

Aerosol Chemistry During the Wet Season in Central Amazonia: The Influence of Long-Range Transport

R. W. TALBOT,^{1,2} M. O. ANDREAE,^{3,4} H. BERRESHEIM,^{3,5} P. ARTAXO,⁶ M. GARSTANG,⁷
R. C. HARRISS,^{1,2} K. M. BEECHER,¹ AND S. M. LI³

The distribution and chemistry of the atmospheric aerosol over the Amazon Basin during the April–May segment of the 1987 wet season are described using ground- and aircraft-based data. Wet season aerosol concentrations and composition are variable in contrast to the remarkably uniform distribution and composition of the predominantly biogenic aerosol that we observed during the 1985 dry season. Four distinct intervals of enhanced aerosol concentration coincided with 3- to 5-day periods of extensive rainfall over central Amazonia. It is hypothesized that a major source of aerosols to the basin was the direct input of northern hemispheric air laden with variable mixtures of Saharan dust, marine aerosol, and possibly biomass combustion products. The enhanced aerosol concentrations over Amazonia were reduced in 1–3 days to 5–10% of their peak levels by large-scale changes in the circulation field with subsequent decoupling from the source region, frequent precipitation, and intermixing of northern and southern hemispheric air masses. The intrusion of northern hemispheric air into the Amazon Basin is linked to the establishment and persistence of the West African Subtropical High (WASH) in a limited region over west Africa. Marine aerosols may be intermixed with the soil dust during transit across the Atlantic or within the sea breeze regime along the northeast coast of South America. It is proposed that a principal source of NO_3^- and SO_4^{2-} associated with the dust is biomass burning south of the Sahara in western Africa.

1. INTRODUCTION

Atmospheric aerosols are important to biosphere-atmosphere cycling of C, N, and S in forest ecosystems. Many reduced N and S gases released to the atmosphere by forest ecosystem metabolic processes are oxidized and subsequently converted to the particulate phase. Vegetation also directly emits particles and volatile organic matter, which is converted into particles via a variety of gas-to-particle conversion pathways. The concentration and chemistry of these biogenic aerosols probably contribute in important ways to regional cloud formation, radiation budgets, and precipitation composition. Vigorous atmospheric circulation in equatorial regions may facilitate exportation of aerosols to the global troposphere.

Few data exist that describe the distribution and chemistry of the atmospheric aerosol over tropical ecosystems. However, west African tropical forests have been found to be a prolific source of natural aerosols, particularly during the wet season [Crozat, 1979; Desalmand *et al.*, 1982]. Decaying vegetation may be a source of small particles which are important nuclei for cloud formation [Schnell and Vali, 1973]. Especially during drier periods, biomass burning is an important source of aerosols to the tropical atmosphere

[Crozat *et al.*, 1978; Cachier *et al.*, 1985; Andreae *et al.*, 1988a] and perhaps the global troposphere [Andreae, 1983; Cachier *et al.*, 1986].

Our measurements indicate that equatorial forests are a source of nutrient-rich particles composed of organic material released from vegetation along with species derived from biogenic gas emissions. During the 1985 dry season in Amazonia, Brazil, we found that organic carbon was the dominant component of the atmospheric aerosol over this extensive tropical wetland and forested region [Talbot *et al.*, 1988]. We also observed a very homogeneous water-soluble aerosol composition on a basinwide scale in the atmospheric mixed layer. On average, we found that the atmospheric concentration of aerosol constituents was enhanced fivefold in mixed layer air over free tropospheric levels. In an earlier aircraft flight over the Guyana tropical forest we observed similar enrichment of aerosol species in mixed layer air [Gregory *et al.*, 1986].

Delmas *et al.* [1978] and Servant *et al.* [1984] observed higher ground-level concentrations of aerosol NO_3^- , NH_4^+ , and SO_4^{2-} in the Ivory Coast tropical forest during the dry season compared to the wet season. In Amazonia, Lawson and Winchester [1978, 1979a, b] found that S, K, and P were associated in ground level coarse particle ($>1 \mu\text{m}$) aerosols and proposed that these aerosols were of biological origin. Later work by Artaxo *et al.* [1988, this issue] during the Global Tropospheric Experiment (GTE) Atmospheric Boundary Layer Experiment (ABLE) 2A and 2B field expeditions has shown that biogenic particles in the atmosphere over the central Amazon region are enriched in K, P, Cl, S, Ca, and Zn.

In this paper we concentrate on the temporal variation in the concentration and chemistry of the atmospheric aerosol over central Amazonia, Brazil, during the 1987 wet season. Our data were obtained during the NASA GTE ABLE 2B expedition conducted in April/May 1987 [see Harriss *et al.*, this issue (a)]. Our wet season results extend those that we obtained during the 1985 Amazonian dry season [Talbot *et*

¹Atmospheric Sciences Division, NASA Langley Research Center, Hampton, Virginia.

²Now at Institute for the Study of Earth, Oceans and Space, University of New Hampshire, Durham.

³Department of Oceanography, Florida State University, Tallahassee.

⁴Now at Max-Planck-Institute for Chemistry, Mainz, Federal Republic of Germany.

⁵Now at School of Geophysical Sciences, Georgia Institute of Technology, Atlanta.

⁶Instituto de Física, Universidade de Sao Paulo, Brazil.

⁷Department of Environmental Sciences, University of Virginia, Charlottesville.

Copyright 1990 by the American Geophysical Union.

Paper number 90JD00639.
0148-0227/90/90JD-00639\$05.00

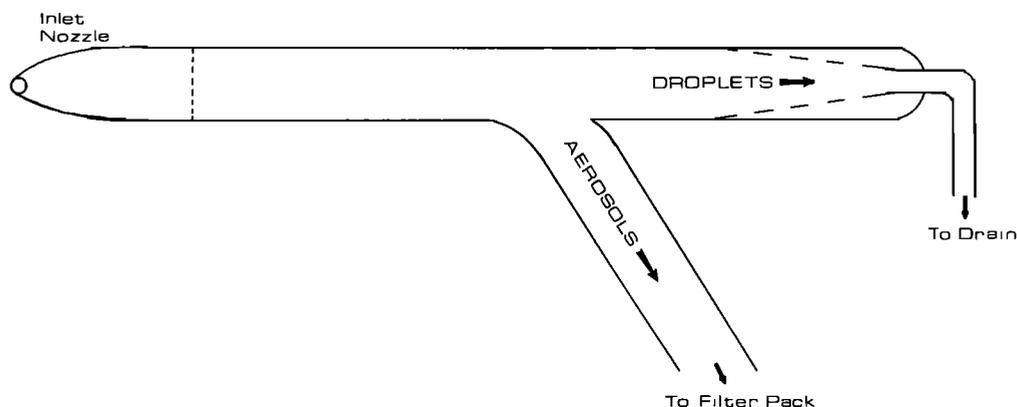


Fig. 1. Schematic representation of atmospheric aerosol sampling system used on NASA Electra research aircraft during ABLE 2B.

al., 1988], as they provide a picture of the atmospheric aerosol regime during extended periods of precipitation which dominate meteorological conditions in central Amazonia from October to June.

