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Field Observation of the Green Ocean Amazon: Neutral Cluster Air Ion Spectrometer (NAIS) Final Campaign Summary

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Executive Summary

The neutral cluster and air ion spectrometer (NAIS) was deployed to the T3 site for Intensive Operations Periods 1 and 2 (IOP1 and IOP2). The NAIS is an instrument that measures aerosol particle and ion number size distributions in the mobility diameter range of 0.8 to 42 nm, corresponding to electrical mobility range between 3.2 and 0.0013 $cm^2 V^{-1} s^{-1}$. New particle formation (NPF) events were detected using the NAIS at the T3 field site during IOP1 and IOP2. Secondary NPF is a globally important source of aerosol number. To fully explain atmospheric NPF and subsequent growth, we need to directly measure the initial steps of the formation processes in different environments, including rain forest. Particle formation characteristics, such as formation and growth rates, were used as indicators of the relevant processes and participating compounds in the initial formation. In a case of parallel ion and neutral cluster measurements, we estimated the relative contribution of ion-induced and neutral nucleation to the total particle formation.

We made direct observations on NPF events at T3 ground site during IOP1. Based on our data, the newly formed particles originated from cluster sizes, and during the course of the day, they grew all the way to potential cloud condensation nuclei sizes. More frequently, we observed the particle formation events that started at larger sizes, possibly associated with their initial production higher up and subsequently transported to the ground level. The small cluster ions (0.8 to 2.0 nm) had a clear and strong diurnal cycle at the T3 site. The cycle presumably is linked to boundary layer development, local precipitation events, and as cluster dynamics.

The NAIS detects both neutral clusters and charged air ions. The air ions are classified according to their natural charge, whereas the neutral particles are artificially charged inside the instrument with a unipolar corona charger and then detected. The NAIS has a high time resolution, varying from 1 second to 3 minute averages. During the GoAmazon campaign, the NAIS was operational during both IOP1 and IOP2, which are representative of the wet and dry seasons, respectively. The data coverage of the NAIS during IOP1 and IOP2 were over 95% and 83%, respectively. The instrument was operated outside the IOPs from January 27 to April 23, 2014, collecting a total of 87 days of available particle number size distribution data. During the IOP2 time period between September 1 and October 10, 2014, measurements began somewhat later because the NAIS underwent repairs (operational from August 26 to October 13, collecting data for a total of 49 days).

In addition to the time series of particles and ions during the GoAmazon campaign IOPs, a similar data set was gathered at the ZF2 measurement station from September 2011 to January 2014 to capture long-term variability of these parameters in the rain forest environment. The ZF2 and T3 sites represent complementary field measurement sites.

Acronyms and Abbreviations

ARM	Atmospheric Radiation Measurement
DMA	Differential Mobility analyzer
GoAmazon	Green Ocean Amazon
IOP	Intensive Operational Period
NAIS	Neutral cluster and Air Ion Spectrometer
NPF	new particle formation
PSM	particle size magnifier
UTC	Coordinated Universal Time

Contents

Figures

1.0 Background

Aerosol particles have global effects on Earth's climate and regional effects on air quality. Aerosol particles are emitted to the atmosphere either directly or they are formed from gas-phase precursors. Secondary pathways can, as a global average, produce 45% of the climatically active particles in terms of their number (Merikanto et al. 2009). Secondary formation of atmospheric nanoparticles is a multi-stage process in which stable clusters form from gas-phase precursors (Kulmala et al. 2013). The initial steps involve molecular clustering, which is followed by cluster activation for enhanced growth (Kulmala et al. 2014). For the subsequent growth, a particularly important step is multi-component condensation of organic vapors (Riipinen et al. 2012; Ehn et al. 2014) that enables the nanoparticles to become large enough to affect the climate via aerosol-cloud interactions (Boucher et al. 2013).

The Amazon is an example of a pristine area where different aerosol formation pathways can be explored with minimal anthropogenic influences (Pöschl et al. 2010; Hamilton et al. 2014). Although secondary aerosol formation is occurring readily in the atmosphere in many environments (Kulmala et al. 2004), the Amazon is one of the areas where the initial steps of nanoparticles formation have not been observed previously from ground-based measurements. Typically, the observations conducted at the ground level in the Amazon have revealed events of sudden appearance of nucleation mode particles in the sizes 10 to 40 nm (Rissler et al. 2010). This feature has been attributed to their formation aloft (Zhou et al. 2002; Pöschl et al. 2010) and subsequent downward mixing. In the Amazon, emissions of volatile organic compounds, their oxidation (Lelieveld et al. 2008), and formation of condensable vapors leading to aerosol activation to cloud droplets and eventually rain is tightly connected and interlinked with meteorological processes, such as boundary-layer development connecting the water cycle and aerosols (Pöschl et al. 2010).

