

TRACE ELEMENTS IN AEROSOLS FROM BACKGROUND AIR POLLUTION MONITORING STATIONS IN THE AMAZON BASIN USING NUCLEAR-RELATED TECHNIQUES

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Abstract

In order to study the natural release of aerosol particles by the Amazon Basin tropical rain forest, the composition and size distribution of biogenic aerosol particles will be analyzed. The role of the atmospheric emissions from the Amazon Basin rain forest in the global atmosphere will be investigated. The atmosphere will be studied in long-term sampling stations in three different locations. The elemental composition of aerosol particles released during biomass burning will also be measured in several different ecosystems, from primary forest to Savannah. One of the main focuses is to identify and quantify important physical and chemical processes in the generation, transformation and deposition of aerosol particles. Also important is to obtain a better understanding of natural aerosol sources concerning identification, their characteristics and strength, to be able to understand the natural chemistry in the atmosphere on a global scale.

Aerosols will be sampled with Stacked Filter Units (SFU) and cascade impactors with detailed size resolution in the sub-micrometer range. The samples will be analyzed by Particle Induced X-ray Emission (PIXE) for measuring 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). The elemental composition will be measured at the new PIXE facility from the University of São Paulo, using a dedicated 5SDH tandem Pelletron nuclear accelerator. The multi-element data obtained from the bulk analysis of the aerosol samples by PIXE will be examined by receptor models.

At the "Serra do Navio" sampling site a very clean background aerosol is being observed. Biogenic aerosol dominates the fine mode mass concentration, with the presence of K, P, S, Cl, Zn, Br, and FPM. Three components dominate the aerosol composition: soil dust particles, the natural biogenic release by the forest, and a marine aerosol component. At the "Cuiabá" site, during the dry season, a strong component of biomass burning is observed. Aerosol mass concentration up to $160 \mu\text{g}/\text{m}^3$ was measured. Absolute Principal Factor Analysis (APFA) showed three components: soil dust (Al, Ca, Ti, Mn, Fe), biomass burning (Soot, FPM, K, Cl) and natural biogenic particles (K, S, Ca, Mn, Zn). The fine mode biogenic component of both sites shows remarkable similarities, although the two sampling sites are 3,000 km apart. Several essential plant nutrients like P, K, S, Ca, Nitrogen and others are transported in the atmosphere as a result of biomass burning processes.

1. SCIENTIFIC BACKGROUND AND SCOPE OF THE PROJECT

The Earth's atmosphere is a vital natural resource that until recently appeared unaffected by human activities, except on local scales. However it has become clear that the worldwide strongly growing anthropogenic activities have impacts on the global atmosphere [1]. It is becoming clear that is necessary to increase our knowledge of the chemical processes that determine the composition of the atmosphere in background areas, and to understand the interactions between atmospheric composition and biological

processes. Also it is necessary to obtain a better understanding of the alterations in the atmospheric composition due to changes in land use in tropical rain forests.

Tropical rain forest covers a large area of the Earth, and is a globally important ecosystem, with large emissions of biogenic gases and particles [2]. The Amazon Basin, with about 4 million square kilometers, plays a very important role in emissions of water vapor, gases and aerosol particles to the global atmosphere. Almost a third of the rain that falls on the Earth's continents is deposited within the narrow equatorial belt, with *important contribution in the global biogeochemical cycling of several elements and species* [3]. The International Global Atmospheric Chemistry Programme (IGAC) and the International Geosphere-Biosphere Programme (IGBP) has recognized the importance of the tropical rain forest ecosystems and several projects are being planned to study characteristics of the atmosphere in these regions. The deposition of biogeochemically important species to the Earth's surface plays an essential role in limiting the atmospheric concentration of many essential nutrients. Important redistribution of both plant nutrients and toxic substances within the biosphere result from such deposition. Both dry and wet deposition pathways must be understood if the biogeochemical cycles of many species are to be quantitatively assessed.

