Sources and Properties of Amazonian Aerosol Particles

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1. Introduction

Aerosol particles in the Amazon Basin have been the focus of numerous field campaigns over the past 20 years (Table 1 and Figure 1). These studies were motivated by a wide range of objectives, the most prominent of which are as follows. (1) The Basin was used as a laboratory to gain baseline knowledge concerning pristine continental aerosol particles, against which the effects of human activities globally could be judged [*Andreae*, 2007]. (2) An understanding was sought of the effects of biomass-burning aerosol particles on human health, such as increased incidences of morbidity, mortality, and asthma [*Ignotti et al.*, 2007]. (3) The effects of aerosol particles on regional climate were investigated, such as changes in rainfall patterns as a consequence of the redistribution of energy and cloud condensation nuclei [*Andreae et al.*, 2004]. (4) The Basin was studied as an integrated ecosystem to understand the feedback and regulation of plant emissions on rainfall and, in turn, of rainfall on plant growth and emissions [*Barth et al.*, 2005; *Keller et al*., 2009]. These topics have in common a need to know the sources and properties of Amazonian aerosol particles, yet an integrated summary of results from previous field campaigns (Table 1) and associated statements of future research priorities have not been prepared previously in a comprehensive review article. This gap in the literature is the motivation for this review and defines its scope: the review's goals are to focus the ongoing activities of researchers already investigating the sources and properties of Amazonian aerosol particles and, by organizing and presenting material of what is already known and what remains to be learned, to invite new researchers to join in critical ways. Complementary general reviews of atmospheric particles, especially with regard to the organic component that is dominant in the Amazon Basin, can be found in *Andreae and Crutzen* [1997], *Jacobson et al.* [2000], *Kanakidou et al*. [2005], and *Fuzzi et al*. [2006].

Aerosol sources located within the Amazon Basin are dominated, with the exception of some urbanized areas and transportation corridors, by natural and anthropogenic emissions from the biosphere. Sources include both high but intermittent biomass-burning emissions (both natural and anthropogenic) and low but more consistent production of primary and secondary biological aerosol particles and components (Figure 2). Primary particles are produced both deliberately by flora (e.g., the release of pollen and fungal spores) and incidentally (e.g., as leaf and soil debris or as suspended microbes). Substantial production of secondary aerosol occurs by the atmospheric oxidation of trace gases to low-volatility compounds. These products can deposit on pre-existing particles or possibly nucleate new particles.

Once in the atmosphere, particles undergo continuous transformations (Figure 3). Processes include (photo)chemical reactions that occur between compounds within the particles as well as interactions that occur between compounds within the particles and those in the gas phase, such as the condensation of low-volatility compounds or reactions with highly reactive gaseous species like the OH radical. Clouds are present at varying abundances almost everywhere and all the time over the Amazon Basin, and most particles are also undergo several cycles of cloud processing during their residence in the Basin. The timescale of cloud cycling of boundary layer air (of the order of hours) is considerably shorter than the residence time of air over the Basin or the deposition lifetime of aerosol particles (of the order of days). Cloud processing can modify particle properties both by chemical reactions in the liquid phase and by interactions between droplets (e.g., collision and coagulation). Particles leave the Amazonian atmosphere by dry deposition to the vegetation surface, by cloud scavenging and precipitation, and by advection out of the region.

In addition to particle sources within the Basin, there are also important and at times dominant long-range natural and anthropogenic sources (Figure 4). The influence of long-range transport is particularly important when in-Basin sources are weak, such as in the wet season, and under these conditions the particle population can be dominated at times both in mass and number by outflow from other areas. The Atlantic Ocean, upwind of the Basin, is a strong source of marine particles that are generated both directly by seaspray as well as indirectly by the conversion of gases, such as the oxidation of dimethyl sulfide (DMS) to form sulfate. Across the Atlantic and further upwind of the Basin, the Saharan desert is the world's largest source of mineral dust. Sub-Saharan Africa is one of the most important sources of smoke from vegetation fires. Although the inflowing air masses that arrive with the trade winds from the Atlantic shed much of their particle burden in transit from Africa and Eurasia, the impact of trans-Atlantic transport on particles measured in the Basin can, nevertheless, be substantial and at times dominant [*Prospero et al.* 1981; *Andreae et al.*, 1990a; *Artaxo et al.*, 1990; *Swap et al.*, 1992; *Formenti et al.*, 2001; *Chen et al.*, 2009]. Furthermore, pollution-derived particles from urban and industrialized areas in southern and eastern Brazil and other South American countries can also be transported into the Basin, especially in the dry season.

During the wet season (Dec-Mar, i.e., summer of the Southern Hemisphere), atmospheric particles are removed relatively fast by wet deposition, and anthropogenic sources such as biomass burning are weak throughout the Basin. This combination of circumstances results in natural processes (including contributions from marine and African sources) as the dominant contributors to the ambient particle populations over large expanses of the Basin and during a significant part of the year. The particle concentrations measured during these conditions are among the lowest found on any continent and are similar to those over the remote oceans [*Andreae*, 2009]. The Basin has been dubbed the "green ocean" because of the similarities in particle concentrations and cloud microphysics between it and remote oceanic regions [*Williams et al.*, 2002]. The Amazon Basin may be the only region on the tropical continents where there remains the possibility to find at times populations of nearly pristine aerosol particles free of direct anthropogenic influences.

Compared to these green-ocean conditions of the wet season, there is stark contrast in the dry season for large regions of the Basin (Jun-Sep, i.e., winter of the Southern Hemisphere). Vast numbers of deforestation fires burn during the dry season, especially along the peripheries of the forest, and large parts of the Basin become among the most polluted places on Earth [*Artaxo et al.*, 2002; *Cardoso et al.*, 2003]. As one effect, the regional energy balance is changed because the high particle concentrations affect the amount and location of solar radiation absorbed by the planet. Simulations using regional climate models show that the changes in energy delivery significantly influence regional patterns of atmospheric circulation and meteorology [*Zhang et* *al.*, 2009]. The high particle concentrations change cloud microphysics and rainfall, with a significant influence on the overall water cycle [*Andreae et al.*, 2004; *IPCC*, 2007; *Rosenfeld et al.*, 2008]. They also influence air quality by degrading visibility and affecting human health [*Reinhardt et al.*, 2001; *Schwartz et al.*, 2002; *Watson*, 2002; *Pope and Dockery*, 2006; *Barregard et al.*, 2006].

The meteorology of South America guides the transport of particle-free or particle-laden air masses into the Amazon Basin. It also affects the rates of wet and dry particle deposition. A brief introduction to the meteorology of South America is therefore provided herein, in the next few paragraphs. A more in-depth presentation can be found in *Satyamurty et al.* [1998], and an introduction to the regional climate of the Amazon Basin (including a history of field campaigns focused on meteorology) can be found in *Nobre et al.* [2004].

The convective activity and the atmospheric circulation of tropical South America are part of a monsoon system [*Zhou et al.*, 1998]. The seasonal cycle of circulation and convection is revealed both by changes in the low-level wind field (cf. Figures 5a and 5b) and in the outgoing longwave radiation (cf. Figures 5c and 5d) for December through February (DJF) compared to June through August (JJA) [*Kalnay et al.*, 1996]. In both the wet and dry seasons, there is considerable inflow coming from the east into the Basin. These air masses originate in the Atlantic, and many of them pass through the semi-arid region of northeastern Brazil before entering central and southern regions of the Basin [*Satyamurty et al.*, 1998].

Several aspects of Figure 5 are important for understanding the variability of particle concentrations and compositions observed in the Amazon Basin. For northern and central Amazonia, the Intertropical Convergence Zone (ITCZ), which is the confluence between the northeastern and southeastern trade winds extending from West Africa to South America (Figures 5a and 5b), has an important influence. The ITCZ reaches the northern coast of South America just south of the equator in DJF but just north of it in JJA. When the ITCZ is located to the south of the equator in DJF (i.e., a large part of the Basin is under the influence of air from the Northern Hemisphere), low-level winds from the global northeast reach the northern coast of

South America and open up the possibility of advection of African dust and biomass-burning particles (e.g., *Artaxo and Hansson* [1995] and *Formenti et al.* [2001]). During the wet season, Figure 5c shows that the outgoing longwave radiation has a minimum over central Amazonia, implying cold cloud tops and hence deep clouds with strong convection and high rates of wet deposition. There is also strong vertical transport and redistribution of particles [*Freitas et al.*, 2000; *Andreae et al.*, 2001]. In comparison, during the dry season the convection weakens and shifts to the northwestern edge of geographical Amazonia (Figure 5d).

In the southern part of the Amazon Basin, northwesterlies prevail during the wet season and are associated with the South Atlantic Convergence Zone (SACZ), a region of enhanced convective activity extending from central Amazonia to the southeast [*Kodama*, 1992]. The SACZ forms a quasi-stationary front with significant intraseasonal variability. As the remains of mid-latitude fronts reach the tropical region [*Garreaud and Wallace*, 1998], the SACZ is reinforced. In the absence of perturbations coming from the south, the SACZ weakens and may disappear. This variability leads to a change in low-level winds from easterlies in the break periods to westerlies in the periods of well-defined SACZ. The active and break periods are related to intraseasonal oscillations [*Nogues-Paegle et al*., 1997] that affect the whole region from the northern coast [*Wang et al*., 2002] to the more continental area [*Jones et al*., 2002]. During the break periods, deep convective systems are commonly isolated, and (given the absence of sinks) particle concentrations are relatively high. By comparison, during SACZ events convection and rainfall are more widespread, the atmosphere is clean, and particle concentrations are relatively low [*Silva Dias et al.*, 2002; *Williams et al.*, 2002].

With the coming of the dry season, the low-level winds in the southern part of the Basin reverse, changing from northwesterlies to southeasterlies (Figures 5a and 5b). These patterns favor the flow of pollution from urban and industrial Brazil into this region of Amazonia, thereby increasing particle concentrations. Alternatively, cold-front southerlies (i.e., winter in the Southern Hemisphere) can bring clean air into the southern part of Amazonia, in some cases extending as far as the equator [*Marengo et al.*, 1997]. At these times, particle concentrations in southern Amazonia can drop considerably. More typically, however, biomass burning in the Basin leads to sustained high particle concentrations regardless of southerlies or southeasterlies [*Artaxo et al.*, 2002].

The diel evolution of the planetary boundary layer (cf. Figure 6) also affects particle concentrations measured at the surface. During the afternoon, the boundary layer is well mixed by strong turbulence that is driven by sensible heat flux from the surface. The depth of this convective boundary layer (CBL) is variable depending on land cover and on the meteorology. By late afternoon CBL depths greater than 1000 m are typical in the Amazon Basin, although variability is high. *Fisch et al*. [2004] observed differences between the wet and dry seasons (e.g., lower than 1500 m in the former and up to 2000 m in the latter) and between forest and pasture landscapes. At sunset, radiative cooling at the surface generates a nocturnal stable layer. The nocturnal boundary layer usually has a depth of a few hundred meters or less. The residual layer above the nocturnal stable layer is typically without turbulence or mixing, although there are exceptions at times when higher-altitude shearing jets are present that induce turnover and hence the cleansing of the residual layer with cleaner higher altitude air. In the absence of cleansing, the nighttime residual layer conserves the properties (such as particle and gas concentrations) of the previous afternoon until the following morning.

These basic characteristics of the daytime and nighttime boundary layers affect particle concentrations measured at the surface. During the afternoon, emissions from the surface get mixed though the whole volume of the convective boundary layer, diluting their concentrations. Moreover, at the top of the boundary layer, turbulence in fair weather conditions maintains an entrainment of cleaner air from higher altitudes while convective clouds associated with rainy weather pump the aerosol particles to higher levels. These effects favor a dip of particle concentrations in the early afternoon. In comparison, the stable nocturnal boundary layer traps emissions in a volume near the surface, thereby favoring higher particle concentrations at night. This effect is amplified during the dry season because biomass burning usually begins at mid-day and continues into the evening hours. The residual layer above the nocturnal boundary layer influences the surface concentrations the following morning because the development of the convective boundary layer mixes the nocturnal layer into the residual layer. As a result, mid-to-late morning surface concentrations (in the absence of nighttime cleansing of the residual layer) can be similar to nighttime concentrations measured the afternoon before in the convective boundary layer [*Rissler et al.*, 2006].

Given the seasonal and regional variability of the contributions from different aerosol sources, the changing transport paths of air masses, and the different removal rates in the dry and wet seasons, the inference (which is supported by the observations) is that Amazonian aerosol particles have considerable variations in space and time, and consequently there is a considerable body of literature to review. Our approach in this review is to follow the life cycle of Amazonian aerosol particles, as outlined in the previous paragraphs. The review begins with a discussion of the primary and secondary sources relevant to the Amazonian particle burden, followed by a presentation of the particle properties that characterize the mixed populations present over the Amazon Basin at different times and places. These properties include number and mass concentrations and distributions, chemical composition, hygroscopicity, and cloud nucleation ability. The review presents Amazonian aerosol particles in the context of natural compared to anthropogenic sources as well as variability with season and meteorology. The review concludes with an outlook and priorities for further research.

2. Sources

Amazonian aerosol particles have a wide range of natural and anthropogenic sources, and the integrated effects of emission and processing in the atmosphere lead to complex internal and external mixtures of particles, even within an apparently homogeneous air mass. The complexity can be usefully dissected by conceptualizing a single particle as composed of components. Depending on their origin, components are usually classified as primary or secondary. Primary components are directly emitted from a source into the atmosphere; secondary components are formed in the atmosphere [*Fuzzi et al.*, 2006]. A single particle composed mainly of primary components can be called a primary aerosol particle, and a single particle composed mainly of secondary components can be called a secondary aerosol particle. After some air mass aging, many, if not most, individual particles can be composed of significant quantities of both types of components. In the Amazon Basin, organic components typically constitute approximately 70 to 90% of the particle mass concentration in both the fine and coarse size fractions [*Graham et al.*, 2003a; *Fuzzi et al.*, 2007].

Examples of primary biological aerosol (PBA) particles emitted in the Amazon Basin include pollen, bacteria, fungal and fern spores, viruses, and fragments of plants and animals [*Elbert et al.*, 2007]. Anthropogenic biomass burning is also an important and at times dominant source at some locations, especially during the dry season. In addition to sources within the Amazon Basin, primary particles are also brought in by long-range transport, such as marine particles from the Atlantic Ocean and desert dust or biomass-burning particles from Africa [*Andreae et al.*, 1990a; *Artaxo et al.*, 1990; *Swap et al.*, 1992; *Formenti et al.*, 2001; *Chen et al.*, 2009]. Regional urban and industrial activities, including traffic and industry in Manaus and other cities and settlements in northeastern and southern Brazil, also have outflow plumes containing combustion-derived particles and dust, and these plumes are significant when a sampling location lies within them.

Examples of components of secondary organic aerosol (SOA) are the low-volatility molecules that result from the reactions of O_3 and OH with biogenic volatile organic compounds (BVOCs), such as isoprene and terpenes. BVOCs are emitted in large quantities to the gas phase by plants. Low-volatility BVOC oxidation products can condense from the gas phase onto pre-existing particles or alternatively can contribute to new particle formation. Liquid-phase reactions inside cloud droplets can also yield low-volatility BVOC oxidation products, serving as another source of organic components in particles for cloud droplets that evaporate [*Blando and Turpin*, 2000; *Lim et al.*, 2005; *Carlton et al.*, 2006].

2.1. Primary Particles

2.1.1. *Primary Biological Particles*

Emissions of primary biological particles are often wind-driven, such as suspension of pollen, plant debris, or soil dust [*Jaenicke*, 2005; *Pöschl*, 2005]. In addition to wind-driven release, certain biological organisms also actively eject materials into the air for reproductive purposes, such as wet-discharged fungal spores [*Elbert et al.*, 2007]. Coarse-mode PBA particles in the Amazon Basin have sizes ranging from several to tens of micrometers and include fragments of plants and insects, pollen grains, algae, fern spores, and fungal spores [*Graham et al.*, 2003a]. Microscopic analysis of collected particles show that, in the absence of African dust and Atlantic marine emissions, morphologically identifiable biological particles dominate both the number-diameter and volume-diameter distributions of the coarse fraction for natural conditions (Figure 7). PBA components like carbohydrates, proteins, and lipids, as well as elemental tracers, have also been detected in the fine fraction [*Artaxo and Hansson*, 1995; *Andreae and Crutzen*, 1997; *Graham et al.*, 2003a]. AMAZE-08 observations by *Sinha et al.* [2009] and *Chen et al.* [2009], however, suggest that PBA components contribute in a minor way to the size class below 1 μm. Nevertheless, the actual number, mass, and size of PBA particles emitted to the fine fraction remain to be fully quantified for Amazonia, and the abundance and composition of PBA particles is highly variable and still poorly characterized, partially because the distinction between biological and other carbonaceous components requires advanced analytical techniques and intensive investigation [*Pöschl*, 2005; *Fuzzi et al.*, 2006; *Despres et al.*, 2007].

Fungi are an especially important source of coarse-mode PBA particles in the Basin. They actively discharge their spores with liquid jets and droplets into the air, and these processes are most active under humid conditions, such as those in the rain forest [*Gilbert*, 2005]. For particles of 1 to 10 μm, fungi are estimated to contribute 25% of the particles during the day and 45% at night. In corroboration, measurements using an ultraviolet aerodynamic particle sizer deployed during AMAZE-08 indicate that fungal spores and other viable PBA particles account for up to 80% of coarse-mode particles during time periods of weak influence from sources outside of the Basin (U. Pöschl, results to be published). Actively wet spore discharging fungi also emit dissolved inorganic salts like potassium chloride and organic substances like sugar alcohols such as mannitol in aqueous jets, and these jets break up to form droplets that can dry as fine particles [*Elbert et al.*, 2007].

2.1.2. *Biomass Burning*

Anthropogenic aerosol particles are generated in great number and mass concentration by biomass burning that is used for deforestation and pasture maintenance [*Andreae et al.*, 1988; *Artaxo et al.*, 1998; *Andreae et al.*, 2002; *Artaxo et al.*, 2002; *Guyon et al.*, 2003b; *Andreae et al.*, 2004; *Freitas et al.*, 2005; *Guyon et al.*, 2005; *Fuzzi et al.*, 2007; *Yokelson et al.*, 2007]. The resulting particles are predominantly carbonaceous, consisting largely of organic carbon but also with significant nearly-elemental soot carbon and minor amounts of inorganic materials. The particles consist of a variety of internal and external mixtures, including soot-carbon fractal aggregates, tarry materials (sometimes present as spherical "tar balls"), grains of inorganic salts (KCl, K_2SO_4, KNO_3, $(NH_4)_2SO_4$, etc.), and ash and char particles [*Allen and Miguel*, 1995; *Andreae et al.*, 1998; *Ferek et al.*, 1998; *Yamasoe et al.*, 2000; *Posfai et al.*, 2003; 2004].

Emission factors from primary deforestation fires and pasture maintenance fires in tropical rain forests range from 6 to 25 g kg^{-1} for total particulate matter (PM) and 7.5 to 15 g kg^{-1} for PM smaller than 2.5 µm ($PM_{2.5}$), expressed as mass of emitted primary particles per mass unit of dry fuel. For Amazonia, the estimates for the emission rates of $PM_{2.5}$ and PM_{10} are 8 and 10 Tg yr^{-1}, respectively [*Yokelson et al.*, 2008]. The pollution plumes can be hundreds of kilometers across and thousands of kilometers long. With an optical thickness regularly exceeding 1.0 in the peak period of biomass burning in the Basin (i.e., September and October), the plumes are clearly visible in satellite images, and biomass burning is the dominant particle source in affected areas. At those times, pollution from biomass smoke typically accounts for >90% of the fine particles and about 50% of the coarse particles. The annual mean of optical thickness over Amazonia and regions nearby is dominated by biomass-burning emissions [*Tegen et al.*, 1997; *Tie et al.*, 2005].

Fire counts observed by satellite over the Amazon Basin in Feb, May, Aug, and Nov 2007 are shown. As expected, the most numerous and intense fires were in the dry season (i.e., Aug 2007 in Figure 8) in the southern part of Amazonia. Fires were also important in northeastern Brazil in Nov 2007, and the prevailing flow patterns carried the biomass-burning emissions into the central and southern Amazon Basin (Figure 5). In the wet season, biomass burning took place along the northern rim of the Basin, and at times local meteorological variability transported the biomass-burning emissions into the Basin.

In addition to biomass burning within South America, emissions from Africa are imported into the Amazon Basin at all times of the year [*Talbot et al.*, 1990; *Andreae et al.*, 1994]. Fires burn in tropical and subtropical Africa year-round, with a maximum early in the year in the Northern Hemisphere and a maximum in the second half of the year in the Southern Hemisphere. At least some of the smoke from these fires is transported across the Atlantic by the trade winds [*Andreae et al.*, 1994]. The particles emitted to the Northern Hemisphere enter northern Amazonia in the wet season and can be important, given the weak baseline production mechanisms of the Amazonian biosphere. In comparison, the contribution from the Southern Hemisphere enters southern Amazonia during the regional burning season of the latter, and the African contribution is therefore typically of less relative importance to Amazonia at that time. In summary, although the highest concentrations of biomass-burning particles are observed in southern Amazonia during the dry season, lower levels of biomass-burning particles can be important intermittently at any time and at most locations of the Basin throughout the year.

2.1.3. *African Mineral Dust*

Saharan dust is a prominent out-of-Basin particle source. The importance of the transatlantic transport of dust was recognized by *Prospero et al*. [1981] and has been observed in several subsequent measurement campaigns [*Swap et al.*, 1992; *Artaxo et al.*, 1998; *Formenti et al.*, 2001]. Imported dust occurs at its highest concentrations in those parts of the Basin that are north of the ITCZ. The maximum dust concentrations at the surface are typically reached around March and April, coinciding with the wet season in the central Basin. A significant fraction of the delivered mineral dust is submicron, as explained by the large transport distance from Africa and the preferential loss of coarse-mode particles along the way. The dust is observed at near-surface stations in pulses of high concentrations that last from one to several days, and, when present, mineral dust often dominates the total particle mass concentrations [*Andreae et al.*, 1990b; *Talbot et al.*, 1990; *Formenti et al.*, 2001; *Worobiec et al.*, 2007].

2.1.4. *Marine Emissions*

Crossing the coast of the Amazon Basin with the trade wind flow, large concentrations of marine aerosol particles are progressively removed by wet and dry deposition as air masses travel deeper into the Basin [*Andreae and Andreae*, 1988; *Talbot et al.*, 1988; *Andreae et al.*, 1990a; 1990b; *Talbot et al.*, 1990; *Worobiec et al.*, 2007]. The contribution by marine particles to the total Amazonian particle mass concentration can remain significant even over the central parts of the Basin. Marine aerosol particles consist in large part of primary seaspray particles, which are composed mainly of coarse-mode inorganic salts mixed with lesser amounts of the primary biological material that was partitioned to the ocean's surface [*O'Dowd et al.*, 2004; *Andreae and Rosenfeld*, 2008]. The sodium and chloride content of the coarse fraction of the particle population in the Amazon Basin is explained almost entirely by marine sources. Marine aerosol particles also have a substantial contribution from secondary processes, such as sulfates produced by the oxidation of dimethyl sulfide and organic material produced by the oxidation of volatile organic compounds [*Ceburnis et al.*, 2008]. Much of the secondary material occurs in the fine mode. In the absence of biomass-burning particles or dust, approximately half of the submicron sulfate fraction is attributable to secondary sulfate produced from marine emissions, especially from DMS oxidation [*Andreae et al.*, 1990a; *Worobiec et al.*, 2007].

Priorities for progress in identifying the sources of primary particles in the Amazon Basin and quantifying their emissions include:

- Characterization and quantification of different types of primary biological, biomass burning, mineral dust, and marine aerosol particles, including long-term trends, seasonal cycles, and diel differences. Identification of their mixing states, including the relative contributions of primary and secondary components.

- Discrimination and quantification of the relative fractions of in-Basin and out-of-Basin sources of all particle types.
- Improved characterization and understanding of Amazonian aerosol particles by application of a combination of advanced measurement techniques, such as bulk and single-particle mass spectrometry, X-ray microanalysis, fluorescence spectroscopy, electron microscopy, and DNA analysis.
- Development of process models describing the emission of primary biological particles from the Amazonian ecosystem and implementation of these process models in regional and global models of atmospheric chemistry, transport, and climate.

