Aerosol Particles in Amazonia: Their Composition, Role in the Radiation Balance, Cloud Formation, and Nutrient Cycles

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The atmosphere above tropical forests plays a very active part in the biogeochemical cycles that are critically important for the processes that maintain the ecosystem, including processes involving the vegetation, soil, hydrology, and atmospheric composition. Aerosol particles control key ingredients of the climatic and ecological environment in Amazonia. The radiative balance is strongly influenced by the direct and indirect radiative forcing of aerosol particles. Nutrient cycling is partially controlled by dry and wet deposition of key plant nutrients. It was observed that the aerosol particles that act as cloud condensation nuclei influence cloud formation and dynamics, having the potential to change precipitation regimes over Amazonia. The 10-year-long record of aerosol optical thickness measurements in Amazonia shows a strongly negative radiative forcing of −37 W m^{-2} averaged over 7 years of dry season measurements in Alta Floresta. There is a strong influence of biomass-burning aerosols on the cloud microphysical properties during the dry season. The connections between the amount of aerosol particles and carbon uptake trough photosynthesis highlighted the close connection between forest natural processes and the aerosol loading in the atmosphere. Climate change combined with socioeconomic drivers could alter significantly the emission of trace gases, aerosols, and water vapor fluxes from the forest to the atmosphere. It is a vital task to quickly reduce Amazonian deforestation rates, and to implement solid and long-term conservation policies in Amazonia.

1. INTRODUCTION

The composition of the tropical atmosphere is controlled by a variety of processes ranging from emissions to the processing, transport, and deposition of trace gases and aerosol particles [*Andreae et al.*, 2002]. The understanding of the chemical composition of the tropical atmosphere requires a knowledge of the natural processes related to the production and emission of naturally released chemical species, as well as the anthropogenic emissions associated with land use changes [*Andreae and Crutzen*, 1997]. Once in the atmosphere, chemical compounds are subject to transport, especially associated with convection that can transport compounds over large heights and distances. Processes associated with deposition of trace gases and aerosol particles occur in both dry and wet regimes, and the removal processes are quite efficient in tropical areas. Photochemical processing is also important, since the interaction with solar

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radiation occurs in both the trace gas and particle phases. Cloud processing of trace gases and aerosols is another important process that is not completely understood. During each of those steps, trace gases and aerosol particles strongly influence the Amazonian climate in many different ways [*Davidson and Artaxo*, 2004; S. T. Martin et al., Sources and properties of Amazonian aerosol particles, submitted to *Reviews of Geophysics*, 2008, hereinafter referred to as Martin et al., submitted manuscript, 2005].

Aerosol particles influence the global and regional climate via changes in the radiative balance of the atmosphere as well as through their influence on the hydrological cycle [*Andreae et al.*, 2004, 2005]. Due to its short residence time in the atmosphere and the fact that its concentration varies in space and time, it is not easy to make an accurate quantitative estimation of the aerosol radiative forcing. According to the latest Intergovernmental Panel on Climate Change report [*IPCC*, 2007], the largest uncertainty in climate forcing arises from a lack of understanding of the effect of aerosols on climate (in particular, the indirect effect, i.e., changes in cloud radiative properties induced by aerosol particles) [*Forster et al.*, 2007]. A better understanding of the role that natural aerosol particles play in climate is also critical to quantitatively assess the environmental changes that regions such as Amazonia are suffering [*Andreae*, 2007]. Natural biogenic aerosol particles emitted by plants play an important role in nutrient cycling in tropical ecosystems [*Artaxo et al.*, 2005]. Tropical ecosystems maintain a delicate nutrient balance characterized by intense internal recycling and depend on atmospheric input of certain nutrients to fulfill certain requirements [*Davidson and Artaxo*, 2004].

Aerosol particles influence the climate in two primary manners: first, the aerosol direct radiative effect, which involves the scattering and absorption of solar radiation by aerosol particles changing the net radiative fluxes in the atmosphere and at the ground; second, the aerosol indirect forcing, which is related with aerosol-induced changes in cloud properties. Clouds are a critical ingredient of the radiation balance, and the increase in aerosol particle population changes the number of cloud condensation nuclei (CCN) and clouds properties such as albedo and lifetime. Aerosols also change the atmospheric thermodynamic properties such as the temperature profile and relative humidity variability over large areas; this is generally called the semidirect effect [*Rosenfeld et al.*, 2008].

Significant changes related to human activities are occurring in Amazonia [*Nobre et al*., 2004] that could have global effects on the carbon balance, the concentrations of greenhouse gases and aerosol particles, and on the oxidizing power of the atmosphere. Amazonia is one of the major direct sources of organic aerosols to the global atmosphere. The size and elemental composition of aerosol particles are important variables that influence their role as CCN. Although aerosols are efficiently scavenged by precipitation, the long-range transport of only a small fraction of Amazonian aerosols could make a major contribution to the global budget of the free troposphere [*Andreae and Crutzen*, 1997].

In pristine areas of Amazonia, background primary and secondary biogenic aerosol concentrations are usually very low [*Artaxo et al.*, 2002; *Zhou et al.*, 2002; *Roberts et al.*, 2001], comparable to the observed levels in other remote locations of the planet. The two main sources of natural aerosol particles are the direct emission of primary particles (mostly biogenic), and second, the oxidation of volatile organic compounds (VOC) emitted by the vegetation [*Guenther et al*., 1995; *Claeys et al*., 2004]. Once in the atmosphere, VOCs are subject to chemical and photochemical transformations that can convert some of them to aerosol particles. Due to this predominantly biogenic character of particle formation, aerosol particles in Amazonia are mostly organic [*Graham et al.*, 2003a, 2003b].

Amazonia has been subject to an intensive process of land use change in the last 40 years [see *Alves et al*., this volume; *Soares-Filho et al.*, 2006]. These land use changes are mostly in the region known as the "arc of deforestation," in the southern and eastern parts of the Amazon basin. Large areas are converted from natural forests to pasture or large-scale arable agriculture [see *Walker et al*., this volume]. The main tool used by farmers to remove biomass is fire [*Bowman et al*., 2009]. At the beginning of the dry season, the number of large fires detected by remote sensing tools rises to a few thousand per day. Most of biomass burning takes place in the states of Rondônia, Mato Grosso, and Pará, following road building during the 1970s and 1980s. During the months of August to October, a large part of Amazonia and South America is covered with smoke. This heavy smoke covering millions of square kilometers has profound effects on the radiation balance, cloud formation, and the health of the Amazonian population. Aerosols from biomass burning can travel over large distances [*Andreae et al.*, 2001] and influence areas far from the source regions [see *Longo et al*., this volume].

Biomass-burning aerosols have very complex physical and chemical properties [*Andreae*, 1991]. Several experiments as part of the Large-Scale Biosphere-Atmosphere Experiment in Amazonia (LBA) were dedicated to the study of aerosol particle properties and their effects on the Amazonian ecosystem. A few of these includes the LBA/Cooperative LBA Airborne Regional Experiment (CLAIRE) 1998 and 2001 and LBA/European Studies on Trace gases and Atmospheric Chemistry (EUSTACH) [*Andreae et al.*, 2002]. During the

dry-to-wet season transition in 2002, the LBA/smoke aerosols, **c**louds, rainfall, and **c**limate (SMOCC) campaign was conducted in Rondônia during September to November [*Andreae et al.*, 2004]. The main objective of the experiment was to characterize the optical, physical, and chemical properties of biomass-burning aerosol particles. A review article by *Fuzzi et al.* [2007] synthesized LBA/SMOCC results and the complexities of aerosol particles emitted through biomass burning in tropical areas [see also *Longo et al.*, this volume]. Recently, the Amazonian Aerosol Characterization Experiment-08 experiment has focused on understanding the natural biogenic aerosol properties (Martin et al., submitted manuscript, 2008), with a field measurement campaign in January–March 2008 in Manaus.