The Amazon Basin has the world's largest rain forest and is a region with intense convective activity, resulting in rapid vertical mixing of biogenic gases and aerosols to high altitudes where they can be transported over long distances and have an impact on the global Tropospheric chemistry. Estimates of the global production rate of organic aerosols show that the forest vegetation is the principal source of atmospheric organic particles. Long-range transport of continental tropical carbonaceous aerosol may have an impact on the remote troposphere [4, 5]. Only few studies involving aerosol elemental concentration measurements have been conducted in tropical rain forests [4, 5, 6, 7, 8, 9, 10, 11, 12], many of them with very limited sampling time.

The continuous natural release of aerosols by plants is perhaps globally more important than the emissions of gases and particles during forest burning. Crozat [13] hypothesized that forest is a major natural global source of aerosols. Fish [14] suggested that haze observed in forested areas could be due to submicrometer particles from electrical generation of biogenic aerosol by leaves. Decaying vegetation can generate small particles that can act as cloud nuclei [15]. Wind action on plant leaves can result in mechanical abrasion, generating large biogenic particles [16]. Biological activity of microorganisms on leaf surfaces and forest litter results in airborne particles, and wind-blown pollen grains contribute to coarse fraction particles in forested areas. Particulate material containing Zn, Pb and Cu can be generated by higher plants [17], and plant wax constituents were measured in urban areas and over the Atlantic ocean. Curtin *et al.* [18] found several trace elements, including Na, Mg, Mn and Zn, in exudates from conifer trees. In the Ivory Coast tropical rain forest Crozat [13] observed K enrichment in biogenic aerosols. The transpiration of plants can lead to migration of Ca^{2+} , SO_4^{2-} , Cl^- , K^+ , Mg^{2+} and Na^+ to the atmosphere [19]. Artaxo [20, 21] studied the aerosol elemental composition in the Brazilian Atlantic forest and used receptor modeling to obtain the

biogenic aerosol characteristics, showing that the elemental profiles from aerosols in the Brazilian Atlantic forest are similar to the ones obtained in the dry season in the Amazon Basin.

1.1 The atmospheric emissions during biomass burning

The fast deforestation occurring now in the Amazon Basin is changing the atmospheric composition and will have a regional climatic impact that can affect a large portion of the equatorial region. Alterations of biogenic exchange fluxes are certainly occurring due to changes in land use. The emissions of gases during biomass burning are affecting the global concentrations of gases like CO, CO₂, CH₄, CH₃Cl, N₂O, COS and others [22, 23]. Much less studied is the composition of aerosol particle emissions. A limited effort was done during the 1980 Brushfire Experiment, an international experiment with the participation of the National Center for Atmospheric Research (NCAR) from USA, Max Planck from Germany, and the University of São Paulo, Brazil. The emission of aerosols during the burning of forests was studied at some locations, including the Amazon Basin and in central Brazil. The composition and size distribution of aerosol particles were measured in forest fires in Rondonia, Brazil, with large emissions of potassium, sulfur, silicon, zinc and organic matter [11]. The French-German experiment Dynamique et Chimie de l'Atmosphère en Forêt Equatoriale (DECAFE/FOS) [24] have measured gases and aerosol particle emissions in Savannah fires in Africa in 1988 and 1991. Aerosol samples were collected for the University of São Paulo team, and analyzed for trace elements. The sulfur cycle was extensively studied in DECAFE experiment, and relationship of aerosol sulphate and other gases was determined. There are large differences in the elemental composition between the DECAFE and Brushfire experiment samples, in part due to the very different vegetation burned. For biomass burning emissions, there is no aerosol composition data covering different environments from different ecosystems. This would be important to obtain, especially for fine mode particles and sulfur species.

2. METHODS

Aerosols will be sampled using Stacked Filter Units (SFU) [25]. Coarse particles ($2.0 < d_p < 15 \mu\text{m}$) are collected on a 47-mm-diameter, 8 μm pore-size Nuclepore filter while a 0.4 μm pore-size Nuclepore filter collected the fine particles ($d_p < 2.0 \mu\text{m}$). The flow rate is typically 14 lpm that results in a 50 % cutoff diameter between fine and coarse aerosol fractions of about 2.0 μm [34]. The SFU are fitted with a specially designed inlet which provides a 50 % cutoff diameter of 15 μm [27] so that only inhalable particles are sampled. SFU are loaded with the Nuclepore filters in a clean room at the University of São Paulo, transported in a sealed container, and hand-carried after the sampling. The sampling duration will be 24 to 48 hours in order to have good time resolution to discriminate long-range transport events.

