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TRACE ELEMENTS IN ATMOSPHERIC AEROSOLS FROM BACKGROUND REGIONS AND BIOMASS BURNING FROM THE AMAZON BASIN

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Abstract

TRACE ELEMENTS IN ATMOSPHERIC AEROSOLS FROM BACKGROUND REGIONS AND BIOMASS BURNING FROM THE AMAZON BASIN.

Aerosol particles from the tropical rain forest and from savannah biomass burning were collected in several experiments in the Amazon Basin. The size distribution of atmospheric trace elements was measured under both background and biomass burning conditions. Sampling from aircraft was performed over a large area of the Amazon Basin in August/September 1991. The aerosol mass concentration, black carbon and trace element concentrations were determined for fine and coarse mode aerosol particles. Particle induced X ray emission (PIXE) was used to measure the concentrations of up to 22 elements: Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Rb, Sr, Zr and Pb. Irradiation was carried out at the Van der Graaff accelerator at Florida State University, USA, and at the cyclotron of the University of Ghent, Belgium. A new dedicated PIXE/Rutherford backscattering spectroscopy (RBS) facility at the University of São Paulo is now being used for PIXE analysis. The natural biogenic aerosol emission from the Amazon Basin shows a component with K, P, S, Rb and Zn as characteristic elements for both fine and coarse mode aerosols. Soil dust is the second component, with the presence of Al, Si, Ti, Mn and Fe. There is also the component of aerosols transported over a long range from the Sahara Desert into the interior of the Amazon Basin. During the dry season, when most of the biomass burning occurs, the concentration of inhalable particles exceeds 300 $\mu g/m^3$ in regions far from the direct influence of emissions from biomass burning. Large amounts of fine particles are injected into the atmosphere, where they can travel over long distances. These particles are rich in K, P, S, Ca, Mg, Cl, Si, Zn, Rb, Sr, Zr and other trace elements. The emissions of trace elements and heavy metals into the global atmosphere owing to biomass burning are very significant. For Cr, global biomass burning emits about 5.6×10^9 g/year, for Cu, 1.6×10^9 g/year and for Zn 5.2×10^9 g/year. These values account for 18, 5 and 3.5%, respectively, of the

global anthropogenic inputs. These emissions of heavy metals by biomass burning are currently not considered in global atmospheric heavy metal inventories. Several essential nutrients, such as P, K, S and others, are transported into the atmosphere as a result of biomass burning processes. Most of the particles are water soluble and can be active as cloud condensation nuclei, with the potential to change the cloud formation mechanisms in the Amazon Basin and other regions of the planet.

1. NATURAL EMISSION OF GASES AND AEROSOL PARTICLES BY THE TROPICAL RAIN FOREST

The Earth's atmosphere is a vital natural resource that, until recently, appeared to be unaffected by human activities, except on a local scale. However, it has become clear that worldwide anthropogenic activities have an impact on the global atmosphere. It is important to increase our knowledge of the chemical processes that determine the composition of the atmosphere in background areas, and to understand the impact of tropical forest biomass burning in the global atmosphere.

In order to assess the role of the tropical atmosphere in global atmospheric changes, it is necessary to identify and quantify important physical and chemical processes in the generation, transformation and deposition of aerosol particles in tropical areas. It is also important to obtain a better understanding of natural gases and aerosol sources with regard to their identification, characteristics and strength. The aim is to be able to understand the natural chemistry in the atmosphere on a global scale, thus providing the possibility of identifying and possibly quantifying the influence of global anthropogenic emissions in background areas. The World Climate Research Programme (WCRP), the International Geosphere Biosphere Programme (ICBP) and the International Global Atmospheric Chemistry Programme (IGAC) have recognized that the tropical regions play a key role in changes in the atmospheric composition and should receive top priority in research aimed at studying the background atmospheric composition and global change. The tropical rain forest is a globally important ecosystem, with large emissions of biogenic gases and particles. The Amazon Basin, with about 4 million km^2, plays an important role in emissions of water vapour, gases and aerosol particles into the global atmosphere. The deposition of biogeochemically important species onto the Earth's surface plays an essential role in limiting the atmospheric concentration of many essential nutrients.

In order to study natural and biomass burning emissions, several aerosol and gas sampling programmes were conducted in the Amazon Basin: the brushfire experiment in 1979/1980 [1]; the GTE/United States National Aeronautics and Space Administration (NASA) Amazon Boundary Layer Experiment (ABLE-2A) in the dry season (1985) [2] and the GTE/ABLE-2B experiment in the wet season (1987) [3, 4].

