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Aircraft observations of the interactions of the Manaus plume with aerosols forest during rainy season: a case study

Observações por avião da interação da pluma de Manaus com os aerossóis da floresta durante a estação chuvosa: um estudo de caso

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Abstract

Measurements of aerosol size distribution and cloud condensation nuclei (CCN) efficiency over the Amazon Region close to the city of Manaus, Brazil, were taken during an airborne experiment in February – March (rainy season) 2014 as part of the Green Ocean Amazon Experiment (GOAmazon). A variety of aerosols were observed following the aging along the downwind Manaus plume, in a relatively well preserved portion of Amazon basin. At this work we evaluate how those aerosols interact with the Amazonian environment for the situation observed during 13th march 2014. The aerosols size distribution is studied considering its aging downwind the Manaus plume. The concentration of CCN inside the plume is evaluated and a large enhancement is observed.

Keywords: CCN, Amazon Aerosols, rain forest, urban pollution, hygroscopicities

Resumo

Medidas da distribuição de tamanho de aerossóis e eficiência de núcleos de condensação de nuvens sobre a região amazônica, próximo a cidade de Manaus, foram realizadas durante a estação chuvosa de 2014 (fevereiro e março). Uma variedade de aerossóis foram observados seguindo o envelhecimento da pluma de Manaus vento a baixo, numa região relativamente bem preservada. Neste trabalho avaliamos como estes aerossóis interagem com o ambiente da floresta para a situação de 13 de março de 2014. A distribuição de tamanho de aerossóis é estudada considerando seu envelhecimento na pluma. A concentração de núcleos de condensação de nuvens dentro da pluma é avaliada e observa-se um aumento significativo.

1. Introduction

When anthropogenic aerosols are added to natural aerosol the concentration of cloud condensation nuclei (CCN) in atmosphere can increases and influence the characteristics of clouds formed in such an environment. The indirect radiative effects of aerosols through their role in cloud microphysics are complex and contribute the largest uncertainty in radiative forcing calculations (IPCC, 2007).

As large cities are major source regions of aerosols and its precursors, the urban plume downwind of Manaus can impact the cloud properties of the otherwise pristine Amazonian environment.

The anthropogenic emissions from Manaus areas can result in modifications of the aerosol size distribution (ASD) and particle hygroscopicity (κ) relevant for climate several hundred kilometers downwind. Hence, it is essential to understand how those particles age, mix and transform in conjunction with long-range transported background aerosols, so they can be accurately represented and incorporate into regional and global climate models.

In this work we evaluate aerosol properties during one flight experiment on the Amazon Region close to the city of Manaus, Brazil, observed during an airborne measurements during rainy season of 2014 as part of the Green Ocean Amazon (GOAmazon), Intensive Airborne Experiment in Amazonia (IARA). During IARA in situ ASD and N_{CCN} at different supersaturation (SS) were measured using aircraft, which allowed to estimates the influence of the Manaus plume over the amazon environment.

2. The region of observation

Manaus is an isolated, two million people urban area within an otherwise pristine rain forest. It holds a free trade zone, manufacturing products mostly shipped to all consumer markets in Brazil. Despite being recently integrated to Brazilian electrical energy grid, part of the city electricity is still produced by thermopower plants. Even considering that diesel used in Brazil contains nowadays low sulfur contents, the large fleet is a major pollution source. As a consequence of the persistent easterly winds throughout the year, the city's pollution plume is dispersed over the nearby forest with small edge mixing along the sides (Kuhn et al., 2010).

2.2 The campaign

Data analyzed on this work were taken from the Gulfstream–1 (G–1) aircraft of the Atmospheric Radiation Measurements (ARM), the ARM Aerial Facility (AAF). The AAF G–1 platform was deployed for in situ measurements of trace gas and aerosol properties.

2.3. Field observations in Manaus environment

Observations were performed during the rainy season, on 23rd March (Intensive Operation Period 1, IOP).

2.4. Meteorological conditions during the field observations

The Manaus city is marked by permanent westerly winds during the rainy season, so its pollution plume can be easily followed. During IOP, vertical profiles suggest BLs between 400 m and 600 m amsl around 12 LT, which constrain the dispersion of plume to a relatively small fraction of the troposphere.

3. Flights

The AAF G-1 sampling gases and particles to investigate properties of polluted layers, and to characterize cloud dynamics, thermodynamics, and microphysics. Two types of flight trajectories out of the Manaus airport (located at 3.03 S, 60.03 W), were used. In the first, the plume was crisscrossed at multiple downwind distances, thus sampling the evolution of properties along the plume. An extension of transects beyond plume boundaries yielded a direct comparison between unpolluted and polluted air masses. In the second type of

trajectory, the aircraft flew along a gradient downwind of Manaus to capture the spatial extent of the plume.

