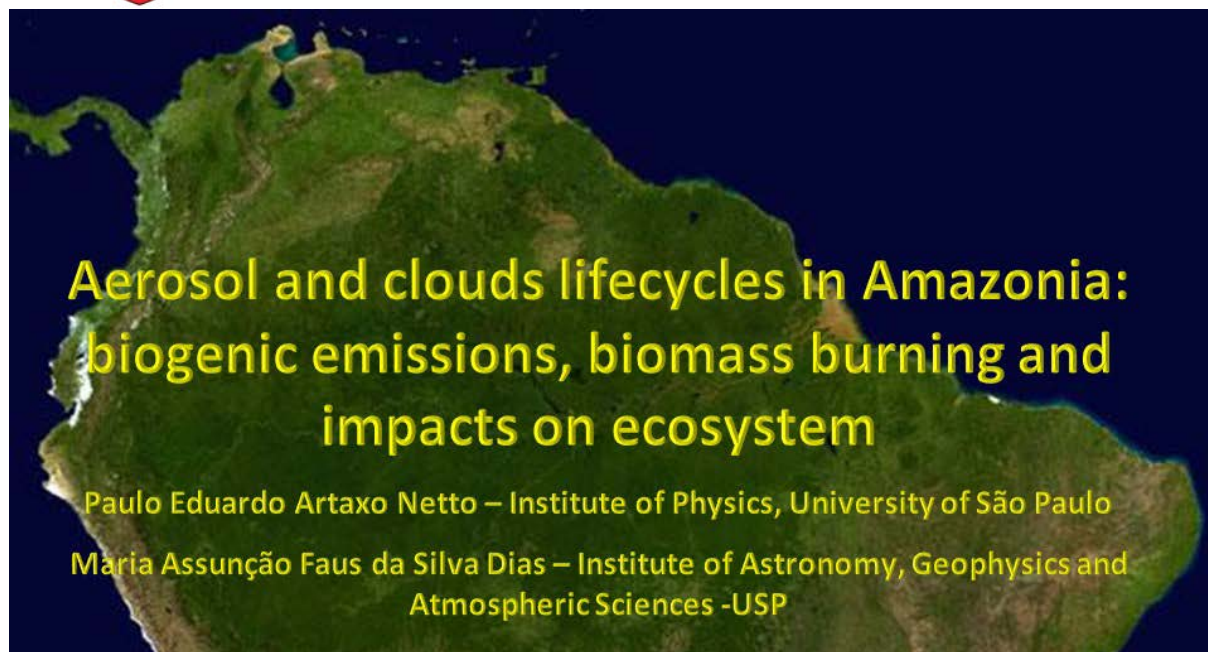




Research Proposal for a FAPESP Thematic Project
August 14, 2017



Brazilian participants

Paulo Eduardo Artaxo Netto	Institute of Physics, University of São Paulo - IFUSP
Maria Assunção F. da Silva Dias	Institute of Astronomy, Geophysics and Atmospheric Sciences, IAG - USP
Luiz Augusto Toledo Machado	CPTEC-INPE
Henrique de Melo Jorge Barbosa	Institute of Physics, University of São Paulo - IFUSP
Luciana Varanda Rizzo	Universidade Federal de São Paulo - UNIFESP
Edmilson Dias de Freitas	Institute of Astronomy, Geophysics and Atmospheric Sciences, IAG - USP
Rachel Ifanger Albrecht	Institute of Astronomy, Geophysics and Atmospheric Sciences, IAG - USP
Alan James Peixoto Calheiros	CPTEC-INPE
Izabelly Carvalho da Costa	CPTEC-INPE
Thiago Souza Biscaro	CPTEC-INPE
Eder Paulo Vendrasco	CPTEC-INPE
Niro Higuchi	Instituto Nacional de Pesquisas da Amazonia - INPA
Samara Carbone	Federal University of Uberlândia, Minas Gerais.
Patrick Schlag	Pos Doc IFUSP – Prof. Paulo Artaxo
Micael Amore Cecchini	Pos Doc IAG-USP – Profa. Assunção
Ramon Campos Braga	Pos Doc IAG-USP Profa. Albrecht

International partners

Scot T. Martin	Harvard University - USA
Meinrat O. Andreae	Max Planck Institute for Chemistry – Germany
Jos Lelieveld	Max Planck Institute for Chemistry – Germany
Hans Christen Hansson	University of Stockholm, Sweden

Ilona Riipinen	University of Stockholm, Sweden
Radovan Krejci	University of Stockholm, Sweden
José Vanderlei Martins	University of Maryland Baltimore County UMBC - USA
Marcos F. Andrade	Laboratorio de Física de la Atmosfera Chacaltaya, Bolivia

Students

Djacinto Aparecido M. Santos Jr.	PhD Student IFUSP – Paulo Artaxo
André Araújo Burger	MSc Student – IFUSP – Paulo Artaxo
Rayner Monteiro dos Santos	MSc Student – IFUSP-CLIAMB – Paulo Artaxo
Janaína M. P. do Nascimento	PhD Student – IFUSP-CLIAMB – Paulo Artaxo
Renata de Araujo Teixeira	PhD Student – IFUSP-CLIAMB – Paulo Artaxo
Rafael da Silva Palácios	PhD Student IFUSP UFMT – Paulo Artaxo
Everlin Pereira Fernandes	MSc Student – IFUSP-CLIAMB – Paulo Artaxo
Bruno Backes Meller	Undergrad Student – IFUSP Prof. Paulo Artaxo
Fernando G. Morais	PhD Student (IFUSP) – Paulo Artaxo
Marco Aurélio Menezes Franco	PhD Student (IFUSP) – Paulo Artaxo
Ana Maria Pereira Nunes	MSc Student - IAG-USP Profa. Assunção
Gláuber Camponogara	PhD Student - IAG-USP Profa. Assunção
Ivan Saraiva	PhD Student - IAG-USP Profa. Assunção
Mercel José dos Santos	PhD Student - IAG-USP Profa. Assunção
Diego Alves Gouveia	PhD IFUSP – Prof. Barbosa
Alex Sandro Alves de Araujo	PhD IFUSP – Prof. Barbosa
Giovanni Souza	MSc IFUSP – Prof. Barbosa
Amanda Vieira dos Santos	MSc IFUSP – Prof. Barbosa
Rebeca Fonseca de Oliveira Pereira	MSc Student – IAG-USP Profa. Albrecht
Camila da Cunha Lopes	MSc Student – IAG-USP Profa. Albrecht
Jessica Cristina dos Santos Souza	Undergrad Student – IAG-USP Profa. Albrecht
Jessé Stenico	Undergrad Student – IAG-USP Profa. Albrecht

Technicians

Fernando Gonçalves Morais	Institute of Physics, USP
Fábio de Oliveira Jorge	Institute of Physics, USP
Simara G. Morais	Institute of Physics, USP
Guilherme Casanova	Institute of Physics, USP
Eduardo Fernandes Gomes	Institute of Astronomy, Geophysics and Atmospheric Sciences, IAG - USP

Contents

Abstract	4
Resumo (in Portuguese)	4
1 - Justification and Rationale	5
1.1 - Biogenic volatile organic compounds emissions	6
1.2 - Secondary Organic Aerosol (SOA) production in Amazonia	7
1.3 - Aerosol characterization in Amazonia	8
1.4 - Cloud Condensation Nuclei (CCN) in Amazonia	10
1.5 - Optical properties and radiative forcing of Amazonian aerosols	11
1.6 - Aerosol production at high altitudes and convective transport by clouds.....	13
1.7 - Precipitation, clouds and climate.....	15
2 – Current Scientific Gaps	17
2.1 – Aerosol Life cycle in Amazonia	17
2.2 - Cloud Life Cycle in Amazonia	18
2.3 – Modeling aerosol-cloud interactions in Amazonia	19
3 - Objectives of the proposed work.....	20
6 - Measurement strategy.....	22
6.1 – Long term measurements at the ATTO – Amazon Tall Tower Observatory ...	22
6.1.2- Clouds observation framework at the ATTO tower.....	24
6.2 – Large scale measurements using ships in western Amazonia	25
6.4 – Large Scale Measurements using the High Altitude HALO plane	27
6.5 – Measurements of Amazonian aerosols in Chacaltaya	29
7 - Bridging Models and Observations	32
7.1 - High Resolution Regional Scale Modeling.....	32
7.2 High resolution cloud dynamics, aerosol and microphysics interactions.....	33
8 - Time schedule of the project	34
9 - Expected results and innovation.....	35
10 – Role of each investigator	35
11 – Data management plan and data sharing.....	36
12 – Major existing equipment to be used in this project.....	37
13 – Major equipment to be acquired in this project.....	37
14 – Results from previous FAPESP Thematic projects.....	38
15 - References.....	39

Abstract

Amazonia is a living laboratory to study critical processes that regulate tropical atmospheric chemistry and physics. The forest is an important global source of aerosols, trace gases and water vapor, and the complex nonlinear processes that regulate these different components are still not fully understood. In this project, we will study Aerosol Life Cycle (ALC), Cloud Life Cycle (CLC), and Cloud-Aerosol-Radiation-Precipitation Interactions (CAPI) in Amazonia, using a combination of approaches that allows innovative research in the tropics. The project comprises 4 measurement efforts: 1) New long term observations at the Amazon Tall Tower Observatory (ATTO); 2) Several fluvial expeditions in the untouched areas of Western Amazonia; 3) A large scale aircraft experiment with the HALO G5 high altitude plane (14 km); and 4) Aerosol and trace gas measurement campaigns at Chacaltaya, Bolivia, 5,240 altitude in the Andes, to study the transport and impact of Amazonian aerosols.

These measurement efforts, going from the ATTO 325 meters tall tower, through fluvial ship and aircraft up into the Andes at the GAW-WMO Chacaltaya station, will allow a large spectrum of critical processes that regulates the links between forest-atmosphere-climate in tropical regions. In these sites and platforms, we will measure, among other things, aerosol optical properties with spectral light scattering and absorption, aerosol size distribution, aerosol composition for organic and inorganic components, aerosol optical depth, radiation balance, cloud condensation nuclei, cloud droplet size, cloud optical depth, and vertical profiles of aerosols, clouds, precipitation and thermodynamic variables. A large set of advanced instrumentation will make these measurements in difficult logistical conditions. High resolution cloud modeling will integrate aerosol, CCN and water vapor for a variety of thermodynamic conditions and will allow integration of organic aerosol analysis with cloud processes. High-resolution BRAMS and WRF-Chem regional modeling will be performed to help understanding regional processes and transport.

With these new datasets and associated modeling efforts, we plan to study cloud-aerosol-precipitation interactions and the feedbacks between biosphere and atmosphere and human activities through deforestation and biomass burning emissions. We expect that these measurements and modeling framework will provide new insights in critical and important processes that regulate tropical atmospheric chemistry and cloud physics. The analysis will also provide insights into how Amazonia is being perturbed by biomass burning emissions and how it influences climate regionally and globally.

Resumo (in Portuguese)

A Amazônia é um excelente laboratório para estudar processos críticos que regulam a física e química atmosférica tropical. A floresta é uma importante fonte global de aerossóis, gases traços e vapor de água, e os complexos processos não lineares que regulam estes diferentes componentes ainda não são totalmente compreendidos. Neste projeto, estudaremos ciclo de vida do aerossol (ALC), o ciclo de vida de nuvens (CLC) e as interações entre nuvens-aerossóis-irradiação e precipitação (CAPI) na Amazônia. Utilizaremos uma combinação de abordagens que permitem pesquisas inovadoras nos trópicos. O projeto envolve 4 esforços de medidas: 1) Novas observações de longo prazo na torre ATTO (Amazon Tall Tower Observatory); 2) Expedições fluviais na Amazônia Ocidental em áreas ainda não investigadas do ponto de vista atmosférico; 3) Um experimento para investigar a atmosfera amazônica em altas altitudes (14 km), com o avião alemão HALO G5; e 4) campanhas de

medição de aerossóis e gases traços transportados da Amazonia para Chacaltaya, Bolívia, a 5.240 metros de altitude nos Andes.

Esses esforços de medição, que vão da torre ATTO de 325 metros de altura, através de navios fluviais e aviões até amostragens nos Andes na estação Chacaltaya, permitirão estudar um grande espectro de processos críticos na ligação entre biosfera-atmosfera-clima em regiões tropicais. Nesses sites e plataformas, nós avaliaremos, entre outras coisas, propriedades ópticas de aerossol como espalhamento e absorção espectral de radiação, distribuição de tamanho de aerossol, composição de aerossol para componentes orgânicas e inorgânicas, profundidade óptica de aerossóis e nuvens, balanço de radiação, núcleos de condensação de nuvens, tamanho de gota de nuvem e perfis verticais de aerossóis, nuvens, precipitação e variáveis termodinâmicas. Um grande conjunto de instrumentação avançada fará essas medidas em difíceis condições logísticas. A modelagem em nuvens em alta resolução integrará aerossóis, CCN e vapor de água para uma variedade de condições termodinâmicas e permitirá a integração de análises de aerossóis com processos em nuvens. A modelagem regional em alta resolução utilizando BRAMS e WRF-Chem serão realizadas para ajudar a entender os processos e o transporte regional.

Com esses novos conjuntos de dados e esforços de modelagem associados, planejamos contribuir no entendimento das interações entre nuvens-aerossol-precipitação e os feedbacks entre biosfera e atmosfera em condições naturais e em condições dominadas por emissões de queimadas. Esperamos que essas medidas e esforços de modelagem proporcionem novos conhecimentos em processos críticos e importantes que regulam a química atmosférica tropical e a física de nuvens e radiação. A análise também fornecerá importantes informações sobre como a Amazônia está sendo perturbada pelas emissões de queima de biomassa e como ela influencia o clima regionalmente e globalmente.

1 - Justification and Rationale

Amazonia is one of the few continental areas where it is still possible to observe pristine atmospheric conditions in the wet season (Andreae et al., 2009, 2016, Artaxo et al., 2013). Wet season atmospheric conditions for aerosol particles resembles background conditions typical of pre-industrialization (Andreae et al., 2009, Davidson et al., 2012, Carslaw, 2017), with particle number concentrations below 300 #/cc and fine mode aerosol concentration at approximately 2-3 $\mu\text{g}/\text{m}^3$ (Rizzo et al., 2013). The Amazon basin functions as a giant biogeochemical reactor to influence regional climate, with both exports and imports of climate-relevant quantities to and from other regions of the Earth (Artaxo et al., 2013). Biogenic emissions of gases and aerosol particles, in combination with high absolute humidity and strong solar radiation, maintain chemical and physical cycles that sustain the aerosol particle population, the cloud field, and the hydrological cycle of the basin (Martin et al., 2016). The biology of the forest has a critical role in regulating atmospheric composition and climate over the region. The Amazonian ecosystem uses the feedstock of plant and microbial emissions in combination with high water vapor, solar radiation, and photo-oxidant levels to produce secondary organic aerosol (SOA) particles and primary biological aerosol particles. Through these emissions, the forest has strong interactions with the atmosphere (Pöschl et al., 2010; Martin et al., 2010a, 2010b, 2016, Artaxo et al., 2013). The hydrologic cycle of the basin is one of the primary heat engines of global circulation (Nobre et al., 2009). This reactor produces the nuclei for clouds and precipitation and partially sustains the hydrological cycle

(Prenni et al. 2009). The Basin has been dubbed the “green ocean” because of the similarities in particle concentrations and cloud microphysics with remote oceanic regions (Roberts et al., 2001; Williams et al., 2002). Figure 1.1 illustrates some of these processes.

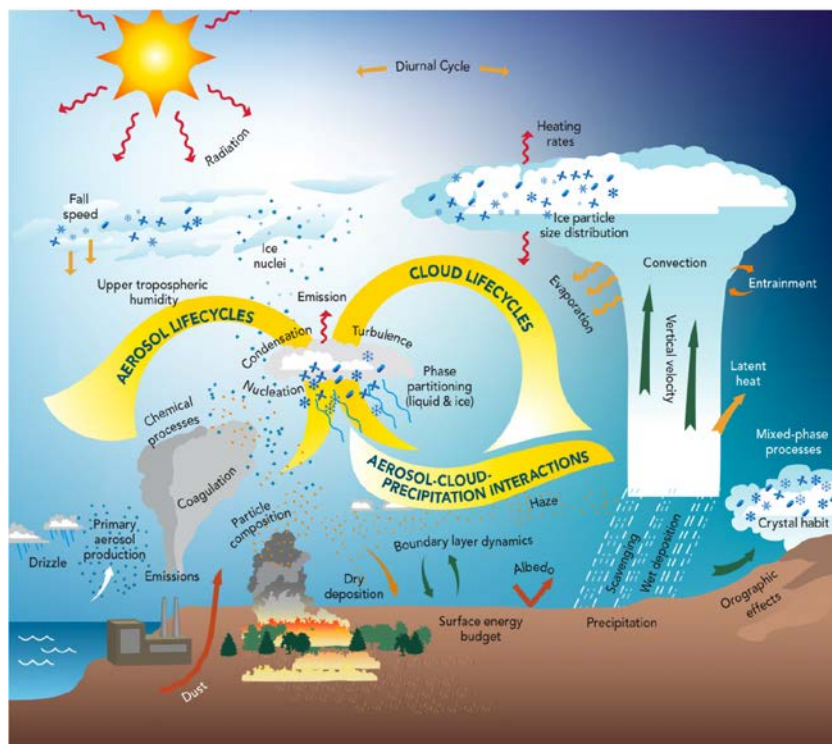


Figure 1.1 - Atmospheric system depiction showing connections between the aerosol life cycle interacting with the cloud life cycle through surface and thermodynamic processes. The role of surface fluxes, including natural and anthropogenic emissions is very important for both life cycles. The role of convection and atmospheric thermodynamic conditions is also critically important. Interaction with solar radiation and convection makes critically important impacts on the ecosystem

At the same time, large-scale biomass burning emissions as well as urban pollution changes this picture with very high aerosol and trace gases concentrations over specific periods of time or at specific locations (e.g. Martin et al., 2016, 2017). The effects of aerosol particles on the radiation balance and cloud microphysical properties, cloud cover, precipitation, lightning, and regional climate over the Amazon are very significant. Several studies from the LBA Experiment (The Large-Scale Biosphere-Atmosphere Experiment in Amazonia) have shown this effect (e.g. Artaxo et al., 2002, Silva Dias et al 2002, Procópio et al., 2004, Albrecht et al., 2011, Davidson, et al., 2012, Martin et al., 2010a, Whitehead et al., 2016 and many others). Moreover, aerosol from biomass burning spreads to the west and south of the Amazon Basin (Freitas et al 2005) and affect CCN population and consequently cloud formation, evolution and rainfall elsewhere in South America (Camponogara et al 2013, 2016). The very low background aerosol concentrations in the wet season, high water vapor levels and intense radiation make the Amazon region particularly susceptible to changes of its trace-level atmospheric composition. The climatic implications for strong tropical aerosol-cloud dynamic interaction are profound, ranging from modulation of local precipitation intensity to modifying large-scale circulations and energy transport associated with deep convective regimes (Andreae et al., 2017).

1.1 - Biogenic volatile organic compounds emissions

Amazonian BVOCs are emitted from plants during growth, maintenance, decay, and consumption (Guenther et al., 2012, Yanez Serrano 2015). Major BVOCs emitted include isoprene (C_5H_8), monoterpenes, sesquiterpenes, ethane, and oxygenated VOCs (OVOCs)

(Jardine, 2015). Tropical forests are the dominant global source of atmospheric BVOCs and the Amazon Basin is a major contributor. The high species diversity in the basin is coupled with an ecological complexity and a seasonality that is very different from temperate regions, yielding significantly different emission trends with different forest types. Isoprene and monoterpene emissions and aerosol concentrations are strongly associated in the Amazon. The OH oxidation pathway is particularly important for BVOC oxidation in the tropics given the high radiation levels and H₂O concentrations. Heald et al. (2010) estimate that the conversion of South American BVOCs into secondary particle mass contributes to 40% of the annual global production of this particle component. Knowledge of the composition, the sources, the chemistry, and the role of the secondary organic components of particles in the atmosphere and earth's climate system is, however, still very limited. Even for the well-studied isoprene compound, recent analysis suggests that state-of-the-art atmospheric chemistry models greatly under predict OH concentrations (Lelieveld et al., 2008), possibly implying important under predicted and underestimated chemistry. Figure 1.1.1 illustrate some of the pathways involved in VOC oxidation and aerosol production.

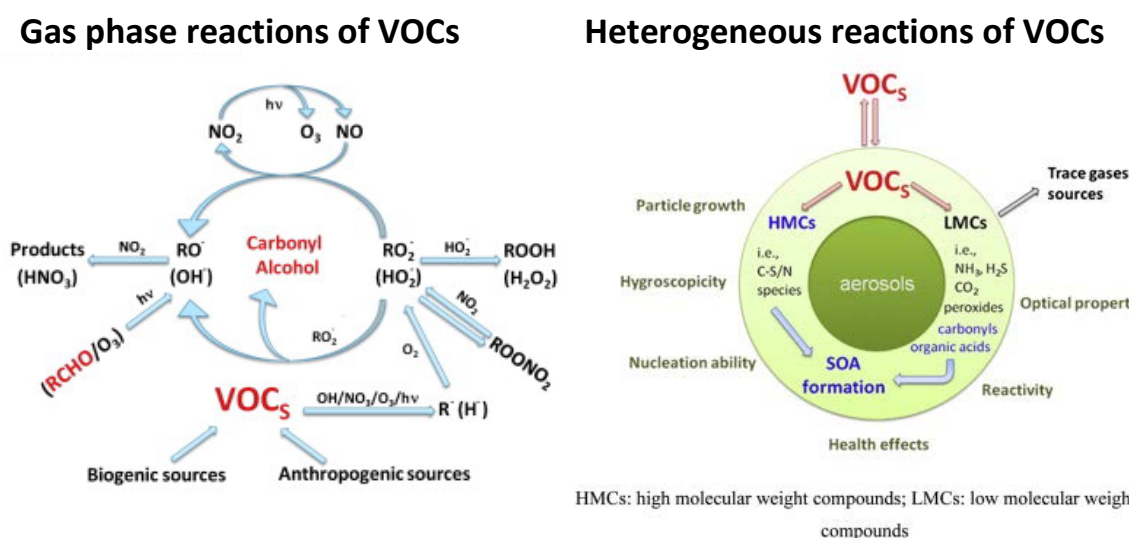


Figure 1.1.1 - Possible pathways for BVOC emissions, processing, oxidation and SOA production in Amazonia. VOCs are the major pathway to aerosol, mostly at the high altitude trough cloud and convection processing (see high altitude aerosol measurements in a latter section).

1.2 - Secondary Organic Aerosol (SOA) production in Amazonia

The most important component of the ALC in the fine mode in Amazonia is SOA. With measurements showing that more than 75% of aerosols in Amazonia in both dry and wet season are SOA, it is important to study its formation, processing and deposition in the region. The production mechanisms for secondary particle components involve many trace gases, in particular BVOCs, aromatics, nitrogen oxides (NO_x), ozone (O₃), hydroxyl radical (OH), and sulfur species including dimethyl sulfide (DMS) and sulfur dioxide (SO₂) (Shrivastava et al., 2017, Chen et al., 2016, Sá et al., 2017). BVOCs and aromatics react with O₃ and OH to produce oxidized organic products, a fraction of which have low enough volatility to condense and serve as particle components. BVOCs, aromatics and NO_x together influence the concentrations of O₃ and OH, thereby influencing the production of BVOC oxidation products. Reactions both in the gas phase and in cloud water are important, as well as vertical

convection to higher altitudes. Previous studies in the pristine Amazon rainforest showed that fine particles (which account for most of the cloud condensation nuclei) consist mostly of secondary organic material derived from oxidized biogenic gases (Pöschl et al., 2010; Martin et al., 2010a; Allan et al., 2014; Chen et al., 2015). Figure 1.2.1 illustrate the process of producing SOA from VOCs through the various NO_x and OH mechanisms.

