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Trace Gases-Aerosol-Cloud Interactions in Amazonia: from Bioaerosols Emissions to Large Scale Impacts (GAIA)

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Abstract

Amazonia is a living laboratory to study critical processes that regulate tropical atmospheric chemistry and physics and influence climate regionally and globally. Long-term observations and process studies are essential to unveil the fate of the Amazonian Forest. We

plan to advance science over critically important processes that influence the hydrological cycle, radiation balance, reactive and greenhouse trace gas exchanges, bioaerosols, and the functioning of the Amazonian ecosystem. This will be done through a combination of 7 components in this proposal: 1) Improvements of the ATTO tower long term continuous measurements, with the addition of new aerosol and trace gas studies, such as using the newly installed automatic elevator (RoLi system) that will allow continuous day and night vertical profiles from the ground up to 325 meters. We will also enhance measurements of nanoparticles (2-10 nanometers) and how they form secondary organic aerosols (SOA) and grow up to Cloud Condensation Nuclei (CCN) sizes; 2) Improvements at the cloud-aerosol interactions with new measurements at the ATTO Campina site, integrating data from the ground up to 16 Km profiling instruments such as the aerosol lidar, cloud radar, precipitation radar, radiometers, and the scanning meteorological radar that is being installed at Balbina. Detailed cloud-aerosol-trace gases interactions over the vertical column will be studied in this component; 3) Extending the ATTO measurements to large-scale basin-wide Amazonian, through extensive ship cruises along the Solimões, Negro, and Amazonas rivers, using the UEA ship as the river platform. The focus on these measurements are distribution and interactions of VOCs, aerosols, greenhouse gases and biological particles; 4) Perform data analysis of the CAFÉ-Brazil experiment, with the HALO plane flying up to 14 Km, and use small aircrafts to perform vertical profiles of aerosols, greenhouse gases, biological particles and other measurements up to 5 Km; 5) Perform detailed biological primary aerosol particle measurements basin-wide, with fluorescence detection of bacteria, fungal spores, plant debris, and other types of bioaerosols, including whole DNA metagenomics sequencing analysis on aerosol particles to identify airborne species that are long-range transported from Africa to Amazonia, and within Amazonia; 6) Integrate large-scale greenhouse gas measurements, such as CO_2 and CH_4, including isotopic determination, trough ship cruises and towers sites along Amazonia; 7) Implement a modeling component integrated with basin-wide remote sensing measurements to help generalize basin-wide properties from fixed point measurements and ship campaigns, improving process understanding.

We plan to unveil the complex trace gases-cloud-aerosol-precipitation interactions with these innovative measurements, more complete datasets, and associated modeling efforts. We will also study the feedback between the biosphere and atmosphere with human activities through deforestation and biomass burning emissions. We expect that these measurements and modeling framework will provide new insights into critical processes that regulate tropical atmospheric chemistry, cloud physics, climate, and the integrated functioning of tropical forests.

Resumo (in Portuguese)

A Amazônia é um laboratório vivo para estudar processos críticos que regulam a química e a física da atmosfera tropical e influenciam o clima regional e global. Observações de longo prazo e estudos de processos são importantes para desvendar possíveis mecanismos de degradação da Floresta Amazônica. Planejamos avançar a ciência sobre processos de importância crítica que influenciam o ciclo hidrológico, o balanço de radiação, as trocas de gases reativos e de efeito estufa, os bioaerossóis e o funcionamento do ecossistema amazônico. Isso será feito por meio de uma combinação de 7 componentes nesta proposta: 1) Melhorias nas medições contínuas de longo prazo da torre ATTO, com a adição de novos estudos de aerossóis e gases traços, com o uso do elevador automático recém-instalado (sistema RoLi) que permitirá perfis verticais diurnos e noturnos contínuos desde o solo até 325 metros. Iremos também medir nanopartículas na faixa de 2 a 10 nanômetros, e estudar como

elas crescem formando aerossóis orgânicos secundários (SOA) e formam núcleos de condensação de nuvens (CCN); 2) Melhorias nos estudos de interações nuvens-aerossol com novas medidas no site ATTO Campina, com perfis verticais do solo até 16 Km de altitude, com instrumentos de perfilagem como o Lidar de aerossóis, radar de nuvens, radar de precipitação, radiômetros e o radar meteorológico de varredura que está sendo instalado em Balbina. As interações nuvem-aerossol-traços de gases sobre a coluna vertical serão estudadas nesta componente; 3) Estender as medições da torre ATTO para toda a bacia amazônica em larga escala, através de extensos cruzeiros de navios ao longo dos rios Solimões, Negro e Amazonas, usando o navio da UEA como plataforma fluvial. O foco dessas medições são a distribuição e interações de VOCs, aerossóis, gases de efeito estufa e partículas biológicas; 4) Realizar análises de dados do experimento CAFE-Brazil com medidas até 14 Km de altitude em toda a Amazônia, e utilizar aeronaves de pequeno porte para realizar perfis verticais de aerossóis, gases de efeito estufa, partículas biológicas e outras medidas até 5 Km; 5) Realizar medidas de partículas de aerossóis biológicas primárias detalhadas em toda a bacia, com detecção de fluorescência em partículas, possibilitando a especiação de bactérias, esporos de fungos, fragmentos de plantas e outros tipos de bioaerossóis. Nesta componente vamos realizar análises metagenômicas de DNA em partículas de aerossol para identificar espécies transportadas atmosfericamente por longas distâncias da África à Amazônia, e dentro da Amazônia; 6) Integrar medições de gases de efeito estufa em larga escala, como CO_2 e CH_4, incluindo determinação isotópica, através de cruzeiros em vários rios e amostragens nas torres do LBA ao longo da Amazônia; 7) Implementar uma componente de modelagem integrada para ajudar a generalizar propriedades em toda a bacia a partir de medições de pontos fixos e campanhas de navios, além de medidas por sensoriamento remoto.

Com essas medidas inovadoras e conjuntos de dados mais completos e esforços de modelagem associados, planejamos desvendar as complexas interações gases-traço-nuvens-aerossol-precipitação. Também estudaremos os feedbacks entre a biosfera e a atmosfera com as atividades antropogênicas através dos impactos do desmatamento e das emissões de queimadas. Esperamos que essas atividades integradas forneçam novos insights sobre processos importantes que regulam a química atmosférica tropical, a física das nuvens, o clima sobre florestas tropicais e o funcionamento integrado do ecossistema amazônico.

1 – Main achievements of previous FAPESP Thematic project (2017/17047-0) and new proposed initiatives

The FAPESP Thematic project that is just finishing in July 2023 had many achievements, advancing the scientific understanding of the functioning of the complex Amazonian Ecosystem significantly. The project had the title "***Aerosol and clouds lifecycles in Amazonia: biogenic emissions, biomass burning and Impacts on the Ecosystem*.**" Among the achievements was the study of long-range transport of aerosols from Amazonia to the high Andes and its impacts on snow and atmospheric chemistry, with two years of operation of a complex aerosol and trace gases measurement system in Chacaltaya at 5240 meters above sea level. We also continuously measure aerosols and trace gases in the 325 meters ATTO tall tower in partnership with Max Planck Institute, INPA, and other Brazilian partners. This was done during the COVID-19 pandemic. We have performed the CAFÉ-Brazil airborne experiment, operating the DLR HALO (High-Altitude Long-Range Observatory) for about two months in Brazil. The HALO collected extensive data from the ground up to 14 Km altitude over most of the Amazon Forest area in the wet season 2022-2023. Finally, we performed a fluvial river campaign, simultaneously with the CAFÉ-Brazil experiment, operating a large University of Amazon State (UEA) boat fully instrumented along the Negro River from Manaus to São Gabriel da Cachoeira. The table below shows a few statistics regarding publications and students involved with this Thematic project. Each publication's titles and full info were listed in the 5 FAPESP complete project reports each year. It is important to emphasize that several high-profile papers were published in the Nature and Science family of journals.

Year	2019	2020	2021	2022	2023	Total
Publications	54	42	47	36	40	219
Masters and Ph.D. Thesis	0	1	1	11	4	17

For this proposal, we are planning to improve measurements as well as to perform new initiatives and activities associated with scientific topics that will be described through the proposal:

1) **Improvements of the ATTO tower's long-term continuous measurements, with the addition of new aerosol and trace gas studies, for instance, using the newly installed automatic elevator (RoLi system) that will allow continuous day and vertical night profiles from the ground up to 325 meters.**

2) **Improvements at the ATTO Campina site measurements with new energy, water vapor, and carbon flux tower, as well as integration of the vertical profiling instruments such as the aerosol lidar, cloud radar, precipitation radar, and the scanning meteorological radar that is being installed at Balbina. Detailed cloud-aerosol-trace gases interactions.**

3) **Extending the ATTO measurements to basin-wide, trough ship cruises along the Amazon, Negro, and Solimões rivers, using the UEA ship as the platform. Focus on the distribution of VOCs, aerosols, greenhouse gases, and biological particles with DNA metagenomic analysis.**

4) **Perform data analysis of the CAFÉ-Brazil experiment, with the HALO plane flying up to 14 Km, and use small aircraft to perform vertical profiles of aerosols, greenhouse gases, biological particles, and other measurements up to 5 Km.**

5) **Perform detailed biological primary aerosol particle measurements basin-wide, with fluorescence detection of bacteria, fungi, spores, and other biological particles.**

6) **Perform whole DNA metagenomics analysis on aerosols to identify airborne species long-range transported from Africa to Amazonia and within Amazonia.**

7) **Integrate large-scale greenhouse gas measurements, mainly CO_2 and CH_4, including isotopic determination, through ship cruises and LBA tower sites along Amazonia.**

8) **Implement a modeling component integrated with remote sensing measurements to help extrapolate basin-wide properties from fixed point measurements and ship campaigns. Use of modeling strategy to improve process understanding.**

2 – The Amazon: a unique integrated laboratory for the atmosphere, aquatic system, and biota

The Amazon is a key region maintaining the regional climate in South America and is also critically important for the global climate (Artaxo, 2019, Lovejoy and Nobre, 2018). It is also one of the few continental areas where it is still possible to observe close-to-pristine atmospheric conditions in the wet season regarding aerosols and trace gases (Andreae et al., 2009, 2016, Artaxo et al., 2013). Wet season atmospheric conditions for aerosol particles resemble background conditions typical of pre-industrialization (Andreae et al., 2009, Davidson et al., 2012, Carslaw, 2017), with particle number concentrations below 300 particles/cc and very low fine mode aerosol concentration ($PM_{2.5}$) at approximately 2-3 µg/m³ (Rizzo et al., 2013, 2018). It is becoming clear that the monitoring of atmospheric conditions in quasi pristine areas should be a priority in atmospheric studies (Kulmala et al., 2023).

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). The natural biogenic emissions of gases and aerosol particles, in combination with high water vapor and strong solar radiation, maintain chemical and physical cycles that sustain the aerosol particle population, the cloud fields, and the hydrological cycle of the basin through very complex mechanisms that only recently started to be studied (Artaxo et al., 2023). The biology of the forest has a critical role in regulating atmospheric composition and climate over the region (Artaxo, 2012). The Amazonian ecosystem uses the feedstock of plant and microbial emissions combined with high water vapor, solar radiation, and photo-oxidant levels to produce secondary organic aerosol (SOA) and primary biological aerosol particles. Through these emissions, the forest has powerful interactions with the atmosphere (Pöschl et al., 2010; Martin et al., 2010, 2017, Artaxo et al., 2013). The intense hydrologic cycle of the basin is one of the primary heat engines of global circulation (Nobre et al., 2009). The emissions of aerosol precursors and the primary biological particles sustain the hydrological cycle (Prenni et al. 2009). Figure 2.1 illustrates some of these integrated processes.