Long-term measurements are essential to provide scientific knowledge on the temporal variability of key atmospheric properties. An important example is the operation of NASA's Aerosol Robotic Network (AERONET) that maintains a network of ground-based Sun photometers in Amazonia [*Holben et al.*, 1998]. The strong change in the radiation fluxes at the surface has important consequences on several aspects of the Amazonian ecosystem functioning. A net surface cooling of around 2–3° can be modeled through the result of the aerosol layer, as well as a heating at the levels of 2–3 km in the atmosphere. This effect stabilizes the vertical temperature profile, reducing convection and the transport of water vapor to upper levels. The radiation field is also strongly affected, with a reduction of direct solar radiation on the surface, and increase in the diffuse solar radiation reaching the forest.

The aim of this chapter is to review the main issues regarding atmospheric chemistry in Amazonia, with emphasis on the role of aerosol particles in the ecosystem functioning including deposition of nutrients and radiation balance.

2. PROPERTIES OF NATURAL PARTICLES IN AMAZONIA

Vegetation has long been recognized as an important source of both primary and secondary aerosol particles [*Artaxo and Hansson*, 1995; Martin et al., submitted manuscript, 2008]. Aerosol particles are responsible for the airborne transport of phosphorus, calcium, sulfur, nitrogen compounds, and other essential nutrients. Only a few studies of natural biogenic aerosols from vegetation in tropical rain forests have been undertaken [*Artaxo et al.*, 1990; *Artaxo and Hansson*, 1995; *Echalar et al.*, 1998; *Artaxo*, 2001]. Biogenic aerosols consist of many different types of particles, including pollen, spores, bacteria, algae, protozoa, fungi, fragments of leaves, excrement, and fragments of insects. This aerosol component is mainly in the coarse size fraction ($dp > 2\ \mu m$). The mechanisms of particle emission are still not well understood. They probably include mechanical abrasion by wind, biological activity of microorganisms on plant surfaces and forest litter, and plant physiological processes such as transpiration and guttation. Fungi, through the release of fungal spores, are a major source of primary biogenic aerosol particles and components, and especially in the humid Amazonian environment, they are expected to be a significant source of aerosol particles [*Elbert et al.*, 2007].

These processes may generate particles containing biogenic-related elements such as Na, Mg, P, S, K, Ca, Zn, and Rb. The transpiration of plants can lead to migration of Ca^{2+}, SO_4^{2-}, Cl^-, K^+, Mg^{2+}, and Na^+ to the atmosphere. The biogenic-related elements (Mg, K, P, S, Zn, Rb, and others) are essential to plants; they are present in the fluids circulating in the plant and are released from the leaves to the atmosphere. Table 1 shows the average elemental concentration for aerosol particles in the wet season in Rondônia [*Artaxo et al.*, 2002]. The very low aerosol concentration under natural conditions is evident from the 2.21 $\mu g\ m^{-3}$ of fine mode mass in the wet season. In particular, an average of 88 $ng\ m^{-3}$ for sulfur is very low for any continental region. Soil dust also appears with very low concentrations, with Fe concentrations of only 34 $ng\ m^{-3}$ in the coarse-mode aerosols. These low elemental concentrations contrast with the high aerosol loading during the dry season in Rondônia.

The optical properties of aerosols in the wet season show a surprisingly strong absorption of radiation in the visible wavelengths by the biogenic particles [*Schmid et al*., 2006; *Guyon et al.*, 2004]. The biogenic aerosols absorb light very efficiently due in part to their morphology and elemental composition, with the presence of humic substances. The absorption efficiency is higher than for biomass-burning particles. This strong absorption effect has important implications in the atmospheric radiative budget and ground-based temperature over large areas of Amazonia [*Hoffer et al.*, 2006; *Schmid et al.*, 2006].

Biogenic volatile organic compounds (BVOC), which following reaction with OH or O_3, are important precursors to secondary organic aerosol production in the Amazon basin, are emitted from plants during growth, maintenance, decay, and consumption. Tropical forests are the dominant global source of atmospheric BVOCs, and the Amazon basin is a major contributor [*Rasmussen and Khalil*, 1988]. The basin contains on the order of 10^5 plant species, each with a unique BVOC emission signature. This high species diversity is coupled with a dramatic ecological complexity and a seasonality that is very different from temperate regions, where BVOC emissions have been studied more extensively. These factors combine to make estimates of BVOC emissions from the whole of Amazonia an important but challenging task. Prior

Table 1. Average Elemental Concentration for Fine- and Coarse-Mode Aerosol Particles in the Wet Season in Rondônia at the Rebio Jaru Large-Scale Biosphere-Atmosphere Experiment in Amazonia (LBA) Tower[a]

	Wet Season Aerosol Composition in Rondônia					
	Fine Fraction			Coarse Fraction		
	Mean	SD	*n*	Mean	SD	*n*
Mass	2.21	1.39	(28)	3.77	1.32	(28)
TC	896.7	504.6	(11)	–	–	–
EC_a	76.6	59.3	(11)	–	–	–
BC_e	276.9	137.0	(28)	–	–	–
Mg	–	–	–	29.0	23.2	(23)
Al	34.5	30.9	(24)	52.5	50.8	(24)
Si	42.5	37.9	(28)	52.0	59.8	(25)
P	5.6	1.9	(28)	23.4	9.3	(28)
S	87.7	87.2	(28)	23.5	14.8	(28)
Cl	6.1	7.9	(11)	8.9	5.4	(28)
K	26.8	26.5	(28)	48.2	14.9	(28)
Ca	8.4	6.1	(28)	12.2	7.5	(28)
Ti	3.8	4.1	(14)	6.5	7.3	(14)
Cr	–	–	–	2.5	1.5	(8)
Mn	0.56	0.36	(28)	0.87	0.57	(28)
Fe	21.6	29.7	(26)	34.2	46.8	(26)
Ni	0.33	–	(1)	–	–	–
Cu	0.42	0.44	(27)	0.32	0.20	(28)
Zn	0.68	0.72	(23)	0.72	0.39	(28)
Br	1.05	–	(1)	–	–	–
Sr	0.16	0.07	(10)	0.26	0.13	(9)
Zr	0.18	0.08	(6)	0.77	0.78	(4)
Pb	0.84	0.92	(18)	0.46	0.17	(3)

[a]Mean and standard deviations (SD) are shown; *n* is the number of samples where the detected elements were observed above detection limit. Mass concentrations are expressed in μg m^{-3}; equivalent black carbon (BC_e) and trace elements concentrations are in ng m^{-3}. Table 1 is adapted from *Artaxo et al.* [2002].

to new studies conducted in the past decade, Amazonian BVOC emission estimates were based on a few measurements conducted by *Zimmerman et al.* [1988] and *Kesselmeier et al.* [2000]. However, due to high biodiversity and spatially different radiation and temperature fields, much more extensive work on VOCs and the production of secondary aerosols are needed in Amazonia.

A prominent contribution of particles from outside the Amazon basin is that made by Saharan dust. The importance of transatlantic transport of dust was recognized by *Prospero et al.* [1981] and has been observed in several subsequent measurement campaigns [*Swap et al.*, 1996; *Artaxo et al.*, 1988, 1998; *Formenti et al.*, 2001]. Imported dust occurs at its highest concentrations in those parts of the basin that are north of the Intertropical Convergence Zone (ITCZ). The maximum dust concentrations at the surface are typically reached around March and April, coinciding with the wet season in the central basin. The dust at ground stations is observed in pulses of high concentrations that last from one to several days. Given the large transport distance from Africa, a significant fraction of the mineral dust depositing in the Amazon basin is submicron.

