

## Long-term monitoring of atmospheric aerosols in the Amazon Basin: Source identification and apportionment

Francisco Echalar, Paulo Artaxo, José Vanderlei Martins, Márcia Yamasoe, and Fábio Gerab

Instituto de Física, Universidade de São Paulo, São Paulo, Brazil

Willy Maenhaut

Institute for Nuclear Sciences, University of Gent, Gent, Belgium

Brent Holben

NASA Goddard Space Flight Center, Greenbelt, Maryland

**Abstract.** Continuous sampling of atmospheric aerosols was carried out at two different sites in the Amazon Basin: Cuiabá (16°S, 56°W), since July 1990, and Alta Floresta (09°S, 56°W), since August 1992. Aerosols were collected on polycarbonate filters mounted in stacked filter units (SFU). Particle-induced X ray emission (PIXE) was used to measure concentrations of up to 26 elements (Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Rb, Sr, Zr, Au, and Pb). Black carbon (BC) and gravimetric mass analyses were also performed. Inhalable particle ground concentrations showed a marked seasonality, with maxima of more than  $100 \mu\text{g m}^{-3}$  in the dry season. The high aerosol optical thickness (AOT) values in this period (up to 4.0 at  $\lambda = 0.440 \mu\text{m}$ ) indicate that the whole air column has a significant aerosol load. Three main types of aerosol sources were identified: biomass burning, natural biogenic emissions, and soil dust resuspension. During the dry season the fine mode aerosol originated predominantly from biomass burning emissions (they were responsible for about 73% of fine aerosol mass), while the coarse mode was dominated by soil dust particles (50 to 60% of the aerosol mass). Crustal elements exhibited a unimodal coarse mode size distribution, while the pyrogenic elements showed a clear submicrometer mode. Black carbon had a submicrometer mode centered at  $0.175 \mu\text{m}$ . During the wet season, biogenic aerosols were the dominant particle type and also the main source of atmospheric P in the region.

### 1. Introduction

During the past two decades it has become increasingly evident that tropical regions are of major importance for the global atmosphere [Andreae and Crutzen, 1997]. Large amounts of biogenic gases and particles are emitted by the tropical rain forest [Harriss *et al.*, 1988; 1990]. Also, more than 80% of biomass burnt worldwide each year originates from tropical regions, and the burning leads to emissions of particles and gases that may affect atmospheric chemistry and climate [Crutzen and Andreae, 1990; Crutzen and Goldammer, 1993; Gash *et al.*, 1996].

The world's largest rain forest is located in the Amazon Basin. Its ecosystems are characterized by intense internal recycling, but they also depend on atmospheric input of certain mineral nutrients [Salati and Vose, 1984; Vitousek and Sanford, 1986]. In the past 20 years, several parts of the Amazon Basin have been under strong pressure by intense immigration and a rapid change of land cover. Forests and adjacent savannas are being cleared, most of the time with the use of fire, and converted into pasture and agricultural fields at a substantial rate [Kaufman *et al.*, 1990; Fearnside, 1991]. The pyrogenic and

natural biogenic emissions from the Amazon Basin may have an impact on global tropospheric chemistry, because the region exhibits intense convective activity [Garstang *et al.*, 1988] which may inject gases and aerosols to high altitudes where they can be transported over long distances. Earlier studies in the region were characterized by short-term intensive sampling campaigns. They focused either on the natural release of biogenic aerosols by tropical rain forests [Lawson and Winchester, 1979; Orsini *et al.*, 1982; Artaxo and Orsini, 1986, 1987; Artaxo *et al.*, 1988, 1990, 1993; Artaxo and Hansson, 1995] or on the characterization of pyrogenic aerosols [Leslie, 1981; Andreae *et al.*, 1988; Ward *et al.*, 1991, 1992; Artaxo *et al.*, 1993; Kaufman *et al.*, this issue].

To increase our knowledge of the chemical processes that determine the composition of the atmosphere in background areas and to understand how changes in land use in tropical forests affect the atmospheric aerosol composition of the region, the Group for Studies of Air Pollution (GEPA) of the University of São Paulo started a long-term monitoring program in July 1990. In its initial phase the program involved the operation of a long-term aerosol sampling station in Cuiabá, a city south of the Amazon Basin. In August 1992 a second monitoring station was added, i.e., in Alta Floresta, a small city in southern Amazonia. Both sites are heavily affected by biomass burning emissions during the dry season (June to October). Parts of the data sets obtained have been presented and



**Figure 1.** Location of the sites in Brazilian Amazonia where continuous monitoring of aerosol particles is being carried out.

discussed previously. Using samples collected up to March 1993, Artaxo *et al.* [1994] performed a preliminary identification of the main sources of the fine particles at Cuiabá and at Alta Floresta and derived source profiles for the fine size fraction. Maenhaut *et al.* [1996a] made a source apportionment for both the fine and the coarse fractions of the Cuiabá aerosol, but they restricted their study to samples collected from July 1992 to September 1993.

The objective of this work is to present a comprehensive picture of the atmospheric aerosol types, their size distribution, and radiative impact in disturbed areas of the Amazon Basin. First, a source identification and up-to-date estimation of the contributions of the main aerosol sources are carried out. In this analysis, use is made of the longest continuous time series available for Cuiabá (samples from July 1990 to August 1995) and for Alta Floresta (samples from August 1992 to March 1995). Results are presented for both the fine and the coarse size fractions, separated by season and by site, covering the whole sampling period to 1995. To provide a more detailed picture of the atmospheric aerosols in the Amazon Basin during the dry season, size distributions obtained during intensive sampling campaigns at Cuiabá and Alta Floresta are also presented. Results from a network of spectral Sun photometers and sky radiometers operated by NASA are also included and discussed in order to extend the ground-based results to the whole atmospheric column and provide evidences of the radiative impact of the aerosol burden in the region.

## 2. Pyrogenic and Natural Biogenic Emission of Particles

Natural emissions of particulate matter by the forests include primary particles, such as pollen, spores, and fragments of leaves, and secondary organic particles formed by gas-to-particle conversion of biogenic trace gases. The primary particles are mostly coarse in size (particle diameter ( $dp$ )  $> 2 \mu\text{m}$ ),

while the secondary aerosol tends to be of submicrometer size. The particle emission mechanisms are still not fully understood but 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. These processes may generate particles which contain elements such as Na, Mg, P, S, K, Ca, Zn, and Rb [Ferry and Ward, 1959; Nemeruyk, 1970; Beauford *et al.*, 1975, 1977]. The biogenic particles can act as cloud condensation nuclei and potentially affect cloud formation mechanisms and cloud dynamics [Maki and Willoughby, 1978; Schnell, 1982; Saxena *et al.*, 1995]. Previous studies have shown that the vegetation is an important natural source of airborne particles [Artaxo *et al.*, 1988, 1990, 1994] and that the biogenic aerosol is responsible for a significant part of the atmospheric phase of the biogeochemical cycles of elements such as carbon [Marenco and Delauney, 1980; Cachier *et al.*, 1985], sulfur [e.g., Lodge *et al.*, 1974; Orsini *et al.*, 1982; Delmas and Servant, 1982, 1983], and potassium [Croizat, 1979].

Every year large amounts of biomass are burnt worldwide, and this leads to significant emissions of gases and particles into the atmosphere. The pyrogenic particles are mainly of submicrometer size and contain high concentrations of organic compounds and black carbon [Cachier *et al.*, 1991; Ward *et al.*, 1992; Artaxo *et al.*, 1993, this issue]. Black carbon emissions by fires are estimated at  $5.65 \text{ Tg yr}^{-1}$  and account for about 45% of the total anthropogenic emissions of black carbon [Liousse *et al.*, 1996]. These emissions could result in regionally significant direct radiative forcing [Holben *et al.*, 1991, 1996a; Hobbs *et al.*, 1997]. The fires are also important sources of aerosol potassium, sulfur, silicon, zinc, halogens, and organic matter [Ward *et al.*, 1992; Artaxo *et al.*, 1993; Cachier *et al.*, 1995; Gaudichet *et al.*, 1995; Maenhaut *et al.*, 1996b]. The trace element fluxes from biomass burning can be significant on a global scale [Echalar *et al.*, 1995]. The pyrogenic particles can be active as cloud condensation nuclei (CCN) [Rogers *et al.*, 1991; Dinh *et al.*, 1994; Kaufman and Fraser, 1997]. They may affect the concentrations and sizes of cloud droplets and thus lead to changes in cloud radiative properties and precipitation patterns [Kaufman and Tanré, 1994; Kaufman *et al.*, this issue].

## 3. Experiments and Analytical Methods

Figure 1 shows the location of the sampling sites. The first one is in Cuiabá ( $16^{\circ}\text{S}$ ,  $56^{\circ}\text{W}$ ), south of the Amazon rain forest, in a cerrado (savanna) region. This sampling site is heavily affected by regional savanna biomass burning. Samples have been continuously collected at this site since July 1990. Data from samples collected until August 1995 were used for the current paper. The second sampling site is in the region of Alta Floresta ( $09^{\circ}\text{S}$ ,  $56^{\circ}\text{W}$ ), near the border of Mato Grosso and Amazonas States. Alta Floresta is located in a primary forest region which is heavily stressed by land clearings and gold mining [Hacon *et al.*, 1995]. The nearest major urban area is more than 500 km away. Samples have been continuously collected at this site since August 1992. Data from samples collected before April 1995 were used in this work. At both sites, wet (austral summer) and dry (austral winter) seasons succeed each other in alternation. The dry season is assumed to begin every year on June 1 and end October 31. Although the actual onset of the dry season varies from year to year, this 5 month period includes the driest months, when most of the deforestation and agricultural fires take place.

Bulk aerosol samples were collected using stacked filter units (SFU) [Parker *et al.*, 1977] fitted with a PM<sub>10</sub> inlet [Hopke *et al.*, 1997]. The collection substrates in the SFU consist of 47 mm diameter polycarbonate membrane filters. An 8 μm pore size filter is used for the collection of the coarse particles (2.0 < dp < 10 μm), while a 0.4 μm pore size filter collects the fine particles (dp < 2.0 μm) [John *et al.*, 1983]. The flow rate was typically 16 L/min, and the sampling time varied from 24 hours during the burning season to a week during the wet season. Particle bounce-off from the filters is not a problem in the Amazon Basin due to the high relative humidity (70–95%). In addition to the SFU samplings, aerosol collections with higher size segregation were performed in different periods at Alta Floresta and at Cuiabá with the aid of cascade impactors. In August 1993 a multi-orifice uniform deposit impactor (MOUDI) was used in Alta Floresta. The eight stages of the MOUDI have  $d_{50}$  size cuts of 18, 3.2, 1.8, 1.0, 0.56, 0.33, 0.175, and 0.093 μm equivalent aerodynamic diameter (EAD); a 0.4 μm pore size Teflon backup filter was used to collect the particles smaller than 0.093 μm. Only the samples from the MOUDI stages with submicrometer size cuts were analyzed. In August 1995, during the Smoke, Clouds, and Radiation–Brazil (SCAR–B) experiment, a Battelle-type, seven-stage, single-orifice, electrically conductive plastic cascade impactor (PIXE International Corp., Tallahassee, Florida) was used in Cuiabá. The seven stages of the particle-induced X-ray emission (PIXE) cascade impactor have  $d_{50}$  size cuts of 16, 8, 4, 2, 1, 0.5, and 0.25 μm EAD; the particles smaller than 0.25 μm were collected on a 0.4 μm pore size polycarbonate filter. The raw elemental size data of the Cuiabá cascade impactor samples were inverted into smooth size distributions, and lognormal curves were then fitted to the latter. The procedures used were described by Hillamo *et al.* [1993] and Havránek *et al.* [1996].