Two monitoring stations are being operated continuously. The first sampling station is situated in Cuiabá, at the Brazilian savanna, south of the Amazon basin rain forest. The Cuiabá sampling station allows the analysis of regional effects of biomass burning emissions due to the location of the station. The site is heavily affected by regional savanna biomass burning. A second aerosol sampling station was installed in November 1991 at the "Serra do Navio," in the Northern part of the Amazon basin. This station is located 190 km North of the equator, in a primary tropical rain forest. The site is relatively free from regional biomass burning emissions, and there are no industrial activities for at least a thousand kilometers around the sampling site.

The samples will be analyzed by Particle Induced X-ray Emission (PIXE). PIXE allows a fast, non-destructive and multielement analysis of the aerosol samples. In the PIXE method [28] the samples will be irradiated by a 2.4 MeV proton beam produced by a nuclear accelerator at the Institute of Physics, University of São Paulo. The PIXE system in São Paulo uses a new Pelletron 5SDH nuclear accelerator dedicated for PIXE analysis. The following 22 elements are normally detected for Amazon Basin aerosol samples: Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Rb, Sr, Zr, and Pb. The detection limit is typically 5 ng m^{-3} for elements with $Z < 20$, and 0.1 ng m^{-3} for $21 < Z < 30$. The precision of the PIXE analysis is better than 5 % for the major elements and about 10 % for elements with concentrations near the detection limit.

The fine and coarse fraction aerosol mass concentrations are obtained through gravimetric analysis of the Nuclepore filters. The filters are weighed before and after sampling in a Mettler M3 electronic microbalance with $1 \mu\text{g}$ sensitivity. Before weighing, the filters are equilibrated for 24 hours at 50 % relative humidity and $20 \text{ }^\circ\text{C}$. Electrostatic charges are controlled by means of ^{210}Po radioactive sources. Detection limit for the aerosol mass concentration is $0.3 \mu\text{g m}^{-3}$. Precision is estimated at about 15 %. Soot carbon concentration will be measured with a reflectance technique using a "Diffusion System" photometer.

2.1 Sources and Transport of Aerosol Particles in the Amazon Basin: Receptor Modeling

The elemental composition of particulate matter sources can be studied with the use of receptor models (*e.g.* Chemical Mass Balance, Stepwise Multiple Regression, Factor Analysis, Cluster Analysis, etc.) [29, 30, 31]. In the present study we are interested in identifying the natural sources of aerosols in the Amazon Basin and extracting their elemental compositions through the use of absolute principal factor analysis (APFA) [32]. APFA offers the possibility to obtain a quantitative source profile instead of only a qualitative factor loading matrix as in traditional applications of factor analysis. The elemental source profiles help in the identification of the factors and can be used to compare the factor compositions with the assumed aerosol sources.

In principal factor analysis [33] a model of the variability of the trace element concentrations is constructed so that the set of intercorrelated variables is transformed into a set of independent, uncorrelated variables. This is done by finding the eigenvalues

and eigenvectors of the correlation matrix. The most prominent eigenvectors (factors) are retained and orthogonally rotated by a VARIMAX rotation. The resulting "factor-loading" matrix represents the correlations between the trace elements and each orthogonal factor. Also "factor scores" are calculated which indicate the relative importance of each factor for the individual samples. The APFA procedure obtains the elemental mass contribution of each identified component by calculating the absolute principal factor scores (APFS) for each sample. The elemental concentrations are subsequently regressed on the APFS to obtain the contribution of each element for each component. These source profiles thus obtained can be compared with values from the literature to gain information on enrichment and atmospheric chemistry processes. The measured aerosol mass concentration can also be regressed on the APFS in order to obtain the aerosol total mass source apportionment.