2. METHODS

The aircraft flight scenarios and scientific rationale for the GTE ABLE 2B field experiment are described elsewhere [Harriss *et al.*, this issue (a)]. Sampling strategy for atmospheric aerosols depended upon individual mission objectives and actual flight legs flown. In some cases during these missions, samples were combined to ensure collection of sufficient material for reliable chemical analysis. Data reported here were obtained during missions designed to study the chemistry and vertical distribution of species in the low-to-middle troposphere associated with various scales of active convection. On one occasion, weather conditions permitted data to be obtained between Manaus in the central Amazon Basin and Belem on the eastern coast of Amazonia.

Ground-based aerosol sampling was conducted at several sites in the central Amazon Basin [Artaxo *et al.*, this issue]. In this paper we utilize data collected atop the 45-m meteorological tower in the Ducke Forest Reserve (2°56'02"S; 59°57'41"W). The sampler was located about 10 m above the mean height of the forest canopy (35 m). It is important to note that the ground-based data represent total elemental concentrations in contrast to the water-soluble concentrations determined from the aircraft sampling.

2.1. Sampling

The atmospheric aerosol over Amazonia was collected using two systems, one for determination of water-soluble components and the other for organic carbon. These systems were of identical design and consisted of an isokinetic inlet directly coupled to a stacked filter unit. We utilized a thin-walled inlet nozzle with a curved leading edge; the design was based on criteria for aircraft engine intakes at low Mach numbers [Andreae *et al.*, 1988b]. This design avoids distortion of the pressure field at the nozzle tip and the resulting problems associated with flow separation and turbulence. Particles were collected on Gelman A/E glass fiber filters for determination of organic carbon content. For the water-soluble species we utilized a Nuclepore (8 μm)/Zefluor

Teflon (1 μm) 90-mm filter pair to fractionate coarse (>1 μm) and fine (<1 μm) particles [John *et al.*, 1983]. Both systems operated at a sampling flow rate that averaged 175 standard liters per minute; the flow was continuously adjusted to maintain isokinetic sampling. A similar sampling system was used for our ABLE 2A dry season measurements [Talbot *et al.*, 1988].

We believe that our data are representative of the wet season atmospheric aerosol over Amazonia with minimal interference from cloud droplet contamination. Because of the frequency of cloud and rain that we expected to encounter in ABLE 2B, the aerosol intake system was modified so that cloud water droplets would be removed from the sampled airstream (Figure 1). This was accomplished by inertial separation of large droplets from the aerosol population. At the point where the intake tube begins to curve to bring the airstream inside the aircraft, a second tube was connected to the main intake tube and extended back in a straight path from the inlet nozzle. The second tube was of the same diameter initially as the main intake tube (2.5 cm ID), but gradually narrowed to a final internal diameter of about 1 cm. This tube was on the order of 20 cm in length and was coupled to another tube of about 1 cm ID which sloped steeply downward and continued on inside the aircraft. Inside the aircraft clear Tygon tubing continued this flow path through a regulating valve and back out of the aircraft through a drain port. Ideally, cloud water droplets would have too much momentum to make the turn into the aircraft, separate from the main airstream and continue in a straight path down the added "impaction tube." Collected water was simply gravity drained from the impaction tube. The use of clear tubing inside the aircraft permitted us to visually observe this draining process. A slight positive pressure was maintained and continuously monitored inside the combined "impaction-drain" tubing. This overpressure minimized aerosol loss from the main sampling stream through the droplet removal system. In practice this droplet removal system was observed to drain collected water readily and we never observed droplets on our aerosol filters. This droplet removal system allowed us to continue aerosol sampling as the aircraft momentarily encountered light to moderate precipitation. In many instances the aircraft flight path was altered slightly to avoid active precipitation.

Ground-based sampling employed stacked 47-mm Nucle-

pore filters of 8- and 0.4- μm pore size to provide information on the coarse ($>2\ \mu\text{m}$) and fine ($<2\ \mu\text{m}$) particle modes, respectively [Parker *et al.*, 1977]. The flow rate was maintained constant at 14 standard liters per minute. Details of the sampling and bulk elemental analysis using particle-induced X ray emission (PIXE) are given elsewhere [Artaxo *et al.*, this issue].

Particle bounce effects can be a problem with stacked filter sampling regimes. However, these problems should have been minimal under the very humid conditions and extremely low filter loadings ($<100\ \mu\text{g}$ per filter) encountered in this study. Our approach was to collect small amounts of aerosols (due to short sampling times of 15–45 min during flight legs) while minimizing postcollection chemical interactions between acidic fine particles and more alkaline larger materials (e.g., soil or sea salt).

2.2. Chemical Analysis

Aerosols collected for determination of water-soluble components (i.e., Na^+ , NH_4^+ , K^+ , Cl^- , NO_3^- , SO_4^{2-} , $\text{C}_2\text{O}_4^{2-}$, CH_3SO_3^- , HCOO^- , CH_3COO^- , and $\text{CH}_3\text{C(O)COO}^-$) were extracted within hours after collection [Talbot *et al.*, 1986]. Field blanks were put through the same handling procedures as actual samples. Ion chromatographic analysis of the sample extracts was generally completed within 24 hours of sample collection following techniques described elsewhere [Talbot *et al.*, 1986, 1988; Andreae *et al.*, 1988b]. The analytical precision of the ion analysis is 3–4%. Using twice the average standard deviation of the blank values, we estimate that our limits of detection are as follows (molar mixing ratio in parts per 10^{12} (ppt)): Na (25), NH_4 (25), K (7), Cl (18), NO_3 (4), SO_4 (8), C_2O_4 (2), CH_3SO_3 (2), HCOO (7), CH_3COO (10), and $\text{CH}_3\text{C(O)COO}$ (2) for an average sampled air volume of $20\ \text{m}^3$. Based on the propagation of the associated errors, the atmospheric concentrations of water-soluble components have an overall uncertainty of about 20%, with the largest uncertainty derived from variation in filter blank levels (10%). On three occasions we operated tandem sampling systems to evaluate the reproducibility of our determination of the atmospheric concentration of water-soluble species. In each of the comparisons the average difference between species concentrations varied from 12% to 22%.