Elemental concentrations were determined by the PIXE spectrometry [Johansson *et al.*, 1995]. Concentration data were obtained for up to 26 elements (Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Rb, Sr, Zr, Au, and Pb). Most of the PIXE analyses were performed at the dedicated 5SDH tandem Pelletron accelerator facility of the University of São Paulo LAMFI (Laboratório de Análise de Materiais por Feixes Iônicos). 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. These detection limits were calculated on the basis of a sampling flow rate of 16 L/min, sampling time of 48 hours and irradiation time of 600 s. The precision of elemental concentration measurements is typically better than 10% but increases up to 20% or more for elements with concentrations near the detection limit.

Aerosol mass concentrations were obtained through gravimetric analysis. Both fine and coarse filters were weighed before and after sampling in a Mettler M3 electronic microbalance with 1 μg sensitivity. Before weighing, filters were kept for 24 hours at 50% relative humidity and 20°C. Electrostatic charges were controlled by means of <sup>210</sup>Po radioactive sources. The detection limit for the aerosol mass concentration is 0.3 μg m<sup>-3</sup>. Precision is estimated at about 15%. Black carbon concentrations were obtained by a light reflectance technique [Andreae, 1983; Andreae *et al.*, 1984; Reid *et al.*, this issue (a)].

From 1992 to 1994, NASA operated a network of spectral Sun photometers and sky radiometers at selected sites in the Amazon Basin [Holben *et al.*, 1996b, 1996c]. Direct Sun measurements were performed at 340, 440, 670, 870, 940, and 1020 nm every 15 min and sky radiance measurements hourly,

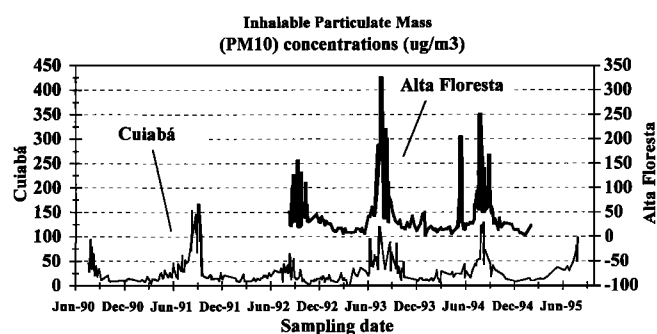


Figure 2. Inhalable particle ( $dp < 10 \mu\text{m}$ ) mass concentrations at Cuiabá and Alta Floresta.

weather permitting. Using precalibrated instruments, direct Sun measurements were converted to aerosol optical thickness (AOT) at each wavelength. AOT values at 440 nm, representing the effect of the aerosol burden over the whole atmospheric column, are compared in this paper with ground-based measurements of aerosol fine particle mass (FPM) concentration.

A multivariate statistical technique, principal component analysis (PCA) [Harman, 1976; Henry, 1991], was used to identify the different sources which contribute to the atmospheric aerosol of Alta Floresta and Cuiabá. First, a model of the variability of the trace element concentrations is constructed so that the set of interrelated measured variables is transformed into a set of independent variables, the principal components. Each variable, measured or resulting from the PCA, is responsible for part of the variance in the data set. The principal components that explain less than one unit of variance are supposed to represent only noise and are excluded before VARIMAX rotation. The PCA gives, on the one hand, “component loadings,” which are the correlation coefficients between the original measured variables and the new principal components and, on the other hand, “component scores,” which are a measure of the relative importance of a component in each sample. The stability of PCA depends strongly on the number of samples included in the analysis [Ito *et al.*, 1986]. Henry [1991] recommends that there should be enough samples to have at least 30 degrees of freedom. We fully complied with this criterion in this work. In our worst case we still had 69° of freedom. The PCA results are quantified using the absolute principal component analysis (APCA) approach developed by Thurston and Spengler [1985]. In APCA, quantitative estimates of the contribution from each component to the atmospheric concentration of an element or of the aerosol are obtained through regressions of the measured concentrations on previously calculated “absolute principal component scores” (APCS). Thus APCA provides an aerosol source apportionment [Thurston and Spengler, 1985].

## 4. Results and Discussion

### 4.1. Bulk Aerosol Concentrations

Figure 2 shows that the inhalable particle mass concentration (IPM, particles with  $dp < 10 \mu\text{m}$ ) is strongly seasonal at both sampling sites, with maxima during August and September every year. In these months the levels are well above the wet season concentrations; they usually peak to over 100 μg m<sup>-3</sup>, with a maximum of more than 300 μg m<sup>-3</sup> in August 1993 for Alta Floresta. The IPM levels are highest late in the

**Table 1.** Averages and Standard Deviations for Atmospheric Concentration (in  $\text{ng m}^{-3}$ ) of Particulate Mass, Black Carbon, and Aerosol Trace Elements in Samples Collected in Alta Floresta From August 1992 to March 1995

Var.	Dry Season						Wet Season					
	Fine Mode			Coarse Mode			Fine Mode			Coarse Mode		
	Mean	s.d.	$N^a$	Mean	s.d.	$N^a$	Mean	s.d.	$N^a$	Mean	s.d.	$N^a$
Mass <sup>b</sup>	47	41	126	34	21	124	5.5	3.5	75	16.4	9.4	77
BC <sup>b</sup>	5.73	6.84	126	...	...	...	0.66	0.41	75	...	...	...
Mg	460	460	42	670	990	75	103	110	37	103	105	46
Al	340	530	126	2200	2200	124	27	23	75	230	240	77
Si	520	440	63	2100	2200	124	74	57	37	270	230	77
P	32	17	46	90	69	124	4.6	7.4	12	86	56	77
S	950	910	126	180	150	124	170	130	75	140	103	77
Cl	67	75	24	58	75	58	2.3	1.3	6	41	24	68
K	1200	1800	126	390	330	124	94	74	75	270	180	77
Ca	40	37	126	230	360	124	8.0	7.2	75	44	28	77
Ti	22	31	126	140	135	124	2.1	1.7	75	17	15	77
V	1.3	1.3	3	5.7	4.6	49	...	...	...	1.05	0.76	20
Cr	37	66	6	4.6	...	1	5.5	3.2	3	2.21	...	1
Mn	3.8	3.5	56	15	20	124	0.53	0.46	45	3.0	3.1	77
Fe	223	330	126	1400	1300	124	24	23	75	190	190	77
Ni	1.6	1.0	42	3.0	4.1	46	1.2	1.7	9	0.49	0.48	28
Cu	2.5	2.9	61	5.4	7.2	44	0.69	0.99	17	1.2	1.7	41
Zn	8.4	9.2	126	4.7	4.2	124	1.19	0.72	75	2.3	1.6	75
Ga	0.86	0.89	29	1.5	1.5	44	0.10	0.12	16	0.59	0.76	20
Ge	0.68	0.69	15	1.5	1.9	18	0.12	0.10	9	0.233	0.015	3
As	0.90	0.75	25	1.8	2.4	31	0.19	0.08	14	0.24	0.17	7
Se	0.62	0.53	38	0.95	0.99	46	0.12	0.16	25	0.21	0.30	19
Br	17	16	27	2.0	2.0	11	1.37	0.42	7	0.68	0.46	17
Rb	6.9	12.3	42	3.1	3.2	53	0.53	0.43	16	0.80	0.54	58
Sr	1.1	1.3	17	4.3	6.8	78	0.21	0.14	10	0.67	0.54	57
Zr	3.8	2.5	2	4.4	3.6	17	0.81	0.68	3	0.90	0.29	12
Au	1.7	1.6	35	1.6	2.0	23	0.18	0.13	20	0.29	0.18	13
Pb	3.3	3.2	126	2.6	2.9	49	0.52	0.30	75	1.0	1.4	47

<sup>a</sup> $N$  is the number of samples in which the element was observed above its detection limit.

<sup>b</sup>Aerosol mass and black carbon (BC) are expressed in  $\mu\text{g m}^{-3}$ .

dry season, when extensive forest and savanna fires are taking place. The lowest dry season peaks are noticed in 1992 for both sites. In that year, one of the longest continuous El Niño-Southern Oscillation (ENSO) events was recorded. The ENSO caused cool and wet conditions over parts of central South America [Bachmeier and Fuelberg, 1996] and thus changed relative humidity and air temperature, which are important parameters affecting fire efficiency [Lobert and Warnatz, 1993].

Table 1 gives the Alta Floresta average particle mass, black carbon (BC), and trace element concentrations for each size fraction and each season. The elements listed account on average for 7% of the fine particle mass and for up to 21% of the coarse particle mass. The remainder of the mass is made up by light elements, black carbon, organic compounds, and water. All trace element concentrations are higher in the dry season than in the wet season. The seasonal difference is most pronounced for the crustal (e.g., Al, Ti, and Fe) and pyrogenic elements (e.g., K, Zn) in the fine size fraction (dry/wet season ratio of 10 or more). In the coarse fraction, there is a large seasonal difference for the crustal elements (dry/wet season ratio larger than 6) but not for the typical pyrogenic elements.

Table 2 presents the Cuiabá average particle mass, BC, and trace element concentrations for each size fraction and each season. Most of the aerosol mass consists of light elements, BC, organic compounds, and water, since the trace elements account only for about 10% of the fine particle mass and for about 23% of the coarse particle mass. The concentrations are higher in the dry season than in the wet season. The differences

are largest for K, Cl, and Br, due to biomass burning emissions. For fine Cl the average concentration is almost 12 times higher in the dry season than in the wet season. For coarse Cl, on the other hand, similar average values are noted in both seasons. The differences for the crustal elements are lower than at Alta Floresta, suggesting that the crustal source has a less marked seasonality in Cuiabá than in Alta Floresta. A comparison of the data from the two locations shows that the particle mass and trace element levels are similar in the coarse size fraction. In the fine fraction, on the other hand, higher concentrations are observed at Alta Floresta than at Cuiabá, especially during the dry season. For both coarse and fine P, average levels are higher at Alta Floresta than at Cuiabá, probably due to the large extent of forested areas in the region of Alta Floresta.

