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SIZE DISTRIBUTION OF BIOGENIC AEROSOL PARTICLES FROM THE AMAZON BASIN

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Abstract—The size distribution and elemental composition of natural background aerosol particles from the tropical rain forest of the Amazon Basin were measured. A 45 m high tower installed in an undisturbed forest near Manaus was used and aerosol was sampled in three levels of the tower. Single orifice Batelle-type cascade impactors with 5 stages and backup filter were used. A total of 50 complete cascade impactor sets was sampled in April and May 1987, during the wet season, when no forest burning occurs. Particle-induced X-ray emission (PIXE) was used to measure elemental concentrations of 20 elements (Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Rb, Sr, Zr and Pb). Absolute factor analysis was used to interpret the large data set of the trace element concentrations and to obtain quantitative elemental source profiles. The identified components in all size ranges comprise biogenic aerosol naturally released by the forest, mineral dust aerosol particles and sea salt. The elements associated with the biogenic component were potassium, phosphorus, sulphur, zinc, strontium and others in smaller proportions. The mineral dust component comprises most of the concentration of aluminium, silicon, calcium, titanium, manganese and iron. Chlorine was found to be associated with the aeolian dust component because of the long-range transport of Sahara dust over the Atlantic ocean, bringing Cl together with mineral dust particles. The size distribution of the elements associated with the windblown dust (Al, Si, Ca, Ti, Mn, Fe and Cl) shows in the coarse mode, with an average aerodynamic diameter of 4 μm . Two different biogenic aerosol components were observed. A fine mode component ($d_{50} \approx 0.5 \mu\text{m}$), characterised by the elements S, Zn and Sr and the mode biogenic component ($d_{50} \approx 3.0 \mu\text{m}$) characterised by the presence of P, K, Cl and Sr. The coarse biogenic (P, K, Cl, Sr) component is predominant at ground level under the forest canopy. The fine mode biogenic component (S, Zn, Sr) is higher at the top level of the tower.

Key word index: Aerosol particles, size distribution, elemental composition, Amazon Basin, biogenic aerosols, receptor modelling, factor analysis.

1. INTRODUCTION

The tropical region has important impacts in the global atmosphere. Tropical rain forest covers a large area of the earth, and is a globally important ecosystem. It is characterised by intense sources of biogenic gases and aerosols (Harriss *et al.*, 1990). The Amazon Basin has the world's largest rain forest, with about 4 million square kilometers, and is a region with intense convective activity (Greco *et al.*, 1990), 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 (Artaxo *et al.*, 1994). The forest vegetation is the principal source of atmospheric organic particles (Simoneit *et al.*, 1990; Cachier *et al.*, 1985; Crozat *et al.*, 1979). Only few studies involving naturally released biogenic aerosols have been conducted in tropical rain forests (Lawson and Winchester, 1978; Artaxo and Orsini, 1986, 1987; Artaxo and Maenhaut, 1990; Artaxo *et al.*, 1988, 1990, 1993a).

The primary biogenic aerosol particles consist of many different types of particles, including pollen, spores, bacteria, algae, protozoa, fungi, fragments of leaves, excrement and fragments of insects (Simoneit, 1989). Also a significant fraction of biogenic particles in forested areas comprises secondary aerosol particles formed by gas-to-particle conversion of organic, nitrogen and sulphur-related gases. These particles are sub-micrometer in size. Bacteria in forested areas are in the size range of 0.5 to 2.5 μm in size (Jaenicke and Mathias-Maser, 1992). Biological particles exhibit cloud influencing properties (Maki and Willoughby, 1978; Schnell, 1982; Vali *et al.*, 1976; Dingle, 1966) and can act as cloud condensation nuclei, potentially affecting the cloud formation mechanisms (Novakov and Penner, 1993). The primary biogenic aerosol particles, even in urban areas, can be as much as 50% in mass, and this shows how the sources of biogenic aerosol particles have been underestimated in the past (Jaenicke and Mathias-Maser, 1992). Aerosol particles are also produced during the photo oxidation of

natural biogenic hydrocarbons as isoprene, monoterpenes and other gases. Their fine mode component also affects the radiative balance in tropical regions.

The continuous natural release of aerosols by plants has been studied by some investigations. Crozat (1979) hypothesised that the forest is a major natural global source of aerosols. Fish (1972) suggested that haze observed in forested areas could be due to submicrometer particles from electrical generation of biogenic aerosol by leaves. Decaying vegetation produce small particles that can act as cloud nuclei (Schnell and Vali, 1973). Wind action on plant leaves can result in mechanical abrasion, generating large biogenic particles with the presence of heavy metals (Beauford *et al.*, 1977). Biological activity of microorganisms on leaf surfaces and forest litter results in airborne particles, and windblown pollen grains certainly contribute to coarse fraction particles in forested areas. Particulate material containing Zn, Pb and Cu was observed to be produced by higher plants (Beauford *et al.*, 1975, 1977). Plant wax constituents were measured in urban areas (Simoneit, 1984) and over the remote marine areas (Simoneit, 1977). The transpiration of plants can lead to migration of Ca^{2+} , SO_4^{2-} , Cl^- , K^+ , Mg^{2+} and Na^+ to the atmosphere. The biogenic related elements (e.g. K, P, S, Zn, Rb) are essential elements in superior plants, and are present in the fluids circulating in the plant and are released from the leaves to the atmosphere during guttation and transpiration (Nemeruyk, 1970).