In 1991, a sampling campaign using a twin engine aircraft collected samples with a heavy 'loading' of biomass burning aerosol particles. In 1990, the Institute of Physics of the University of São Paulo installed two permanent aerosol monitoring stations in the Amazon Basin. One is situated in Cuiabá, in the Brazilian savannah, south of the Amazon Basin rain forest, and the other is in the 'Serra do Navio', in the northern part of the Amazon Basin, in the middle of the Amazon forest.

2. BIOMASS BURNING IN THE AMAZON BASIN TROPICAL RAIN FOREST

Biomass burning is a major source of particulate matter and gaseous emissions into the atmosphere [5, 6]. More than 80% of the emissions from all sources of biomass burning originate in the tropics. The particles emitted have hygroscopic properties [7, 8], affecting cloud formation. The increased number of cloud condensation nuclei (CCN) generates brighter clouds that reflect solar radiation back into space. There is a possibility of a change in the precipitation pattern and composition due to changes in CCN characteristics [9]. A major component of the particles is elemental carbon (also called graphitic carbon, or black carbon), which has strong radiative absorption properties [10].

Estimates of the total biomass consumed on a global basis range from 2 to 10 Pg/year (1 petagram = 10^{15} g) [6]. In terms of total particulate matter (TPM), emissions are around 104 Tg/year (1 teragram = 10^{12} g). For particulate matter in the fine mode ($PM_{2.0}$), emissions are estimated at 49 Tg per year, accounting for about 7% of the global emission rate. For elemental carbon, the emission of 19 Tg/year can account for a very high 86% of the total anthropogenic emissions [11]. Setzer and Pereira [12] estimated that the emissions of TPM from 350 000 independent fires in the Amazon Basin in 1987 were of the same order as those from a large volcanic eruption. In 1991, they observed 418 620 individual fires detected by remote sensing with the Advanced Very High Resolution Radiometer (AVHRR).

3. EXPERIMENTAL METHODS

Aerosol particles were sampled using stacked filter units (SFUs) [13]. The SFU was fitted with a specially designed inlet, which provided a 50% cutoff diameter of 15 μm [14]. Coarse particles ($2.0 < d_p < 15$ μm) were sampled on a 47 mm diameter, 8 μm pore size Nuclepore filter, while a 0.4 μm pore size Nuclepore filter collected the fine particles ($d_p < 2.0$ μm). The flow rate was typically 14 L/min, which resulted in a 50% cutoff diameter between fine and coarse aerosol fractions of about 2.0 μm [15]. The aerosol size distribution was measured with a six stage,

single orifice, Battelle type cascade impactor. At a flow rate of 1 L/min, the 50% cutoff diameters for the cascade impactor stages 5, 4, 3, 2 and 1 are: 4, 2, 1, 0.5 and 0.25 μm, respectively. A Nuclepore backup filter collects particles <0.25 μm. Vaseline coated Kimfol polycarbonate film was used as the impaction surface.

Particle induced X ray emission (PIXE) [15] was used to measure the concentrations of up to 22 elements (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Rb, Sr, Zr and Pb) [17]. The samples were irradiated by a proton beam at the Van der Graaff accelerator at Florida State University, and also at the cyclotron at the University of Ghent, Belgium [18]. A new, dedicated PIXE/Rutherford backscattering spectroscopy (RBS) facility at the University of São Paulo is now being used for PIXE analysis. A 5SDH Pelletron accelerator, coupled with a 135 eV Si(Li) detector, provides good detection limits for aerosol measurements. Gravimetric analysis was performed to obtain the fine ($d_p < 2$ μm) and coarse (2 μm $< d_p <$ 15 μm) mass concentration. The black carbon concentration was determined by a reflectance technique.

The large database collected in these experiments was analysed using receptor modelling [19]. Absolute principal factor analysis [20] was used to extract quantitative source profiles from the variability of the elemental concentrations.