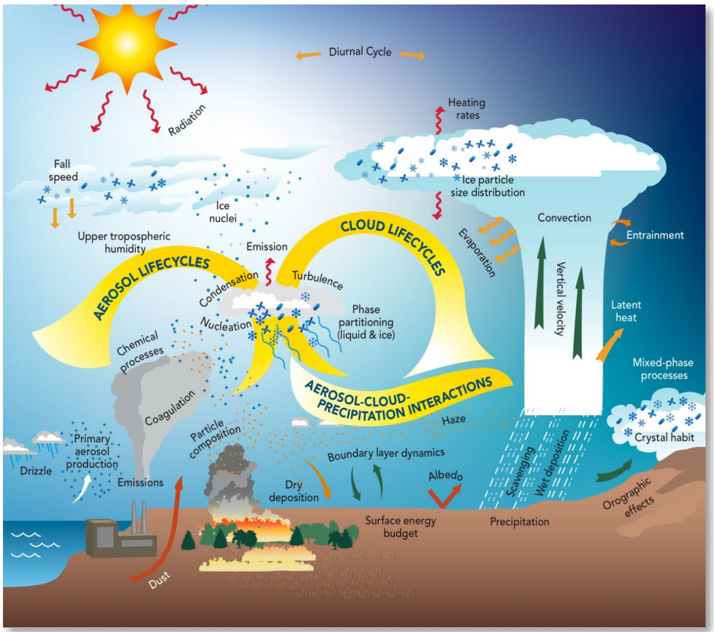


Figure 2.1 - Atmospheric system depiction showing connections between the aerosol life cycle interacting with the cloud life cycle through the surface and thermodynamic processes. The role of surface fluxes, including natural and anthropogenic emissions, is very important for both life cycles. The role of convection and atmospheric thermodynamic conditions is also critically important. Interaction with solar radiation and convection makes important impacts on the ecosystem. (DOE Atmospheric System Research)

Anthropogenic disturbances are changing the Amazonian forests very significantly. The deforestation rate of 12.000 Km² in 2022 illustrates the recent destruction of the primary forest. After the deforestation, smoke associated with large-scale biomass burning emissions changes the pristine picture with very high aerosol and trace gases concentrations over the months of August to December, covering large amounts of forest area. 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., 2010, 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), affecting the CCN population and, consequently, cloud formation, evolution, and rainfall elsewhere in South America. 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 in 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., 2015). Recent work on long-range Sahara dust transport (Holanda et al., 2023) shows that these events can also bring together black carbon and biogenic aerosol particles from Africa to Brazil. In long-time scales, the transport of nutrients from Africa to Brazil can impact biogeochemical cycles.

2.1 – The forest emissions of biogenic volatile organic compounds (BVOC)

Central to the interactions of forest with climate in Amazonia are the biogenic Volatile organic compounds (BVOCs) (Yanez Serrano 2020). Amazonian BVOCs are emitted from plants during growth, maintenance, decay, and consumption (Guenther et al., 2012). 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 with the Amazon. The OH oxidation pathway is particularly important for BVOC oxidation in the tropics, given the high radiation levels and H_2O 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. However, knowledge of the composition, the sources, chemistry, and the role of the secondary organic components of particles in the atmosphere and Earth's climate system still needs to be improved. Even for the well-studied isoprene compound, recent analysis suggests that state-of-the-art atmospheric chemistry models considerably underpredict OH concentrations (Lelieveld et al., 2008), possibly implying important under-predicted and underestimated chemistry.

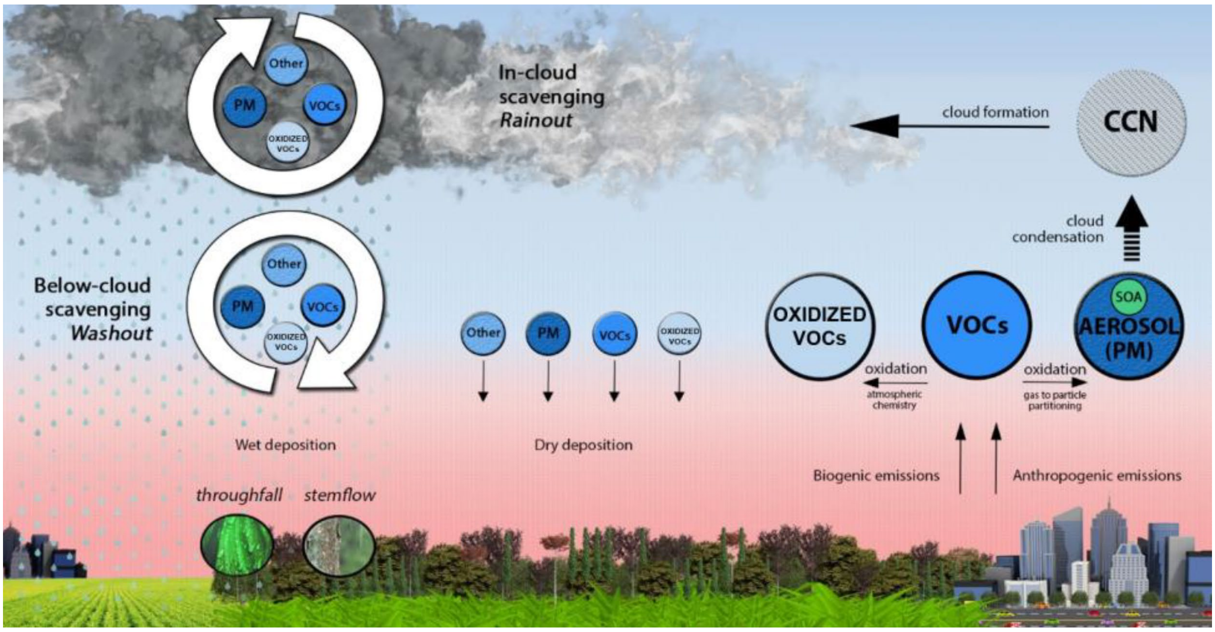


Figure 2.1.1 – The complex role of volatile organic compounds (VOCs) in the Amazonian atmosphere and biosphere, showing the different drivers, cloud processing, and deposition (Yáñez-Serrano et al., 2020).

2.2 - Secondary Organic Aerosol production, particle formation, CCN, and clouds

Organic aerosols dominate the aerosol composition, responsible for 70-85% of the aerosol mass. Two organic aerosol components exist in tropical forests: Primary biological aerosol particles (PBAP) and secondary organic aerosols (SOA). PBOA dominates the coarse mode, consisting of pollen, fungus, bacteria, and plant debris. SOA production involves a variety of trace gases, such as BVOCs, aromatics, nitrogen oxides (NOx), ozone (O_3), hydroxyl radical (OH), and sulfur species, including dimethyl sulfide (DMS) and sulfur dioxide (SO_2) (Shrivastava et al., 2017, Chen et al., 2016, Sá et al., 2018). BVOCs and aromatics react with O_3 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 NOx together influence the concentrations of O_3 and OH, thereby influencing the production of BVOC oxidation products (Yanez Serrano et al., 2022). Reactions in the gas phase and cloud water and vertical convection to higher altitudes are essential. Previous studies in the pristine Amazon rainforest showed that fine particles (which account for most of the cloud condensation nuclei) consist primarily 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). There is a complex pathway for oxidation products of isoprene and proxies of biomass burning emissions (such as black carbon and acetonitrile), with NO controlling the chemical route in Amazonia (Langford et al., 2022). Figure 2.2.1 shows some complex mechanisms involved in atmospheric transport and processing (Kadir et al., 2023).

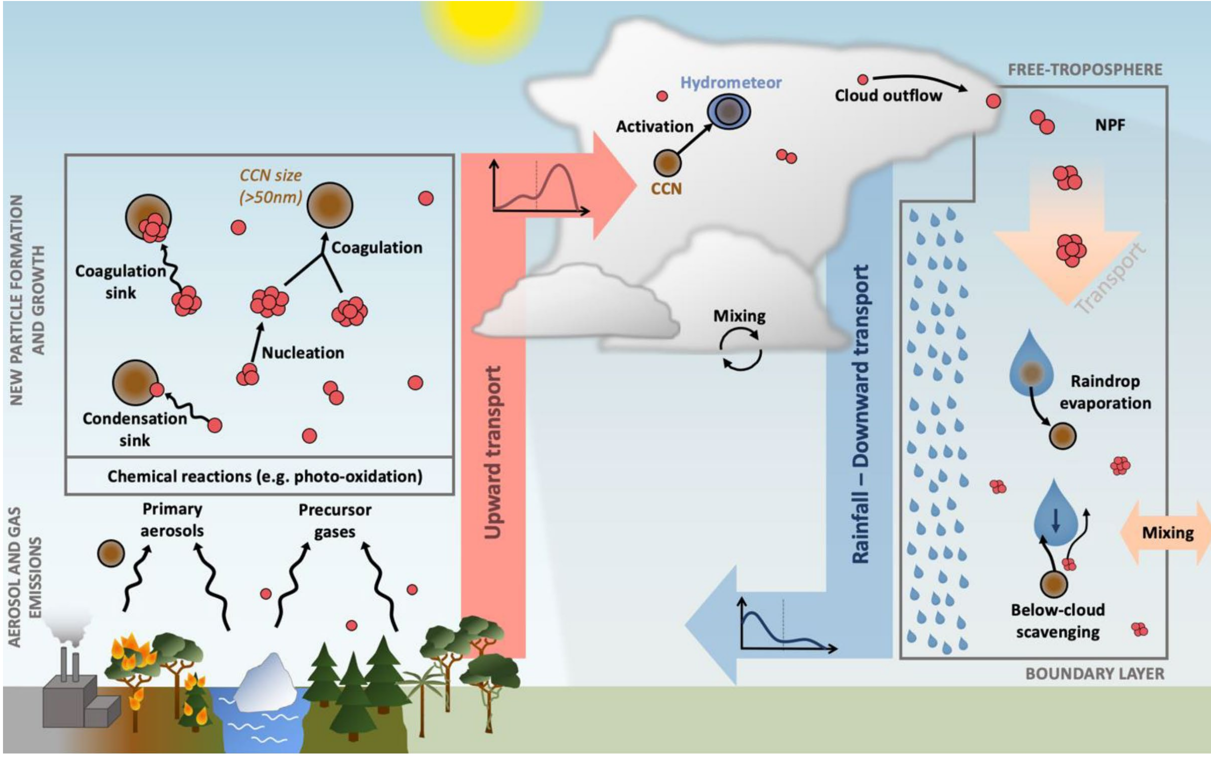


Figure 2.2.1 - Schematic of the links between VOCs, nucleation, transport, cloud processing, downdrafts, and precipitation, as explored in Kadir et al., 2023.

2.3 – Primary biogenic aerosol particles in Amazonia

Aerosols of biological origin, also called bioaerosols or primary biological aerosol particles (PBAP), play a vital role in the Earth system, particularly in the interactions between the atmosphere, biosphere, climate, and public health (Pöhlker et al., 2012, Pöschl et al., 2010, Prass et al. 2021). The atmosphere connects habitats across multiple spatial scales via the airborne dispersal of microbial cells, propagules, and biomolecules, and atmospheric microorganisms have been implicated in various biochemical and biophysical transformations (Šantl-Temkiv et al., 2022). They are defined as solid or liquid airborne particles of biological origin emitted directly from the biosphere into the atmosphere. Airborne bacteria, fungal spores, archaea, algae, pollen, and other bioparticles are essential for the reproduction and spread of organisms across various ecosystems (Pöhlker et al., 2012). They can cause or enhance human, animal, and plant diseases. They represent a highly diverse particle mixture in terms of size (i.e., a few nanometers up to hundreds of micrometers), morphology, taxonomy, and metabolic state occurring in the atmosphere throughout the globe. They are primarily emitted passively through mechanical momenta, such as air currents or rain splash, but also actively through humidity-triggered spore discharge mechanisms. Moreover, they can serve as nuclei for cloud droplets, ice crystals, and precipitation, thus influencing the hydrological cycle and climate. The sources, abundance, composition, and effects of biological aerosols and the atmospheric microbiome are, however, not yet well characterized and constitute a significant gap in the scientific understanding of the interaction and co-evolution of life and climate in the Earth system (Frolich-Nowoisky et al., 2016, Pöschl and Shiraiwa, 2015). Once emitted, the atmospheric lifetime of PBAP depends on particle properties (i.e., size, density, hygroscopicity), their mixing state (e.g., their potential adhesion to other aerosol particles), as well as meteorological conditions (i.e., wind speed and precipitation), and typically ranges from a few minutes up to days (Löbs et al., 2020).

Given that most fungi in the air occur as dormant spores, it is likely that the active fraction of atmospheric microbiota is comprised almost exclusively of bacteria that possess adaptation to atmospheric environmental stress (Archer et al. 2022). Putative generation times for bacteria in cloud water have been estimated at 3.6–19.5 days, which fits within their modeled airborne residence times and allows for long-range transport.

Bioaerosols are regarded as important factors in climatology, ecology, agriculture, and public health, with their role and relevance in these fields still being a matter of active research (Prass et al., 2021). In terms of atmospheric processes and relevance for the Earth's climate system, the ability of PBAP to act as (giant) CCN and ice nuclei (INP), as well as their interaction with chemical processes in the atmosphere, have been shown (Prenni et al., 2009). Figure 2.3.1 shows the major bioaerosol components in the atmosphere, with the extensive range of bioaerosols and their atmospheric processing microbiology (see also Artaxo and Hansson, 1995).