2.2. Secondary Gas-to-Particle Conversion

The production mechanisms for secondary particle components involve many trace gases, in particular biogenic volatile organic compounds, nitrogen oxides (NO_x), ozone (O_3), hydroxyl radical (OH), and sulfur species including dimethyl sulfide (DMS) and sulfur dioxide (SO_2) [*Andreae and Andreae*, 1988; *Jacob and Wofsy*, 1988; *Andreae et al.*, 1990a; *Browell et al.*, 1990; *Jacob and Wofsy*, 1990; *Kesselmeier et al.*, 2000; *Andreae et al.*, 2002]. DMS and SO_2 are oxidized to form particle sulfate. BVOCs 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 and NO_x together influence the concentrations of O_3 and OH, thereby influencing the production of BVOC oxidation products. Reactions both in the gas phase and in cloud waters are important. BVOCs and NO_x emitted by biomass burning can lead to regional concentrations of these trace gases and O_3 that are similar to those found in industrially polluted regions.

For natural conditions (i.e., as defined by the influences in Figure 4), Figure 9 shows that NO_x and O_3 concentrations in the convective boundary layer over Amazonia are low, of the order of 20 - 200 ppb and 5 - 20 ppb, respectively. At night, NO_x concentrations within the canopy space can increase by a factor of approximately 10 because the trace gases emitted from the soils get trapped by the nocturnal inversion (Figure 10). At the same time, O_3 concentrations can drop to nearly 0 ppb in the canopy space, both because of reactions between NO and O_3 and because of deposition to the vegetation surfaces. The enrichment of trace gases in the nocturnal boundary layer, especially in topographic depressions, can persist into the morning hours, thus providing conditions that may be conducive to intense photochemical processes, possibly including the formation of low-volatility products relevant to particle growth.

2.2.1. *Biogenic Volatile Organic Compounds*

Biogenic volatile organic compounds are emitted from plants during growth, maintenance, decay, and consumption, and average emission rates account for more than 2% of net primary productivity in the Basin and other regions [*Zimmerman et al.*, 1988]. The Amazon Basin contains on the order of 10^5 plant species, each having unique signatures of BVOC emissions. Estimates of BVOC emissions from the whole of the Amazon Basin represent a challenging but important task. Prior to new studies conducted in the past decade, Amazonian BVOC emission estimates were based on a few measurements conducted by *Zimmerman et al.* [1988]. *Kuhn et al*. [2007] and *Karl et al*. [2007] have shown that under some circumstances for specific compounds models of biogenic emissions accurately simulate measured BVOC fluxes in the region. The substantial progress in understanding Amazonian BVOC emissions and the major remaining uncertainties are described in detail by *Kesselmeier et al.* [2009].

The specific BVOC compounds emitted and their relative rates of emissions vary widely by plant species and environmental conditions. Major BVOCs emitted include isoprene (C_5H_8), monoterpenes (i.e., compounds composed of two isoprene moieties), sesquiterpenes (i.e., three isoprene moieties), ethane, and oxygenated VOCs (OVOCs). BVOC emissions typically increase exponentially with temperature, doubling every 5 to 15 K depending on the compound. For some compounds such as isoprene, emissions also increase with available sunlight, and light-dependent monoterpene emissions result in a pronounced diel flux and mixing ratio cycle (Figure 11). The controlling mechanisms for emission differ among compounds. Some are emitted immediately following production, and others are stored in plant tissues. Examples of directly emitted compounds include isoprene and some types of monoterpenes and sesquiterpenes. Examples of stored compounds include other types of monoterpenes and sesquiterpenes, some oxygenated terpenes, and some components of plant oils. The factors influencing the emission of stored compounds to the atmosphere are complex, depending on molecular vapor pressure, animal herbivory, and plant phenology, moisture, or stress.

Although tropical forests are the dominant global source of atmospheric BVOCs and the Amazon Basin is a major contributor [*Rasmussen and Khalil*, 1988], BVOC emissions have been studied more extensively in temperate regions. The high species diversity in the Amazon Basin is coupled with an ecological complexity and a seasonality, however, that is very different from temperate regions, yielding significantly different emission trends with different forest types. For example, because of the consistently high temperatures over the Amazon Basin, BVOC emissions do not exhibit large seasonality there. Isoprene and monoterpene emissions and concentrations are also strongly correlated in the Amazon (Figure 11), in contrast to their anticorrelated behavior in temperate forests. Isoprene concentrations are highest at midday in temperate forests while monoterpene concentrations are highest at night, corresponding to their emission into a shallow boundary layer. Monoterpene release by plants in those forests is dominated by the emission of stored compounds, and the diel monoterpene emission pattern is therefore significantly different from that of isoprene, which favors released during time period of intense sunlight. The explanation for the different diel monoterperene emission pattern in the Amazon rain forest is not yet fully known.

Emissions of BVOCs have been incorporated into global chemical transport models, and the contribution of low-volatility BVOC oxidation products to the mass concentration of organic particles has been predicted [*Chung and Seinfeld*, 2002; *Tsigaridis and Kanakidou*, 2003; *Hoyle et al.*, 2007; *Heald et al.*, 2008]. *Heald et al.* [2008] estimate that the conversion of South American BVOCs into secondary particle mass contributes 40% of the annual global production of this particle component. Simulated concentrations over the Amazon Basin vary from 0.6 to 3 μg m^{-3} and peak in the dry season, corresponding to decreased wet deposition during that time period.

A significant underestimate by models of ambient organic particle concentrations, as reported for a number of anthropogenically influenced environments, has been attributed to underestimated conversion yields of oxidized VOCs to secondary particle mass [*Volkamer et al.*, 2007]. The models employ laboratory-based yields of a few percent for isoprene and 10-15% for most terpenes [*Chung and Seinfeld*, 2002; *Henze and Seinfeld*, 2006]. These yields, however, may significantly underestimate what occurs over longer timescales in the atmosphere [*Ng et al.*, 2006]. Recent laboratory studies carried out for BVOC concentrations present in the atmosphere have also shown that yields of secondary particle mass are higher than those obtained by the extrapolation of earlier laboratory results carried out at higher BVOC concentrations [*Shilling et al.*, 2008], and this finding could potentially bring models and observations into closer agreement. A mass-balance approach, based on analysis of the fate of BVOCs as either (i) oxidation to CO and CO_2 or (ii) deposition with the remainder assumed to form particle mass, leads to much higher production estimates of secondary particle mass [*Goldstein and Galbally*, 2007]. For the Amazon Basin, the underpredictions by chemical transport models of observed concentrations appear significantly less (e.g., 35% reported by *Chen et al*. [2009]) than those reported for anthropogenically influenced regions of the world.

Further refinement of the mass-balance approach requires better measurements of BVOC oxidation products and their rates of wet and dry deposition. Studies of plant signaling, defense, and food and flavor chemistry have led to the detection of thousands of individual BVOCs [*Hines*, 2006], yet only a few of these have been studied for their emission rates, their atmospheric chemistry, and their contribution to secondary particle mass. Given the limitations in knowledge of emissions rates, oxidation pathways, and yields of particle mass, monoterpene, sesquiterpene, and other BVOC emissions are generally lumped into a few categories for both emission modeling and atmospheric chemistry modeling. As analytical techniques have improved in recent decades, a much broader array of highly reactive and oxygenated BVOCs and their oxidation products has been identified [*Holzinger et al.*, 2005; *Lee et al.*, 2006; *Surratt et al.*, 2008]. The hardest to measure of the directly emitted compounds tend to be the most reactive, and these same compounds, following multi-step oxidation in the atmosphere, often have the highest potential for the production of secondary particle components [*Ng et al.*, 2006]. Sesquiterpenes, which have atmospheric lifetimes of a few minutes or less, are the best known example [*Ciccioli et al.*, 1999].

One approach to estimate BVOC emissions for incorporation in chemical transport models is a bottom-up calculation constrained by leaf, branch, or canopy-scale fluxes [*Guenther et al.*, 1995]. Bottom-up models are based primarily on enclosure measurements that characterize emissions associated with the foliage of an individual plant species. The resulting emission factors are combined with the distributions of plant species to estimate landscape-level emission rates. This approach, however, is difficult to apply in the Amazon Basin because of the high species diversity. Direct BVOC emission measurements are available for less than 0.2% of the 10^5 plant species in the Amazon Basin [*Harley et al.*, 2004], and investigations performed to date consist of only a few measurements per plant species, with analyses that include only a limited subset of all BVOCs. Even so, *Harley et al.* [2004] demonstrate the utility of this approach for specific Amazonian landscapes. Accurate characterization of the species-dependent emission rates typically requires a large number of measurements over different seasons and locations due to the substantial variability. Representative measurements also require sampling of upper canopy foliage, which is often difficult to access in rain forests. Above-canopy flux measurements, summarized by *Kesselmeier et al.* [2009] for Amazonia, provide an alternative approach to the parameterization of bottom-up models.

Another approach to estimate BVOC emissions for use in chemical transport models is a top-down calculation driven by satellite observations of the atmospheric distribution of formaldehyde (HCHO). Formaldehyde is a high-yield oxidation product of isoprene and thus enhancements above the methane background can be used as a proxy for this emission source [*Palmer et al.*, 2003]. The first global top-down study by *Shim et al.* [2005] provided a global emission estimate that was approximately 13% higher than the bottom-up estimate of *Guenther et al*. [1995]. The estimates by *Shim et al.* [2005], however, were 35% lower for South America than those of *Guenther et al*. [1995]. Recent top-down investigations have begun to specifically focus on the Amazon Basin. *Palmer et al.* [2007] examined seasonal variations and found a good correlation between satellite HCHO column measurements and isoprene concentrations measured above the eastern part of the Basin. A related top-down study by *Barkley et al*. [2008] estimated isoprene emissions that were 35% lower than the bottom-up estimates of *Guenther et al.* [2006]. This result differed from that of *Stavrakou et al*. [2009], who estimated in a top-down study that isoprene emissions were slightly higher than the bottom-up emission estimate of *Guenther et al.* [2006]. The bottom-up and top-down models, nevertheless, can be considered to generally agree because each has an uncertainty factor of about two.

After emission, some fraction of the oxidized BVOCs yields secondary particle mass. *Went* [1960] first proposed that BVOCs are oxidized creating the blue haze observed in the atmosphere above many forested regions. BVOCs are usually oxidized by OH, O_3, or NO_3 in the atmosphere. The OH pathway is particularly important for BVOC oxidation in the tropics given the high light levels and H_2O concentrations. The NO_3 pathway is minor for the usual conditions in the Amazon Basin because there is low O_3 at night and low NO_2 in the day so that the rate of formation by $O_3 + NO_2$ is slow. However, 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 still extremely limited. Even for the well-studied compound isoprene, recent analysis suggests that state-of-the-art atmospheric chemistry models greatly underpredict OH concentrations [*Lelieveld et al.*, 2008], possibly implying important missing chemistry [*Karl et al*., 2009]. The OH concentrations measured in flights by *Lelieveld et al.* [2008] over the Amazon forest ($5.6 \pm 1.9\ 10^6$ molec cm^{-3} in the boundary layer and $8.2 \pm 3.0\ 10^6$ molec cm^{-3} in the free troposphere) were significantly higher than anticipated from model calculations, suggesting an overlooked pathway mediated by organic peroxy radicals for OH production (Figure 12). Isoprene emissions estimated using inverse modeling can be artificially low for times during which OH is underestimated [*Kuhn et al.*, 2007]. In addition, higher OH estimates change greatly our understanding of photochemistry in the tropics and the rate of transformations of BVOCs into particle mass.

Opportunities for progress in identifying the correct BVOC precursors to secondary particle mass include:

- *Emissions*: Develop approaches for measuring total non-methane BVOC and improve approaches for measuring total oxidant reactivity (principally OH and O_3); quantify the major contributions of known compounds to these totals and investigate any residuals; and characterize and understand regional and seasonal variations in canopy-scale emissions using airborne and tower networks of above-canopy flux measurements and satellite observations.
- *Oxidation*: Develop conceptual approaches to utilize mass balance of organic material in the atmosphere as a diagnostic tool to test current understanding, to predict compounds that should be in the atmosphere (oxidation products), and to search for them in a systematic way. Explore by experimental or modeling studies the possible formation of low-volatility products relevant to particle production by intense photochemical processes at daybreak, following the enrichment of trace gases in the nocturnal boundary layer.
- *Measurements*: Develop laboratory, in situ, and remote sensing techniques to scan the atmosphere for currently unmeasured compounds, to observe sums of compounds by functional classes and compare with measured individual species, to more broadly utilize comprehensive separation technology, and to quantify wet and dry atmospheric deposition of gas-phase and particle-phase organic molecules. Focus on the broad array of semi-volatile organic species present in the atmosphere, the majority of which are likely oxidation products of primary BVOC emissions and can potentially condense at secondary organic components of particles.
- *Modeling*: Develop models to constrain the chemical influence and fate of products from atmospheric BVOC reactions, to assess the importance of additional organic compounds for atmospheric photochemistry and secondary particle mass, and to represent the full range of

BVOCs and their gas-phase and particle-phase products in chemistry and climate simulations.

2.2.2. *New Particle Formation*

Near-surface measurements of particle number-diameter distributions and ion number-mobility distributions at many terrestrial sites around the globe—but excluding the Amazon Basin—show that new particle formation occurs frequently [*Hörrak et al.*, 1994; *Kulmala et al.*, 2004; *Laakso et al.*, 2007; *Iida et al.*, 2008]. Evidence for these events (outside the Amazon) is the appearance of neutral and electrically charged nanoparticles at diameters well below 10 nm and their subsequent growth to larger diameters. Measurements carried out in parallel at sites located distances of several hundred kilometers apart show that the events are often regional [*Stanier et al.*, 2004; *Vana et al.*, 2004; *Komppula et al.*, 2006]. In contrast, measurements in the Amazon Basin provide little evidence for near-surface regional-scale production of new particles [*Zhou et al.*, 2002; *Krejci et al.*, 2003; 2005; *Rissler et al.*, 2004; 2006]. Whereas in other continental locations, 3-nm particles are regularly observed at near-surface measurement sites and also seen to grow into the Aitken mode size range above 30 nm, in the Amazon Basin the smallest particles typically have sizes of 10 to 20 nm and continuous growth to larger diameters is rarely observed. Growth rates for Amazonia under pristine conditions have been reported as 5 nm hr^{-1} in one location for a limited set of measurements [*Zhou et al.*, 2002], implying that the observed ultrafine particles nucleated 2 to 4 hr prior to the observations. Further observations are needed, however, to define the possible variability of growth rates in the Amazon Basin.

A constraint on an observable new-particle mode is that freshly nucleated particles must grow more quickly than they are scavenged by coagulation with preexisting larger particles. The likelihood of satisfying this condition increases both with faster growth rates of the nucleated particles and with lower concentrations of preexisting particles [*McMurry and Friedlander*, 1979; *Kerminen and Kulmala*, 2002; *McMurry et al.*, 2005; *Lehtinen et al.*, 2007]. The constraint on an observable new-particle mode is typically satisfied for growth rates of 1 to 10 nm hr^{-1}. The condensation of sulfuric acid vapor typically accounts for only a fraction of this growth [*Fiedler* *et al.*, 2005; *Stolzenburg et al.*, 2005], with most of the balance due to condensation of VOC oxidation products [*Smith et al.*, 2008]. Under the assumption that the growth rate of 5 nm hr^{-1} reported by *Zhou et al*. [2005] can be broadly extrapolated to the Amazon Basin (i.e., both geographically and seasonally), the discussed constraint should be satisfied and nucleation events should therefore be observable as new-particle modes, and their absence in observations therefore suggests that near-surface nucleation is not widespread.

Some evidence for the Amazon Basin shows that nucleation occurs at high altitudes and that the entrainment of these particles to the near-surface layer explains surface observations of ultrafine particles. Aircraft measurements above Suriname in northern Amazonia observed enhanced ultrafine number concentrations at 2 to 4 km in regions of cloud outflow (Figure 13), suggesting nucleation [*Krejci et al.*, 2003; 2005], which is in broad agreement with observations of cloud outflow from other locations worldwide [*Perry and Hobbs*, 1994; *Clarke et al.*, 1998; 1999]. For comparison, measurements of vertical profiles with a tethered balloon in Melpitz, Germany, showed that nucleation first occurred aloft in the residual layer prior to breakup of the nocturnal inversion and then continued in the mixed layer during and after break-up, all in the absence of clouds [*Stratmann et al.*, 2003]. Over the boreal forests of Finland, a similar measurement program showed that nucleation occurred within the boundary layer but not aloft [*Laakso et al.*, 2007]. In the Amazon Basin, nocturnal events of apparent nucleation, which cannot be explained by outflow from deep convective clouds, have also been observed intermittently for short periods [*Rissler et al.*, 2004; 2006]. Diel patterns of intermittent nucleation were similar for a wide variety of conditions, including periods of intensive biomass burning as well as natural conditions. Nucleation-mode particle concentrations were highest at sunrise and sunset, with average concentrations exceeding 1000 cm^{-3} (Figure 14). Similar nocturnal events were observed in an Australian eucalypt forest [*Suni et al.*, 2008].

The implications of these observations for the Amazon Basin compared to those worldwide are that several different chemical processes may be capable of separately inducing nucleation and growth and, further, that these different processes may occur in different regions of the atmosphere. Further research, however, may yet succeed in unifying these presently disparate observations by using a mechanistic approach to the problem. A comprehensive analysis of particle formation events recorded at continental locations around the world shows that the nucleation rates J, which quantify the rates at which stable molecular clusters are produced, satisfy the following empirical expression [*Riipinen et al.*, 2007; *Kuang et al.*, 2008]: $J = k[H_2SO_4]^p$ for $1 < p < 2$, where $[H_2SO_4]$ is the sulfuric acid vapor concentration and k is a kinetic prefactor that varies from location to location. Mechanisms responsible for the variability in k are not yet understood. One hypothesis is that k accounts for the concentrations of species that co-nucleate with sulfuric acid. *Laaksonen et al*. [2008] have proposed that BVOC oxidation products may be important co-nucleating species over forested regions.

Of critical importance for the application of this nucleation equation to the near-surface layer over the Amazon Basin are the weak sulfur sources within the Basin, which lead to sulfur dioxide concentrations of typically 20 to 30 ppt [*Andreae and Andreae*, 1988; *Andreae et al.*, 1990a]. This value is more than an order of magnitude lower than the values commonly found under remote conditions over the continents of the Northern Hemisphere. This concentration of SO_2 plausibly implies correspondingly low gas-phase H_2SO_4 concentrations, although no direct observations have ever been made in the Amazon Basin to provide confirmation. Simulated peak daytime near-surface H_2SO_4 concentrations are less than 5×10^5 cm^{-3} (0.019 ppt) [*Spracklen et al.*, 2005]. By the above nucleation equation, this modeled H_2SO_4 concentration is too low to result in near-surface particle formation because preexisting particles will scavenge any incipient molecular clusters before they grow to new particles [*Spracklen et al.*, 2006]. An alternative mechanism to the H_2SO_4 pathway, namely ion-mediated nucleation, is also modeled as an unimportant source of nuclei over the Basin [*Yu et al.*, 2008].

In comparison to the absence of predicted new particle formation in the near-surface region of the Basin, models predict that new particle formation upwind or aloft, in particular within the upper troposphere followed by growth and entrainment into the near-surface layer, contributes significantly to the Amazonian particle number concentrations, especially during the wet season [*Spracklen et al.*, 2005]. Although sufficient for nucleation at higher altitudes, modeled H_2SO_4 concentrations are insufficient to explain the subsequent rate of particle growth observed in convective outflow over the Basin, suggesting that other gas-phase species such as the oxidation products of BVOCs may have a role [*Ekman et al.*, 2008].

Opportunities for progress to better constrain and quantify mechanisms of new particle formation over the Amazon include:

- *H_2SO_4 concentration*: Perform simultaneous observations of sulfuric acid vapor concentrations, particle nucleation rates, and particle number-diameter distributions over the Amazon Basin. Establish whether the apparent absence of surface level nucleation in the Basin is consistent with our understanding of the atmospheric conditions that lead to new particle formation in other locations [*McMurry et al.*, 2005].
- *Particle growth rates*: Observe and explain growth rates of ultrafine particles over the Basin. Quantify the contribution of sulfuric acid and BVOC oxidation products to the observed rates.
- *Ion mobility distributions*: Deploy an air-ion spectrometer [*Hõrrak et al.*, 1994; *Mirme et al.*, 2007] in the Basin to give information on very small particles (diameter < 3 nm) and the earliest steps of new particle formation.
- *Modeling*: Develop models to assess the contribution of different nucleation mechanisms to aerosol particles in the Basin. Evaluate candidate mechanisms by comparisons of model predictions made using these mechanisms against past and newly available observations.

3. Properties

3.1. Mass Concentration

The mass concentrations of particles in the Amazon Basin vary strongly with season and location, modulated to the largest extent by the influence of in-Basin biomass burning with other important influences by the episodic long-range transport of African dust and biomass burning [*Artaxo et al.*, 2002; *Guyon et al.*, 2003b]. Figure 15 shows the time series of PM_{10}

concentrations for central Amazonia (Balbina), eastern Amazonia (Santarem), and southern Amazonia (Alta Floresta). These locations are influenced seasonally in varying degrees by in-Basin biomass-burning emissions. In Alta Floresta (Aug 1992 to Feb 2005), two different prevailing regimes of mass concentration occur. In the wet season, in the absence of biomass burning, the PM_{10} concentration is 9 to 12 $\mu g\ m^{-3}$, with a fine fraction of 2 to 3 $\mu g\ m^{-3}$. In the dry season, the PM_{10} concentration approaches 300 to 600 $\mu g\ m^{-3}$, producing an optical thickness of more than 4 at 500 nm [*Schafer et al.*, 2008].

For comparison, in central Amazonia where the influence of biomass burning is less, the mass concentration is low even in the dry season (Figure 15, Balbina), with an average PM_{10} concentration of 11 $\mu g\ m^{-3}$ (Oct 1998 to Feb 2005). The typical concentration of fine particles increases from 2 $\mu g\ m^{-3}$ in the wet season to 4 $\mu g\ m^{-3}$ in the dry season. The corresponding fine-mode black-carbon-equivalent (BC_e; cf. §3.3.1) mass concentration ranges from 100 to 150 ng m^{-3} during the wet season and from 600 to 800 ng m^{-3} during the dry season. Abrupt pulses of relatively high mass concentration can occur in both the fine and coarse fractions (Figure 15, Balbina), and these pulses are explained by African dust outflow that reaches the observation site.

The influence of biomass burning on mass concentration in Santarem in eastern Amazonia is intermediate compared to Balbina and Alta Floresta. From March 2000 to January 2005 in Santarem, the PM_{10} concentration increases from about 10 $\mu g\ m^{-3}$ in the wet season to 40 $\mu g\ m^{-3}$ in the dry season. The fine fraction, typically as low as 2 $\mu g\ m^{-3}$ in the wet season, reaches 20 to 30 $\mu g\ m^{-3}$ in the dry season. During the wet season, the ratio of the fine-to-coarse fraction is lower for Santarem than Balbina, possibly suggesting an increased relative importance of out-of-Basin coarse-mode particles over eastern Amazonia. Figure 15 also shows that the influence of biomass burning is strongest in the fourth quarter of each year at Santarem whereas it is strongest in the third quarter at Alta Floresta. These seasonal patterns match those of vegetation fires in eastern and southern Amazonia, respectively, as is apparent in Figure 8.

Figure 16 (top) shows that the particle mass-diameter distribution is dominated for natural conditions by coarse-mode particles, corresponding to primary biological particles possibly coated by secondary material. These data were obtained by gravimetric analysis of the stages of a Multi-Orifice Uniform Deposit Impactor (MOUDI) during Mar and Apr 2008 as part of AMAZE-08 in central Amazonia [*Martin et al.*, 2009]. The selected data correspond to time periods during which the influence of sources outside of the Amazon Basin was weak [*Chen et al.*, 2009]. *Artaxo and Hansson* [1995] applied principal component analysis to the elemental composition and the mass concentrations recorded on five stages of a cascade impactor for various levels within the canopy and found that the concentrations of potassium and phosphorus, indicative of primary particles, were prominent in the coarse fraction, especially during the night.