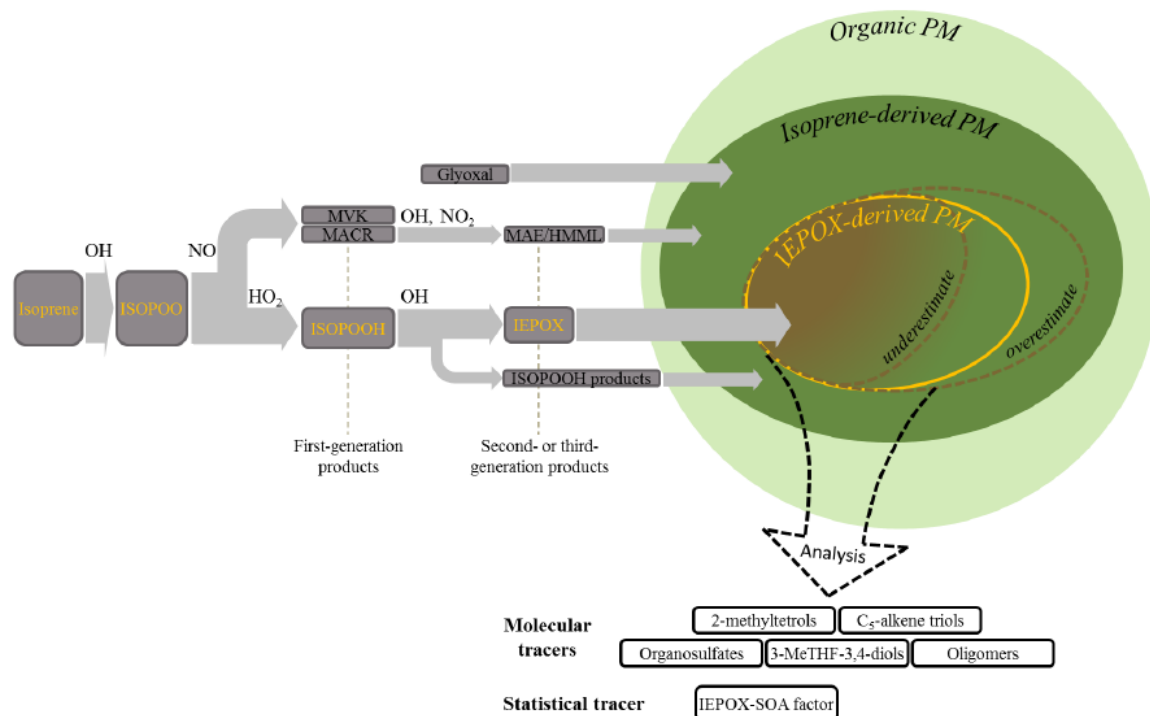


Figure 1.2.1 - Schematic diagram for the production of isoprene epoxydiols (IEPOX)-derived PM from the photooxidation of isoprene. Organic peroxy radicals (ISOPOO), produced by OH attack and O₂ addition to isoprene, are scavenged along NO or HO₂ pathways. By the HO₂ pathway, organic hydroperoxides (ISOPOOH) are first-generation products that react with additional OH to produce IEPOX. The IEPOX species undergo reactive uptake into particles, ultimately producing IEPOX-derived particulate matter (from Sá et al., 2017).

1.3 - Aerosol characterization in Amazonia

Several recent large experiments in Amazonia such as AMAZE-2008, BUNIACCIC, ACRIDICON-CHUVA and GoAmazon2014/15 have characterized key properties of Amazon aerosols. The aerosol size distribution varies significantly between wet and dry season, as can be observed in the figure 1.3.1. In the dry season, aerosol are mostly in the accumulation mode, while at the wet season, we can see the presence of Aitken mode. It is remarkable how the ultrafine particle mode are really rare in Amazonia, and Aitken and accumulation modes are present all the time in the wet season while in the dry season we only observe the accumulation mode with average diameter at 150 nm. In the wet season (Figure 1.3.1 left), it is also possible to observe photochemical particle processing in early afternoon. In the dry season, long range transported biomass burning aerosol (aged) dominates the aerosol population in the accumulation mode.

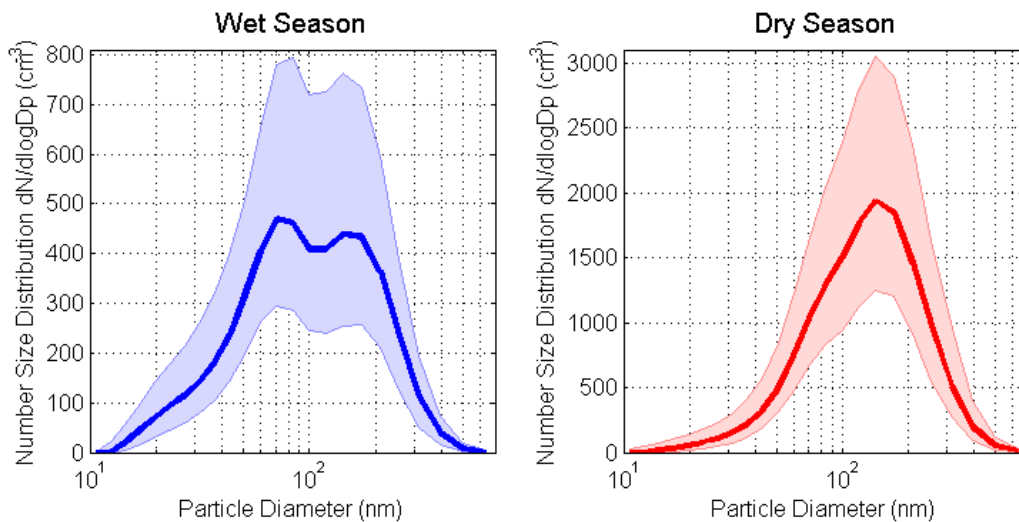


Figure 1.3.1 - Median particle number size distributions for wet and dry seasons at the TT34 – ZF2 forest site in Central Amazonia (Rizzo et al., 2017).

To fully characterize the aerosol, it is important also to measure the organic and inorganic composition. The figure 1.3.2 shows the composition of the inorganic component collected in filters and analyzed by X-ray Fluorescence. Averages for the fine mode at 3 sites in Central Amazonia (ZF2, Tiwa Hotel, Manacapuru) are shown. Sulfur concentrations are low on average (100 ng/m^3).

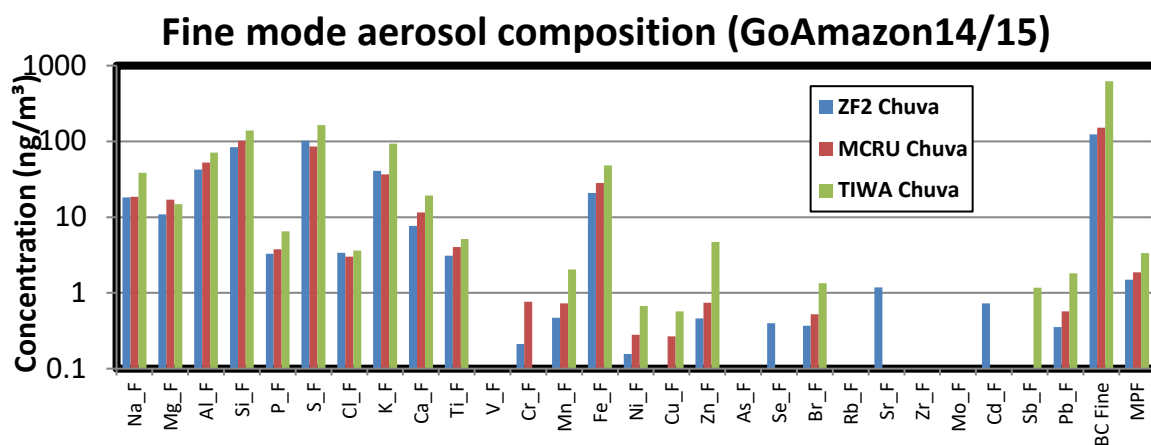


Figure 1.3.2 - Composition of the inorganic aerosol component in the wet season collected in filters and analyzed by X-Ray fluorescence. Averages for the fine mode aerosol at 3 sites (ZF2, Tiwa Hotel, Manacapuru) in Central Amazonia are shown. (Data from Andre Burger, 2017).

In particular, detailed measurements from the organic submicron component was performed using Aerodyne AMS and other techniques (Chen et al., 2009, de Sá et al., 2017). Patterns in the mass spectra closely resembled those of secondary-organic aerosol particles formed in environmental chambers from biogenic precursor gases (Martin et al., 2017). High-resolution mass spectra of SOA particles for the oxidation of isoprene, the monoterpene α -pinene, and the sesquiterpene β -caryophyllene, can be linearly combined to largely reproduce the patterns observed in Central Amazonia (de Sá et al., 2017). The organic mass concentration of the Amazonian ecosystem had an average value of $0.6 \mu\text{g m}^{-3}$ and an average elemental oxygen-to-carbon (O:C) ratio of 0.42. This average O:C ratio is also similar to that of

laboratory SOA particles at realistically-low precursor concentrations. Speciation studies by chromatography show the presence of methyltetrols, which are produced by the oxidation of isoprene (Clayes et al., 2004). Figure 1.3.3 shows the time series and average composition for wet and dry seasons in the three sites in the GoAmazon2014/15 experiment.

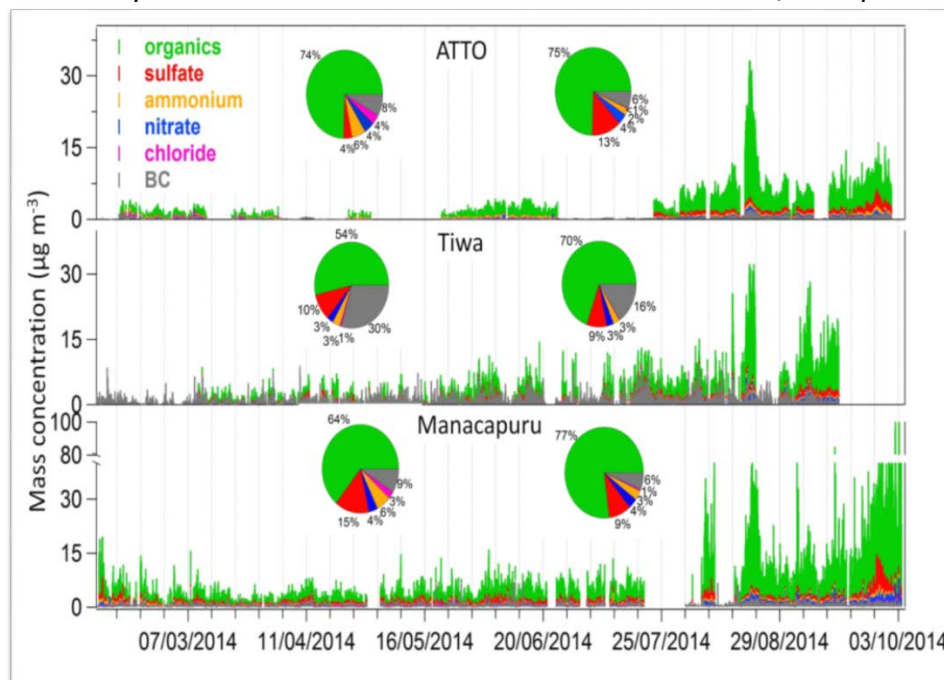


Figure 1.3.3 – AMS measurements from the GoAmazon experiment shows that organic aerosol dominates the fine mass, comprising 55-75% of aerosol mass. BC and sulfate are the second most important component. There is a strong similarity in aerosol composition for the several GoAmazon sampling sites.

1.4 - Cloud Condensation Nuclei (CCN) in Amazonia

One of the links between the aerosol and cloud components happens via the CCN activity of aerosol particles (Pöhlker et al., 2017, Thalman et al., 2017). The undisturbed central Amazon presents very low CCN particle concentrations, on the order of 150-250 CCN cm⁻³ (Pöhlker, M. et al., 2016, Artaxo et al., 1994). These particles are mainly natural primary biogenic particles (coarse mode) as well as SOA produced from oxidation of naturally emitted VOCs (fine mode). Figure 1.4.1 shows a time series of one-year measurements of kappa (the hygroscopicity parameter) at the ATTO 85 meters tower, showing remarkable constant Kappa value of about 0.22, despite large changes in aerosol composition and size distribution during the dry season. Urban emissions within the basin also perturb the natural environment. During repeated aircraft transects of the urban plume, Kuhn et al. (2010) observed that, within the plume core of the Manaus outflow, aerosol concentrations were strongly enhanced, with particle number concentrations reaching 30,000 cm⁻³ compared to background conditions of 300 cm⁻³. Furthermore, only about 15% of the plume particles served as CCN, compared to 60 to 80% in background conditions. Moreover, the CCN concentrations increased with plume age, indicative of the condensation of water-soluble and water-insoluble species on particles. Recently Crooks et al, 2017 proposed a new scheme for the co-condensation of semi-volatile organics into multiple aerosol particle modes. In this new parametrization, the dynamic condensation parameterisation differs from equilibrium absorptive partitioning theory by calculating time dependent condensed masses that depend on the updraft velocity, making the link between SVOC and cloud droplets production.

The sources of CCN in Amazonia are not yet fully understood, since no new particle formation is observed at the ground (Artaxo et al., 2011). Recently it was found out that new

particle formation actually happens unexpectedly in high altitudes (10-14 Km), and it is a key source of CCN in Amazonia (Andreae et al., 2017) (see section 1.6). The high altitude aircraft campaign will study the CCN activity of these newly formed particles, and how they can repopulate the CCN concentrations at cloud base and at the ground. The link between hygroscopicity parameter, particle growth and optical properties will be studied using imaging polar nephelometer at varying relative humidity. The hygroscopicity parameter K will be measured using size selective CCN measurements and the integration with the imaging polar nephelometer with varying relative humidity will link with the optical properties, since refractive index will be measured in parallel.

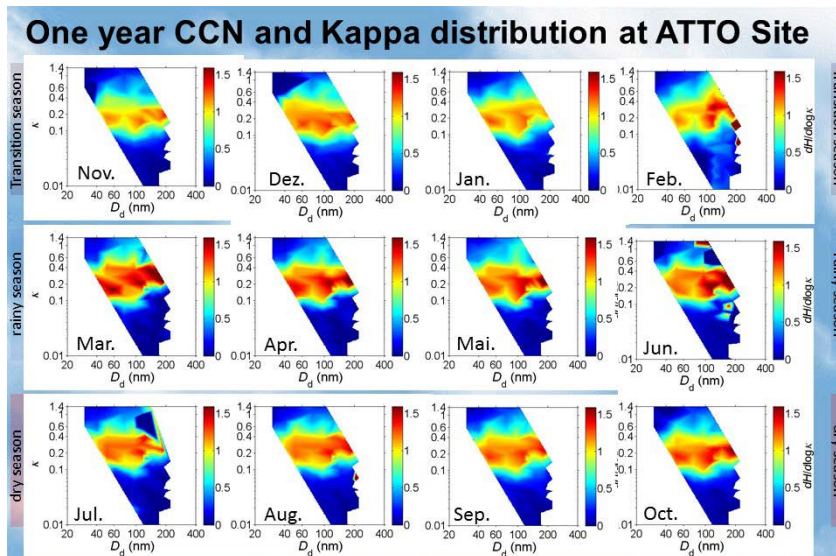


Figure 1.4.1 – CCN and hygroscopicity parameter K measured for one full year at the ATTO site. The dominant K values are around a low value of 0.22, showing the dominance of organic aerosol on CCN activity in Amazonia. Very low seasonal variability as well as variability according to the particle size was observed. From Pöhlker et al., 2016, Thalman et al., 2017.

Larger super micron particles are dominated by primary biological aerosol particles (PBAP) released from rainforest biota (Elbert et al., 2007; Pöschl et al., 2010; Huffman et al., 2012), which can play a significant role as ice nuclei (Prenni et al., 2009). These PBAP consist of wind driven particles, such as pollen, bacteria, and plant debris, as well as actively ejected material, such as fungal and plant spores (Laskin et al., 2017).

1.5 - Optical properties and radiative forcing of Amazonian aerosols

The optical properties of carbonaceous aerosols, including natural biogenic aerosol particles as well as secondary organic aerosol (SOA) and biomass burning emissions is not well constrained (Wang et al., 2016, Laskin et al., 2015). The balance between scattering and absorption is a key property of aerosol, expressed as single scattering albedo (SSA). The SSA, surface reflectance and aerosol optical depth are the ingredients that determines the aerosol radiative forcing. The radiative impact of organic aerosols (OA) is a large source of uncertainty in estimating the global direct radiative effect (DRE) of aerosols (Forster et al., IPCC 2007). This impact includes not only light scattering but also light absorption from a subclass of OA referred to as brown carbon (BrC). Several papers have observed strong absorption properties of natural biogenic aerosols (Schafer et al., 2008; Rizzo et al. 2011). Because of the intricate shapes and composition, biogenic particles can absorb significant amounts of visible light (Andreae and Gelencsér, 2006; Després et al., 2012). Rizzo et al. (2013) showed that the aerosol single scattering albedo (SSA) in a forest site in Amazonia is rather constant along the

year even though there is large variability in the presence of biomass burning aerosols. The surface radiative forcing of aerosol particles have important effects on the ecosystem, including the reduction of total radiation fluxes and increasing the ratio of diffuse to direct radiation (Oliveira et al., 2007, Rap et al., 2015). These changes directly affect plant photosynthetic rate (Cirino et al., 2014), specially on tropical forests, where enhanced photosynthesis was observed to reach 30-40 % of Net Ecosystem Exchange (NEE) for ZF2 station (forest site north of Manaus), Santarém, and Rondônia. The overall effects of aerosols on photosynthetic rate in the whole Amazonia was studied recently in our work published in Rap et al. (2015), and we found that BBA (Biomass Burning Aerosol) increases overall Amazon basin annual mean diffuse radiation by 3.4–6.8% and net primary production (NPP) by 1.4–2.8%. The enhancement of Amazon basin NPP by 78–156 Tg C is very high and equivalent to 33–65% of the annual regional carbon emissions from biomass burning.

Figure 1.5.1 below shows a time series of aerosol light scattering and light absorption from 2008 to 2016 (Saturno et al., 2017). It is very clear that a strong seasonal cycle can be observed and that the year-to-year variability in aerosol optical properties is very high. In this figure, the El Nino index is also shown, indicating the strong influence of large-scale climatic forcers in aerosol properties in Amazonia, through variability in precipitation and biomass burning.

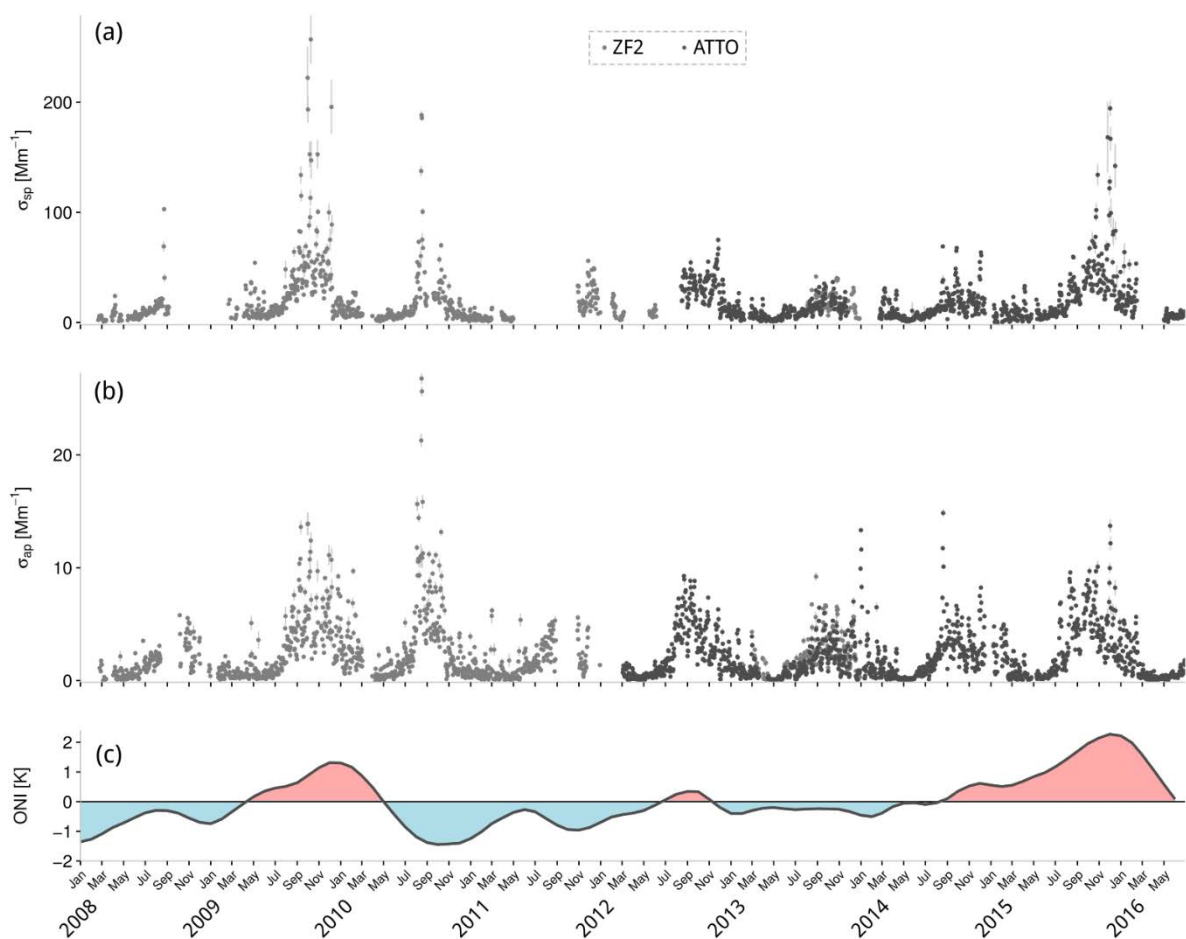


Figure 1.5.1 - Long time series (2008-2016) of aerosol light scattering (a) and light absorption (b) in Central Amazonia. El Nino index is also shown (c), indicating the strong influence of large-scale climatic forcers in aerosol properties in Amazonia.

Other long-term measurement network that IFUSP is running in partnership with NASA Goddard is the AERONET network. Figure 1.5.2 shows the time series of aerosol optical depth (AOD) measured from 2000 to 2017 using the AERONET sunphotometer network in several sites in Amazonia. These 17 years of continuous measurements in difficult conditions in Amazonia shows a lot of information in addition to AOD, such as aerosol size distribution from Almuçantar inversions, refractive index, single scattering albedo and scattering and absorption Ångström exponent, among other key aerosol properties.