Long-term data on aerosol and ion number concentration measurements in the Amazon environment are valuable for gaining a deeper understanding of the aerosol-cloud interactions and processes leading to aerosol formation as well as identifying and quantifying the complex feedback mechanisms between the biosphere and the atmosphere. In this work, we used novel instrumentation including the neutral cluster and air ion spectrometer (NAIS) (Kulmala et al. 2007; Mirme and Mirme 2013) and the particle size magnifier (PSM) (Vanhanen et al. 2010) to collect measurements of the freshly formed atmospheric clusters and particles within the Amazon region. The NAIS measures size distributions of 0.8 to 47 nm neutral and charged aerosol particles and clusters, whereas the PSM detects aerosol particles down to 1 nm in size. The observations were conducted during intensive observational periods 1 and 2 (IOP1 and IOP2) as well as during the time between the two IOPs. The measurements were conducted at the T3 site. Additional measurements were conducted at the ZF2 site.

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2.0 Notable Events or Highlights

The location of our measurements was the ARM site is T3, which is a terrestrial site 60 km west of Manaus, Brazil (3°12'47.82"S, 60°35'55.32"W). The campaign dates were from January 27 to April 23, 2014 for IOP1 (the wet season) and August 15, 2014 to October 15, 2014, for IOP2 (the dry season).

As a part of the GoAmazon campaign, NAIS (1 to 40 nm) and PSM (1 to 3 nm) instruments performed size distribution measurements at the smaller end of aerosol particle distribution. These instruments are able to measure the size distribution of aerosol particles below the size range of standard scanning mobile particle sizer instruments. Both the NAIS and the PSM instruments provide valuable information on the atmospheric neutral and charged cluster dynamics, and their formation processes down to a size of 1 nm (Kulmala et al. 2012, 2013). Figure 1 shows the instrument location at T3.

Figure 1. Photograph from the T3 field site. The NAIS instrument is inside the white box next to the container. The yellow arrow points to the NAIS inlet tube.

Selected highlights are briefly discussed below:

- We observed an increasing number of new particle formation (NPF) events at the T3 field site during IOP1. We were able to follow the growth from cluster sizes to potential cloud condensation nuclei sizes. The most active days were January 30, 2014 and March 13, 2014 (see Figure 2 and Figure 3).

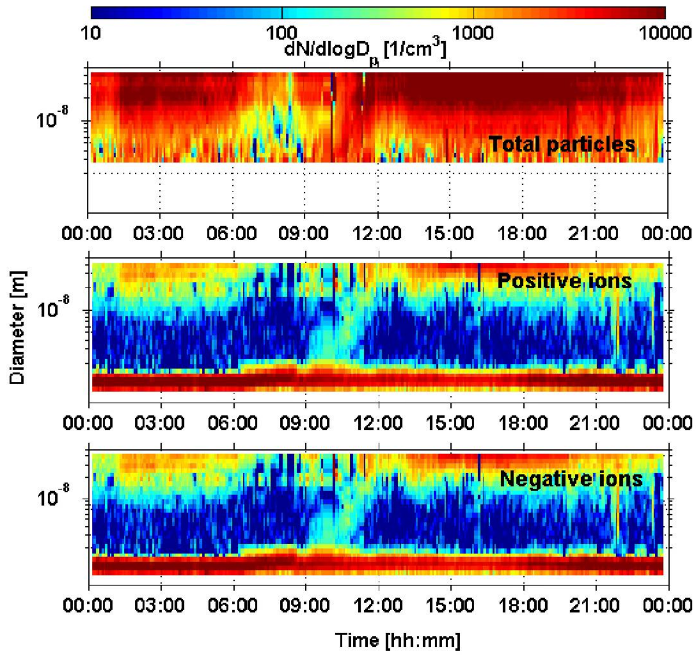

Figure 2. Particle and ion number size distribution measured with the NAIS on January 31, 2014, at the Manaus T3 Site during IOP1. Time in UTC, local time is UTC-4. The NPF event started at 9:00 UTC time, when both particles and ions are detected with the instrument. Subsequently, the particles grow towards larger (climatically relevant) sizes within the next few hours.