The mass-diameter distribution shifts from the coarse to the fine fraction during times of strong influence by in-Basin biomass burning. Figure 16 (bottom) shows MOUDI measurements recorded during LBA-SMOCC in southwestern Amazonia (Rondônia). The total particle mass concentration was 154 µg m^{-3}, emphasizing the overwhelming influence of biomass-burning particles compared to any other types at the time of sampling. The figure shows that the mass-diameter distribution is heavily loaded in the accumulation mode, with a mass median aerodynamic diameter between 0.33 and 0.56 µm. Also apparent, however, is that the mass concentrations in the coarse fraction are similar between the top and bottom panels of Figure 16. The implication is that the mass concentration of coarse-mode natural biogenic particles (possibly coated by secondary materials) is similar in the dry and wet seasons as well as for different locations within the Amazon Basin.

Opportunities for progress to identify and quantify the processes that control mass concentrations and mass-diameter distributions of particles over the Amazon Basin include:

- Measurements show that long-range transport aerosol particles from Africa and the Atlantic Ocean are present within the Amazon Basin and at times dominate observations, yet the concentrations and distribution of these out-of-Basin particles are not fully understood, especially in relation to the extensive cloud development and opportunities for wet scavenging within the Basin. Long-term measurements at a minimum of three locations across the Basin along an east-west transect are therefore motivated to provide observations of concentration gradients. Such a data set would provide insights into the sources of particles from outside the Basin as well as sinks within the Basin, thereby placing new constraints on the processes affecting particle lifetime.

- Systematic measurements of the size-segregated composition of aerosol particles are scarce in the Amazon Basin. Such measurements are particularly needed in the remote and seasonally pristine areas in the western half of the Basin.

3.2. Number-Diameter Distribution

At times of prevailing natural conditions, particle number concentrations in the mixed layer of the Basin are 200 to 400 cm^{-3}, lower by 10 to 100 times than continental and urban concentrations elsewhere in the world having high anthropogenic influence. The baseline, low number concentrations in the Basin are derived from biogenic sources that have low variability and low concentration throughout the year, and strong deviations from this norm that are episodically observed at near-surface stations must be explained either by local pollution or long-range transport from distant sources [*Pauliquevis et al.*, 2007].

The particle number-diameter distributions have been measured in the Basin by techniques such as mobility particle sizing (diameter range of 3 to 850 nm) (e.g., *Zhou et al.* [2002]), optical particle counting (diameters 100 nm and larger) (e.g., *Krejci et al.* [2003]), and aerodynamic particle sizing (diameters of 500 nm and larger) (e.g., *Rissler et al.* [2006]). A typical number-diameter distribution measured by a differential mobility particle sizer (DMPS) operated during a period of natural conditions (i.e., CLAIRE-98 wet season in central Amazonia) is shown in Figure 17a [*Zhou et al.*, 2002]. Aitken and accumulation modes are apparent at 70 and 150 nm, respectively. The histograms of Aitken, accumulation-mode, and particle number concentrations for the observations from CLAIRE-98 are shown in Figures 17b-d, and the modal statistics are summarized in Table 2. The characteristic diameters of the nucleation, Aitken, and accumulation modes are well confined within diameter ranges, and the modes are clearly separated (Table 2). These results of CLAIRE-98 are comparable to those of CLAIRE-01 [*Rissler et al.*, 2004] at the same site and of AMAZE-08 about 100 km away (Figure 18, top) (E. Swietlicki, unpublished data).

In Figure 17 a Hoppel gap, which is persistent in Amazonian number-size distributions, separates the Aitken and accumulation modes and differentiates to a large extent those particles that have been subjected to in-cloud processing from those that have not. According to *Hoppel et al*. [1986; 1994], cloud-droplet activation on Aitken particles is followed by the uptake and reaction of soluble gases to form low-volatility products, and net diameter growth therefore occurs upon evaporation of the cloud droplets. In the Amazon Basin, the soluble gases that become fixed are expected mostly to be BVOCs, which then react in the cloud waters to form at least in part low-volatility BVOC oxidation products (i.e., SOA particle components) [*Blando and Turpin*, 2000; *Lim et al.*, 2005; *Carlton et al.*, 2006]. Alternative mechanisms for converting Aitken particles into the accumulation mode, such as out-of-cloud coagulation and condensational growth, are too slow in clean atmospheres such as the green-ocean Amazon.

In fair weather, a diel pattern during CLAIRE-98 in the number concentration of the two modes, specifically that the Aitken number concentration decreased while that of the accumulation mode increased as the day progressed, was linked to the diel variation of the lower atmosphere (Figure 6) [*Zhou et al.*, 2002]. Cloud processing above the boundary layer leads to the depletion of Aitken particles and to the growth of accumulation particles in that layer, and this cloud convective layer mixes into the surface layer during the day, strongly influencing observations there. In rainy weather, the behavior is interrupted by strong scavenging of particle number, volume, and mass, followed by quick recovery (due to regional mixing) after rainfall ceases.

Measurements in CLAIRE-98 showed that the diameters of both the Aitken and accumulation modes continuously increased from sunrise to sunset, with few exceptions. The growth of the accumulation-mode particles was attributed to cloud processing, with downward mixing of these larger particles throughout the day. The growth of the Aitken particles (ca. 5 nm hr^{-1}) was plausibly explained by the condensation of low-volatility vapors resulting from the photooxidation of BVOCs (i.e., SOA production) (cf. §2.2.1). Much of this particle growth was proposed to occur in the boundary layer itself. Rainfall temporarily halted the steady diameter growth of the Aitken particles. This observation suggested a down-mixing of somewhat smaller Aitken particles in association with cold downdrafts.

These near-surface observations in central Amazonia during CLAIRE-98, CLAIRE-01, and AMAZE-08 were made on air masses that had spent several days within the Basin and thus were highly processed (e.g., ecosystem emissions, cloud cycles, and so forth). For contrast, measurements made during flights over Suriname and French Guyana during CLAIRE-98 were taken as air masses first entered the Amazon Basin and to some extent can be considered the initialization conditions for processing within the Basin [*Krejci et al.*, 2003]. The average number-diameter distribution from the lowest flight level within the well-mixed boundary layer (0.2 - 1.2 km) is shown in Figure 18 (bottom). This distribution is depleted in Aitken particles compared to the near-surface measurements described for central Amazonia (e.g., CLAIRE-98) (cf. top and bottom of Figure 18), offering evidence for the formation of Aitken particles within the Basin.

The particle number-diameter distribution changes greatly at locations in the Basin that are influenced by in-Basin biomass burning [*Reid et al*., 2005]. The mode of the distribution is typically between 100 and 160 nm. The higher values are found in more aged pollution plumes as a result of coagulation with other particles and the condensation from the gas phase of low-volatility species. Given these high concentrations, when they are present, aerosol particles resulting from in-Basin biomass burning dominate the overall features of the Amazonian aerosol, tending to minimize the impact of other processes such as particles imported from outside of the Basin or the processes of the natural Amazonian biosphere.

Measurements using a DMPS conducted at a ground site during LBA-SMOCC showed a single number median diameter of 135 nm for fresh smoke and average particle number concentrations of 10,500 cm^{-3} for the diameter range of 30 to 850 nm [*Rissler et al.*, 2006]. The number concentration of particles in the nucleation mode (i.e., from 3 to 30 nm diameter) was also relatively high, averaging 800 to 1000 cm^{-3}, although their presence was intermittent. Airborne measurements of particle number-diameter distributions were also performed [*Guyon et al.*, 2005], and the geometric mean diameters were 110 ± 15 nm in 69 plumes within the boundary layer and 139 ± 17 nm for 50 smoke plumes detrained above the boundary layer, mostly from non-precipitating clouds. Biomass-burning particles that enter higher altitudes and escape wet deposition can be exported out of the Basin, affecting particle number and mass concentrations in distant regions of the Southern Hemisphere.

Opportunities for progress in identifying and quantifying the processes that control the number concentration and the number-diameter distribution of particles in the Amazon Basin include:

- *Processes*: The processes that control the number-diameter distribution over the Amazon Basin are not fully understood. The consistent appearance of the Aitken and accumulation modes in confined diameter windows points to the importance of in-cloud processing, but the source and sink rates of Aitken and accumulation-mode particles, as well as the vertical structure and mixing of particles (including quantifying of cloud convective mixing and associated downdrafts as mechanisms for entraining particles into the boundary layer), must yet be quantified.
- *Measurements and Instrumentation:* Long-term (i.e., years) size-distribution measurements in a pristine environment using an air ion spectrometer, twin scanning mobility particle sizers, ultraviolet and normal aerodynamic particle sizers, preferably both just over the canopy as well as higher on a tall tower to observe vertical gradients. Size-resolved particle number fluxes by eddy-covariance techniques, preferably at several altitudes in a high tower. Tethered balloons equipped with condensation particle counters having various lower-size cutoff diameters or diffusion batteries to locate altitudes having increased new particle formation. Ground-based long-term LIDAR measurements for vertical profiling. These instruments should be applied to closure studies between (i) number and mass, (ii) the

number-diameter distribution and light scattering, and (iii) the number-diameter distribution (including hygroscopic properties) and the concentration of cloud condensation nuclei.

3.3. Chemical Composition

Aerosol particles in the Amazon Basin are composed mainly (i.e., 80 to 90%) of organic carbon (OC), although African mineral dust and Atlantic sea salt can dominate the mass concentration for short periods [*Talbot et al.*, 1990].

3.3.1. Organic Component

At times of prevailing natural conditions, typical OC concentrations are 1 $\mu g{\cdot}C\ m^{-3}$ or less in the fine fraction and 1 to 3 $\mu g{\cdot}C\ m^{-3}$ in the coarse fraction [*Formenti et al.*, 2001; *Graham et al.*, 2003a; *Guyon et al.*, 2003b]. A ratio of 1.6 to 1.7 for OM:OC (i.e., the ratio of organic mass to organic carbon) is estimated from the high-resolution mass spectra of *Chen et al.* [2009] and in agreement with value of *Fuzzi et al*. [2007]. Molecular characterization by chromatography has been carried out for the water-soluble organic fraction [*Graham et al.*, 2003b; *Claeys et al.*, 2004]. Results are shown in Figure 19 for particles collected on fine-fraction filters [*Decesari et al.*, 2006]. The identified composition (0.10 $\mu g{\cdot}C\ m^{-3}$) is less than 20% of the water-soluble organic carbon (WSOC; 0.85 $\mu g{\cdot}C\ m^{-3}$), indicating the presence of a wide range of other unidentified organic compounds. Dicarboxylic and hydroxyacids are persistently higher during the day compared to night, consistent with the condensation from the gas phase of BVOC oxidation products [*Graham et al.*, 2003a]. Likewise, the detection of methyltetrols is a signature of isoprene photooxidation [*Claeys et al.*, 2004]. BVOC oxidation products may also explain the elevation of fine-fraction OC concentrations by 1.4 ± 0.2 times during the day compared to the night (i.e., greater plant emissions and greater sunlight during the day), although enhanced convective downward mixing of particles from aloft can also play an important role [*Graham et al.*, 2003a, b; *Claeys et al.*, 2004]. The presence of low levels of anhydrosugars (such as levoglucosan, mannosan, and galactosan) even during time periods for which natural conditions prevail demonstrates an influence of biomass burning [*Graham et al.*, 2003a], possibly indicative of the long-range transport of emissions from African fires.

For AMAZE-08, *Chen et al.* [2009] report the results of real-time mass spectrometric measurements of submicron particles for a time period of natural conditions. During periods of weak out-of-Basin influence, patterns and identifier peaks in the mass spectra closely resemble those of secondary particle components formed by the oxidation of BVOCs in environmental chambers. Most of the mass concentration of submicron organic particles is attributed for the period of AMAZE-08 to the condensation of BVOC oxidation products as secondary particle components, with a smaller amount to highly oxidized materials representative of humic-like substances (HULIS) that arrive by long-range transport (e.g., aged biomass burning emissions from Africa).

In regard to the coarse fraction, *Graham et al*. [2003a] quantify sugars, sugar alcohols, and fatty acids, providing strong evidence for the release of primary biological particles into the forest atmosphere. Trehalose, mannitol, arabitol, and the fatty acids are more prevalent at night, coinciding with a nocturnal biological activity that increases the release rates of yeasts and other small fungal spores. Glucose, fructose, and sucrose are persistently higher during the day, coinciding with a daytime increase in large fungal spores, fern spores, pollen grains, and, to a lesser extent, plant fragments, as driven by lowered relative humidity and enhanced wind speeds and convective activity during the day. Although mass emissions are reduced at night, coarse-fraction OC concentrations are, nevertheless, elevated at night compared to day by a mean factor of 1.9 ± 0.4, which is explained by trapping of emitted particles in the nocturnal boundary layer.

The organic compounds constituting the natural particles can be light absorbing. Although black-carbon-equivalent (BC_e) concentration is typically less than 0.1 $\mu g\ m^{-3}$ and represents under 5% of the total carbon concentration [*Formenti et al.*, 2001; *Graham et al.*, 2003b], the BC_e concentrations are higher than those of elemental carbon (EC), implying that organic components are contributing to the absorption of light [*Guyon et al.*, 2003a; c]. Because these compounds have a steep increase of light absorption with decreasing wavelength, resulting in a brown color of the filter samples, they have been termed “brown carbon” [*Andreae and Gelencser*, 2006]. The light-absorbing material is mainly in the coarse fraction and can be explained mostly by chromophores present in primary biological particles and certain BVOC oxidation products.

In addition to natural particles, the composition of biomass-burning particles in the Basin has also been extensively studied [*Penner et al.*, 1991; *Andreae*, 1993; *Falkovich et al.*, 2005; *Decesari et al.*, 2006; *Fuzzi et al.*, 2007]. The biomass-burning particles, found mostly in the fine size fraction (cf. §3.2), are most predominant in southern Amazonia and downwind of it (Figure 5), but even remote areas in northern and central Amazonia are subject to the large-scale transport of biomass-burning emissions [*Pauliquevis et al.*, 2007]. The particles consist of 85 to 90% organic carbon [*Talbot et al.*, 1990; *Yamasoe et al.*, 2000; *Soto-García et al.*, 2009], of which more than half is water soluble [*Graham et al.*, 2002; *Mayol-Bracero et al.*, 2002; *Decesari et al.*, 2006]. The balance of 10 to 15% has typically been operationally defined as black carbon (i.e., apparent elemental carbon). The OC and BC_e concentrations have a diel variability arising both from variations in the thickness of the atmospheric boundary layer and the frequency of fires [*Soto-García et al.*, 2009]. Biomass-burning particles also are an important source of water-soluble organic nitrogen, including urea and several amino acids. For example, *Mace et al*. [2003] report concentrations of 0.9 $\mu g{\cdot}N\ m^{-3}$ in the dry season compared to 0.05 $\mu g{\cdot}N\ m^{-3}$ in the wet season (i.e., a 20-fold difference).

Figure 20 shows the composition of biomass-burning particles in Amazonia, as obtained from a synthesis of data from multiple complementary techniques (e.g., gas-chromatography mass spectrometry (GC-MS) and high-pressure liquid chromatography (HPLC)) [*Mayol-Bracero et al.*, 2002].The organic compounds are a complex mixture of differing molecular structures, physical properties, and reactivities [*Jacobson et al.*, 2000]. Molecular speciation using GC-MS accounts for about 10% of the WSOC. The identified species are mostly pyrolysis products of cellulose, hemicellulose, and lignin [*Graham et al.*, 2002; *Zdrahal et al.*, 2002; *Claeys et al.*, 2004; *Trebs et al.*, 2005; *Decesari et al.*, 2006]. Levoglucosan, a primary vegetation combustion product, is the single most abundant compound identified [*Schkolnik et al.*, 2005; *Fuzzi et al.*, 2007]. It is enriched in samples collected at night compared to those from the day, reflecting the shift from flaming fires in the day to smoldering fires at night [*Fuzzi et al.*, 2007]. The balance of about 90% that eludes analysis by molecular chromatography is expected to be chemical compounds that are predominantly of high molecular weight [*Mayol-Bracero et al.*, 2002; *Hoffer et al.*, 2006]. The HPLC results show that neutral molecules, mono and dicarboxylic acids, and polycarboxylic acids represent about 70% of the WSOC. *Decesari et al.* [2006] propose model compounds to reproduce quantitatively the average chemical structure of the WSOC, with the intention that these model compounds can be used as best-guess surrogates in microphysical models.

Fuzzi et al. [2006] provide excellent recommendations in a general context on the research needs for improved chemical characterization of organic aerosol particles. For the specific context of the Amazon, high-priority opportunities for increased chemical characterization include:

- *Sampling*: Use of sampling techniques such as denuders and real-time measurements that reduce positive and negative sampling artifacts. As necessary, the development of new techniques for these purposes. Control of sampling with wind direction to facilitate the interpretation of the results. Increased use of size-segregated sampling.
- *Analysis*: Development of analytical methods to improve chemically resolved mass balance, including both at the molecular and common-property levels. Innovation of new methods for the analysis of high-molecular weight compounds. Determination of the composition of light-absorbing material. Development of analytical techniques for airborne measurements having a similar capability as ground-level measurements.
- *Composition*: Identification and characterization of the molecules and the molecular families constituting the water-<u>in</u>soluble organic fraction. Determination of the relative contributions of humic-like substances and BVOC oxidation products to OC mass concentrations during the wet and dry seasons. Characterization and improvement of the understanding of water-soluble organic nitrogen in biomass-burning particles.

3.3.2. *Inorganic Component*

Table 3 summarizes the composition of the inorganic component of Amazonian aerosol particles. The inorganic component typically constitutes 10 to 20% of the total mass in the fine fraction and less in the coarse fraction, with the balance largely from organic components and at times African dust and African and South American biomass burning.

The two most commonly applied techniques for the study of the inorganic composition have been off-line ion chromatography and proton-induced X-ray emission (PIXE). Samples were collected for at least half a day up to several days on a single filter to obtain sufficient quantities for analysis. The SMOCC campaign (Table 1) introduced the use of selective semi-continuous measurements of water-soluble inorganic compounds (i.e., SO_4^{2-}, NO_3^-, Cl^-, and NH_4^+) via a steam-jet aerosol collector and their gaseous precursors (i.e., SO_2, HNO_3, HCl, NH_3) by use of a rotating wet-annular denuder [*Trebs et al.*, 2004; 2005]. In AMAZE-08, an Aerodyne aerosol mass spectrometer was employed to quantify non-refractory sulfate, nitrate, and ammonium every 5 min (Figure 21) [*Chen et al.*, 2009].

Sulfate is the major water-soluble inorganic anion and is primarily distributed in the fine fraction. Sulfate is formed by the reactions of DMS, H_2S, and CS_2 emitted by plants and microorganisms within the Amazon Basin, and the in-Basin source contributes approximately 0.05 $\mu g\ m^{-3}$ to the sulfate mass concentration [*Andreae et al.*, 1990a]. Even for natural conditions, however, sulfate concentrations averaged over several weeks are 3 to 5 times greater than this in-Basin contribution (Table 3) [*Artaxo et al.*, 1990; *Formenti et al.*, 2001; *Artaxo et al.*, 2002; *Fuzzi et al.*, 2007]. Part of the explanation is that marine DMS is persistently imported from the Atlantic Ocean, representing an integral part of the natural processes of the Basin. Episodic importation of sulfate included as part of African dust and biomass-burning particles also occurs, and these episodes can increase sulfate concentrations in both the fine and coarse fractions by factors of 2 to 3 or more [*Talbot et al.*, 1990; *Formenti et al.*, 2001]. In the dry season, the average sulfate concentration increases by a factor of 2 to 3 (Table 3), even for nominally clean conditions. The increase is attributed in large part to the reduction in wet deposition as well as to the presence of biomass-burning particles diluted throughout the Basin, rather than a change in biogenic gaseous emissions or shifts in the patterns of imported sulfate precursors [*Artaxo et al.*, 1988; *Talbot et al.*, 1988; *Artaxo et al.*, 2002; *Graham et al.*, 2003a; *Fuzzi et al.*, 2007]. At sampling locations and times repeatedly and heavily affected by biomass burning (e.g., in plumes), the average sulfate concentration can increase by a factor of 10 or more.

Nitrate is found predominately in the coarse fraction of natural Amazonian aerosol particles and, compared to sulfate, is a minor component by mass (Table 3) [*Talbot et al.*, 1988; 1990; *Graham et al.*, 2003a; *Fuzzi et al.*, 2007]. The ambient HNO_3 concentrations are too low and the temperatures typical of the Basin are too high to favor elevated concentrations of particle-phase nitrate. Significant nitrate enrichment to the fine fraction occurs, however, for locations affected by in-Basin biomass burning, which can be attributed to increased NO_x emissions followed by oxidation and subsequent condensation of HNO_3 [*Trebs et al.*, 2004]. At these times, the nitrate and sulfate concentrations can be nearly equal [*Trebs et al.*, 2005].

Ammonium concentrations in the fine and coarse fractions are typically more than sufficient during both the wet and dry seasons to neutralize the nitrate and sulfate in each size fraction, particularly at night [*Talbot et al.*, 1988; *Graham et al.*, 2003a; *Mace et al.*, 2003; *Fuzzi et al.*, 2007]. *Trebs et al*. [2005] conclude that organic anions, such as formate, acetate, and oxalate, must balance the ammonium in the particles, implying the presence of species such as ammonium oxalate. At locations having a strong influence from biomass burning, ammonium concentrations, along with those of nitrate, are elevated in the fine fraction during the night [*Trebs et al.*, 2004], which can be explained by higher concentrations of HNO_3 and NH_3 in the vicinity of biomass burning and favorable gas-to-particle partitioning for higher relative humidity and cooler temperatures.

The mass concentrations of crustal elements (e.g., Al, Si, Fe, Ti, and Mn) are low in the absence of episodic inputs of African mineral dust (cf. Table 3) [*Talbot et al.*, 1990; *Formenti et al.*, 2001; *Graham et al.*, 2003a; *Trebs et al.*, 2005]. During episodes, their concentrations are equally distributed in the fine and coarse fractions, representing a fractionation that is significantly different from the dominance of the coarse mode near source regions. After long transport over the Atlantic Ocean, the coarse mode of these elements is lost preferentially over the fine mode, resulting in the observations seen for the Amazon, including an important tail into the accumulation mode. Wet-season concentrations are several times higher than those during the dry season, which is explained by a shift of the ITCZ that favors the transport to the Amazon Basin of air from the Saharan region (cf. §1).

The mass concentrations of P, K, and Zn in the coarse fraction are derived primarily from PBA particles [*Artaxo and Hansson*, 1995; *Guyon et al.*, 2003c], as evidenced by their greater concentrations underneath the forest canopy compared to above it and by their greater concentrations at night than day (cf. §3.3.1). Although the long-range transport of phosphorus from African dust is important for the fertilization of the Basin on long timescales [*Mahowald et al.*, 2005], the incremental concentrations are low and difficult to detect compared to the in-Basin cycling of phosphorus by PBA particle emission and deposition [*Formenti et al.*, 2001]. For natural conditions, K occurs almost exclusively in the coarse fraction, but a fine mode appears at times of biomass-burning influence, increasing the mass concentration by a factor of 10 or more in fresh plumes downwind of biomass burning [*Echalar et al.*, 1998; *Yamasoe et al.*, 2000; *Artaxo et al.*, 2002; *Maenhaut et al.*, 2002; *Trebs et al.*, 2005]. Fire is therefore important for the recycling of K throughout the Basin, and its presence in the fine fraction is often used as a marker for the influence of biomass burning.

High priorities for the improved chemical characterization of the inorganic component of Amazonian aerosol particles include:

- *Instrumentation*: Positive and negative sampling artifacts can be introduced, especially for semi-volatile compounds like ammonium nitrate, by swings in temperature and relative humidity during the long sampling periods required by many techniques when employed at the low particle mass concentrations characteristic of the Amazon Basin [*Solomon and*

Sioutas, 2008; *Trebs et al.*, 2008]. The development of instrumentation that can overcome these obstacles in tropical environments is needed.