The emission of sulphur gases from terrestrial plants is well-established. Vegetation is known to emit a large number of sulphur containing gases like COS (carbonyl sulphide), DMS (dimethylsulphide), DMDS (dimethyldisulphide), SO_2 (sulphur dioxide), H_2S (hydrogen sulphide), CS_2 (carbon disulphide), MeSH (methylmercaptan) and other. The emission of DMS from trees was observed by Lovelock (1974). Generally the emission rate for sulphur gases increases with temperature (Fall *et al.*, 1988). The vegetation is believed as the major worldwide sink for COS (Brown and Bell, 1986). The flux of DMS, H_2S and MeSH was measured in the Amazon Basin during ABLE-2A by a gradient/flux technique (Andreae and Andreae, 1988). Measurements of the sulphur cycle compounds in the African rain forest by Bingemer *et al.* (1992), showed similar concentration to those measured in Amazonia. Many of these sulphur compounds are rapidly oxidised to sulphate in the very humid and hot atmosphere of the Amazon forest. As sulphur is also present in the fluids of the vegetation together with P, K, Ca and other biogenic elements, it is also emitted directly during the natural processes of transpiration and guttation. Due to their organic matrix, biogenic aerosol particles could make most of the cloud condensation nuclei in tropical regions (Novakov and Penner, 1993).

To improve our understanding of chemical processes and atmospheric characteristic of the Amazon Basin, the Amazon Boundary Layer Experiment

(ABLE-2B) was conducted during April and May of 1987, during the wet season (Harriss *et al.*, 1990). The experiment was a part of the NASA Global Tropospheric Experiment (GTE), and was the first large atmospheric characterisation experiment held in the Amazon Basin during the wet season.

This study is the first to obtain detailed size distribution of biogenic aerosol particles. These elemental size distribution measurements complement the data obtained using stacked filter unit (SFU) during the ABLE-2B and ABLE-2A experiments (Artaxo *et al.*, 1988, 1990; Artaxo and Maenhaut, 1990). Vertical profiles of the elemental concentration of aerosol particles at ground level, canopy level and the free troposphere were performed, with sampling in a 45 m high tower inside the jungle. Five stage cascade impactors (CI) have collected aerosol particles in five size ranges and a Nuclepore backup filter collects particles smaller than $0.25 \mu\text{m}$. The aerosol source composition as function of particle size was obtained, and also the quantitative source attribution along the vertical profile inside the forest canopy. The elemental composition and size distribution of the land dust and two natural biogenic components were obtained.

2. SAMPLING SITE DESCRIPTION AND METEOROLOGICAL CONDITIONS

The sampling site was located in the central part of the Amazon Basin, at the Ducke Forest Reserve ($2^\circ 57'S$; $59^\circ 57'W$; elevation 120 m above MSL), located about 25 km Northeast of Manaus. A total of 50 cascade impactor samples was collected from March 10 to April 22, 1988. Sampling was performed at three levels on a meteorological tower. The Ducke reserve tower is 45 m high whereas the height of the dense forest canopy is about 35 m. The tower was designed and installed in order to have minimum effect on the surrounding jungle. Platforms at three different levels on this tower were used for aerosol sampling. Near the ground sampling was performed at 1.5 m height. In order to emphasise the contribution from the leaves and the forest canopy, we sampled near the canopy level at 28 m height. At the top level of the tower (45 m) sampling was done about 17 m above the average forest canopy level. The forest around the tower is virgin and undisturbed. At the top level of the tower, 25 CI sets were collected, at the canopy level 10 CI sets, and at the ground level 15 CI sets were collected, making a total of 50 CI samples of 5 stages each plus backup filter.

The dominant wind direction (over 90% of the time) is northeast to east, so the urban plume of Manaus cannot reach the sampling site most of the time. High-resolution (5 min) wind direction and wind velocity measurements from the NCAR PAM stations were also available. As ozone, carbon monoxide, and nitrogen oxides were measured continuously at the Ducke Tower by other groups, it was possible to have

a continuous check if any possible contamination from the urban plume of Manaus was present at the aerosol sampling site. All these indicators together suggested that there were three possible urban air intrusion episodes during the sampling period. The samples corresponding to these events were eliminated from the data set in order to have samples collected exclusively from background air masses. Temperature during the sampling campaign were between 22 and 34°C. High precipitation rate was observed at the Ducke Reserve with 199.1 mm in April and 105.2 mm in May. During April and May, we had only 5 days without precipitation at the sampling sites. Forest burning in the Amazon Basin did not occur during the experiment as observed from satellite pictures and aircraft observations, so the sampled biogenic aerosol component should consist only of natural release from the vegetation and not of emissions from forest burning. Artaxo *et al.* (1993) gives a detailed study of biomass burning emissions in the Amazon Basin.