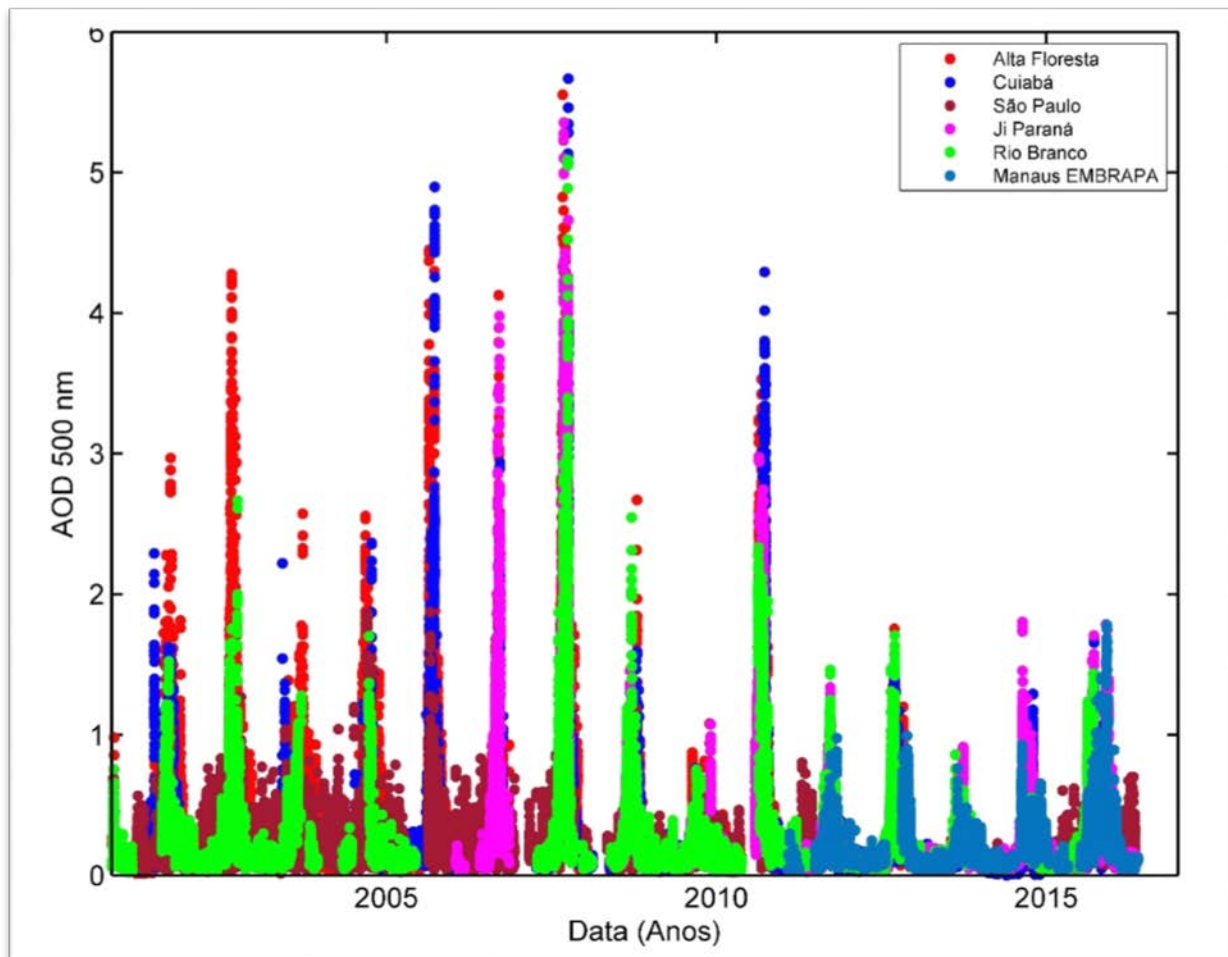


Figure 1.5.2 Time series of aerosol optical thickness measured from 2000 to 2017 using the AERONET sunphotometer network in several sites in Amazonia. AERONET Level 2 data with inversion algorithm 3.0. (Compilation done by Rafael Palácios, 2017)

1.6 - Aerosol production at high altitudes and convective transport by clouds

A lack of evidence for new particle formation at ground-based measurements (Zhou et al., 2002; Rissler et al., 2004; Martin et al., 2010a) in Amazonia implies that nucleation processes could occur at higher altitudes, and new particles are entrained into the boundary layer from aloft (Krejci et al., 2005; Wang et al., 2016, Andreae et al., 2017). This particular relationship between VOC forest emissions, vertical transport due to deep convection and aerosol aging at high altitudes is an important particle production mechanism unveiled recently as a result of the GoAmazon2014/15 and the ACRIDICON-CHUVA measurements and this project will address this issue further. Figure 1.6.1 shows aerosol size distribution and total

particle concentration as a function of altitude measured with the G1 aircraft up to 5.5 Km (Wang et al., 2017). It is clear that concentrations increase with altitude, indicating that the source of these particles is at high altitudes. As many of the aerosol particles go through cloud processing several times before they are deposited, and Amazonia has one of the most intense hydrological cycles in terrestrial ecosystems, this mechanism is critically important to sustain the concentrations of aerosol particles and CCN in tropical regions.

Vertical profile of particle size distribution under pristine condition during wet season

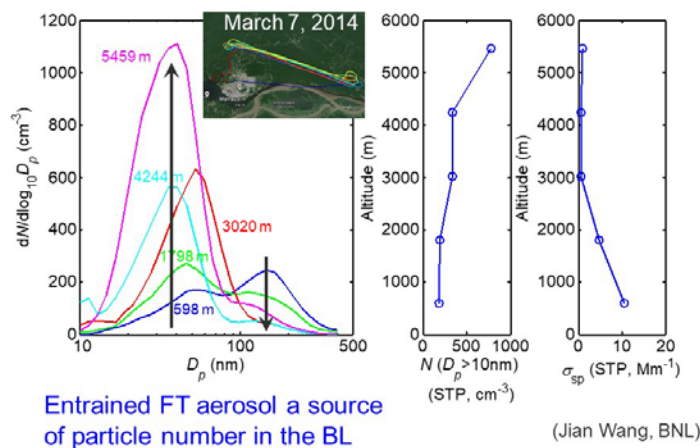


Figure 1.6.1 – Measurements of aerosol size distribution shows an increase in concentrations and decrease in size for higher altitudes, normally the opposite what could be observed if the source of particles are on lower altitudes. (Wang et al., 2017)

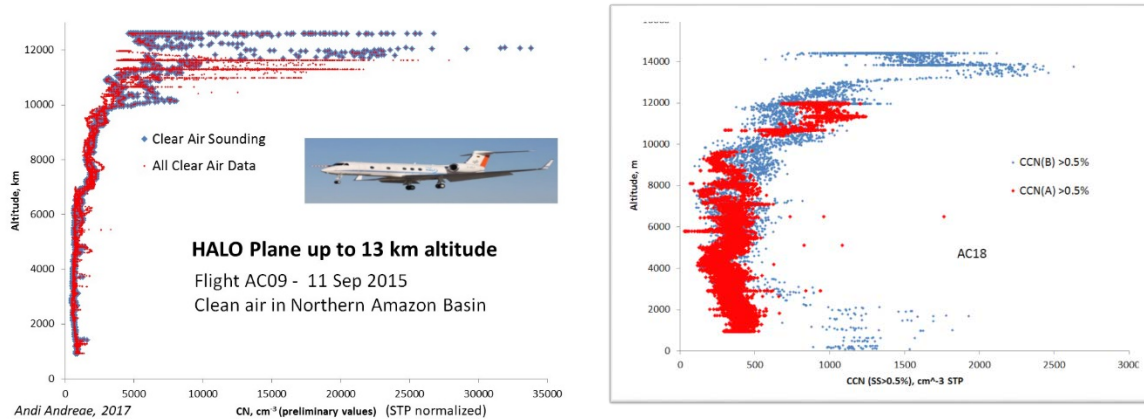


Figure 1.6.2 – Vertical profile up to 13 km of particle concentrations measured by the HALO high altitude plane. It was found very high concentrations of more than 22,000 particles per cc at 12 km altitude. The picture on the right is the vertical profile of CCN, also showing high concentrations above 10 Km.

Fortunately, we also had an experiment with the HALO high altitude plane in the dry season of 2014. Figure 1.6.2 shows the vertical profile up to 13 km of particle concentrations measured by the HALO high altitude plane. Very high concentrations of more than 22,000 particles per cc were found at 12 km altitude. The picture on the right is the vertical profile of CCN, also showing very high concentrations for higher altitude. The enhanced particle concentrations in the ultrafine (UF) size range (here defined as particles smaller than 90 nm), on the other hand, cannot be explained by transport from the lower troposphere, since they far exceed typical concentrations in the PBL and generally are too short-lived to survive deep convection and long-range transport. Therefore, nucleation and new particle formation (NPF)

from gas phase precursors brought into the UT by the outflow from deep convection have been proposed as the source of these enhanced particle concentrations (Andreae et al., 2017). No enrichment of sulfate was observed at high altitudes, and possibly extremely low volatility organic compounds (ELVOCs) are involved. The production of particles in the UT may be a key component of the atmospheric budget of optically and cloud-microphysically active aerosols, especially in pristine or relatively unpolluted regions. In turn, the concentrations of aerosols in the PBL have a pronounced influence on the characteristics of convection and thereby influence cloud radiative forcing and atmospheric dynamics (Cechini et al., 2017). Figure 1.6.3 shows a conceptual framework that will be tested with measurements in this project (adapted from Wang et al., 2017, Andreae et al., 2017).

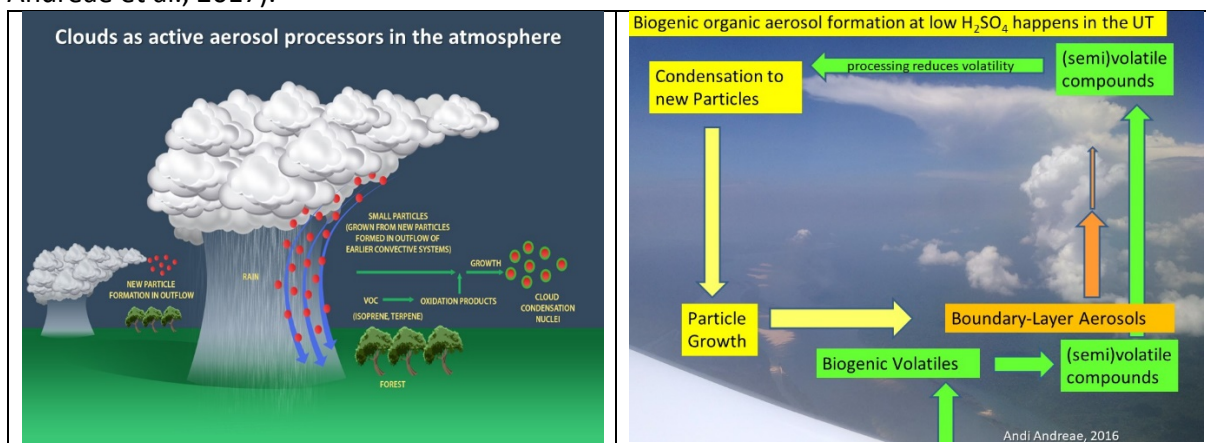


Figure 1.6.3 - On the left panel we can observe how clouds can be active aerosol processors in the atmosphere of Amazonia. The right panel shows how semi volatile compounds produced by the vegetation can be transported to the upper atmosphere by convection, where it condenses due to low temperature and is processed and oxidized to less volatile compounds and produce particles that are brought to the low atmosphere by precipitation.

In this proposal, we are planning a dedicated airborne campaign to further clarify the processes that leads to this aerosol production in high altitude, and how VOCs and organic aerosols are vertically transported through cloud processes. This campaign will be done in partnership with Max Planck Institute and INPE.

1.7 - Precipitation, clouds and climate

In our current understanding of the Earth's climate system and its man-made perturbation, the multi-scale and feedback rich life cycles of clouds represent one of the largest uncertainties (Boucher et al., 2013, Stevens et al., 2016). Accordingly, the adequate and robust representation of cloud properties is an Achilles' heel in climate modeling efforts (Bony et al., 2015). The picture is aggravated because the Amazon has different cloud regimes in the several sub regions (Saraiva et al, 2016), a feature not well represented by Global Climate Models (GCMs). The microphysical link between clouds and aerosol has been the subject of manifold and long-term research efforts in Amazonia (Wendish et al., 2016, Cechini et al., 2016). Overall, the observations indicate that increasing aerosol concentrations can have substantial impacts on spatial and temporal rainfall patterns in the Amazon (e.g., Martins et al., 2009a; Reutter et al., 2009) but may also alter cloud dynamics (e.g. Camponogara et al, 2017). Also, surface patterns such as deforestation and urban areas may affect cloud patterns (e.g. Dos Santos et al, 2014, Saad et al, 2010, Albrecht et al. 2011). In view of the globally

increasing pollution levels and the ongoing deforestation in the Amazon, pollution-triggered perturbations of the hydrological cycle are discussed as potential major threats to the Amazonian ecosystem, its forest structure, stability, and integrity (e.g., Wright et al., 2017, Coe et al., 2013; Junk, 2013).

Especially in Amazonia, clouds are critically important to sustain a vigorous hydrological cycle, recycling water vapor and strongly influencing most of atmospheric properties (Martin et al., 2010). Specific cloud life cycle research areas are categorized into three broad areas: *dynamics*, the atmospheric motions that generally dominate cloud life cycles; *microphysics*, the properties of cloud droplets/ice particles and rain and snow hydrometeors and the processes that determine these properties and their interactions; and *radiation*, the impacts of cloud amount and properties on absorption, emission, and transport of shortwave and longwave radiation (Rosenfeld et al., 2008, Boucher et al., 2013). There are a significant number of studies that highlight the mechanisms by which interactions among land surface processes, atmospheric convection, and biomass burning may alter the timing of wet season onset and provide a mechanistic framework for understanding how deforestation extends the dry season and enhances regional vulnerability to drought (Wright et al., 2017).

Climate models have great difficulty in simulating deep convective clouds and their radiative effects. The convective activity and the atmospheric circulation of tropical South America are part of an American monsoon system, and any changes in tropical precipitation can have significant, potentially global consequences because of non-linear interactions of tropical waves with tropical precipitation in the Amazon, leading also to possible changes in the tropical Atlantic intertropical convergence zone (ITCZ). Models of future climate accounting for human activities have suggested a possible drying in Amazonia, especially in the eastern region (Nobre et al., 1991; Boisier et al., 2015). Significant changes in the amounts and patterns of precipitation in the basin can have far-reaching consequences because of the nonlinear, multi-scale interactions that affect clouds, precipitation, and atmospheric circulation, leading for example to possible modifications in the annual migration of the Intertropical Convergence Zone (ITCZ) (Wang and Fu, 2007). The hydrological cycle in the Amazon basin has changed over the past 2 decades, but the causes are not fully identified and understood (Davidson et al., 2012; Gloor et al., 2013). Moreover, Camponogara et al (2017), among others, show the potential impact of increased concentration of CCN upon cloud dynamics, essentially the strength of cloud scale updrafts and downdrafts, thus affecting the organization of mesoscale convective systems.

The GoAmazon and Acridicon-chuva experiments strongly improved our knowledge on aerosol-cloud-precipitation interaction, several papers discuss the different parameterizations, the effect of Manaus plume, and the sensitivities of these interactions. However, mostly all of these studies deal with only aerosol concentration and preliminary studies show how important is the aerosol size distribution and hygroscopicity. The aerosol effect depends from these parameters because the cloud droplet formation can be saturated as function of the water vapor availability. All these features need to be better describe and modeled to provide a clear picture of the CAPI. A critical issue for GCMs which is still open is that GCMs is not able to organize convection. This GCM deficiency could have an important impact in climate simulation. Amazonia is the perfect place to study cloud organization over land because of the green ocean behavior and the long lived tropical squall lines.

2 – Current Scientific Gaps

Although there were important and large scientific experiments in Amazonia over the last 30 years studying atmospheric properties, the system is very complex. The interactions between the forest biology and atmospheric properties and climate is very strong. We will describe below the main scientific gaps we observe in Amazonia for the several project components.

2.1 – Aerosol Life cycle in Amazonia

Aerosol Life Cycle in Amazonia: Aerosol particles exert important influences on climate and climate change by scattering and absorbing solar radiation with strong impact on radiation balance and by influencing the properties of clouds (Boucher et al., 2013). The aerosol life cycle determines the spatial and temporal distribution of atmospheric particles and their chemical, microphysical, and optical properties. It is essential to improve understanding of the roles of aerosols in the climate system and specifically to decrease uncertainty in radiative forcing by aerosols (Forster et al., 2007). Current aerosol models require improvement in several areas: emissions, mechanisms, new particle formation events, aerosol physical-chemical changes under biogenic and anthropogenic influences. The research that will be conducted by this proposal is usefully distinguished into four topical areas or elements: new particle formation, aerosol growth and aging, the direct radiative impacts of aerosol, and separating the natural vs. anthropogenic aerosol influences on aerosol properties.

New particle formation: Besides primary emissions of primary aerosol particles, another important source of atmospheric aerosols is the chemical conversion of gas-phase atmospheric precursors to new particles through nucleation and growth (Kulmala et al., 2012). New particle formation (NPF), which affects the number concentration of particles, is a key process in aerosol dynamics that governs size-distributed composition and in turn the aerosol optical and cloud nucleating properties. Freshly nucleated particles are approximately a few nanometers in size and thus are too small to directly influence climate. But these particles can quickly grow, even over the course of a single day, to reach the size where they can serve as cloud condensation nuclei (CCN) or appreciably scatter light (Heald et al., 2010). A strong coupling between nucleation and growth rates of nucleated particles further contributes to the climate importance of new particle formation. As particles grow their ability to serve as CCN or Ice Nuclei (IN) depends strongly on the size and composition of the particle (Fuzzi et al., 2007). Amazonia is a place where NPF is rarely observed naturally on the ground (Artaxo et al., 2009), but new observations indicate that these particles could be formed at high altitudes (Wang et al., 2016, Andreae et al., 2017). Which is the partitioning for the different particles types in Amazonia? Is *high altitude NPF the only source of new aerosol particles in Amazonia?*

Aerosol Aging and Mixing State: Aging of aerosols consists of modification of the composition, size, and surface properties of aerosol particles in the atmosphere by coagulation, condensation, and surface reactions. These processes are important as they affect the optical and cloud nucleating properties of the aerosol. Measurements downwind of urban sources of aerosol particles and precursor gases have shown that the mass

concentration of secondary organic aerosol (SOA) can be several-fold greater than can be explained on the basis of current model calculations using observed precursor concentrations. The dependence of SOA formation on sulfur and nitrogen oxides and other factors will be examined in this proposal. This information is essential to the development of comprehensive chemical mechanisms that can contribute to a better understanding of the remote aerosol life cycle (Heald et al., 2008, 2010, Carslow et al., 2016). *How aerosol aging, hygroscopicity and mixing state influence particle properties?*

Direct radiative impacts of aerosol: Scattering and absorption of solar radiation by aerosols modify the amount of incoming solar radiation taken up by Earth, and modify the vertical distribution of that absorption and the resultant heating profile of the atmosphere. (Trenberth et al., 2009). Aerosol optical properties depend strongly on particle size, composition, mixing state and morphology, all properties that will be measured in this study. Previous studies from the IFUSP group shows a large effect from biomass burning aerosol on regional direct radiative balance (Procópio et al., 2003, 2004, Sena et al., 2013). Aerosols containing black and “brown” carbon absorb radiation in the visible spectral region (Martins et al., 1998, 2009, 2010) and have a potential large impact on climate by reducing the net solar radiative flux at the Earth’s surface and by warming the air in their vicinity, possibly leading to cloud evaporation (Koren et al., 2012). Measurements at the ATTO tower and at the 7 NASA/AERONET and SunRadNet sites in Amazonia will help to understand the processes that controls the effects of aerosols on the radiation balance in Amazonia. *What is the large-scale effect of aerosol radiative forcing in carbon uptake, changes in the vertical dynamics and other key effects in Amazonia?*

Natural vs. Anthropogenic Influences on Aerosol Properties: Measurements that distinguish natural and anthropogenic influences on aerosol properties are needed to determine the anthropogenic perturbation to radiative forcing (Carslow et al., 2016, Andreae et al., 2007, Artaxo et al., 2008). Both anthropogenic and natural processes contribute to the atmospheric aerosol loading, influencing the new particle formation, the role of anthropogenically enhanced levels of atmospheric oxidants in SOA formation from natural and more volatile organic emissions, and the oxidation reactions on aerosol surfaces and within cloud droplets. *How does the long range transport of aerosols from Africa and outside the basin influence atmospheric chemistry in Amazonia?*

2.2 - Cloud Life Cycle in Amazonia

Atmospheric dynamics: Convection and advection air motions play a central role in cloud life cycles. Whereas emissions and nucleation lead to new aerosol particles, it is primarily atmospheric dynamical conditions in a sufficiently humid environment that lead to new cloud particles. To simulate cloud life cycles, models must adequately represent the strength and depth of updrafts and downdrafts. Important parameters in clouds include vertical air velocity variability, the structure of turbulent motions, the skewness of the vertical air velocity distribution, and atmospheric stability profiles. This proposal will use a multi-instrument and -platform approach to study the variability of these dynamical parameters with the cloud microphysics in order to reveal the important linkages between boundary layer dynamics, radiation, cloud formation, and cloud composition, all of which are crucial to understanding the life cycle of these clouds. Entrainment of environmental air into clouds is a

key process that is poorly understood and we will use a modeling approach to better understand processes that regulates entrainment in shallow clouds in Amazonia (Williams et al., 2002; Braga et al. 2017a, 2017b). *How do boundary layer dynamics, radiation, cloud formation, and cloud composition affect cloud life cycle?*

Cloud microphysics: Accurate knowledge of the hydrometeor number, size, surface area, volume or mass, dispersion, skewness, and phase are required in order to understand basic cloud processes such as the microphysical evolution through competition for available water vapor, formation of precipitation-sized particles, sedimentation, and collisions among cloud particles. Within mixed-phase clouds both liquid and ice size distributions exist within the same cloud system, interacting and coevolving through myriad, complex mechanisms, also being responsible for the main charging mechanism in cloud electrification.. Several key parameters in cloud model simulations remains very poorly constrained by measurements. *How to reconcile models and measurements of cloud droplet size and CCN concentrations for liquid and mixed phase clouds?*

Cloud processing of particles: Aerosols and clouds are inextricably coupled throughout their life cycles in processes that dictate cloud formation and development, spatial coverage, persistence, and precipitation efficiency (Khain, 2009). Cloud processing of aerosols plays an important role in aerosol chemical and microphysical properties through aqueous-phase chemistry, aerosol removal and vertical redistribution mediated by precipitation and vertical motions, especially in a convective region as Amazonia. The light-absorbing properties of aerosols through BC and BrC can have a strong influence on cloud dynamics through heating. Typically, the presence of absorbing aerosol is thought to change atmospheric stability, suppress vertical motion, and decrease cloud formation (Gonçalves et al., 2014, Koren et al., 2004). On the other side, recent studies shows that clouds are active in processing aerosol particles, changing their hygroscopicity, size, and properties, but the mechanisms are unclear. *How do cloud processing of aerosols change their physico-chemical properties?*

2.3 – Modeling aerosol-cloud interactions in Amazonia

In the tropical to equatorial regions, particularly in the heart of the Amazon Basin, clouds play a major role in several processes spanning a range of scales. They are also the main cause of uncertainty in numerical modeling of the atmosphere from activities that range from weather forecasting, to seasonal forecasting, to climate projections and in a broader sense to Earth Climate System Modeling. As shown by recent modeling studies (Raupp and Silva Dias, 2009, 2010) the diurnal life cycle of clouds in atmospheric models is the basis for resonant wave interactions that excite slower modes in the atmosphere (e.g. Madden-Julian Oscillation).