- *Processes*: Investigations are needed (1) that target the influence of water-soluble organic compounds on the gas-particle partitioning of inorganic species, (2) that test for the possible presence of organosulfates and understand how these compounds may be tracers for the sources of particle components, and (3) that focus on connections between inorganic compounds and their physical properties (e.g., their activity as cloud condensation nuclei).
- *Measurements*: Long-term measurements are needed for a better understanding of seasonal and annual variability, especially the effects of the long-range transport of African dust and biomass-burning emissions. Measurements with improved geographic coverage are needed from the eastern edge of the Basin into central parts to quantify gradients in African and Atlantic aerosol particles and thereby to understand better their influence.

3.4. Hygroscopicity

The hygroscopic properties of submicron Amazonian aerosol particles have been studied both for the wet season during periods of weak out-of-Basin influence [*Zhou et al.*, 2002] and for the dry season at times of strong in-Basin biomass burning [*Rissler et al.*, 2004; 2006]. Irrespective of season and the air mass type, the hygroscopic diameter growth factor measured at 90% relative humidity (RH) by use of a tandem differential mobility analyzer is typically 1.05 to 1.35, with few exceptions. This finding differentiates Amazonia from rural sites on other continents, for which “highly hygroscopic” particles having growth factors of 1.7 that approach those of inorganic salts are observed for at least a fraction of the particles [*Swietlicki et al.*, 2008]. The consistently high organic fraction of submicron particles (cf. §3.3) can explain the absence of highly hygroscopic particles in the Amazon Basin. As the exception, “highly hygroscopic” particles have, however, been observed in the Amazon Basin at least once [*Zhou et al.*, 2002], plausibly corresponding to the presence of marine particles imported with an Atlantic air mass [*Formenti et al.*, 2001]

For natural conditions, “moderately hygroscopic” particles dominate the submicron particle population in the Amazon Basin [*Zhou et al.*, 2002; *Rissler et al*. 2004]. *Zhou et al.* [2002] find that the hygroscopic growth factor increases from 1.17 at 35 nm to 1.32 at 264 nm. Ammonium-bisulfate dry-volume fractions ranging from 0.17 at 35 nm to 0.27 at 265 nm, with the balance of the dry-volume fraction corresponding to an insoluble core, can equivalently represent the observed hygroscopic growth. This equivalent representation must not, however, be interpreted as implying that the water-soluble components of the real particles are composed solely of ammonium and bisulfate ions. Water-soluble organic compounds constitute a large fraction of the particle components (cf. Figures 19 and 20), although many of the substances, such as the larger dicarboxylic acids, fulvic acids [*Svenningsson et al.*, 2006], or humic-like substances (HULIS) [*Ziese et al.*, 2008], have low water uptake. There is also the important possibility of a difference between the WSOC fraction measured for relatively dilute aqueous solution (i.e., as represented in Figures 19 and 20) and that relevant to the lower water activity of 90% RH at which measurements of hygroscopic growth have been made.

For locations strongly influenced by fresh in-Basin biomass burning, an external mixture of “moderately hygroscopic” (as described above) and “barely hygroscopic” particles is observed [*Rissler et al.*, 2006; *Vestin et al.*, 2007]. “Barely hygroscopic” particles have growth factors from 1.06 at 20 nm to 1.12 at 440 nm, corresponding to inorganic-equivalent dry-volume fractions of approximately 0.07. The “barely hygroscopic” particles can dominate the number balance of the external mixture by a factor of 5 to 10, with a larger fraction at smaller particle sizes. Open-air biomass burning produces particles largely composed of organic components that have a limited propensity for water uptake.

In an advance compared to the inorganic-equivalent representation for hygroscopicity, *Mircea et al*. [2005] provide a more comprehensive treatment of chemical hygroscopic closure that includes a treatment of the organic component. Growth factors observed during LBA-SMOCC (i.e., moderately to strongly influenced by biomass burning) for particles 420 nm and smaller were compared with the predictions of a water-uptake model that incorporated the size-segregated chemical composition. Inorganic components accounted for about 10% of the size-segregated mass concentrations, and the balance was carbonaceous [*Fuzzi et al.*, 2007]. Water-soluble organic compounds constituted 50 to 60% of the mass concentrations [*Decesari et al.*, 2006]. The water-uptake model used for the hygroscopic closure simplified the organic composition by choosing nine model compounds derived from functional group analyses and other analytical techniques (cf. further description in §3.3). The model, combined with the size-segregated chemical composition, accurately predicted the measured growth factors for an assumption of limited solubility of the organic compounds at 90% RH. Closure could not be obtained for other candidate assumptions, including complete solubility or complete insolubility.

Priorities for progress to better constrain the hygroscopic behavior of Amazonian aerosol particles include:

- Amazonian biomass-burning particles were extensively characterized during the LBA-SMOCC experiment in 2002. Less is known about natural particles in pristine rain forest environments, essentially based on 18 days of data from *Zhou et al*. [2002] and 2 days of data from *Rissler et al*. [2004], and further measurements are therefore highly motivated with a focus on understanding temporal and geographic variability.
- The links between size-segregated chemical composition and hygroscopic behavior, for instance by implementing new instrumentation such as aerosol mass spectrometers, as well as between hygroscopic growth and radiative properties, for instance by conducting experiments using humidity-controlled nephelometers, should be pursued.
- The presence of PBA particles should be quantified by utilizing the ability of a tandem differential mobility analyzer to determine the hygroscopic growth of individual particles. The intermittent appearance of externally mixed "barely hygroscopic" particles during the wet-season of *Zhou et al*. [2002] (i.e., a frequency of occurrence of 5-9% in the Aitken mode and 11-14% in the accumulation mode) might indicate an influence from PBA particle sources, although anthropogenic pollution was not entirely ruled out as having an influence during that study.

3.5. Cloud Condensation Nuclei

The concentrations of cloud condensation nuclei (CCN), in the absence of an influence by in-Basin biomass burning, are 250-300 cm^{-3} for 1% supersaturation [*Roberts et al.*, 2001; 2002; *Andreae et al*., 2004; *Gunthe et al*., 2009]. These CCN concentrations are comparable to the total particle concentration, and they are lower than typically observed for other rural sites worldwide, a finding which is indicative of strong anthropogenic influence for most other continental locations worldwide [*Andreae*, 2007; 2009]. The CCN properties for natural conditions can be described in large part by an effective CCN hygroscopicity parameter $\kappa = 0.15$ [*Gunthe et al*., 2009]. Such particles are sufficiently hygroscopic that they activate at supersaturations of 0.1 to 0.3% that typically occur within clouds [*Zhou et al.*, 2002; *Svenningsson et al.*, 2006]. For comparison, κ is larger by a factor of two for other continental locations [*Andreae and Rosenfeld*, 2008]. The lower κ in Amazonia is consistent with properties of SOA material reported in laboratory studies [*King et al*., 2007, 2009; *Prenni et al*., 2007] and with the report of *Chen et al*. [2009] for AMAZE-08 that high proportions of secondary organic matter constitute the components of submicron particles. Supermicron PBA particles present at relatively low number concentrations can also be important under some circumstances by serving as “giant” CCN, which activate at supersaturations below 0.1% because of their large diameters and enhance the collision-coalescence stage of precipitation formation, especially under polluted conditions [*Yin et al.*, 2000]. PBA particles are also an important source of ice nuclei in the Basin [*Prenni et al*., 2009].

In stark contrast to the low CCN concentrations observed for natural conditions, regions affected by in-Basin biomass burning can have CCN concentrations on the order of 10,000 cm^{-3} or more (Figure 22) [*Roberts et al.*, 2003; *Rissler et al.*, 2004; *Vestin et al.*, 2007]. The contribution to CCN number concentration arises not only from the increase in particle number concentration but also by increases in mode diameter and in water-soluble fraction, both of which further favor CCN activation [*Mayol-Bracero et al.*, 2002; *Decesari et al.*, 2006; *Fuzzi et al.*, 2007]. For the observations of LBA-SMOCC, *Mircea et al.* [2005] show that CCN closure in the supersatured regime (i.e., greater than 100% RH) is best achieved by assuming complete water solubility of organic species at the high water activities of CCN activation.

The difference in CCN concentrations between natural and anthropogenically influenced conditions has significant consequences on the microphysical properties of clouds, particularly the average droplet diameter, the maximum in-cloud supersaturation, and the precipitation dynamics. Microphysical properties are most susceptible to increasing CCN concentrations for low base concentrations, such as those of natural Amazonia. The input of additional particles from biomass burning greatly alters the pathways of cloud development. When smoke plumes spread over large areas, shallow clouds are inhibited, causing a reduction in cloud cover [*Koren et al.*, 2004; 2005]. *Feingold et al.* [2005] and *Jiang and Feingold* [2006] also suggest that the extinction of radiation by elevated particle concentrations in the middle troposphere reduces the surface heat flux, thereby stabilizing the boundary layer and further reducing cloud cover. Furthermore, enhanced CCN concentrations that result from biomass burning reduce the cloud droplet diameter below the collision-coalescence threshold, an effect which reduces warm-cloud precipitation [*Andreae et al.*, 2004]. Another potential effect of increased CCN concentrations, which is to enhance cloud albedo as a result of both the smaller droplet diameter and more numerous droplets [*Twomey*, 1977], is small or negligible in the Amazon Basin because the clouds are usually already optically thick [*Platnick and Twomey*, 1994; *Roberts et al.*, 2003].

The range of microphysical regimes observed in the Basin, including blue-ocean, green-ocean, smoky-, and pyro- clouds, is illustrated by the mass-diameter distributions of liquid water content (Figure 23) [*Andreae et al.*, 2004]. There is a narrowing of the distributions and a slowing of their rate of broadening with increasing height for the progressively more particle-rich regimes from *A* to *D*. For low CCN concentrations (panels *A* and *B*), the droplet distributions over the ocean and the Amazon Basin grow and broaden in a similar manner as the parcel rises. In contrast, for the very high CCN concentrations of pyroclouds (panel *D*) that form in the invigorated updrafts of the smoke plume over an active fire, the droplet distribution stops growing once the air parcel rises above a critical altitude (e.g., above 2800 m in Figure 23). The stunted growth is explained by reduced in-cloud supersaturation that inhibits droplet growth and has the consequence of suppressing precipitation. Pyro-clouds embedded in a smoky atmosphere also reduce ground heating by blocking sunlight, and they therefore reduce convective vigor and precipitation. Smoky clouds (panel *C*) represent an intermediate case compared to green-ocean and pyro- clouds. In smoky clouds, the altitude for the onset of precipitation, which corresponds to a modal diameter of the liquid water content that is greater than an approximate threshold of 24 μm, shifts higher compared to natural conditions. The consequence is that rain either does not occur or occurs from higher altitudes, including more precipitation initiated through the ice phase. Consequently, there is a vertical redistribution of released heat and, in the case of ice, invigorated convection and lightning [*Rosenfeld et al.*, 2008].

The need to understand how the CCN activity of organic particles evolves and the associated implications for direct and indirect radiative forcing have been highlighted in general reviews [*Kanakidou et al.*, 2005; *Fuzzi et al.*, 2006; *McFiggans et al.*, 2006; *Andreae and Rosenfeld*, 2008]. Specific priorities for better understanding and predicting the CCN activity of Amazonian aerosol particles and their effects on climate include:

- *Measurements*: Data sets of CCN activity in the Basin are restricted both in time and space and can be considered sparse in comparison to the seasonal and spatial heterogeneities of the Amazon Basin. Most measurements in available data sets were carried out during intensive campaigns lasting a short number of weeks at a single location [*Roberts et al.*, 2002; *Rissler et al.*, 2004; *Vestin et al.*, 2007; *Gunthe et al.*, 2009]. Long-term and distributed measurements are therefore highly motivated so that seasonal and annual cycles can be related to other particle parameters, such as chemistry and diameter, as well as to different meteorological conditions.
- *Organic components of CCN activity*: Particle CCN activity depends both on the physical and chemical properties of the particles, especially particle diameter and the water-soluble fraction [*McFiggans et al.*, 2006]. The understanding of the mechanistic processes of CCN activation, in particular variability in space and time of the organic chemical speciation and

most importantly in the water-soluble fraction of Amazonian aerosol particles (cf. §3.3.1), must be improved.

- *Modeling*: Models should be developed that connect the sources of particles and their components with CCN activity, including the further evolution as cloud droplets. This approach should be added to cloud-resolving models, such as the Regional Atmosphere Modeling System (RAMS) or the Weather and Research Forecasting (WRF) model, and large-eddy simulations should be carried out. Interconnections among vegetation, particle chemistry, cloud microphysics, and dynamics can thereby be investigated.

4. Outlook and Future Priorities

The Amazonian rain forest, having a present area of greater than 5×10^6 km^2 and corresponding to 85% of its original extent, is the largest on Earth. The future of Amazonian aerosol particles is directly connected to the fate of this forest. Before the 1950s, less than 1% of the area had been deforested. At present, approximately 0.02×10^6 km^2 is deforested annually, mostly accomplished by biomass burning. The fate of the forest depends largely on two interacting factors, including (1) the rates and the types of human development in the region and (2) the influences of ongoing global climate change [*Laurance and Williamson*, 2001; *Cardoso et al.*, 2003; *Lewis*, 2006; *Soares-Filho et al.*, 2006; *Malhi et al.*, 2008; *Nepstad et al.*, 2008]. At present, deforestation is largely illegal and driven by business opportunities for cattle ranchers and soy farmers. In the near future, several of the ruling governments plan to construct a network of crisscrossing roads through the forest (Figure 24). Road access, especially when paved, is the first step of illegal deforestation [*Laurance et al.*, 2001; *Nepstad et al.*, 2008], and forest fragmented by human activities is also more susceptible to drying and subsequent burning, resulting in positive feedbacks between deforestation and wildfire [*Laurance and Williamson*, 2001; *Cardoso et al.*, 2003]. Deforestation by business-as-usual policies is expected to drive cumulative loss of the forest from 15% in 2003 of the original extent to 40% by 2050 (Figure 25) [*Soares-Filho et al.*, 2006]. Even with good governance, however, ongoing global climate change might nevertheless induce large-scale drought [e.g., *Marengo et al.*, 2008]. According to some coupled climate-carbon models, droughts might lead to the loss of most forest in the Amazon Basin and to the conversion to savanna grassland or desert, regardless of activities within political Brazil [*Betts et al.*, 2004; *Saleska et al.*, 2007; *Cox et al.*, 2008]. Some models also indicate that a "tipping point" might occur for a fractional forest cover that falls below a critical value, meaning that deforestation might serve as a feedback that decreases rainfall and thus accelerates deforestation and further reductions in rainfall (i.e., at present total evapotranspiration approaches 50% of total precipitation), ultimately transforming the region from a rain forest biome to a savanna [*Silva Dias et al.*, 2002; *Oyama and Nobre*, 2003; *Nobre et al.*, 2004]. A possible fate of large-scale forest loss has paradigm-shifting implications for the concentrations and the properties of particles as well as for meteorology, cloud type, and rainfall intensity.

According to projections by *Streets* [2007] of the future of the Amazon Basin, emissions of both sulfur dioxide and organic carbon can be expected to grow by 2050 because of high population and economic growth and limited environmental protection. *Heald et al*. [2008] project that a 20% mass increase in the production of particle components resulting from BVOC oxidation can be expected for a business-as-usual scenario, mostly because of changes in climate and emissions over South America by 2100. Nevertheless, this estimate includes uncertain changes in BVOC emissions and land-use patterns. Deforestation, for example, might reduce BVOC emissions and therefore reduce some of the increases projected by *Heald et al*. Another possibility is that future anthropogenic pollution, in addition to the direct release of primary particles, might enhance oxidant levels and thereby might significantly alter prevailing BVOC oxidation pathways, including the fraction of low-volatility products formed (i.e., those that go on to compose particle components). Changes in land cover might also influence the abundance and the properties of PBA particles and components. The CCN concentrations in the Basin might increase year-round, driven both by higher anthropogenic emissions of primary particles and by greater concentrations of sulfuric acid that might increase the rate of new particle formation. These many possible changes in the climatology of Amazonian aerosol particles might in turn significantly influence aerosol-cloud-climate interactions in the Basin, providing a feedback to rainfall, vegetation, and climate change [*Barth et al*., 2005; *Andreae and Rosenfeld*, 2008]. *Davidson and Artaxo* [2004] review the links in the Amazon Basin between biology and atmospheric properties.

Potentially of particular importance presently and in the future is the export of particles from the Amazon Basin to other parts of the world following deep convection [*Andreae et al.*, 2001], especially during the dry season when biomass-burning particle concentrations can be very high. Because the ITCZ cuts across the Basin and the general circulation involves the meridional transport of air towards it from both north and south, a substantial fraction of the Amazonian particles experiences deep convection. Whatever does not get scavenged in that process can be injected into the middle and upper troposphere, and some particles can also enter the stratosphere via the Brewer-Dobson circulation. These particles can be transported over long distances between continents. Transported microorganisms provide a clear demonstration of these processes [*Griffin et al.*, 2006; *Griffin*, 2008]. The Amazon Basin may thus represent a significant global source both of particles and of gaseous precursors to secondary particle components, and these contributions may have a strong influence on the pristine, low concentrations that are typically found in the upper troposphere and the stratosphere.

4.1. Priorities for Improved Models

Few modeling studies have focused on aerosol particles in the Amazon Basin. In particular, very little effort has been invested in understanding particle sources for natural conditions when concentrations are low. Regional models have been used, however, to characterize the importance of biomass burning to particle number and mass concentrations as well as to climate [*Freitas et al.*, 2005; *Liu*, 2005; *Martins and Pereira*, 2006]. An intercomparison of global models showed that model skill over Amazonia for the annual-average aerosol optical thickness misleadingly appeared to be good because of compensation by an underestimate of optical thickness for regions influenced by biomass burning and an overestimate for periods during which natural conditions prevail [*Kinne et al.*, 2003]. The overestimate for natural conditions was puzzling because the global models did not include emissions of PBA particles and had very rudimentary descriptions, if any, of the production of particle-phase BVOC oxidation products. The bias for natural conditions must therefore be attributed to one or more of an overestimate of out-of-Basin particles into the region, an underestimate of their in-Basin deposition rates, or a poor characterization of the optical properties of Amazonian particles. *Kanakidou et al*. [2005] summarized the uncertainties and challenges related to global climate model simulations of organic aerosol particles. There is a critical need to validate these model estimates with observations over the Amazon Basin.

High priority research needs for improving the regional modeling of Amazonian aerosol particles include:

- Development of model schemes for emissions of PBA particles in the Amazon Basin. These schemes are entirely absent in state-of-the-art chemical transport models.
- Investigation and implementation of models at the scale necessary to capture how vegetative heterogeneity within the rain forest canopy affects BVOC and PBA particle emissions.
- Inclusion in models at the level of compounds or families of all BVOC emissions contributing to the secondary components of particles. Amazonian vegetation might be a more or less efficient emitter of specific compounds compared to other locations for which standard emissions in models have been calibrated.
- Incorporation in models of new BVOC chemistry, such as the reactions of isoprene in the chemical regimes prevailing in the Amazon Basin (e.g., pristine low NO_x) and more generally of organic peroxy radicals.
- Attention in models, validated by measurements, of how efficiently particles over the Amazon Basin are removed by precipitation and how this sink term is affected by the processing and alteration of particles during their residence in the atmosphere.

4.2. Priorities for Improved Measurements

There have been many technological advances in the past 10 years for the characterization of aerosol particles globally, yet many of the new instruments have yet to be deployed in the Amazon Basin. The logistical difficulties there have constrained measurements temporally, spatially, and technically to levels insufficient for fully accurate descriptions of Amazonian aerosol particles and the processes affecting them. These difficulties notwithstanding, new instruments, defined in the context of Amazonian aerosol particles both as truly new instruments in the broad scientific community and as more familiar instruments that have never been deployed before for studies in the Basin, should be used to obtain more precise and accurate measurements of key properties of Amazonian particles. The instruments should be deployed to provide complementary information on complex properties, especially related to particle chemical composition. Chemical information can be employed, in conjunction with models, to understand mechanisms of particle formation and subsequent aging processes. In addition, more creative applications should be made of realtime displays and airborne remote sensors for better placement of aircraft, particularly when sampling clouds.

The particle properties that are the most uncertain and thus limit our ability to assess their environmental effects include (1) the molecular composition of the organic component as quantitative tracers of sources and age, (2) the hygroscopicity and mixing states as affected by atmospheric processing, (3) the activity as cloud and ice nuclei, and (4) the optical activity (i.e., extinction, absorption, single scattering albedo, and asymmetry factor). State-of-the-art measurement systems have the potential to significantly reduce the uncertainties surrounding these properties. For example, improvements in measuring the chemistry of aerosol particles with instruments like aerosol mass spectrometers [*Prather et al.*, 1994; *Jayne et al.*, 2000] have provided a detailed look into the chemistry of particles, yet an aerosol mass spectrometer went to Amazonia for the first time only recently as part of AMAZE-08 [*Chen et al.*, 2009]. Similarly, single- and multi- column continuous flow CCN counters, including instruments with size-selective inlets, have opened new possibilities for measurements of particle hygroscopicity [*Roberts and Nenes*, 2005], and these state-of-the-art instruments also were deployed in Amazonia for the first time in 2008 as part of AMAZE-08. There are many other instruments that are currently under development or that have only just been made operational. For example, the single-particle soot photometer is the first instrument to measure the mass in single, light-absorbing carbon particles, and its use in studying biomass-burning particles could improve the connections between the particle chemistry and the particle radiation field [*Baumgardner et al.*, 2004; *Schwarz et al.*, 2006; *Moteki et al.*, 2007]. When an aircraft is available, airborne lidar and radar with realtime displays could be used to locate optimum areas for cloud penetration to understand and quantify the effects of cloud processing on particles.

The use of measurements to understand the evolution of Amazonian aerosol particles, their interactions with clouds and radiation, and their impacts on climate must ultimately be facilitated by global climate models coupled with chemical transport models. These models, however, must be based on particle properties and processes defined by a combination of laboratory and ambient measurements, and significant uncertainties exist in the treatments presently employed for Amazonian aerosol particles (cf. §4.1), traceable in part to an insufficient set of measurements. As an example of how the uncertainty in measurements propagates into models, the aerosol optical thickness predicted by several different global models varies by more than 50% in the Basin [*Kinne et al.*, 2003; 2006]. These intermodel differences are primarily attributable to uncertainties in the parameterization of particle composition and residence time [*Textor et al.*, 2006]. Reductions in these uncertainties requires knowledge, to be gained through well-designed measurement programs, that focus on closure studies, where the closure to be achieved is between the predicted and measured particle properties, particularly their hygroscopic, chemical, microphysical, and optical properties. Another important closure, though less precisely defined, is that of particle residence time, meaning an evaluation of sources and sinks.

As one example, obtaining closure for CCN activation requires measuring the chemical composition of particles as a function of size and accurately predicting the number-diameter distribution of CCN as a function of supersaturation that is measured with in situ instrumentation. *Gunthe et al*. [2009] provide one example for studies in the Amazon Basin. The results of this type of closure should be incorporated into modules of cloud microphysics to improve how the aerosol indirect effect is forecast. As a second example, radiation closure requires ground-based and satellite measurements of optical thickness, at multiple wavelengths, to compare with the optical thickness derived from measurements of the chemical and optical properties of particles over a range of altitudes and geographic locations.

The ideal field program to implement the above closure studies would span several dry and wet seasons in the Basin, would require in situ and remote sensing measurements from multiple ground-based and airborne platforms, and would be complemented by satellite observations from which particle and trace gas properties are derived. Autonomous, unmanned aerial vehicles have recently been used to make measurements of particles over the Indian Ocean [*Corrigan et al.*, 2008] and could be extremely useful in the Basin. Establishment of a tall-tower atmospheric observatory could greatly facilitate long-term, high-quality measurements of particle properties, gaseous tracers, and meteorological variables. A tall tower could additionally provide the opportunity to measure vertical profiles through the atmosphere over the forest.

Concluding remarks. The goal of this review was to provide a synthesis of aerosol research in the Amazon Basin, most of which has been published during the last two decades. By integrating the information that has been published by a considerable number of authors in a variety of journals over a long span of time, we sought to provide a comprehensive picture about what is known about Amazonian aerosol particles and to bring together the various aspects that are now scattered throughout the literature. We intend that this effort will facilitate an understanding of the current state of knowledge on Amazonian aerosol particles specifically and tropical continental aerosol particles in general and will thereby enhance future research in this area. This review barely touched on the transformations of particles by interactions between particle- and gas-phase species (i.e., condensation, evaporation, and reactions), reactions within the particles, and cloud processing of particles. These processes, though undoubtedly important, have hardly been researched in the Amazon Basin. We therefore identify future studies designed toward these ends as being urgently needed.