The observation described here started about 10:00 local time (LT) (UTC – 4), with around 3 hours run time. The flight was accomplished at approximately constant altitude and aircraft speed.

3.2 Instruments

For the work shown here, only a few instruments were used and are briefly described below.

Cloud Condensation Nuclei Counter - CCNC

A double-column continuous-flow stream-wise thermal gradient CCN chamber (DMT CCNC-200; Roberts and Nenes, 2005; Lance et al., 2006) was used to measure the total polydisperse N_{CCN} as a function of time and *SS*. It was operated at a large gradient of temperature, decreasing incertitude in *SS*. *SS* inside CCNC were calibrated at the ground using $(NH_4)_2SO_4$ particles and Köhler theory. For column A, *SS* was set at 0.23% and at 0.50% for column B.

Fast Integrated Mobility Spectrometer – FIMS

A FIMS was used to measures the ASD (Olfert et al., 2008) with high spatial resolution and was operated with 26 size channels ranging from 16 to 440 nm. During the experiment FIMS was also operated downstream of a constant pressure inlet.

Aerosol Mass Spectrometer – AMS

An Aerodyne high-resolution time of flight mass spectrometer (Tof-AMS) was used to collect data during the GoAmazon field campaign.. The AMS sampled behind a nafion diffusion dyer, which reduced the RH of the airstream to 30 – 40%. A pressure controlled inlet was used to maintain a constant pressure upstream of the AMS aerodynamic lens. The inlet closely followed the design in Bahreini et al. (2008). The inlet pressure was set to approximately 620 mbar during the campaign. The instrument was run only in V-MS mode. Therefore, there is no ASD data.

4. Results and discussion

For this work the data were cleaned and processed in order to remove erroneous data and also to identify stretches approximately homogeneous. The procedure was complex due to the absence of a standardized procedure applicable for the investigation. In our case, we considered a homogenous section when FIMS concentration remained approximately constant. Sudden modification could reveal an inhomogeneous spot, and was considered as such. The inhomogeneous spot properties and particularities are not analyzed here.

4.1. Aerosol size distribution and CCN properties

Figure 1 shows the results from observations performed during 13th March 2014.

Manaus aerosol number concentrations (N_{CN}) is clearly contrasting with the background N_{CN}. In fact, due to low crosswind dispersion, it was possible to use data collected at either edge of transects to characterize the Manaus background air condition. The ratio between in plume and background aerosols could vary by a factor of 5, implying that even a slight diffusion of the plume air into the surroundings yields a detectable enhancement.

During the flights the Manaus plume could produce N_{CN} as high as 100.000 particles cm^{-3} (not shown on the picture due to the average).

At points of prevailing natural conditions, where aerosols were likely derived from forest sources, N_{CN} measured by FIMS was 400 – 650 cm^{-3}, and presents low variation in both sides of the plume.

The N_{CCN} measurements for the same situation have shown discernible enhancement with respect to background conditions, as is shown in Figure 1. In fact, during IOP N_{CCN} is subject to atmospheric fluctuations and there is a substantial increase during transects of Manaus plume. The results agree with previous

measurements of Kunh et al. (2010), who found an enhancement of CCN0.6 inside the Manaus plume. The result suggests that most particles transported from urban surface to altitudes where measurements took place were not effective CCNs under measured *SS*. This was probably due to the combination of the relatively small particle size and particle compositions dominated likely by organics from fresh fossil fuel combustion that in general are significantly less hygroscopic than inorganic salts such as sulfate, as was observed in Almeida et al. (2014).

During the flight mean N_{CCN} on the transections varied from 120 cm^{-3} (mean) to 180 cm^{-3} at 0.23% *SS*, and from 190 cm^{-3} to 320 cm^{-3} at 0.50% *SS* outside the plume. Inside the plume those values increased to 230 cm^{-3} (mean) to 630 cm^{-3} at 0.23% *SS*, and from 400 cm^{-3} to 1160 cm^{-3} at 0.50% *SS*.

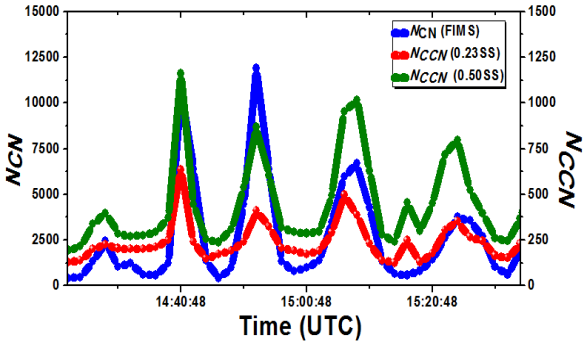

Figure 1. NCN and NCCN as a function of UTC time during measurements performed 13th March 2014 close to Manaus.

