biomass burning in Brazil, where the aerosol was primarily composed of organic compound and hygroscopic growth was inhibited, the found this model performed poorly, as it is based on the equilibrium growth of aerosols with RH and therefore will not be accurate for deliquescent samples.[9]

Tardif,[2] performing the same modeling we applied in this work, found values of $\mathbf{a = 0.43}$ and $\mathbf{b = 0.72}$ for a fixed level of relative humidity of 70%. Im[7] adopted a fixed value $\mathbf{a = 1}$ and a minimum value of RH of 30% and in their studies they found $\mathbf{b = 0.38}$ for polluted continental, continental and marine aerosols. In this work, we adopted different levels for minimum RH for each day and this methodology probably introduced another source of variability to the data. Also, we had only feel points to adjust de model, what diminishes its statistical relevance.

Here it is not possible to assume a well-mixed boundary layer, because there is no cloud cap and the breeze onset is expected to bring another population of aerosols. As pointed by Tardif,[2] without a complete knowledge of the aerosols present in the LIDAR volume, it iss not possible to differentiate if the changes in backscattering are due to the effect of the hygroscopic growth or due changes in aerosol concentration. But, it is possible to assume a well mix aerosol population inside the breeze in its evolution in time, as the bassckscattering values do not have dramatic changes after a few hours. In this case, the behavior of the backscattering pattern inside the breeze is supposed to be due to the increasing in the liquid content in the aerosol particles. 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 an intensive radiosounding campaign coupled to a water vapor Raman LIDAR.

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