Atmospheric aerosol was collected on precombusted glass fiber filter (Gelman type A/E) for determination of the total organic carbon content [Talbot *et al.*, 1988]. Samples were frozen immediately after collection and thawed just before analysis. Our limit of detection for total aerosol organic carbon is about 0.25 ppb C (parts per 10^9). Overall uncertainty in the atmospheric concentration of organic carbon is about 15%, and like the soluble components originates primarily from variation in blank levels.

3. RESULTS AND DISCUSSION

3.1. Seasonal Variations in Aerosol Composition

The mean water-soluble ionic composition of the wet season atmospheric aerosol in the mixed layer and free troposphere over central Amazonia is presented in Table 1. It is apparent that organic carbon is the dominant component of the wet season atmospheric aerosol, as it was during the

TABLE 1. Atmospheric Concentration of Selected Aerosol Constituents Over Amazonia

Constituent	Mixed Layer	Free Troposphere	Ratio of Mixed Layer to Free Tropospheric Concentrations
POC	$12,000 \pm 6500$	3000 ± 1300	4.0
NH_4	92 ± 74	37 ± 28	2.5
Na	139 ± 121	27 ± 17	5.2
K	25 ± 16	7.3 ± 5.2	3.4
SO_4	81 ± 55	11 ± 3.5	7.4
NO_3	58 ± 52	11 ± 5.2	5.3
Cl	110 ± 129	16 ± 10	6.9
C_2O_4	8.8 ± 5.0	2.4 ± 1.4	3.7

POC, particulate organic carbon. Atmospheric concentration is stated in ppt. Values are the mean ± 1 standard deviation (mixed layer, $n = 16$; free troposphere, $n = 12$).

dry season [Talbot *et al.*, 1988]. Organic aerosols are known to be released from vegetation directly [Zenchelsky and Youssefi, 1979] and can also form readily in the atmosphere from biogenic volatile organic matter [Duce, 1978; Duce *et al.*, 1983]. Our ABLE 2A and 2B data suggest that copious production of biogenic aerosol takes place during both the dry and wet seasons in Amazonia, Brazil. Cachier *et al.* [1985] observed similar concentrations and seasonal variation of aerosol carbon over west African tropical forests.

The mean chemical composition of the atmospheric aerosol over central Amazonia during the April/May segment of the 1987 wet season is presented in Figure 2. In both mixed layer and free tropospheric air, Cl^- was the dominant anionic aerosol component. Sodium ion was the principal aerosol cation in the mixed layer, but NH_4^+ dominated in the free troposphere. Sulfate and NO_3^- occurred at comparable levels from 0.15 to 5 km altitude, averaging about 70 ppt in the mixed layer and 10 ppt in the free troposphere. For the major aerosol components (Table 1), we observed an average concentration enrichment in mixed layer air of 4.8 times midtropospheric levels at 5 km. These data indicate that the ecosystems comprising Amazonia are an important regional source of some aerosol chemical constituents. Factor analysis of the ground-based aerosol elemental data indicates that the dominant source of the atmospheric aerosol over Amazonia is biogenic particles [Artaxo *et al.*, this issue]. Species enriched in these biogenic particles include K, P, S, Cl, Ca, and Zn. Reduced N and S gases released from the forest ecosystem [Andreae *et al.*, this issue (a); Bakwin *et al.*, this issue] also contribute to NO_3^- and SO_4^{2-} in aerosols over Amazonia.

The seasonal comparison of aerosol composition over central Amazonia is shown in Figure 2 for the 1985 dry season and 1987 wet season. On the average the atmospheric concentration of aerosol constituents was a factor of 2 lower in the wet season than in the dry season. Potassium, NH_4^+ , and the organic ions showed the greatest decrease in mixed layer concentration (mean of 3.6 times lower) from the dry to the wet season. The Cl^- concentration, on the other hand, averaged nearly 4 times higher in the wet season than in the dry season. We did not observe, however, a significant seasonal change in the Na^+ concentration. Chloride and Na^+ showed very interesting temporal variation in the