At each of the two sites (Tables 1 and 2), the dry season averages of fine and coarse particulate matter concentrations are substantially larger than the wet season averages. Low precipitation rates, which reduce wet deposition and are favorable for soil dust resuspension, coupled with the maximum intensity of an important aerosol source in the region, i.e., the deforestation and agricultural fires [Artaxo et al., 1994; Maenhaut et al., 1996a], are responsible for the large dry season values. Dry season FPM average concentration is higher at Alta Floresta than at Cuiabá, suggesting that the impact from the fires on the atmosphere is more important at Alta Floresta than at Cuiabá. Average concentrations of FPM and coarse particle mass (CPM) are higher than those observed during the ABL (Amazon Boundary Layer Experiment) 2A and 2B ex-

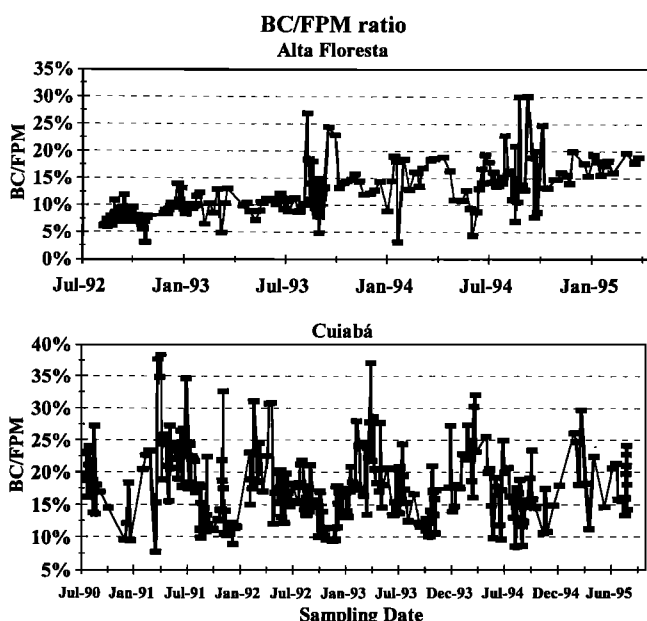
**Table 2.** Averages and Standard Deviations for Atmospheric Concentration (in  $\text{ng m}^{-3}$ ) of Particulate Mass, Black Carbon, and Aerosol Trace Elements in Samples Collected in Cuiabá From July 1990 to August 1995

Var.	Dry Season						Wet Season					
	Fine Mode			Coarse Mode			Fine Mode			Coarse Mode		
	Mean	s.d.	$N^a$	Mean	s.d.	$N^a$	Mean	s.d.	$N^a$	Mean	s.d.	$N^a$
Mass <sup>b</sup>	17	14	149	32	24	154	3.9	1.8	127	11.7	5.7	131
BC <sup>b</sup>	2.6	1.7	149	...	...	...	0.72	0.33	127	...	...	...
Al	102	72	149	1240	740	154	33	30	127	407	270	131
Si	230	160	149	2700	1600	154	96	67	127	970	590	131
P	4.7	3.4	41	17	12	154	2.3	1.4	80	18	13	131
S	400	260	149	160	180	154	170	110	127	65	39	131
Cl	12	19	58	24	20	154	1.03	0.93	13	14.3	9.1	131
K	440	310	149	580	440	154	80	44	127	200	100	131
Ca	29	18	149	460	330	154	12.0	6.5	127	180	120	131
Ti	10.8	6.9	149	120	78	154	3.4	2.6	127	36	26	131
V	1.17	0.99	46	5.2	3.8	111	0.49	0.43	43	1.6	1.4	91
Cr	2.4	1.5	14	2.2	1.8	29	1.2	2.3	20	1.16	0.88	25
Mn	1.5	1.0	149	15	11	154	0.57	0.37	127	3.4	2.5	131
Fe	190	120	149	1800	1000	154	82	60	127	790	470	131
Ni	0.39	0.53	50	1.09	0.78	34	0.19	0.10	39	0.55	0.40	25
Cu	1.11	0.80	149	4.8	5.3	129	0.73	0.72	127	2.4	3.5	94
Zn	5.25	3.04	149	6.6	6.6	151	2.0	1.1	127	4.5	8.3	107
Se	0.27	0.14	45	...	...	...	0.104	0.062	30	...	...	...
Br	3.5	2.5	94	2.5	2.8	81	0.64	0.30	35	0.68	0.51	42
Rb	1.5	1.1	56	3.3	2.4	120	0.32	0.21	20	0.91	0.68	81
Sr	0.34	0.31	33	3.0	2.0	110	0.16	0.14	22	0.96	0.68	77
Zr	0.56	0.34	32	5.3	3.6	67	0.104	0.055	12	1.44	0.92	33
Pb	3.0	2.2	149	5.8	4.2	154	1.24	0.86	127	3.8	3.0	131

periments [Artaxo et al., 1988, 1990]. Cuiabá and Alta Floresta are heavily disturbed sites, whereas the ABLE measurements were carried out in undisturbed areas.

Black carbon concentrations in the fine fraction during the dry season are high at both sites (Tables 1 and 2), in agreement with the great number of fires in this period. Wet season values are nonetheless significant (about  $700 \text{ ng m}^{-3}$  at both sites). It has been argued that this equivalent black carbon corresponds to other light-absorbing material, such as naturally emitted

humic-like substances [Andreae and Crutzen, 1997]. It is important to stress, however, that biomass burning may occur throughout the year. Wood burning is still an important energy source in developing countries [Lioussé et al., 1996] and is used as such in several regions of Brazil [Andreae, 1991]. Though wet season black carbon optical measurements should be confirmed by data from other independent methods when available, emissions from wood burning may explain the high BC concentrations observed in the wet season. Figure 3 shows the time series of the BC/FPM ratio. At Cuiabá this ratio shows a high variability, oscillating between 10 and 37%, with an average value of  $17.9 \pm 5.9\%$ . Alta Floresta BC/FPM values are approximately in the same range, but they show a smaller variability and exhibit a roughly linearly growing trend with time. This trend suggests an increase of light-absorbing material in the atmosphere of Alta Floresta throughout the sampling period. A possible explanation for the increase of black carbon could be an increase in biofuel consumption. However, this was not the case for the region around Alta Floresta. Besides black carbon, other possible light-absorbing materials in atmospheric particles are minerals such as magnetite, or naturally emitted humic-like substances [Andreae and Crutzen, 1997]. The latter are certainly emitted by the rain forest into the atmosphere of Alta Floresta, but there is no reason for a steady increase of these materials. Thus the temporal increase of the BC/FPM ratio at Alta Floresta remains unexplained and deserves further research.



**Figure 3.** Black carbon to fine particle mass concentration ratio (BC/FPM) at Cuiabá and Alta Floresta.

**4.2. Aerosol Source Identification and Source Apportionment**

The aerosol data sets were analyzed by absolute principle component analysis (APCA). For each site, the data set was divided by season (fine/wet) and by size fraction (fine/coarse). In a first step, aerosol sources for each subset were identified

**Table 3.** VARIMAX-Rotated Principal Component Loading Matrix for Alta Floresta Dry Season Samples

Var.	Dry Season						
	Fine Mode			Coarse Mode			
	Biomass Burning	Soil Dust	Comm. <sup>a</sup>	Soil Dust	Biomass Burning	Biogenic Aerosol	Comm. <sup>b</sup>
Mass	0.82	0.30	0.76	0.76	0.09	0.48	0.82
BC	0.92	0.32	0.95	...	...	...	...
Al	0.23	0.96	0.98	0.91	0.36	0.03	0.97
Si	...	...	...	0.89	0.40	0.05	0.95
P	...	...	...	-0.08	0.13	0.96	0.94
S	0.95	0.06	0.90	0.28	0.56	0.65	0.81
K	0.93	0.26	0.93	0.41	0.68	0.57	0.95
Ca	0.81	0.42	0.82	0.30	0.87	0.23	0.90
Ti	0.33	0.94	0.99	0.93	0.34	0.04	0.98
Mn	...	...	...	0.54	0.79	0.12	0.94
Fe	0.22	0.97	0.98	0.92	0.31	0.06	0.95
Zn	0.94	0.24	0.95	...	...	...	...
Pb	0.86	0.27	0.81	...	...	...	...
$\lambda^b$	5.77	3.31		4.55	2.68	1.96	

<sup>a</sup>Comm. is the communality of the variable.

<sup>b</sup> $\lambda$  is the eigenvalue of the principal component.

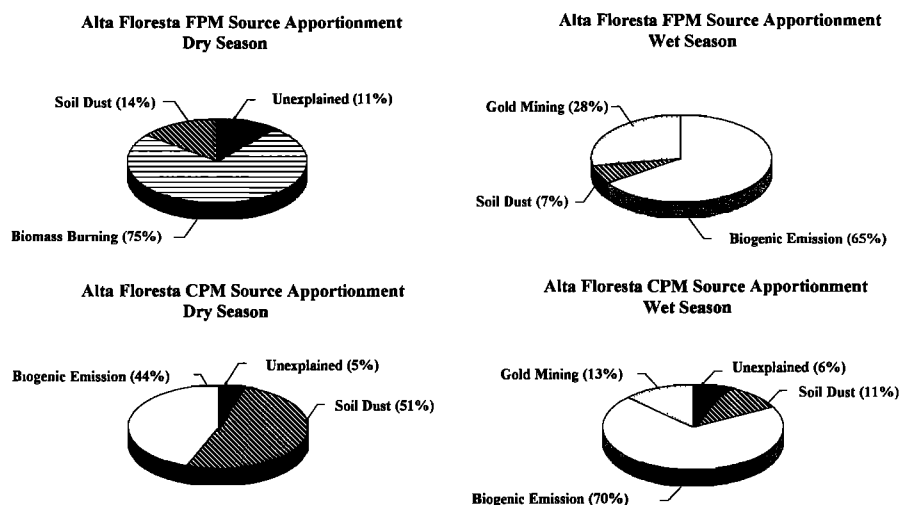
with principal component analysis (PCA), only including variables with less than 20% of the data below the detection limit (missing values). Then, APCA was used to quantitatively apportion the source contributions.

**4.2.1. Alta Floresta aerosols in the dry season.** Table 3 shows the PCA results for the Alta Floresta dry season aerosols. In the fine fraction, two components were identified, which together explain 91% of the total variance in the data set. The communalities listed in the table indicate which fraction of the variance is explained for each variable. For the fine particles, most variables have communalities higher than 0.80.

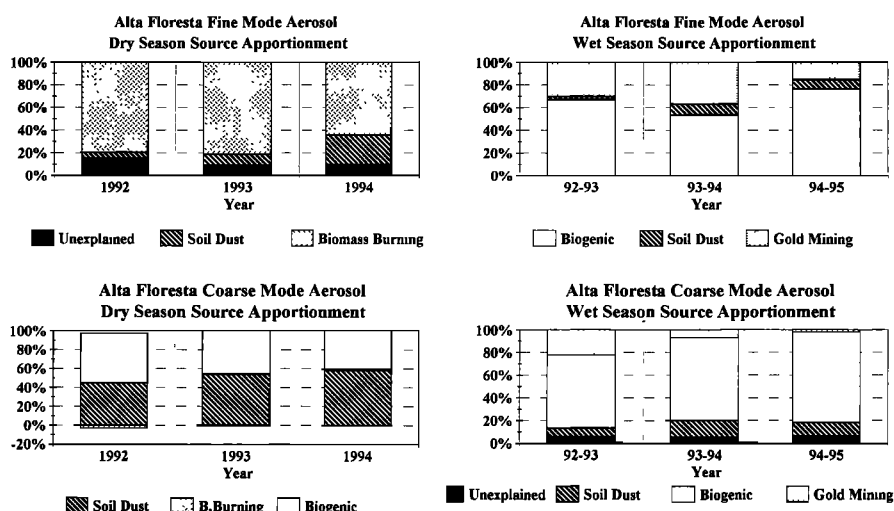
The variables S, Zn, K, BC, Pb, Mass, and Ca are associated with the first component in the fine fraction, and Fe, Al, and Ti with the second one (Table 3). The second component clearly represents soil dust resuspension, as indicated by the typical crustal elements associated with it. The first one is associated with biomass burning emissions: S, K, and Zn can be related

with both biomass burning [Artaxo *et al.*, 1993; Yamasoe, 1994; Gaudichet *et al.*, 1995; Echalar *et al.*, 1995; Maenhaut *et al.*, 1996b] and biogenic emissions [Artaxo *et al.*, 1988, 1990], but the presence of BC in this component clearly points to an association with biomass burning. Calcium has also been found in emissions of fires [Yamasoe, 1994; Gaudichet *et al.*, 1995; Echalar *et al.*, 1995; Maenhaut *et al.*, 1996b]. The Pb in the fine mode aerosol from Alta Floresta is probably associated with the gold mining activity [Hacon *et al.*, 1995; Fernandes, 1997; Gerab *et al.*, 1998]. This is confirmed by the significant correlation between Pb and Au (correlation coefficient = 0.74; significance level = 0.001).