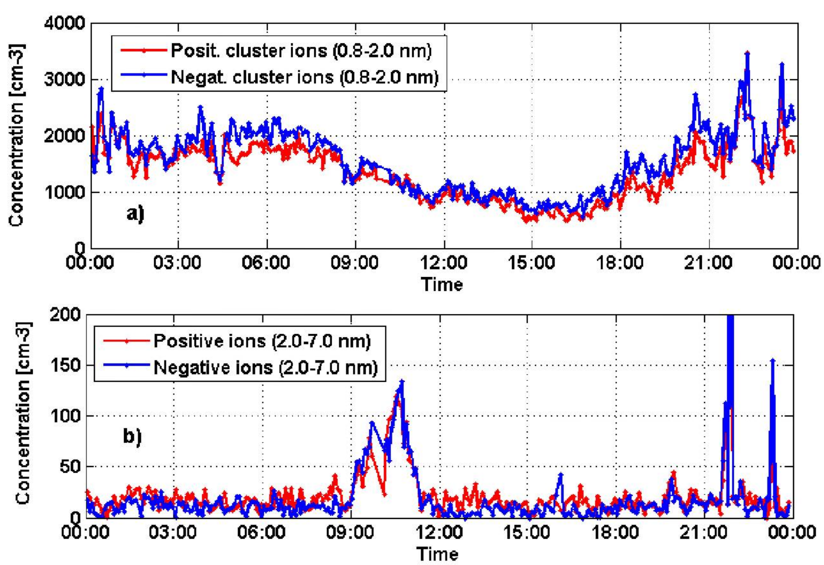

Figure 3. Number concentration of a) positive and negative cluster ions and b) intermediate negative and positive ions measured with the NAIS on January 31, 2014, at the Manaus T3 site. Time in UTC, local time is UTC-4. The cluster ion concentration at 0.8 to 2.0 nm size range decreased as the ions grew into the intermediate ions size range 2.0 to 7.0 nm during the particle growth period (9:00 to 11:30 UTC).

- During the IOP2, signals from biomass burning aerosol were apparent at ground level. Strong rain episodes affected the observed number size distribution with the NAIS. September 17, 2014, was an eventful day during IOP2. First, we observed an increase in the cluster ion concentration shortly after sunrise. Second, we observed a weak nucleation event. Then, we detected what were probably anthropogenic pollution plumes from Manaus, and finally, a rain event in the evening caused by the heavy rain associated episode.
- Our measures on aerosol particle number size distributions at ZF2 and T3 sites with different forest environments showed very clear differences because the canopy is a large sink for nanoparticles.

NAIS data for IOP1 and IOP2 are available at the ARM T3 site:

- GoAmazon IOP1 – February 22 to March 23, 2014. Good-quality NAIS data is available between January 27 and April 23, 2014, a total of 87 days for IOP1. Data coverage during IOP1 was 83%.
- GoAmazon IOP2 – September 06 to October 04, 2014. Good-quality NAIS data is available between August 26 and October 13, 2014, a total of 49 days for the IOP2. The NAIS data coverage during IOP2 was 73%.

Supporting data sets are available at the ARM T3 and ZF2 sites:

- Good-quality PSM data set is available for the period from August 26 to October 13, 2014, a total of 49 days for IOP2. The PSM provides size distribution of particles and clusters in size range 1 to 3 nm.
- We measured a continuous time series of aerosol particle number size distribution inside the rain forest canopy at ZF2 site from September 2011 to January 2014. Instruments used were the NAIS (0.8 to 40 nm) and a differential mobility particle sizer (6 to 800 nm), and assorted other meteorological instrumentation.

3.0 Lessons Learned

Operating scientific instrumentation in the tropics is challenging. Some of the problems encountered during the IOPs were issues we have experienced before and therefore expected. Our previous experience and good practices at ZF2 proved valuable. First, the NAIS instrument was operated in a heated cabin outside of the measurement containers. This was done to avoid losses in the sampling lines (Figure 1). The configuration allows the inlet line to be as short as possible. The instrument needed to be heated to a temperature above ambient to avoid water condensation inside the instrument. The performance of this setup was established before IOP1 because the setup had been measuring at the ZF2 site some 50 km north of Manaus during previous years.

The staff at Instituto Nacional de Pesquisas da Amazônia in Manaus, Brazil, had experience with the NAIS instrument from previous deployments, so supporting staff with knowledge of how to operate and troubleshoot the instrument were available. This proved to be helpful when the instrument malfunctioned between the two IOP periods. The collaboration allowed us to remotely troubleshoot the malfunction and bring spare parts for the required repairs before IOP2.