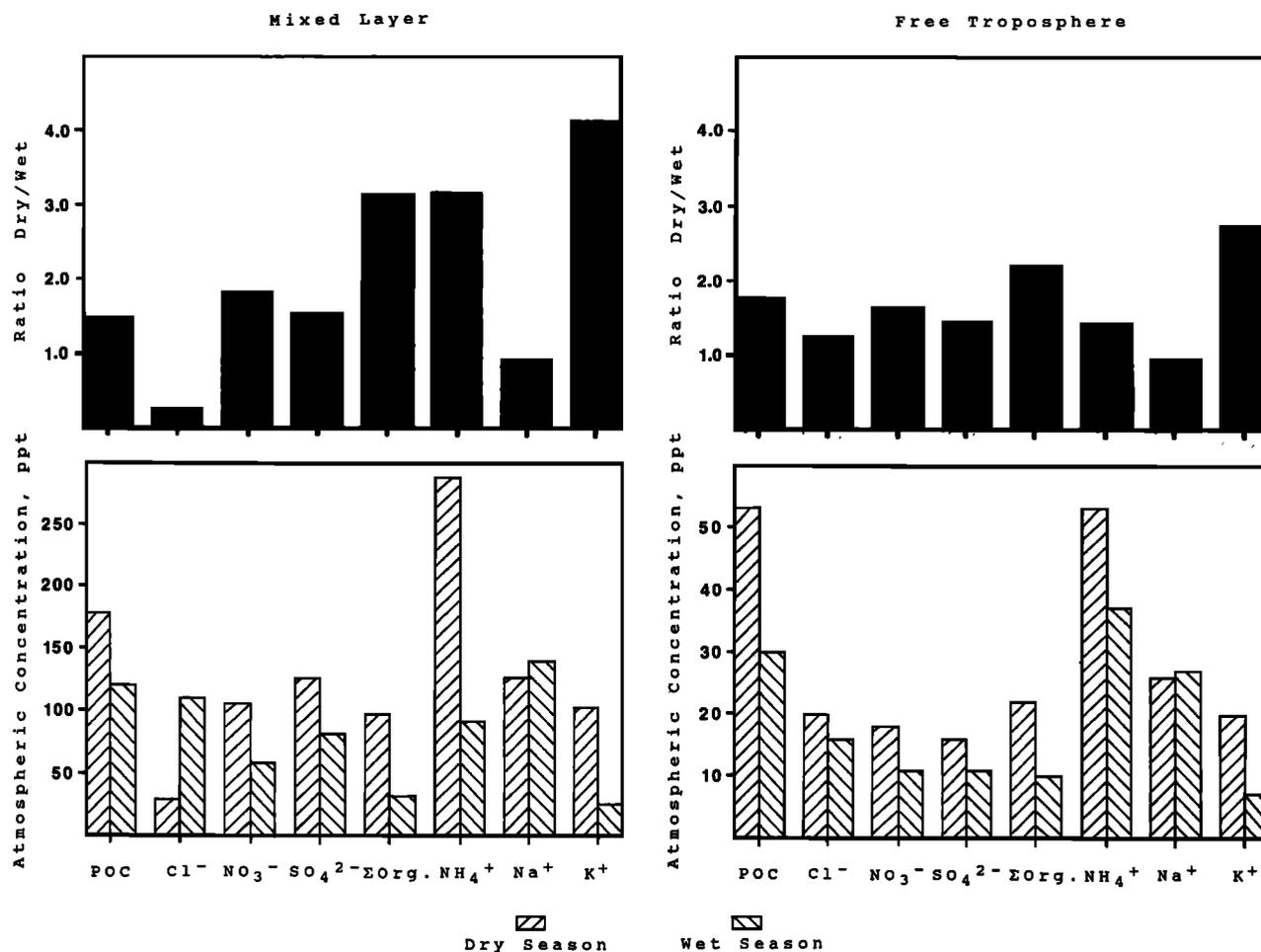


Fig. 2. Comparison of aerosol constituent mean concentrations between the 1985 dry season and 1987 wet season in the mixed layer and free troposphere over central Amazonia, Brazil. The dry-to-wet season concentration ratios are also shown.

mixed layer during our wet season study period. We will describe this behavior in a later section of this paper.

It is apparent from the seasonal comparison presented in Figure 2 that there are major differences in aerosol composition in mixed layer air over the Amazon Basin between the dry and wet periods. To examine these differences more closely, we have summarized the aerosol constituent mean concentrations in terms of seasonal anionic-cationic charge balances (Table 2). During the dry season the ratio $\text{NO}_3^- + \text{SO}_4^{2-}/\text{NH}_4^+$ averaged 1.2, indicating that the aerosol was nearly acid-base neutral, apparently due to an abundant supply of NH_3 and subsequent incorporation of NH_4^+ into the particulate phase [Talbot *et al.*, 1988]. In the wet season the ratio averaged 2.4, which implies that the atmospheric aerosol might be quite acidic. However, our precipitation data suggest that Ca^{2+} and Mg^{2+} are important missing cations in this charge balance [Andreae *et al.*, this issue (b)]. Furthermore, it is unlikely that the wet season aerosol is strongly acidic, since the rain water is less acidic during the wet season (pH 5.3) than during the dry season (pH 4.6).

Figure 3 shows a plot of the relationship between anionic and cationic components in the fine and coarse aerosol fractions in the mixed layer during ABLE 2B. For both aerosol fractions there are several data points that show a large excess of anions compared to cations. The majority of

the data points are much closer to a 1:1 anion to cation ratio. The data indicating large disparity between anions and cations correspond to samples collected on April 13, 20, and 29 and May 7 (missions 4, 9, 16, and 21). For these intervals

TABLE 2. Amazon Basin Aerosol Compositional Summary: Dry Versus Wet Season Comparison in Atmospheric Mixed Layer

	Dry	Percent of Total	Wet	Percent of Total
SO_4	10	47	7.2	45
NO_3	4.4	20	2.6	16
Cl	1.2	6	4.9	31
Σ_{Org}	5.9	27	1.2	8
Σ	21.5		15.9	
NH_4	12	56	4.1	26
Na	5.3	25	6.2	39
K	4.3	20	1.1	7
"X"	≈ 0	≈ 0	4.5	28
Σ	21.6		15.9	

Atmospheric concentrations are stated in nanoequivalents per cubic meter. Values for the dry season were taken from Table 2 of Talbot *et al.* [1988]. Σ_{Org} is composed of oxalate, methyl sulfonate, formate, acetate, and pyruvate. "X" represents the missing ion concentration(s) as estimated from the charge balance (see text).

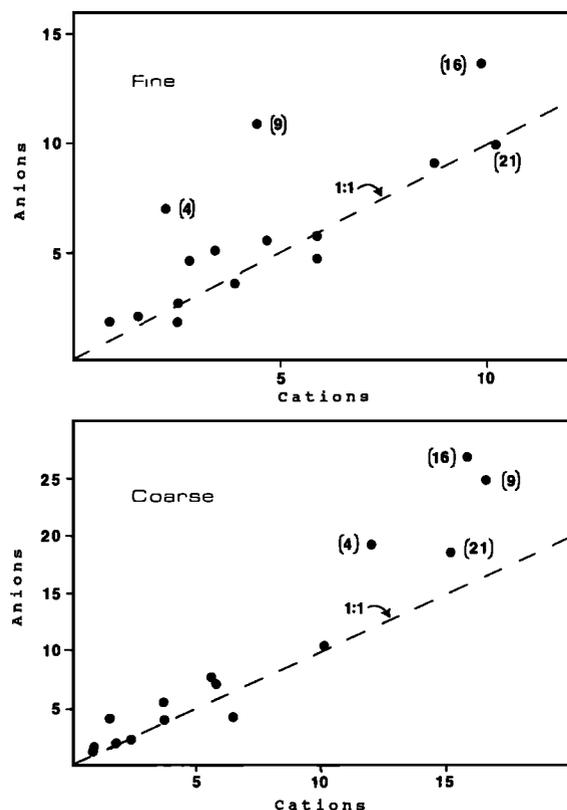


Fig. 3. Anionic-cationic relationship in fine ($< 1 \mu\text{m}$) and coarse ($> 1 \mu\text{m}$) mode aerosols in mixed layer air. Selected mission numbers are indicated in parentheses.

our data show a deficiency in cations which averaged a factor of 2 in the fine fraction and 1.5 for the coarse fraction. The composition of the atmospheric aerosol in mixed layer air over the Amazon Basin during these four periods appears to be quite different from the wet season average aerosol composition.

3.2. Temporal Variability

3.2.1. Aerosol constituents. To examine the periodic perturbations in the wet season aerosol regime, we utilize time series plots for selected species in the atmospheric mixed layer (Figure 4). For the water-soluble aerosol components, large increases in atmospheric concentration were observed on April 13, 20, and 29 and May 7. In Figure 4 we show the temporal variation in coarse particle NO_3^- , SO_4^{2-} , and Cl^- and the corresponding variation in Cl/Na ratio. These data suggest that the dramatic increases in concentration might reflect the transport of marine aerosols into the central Amazon Basin. However, during these episodes the NO_3^- concentration was much greater than background levels typically found over the tropical Atlantic Ocean (about 80 ppt) [Savoie *et al.*, 1989]. During these same periods the concentrations of NH_4^+ and SO_4^{2-} were enhanced 2–5 times in the fine particle fraction (Figure 4). We have examined the aerosol data to see if the apparent temporal variability in aerosol species concentration could be directly related to precipitation dynamics. However, a comparison of aerosol SO_4^{2-} concentration in the mixed layer, for example, with rainfall amount in the central

Amazon Basin during the study period [Greco *et al.*, this issue; Garstang *et al.*, 1990] shows no direct anticorrelation between these two variables.