The complex interactions between clouds and aerosol have been the focus of many studies in the last decade or so (for instance Silva Dias et al., 2013). Local observations, remote sensing data and modeling have been combined to unravel the microphysical, thermodynamic and dynamic causes for observed behavior and to reduce uncertainty of model simulations. However, still no final answer has been reached on the overall, global impact of aerosols on clouds, and to a certain extent of clouds on aerosol. The reason for this is that the main process describing the microphysical processes, the formation of the different hydrometeors still lack in representation of the observable cloud processes. From another side, turbulence has very

important implications in the entrainment, microphysical process, energy exchange and this is roughly represented. Important effort should be done to improve these representations to make the model more representative of the observable fields. Over land, in tropical region, models have several issues in the rainfall localization and timing. Without the improvement of these specific topics, models will not be able to represent correctly the aerosol-cloud-rainfall interaction. One example of complex interactions happens during the transition between the dry and the wet season, when deep convection may be seen as quite continental showing severe convective features (Nunes et al 2016, Saraiva et al 2016) including deeper cloud cores (Silva Das et al 2002, Carvalho et al 2002) and strong electrification (Williams et al 2002, Albrecht et al 2011).

The region of Central Amazonia is particularly well suited to study the evolution of tropical convective systems and their regional and global upscale feedbacks since it experiences a wide range of convective storm types and environmental conditions throughout the year. There is a strong seasonal cycle in rainfall with a maximum in March-April (wet season) and a minimum in August-September (dry season). Large mesoscale convective systems are predominant in the wet season while more isolated but intense thunderstorms are predominant in the dry to wet transition season (Machado et al., 1998; Machado et al., 2004; Romatschke and Houze, 2010; Rasmussen and Houze, 2011, Nunes et al 2016). In the wet to dry season, large squall lines originated at the Northern Coast of South America propagate throughout the Central Basin (Alcântara et al., 2011). Local circulation is formed where contrasting surface features impose local gradients of temperature, moisture and atmospheric pressure. In the Amazon, these conditions have been associated with deforestation (Silva Dias et al., 2009, Saad et al., 2010) and with large rivers (Silva Dias et al., 2004, Lu et al., 2005, Dos Santos et al 2014)). Paiva et al. (2011) have shown that rainfall is reduced over large Amazonian rivers, and the authors point to reduced surface sensible heat and the occurrence of local circulation as the reason. Thus, the Central Amazonia region is a natural laboratory, not only to observe the characteristics of the tropical continental convective life cycle, but also to study cloud-aerosol precipitation interactions, and the role of land surface processes. *How do aerosol-cloud interactions influence precipitation in pristine conditions in Central Amazonia?*

3 - Objectives of the proposed work

This project aims to study aerosols and clouds lifecycles, and their impact in the Amazonian ecosystem using long-term measurements at ATTO, river ships, the high altitude HALO airplane and measurements in the Andes (Chacaltaya). The **key scientific questions** to be answered by this project are composed by four main topics:

1. **Secondary Organic Aerosol (SOA) formation: Interactions of biogenic and anthropogenic emissions**
 - 1a. What are the chemical and physical processes that controls and affect the production of SOA in Amazonia?
 - 1b. How are (organic) particles produced (e.g., nucleation and SOA formation)?
 - 1c. What are the potential roles of primary particles (fungal spores, bacteria, and leaf cuticle) as cloud condensation nuclei (CCN)?
 - 1d. In the pristine Western Amazon, what is the contribution of the IEPOX-SOA component? What is the role of ELVOCs?

1e. How SOA is formed and oxidized at high altitude (>12 Km)?

2. Links between particle size distributions, optical properties, and cloud condensation nuclei (CCN) activity

2a. What are the main characteristics of the life cycle of aerosols in the Amazon, and what are the impacts of the biogenic, soil dust and long range transported biomass-burning emissions on the atmospheric chemistry in Central Amazonia?

2b. What is the impact of “Brown Carbon” (BrC) in aerosol absorption in pristine conditions? How is the partition between BrC versus BC absorption as function of biomass burning aging in Central and Western Amazonia?

2c. How is the aerosol absorption in Central Amazonia linked with the atmospheric conditions leading to cloud formation, evolution and lifetime?

2d. What is the influence of primary and secondary organic aerosols on the cloud condensation nuclei (CCN) activity and in the hygroscopicity parameter Kappa?

3. Biogenic Volatile Organic Compound (BVOC) emissions and impact on atmospheric chemistry and aerosol production

3a. What are the characteristics of BVOCs emission from vegetation, and how do they vary with season and climate conditions?

3b. In BVOC emission, what are the roles of the physical environment (e.g., temperature, rainfall, radiation, and nutrients), environmental perturbations (e.g., drought, nutrient deposition, and temperature extremes)?

3c. What is the relative contribution of isoprene oxidation products ISOPOOH (low NO_x conditions) and MVK+MACR in Western Amazonia?

4. Impact of aerosol particles on cloud processes and precipitation in Amazonia.

4a. What is the more appreciated microphysical parameterization for shallow and deep convective clouds in the Amazonas region?

4b. What are the physical parameters that improve model rainfall field descriptions in the Amazonas region.

4c. What are the physical parameters controlling the Amazonas cloud organization and life cycle in convective parameterization models?

4d. What are the controls of cloud microphysics, aerosols and cloud dynamics upon cloud life cycle and rainfall in the Amazon region?

4e. What are the controls of clouds mixed phase in the Amazonas region?

4f. What are the relative roles into convective cloud intensity and severity in the Amazon of large scale dynamics, local thermodynamics and aerosol concentration?

4g. What controls the transition between shallow and deep convection in the Amazon region and why models do not simulate this transition properly?

4h. How is the diurnal cycle of convective activity linked to aerosol variability?

4i. What is the relative impact of surface features, such as large rivers and land use features and patterns of deforestation, and aerosols upon convective cloud life-cycle?

We propose to use long-term observations from the ATTO tower, boat measurements in Western Amazonia, observations over the Andes, high altitude aerosol and trace gas measurements using the HALO plane to answer these questions. We will use a range of instruments, remote sensing from surface and satellite based sensors, and numerical modeling of physical processes acting in several spatial and temporal scales as tools to advance the understanding of the underlying processes expressed in the above questions.

The main theme uniting these objectives is the development of a data-driven knowledge base for predicting how the present-day energy and mass flows in the Basin might change by internal forcing prevent from projected changes in the Basin. Our ultimate goal is to estimate future changes in direct and indirect radiative forcing, energy distributions, regional climate, and feedbacks to global climate by improving our understanding of the basic processes controlling the atmospheric chemistry, clouds, precipitation, mass and energy fluxes in Amazonia.

6 - Measurement strategy

We are proposing a complex set of measurements with different strategies, involving long-term ATTO tower measurements, Riverboat, Andes and large-scale aircraft. A coherent set of instruments to characterize aerosols CCN, clouds and trace gases will be used in each of the sampling sites. In this section, we describe the measurement strategy to achieve our project goals.

6.1 – Long term measurements at the ATTO – Amazon Tall Tower Observatory

The ATTO site is located in one of the most pristine sites in continental areas in the world, with coordinates at S 02° 8' 38.8", W 58° 59' 59.5". At this site, a German-Brazilian scientific cooperation had already built four 85 meters towers and a 325 meters tall tower. The site is already fully operational and aerosol and trace gases are being measured in one of the 85 meters auxiliary towers, and also starting in July 2017 at the 325 m tall tower. The site is operated by INPA – The Brazilian Institute for Research in Amazonia, and have support from the Max Planck Institute, UEA (Universidade Estadual do Amazonas) and many other partners, including USP and INPE. Figure 6.1.1 shows the location of the ATTO tower, and part of the measurement strategy, adapted from Fuentes et al., 2016.

The figure 6.1.2 illustrates the 325 meters tower. Aerosol physical and chemical properties are being measured at ATTO including the organic aerosol composition with an Aerosol Chemical Speciation Monitor (ACSM) from IFUSP. The site will be kept with continuous measurements in a long-term basis (>20 years). The pristine condition of this site makes it perfect for background aerosol characterization. The site is very difficult to access, requiring many hours for a trip from Manaus to the ATTO site. Table 6.1.1 shows the main continuous measurements that will be done at the ATTO tower continuously. Most of the instruments are already operational, and this proposal will provide a few extra key measurements such as the cloud radar, the Micro Pulse Lidar, the real time total organic carbon analyzer, the AE33 Aethalometer, and a few others.

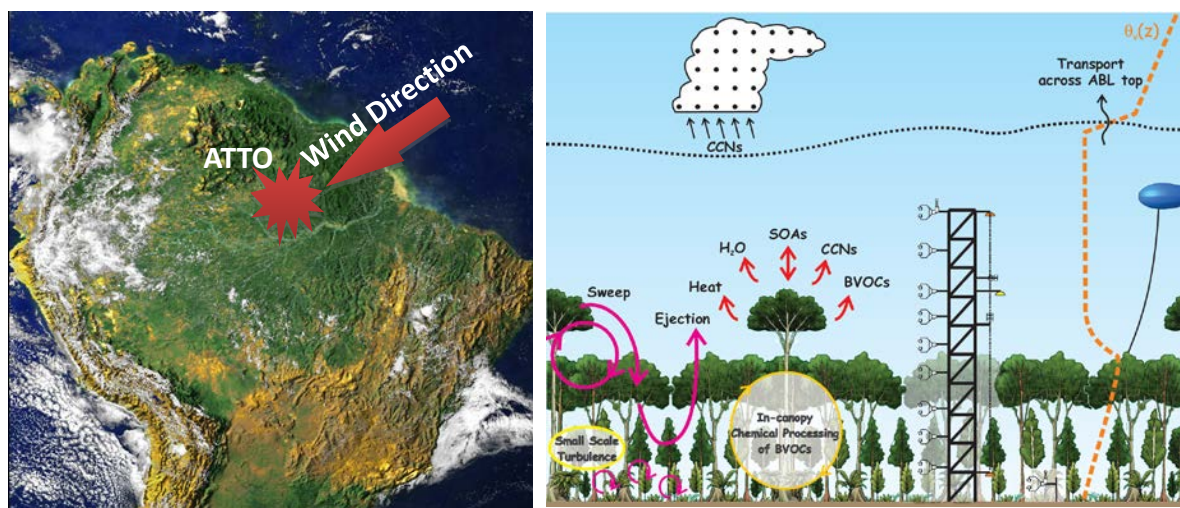


Figure 6.1.1 – Location of the ATTO tower in central Amazonia, downwind of very pristine areas. At the right figure, an illustration showing the exchange of trace gases and aerosols and the role of canopy and PBL in regulating concentrations and processes (adapted from Fuentes et al., 2016).

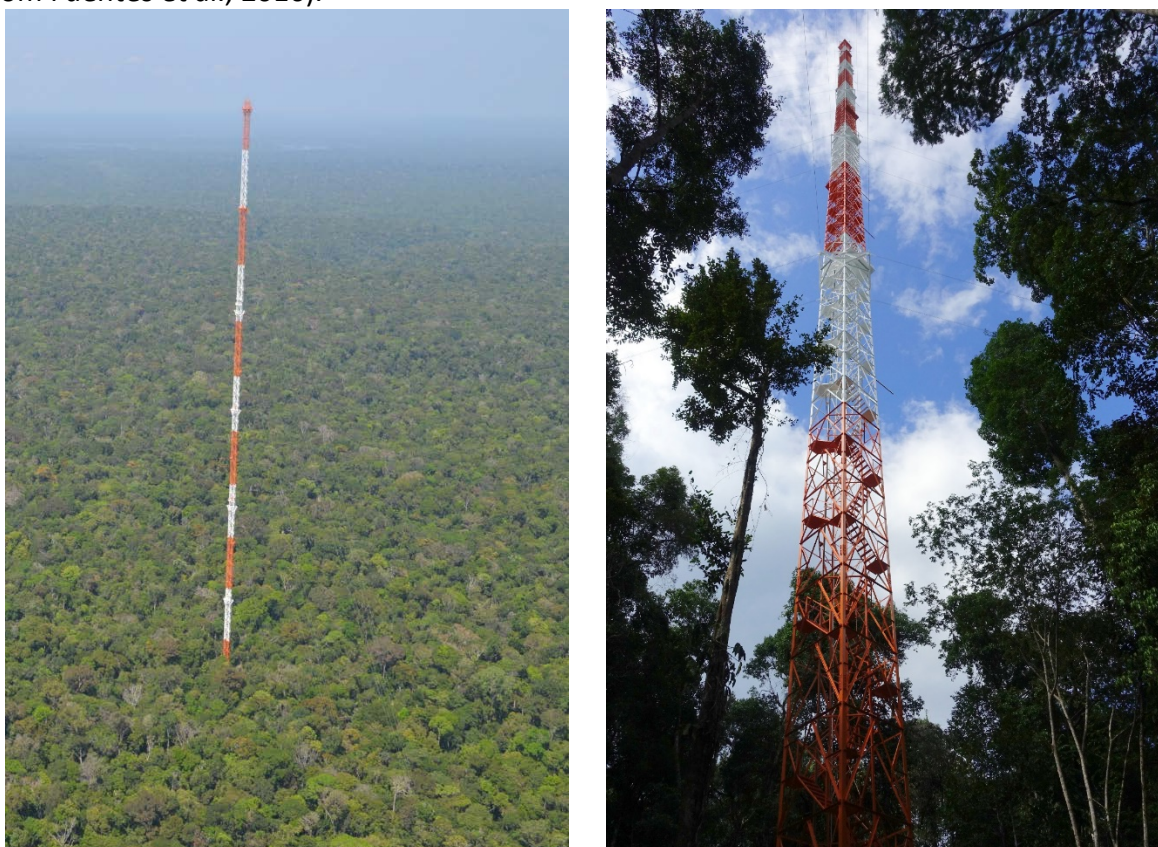


Figure 6.1.2 – The ATTO 325 meters tall tower in Central Amazonia. Top of the tower as well as profiles for trace gases and aerosols will be measured continuously over 5 years of this project.

Vertical profile of trace gases and aerosols will be measured along the tower, and data will be analyzed together with 8 levels of vertical fluxes measurements. A Micro Pulse Lidar in parallel with a cloud radar will provide the vertical profiles up to 12 Km. Detailed physicochemical properties will also be measured along the vertical, to analyze the role of the forest canopy at low levels and how it influences atmospheric composition up to 325 meters.

We will deploy for specific campaign instrumentation such as API-ToF-MS, CIMS-ToF-MS, PTR-MS to measure detailed organic gas composition in parallel with aerosol measurements, including size selective CCN concentration. Brown Carbon and Black Carbon will be measured as well as solar radiation fluxes and aerosol optical thickness with CIMEL sunphotometers to study radiation balance at the ATTO tower.

Table 6.1.1 - Instrumentation to measure aerosol, trace gases, radiation fluxes and cloud properties at the ATTO site.

Cloud Condensation Nuclei CCN from DMT – Droplet Measurement Technology.	8 levels of Eddy Correlation System – Latent and sensible heat fluxes
Nephelometer from TSI model 3563	Microwave Radiometer
Particle/Soot Absorption Photometer – PSAP – Spectral light absorption	Downwelling Radiation
TSI 3776 Condensation Nuclei Counter	Shaded Black and White Pyranometer
Aerodyne Aerosol Chemical Speciation Monitor (ACSM)	Normal Incidence Pyrheliometer
Photo-Acoustic Soot Spectrometer (PASS-3)	8 levels of 3D Anemometers
Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)	Vaisala Ceilometer (range ~7 km)
PTR-MS Real-time VOC	Vertically Pointing Cloud Radar (94 or 35 GHz)
Carbon Monoxide Analyzer from Picarro	Total Sky Imager for cloud cover
Trace Gas – O ₃ , SO ₂ , NO _x	Narrow Field of View Radiometer
Optical particle counter - OPC	Precision Infrared Radiometer
Aethalometer AE33 for Black Carbon	Precision Spectral Radiometer
Single Particle Soot Photometer (SP2)	Infrared Thermometer
Total column of CO, CO ₂ and CH ₄	Surface Meteorology
Cimel Sun Photometer for AOD	Barometer
Micropulse Lidar with Dual Polarization	Temperature and Humidity Sensor
Doppler Cloud Lidar	Upwelling Radiation
Scanning Mobility Particle Sizer (SMPS)	Multiangle Absorption Photometer (MAAP)
Disdrometers (JOSS, Parsivel and Thies)	Real time total carbon analyzer
SIPAM S-band radar	1290 MHz Radar Wind Profiler (RWP)

6.1.2- Clouds observation framework at the ATTO tower

Our general aim is to better understand the processes that modulate the formation of shallow convection and its subsequent conversion to deep convection in Central Amazonia, and what role do aerosols have in this process. The instruments deployed at the ATTO site will include: (1) remote sensing ranging profilers with LIDAR, ceilometer, microwave radiometer, thermal infrared imager, and a vertical pointing cloud and rain radars; (2) ground observation with disdrometers (JOSS, Parsivel and Thies) and three different meteorological weather stations with P, T, RH, wind and radiation; and (3) column integrated measurements including a multi filter shadow band radiometer, a Cimel AERONET sunphotometer and a Global Navigation Satellite System (GNSS) receiver.

Continuous and long-term observations such as these, combined with satellite images and SIPAM volume scan radar data, enable the characterization of the diurnal cycle of different cloud types, convection and precipitation structure and life cycle. For instance, the

use of the GNSS stations allowed Adams et al. (2011) to describe the typical pattern of afternoon deep convection events. Figure 9 shows an example of PWV time series derived from the GNSS station at INPA. It is clear that there is an increase in PWV before the precipitation event, a decrease afterwards, and that there are concurrent variations of cloud top and surface temperatures. The authors, however, could say very little on the possible role played by moisture convergence during the event. This could be tackled by the integrated use of all GNSS stations and soundings that will allow precise derivation of the vapor convergence and transport while different types of radars observe the formation of deep convection over the sites. Vertical profiles of diabatic heating associated with continental and maritime convective systems derived from sounding (Yanai et al., 1973) and radar (Schumacher et al., 2004) measurements, and the connection of heating profiles to the reversal of the large-scale wind pattern derived from the radiosonde network, wind profilers, and reanalysis data will shed light on how convective heating is linked to moisture transport to central Amazonia.

Looking at the water vapor content in the atmosphere is only one aspect of the shallow to deep transition. At the same time, ceilometers and Lidar will provide the vertical thermodynamic profile, distribution of aerosols and position of cloud base/top. This was done by Bourayou et al. (2011) with data from CHUVA's campaign in Fortaleza. They compared cloud base heights from MP3000, MRR and LIDAR and found that only the LIDAR gives a reasonable measurement. On the other hand, as pointed out by Barbosa et al. (2012), it is very hard to translate the time evolution of a single vertical profile to what shall be the horizontal distribution of clouds and their instantaneous development stage. This caveat can be overcome combining these vertical profiles with the spatial distribution of precipitation from SIPAM radar (Giangrande et al. 2013; Kumar et al. 2015; Giangrande et al., 2016). These combinations provide mass fluxes of cloud particles and precipitation which are parameters more directly comparable to those of cloud models (Kumar et al. 2015). The proposed detailed measurements will also be able to address fundamental aspects of aerosol and cloud microphysics and can be used to advance our understanding of the aerosol-cloud interactions.

6.2 – Large scale measurements using ships in western Amazonia

Western Amazonia is still a region where no systematic atmospheric measurements were done, mostly because of difficult logistics. However, of course is an important region in Amazonia, with most of the area consists of untouched forest. In addition, the climatology is different from Central and Eastern Amazonia, which could make the forest emissions different from the Santarem and Manaus regions, for instance. Figure 6.2.1 below indicates this region, and where fluvial measurement campaigns will analyze atmospheric properties.



Figure 6.2.1 – Main Rivers in the Western Amazonia, where instrumented boat campaigns will analyze atmospheric properties in pristine regions.

The boat for this component will be provided by USP that recently bought a large laboratory ship to assist in terms of health the Madeira River population and to do research in this difficult to access Amazonia region. The ship belongs to the ICB (Instituto de Ciências Biomédicas da USP) is based in Porto Velho, and every 15 days it goes down the Madeira River in very undisturbed regions in Western Amazonia.



Other boat platform that we plan to use is the new UEA (Universidade Estadual do Amazonas), a river research vessel planned from the original project to be research ship for Amazonia. The figure bellow shows an illustration of the ship that is under construction in the final stage in Manaus, and should be ready in mid-2018. We can alternatively also rent local boats in other rivers in the basin to complement measurements for this component of the project. The person responsible for this UEA platform is Prof. Rodrigo de Souza.



The main objectives of this component will be to characterize VOC emissions and the SOA production in this remote region. The very high biodiversity in Amazonia and the species dependence of VOC emissions makes necessary to measure atmospheric properties in different conditions and regions. We will use this boat of opportunity to expand atmospheric studies to Western Amazonia. The most important instrumentation for this boat campaign will be: 1) PTR-MS for VOC measurements; 2) ACSM for organic aerosol characterization; 3) SMPS for aerosol size distribution in the range of 7 to 500 nm; 4) Optical properties measurements (Nephelometer and Aethalometer); 5) Total organic carbon analyzer, 6) trace gases (CO, CO₂), 7) Micro pulse Lidar, among others.

6.4 – Large Scale Measurements using the High Altitude HALO plane

One of the most striking findings from the HALO aircraft campaign during GoAmazon was that aerosol particles in Amazonia are produced not at the ground, as in marine and other terrestrial ecosystems, but at high altitudes of about 12-14 Km (Wand et al., 2017, Andreae et al., 2017). The mechanism for this particle production is not clear, and one of the ideas is illustrated in figure 1.6.4, that discuss semivolatile compounds being transported to the upper atmosphere by convection, where it condenses due to low temperature. The particles produced are processed and oxidized to less volatile compounds and produce particles that are brought to the low atmosphere by clouds with active precipitation (Wang et al., 2016, Andreae et al., 2017). We will need a special configuration of the plane to measure precursors and nanoparticles.

In order to detail the measurements we plan to do an experiment called Chemistry of the Atmosphere: Field Experiment in Brazil - (CAFE-Brazil), that is a partnership between Max Planck Institute for Chemistry (coordinator), Karlsruhe Institute of Technology, Research Center Jülich, University of Frankfurt, University of Mainz, Centre for Weather Forecasting and Climate studies (CPTEC-INPE) and the University of Sao Paulo. We are planning to bring to Brazil again the High Altitude Atmospheric Observatory (HALO) G5 airplane. To study particle and trace gas at 14 km altitude, it is necessary to use a platform that can reach such high

altitude, and we will use the HALO German plane. This mission has already been approved by DLR and HALO Science committee.