3. RESULTS

A large number of samples were collected at the Cuiabá sampling station, from which results from 106 fine mode aerosol samples are presented here. For Serra do Navio, 28 fine mode samples were analyzed. Figure 1 presents the time series of the fine, coarse

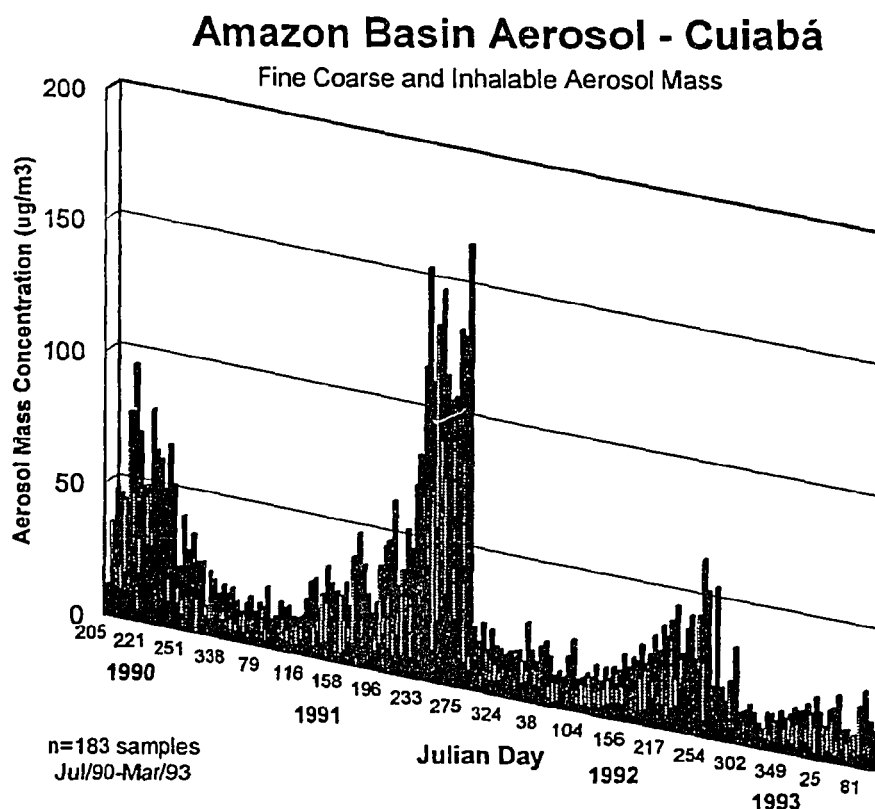


Figure 1. Time Series of the Fine, Coarse and Inhalable Particulate Matter Concentrations for the Cuiabá Site

and inhalable particulate matter concentration for the SFU samples collected at the Cuiabá sampling site. There is very clearly a large increase in aerosol loading in the atmosphere during the biomass burning season. From an inhalable particulate matter (IPM) concentration of $20 \mu\text{g}/\text{m}^3$ during the wet season, the concentration goes as high as $160 \mu\text{g}/\text{m}^3$ during the biomass burning season. Aircraft measurements over large areas of the Amazon Basin show very high average IPM concentrations up to $300 \mu\text{g}/\text{m}^3$. These high concentrations are observed in areas as large as 2 millions square kilometers, using aircraft and remote sensing measurements. Soot carbon concentrations are also high. Table I presents the average elemental concentrations for the fine mode aerosol from both sampling sites. FPM represent the fine mode aerosol mass concentration in $\mu\text{g}/\text{m}^3$. Sulphur at Cuiabá, with an average concentration of $426 \pm 365 \text{ ng}/\text{m}^3$ appears higher than in other studies of aerosol composition in the Amazon Basin [6, 7], due to the high influence of biomass burning emissions. At the Serra does Navio site, the fine mode sulfur concentration ($726 \pm 411 \text{ ng}/\text{m}^3$) is partially a result of marine air masses entering the Amazon basin region. The average concentrations of heavy metals like Zn, Cu, Ni, Sr, Zr, Rb, Pb, *etc.* are very low, showing the absence of atmospheric anthropogenic emissions at the sites, with the exception of biomass burning.

