

## Physical and chemical properties of aerosols in the wet and dry seasons in Rondônia, Amazonia

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[1] As part of the Large-Scale Biosphere-Atmosphere Experiment in Amazonia (LBA), a large study of aerosol and trace gas properties was conducted in Amazonia during the dry and wet seasons and for pasture and primary forest sites. Aerosol mass and elemental composition were measured on filters for both fine ( $d_p < 2.0 \mu\text{m}$ ) and coarse ( $2.0 < d_p < 10 \mu\text{m}$ ) modes using polycarbonate filters mounted in stacked filter units (SFU).

Measurements with real-time aerosol and trace gas monitors were made, parallel to the collection of filter samples to obtain high time resolution (1–30 min). Black carbon equivalent (BCE) was determined with a multiwavelength Aethalometer as well as by reflectance in the fine-mode SFU samples. Elemental carbon as well as total and organic carbon was measured using an R&P 5400 real-time carbon monitor.  $\text{PM}_{10}$  aerosol mass concentrations were determined with a Tapered Element Oscillating Monitor (TEOM) monitor every 5 min.  $\text{CO}$ ,  $\text{O}_3$ ,  $\text{NO}$ , and  $\text{NO}_2$  were analyzed with Thermo Environment monitors with trace level capabilities for  $\text{NO}_x$ . A TSI 3010 particle counter measured the total particle number concentration ( $d_p > 10 \text{ nm}$ ) every minute. A 55-m tower in a primary forest in Rondônia and a facility built on a pasture site were used to operate the instrumentation for 4 months in the wet season and 3 months in the dry season of 1999. Particle-Induced X-Ray Emission (PIXE) was used to measure the concentration of 22 trace elements for fine- and coarse-mode aerosol. During the wet season, very clean atmospheric conditions were observed at both sites. Particle concentration averaged  $900 \text{ cm}^{-3}$ , black carbon averaged  $250 \text{ ng m}^{-3}$ , and mean fine-mode mass concentration was  $2.9 \mu\text{g m}^{-3}$ . In sharp contrast, very high concentrations were observed in the dry season for all the parameters at both sampling sites. Aerosol mass concentrations up to  $250 \mu\text{g m}^{-3}$  and particle number concentrations of more than  $40,000 \text{ cm}^{-3}$  were observed, while the  $\text{CO}$  peaked at 8 ppm and  $\text{NO}_2$  reached 16 ppb. High correlation was observed between BCE,  $\text{NO}_2$ ,  $\text{CO}$ , aerosol particle number, mass, aerosol optical thickness, and other properties linked to biomass-burning emissions. Relatively large concentrations of phosphorus in the coarse-mode particles were observed, especially at nighttime. The emission of biogenic particulate P could have an effect in the nutrient cycling of this essential and key nutrient, which is present almost exclusively in the form of aerosol particles. Phosphorus is exchanged in the aerosol phase and mostly for coarse-mode particles and during nighttime.

*INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0315 Atmospheric Composition and Structure: Biosphere/atmosphere interactions; 1610 Global Change: Atmosphere (0315, 0325); *KEYWORDS:* Amazonia, global change, aerosol, biomass burning, trace gases

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## 1. Introduction

[2] The Large-Scale Biosphere-Atmosphere Experiment in Amazonia (LBA) is a large scientific effort aiming to understand the functioning of the Amazonian ecosystem in an integrated way. LBA is investigating the close linkages between trace gas exchange, aerosols, clouds, the hydrological system, and ultimately the sustainability of the Amazon Basin ecosystem [Artaxo *et al.*, 2001]. Atmospheric processes are key in several ecosystem processes and in the regulation of the Amazon basin climatic system. In particular, aerosol particles are essential participants in ecosystem biogeochemistry and nutrient cycling, as well as in cloud formation mechanisms. Biogenic trace gases emitted by the vegetation are important in the regulation of the oxidizing power of the tropical and global atmosphere [Andreae and Crutzen, 1997]. The Amazon region is a key tropical ecosystem that is suffering profound changes in land use [Artaxo, 2001; Artaxo *et al.*, 2001]. Although only about 11–14% of the original forest area is now deforested, the vast area of the forest (5.5 million km<sup>2</sup>) and the rapid land use change are responsible for the regional climatic effects that only recently are being noticed in Rondônia State and in the southern region of Para State [Silva Dias *et al.*, 2002].

[3] Over the last decade, a growing amount of attention has been focused on the climatic effects of atmospheric aerosols [Hobbs *et al.*, 1997; Penner *et al.*, 2001]. The aerosol effect on climate is very complex, and depends on several aerosol microphysical properties, such as composition, size distribution, morphology, hygroscopic behavior and other properties [Jacobson, 2001]. Some aerosol types absorb radiation, and thereby warm the atmosphere, and scatter light back to space, which cools the surface. Aerosols can also reduce precipitation from clouds, which enhance their lifetime, contributing to the cooling effect. Or, if they absorb radiation, and warm an atmospheric layer, they may reduce cloud formation, which would warm the Earth [Rosenfeld, 1999]. The organic and inorganic aerosol components are very active in terms of heterogeneous interactions with the gaseous component and through cloud processing. The aerosol indirect climatic effects resulting from increased cloud condensation nuclei (CCN) concentrations are expected to be very important in tropical regions, particularly because of the very high biomass-burning emissions [Roberts *et al.*, 2001]. In view of the very low natural CCN concentration in Amazonia [Roberts *et al.*, 2001; Zhou *et al.*, 2002], it does not take very high amounts of additional aerosol particles to cause a significant increase in the cloud droplet number concentration and a corresponding decrease in the cloud droplet size [Rosenfeld, 1999].

[4] A large part of the natural aerosol component over tropical forests is thought to come from the oxidation of biogenic volatile organic carbon compounds (VOCs), particularly terpenes, to low-volatility compounds that condense into aerosol particles [Kesselmeier *et al.*, 2000]. Recent estimates suggest that over 1000 Tg C is emitted annually in the form of biogenic VOCs, of which an estimated 30% are potential aerosol precursor substances [Andreae and Crutzen, 1997; Kesselmeier *et al.*, 2000]. Aerosol particles also are important in gas phase chemistry.

They change the visible and UV radiation flux [Schafer *et al.*, 2002] and spectral radiance [Yamasoe *et al.*, 1998], directly affecting photochemistry. Submicrometer particles are very efficient in scattering UV as well as visible radiation. They alter the actinic flux and thereby affect the production of the OH radical that is key in tropical photochemistry [Crutzen and Andreae, 1990].

[5] Moist tropical forests account for a substantial component of terrestrial plant productivity and several lines of evidence suggest that they may be sequestering significant amounts of anthropogenic CO<sub>2</sub> at the present time. But there is also some evidence that the productivity of many of these forests is limited by their low phosphorus availability. This has led to some suggestions that moist tropical forests may not be able to increase their growth in response to increases in atmospheric carbon dioxide concentrations to the same degree as would be the case if rates of P availability were higher. Atmospheric inputs of mineral elements into tropical rain forests may constitute an important input of plant nutrients, especially for soils of low inherent fertility [Artaxo *et al.*, 1997, 2001]. Such atmospheric inputs are traditionally divided into wet deposition (input of mineral elements dissolved in rainwater) and dry deposition (inputs from deposited aerosol particles or as dust). When attempting to deduce nutrient external inputs into a forest, a further complication may be that tropical forests themselves produce aerosols. We know virtually nothing about the source of airborne phosphorus. This is especially the case as continental atmospheric P deposition cannot be supported by long-term transport of P from tropical oceans as these typically have very low P concentrations in their surface waters. One possibility is the intrusion of dust from desert regions. The importance of dust as a nutrient source is likely for West African rain forests, but the significance of occasional long-term transport of Saharan dust into Amazonia still has to be addressed [Swap *et al.*, 1996; Formenti *et al.*, 2001].

[6] The objective of this paper is to describe extensive measurements of physical and chemical properties of atmospheric aerosols in Rondônia, collected during two intensive measurements programs during dry and wet season as part of the LBA experiment. This is the first study of aerosol and trace gas characterization in Amazonia to combine high time resolution measurements with more traditional filter based aerosol characterization. The use of parallel measurements for dry and wet season in pasture and forest sites is also unique in this study. Rondônia was chosen because it contains areas with pristine primary forests, as well as extensive pasture areas with heavy land use change. Our strategy was to conduct similar aerosol and trace gas measurements in nearby pasture and primary forest areas to assess differences in aerosol composition and physical properties. Filter sampling and real-time aerosol and trace gases monitors allowed the study of rapid process in the Amazonian atmosphere. Several other cooperative studies complement the results discussed in this paper, such as the work by Graham *et al.* [2002], Mayol-Bracero *et al.* (Chemical characterization of fine carbonaceous aerosols over the Amazon Basin during the dry season: Black carbon, organic carbon and water-soluble organic compounds, submitted to *Journal of Geophysical Research*, 2001, hereinafter referred to as Mayol-Bracero *et al.*,

submitted manuscript, 2001), *Mayol-Bracero et al.* [2002], *Schafer et al.* [2002], *Zhou et al.* [2002], *Andreae et al.* [2001, 2002], *Silva Dias et al.* [2002], and others. The first two papers discuss the organic aerosol component. Schafer et al. works on the aerosol radiative properties with data from the LBA Sun photometer network. Zhou et al. discuss the hygroscopic properties of Amazon basin aerosols and also detailed size distribution measurements. Andreae et al. discuss the overall LBA/EUSTACH results, and Silva Dias et al. discuss key meteorological aspects of the sampling campaigns in Rondônia.