4. CHARACTERIZATION OF AEROSOL PARTICLES EMITTED DURING BIOMASS BURNING

Aerosol particles were collected in forest fires in Mato Grosso, Brazil, in the southern part of the Amazon Basin. Cascade impactors and SFUs were used to collect particles near the fires, and their elemental composition was measured by PIXE [21]. There was no separation of the flaming and smouldering phases. Figure 1 presents size distributions for sulphur and potassium. Most of the elements are emitted mainly in the fine mode as a result of the combustion process, but there is a clear coarse mode for some elements, especially K. Calcium, Fe, Si and Ti are emitted, mainly in the coarse mode, with a small component in the fine mode. This effect is probably the result of two different processes: (1) gas to particle conversion, bringing the production of particles less than 2μm; and (2) strong convective dispersion of ash, semi-burned material and soil dust. Owing to the low settlement velocities, the fine fraction particles ($d_p < 2.0$ μm) can be transported for long distances. Figure 2 shows the variability of the elemental composition for different samples collected at different sites. Some elements show large variability, such as K which varies from 0.03 to 1% of the total aerosol mass loading. Sulphur has a rather constant emission factor of about 0.1% in mass.

In September 1991, a twin engine INPE 'Bandeirante' aircraft was used to collect aerosol samples in flights covering the Amazon Basin and the Cerrado region. Cascade impactors and SFUs were used to collect particles from an isokinetic sam-

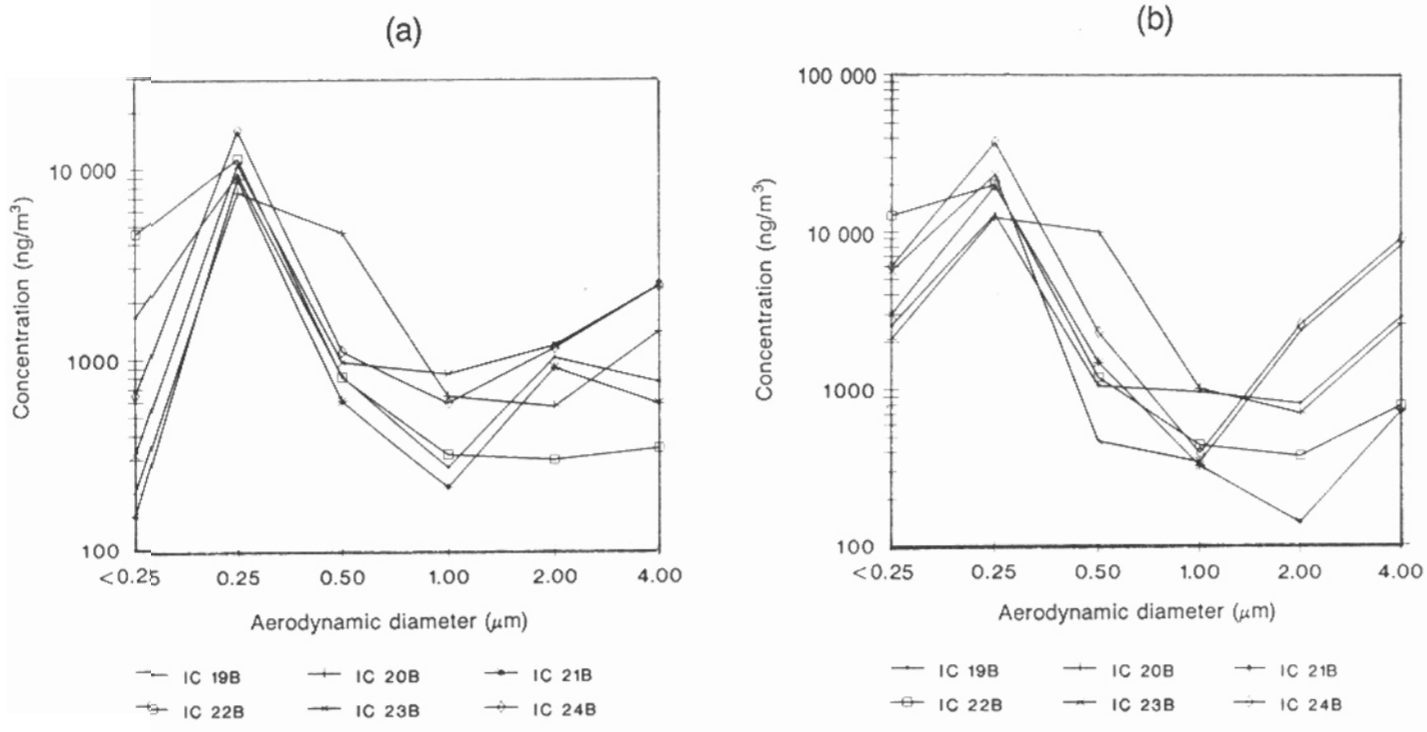

FIG. 1. Size distribution of (a) S and (b) K in aerosol particles emitted in biomass burning plumes.