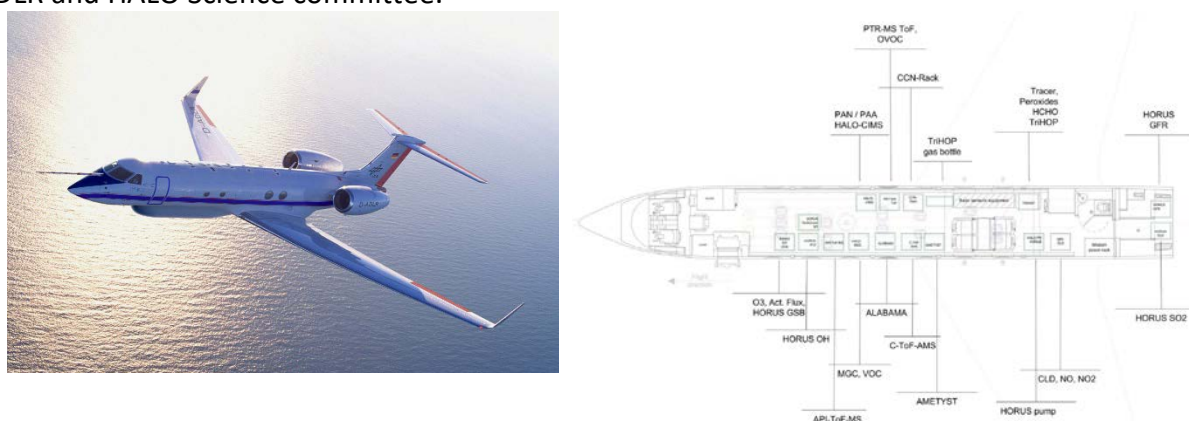


Figure 6.4.2 – The HALO plane and its configuration to be used in the CAFÉ-Brazil experiment.

Institute	PI	Instrument	Species/Parameters
DLR-FX		BAHAMAS	P, T, wind, humidity, position, altitude
DLR-FX		SHARC	H ₂ O mixing ratio (gas phase)
FZ-Jülich	B. Bohn	HALO-SR	Actinic Flux (photolysis rates)
KIT	A. Zahn	FAIRO	O ₃
MPIC	J. Crowley	HALO-CIMS	PAN/PAA, SO ₂ , ClNO ₂ , HCl
MPIC	J. Williams	HALO-MGC	NMVOC
MPIC	H. Fischer	TRIHOPE	H ₂ O ₂ , tot. peroxides, CO, HCHO, CH ₄ /CO ₂
MPIC	J. Williams	PTR-ToF-MS	OVOC
MPIC	H. Harder	HORUS-OH	OH, HO ₂ (RO ₂)
MPIC	H. Harder	HORUS-SO ₂	SO ₂
Univ. Mainz/MPIC	J. Schneider	AMETYST	Aerosol number and size distributions
Univ. Mainz/MPIC	J. Schneider	C-ToF-AMS	Aerosol composition (non-refractory)
Univ. Mainz/MPIC	S. Borrmann	ALABAMA	Single particle composition (incl. refractory)
Univ. Frankfurt	J. Curtius	CI-APi-ToF-MS	H ₂ SO ₄ (g), ions, clusters, HOM/ELVOC
MPIC	U. Pöschl	CCN-Rack, BC	CCN-Counters, SP2, aerosol impactor
MPIC	H. Fischer	CLD	NO, NO ₂

The PTR-ToF-MS coupled to the C-ToF-AMS and the ALABAMA instrument will allow single and bulk particle organic composition in addition to the precursor VOCs. The APi-ToF-MS will measure the ELVOCs (extremely low volatility organic compounds), which were recently identified to directly produce particles without sulfur or nitrogen compounds.

6.5 – Measurements of Amazonian aerosols in Chacaltaya

Chacaltaya is a unique site, very close to Amazonia (100 km), but at the Andes Mountains at 5,240 meters of altitude, where an international team with our Brazilian participation is planning a large experiment in 2018 to study the impact of Amazonian aerosols into the Andes. The station is located at the Bolivian Andes (16°21.014'S, 68°07.886'W, 5240 masl). The station is also a WMO Global Atmosphere Watch station under the code CHC/GAW. Figure 6.5.1 shows the location of the Chacaltaya station in the Andes and several air mass trajectories showing that some of the trajectories came directly from Amazonia that is less than 100 km from Chacaltaya. On the same picture on the right, it is possible to observe a photo of the sampling station, where aerosol and trace gases will be sampled in an international experiment in 2018. The opposite is also true; air mass from Bolivia penetrates the Amazon basin. Trajectories histories from several flights during Acridicon-CHUVA show air coming directly from the Andes toward the Amazon basin as shown in Figure 6.5.2 (Andreae et al. 2017).

Rose et al. (2015) measured and identified several events of new particle formation in Chacaltaya, and they are enhanced for air masses coming from Amazonia, for unknown reasons. Recent work from Pérez-Ramírez et al., 2017 analyzing AERONET data from Amazonia and the Andes shows clearly the impact of Amazonian aerosols in Chacaltaya. Episodic intrusions of air masses from Amazonia shows different single scattering albedo attributed to changes in water vapor.

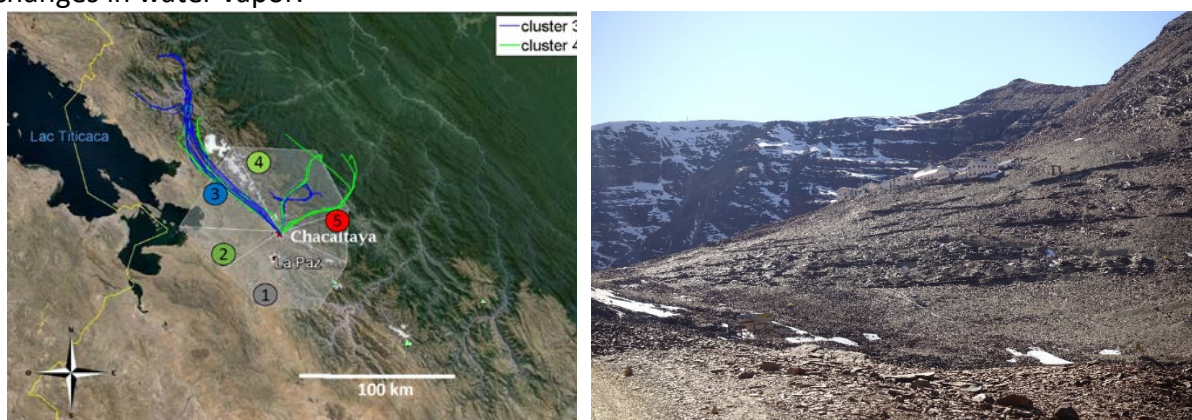


Figure 6.5.1 – Location of the Chacaltaya station in the Andes and several air mass trajectories showing that some of the trajectories came directly from Amazonia, that is less than 100 Km from Chacaltaya. Right: photo of the station, where aerosol and trace gases will be sampled in an international experiment.

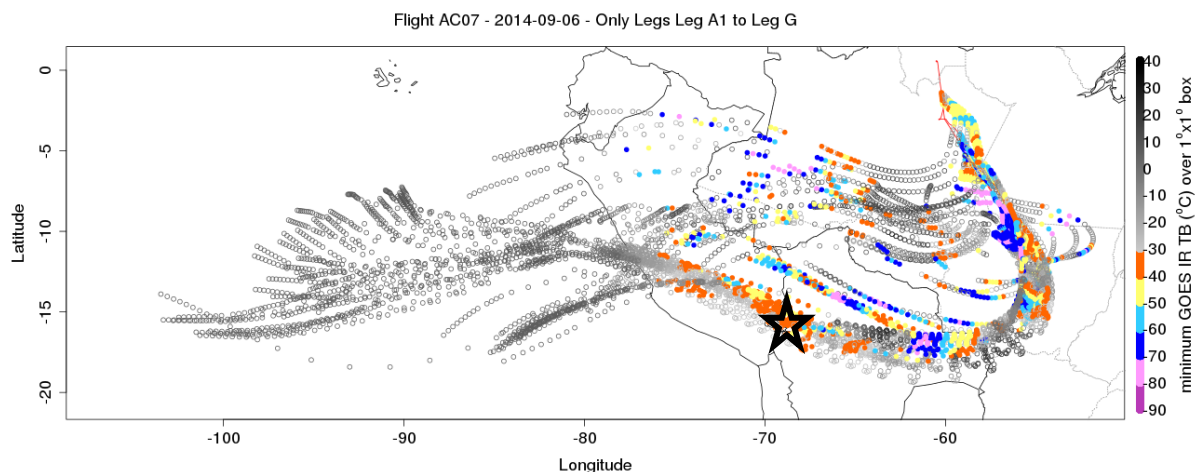


Figure 6.5.2 – Air mass backtrajectories (up to 5 days) from FLEXPART (“FLEXible PARTICle”) Lagrangian Particle Dispersion Model version 9.02 (Stohl et al., 2005) for several leg segments of flight AC07 during Acridicon-CHUVA. Air mass position is colored by the value of cloud-top temperatures from GOES-13 weather satellite images (Andreae et al., 2017). Location of the Chacaltaya station is highlighted by a star symbol in the figure.

Several previous studies shows that Chacaltaya received natural biogenic aerosols from Amazonia during the wet season (December to July), and biomass burning during the dry season (August-December). The picture in Figure 6.5.3 shows the Chacaltaya station under a severe plume of biomass burning aerosol going up to the Andes. The deposition of black carbon on the Andes snow could change significantly the surface albedo, increasing temperature and melt snow.

An international team will study the transport and aerosol properties at Chacaltaya Station in 2018. The team is led by the Laboratorio de Física de la Atmósfera, IIF-UMSA (Marcos Andrade) with the participation of the University of Helsinki (Markku Kulmala), University of Grenoble (Paolo Laj), University of Stockholm (Radovan Krejci), University of Sao Paulo (Paulo Artaxo), Leibniz Institute for Tropospheric Research (Alfred Wiedensohler) and others. The group will deploy a large set of instruments that are shown in Table 6.5.1. In particular, an APiTOF-AMS, a CIMS with Figaero inlet and an ACSM will study the composition of aerosol and trace gases with high time resolution. A PTR-MS-ToF will measure VOC concentrations and several instruments will investigate particle nucleation (NAIS, PSM, and SMPS). The Bolivian group lead by Prof. Marcos Andrade will coordinate the operation of this large set of measurements. The local Bolivian scientists will be Dr. Ricardo Forno and Dr. Luis Blacutt, among others. The team operates the station for more than 5 years already.

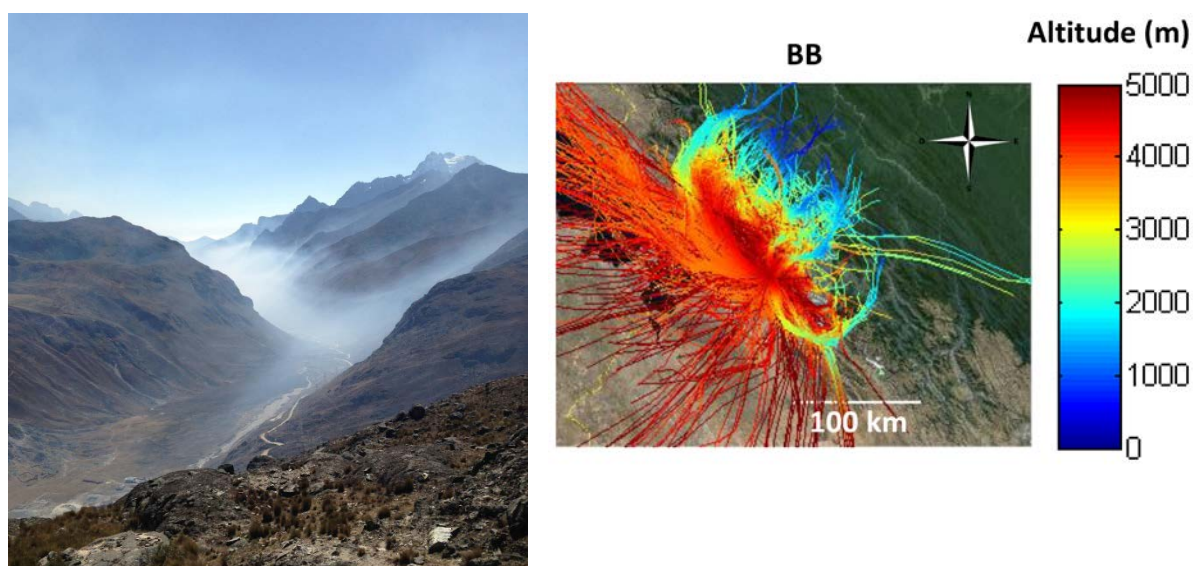


Figure 6.5.3 – Left: Chacaltaya station under a severe plume of biomass burning aerosol going up to the Andes and originated in Amazonia. Right: Air mass trajectories during the dry season, showing that part of the complex air mass trajectories are coming from Amazonia.

Table 6.5.1 - Instrumentation to be deployed at Chacaltaya 2018 experiment

Instrument	measured parameter	PI	Institute
Hi-Vol Sampler	Aerosol Chemistry	M. Andrade/J.L. Jaffrezo	LFA/CNRS-IRD
Meteo Station	RH,T,P, wind dir & speed	M. Andrade /P.Ginot	LFA/IRD
Horiba	CO	F. Velarde	LFA
Picarro	CO ₂ /CH ₄ /H ₂ O/CO	M. Andrade/M. Ramonet	LFA/CNRS-IRD
Picarro	CO ₂	M. Andrade/M. Ramonet	LFA/CNRS-IRD
Thermo	O ₃	M. Andrade/P. Cristofanelli	LFA/CNR
Aurora Neph	Aerosol scatt; coef	M. Andrade/J. Nicolas	LFA/CNRS-IRD
TSI 3 wav. neph	Aerosol scattering	Paulo Artaxo	Inst. Physics USP
MAAP	Aerosol abs. coef	M. Andrade/A. Wiedensohler	LFA/TROPOS
SMPS	Aerosol size distribution	M. Andrade/Wiedensohler/R. Krejci	LFA/TROPOS/SU
Total CN	Aerosol number	M. Andrade/Wiedensohler/R. Krejci	LFA/TROPOS/SU
Aethalometer AE31	Aerosol abs. coef	M. Andrade/J.Nicolas	LFA/CNRS-IRD
NAIS	Ultra-fine particles	M. Andrade/ K. Sellegri	LAF/CNRS-IRD
APiTOF AMS	Positive and negative ions	Federico Bianchi	UHEL
CI-APiTOF AMS	H ₂ SO ₄ , MSA, HOMs	Federico Bianchi	UHEL
PSM	ultra fine particles	Karine Sellegri	CNRS
SP2	BC size distribution	Patrick Ginot	CNRS-IRD
ACSM	organic/inorganic aerosols	Paulo Artaxo	Inst. Physics USP
FIGAERO - CIMS	VOCs	Claudia Mohr	Stockholm
PTR-ToF-MS	VOCs	Armin Hansel	Innsbruck

AERONET sun photometers in several locations in Amazonia (Porto Velho, Rio Branco and low lands Bolivia) as well as in Chacaltaya, La Paz and El Alto will measure aerosol optical properties and optical depth. High-resolution modeling with WRF-Chem will derive air mass trajectories from Central Amazonia to the Andes. MODIS remote sensing aerosol products as well as CALIPSO and MISR will help in the identification of transport episodes from Amazonia to Chacaltaya.

7 - Bridging Models and Observations

For a better understanding of cloud life cycle, it is critical that long-term measurements of cloud and related atmospheric properties be performed. Therefore, the CLOUDS AND MODELING component of this proposal will have an observation framework where continuous measurements capable of relating atmospheric thermodynamics, aerosols and cloud properties to the diurnal cycle of convection in Central Amazonia will be performed allowing for water vapor-cloud-convection-aerosol-climate feedbacks to be investigated. These measurements will be carried out at the ATTO tower.

At the same time, a myriad of physical, chemical and meteorological processes that range from the sub micrometer to the continental scale drives the clouds' life cycle making modeling of clouds in Amazonia very challenging. To address this issue models were developed to represent different scales, ranging from parcel to meso (or even global) scales. Therefore, the adequate representation of the entire life cycle of clouds demands several different models, and we propose to use parcel, LES and regional models. Regional models (BRAMS and WRF) with various land surface scenarios under different aerosol loads and large-scale dynamic lateral boundary conditions will also be used to shed light on the scientific questions raised in the previous sections. High-resolution simulations will also be used for forward and backward trajectories of air parcels to help the interpretation of data collected in the ATTO tower as well as in the boat and aircraft measurements component. While modeling will serve as research tool for the overall objectives of this proposal, it is expected that model improvements will also be obtained by the comparison/validation of model output to the special measurements proposed in this project. Tools for model-data comparison will be used for this purpose. Models like CR-SIM (Cloud resolving models radar simulator) developed by McGill is one of these several tools available to compare model to observation. In addition, it has been demonstrated that local vertical profiles of precipitation can be combined with scanning radars for convection area representation, and directly model comparison (Giangrande et al. 2013, 2016; Kumar et al. 2015).

7.1 - High Resolution Regional Scale Modeling

For regional scale modeling, the Brazilian developments on the Regional Atmospheric Modeling System (Freitas et al., 2009, 2017) will be used, as well as the WRF-Chem model. Among other important features, the BRAMS model is equipped with a simplified photochemical module (Freitas et al., 2005) which is intended to perform high-resolution simulations of air quality features, especially over urban areas and its vicinities, where vehicular and industrial emissions are the main source of pollution. The model also has an appropriate parameterization for urban areas. The Town Energy Budget (Masson, 2000), which was modified to be compatible with the SPM parameterization and be able to represent the vehicular behavior in large urban areas, such as São Paulo and Rio de Janeiro. Most of the modifications are described in Freitas et al., 2007. This is a very important feature, since the urban area of Manaus is surrounded by large water bodies (Negro and Amazonas rivers), being subjected to river breeze circulations. In addition, some evidences show that urban heat island effects in Manaus are very dependent on vehicular emissions of heat and moisture. Souza and Alvalá (2012) found that the strength of UHI are marked by two periods of the day, being one in the morning (around 8:00 Local time) and the other in the afternoon (around 16:00 Local time). These hours are coincident with rush hours in many large urban cities and the diurnal

cycle presented in the paper (Souza and Alvalá, 2012) fits very well with that characteristic. The same cycle is used for latent heat fluxes and for pollutant emissions in the SPM (CO, SO₂, PM_{2.5}, NO_x, and VOC).

Modeling of the local circulations in the area will be performed with BRAMS, with SPM and TEB activated, operating in high spatial resolution (≤ 500 m) to account for the steep gradients in a realistic way. BRAMS output will be used to calculate high resolution forward and backward trajectories to the ATTO tower, as well as in the Western Amazonia boat measurements. An operational version of BRAMS for the area is already running at the MASTER Lab at USP with 2 km resolution.

7.2 High resolution cloud dynamics, aerosol and microphysics interactions

As shown by Camponogara et al (2017) aerosols may impact cloud dynamics through complex cloud microphysics processes involving water and ice formation. Injection of aerosols in different levels in the atmosphere by biomass burning or by deep convection (Wang et al, 2017) is produced by atmospheric large scale circulations that will advect aerosol particles over very large distances (Freitas et al 2005). Upon entering a convective system, either through cloud base or in specific levels, aerosol may act as CCN and disturb cloud microphysical processes. Depending upon the large scale setting the effect of different aerosol features may be different as has been seen in several previous studies. Looking for patterns of response in specific situations in the Amazon will be the goal of a series of numerical simulations. Specifically, the efficiency of the collision-coalescence efficiency, and the efficiency of ice processes such as riming are fundamental issues that may be affected by the three dimensional nature of aerosol concentration in the cloud environment. The efficiency of these processes may actually affect the strength of cloud scale updrafts and downdrafts, thus affecting cloud life cycle and rainfall production.

Patterns of evolution given by models and observations will be addressed through the methodology proposed by Cecchini et al (2017). Microphysical processes are analyzed by considering the space and time evolution of droplet concentration in the lower part of the convective core and up into the transition into the mixed layer. Cecchini et al (2017) considered observations of cloud distributions in deep convection in the Amazon represented by Gamma functions. The associated parameters are used to define a phase space where processes like condensation and collision growth may be analyzed directly. Generalization of this approach will be used based on model parameters related to size distribution of the different hydrometeors and their evolution in specific phase spaces defining the life-cycles and microphysical processes under different aerosol scenarios. This novel approach will potentially highlight and summarize the different microphysical processes at play in a given convective core and may be associated with changes in cloud dynamics.

Simulation with different microphysical parameterization, as well with 3D turbulence and different cloud mixing length will also be evaluated. In cloud resolving models microphysics and turbulence parameterization play a very important role and models are very sensitivity. The combination of different parameterizations, the observed data from filed campaign and model simulators will provide a framework to study the main issues in models to reproduce convection in Amazonas. With, the best model adjustment and parameterization aerosol-cloud-rainfall interaction will be studied with more confidence and precision. Also these model-observation facilities will provide information to understand cloud life cycle,

cloud organization, the transition from shallow to deep convection, the effect of the surface on cloud formation from cloud to large scale organization and the complex interaction deforestation-aerosol-cloud-rainfall.

8 - Time schedule of the project

This project is scheduled to start in January-June 2018 and last for 5 years. The team already started preparation for this project, since we are deeply involved in the planning for the ATTO tower measurements, and other components. In 2018, we will start the ATTO data collection and analysis that is planned to continue for the whole duration of the project. In 2019, we plan to do the boat field campaign in the Western Amazonia, followed by the data analysis and modelling component. We will have a large aircraft campaign in 2020. In 2022 and 2023, we will focus on data analysis, modeling of the measurements and publications. Most of ATTO instrumentation was already ordered with funds from other projects. We just included here additional essential instrumentation that will be necessary to achieve our goals.

Project Activity	2018	2019	2020	2021	2022	2023
ATTO tower measurements	XXXXXX	XXXXXX	XXXXXX	XXXXXX	XXXXXX	XXXXXX
Analysis and consolidation of ATTO measurements		XXXXXX	XXXXXX	XXXXXX	XXXXXX	XXXXXX
Chacaltaya measurement campaign	XXXXXX	XXXXXX				
Development of the aerosol-cloud interaction modelling component.		XXXXXX	XXXXXX	XXXXXX		
Western Amazonia Boat Component		XXXXXX	XXXXXX			
ATTO and Boat modeling component data analysis			XXXXXX	XXXXXX	XXXXXX	XXXXXX
Cloud measurements integration with modeling			XXXXXX	XXXXXX	XXXXXX	
Western-Central Amazonia Boat Component				XXXXXX	XXXXXX	
CAFÉ-Brazil HALO Experiment			XXXXXX			
Data integration HALO and ATTO measurements				XXXXXX	XXXXXX	
Students thesis defense			XXXXXX	XXXXXX	XXXXXX	XXXXXX
Publications and scientific conference participations				XXXXXX	XXXXXX	XXXXXX

It is important to emphasize that two of the activities are heavily international in nature and depends on difficult logistical issues, some changes in this schedule can happen. This is particularly true for the HALO aircraft campaign. The campaign was already approved at the DLR and Max Planck level, there are financial resources to bring and operate the plane in Brazil, but technical challenges are large, and small displacements in this schedule can happen.