## 2. Aerosol Sampling and Analysis

[7] The objective of this paper involves the characterization of aerosols over two types of land use in Amazonia: pasture and primary rain forest. The sampling site in the state of Rondônia that was used as representative of a pasture environment is called “Fazenda Nossa Senhora Aparecida,” located at 10°45′44″S, 62°21′27″W, 315 m asl. For the primary rain forest site, an ecological reserve called “Reserva Biológica Jarú,” located at 10°04′55″S, 61°55′48″W, 110 m asl was used. Both sites are located within Rondônia, Brazil, a region where extensive deforestation has occurred in recent years (for further details on the sampling campaigns and sites, see *Andreae et al.* [2002] and *Silva Dias et al.* [2002]).

[8] Two types of aerosol measurements were performed for each site: Filters collecting aerosol samples integrated over 24 to 48 hours and real-time aerosol monitors with high time resolution. Bulk aerosol samples were collected using stacked filter units (SFU) [*Hopke et al.*, 1997] fitted with a PM<sub>10</sub> inlet. The SFU collects particles on 47 mm diameter polycarbonate membrane filters. An 8 μm pore size filter collects coarse particles (2.0 < dp < 10 μm) while a 0.4 μm pore size filter collects fine particles (dp < 2.0 μm). The flow rate was typically 16 liters per minute, and sampling time varied from 24 to 48 hour. The aerosol size distribution was measured with the use of the MOUDI (MultiOrifice Uniform Deposit Impactor) cascade impactor. The MOUDI separates aerosol particles in 8 stages with d<sub>50</sub> size cuts at 18, 3.2, 1.8, 1.0, 0.56, 0.33, 0.175, and 0.093 μm equivalent aerodynamic diameter (EAD); a Teflon backup filter collects the particles smaller than 0.093 μm. The MOUDI used in this experiment rotates the sample continuously to obtain a homogeneous aerosol deposit. Trace gas measurements were performed with Thermo Environment continuous trace gas monitors. Ozone, CO, NO and NO<sub>2</sub> were measured in parallel with the aerosol components. The NO and NO<sub>2</sub> instrument is a trace level monitor, required to measure the very low values in the Amazon atmosphere during the wet season.

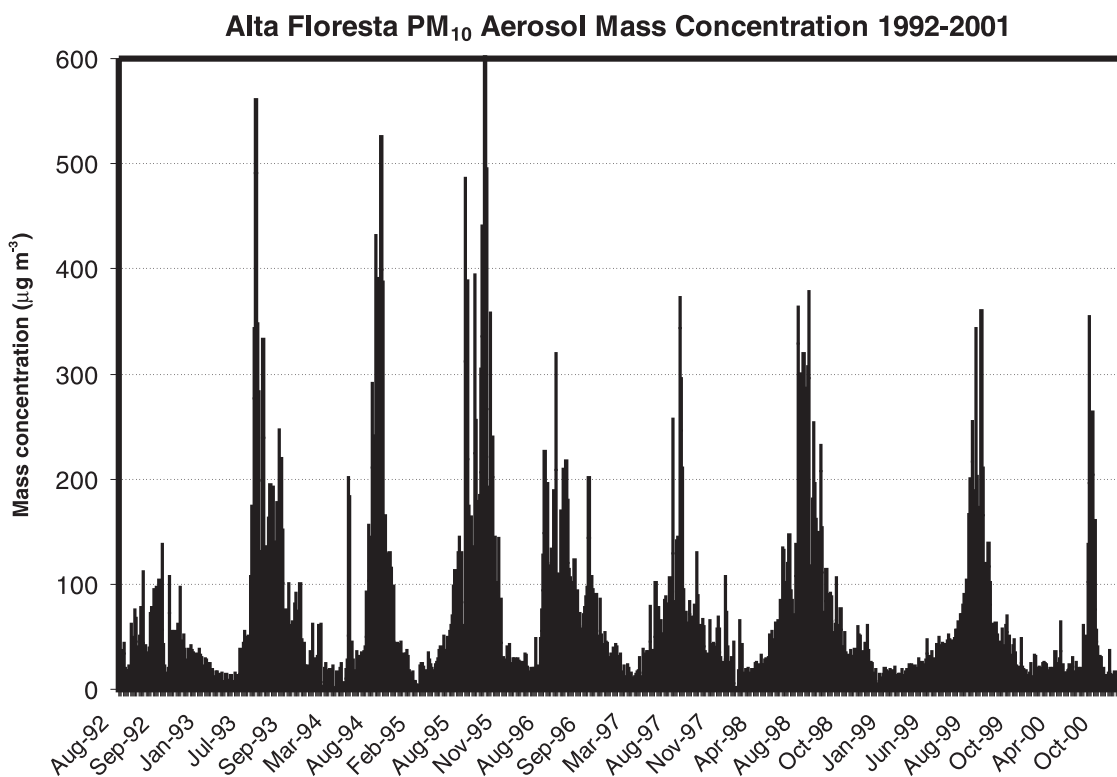
[9] Elemental concentrations for the SFU filters and MOUDI samples were obtained by Particle-Induced X-Ray Emission (PIXE) analysis [*Artaxo and Orsini*, 1987; *Johansson et al.*, 1995]. It was possible to determine the concentrations of up to 21 elements (Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Rb, Sr, Zr, and Pb). A dedicated 5SDH tandem Pelletron accelerator facility at the University of São Paulo LAMFI (Laboratório de Análise de Materiais por Feixes Iônicos) was used for the PIXE analyses. Detection limits were typically 5 ng m<sup>-3</sup> for

elements in the range 13 < Z < 22 and 0.4 ng m<sup>-3</sup> for elements with Z > 22. Precision of elemental concentration measurements is typically better than 7%, reaching up to 20% for elements with concentrations near the detection limit.

[10] Mass concentrations were obtained through gravimetric analysis. Both fine and coarse filters from the SFU were weighed before and after sampling in a Mettler M3 electronic microbalance with 1 μg sensitivity. Before weighing, filters were kept for 24 h at 50% relative humidity and 20°C. Electrostatic charges were controlled by means of <sup>210</sup>Po radioactive sources. Detection limit for the aerosol mass concentration is 0.3 μg m<sup>-3</sup>. Precision is estimated at about 10%. Black carbon concentrations on the fine fraction of the SFU filters were obtained by a light reflectance technique [*Martins et al.*, 1998a, 1998b]. Gravimetric and black carbon analyses were also performed on the MOUDI samples.

[11] A real-time aerosol mass monitor, the Tapered Element Oscillating Monitor (TEOM) series 1400a from Rupprecht & Patashnick Inc., was used to measure PM<sub>10</sub> aerosol concentration with a time resolution of 10 min. The TEOM was operated at 50°C heating in the inlet. It is well known that the TEOM sampler always yields smaller values for mass concentrations than gravimetric methods, because of the slight heating that the instrument must apply to the aerosol flow. Some of the volatile component can be lost during analysis in the TEOM monitor. The organic and elemental carbon concentrations were measured in the PM<sub>2.5</sub> fraction by a thermal technique using the real-time Ambient Carbon Particulate Monitor, series 5400 from Rupprecht & Patashnick, Inc. The organic carbon component was defined as the carbon content when a controlled combustion at 350°C was applied. The total carbon was obtained by combusting the aerosol at 700°C. Elemental carbon is derived as the difference between total and organic carbon. A high-time-resolution black carbon monitor, the Aethalometer, manufactured by Magee Scientific Co., measured the black carbon concentration. Because of the nature of the absorption measurement used by the Aethalometer, it is a measurement not specific for black carbon. Therefore, we will call the black carbon measured by the Aethalometer and by the reflectometer as “BCE,” short for black carbon equivalent. The Aethalometer was operated with 5 min time resolution, and an absorption cross section of 10 m<sup>2</sup> g<sup>-1</sup> was used to convert optical absorption to black carbon equivalent [*Martins et al.*, 1998a, 1998b]. The aerosol light scattering was measured using the 3-wavelength TSI Model 3563 nephelometer. The nephelometer and the Aethalometer were operated with a 2-μm aerosol inlet. The TSI CPC 3010 was operated with a time resolution of 1 min, and an inlet with a cut-off of 2 μm. All the sampling instruments were operating with Hastings precision mass flowmeters to measure the airflow rate with an accuracy of about 2%. The time resolution of the aerosol and trace gas instruments enabled the study of diurnal cycles and allowed the identification of the factors that control the diurnal variability, complementing the 24 or 48 hour bulk aerosol filter analyses.