Marine aerosol particles consist largely of primary sea spray 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 [*Andreae and Rosenfeld*, 2008]. Marine emissions dominate the particle population that enters the Amazon basin with the trade wind flow, being progressively removed by wet and dry deposition as an air mass travels deeper into the basin. Even so, the relative contribution by marine particles to the total Amazonian particle mass concentration remains significant even over the central parts of the basin during the wet season. This can be explained by the large concentrations of marine particles present in the air, as it crosses the coast and the relatively slow rates at which aerosol particles are

removed over the continent [*Andreae and Andreae*, 1988; *Worobiec et al.*, 2007].

3. PROPERTIES AND EFFECTS OF BIOMASS-BURNING AEROSOL PARTICLES IN AMAZONIA

The very clean atmospheric conditions that predominate in the wet season in Amazonia change strongly in the dry season with large emissions of biomass-burning aerosols [*Andreae et al.*, 2002; *Echalar et al*., 1995]. Typical aerosol number concentrations for the wet season is around 200–300 particles cm^{-3}. In the dry season, particle number concentrations jump to 10,000–20,000 cm^{-3} [*Artaxo et al.*, 2002]. Aerosol mass concentrations (PM_{10}) for the wet season are typically 10–12 $\mu g\ m^{-3}$, while in the dry season, they can reach extremely high values of 600 $\mu g\ m^{-3}$. This large aerosol concentration can lead to important effects on human health, clouds, and radiation balance [*Kaufman et al.*, 1998]. Figure 2 shows the deforestation rate in square kilometers per year from the late 1970s to 2008. Large year-to-year variability is observed, due to climatic and to socioeconomic drivers. Figure 1 shows that deforestation has declined significantly from 2004 to 2007, but an increase in 2008 has changed the recent trend. A deforestation of 10,000 to 20,000 km^2 per year from burning injects huge amounts of particles in the atmosphere [*Yokelson et al.*, 2007, 2008]. Emission factors from primary deforestation fires and pasture maintenance fires in tropical rainforests range from 6 to 25 $g\ kg^{-1}$ for total PM and 7.5 to 15 $g\ kg^{-1}$ for $PM_{2.5}$, 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 a^{-1}, respectively [*Yokelson et al.*, 2008].

One of the consequences of these large areas being burned is the emission of large number of particles and the high aerosol concentrations [*Artaxo et al.*, 2002; *Andreae and Merlet*, 2001; *Hoffer et al.*, 2006]. Table 2 shows the average elemental concentrations for aerosol particles in the dry season in Rondônia collected at the Rebio Jaru LBA tower. Fine-mode sulfur now appears at a high concentration of 533 ng m^{-3}, which is six times higher than the values measured in the wet season, a similar increase as black carbon. Fine-mode potassium concentration increases by 18 times. This large change in trace element concentrations affects the biogeochemical cycles of several key nutrients such as phosphorus.

The Amazonian atmosphere is not isolated from the global atmosphere and interacts strongly with the nearby continents and oceans [*Andreae et al.*, 2001]. *Swap et al.* [1996] showed that particles originating in the Sahara desert reach Amazonia and can be important in terms of nutrient cycling over large time spans. *Formenti et al.* [2001] also observed Saharan dust particles over the northern part of Amazonia and in Suriname, indicating that this process is more important than previously thought. The biomass-burning emissions of soluble iron also have important implications in the South Atlantic primary production, since Fe is critically important to the ocean biogeochemistry and is emitted in large amounts by biomass burning [*Luo et al*., 2008]. Mercury emissions from biomass burning can be significant, as was observed in the study of *Artaxo et al.* [2000], with airborne measurements of Hg and black carbon showing a strong relationship. It is

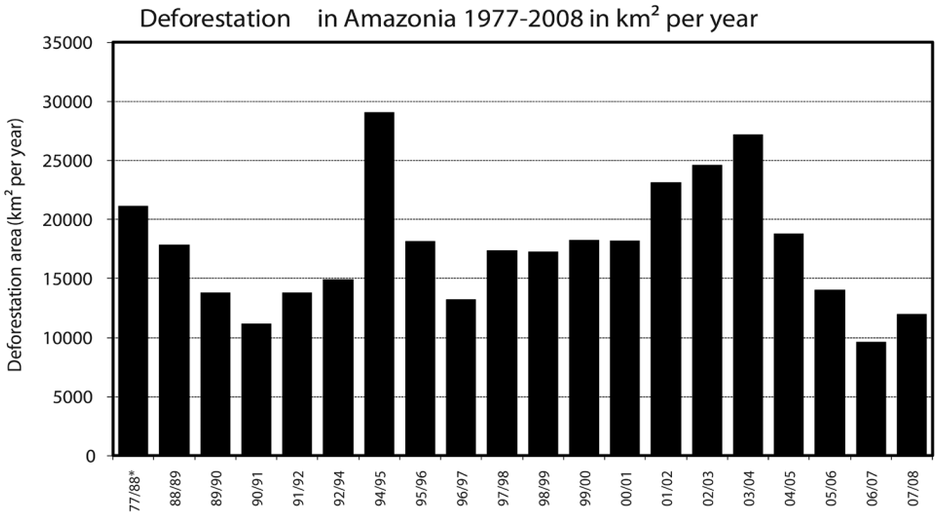

Figure 1. Deforestation rate in the Brazilian Amazonia in $km^2\ a^{-1}$ from late 1970s to 2008. It is possible to observe a decrease in deforestation rate from 2003 to 2007, followed by an increase in 2008.

Table 2. Average Elemental Concentration for Aerosol Particles in the Dry Season in Rondônia at the Rebio Jaru LBA Tower[a]

	Dry Season Aerosol Composition in Rondônia					
	Fine Fraction			Coarse Fraction		
	Mean	SD	*n*	Mean	SD	*n*
Mass	33.5	21.8	(79)	6.6	3.0	(79)
TC	17.1	8.8	(28)	–	–	–
EC_a	0.74	0.34	(28)	–	–	–
BC_e	1.82	1.30	(79)	–	–	–
Al	91.0	63.7	(72)	80.0	50.0	(26)
Si	118.8	97.1	(80)	95.0	133.8	(62)
P	26.7	11.7	(81)	47.7	23.0	(81)
S	532.7	288.8	(81)	59.8	26.6	(81)
Cl	20.4	11.5	(19)	10.1	5.1	(20)
K	506.2	355.3	(81)	93.1	56.2	(79)
Ca	25.1	26.2	(75)	52.3	74.8	(76)
Ti	6.0	4.4	(50)	8.0	5.3	(42)
Cr	10.3	6.2	(15)	15.4	17.1	(31)
Mn	1.87	1.40	(79)	4.5	5.5	(79)
Fe	28.6	28.0	(81)	48.0	47.5	(77)
Cu	1.66	1.57	(67)	3.9	6.1	(24)
Zn	4.3	3.5	(81)	4.3	4.8	(7)
Br	10.2	6.4	(43)	25.1	15.6	(5)

[a]Mean and standard deviations (SD) are shown; *n* is the number of samples where the detected elements were observed above detection limit. Mass concentrations are expressed in $\mu g\ m^{-3}$; equivalent black carbon (BC_e) and trace elements concentrations are in $ng\ m^{-3}$. Table 2 is adapted from *Guyon et al.* [2003a, 2004b].

always important to emphasize that the burning of the forest not only releases particles, but also large amounts of important trace gases, which include, VOCs, methane, CO, CO_2, and several other key species [*Karl et al.*, 2007a, 2007b; *Yokelson et al.*, 2007, 2008; *Andreae and Merlet*, 2001].