3. EXPERIMENTAL METHODS

3.1. Sampling procedure

Aerosols were sampled using 5-stage single orifice Batelle-type cascade impactors (CI) (PIXE International Corp., Tallahassee, FL, U.S.A.), which are made of electrically conductive plastic. The CI operated at a flow rate of 1 lpm, and sampling time was typically 24 h. The five-stage cascade impactors have collected aerosol particles with aerodynamic diameters of stage 5: 4 μm ; stage 4: 2 μm ; stage 3: 1 μm ; stage 2: 0.5 μm ; stage 1: 0.25 μm ; and a so-called stage 0 consisting of a Nuclepore backup filter that collects particles smaller than 0.25 μm . The aerosol particles were deposited over a thin (1.5 μm) Kimfol plastic film. To maintain high particle collection efficiencies during sampling, avoiding bounce-off of particles, a coating of vaseline was used in all stages. The Nuclepore backup filter consists of a 0.4 μm pore-size polycarbonated filter. Blanks substrates have followed all the procedures as collected samples. It is important to notice that the values obtained for the aerodynamic diameters might be affected by the ambient relative humidity. It was collected 25 CI at the top level of the tower, 10 CI at the canopy level, and at the ground level 15 CI sets were collected, making a total of 50 CI samples of 5 stages each.

3.2. Elemental analysis

The elemental concentrations were measured by particle-included X-ray emission (PIXE) (Johnson and Campbell, 1988). The samples were irradiated by 2.5 MeV proton beam, supplied by the Pelletron nuclear accelerator of the Department of Nuclear Physics of the University of Lund, Sweden. The following 20 elements were detected in the samples: Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Rb, Sr, Zr, and Pb. The detection limit was typically 1 ng m^{-3} for elements with $Z < 20$, and 0.05 ng m^{-3} for $21 < Z < 30$. The precision of the PIXE analysis was better than 7% for the major elements and about 15% for elements with concentrations near the analytical detection limit.

4. ABSOLUTE PRINCIPAL FACTOR ANALYSIS

In order to separate the different components of natural biogenic aerosols using the elemental com-

position, absolute principal factor analysis (APFA) was used (Thurston and Spengler, 1985; Hopke, 1985; Gordon, 1988). APFA offers the possibility to obtain a quantitative elemental source profile instead of only a qualitative factor loading matrix as in traditional applications of factor analysis. The absolute elemental source profiles help in the identification of the factors and can be used to compare the factor composition with the assumed aerosol sources.

In principal factor analysis 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 (Hopke, 1985). The APFA procedure obtains the quantitative source apportionment 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 component (Thurston and Spengler, 1985).

5. RESULTS AND DISCUSSION

It is important to emphasise that the sampling campaign occurred in May and April, during the wet season, with no biomass burning occurring in the whole Amazon Basin, due to the very high precipitation rate. For the 60 days of the experiments, precipitation was observed in the sampling area in 55 days (Greco *et al.*, 1990). Most of the rain events exhibited a high precipitation rate and lasted for 1 or 2 h. The average rainfall amount was 6.3 mm (day)^{-1} at the Ducke Reserve (Andreae *et al.*, 1990). Because of this high precipitation rate and also from observations at all ground sampling sites, aircraft flights, and satellite images, it was concluded that the biogenic aerosol component in this study is not related to forest burning, but to the natural release of aerosol particles by the vegetation.

Two variants of factor analysis were performed to extract information from the extensive data set. In the first part of the analysis, the individual stages of the CI measured at the three levels of the tower were combined. Through this procedure factors for each size range regardless of the height in the forest canopy were obtained. Later, in the second part of the analysis the individual differences along the three levels of the tower were analysed, with all the size ranges together. The reason for the two procedures was to obtain a

more complete and robust statistical analysis, while obtaining the information regarding both, particle size and height in the forest canopy.

Table 1 presents the average elemental concentration observed for each cascade impactor stage, and the backup filter. The column "Total" represents the sum of the elemental concentration for impactor stages including the backup filter. The average size distribution for the various measured elements shows some of the properties of natural biogenic aerosol. Sulphur, for example, is mainly in the fine mode, at the 0.25 μm size fraction, but also a coarse sulphur component at a diameter of 2.0 μm can be observed. Several elements like Ca, Ni, Cu, Zn, Pb, Br, Sr and Zr have bimodal distributions or are present with almost constant concentrations among the various size fractions. The land dust related elements (Al, Si, Ti, Mn and Fe) are mainly in the coarse mode, as expected. The total aerosol elemental concentration values are similar to the observed during the wet season in other regions of the Amazon Basin (Artaxo *et al.*, 1993). The "Total" sulphur concentration of 115 ng m^{-3} is low compared with other background regions of the world. Also levels of heavy metals are low, as can be observed for total zinc at 1.51 ng m^{-3} .