9 - Expected results and innovation

The large scope of the scientific agenda of this project will allow a better understanding of critical issues in tropical forests that has strong impacts far from the Amazonian region. The proposed studies of the effects of aerosols and clouds on the radiation balance can help to constrain this critically important area, since there it is difficult to obtain closure of the radiation balance at the ground in Amazonia. The photochemistry of tropical areas is another area that will also gain from the results of this project, since we will study the mechanisms of ozone production from NO_x and VOCs emissions from natural biogenic and biomass burning emissions. The joint use of LIDAR's, radiosondes, cloud radars and meteorological radars is a powerful tool in tropical area experiments, because it allows a detailed picture of the vertical profile of trace gases and aerosols. This will be combined with experiments studying the vertical distribution of trace gases and aerosols at the 325 meters ATTO tall tower. This powerful combination of measurements will add a critical knowledge on the vertical structure of the aerosol-clouds interactions, together with the thermodynamics and dynamical atmospheric conditions that are relevant for cloud modeling in tropical regions. We will integrate these detailed measurements with the modeling strategy in order to have a more comprehensive understanding of tropical forest ecosystems. The cloud and aerosol interactions will be integrated in regional models such as WRF-Chem and BRAMS, to increase our capability to predict precipitation in tropical areas.

10 – Role of each investigator

Paulo Artaxo – Overall coordination of the measurement component, responsible for the ATTO tower continuous measurements over the 5 years duration of the project, as well as to arrange logistics for the Boat experiment in Western Amazonia, the sampling campaign in Chacaltaya, and the HALO airplane campaign in 2020. Responsible for the AERONET operation and data analysis.

Maria Assunção Faus da Silva Dias – Overall coordination of the modeling activities, and development of the aerosol-cloud interaction component. Integration of the modelling component with measurements.

Luiz Augusto T. Machado – Responsible for the Cloud Radar observations, as well as measurements associated to aerosol-cloud interactions. Responsible to collect data from the SIPAM Precipitation radar that runs in Manaus. Also responsible for remote sensing of clouds and precipitation. Coordinator for the HALO 2020 aircraft campaign.

Rachel Albrecht – Cloud and Precipitation Radar observations at ATTO, and measurements associated with cloud-aerosol-precipitation interactions, as well as their impacts on deep convection and cloud electrification.

Henrique Melo Jorge Barbosa – Responsible for measurements with the Raman Lidar and CCN measurements, as well as operation and data analysis with the Polar Nephelometer to retrieve optical properties and size distribution from scattering measurements.

Luciana Varanda Rizzo – Responsible for the aerosol optical properties measurements and its coupling with size distribution and composition. Also responsible for the data compilation and data base maintenance.

Samara Carbone – Responsible for the operation and data analysis of the 4 Aerosol Mass Spectrometers that will be run in this project.

Edmilson Dias de Freitas – responsible for the BRAMS and WRF-Chem modeling simulations and integration with the measurements.

Niro Higuchi – responsible for the ATTO tower operation, and data integration with other groups at the INPA and UEA.

Meinrat O. Andreae – They coordinate the German component of the ATTO tower operation. Andi Andreae is also co-responsible for the HALO plane campaign.

Jos Lelieveld and Meinrat O. Andreae – They are the German coordinators of the CAFÉ experiment with the German HALO high altitude plane.

Hans Christen Hansson, Ilona Riipinen, Radovan Krejci – They will be partners in the Chacaltaya experiment as well as in the modeling of aerosol-cloud-VOCs interactions.

Scot T. Martin – Will collaborate in the VOCs and organic aerosols for the Western Amazonia boat experiment.

José Vanderlei Martins – Will collaborate in the aerosol optical properties, in special the measurement of refractive index and other inversion products using the Polar Imaging Nephelometer.

11 – Data management plan and data sharing

All data collected in this project will be freely distributed and available for the whole science community. As part of the LBA experiment, this project follows the open data policy of the LBA program for all the components (data from the ATTO tower, Chacaltaya, boat experiments and HALO aircraft. As an example, all data from the GoAmazon2014/15 FAPESP Thematic project is being shared in our FTP site: <http://lfa.if.usp.br/ftp/public/>. At the US Department of Energy DoE web site, our data as well as all the other large projects that are part of the GoAmazon2014/15 is being shared using a common and easy to use data sharing platform: <http://www.archive.arm.gov/discovery/>.

For this project, we plan to have a close partnership with Prof. Pedro Luiz Pizzigatti Corrêa, from POLI-USP, who is a specialist on data sharing. We plan to use tools developed by the group of Pedro Corrêa, based on DataONE initiative to facilitate collaboration on data analysis. DataONE focuses on both the research and education, with a cyberinfrastructure that supports sharing, conservation, preservation and open access to scientific data. DataONE's cyberinfrastructure promotes institutional cooperation with national and international coverage, multidisciplinary and interdisciplinary participation. We will also use the ARM Climate Research Facility from DoE. The ATTO and Chacaltaya stations are part of the WMO

Global Atmospheric Watch (GAW) program that also has its own data repository. Our previous data from FAPESP projects are stored at the GAW dataset, under the Manaus station name.

12 – Major existing equipment to be used in this project

In the last 25 years, several successful FAPESP thematic projects allowed the acquisition of several key equipment that were well maintained and are in operation to collect and analysis samples for this project. At the ATTO tower, the INPA-MPI collaboration already installed most of the instruments. At Chacaltaya, the existing collaborations between Bolivians, French, German and Swedish investigators already has in operation a large sweep of instruments that will be complemented with the proposal. At the University of São Paulo, some of the available equipment that will be used in this project includes:

- 1 – Four Aerodyne Aerosol Mass Spectrometers - Aerosol Chemical Speciation Monitors (ACSM), 3 of which uses a quadrupole and one a ToF detector. All four will be available for this project in all field measurements.
- 2 – Ionicon PTR-MS - Proton Transfer Reaction Mass Spectrometer, with a quadrupole for VOC measurements.
- 3 – Integrating Nephelometers - IFUSP has 3 Aurora and TSI 3 lambdas nephelometers
- 4 – Aethalometers – IFUSP has 2 Aethalometers but from an old version (AE10).
- 5 – SMPS – Scanning Mobility Particle Sizer – IFUSP has 3 TSI SMPS 3080.
- 6 – CCN Counter – Droplet Measurement Technology CCN 100 counter.
- 7 - Sunphotometers – IFUSP has 4 CIMEL sunphotometers, and NASA/AERONET provides another 5 instruments for a total AERONET network of 7 sites, with some spare instruments.
- 8 – X-Ray Fluorescence Analyzer – IFUSP has one PanAnalytical Epsilon 5 XRF analyzer
- 9 – Sunset EC/OC analyzer and associated laboratory.
- 10 – Raymetrics Raman Lidar with water vapor channel to measure vertical aerosol distribution and water vapor.
- 11 – Filter collection systems using Teflon, quartz or Nuclepore filters.
- 12 – 1290 MHz Radar Wind Profiler (RWP) – This radar was recently acquired by the ATTO project. It is a Doppler radar originally designed to measure the vertical distribution of winds in clear air (Bragg scattering) along the planetary boundary layer and lower troposphere, but it also measures precipitation if the radar resolution volume contains hydrometeors or other scatterers larger than 0.1 mm (Rayleigh scattering).

13 – Major equipment to be acquired in this project

In addition to the large set of existing instruments, as well as instruments that will be provided by our international partners such as CIMS, API-ToF-MS, Figaero-ToF-MS and others, we need a few extra equipment listed below.

1 – Vertically Pointing Cloud Radar (94 GHz or 35 GHz)

In other to measure vertical distribution of cloud properties, a cloud radar will be acquired. Cloud properties can be measured with millimeter radars such as 35 GHz (8.6 mm) (e.g., Williams et al. 2016) or 94 GHz (3.2 mm) (e.g., Delanoë et al., 2016) and combined with the precipitation measurements of the already available RWP we will be

able to study the transition from clouds to precipitation and their interactions with aerosols.

2 – Micro Pulse Polarized Lidar

This Lidar will measure the vertical profile of aerosol concentrations up to 15 Km, with a low altitude (0.3 Km) to start the measurements. It is an instrument with polarization and constructed in a way to be deployable in Amazonian conditions. The polarization helps separate different types of particles from the particle geometry and composition. The data will be instantly processed and available at the MPL Network site.

3 – Magee Scientific Aethalometer AE33

The Aethalometer AE33 instrument measures the aerosol absorption in 7 wavelengths, with a special algorithm to compensate for filter loading. It is a robust instrument. It allows to measure the Absorption Angstrom Exponent to help discriminate between brown and black carbon.

4 – High resolution Aerosol Mass Monitor

This instrument measure the total aerosol mass concentration with high time resolution and will complement measurements of SMPS, ACSM, and Aethalometer to obtain mass closure. The measure of total aerosol mass with high resolution must be done in parallel with the organic aerosol mass and size distribution to allow quantification of aerosol mass produced.

5 - Thermo Scientific Partisol 2025i-D

The Partisol allows the automatic collection of aerosol fine and coarse modes on Teflon filters and allow XRF trace element analysis. We can also use Quartz filters to determine EC/OC amounts using a Sunset OC/EC analyzer.

6 – Total Aerosol Carbon Analyzer

This new instrument from Magee Scientific allows the measurements of total aerosol carbon contents in real time with 30 minutes time resolution. It is essential to characterize the dynamics of secondary organic aerosol formation. The instrument is the first that is field deployable with no carrier gas needed, what is essential for operation in difficult Amazonian conditions.

7 – High Performance computational cluster

Equipment essential for the project modeling component. The air mass trajectory calculation system, the regional WRF-Chem modeling and the aerosol-cloud modeling component needs a dedicated workstation to the duration of the project. We plan to acquire a SGI workstation for this task.

14 – Results from previous FAPESP Thematic projects

Most of the basic team of this project have already worked together in several previous large FAPESP thematic projects. In addition, our international partners (Harvard, Stockholm, Max Planck etc.) has a long history of successful scientific collaboration along the last 25 years.

We had run several FAPESP Thematic projects and we have other projects close to the thematic of this proposal. They are:

- 1) Thematic project FAPESP 2013/05014-0 – GoAmazon - **Interactions between urban and forest emissions in Manaus, Amazonia: The Brazilian component of GoAmazon**. PI: Paulo Eduardo Artaxo Netto, Co-PI: Maria Assunção Faus da Silva Dias – IAG-USP. In the completing phase, run from Nov. 2013-Nov. 2017. The project studied the Manaus pollution plume and its interaction with the natural biogenic component. In the last 4 years, it produced about 120 papers, of which 7 of them in Nature and Nature Comm. More than 25 students have worked their PhD and MSc using data from the GoAmazon project.
- 2) Thematic Project FAPESP 2008/58100-2, Title: **AEROCLIMA - Direct and indirect effects of aerosols on climate in Amazonia and Pantanal**. Period from 01/06/2009 to 31/05/2013. We studied the large scale effects of aerosol on the Amazonian radiation budget in Amazonia and Pantanal. About 60 papers were published with the results from AEROCLIMA, several of them in Science and Nature.
- 3) Thematic Project FAPESP 1997/11358-9: **Interações físicas e químicas entre a biosfera e a atmosfera da Amazônia no experimento LBA**, PI: Paulo Artaxo, Co-PI: Maria Assunção F. Silva Dias, with a participation of about 18 collaborators, allowed to established the LBA scientific agenda in several measurements stations in Amazonia, including Santarem, Manaus, Porto Velho and Alta Floresta. Period from July 1998 to July 2002.
- 4) Thematic project FAPESP 1990/2950-2 - **Caracterização de gases e partículas de aerossóis da atmosfera Amazônica e seu relacionamento com processos de transporte e emissões em queimadas**. Period: August 1991 to August 1995. This Project studied the impact of biomass burning emissions on atmospheric chemistry in Amazonia. 2 papers on Science in 1992 and more than 20 publications. The PI was Artaxo.
- 5) LBA Millennium Institute - **Integração de abordagens do ambiente, uso da terra e dinâmica social na Amazônia: as relações homem-ambiente e o desafio da sustentabilidade – MilênioLBA2**. The team participated in the two MCT/CNPq Millennium Institutes from 2001 to 2008, with integrated studies in Amazonia. PI: P. Artaxo. Large interdisciplinary team integrated physical processes measurements with socio-economic issues in Amazonia. It was responsible for a large number of new PhD students in Amazonia (more than 30) and a large number of scientific papers, with more than 80 publications.

15 - References

- Adams, D. K., R. M. S. Fernandes, E. R. Kursinski, J. M. Maia, L. F. Sapucci, L. A. T. Machado, I. Vitorello, J. F. G. Monico, K. L. Holub, S. I. Gutman, N. Filizola and R. A. Bennett, 2011: A dense GNSS meteorological network for observing deep convection in the Amazon, *Atmos. Sci. Lett.*, 12, 2, pp. 207-212.
- Albrecht, Rachel I., Morales, Carlos A., Silva Dias, Maria A. F. 2011, Electrification of precipitating systems over the Amazon: Physical processes of thunderstorm development. *Journal of Geophysical Research.*, v.116, D08209.
- Albrecht, Rachel I., Morales, Carlos A., Silva Dias, Maria A. F. 2011, Electrification of precipitating systems over the Amazon: Physical processes of thunderstorm development. *Journal of Geophysical Research.*, v.116, p.D08209.
- Albrecht, Rachel I.; Morales, Carlos A.; Silva Dias, Maria A. F.. 2011. Electrification of precipitating systems over the Amazon: Physical processes of thunderstorm development. *Journal of Geophysical Research.*, v.116, D08209
- Allan, J. D., Morgan, W. T., Darbyshire, E., Flynn, M. J., Williams, P. I., Oram, D. E., Artaxo, P., Brito, J., Lee, J. D., and Coe, H.: Airborne observations of IEPOX-derived isoprene SOA in the Amazon during SAMBBA, *Atmos. Chem. Phys.*, 14, 11393–11407, doi:10.5194/acp-14-11393-2014, 2014.

- Andrade, M. et al. (2015) Puesta en marcha de una nueva estación de monitoreo climático en los andes centrales de Bolivia: la estación GAW/Chacaltaya. *Revista Boliviana de Física* vol.26 no.26 La Paz jun. 2015.
- Andreae et al., A. M.: The Amazon Tall Tower Observatory (ATTO): overview of pilot measurements on ecosystem ecology, meteorology, trace gases, and aerosols, *Atmos. Chem. Phys.*, 15, 10723–10776, doi:10.5194/acp-15-10723-2015, 2015.
- Andreae, M. and Rosenfeld, D.: Aerosol–cloud–precipitation interactions, Part 1. The nature and sources of cloud-active aerosols, *Earth-Sci. Rev.*, 89, 13–41, doi:10.1016/j.earscirev.2008.03.001, 2008.
- Andreae, M. O. Aerosols before pollution. *Science* 315, 50–51 (2007).
- Andreae, M. O., and Coauthors, 2017: Aerosol characteristics and particle production in the upper troposphere over the Amazon Basin. *Atmos. Chem. Phys. Discuss.*, 1–95, doi:10.5194/acp-2017-694.
- Andreae, M. O., Artaxo, P., Fischer, H., Freitas, S. R., Gregoire, J. M., Hansel, A., Hoor, P., Kormann, R., Krejci, R., Lange, L., Lelieveld, J., Lindinger, W., Longo, K., Peters, W., Reus, M. d., Scheeren, B., Silva Dias, M. A. F., Ström, J., van Velthoven, P. F. J., and Williams, J., Transport of biomass burning smoke to the upper troposphere by deep convection in the equatorial region: *Geophys. Res. Lett.*, 28, 951-954, 2001
- Andreae, M. O., Correlation between cloud condensation nuclei concentration and aerosol optical thickness in remote and polluted regions, *Atmos. Chem. Phys.*, 2009, 9, 543–556.
- Andreae, M. O., Rosenfeld, D., Artaxo, P., Costa, A. A., Frank, G. P., Longo, K. M., and Silva-Dias, M. A. F., Smoking rain clouds over the Amazon: *Science*, 303, 1337-1342, 2004
- Andreae, M. O.; Artaxo, P et al., “Biogeochemical cycling of carbon, water, energy, trace gases, and aerosols in Amazonia: The LBA-EUSTACH experiments,” *J. Geophys. Res.* 2002, 107, 8066.
- Andreae, M. O.; Gelencser, A. "Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols," *Atmos. Chem. Phys.* 2006, 6, 3131.
- Andreae, M. O.; Rosenfeld, D.; Artaxo, P.; Costa, A. A.; Frank, G. P.; Longo, K. M.; Silva-Dias, M. A. F., “Smoking rain clouds over the Amazon,” *Science* 2004, 303, 1337-1342.
- Artaxo, P., F. Gerab, M. A. Yamasoe, J. V. Martins, Fine Mode Aerosol Composition in Three Long Term Atmospheric Monitoring Sampling Stations in the Amazon Basin. *J. Geophys. Res.*, 99, D11, Pg. 22.857-22.868, 1994.
- Artaxo, P., F. Gerab, M.L.C. Rabello, Elemental composition of aerosol particles from two background monitoring stations in the Amazon Basin, *Nuclear Instruments and Methods in Physics Research*, B75, 277-281, 1993.
- Artaxo, P., H-C Hansson, Size distribution of biogenic aerosol particles from the Amazon basin. *Atmospheric Environment*, 29, 3, 393-402, 1995.
- Artaxo, P., J. V. Martins, M. A. Yamasoe, A. S. Procópio, T. M. Pauliquevis, M. O. Andreae, P. Guyon, L. V. Gatti, A. M. C. Leal. Physical and chemical properties of aerosols in the wet and dry season in Rondônia, Amazonia. *Journal of Geophysical Research*, Vol. 107, No. D20, 8081 - 8095, 2002.
- Artaxo, P., L. V. Rizzo, M. Paixao, S. de Lucca, P. H. Oliveira, L. L. Lara, K. T. Wiedemann, M. O. Andreae, B. Holben, J. Schafer, A. L. Correia, and T. M. Pauliquevis. Aerosol particles in Amazonia: Their composition, role in the radiation balance, cloud formation and nutrient cycles. In: *Amazonia and Global Change*, Ed. M. Keller, M. Bustamante, J. Gash, P. S. Dias. AGU Geophysical Monograph 186, 235-254, 2009.
- Artaxo, P., Martins, J. V., Yamasoe, M. A., Procópio, A. S., Pauliquevis, T. M., Andreae, M. O., 1397 Guyon, P., Gatti, L. V., and Leal, A. M. C., Physical and chemical properties of aerosols in the wet and dry season in Rondonia, Amazonia: *J. Geophys. Res.*, 107, 8081, doi:10.1029/2001JD000666, 2002. 1400
- Artaxo, P., Rizzo, L. V., Brito, J. F., Barbosa, H. M. J., Arana, A., Sena, E. T., Cirino, G. G., Bastos, W., Martin, S. T., and Andreae, M. O.: Atmospheric aerosols in Amazonia and land use change: from natural biogenic to biomass burning conditions, *Faraday Discuss.*, 165, 203–235, doi:10.1039/c3fd00052d, 2013.
- Artaxo, P.; Maenhaut, W.; Storms, H.; Van Grieken, R.; "Aerosol characteristics and sources for the Amazon basin during the wet season *J. Geophys. Res.*, 95, D10, 16971-16985, 1990.
- Artaxo, P.; Martins, J. V.; Yamasoe, M. A.; Procopio, A. S.; Pauliquevis, T. M.; Andreae, M. O.; Guyon, P.; Gatti, L. V.; Leal, A. M. C., “Physical and chemical properties of aerosols in the wet and dry seasons in Rondonia, Amazonia,” *J. Geophys. Res.*, 2002, 107, 8081.
- Barbosa, H. M. J., Barja, B., Pauliquevis, T., Gouveia, D. A., Artaxo, P., Cirino, G. G., Santos, R. M. N., and Oliveira, A. B.: A permanent Raman lidar station in the Amazon: description, characterization, and first results, *Atmos. Meas. Tech.*, 7, 1745–1762, doi:10.5194/amt-7-1745-2014, 2014.
- Ben-Ami, Y., Koren, I., Rudich, Y., Artaxo, P., Martin, S. T., and Andreae, M. O.: Transport of North African dust from the Bodélé depression to the Amazon Basin: a case study, *Atmos. Chem. Phys.*, 10, 7533–7544, doi:10.5194/acp-10-7533-2010, 2010.
- Betts, A. K, Silva Dias, M. A. F 2010 Progress in Understanding Land-Surface-Atmosphere Coupling from LBA Research. *Journal of Advances in Modeling Earth Systems.* , v.2, p.1 - 20.
- Betts, A. K., and C. Jakob: Study of diurnal cycle of convective precipitation over Amazonia using a single column model, *J. Geophys. Res.*, 107, 4732, 2002a
- Betts, A.; Jakobs, C. Evaluation of the diurnal cycle of precipitation, surface thermodynamics, and surface fluxes in the ECMWF model using LBA data. *J. Geophys. Res.*, 107(D20), 2002b
- Bianchi, F., et al., New particle formation in the free troposphere: A question of chemistry and timing: *Science*, 352, 1109-1112, doi:10.1126/science.aad5456, 2016.
- Bony, S., Stevens, B., Frierson, D. M. W., Jakob, C., Kageyama, M., Pincus, R., Shepherd, T. G., Sherwood, S. C., Siebesma, A. P., Sobel, A. H., Watanabe, M., and Webb, M. J.: Clouds, circulation and climate sensitivity, *Nat. Geosci.*, 8, 261–268, doi:10.1038/ngeo2398, 2015.
- Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V. M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S. K., Sherwood, S., B., S., and Zhang, X. Y.: *Clouds and Aerosols*, Cambridge, United Kingdom and New York, NY, US, 571–658, 2013.
- Braga, R. C., Rosenfeld, D., Weigel, R., Jurkat, T., Andreae, M. O., Wendisch, M., Pöhlker, M. L., Klimach, T., Pöschl, U., Pöhlker, C., Voigt, C., Mahnke, C., Borrmann, S., Albrecht, R. I., Mollerker, S., Vila, D. A., Machado, L. A. T., and Artaxo, P.: Comparing parameterized versus measured microphysical properties of tropical convective cloud bases during the ACRIDICON–CHUVA campaign, *Atmos. Chem. Phys.*, 17, 7365-7386, <https://doi.org/10.5194/acp-17-7365-2017>, 2017.
- Brito, J., Rizzo, L. V., Morgan, W. T., Coe, H., Johnson, B., Haywood, J., Longo, K., Freitas, S., Andreae, M. O., and Artaxo, P.: Ground-based aerosol characterization during the South American