Figure 4 shows the average APCA source apportionment for the fine and coarse Alta Floresta aerosol during the dry season. The model explained 89% of the Fine Particle Mass concentration (FPM). The results show the importance of the biomass burning emissions. APCA attributed 75% of the FPM to bio-



**Figure 4.** Source apportionment for Alta Floresta fine (FPM) and coarse (CPM) mode aerosol in terms of average percentage of the observed aerosol mass. Average results for the entire sampling period.



**Figure 5.** Source apportionment for Alta Floresta fine and coarse mode aerosol in terms of average percentage of the observed aerosol mass. Results separated per year.

mass burning emissions and 14% to soil dust (Figure 4). Though the estimated biomass burning contribution may include some contributions from gold mining and from biogenic aerosol, it is certainly the main source of fine particles during the dry season. As far as the trace elements during the dry season are concerned, biomass burning was the main source of fine particulate S, with more than 80% of the fine S, on average, attributed to it. APCA allowed us also to estimate the seasonal source contributions for each year, as shown in Figure 5 for Alta Floresta. It can be seen that the biomass burning contributions to FPM are similar in the 1992 and 1993 dry seasons (about 80% of FPM) and decrease to about 60% of FPM in the 1994 dry season.

In the coarse fraction of the Alta Floresta dry season aerosol, three components were identified (Table 3); together they explained 92% of the total data variance. As seen in Table 3, the first component has high loadings for typical crustal elements (Al, Si, Ti, and Fe); it clearly represents soil dust resuspension. Calcium, potassium, and sulfur are associated with

the second component. The presence of K and S suggests biomass burning emissions, which contain also Ca and Mn [Yamasoe, 1994; Echalar et al., 1995]. Besides, the second component was significantly correlated with fine BC (correlation coefficient = 0.62, significance level = 0.001); this allows a clear identification as emissions from biomass burning. The third component is highly loaded with P, S, and K and was attributed to biogenic emissions.

Figure 4 shows the APCA results for the Alta Floresta dry season coarse mode aerosol (CPM). The contribution from biomass burning to the CPM is very small (<0.01%); CPM is dominated almost equally by biogenic and crustal aerosol. The yearly seasonal estimates show, however, a slight increase of soil dust importance since 1992 (Figure 5). With regard to the trace elements, one quarter of the coarse S average concentration was attributed to soil dust particles, and three quarters to biogenic emissions. This latter source alone explained about 93% of the coarse P average concentration.

**4.2.2. Alta Floresta aerosol in the wet season.** Table 4

**Table 4.** VARIMAX-Rotated Principal Component Loading Matrix for Alta Floresta Wet Season Samples

Var.	Wet Season							
	Fine Mode				Coarse Mode			
	Biogenic Aerosol	Soil Dust	Gold Mining	Comm. <sup>a</sup>	Soil Dust	Biogenic Aerosol	Gold Mining	Comm. <sup>a</sup>
Mass	0.85	0.12	0.39	0.88	0.22	0.83	0.40	0.90
BC	0.85	0.35	0.14	0.87	...	...	...	...
Al	0.14	0.93	0.02	0.88	0.98	0.10	0.04	0.96
Si	...	...	...	...	0.90	0.27	0.14	0.91
P	...	...	...	...	0.03	0.95	0.22	0.95
S	0.85	-0.11	0.33	0.84	-0.02	0.66	0.63	0.83
K	0.93	0.20	0.13	0.92	0.16	0.94	0.14	0.93
Ca	0.86	0.23	0.13	0.82	0.47	0.79	0.11	0.86
Ti	0.24	0.92	0.15	0.92	0.98	0.09	0.07	0.97
Mn	...	...	...	...	0.14	0.30	0.90	0.92
Fe	0.18	0.92	0.20	0.93	0.96	0.14	0.04	0.95
Zn	0.85	0.35	-0.11	0.85	...	...	...	...
Pb	0.27	0.26	0.90	0.94	...	...	...	...
$\lambda^b$	4.67	2.99	1.19	...	3.97	3.73	1.48	...

**Table 5.** VARIMAX-Rotated Principal Component Loading Matrix for Cuiabá Dry Season Samples

Variable	Dry Season								
	Fine Mode					Coarse Mode			
	Soil Dust	Biomass Burning	Urban 1	Urban 2	Comm. <sup>a</sup>	Soil Dust	Biomass Burning	Biogenic Aerosol	Comm. <sup>a</sup>
Mass	0.28	0.92	0.09	0.03	0.93	0.67	0.55	0.45	0.95
BC	0.40	0.85	0.11	0.19	0.92	...	...	...	...
Al	0.91	0.26	0.17	0.01	0.93	0.85	0.47	0.14	0.96
Si	0.82	0.13	-0.02	0.32	0.80	0.79	0.48	0.27	0.93
P	...	...	...	...	...	0.16	0.09	0.92	0.89
S	0.09	0.90	0.10	0.14	0.84	0.36	0.46	0.72	0.86
Cl	...	...	...	...	...	0.21	0.90	0.16	0.89
K	0.31	0.92	0.14	0.05	0.96	0.64	0.52	0.51	0.94
Ca	0.67	0.51	0.12	0.16	0.75	0.64	0.49	0.31	0.75
Ti	0.88	0.33	0.23	0.05	0.93	0.85	0.40	0.26	0.95
Mn	0.68	0.44	0.38	0.07	0.80	0.60	0.59	0.41	0.88
Fe	0.91	0.23	0.20	0.10	0.93	0.91	0.32	0.16	0.95
Cu	0.20	0.20	0.15	0.93	0.97	...	...	...	...
Zn	0.42	0.69	0.41	0.20	0.86	...	...	...	...
Pb	0.25	0.19	0.92	0.14	0.97	0.86	-0.04	0.26	0.81
$\lambda^b$	4.65	4.46	1.36	1.13	...	5.46	2.92	2.38	...

presents the PCA results for the Alta Floresta wet season aerosol. In the fine fraction, three components could be identified, explaining 92% of the data variance. The first component is highly loaded with K, Ca, BC, Mass, S, and Zn. The trace elements may be associated with biomass burning but also with biogenic emissions. The presence of BC would relate this component to biomass burning. However, this source is certainly much less intense during the wet season than during the dry season (as shown by the BC seasonal averages in Table 1), whereas biogenic emissions by the forest are very significant [Artaxo *et al.*, 1990]. It was therefore concluded that the first component is mainly associated with biogenic emissions. The presence of BC indicates either that the biogenic aerosol component has light-absorbing characteristics or that a small biomass burning component is always present at Alta Floresta, even during the wet season. The second component represents clearly soil dust resuspension, as only crustal elements (Al, Ti, and Fe) are related to it. The third component is loaded mainly with Pb. This component was significantly correlated with Au (correlation coefficient = 0.70, significance level = 0.01) and seems thus associated with gold mining. This activity is not seasonal and a component associated with it should also exist during the dry season. However, during the dry season, biomass burning provides a much larger contribution to the atmospheric aerosol burden, so no separate gold-mining-related component could be distinguished.

Figure 4 shows the wet season average source contributions estimated by APCA. There is clearly a strong domination of biogenic emissions, with an average contribution of 65% to FPM. Emissions from gold mining activities contribute with about 28% to FPM, while the soil dust contribution is small (about 7% of FPM). As shown in Figure 5, gold mining contributions represented about 35% of the FPM in 1992 and 1993 and decreased to 14% in 1994. This is linked to the reduction of gold mining activities in the region [Fernandes, 1997]. As far as the trace elements are concerned, the natural emissions by the forest were the main source of particulate fine sulfur; they were responsible for about 70% of the average fine S concentration.

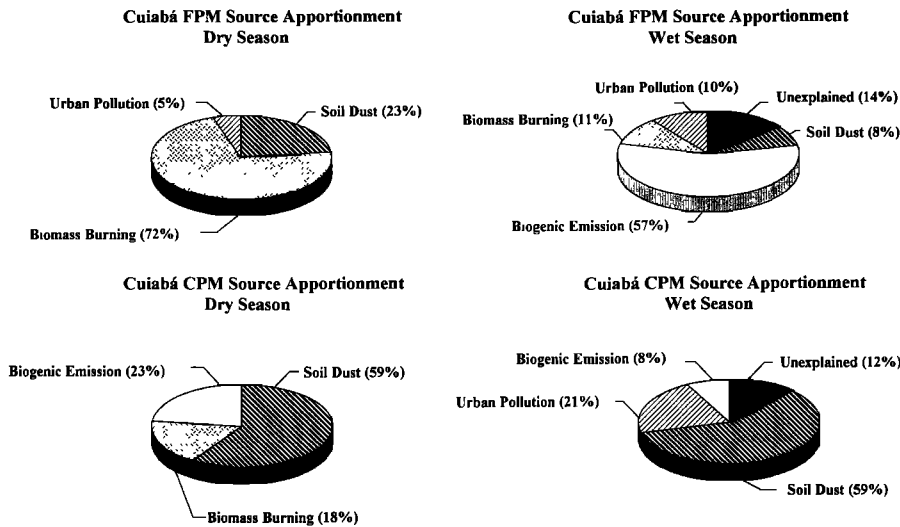
The PCA for the Alta Floresta wet season coarse mode aerosol resulted in three components (Table 4), explaining 92% of the data variance. The first component can be associated with soil dust resuspension, as crustal elements (Ti, Fe, Al, Si, and Ca) are correlated with it. The second component is highly loaded with P, K, mass, S, and Ca. The presence of P suggests biogenic emissions, which may contain also S, K, and Ca [Artaxo *et al.*, 1990]. The second component was therefore attributed to the natural biogenic emissions of coarse particles. The third component is mainly loaded with Mn and S. The latter element is present in emissions from gold mining facilities in Alta Floresta [Gerab, 1996]. Besides, this component was significantly correlated with Au (correlation coefficient = 0.67, significance level = 0.01). Thus it was attributed to emissions from gold mining.

The APCA for the wet season coarse aerosol indicated that the biogenic emissions are dominating, with an average contribution of 70% to CPM (Figure 4). Soil dust and gold mining have similar contributions, 11 and 13% of CPM, respectively. There is a clear decrease of gold mining contributions from 1992 to 1995 wet season (Figure 5), due to the reduction of gold mining activities in the Alta Floresta region. Coarse mode S was, on average, for about one quarter attributed to soil dust. Biogenic emissions were responsible for three quarters of the average coarse S concentration and for about 90% of the average coarse P concentration.

**4.2.3. Cuiabá aerosol in the dry season.** Table 5 gives the PCA results for fine and coarse mode dry season Cuiabá aerosol. In the fine fraction data set, four components were identified, explaining 89% of the data variance. The first component is highly loaded with crustal elements (Fe, Si, Ti, Ca, and Mn) and is attributed to soil dust resuspension. The second component is loaded with K, mass, S, BC, Zn, and Ca and can be associated with biomass burning, as discussed for the first component of the Alta Floresta dry season fine fraction PCA. The third component is related to Pb and Zn, and the fourth to Cu. These components are almost certainly associated with emissions from the nearby urban area of Cuiabá.