In order to investigate the different components present for each size fraction, factor analysis was performed. Table 2 presents the VARIMAX rotated factor loading matrix for each of the 5 size fractions and for the total aerosol concentration data. For all 6 factor analyses, only two factors were statistically significant, and the communalities were typically 85% for most of the elements and all the analyses. These two factors were biogenic and mineral dust aerosol particles. For the 0.25 μm size fraction, the biogenic component is the first factor, explaining 53% of the data variability, and is characterized, by high loadings for P, S, Zn, Fe and Sr. The second component is the land dust related particles, explaining 27% of the data variability, with the presence of Al, Si, Fe and Sr. The biogenic component is the most important source of data variability only for the 0.25 μm size fraction, while mineral dust dominates the other coarser size fraction. For most of the factor analysis, the biogenic component is characterised by high loadings for P, S, K, Ca, Zn and Sr. The land dust components are characterised by Al, Si, Ti, Fe, with occasional presence of K, Ca, Mn and Sr. Potassium is present in both biogenic and mineral dust aerosol particles, with amounts depending on the size range. For the 0.25 μm size range K is almost entirely associated with biogenic particles. For the 1.0 μm size range K is mainly associated with mineral dust. For the 2.0 μm size range K is equally distributed in the two factors, while for the coarser fraction (4.0 μm), K is exclusively associated with the biogenic component. Zinc is always associated with biogenic material also observed in other aerosol studies in the Amazon Basin (Artaxo *et al.*, 1988, 1990, 1993b). However, the mechanisms for or the reason for the release of Zn by the vegetation

is not clear. Zn is present in the bulk vegetation at levels of 100 to 160 ppm (Bowen, 1979) a value that is compatible with the aerosol composition that was observed in this study.

Chlorine is a special case for the elements included in the factor analysis presented in Table 2. It is always mostly associated with the mineral dust component, although the presence of chlorine in land dust is very low. The observed chlorine is actually sea-salt derived. Although the sampling site is about 2000 km from the sea coast, it was demonstrated that there is a fast transport of marine aerosol air masses into the interior of the Amazon Basin during the wet season (Greco *et al.*, 1990). The long-distance transport also brings along mineral dust particles that originate in the Sahara desert. Detailed analysis of this long-range transport from the meteorological and chemical point of view can be obtained in Swap *et al.* (1989, 1991).

Absolute principal factor analysis calculations were performed in order to obtain absolute source apportionment for each of the separated components for each element. Table 3 shows the absolute source apportionment for the biogenic component. In this table absolute concentrations in ng m^{-3} associated with the biogenic component is presented. It can be observed from Table 3 that biogenic K, P, S and Zn show a bimodal size distribution. There is a small contribution in this biogenic component from mineral dust related elements like Al, Si, Ti, Fe and Mn. There are two reasons for this presence. First, as elements Si and Mn are also present in the vegetation at significant amounts, being released together with other biogenic elements. Furthermore, due to some joint variability because of meteorological conditions, it is common to observe some co-variability of most of the elements in factor analysis. The boundary layer high over the Amazon Basin during daytime remains at about 600–1200 m, and at nighttime is lower to less than 200 m high. This meteorological forcing produces joint co-variability for all the elements present in aerosol particles.

Table 4 presents calculations derived from Table 3, showing the percentage of each element concentration that was apportioned to the biogenic component. For example, 92.2% of the S concentration in the 0.25 μm size range were apportioned to the biogenic component, whereas K is 99.8% in the biogenic component of this size range. For the total concentration, some elements are present mostly in the biogenic component: P (98.1%), S (86.9%), K (74.4%), Zn (96.6%) and Sr (68.4%). Others have a somewhat split source attribution, like Al (31.7%), Si (26.5%), Ca (45.0%), Ti (34.7%), Mn (33.1%), Fe (27.8%). A special case is chlorine. A significant fraction of chlorine for the various stages (35.1%, 39.2%, 35.5%, 34.7%, 42.9%) was attributed to the biogenic component. The remaining of the Cl is sea-salt derived. This biogenic chlorine was also observed in other Amazon Basin aerosol studies (Artaxo *et al.*, 1993). Chlorine is present in the vegetation at fractions from 0.2% to 0.6%

Table 1. Average aerosol elemental concentrations in ng m^{-3} for each cascade impactor stage in ABL-E-2B. * Backup filter and the five individual stages