After emission from the biosphere, bioaerosol particles interact with other aerosol particles and trace gases in the atmosphere and can be involved in forming clouds and precipitation. After dry or wet deposition to the Earth's surface, viable bioparticles can contribute to biological reproduction and further emission. This feedback can be remarkably efficient when coupled with the water cycle (bioprecipitation) (Frolich-Nowoisky et al., 2016). Figure 2.3.2 shows the potential anthropogenic impacts on atmospheric microbiology. The recent COVID-19 pandemic and its atmospheric transport pathways demonstrated the importance of a better understanding of processes regulating bioaerosols transport and deposition in the atmosphere. Direct and indirect anthropogenic forcing of the atmosphere will potentially impact the emission and deposition flux, atmospheric burden, biodiversity, and biophysical and metabolic activity of microorganisms in the atmosphere (Šantl-Temkiv et al., 2022).

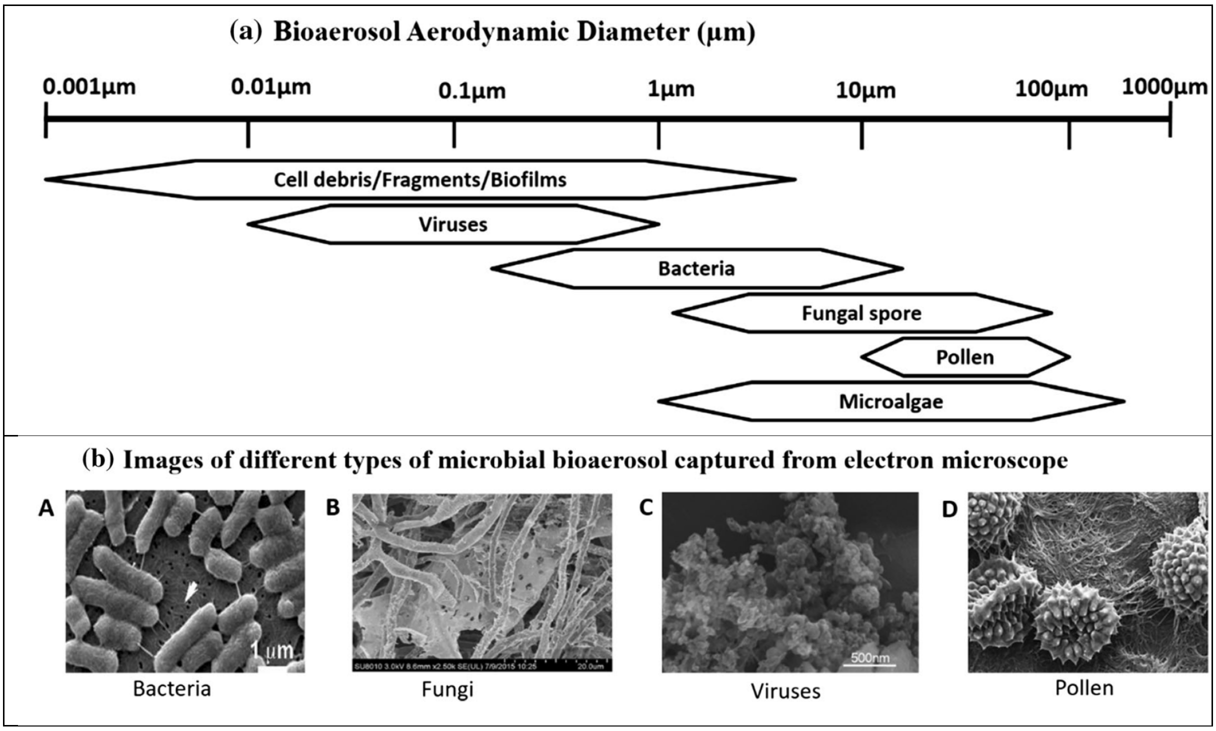


Figure 2.3.1 - Major bioaerosol components present in the atmosphere. A) bioaerosol aerodynamic diameter; b) images of different types of bioaerosol microorganisms view from an electron microscope.

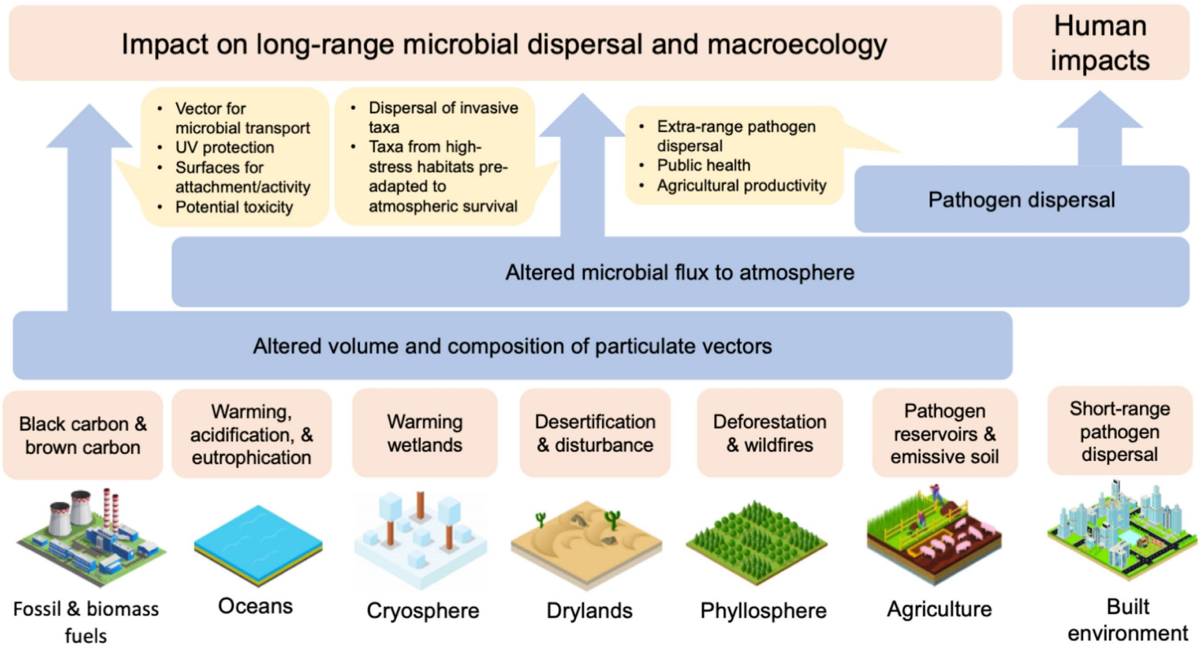


Figure 2.3.2 - Potential anthropogenic impacts on atmospheric microbiology. Direct and indirect anthropogenic forcing of the atmosphere will potentially impact the emission and deposition flux, atmospheric burden, biodiversity, and biophysical and metabolic activity of microorganisms in the atmosphere (Šantl-Temkiv et al., 2022).

The number of studies that have investigated the properties and concentrations of bioaerosols in Amazonia is still relatively small (Artaxo et al., 2022, Martin et al., 2010; Whitehead et al., 2016; Andreae et al., 2015; Womack et al. 2015). In Amazonia, the aerosol coarse mode, which comprises most of the PBAP mass (i.e., fungal spores, pollen, fragments of a different kind), is comparatively constant throughout the year (Moran-Zuloaga et al. 2018), which suggests a weaker seasonality in the overall abundance of the coarse mode PBAP population as compared to the accumulation mode aerosol. Little is known about the bioaerosol composition. Until now, particle identification has been limited to a few microscopic and molecular genetic analyses, which identified fungal spores as a predominant fraction of primary biological particulate mass. Souza et al. (2019) conducted a metagenomic DNA extraction and analysis on Amazonian aerosol samples. They revealed that fungi and bacteria are a significant fraction of the organisms present in Amazonian bioaerosol. These organisms are known to disperse via cells and/or spores through the atmosphere. Metagenomic DNA was extracted from air samples, and the prokaryotic diversity was determined by simplified 16S rRNA gene sequencing. During the wet season, 35 % of the coarse-mode air particulate matter was found to be fungal spores in the Amazonian tropical rainforest region. Huffman et al. (2012) suggest that fungi and/or fungal spores tend to dominate the bioparticle population.

The discipline of aeromicrobiology has benefitted enormously from recent methodological advances that overcome the limitations associated with studying ultra-low biomass habitats. We view this as beneficial in a broader sense to an improved ecological understanding of how the atmosphere links to surface terrestrial and marine biomes. Specifically, the ability to accurately estimate microbial diversity in the air has revealed that the atmospheric boundary layer supports a highly diverse but nonrandomly assembled microbiota. This is especially true in tropical forests such as Amazonia.

2.4 – Metagenomics analysis of bioaerosol particles in Amazonia

With the advance in sequencing techniques, it is possible today to perform a metagenomics analysis of viruses and bacteria in airborne aerosol particles (Archer et al., 2023, Hall et al., 2013, Fröhlich-Nowoisky, 2016, Yoo et al., 2017, Santos junior et al., 2016). Bioaerosols are a diverse and complex dispersed fraction of organic aerosols originating from different biological materials such as bacteria, fungi, viruses, debris, or by-products of biological activities, metabolites, peptidoglycans, endotoxins, mycotoxins, biopolymers, and micro molecules (Després et al., 2012, Ghimire et al., 2019). A few studies in this emerging area have identified diverse groups of microorganisms in atmospheric aerosols after sampling, DNA extraction, DNA amplification, sequencing, and bioinformatics analysis (Mota de Oliveira et al., 2022). Those studies have identified various bacteria, actinomycetes, and fungi, providing considerable information concerning the presence of possible pathogens and allergens in the atmosphere and providing a better understanding of the links between airborne bioaerosols and biodiversity. Bioaerosols can be measured using real-time techniques (Ghimire et a., 2019) or analyzed from material collected in aerosol filters (Prass et al., 2021). Bioaerosols can be found over a large particle size range from 10 nanometers to 100 micrometers (Moran-Zuloaga et al., 2018). This very large range makes collecting and analyzing a broad range of bioaerosols difficult.

Fluorescence detection methods based on microbe-induced fluorescence are used to constantly observe viable bioaerosols, as we plan to do in this project. Ultraviolet-Laser-Induced Fluorescence/Wideband Integrated Bioaerosol Sensor (UV-LIF-WIBS) is a common method for directly detecting bioaerosols (Pöhlker et al., 2012).

The bioaerosol diversity at the ATTO site was studied in relation to overall atmospheric conditions, such as the contrast between the clean wet season background and the day versus night contrast, as well as different heights above the forest canopy (Prass et al., 2021). Prass used fluorescence in situ hybridization (FISH), a molecular biological staining technique. They found that wet season bioaerosol number concentrations in the range of 1–5 × 105 m−3 account for > 70 % of the coarse mode aerosol. Eukaryotic and bacterial particles predominated, with fractions of ∼ 56 % and ∼ 26 % of the intact airborne cells. Archaea occurred at very low concentrations. Vertical profiles exhibit a steep decrease in bioaerosol numbers from the understory to 325 m height on the Amazon Tall Tower Observatory (ATTO), with a stronger decrease in Eukarya than Bacteria.

Aerosol filter samples were collected, and their DNA was extracted for shotgun sequencing analysis. The results show that the main influencing factor over the air microbiome in the Amazon rainforest is the altitude (Fig. 1). Samples collected at the height of 325 m were separately clustered from the rest of the samples at 42 m. This result suggests that the distance to the sources substantially influences the microbial diversity of airborne particles (Ghimire et al., 2019, Womack et al., 2010).

Analyzing the microbiome of the upper troposphere (8-15 Km altitude), DeLeon-Rodrigues (2013) shows that airborne microorganisms play an important role in cloud formation and precipitation. Quantitative PCR and microscopy revealed that viable bacterial cells represented, on average, around 20% of the total particles in the 0.25- to 1-µm diameter range and were at least an order of magnitude more abundant than fungal cells, suggesting that bacteria represent an important and underestimated fraction of micrometer-sized atmospheric aerosols. These bioparticles are efficient ice nuclei and are important in forming and developing deep ice clouds. They conclude that the microbiome is a dynamic and underappreciated aspect of the upper troposphere, potentially impacting the hydrological cycle, clouds, and climate.

One study of sequencing bioaerosols at the ATTO tower for a few filters shows that Eukaryota largely dominates the bioaerosol population, followed by Bacteria, Archaea, and viruses (Barbosa et al., 2022). The taxonomic analysis at the phylum level shows a large variety of species, with Streptophyta, as the most abundant phylum having more than 80%, followed by the bacterial group of Proteobacteria and the phylum Chordata. According to this result, most of the families correspond to plant sequences. Few groups of bacteria and fungi appear among the most abundant classification families (Sánchez Parra et al., 2020). Amato et al., 2019 have explored the analysis of the metatranscriptomic exploration of microbial functioning in clouds. Archer et al., 2023, have discussed a global biogeography of atmospheric microorganisms that reflects diverse recruitment and environmental filtering. They show that atmospheric transport is critical to the dispersal of microorganisms between habitats, underpinning resilience in terrestrial ecosystems globally (Archer et al., 2023, Santos Junior et al., 2016).