In retrospect, we would have needed a setup that would prevent the instruments from overheating. This caused us to replace a computer and some components between the IOPs. Based on past experience, having enough spare parts on hand was vital because transporting them to Manaus had proven quite challenging previously.

Based on previous field experience, we knew that a reliable Internet connection for monitoring instrument performance online was a very good way to improve the data quality. The bandwidth and connection provided by ARM Mobile Facility 1 at T3 was vital to the success of the measurement campaign. Without an Internet connection, the NAIS would not have been operational during the second IOP because of multiple malfunctions that occurred between the IOPs. Challenging environmental conditions—high ambient temperature differences between day and night, heavy rain episodes, pollution plumes, power outages, and high voltage transients in the electrical supply—all contributed to issues with the instrumentation.

Another lesson learned related to the instrument setup was that the NAIS needs to be installed so that humidity does not build up in the instruments when it is powered off (e.g., between IOPs). When powered off for a long time period (i.e., several days), water vapor will collect in porous insulators within the instrument, and it takes a very long time for this water to be eliminated. This issue caused large background current readings in the electrometers, which is the primary measurement from which the number size distribution is determined. We observed that, after power had been off for a while, data quality was poorer.

Overall, the collaboration among the partners during the IOPs was a key to the successful campaign. Without a trained local support group, the measurements like GoAmazon would not have been possible.

4.0 Results

We made direct observations of NPF events at T3 ground site, especially during IOP1. Some of the newly formed particles were observed down to cluster sizes, and during the course of the events, they grew all the way to potential cloud condensation nuclei sizes, where they may have climatic relevance. More frequently, we observed particle formation events that started with larger particle sizes. This indicates that these particles were formed aloft and transported downwards, possibly via downdrafts associated with precipitation.

Figure 2 shows the particle and ion number size distribution measured with the NAIS on January 31, 2014, at the T3 site. The particle formation event started at 9:00 UTC both in the particle and ion channels, and the particles grew to larger sizes during the next few hours. Overall, the analysis revealed that the distinct shape of NPF and growth events was observed more frequently on the contour plot series of number size distribution during the wet season compared to the dry season. During the particle growth period from 9:00 to 11:30 UTC as seen in Figure 3, the cluster ion concentration within the 0.8 to 2.0 nm size range decreased as the ions grew to intermediate size within the 2.0 to 7.0 nm range.

The small cluster ions (0.8 to 2.0 nm) had a clear and strong diurnal cycle at the T3 site. This cycle results from cluster and boundary-layer dynamics, which provide vertical mixing. The importance of photochemistry in cluster production and growth is indirectly observed during particle growth events in the day-time hours. Our results indicate initial formation aloft and subsequent mixing down to ground level. However, a more detailed analysis is needed to confirm this hypothesis.

During the GoAmazon IOP2 campaign (dry season), we collected data over a total of 49 days. According to the preliminary analysis, we observed 29 days with rain events, 3 days with NPF events, and 14 days with evening anthropogenic plume-type events. As is typical for rain forest environments, intense rain episodes are frequently visible in the ion data, particularly in the negative polarity. These bursts of intermediate ions are usually associated with the balloelectric effect (i.e., generation of the ions by the splashing of water) (Tammet et al. 2009). During the rain events, the concentrations of intermediate ions increased rapidly to very high levels, but no subsequent growth was observed. Therefore, based on the temporal evolution of the ion population, this phenomenon can be distinguished from the secondary aerosol formation.

The NAIS data set is valuable for exploring the spatial and temporal variability of nanoparticle and ion size distributions in the Amazon region, when both ZF2 and T3 data sets are compared. The differences between the dry and wet seasons provide insights into the aerosol physical and meteorological processes that drive the nanoparticle concentrations. A deeper analysis on NPF events and growth can be used to estimate required source strengths of the condensable vapors, which can be compared with previous results (Manninen et al. 2010).

5.0 Public Outreach

Public outreach was conducted jointly with the GoAmazon consortium.

6.0 Publications

6.1 Journal Articles/Manuscripts

A joint publication (led by Jian Wang, Brookhaven National Laboratory) is being prepared.

A publication on ground-based observations of nanoparticles at the T3 and ZF2 sites (led by Tuukka Petäjä, University of Helsinki) is being prepared.

6.2 Meeting Abstracts/Presentations/Posters

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Wimmer, D, et al. 2014. "Measuring with Condensation Particle Counters under Challenging Environments." Presented at the International Aerosol Conference 2014, Busan, South Korea. (*Short abstract, poster presentation*.)

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