The ground-based aerosol collections at the Ducke tower site provide a continuous record of mixed layer aerosol concentration and selected elemental total composition from April 8, to May 14, 1987 [see Artaxo *et al.*, this issue]. In Figure 5 the aerosol mass, Cl, and Si concentrations are shown for the April/May study period. For Cl the temporal record mimics what was observed from the aircraft sampling; four periods of greatly elevated concentration compared to very low levels the rest of the time. There sometimes appears to be a time lag of a day between observance of peak concentration of aerosol species at ground level and from aircraft higher up in the boundary layer. This phenomenon is partially an artifact of the ground-based sampling protocol. Sample collection intervals were usually of 24 hours duration which began and ended around 1200 LT. The daily concentrations are plotted in Figure 5 on the day that sampling ended. Also, the geographic area sampled by the aircraft was in many instances displaced 100 km or more from the location of the ground-based sampling site. It is likely then that the aircraft intercepted air masses on certain days that were not directly affecting the ground site at that same time. The aircraft tended to operate in clear areas that were often surrounded by precipitating systems. The patchiness of these precipitating systems in the basin might strongly affect the dynamics of aerosols collected at a stationary point. All of the factors described above probably add to the apparent discrepancies in daily aerosol concentration and composition between our ground-based and aircraft data.

A key to interpretation of the aerosol data was the temporal signal observed for crustal-related elements (e.g., Al, Si, Fe, Ti, Ca). The temporal variation in Si is used in Figure 5 to illustrate the salient features of the crustal (soil dust) signal. With the exception of April 20, the time series trends in Si and Cl are essentially identical. Such a signal of soil dust was not expected during the wet season. During the ABLE 2A dry season experiment the soil dust concentration over central Amazonia was negligible compared to that of biogenic aerosols [Artaxo *et al.*, 1988]. Wet season conditions should greatly suppress any potential regional source of soil dust in Amazonia.

The episodic occurrence of enhanced soil dust concentrations in the wet season most likely represents long-range transport of this material to the central Amazon Basin. The distribution of Si between the fine and coarse fractions indicates that a substantial fraction of the soil dust material was in the submicron particle size range (Figure 5). This evidence supports the hypothesis, discussed in the next section, for a source of this soil dust outside of Amazonia. Saharan dust is known to dominate eolian material in the troposphere over the tropical Atlantic Ocean [Prospero, 1981]. Furthermore, Prospero *et al.* [1981] have shown that large quantities of soil dust are frequently carried out of north Africa across the Atlantic to South America. During ABLE 1, we determined the vertical distribution and chemical composition of intense plumes of Saharan dust in the vicinity of Barbados, West Indies [Talbot *et al.*, 1986]. On one occasion we intercepted the dust near South America just off the coastline of Guyana. Parkin *et al.* [1972] have reported measurements of Saharan dust from shipboard

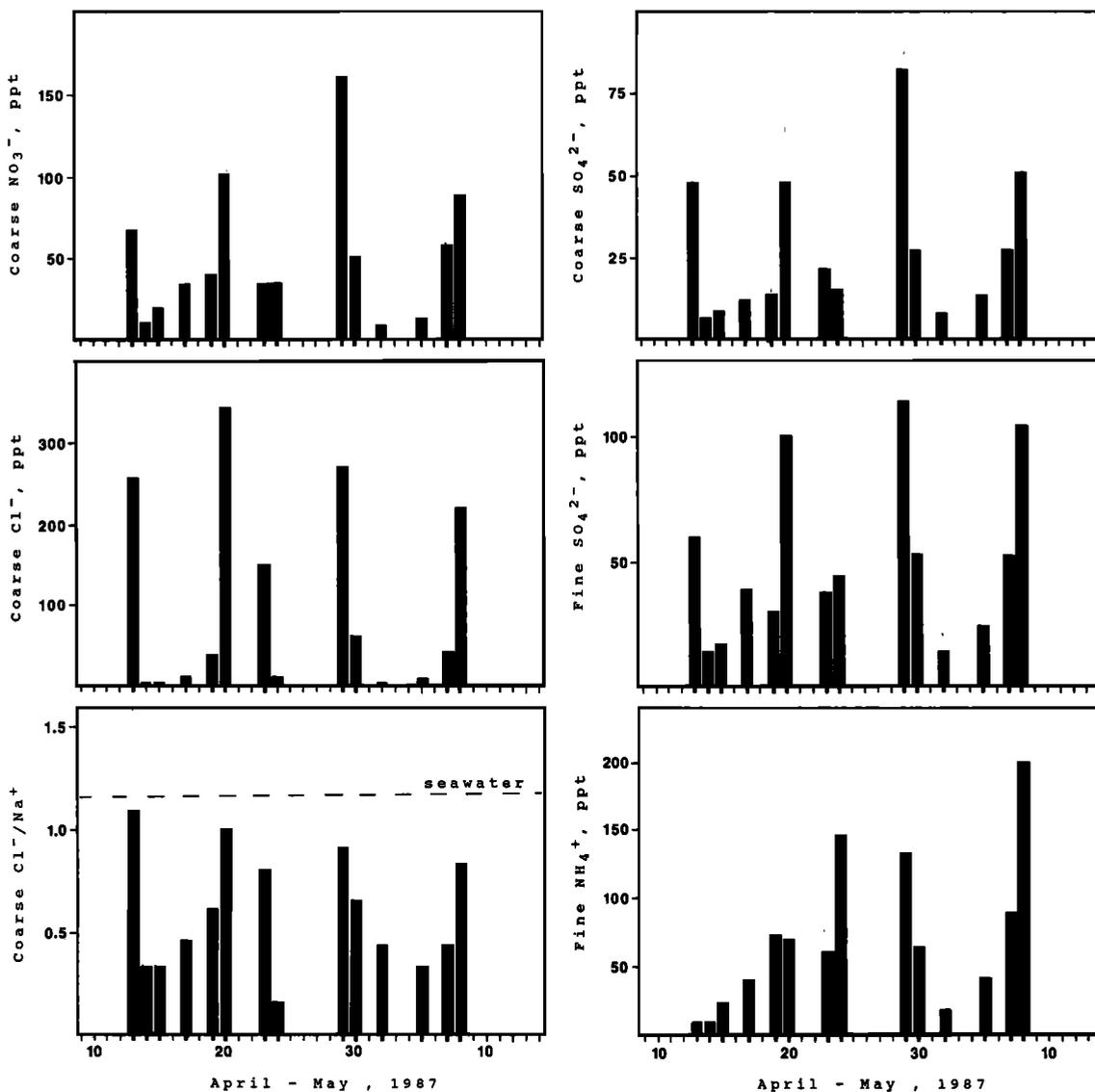


Fig. 4. Temporal variation of selected fine ($<1 \mu\text{m}$) and coarse ($>1 \mu\text{m}$) mode aerosol constituents in mixed layer air. The ratio Cl^-/Na^+ is shown for coarse mode aerosols. The indicated value of 1.16 for Cl^-/Na^+ molar ratio in seawater is based on concentrations reported by Wilson [1975].

