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Assessment of Aerosol Hygroscopic Growth Using an Elastic LIDAR and BRAMS Simulation in Urban Metropolitan Areas

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Abstract. Atmospheric aerosol particles have received attention recently due to their importance in climate change and cloud formation. The influence of these particles on Earth's radiative budget depends on a number of factors, including their size distribution and chemical composition. The size increase of aerosol particles due to water vapor uptake has important implications for the direct scattering of radiation and cloud droplets formation. The extent of the particles' affinity for water vapor depends on chemical composition and atmospheric parameters such as water vapor availability. We used a single-wavelength backscatter LIDAR (532 nm) and relative humidity profiles obtained with radiosondes to study particle growth over the Sao Paulo metropolitan region, Brazil, under different conditions of water vapor availability. On this day we had a breeze onset over the urban area, potentially bringing marine aerosols and humidity from the Atlantic Ocean. To simulate the breeze onset and path of the air parcels, we ran the BRAMS model. To infer the hygroscopic growth factor, we developed a fitting model algorithm, already proposed in the literature, calculating the backscatter coefficient at 532 nm for twenty-minute periods after and during the breeze onset and comparing backscattering at various altitude levels with a reference backscattering at the lowest relative humidity below the breeze, i.e., below the base of a cloud cap. In addition, we compared the twenty minutes backscattering profiles inside the breeze to a reference twenty minutes backscattering profile before the breeze, expecting to infer some aerosol hygroscopic properties over the Sao Paulo urban area.

Keywords: LIDAR, BRAMS, Hygroscopic growth, Aerosols.
PACS: 92.60.Mt

INTRODUCTION

The ability of an aerosol population to work as CCN (Cloud Condensation Nuclei) and form cloud droplets depends on the chemical nature of aerosol populations, and functioning as a CCN is common in more hygroscopic aerosols. Hygroscopicity describes the ability of an aerosol particle to grow its liquid water content when water vapor is available in the atmosphere, i.e., it is related to relative humidity levels. As clouds contribute to the enhancement of earth albedos, the indirect effect leads to radiative cooling of the global system. There are some particles like black carbon which work more as absorbers than CCNs, leading to a decrease in total earth albedo [1].

As the relative humidity (RH) of the environment increases, condensation of water vapor may occur over aerosols. This phenomenon leads to an increase of particle size and consequently causes changes in size and refractive index. Therefore, significant variations in the backscattering signal detected with a LIDAR are expected when changes in RH are observed [2]. This is particularly true for high RH levels, where hygroscopic growth of aerosols is more pronounced [3].

LIDAR has several advantages over other methods on measuring hygroscopic growth. Foremost, this remote sensing system is able to measure changes in backscattering under unperturbed atmospheric conditions, and the range of measurements can be extended 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 one day of breeze onset over the metropolitan region in 2011, aiming to evaluate relative differences in backscattering of aerosol populations 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 radiosonde, and chose a reference

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backscattering twenty minutes profile before the breeze onset to compare with twenty minutes profiles during the passage of the breeze. Thereafter, we used a fitting model of aerosol hygroscopic growth factor described on the literature to compare the aerosol population above the breeze subjected to different levels of relative humidity. For that, we used a cloud cap formation at the lower limit of the breeze as a calibration point.

METHODOLOGY

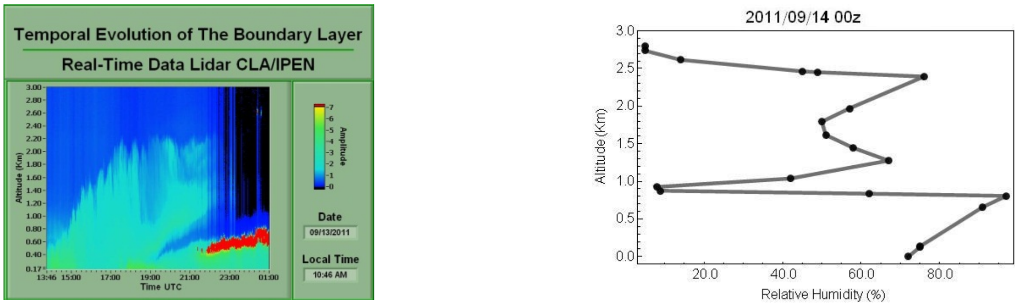
LIDAR

We selected one day (13 September 2011), in which we could verify and characterize a breeze onset over the metropolitan region of São Paulo. This onset could be verified using the LIDAR system located at the Nuclear and Energy Research Institute (IPEN), in São Paulo.