Element	<0.25 μm	0.25 μm	0.5 μm	1.0 μm	2.0 μm	4.0 μm	Total
Al	—	2.59 \pm 1.46 (50)	16.5 \pm 20.1 (50)	36.7 \pm 35.9 (50)	55.8 \pm 71.6 (50)	20.9 \pm 21.1 (50)	132 \pm 129
Si	16.0 \pm 13.5 (50)	1.6 \pm 1.2 (50)	34.1 \pm 41.5 (50)	75.6 \pm 79.7 (50)	126 \pm 169 (50)	38.2 \pm 44.1 (50)	276 \pm 297
P	—	1.16 \pm 0.71 (50)	2.19 \pm 2.55 (50)	1.35 \pm 0.63 (50)	6.74 \pm 6.33 (50)	5.75 \pm 5.37 (50)	17.2 \pm 10.7
S	1.23 \pm 0.61 (6)	41.4 \pm 34.3 (50)	35.5 \pm 52.5 (50)	11.2 \pm 8.1 (50)	17.2 \pm 18.3 (50)	10.0 \pm 6.4 (50)	115 \pm 95
Cl	1.75 \pm 0.77 (44)	0.91 \pm 0.57 (13)	4.95 \pm 8.03 (18)	6.17 \pm 8.72 (13)	25.6 \pm 30.8 (13)	18.3 \pm 16.6 (50)	52.2 \pm 54.7
K	0.55 \pm 0.10 (1)	8.59 \pm 7.53 (50)	8.07 \pm 9.54 (50)	7.42 \pm 6.31 (50)	24.3 \pm 23.3 (50)	16.7 \pm 13.7 (50)	65.1 \pm 40.9
Ca	2.29 \pm 0.40 (1)	0.32 \pm 0.29 (12)	3.03 \pm 4.03 (50)	7.22 \pm 7.83 (50)	18.8 \pm 30.7 (50)	10.1 \pm 10.8 (50)	39.3 \pm 47.4
Ti	—	0.15 \pm 0.08 (4)	1.58 \pm 1.82 (50)	2.97 \pm 3.05 (50)	5.19 \pm 6.79 (50)	1.86 \pm 1.62 (50)	11.6 \pm 11.5
V	—	0.49 \pm 0.35 (10)	0.17 \pm 0.03 (2)	—	0.16 \pm 0.01 (2)	0.24 \pm 0.18 (3)	—
Cr	0.62 \pm 0.24 (2)	0.13 \pm 0.04 (3)	0.19 \pm 0.04 (1)	0.084 \pm 0.016 (1)	0.22 \pm 0.04 (1)	—	—
Mn	—	—	0.19 \pm 0.21 (50)	0.43 \pm 0.44 (50)	0.75 \pm 1.05 (50)	0.19 \pm 0.18 (50)	1.56 \pm 1.64
Fe	0.29 \pm 0.21 (11)	0.38 \pm 0.29 (50)	10.0 \pm 12.7 (50)	22.4 \pm 24.7 (50)	39.7 \pm 53.0 (50)	10.3 \pm 11.6 (50)	82.7 \pm 89.8
Ni	0.25 \pm 0.05 (1)	0.20 \pm 0.25 (37)	0.14 \pm 0.12 (8)	0.062 \pm 0.021 (50)	0.099 \pm 0.049 (6)	0.18 \pm 0.12 (10)	—
Cu	0.21 \pm 0.05 (11)	0.099 \pm 0.086 (39)	0.22 \pm 0.11 (4)	0.20 \pm 0.20 (4)	0.84 \pm 1.45 (9)	0.24 \pm 0.31 (31)	—
Zn	0.31 \pm 0.21 (6)	0.35 \pm 0.43 (50)	0.26 \pm 0.41 (50)	0.52 \pm 0.73 (24)	0.27 \pm 0.20 (50)	0.39 \pm 0.29 (50)	1.51 \pm 1.27
Pb	—	0.59 \pm 0.65 (12)	0.73 \pm 1.18 (10)	0.29 \pm 0.22 (6)	0.66 \pm 0.37 (4)	0.55 \pm 0.11 (1)	—
Br	—	0.17 \pm 0.19 (32)	0.19 \pm 0.20 (21)	0.099 \pm 0.032 (9)	0.15 \pm 0.12 (9)	0.11 \pm 0.04 (14)	—
Rb	—	0.11 \pm 0.03 (32)	0.13 \pm 0.04 (6)	0.18 \pm 0.11 (9)	0.29 \pm 0.39 (16)	0.13 \pm 0.04 (15)	—
Sr	—	0.23 \pm 0.13 (50)	0.24 \pm 0.23 (50)	0.22 \pm 0.12 (50)	0.35 \pm 0.34 (50)	0.26 \pm 0.10 (50)	1.29 \pm 0.68
Zr	—	0.15 \pm 0.03 (1)	0.26 \pm 0.10 (8)	0.33 \pm 0.22 (16)	0.53 \pm 0.33 (11)	0.30 \pm 0.12 (16)	—

* Average and standard deviation are shown. Numbers in parentheses are the number of samples in which the concentration of the element was above the detection limit. Only samples with values above the detection limit were used in calculating the averages. Total is the sum of the elemental concentration for all 5 cascade impactor stages plus the backup filter.

Table 2. Factor analysis results for the Amazon Basin Cascade impactor data: VARIMAX rotated factor loading matrices for the two retained factors for cascade impactor stage. * "Biog." denominate the natural biogenic component whereas "mineral dust" indicates the land dust component for each cascade impactor stage