collections along a transect from the African coastline across the Atlantic and thence southward to 35°S latitude following the Brazilian coastline. Concentrations of Saharan dust in the marine boundary layer off northern Brazil were in the $1\text{--}25 \mu\text{g m}^{-3}$ range during February–March 1971. Several years of ground-based sampling of aerosols at Cayenne, French Guiana ($4^\circ50'\text{N}$; $52^\circ22'\text{W}$), has shown that the annual maximum importation of Saharan soil dust to South America occurs from February to May [Prospero *et al.*, 1981]. Saharan dust concentrations in excess of $100 \mu\text{g m}^{-3}$ have been observed at Cayenne during the month of March, but values of $20\text{--}40 \mu\text{g m}^{-3}$ are typical. The time interval of maximum dust transport to South America from Africa brackets the ABLE 2B study period in Amazonia.

3.2.2. Meteorological conditions. Climatologically, ABLE 2B was conducted at the end of the wet season. Mean rainfall in the central Amazon Basin for the month of April 1987 suggested that conditions were drier than normal [Greco *et al.*, this issue]. Substantial variability in atmo-

spheric organization can be seen in the production of rainfall as measured at 4 stations over a 1000 km^2 area NNE of Manaus. Figure 6 shows four extended episodes of rainfall during the period April 9, to May 14, 1987. The four major episodes of enhanced aerosol concentration (Figures 4–6) generally coincide with rain events. This coincidence suggests that atmospheric circulations producing the rainfall in the central Amazon Basin might also be responsible for the temporal variations in aerosol composition and concentration.

Examination of the large-scale circulation patterns and cloud fields over the tropical Atlantic and adjacent continents of Africa and South America shows that the relative positions of the lower tropospheric subtropical anticyclones play an important role in both the transport of air into and the organization of rain-producing systems in the Amazon Basin. The combination of the West African Subtropical High (WASH) with ridging over the central tropical Atlantic, often coinciding with a decay of the South Atlantic Subtrop-

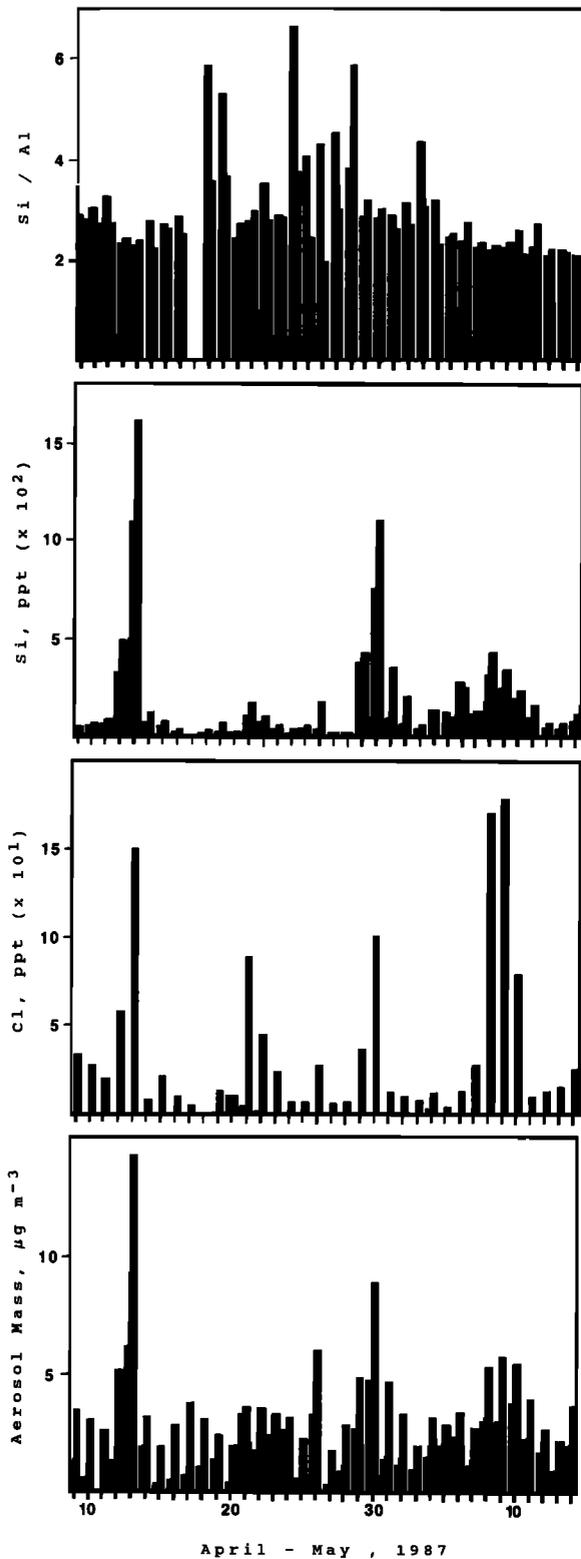


Fig. 5. Temporal variation of selected aerosol characteristics determined at 45-m altitude from micrometeorological tower at Ducke Forest Reserve. For each day we show the concentration in the fine mode (first bar) and the coarse mode (second bar). Chloride was detected in the fine mode in only a few instances.

ical High (SASH), results in flow which may extend from west Africa to the Amazon Basin. Strong low-level onshore (NEly) flow onto the NE coast of the Guianas and Brazil occurs under such circumstances. Horizontal velocity fields

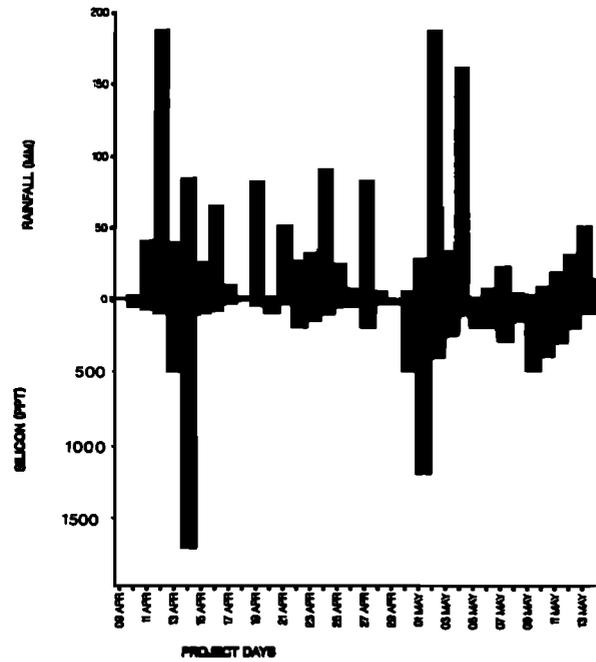


Fig. 6. Mean daily rainfall at four stations within 100 km of Manaus together with aerosol Si concentrations as depicted in Figure 5 for the period April 9, to May 13, 1987.