Figure 6 shows the average contribution estimated by APCA



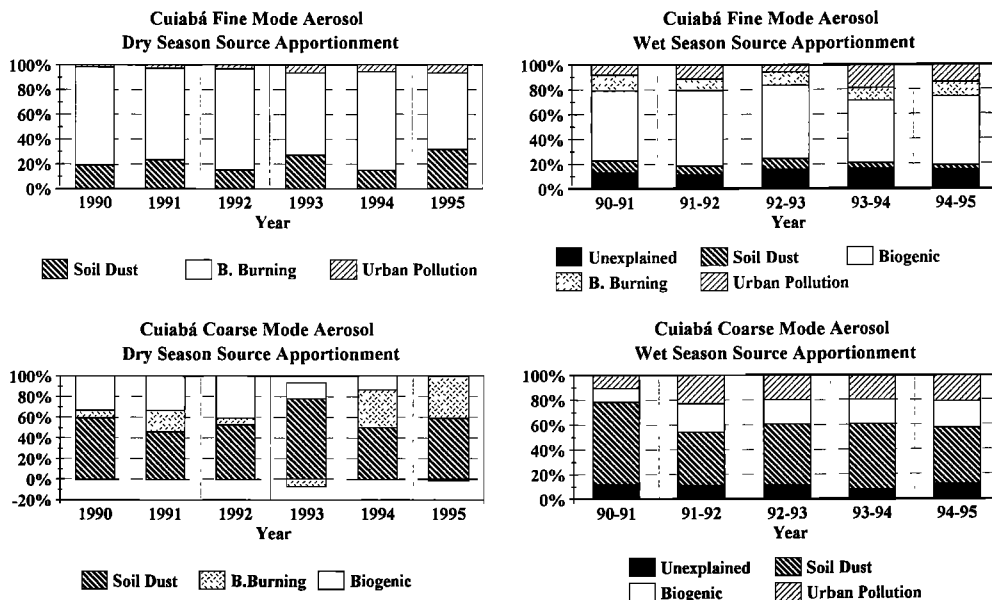


**Figure 6.** Source apportionment for Cuiabá fine (FPM) and coarse (CPM) mode aerosol in terms of average percentage of the observed aerosol mass. Average results for the entire sampling period.

for each component to the FPM. The urban components account for only some 5% of FPM. As at Alta Floresta, biomass burning is clearly the dominant source, with an average contribution of about 72% to FPM. Soil dust is responsible, on average, for about one fifth of FPM. Similarly, as at Alta Floresta, biomass burning was the main source for fine particulate sulfur (62% on average). Figure 7 shows the APCA Cuiabá source contribution estimates in each season, for each year, and each size fraction. The contribution from biomass burning to the fine mode aerosol is quite constant over the various years, while the contributions from urban pollution have been increasing with time, though they are always lower than 10% of FPM.

PCA identified three components in the coarse size fraction, explaining 90% of the data variance (Table 5). The first com-

ponent is highly loaded with crustal elements (Fe, Al, Si, and Ti) and is attributed to soil dust resuspension. Also Pb, K, Ca, mass, and Mn are correlated with this component. The presence of Pb suggests that pollution aerosol from the urban area of Cuiabá may also be included in this component. The other three elements (K, Ca, Mn) are present in soil dust, but also in biomass burning or biogenic emissions. Values attributed to this component should be considered as an upper limit for the soil dust contribution. Mass, Cl, K, and Mn are associated with the second component. This component is attributed to biomass burning emissions, but it must be noted that it may have some contribution from biogenic emissions. The third component is loaded with P, S, K, and Mn. The presence of P indicates that this component should be attributed to biogenic emissions.



**Figure 7.** Source apportionment for Cuiabá fine and coarse mode aerosol in terms of average percentage of the observed aerosol mass. Results separated per year.

**Table 6.** VARIMAX-Rotated Principal Component Loading Matrix for Cuiabá Wet Season Samples

Variable	Wet Season										
	Fine Mode						Coarse Mode				
	Soil Dust	Biogenic Aerosol	Biomass Burning	Urban 1	Urban 2	Comm. <sup>a</sup>	Soil Dust	Urban 1	Biogenic	Urban 2	Comm. <sup>a</sup>
Mass	0.25	0.86	0.18	0.18	0.14	0.89	0.89	0.24	0.11	0.25	0.92
BC	0.28	0.46	0.69	0.21	0.14	0.83	...	...	...	...	...
Al	0.92	0.16	0.10	0.28	-0.03	0.96	0.97	0.10	-0.04	0.12	0.96
Si	0.77	0.15	0.22	-0.05	0.38	0.81	0.95	0.09	0.02	0.14	0.94
P	...	...	...	...	...	...	-0.22	0.29	0.86	-0.11	0.89
S	0.17	0.85	0.26	0.11	0.18	0.86	0.55	0.03	0.70	0.20	0.83
Cl	...	...	...	...	...	...	0.17	0.90	0.29	0.03	0.92
K	0.35	0.75	0.43	0.09	0.05	0.89	0.92	0.18	0.27	0.06	0.95
Ca	0.54	0.34	0.20	0.47	0.15	0.69	0.63	0.58	0.02	0.15	0.76
Ti	0.87	0.27	0.22	0.28	0.01	0.96	0.97	0.08	-0.01	0.17	0.97
Mn	0.58	0.29	0.62	0.15	-0.03	0.82	0.88	0.28	-0.01	0.16	0.87
Fe	0.90	0.24	0.18	0.15	0.05	0.93	0.95	0.08	-0.03	0.20	0.95
Cu	0.08	0.20	0.16	0.07	0.93	0.94	...	...	...	...	...
Zn	0.14	0.31	0.79	0.24	0.26	0.86	...	...	...	...	...
Pb	0.31	0.18	0.28	0.85	0.05	0.93	0.26	0.07	0.00	0.96	0.99
$\lambda^b$	4.05	2.78	2.04	1.30	1.18	...	6.92	1.44	1.40	1.19	...

As seen in Figure 6, soil dust resuspension is the main source of the Cuiabá dry season coarse aerosol. It is, on average, responsible for about 59% of the CPM. Biogenic emissions and biomass burning contribute about 23 and 18%, respectively. The evolution from year to year of the source apportionment (Figure 7) shows that the contribution from soil dust has remained constant and close to 50% since 1990, with the exception of the 1993 dry season, when it was almost 89%. The estimated contribution from biomass burning in the coarse mode is small (up to 16% of CPM) compared to the fine fraction results, but it is higher than for the Alta Floresta dry season data set (Figure 5). Usually, more than 80% of the biomass burning particle mass is found in the fine fraction [e.g., *Artaxo et al.*, this issue]. The coarse pyrogenic aerosol at Cuiabá may predominantly originate from savanna fires nearby Cuiabá or even from local sources using wood as fuel.

**4.2.4. Cuiabá aerosol in the wet season.** Table 6 shows the PCA results for the Cuiabá aerosol in the wet season. In the fine mode, five components were identified, explaining 87% of the data variance. The first component is heavily loaded with crustal elements (Al, Fe, Ti, Si, Mn, and Ca) and is attributed to soil dust resuspension. The second component is associated with FPM, S, and K, and also BC, while the third one is correlated with Zn, BC, Mn, and K. The trace elements of these two components can be associated with biomass burning and with biogenic emissions. BC, which normally enables one to discriminate between the two types of emissions, is present in both components. During the wet season, FPM variability is controlled by biogenic aerosol rather than by biomass burning. For this reason, the second component was attributed to biogenic emissions, with a small contribution from biomass burning. Since BC is mostly correlated with the third component, this component was attributed to biomass burning; it probably represents the local utilization of wood as fuel. The fourth component is loaded with Pb and Ca, and the fifth with Cu. Both are associated with pollution emissions from the nearby Cuiabá urban area.

Figure 6 shows the APCA results for the Cuiabá fine and coarse mode aerosol during the wet season. It is evident that biogenic emissions are the dominant source of fine particles,

with a contribution of about 60% to FPM. Soil dust, urban pollution, and biomass burning are responsible for 8, 10, and 11% of FPM, respectively. However, the contribution from Cuiabá urban pollution may reach up to 20% of FPM in some years (Figure 7). About 80% of the fine sulfur was, on average, attributed to the second and third fine fraction components (biogenic emissions and biomass burning).

The PCA of the Cuiabá wet season coarse mode aerosol resulted in four components, explaining 92% of the data variance (Table 5). The first component is clearly associated with soil dust resuspension, as it is highly loaded with crustal elements (Al, Ti, Fe, K, Mn, and Si). It is also strongly loaded with CPM, and S and Ca are also correlated with it. The second component is loaded with Cl and Ca. From this result, it is unclear which source should be assigned to this component. However, as the second component was weakly, but significantly, correlated with the first urban pollution component of the fine fraction (correlation coefficient = 0.44, significance level = 0.001), it was assigned to urban pollution. The third component is correlated with P and S and is associated with biogenic emissions. The fourth component is only highly loaded with Pb and represents a second urban pollution component. The APCA results (Figure 6) show that also in the wet season, soil dust is the dominant source of coarse particles, with an average contribution of almost 60% of CPM. Urban pollution is responsible, on average, for one fifth of the coarse particle mass. Coarse S was mainly apportioned to soil dust and biogenic emissions; their average contributions were about 40 and 50%, respectively. Biogenic emissions were responsible for more than 80% of the average coarse P concentration.

**4.2.5. Cuiabá results compared to a previous source apportionment.** For the Cuiabá aerosol, a source apportionment was already performed by *Maenhaut et al.* [1996a]. These authors restricted their study to samples collected in the 14-month continuous period from July 1992 to September 1993, while the results presented here refer to a 5-year period, separated in wet and dry seasons. On the other hand, the data sets of *Maenhaut et al.* [1996a] comprised more elements than in the current work, and besides APCA, also chemical mass balance (CMB) [*Gordon*, 1988] was used for the source apportionment.

tionment. *Maenhaut et al.* [1996a] identified five components in the fine size fraction. The first component was associated with soil dust; the second one essentially represented biomass burning, but some contribution from natural biogenic emissions could not be excluded. Then, there were two urban pollution components, one characterized by V, S, and Pb and the other by As; and finally, there was a unique component associated only with Na. These results are similar to the Cuiabá fine aerosol fraction results presented in this paper: for the dry season, components associated with soil dust, biomass burning, and urban pollution. The longer time series and the separation between wet and dry season enabled us to identify the biogenic component in the wet season. Our analysis could not identify a Na component, because Na was not measured in the current work. Overall, the APCA results of *Maenhaut et al.* [1996a] agree well with ours, especially for the dry season, though the biomass burning contribution (56% of FPM) is probably underestimated by *Maenhaut et al.* [1996a], while possibly our work underestimates soil dust contributions during the wet season. In their CMB apportionment, *Maenhaut et al.* [1996a] found that biomass burning contributed 72% of FPM, which agrees well with our dry season APCA estimate.

In the coarse mode, *Maenhaut et al.* [1996a] identified four components: a soil dust component, a Ca + Mg component, an urban pollution component, and a Na unique component, counterpart of the fine Na component. The source of the Ca + Mg component was unclear, but one suggestion was that it may be related to natural biogenic emissions. Our PCA indicated the possibility of a Ca unique component in the coarse mode (Mg was not included for the PCA), but this component had an eigenvalue of only about 0.80 (in both wet and dry season), and it was therefore not retained. *Maenhaut et al.* [1996a] estimated that the contribution from soil dust to CPM was 75% (CMB result). Our APCA estimates for the soil dust contribution to CPM are 59% in both seasons. Thus both studies agree that soil dust accounts for most of the coarse aerosol mass at Cuiabá.