[12] Local meteorological variables were obtained from a weather station at each sampling site. The large-scale circulation was characterized using the RAMS model configured to run in high resolution over the state of Rondônia.



**Figure 1.** Alta Floresta PM<sub>10</sub> aerosol concentration observed in a LBA atmospheric monitoring station operated continuously since 1992.

Air mass trajectories allowed the study of the large-scale circulation [Silva Dias *et al.*, 2002].

### 3. Results and Discussion

#### 3.1. Fine- and Coarse-Mode Aerosol Mass Concentrations

[13] The concentration of aerosol and trace gases in Amazonia undergoes a large increase during the dry season, changing from very low concentrations to levels exceeding values observed in polluted urban areas [Artaxo *et al.*, 1994, 1998]. Figure 1 show the time series of PM<sub>10</sub> aerosol concentration observed at a monitoring station operated in Alta Floresta, situated in the North of Mato Grosso State since 1992. Alta Floresta is on the border between primary forest and areas with heavy land use change, suffering heavily from biomass-burning emissions. During the wet season, typical aerosol mass concentration for PM<sub>10</sub> is about 10–15  $\mu\text{g m}^{-3}$ . During the dry season months (August–October), PM<sub>10</sub> concentrations reach very high values of about 400–600  $\mu\text{g m}^{-3}$ , even if the sampling site is not in the immediate vicinity of any large emissions [Artaxo *et al.*, 2000]. It is possible to observe in Figure 1 the yearly variability in the maximum concentrations during the dry season due to climatic and economic factors and also due to changes in precipitation patterns. A strong drought in Amazonia during most of 1995 caused this year to be the largest in fire counts, deforestation rate and atmospheric concentrations of biomass-burning-related species. The years 1996 to 1999 were characteristic of economic slowdown in Amazonia, caused by enforcement of new regulations on land use change. During the dry season, fine-mode

aerosol concentrations can reach very high values, in the vicinity of 200–300  $\mu\text{g m}^{-3}$ , reducing significantly atmospheric visibility (down to 100–300 m), and producing a potentially significant local aerosol radiative forcing.

[14] A large number of parameters were measured in this study. Table 1 shows a summary of the physical and chemical properties of the dry and wet season aerosol measured in Rondônia. It is easy to observe the large differences between the wet and dry season atmospheric composition, as well as the high values observed when Rondônia is at the peak of the biomass-burning impact. One key aerosol property related to climate is the total aerosol number concentration that is an indication of the cloud condensation nuclei number [Zhou *et al.*, 2002; Roberts *et al.*, 2001]. During the wet season, clean, almost maritime-like conditions prevail, with an average of 890  $\text{cm}^{-3}$ . This number increases by a factor of 10 on average during the dry season, reaching 40,000  $\text{cm}^{-3}$  at the peak of the dry season, an increase by a factor of 50 over natural wet season conditions. Most of this aerosol is organic in nature and the particles are effective as cloud condensation nuclei [Novakov and Corrigan, 1996].

[15] In Rondônia, large differences in aerosol mass concentration were observed for both pasture and forest sites, similar to the results from Alta Floresta. Figure 2 shows the wet season aerosol mass concentrations at the pasture site and Figure 3 shows the mass concentration for the dry season. The average aerosol concentrations in the wet season are very low. During nighttime, much higher concentrations of coarse-mode biogenic aerosol are observed at both forest and pasture sites. This is clearly visible in Figure 2, where sampling was done separating daytime and nighttime samples. The higher coarse-mode samples were collected

**Table 1.** Summary of Physical and Chemical Properties of Dry and Wet Season Aerosols in Rondônia<sup>a</sup>

Specie or property	Unit	Dry season average	Dry season std. dev.	Dry season peak	Wet season average	Wet season std. dev.
PM <sub>10</sub> TEOM	μg m <sup>-3</sup>	55.2	52.2	250	10.1	5.5
PM <sub>10</sub> SFU	μg m <sup>-3</sup>	83.0	63.7	300	8.6	3.9
Fine mass (FPM)	μg m <sup>-3</sup>	66.9	55.1	250	2.9	1.71
Coarse mass (CPM)	μg m <sup>-3</sup>	17.8	11.7	50	5.7	3.1
Particle number (CPC)	#/cc	8045	5668	40000	890	920
Organic carbon (OC)	μg m <sup>-3</sup>	10.1	9.4	55	2.04	0.89
Elemental carbon (EC)	μg m <sup>-3</sup>	3.8	4.2	25	0.47	0.37
Total carbon (TC)	μg m <sup>-3</sup>	13.9	13.2	80	2.31	0.95
Black carbon (BCE)	μg m <sup>-3</sup>	3.60	2.99	18.0	0.19	0.22
Sulfur fine mode	ng m <sup>-3</sup>	1104	870	2500	83.9	78.8
Potassium fine mode	ng m <sup>-3</sup>	1109	865	3000	33.5	28.5
NO <sub>2</sub>	ppb	2.73	1.59	16.0	0.67	0.61
CO	ppm	0.89	0.74	8.0	0.15	0.18
AOT 500 nm <sup>b</sup>	–	0.91	0.56	3.3	0.13	0.11
Angstrom Exponent	–	1.70	0.49	2.5	0.72	0.53

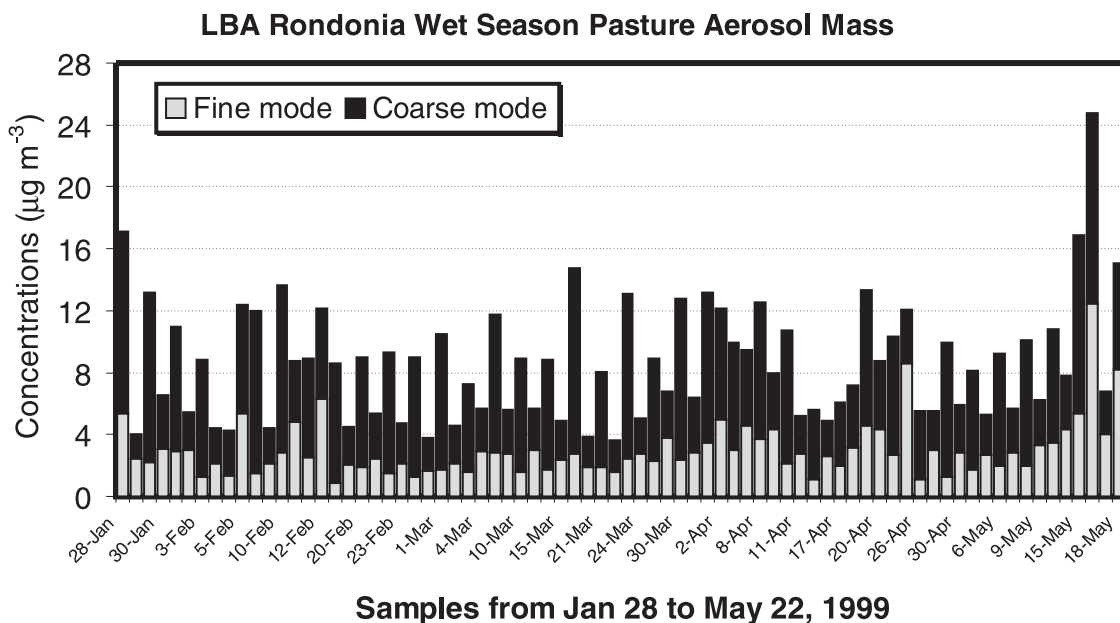
<sup>a</sup>The averages were calculated with the highest time resolution provided by the measurements. Typically the averages were calculated for 5–30 min integration time.

<sup>b</sup>AOT is the aerosol optical thickness at 500 nm measured with the AERONET Sun photometer network. The Angstrom Exponent is derived from the Sun photometer aerosol optical thickness and is an indication of the aerosol size distribution.