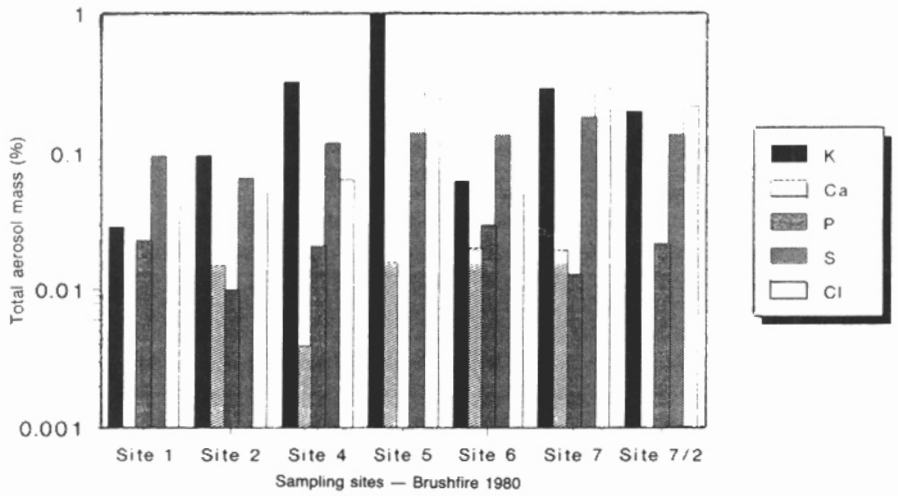

FIG. 2. Variability in the elemental composition for several biomass burning samples collected in the Amazon Basin.

pling probe. The flight time was 3–4 h at an altitude of 1000–3000 m. It is important to recognize that the collected aerosol particles are 'aged' biomass burning emissions and represent an average composition on a basin wide scale. During the week of the experiment, the number of fires in the Amazon Basin region was monitored using the AVHRR on channel 3.75 μm, with 1 km resolution, on board the NOAA-11 satellite. In the week of the sampling programme, 88 414 fires were detected in the Amazon Basin and in the month of September, 237 599 individual fires were detected. The 1991 season was very dry in the Amazon Basin. There were 105 days with no rain in Brasilia, just before the collection period. Table I presents the elemental, black carbon and aerosol mass concentrations for each of the individual samples. Also shown is the region in which the sample was collected. Most of the elements show high concentration variability, suggesting the regional influence of biomass burning plumes. The mass concentrations varied from 30 $\mu g/m^3$ for areas not affected much by biomass burning, to more than 370 $\mu g/m^3$ for regions with intense biomass burning. These concentrations represent the average of large areas, with no impact of any particular plume. The visibility for the whole area of the Amazon Basin was typically 500–1000 m, with regions with 200 m visibility, which keeps most of the airports in the region closed during the biomass burning season (August and September). Figure 3 presents the measured sulphur concentrations. For regions where high biomass burning was occurring, S concentrations were a very high 3000 ng/m^3, going down to less than 500 ng/m^3 for less affected regions. In the wet season, with no biomass burning, the S concentration averaged 120 ng/m^3 [22]. Another biomass burning experiment that we were involved in was

TABLE I. AMAZON BASIN BIOMASS BURNING AEROSOL COMPOSITION IN 1991

(aerosol mass, soot carbon and elemental concentrations expressed in ng/m^3)