	Aerodynamic diameter												Total			
	0.25 μm			0.50 μm			1.0 μm			2.0 μm				4.0 μm		
	Biog.	Mineral Dust	Biog.	Mineral Dust	Biog.	Mineral Dust	Biog.	Mineral Dust	Biog.	Mineral Dust	Biog.	Mineral Dust		Biog.	Mineral Dust	
Al	(-0.03)	0.93	0.25	0.96	0.93	0.11	0.93	0.35	0.98	0.98	(-0.04)	0.98	(-0.04)	0.98	(0.16)	
Si	(0.08)	0.88	0.30	0.95	0.98	(0.07)	0.92	0.33	0.98	(0.07)	(-0.05)	0.97	(-0.05)	0.97	(0.13)	
P	0.83	(0.22)	0.93	0.31	(0.04)	0.86	(0.04)	0.91	(-0.20)	(-0.07)	0.94	(-0.07)	0.94	(-0.07)	0.85	
S	0.96	(0.08)	0.94	0.31	(0.21)	0.81	0.58	0.76	(0.27)	0.43	0.83	0.43	0.83	0.43	0.79	
Cl	—	—	—	—	0.73	(0.25)	0.74	(0.05)	0.40	0.70	(0.09)	0.70	(0.09)	0.70	(0.18)	
K	0.94	(-0.05)	0.89	0.43	0.93	0.27	0.65	0.70	(0.04)	0.61	0.94	0.61	0.94	0.61	0.73	
Ca	—	—	0.39	0.89	0.96	0.19	0.86	0.43	0.73	0.86	(0.08)	0.73	(0.08)	0.86	0.37	
Ti	—	—	0.32	0.94	0.98	0.15	0.92	0.38	0.98	0.86	(0.03)	0.98	(0.03)	0.97	(0.22)	
Mn	—	—	0.32	0.93	0.97	(0.13)	0.92	0.37	0.91	0.97	(0.16)	0.91	(0.16)	0.97	(0.22)	
Fe	0.67	0.51	0.25	0.96	0.99	0.11	0.93	0.32	0.98	0.98	(-0.02)	0.98	(-0.02)	0.98	(0.16)	
Zn	0.86	(-0.23)	0.96	(0.16)	—	—	0.30	0.70	0.53	(0.12)	0.41	(0.12)	0.41	(0.12)	0.75	
Sr	0.74	0.42	0.91	0.36	0.75	0.49	0.71	0.64	—	0.66	—	0.66	—	0.66	0.62	
P. Var. (%)	53	27	44	53	70	17	58	30	52	54	24	54	24	54	24	

* Factor loadings that are smaller than 3 standard deviations are shown in parentheses. P. Var. (%) is the percentage of the variance explained by each factor.

Table 3. Absolute concentrations in ng m^{-3} apportioned by the APFA procedure to the biogenic aerosol component for each cascade impactor stage

	Aerodynamic diameter					
	0.25 μm	0.5 μm	1.0 μm	2.0 μm	4.0 μm	Total
Al	—	3.19 ± 0.18	10.6 ± 0.91	27.8 ± 1.08	—	41.9 ± 2.7
Si	—	7.66 ± 0.31	17.4 ± 2.1	64.5 ± 3.5	-5.28 ± 1.61	72.9 ± 7.56
P	0.76 ± 0.05	1.51 ± 0.04	1.25 ± 0.05	6.49 ± 0.29	6.88 ± 0.22	16.9 ± 0.81
S	38.1 ± 1.3	29.1 ± 0.8	10.6 ± 0.7	13.9 ± 0.6	7.67 ± 0.47	100.2 ± 6.4
Cl	—	1.74 ± 0.78	2.42 ± 0.84	9.10 ± 2.53	6.36 ± 2.28	22.4 ± 5.4
K	8.58 ± 0.36	5.25 ± 0.14	3.27 ± 0.25	17.9 ± 0.7	16.9 ± 0.7	48.4 ± 1.9
Ca	—	0.89 ± 0.07	2.18 ± 0.26	10.9 ± 1.0	1.37 ± 1.05	17.7 ± 2.5
Ti	—	0.37 ± 0.02	0.86 ± 0.05	2.72 ± 0.08	0.12 ± 0.05	4.04 ± 0.17
Mn	—	0.046 ± 0.003	0.12 ± 0.01	0.38 ± 0.02	0.018 ± 0.010	0.52 ± 0.03
Fe	0.20 ± 0.02	1.95 ± 0.09	5.19 ± 0.39	18.7 ± 0.99	-1.09 ± 0.34	22.9 ± 1.59
Ni	0.21 ± 0.03	—	—	—	—	—
Cu	0.082 ± 0.009	—	—	—	—	—
Zn	0.38 ± 0.03	0.24 ± 0.01	0.35 ± 0.07	0.20 ± 0.02	0.19 ± 0.03	1.46 ± 0.12
Br	0.17 ± 0.02	0.057 ± 0.014	—	0.12 ± 0.02	0.048 ± 0.01	—
Rb	—	—	0.11 ± 0.02	0.17 ± 0.01	0.055 ± 0.011	—
Sr	0.12 ± 0.01	0.14 ± 0.01	0.14 ± 0.01	0.25 ± 0.01	0.12 ± 0.05	0.89 ± 0.04
Zr	—	—	0.17 ± 0.02	0.38 ± 0.04	0.28 ± 0.06	—
Pb	0.46 ± 0.08	—	—	—	—	—

Table 4. Percentage of element mass apportioned on each site to the biogenic aerosol component in the ABLE-2B experiment for the cascade impactor data set