The IPEN LIDAR system 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 (Brillant by Quantel SA) operating at the second harmonic frequency (SHF), 532 nm, with a fixed repetition rate of 10 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). Data are averaged each 2 minutes and then summed over a period of about 20 min, with a spatial resolution of 3.75 to 15 m.

RH Profiles from Radiosonde

To obtain information about the relative humidity profiles, we used data from radiosondes, which are launched twice a day, at 1200 UTC and 0000 UTC and are distant about 10km from where the LIDAR is located. It is possible to calculate relative humidity profiles using the temperature, the dew point temperature and Clausius-Clapeyron equation (the values of RH obtained this way are provided as products of these measurements).



(a) (b)

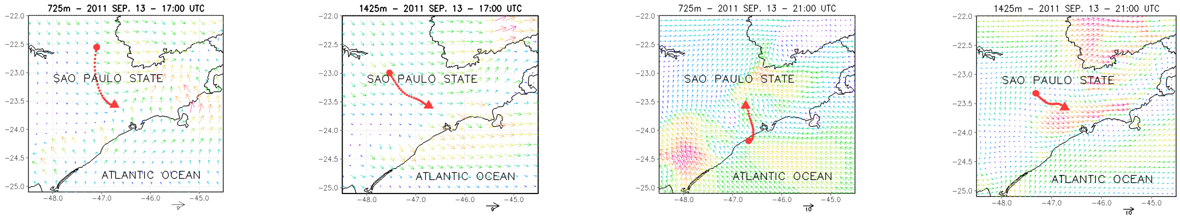
FIGURE 1. (a) Range corrected plot of the LIDAR signal showing the breeze onset with a cloud cap formation at about 600m beginning at 2200 UTC. (b) Radiosonde plot for 14 Sept. 2011, showing a sharp increase in RH until about 900 meters.

DATA ANALYSIS

BRAMS

BRAMS (Brazilian developments on the Regional Atmospheric Modeling System) is the Brazilian adaptation for the version 5.04 of RAMS (Regional Atmospheric Modeling System), a numerical prediction model that simulates atmospheric circulations, for applications from operational weather forecasting to air quality regulatory uses [5].

We used the model for this day in 2011 using a horizontal grid of 25 km, aiming to model characteristics of the sea breeze onset over the metropolitan region. Figure 2 (a) and (b) show the origins of the air parcels over the metropolitan region for two moments of interest, with the circle indicating the origin and the triangle indicating the destiny. These two moments were taken before the breeze at two different levels of altitude, indicating that the air mass is coming from the continent. Figure 2 (c) and (d) shows the simulations at the moment of the breeze onset, at two different levels of altitude, showing that above the breeze the air mass is coming from the continent and for the level of the breeze the air mass is coming from the ocean.



(a) (b) (c) (d)

FIGURE 2. BRAMS generated simulations of the air parcel's trajectory at 725m (a) and 1425m (b) before the breeze onset, showing the air mass comes from the continent, and the air parcel's trajectory at 725m (c) and 1425m (d) during the breeze onset showing the air mass is coming from the continent at a higher altitude and from the ocean at the breeze altitude.

These BRAMS analyses were performed to gain information about the origin of the breeze detected by the LIDAR and consequently of the aerosol population and characteristics of relative humidity. To give us more information about the relative humidity, we performed a simulation of the differences in relative humidity (%) before and after the breeze onset. Figure 3(a) shows these differences. It is possible to see that the breeze onset is bringing more humidity and increasing around 25 – 30% the relative humidity at the metropolitan region.

Backscattering Comparisons

Using the LIDAR and analysis routines, we calculated the backscattering pattern averaged every twenty minutes during breeze onset. These profiles were compared with one standard profile (β*ref*) chosen in a moment before the breeze. To choose this profile, we considered the absence of clouds and proximity to a period where the boundary layer was next to maximum altitude, to give 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.