The variability of the elemental concentrations was analyzed using the Absolute Principal Factor Analysis (APFA) technique. For the fine mode Cuiabá aerosol samples, only three factors account for most of the trace element variability. Table II presents the VARIMAX rotated factor loading matrix. The first factor with high loading for Al, Ca, Ti, Mn, Fe and Zn represents soil dust aerosol particles. The second factor with high loading for Soot, FPM, K and Cl represents biomass burning aerosol. The third factor with high loading for S, K, Ca, Zn and Mn represents biogenic aerosol particles. The communality for each element is shown in the last column, and is very high for all variables. Table III shows the VARIMAX rotated factor loading matrix for the Serra do Navio sampling site. Three factors were statistically significant. The first factor has high loadings for Al, Si, Ca, Ti, Mn, and Fe, clearly representing soil dust aerosol particles. The second factor has high loadings for K, S, Zn, Br, FPM, and Cl, representing biogenic aerosol particles. The third factor has significant loadings only for chlorine, representing marine aerosol particles. The Serra do Navio sampling site is about 500 km away from the ocean, and the prevailing wind direction is from the sea coast to the sampling site. Communality for all elements in Table III is very high.

The APFA procedure allows obtaining absolute source profiles in units of ng/m^3 . Figure 2 shows the elemental source profiles for the APFA Cuiabá data set. Potassium has high elemental concentrations for the three factors, because it has contribution from soil dust, biomass burning and biogenic aerosol emissions. Fine mode chlorine is emitted mainly by biomass burning. Rubidium and strontium were shown to be emitted by biogenic and biomass burning processes. Pb and Cr are present mainly in the biogenic and soil dust components. Zr is similarly distributed along the three components.

Table I - AVERAGE ELEMENTAL CONCENTRATION IN NG/M³ AND STANDARD DEVIATION (σ) FOR FINE MODE AEROSOL COLLECTED AT TWO BACKGROUND SAMPLING STATIONS IN THE AMAZON BASIN.

The station at "Cuiabá" is situated in a region with influence of regional biomass burning. The "Serra do Navio" sampling site is in a primary forest location far away from biomass burning regions. n is the number of samples with elemental concentration above the detection limits.

Element	Cuiabá	n	Serra do Navio	n
Al	104±107	106	209±232	28
Si	148±146	79	479±558	28
P	10.6±7.5	67	15.1±10.9	19
S	426±365	106	726±411	28
Cl	11.9±12.9	106	15.5±8.2	28
K	369±379	206	375±182	28
Ca	32.4±25.8	106	62.0±39.5	28
Ti	7.68±8.07	106	15.9±17.7	28
V	-	-	1.20±0.22	5
Cr	3.79±2.97	55	2.51±0.92	13
Mn	4.00±3.29	106	4.13±2.04	28
Fe	199±183	106	140±127	28
Ni	0.57±0.57	1	0.17±0.17	1
Cu	1.53±1.14	44	1.32±0.81	9
Zn	6.39±5.09	106	3.69±1.59	28
Se	0.35±0.23	5	5.26±3.15	1
Br	5.62±5.89	86	5.26±3.15	28
Rb	1.32±1.02	41	1.07±0.54	19
Sr	0.70±0.59	49	1.06±0.56	20
Zr	1.22±0.91	5	1.39±0.59	23
Pb	1.68±1.35	87	0.82±0.21	16
FPM(*)	10.9±10.7	106	14.2±6.9	28
Soot (**)	2.10±1.79	106	-	-

(*) FPM is the fine mode aerosol mass concentration in $\mu\text{g}/\text{m}^3$.

(**) Soot carbon concentration is expressed in $\mu\text{g}/\text{m}^3$.

Figure 3 shows the elemental composition of the biogenic component for the two sampling sites. P, Ca, Cr, Zn, Br, Rb, Sr, Zr and Pb show similar concentrations for both sampling sites, despite the three thousand kilometers distance between the two sampling stations. S, Cl, K and FPM show higher concentrations for the Serra do

Table II - FACTOR ANALYSIS RESULTS FOR THE FINE MODE CUIABÁ AEROSOL SAMPLES.

(Varimax rotated factor loading matrix, with three factors statistically significant.)