4. AEROSOL OPTICAL DEPTH MEASUREMENTS THROUGH THE AERONET NETWORK IN AMAZONIA

One of the strong points in the atmospheric chemistry component of LBA is the long-term aerosol monitoring stations operated in Amazonia for the last 10 years. One of the set of measurements was obtained using Sun photometers within AERONET. AERONET is a globally distributed network of well-calibrated and standardized Sun photometers, maintained by NASA and expanded by national and international collaborations [*Holben et al.*, 1998]. A CIMEL Sun-sky radiometer (manufactured by CIMEL Electronique, France) and total radiation sensors and photosynthetic active radiation sensors are deployed at several sites in Brazil for measuring the aerosol optical thickness (AOT) and solar flux in the total solar spectrum. Currently, in Brazil, there are seven Sun photometers in operation: Ji Paraná (JP) (Rondonia (RO)), Alta Floresta (AF) (Mato Grosso (MT)), Cuiabá (CB) (MT), Rio Branco (Acre), Campo Grande (Mato Grosso do Sul (MS)), São Paulo (SP), and Petrolina (Pernambucco), but several other sites such as Balbina (BA) (Amazonas), Belterra (Pará (PA)), Santarém (PA), Brasília (Distrito Federal), and Reserva Biológica Jaru (RO) have been operated [*Schafer et al*., 2008]. The CIMEL Sun-sky radiometer measures radiances in several wavelengths, in near real-time and provides, from direct Sun observations, aerosol properties such as AOT, column water vapor and, from sky observations, aerosol size distribution, absorption properties, and other key aerosol properties [*Holben et al.*, 1998]. This network is the only long-term project (with a record including observations from more than 11 years at some locations) ever to have provided ground-based remotely sensed column aerosol properties for this critical region. The monitoring sites generally include measurements from 1999 through the present day, but some sites have measurement records that date back to the initial days of the AERONET program in 1993 [*Schafer et al*., 2008].

All LBA-AERONET sites exhibit similar seasonal trend in atmospheric properties with very low aerosol loading dur-

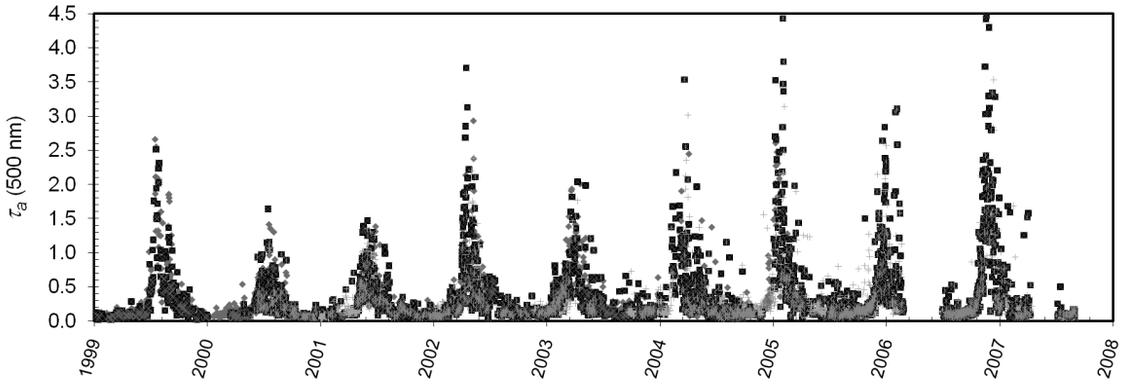

Figure 2. Time series of daily average of aerosol optical thickness (AOT) in Ji Paraná (JP), Alta Floresta (AF), Balbina, and Cuiabá. Note the very high increase in AOT every year in the dry season.

ing the wet season (January–June). As the dry season starts, aerosols from biomass burning increase very significantly the amount of aerosols in the atmosphere. Figure 2 shows the time series of daily average AOT (at 500 nm) in JP, AF, BA, and CB. In the wet season, very low AOT (around 0.1–0.2) is observed for all sites. BA shows some increase during the dry season, but this increase is modest. During the dry season, values as high as 3.5 are frequently observed in AF, CB, and JP. These are the highest AOT values observed for all worldwide AERONET sites.

The AERONET Sun photometers also measure continuously the total column water vapor (CWV) [*Yamasoe et al*., 1998]. Figure 3 shows the time series of the water vapor column in AF (Mato Grosso), JP (Rondonia), and Belterra (near Santarem, Pará). In the northern part of the basin, water vapor changes slightly on an annual basis, as can be seen for the BA CWV data, since the dry season is not very strong in this part of the basin. In the southern part of Amazonia, the annual cycle of water vapor is much more pronounced, with water vapor sometimes as low as 1.5 cm [*Schafer et al.*, 2008].

Figure 4 shows an analysis of the average seasonality with the weekly average values of AOT sites in the northern part of Amazonia, as well as sites in the cerrado and in forest sites in the southern part of the basin. It is clear that in the south, the most intense biomass burning occurs in August and September, while in the northern part, the most impacted period is November–December [*Schafer et al.*, 2008].

Aerosol forcing is the most important uncertainty in global and regional climate change [*IPCC*, 2007], making it important to reduce uncertainties in the parameters relevant to the radiative forcing calculations to get aerosol optical depth with high spatial resolution. Currently, using the Moderate Resolution Imaging Spectroradiometer (MODIS) sensor from AQUA and TERRA satellites, efforts are being undertaken for getting AOT with a high spatial resolution of 1 km × 1 km in Amazonia. For this purpose, models of aerosol optical properties divided in single-scattering albedo ranges were obtained using AERONET data. This approach involves obtaining AOT not just for a few AERONET sites, but for the whole area of Amazonia. The main problem is the high cloud

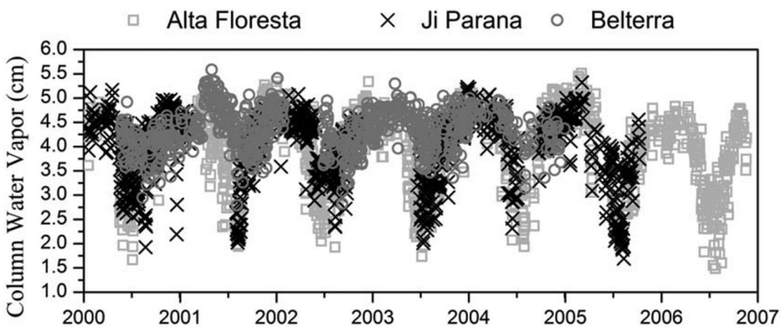

Figure 3. Time series of daily average of columnar water vapor (in centimeters of water vapor) in AF (Mato Grosso), JP (Rondonia), and Belterra (near Santarem, Pará). Data from the AERONET Sun photometer network.

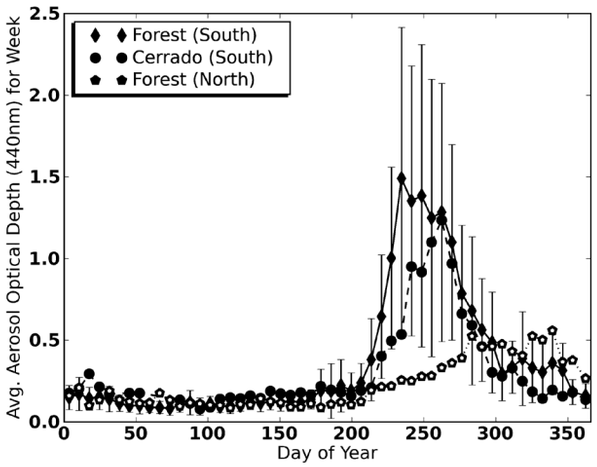

Figure 4. Weekly averages of aerosol optical depth (at 440 nm) for regionally grouped sites in Amazonia [modified after *Schafer et al.*, 2008]. The increase in aerosols due to biomass burning is more pronounced in the southern part of the Amazon Basin.

cover, but improved algorithms are being developed to increase the accuracy of AOT measurements using MODIS.