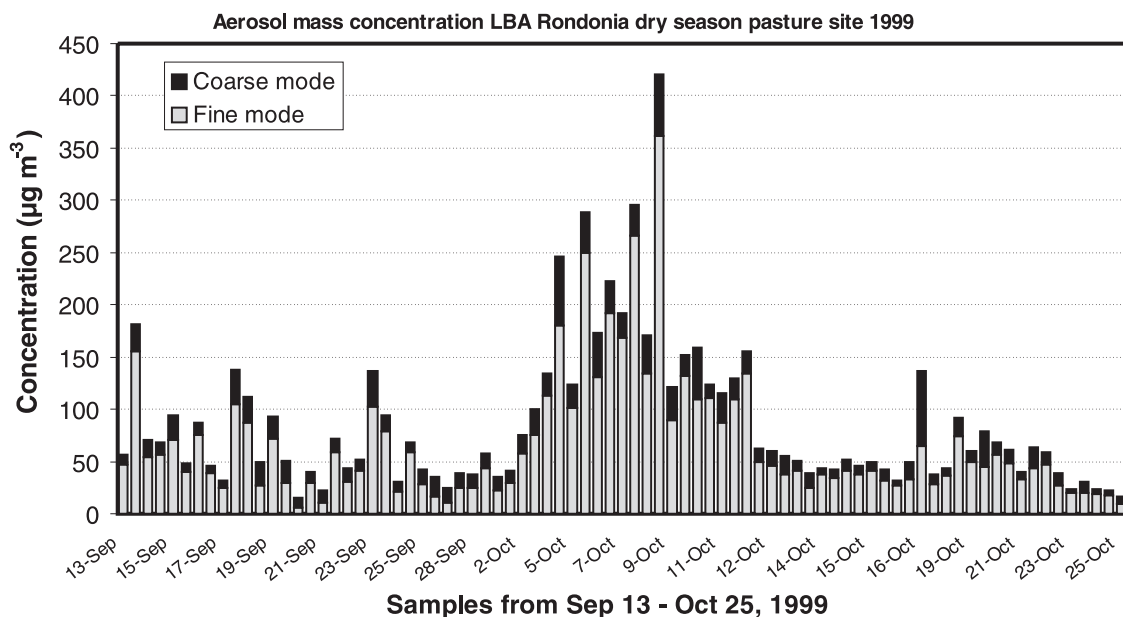
during nighttime. In the dry season, aerosol concentrations of up to 400 μg m<sup>-3</sup> can be observed (Figure 3), with fine-mode concentrations up to 250–350 μg m<sup>-3</sup>. It is important to emphasize that these high concentrations during the dry season occur not only in Alta Floresta or Rondônia, but that the haze plume extends over a large area, of up to 5–8 million km<sup>2</sup> [Artaxo *et al.*, 1998]. This very high atmospheric aerosol loading has extensive influence on cloud condensation nuclei concentrations [Roberts *et al.*, 2001], with the potential to suppress precipitation in tropical areas [Rosenfeld, 1999].

[16] Figure 4 shows the aerosol mass concentrations for the Rondônia Forest (Rebio Jaru Tower) site during the wet season. At the tower, aerosol sampling was performed

at three levels to allow the analysis of the aerosol deposition and gradients in the forest canopy. Figure 5 shows the aerosol mass concentrations for the Rondônia Forest site during the dry season, also showed separately for the three levels of the tower. Comparing the measurements at the forest site with the pasture site during the dry season, the forest shows much lower concentrations, indicating that the forest site is indeed relatively far from biomass-burning emissions. Even so, PM<sub>10</sub> concentrations peaked at a relatively high 80–100 μg m<sup>-3</sup> levels. FPM at the two top levels in the forest tower averaged 33 μg m<sup>-3</sup> while at the ground it averaged 28 μg m<sup>-3</sup>, indicating the possibility of biomass-burning aerosol deposition in the forest canopy. Due to the high concentrations and high



**Figure 2.** Aerosol mass concentration for the Rondônia pasture site during the wet season. Average concentrations were FPM = 3.0 μg m<sup>-3</sup>, CPM = 5.7 μg m<sup>-3</sup>, PM<sub>10</sub> = 8.7 μg m<sup>-3</sup>, and black carbon equivalent (BCE) = 0.47 μg m<sup>-3</sup>.



**Figure 3.** Aerosol mass concentration for the Rondônia pasture site during the dry season. Average concentrations were FPM =  $67.3 \mu\text{g m}^{-3}$ , CPM =  $18.4 \mu\text{g m}^{-3}$ ,  $\text{PM}_{10}$  =  $85.7 \mu\text{g m}^{-3}$ , and BCE =  $7.6 \mu\text{g m}^{-3}$ .

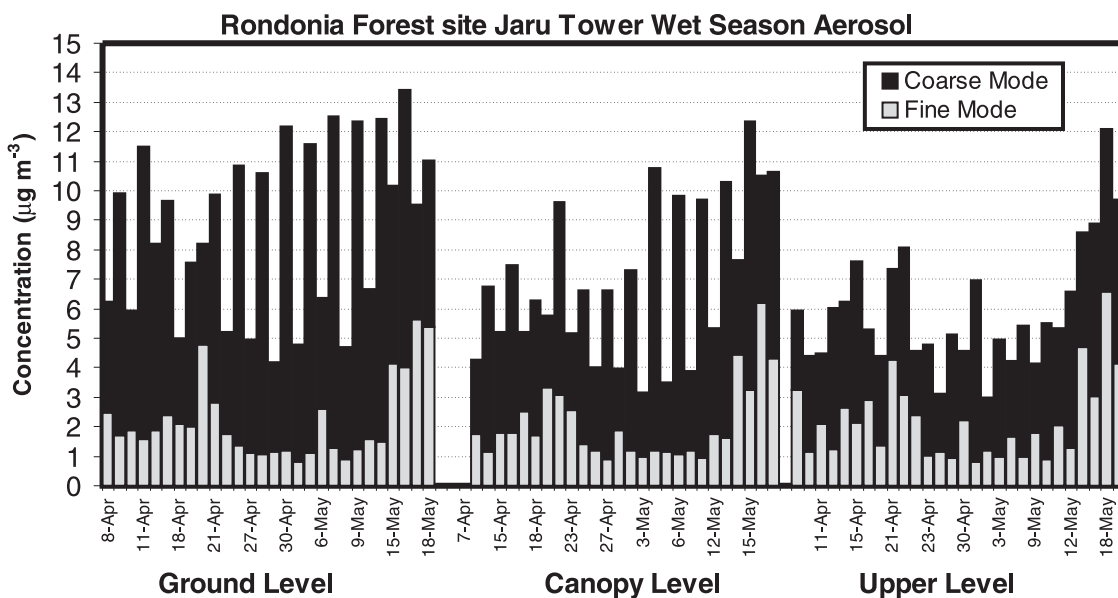
precipitation rate, the dry and wet depositions in Amazonia are very important to the nutrient cycling. Other studies have shown the long-range transport of biomass-burning aerosol in Amazonia [Andreae et al., 2001, 2002; Longo et al., 1999], and from Africa to South America [Swap et al., 1996].

**3.2. Real-Time Aerosol Monitors**

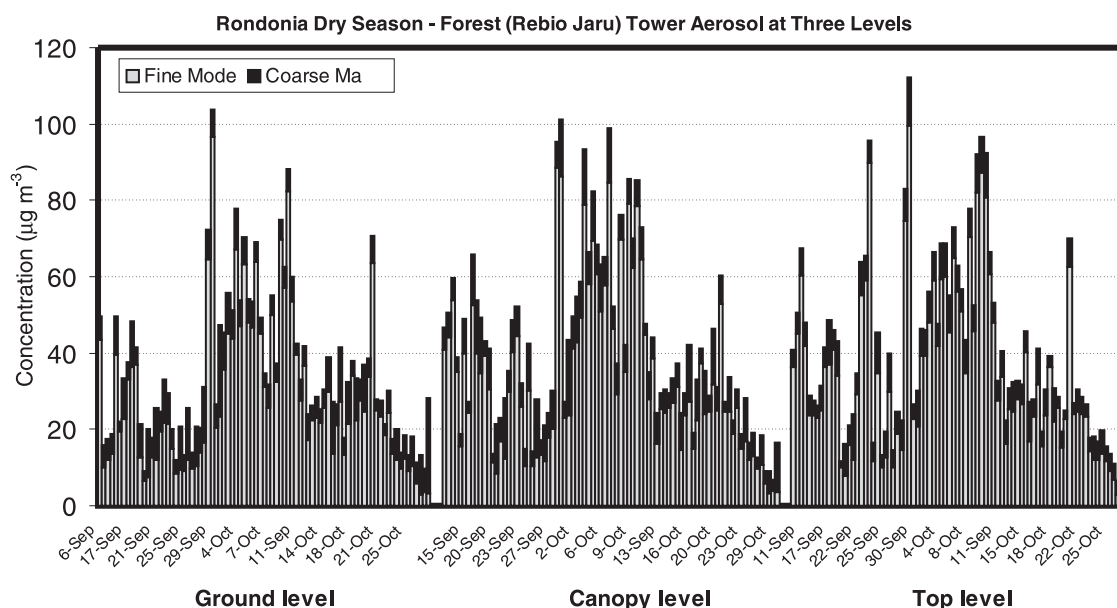
[17] The detailed diurnal pattern and short-term temporal variability of the aerosol concentrations can not be observed

using 12 or 24 hours filter sampling. Real-time monitors with a time resolution of 5–30 min are required. TEOM  $\text{PM}_{10}$  aerosol monitor, Aethalometer, real-time carbon monitor, CPC and nephelometer were used to measure aerosol physical properties in the dry and wet season for both sampling sites, pasture and forest.

[18] Figure 6 presents the measured  $\text{PM}_{10}$  aerosol mass concentrations for the dry season pasture site by two different techniques: gravimetric by the SFU and the TEOM monitor. The TEOM heats the sample air stream to  $50^\circ\text{C}$ , in



**Figure 4.** Aerosol mass concentrations for the Rondônia Forest (Rebio Jaru Tower) site during the wet season. At the tower site, aerosol sampling was performed at three levels of the tower. Overall average concentrations were FPM =  $2.2 \mu\text{g m}^{-3}$ , CPM =  $5.1 \mu\text{g m}^{-3}$ ,  $\text{PM}_{10}$  =  $7.3 \mu\text{g m}^{-3}$ , and BCE =  $0.28 \mu\text{g m}^{-3}$ .