#### 4.3. Aerosol Elemental Mass Size Distributions

Aerosol collections with cascade impactors require much more local logistics than SFU samplings. Therefore such collections were only performed during two short periods, i.e., in August 1993 (at Alta Floresta) with a MOUDI and in August 1995 (at Cuiabá) with a PIXE cascade impactor. Results from the MOUDI samplings at Alta Floresta are presented in Figure 8. For each cascade impactor sample, the size distribution of black carbon and some trace elements are shown. Concentrations are available only for the stages with  $d_{50} \leq 1 \mu\text{m}$ . The size distributions of the species that are related to biomass burning emissions (black carbon, K, S, Cl, and Zn) exhibit a clear fine mode, which peaks at either  $0.175 \mu\text{m}$  or  $0.33 \mu\text{m}$ . This result is in agreement with airborne observations of fire plumes during the SCAR-B experiment [*Artaxo et al.*, this issue]. The size distributions of the crustal elements (Al, Si, and Fe), on the other hand, are monotonically ascending, suggesting a mode with aerodynamic diameter larger than  $1 \mu\text{m}$ . Phosphorous, an element typically associated with biogenic emissions, could not be detected. Figure 9 presents results from the PIXE cascade impactor samplings at Cuiabá. The size distributions shown were obtained by inversion of the raw impactor data and subsequent fitting of multimodal lognormal curves. The five curves for each element represent five different samples. For four of the elements shown, i.e., K, S, Cl, and

Zn, biomass burning is the main contributor to the submicrometer size fraction. The size distributions for three of these elements (K, S, Cl) exhibit a fine mode centered at about  $0.10\text{--}0.13 \mu\text{m}$  and another one centered at  $0.60\text{--}0.70 \mu\text{m}$ . For Zn there are also submicrometer modes but with peak shifts from sample to sample. A possible explanation for this behavior is that fine Zn at Cuiabá is not only associated with biomass burning emissions but also with urban pollution emissions. It is further noteworthy that the size distributions of K, Cl, and Zn exhibit a significant coarse mode, which is centered at about  $7.5 \mu\text{m}$  and is probably due to biogenic and/or soil dust particles. The crustal elements (Al, Si, and Fe) are present in a single coarse mode, centered at about  $6.5\text{--}7.8 \mu\text{m}$ . Phosphorous also exhibits only a single coarse mode, which is centered at about  $8.0 \mu\text{m}$ .

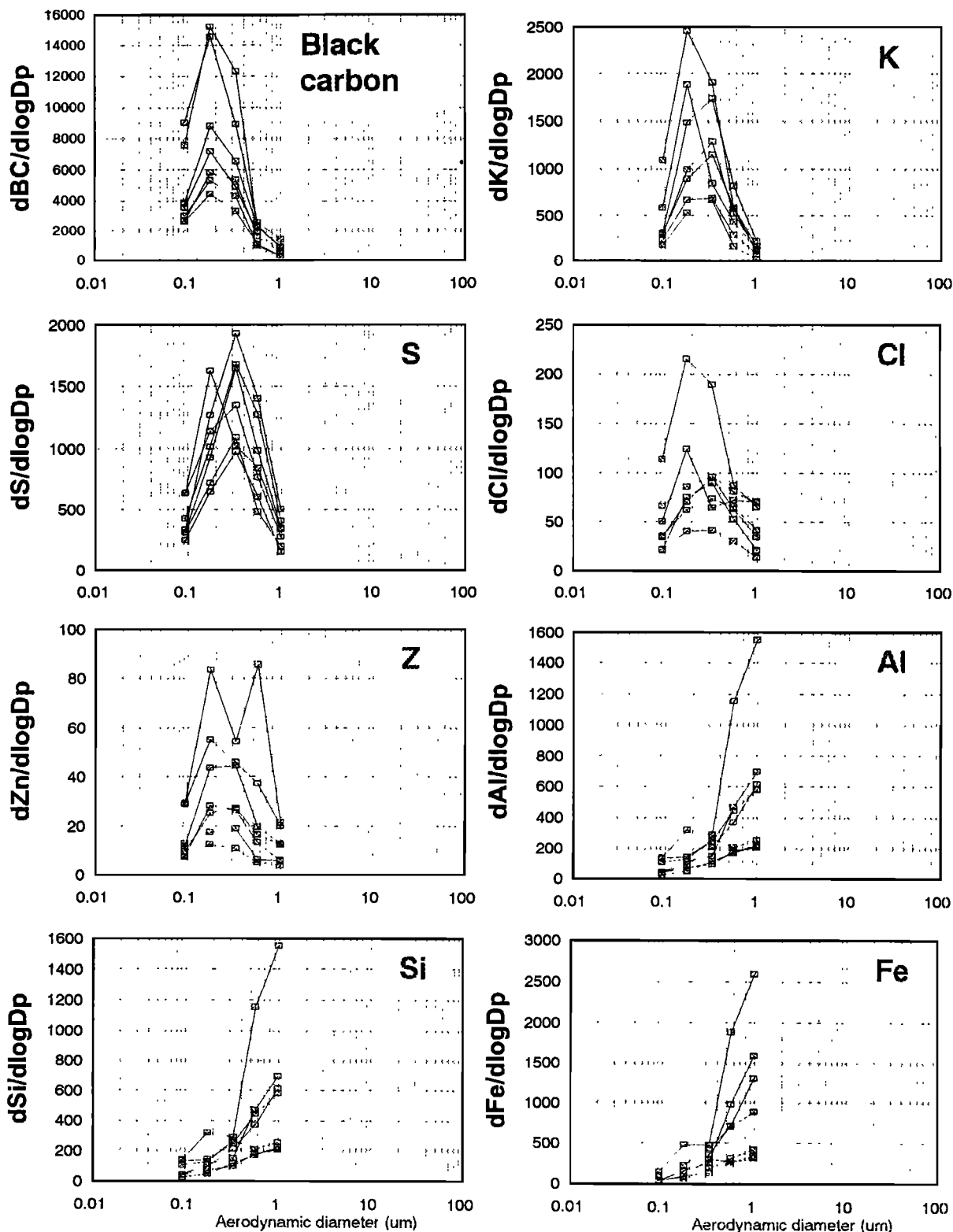
#### 4.4. Aerosol Optical Thickness (AOT)

Figure 10 shows an intercomparison between AOT at  $\lambda = 0.440 \mu\text{m}$  and FPM concentrations at ground level in Cuiabá and in Alta Floresta. At each site the measurements of the two parameters were performed in parallel. AOT measurements were made with 15 min time resolution along the daily solar cycle, whereas FPM measurements represent averages over one or more days and nights, depending on the filter loading. The AOT and FPM results for Cuiabá and Alta Floresta show a well-marked biomass burning season, with good agreement between both types of measurements. At Alta Floresta, AOT and FPM are nicely correlated with each other, indicating a well-mixed aerosol burden in the mixing layer. It is important to note that the Sun photometer measures the AOT integrated over the entire vertical atmospheric column, while FPM is measured exclusively at ground level. The good agreement between FPM and AOT throughout the sampling period associates the aerosol ground measurements to the aerosol load of the whole atmospheric column and suggests that these local measurements reflect the situation of a larger scale.

During the biomass burning season the AOT values ( $\lambda = 0.440 \mu\text{m}$ ) at Cuiabá were up to 2.5, while even higher values (up to 4.0) were observed at Alta Floresta. At both sites the dry season AOT values were much higher than the usual background values of the wet season (which were below 0.3). The dry season values of AOT are significantly important in terms of the radiative balance of the atmosphere; they suggest a significant backscattering by aerosols of solar radiation back to space and also a possible influence on the spectral redistribution of energy at the surface. During the dry season, biomass burning particles constitute the main fraction of fine mode aerosols in the region. *Reid et al.* [this issue] found that the single-scattering albedo of biomass burning aerosols is about 0.82; that is, they absorb 18% of the incident radiation. This fact, together with the observed high values of optical depth, suggests that there may be a possibly significant radiative heating of the atmosphere by biomass burning aerosols over the Amazon Basin during the dry season.

### 5. Summary and Conclusions

Reflecting the seasonal pattern of rainfall in the region, the atmospheric concentrations of inhalable particles in the Amazon Basin exhibit a marked seasonality, with maximum values up to  $300 \mu\text{g m}^{-3}$  or more during August/September every year. Trace element concentrations also show this seasonality, with higher values in the dry season.



**Figure 8.** Size distribution of black carbon and trace elements at Alta Floresta, August 1993. MOUDI samples.

Three main types of aerosol sources were identified: biomass burning, natural biogenic emission by the vegetation, and soil dust resuspension. These are the main sources identified by previous ground-based studies [Artaxo *et al.*, 1988, 1990, 1994; Maenhaut *et al.*, 1996a] and also by an airborne large-scale

source apportionment study [Artaxo *et al.*, this issue] in the region. In the current study we also identified minor contributions from local urban sources, which may account for up to 30% of the aerosol mass in the wet season of some years. A comparison with other quantitative results [Maenhaut *et al.*,

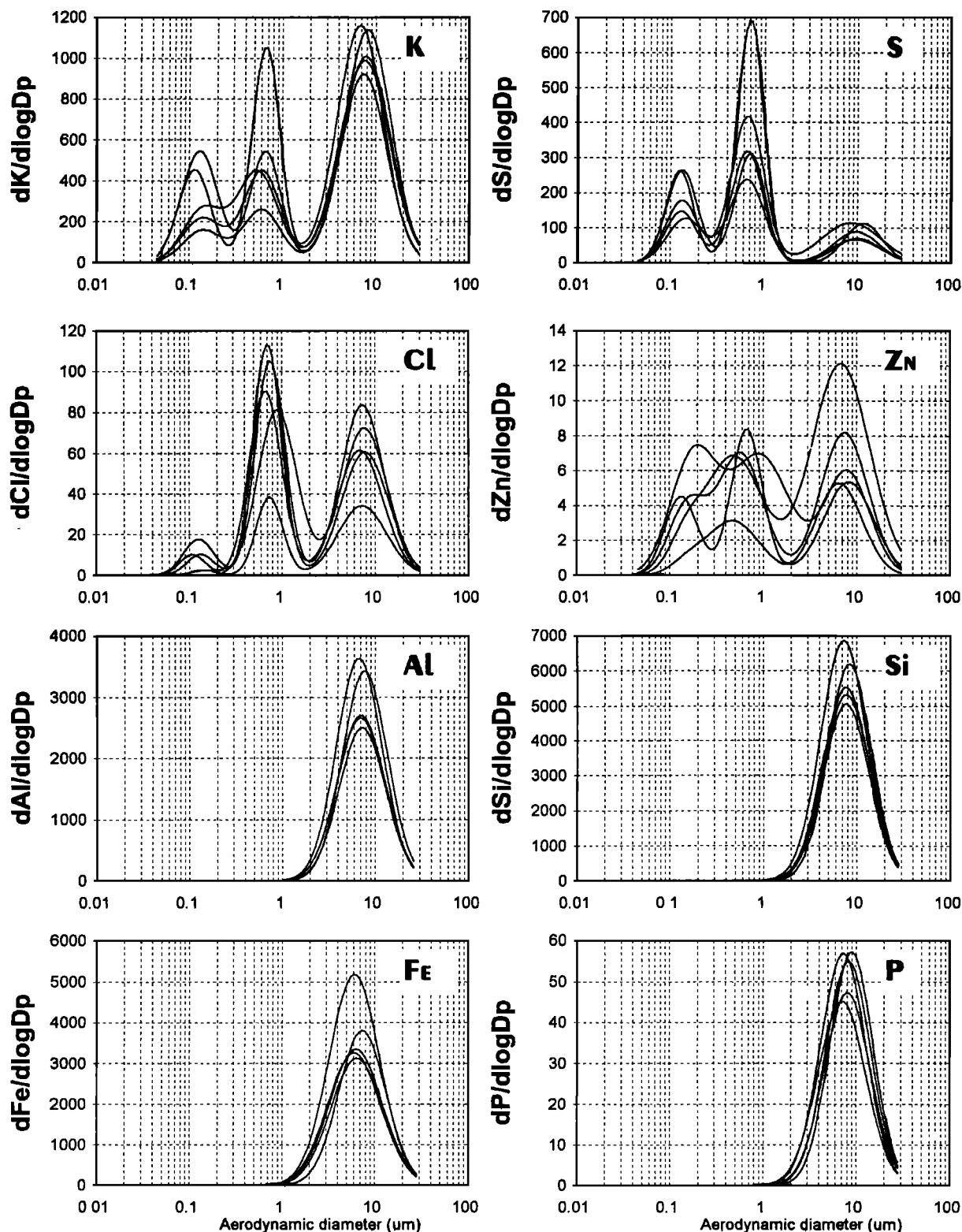
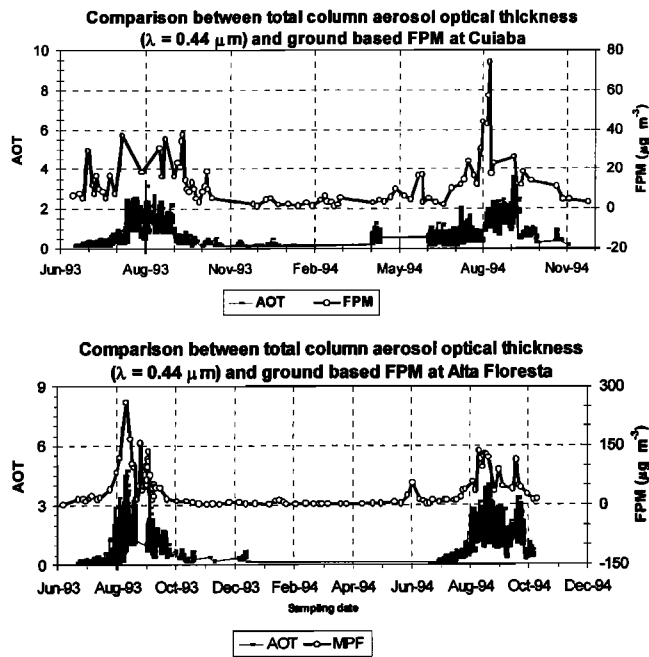