The work of Souza et al. (2019) used a very simplified metagenomic DNA method using 16S rRNA gene sequencing. They combined environmental metagenomic extraction, 16S rRNA gene sequencing, and bioinformatics are helping to uncover prokaryotic environmental communities. In this work, most of the DNA was unclassified, and for Bacteria, figure 2.4.2 shows the OTU clusters subjected to taxonomic classification using the Silva 16S database v.132.

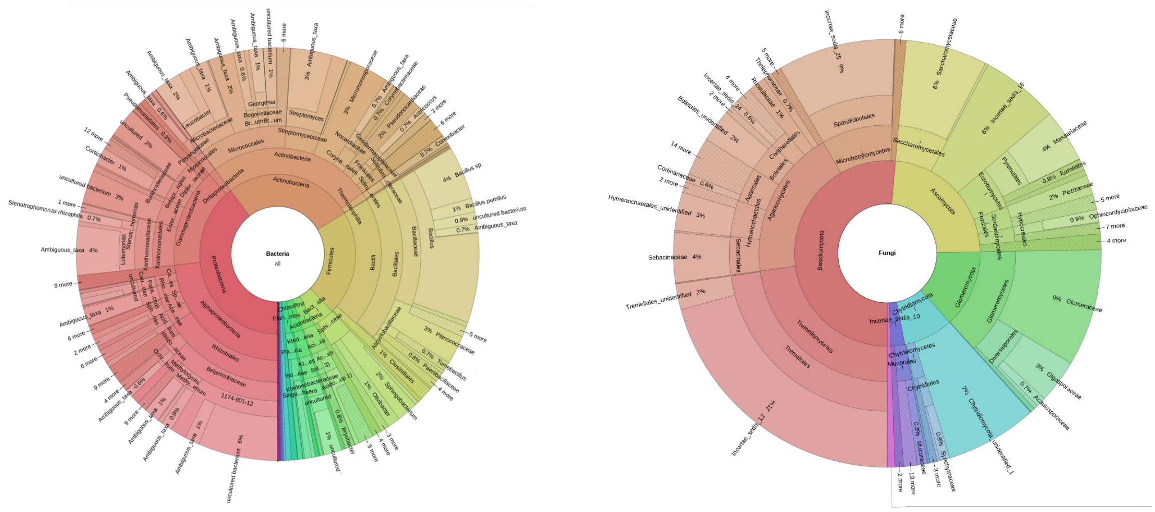


Figure 2.4.2 - Composition of the prokaryotic community (left) and classification of the reads using the 18S SILVA database (right) for bioaerosols from Amazonia. From Souza et al. (2019).

2.5 – The Amazonian aerosol-cloud-precipitation interactions

The hydrological cycle is critically important for the Amazonian ecosystem since many processes that regulate the functioning of Amazonia depend on precipitation, including the carbon cycle (Brando et al., 2019, Artaxo et al., 2022). The Amazon is one of the few continental regions where anthropogenic sources do not dominate atmospheric aerosol particles and their effects on climate. One of the links between the aerosol and cloud components happens via the Cloud Condensation Nuclei (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 et al., 2016, Artaxo et al., 1994). These particles are mainly natural primary biogenic particles (coarse mode) as well as SOA produced from the oxidation of naturally emitted VOCs (fine mode).

The sources of CCN in Amazonia are not yet fully understood (Artaxo et al., 2011). The Acridicon-Chuva experiment found that new particle formation could happen unexpectedly in high altitudes (10-14 Km), and it could be a relevant source of CCN in Amazonia (Andreae et al., 2015). Another important source of particles is in the canopy forest itself, produced from the oxidation of VOCs, especially isoprene and monoterpenes.

Among natural sources, most studies on the impact of aerosols on precipitation focus on the modal cloud condensation nuclei (CCN) size region, usually for particles larger than 0.1 micrometers (Cecchini et al., 2016, 2017), and until recently, little attention has been given to the two extremes of sizes: ultrafine particles (UFP – ultrafine particles) and the giant nuclei of CCNs, which are smaller than 50 nm and larger than 10 μm, respectively. Recent studies show that UFP rapidly grows to CCN sizes both in regions with anthropogenic influence and in remote regions of the Amazon (Franco et al., 2022; Zaveri et al., 2022; Zhao et al., 2022) and alter the formation of shallow convective clouds, suppress precipitation, and can enhance the transition to deep convective clouds (Fan et al., 2018). Figure 2.5.1 illustrate this mechanism. However, Fan et al. (2018) show observational data and numerical simulations of deep convective clouds over the Amazon where UFP are ingested and activated to form additional cloud droplets in which excess supersaturation condenses and forms additional cloud water and latent heating, invigorating convection. The giant CCNs, which in the Amazon are normally associated with emissions from biogenic sources (Pöschl et al., 2010; Martin et al., 2010; Barbosa et al., 2022), accelerate the collision and coalescence process, resulting in shallow clouds with a predominant warm phase and rapid precipitation formation (Martin et al., 2017).

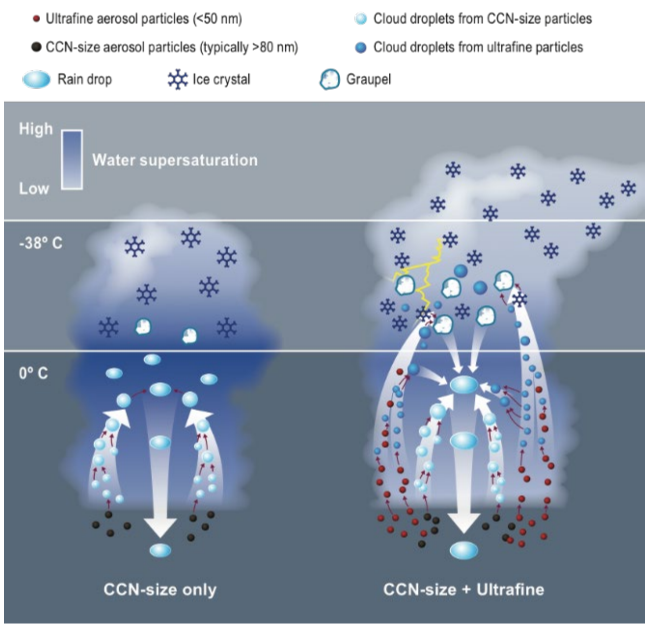


Figure 2.5.1 – The role of ultrafine aerosol particles enhancing cloud droplets formation. In clouds that lack UAP<50 (left), the clouds are highly supersaturated as a result of fast drop coalescence that forms warm rain and reduces the integrated droplet surface area available for condensation. With added UAP<50 (right, red dots), an additional number of cloud droplets are nucleated above the cloud base, which lowers supersaturation drastically by enhanced condensation, releasing additional latent heat at low and middle levels, thus intensifying convection (Fan et al., 2018).

Considering these recent findings on the effects of the two far-end modes of CCN size distribution in the clouds, we propose to study how UFP and giant CCNs influence cloud precipitation and raindrop size distribution at ATTO. We will leverage the extensive set of precipitation and aerosol measurements at the ATTO-Campina site and ATTO towers, with the addition of data analysis from disdrometers at the ATTO Campina site.

We will investigate the "aerosol invigoration" hypothesis in low and high UFP scenarios. The investigation will combine the radar measurements from the cloud radar MIRA-35C, Radar Wind Profiler (RWP), and XPOL with the aerosol, CCN, and DSD measurements. Lightning observations from satellite and ground-based location systems will also be used as they can also provide a proxy for updraft strength. Combining MIRA-35C and RWP datasets into a unified cloud mask of precipitating clouds to provide cloud-type profile classification will aid the investigation of this hypothesis. Moreover, the RWP provides an estimate of convective cloud vertical velocity and mass flux profiles that cannot only tell the strength of the updrafts and downdrafts of storms but also assist in the estimates of UFP and trace gases transports to the surface.

2.6 - 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, especially in tropical forests (IPCC AR5 2021, Artaxo et al., 2022, Wang et al., 2016). 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 determine the aerosol radiative forcing. The radiative impacts of organic aerosols (OA) are a large source of uncertainty in estimating the global direct radiative effect (DRE) of aerosols (IPCC, 2021). These impacts include light scattering and absorption from a subclass of OA called brown carbon (BrC). Several papers have observed strong absorption properties of natural biogenic aerosols (Morais et al., 2022). Because of the intricate shapes and composition, biogenic particles can absorb significant amounts of visible light. The surface radiative forcing of aerosol particles has important effects on the ecosystem, including reducing total radiation fluxes and increasing the diffuse to direct radiation ratio. These changes directly affect plant photosynthetic rate (Cirino et al., 2014), especially in tropical forests, where enhanced photosynthesis reached 30-40 % of Net Ecosystem Exchange (NEE) for the ZF2 site, Santarém, and Rondônia. Figure 2.6.1 shows the long time series (2000-2023) of aerosol optical depth (AOD) being made continuously by our group at five sites in Amazonia, including the ATTO tower and Manaus. It shows a strong seasonality of aerosol particles in Amazonia resulting from biomass burning emissions. Also, we can see the large year-to-year variability. These high AODs strongly impact carbon uptake due to the increase in diffuse radiation.

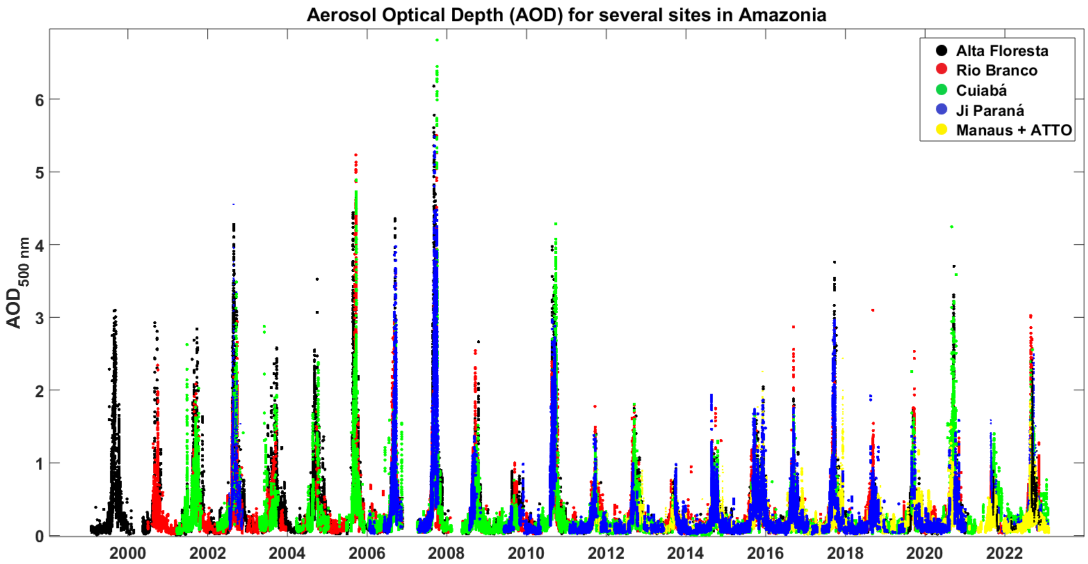


Figure 2.6.1 - Long time series (2000-2023) of aerosol optical depth (AOD) being made continuously by our group at five sites in Amazonia, including the ATTO tower and Manaus. These AERONET measurements are being made in collaboration with NASA Goddard.

These 23 years of continuous measurements under challenging conditions in Amazonia show much information in addition to AOD, such as aerosol size distribution from Almucantar inversions, refractive index, single scattering albedo, and scattering and absorption Ångström exponent, among other fundamental aerosol properties. The recent work of Morais et al., 2022, summarizes the latest findings. We plan to continue these measurements as part of this proposal and the effort to have aerosol information basin wide.

2.7 – Properties of aerosols at high altitudes as measured in the CAFÉ-Brazil experiment

From some previous experiments, we learned about the possibility of aerosol particles in Amazonia being formed in the high altitude (>12 Km) Upper Troposphere (UT) region (Andreae et al., 2018, Wang et al., 2016). The production of particles in the UT may be a vital 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 convection characteristics, thereby influencing cloud radiative forcing and atmospheric dynamics (Cecchini et al., 2017). These mechanisms can be illustrated in Figure 2.7.1, which shows a conceptual framework that was tested with measurements from the CAFÉ-Brazil experiment, which is undergoing data analysis as part of this proposal.