Site	Mass	Soot	Mg	Al	Si	P	S	Cl	K	Ca	Ti	V	Mn	Fe	Cu	Zn	Ga	Pb	Br	Rb	Sr	Zr
Brasilia	60 000			3539	4402	106	604	146	875	1025	423		56.9	3085							8.06	15.0
P. Emas 1	128 000			9643	7452	69	1126	192	1 535	1649	1145	19	69.3	7817			4.36				10.55	36.9
P. Emas 2	94 000			7558	5490	95	902	199	1 369	980	1065	25	71.2	6449			5.56				8.14	22.8
Goiânia	134 000		1346	5344	3381		631	292	1 141	925	369		243.1	3463								
Barr. (Ba) 1	101 000			2127	3433		505	131	609	700	195		33.2	1435								
Barr. (Ba) 2	37 000			1590	2883		492		765	773	150		26.8	1089	6.9	7.4					4.84	9.0
P. Nacional	80 000	7 010	426	2223	4009	124	871	131	1 604	1348	185	3	71.3	1663	5.0	11.7			8.33	4.84	13.27	
Maraba 1	289 000	26 230	4337	1629	4252	1509	2645	2265	11 061	9253	112		370.9	1350	16.7	45.3	1.82		37	17.6	83.64	
Maraba 2B	171 000	10 780	502	645	2422	160	2596	908	3 042	2892	93		53.8	759	11.1	24.9			23.7		27.11	
Maraba 3	188 000	14 270	555	589	1083	116	2839	165	3 189	1585	41		52.8	384		20.3			27.3	9.06	12.82	
Santarem	28 000			216	399		1269		865	129	13		3.3	141		7.3						
Manaus	94 000	7 610	137	443	798		1960	28	1 434	138	38		6.9	369	2.1	11.7			9.94			
Manaus B	342 000	11 150	864	1375	5930	0364	2775	2153	3 613	4966	271		99.1	1424	8.5	25.8			18.8	7.49	43.51	
P. Velho 1	207 000	16 370		1062	1487		3286	121	2 484	276	88		17.2	796		22.8			14.6	15.2		
P. Velho 2	106 000			4044	4724		1444		1 789	479	375		45.4	2944		14.1						16.6
P. Velho 1B	370 000	15 140	1485	1615	5665	370	3210	1346	4 469	7693	141		175.9	1410		27.1			14	10.3	67.99	
Cuiaba	181 000	13 940	468	7241	9113	253	1755	106	2 479	1522	933	19	149.5	6234	9.6	34.3	4.94		12.7		20.71	21.0
C. Grande	49 000			737	1618		1489		673	210	85		13.3	729	4.0	11.5		7.68				
Ponta Pora	248 000			207	525		311		172	88	33		4.1	241				8.59				