- Biomass Burning Analysis (SAMBBA) field experiment, *Atmos. Chem. Phys.*, 14, 12069–12083, doi:10.5194/acp-14-12069-2014, 2014.
- CAMPONOGARA, GLÁUBER; SILVA DIAS, MARIA ASSUNÇÃO FAUS; CARRIÓ, GUSTAVO G.. 2017. Biomass burning CCNs enhance the dynamics of a Mesoscale Convective System over the La Plata Basin: a numerical approach In *ATMOSPHERIC CHEMISTRY AND PHYSICS DISCUSSION (ONLINE)*. , v.1, 1-29
- Carlsaw, K. S. *et al.* Large contribution of natural aerosols to uncertainty in indirect forcing. *Nature* **503**, 67–71 (2013).
- Carlsaw, K. S., Gordon, H., Hamilton, D. S., Johnson, J. S., Regayre, L. A., Yoshioka, M., and Pringle, K. J., Aerosols in the pre-industrial atmosphere: Current Climate Change Reports, 3, 1-15, doi:10.1007/s40641-017-0061-2, 2017.
- CARVALHO, L. M. V.; JONES, C.; SILVA DIAS, M.A.F.. 2002. Intraseasonal large-scale circulations and mesoscale convective activity in tropical South América during the TRMM-LBA campaign. In *Journal of Geophysical Research*. , v.107, 9.1-9.20
- Cecchini, M. A., Machado, L. A. T., Wendisch, M., Costa, A., Krämer, M., Andreae, M. O., Afchine, A., Albrecht, R. I., Artaxo, P., Borrmann, S., Fütterer, D., Jäkel, E., Klimach, T., Mahnke, C., Martin, S., Minikin, A., Moller, S., Pardo, L. H., Pöhlker, C., Pöhlker, M., Pöschl, U., Rosenfeld, D., and Weinzierl, B.: Illustration of microphysical processes in Amazonian deep convective clouds in the Gamma phase space: Introduction and potential applications *Atmos. Chem. Phys.*, 2017.
- CECCHINI, MICAEL A.; MACHADO, LUIZ A. T. ; COMSTOCK, JENNIFER M. ; MEI, FAN ; WANG, JIAN ; FAN, JIWEN ; TOMLINSON, JASON M. ; SCHMID, BEAT ; ALBRECHT, RACHEL ; MARTIN, SCOT T. ; ARTAXO, PAULO . Impacts of the Manaus pollution plume on the microphysical properties of Amazonian warm-phase clouds in the wet season. *Atmospheric Chemistry and Physics (Online)*, v. 16, p. 7029-7041, 2016.
- Cesana, G., and H. Chepfer (2012), How well do climate models simulate cloud vertical structure? A comparison between CALIPSO-GOCCP satellite observations and CMIP5 models, *Geophys. Res. Lett.*, 39, L20803.
- Chen, Q., Farmer, D. K., Rizzo, L. V., Pauliquevis, T., Kuwata, M., Karl, T. G., Guenther, A., Allan, J. D., Coe, H., Andreae, M. O., Pöschl, U., Jimenez, J. L., Artaxo, P., and Martin, S. T.: Submicron particle mass concentrations and sources in the Amazonian wet season (AMAZE-08), *Atmos. Chem. Phys.*, 15, 3687–3701, doi:10.5194/acp-15-3687-2015, 2015.
- Chen, Q.; Farmer, D. K.; Schneider, J.; Zorn, S. R.; Heald, C. L.; Karl, T. G.; Guenther, A.; Allan, J. D.; Robinson, N.; Coe, H.; Kimmel, J. R.; Pauliquevis, T.; Borrmann, S.; Pöschl, U.; Andreae, M. O.; Artaxo, P.; Jimenez, J. L.; Martin, S. T. "Mass spectral characterization of submicron biogenic organic particles in the Amazon Basin," *Geophys. Res. Lett.*, 2009, 36, L20806.
- Cirino, G. G., R. A. F. Souza, D. K. Adams, and P. Artaxo. The effect of atmospheric aerosol particles and clouds on net ecosystem exchange in the Amazon. *Atmos. Chem. Phys.*, 14, 6523 – 6543, doi:10.5194/acp-14-6523-2014, 2014.
- Claeys, M.; Graham, B.; Vas, G.; Wang, W.; Vermeylen, R.; Pashynska, V.; Cafmeyer, J.; Guyon, P.; Andreae, M. O.; Artaxo, P.; Maenhaut, W., "Formation of secondary organic aerosols through photooxidation of isoprene," *Science* 2004, 303, 1173-1176.
- Clarke, A. D. *et al.* Nucleation in the equatorial free troposphere: favorable environments during PEM-Tropics. *J. Geophys. Res.* **104**, 5735–5744 (1999).
- Clarke, A. D. *et al.* Particle production in the remote marine atmosphere: cloud outflow and subsidence during ACE 1. *J. Geophys. Res.* **103**, 16397–16409 (1998).
- Crooks, M., Paul Connolly¹, and Gordon McFiggans¹. A parameterisation for the co-condensation of semi-volatile organics into multiple aerosol particle modes. *Geosci. Model Dev. Discuss.*, <https://doi.org/10.5194/gmd-2017-123>.
- Davidson, E. A., de Araujo, A. C., Artaxo, P., Balch, J. K., Brown, I. F., Bustamante, M. M. C., Coe, M. T., DeFries, R. S., Keller, M., Longo, M., Munger, J. W., Schroeder, W., Soares-Filho, B. S., Souza Jr., C. M., and Wofsy, S. C.: The Amazon basin in transition, *Nature*, 481, 321–328, doi:10.1038/nature10717, 2012.
- Delanoë, J., and co-authors, BASTA: A 95-GHz FMCW Doppler Radar for Cloud and Fog Studies. *J. Atmos. Ocean. Technol.*, 33, 1023–1038, doi:10.1175/JTECH-D-15-0104.1, 2016.
- DOS SANTOS, MERCEL J.; Silva Dias, Maria A. F.; Freitas, Edmilson D.. 2014. Influence of Local Circulations on Wind, Moisture and Precipitation Close to Manaus City, Amazon Region - Brazil In *Journal of Geophysical Research: Atmospheres*. , v.119
- Doughty, C. E.; Flanner, M. G.; Goulden, M. L. "Effect of smoke on subcanopy shaded light, canopy temperature, and carbon dioxide uptake in an Amazon rainforest," *Global Biogeochemical Cycles*, 2010, 24.
- Draxler, R. and Hess, G.: An Overview of the HYSPLIT_4 Modelling System for Trajectories, Dispersion, and Deposition, *Aust. Meteorol. Mag.*, 47, 295–308, 1998.
- Dunne, E. M., *et al.*, Global atmospheric particle formation from CERN CLOUD measurements: *Science*, 354, 1119-1124, doi:10.1126/science.aaf2649, 2016.
- Durieux, L.; Machado, L. A. T.; Laurent, H., "The impact of deforestation on cloud cover over the Amazon arc of deforestation," *Remote Sensing of Environment* 2003, 86, 132-140.
- Ehn, M., *et al.*, A large source of low-volatility secondary organic aerosol. *Nature*, 506, 476-479, doi:10.1038/nature13032, 2014.
- Ekman, A. M. L., Krejci, R., Engström, A., Ström, J., de Reus, M., Williams, J., and Andreae, M. O., Do organics contribute to small particle formation in the Amazonian upper troposphere? *Geophys. Res. Lett.*, 35, L17810, doi:10.1029/2008GL034970, 2008.
- Fan, J. W.; Zhang, R. Y.; Tao, W. K.; Mohr, K. I., "Effects of aerosol optical properties on deep convective clouds and radiative forcing," *J. Geophys. Res.* 2008, 113, D08209.
- Farmer, D. K.; Chen, Q.; Kimmel, J. R.; Docherty, K. S.; Nemitz, E.; Artaxo, P.; Cappa, C. D.; Martin, S. T.; Jimenez, J. L. "Chemically-resolved particle fluxes over tropical and temperate forests," 2013, in press.
- Feingold, G, WL Eberhard, DE Veron, and M Previdi. 2003. "First measurements of the Twomey indirect effect using ground-based remote sensors." *Geophysical Research Letters*, 30, 1287, doi:10.1029/2002GL016633.
- Feingold, G., and H. Siebert, Cloud-aerosol interactions from the micro to the cloud scale. Chapter in Strungmann Forum report, vol. 2. Heintzenberg, J., and R. J. Charlson, eds. 2009. *Clouds in the Perturbed Climate System: Their Relationship to Energy Balance, Atmospheric Dynamics, and Precipitation*. Cambridge, MA: The MIT press
- Feingold, G., S. Tzivion and Z. Levin, 1988: The evolution of raindrop spectra. Part I: stochastic collection and breakup. *J. Atmos. Sci.*, 45, 3387 – 3399.

- Feingold, G.; Jiang, H. L.; Harrington, J. Y., "On smoke suppression of clouds in Amazonia," *Geophys. Res. Lett.* 2005, 32, L02804.
- Fitzjarrald, D. R., R. K. Sakai, O. L. L. Moraes, R. Cosme de Oliveira, O. C. Acevedo, M. J. Czikowsky, and T. Beldini (2008), Spatial and temporal rainfall variability near the Amazon-Tapajós confluence, *J. Geophys. Res.*, 113, G00B11, doi:10.1029/2007JG000596.
- Forster, P., Ramaswamy, V., Artaxo, P., Bernsten, T., Betts, R., Fahey, D. W., Haywood, J., Lean, J., Lowe, D. C., Myhre, G., Nganga, J., Prinn, R. G., Schulz, M., Van Dorland, R., and Van Dorland, R.: Changes in Atmospheric Constituents and in Radiative Forcing Chapter 2., Cambridge University Press, 2007.
- FREITAS, E. D. 2008. Modelagem numérica da atmosfera em regiões urbanas: Aplicações em estudos e prognósticos sobre a qualidade do ar. *Boletim da Sociedade Brasileira de Meteorologia*, 32, 19-27.
- Freitas, E. D., C. M. Rozoff, W. R. Cotton, P. L. Silva Dias 2007, Interactions of an urban heat island and sea-breeze circulations during winter over the metropolitan area of São Paulo, Brazil. *Boundary-Layer Meteorology*, 122, 1, 43-65.
- Freitas, S. R., Longo, K. M., Silva Dias, M. A. F., Chatfield, R., Silva Dias, P., Artaxo, P., Andreae, M. O., Grell, G., Rodrigues, L. F., Fazenda, A., and Panetta, J.: The Coupled Aerosol and Tracer Transport model to the Brazilian developments on the Regional Atmospheric Modeling System (CATT-BRAMS) – Part 1: Model description and evaluation, *Atmos. Chem. Phys.*, 9, 2843–2861, doi:10.5194/acp-9-2843-2009, 2009.
- FREITAS, SAULO R.; et al., 2017. The Brazilian developments on the Regional Atmospheric Modeling System (BRAMS 5.2): an integrated environmental model tuned for tropical areas In *Geoscientific Model Development*, v.10, 189-222. 2017.
- FREITAS, Saulo Ribeiro de; LONGO, Karla Maria; SILVA DIAS, M.A.F.; DIAS, Pedro Leite da Silva; CHATFIELD, Robert; PRINS, Elaine; ARTAXO, Paulo; GRELL, George; RECUERO, F.. 2005. Monitoring the Transport of Biomass Burning Emissions in South America. In *Environmental Fluid Mechanics (London)*, v.5, 135-167
- Freitas, E. D., L. D. Martins, P. L. Silva Dias, M. F. Andrade 2005 A simple photochemical module implemented in RAMS for tropospheric ozone concentration forecast in the metropolitan area of São Paulo, Brazil: Coupling and validation. *Atmospheric Environment*, 39, 6352-6361.
- Freud, E., Rosenfeld, D., Andreae, M. O., Costa, A. A., and Artaxo, P.: Robust relations between CCN and the vertical evolution of cloud drop size distribution in deep convective clouds, *Atmos. Chem. Phys.*, 8, 1661–1675, doi:10.5194/acp-8-1661-2008, 2008.
- Giangrande, S. E., and co-authors, Convective cloud vertical velocity and mass-flux characteristics from radar wind profiler observations during GoAmazon2014/5. *J. Geophys. Res. Atmos.*, 121, 12, 891-12,913, doi:10.1002/2016JD025303, 2016.
- Giangrande, S. E., Feng, Z., Jensen, M. P., Comstock, J., Johnson, K. L., Toto, T., Wang, M., Burleyson, C., Mei, F., Machado, L. A. T., Manzi, A., Xie, S., Tang, S., Silva Dias, M. A. F., de Souza, R. A. F., Schumacher, C., and Martin, S. T., Cloud Characteristics, Thermodynamic Controls and Radiative Impacts During the Observations and Modeling of the Green Ocean Amazon (GoAmazon2014/5) Experiment: *Atmos. Chem. Phys. Discuss.*, 2017, 1-41, doi:10.5194/acp-2017-452, 2017.
- Giangrande, S. E., S. Collis, J. Straka, A. Protat, C. Williams, and S. Krueger, 2013: A Summary of Convective-Core Vertical Velocity Properties Using ARM UHF Wind Profilers in Oklahoma. *J. Appl. Meteorol. Climatol.*, 52, 2278–2295, doi:10.1175/JAMC-D-12-0185.1.
- Gilardoni, S., Vignati, E., Marmer, E., Cavelli, F., Belis, C., Gianelle, V., Loureiro, A., Artaxo, P., Sources of carbonaceous aerosol in the Amazon basin. *Atmospheric Chemistry and Physics*, 11, 2747-2764, 2011.
- Gloor, M., Barichivich, J., Ziv, G., Brienen, R., Schongart, J., Peylin, P., Cintra, B. B. L., Feldpausch, T., Phillips, O., and Baker, J.: Recent Amazon climate as background for possible ongoing and future changes of Amazon humid forests, *Global Biogeochem. Cy.*, 29, 1384–1399, doi:10.1002/2014gb005080, 2015.
- Gonçalves, W.A., L. A. T. Machado, and P.-E. Kirstetter. Influence of biomass aerosol on precipitation over the Central Amazon: an observational study. *Atmos. Chem. Phys.*, 15, 6789-6800, 2015, https://doi.org/10.5194/acp-15-6789-2015.
- Gordon, H., et al., Reduced anthropogenic aerosol radiative forcing caused by biogenic new particle formation: *Proc. Natl. Acad. Sci.*, 113, 12, 053-12,058, doi:10.1073/pnas.1602360113, 2016.
- Graham, B. P. Guyon, P. E. Taylor, P. Artaxo, W. Maenhaut, M. M. Glosky, R. C. Flagan, M. O. Andreae, Organic compounds present in the natural Amazonian aerosol: Characterization by gas chromatography–mass spectrometry. *J. Geophys. Res.*, 108, No. D24, 4766. doi: 10.1029/2003JD003990, 2003.
- Graham, B.; Guyon, P.; Maenhaut, W.; Taylor, P. E.; Ebert, M.; Matthias-Maser, S.; Mayol-Bracero, O. L.; Godoi, R. H. M.; Artaxo, P.; Meixner, F. X.; Moura, M. A. L.; Rocha, C.; Van Grieken, R.; Glosky, M. M.; Flagan, R. C.; Andreae, M. O., "Composition and diurnal variability of the natural Amazonian aerosol," *J. Geophys. Res.* 2003, 108.
- Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions, *Geosci. Model Dev.*, 5, 1471–1492, doi:10.5194/gmd-5-1471-2012, 2012.
- Gunthe, S. S.; King, S. M.; Rose, D.; Chen, Q.; Roldin, P.; Farmer, D. K.; Jimenez, J. L.; Artaxo, P.; Andreae, M. O.; Martin, S. T.; Poschl, U. "Cloud condensation nuclei in pristine tropical rainforest air of Amazonia: size-resolved measurements and modeling of atmospheric aerosol composition and CCN activity," *Atm. Chem. Phys.*, 2009, 9, 7551.
- Guyon, P., B. Graham, J. Beck, O. Boucher, E. Gerasopoulos, O. L. Mayol-Bracero, G. C. Roberts, P. Artaxo, and M. O. Andreae, Physical properties and concentration of aerosol particles over the Amazon tropical forest during background and biomass burning conditions. *Atm. Chem. Phys.*, 3, 951 – 967, 2003.
- Hallquist, M.; et al., "The formation, properties and impact of secondary organic aerosol: current and emerging issues," *Atm. Chem. Phys.*, 2009, 9, 5155.
- Heald, C. L., J. H. Kroll, J. L. Jimenez, K. S. Docherty, P. F. DeCarlo, A. C. Aiken, Q. Chen, S.T. Martin, D. K. Farmer, P. Artaxo, A. J. Weinheimer. A simplified description of organic aerosol elemental composition and implications for atmospheric aging. *Geophysical Research Letters*, Vol. 37, Article number L08803, 5 PP., 2010.
- Heald, C. L.; Henze, D. K.; Horowitz, L. W.; Feddes, J.; Lamarque, J.-F.; Guenther, A.; Hess, P. G.; Vitt, F.; Seinfeld, J. H.; Goldstein, A. H.; Fung, I., "Predicted change in global secondary organic aerosol concentrations in response to future climate, emissions, and land use change," *J. Geophys. Res.* 2008, 113, D05211.
- Houze 1993: *Cloud Dynamics*. International Geophysics Series, 53, Academic Press.
- Hu, W. W., et al., Characterization of a real-time tracer for isoprene

- 1630 epoxydiols-derived secondary organic aerosol (IEPOX-SOA) from aerosol mass spectrometer measurements: *Atmos. Chem. Phys.*, 15, 11807-11833, doi:10.5194/acp-1632-15-11807-2015, 2015.
- Huete, A. R.; Didan, K.; Shimabukuro, Y. E.; Ratana, P.; Saleska, S. R.; Hutyrta, L. R.; Yang, W. Z.; Nemani, R. R.; Myneni, R., "Amazon rainforests green-up with sunlight in dry season," *Geophys. Res. Lett.* 2006, 33, L06405.
- Huffman, J. A., Sinha, B., Garland, R. M., Snee-Pollmann, A., Gunthe, S. S., Artaxo, P., Martin, S. T., Andreae, M. O., and Pöschl, U.: Size distributions and temporal variations of biological aerosol particles in the Amazon rainforest characterized by microscopy and real-time UV-APS fluorescence techniques during AMAZE-08, *Atmos. Chem. Phys.*, 12, 11997-12019, doi:10.5194/acp-12-11997-2012, 2012.
- IPCC, 2013 – Intergovernmental Panel Climate Change – Working Group I: The Physical Science Basis of Climate Change. <http://ipcc-wg1.ucar.edu/wg1/wg1-report.html>
- Jardine, K. J., Monson, R. K., Abrell, L., Saleska, S. R., Arneth, A., Jardine, A., Ishida, F. Y., Serrano, A. M. Y., Artaxo, P., Karl, T., Fares, S., Goldstein, A., Loreto, F., and Huxman, T.: Within-plant isoprene oxidation confirmed by direct emissions of oxidation products methyl vinyl etone and methacrolein, *Glob. Change Biol.*, 18, 973-984, doi:10.1111/j.1365-2486.2011.02610.x, 2012.
- Jiang, J. H. et al., Clean and polluted clouds: Relationships among pollution, ice clouds, and precipitation in South America. *Geophys. Res. Lett.* 35, L14804 (2008).
- Jimenez, J. L et al., Evolution of Organic Aerosols in the Atmosphere, *Science*, 2009, 326, 1525-1529.
- Kanakidou, M.; Tsigaridis, K.; Dentener, F. J.; Crutzen, P. J., "Human-activity-enhanced formation of organic aerosols by biogenic hydrocarbon oxidation," *J. Geophys. Res.* 2000, 105, 9243-9254.
- Karl, T.; Guenther, A.; Turnipseed, A.; Tyndall, G.; Artaxo, P.; Martin, S. "Rapid formation of isoprene photo-oxidation products observed in Amazonia " *Atmospheric Chemistry and Physics*, 2009, 9, 7753.
- Keller, M., Bustamante, M., Gash, J., and Dias, P.: Amazonia and Global Change, Vol. 186, American Geophysical Union, Wiley, Washington, DC, 2009.
- Kesselmeier, J.; Guenther, A.; Hoffmann, T.; Warnke, J. Natural volatile organic compound (VOC) emissions from plants and their roles in oxidant balance and particle formation. In Amazonia and Global Change; Keller, M., Gash, J., Silva Dias, P., Eds., 2009.
- Khain, AP. 2009. "Notes on state-of-the-art investigations of aerosol effects on precipitation: a critical review." *Environmental Research Letters*, 4, 015004, doi:10.1088/1748-9326/4/1/015004.
- Koch, D.; et al., "Evaluation of black carbon estimations in global aerosol models," *Atmos. Chem. Phys.* 2009, 9, 9001-9026.
- Koren, I., Altartatz, O., Remer, L. A., Feingold, G., Martins, J. V., and Heiblum, R. H.: Aerosol-induced intensification of rain from the tropics to the mid-latitudes, *Nat. Geosci.*, 5, 118-122, 2012.
- Koren, I., Dagan, G., and Altartatz, O.: From aerosol-limited to invigoration of warm convective clouds, *Science*, 344, 1143-1146, 2014.
- Koren, I., Kaufman, Y. J., Remer, L. A., and Martins, J. V.: Measurement of the effect of Amazon smoke on inhibition of cloud formation, *Science*, 303, 1342-1345, doi:10.1126/science.1089424, 2004.
- Koren, I.; Martins, J. V.; Remer, L. A.; Afargan, H., "Smoke invigoration versus inhibition of clouds over the Amazon," *Science* 2008, 321, 946-949.
- Krejci, R. et al. Evolution of aerosol properties over the rain forest in Surinam, South America, observed from aircraft during the LBA-CLAIRE 98 experiment. *J. Geophys. Res.* 108, 4561 (2003).
- Krejci, R. et al. Spatial and temporal distribution of atmospheric aerosols in the lowermost troposphere over the Amazonian tropical rainforest. *Atmos. Chem. Phys.* 5, 1527-1543 (2005).
- Kuhn, U.; Andreae, M. O.; Ammann, C.; Araujo, A. C.; Brancaleoni, E.; Ciccioli, P.; Dindorf, T.; Frattoni, M.; Gatti, L. V.; Ganzeveld, L.; Kruijft, B.; Lelieveld, J.; Lloyd, J.; Meixner, F. X.; Nobre, A. D.; Pöschl, U.; Spirig, C.; Stefani, P.; Thielmann, A.; Valentini, R.; Kesselmeier, J., "Isoprene and monoterpene fluxes from Central Amazonian rainforest inferred from tower-based and airborne measurements, and implications on the atmospheric chemistry and the local carbon budget," *Atmos. Chem. Phys.* 2007, 7, 2855-2879.
- Kuhn, U.; Ganzeveld, L.; Thielmann, A.; Dindorf, T.; Schebeske, G.; Welling, M.; Sciare, J.; Roberts, G.; Meixner, F. X.; Kesselmeier, J.; Lelieveld, J.; Kolle, O.; Ciccioli, P.; Lloyd, J.; Trentmann, J.; Artaxo, P.; Andreae, M. O. "Impact of Manaus City on the Amazon Green Ocean atmosphere: ozone production, precursor sensitivity and aerosol load," *Atm. Chem. Phys.*, 2010, 10, 9251.
- Kulmala, M. et al. Deep convective clouds as aerosol production engines: role of insoluble organics. *J. Geophys. Res.* 111, D17202 (2006).
- Kulmala, M. et al. Direct observations of atmospheric aerosol nucleation. *Science* 339, 943-946 (2013).
- Kulmala, M.; Artaxo, P. et al., "General overview: European Integrated project on Aerosol Cloud Climate and Air Quality interactions (EUCAARI) integrating aerosol research from nano to global scales," *Atmos. Chem. Phys.*, 2011, 11, 13061.
- Kumar, V. V. C. Jakob, A. Protat, C. R. Williams, and P. T. May, 2015: Mass-Flux Characteristics of Tropical Cumulus Clouds from Wind Profiler Observations at Darwin, Australia. *J. Atmos. Sci.*, 72, 1837-1855, doi:10.1175/JAS-D-14-0259.1.
- Laurent, H.; Machado, L. A. T.; Morales, C. A.; Durieux, L., "Characteristics of the Amazonian mesoscale convective systems observed from satellite and radar during the WETAMC/LBA experiment," *J. Geophys. Res.* 2002, 107, 8054.
- Lawrence, D. and Vandecar, K.: Effects of tropical deforestation on climate and agriculture, *Nature Climate Change*, 5, 27-36, doi:10.1038/nclimate2430, 2015.
- Lelieveld, J.; Butler, T. M.; Crowley, J. N.; Dillon, T. J.; Fischer, H.; Ganzeveld, L.; Harder, H.; Lawrence, M. G.; Martinez, M.; Taraborrelli, D.; Williams, J., "Atmospheric oxidation capacity sustained by a tropical forest," *Nature* 2008, 452, 737-740.
- Lim, H. J.; Carlton, A. G.; Turpin, B. J. "Isoprene forms secondary organic aerosol through cloud processing: Model simulations," *Environmental Science & Technology*, 2005, 39, 4441.
- Lin, J. C.; Matsui, T.; Pielke, R. A.; Kummerow, C., "Effects of biomass-burning-derived aerosols on precipitation and clouds in the Amazon Basin: a satellite-based empirical study," *J. Geophys. Res.* 2006, 111, D19204.
- Lu, L., Denning, A S., Silva Dias, M.A.F., Silva Dias, P. L. , Longo, Marcos, Freitas, S. R. , Saatchi, S Mesoscale circulations and atmospheric CO2 variations in the Tapajós region, Pará, Brazil. *Journal of Geophysical Research.* , v.110, p.1 - 17, 2005
- Machado, L. A. T., H. Laurent, and A. A. Lima (2002), Diurnal march of the convection observed during TRMM-WETAMC/LBA, J.