	Aerodynamic diameter					
	0.25 μm	0.5 μm	1.0 μm	2.0 μm	4.0 μm	Total
Al	—	19.4 ± 1.07	28.9 ± 2.5	49.9 ± 1.9	—	31.7 ± 2.1
Si	—	22.4 ± 0.9	23.0 ± 2.8	48.7 ± 2.8	-13.8 ± 4.2	26.5 ± 2.7
P	65.2 ± 4.5	69.2 ± 1.8	92.5 ± 3.7	96.3 ± 4.3	120 ± 4	98.1 ± 4.7
S	92.2 ± 3.3	81.9 ± 2.3	95.2 ± 6.2	81.5 ± 3.8	76.7 ± 4.7	86.9 ± 5.6
Cl	—	35.1 ± 15.8	39.2 ± 13.5	35.5 ± 9.9	34.7 ± 12.4	42.9 ± 10.3
K	99.8 ± 4.2	65.0 ± 1.7	43.8 ± 3.3	73.8 ± 3.0	101 ± 4	74.4 ± 2.9
Ca	—	29.4 ± 2.4	30.3 ± 3.6	58.3 ± 5.5	13.5 ± 10.4	45.0 ± 6.5
Ti	—	23.6 ± 1.1	28.8 ± 1.8	52.3 ± 1.5	6.15 ± 2.72	34.7 ± 1.5
Mn	—	23.9 ± 1.6	28.8 ± 2.9	51.3 ± 2.3	9.75 ± 5.46	33.1 ± 2.0
Fe	53.8 ± 6.2	19.4 ± 0.9	23.2 ± 1.7	47.0 ± 2.5	-10.6 ± 3.4	27.8 ± 1.9
Ni	104 ± 13	—	—	—	—	—
Cu	82.1 ± 8.6	—	—	—	—	—
Zn	109 ± 8	92.8 ± 2.9	66.4 ± 13.3	74.6 ± 5.8	49.4 ± 8.3	96.6 ± 7.8
Br	106 ± 13	30.5 ± 7.7	—	78.9 ± 10.9	44.8 ± 6.5	—
Rb	—	—	63.7 ± 13.3	59.5 ± 3.6	41.6 ± 8.5	—
Sr	53.7 ± 4.5	59.9 ± 2.1	64.7 ± 4.0	69.7 ± 3.1	45.4 ± 8.2	68.4 ± 3.5
Zr	—	—	51.3 ± 6.9	72.1 ± 7.9	91.9 ± 20.4	—
Pb	77.8 ± 12.7	—	—	—	—	—

in mass (Bowen, 1979), so it can be released to the atmosphere in similar ways as the other biogenic elements like P and K. Analysis of the aerosol mass concentration shows that biogenic particle account for 55–95% of the airborne aerosol mass concentrations in the Amazon Basin (Artaxo *et al.*, 1988, 1990).

The second part of the factor analysis calculation involves the analysis of the aerosol composition as a function of height in the tower related to the forest canopy. Aerosol sampling was performed at three levels of the jungle tower; Ground level at 1.5 m height; forest canopy level at 28 m height; and top level of the tower at 45 m height. In this analysis. All cascade

impactor stages were joined together, and a global factor analysis was performed with 250 cases, corresponding to the 50 cascade impactors and the 5 stages. Table 5 presents the VARIMAX rotated factor loading matrix for this factor analysis. Three factors were statistically significant. The first factor is the mineral dust component, with the presence of Al, Si, Ca, Ti, Mn, Fe, Sr and Cl. The second factor is interpreted as a biogenic related component, with the presence of S, Zn and Sr. The third factor represents another biogenic component, with high loadings for P, K, Cl and Sr. The eigenvalue of this third component is a high 1.93, explaining 16.1% of the data variability. The

Table 5. VARIMAX rotated factor loading matrix for factor analysis using all individual cascade impactor stages in the same analysis. A total of 250 samples and 12 variables were analysed. Three factors were statistically significant. The data variability explained by the three factor model was 89.8%*

Element	Factor 1 Mineral dust	Factor 2 Biogenic: S, Zn, Sr	Factor 3 Biogenic: P, K, Cl, Sr	Commun.
Al	0.97	(0.06)	(0.14)	0.97
Si	0.97	(0.05)	(0.15)	0.97
P	(0.13)	(0.13)	0.97	0.97
S	(0.09)	0.91	(0.11)	0.84
Cl	0.71	(0.01)	0.34	0.62
K	0.57	0.29	0.74	0.95
Ca	0.90	(0.18)	(0.21)	0.88
Ti	0.97	(0.18)	(0.15)	0.99
Mn	0.96	(0.16)	(0.15)	0.98
Fe	0.98	(0.05)	(0.14)	0.99
Zn	(0.06)	0.88	(0.11)	0.79
Sr	0.64	0.52	0.39	0.83
Eigenv.	6.78	2.07	1.93	
Perc. Var.	56.5%	17.3%	16.1%	

* The column "Commun." indicates the communalities that express the total percentage of the original variance explained by the three factors for each element.

communalities in the last column of Table 5 show the adequacy of the factor analysis model in this calculation, explaining typically 90% of the data variability for all elements. Due to the high temporal variability, the mineral dust component explains 56.5% of the data variability, whereas the two biogenic components explain 17.3 and 16.1% of the data variability. It is important to notice that it was observed in this analysis two statistically significant biogenic aerosol components. The factor 2 with the presence of S, Zn and Sr is present mainly in the fine mode, whereas the factor 3, with P, K, Cl and Sr is present in the coarse mode. These two biogenic components have different elemental composition. Most of the chlorine is associated with the mineral dust component as discussed previously, but some chlorine in this analysis also appears significantly in the second biogenic component, as already indicated in the APFA calculations presented in Table 4. This second factor analysis shows a more clear separation for the components than the factor analysis for each stage done previously.