5. DIRECT RADIATIVE FORCING OF AEROSOL PARTICLES AND EFFECTS ON THE AMAZONIAN ECOSYSTEM

The deforestation processes have been disturbing the Amazonian forest ecosystems in several different ways [*Ometto et al.*, 2005]. As an example, the conversion of natural forest to pasture changes the energy and water balances and, consequently, can alter the atmospheric water content and precipitation patterns [*Silva Dias et al.*, 2002]. These processes release a large amount of aerosol particles to the atmosphere, leading to strong changes in the surface radiation balance [*Chand et al.*, 2006; *Schafer et al.*, 2008; *Procópio et al.*, 2004]. The interaction between the downward solar radiation with these aerosol particles and clouds affect directly the atmospheric radiative budget reducing the direct incoming solar radiation and increasing its diffuse fraction [*Schafer et al.*, 2002a, 2002b; *Niyogi et al.*, 2004; *Gu et al.*, 1999, 2003]. LBA made long-term measurements of detailed aerosol optical properties using several AERONET Sun photometers and radiometer in several sites [*Eck et al*., 1998]. The reduction in the surface radiation fluxes was monitored for AF, and Figure 5 shows a time series of 9 years of aerosol direct radiative forcing in AF (located in the northern portion of the Mato Grosso state), a representative area of biomass-burning influence. Instantaneous direct radiative forcing of up to −300 w m^{-2} (the radiative forcing is negative because this effect subtracts solar radiation at the ground) is observed most of the years during the dry season. The observations over many years of dry season measurements resulted in an average surface radiative forcing of –37 W m^{-2} [*Procópio et al.*, 2004], which is a very significant cooling effect in the surface level.

As shown by *Oliveira et al.* [2007], a small increase in atmospheric aerosol loading increases the fraction of diffuse versus direct radiation. Therefore, the vegetation increases the efficiency of the use of solar radiation and consequently

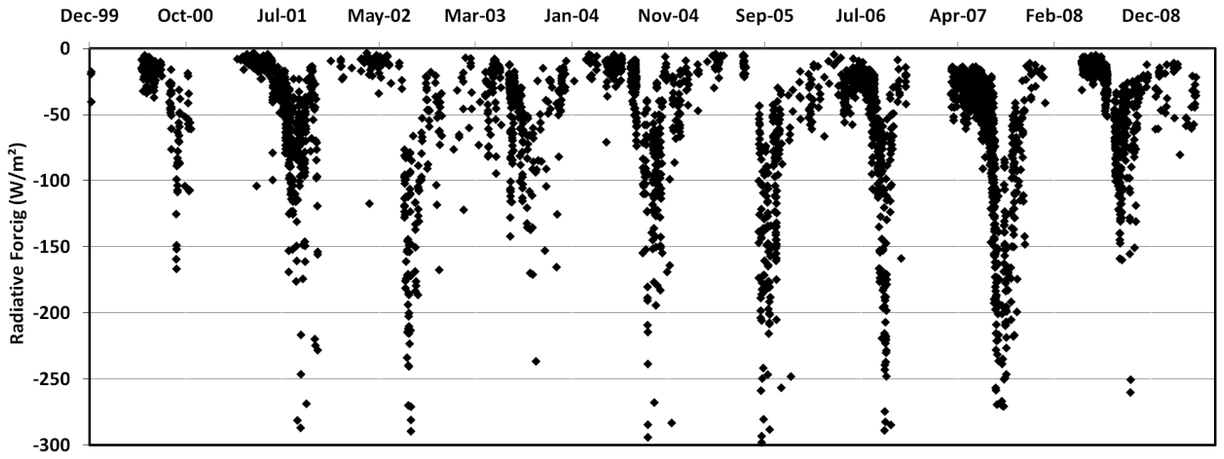

Figure 5. Instantaneous direct aerosol forcing (in W m^{-2}) measured over 9 years in AF, Mato Grosso state, a region heavily impacted by biomass-burning emissions. Note the very large instantaneous forcing of up to −300 W m^{-2} measured almost every year during the dry season.

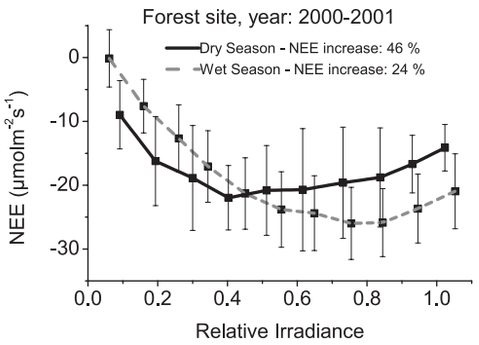

Figure 6. Net ecosystem exchange (carbon flux in μmol m^{-2} s^{-1}) as a function of aerosol loading in the atmosphere expressed as the relative irradiance index. Relative irradiance 1 means no aerosols, with increase in aerosols as the radiation decreases. Note that negative values of NEE correspond to net ecosystem uptake and that relative irradiance index decreases with increasing AOT [*Oliveira et al.*, 2007].

increases the net primary productivity (NPP, defined as the net flux of carbon from the atmosphere into the vegetation per unit of time), but up to a certain point. From clean conditions, characterized as with AOT, around 0.1 at 500 nm to AOT of 1.2, the NPP increases by 30–50% in the dry season and 24% in the wet season in Rondônia [*Oliveira et al.*, 2007]. As soon as the AOT becomes larger than 1.2 at 500 nm, the reduction in the total flux starts to shut off carbon assimilation, and for AOT of about 3–4, the vegetation stops assimilating carbon due to the large reduction in the solar radiation flux, as can be seen in Figure 6.

As the area of the biomass-burning plumes in South America, Africa, and Southeast Asia is very large, the effect of biomass-burning aerosols on the carbon exchange is an important factor in the Southern Hemisphere [*Artaxo and Andreae*, 2007]. Particles emitted through vegetation fires absorb solar radiation very efficiently because of the large amount of black carbon in these particles [*Artaxo et al.*, 1988, 1998].

Taking into account that due to biomass-burning emissions there are large areas in Amazonia where there is a significant aerosol loading for about 4 months, the effects of aerosols on the carbon uptake by the Amazonian forest is possibly very significant. The effects observed in Amazonia are certainly also present in tropical forests of Africa and Southeast Asia because of the similar biomass-burning conditions and type of forest. Aerosol particles from biomass burning affect the global carbon budget, and quantification at a global scale would bring important extra knowledge of the global carbon cycle. Additionally, it was observed that large amounts of ozone are formed from biomass-burning emissions [*Longo et al.*, 1999]. The ozone levels observed, which are sometimes higher than 100 ppb, can play an important role in the primary productivity of Amazonian vegetation because some species and cultivars are sensitive to high levels of ozone [*Bulbovas et al.*, 2007]. Ozone is also a greenhouse gas and changes photochemistry significantly, potentially altering the production of secondary organic aerosols in ways still to be studied (Martin et al., submitted manuscript, 2008).

6. AEROSOLS AND CLOUDS: NUCLEATION PROPERTIES OF BIOGENIC AND BIOMASS-BURNING PARTICLES

Clouds are a critically important component of the Amazonian ecosystem because of their effect on the hydrological cycle and the radiation balance. The formation of clouds in the Amazon basin takes place under conditions of high water vapor availability, low CCN concentrations, high temperature, and solar radiation, which are quite different from other continental areas of the world [*Williams et al.*, 1997; *Kaufman and Koren*, 2006]. Precipitating clouds are generally divided in two classes: low-level stratus type clouds (up to 2–5 km in altitude) and high-level convective systems (more than 6 km altitude). In Amazonia during the wet season, most cloud fields are comprised of the so-called low-level warm clouds. High convective systems are responsible for most of the precipitation, and they form and develop under special thermodynamic conditions [*Silva Dias et al.*, 2002]. However, even during the dry season, the warm clouds are present in fair weather cumuli as well as in precipitating clouds. This seasonal difference is mainly driven by large-scale phenomena, which control the wet/dry season patterns.