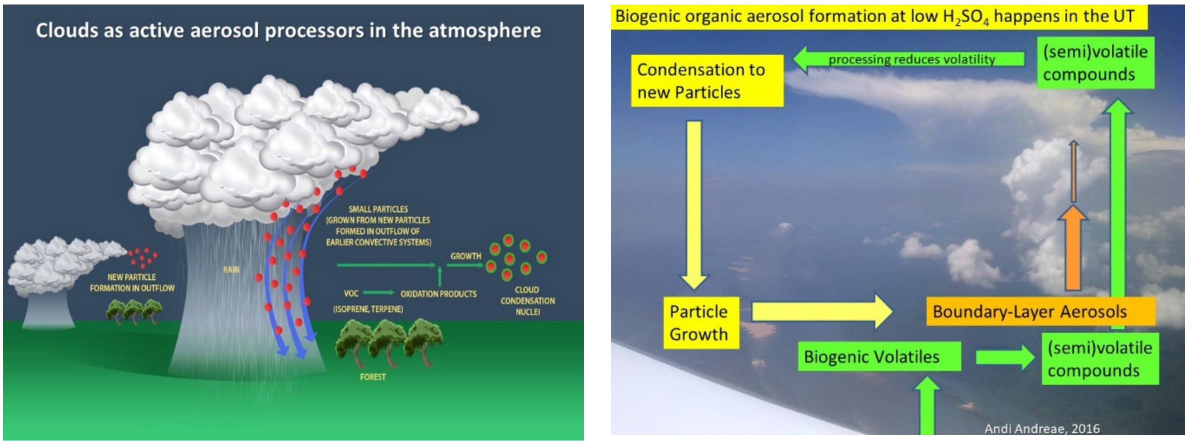


Figure 2.7.1 - On the left panel, we can observe how clouds can be active aerosol processors in the atmosphere of Amazonia. The right panel shows how semi-volatile compounds produced by the vegetation can be transported to the upper atmosphere by convection, where it condenses due to low temperature and is processed and oxidized to less volatile compounds and produce particles that are brought to the lower atmosphere by precipitation (Andreae et al., 2018).

In December 2022 and January 2023, we carried out the experimental CAFÉ-Brazil (*Chemistry of the Atmosphere Field Experiment – Brazil*) campaign. Very few large-scale measurements of gases and aerosols in the Amazon are made with aircraft, especially at high altitudes. The main objective of CAFE-Brazil is to understand the processes that could produce high concentrations of aerosols in the upper atmosphere, to study the photochemistry of tropospheric oxidants that form secondary aerosols in the Amazon rainforest in polluted and clean regions, to validate satellite estimates and numerical prediction models weather, and understand the cloud-aerosol-precipitation interaction. The CAFE-Brazil experiment employed the research aircraft (HALO – *High-Altitude Long-Range Observatory*). It was headquartered in Manaus and carried out flights over the forest throughout the Amazon. The flights investigated sources of high concentrations of nanoparticles, how emissions of volatile compounds from the forest influence oxidation chemistry, and how this process relates to the formation of aerosols and their abundance at high altitudes. The HALO measurements were coordinated with measurements at the Amazon Tall Tower Observatory (ATTO), located near Manaus, in a remote location within the forest. Some instruments performing measurements at ATTO were replicated at HALO. In this way, it will be possible to relate the measurements at the ATTO and directly above the forest with the surface aerosol formation processes and sources of volatile compounds and the oxidation and aerosol processes in the upper atmosphere.

2.8 – The FLOAT-Amazon, the ship research campaign

Measurements covered the atmosphere and terrestrial surface components during the previous FAPESP Thematic Project. The rivers were recently covered with a UEA research vessel campaign between January 10 and 21, 2023. This was an unprecedented experiment that was carried out on the Rio Negro, aboard a research vessel (Figure 2.8.1). This experiment took place simultaneously with CAFE-Brazil, characterizing the properties of the atmosphere in a preserved and little scientifically explored region. The cruise consisted of a round trip from Manaus to São Gabriel da Cachoeira, in the extreme northwest of the state of Amazonas, covering a distance of approximately 800 km each way. Air and water samples were collected in this virtually untouched region of the Brazilian Amazon. The experiment was carried out in partnership with the State University of Amazonas (UEA) and collaborators from the Max Planck Institute, the University of California-Irvine, and other researchers from IPEN and IAG-USP.

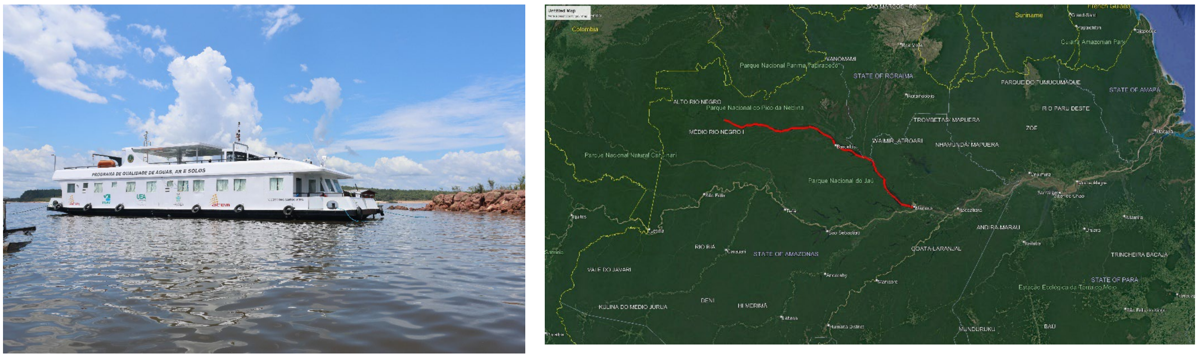


Figure 2.8.1 – Research vessel from the University of Amazonas State (UEA), and ship trajectory from Manaus to São Gabriel da Cachoeira in Western Amazonia, a region never explored from ground-based measurements.

Figure 2.8.2 – Instrumentation installed and operated in the UEA Research vessel for the FLOAT-CAFE-Brazil experiment.

The extensive instrumentation includes aerosol size distribution, aerosol chemical composition, scattering and absorption of radiation, total aerosol particle number, biogenic aerosol fluorescence, collection of biological particles for microscopy analysis, ozone, CO_2, CH_4, and carbon isotopes in CO_2 and CH_4, VOCs measurements using Proton Reaction Mass Spectrometer and many others. We also measured CO_2 and CH_4 in the river water and studied the transfer function to the atmosphere.

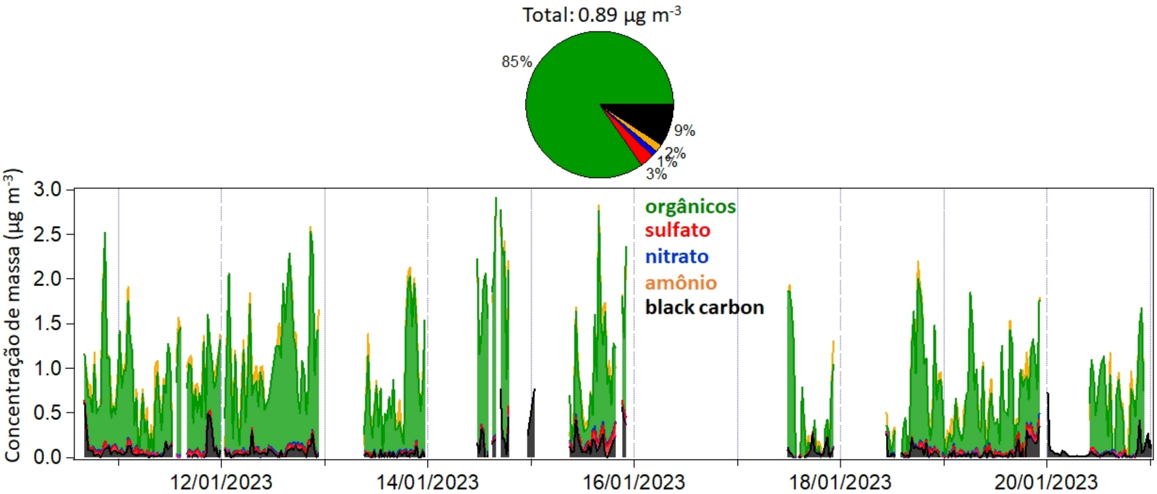


Figure 2.8.3 – Mass concentration of the main species that make up PM1, as measured in the FLOAT experiment: organics, sulfate, nitrate, ammonium, and BC. The values presented correspond only to the period in which the boat was in motion to avoid contamination.

This extensive data set will be analyzed as part of this proposal during 2023 and 2024. We plan to integrate the river measurements with the HALO aircraft and ATTO Tower measurements to have a comprehensive view of trace gases and aerosols in Amazonia.

2.9 Greenhouse gases (GHG) concentrations and fluxes at ATTO and basin wide

The carbon cycle in Amazonia is intrinsically linked to the climate of the region (Artaxo et al., 2023, Aragão et al., 2018), and several recent papers show that deforestation and forest degradation are changing the carbon allocation in Amazonia (Lapola et al., 2023, Gatti et al., 2021). The Amazon Forest stores about 150 Pg C in the soil and biomass, acting as a net carbon sink by removing, on average, 0.4 Pg C y^{-1} from the atmosphere (Malhi et al., 2015; Science Panel for the Amazon, 2021). About 17% of the planet's photosynthesis takes place in the Amazon. The carbon cycle in Amazonia is intrinsically linked with warm and humid conditions sustaining one of Earth's most productive natural ecosystems. The Amazon Forest interacts with the atmosphere in many different ways. Winds bring moisture from the tropical Atlantic Ocean into the Amazon, resulting in precipitation, which is partially recycled through evapotranspiration. Deep tree roots and the biological regulation of water flux through, resulting in a steady flow of water vapor into the atmosphere (Saleska et al., 2016). The recycling ratio in the Amazon Basin is in the range of 24% to 35% (Zemp et al., 2014). Part of the recycled water vapor precipitates over the rainforest and is transported to neighboring regions, making Amazonia a major water vapor source to other South American regions, especially regions with high food production in Central Brazil. There is a close link between the water and carbon cycles in Amazônia (Brando et al., 2019). The hydrological cycle in Amazonia is complex, controlled by water vapor fluxes, aerosol-cloud interaction, and atmospheric thermodynamics (Machado et al., 2018). Tropical forests have a critical role in supporting biodiversity, storing carbon, regulating the water cycle, influencing the radiation balance via albedo, and having an important role in human well-being.

When forest cover and structure change due to land use and climate change forcings, shifts in biophysical processes occur, affecting ecosystem services related to water, carbon, and energy balances (Hubau et al., 2020, Brienen et al., 2015, Aragão et al., 2018). There are several important links between carbon stored in the ecosystem and the science topics we will work on in this proposal. The first is a detailed study of how VOC emissions, nucleation of particles, CCN, and clouds impact precipitation and radiation balance. Light scattering by aerosols and clouds also increases the fraction of diffuse radiation, enhancing photosynthesis efficiency and carbon uptake by vegetation canopies (Cirino et al., 2014; Malavelle et al., 2019). This analysis shows that aerosols and biogenic emissions have an important role in carbon processing by the Amazonian ecosystem. The recent work of Lapola et al., 2023 shows the importance of forest degradation in addition to deforestation. They have simulated the Business-as-usual scenarios versus a Governance scenario when deforestation halts at 203. The result is Figure 2.9.1., where we can observe that carbon loss from the ecosystem varies significantly with the future scenario for Amazonia.

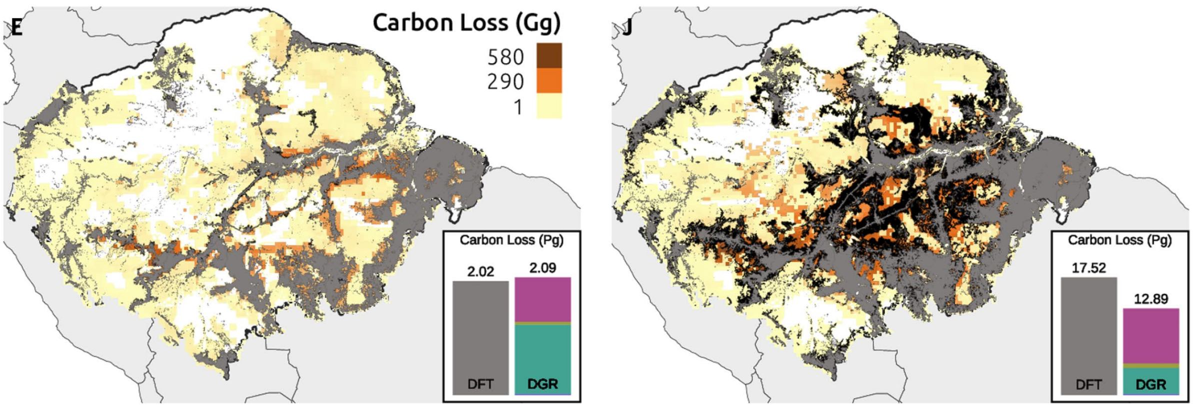


Figure 2.9.1 - First-order 2019-2050 projections of carbon losses from Amazon Forest degradation through its main drivers. The left map shows the governance scenario, and the right map the business-as-usual scenario. From Lapola et al., 2023.