using the European Center for Medium Range Weather Forecast (ECMWF) global data set together with commercial aircraft and satellite wind observations have been produced by T. Krishnamurti (personal communication, 1989). The data at 850, 500 and 200 mbar for the ABLE 2B experiment were used to determine the streamline fields over the tropical Atlantic, Africa, and South America. The 850- and 500-mbar pressure levels bracket the atmospheric layer where transport of Saharan dust occurs across the tropical North Atlantic [Prospero and Carlson, 1981]. During ABLE 1 we observed massive plumes of Saharan dust over Barbados consistently near the 850-mbar level [Talbot et al., 1986].

The 200-mbar (≈ 10 km) flow during April and May 1987 over the tropical North Atlantic, northern South America, and northwest Africa was persistently westerly. The 500 mbar (≈ 6.5 km) flow showed substantial variation in the location and intensity of WASH and SASH. Figure 7 shows examples of the 500-mbar streamline fields for three days in April 1987. When the center of the WASH was located geographically within the rectangle (5°E to 20°W longitude and 10°N to 25°N latitude) depicted in Figures 7a and 7b, anticyclonic flow at 500 mbar emerged from west Africa and extended across the tropical Atlantic into Amazonia. When the WASH center was displaced from the rectangle (Figure 7c), complex circulation fields existed over the tropical Atlantic with the SASH injecting southern hemispheric air into Amazonia.

Table 3 gives the dates and durations (days) in April and the first half of May 1987 when the WASH was centered geographically in the rectangle (Figure 7) over West Africa. These observations indicate that, once centered in this region, the WASH persisted for continuous multiday time periods. In April and May 1987 this persistence ranged from 4 to 7 days with a persistent 7 days occurring on three out of the five occasions. Under such a persistent flow regime,

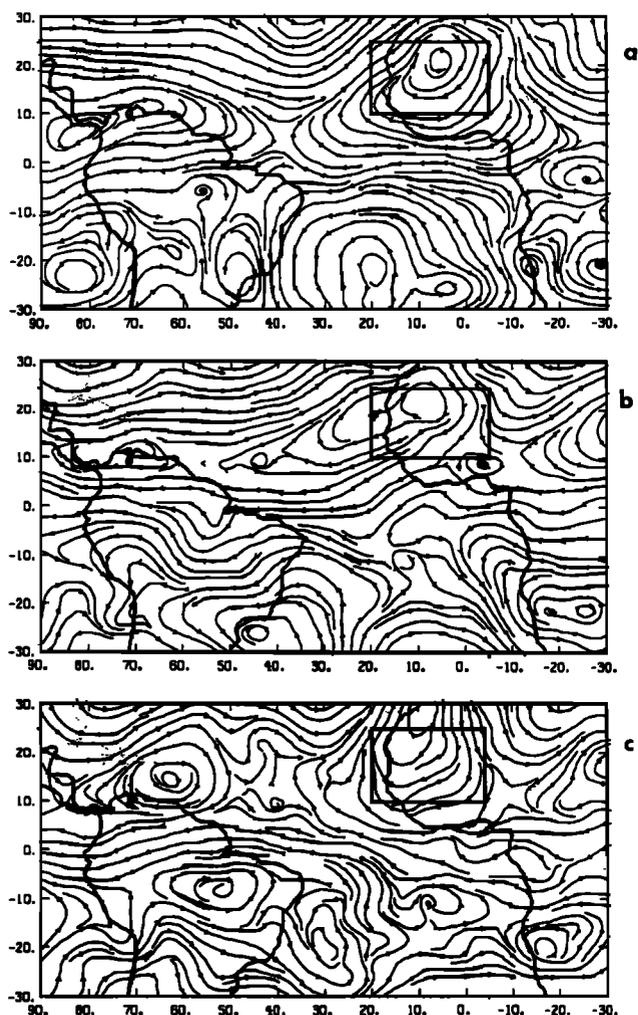


Fig. 7. Streamlines at 500 mbar over the tropical Atlantic and adjacent continents of Africa and South America based on the ECMWF analysis and additional observations incorporated by T. Krishnamurti (personal communication, 1989) at 1200 UT on a 2.5×2.5 latitude/longitude grid. Rectangle over west Africa covers 5°E to 20°W longitude and 10°N to 25°N latitude. (a and b) Days when the geographic center of the 500 mbar WASH is contained in the rectangle and (c) when the WASH center lies outside of the rectangle.

steady state is approached and the streamlines approximate trajectories.

Prospero *et al.* [1981] estimate a transit time of 4–5 days for soil dust from west Africa (Sahara) to cross the ≈ 4000 km

TABLE 3. Occurrences and Durations of the WASH Centered at 500 mbar in the Rectangle 5°E to 20°W Longitude and 10°N to 25°N Latitude (See Figure 7) During the Period April 1, to May 15, 1987

Dates of Occurrence	Duration, days	Dates of Aerosol Si and Cl Concentration Pulses (Figure 5)
April 4–10	7	April 12–14
April 14–17	4	April 21–22
April 21–27	7	April 29 to May 1
April 30 to May 5	6	May 8–10
May 7–13	7	end of observations, May 14

Atlantic fetch to South America. If the establishment and persistence of the WASH control the dust transport, then durations of 4–7 days for the optimal circulation field are reasonable. Dates of pulses of Si and Cl seen in the central Amazon Basin lag the end of such WASH transport phases by 2–4 days (Table 3; Figures 4 and 5).

The 850 mbar (≈ 1.2 km) streamline fields do not show the persistent patterns seen at 500 mbar. The most coherent sequence in the 850-mbar streamline field during ABLE 2B was from April 5–9 (Figure 8). Flow into the central Amazon Basin at 850 mbar varies from NElies (Figure 8a) to easterly (Figures 8d and 8e) to SELies (Figures 8b and 8c). We believe that marine aerosols were advected into the central Amazon Basin primarily below 2-km altitude, whereas the soil dust may have been transported at altitudes up to 3 km. Large-scale convective circulations and subsidence over Amazonia appear to confine the importation of long-range transported aerosols to the boundary layer.