Variable	Factor 1	Factor 2	Factor 3	Communality
	Soil Dust	Biomass Burning	Biogenic particles	
Al	0.87	0.36	0.29	0.97
S	0.41	0.36	0.78	0.91
Cl	0.14	0.90	0.19	0.88
K	0.45	0.59	0.61	0.93
Ca	0.73	0.18	0.58	0.91
Ti	0.93	0.24	0.24	0.99
Mn	0.71	0.39	0.53	0.94
Fe	0.91	0.23	0.31	0.97
Zn	0.58	0.40	0.63	0.89
FPM	0.25	0.91	0.25	0.96
Soot	0.39	0.85	0.26	0.95

Table III - FACTOR ANALYSIS RESULTS FOR THE FINE MODE SERRA DO NAVIO AEROSOL SAMPLES.

(Varimax rotated factor loading matrix, with three factors statistically significant.)

Element	Factor 1	Factor 2	Factor 3	Communality
	Soil Dust	Biogenic Aerosol	Marine Aerosol	
Al	0.96	-0.14	-0.19	0.99
Si	0.97	-0.14	-0.16	0.99
S	-0.28	0.94	0.01	0.96
Cl	-0.24	0.64	0.70	0.97
K	0.00	0.99	0.03	0.98
Ca	0.94	0.03	0.02	0.88
Ti	0.97	-0.17	-0.14	0.99
Mn	0.91	-0.14	0.28	0.93
Fe	0.99	-0.11	-0.09	0.99
Zn	-0.05	0.97	0.17	0.96
Br	-0.08	0.88	0.16	0.80
FPM	-0.14	0.96	0.02	0.93

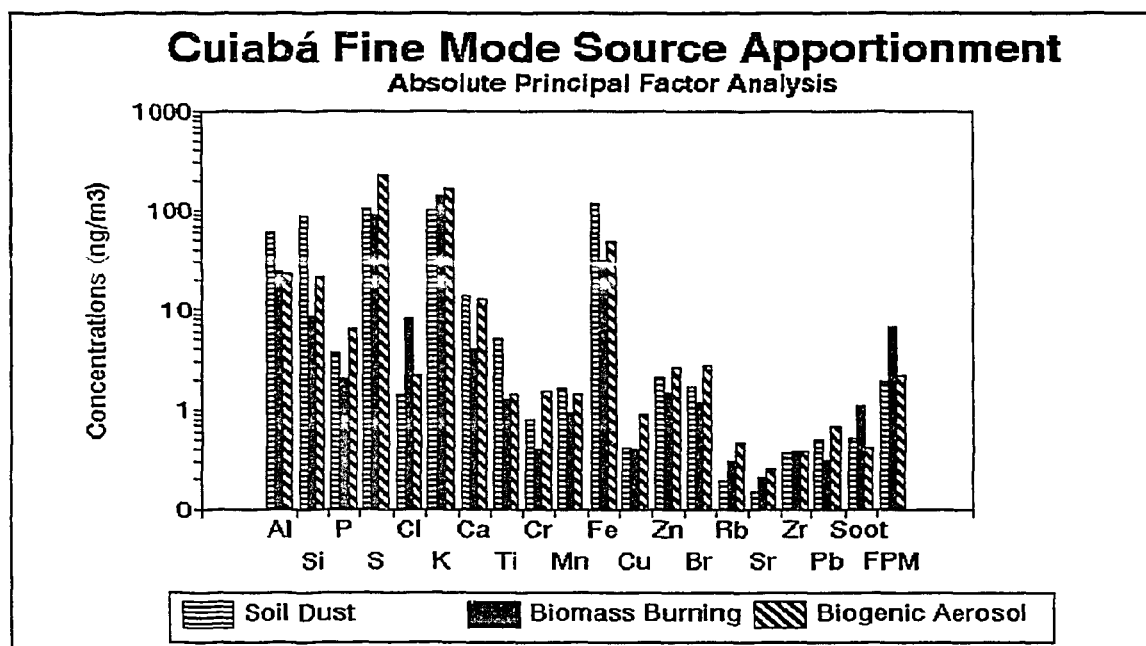


Figure 2 - Elemental Source Profiles for the Cuiabá Absolute Principal Factor Analysis Calculations.