In such an environment where warm clouds play an important role in the hydrological cycle, the concentration of atmospheric CCN and ice nuclei and updraft velocities are the critical characteristic of the atmosphere in the formation and properties of convective systems [*Prenni et al.*, 2009; *McFiggans et al.*, 2006]. A surprising result obtained during the LBA/CLAIRE intensive campaign (March–April 1998, in central Amazonia) is that, when free of anthropogenic emissions, the typical concentration of CCN in Amazonia is very low at about 200 cm^{-3} at 1% supersaturation (SS) [*Roberts et al.*, 2000, 2001] (Figure 7). During the dry season, CCN concentrations at 1% SS goes to very high values of around 3000 cm^{-3}. It means that, under natural conditions, the typical concentration of natural biogenic CCN in Amazonia resembles more those ones found in oceanic than in continental areas. Typical oceanic CCN concentrations are about 100–200 per cm^3, whereas typical background

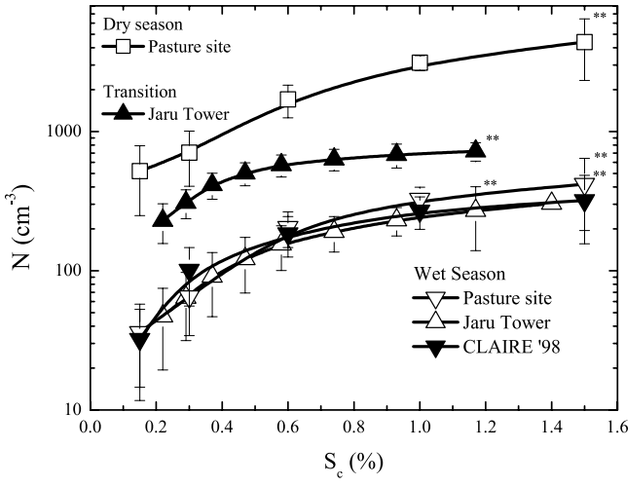

Figure 7. Number of cloud condensation nuclei particles versus supersaturation for wet season, transition, and dry season in several regions in Amazonia. Data from *Roberts et al.* [2001].

continental values are within 600–1000 cm^{-3} [*Pruppacher and Klett*, 1998]. It was found also that a significant fraction of the aerosol particles in Amazonia (40–60%) actually could act as CCN [*Gunthe et al*., 2009]. This is due to their relatively large size and the predominantly water-soluble chemical composition [*Roberts et al.*, 2001].

Because of these two features, the low CCN concentration and the predominance of warm clouds, the Amazon basin has been called "the green ocean" [*Williams et al.*, 2002]. The comparison of cloud structure between oceanic and Amazonian conditions is valid, as pointed out in the pioneer study of *Squires* [1956]. As over oceans, clouds in the Amazon basin typically present low vertical development and are very efficient at producing rain quickly. The role of the low CCN concentration is important because it yields cloud droplets that are initially already large, with a fast and efficient growth mechanism through water vapor diffusion. The large radii of these droplets improve the effectiveness of the collision-coalescence phase of rain formation, making the production of precipitation very efficient.

Most of these CCN particles are biogenic in origin. Most of them originate from gas-to-particle conversion of VOCs emitted naturally by the vegetation [*Guenther et al.*, 1995; *Claeys et al.*, 2004], while some of the larger particles are primary biological particles. The composition of such particles is mainly organic [*Artaxo et al.*, 1990, 1994, 1998], with very low concentrations of sulfur and heavy metals. Further, they are rich in water-soluble organic compounds (WSOC), and the presence of soluble matter helps in the effectiveness of an aerosol particle in acting efficiently as a CCN [*Rissler et al.*, 2006].

The scenario is completely different in regions dominated by biomass-burning activity. In these affected areas, the particle population rises from a few hundred per cubic centimeter [*Zhou et al.*, 2002] to concentration levels as high as 10,000 cm^{-3} during most of the dry season [*Rissler et al.*, 2006]. This extra loading of particles is released basically in the fine-mode fraction [*Guyon et al.*, 2005; *Artaxo et al.*, 2002], which makes the biomass-burning aerosols very susceptible to large-scale transport by winds, and influencing large areas that are free of local biomass-burning emissions. The chemical composition of these biomass-burning particles, as demonstrated in many studies, is predominantly organic matter distributed in a myriad of components, which makes them very efficient as CCN [*Mircea et al.*, 2005; *Graham et al.*, 2003a, 2003b; *Mayol-Bracero et al.*, 2002; *Falkovich et al.*, 2005; *Decesari et al.*, 2006; *Fuzzi et al.*, 2007]. Actually, WSOC affect the CCN properties by contributing to the solute material, altering the surface tension of the growing droplet and affecting the mechanisms responsible for the growth of activated droplets [*Vestin et al.*, 2007; *Sun and Ariya*, 2006]. The complete understanding of these properties are largely unknown even for the relevant WSOC species found in the atmosphere and are currently an area of intense research [*Svenningsson et al.*, 2006; *McFiggans et al.*, 2006].

This large increase in CCN and droplet concentrations has profound effects in cloud microphysics properties [*Freud et al.*, 2008]. *Roberts et al.* [2003] used a one-dimensional cloud parcel model to assess the impact of biomass-burning aerosols on cloud properties. They found that properties such as cloud droplet effective radius and maximum SS are more sensitive at low CCN concentrations, which would lead to larger interannual variation of cloud properties during the wet season than the dry season. However, the authors also conducted measurements of CCN spectra, and they observed few differences between forested and deforested regions during the wet season and that the resulting modifications of cloud properties are small compared to those between wet and dry seasons. They postulate that, for the case of the wet season, differences in surface albedo between forested and deforested regions may dominate the impact of deforestation on the hydrological cycle and convective activity during the wet season. The large population of droplets and the inhibited growth mechanisms can make it difficult for droplets to reach the 25-μm threshold for precipitation, leading to a larger cloud fraction that evaporates instead of precipitates [*Andreae et al.*, 2004]. The most extreme example of the influence of biomass-burning aerosol particles is the formation of pyroclouds (i.e., clouds that form in the smoke

plume over an active fire). The pyroclouds feed directly on the smoke and heat from the fires. They receive conflicting impacts: on extreme concentrations of CCN suppress the onset of precipitation and the fire-generated heat invigorates the updrafts and further suppresses warm rain processes [*Andreae et al.*, 2004, 2008].

Microphysical properties of those pyroclouds are very different to natural Amazonian clouds. First, a pyrocloud will be difficult to precipitate due to the strong inhibition of the collision-coalescence process that makes droplets grow to a size where they could precipitate. As the cloud droplet concentration is very high, the effective cloud droplet diameter (D_{eff}) is significantly reduced, and the smaller the D_{eff}, the less efficient is the evolution of the droplet size distribution at higher altitudes into the cloud. On the other hand, clouds in oceanic areas ("Blue Ocean") and in the green ocean presents an effective droplet-growing process along the vertical coordinate. This is not the case of pyroclouds, as is shown in Figure 8 [*Andreae et al.*, 2004]. It is possible to observe that for blue and green ocean clouds, there is a pronounced broadening in the droplet size distribution as the cloud becomes higher, a clear consequence of the growth of droplets due to the collision and coalescence process. This growth cannot be observed for the pyrocloud measurements. Instead, the droplet distribution of the pyrocloud does not grow significantly after 2800 m height, a consequence of the saturation effect of too many particles and droplets. Figure 8c shows an intermediate case: A cloud formed under smoky