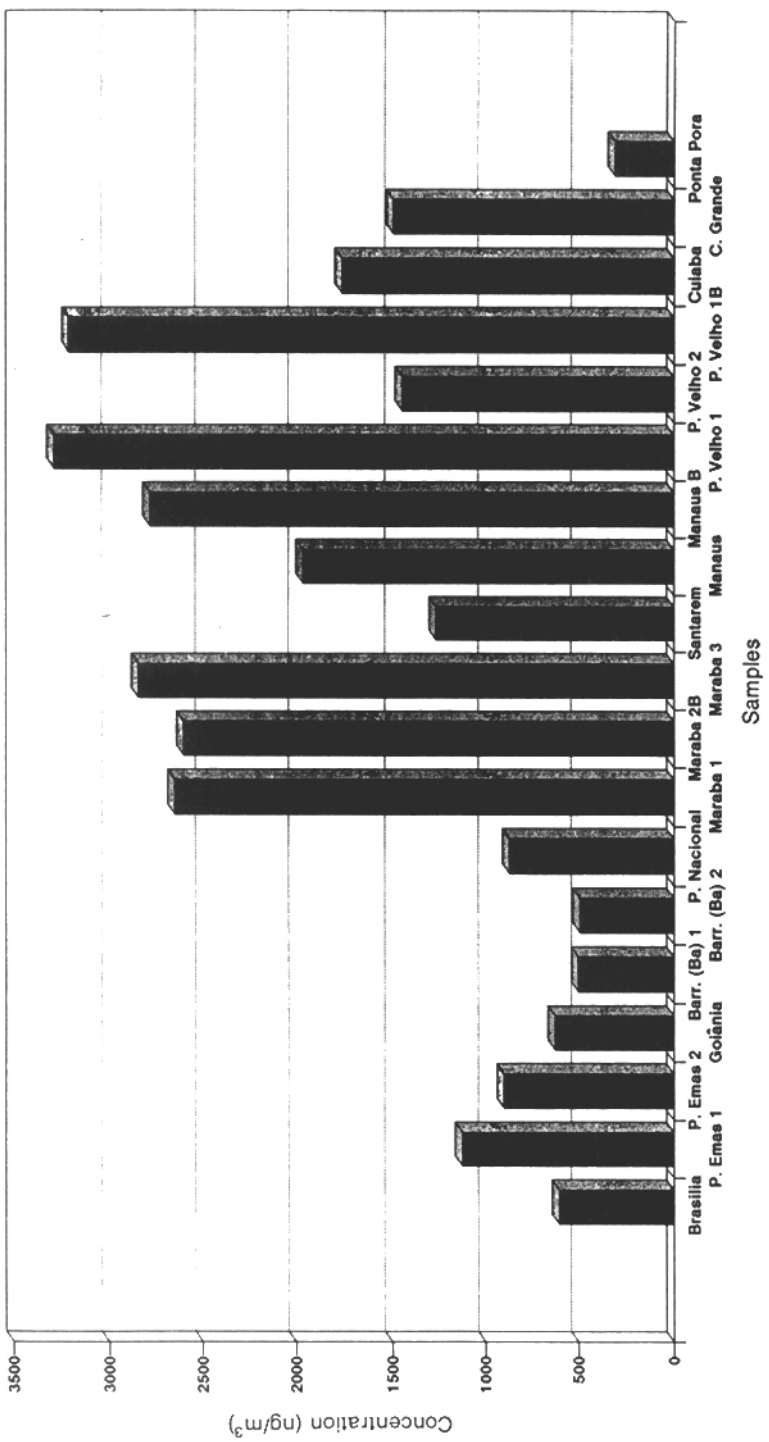

FIG. 3. Aerosol sulphur concentration (in ng/m^3) for samples collected in aircraft flights over the Amazon Basin in the biomass burning season in September 1991.

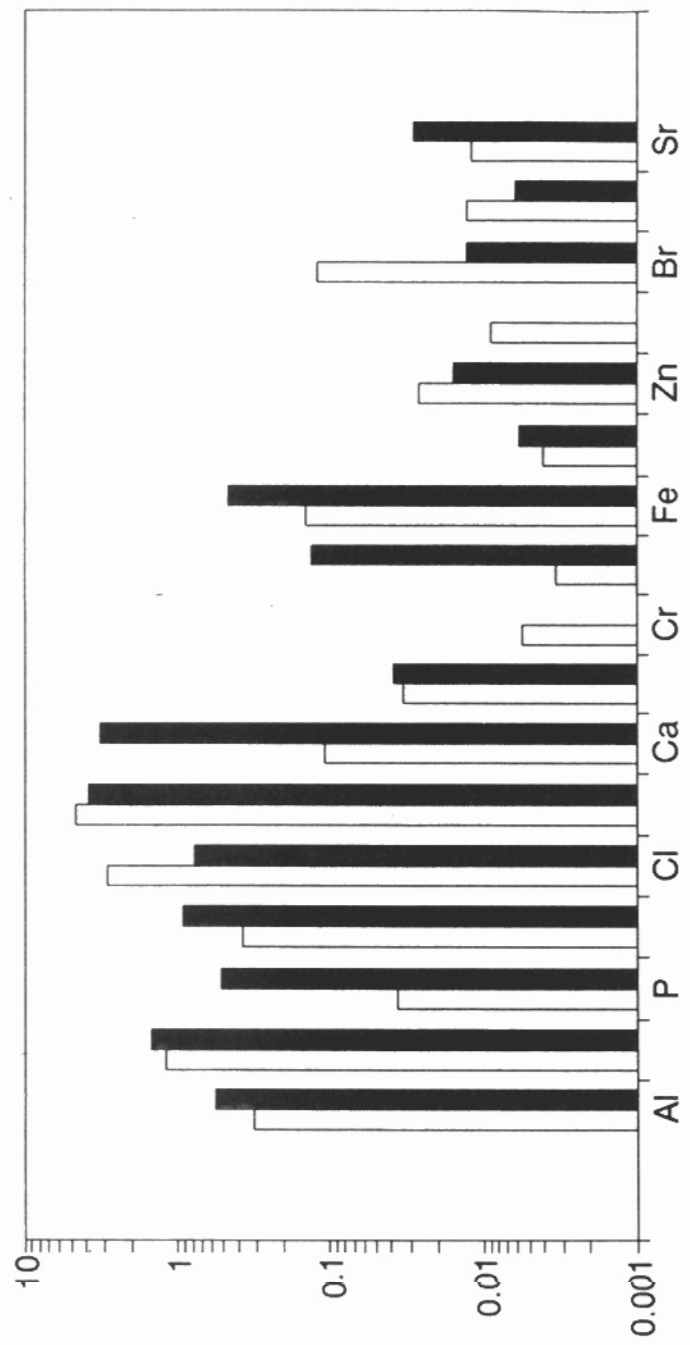

FIG. 4. Comparison of the elemental composition of African and Amazon Basin biomass burning emission (□: average DECAFE II; ■: average Amazon 1991).