The ASD shows that most of the particles added by pollution are initially smaller than 40 nm in diameter, but they grow by coagulation and diffusion, as is shown in Figure 2. Usually, the ASD observed on typical background conditions over the rain forest has a large number of particles in the Aitken mode. In natural condition, the ASD presents a discernible accumulation mode at 150 nm and seems to be separated from the Aitken mode at 95 nm, as was also observed by Zhou et al. (2002).

5. Aerosol hygroscopicities

5.1. Estimation of hygrospcopicity (κ) values

To evaluate κ we assume that the aerosol is internally mixed (i.e., particles of a given size have similar composition) and that κ size-dependency does not vary enough to inhibit activation at larger sizes. First, we look for the diameter above which the integrated ASD (FIMS) equal the measured N_{CCN}. Then κ can be calculate using the procedure applied in Juranyi et al. (2010), which consist of insert the *SS* measured with CCNC and the just calculated diameter (D_c) on the equation

$$SS_{crit} = \left(\frac{256\sigma^3 M_w^3}{27\kappa R^3 T^3 \rho_w^3}\right)^{1/2} D_c^{-3/2}$$

for the ambient particles. Although this estimation of κ can helps in the interpretation of aerosols activity, it is subject to some errors in the *SS* set point, especially at low *SS*, which can results in a relatively large level of uncertainties in κ values.

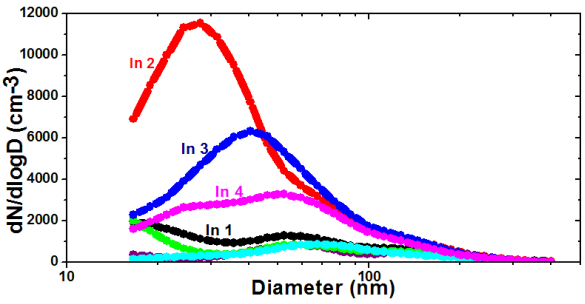

Figure 2. Aerosol size distribution following the downwind distance travelled from Manaus. The ASDs observed inside the pollution plume are indicated as IN, according to the time travelled downwind Manaus.

As pointed out by Paramonov (2013), the measured κ is probably representative of the particle properties near the activation diameter D_c, and thus different *SS* values give indication of the frequency of different κ as a function of size.

5.2 κ observed values

Figure 3 shows the values κ calculated at 0.23% and 0.50% supersaturation. For illustration values of N_{CN} are also indicated to allow a better understanding of observed values. While penetrating inside the plume κ tends to decrease. This is probably a result of what was stated above, resulting from the fact the particles

originated from urban pollution are less efficient to become *CCN*, due to their low κ values. Outside the plume values tends to be more homogeneous and presents values between 0.10 and 0.20, in good agreement with values from Gunthe et al., 2009

It also can be shown that values observed at north side of the plume are larger than those observed at south side. This is probably a result from the fact that while infusing inside the plume the particles from forest increase mean values of κ. The opposite is expected for the south side, but fluvial breeze can produce a different circulation flow, affecting the plume dispersion.

Values of κ are larger for lower *SS*. This is consistent with the fact that that in general at higher *SS* particles smaller and less active as *CCN* are activated. Due to the large amount of organic compounds, those particles are expected to have lower hygroscopicities.

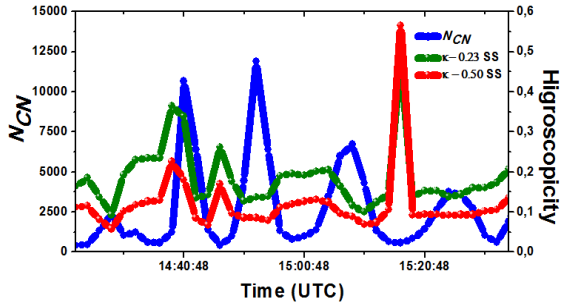

Figure 3. Values of κ derived for 13th March 2014 following the downwind distance travelled from Manaus.

6. Concluding remarks

Particles from urban pollution can be composed of freshly emitted soot, which are hydrophobic or limited in hygroscopicity and less able to contribute to the CCN population in the atmosphere (Weingartner et al., 1997; Meyer and Ristovski, 2007; Zhang et al., 2008; Tritscher et al., 2011). Almeida et al. (2014) have shown that urban aerosols in São Paulo, Brazil are composed of weakly active organic compounds, which suggest that aerosols form Manaus can have the same properties. Soot agglomerates, nevertheless, become more hygroscopic when coated, partly or fully, by organic or inorganic material (Wittbom et al., 2014), the process, then, enable them to act as more effectively as CCN.

We intend to study how the addition of Black Carbon (BC) can affect the determination of κ of the plume. For this, long term observation of aerosol composition and activation efficiency need to be studied and parametrized for the calculation performed here.

7. Acknowledgments

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