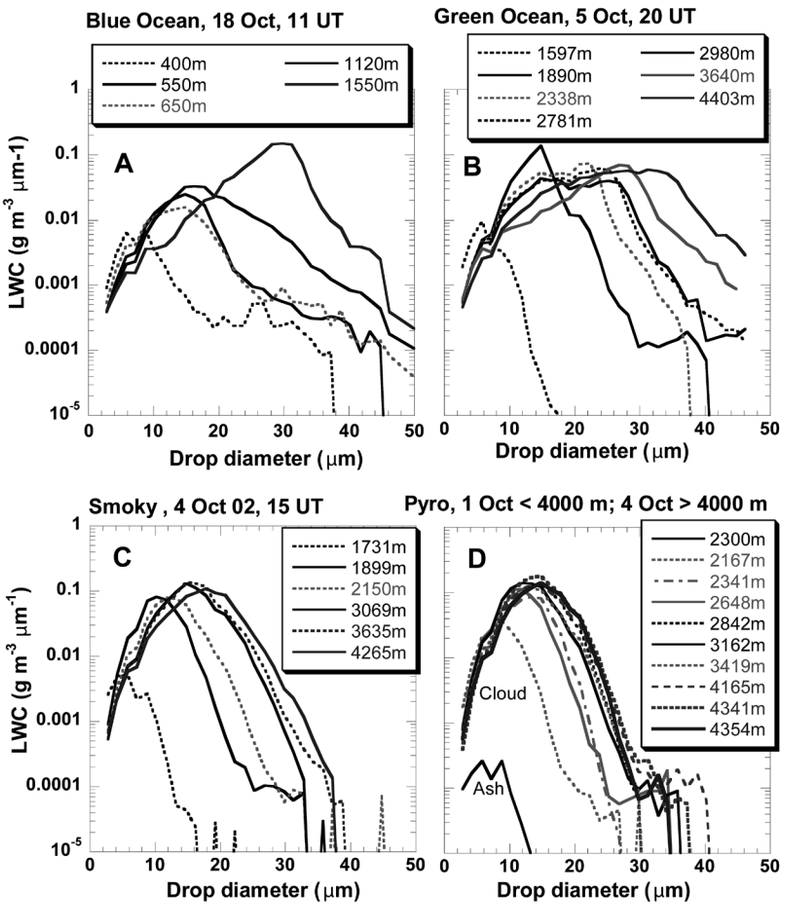

Figure 8. The evolution of cloud drop diameter distribution (DSD) with height in growing convective clouds, in the four aerosol regimes of (a) blue ocean, off the northeast Brazilian coast (4°S 38°W); (b) green ocean in the clean air at the western tip of the Amazon (6°S 73°W); (c) smoky clouds in Rondônia (10°S 62°W); and (d) pyroclouds. The lowest DSD in each plot represents conditions at cloud base, except in Figure 8d, where a size distribution for large ash particles outside of the cloud is also shown. Note the narrowing of CDSD and the slowing of its rate of broadening with height for the progressively more aerosol-rich regimes from Figures 8a to 8d [*Andreae et al.*, 2004].

conditions. Despite the fact that it is possible to observe an evolution and broadening of droplet spectra, it is slower than in the blue and green ocean conditions. The consequence is that warm rain, if it really occurs, will be at higher altitudes than in the natural case.

Another important effect of biomass smoke in cloud properties is the change in atmospheric thermodynamic conditions [*Koren et al.*, 2008]. The increase of black carbon loaded particles in the mid-atmosphere increases the stability of the lower troposphere. The presence of these absorbing particles results in a strong cooling at the surface, with a heating effect at altitudes around 3–4 km. This has been observed to suppress cloud formation during the biomass-burning season in the Amazon basin by remote sensing observations [*Koren et al.*, 2004]. Modeling calculations show that the suppression is possible under certain conditions [*Jiang et al.*, 2006]. This effect of the inhibition of low cloud formation was well documented by *Koren et al.* [2004, 2008].

Figure 9 shows that cloud cover for low clouds is strongly dependent on the atmospheric aerosol loading, expressed as AOT at 500 nm. For "natural" aerosol conditions (AOT <0.1), the average cloud cover in this study was about 40%. As the amount of aerosol increases, the cloud cover is strongly reduced. For AOT larger than 1.2, virtually all low clouds are inhibited [*Koren et al.*, 2004]. This value of AOT is frequently reached in the dry season, as can be seen in Figure 6. The mechanisms for this cloud suppression could be the high BC content of biomass-burning aerosol coupled with high solar radiation, in addition to the smaller droplet size. The small cloud droplets tend to evaporate if the core has high absorption properties [*McFiggans et al.*, 2006].

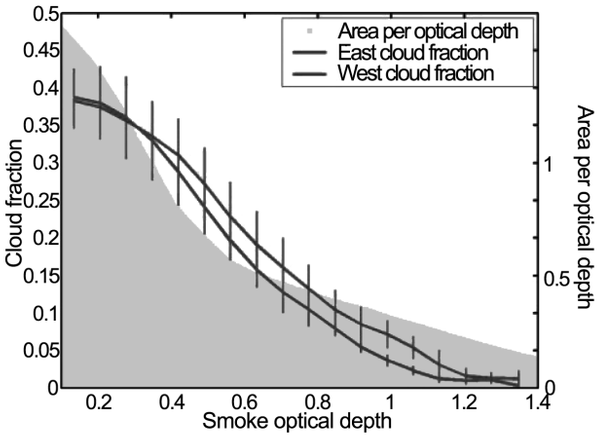

Figure 9. Cloud cover in Amazonia as a function of atmospheric aerosol loading, expressed as AOT at 500 nm.

7. DRY AND WET DEPOSITION OF TRACE ELEMENTS AND IMPACTS ON NUTRIENT CYCLING

The atmosphere plays an important role in nutrient cycling through wet and dry deposition of trace elements that are critically important to the forest ecosystem. Rain is an efficient scavenging process for atmospheric pollutants, and nutrients present in the atmosphere can be removed both in the gas and aerosol phases. Dry and wet deposition provides a key pathway for essential nutrients to reach terrestrial and aquatic ecosystems [*Galloway et al.*, 1984]. Wet deposition could have several harmful effects on various ecosystems with possible acidification due to high nitrate and sulfate values and therefore can have important impacts in the biogeochemical cycles. Systematic measurements of deposition provide a simple way to evaluate the influence of human activities on the atmospheric composition and to improve the knowledge of physicochemical processes related to the atmospheric transport and deposition of nutrients and pollutants and their impacts on the ecosystems [*Galloway et al.*, 1984; *Lara et al.*, 2001]. Various factors can affect the chemical composition of precipitation, including local emissions, regional-scale pollutant and nutrient transport processes, sea level and meteorological conditions. In addition, the rainwater chemical composition is also directly controlled by in-cloud and below-cloud scavenging of the atmospheric aerosols and trace gases derived from natural or anthropogenic sources [*Seinfeld and Pandis*, 2006]. The aqueous cloud droplet environment is also adequate for the absorption of soluble trace gases, working as a catalytic factor to many chemical reactions possible only in aqueous media [*Hegg et al.*, 1984], and biotransformation by microorganisms. On the other hand, during the below-cloud processes, the falling raindrops scavenge the airborne aerosols present in the atmosphere [*Pruppacher and Klett*, 1998]. This mechanism of aerosol removal is one of the major processes by which the atmosphere is cleansed [*Wallace and Hobbs*, 2006]. Dry deposition is the transport of gaseous and particulate species from the atmosphere onto surfaces in the absence of precipitation. The dry deposition flux is commonly described as a function of the "deposition velocity." Obviously, the larger and/or heavier the particle, the greater is its deposition velocity [*Seinfeld and Pandis*, 2006], and consequently, the coarse-mode particles ($d > 2.5$ μm) will be more subject to gravitational settling than smaller ones. In Amazonia, very different patterns of deposition occur in the dry and wet seasons. For both dry and wet seasons, the atmospheric cycling of phosphorus is critically important for the maintenance of the Amazonian ecosystem [*Mahowald et al.*, 2005].