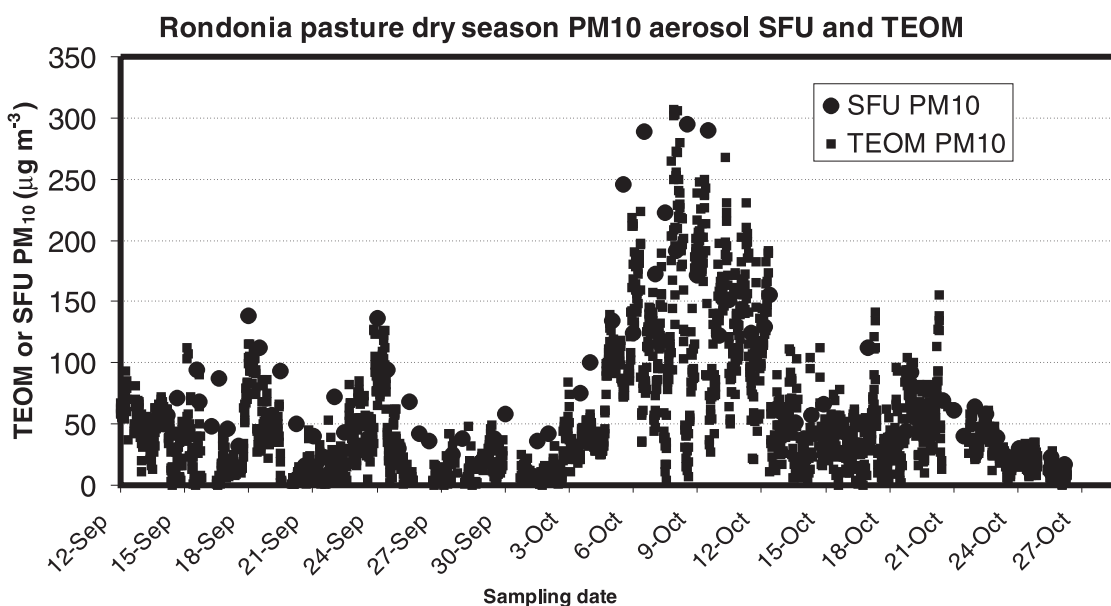


**Figure 5.** Aerosol mass concentrations for the Rondônia Forest (Rebio Jaru Tower) site during the dry season. Average concentrations were  $\text{FPM} = 31.7 \mu\text{g m}^{-3}$ ,  $\text{CPM} = 7.2 \mu\text{g m}^{-3}$ ,  $\text{PM}_{10} = 38.9 \mu\text{g m}^{-3}$ , and  $\text{BCE} = 1.78 \mu\text{g m}^{-3}$ .

order to avoid condensation of water in the measuring element. This slight heating produces losses of some volatile compounds. In the dry season experiment, SFU  $\text{PM}_{10}$  averaged  $83 \mu\text{g m}^{-3}$  while the TEOM averaged  $54 \mu\text{g m}^{-3}$ , corresponding to a loss of about 35% of the volatile aerosol mass. This number is similar to the values observed in urban areas, and can be interpreted as a measure of the aerosol volatility. The concentrations reach their highest values at nighttime and early morning, due to the very low boundary layer height at night (sometimes below 150 m). At 3:00 P.M. in general the diurnal minimum in concentration is observed, because the mixing layer height is generally

1500–1800 m at midafternoon (G. Fisch et al., Convective boundary layer over pasture and forest sites in Amazonia, submitted to *Journal of Geophysical Research*, 2001). Most of the fires occur at the end of the afternoon in Amazonia.

[19] Since biomass burning is the dominant aerosol and trace gas source during the dry season, it is expected that trace gases that are highly associated with biomass burning such as carbon monoxide (CO) and nitrogen dioxide ( $\text{NO}_2$ ) show strong correlations with the pyrogenic aerosol component [Longo et al., 1999]. Figure 7 shows the dry season Aethalometer black carbon concentration and  $\text{NO}_2$  while Figure 8 shows the joint variability of BCE with CO. The



**Figure 6.** Dry season  $\text{PM}_{10}$  aerosol mass concentrations for the pasture site measured by two different techniques: Gravimetric by SFU and the TEOM monitor.

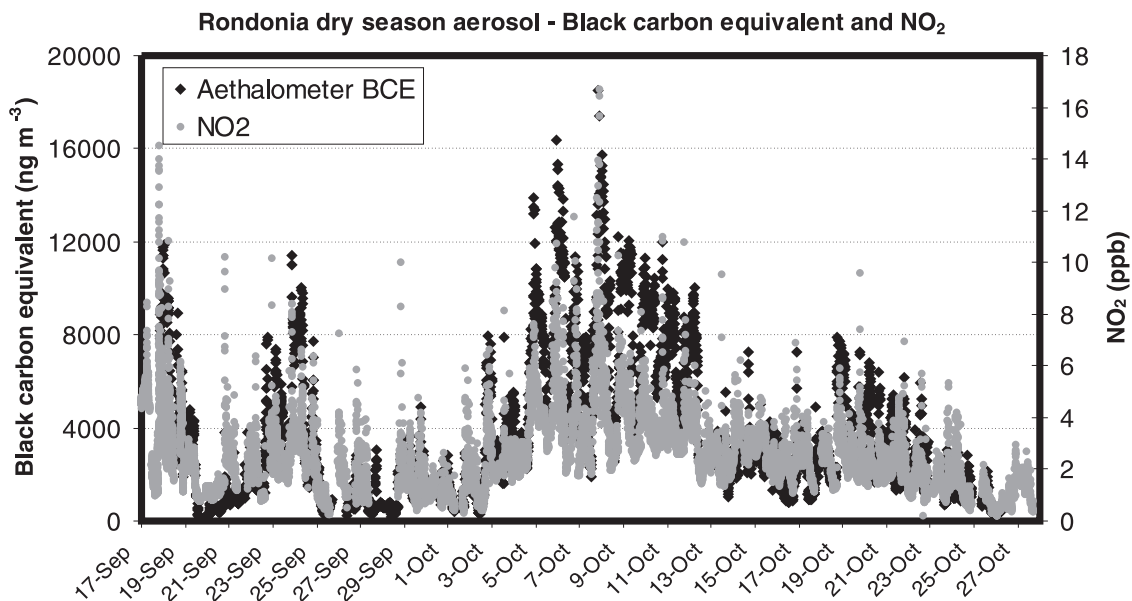


Figure 7. Dry season Aethalometer black carbon equivalent and NO<sub>2</sub> concentrations for the pasture site.

values observed for black carbon in this study are similar to measurements during the SCAR-B experiment [Martins *et al.*, 1998a, 1998b; Artaxo *et al.*, 1998]. During the peak of the dry season, CO concentrations can reach 8 ppm, while NO<sub>2</sub> concentrations reach 16 ppb, values comparable to urban air pollution episodic conditions. The large simultaneous increase in aerosol, CO and NO<sub>2</sub> at the end of September and mid-October is evident. CO has an estimated lifetime of 7–10 days in Amazonia, longer than the estimated value for the fine-mode black carbon aerosol, and NO<sub>2</sub> is very reactive under the Amazonian conditions. In spite of the large difference in residence times between these three species, their concentration time series are fairly similar, indicating that the source term dominates their atmospheric concentration.

[20] The high black carbon concentration (BCE) in the fine mode of 16  $\mu\text{g m}^{-3}$  and its large absorption cross section makes this species critical in the vertical column radiative forcing. Recent results by Jacobson [2001] indicate a strong heating effect due to internally mixed BC, with a global mean forcing of up to +0.62  $\text{W m}^{-2}$  from BC. This would make BCE the second most important climate warming agent after CO<sub>2</sub>. In addition, this high level of BCE changes the vertical structure of the atmosphere, making it more stable. In a parallel study, Schafer *et al.* [2002] showed that for Rondônia, a deficit of about  $-80 \text{ W m}^{-2}$  of PAR was observed for one unit of optical depth. When the optical depth reaches 2.5–3, as in this study, PAR deficit can reach  $-200 \text{ W m}^{-2}$ . This very high value probably has significant effects on the Amazonian ecosystem.