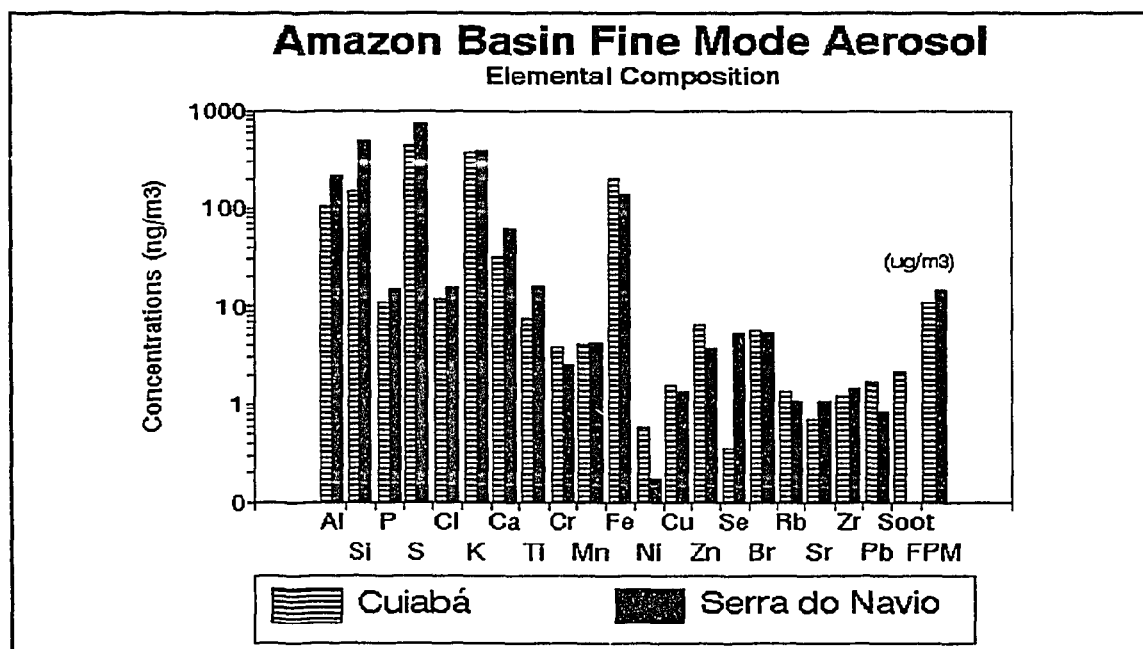


Figure 3 - Elemental Composition of the Biogenic Component for the Two Background Aerosol Sampling Sites. P, Ca, Cr, Zn, Br, Rb, Sr, Zr and Pb show similar concentrations for both sampling sites, despite the three thousand kilometers distance.

Navio sampling site. In Cuiabá, due to the high soil dust load in the atmosphere, it is possible to observe Al, Si, Ti and Fe in the biogenic elemental profile. The biogenic elemental source profile is similar to the one observed near Manaus during the ABLE-2B experiment [42], and in other forested regions of Brazil [9]. This agreement is surprising because of the large differences in ecosystems and plant species in the Amazon Basin. This biogenic aerosol component consists of many different types of particles, such as algae, fungus, plant debris and other particles difficult to identify [34, 35, 36]. Together with the soil dust component, it was possible to observe long-range transported aerosol particles from the Sahara desert into the Amazon Basin, with this component being mixed with the marine aerosol component at the Serra do Navio sampling site.

4. PLANS FOR FUTURE WORK

We will continue to operate the Amazon Basin sampling stations, collecting continuously fine and coarse mode aerosol particles. New instruments are being added to the network, including a Multi-Orifice Uniform Deposit Impactor (MOUDI) cascade impactor and an instrument to continuously measure soot carbon, the Aethalometer. At the analytical side, we will perform elemental analysis using our PIXE system, and also ion chromatography will be used to measure ionic components, including organic acids. New developments in receptor modelling, using hierarchical cluster analysis will be used to help in the data interpretation. Also some aircraft flights will be performed over large areas of the Amazon Basin, in order to obtain a basin-wide picture of aerosol spatial distribution.

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