Determining the Hygroscopic Growth Factor above the Cloud Cap

We selected three 20-minute data sets around the time when the radiosonde was launched, and calculated relative humidity using water vapor mixing ratio from LIDAR and temperature and pressure from radiosonde. After obtaining the backscattering and the relative humidity, we calculated the hygroscopic growth factor, $f_{RH}=\frac{\beta_a}{\beta_0}$, where β_a is the backscattering value for aerosol subjected to a RH value higher than the reference one, and β_0 is a reference backscattering value for a chosen level of RH. In this work we used the particular value of β_0 of 72% for 09/13/2011. This reference level was the lowest value of the relative humidity for the data studied. In sequence, we fitted a curve to the data, in the form [6]:

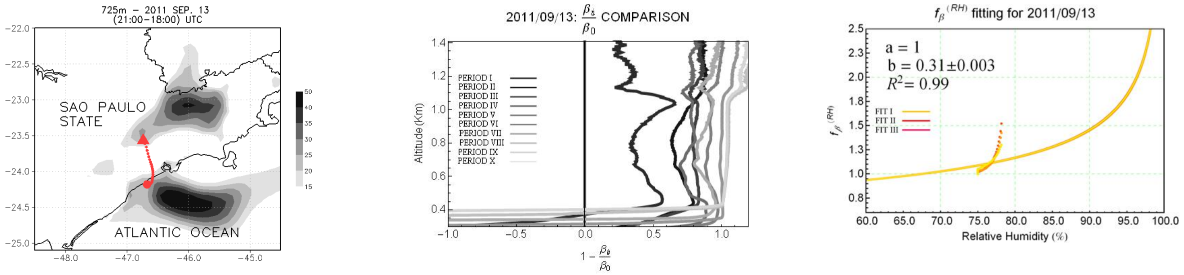
$$f_{RH}=a\left(1-\left(\frac{RH}{100}\right)\right)^{-b} \qquad (1)$$

where *a* represents the total light scattering for the dry aerosol and *b* is a fitting parameter used to describe the variation of the data set.

We took three periods of 20 minutes each and calculated the hygroscopic growing factor using a lowest level of RH = 72% and set the cloud base as the stop point. We fit results to the curve described by equation 1. As the radiosonde is launched 10 km from the LIDAR and showed increasing relative humidity in the altitudes of interest, we used an interpolation function to obtain the relative humidity at the same heights of the LIDAR data.

RESULTS AND DISCUSSION

The displacement calculated using a reference backscattering profile is shown below. As we have a cloud cap, for altitudes above the breeze, the backscatter values go quickly to high values. At the same altitude of the breeze, the ratio $\frac{\beta_i}{\beta_{ref}}$ goes to smaller values, showing there is progressive cleaning of the aerosols by the breeze. No effects of the relative humidity causing the hygroscopic growth could be verified, but it may be due to the presence of the cloud cap causing the attenuation of the LIDAR signal. Other works [7] using breeze onset datasets show that it is possible to see some changes in the backscattering pattern suggesting a growth of the aerosol population.



(a) (b) (c)

Figure 3. (a) Difference (%) between the RH level at 725m during the breeze onset, showing the increase of about 25-30% in the metropolitan region. (b) Displacement calculated using a reference backscattering profile outside the breeze and 10 profiles during the breeze onset. Note the progressive decrease of backscattering values. (c) Fitting curve for 09/13/2011 for the three twenty-minute periods considered for analysis.

For the fitting above the cloud cap formation, the highest point to be considered for 09/13/2011 was chosen using the images of the backscattering pattern for this day, combined with the relative humidity obtained by the radiosonde. The maximum RH in altitude for 09/13/2011 is 95%, and then we have a ratio $\frac{\beta_{95\%}}{\beta_{72\%}}$. As the cloud cap formation is about 400m to 500m from the ground, only a few points could be used for analysis. With these few available points, we fixed the value of a equal to one and adjusted b, as done by Im [7], finding a value for b equal to 0.31.

As we considered only a few points above the cloud base, it was difficult to conclusively derive the backscattering pattern. Thus we used points when we had clear sky during the breeze period as reference point for Klett analysis.

For one experiment made in western North Carolina, Im et al. [7] encountered values for b equal to 0.38±0.03 with an R^2=0.94 for polluted continental air masses, 0.37±0.05 with an R^2 = 0.84 for continental air masses, and 0.38±0.05 with an R^2 = 0.85 for marine air masses; the hygroscopic growth factor was calculated to be 1.61 for polluted continental air masses, 1.61 for marine air masses, and 1.59 for continental air masses, indicating that the hygroscopic growth factor is nearly constant for the three different air masses. They used a ratio from $\frac{\beta_{80\%}}{\beta_{30\%}}$ in their work, maintaining a constant and equal to 1 [7].

Further developments of this work include a better understanding of the behavior of the breeze using more BRAMS simulations, like radiosonde simulations at the same site, sources of marine aerosols, wind speed and vertical wind speed, and an implementation of a new Raman LIDAR system to measure the relative humidity at the same site.

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