Figure 9. Size distribution of trace elements at Cuiabá, August 1995. PIXE cascade impactor data.

1996a] obtained for one of the sites studied in this work (Cuiabá) showed agreement in the estimates of the main source contributions. The longer time series and the separation of the analysis by season allowed us to identify with more detail the main aerosol sources at Cuiabá.

The contributions from each source to the atmospheric

aerosol were different in the dry and wet seasons and for the fine and coarse size fractions, and they showed also some variation from year to year. Biomass burning emissions dominate the fine mode aerosol at both sites during the dry season, which is the period of extensive fires. During this season, the coarse mode at both sites is dominated by soil dust particles.



**Figure 10.** Comparison between aerosol optical thickness at  $\lambda = 0.440 \mu\text{m}$  and fine particle mass (FPM) concentration at ground level in Cuiabá and Alta Floresta.

Biogenic aerosols are the dominant particle type in both size fractions during the wet season, except for the Cuiabá coarse aerosol, where soil dust predominates. The biogenic aerosols are the result of natural emissions by the vegetation. Both fine and coarse particulate sulfur originated mainly from biomass burning and natural biogenic emissions. Phosphorous was mainly present in the coarse mode and originated in most cases from the biogenic source. Internal recycling should thus be important for the supply of this nutrient in the ecosystems of the Amazon Basin.

The cascade impactor results for the dry season were consistent with the picture of aerosol sources obtained from bulk analysis. The crustal elements were present in a single coarse mode, while the elements with important contributions from biomass burning exhibited clear submicrometer modes.

Aerosol optical thickness (AOT) values showed a temporal evolution similar to the evolution of the fine particle mass concentrations measured in parallel. AOT values during dry season were significantly high to have an effect on the atmospheric radiative balance. The high values suggest significant backscattering of solar radiation by aerosols and possibly also a radiative heating of the atmosphere, due to the absorbing properties of the biomass burning particles.

**Acknowledgments.** We would like to thank José Roberto Chagas, Saturnino José da Silva Filho, Edilson Bernardino de Andrade, and INPE for support during the sampling program in the Amazon Basin. We thank Alcides C. Ribeiro, Ana L. Loureiro, and Tarsis Germano for assistance during sampling and PIXE analysis. This work was financed through grant 90/2950-2 and 96/2672-9 from FAPESP-Fundação de Amparo à Pesquisa do Estado de São Paulo. F. Echalar acknowledges a postdoctoral grant from CNPq (process 150048/96-3). W. Maenhaut acknowledges the financial support from the Belgian State—Prime Minister's Service—Federal Office for Scientific, Technical and Cultural Affairs (OSTC) and from the "Fonds voor Wetenschappelijk Onderzoek—Vlaanderen."

## References

- Andreae, M. O., Soot carbon and excess fine potassium: Long-range transport of combustion derived aerosols, *Science*, **230**, 1148–1151, 1983.
- Andreae, M. O., Biomass burning: Its history, use and distribution and its impact on environmental quality and global climate, in *Global Biomass Burning: Atmospheric, Climatic and Biospheric Implications*, edited by J. S. Levine, pp. 3–21, MIT Press, Cambridge, Mass., 1991.
- Andreae, M. O., and P. J. Crutzen, Atmospheric aerosols: Biogeochemical sources and role in atmospheric chemistry, *Science*, **276**, 1052–1058, 1997.
- Andreae, M. O., T. W. Andreae, R. J. Ferek, and H. Raemdonck, Long-range transport of soot carbon in the marine atmosphere, *Sci. Total Environ.*, **36**, 73–80, 1984.
- Andreae, M. O., et al., Biomass burning emissions and associated haze layers over Amazonia, *J. Geophys. Res.*, **93**, 1509–1527, 1988.
- Artaxo, P., and H.-C. Hansson, Size distribution of biogenic aerosol particles from the Amazon Basin, *Atmos. Environ.*, **29**, 393–402, 1995.
- Artaxo, P., and C. Orsini, The emission of aerosol by plants revealed by three receptor models, in *Aerosols: Formation and Reactivity*, edited by G. Israel, pp. 148–151, Pergamon, New York, 1986.
- Artaxo, P., and C. Orsini, PIXE and receptor models applied to remote aerosol source apportionment in Brazil, *Nucl. Instrum. Methods Phys. Res.*, **B22**, 259–263, 1987.
- Artaxo, P., H. Storms, F. Bruynseels, R. Van Grieken, and W. Maenhaut, Composition and sources of aerosols from the Amazon Basin, *J. Geophys. Res.*, **93**, 1605–1615, 1988.
- Artaxo, P., W. Maenhaut, H. Storms, and R. Van Grieken, Aerosol characteristics and sources for the Amazon Basin during the wet season, *J. Geophys. Res.*, **95**, 16,971–16,985, 1990.
- Artaxo, P., M. Yamasoe, J. V. Martins, S. Kocinas, S. Carvalho, and W. Maenhaut, Case study of atmospheric measurements in Brazil: Aerosol emissions from Amazon Basin biomass burning, in *Fire in the Environment: The Ecological, Atmospheric, and Climatic Importance of Vegetation Fires*, edited by P. J. Crutzen and J.-G. Goldammer, pp. 139–158, John Wiley, New York, 1993.
- Artaxo, P., F. Gerab, M. A. Yamasoe, and J. V. Martins, Fine mode aerosol composition in three long-term atmospheric monitoring sampling stations in the Amazon Basin, *J. Geophys. Res.*, **99**, 22,857–22,868, 1994.
- Artaxo, P., E. T. Fernandes, J. V. Martins, M. A. Yamasoe, P. V. Hobbs, W. Maenhaut, K. M. Longo, and A. Castanho, Large-scale aerosol source apportionment in Amazonia, *J. Geophys. Res.*, this issue.
- Bachmeier, A. S., and H. E. Fuelberg, A meteorological overview of the TRACE A period, *J. Geophys. Res.*, **101**, 23,881–23,888, 1996.
- Beauford, W., J. Barber, and A. R. Barringer, Heavy metal release from plants into the atmosphere, *Nature*, **256**, 35–37, 1975.
- Beauford, W., J. Barber, and A. R. Barringer, Release of particles containing metals from vegetation into the atmosphere, *Science*, **195**, 571–573, 1977.
- Cachier, H., P. Buat-Ménard, M. Fontugne, and J. Rancher, Source terms and source strengths of the carbonaceous aerosol in the tropics, *J. Atmos. Chem.*, **3**, 469–489, 1985.
- Cachier, H., J. Ducret, M. P. Brémond, V. Yoboué, J. P. Lacaux, A. Gaudichet, and J. Baudet, Biomass burning in a savanna region of the Ivory Coast, in *Global Biomass Burning: Atmospheric, Climatic and Biospheric Implications*, edited by J. S. Levine, pp. 174–180, MIT Press, Cambridge, Mass., 1991.
- Cachier, H., C. Lioussé, P. Buat-Ménard, and A. Gaudichet, Particulate content of savanna fire emissions, *J. Atmos. Chem.*, **22**, 123–148, 1995.
- Crozat, G., Sur l'émission d'un aérosol riche en potassium par la forêt équatoriale, *Tellus*, **31**, 52–57, 1979.
- Crutzen, P., and M. O. Andreae, Biomass burning in the tropics: Impact on atmospheric chemistry and biogeochemical cycles, *Science*, **250**, 1669–1678, 1990.
- Crutzen, P. J., and J. G. Goldammer (Eds.), *Fire in the Environment: The Ecological, Atmospheric and Climatic Importance of Vegetation Fires*, John Wiley, New York, 1993.
- Delmas, R., and J. Servant, The origins of sulfur compounds in the atmosphere of a zone of high productivity (Gulf of Guinea), *J. Geophys. Res.*, **87**, 11,019–11,026, 1982.
- Delmas, R., and J. Servant, Atmospheric balance of sulfur above an equatorial forest, *Tellus, Ser. B*, **35**, 110–120, 1983.