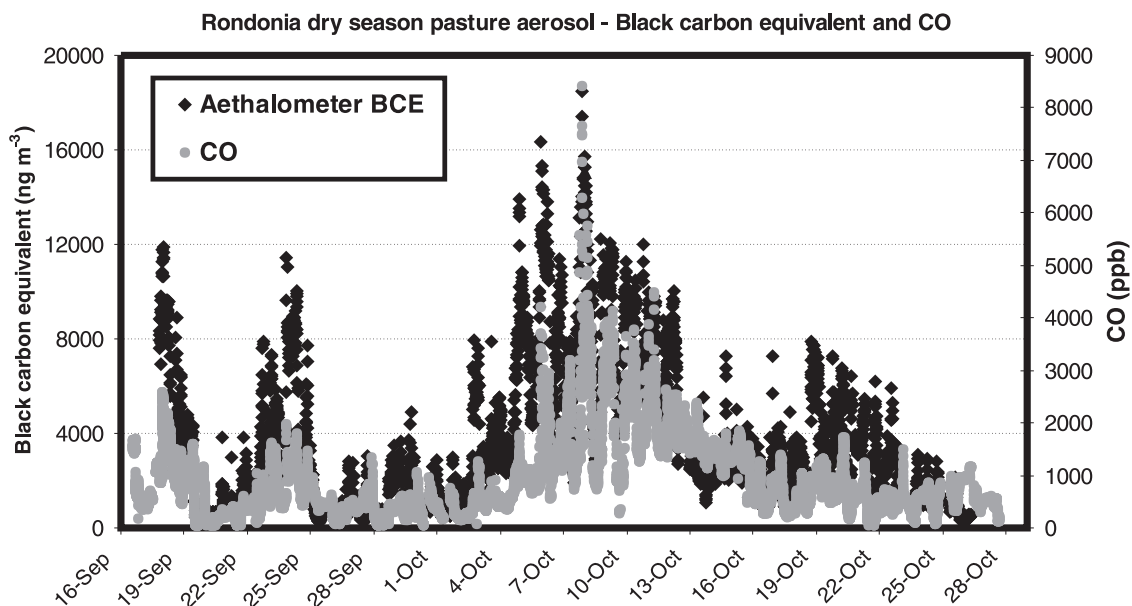
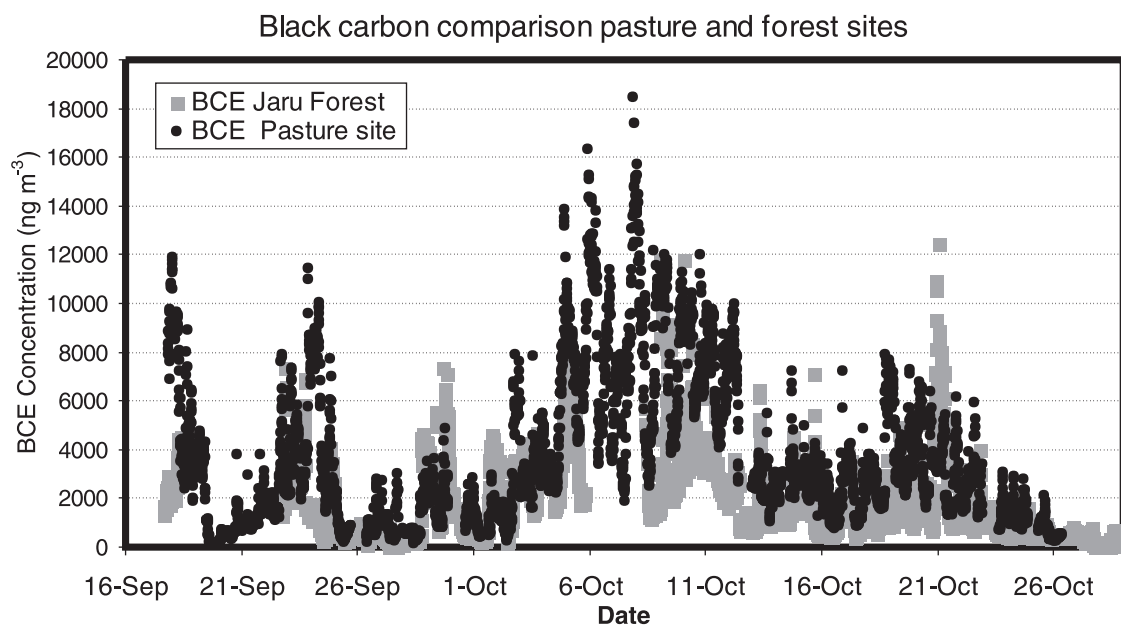


Figure 8. Dry season Aethalometer black carbon equivalent and CO concentrations for the pasture site.





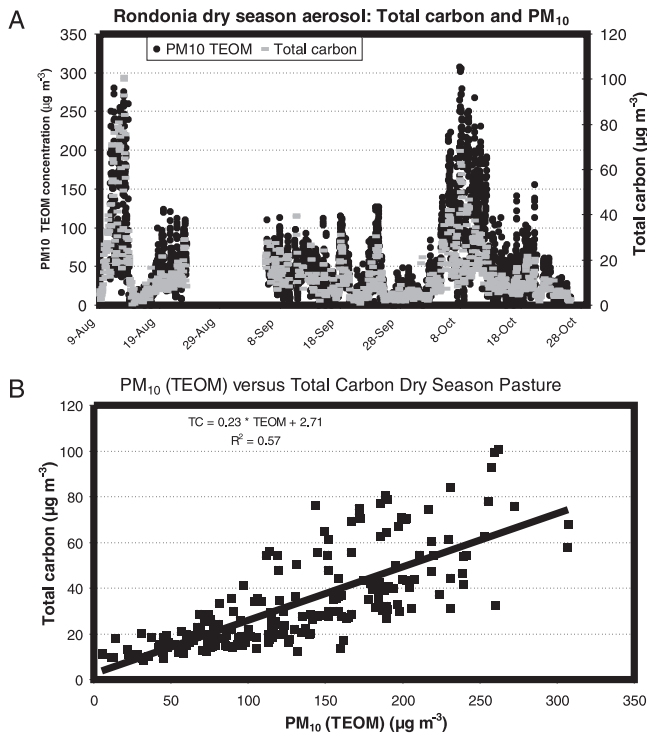
**Figure 9.** Comparison of black carbon equivalent measurements for the pasture and forest sites during the dry season.

[21] The high concentrations measured in the pasture site during the dry season actually represent regional biomass-burning aerosol concentrations, and not a very local effect. To make this point clear, Figure 9 shows a comparison of black carbon equivalent measurements for the pasture and forest sites during the dry season. The sites are about 50 km apart each other, so local emissions in the pasture do not directly affect the forest site. At the pasture site, concentrations are higher, but the large-scale picture can be observed in Figure 9. The episode from 2 to 11 October raised both pasture and forest sites BCE concentrations significantly, indicating that this is a regional large-scale biomass-burning episode.

[22] It was already shown in previous studies of pyrogenic aerosol that the single largest aerosol constituent is the carbonaceous component [Cachier, 1998; Penner *et al.*, 1992; Andreae *et al.*, 1988]. In this study, a real-time carbonaceous aerosol monitor was operated in parallel with the other instruments. Figure 10a shows the time series of total carbon (TC) concentration for aerosol particles less than  $2.5 \mu\text{m}$  versus the TEOM  $\text{PM}_{10}$  aerosol mass. It is evident the very high correlation, indicating that association of most of the aerosol mass with carbonaceous material. The average  $\text{PM}_{10}$  TEOM concentration was  $55.2 \mu\text{g m}^{-3}$  for the whole period, while the total carbon concentration averages  $13.9 \mu\text{g m}^{-3}$ , with most of it ( $10.1 \mu\text{g m}^{-3}$ ) consisting of organic carbon. Figure 10b shows the regression between aerosol mass and total carbon, with a ratio of 23%. The carbonaceous aerosol can be obtained from the carbon concentration, through a factor of 1.8 to account for the oxygen and hydrogen mass in the carbonaceous fraction. This brings the carbonaceous aerosol at about 50% of the aerosol mass. Sulfates, nitrates, soil dust and water makes up the remaining aerosol mass. Graham *et al.* [2002] measured organic aerosols at the same site with a high-volume  $\text{PM}_{10}$  sampler, giving results for TC in the range of  $4.4\text{--}82.3 \mu\text{g m}^{-3}$ , a very similar range than our measure-

ments. Since we used a  $2.5 \mu\text{m}$  aerosol inlet, the general agreement in the results shows that the coarse-mode carbonaceous aerosol mass is not very significant, particularly for the periods where biomass-burning aerosol which is mostly in the fine fraction, dominates the aerosol concentration. Mayol-Bracero *et al.* (submitted manuscript, 2001), Mayol-Bracero *et al.* [2002] measured the water-soluble organic carbon (WSOC) fraction at 45–75% of the OC, a very high value. This fact helps to explain the cloud condensation nuclei properties of these biomass-burning aerosols. One single water-soluble compound (Levoglucosan) accounts for a significant fraction (ca. 6%) of the WSOC. In addition, dicarboxylic acids were observed in significant amounts in Amazonian aerosol [Kubátová *et al.*, 2000; Zbynek *et al.*, 2001]. An in-depth discussion of the temporal variations of TC and its various components (WSOC, non-WSOC, BCE) is provided by Mayol-Bracero *et al.* (submitted manuscript, 2001) and Graham *et al.* [2002].