the DECAFE (Dynamique et chimique de l'atmosphère en forêt équatoriale) experiment. Figure 4 presents a comparison of the elemental composition of DECAFE II samples and the 1991 Amazon Basin biomass burning samples. The elemental profiles from the savannah samples from DECAFE II and the Amazon Basin are, in general, similar. Potassium, Cl, Zn, Rb and Br are higher for the African emissions. These elements are known to constitute the natural biogenic emission from tropical rain forests. Soil dust elements are higher in samples collected in the Amazon Basin, perhaps due to the strong dry season. Sulphur accounts for 0.2–1.0% of the fine aerosol mass concentration. The major elements emitted are K, Cl, Si, Al, Ce, S, Fe and Br, with traces of Cu, Zn, Sr, Rb, As, Mn, Ti and Cr.

The size distribution shows that typical soil dust related elements like Fe, Al, Si, Ti and Mn are observed, mainly in the coarse mode fraction. Chlorine, S and Br are mainly observed in the fine mode. This is probably a result of gas to particle conversion mechanisms for these elements.

From these data it is possible to deduce that emissions of trace elements and heavy metals into the global atmosphere due to biomass burning are very high. For Cr, biomass burning emits about 5.6×10^9 g/year; for Cu, 1.6×10^9 g/year; and for Zn, 5.2×10^9 g/year. These values correspond to 18, 5 and 3.5%, respectively, of the global anthropogenic inputs. These emissions of heavy metals by biomass burning are currently not being taken into account in global emission heavy metal inventories.

5. BACKGROUND AEROSOL MONITORING IN THE AMAZON BASIN

For the samples collected in background sites, the concentration of naturally released biogenic elements like K, S, P, Cl, Ca and Zn dominates the elemental concentrations [2, 3, 22]. Dry and wet season results show some remarkable differences. The concentration of soil dust related elements (Al, Si, Ti, Mn and Fe) is five times larger in the wet season than in the dry season, and this was attributed to the long range transport of dust from the Sahara Desert to the interior of the Amazon Basin. Biogenic aerosol related elements (e.g. P, S, Cl, K and Zn) in the fine mode showed strongly reduced concentrations in the wet season. Fine mode sulphur concentrations averaged 80 $\mu g/m^3$ in the wet season and 260 $\mu g/m^3$ in the dry season. Factor analysis of these databases show that generally two factors explain more than 90% of the data variability for most of the sampling sites in the dry and wet seasons. These factors were soil dust (represented mainly by Al, Si, Ti, Mn and Fe) and biogenic related aerosol (with K, P, Cl, S, Zn and the aerosol mass concentration). The aerosol mass source apportionment indicated that biogenic particles account for 55–95% of the airborne concentrations for both the dry and wet seasons.