- Geophys. Res., 107, 8064.
- Machado, L. A. T., Silva Dias, M. A. F et al., THE CHUVA PROJECT How Does Convection Vary across Brazil?, *B. Am. Meteorol. Soc.*, 95, 1365–1380, doi:10.1175/bams-d-13-00084.1, 2014.
- Machado, L. A. T., W. B. Rossow, R. L. Guedes, and A. W. Walker, 1998: Life cycle variations of mesoscale convective systems over the Americas. *Mon. Wea. Rev.*, 126, 1630-1654.
- Machado, L. A. T.; Laurent, H.; Dessay, N.; Miranda, I., "Seasonal and diurnal variability of convection over the Amazonia: A comparison of different vegetation types and large scale forcing," *Theor. Appl. Climatol.* 2004, 78, 61-77.
- Marengo, J. A., Tomasella, J., Alves, L. M., Soares, W. R., and Rodriguez, D. A.: The drought of 2010 in the context of historical droughts in the Amazon region, *Geophys. Res. Lett.*, 38, 1–5, doi:10.1029/2011GL047436, 2011.
- Martin, S. T., Andreae, M. O., Artaxo, P., Baumgardner, D., Chen, Q., Goldstein, A. H., Guenther, A., Heald, C. L., Mayol-Bracero, O. L., McMurry, P. H., Pauliquevis, T., Pöschl, U., Prather, K. A., Roberts, G. C., Saleska, S. R., Dias, M. A. S., Spracklen, D. V., Swietlicki, E., and Trebs, I.: Sources and properties of Amazonian aerosol particles, *Rev. Geophys.*, 48, RG2002, doi:10.1029/2008rg000280, 2010b.
- Martin, S. T., Artaxo, P., Machado, L. A. T., Manzi, A. O., Souza, R. A. F., Schumacher, C., Wang, J., Andreae, M. O., Barbosa, H. M. J., Fan, J., Fisch, G., Goldstein, A. H., Guenther, A., Jimenez, J. L., Pöschl, U., Silva Dias, M. A., Smith, J. N., and Wendsch, M.: Introduction: Observations and Modeling of the Green Ocean Amazon (GoAmazon2014/5), *Atmos. Chem. Phys.*, 16, 4785–4797, doi:10.5194/acp-16-4785-2016, 2016.
- Martin, S. T.; Andreae, M. O.; Althausen, D.; Artaxo, P.; Baars, H.; Borrmann, S.; Chen, Q.; Farmer, D. K.; Guenther, A.; Gunthe, S.; Jimenez, J. L.; Karl, T.; Longo, K.; Manzi, A.; Pauliquevis, T.; Petters, M.; Prenni, A.; Pöschl, U.; Rizzo, L. V.; Schneider, J.; Smith, J. N.; Swietlicki, E.; Tota, J.; Wang, J.; Wiedensohler, A.; Zorn, S. R. "An Overview of the Amazonian Aerosol Characterization Experiment 2008 (AMAZE-08)," *Atm. Chem. Phys.*, 2010, 10, 11415.
- Martin, S. T.; Andreae, M. O.; Artaxo, P.; Baumgardner, D.; Chen, Q.; Goldstein, A. H.; Guenther, A.; Heald, C. L.; Mayol-Bracero, O. L.; McMurry, P. H.; Pauliquevis, T.; Pöschl, U.; Prather, K. A.; Roberts, G. C.; Saleska, S. R.; Silva-Dias, M. A.; Spracklen, D. V.; Swietlicki, E.; Trebs, I., "Sources and Properties of Amazonian Aerosol Particles," *Rev. Geophys.* 2010, 48, RG2002.
- Martinez, I. S.; Peterson, M. D.; Ebben, C. J.; Hayes, P. L.; Artaxo, P.; Martin, S. T.; Geiger, F. M. "On molecular chirality within naturally occurring secondary organic aerosol particles from the central Amazon Basin," *Physical Chemistry Chemical Physics*, 2011.
- Martins, J. A., Silva Dias, M.A.F., Gonçalves, F. L. T. Impact of biomass burning aerosols on precipitation in the Amazon: A modeling case study. *Journal of Geophysical Research.*, v.114, p.D02207 -, 2009.
- Martins, J. A.; Dias, M., "The impact of smoke from forest fires on the spectral dispersion of cloud droplet size distributions in the Amazonian region," *Environmental Research Letters* 2009, 4.
- Martins, J.V., Marshak, A., Remer, L.A., Rosenfeld, D., Kaufman, Y.J., Fernandez-Borda, R., Koren, I., Correia, A.L., Zubko, V., and Artaxo, P. Remote sensing the vertical profile of cloud droplet effective radius, thermodynamic phase, and temperature. *Atmospheric Chemistry and Physics*, 11, 9485–9501, 2011.
- McComiskey, A. and G. Feingold, 2012: The scale problem in quantifying aerosol indirect effects, *ACP*, v. 12, pp. 1031-1049.
- McFiggans, G., Artaxo, P., Baltensperger, U., Coe, H., Facchini, M. C., Feingold, G., Fuzzi, S., Gysel, M., Laaksonen, A., Lohmann, U., Mentel, T. F., Murphy, D. M., O'Dowd, C. D., Snider, J. R., and Weingartner, E.: The effect of physical and chemical aerosol properties on warm cloud droplet activation, *Atmos. Chem. Phys.*, 6, 2593–2649, doi:10.5194/acp-6-2593-2006, 2006.
- Negri, A. J.; Adler, R. F.; Xu, L. M.; Surratt, J., "The impact of Amazonian deforestation on dry season rainfall," *J. Clim.* 2004, 17, 1306-1319.
- Ng, N. et al., An Aerosol Chemical Speciation Monitor (ACSM) for Routine Monitoring of the Composition and Mass Concentrations of Ambient Aerosol, *Aerosol Sci. Tech.*, 45, 780–794, doi:10.1080/02786826.2011.560211, 2011.
- Nobre, C. A., Sellers, P. J., and Shukla, J.: Amazonian deforestation and regional climate change, *J. Climate*, 4, 957–988, 1991.
- NUNES, ANA M. P.; Silva Dias, Maria A. F.; ANSELMO, EVANDRO M.; Morales, Carlos A.. 2016. Severe Convection Features in the Amazon Basin: A TRMM-Based 15-Year Evaluation In *Frontiers in Earth Science.*, v.4, 2016/000037
- Oliveira, P. H. F.; Artaxo, P.; Pires, C.; Lucca, S.; Procópio, A.; Holben, B.; Schafer, J.; Cardoso, L. F.; Wofsy, S. C.; Rocha, H. R., "The effects of biomass burning aerosols and clouds on the CO2 flux in Amazonia," *Tellus* 2007, 59B, 338-349.
- Pérez-Ramírez, D., Andrade, M., Eck, T., Stein, A., O'Neill, N., Lyamani, H., Gassó, S., Whiteman, D.N., Veselovskii, I., Velarde, F., Alados-Arboledas, L., Multi year aerosol characterization in the tropical Andes and in adjacent Amazonia using AERONET measurements, *Atmospheric Environment* (2017), doi: 10.1016/j.atmosenv.2017.07.037.
- Petersen, W. A., R. Fu, M. Chen, and R. Blakeslee, 2006: Intraseasonal forcing of convection and lightning activity in the southern Amazon as a function of cross-equatorial flow. *J. Climate*, 19, 3180–3196.
- Pöhlker, C.; Wiedemann, K. T.; Sinha, B.; Shiraiwa, M.; Gunthe, S. S.; Smith, M.; Su, H.; Artaxo, P.; Chen, Q.; Cheng, Y.; Elbert, W.; Gilles, M. K.; Kilcoyne, A. L. D.; Moffet, R. C.; Weigand, M.; Martin, S. T.; Pöschl, U.; Andreae, M. O. "Biogenic Potassium Salt Particles as Seeds for Secondary Organic Aerosol in the Amazon," *Science*, 2012, 337, 1075.
- Pöhlker, M. L., et al., Long-term observations of cloud condensation nuclei in the Amazon rain forest – Part 2: Near-pristine episodes, ultrafine particle bursts, biomass burning and long range transport events, in preparation, 2017a.
- Pöschl, U.; Martin, S. T.; Sinha, B.; Chen, Q.; Gunthe, S. S.; Huffman, J. A.; Borrmann, S.; Farmer, D. K.; Garland, R. M.; Helas, G.; Jimenez, J. L.; King, S. M.; Manzi, A.; Mikhailov, E.; Pauliquevis, T.; Petters, M. D.; Prenni, A. J.; Roldin, P.; Rose, D.; Schneider, J.; Su, H.; Zorn, S. R.; Artaxo, P.; Andreae, M. O. "Rainforest aerosols as biogenic nuclei of clouds and precipitation in the Amazon," *Science*, 2010, 329, 1513.
- Prenni, A. J.; Petters, M. D.; Kreidenweis, S. M.; Heald, C. L.; Martin, S. T.; Artaxo, P.; Garland, R. M.; Wollny, A. G.; Poeschl, U. "Relative roles of biogenic emissions and Saharan dust as ice nuclei in the Amazon basin," *Nature Geoscience*, 2009, 2, 402.
- Procópio, A. S.; Artaxo, P.; Kaufman, Y. J.; Remer, L. A.; Schafer, J. S.; Holben, B. N., "Multiyear analysis of Amazonian biomass burning smoke radiative forcing of climate," *Geophys. Res. Lett.* 2004, 31, L03108.
- Procópio, A. S.; Remer, L. A.; Artaxo, P.; Kaufman, Y. J.; Holben, B. N., "Modeled spectral optical properties for smoke aerosols in Amazonia," *Geophys. Res. Lett.* 2003, 30, 2265.

- Raasch, S. and M. Schröter, 2001: PALM - A large-eddy simulation model performing on massively parallel computers. *Meteorol. Z.*, 10, 363-372.
- Rapp, A. D., C. D. Kummerow, and L. Fowler (2011), Interactions between warm rain clouds and atmospheric preconditioning for deep convection in the tropics, *J. Geophys. Res.*, 116, D23210.
- Rasmussen, K. L., and R. A. Houze, 2011: Orographic convection in subtropical South America as seen by the TRMM satellite. *Mon. Wea. Rev.*, 139, 2399-2420.
- Raup and Silva Dias 2009: Resonant Wave Interactions in the Presence of a Diurnally Varying Heat Source. *J. Atmos. Sci.*, 66, pp. 3165.
- Remer, L. A., Mattoo, S., Levy, R. C., & Munchak, L. (2013). MODIS 3 km aerosol product: algorithm and global perspective. *Atmospheric Measurement Techniques Discussions*, 6, 69-112.
- Reutter, P., Su, H., Trentmann, J., Simmel, M., Rose, D., Gunthe, S. S., Wernli, H., Andreae, M. O., and Pöschl, U.: Aerosol and updraft-limited regimes of cloud droplet formation: influence of particle number, size and hygroscopicity on the activation of cloud condensation nuclei (CCN), *Atmos. Chem. Phys.*, 9, 7067–7080, doi:10.5194/acp-9-7067-2009, 2009.
- Riipinen, I., Yli-Juuti, T., Pierce, J. R., Petaja, T., Worsnop, D. R., Kulmala, M., and Donahue, N. M., The contribution of organics to atmospheric nanoparticle growth: *Nature Geoscience*, 5, 453-458, doi:10.1038/ngeo1499, 2012.
- Rissler, J., Swietlicki, E., Zhou, J., Roberts, G., Andreae, M. O., Gatti, L. V., and Artaxo, P.: Physical properties of the submicrometer aerosol over the Amazon rain forest during the wet to-dry season transition – comparison of modeled and measured CCN concentrations, *Atmos. Chem. Phys.*, 4, 2119–2143, doi:10.5194/acp-4-2119-2004, 2004.
- Rissler, J., Vestin, A., Swietlicki, E., Fisch, G., Zhou, J., Artaxo, P., and Andreae, M. O.: Size distribution and hygroscopic properties of aerosol particles from dry-season biomass burning in Amazonia, *Atmos. Chem. Phys.*, 6, 471–491, doi:10.5194/acp-6-471-2006, 2006.
- Rizzo, L. V. et al., "Multi-year observation of submicrometer particle number size distributions at a primary forest site in Amazonia", *in preparation*, 2017.
- Rizzo, L. V., Artaxo, P., Karl, T., Guenther, A. B., Greenberg, J. P. Aerosol properties, in-canopy gradients, turbulent fluxes and VOC concentrations at a pristine forest site in Amazonia. *Atmospheric Environment*, Vol. 44, Issue 4, 503-511, 2010.
- Rizzo, L. V.; Artaxo, P.; Müller, T.; Wiedensohler, A.; Paixão, M.; Cirino, G. G.; Arana, A.; Swietlicki, E.; Roldin, P.; Fors, E. O.; Wiedemann, K. T.; Leal, L. S. M.; Kulmala, M. "Long term measurements of aerosol optical properties at a pristine forest site in Amazonia," *Atmos. Chem. Phys. Discuss.*, 2012, 12, 23333.
- Rizzo, L. V.; Correia, A. L.; Artaxo, P.; Procópio, A. S.; Andreae, M. O. "Spectral dependence of aerosol light absorption over the Amazon Basin," *Atmospheric Chemistry and Physics*, 2011, 11, 8899.
- Roberts, G. C. P. Artaxo, J. Zhou, E. Swietlicki, M. O. Andreae, Sensitivity of CCN spectra on chemical and physical properties of aerosol: A case study from the Amazon Basin *J. Geophys. Res.* 107, No. D20, 8070 - 8088, 2002.
- Roberts, G. C., Nenes, A., Seinfeld, J. H., and Andreae, M. O.: Impact of biomass burning on cloud properties in the Amazon Basin, *J. Geophys. Res.-Atmos.*, 108, ACC 9-1-ACC 9-19., doi:10.1029/2001jd000985, 2003.
- Roberts, M. C., Andreae, M. O., Zhou, J. C., and Artaxo, P.: Cloud condensation nuclei in the Amazon Basin: "Marine" conditions over a continent?, *Geophys. Res. Lett.*, 28, 2807–2810, 2001.
- Romatschke, U., and R. A. Houze, 2010: Extreme summer convection in South America. *J. Climate*, 23, 3761-3791.
- Rose, C., K. Sellegri, F. Velarde, I. Moreno, M. Ramonet, K. Weinhold, R. Krejci, P. Ginot, M. Andrade, A. Wiedensohler, P. Laj (2015). Multiple daytime nucleation events at the high altitude station of Chacaltaya (5240 m a.s.l.), Bolivia – AE Vol. 102, February 2015, Pages 18-29.
- Rosenfeld, D., Andreae, M. O., et al., Global observations of aerosol-cloud-precipitation-climate interactions: *Rev. Geophys.*, 52, 750-808, doi:10.1002/2013RG000441, 2014.
- Rosenfeld, D., U. Lohmann, G. B. Raga, C. D. O'Dowd, M. Kulmala, S. Fuzzi, A. Reissell, and M. O. Andreae, Flood or drought: How do aerosols affect precipitation?, *Science*, 2008, 321, 1309–1313.
- Saad, Sandra I.; da Rocha, Humberto R.; Silva Dias, Maria A. F.; Rosolem, Rafael. 2010. Can the Deforestation Breeze Change the Rainfall in Amazonia? A Case Study for the BR-163 Highway Region In *Earth Interactions*. , v.14, 1-25
- SARAIVA, I.; Silva Dias, M. A. F.; MORALES, C. A. R.; SARAIVA, J. M. B.. 2016. Regional Variability of Rain Clouds in the Amazon Basin as Seen by a Network of Weather Radars In *Journal of Applied Meteorology and Climatology*. , v.55, 2657-2675
- Schafer, J. S.; Eck, T. F.; Holben, B. N.; Artaxo, P.; Duarte, A. F. "Characterization of the optical properties of atmospheric aerosols in Amazonia from long-term AERONET monitoring (1993-1995 and 1999-2006)," *J. Geophys. Res.*, 2008, 113.
- Schneider, J.; Freutel, F.; Zorn, S. R.; Chen, Q.; Farmer, D. K.; Jimenez, J. L.; Martin, S. T.; Artaxo, P.; Wiedensohler, A.; Borrmann, S. "Mass-spectrometric identification of primary biological particle markers and application to pristine submicron aerosol measurements in Amazonia," *Atm. Chem. Phys.*, 2011, 11, 11415.
- Sena, E.T., Artaxo, P., and Correia, A.L. Spatial variability of the direct radiative forcing of biomass burning aerosols and the effects of land use change in Amazonia. *Atm. Chem. Phys.*, 13, 1261–1275, 2013.
- Shipway and Hill, 2012, Diagnosis of systematic differences between multiple parametrizations of warm rain microphysics using a kinematic framework, *Q.J.R. Meteorol. Soc.* doi: 10.1002/qj.1913.
- Shrivastava, Manish et al., Recent advances in understanding secondary organic aerosol: Implications for global climate forcing. *Review of Geophysics*, 2017.
- Silva Dias, M. A. F., Avissar, R., Silva Dias, P. 2009 Modeling the Regional and Remote Climatic Impact of Deforestation In: *Amazonia and Global Change. Geophysical Monographs Series ed.: American Geophysical Union*, v.186, p. 255-264.
- Silva Dias, M. A. F., Dias, J., Carvalho, L. M. V., Freitas, E. D., Silva Dias, P. L. 2013, Changes in extreme daily rainfall for São Paulo, Brazil. *Climatic Change*. 116:705–722.
- Silva Dias, M.A.F., et al., 2002 Clouds and rain processes in a biosphere atmosphere interaction context in the Amazon Region. *J. Geophys. Res.*, 107, 46.1 - 46.23, 2002.
- Silva Dias, M.A.F., Silva Dias, P. L. Longo, M., Fitzjarrald, D. R. , S., A, Denning River breeze circulation in eastern Amazon: observations and modeling results. *Theoretical and Applied Climatology*. , v.78, 111 - 121, 2004.

- Stohl, A., Forster, C., Frank, A., Seibert, P., and Wotawa, G., Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2: Atmos. Chem. Phys., 5, 2461-2474, 2005.
- Thalman, R., et al., CCN activity and organic hygroscopicity of Amazonian aerosols – seasonal and diel variations and impact of anthropogenic emissions, in preparation, 2017.
- Tröstl, J., et al., The role of low-volatility organic compounds in initial particle growth in the atmosphere: Nature, 533, 527-531, 1941 doi:10.1038/nature18271, 2016.
- Tunved, P., Hansson, H.-C., Kerminen, V.-M., Strom, J., Dal Maso, M., Lihavainen, H., Viisanen, Y., Aalto, P.P., Komppula, M., Kulmala, M., High natural aerosol loading over boreal forests, Science, 2006 312, 261–263.
- Twomey, S.: The nuclei of natural cloud formation—part II: The supersaturation in natural clouds and the variation of cloud droplet concentration, Geofis Pura e Appl, 43, 243–249, 1959.
- Wang, Q., Saturno, J., Chi, X., Walter, D., Lavric, J. V., Moran-Zuloaga, D., Ditas, F., Pöhlker, C., Brito, J., Carbone, S., Artaxo, P., and Andreae, M. O., Modeling investigation of light-absorbing aerosols in the Amazon Basin during the wet season: Atmos. Chem. Phys., 16, 14,775-14,794, doi:10.5194/acp-16-14775-2016, 2016.
- Wendisch, M., Pöschl, U., Andreae, M. O., Machado, L. A. T., Albrecht, P. Artaxo et al., ACRIDICON-CHUVA Campaign: Studying Tropical Deep Convective Clouds and Precipitation over Amazonia Using the New German Research Aircraft HALO, B. Am. Meteorol. Soc., 97, 1885–1908, doi:10.1175/bams-d-14-00255.1, 2016.
- Whitehead, J. D., Darbyshire, E., Brito, J., Barbosa, H. M. J., Crawford, I., Stern, R., Gallagher, M. W., Kaye, P. H., Allan, J. D., Coe, H., Artaxo, P., and McFiggans, G.: Biogenic cloud nuclei in the central Amazon during the transition from wet to dry season, Atmos. Chem. Phys., 16, 9727–9743, doi:10.5194/acp-16-9727-2016, 2016.
- Williams, C. R., R. M. Beauchamp, and V. Chandrasekar, Vertical Air Motions and Raindrop Size Distributions Estimated Using Mean Doppler Velocity Difference From 3- and 35-GHz Vertically Pointing Radars. IEEE Trans. Geosci. Remote Sens., 54, 6048–6060, doi:10.1109/TGRS.2016.2580526, 2016.
- Williams, C. R., W. L. Ecklund, and K. S. Gage, Classification of Precipitating Clouds in the Tropics Using 915-MHz Wind Profilers. J. Atmos. Ocean. Technol., 12, 996–1012, doi:10.1175/1520-0426(1995)012<0996:COPCIT>2.0.CO;2, 1995.
- Williams, E.; Machado, L.; Artaxo, P.; et al., Contrasting convective regimes over the Amazon: Implications for cloud electrification, J. Geophys. Res., 2002, 107, 8082.
- Wright, J.S., Rong Fu, John R. Worden, Sudip Chakraborty, Nicholas E. Clinton, Camille Risi, Ying Sun, and Lei Yin. Rainforest-initiated wet season onset over the southern Amazon. PNAS doi/10.1073/pnas.1621516114, 2017.
- Wu, C.M., Stevens, B and Arakawa, A.: What Controls the Transition from Shallow to Deep Convection? J. Atmos. Sci, 66, 1793-1806, 2009.
- Yáñez-Serrano, A. M., Nölscher, A. C., Williams, J., Wolff, S., Alves, E., Martins, G. A., Bourtsoukidis, E., Brito, J., Jardine, K., Artaxo, P., and Kesselmeier, J.: Diel and seasonal changes of biogenic volatile organic compounds within and above an Amazonian rainforest, Atmos. Chem. Phys., 15, 3359–3378, doi:10.5194/acp-15-3359-2015, 2015.
- Zhang, Z., Engling, G., Zhang, L., Kawamura, K., Yang, Y., Tao, J., Zhang, R., Chan, C.-Y., and Li, Y.: Significant influence of fungi on coarse carbonaceous and potassium aerosols in a tropical rainforest, Environ. Res. Lett., 10, 034015, doi:10.1088/1748-9326/10/3/034015, 2015.