[23] The atmospheric composition over Amazonia in the wet season gives a completely different picture. Figure 11 shows the time series from 25 January to 25 May of  $\text{PM}_{10}$ , BCE, aerosol particle number and total carbon. During the wet season, natural biogenic aerosol, even at very low concentrations, dominates the aerosol composition [Artaxo and Hansson, 1995; Echalar *et al.*, 1995; Artaxo *et al.*, 1988, 1990; Gerab *et al.*, 1998]. However, a careful analysis of Figures 11b and 11c shows “spikes” in the BCE and CPC measurements, which can be attributed to the large number of charcoal factories in the surrounding region. The nearest charcoal production unit is located at about 10 km from the sampling site, but its influence is clearly observed in the data. Generally, peaks in BCE,  $\text{NO}_2$ , particle number and mass are clearly observed at the beginning of the night. From an average BCE value of  $190 \text{ ng m}^{-3}$  peaks of short duration up to  $1,400 \text{ ng m}^{-3}$  were frequently observed. Aerosol particle numbers jumps from 600 to  $6,000 \text{ \# cm}^{-3}$  or more. This is evidence that



**Figure 10.** (a) Time series of the dry season PM<sub>10</sub> aerosol mass and PM<sub>2.5</sub> total carbon for the pasture site. (b) Relationship between PM<sub>10</sub> aerosol mass and PM<sub>2.5</sub> total carbon for the pasture site during the period when biomass burning impacts were at their maximum strength (10–13 August 1999 and 6–12 October 1999).

for some regions in Amazonia, the anthropogenic perturbation is now already so intense, that the atmospheric background concentrations are disturbed even in the wet season.

### 3.3. Elemental Composition of the Wet and Dry Season Aerosol in Rondônia

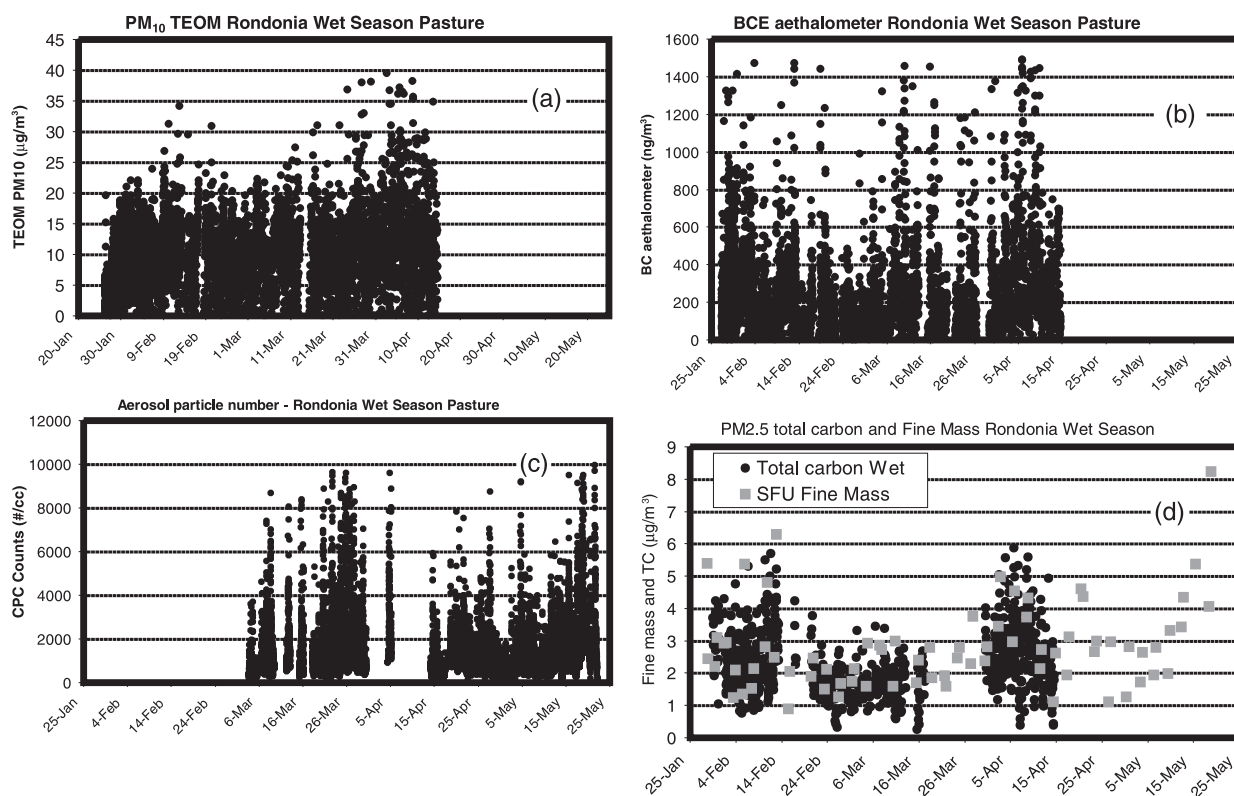
[24] During the wet season, the high precipitation rate makes forest fires and biomass burning impossible, so the natural biogenic aerosol dominates the picture. Table 2 shows the average elemental composition for the fine- and coarse-mode aerosol collected at the pasture and forest sampling sites. Very low concentrations for most of the trace elements were observed. Sulfur, for instance appears at 84 and 88 ng m<sup>-3</sup> levels for both sites, a value similar to those found during the wet season in the central Amazon Basin [Gerab *et al.*, 1998; Andreae *et al.*, 1990; Roberts *et al.*, 2001], but much smaller than observed in Equatorial Africa [Gaudichet *et al.*, 1995]. A careful analysis of the relationship between size and composition of the aerosol and its CCN properties shows that, even at this very low concentration of sulfate, this component still can account for much of the cloud droplet nucleation capacity of the wet-season aerosol [Roberts *et al.*, 2001]. This analysis also indicates, however, the possibility that soluble organic particles could make a substantial contribution to the CCN activity in Amazonia during the wet season. The concentration of soil-dust-related elements such as Al, Si,

Ti, Mn and Fe are also very low, indicating that the forest cover in Amazonia suppress efficiently the soil dust resuspension. The soil dust concentration is enhanced at the pasture site, as expected, and the concentrations were similar to values observed in the Brazilian savannah by Maenhaut *et al.* [1996]. The high precipitation rate in the wet season efficiently removes soil dust aerosol particles. The concentration of heavy metals such as Pb, Cu and Ni are very low. Zinc is associated with natural biogenic particles [Artaxo *et al.*, 1988; Artaxo and Hansson, 1995; Maenhaut *et al.*, 1996]. In the Northern part of Amazonia, it is frequent the observation of air masses loaded with Sahara dust, in episodic events, mostly from February to May, when the ITCZ crosses the central part of the Amazon basin [Swap *et al.*, 1996; Gerab *et al.*, 1998]. These events were observed in Santarém and Manaus, with concentrations up to 80 µg m<sup>-3</sup> of soil dust, but they were never observed in Rondônia.

[25] One particular important issue evident in Table 2 is the presence of a significant coarse-mode phosphorus concentration. Phosphorus is a key and limiting nutrient in the Amazonian ecosystem. It is noticeable that coarse-mode phosphorus appears in concentrations higher than Ca, a much more abundant element in the Amazonian soils and biota. These high P concentrations in the coarse mode are further enhanced at nighttime aerosol samples. This could be an important ingredient in the nutrient cycling in Amazonia, since rainwater measurements shows that P wet deposition is very small or negligible. Further studies on this issue are under way.

[26] Table 3 shows the dry season average aerosol elemental composition. Much higher concentrations than in the wet season were observed. Sulfur appears in concentrations of 500 to 1000 ng m<sup>-3</sup>, and increase of seven times over the natural biogenic component in the wet season. Also, soil dust related elements show much higher concentrations because of the dryer climate and the enhanced convection during biomass burning [Freitas *et al.*, 2000]. Concentrations at the pasture site are higher than in the forest, because biomass-burning emissions are more intense near the pasture site. At the forest site, mostly large-scale regional biomass burning is observed, with very few local events. The measured ambient values are similar to measurements obtained from direct emissions with aerosol collected over fires in cerrado and forest [Yamasoe *et al.*, 2000]. The values observed in this study for the dry season are similar to the ones observed in Africa [Gaudichet *et al.*, 1995], and previously in Brazil [Hacon *et al.*, 1995; Echalar *et al.*, 1998].