- Dinh, P. V., J. P. Lacaux, and R. Serpouly, Cloud-active particles from African savanna combustion experiments, *Atmos. Res.*, **31**, 41–58, 1994.
- Echalar, F., A. Gaudichet, H. Cachier, and P. Artaxo, Aerosol emissions by tropical forest and savanna biomass burning: Characteristic trace elements and fluxes, *Geophys. Res. Lett.*, **22**, 3039–3042, 1995.
- Fearnside, P. M., Greenhouse gas contributions from deforestation in Brazilian Amazonia, in *Global Biomass Burning: Atmospheric, Climatic and Biospheric Implications*, edited by J. S. Levine, pp. 92–105, MIT Press, Cambridge, Mass., 1991.
- Fernandes, E., *Mercúrio Atmosférico na Amazônia e Estudo de Caso em Alta Floresta*, (in Portuguese), M.S. thesis, Inst. of Phys., Univ. of São Paulo, São Paulo, Brazil, 1997.
- Ferry, J. F., and H. S. Ward, *Fundamentals of Plant Physiology*, Macmillan, Indianapolis, Indiana, 1959.
- Garstang, M., et al., Trace gas exchanges and convective transports over the Amazonian rain forest, *J. Geophys. Res.*, **93**, 1528–1550, 1988.
- Gash, J. H. C., C. A. Nobre, J. M. Hoberts, and R. L. Victoria (Eds.), *Amazon Deforestation and Climate*, John Wiley, New York, 1996.
- Gaudichet, A., F. Echalar, B. Chatenet, J. P. Quisefit, G. Malingre, H. Cachier, P. Buat-Ménard, P. Artaxo, and W. Maenhaut, Trace elements in tropical African savanna biomass burning aerosols, *J. Atmos. Chem.*, **22**, 19–39, 1995.
- Gerab, F., *Técnicas Analíticas Nucleares Aplicadas à Medida em Larga Escala de Aerossóis Atmosféricos na Região Amazônica*, (in Portuguese), Ph.D. thesis, Inst. of Phys., Univ. of São Paulo, São Paulo, Brazil, 1996.
- Gerab, F., P. Artaxo, E. Swietlicki, and J. Pallon, Scanning proton microprobe applied to analysis of individual aerosol particles from Amazon Basin, *Nucl. Instrum. Methods Phys. Res., Sect. B*, in press, 1998.
- Gordon, G. E., Receptor models, *Environ. Sci. Technol.*, **22**, 1132–1142, 1988.
- Hacon, S., P. Artaxo, F. Gerab, M. A. Yamasoe, R. C. Campos, L. F. Conti, and L. D. Lacerda, Atmospheric mercury and trace elements in the region of Alta Floresta in the Amazon Basin, *Water Air Soil Pollut.*, **80**, 273–283, 1995.
- Harman, H., *Modern Factor Analysis*, Univ. of Chicago Press, Chicago, Ill., 1976.
- Harriss, R. C., et al., The Amazon Boundary Layer Experiment: Wet season 1987, *J. Geophys. Res.*, **95**, 16,721–16,736, 1990.
- Havráněk, V., W. Maenhaut, G. Ducastel, and J. E. Hanssen, Mass size distributions for atmospheric trace elements at the Zeppelin background station in Ny Ålesund, Spitsbergen, *Nucl. Instrum. Methods Phys. Res., Sect. B*, **109/110**, 465–470, 1996.
- Henry, R. C., Multivariate receptor models, in *Receptor Modeling for Air Quality Management*, edited by P. K. Hopke, pp. 117–147, Elsevier, New York, 1991.
- Hillamo, R. E., V.-M. Kerminen, W. Maenhaut, J.-L. Jaffrezo, S. Balachandran, and C. I. Davidson, Size distributions of atmospheric trace elements at Dye 3, Greenland, I, Distribution characteristics and dry deposition velocities, *Atmos. Environ.*, **27A**, 2787–2802, 1993.
- Hobbs, P. V., J. S. Reid, R. A. Kotchenruther, R. J. Ferek, and R. Weiss, Direct radiative forcing by smoke from biomass burning, *Science*, **275**, 1776–1778, 1997.
- Holben, B. N., Y. J. Kaufman, D. Tanré, and D. Ward, Optical properties of aerosol from biomass burning in the tropics: BASE-A, in *Global Biomass Burning*, edited by J. S. Levine, pp. 403–411, MIT Press, Cambridge, Mass., 1991.
- Holben, B. N., T. F. Eck, A. Pereira, and I. Slutsker, Effect of dry season biomass burning on Amazon Basin aerosol concentrations and optical properties, 1992–1994, *J. Geophys. Res.*, **101**, 19,455–19,464, 1996a.
- Holben, B. N., A. Setzer, T. F. Eck, A. Pereira, and I. Slutsker, Effect of dry-season biomass burning on Amazon Basin aerosol concentrations and optical properties, 1992–1994, *J. Geophys. Res.*, **101**, 19,465–19,481, 1996b.
- Holben, B. N., et al., Automatic sun and sky scanning radiometer system for network aerosol monitoring, *Remote Sens. Environ.*, in press, 1996c.
- Hopke, P. K., Y. Xie, T. Raunemaa, S. Biegalski, S. Landsberger, W. Maenhaut, P. Artaxo, and D. Cohen, Characterization of the Gent stacked filter unit PM<sub>10</sub> sampler, *Aerosol Sci. Technol.*, **27**, 726–735, 1997.
- Ito, K., T. J. Kneip, and P. J. Liroy, The effects of number of samples and random errors on the factor analysis/multiple regression (FA/MR) receptor modelling technique, *Atmos. Environ.*, **20**, 1433–1440, 1986.
- Johansson, S. A. E., J. L. Campbell, and K. G. Malmqvist (Eds.), *Particle-Induced X-Ray Emission (PIXE) Spectrometry*, John Wiley, New York, 1995.
- John, W., S. Hering, G. Reischel, and G. Sasaki, Characteristics of Nuclepore filters with large pore size, II, Filtration properties, *Atmos. Environ.*, **17**, 373–382, 1983.
- Kaufman, Y. J., and R. S. Fraser, Confirmation of the smoke particles effects on clouds and climate, *Science*, in press, 1997.
- Kaufman, Y. J., and D. Tanré, Variations in cloud supersaturation and the aerosol indirect effect on climate, *Nature*, **369**, 45–48, 1994.
- Kaufman, Y. J., C. J. Tucker, and I. Fung, Remote sensing of biomass burning in the tropics, *J. Geophys. Res.*, **95**, 9927–9939, 1990.
- Kaufman, Y. J., et al., Smoke, Clouds, and Radiation–Brazil (SCAR-B) experiment, *J. Geophys. Res.*, this issue.
- Lawson, D. R., and J. W. Winchester, Atmospheric sulfur aerosol concentrations and characteristics from the South American continent, *Science*, **205**, 1267–1269, 1979.
- Leslie, A. C. D., Aerosol emissions from forest and grassland burnings in the southern Amazon Basin and central Brazil, *Nucl. Instrum. Methods*, **181**, 345–351, 1981.
- Liousse, C., J. E. Penner, J. J. Walton, H. Eddleman, C. Chuang, and H. Cachier, Modeling biomass burning aerosols, in *Biomass Burning and Global Change*, edited by J. S. Levine, pp. 492–508, MIT Press, Cambridge, Mass., 1996.
- Lober, J. M., and J. Warnatz, Emissions from the combustion process in vegetation, in *Fire in the Environment: The Ecological, Atmospheric, and Climatic Importance of Vegetation Fires*, edited by P. J. Crutzen and J. G. Goldammer, pp. 15–37, John Wiley, New York, 1993.
- Lodge, J. P., P. A. Machado, J. B. Pate, D. C. Sheesley, and A. F. Wartburg, Atmospheric trace chemistry in the American humid tropics, *Tellus*, **26**, 250–259, 1974.
- Maenhaut, W., G. Koppen, and P. Artaxo, Long term atmospheric aerosol study in Cuiabá, Brazil: Multielemental composition, sources, and impacts of biomass burning, in *Biomass Burning and Global Change*, edited by Joel Levine, pp. 637–652, MIT Press, Cambridge, Mass., 1996a.
- Maenhaut, W., I. Salma, J. Cafmeyer, H. J. Annegarn, and M. O. Andreae, Regional atmospheric aerosol composition and sources in the eastern Transvaal, South Africa, and impact of biomass burning, *J. Geophys. Res.*, **101**, 23,631–23,650, 1996b.
- Maki, L. R., and K. J. Willoughby, Bacteria as biogenic source of freezing nuclei, *J. Appl. Meteorol.*, **17**, 1049–1053, 1978.
- Marenco, A., and J. C. Delauney, Experimental evidence of natural sources of CO from measurements in the troposphere, *J. Geophys. Res.*, **85**, 5599–5613, 1980.
- Nemeruyk, G. E., Migration of salts into the atmosphere during transpiration, *Sov. Plant Physiol.*, **17**, 560–566, 1970.
- Orsini, C., P. Artaxo, and M. Tabacnick, Preliminary data on atmospheric aerosols of the Amazon Basin, *Atmos. Environ.*, **16**, 2177–2181, 1982.
- Parker, R. D., G. H. Buzzard, T. G. Dzubay, and J. P. Bell, A two stage respirable aerosol sampler using Nuclepore filters in series, *Atmos. Environ.*, **11**, 617–621, 1977.
- Reid, J. S., P. V. Hobbs, C. Liousse, J. V. Martins, R. E. Weiss, and T. F. Eck, Comparisons of techniques for measuring shortwave absorption and the black carbon content of aerosols from biomass burning in Brazil, *J. Geophys. Res.*, this issue (a).
- Reid, J. S., P. V. Hobbs, R. J. Ferek, D. R. Blake, J. V. Martins, M. R. Dunlap, and C. Liousse, Physical, chemical, and optical properties of regional hazes dominated by smoke in Brazil, *J. Geophys. Res.*, this issue (b).
- Rogers, C. F., J. G. Hudson, B. Zielinska, R. L. Tanner, J. Hallet, and J. G. Watson, Cloud condensation nuclei from biomass burning, in *Global Biomass Burning: Atmospheric, Climatic and Biospheric Implications*, edited by J. S. Levine, pp. 431–440, MIT Press, Cambridge, Mass., 1991.
- Salati, E., and P. B. Vose, Amazon Basin: A system in equilibrium, *Science*, **225**, 129–138, 1984.
- Saxena, P., L. M. Hildemann, P. McMurry, and J. Seinfeld, Organics alter hygroscopic behavior of atmospheric particles, *J. Geophys. Res.*, **100**, 18,755–18,770, 1995.

- Schnell, R. C., Kenyan leaf litter: A source of ice nuclei, *Tellus*, *34*, 92–95, 1982.
- Thurston, G. D., and J. D. Spengler, A quantitative assesment of source contributions to inhalable particulate matter pollution in metropolitan Boston, *Atmos. Environ.*, *19*, 9–25, 1985.
- Vitousek, P. M., and R. L. Sanford, Nutrient cycling in moist tropical forest, *Annu. Rev. Ecol. Syst.*, *17*, 137–167, 1986.
- Ward, D. E., A. W. Setzer, Y. J. Kaufman, and R. A. Rasmussen, Characteristics of smoke emissions from biomass fires of the Amazon region—BASE-A experiment, in *Global Biomass Burning: Atmospheric, Climatic and Biospheric Implications*, edited by J. S. Levine, pp. 394–402, MIT Press, Cambridge, Mass., 1991.
- Ward, D. E., R. Susott, J. Kauffman, R. Babbitt, B. N. Holben, Y. J. Kaufman, A. Setzer, R. Rasmussen, D. Cumming, and B. Dias, Emissions and burning characteristics of biomass fires for cerrado and tropical forest regions of Brazil—BASE-B experiment, *J. Geophys. Res.*, *97*, 14,601–14,619, 1992.
- Yamasoe, M. A., *Estudo da composição elementar e iônica de aerossóis emitidos em queimadas na Amazônia*, (in Portuguese), M.S. thesis, Inst. of Phys., Univ. of Sao Paulo, Brazil, 1994.
- P. Artaxo (corresponding author), F. Echalar, F. Gerab, J. V. Martins, and M. Yamasoe, Instituto de Física, Universidade de São Paulo, Caixa Postal 66318, CEP 05315-970, São Paulo, Brazil. (e-mail: artaxo@if.usp.br.)
- W. Maenhaut, Institute for Nuclear Sciences, University of Gent, Proeftuinstraat 86, B-9000 Gent, Belgium. (e-mail: maenhaut@inwchem.rug.ac.be.)
- B. Holben, NASA Goddard Space Flight Center, Greenbelt, MD 20771. (e-mail: brent@kratmos.gsfc.nasa.gov.)

(Received November 10, 1997; revised May 7, 1998;  
accepted May 13, 1998.)