The 325 meters ATTO tower is an ideal platform for greenhouse gas measurements, representing a significant footprint for emissions and sinks (Botia et al., 2020, 2021). Concentrations of CH_4, CO_2, and N_2O are being measured, as well as carbon isotopic composition, to help constrain the measured concentrations. The CAFÉ-Brazil flights have done the very first large-scale measurements of high-precision CH_4 and N_2O measurements basin-wide. We will also perform GHG measurements at the boat campaigns, including isotopic composition, to estimate CH_4 emissions from the flooded areas in Amazonia. Satellite measurements using AIRS, OCO2, GOSAT, TROPOMI, and the new platforms launched over the next two years will provide a significant basis for GHG in Amazonia. This unique large data set, coupled with a modeling component with the WRF-GHG regional modeling, will provide new insights into GHG cycling over Amazonia.

2.10 - Modeling the aerosol-clouds-trace gases and transport over Amazonia.

There are several efforts of observation and modeling to improve the knowledge of the different processes that evolved in the biogeochemical and physical cycles between the Earth's surface and atmosphere. Many of these efforts aim to produce new datasets to validate models, remote sensing observation, and a better understanding of processes. Unfortunately, these important efforts do not have any integrated observation-modeling sites in the tropical region. The tropics, mainly the continental tropics, such as Amazonia, have an important impact on the Earth's climate system, but the biogeochemical and physical cycles have critically important unknown processes.

Therefore, this proposal presents a virtual-modeling laboratory to explore physics-chemistry-biological processes occurring in Amazonas. These include aerosol-cloud precipitation interactions, carbon cycling, and long-range transport, among many other topics. Long-term observations at ATTO and the LBA towers and intensive campaigns such as ACRIDICON-CHUVA and CAFÉ-Brazil collect critically important information to feed several models for a better understanding of processes in Amazonia.

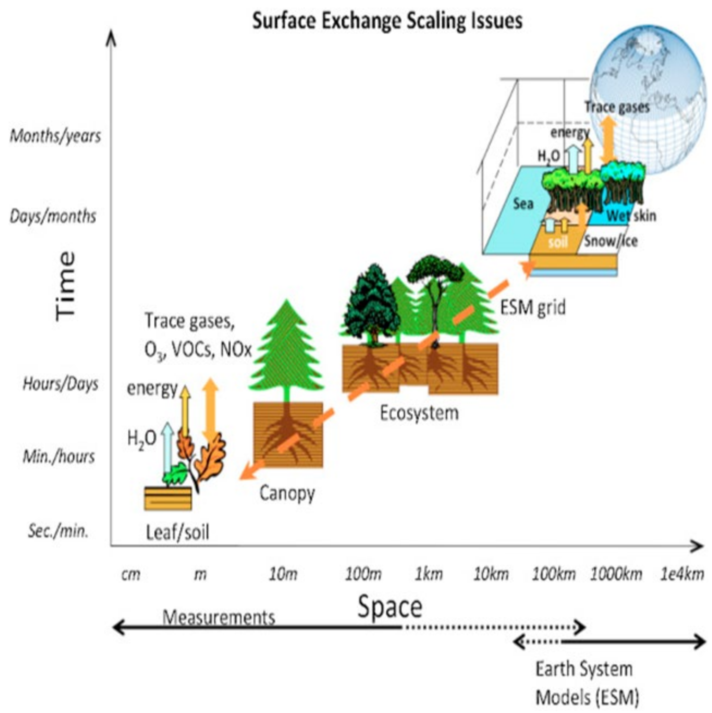


Figure 2.10.1 – Schematic representation of the time-space scales of models that will be used in this proposal. From LES (Large Eddy Simulation) models going through regional models like WRF-CHEM-GHG going up to Global Earth System models such as NCAR CESM2.

WRF-Chem is the Weather Research and Forecasting (WRF) model coupled with Chemistry. The development of WRF-Chem is a collaborative effort among the community. The WRF Model is a state-of-the-art mesoscale numerical weather prediction system designed for atmospheric research and operational forecasting applications. It features two dynamical cores, a data assimilation system, and a software architecture supporting parallel computation and system extensibility. The model serves various meteorological applications across scales from tens of meters to thousands of kilometers. The Greenhouse Gas model (WRF-GHG) was developed to allow for passive tracer transport simulations of CO_2, CH_4, and CO. Several flux models and emission inventories are used for an estimation of the emission and consumption fluxes of CO_2, CH_4, and CO. These flux models are either coupled online, i.e., their code is implemented into the WRF-GHG code or is operated as preprocessors for WRF-GHG to calculate fluxes for the different source and sink processes for CO_2, CH_4, and CO.

The CESM2 (Community Earth System Model 2) is an open-source community coupled model consisting of ocean, atmosphere, land, sea-ice, land-ice, river, and wave models that exchange states and fluxes via a coupler. It contains many substantial science and infrastructure improvements and capabilities since its previous version, the CESM. The CESM2 (Danabasoglu et al., 2020) was developed for participation in CMIP6 (Eyring et al., 2016).

The modeling component and the buildup of the Amazonian-virtual-laboratory will help to work on several key questions in this proposal, such as

1) Bioaerosol regional and long-distance transport
2) New particle Formation in the Upper troposphere and its transport through convection and downdrafts associated with clouds.
3) New particle formation on the surface from VOCs, catalyzed by NOx, O3, and OH.
4) The transport of gases to the upper troposphere by convective processes.
5) Aerosol and shallow cloud formation from VOCs precursors, followed by CCN formation.
6) Convective invigoration
7) Shallow to deep convective cloud transitions
8) Deforestation and carbon cycle, through WRF-GHG simulations and maps with the distribution of carbon density in Amazonia.

This modeling component will be done through collaborations with FAPESP Thematic project 2022/07974-0 coordinated by Luiz Augusto Toledo Machado in partnership with Chinese researchers. Additionally, joint efforts with the Max Planck Institute for Chemistry in Mainz, Jena, Germany, on the CAFÉ-Brazil modeling efforts and Prof. Jordi-Villa in the Wageningen University with the Cloud-Roots experiment will complement these efforts.

3 – Current scientific gaps and issues to be addressed

Although there were important and large scientific experiments in Amazonia over the last three decades studying atmospheric properties, the system is very complex, and we have major knowledge gaps. The interactions between forest biology, atmospheric properties, and climate are strong but still far from being understood. We will describe below the main scientific gaps we observe in Amazonia for several project components.

3.1 – Aerosol Life Cycle in Amazonia

Aerosol properties, life cycle, and aging in Amazonia: From primary biogenic particle emissions to secondary aerosol production, we still do not fully understand the role of aerosols in cloud formation, in the radiation balance and the biogeochemistry of nutrients at the basin.

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. Current aerosol models require improvement in several areas: emissions, mechanisms, new particle formation events, and aerosol physical-chemical changes under biogenic and anthropogenic influences. The research that this proposal will conduct is usefully distinguished into four topical areas or elements: nucleation and new particle formation, aerosol growth and aging, the direct radiative impacts of aerosol, and separating the natural vs. anthropogenic aerosol influences on aerosol properties. ***In which conditions do nucleation and new particle formation happen in Amazonia?***

The aging of aerosols consists of modifying the composition, size, and surface properties of aerosol particles in the atmosphere by coagulation, condensation, and surface reactions. These processes are essential as they affect the aerosol's optical and cloud nucleating properties. The dependence of SOA formation on sulfur and nitrogen oxides and other factors will be examined in this proposal. This information is essential to developing comprehensive chemical mechanisms that can contribute to a better understanding of the remote aerosol life cycle. ***How do 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. Aerosol optical properties depend strongly on particle size, composition, mixing state, and morphology, all properties that will be measured in this study. Aerosols containing black and "brown" carbon absorb radiation in the visible spectral region and heat the environment. Scattering organic aerosols have a cooling effect on the surface. Measurements at the ATTO tower and the 7 NASA/AERONET and SunRadNet sites in Amazonia will help to understand the processes that control the effects of aerosols on the radiation balance in Amazonia. ***What is the large-scale impact of aerosol radiative forcing on 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. 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 the ecosystem and atmospheric chemistry in Amazonia?***

The bioaerosol component can consist of living organisms that can be transported long-range, and we still need to quantify the potential of bioaerosol to influence biodiversity over large time scales. ***How does metagenomics analysis provide information on living species in the bioaerosol?***

3.2 - Cloud life cycle in Amazonia

Atmospheric dynamics: Convection and advection air motions are central to cloud life cycles. Whereas emissions and nucleation lead to new aerosol particles, it is primarily atmospheric dynamical conditions in a sufficiently humid environment that led to new cloud droplets. 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 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 needs to be better understood, and we will use a modeling approach to better understand processes that regulate entrainment in shallow clouds in Amazonia. ***How do boundary layer dynamics, radiation, cloud formation, and composition affect the cloud life cycle?***

Cloud microphysics: Accurate knowledge of the hydrometeor number, size, surface area, volume or mass, dispersion, skewness, and phase are required 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 still need to be better 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. 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 such as Amazonia. The light-absorbing properties of aerosols through BC and BrC can strongly influence cloud dynamics through heating. On the other side, recent studies show that clouds are active in processing aerosol particles, changing their hygroscopicity, size, and properties, but the mechanisms are unclear. ***How does cloud processing of aerosols change their physicochemical properties*?**

3.3 – Greenhouse gases budgets in Amazonia

The Amazon is an important source of greenhouse gases to the global atmosphere through deforestation and forest degradation, and through photosynthesis can be an important sink. Measurements of CH_4 and CO_2 at ATTO over the last few years have shown interesting and not fully understood dynamics (Botia et al., 2020, 2021). Isotopic measurements have shown variabilities that are difficult to attribute. Through large-scale measurements along the main rivers and CH_4 and N_2O during the CAFÉ-Brazil, coupled with a carefully designed modeling component, it could be possible to understand greenhouse gas emissions over the Amazon better. ***How are Amazonia's CO_2 and CH_4 emissions and sinks balanced overall?***

3.4 – 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. 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. The Central Amazonia region is a natural laboratory to observe the characteristics of the tropical continental convective life cycle and model cloud-aerosol precipitation interactions and the role of land surface processes. ***How do aerosol-cloud interactions influence precipitation in pristine conditions in Central Amazonia?***

4 - Objectives of the proposed work

As discussed in the previous topics, this project aims to study trace gases, aerosols, and clouds lifecycles and their impact on the Amazonian ecosystem using long-term measurements at ATTO, river ships, analysis of the high-altitude HALO airplane, and the modeling component. The **key scientific questions** to be answered by this project can be clustered in six main topics:

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

1a. What are the characteristics of BVOCs emission from vegetation, and how do they vary along the basin, with seasons and climate conditions?

1b. How do different VOCs and environmental conditions affect aerosol particle production?

Hypothesis: The emission of VOCs by the forest vegetation is critical to aerosols and cloud properties over Amazonia. We need long-term and extensive spatial coverage of key VOCs, such as isoprene over the Amazon region. The particle production mechanisms, both at the surface and high altitudes, still need to be fully understood. It would be important to have the seasonal and diurnal variability of isoprene and organic aerosol measurements to understand the VOCs-aerosol production mechanisms fully.

Methodology: We will help perform long-term isoprene measurements at the ATTO tower and isoprene basin-wide in the ship cruises to understand better the VOCs distribution and time variability over the basin. We will run two Aerodyne Aerosol Chemical Speciation Monitors (ACSM) in parallel with PTR-MS VOCs measurements at the top of the 325 ATTO tower and in the ground to better understand the mechanisms of these complex links. The data analysis of measurements from the HALO plane at the CAFÉ-Brazil experiment will allow studying these processes at high altitudes (up to 14 Km).

2. Secondary Organic Aerosol (SOA) formation: Interactions of biogenic and anthropogenic emissions

2a. What are the chemical and physical processes that control and affect the production of SOA in Amazonia?