[27] One very important and interesting feature is the total aerosol and phosphorus distribution along the forest canopy at the Jaru tower in the wet season. Figure 12 shows the aerosol mass concentrations for the wet season for each of the three levels of the Jaru Tower. The fine particle concentrations are virtually identical for the three levels, with no evidence of production or deposition. For the coarse-mode aerosol, however, a strong concentration gradient was observed, with highest concentrations at the bottom of the tower. This is evidence that internal forest canopy process could be responsible for a large fraction of the aerosol concentration in pristine forest sites. This enhancement of aerosols at the tower ground level is



**Figure 11.** Time series of aerosol characteristics for the wet season in Rondônia. (a) PM<sub>10</sub> aerosol mass concentration measured with the TEOM monitor; (b) Black carbon; (c) Aerosol particle number; and (d) Total carbon and fine mode aerosol mass.

sharply more pronounced during nighttime. Figure 13 shows phosphorus concentration for each of the three levels of the Jaru Tower during the wet season, separating daytime from nighttime aerosol samples. The fine-mode phosphorus concentration is essentially constant along the forest canopy. The coarse-mode phosphorus concentration

increases from  $16 \text{ ng m}^{-3}$  at the top of the tower at daytime to a high  $68 \text{ ng m}^{-3}$  at nighttime under the canopy. It is unclear which biogenic mechanism could be responsible for this nighttime coarse-mode phosphorus enhancement. Similar patterns were also observed in the Santarém and Manaus towers. This phenomenon is even

**Table 2.** Wet Season Fine and Coarse Mode Average Aerosol Elemental Composition for the Pasture and Forest Sites<sup>a</sup>

	Pasture site				Forest site			
	Fine mode		Coarse mode		Fine mode		Coarse mode	
	Average	Std. dev.	Average	Std. dev.	Average	Std. dev.	Average	Std. dev.
Mass	2.94	1.71	5.70	3.07	2.17	1.34	5.09	2.60
BCE	477	240	–	–	281	14	–	–
Al	43.9	37.8	92.3	105.1	35.69	29.5	44.2	43.9
Si	82.2	58.1	135.8	156.2	37.83	33.9	50.1	54.63
P	7.40	2.53	37.9	23.3	4.83	1.89	34.0	23.7
S	83.9	78.8	24.0	14.1	88.1	88.7	28.2	15.9
Cl	5.50	4.94	14.3	12.8	5.07	6.15	9.37	5.30
K	33.5	28.47	76.7	42.1	26.2	26.6	73.7	45.8
Ca	7.69	4.89	18.3	12.2	6.39	5.48	10.8	6.07
Ti	3.95	4.59	8.57	11.51	3.51	3.46	5.40	6.22
Cr	1.89	0.48	4.67	3.18	–	–	3.02	1.95
Mn	0.74	0.59	1.90	1.58	0.56	0.34	0.82	0.51
Fe	26.0	34.0	64.4	77.1	21.4	27.8	30.1	40.7
Ni	0.30	0.20	0.44	0.41	0.25	0.07	0.40	0.03
Cu	0.22	0.21	0.21	0.10	0.33	0.38	0.31	0.25
Zn	0.64	0.67	0.91	0.61	0.78	0.84	1.00	0.72
Sr	0.21	0.09	0.35	0.20	0.15	0.07	0.28	0.19
Zr	0.43	0.38	0.83	0.62	0.43	0.52	0.81	0.71
Pb	1.08	1.23	0.79	0.66	0.92	0.97	0.50	0.24

<sup>a</sup> Averages and standard deviations are shown. Elemental concentrations and BCE are expressed in  $\text{ng m}^{-3}$ . Aerosol mass is expressed in  $\mu\text{g m}^{-3}$ . Only concentrations measured above analytical detection limits were used in calculating the averages and standard deviations.

**Table 3.** Dry Season Fine and Coarse Mode Average Aerosol Elemental Composition for the Pasture and Forest Sites<sup>a</sup>

	Pasture site				Forest site			
	Fine mode		Coarse mode		Fine mode		Coarse mode	
	Average	Std. dev	Average	Std. dev	Average	Std. dev	Average	Std. dev
Mass	66.9	55.1	17.8	11.7	33.4	20.8	6.5	6.1
BCE	7420	6110	–	–	1830	1310	–	–
Al	274	269	502	586	88.5	50.6	79.3	39.6
Si	364	420	875	993	119	83	89.8	110.7
P	32.8	20.8	68.2	29.0	26.7	11.95	46.9	26.4
S	1104	870	155	121	529	272	59.3	27.4
Cl	62.2	56.6	28.2	19.4	20.4	9.1	10.1	13.6
K	1109	865	236	150.5	506	316	88.3	55.47
Ca	50.8	36.5	135	111	25.1	21.5	45.8	60.4
Ti	32.7	30.4	65.1	71.6	6.01	4.95	7.99	4.83
Cr	9.54	10.14	12.9	42.8	10.3	5.41	15.4	18.0
Mn	4.95	3.43	16.1	13.7	1.87	1.30	3.92	3.35
Fe	162	181	455	501	28.2	24.7	47.5	37.8
Cu	2.09	4.19	3.34	5.29	1.61	1.35	3.94	4.16
Zn	8.33	6.38	4.18	3.21	4.17	3.02	2.85	1.45
Br	21.9	16.6	47.4	35.9	10.2	6.2	25.1	13.8

<sup>a</sup>Elemental concentrations and BCE are expressed in ng m<sup>-3</sup>. Aerosol mass is expressed in µg m<sup>-3</sup>. Only concentrations measured above analytical detection limits were used in calculating the averages.

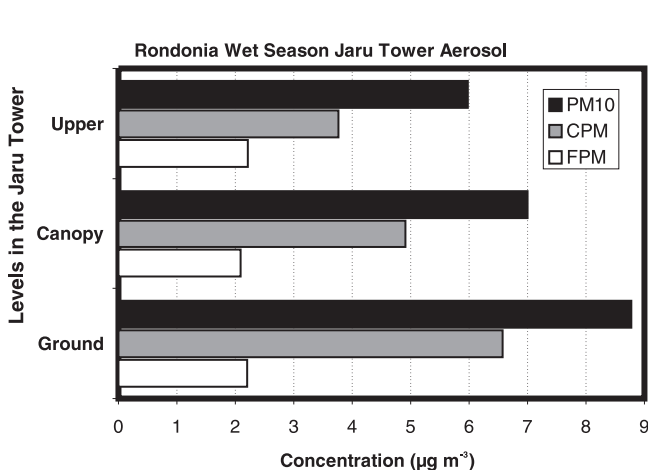
more important if we call the attention that P is the limiting nutrient for the Amazonian forest and that P is not present at concentrations above 1 ppb in Amazonian precipitation. Phosphorus is exchanged in the ecosystem exclusively in the aerosol phase and mostly for coarse-mode particles and during nighttime.

#### 4. Conclusions

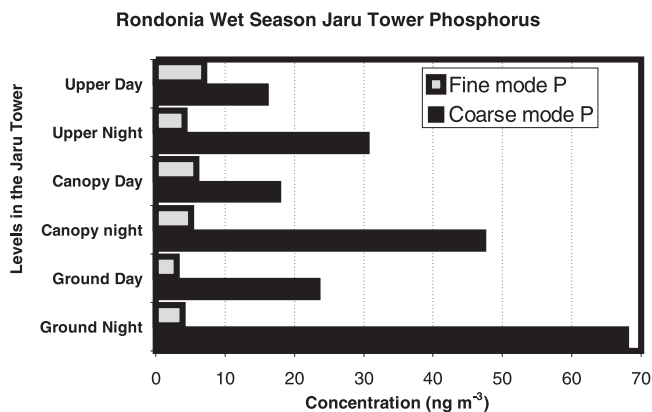
[28] A large difference in aerosol concentration and composition was observed between dry and wet season and between forest and pasture sites in Rondônia. In the wet season, very low aerosol and trace gas concentrations were observed. But, short episodes with plumes from charcoal factories were observed in the wet season, reflecting the occupation and anthropogenic influence in parts of Amazonia already significantly deforested, such as Rondônia. Particle number concentrations measured at 600–900 cm<sup>-3</sup> are low, but are a bit higher than remote marine conditions values. Consequently, the CCN concentrations are also low

[Roberts et al., 2001; Zhou et al., 2002], making cloud properties over Amazonia in the wet season similar to the remote marine environment. Most of the aerosol mass consists of organic compounds, with low sulfate concentrations.

[29] On the other hand, in the dry season we observed a heavily polluted atmosphere at both pasture and forest sampling sites. The large impact of pyrogenic aerosols changes the radiation balance of the atmosphere, with effects on the Amazonian ecosystem. Aerosol particle concentrations up to 40,000 cm<sup>-3</sup> were observed, in sharp contrast with the low aerosol abundance in the wet season. High correlation was observed between BCE, NO<sub>2</sub>, CO, aerosol particle number and mass, aerosol optical thickness and other properties linked to the biomass emissions. Most of the aerosol mass is organic and a large fraction is water-soluble compounds, as observed in the dry season in a parallel study [Mayol-Bracero et al., submitted manuscript, 2001; Graham et al., 2002]. This water-soluble component



**Figure 12.** Aerosol mass concentrations for the wet season for each of the three levels of the Jaru Tower.



**Figure 13.** Phosphorus concentration for each of the three levels of the Jaru Tower (upper, canopy, and ground levels) during the wet season, separating daytime from nighttime aerosol samples.

is necessary to explain the CCN activity observed in Rondônia and other areas of Amazonia, also in parallel studies [Roberts *et al.*, 2001; Zhou *et al.*, 2002].

[30] More research is needed to investigate the strong enhancement of coarse-mode phosphorus observed at nighttime samples for both forest and pasture sites. It could have important implications in the Amazon basin nutrient cycling. Also investigation on the role of natural biogenic aerosol particles in nucleating clouds is relevant, since an enhanced anthropogenic activity in Amazonia could alter the cloud formation mechanism, with implications far from the Amazon region.

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