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List of Tables

Table 1. Major international measurement campaigns of Amazonian aerosol particles. Beginning in the 1980s, large-scale measurement campaigns focusing on the atmospheric sciences began in the Amazon Basin, and many of them had at least a component devoted to aerosol research. Over the same time span, these limited-term campaigns have been complemented by some extended measurement series at fixed ground stations.

Table 2. Statistics of particle number-diameter distributions measured during CLAIRE-98.

Table 3. Study-average concentrations (ng m^{-3}) measured for fine- and coarse-mode inorganic species during the wet and dry seasons of the Amazon Basin. Time periods of the studies are categorized as weakly, moderately, or strongly affected by biomass burning. Time periods are also categorized for the relative influence of sources from outside the Basin. Table entries are rounded to two significant figures for presentation. *Notes: Fine fraction is defined as aerodynamic diameter under 1 μm for studies b and o, under 2 μm for studies a, c, f, g, h, i, j, k, & m, and under 2.5 μm for studies n and p. Coarse fraction is defined as aerodynamic diameters from 2 to 10 μm for studies f, g, h, i, j, k, & m, of 2 to 15 μm for studies a & c, under 10 μm for study l, and greater than 1 μm for study b. Total suspended particulate matter is used as an approximation of coarse-mode mass for studies d and e.* For studies *n* and *p*, medians rather than means are listed. *The sulfate column includes that measured by ion chromatography as well as that inferred on an equivalent basis from PIXE measurements of elemental sulfur (i.e., elemental sulfur attributed entirely to sulfate in the analysis).* References: a. *Artaxo et al*. [1988]; b. *Talbot et al.* [1988]; c. *Artaxo et al* [1990]; d. *Talbot et al*. [1990]; e. *Artaxo et al* [1998]; f. *Echalar et al*. [1998]; g. *Gerab et al.*[1998]; h. *Formenti et al*. [2001]; i. *Artaxo et al*. [2002]; j. *Maenhaut et al*. [2002]; k. *Guyon et al.* [2003c]; l. *Mace et a*l. [2003]; m. *Graham et al*. [2003a]; n. *Trebs et al*. [2005]; o. *Fuzzi et al*. [2007]; p. *Trebs et al*. [2008].

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Figure 1. Map showing the geographic boundaries of the Amazon forest (red line), the Amazon-Tocantins River Basins (purple line), and Brazilian Legal Amazon (orange line). The political boundaries of South America are shown as black lines. Cities often mentioned in the literature of Amazonian aerosol particles are indicated. Specific research sites for some of the campaigns listed in Table 1 are also highlighted, including (1) FNS and RBJ in Rondônia in southwestern Amazonia, (2) K34, TT34, and C14 north of Manaus in central Amazonia, (3) Balbina in central Amazonia, and (4) Caxiuanã in eastern Amazonia.

Figure 2. Scanning electron micrographs of primary biological particles collected in the Amazon Basin. Source: unpublished results of the Max Planck Institute for Chemistry, Mainz, Germany, for particles collected during the project European Studies on Trace Gases and Atmospheric Chemistry as a Contribution to the Large-Scale Biosphere-Atmosphere Experiment in the Amazon Basin (LBA-EUSTACH) in 1999.

Figure 3. Illustration of vertical mixing processes that affect the particle number-diameter distribution of aerosol particles in the Amazon Basin. The lowest 4 km represents daytime conditions with a fully developed mixing layer and shallow convection in the transition layer. Upward transport is controlled by the deep convection and by the fair-weather cumulus clouds. Subsidence dominates the large-scale downward transport. Shown also are wind roses from *Krejci et al.* [2003]. The original figure in *Krejci et al.* [2003] was prepared to describe observations over Suriname but the processes depicted are applicable to the wider Amazon Basin.

Figure 4. Source classification scheme for Amazonian aerosol particles. Although emissions from the Amazonian biosphere are active at all times and have low variability year-round, they are relatively weak, and particles and their components can be dominated at times by influences from outside of the Amazon Basin, such as from Saharan dust,

African biomass burning, or Atlantic marine emissions. The dust and marine emissions are a natural contribution because they were present in the year 1750. At other times, anthropogenic influences such as in-Basin biomass burning in the dry season can dominate the type, the number and, the mass concentrations of Amazonian aerosol particles. In this figure, "favored" suggests conditions of greater probability, although all influences are possible under most conditions. For example, in the wet season biomass burning can still influence some observations, such as sampling sites downwind of the border area of Brazil and Guyana and Suriname, which engage in biomass burning during the northern Amazonian wet season (cf. Figure 8).

Figure 5. (a) December-through-February (DJF) wind vector and wind speed (m s^{-1}) at 1000 hPa for South America. (b) Same as *a* but averaged for June through August (JJA). (c) DJF outgoing longwave radiation (W m^{-2}). (d) Same as *c* but for JJA. The horizontal black line shows the position of the equator. Data represent the average reanalysis of 1988-2007 from the National Centers for Environmental Prediction (NCEP).

Figure 6. Illustration of the typical diel evolution of the lower troposphere in the Amazon Basin. Adapted from *Rissler et al.* [2006].

Figure 7. (A) Number-diameter and (B) volume-diameter distributions for giant Amazon aerosol particles (> 4 μm), as determined by light microscopic analysis of samples collected on glass slides. Shown are particle totals, biological particle totals, and percentage contribution of biological particles to the total. Data are from M. Andreae (personal communication).

Figure 8. Fire locations in Feb, May, Aug, and Nov 2007 based on the MODIS Collection-5 Active Fire Product [*Giglio et al.*, 2006]. Red (scaled from 0 to 300) is the number of fire pixels, with corrections for cloud cover. Some boxes have more than 300 fire pixels (e.g., the maximum value for the scenes shown is 1144 fire pixels). Figure prepared by S.T. Martin and C.L. Heald.

Figure 9. (top) Mean diel NO, NO_2, and O_3 concentrations at the LBA-EUSTACH primary forest site Reserva Biológica Jarú (RBJ) and at the LBA-EUSTACH pasture site Fazenda Nossa Senhora Aparecida (FNS) in 1999. Both sites are located in Rondônia (cf. Figure 1). Measurements were taken 3.5 m above ground at the pasture site and 20 m above the rain forest canopy. Data are presented as 1-hour medians over 27 days of the LBA-EUSTACH-1 campaign (mostly natural conditions) and 46 days of the LBA-EUSTACH-2 campaign (strongly influenced by biomass burning). (bottom, left) A midday CO profile taken from a flight out of Manaus on July 18 over remote forest in 1985 for natural conditions (ABLE-2A). (bottom, right) Temporal variation of the CO mixing ratio at 30 min intervals at the FNS surface station during LBA-SMOCC in 2002. Adapted from *Sachse et al*. [1988], *Andreae et al*. [2002], and *Chand et al.* [2006].

Figure 10. Mean vertical profiles of NO, NO_2, and O_3 in the forest canopy. Profiles represent an average over 43 days in Rondônia at 14:00 local time during LBA-EUSTACH-2. Data points of concentrations are medians (0.5-quantile), and corresponding variations are indicated by horizontal bars (left end: 0.25-quantile, right end: 0.75-quantile). Variation of the NO concentration above 5 m is smaller than symbol size. Adapted from *Andreae et al.* [2002].

Figure 11. (top) Diel fluxes and (bottom) mixing ratios of isoprene and monoterpenes measured in central Amazonia (tower C14; cf. Figure 1) between 14 and 29 September 2004. Black lines represent the study mean and gray area represents the standard deviation. Adapted from *Karl et al*. [2007].

Figure 12. Scatter plots between the amount of OH observed from aircraft and that modeled for the boundary layer over Suriname in October 2005. (a) The standard model. (b) An updated model including the role of organic peroxy radicals. The solid lines indicate ideal agreement and the dashed lines the ±40% range, based on the measurement accuracy. (c) Percentage difference in the annual mean OH, as calculated using the

updated model compared to the standard model (the arrow indicates the location of Suriname). The aircraft measurements were performed in October 2005 over the pristine forests of Suriname, Guyana, and Guyane (French Guiana). Adapted from *Lelieveld et al.* [2008]. [permission pending]

Figure 13. Particle number-diameter distributions observed over southern Suriname. N_6, N_{18}, and N_{120} represent the number concentration of particles larger than 6, 18, and 120 nm, respectively. The difference $N_{6\text{-}18}$, equal to $N_6 - N_{18}$, quantifies the concentration of nucleation-mode particles. The arrows on the left mark the altitude where the average number-diameter distributions on the right were measured. The error bars on the average distributions represent lower and upper quartiles. Adapted from *Krejci et al.* [2005].

Figure 14. Diel variation in the number concentration of nucleation-mode (viz. < 25 nm) particles. Shown are averages for periods of LBA-SMOCC that were weakly, moderately, and strongly influenced by biomass burning. Adapted from *Rissler et al.* [2006].

Figure 15. Time series of particle mass concentrations in Balbina (central Amazonia), Santarem (eastern Amazonia), and Alta Floresta (southern Amazonia). Data are shown as stack bar plots of fine (red; < 2 μm) and coarse (blue; 2 to 10 μm) fractions. The PM_{10} concentration is the sum of the two. The measurement protocol follows that of the US EPA for weighing filters, and some water was therefore also included in the mass measurement. The relative mass of water, however, was small because of the low hygroscopic growth factors of Amazonian aerosol particles. Adapted and updated from *Artaxo et al*. [2002].

Figure 16. Particle mass-diameter distributions from gravimetric analysis of MOUDI stages. (top) Examples of three distributions collected during AMAZE-08 at times when natural conditions prevailed. Each distribution represents one week of data (10 to 16 Mar 2008; 22 to 31 Mar 2008; and 9 to 17 Apr 2008). (bottom) Representative

distribution collected during LBA-SMOCC during time periods strongly influenced by biomass burning. A bin label such as “3.2 to 10 µm” represents a mass filter having 50% cutpoints at 3.2 and 10 µm. Data are from P. Artaxo (personal communication).

Figure 17. (A) A typical particle number-diameter distribution observed for natural conditions in central Amazonia during CLAIRE-98. (B-D) Whole-campaign histograms of Aitken, accumulation-mode, and total particle number concentrations. Natural conditions mostly prevailed. Adapted from *Zhou et al.* [2002].

Figure 18. Average particle number-diameter distributions observed in the Amazon Basin for periods during which natural conditions prevailed. (top) CLAIRE-98 (Balbina, 18 days of data) [*Zhou et al.*, 2002], CLAIRE-01 (Balbina, two days of data) [*Rissler et al.*, 2004], and AMAZE-08 (approximately equidistant to Manaus and Balbina, 22 Feb - 12 Mar 2008) [E. Swietlicki, personal communication]. (bottom) Airborne measurements over Suriname during CLAIRE-98 [*Krejci et al.*, 2003]. Altitude ranges were from (1) 0.2 to 1.2 km and (2) 1.2 to 2.4 km.

Figure 19. Mean composition of the (identified) water-soluble organic carbon. Fine-mode filter samples collected in the different periods of the LBA-SMOCC campaign were examined by chromatography. Key: monocarboxylic acids (MCA), oxalic acid, C_3-C_6 dicarboxylic acids (DCA C3-C6), tricarboxylic acids (TCA), aromatic acids (ArAcids), aromatic aldehydes (ArAld), anhydrosugars (AnSugars), sugar-alcohols, sugars, and 2-methyltetrols (MeTetrols). Adapted from *Decesari et al.* [2006].

Figure 20. Total-carbon apportionment for biomass-burning particles collected during the dry season (LBA EUSTACH). Total carbon is divided into black carbon and organic carbon; organic carbon is partitioned into water-insoluble and water-soluble fractions; the portion of the water-soluble fraction that is elutable and identifiable by HPLC is indicated; the fraction of that eluate that is identifiable by GC-MS is indicated; and the partitioning of that fraction into the chemical species is shown. This final fraction

is also represented in Figure 19. “BC water” is based on thermal analysis for black carbon after washing the sample with water. Adapted from *Mayol-Bracero et al.* [2002].

Figure 21. Two-day time series of chemically apportioned, submicron particle mass concentrations measured on February 17 and 18, 2008, in central Amazonia as part of AMAZE-08. Measurements were made using an Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-TOF-AMS) during a time period for which natural conditions prevailed. (a) Time series of organic mass fraction. (b) Time series of submicron mass concentrations of organic material (green), sulfate (red), ammonium (orange), nitrate (blue), and chloride (magenta). The measurement window of the AMS is approximately 60 to 600 nm in vacuum aerodynamic diameter. Data are from S.T. Martin (personal communication).

Figure 22. Spectra of CCN activation in the Amazon Basin for a range of natural to very polluted conditions. The highest concentrations occur for locations heavily influenced by in-Basin biomass burning. Adapted from *Roberts et al.* [2003].

Figure 23. The evolution of the mass-diameter distribution of cloud drops for increasing height in growing convective clouds, in regimes of (A) blue ocean, off the northeast Brazilian coast (4°S, 38°W), (B) green ocean (i.e., natural conditions), at the western tip of the Amazon (6°S, 73°W), (C) smoky clouds in Rondonia (10°S, 62°W), and (D) pyro-clouds. The lowest-altitude distribution in each plot represents conditions at cloud base, except in *D* for which a distribution for large ash particles outside of the cloud is also shown. Adapted from *Andreae et al.* [2004]. [permission pending]

Figure 24. Amazon highways. The government of Brazil plans that the core region of the Amazon will be accessible by all-weather highways (yellow) in the future. Although these investments in transportation promise to lower the production costs of ranching and farming, they potentially threaten to stimulate deforestation. Source: D.C. Nepstad, *A Report to the World Wide Fund for Nature (WWF)*, 2007.

Figure 25. Simulations of forest cover for the year 2050. (a) Scenario of business as usual. (b) Scenario of good governance. Adapted from *Soares-Filho et al.* [2006]. [permission pending].

Dates		Campaign	Location & Description	Ground	Airborne	Science Focus	Reference
Jul-Aug 1985	ABLE-2A	*Amazon Boundary-Layer Experiment - dry season*	dry season in Amazonia	Y (Manaus)	Y (ER-2 and Electra)	trace gas and meteorological measurements	[*Harriss et al*., 1988]
Apr-May 1987	ABLE-2B	*Amazon Boundary-Layer Experiment - wet season*	wet season in Amazonia	Y (Manaus)	Y (ER-2 and Electra)	trace gas and meteorological measurements	[*Harriss et al*., 1990]
Aug-Sep 1995	SCAR-B	*The Smoke, Clouds, and Radiation-Brazil*	dry season in disturbed southern Amazonia (Brasilia and Alta Floresta in southern Amazonia)	Y	Y (ER-2 and Convair)	effects of biomass burning on climate (direct effect)	[*Kaufman et al.*, 1998]
Mar-Apr 1998	CLAIRE-98	*The Cooperative LBA Airborne Regional Experiment*	wet season in undisturbed central Amazonia (Balbina in central Amazonia)	Y		chemistry and physics of biogenic aerosols	[*Avissar et al*., 2002]
Apr-May 1999 Sep-Oct 1999	LBA-EUSTACH	*European Studies on Trace Gases and Atmospheric Chemistry as a Contribution to the Large-Scale Biosphere-Atmosphere Experiment in Amazonia*	wet season in disturbed western Amazonia (FNS and RBJ in Rondônia in southwestern Amazonia; K34 and C14 in central Amazonia; Caxiuanã in eastern Amazonia)	Y	Y (Bandeirante)	chemistry and physics of wet season biogenic aerosols; trace gas emissions and reactivity	[*Andreae et al*., 2002]
Jul 2001	CLAIRE-01	*The Cooperative LBA Airborne Regional Experiment*	transition of wet-to-dry season in undisturbed central Amazonia (Balbina)	Y	Y (Bandeirante)	chemistry and physics of biogenic aerosols; trace gas emissions and reactivity	
Sep-Nov 2002	LBA-SMOCC	*The Large-Scale Biosphere-Atmosphere Experiment in Amazonia: Smoke, Aerosols, Clouds, Rainfall, and Climate field campaign*	dry season biomass burning experiment in pasture and forest sites in disturbed Amazonia (FNS and RBJ Rondônia)	Y	Y (Bandeirante)	effects of biomass burning on clouds (indirect effect)	[*Fuzzi et al*., 2007]
Aug-Sep 2004	TROFFEE	*Tropical Forest and Fire Emissions Experiment*	dry season in disturbed southern Amazonia (Alta Floresta)		Y (Bandeirante)	measurements of aerosol and trace gas emission factors from biomass burning	[*Yokelson et al*., 2007]
Feb-Mar 2008	AMAZE-08	*Amazonian Aerosol Characterization Experiment*	wet season in undisturbed central Amazonia (TT34 north of Manaus in central Amazonia)	Y	N	chemistry and physics of biogenic aerosols; trace gas emissions and reactivity	*Martin et al*. [in preparation]

Table 1

Mode	Frequency of Occurrence (%)	Number Concentration (# cm^{-3})			Geometric Mean Diameter (nm)	Geometric Standard Deviation
		Mean ± Standard Deviation	Geometric Mean	Median		
Ultrafine	18	92±99	55	48	24±10	1.31±0.15
Aitken	100	239±154	200	200	68±12	1.40±0.14
Accumulation	100	177±115	137	146	151±22	1.40±0.10

Table 2

	In-Basin Biomass Burning	Out-of-Basin Dust, Marine, African Biomass Burning	SO_4^{2-}	NO_3^-	NH_4^+	K	P	Ca	Zn	Cl	Na	Al	Si	Ti	Mn	Fe	Reference
Wet Season																	
Fine	weakly	weakly	170-260	60-140	31-340	18-34	2.8-7.4	5.0-12	0.25-0.78	0.75-65	26-49	23-62	38-160	1.5-5.0	0.22-0.86	12-37	c,h,i,k, n,o,p
	weakly	strongly	690	-	-	200	13	120	1.6	180	270	690	1400	46	5.5	340	h
	moderately	moderately	510-1000	21	140	80-220	1.9-14	8.0-40	1.2-2.3	1.0-37	51-190	33-130	27-300	2.1-11	0.53-3.6	24-82	f,g,j
Coarse	weakly	weakly	38-140	25-36	5.0-13	39-110	8.0-38	11-39	0.60-1.6	8.9-160	54-91	44-92	50-220	4.0-8.6	0.40-1.9	25-64	c,d,h,i, k,l,o
	weakly	strongly	430	-	-	260	37	280	1.9	1100	660	1100	2100	72	10	540	h
	moderately	moderately	200-420	-	-	140-270	7.6-87	44-230	2.2-7.4	14-1100	82-2100	100-450	230-970	17-46	2.4-59	190-790	f,g,j
Dry Season																	
Fine	weakly	weakly	320-780	14-68	5.0-180	35-160	4.0-6.6	4.0-9.2	0.93-2.6	6.0-8.2	46	8.0-16	25	0.90-1.5	0.19-0.80	6.5-11	a,b,m
	strongly	weakly	1600-3300	1000-1400	860-1600	510-1100	27-33	25-51	4.2-8.3	20-62	-	89-270	120-360	6.0-33	1.9-5.0	28-160	i,k,n,o
	strongly	strongly	1200-2900	-	-	440-1300	4.7-32	1.5-53	5.3-8.6	12-110	96	100-640	230-540	11-28	1.5-3.8	190-270	f,j
Coarse	weakly	weakly	51-200	110-210	27-40	33-100	8.9-32	11-18	0.48-1.8	51-81	-	19-35	33-66	1.4-3.5	0.32-0.60	11-23	a,b,m
	strongly	weakly	180-470	48-520	97-170	88-240	47-68	14-52	2.9-4.3	10-28	-	79-500	90-880	8.0-65	3.9-16	48-460	i,k,l,o
	strongly	strongly	480-3600	-	-	390-1600	17-140	0.86-1300	4.7-11	24-220	-	1200-3100	2100-3100	110-140	14-68	1100-1800	e,f,j

Table 3

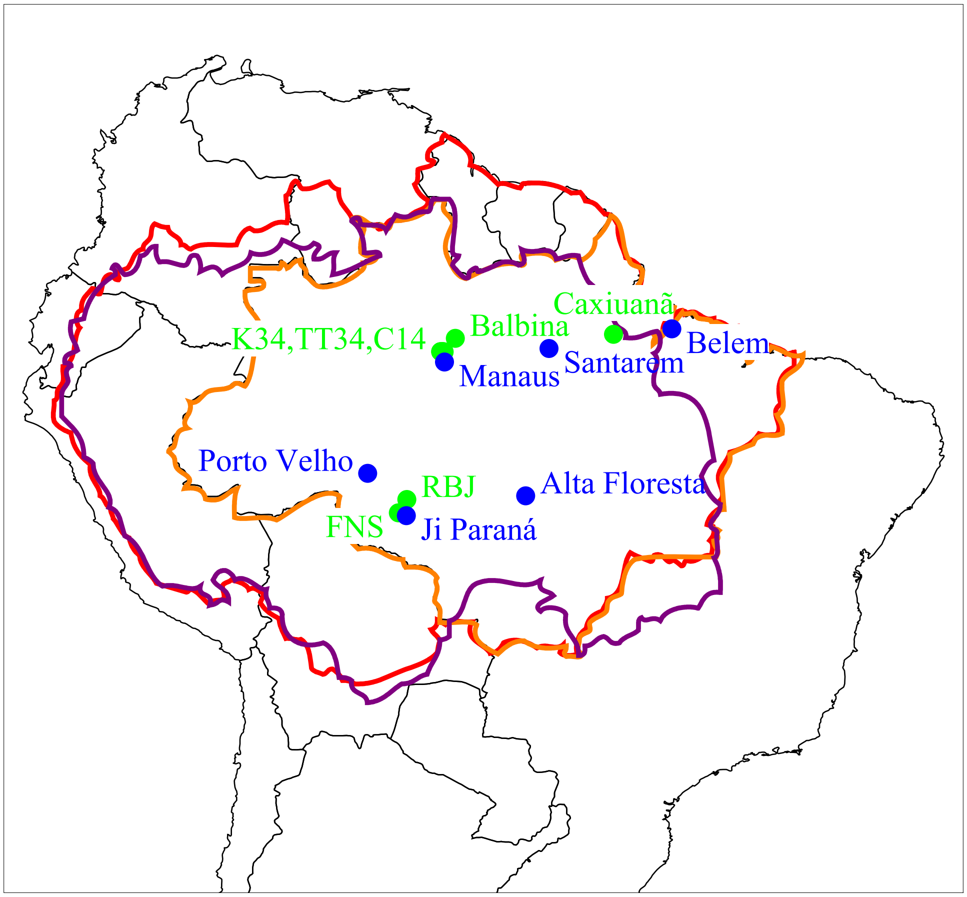

Caxiuanã
Balbina
K34,TT34,C14
Belem
Santarem
Manaus
Porto Velho
RBJ
Alta Floresta
FNS
Ji Paraná