With the two factor analysis procedures, it was possible to separate the contributions from each cascade impactor size range and also from each of the three levels in the forest canopy tower. In order to compare the contributions from the several components as a function of size and sampling height in the tower, absolute factor scores were calculated. These factor scores represent the absolute contribution of each sample for the particular factor analysis. They can be analysed from the point of view of size range and also from the point of view of the three levels in the jungle tower. Figure 1 shows the absolute factor scores for each of the three discriminated factors as a function of the aerodynamic diameter. In this plot it is possible

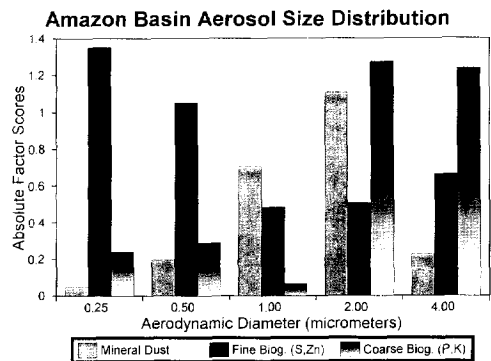


Fig. 1. The size distribution of the discriminated aerosol sources. Absolute factor scores for each of the three factors as function of the aerodynamic diameter.

to observe that the mineral dust component peaks at size ranges of 2.0 μm , with very little contribution for the fine mode at 0.25 and 0.50 μm . The biogenic component rich in S, Zn and Sr is present mainly in the fine mode, for particles less than 1.0 μm , whereas the second biogenic component with dominant presence of P and K is mainly in the 2.0 and 4.0 μm size ranges. The size distributions of the three components are clearly different.

The average factor scores were calculated as a function of sampling height at the three levels in the jungle tower (Fig. 2). The mineral dust component is almost negligible at the ground level, being dominant at the canopy and top levels of the tower. This is expected, since the land dust component is transported mainly over long range and must come from the free atmosphere. Also the forest floor is covered

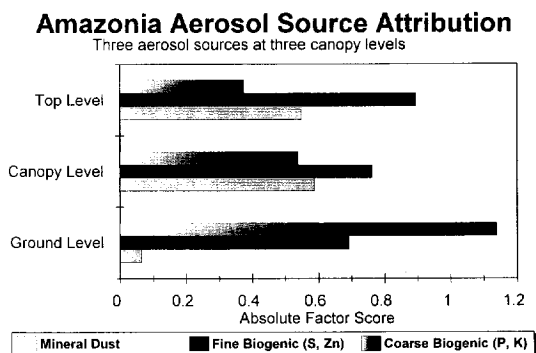


Fig. 2. Vertical distribution of source strength for the three discriminated factors. Average absolute factor scores for each of the three components and each of the three heights in the tower.

with litter consisting mainly of dead leaves and plant debris and is very humid, making it even more difficult to resuspend soil dust particles. The wind velocity under the canopy, at 1.5 m high is always virtually zero, making it difficult to have a significant wind-blown dust component under the forest canopy. The fine mode biogenic component with S and Zn is shown in Fig. 2 as being slightly higher at the top and canopy levels. The coarse mode biogenic component with P and K is dominant at the ground level of the jungle tower. It should represent mainly pollen grains, plant debris, and fragments of leaves. Analysis of individual aerosol particles from the Amazon Basin using nuclear microprobe shows similar compositions for groups of particles. A similar association was observed between P, K, Cl, Zn and other elements (Artaxo *et al.*, 1993b).

6. CONCLUSIONS

The emission of biogenic particles by the Amazon Basin tropical rain forest accounts for a significant part of the airborne elemental aerosol concentration during the wet season. Only two types of particles were identified by factor analysis based on measurements of trace elements by PIXE: dust from long-range transport and biogenic aerosol naturally released by the forest. Sulphates are also present and associated in the fine fraction with the biogenic aerosol component. Measurement at three levels of the Ducke Tower showed that mineral dust is more predominant at the top level of the tower (45 m) and in the canopy level (28 m) than at the ground level (1.5 m), indicating an influx of mineral dust particles from free atmosphere to the forest canopy. The coarse mode biogenic aerosol component (with most of the P and K) exhibits an increasing concentration trend from the top level to the ground level of the tower, indicating that the production of coarse mode biogenic aerosol particles occurs mainly in and under the forest canopy. The fine mode biogenic component

(comprising S, Zn and others) is almost evenly distributed over the height of the forest. Meteorological analysis points to the Sahara desert as the probable source of this long-range transported mineral dust component.

It is evident from the present study that primary and secondary biogenic aerosol particles are responsible for most of the airborne aerosol concentration in the Amazon Basin region. Considering the vast area of tropical rain forest and the concentration measured in this work, biogenic particles should be important in the global aerosol budget and in the global biogeochemical cycles of various elements. Their fine mode component can affect the radiative balance in tropical regions. Due to their organic matrix, they could represent a large number of the cloud condensation nuclei in tropical regions, having the potential to influence the hydrological cycle in tropical regions.

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