Previous studies on wet deposition conducted in pristine areas in the Amazon basin [*Pauliquevis et al.*, 2007; *Williams et al.*, 1997; *Andreae et al.*, 1990; *Stallard and Edmond*, 1981] have reported low anthropogenic influence in the rainwater composition. A significant fraction of aerosol emitted by biomass burning is soluble as showed by *Yamasoe et al.* [2000]. As should be expected in pristine areas, rainwater composition is characterized by lower concentrations of the major compounds than those values found in areas subjected to biomass-burning emissions and anthropogenic activities [*Lara et al.*, 2001; *Trebs et al.*, 2006]. The natural acidity (pH ranging from 4.9 to 5.2) is commonly linked to the organic acids, mainly acetic and formic acids. In contrast, in areas with anthropogenic activities and land use changes, especially during the dry season, this scenario is completely different, with important changes in concentrations of sulfate and nitrate, in addition to the organic acids [*Artaxo et al.*, 2003].

There are significant differences in the concentrations of most rainwater compounds between dry and wet seasons (Figure 10), including H^+, K^+, Mg^{2+}, Ca^+, Cl^-, NO_3^-, and SO_4^{2-}. The significant increase of the average concentrations of NO_3^- could be attributed only to an increase in natural biogenic emissions or reduced deposition due to reduced precipitation. Since the compounds originating from biomass-burning emissions have shown no significant differences between the two seasons possibly in this site, there are no anthropogenic interferences, and it could be assumed to be a pristine area. The dominant compound in the rainwater composition is H^+ during both dry and wet seasons. Despite this during the whole year, both dry and wet seasons, the acidity in this area is always associated with the organic compounds. The organic compounds are dominated by acetate that is up to 10 times higher than formate and oxalate, which was expected since biogenic emissions are one of the main sources of acetate in pristine forest areas.

On the other hand, the rainwater composition changes completely when there are influences of a heavy polluted atmosphere due to the biomass burning. Most of the compounds emitted by biomass burning and soil emissions such as SO_4^{2-}, NH_4^+, K^+, Cl^-, Ca^{2+}, and others become significantly enriched in Rondônia during the dry season. The observed volume-weighted mean concentrations are significantly higher than the ones observed in clean areas such as BA.

Nitrogen is a fundamental nutrient for ecosystems. However, because of human activities, the input of nitrogen on terrestrial ecosystems has dramatically increased in the last 50 years. In the tropics, the increase in N deposition is related with the widespread use of biomass burning as a tool for land use change. When wood is burned, the biomass-associated N is volatilized, and a large fraction is emitted

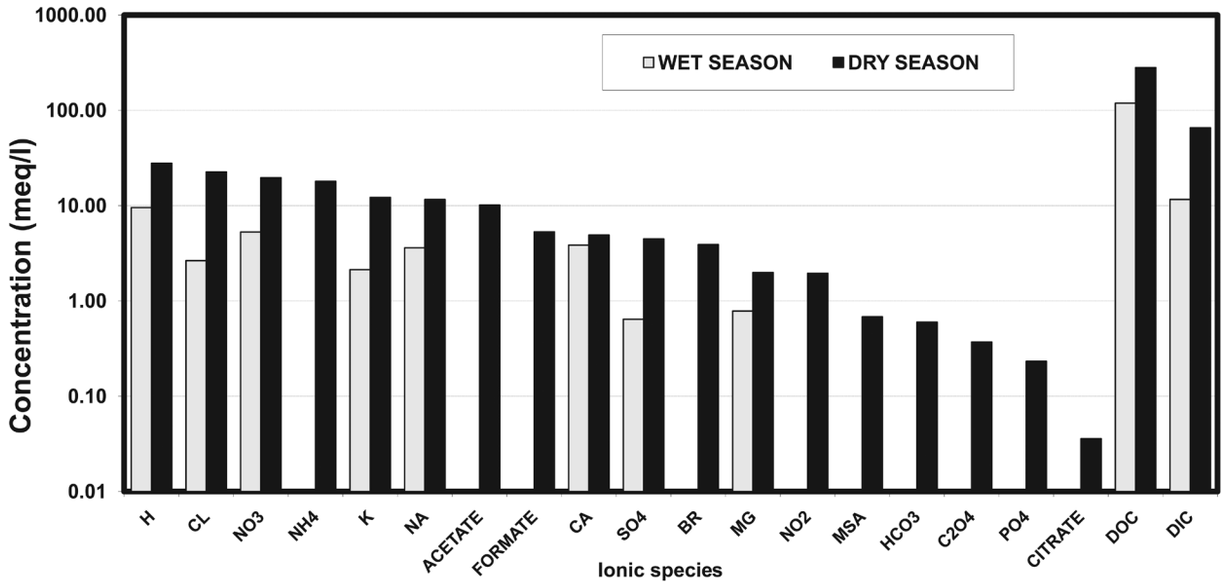

Figure 10. Rainwater composition in Rondônia, a region impacted by biomass burning, for the wet and dry seasons. Concentrations expressed in μeq L^{-1}, except dissolved organic carbon and dissolved inorganic carbon, both expressed in μM L^{-1}.

in the form of gaseous NH_3, which may result in considerable N losses from tropical ecosystems [*Kauffman et al.*, 1998]. As biomass burning is also a rich source of NO_x (= [NO] + [NO_2]), there is a concern on the role of these emissions on the N cycle [*Trebs et al.*, 2006]. An important implication is the alteration of the dominant form of nitrogen deposition from nitrate to ammonium [*Lara et al.*, 2001]. It is particularly critical because, once deposited, ammonium releases acidity since the nitrogen is either accumulated in organic nitrogen form or nitrified and leached as nitrate.

8. CONCLUSIONS

This review of LBA science in terms of atmospheric chemistry shows a lot of progress in understanding the role of atmospheric chemistry as a main driver of the ecosystem functioning in several key areas of Amazonia. The land use changes are altering the atmospheric emissions and deposition as well as the radiative balance and the hydrological cycle over large areas. The changes in land use also alter the surface albedo that is a critical parameter in the atmospheric radiative balance. The large aerosol concentrations reported in this chapter have important health effects on the Amazonian population, and this effect must be taken into account in designing policies to reduce deforestation and biomass-burning emissions.

During the wet season, Amazonia is one of the very few regions on our planet where we can still observe "natural" conditions in terms of aerosol particles. This helps the understanding of how a pristine atmosphere works before humans started to change atmospheric composition in a significant way. The study of natural biogenic particles and their role in cloud droplet nucleation and precipitation formation is a key area that just started to emerge. The coupling between the atmosphere, vegetation, and climate is very strong in Amazonia, and we have major gaps in understanding the complex relationship of biosphere-atmosphere interactions. We need more long-term environmental measurements at a spatial scale compatible with the area of Amazonian forest. Very few measurements of atmospheric properties have been taken in the western part of Amazonia (between Tefé and São Gabriel da Cachoeira), and the high precipitation rate and carbon uptake in this area indicated processes that are different from eastern Amazonia.

Opportunities for progress in identifying the sources of primary particles in the Amazon basin and quantifying their emissions include (1) characterization and quantification of different types of primary biological, biomass burning, mineral dust, and marine aerosol particles, including long-term trends, seasonal cycles, diurnal variability; (2) discrimination and quantification of local, regional, and long-range sources of all particle types; (3) 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; (4) 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 (Martin et al., submitted manuscript, 2008). We need studies on the hygroscopic behavior of Amazonian aerosol particles, since this is critically important for CCN activation as well as for radiative forcing. We also need to understand new particle formation mechanisms in Amazonia and to study the radiation balance and the relationship between aerosol particles and clouds at the large scale using advanced remote sensing techniques.

The main message of this chapter for public policies is that deforestation is not only changing carbon pools, but has profound impacts on the functioning of the Amazonian ecosystem itself. The changes in the hydrological cycle through changes in CCN population have implications for the whole South American continent as well as teleconnections with regions far from Amazonia. The changes in cloud structure have important implications on water vapor transport over large areas. The Amazonian region plays a critical role in global climate change, as well as the possible effects that the changing climate could have over tropical forest ecosystems [*IPCC*, 2007]. These points make a strong argument for reducing deforestation rates as quickly as possible and to look at Amazonia as a key region in regulating planetary climate from the atmospheric chemistry point of view.

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