Figure 1

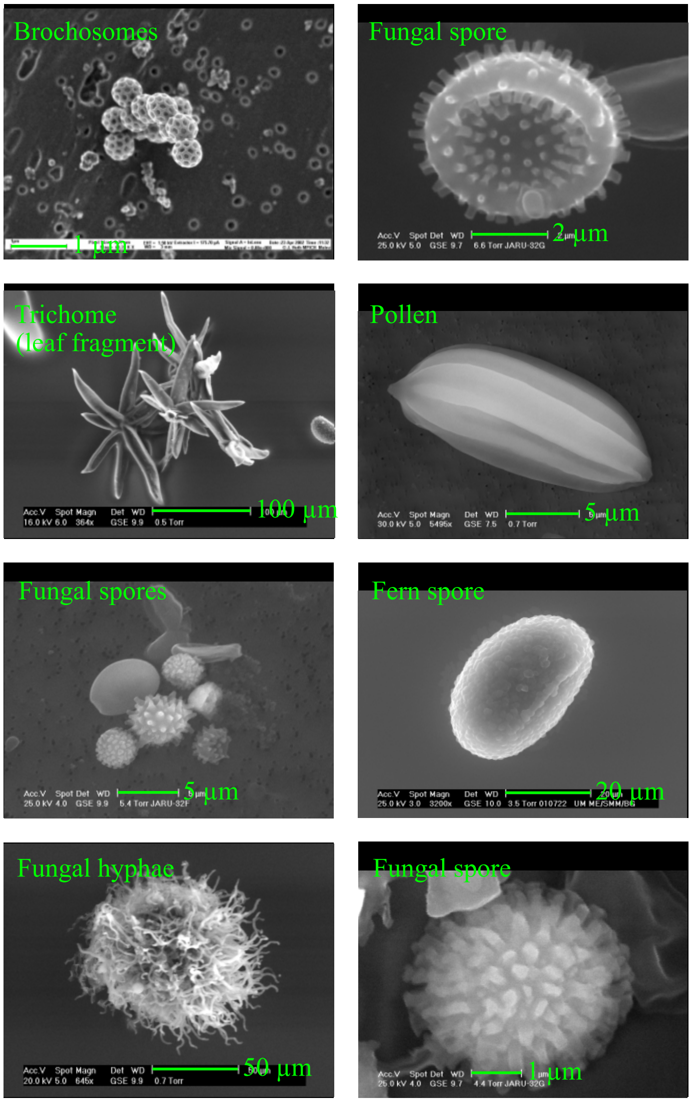
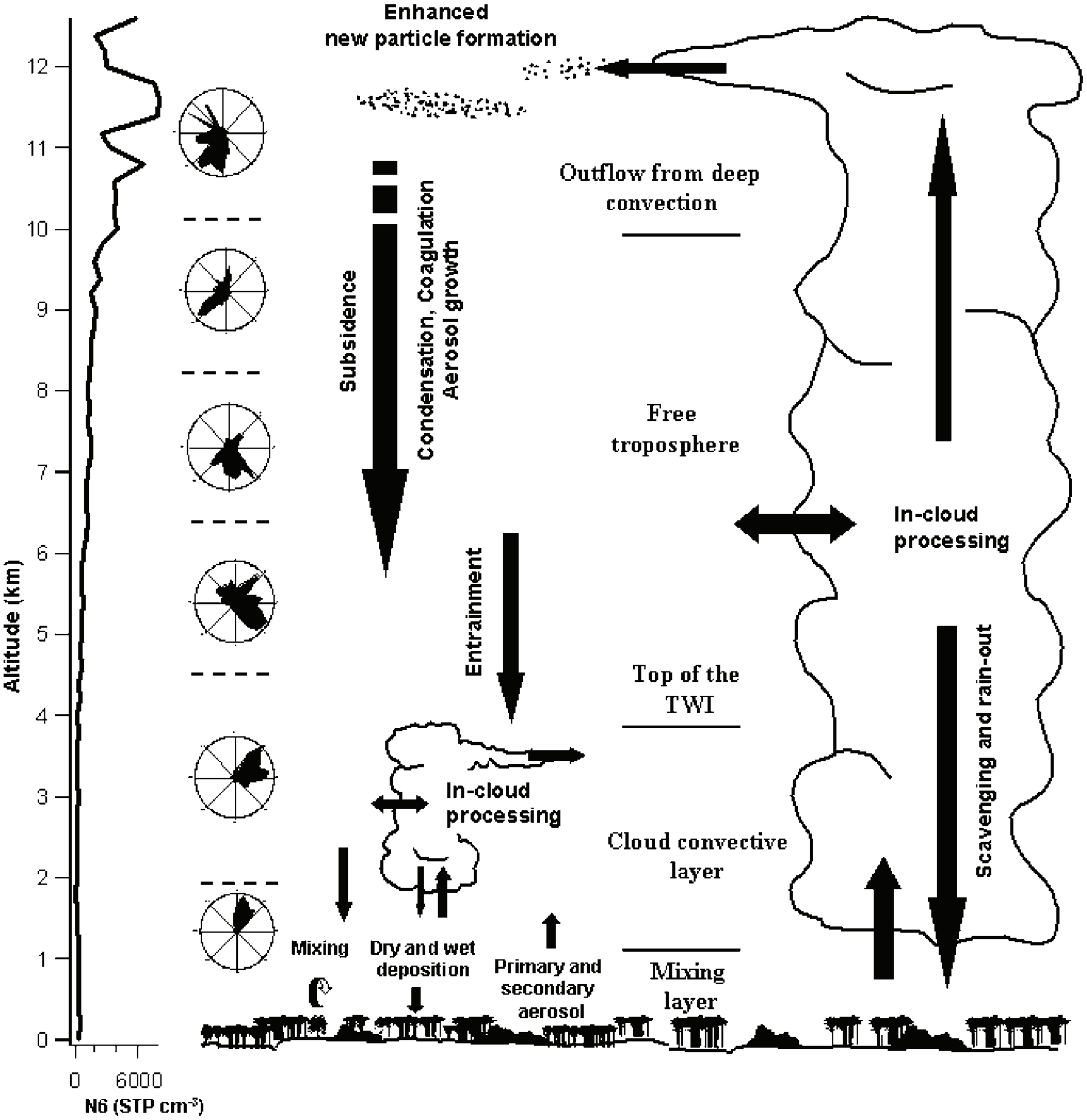

Brochosomes
1 µm
Fungal spore
2 µm
Trichome
(leaf fragment)
100 µm
Pollen
5 µm
Fungal spores
5 µm
Fern spore
20 µm
Fungal hyphae
50 µm
Fungal spore
1 µm

Figure 2



Enhanced new particle formation
Outflow from deep convection
Subsidence
Condensation, Coagulation Aerosol growth
Free troposphere
In-cloud processing
Entrainment
Top of the TWI
Scavenging and rain-out
In-cloud processing
Cloud convective layer
Mixing
Dry and wet deposition
Primary and secondary aerosol
Mixing layer
Altitude (km)
0
1
2
3
4
5
6
7
8
9
10
11
12
0
6000
N6 (STP cm^{-3})

Figure 3

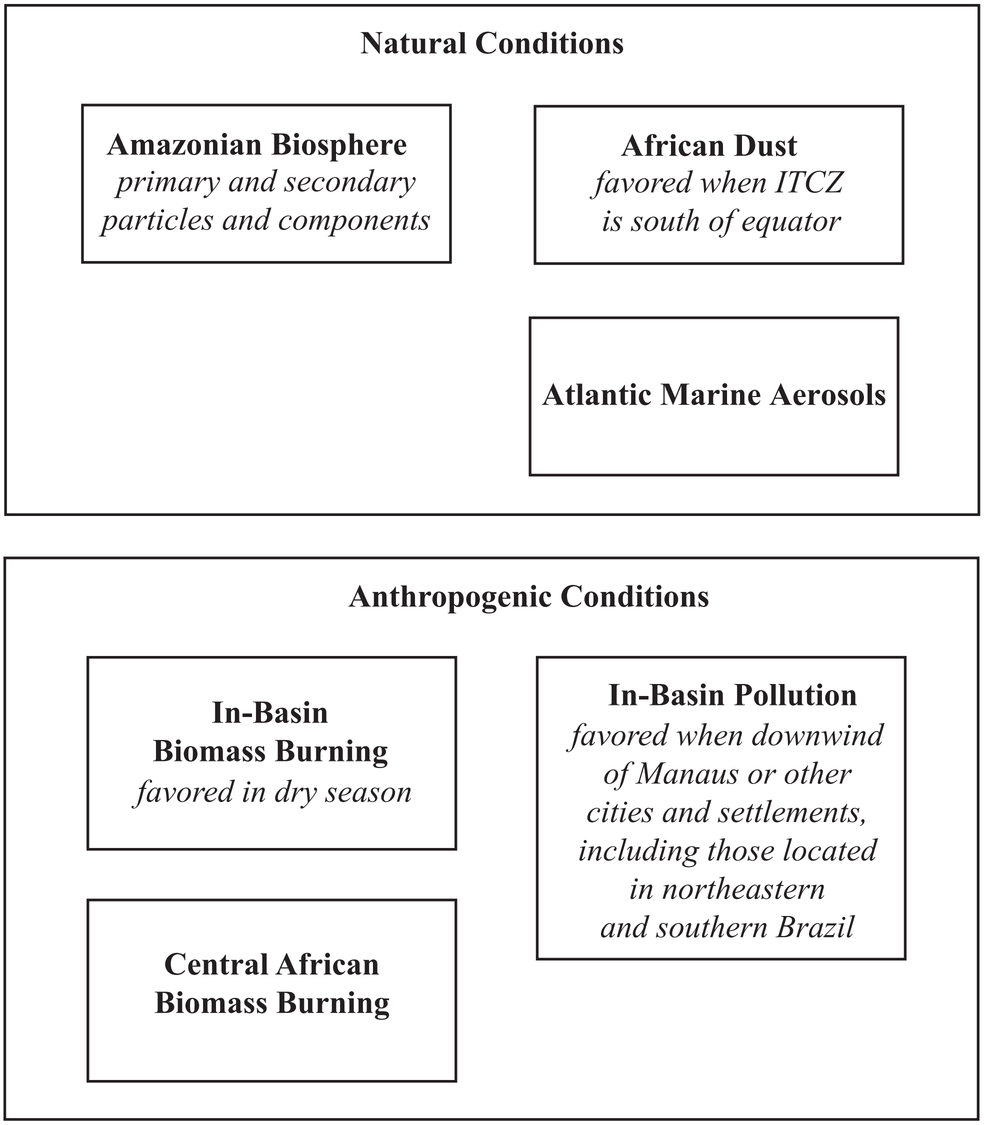
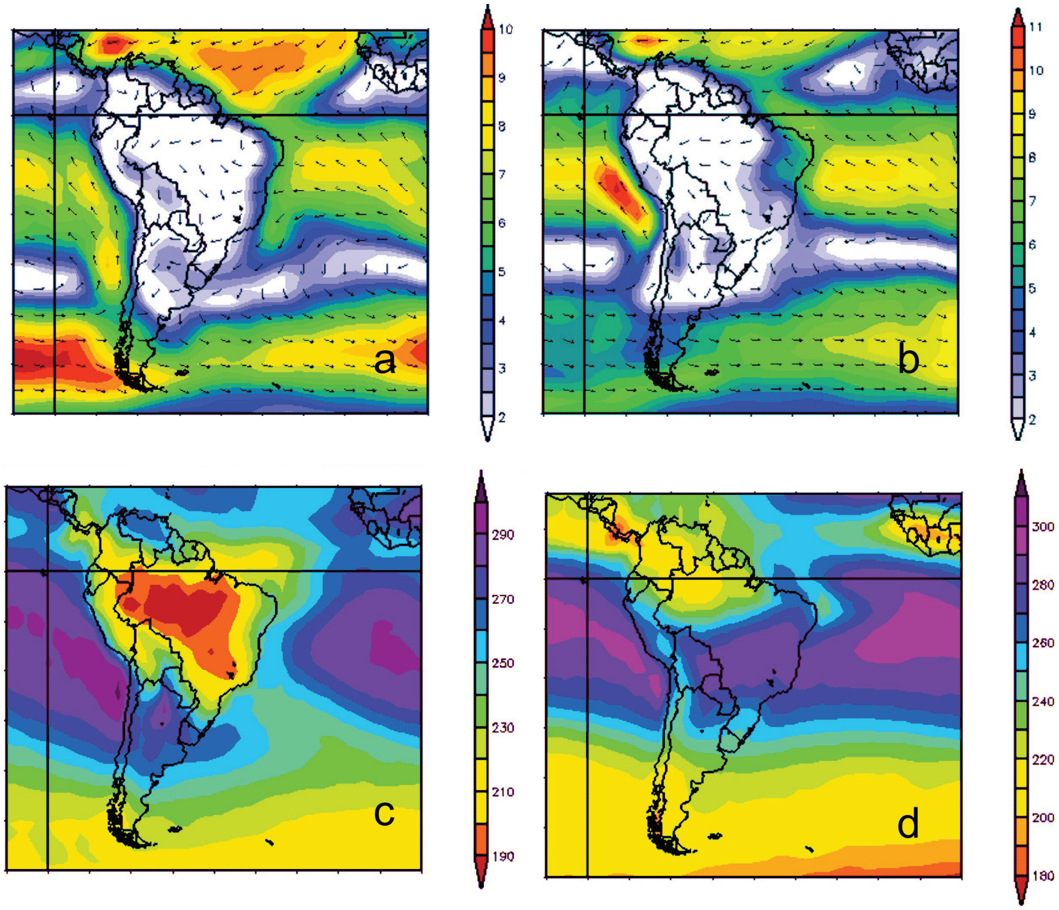
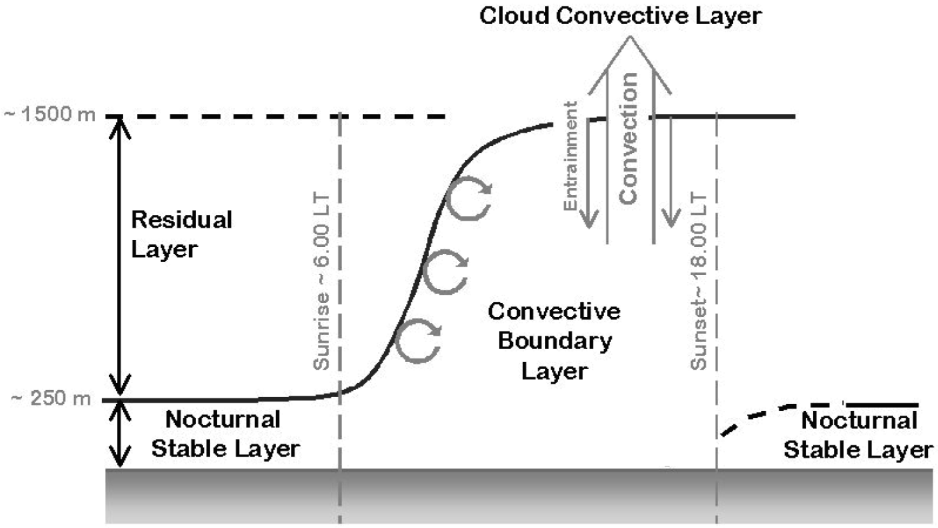

Natural Conditions
Amazonian Biosphere
primary and secondary particles and components
African Dust
favored when ITCZ is south of equator
Atlantic Marine Aerosols
Anthropogenic Conditions
In-Basin Biomass Burning
favored in dry season
In-Basin Pollution
favored when downwind of Manaus or other cities and settlements, including those located in northeastern and southern Brazil
Central African Biomass Burning

Figure 4



a
10
9
8
7
6
5
4
3
2
b
11
10
9
8
7
6
5
4
3
2
c
290
270
250
230
210
190
d
300
280
260
240
220
200
180

Figure 5



Cloud Convective Layer
~ 1500 m
~ 250 m
Residual Layer
Sunrise ~ 6.00 LT
Entrainment
Convection
Sunset~ 18.00 LT
Convective Boundary Layer
Nocturnal Stable Layer
Nocturnal Stable Layer

Figure 6

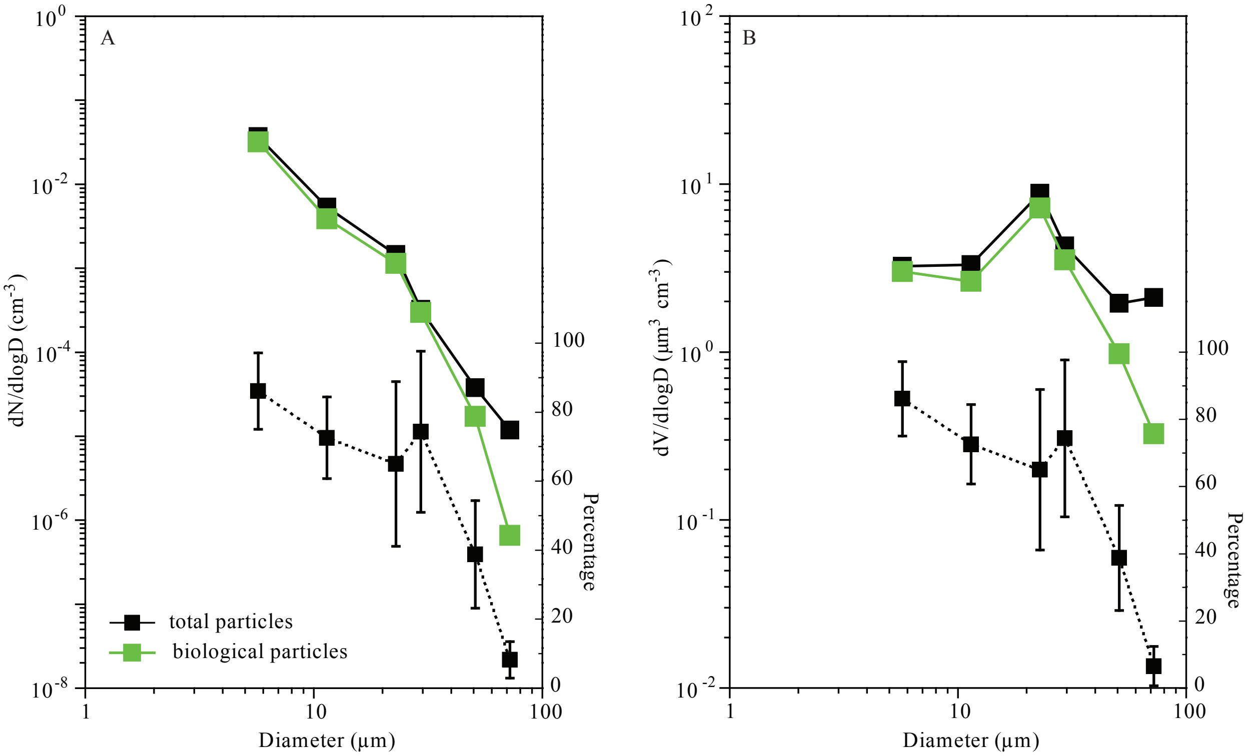
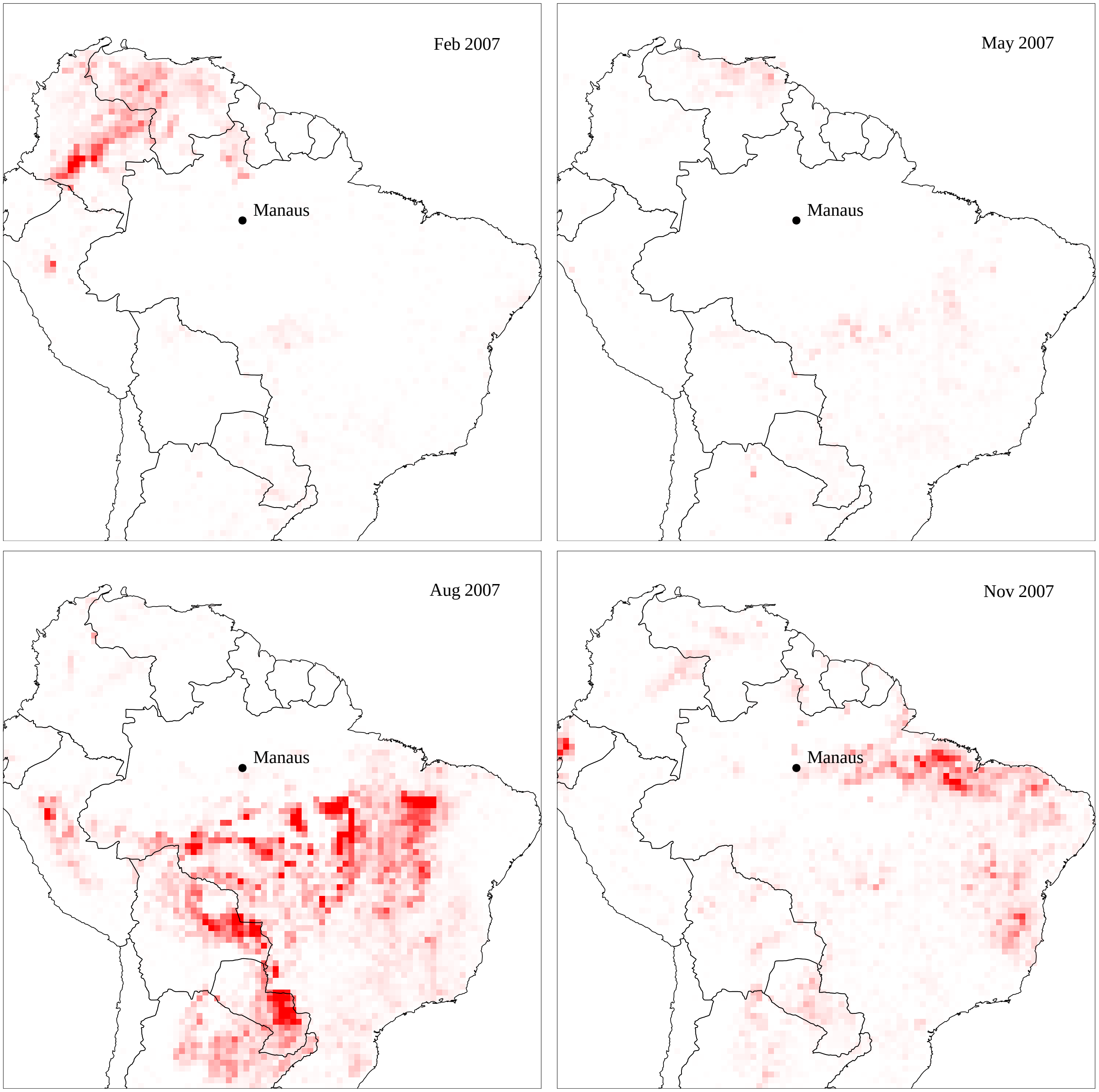

A
B
10^0
10^{-2}
10^{-4}
10^{-6}
10^{-8}
dN/dlogD (cm^{-3})
10^2
10^1
10^0
10^{-1}
10^{-2}
dV/dlogD (µm^3 cm^{-3})
100
80
60
40
20
0
Percentage
1
10
100
Diameter (µm)
total particles
biological particles