2b. How are the mechanisms of organic particles production (e.g., nucleation and SOA formation)?

2c. What are the potential roles of primary biological particles (fungal spores, bacteria, and leaf cuticles) as giant cloud condensation nuclei (GCCN)?

2d. How new particle formation occurs at high altitudes (>12 Km) and at the ground?

Hypothesis: We already know that SOA is responsible for about 70-80 % of accumulation mode aerosol mass over Amazonia. But we do not fully understand the evolution and particle dynamics of SOA. From new particle formation to the bursts of 20-40 nanometer particles commonly observed in Amazonia, we have to find the mechanisms behind SOA and CCN formation over Amazonia. The role of chemical pathways and atmospheric transport still must be unveiled.

Methodology: We will help to perform long-term measurements of SOA and CCN at the ATTO tower, as well as in the analysis of the CAFÉ-Brazil rich dataset. The chemical characterization of CCN active particles and their hygroscopicity and size allows a detailed analysis of their formation pathways. The particle nucleation package with CIMS (Chemical Ionization Mass Spectrometry) coupled with NAIS (Neutral Cluster and Air Ion Spectrometer), particle magnifier, and nano SMPS that allows the detection of cluster ions and nanoparticles above 2 nanometers will help to unveil these mechanisms.

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

3a. 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?

3b. How do primary and secondary organic aerosols influence cloud condensation nuclei (CCN) activity?

Hypothesis: Aerosol particle size distribution dynamics must be better understood, especially the links with optical properties. Light scattering by organic aerosols and light absorption by black carbon is critical to the variability of single scattering albedo (SSA) that controls radiative forcing. Light absorption by brown carbon (BrC) particles can be significant in Amazônia. The optical properties of super micrometer particles associated with natural primary biogenic particles and Sahara dust transport need a better understanding. Deposition of phosphorus from African transport can be important for the nutrient cycle over Amazonia.

Methodology: We will do aerosol size distribution coupled with aerosol optical properties at several levels of the ATTO tower and at the RoLi continuous elevator along the vertical profile. Spectral dependence of aerosol absorption will be measured, including UV and visible wavelengths, to determine BrC concentrations. The

NASA/AERONET network will also measure BrC, aerosol radiative forcing, scattering, and absorption.

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

4a What are the microphysical properties of the clouds, and how to improve cloud processes parameterization for shallow and deep convective clouds in the Amazonas region?

4b What are the physical parameters controlling the Amazonas cloud organization and cloud life cycle?

4c How do aerosol thermodynamics and dynamics control the Amazon region's cloud life cycles and rainfall?

4d What physical processes control the transition between shallow and deep convection in the Amazon region,

Hypothesis: Shallow and deep-convective clouds have different parametrization, and this is one of the main issues in the representation of cloud organization, development, and rainfall, and not well represented in numerical models. In addition, models do not consider aerosol size distribution, which could strongly impact the Amazonian clouds, one environment with high specific humidity. Aerosol concentration is very important during the wet season; however, during the dry season, the high aerosol concentration has a smaller impact as the capability of the clouds to produce more droplets is limited. Cloud organization depends on accurately representing gravity waves in atmospheric models to produce cloud clusters.

Methodology: The combination of 3D dual polarimetric radars and vertical pointing radars provides hydrometeor classification and vertical velocities, with ceilometers, lidars, and cloud radars to provide cloud description, and boundary layer evolution would help to characterize the cloud processes. This information, combined with large eddy simulation and cloud-resolving models, will allow us to test the different hypotheses. The SMPS from Campina and ATTO sites will provide the particle size distribution to test in the models and constrain the simulations.

5 – Primary biogenic aerosol particles and metagenomics identification

5a What is the role of primary biogenic aerosol particles in the radiation balance and giant CCN population?

5b What is the distribution between different living organisms (Eukaryota, bacteria, archaea, virus, etc.) on aerosol in the Amazonian atmosphere?

5c What are different species' seasonal and diurnal variability and spatial distribution on atmospheric bioaerosols?

5d Does metagenomics of aerosol particles able to identify species in Amazonian aerosols?

Hypothesis: Primary biogenic aerosol particles are responsible for 80% of the coarse-mode aerosol mass, so they should play a significant role in trace element deposition, especially phosphorus, nitrogen, and potassium. The advanced metagenomic analysis will allow species identification for long-range transported bioaerosols within the basin and along the African-South America route.

Methodology: We will use a novel biogenic aerosol particle detector using Fluorescence to measure living organisms in aerosol particles over the ATTO tower and on a large scale using the ship cruises. We will perform a detailed metagenomic analysis of aerosol particles to identify at the specie level the presence of microorganisms, algae, fungi, and viruses.

6. The Large-scale greenhouse gases distribution, emissions, and sinks.

6a What is the basin wide CO_2 and CH_4 concentrations and fluxes?

6b Does the joint combination of measurements at the ATTO and LBA towers, river systems, and modeling provide a large-scale picture of GHG over Amazonia?

6c Do regional and global models correctly describe the carbon cycle balance among the different processes?

Hypothesis: The hydrological cycle and the carbon cycle are closely linked in tropical forests. Water and radiation are critical ingredients for photosynthesis. GHG measurements by the ATTO and LBA towers agree with satellite retrievals. However, local processes in the dense forest, varzea, river, and deforestations regions have a completely different carbon contribution from the different players that are well described by the large eddy simulations. Radiation, cloud, and GHG, locally, have a strong interaction. All these specific processes related to stomates aperture, radiation, temperature, and GHG concentration can be well parametrized using the instrumentation, including isotopic measurements.

Methodology: We will help perform continuous greenhouse gas measurements at several levels of the ATTO tower, including isotopic composition. We will perform CO2 and CH4 isotopes on the ship cruises. We will also do extensive remote sensing of GHG using AIRS, OCO2, GOSAT, TROPOMI, and the new platforms launched over the next few years. The first-time measurements of large-scale CH4 and N2O performed over Amazonia during CAFÉ-Brazil is a great opportunity to study the links between land use change and greenhouse gas emissions and sinks. The coupling with regional and global Earth System models will help to understand processes critically important for greenhouse gas cycling in Amazonia. Simulations using Large Eddy Simulation (LES) models with the different carbon cycle components will be evaluated in detail and contained by the observations.

We propose to use long-term observations from the ATTO tower, cruises along key rivers in boat measurements in Amazonia, and analysis of high-altitude aerosol and trace gas measurements during CAFÉ-Brazil, coupled with a modeling component, to answer these critical questions.

5 – Implementation strategy

We are proposing a complex set of measurements with different strategies involving long-term ATTO tower and CAMPINA/ATTO measurements, riverboats, and analysis of large-scale aircraft measurements. Each sampling site will use a coherent set of instruments to characterize aerosol particles, CCN, clouds, and trace gases. This section describes the measurement strategy to achieve our project goals.

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

A unique laboratory, the ATTO tower site is located in one of the world's most pristine sites in continental areas, with coordinates at S 02° 8' 38.8", W 58° 59' 59.5" (Andreae et al., 2015). A German-Brazilian long-term scientific cooperation is operating a 325 meters tall tower at this site. INPA operates the site – The Brazilian Institute for Research in Amazonia and has support from the Max Planck Institute, UEA (Universidade Estadual do Amazonas), and many other partners, including USP and INPE. Figure 5.1.1 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 on a long-term basis (>20 years). The pristine condition of this site makes it perfect for background aerosol characterization. The full set of measurements can be found in Andreae et al., 2015.

Vertical profiles of trace gases and aerosols are being measured along the tower. A recently installed elevator (RoLi system) allows continuous measurements along the whole tower through an elevator carrying about 200 Kg of equipment. Detailed physicochemical properties are being measured along the vertical, aiming to analyze the role of the forest canopy emissions at low levels and how it influences atmospheric composition up to 325 meters.

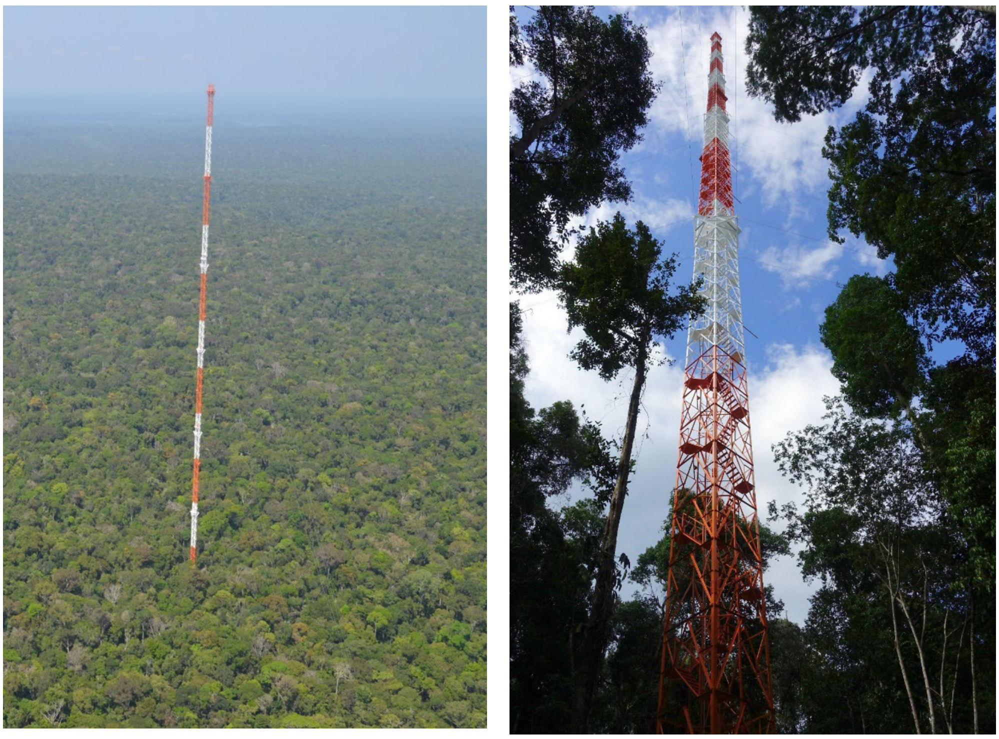
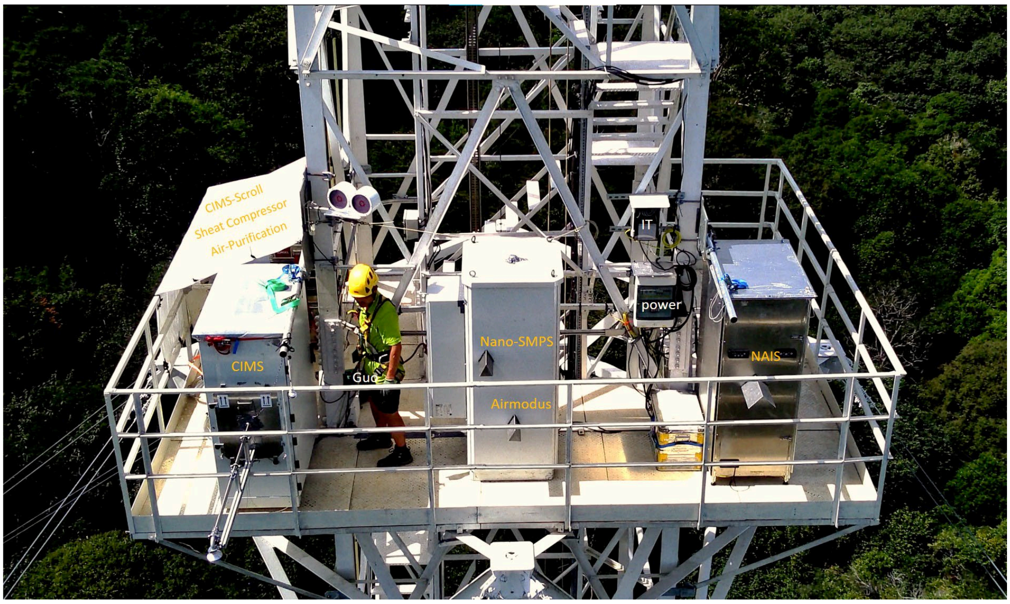


Figure 5.1.1 – The ATTO 325 meters tall tower in Central Amazonia. Aerosols and trace gases are being measured continuously at the top of the tower and profiles for trace gases and particles.

Recently it was installed at ATTO, a platform at 80 meters high, dedicated to the stud of processes that could occur close to the canopy, such as VOC emissions and aerosol nucleation. The picture shows the operation of nanoparticle measurements such as Nano-SPS, NAIS, CIMS, Airmodus particle growth system, and others. A picture of this platform is shown in Figure 5.1.2 below.