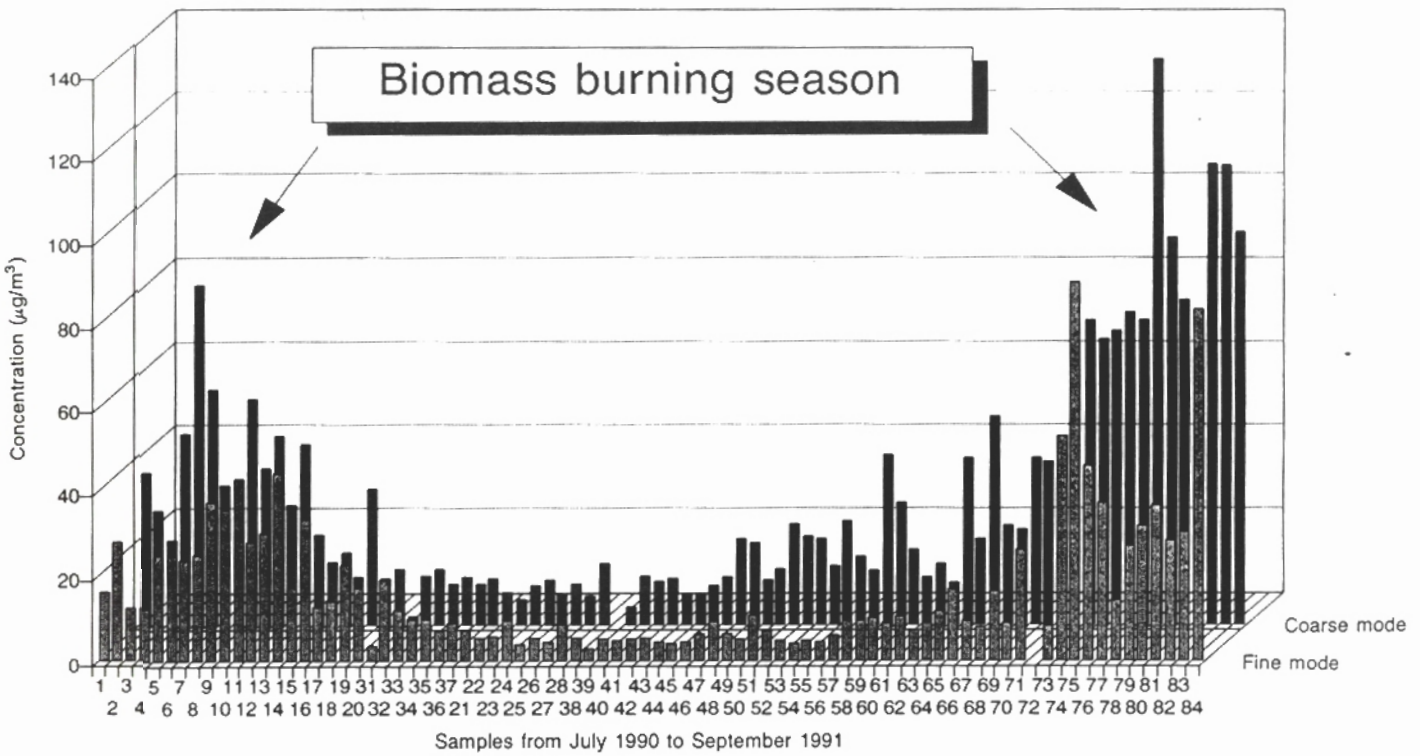

FIG. 5. Fine and coarse mode aerosol mass concentrations for the background monitoring station in Cuiabá, south of the Amazon Basin.

Since August 1990, the University of São Paulo has been operating a long term aerosol sampling station in Cuiabá, located south of the Amazon Basin. Most of the air masses from Rondonia, where large emissions of biomass burning occur, are transported about 1000 km to Cuiabá. Figure 5 shows the fine and coarse mode aerosol mass concentration for more than one year of continuous aerosol sampling in Cuiabá. The biomass burning season is in August/September. The large increases in aerosol concentrations during the burning season are clear. From a background concentration of less than 20 $\mu g/m^3$, the aerosol mass concentration increases to more than 140 $\mu g/m^3$ during the biomass burning season, even very far from the source. The black carbon concentration has strong variability. From a background value of 0.6 $\mu g/m^3$, black carbon goes to 7 $\mu g/m^3$ during the burning season. Several trace elements, such as S, K, Ca, Zn, Rb and others, also undergo a change in their concentrations as a result of regional biomass burning.

6. CONCLUSIONS

Aerosol concentrations in the whole area of the Amazon Basin during the biomass burning season are very high. Large amounts of fine particles are injected into the atmosphere, where they can travel for long distances. Several essential nutrients are transported in the atmosphere as a result of biomass burning processes. Most of the particles are water soluble and are active as CCN, with the potential to change the cloud formation mechanisms in the Amazon Basin.

At the biomass burning plumes, carbon accounts for more than 80% of the airborne mass, while N and O account for up to 18%. Potassium, P, S, Ca, Cl and Si are the dominant elements emitted. There are also significant emissions of Fe, Zn, Ca, Cl, Mn, Zn, Br and Rb. In the fine mode, K accounts for $4.6 \pm 2.6\%$ of the fine particle mass. Chlorine accounts for $2.9 \pm 1.9\%$, Zn for $0.026 \pm 0.014\%$ and S for $0.37 \pm 0.31\%$. Some elements, like Si, Ti, Fe and Ca, are emitted predominantly in the coarse mode, while others, like S, Br and Cl, are emitted in the fine mode. There are large differences in emissions between the different fire conditions (smouldering or flaming), and there is great variability between sites for some elements. The size distributions show clear patterns for elements predominantly emitted in the fine mode (S, Cl, K and Br), with a mass median aerodynamic diameter (MMAD) of about 0.3 μm, while in the coarse mode (Fe, Si, Al and Ca) the MMAD is about 4.0 μm. Taking into account the worldwide biomass burned and the average emission factors measured for zinc, biomass burning can account for 5–30% of the anthropogenic global emissions of zinc. For elements like K, P, Ni, Cu, Zn and Rb, biomass burning can be responsible for a significant fraction in the worldwide emissions.

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