Figure 7



Feb 2007

May 2007

Aug 2007

Nov 2007

Figure 8

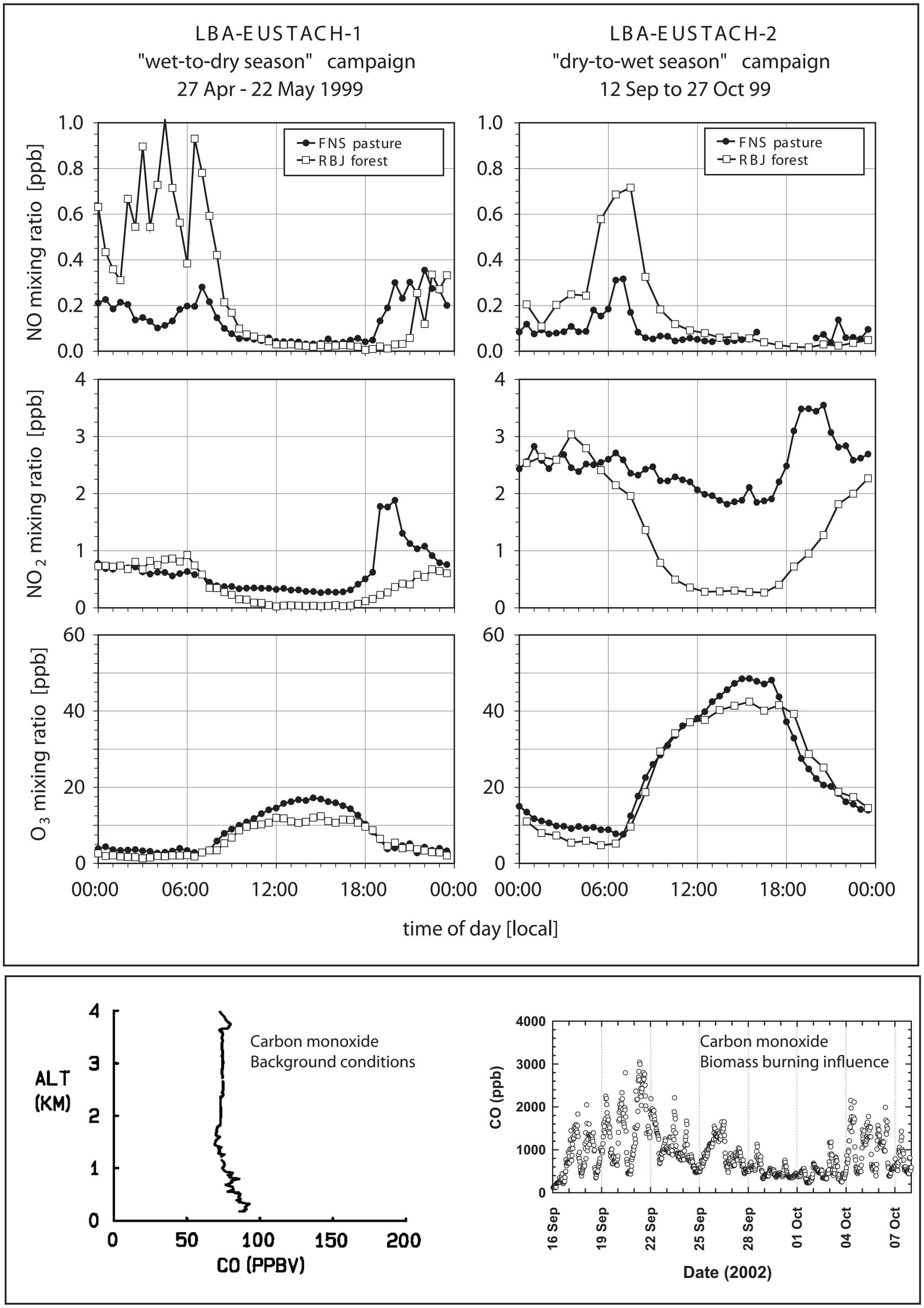

LBA-EUSTACH-1
"wet-to-dry season" campaign
27 Apr - 22 May 1999
LBA-EUSTACH-2
"dry-to-wet season" campaign
12 Sep to 27 Oct 99
FNS pasture
RBJ forest
NO mixing ratio [ppb]
1.0
0.8
0.6
0.4
0.2
0.0
NO$_2$ mixing ratio [ppb]
4
3
2
1
0
O$_3$ mixing ratio [ppb]
60
40
20
0
00:00
06:00
12:00
18:00
00:00
time of day [local]
ALT (KM)
CO (PPBV)
0
50
100
150
200
Carbon monoxide
Background conditions
CO (ppb)
4000
3000
2000
1000
0
Carbon monoxide
Biomass burning influence
16 Sep
19 Sep
22 Sep
25 Sep
28 Sep
01 Oct
04 Oct
07 Oct
Date (2002)

Figure 9

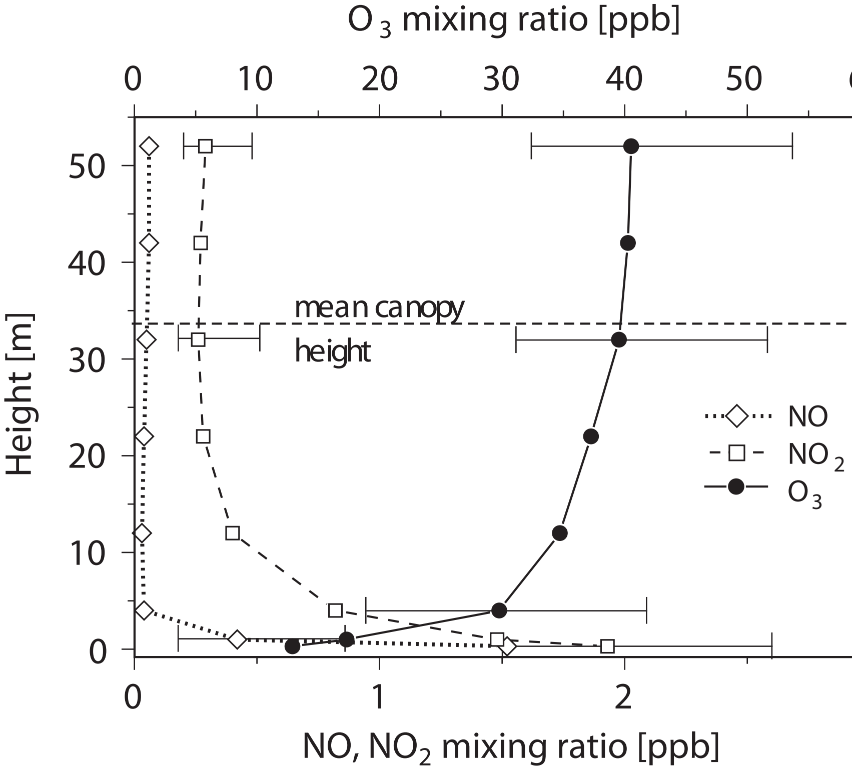

O 3 mixing ratio [ppb]
0
10
20
30
40
50
60
Height [m]
0
10
20
30
40
50
mean canopy
height
NO
NO 2
O3
0
1
2
3
NO, NO2 mixing ratio [ppb]

Figure 10

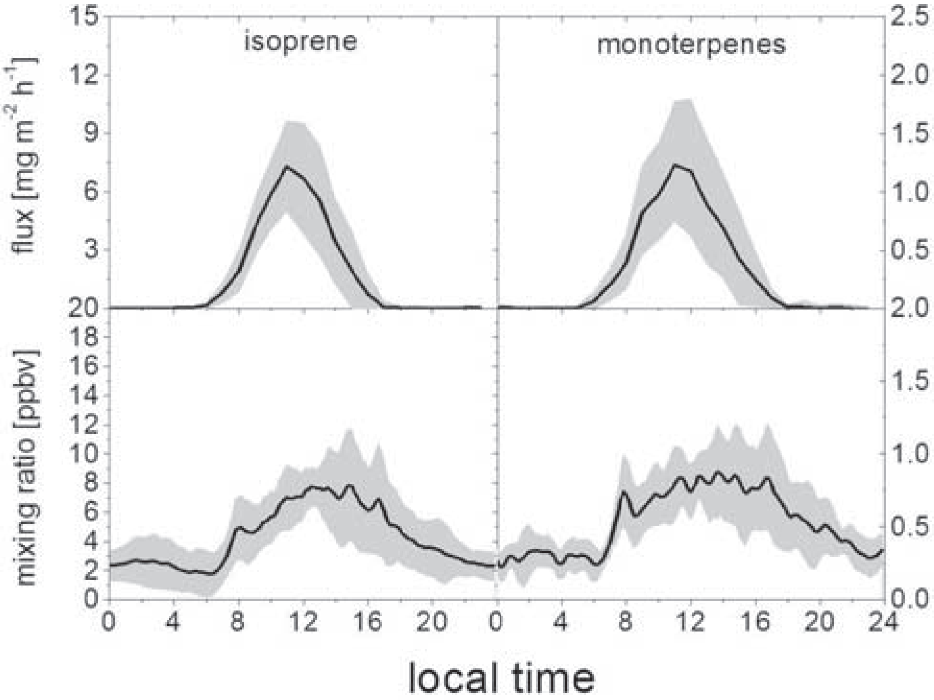

isoprene
monoterpenes
flux [mg m^{-2} h^{-1}]
mixing ratio [ppbv]
15
12
9
6
3
20
18
16
14
12
10
8
6
4
2
0
2.5
2.0
1.5
1.0
0.5
2.0
1.5
1.0
0.5
0.0
0
4
8
12
16
20
0
4
8
12
16
20
24
local time

Figure 11

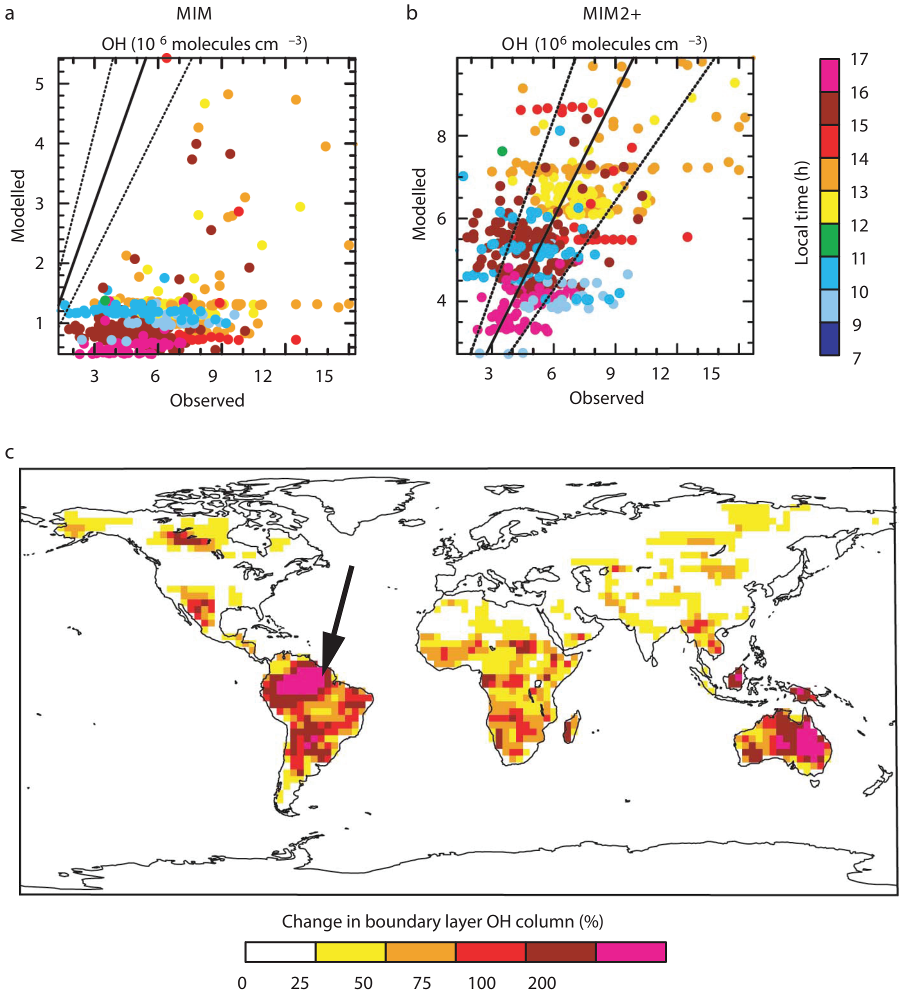

a
MIM
OH (10 6 molecules cm −3)
Modelled
Observed
1
2
3
4
5
3
6
9
12
15
b
MIM2+
OH (10⁶ molecules cm −3)
Modelled
Observed
4
6
8
3
6
9
12
15
Local time (h)
17
16
15
14
13
12
11
10
9
7
c
Change in boundary layer OH column (%)
0
25
50
75
100
200

Figure 12

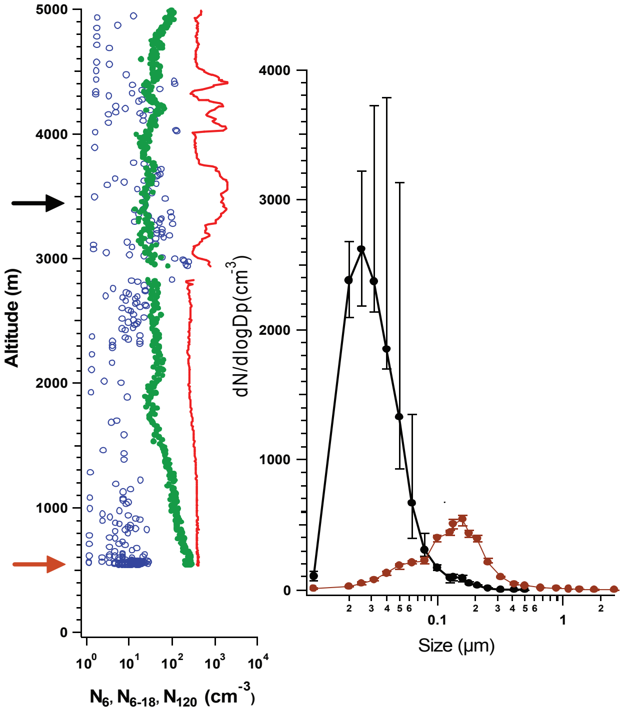

5000
4000
3000
2000
1000
0
Altitude (m)
10^0
10^1
10^2
10^3
10^4
N_6, N_{6-18}, N_{120} (cm^{-3})
4000
3000
2000
1000
0
dN/dlogDp(cm^{-3})
2
3
4
5
6
0.1
2
3
4
5
6
1
2
Size (µm)

Figure 13

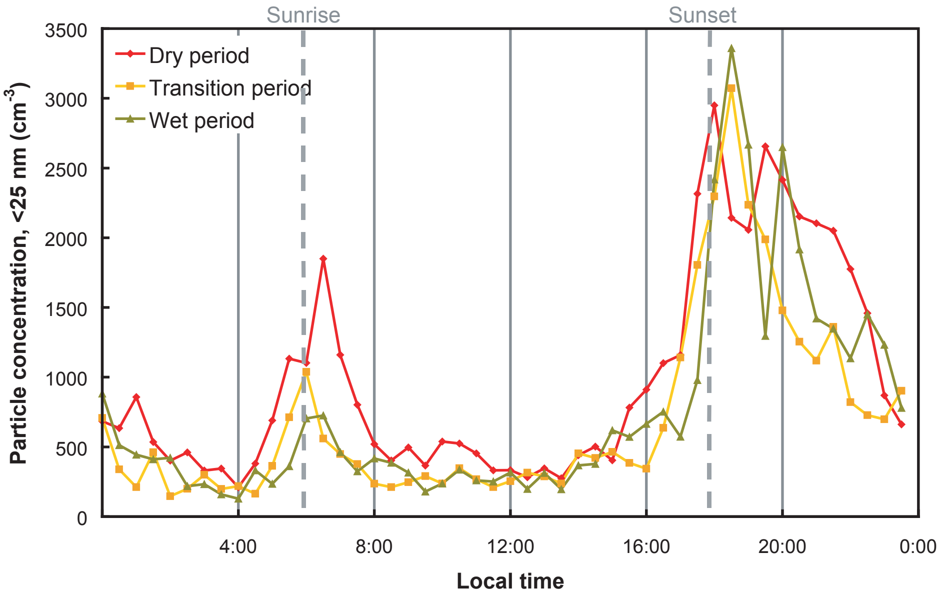
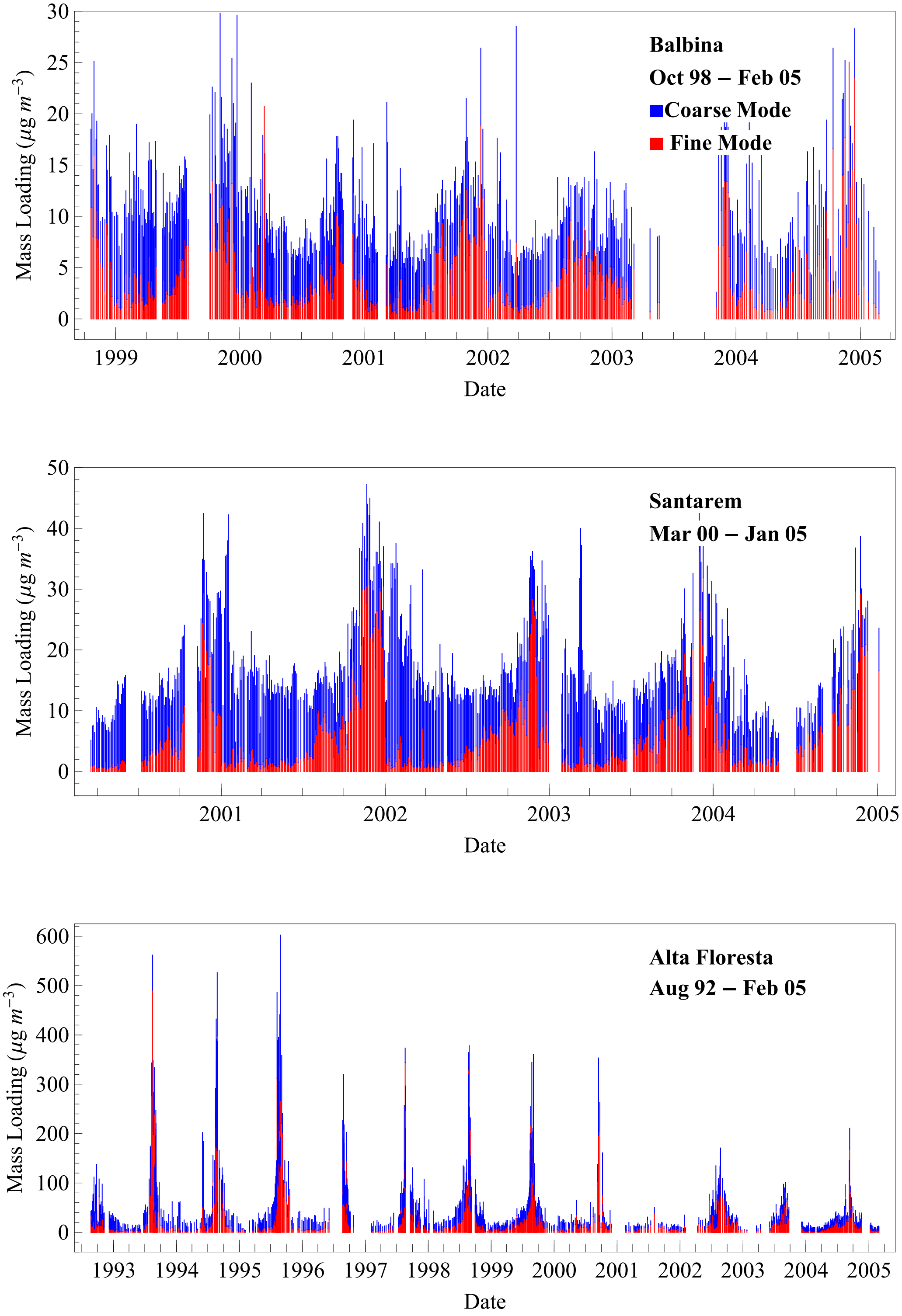

Sunrise
Sunset
Dry period
Transition period
Wet period
Particle concentration, <25 nm (cm^{-3})
3500
3000
2500
2000
1500
1000
500
0
4:00
8:00
12:00
16:00
20:00
0:00
Local time

Figure 14



Balbina
Oct 98 – Feb 05
🟦Coarse Mode
🟥 Fine Mode
Mass Loading ($\mu g\ m^{-3}$)
0
5
10
15
20
25
30
1999
2000
2001
2002
2003
2004
2005
Date
Santarem
Mar 00 – Jan 05
Mass Loading ($\mu g\ m^{-3}$)
0
10
20
30
40
50
2001
2002
2003
2004
2005
Date
Alta Floresta
Aug 92 – Feb 05
Mass Loading ($\mu g\ m^{-3}$)
0
100
200
300
400
500
600
1993
1994
1995
1996
1997
1998
1999
2000
2001
2002
2003
2004
2005
Date

Figure 15

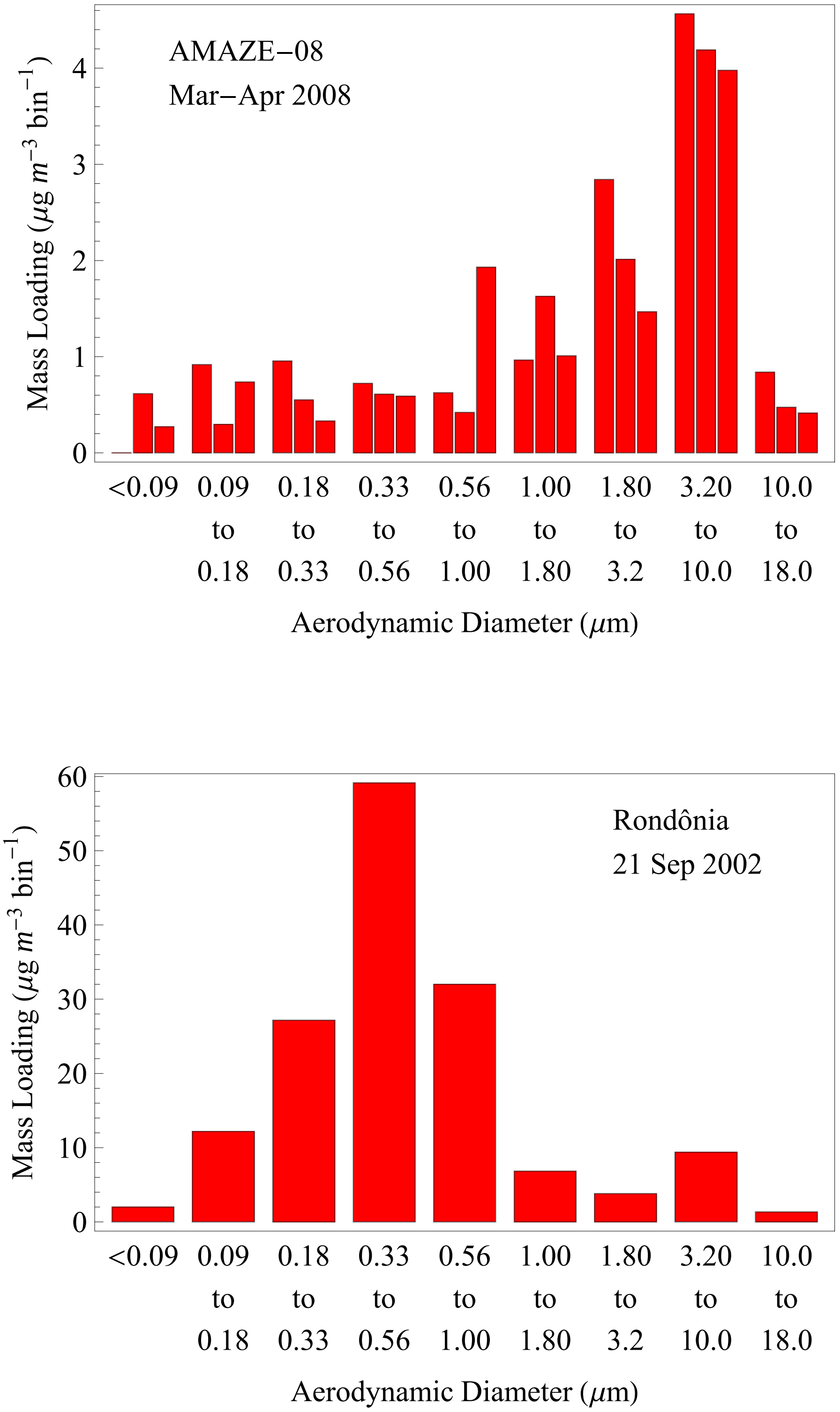

AMAZE−08
Mar−Apr 2008
Mass Loading ($\mu g\ m^{-3}$ bin^{-1})
0
1
2
3
4
<0.09
0.09 to 0.18
0.18 to 0.33
0.33 to 0.56
0.56 to 1.00
1.00 to 1.80
1.80 to 3.2
3.20 to 10.0
10.0 to 18.0
Aerodynamic Diameter (μm)
Rondônia
21 Sep 2002
Mass Loading ($\mu g\ m^{-3}$ bin^{-1})
0
10
20
30
40
50
60
<0.09
0.09 to 0.18
0.18 to 0.33
0.33 to 0.56
0.56 to 1.00
1.00 to 1.80
1.80 to 3.2
3.20 to 10.0
10.0 to 18.0
Aerodynamic Diameter (μm)