5.2- The Campina-ATTO remote sensing site

The ATTO site is in a closed forest, so operating remote sensing instruments on the ground is impossible. For that, as part of our previous FAPESP Thematic project, we set up a new ATTO site called CAMPINA-ATTO, located about 4 Km from the main tower but is a natural open forest area. The schematics of instruments being operated at the Campina-ATTO site are shown below in Figure 5.2.1. It includes clouds and precipitation radars, Polarized Aerosol Lidar, disdrometers, ceilometers, radiometers, and wind profiles, among other instruments.

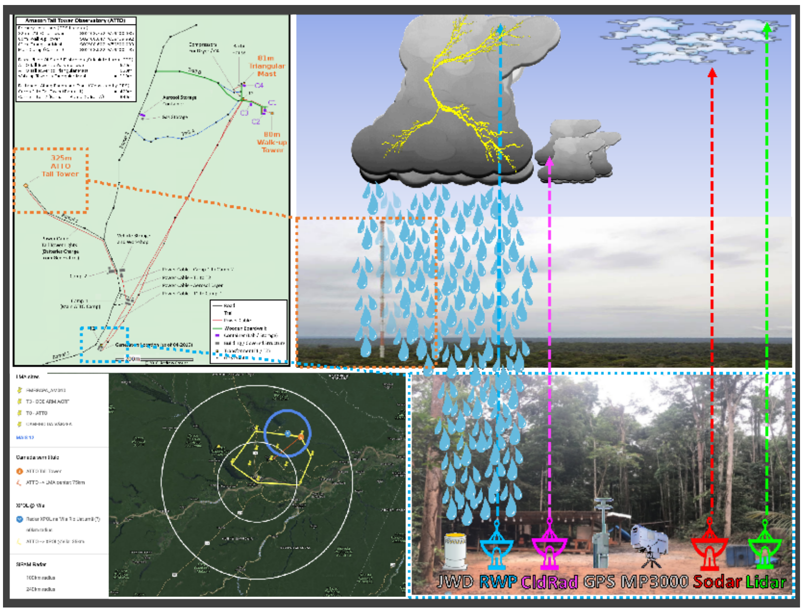


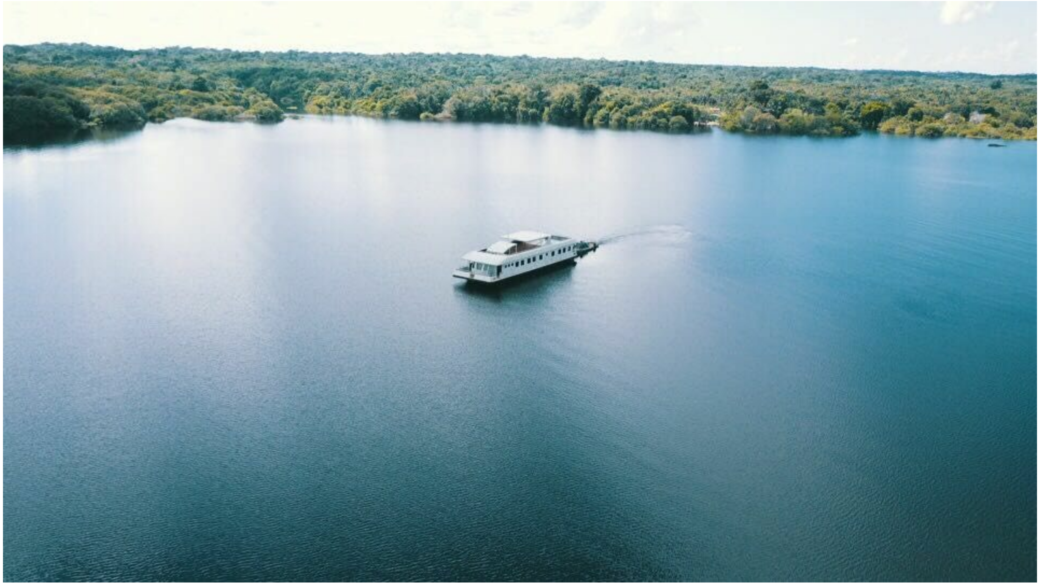
Figure 5.2.2 shows a picture of the site in 2023, with all instruments fully operational. It also shows the location of the Campina-ATTO site.

5.3 – Large scale measurements using ships in rivers in Amazonia

Many regions in Amazonia, such as western Amazonia, are still without systematic atmospheric measurements, primarily because of complex logistics. The LBA project operates a set of 7 towers in remote locations, but it is an undersampled region for a region with 5.5 million square km. Climatology from Western Amazonia differs greatly from Eastern, which could make the forest emissions different from the Santarem, Manaus, Tefé, and São Gabriel da Cachoeira regions. Figure 5.3.1 below indicates this region and where fluvial measurement campaigns will analyze atmospheric properties.

Figure 5.3.1 – Main Rivers in the Western Amazonia, where instrumented boat campaigns will analyze atmospheric properties in pristine regions.

Another boat platform that we plan to use is the new UEA (Universidade Estadual do Amazonas), a river research vessel. The figure below shows a picture of the research ship. Alternatively, we can rent local boats in other rivers in the basin to complement measurements for this project component. The persons responsible for this UEA platform are Prof. Rodrigo de Souza and Prof. Sérgio Duvoisin.



The main objectives of this component will be to characterize VOC emissions and SOA production in this remote region. The very high biodiversity in Amazonia and the species' dependence on VOC emissions make measuring atmospheric properties in different conditions and regions necessary. We will use this 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, CO_2), 7) Micro pulse Lidar, among others.

5.4 – Analysis of the data collected during the CAFÉ-Brazil Experiment

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

The CAFÉ-Brazil has just been successfully completed, collecting 143 hours of data covering 85,000 km transects over the Amazon in December 2022 and January 2023. The collected dataset will be analyzed in the framework of this proposal over the next 2 to 3 years, and the measurements will be integrated into the dataset for this project. The DLR Gulfstream 550 was totally equipped with the instrumentation described in Figure 6.4.1, which has state-of-the-art instrumentation with very low detection limits and high precision. Figures 6.4.2 and 6.4.3 illustrate the aircraft and the team of Brazilians and Germans responsible for running the CAFÉ-Brazil experiment.

Figure 6.4.1 – The HALO plane doing a research flight. The aircraft was based in Manaus during the CAFÉ-Brazil experiment, and flights covered most of the Amazon region.

Figure 6.4.2 – Photo from inside the plane and the German and Brazilian teams responsible for running the experiment.

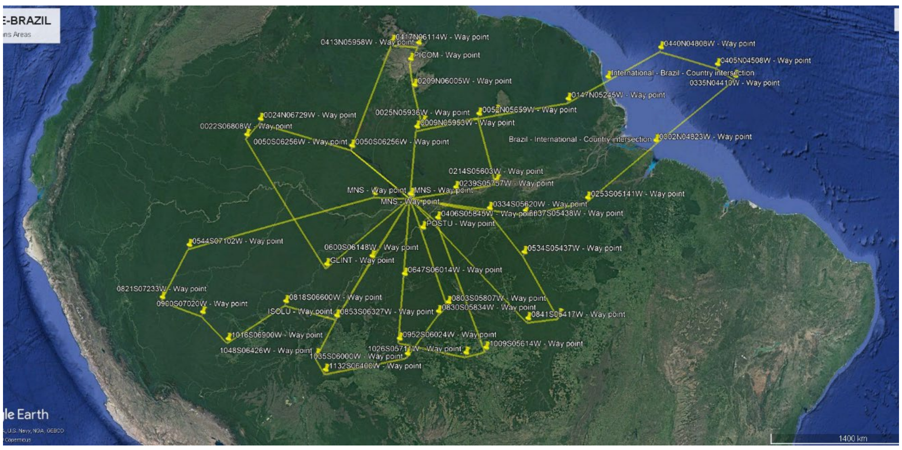


Figure 6.4.4 - Region covered by the HALO flights, sampling most of the Amazon Rainforest. Many vertical profiles were made from 150 to 14,000 meters at almost all of the Amazon.

Figure 6.4.3 - Instruments in operation on the HALO plane for the CAFE-Brazil experiment.

Institute	Instrument	Species/Parameters
GU Frankfurt	CI-API-ToF MS	H_2SO_4, HOMs/ELVOCs, clusters
MPIC	HALO-CIMS	PAN/PAA, SO_2, $ClNO_2$, HCl
MPIC	HALO-MGC	NMVOC
MPIC	PTR-MS TOF	OVOC
MPIC	TRIHOP	Total Peroxides, H_2O_2, HCHO
MPIC	NOAH/ATTILA-CLD	NO, NO_2, CO, CH_4
MPIC	HORUS	OH/HO_2
MPIC	LIF-SO2	SO_2
MPIC	C-ToF-AMS	Aerosol composition (non-refractory)
MPIC	FASD (CPCs, UHSAS, OPC)	Aerosol number and size distribution
MPIC	CCN-Rack (CCNC, SP2, impactor)	CCN, BC, aerosol impactor
KIT	FAIRO	O_3
FZ-Jülich	HALO-SR	Actinic Flux
DLR-FX	BAHAMAS	P, T, wind, humidity, TAS, aircraft position, altitude
DLR-FX	SHARC	H_2O mixing ratio (gas phase)

The flights were performed from 150 meters above the ground to 14,000 meters above sea level, at the top of the troposphere. This is the first measurement of greenhouse gases at that altitude over the Amazon, and several reactive gases have been measured for the first time in tropical forests. Figure 6.4.4 shows the extensive region covered by the HALO research flights, which included regions on the Atlantic Ocean up to the Western border of Amazonia.

The large data set collected during the recent CAFÉ-Brazil experiment will be analyzed during the first two years of this proposal. The analysis will integrate CAFÉ-Brazil, Float, and ATTO tower measurements in an integrated way, and the unique data sets will be used in the modeling component.

6 – Synergies with other research projects

This project will be run in collaboration with other Brazilian and international projects. The integration will benefit both projects. We took special care for not duplicate research efforts, and most of the projects will profit from shared data, instruments, and modeling analysis.

FAPESP Thematic project, "***Synergistic effects of climate change and land use on carbon source and sink of Amazon Forest ecosystem***," coordinated by Luiz Augusto Machado from IFUSP and XU Xiyan, from the Institute of Atmospheric Physics, Chinese Academy of Sciences. This project was recently approved through the FAPESP-NSFC call for proposals. It will link with this proposal in the modeling component.

FAPESP JP Project 2022/13257-9, from Micael Amore Cecchini. The title is "**Correlacionando o nivel de organização de nuvens convectivas aos ciclos hidrológicos e de aerossóis na Amazonia**" (CLOUDORG). This project aims to obtain a better understanding of the transition of low clouds to deep convection in Amazônia, one aspect that is poorly represented in regional and global climate models.

The project **"EValuation of the Impact of Atmospheric Deposits at the Scale of the AMazonian Basin (EVIDAM)"** will look at dust transported from Africa to Brazil. It will use advanced isotopic measurements to trace dust regions. It is coordinated by Damien Guinoiseau, University of Paris Saclay – CNRS. The proposal was submitted in 2022 and is being analyzed at CNRS.

Proposal for the ERC synergy grant **"BIODUST: Effects of BIOcrusts on global DUST cycling, biogeochemical processes, climate, and health**." Proposed by Bettina Weber, University of Graz, Austria. It also looks at aerosols transported from Africa but looks at bioaerosols. The proposal was submitted at the end of 2022 and is under review.

7 - Expected results and innovation

The broad 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, precipitation, and carbon cycling can help constrain this critically important area. The mechanisms of new particle formation at the ground and the high troposphere are very different from what we observe in boreal forests.

The joint use of vertical profiles from the ATTO tower, with the Campina-ATTO instrumentation with LIDARs, radiosondes, cloud, precipitation, and meteorological radars, is a powerful tool in tropical area experiments because it allows a detailed picture of the vertical distribution of trace gases and aerosols and its links with convection and cloud transport. The metagenomics analysis of aerosol particles also has the potential to find new links between atmospheric sciences and the biosphere.

The joint observation of greenhouse gases and carbon isotopic composition in multiple platforms has the potential to reveal novel aspects of the complex interactions between the forest canopy and the atmosphere.

Using high-resolution LES models coupled with aerosol-cloud interactions studies, the extensive modeling component can bring new insights into the complex aerosol-hydrological cycle links. The use of regional mesoscale models such as WRF-Chem-GHG and global Earth System models can also help integrate the large amount of information collected in this project. The international partners in this project and the Amazonian institution's scientists can provide an intense interaction of different views about Amazonian Science.

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