Figure 16

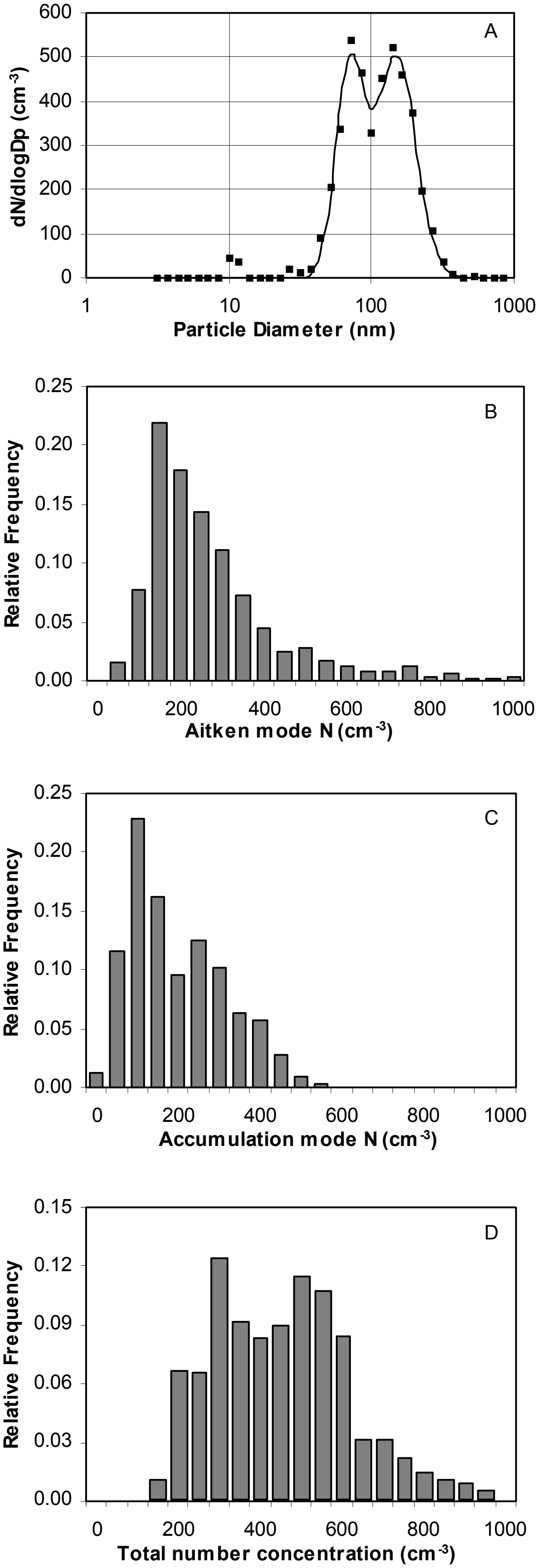

A
dN/dlogDp (cm-3)
600
500
400
300
200
100
0
1
10
100
1000
Particle Diameter (nm)
B
Relative Frequency
0.25
0.20
0.15
0.10
0.05
0.00
0
200
400
600
800
1000
Aitken mode N (cm-3)
C
Relative Frequency
0.25
0.20
0.15
0.10
0.05
0.00
0
200
400
600
800
1000
Accumulation mode N (cm-3)
D
Relative Frequency
0.15
0.12
0.09
0.06
0.03
0.00
0
200
400
600
800
1000
Total number concentration (cm-3)

Figure 17

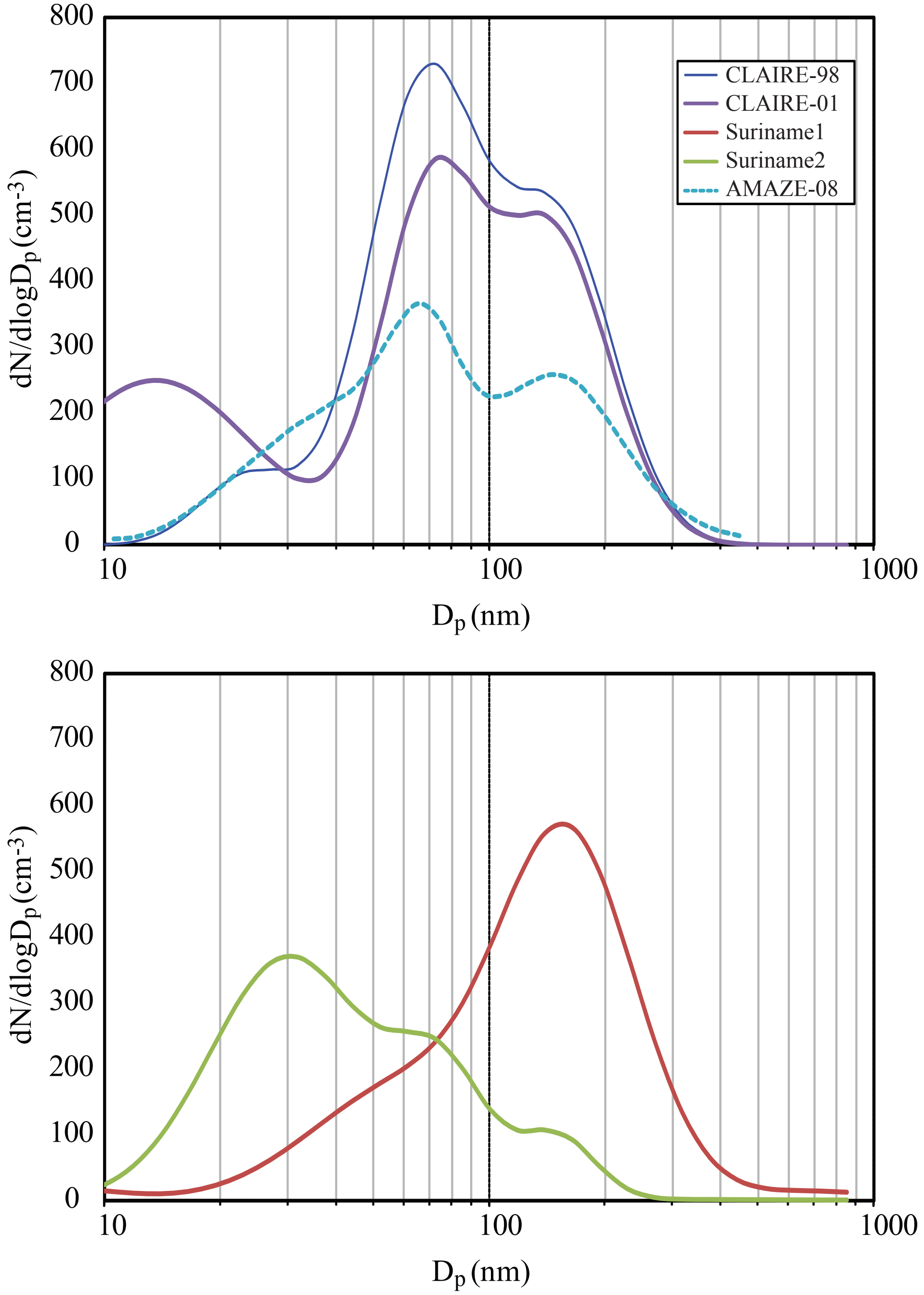
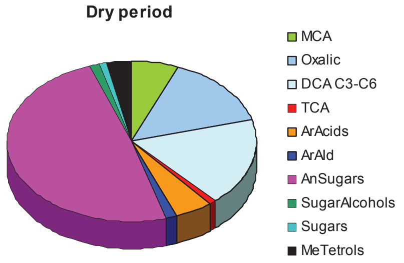
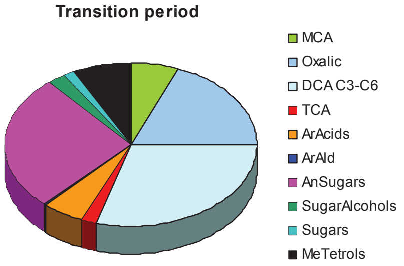

800
700
600
500
400
300
200
100
0
dN/dlogD$_p$ (cm^{-3})
10
100
1000
D$_p$ (nm)
CLAIRE-98
CLAIRE-01
Suriname1
Suriname2
AMAZE-08
800
700
600
500
400
300
200
100
0
dN/dlogD$_p$ (cm^{-3})
10
100
1000
D$_p$ (nm)

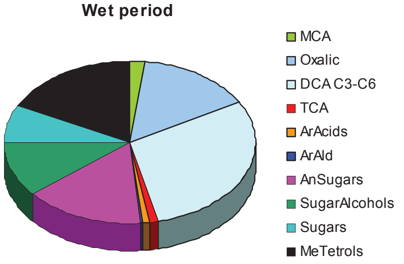
Figure 18



Total identified organic compounds: 6.1 µg m^{-3} (2.5 µg C m^{-3})
WSOC: 19.5 µg C m^{-3}
OC: 26.9 µg C m^{-3}

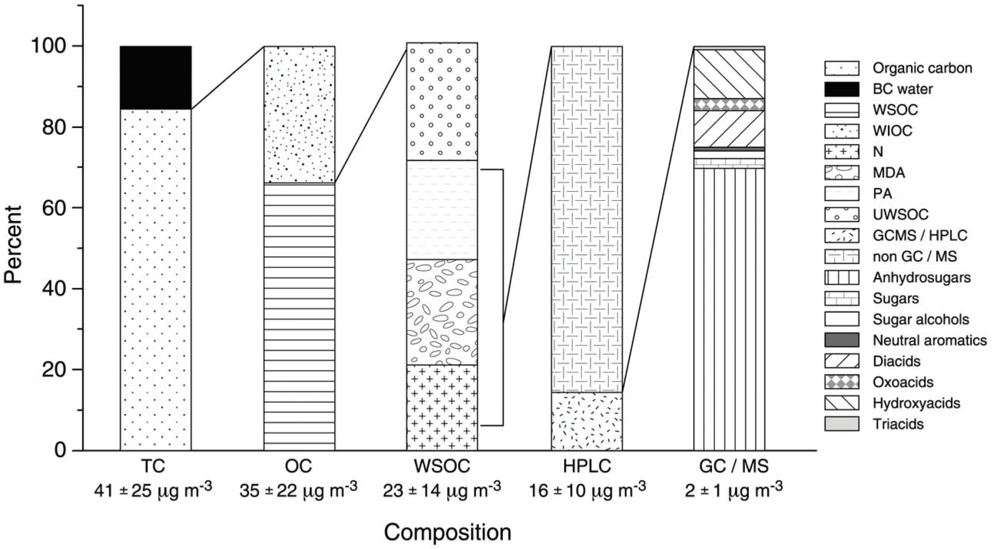


Total identified organic compounds: 1.6 µg m^{-3} (0.6 µg C m^{-3})
WSOC: 6.4 µg C m^{-3}
OC: 8.0 µg C m^{-3}



Total identified organic compounds: 0.25 µg m^{-3} (0.10 µg C m^{-3})
WSOC: 0.85 µg C m^{-3}
OC: 1.42 µg C m^{-3}

Figure 19



Percent
0
20
40
60
80
100
TC
41 ± 25 µg m-3
OC
35 ± 22 µg m-3
WSOC
23 ± 14 µg m-3
HPLC
16 ± 10 µg m-3
GC / MS
2 ± 1 µg m-3
Composition
Organic carbon
BC water
WSOC
WIOC
N
MDA
PA
UWSOC
GCMS / HPLC
non GC / MS
Anhydrosugars
Sugars
Sugar alcohols
Neutral aromatics
Diacids
Oxoacids
Hydroxyacids
Triacids

Figure 20

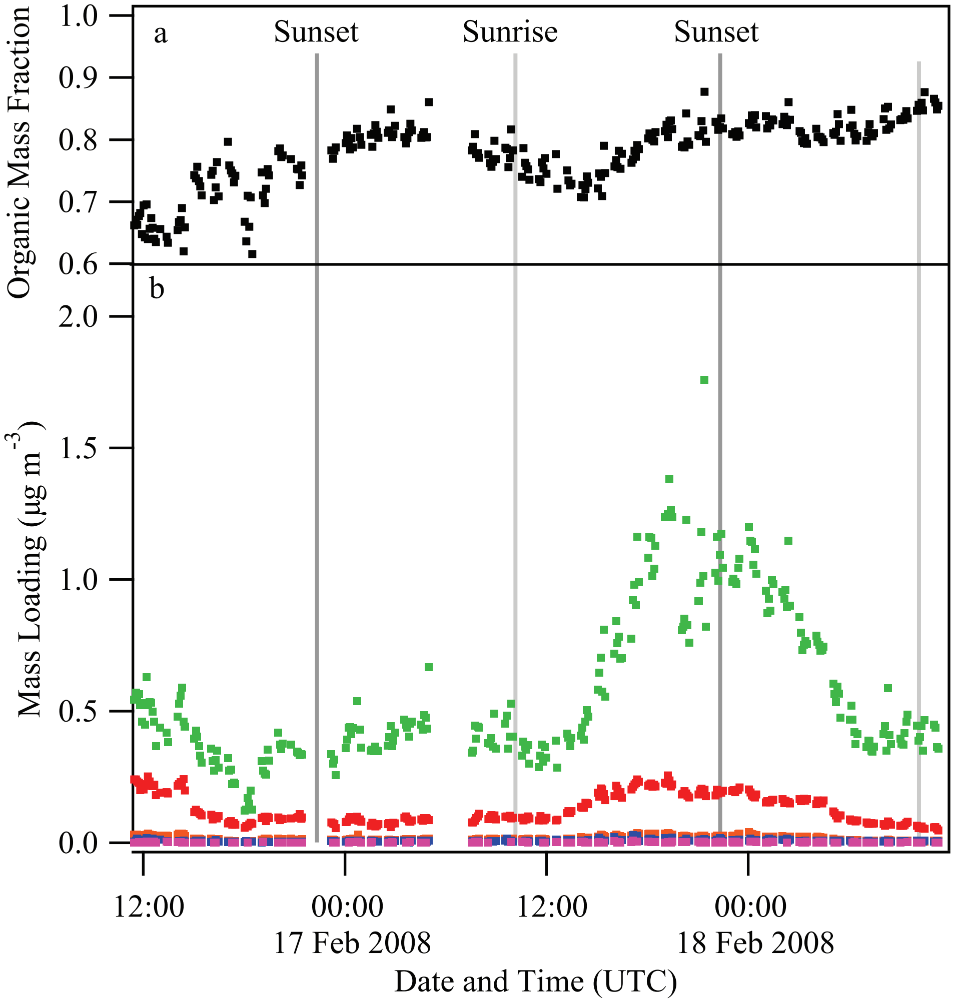

a
Sunset
Sunrise
Sunset
Organic Mass Fraction
1.0
0.9
0.8
0.7
0.6
b
2.0
1.5
1.0
0.5
0.0
Mass Loading (µg m-3)
12:00
00:00
17 Feb 2008
12:00
00:00
18 Feb 2008
Date and Time (UTC)

Figure 21

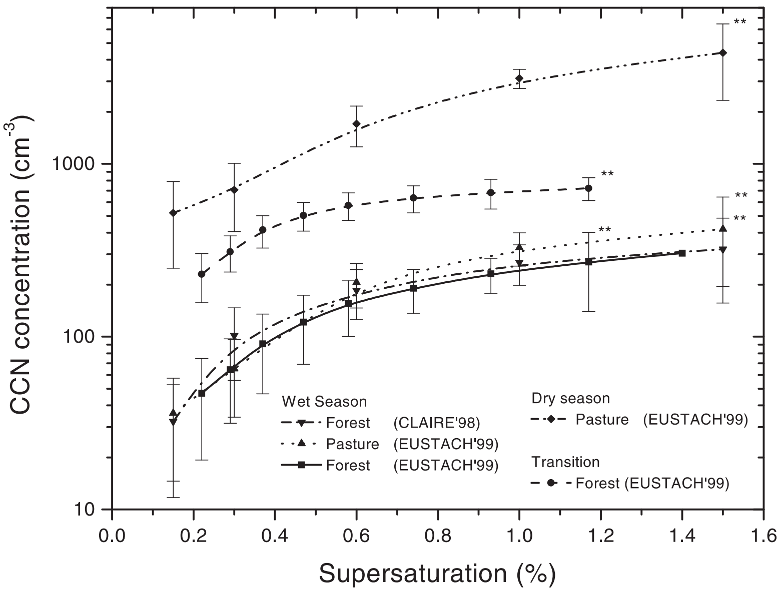

CCN concentration (cm^{-3})
10
100
1000
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
Supersaturation (%)
Wet Season
Forest (CLAIRE'98)
Pasture (EUSTACH'99)
Forest (EUSTACH'99)
Dry season
Pasture (EUSTACH'99)
Transition
Forest (EUSTACH'99)
**
**
**
**
**

Figure 22

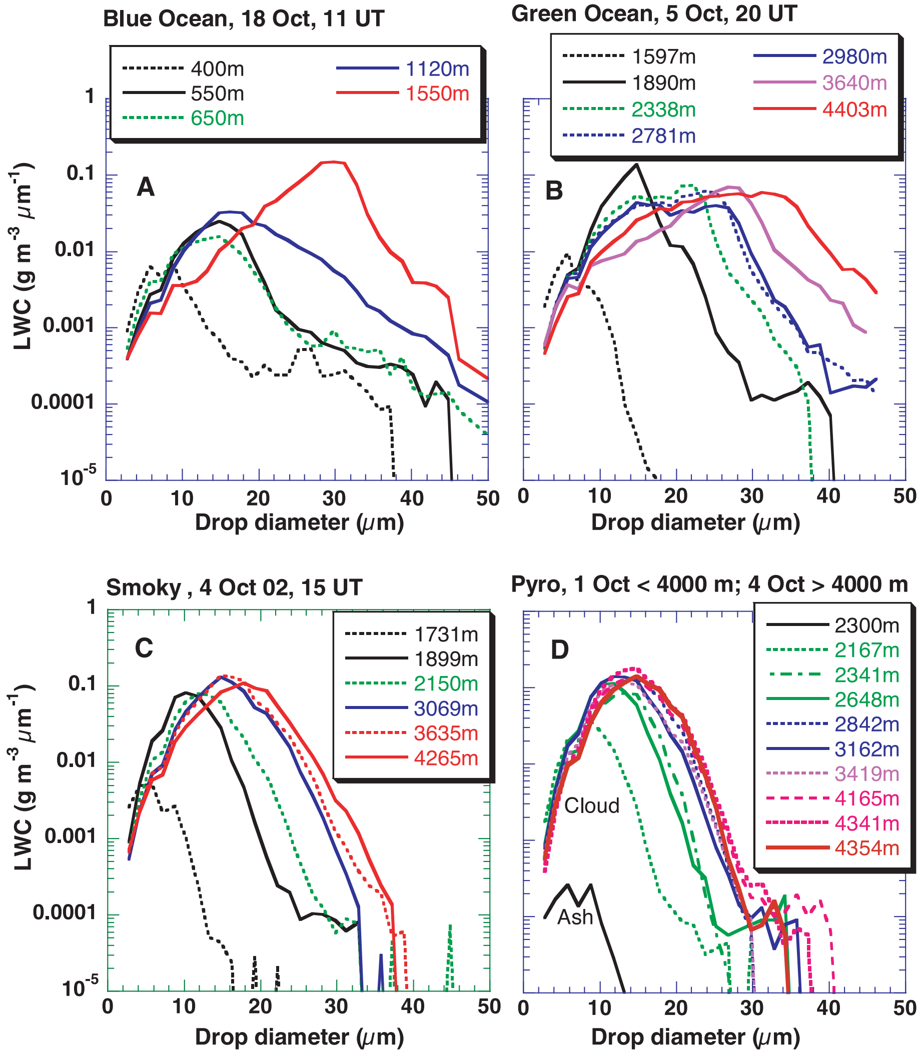

Blue Ocean, 18 Oct, 11 UT
400m
550m
650m
1120m
1550m
A
Green Ocean, 5 Oct, 20 UT
1597m
1890m
2338m
2781m
2980m
3640m
4403m
B
Smoky , 4 Oct 02, 15 UT
1731m
1899m
2150m
3069m
3635m
4265m
C
Pyro, 1 Oct < 4000 m; 4 Oct > 4000 m
2300m
2167m
2341m
2648m
2842m
3162m
3419m
4165m
4341m
4354m
D
Cloud
Ash
LWC (g m^{-3} μm^{-1})
1
0.1
0.01
0.001
0.0001
10^{-5}
0
10
20
30
40
50
Drop diameter (μm)

Figure 23

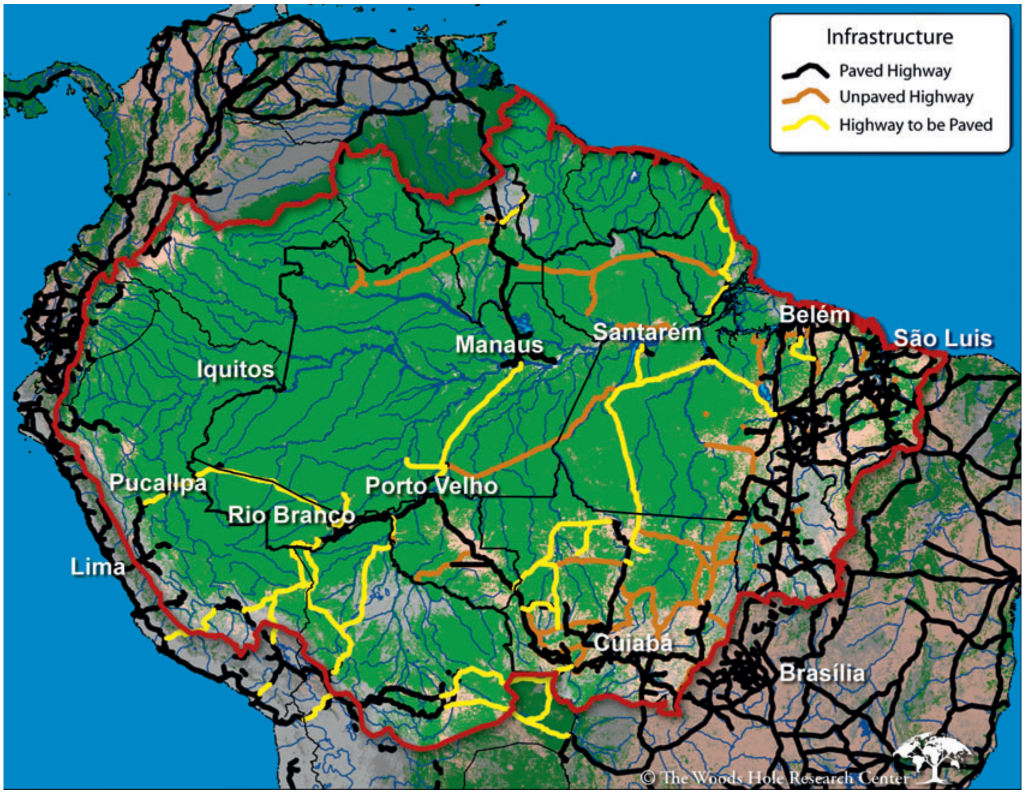

Infrastructure
Paved Highway
Unpaved Highway
Highway to be Paved
Belém
São Luis
Santarém
Manaus
Iquitos
Pucallpa
Porto Velho
Rio Branco
Lima
Cuiabá
Brasília
© The Woods Hole Research Center

Figure 24

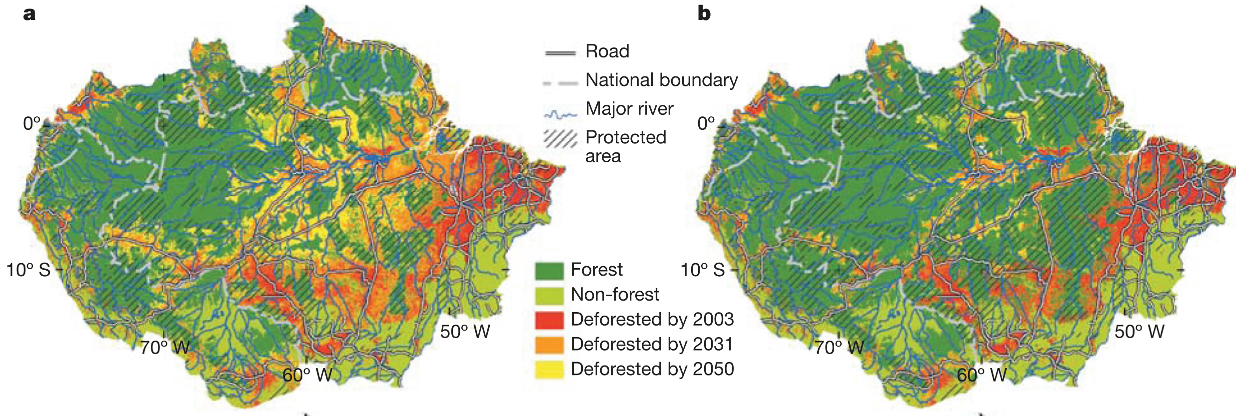
a
b
Road
National boundary
Major river
Protected area
Forest
Non-forest
Deforested by 2003
Deforested by 2031
Deforested by 2050
0°
10° S
70° W
60° W
50° W

Figure 25