3.3. A Case Study: Mission 16 (April 29, 1987)

3.3.1. *Aerosol chemistry.* The mission 16 flight pattern provided a four-altitude (0.15, 1.2, 1.9, and 4.3 km) profile of aerosol chemistry over a pristine forested region of Amazonia. Flight paths at the two intermediate altitudes were as close as possible to cloud base and cloud top. An inversion was located just above the cloud tops at 2.5 km altitude (Figure 9). Below this altitude the aerosol number density (aerodynamic diameter $>0.12 \mu\text{m}$) was relatively constant and greatly enhanced ($80\text{--}90$ particles cm^{-3}) all the way to down to the lowest flight altitude of 0.15 km. The number density was typically in the range $10\text{--}30$ particles cm^{-3} in the wet season atmospheric mixed layer.

The atmospheric concentrations of soluble aerosol constituents were nearly constant from the ground to 2-km altitude and elevated by factors of 2–4 over typical mixed layer levels. Acetate, which typically was below detection (<10 ppt), was enhanced in concentration up to 30 ppt. The ground-based concentrations of soil dust related species showed a sharp factor of 10 increase from April 29 to May 1 (Figure 5). This large increase in concentration was much greater above the forest canopy in the mixed layer than below the canopy [Artaxo *et al.*, this issue]. Comparison of aircraft and ground-based data for species like K, Na, and Cl showed much higher concentrations at 150 m than at 45 m. These data indicate that soil dust was advected from aloft and that it was not generated by local resuspension.

Using the ground-based data we have computed the mean aerosol concentrations of selected species in the fine and coarse particle fractions between April 29 and May 1. The average background concentration, determined using time periods outside of the intervals of elevated concentration, has been subtracted and the net mean enhancement is given in Table 4 for selected species. The background corrections were generally of the order of 10%, but averaged 30% for K, Ca, and Mg. The atmospheric concentrations reported in Table 4 represent an estimate of the composition of the material input to the central Amazon Basin during the last few days of April 1987. The composition of this material suggests that soil dust is a major component.

If we assume that this material is soil dust, the mass concentration of the various species in the dust can be

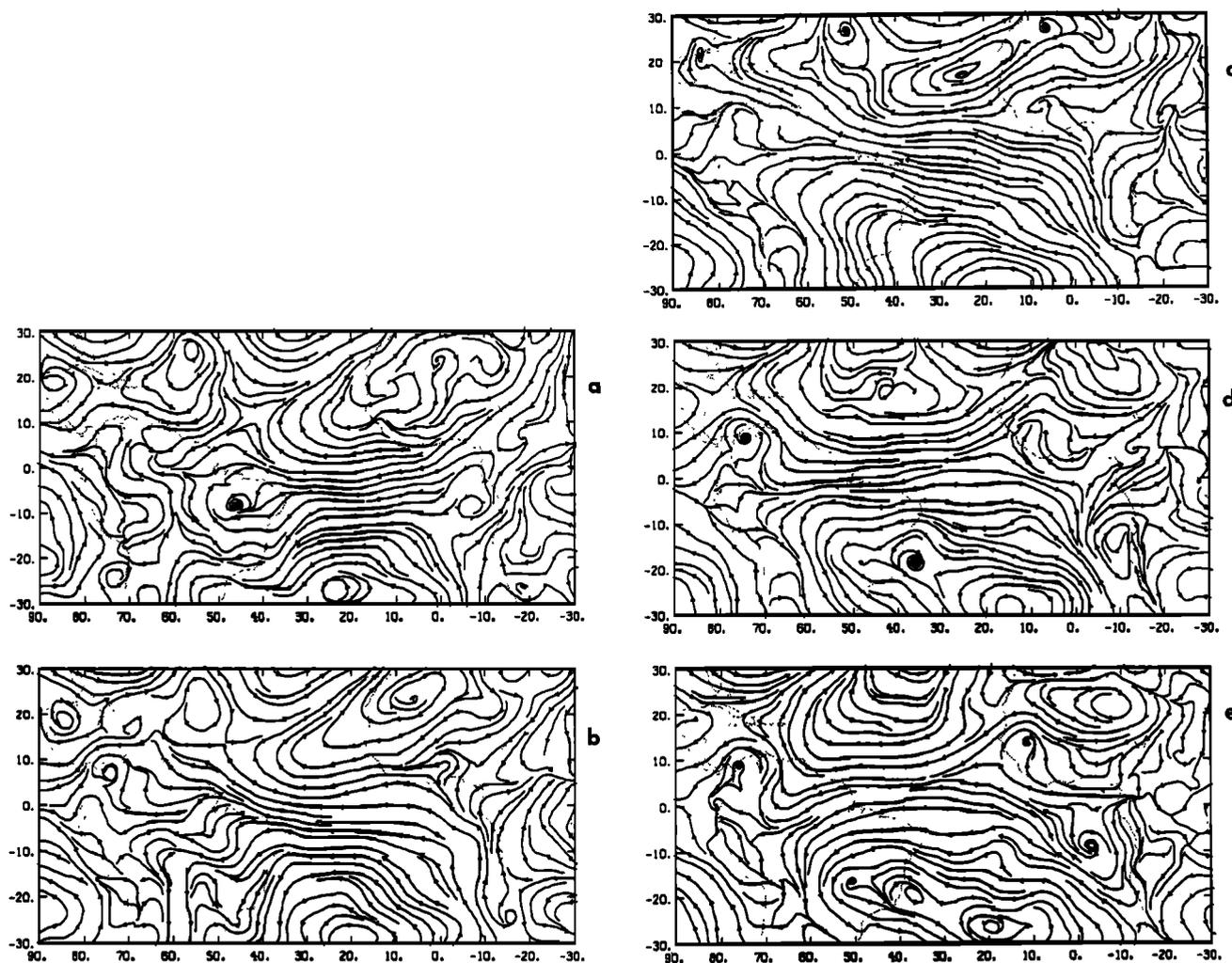


Fig. 8. Streamlines at 850 mbar from the same source as Figure 7: (a) April 5, (b) April 6, (c) April 7, (d) April 8, and (e) April 9, 1987.

estimated using the aerosol mass concentration. As for the elemental constituents, the mean net enhancement in aerosol mass can be calculated for the April 29 to May 1 period. For the coarse fraction we estimate a concentration of $3.19 \mu\text{g m}^{-3}$, and for the fine fraction $1.52 \mu\text{g m}^{-3}$. Using these values for the soil dust concentration, the mass concentrations of selected species were calculated and are shown in Table 4 along with values for average world soils [Bowen, 1966] and Amazonian soils [Kronberg *et al.*, 1979]. There is generally good agreement between our estimated soil dust composition and the average world soil composition, suggesting that a major fraction of the material input to the Amazon Basin was indeed soil dust. However, Amazonian soils are highly weathered, and exhibit a distinct deficit of alkali and alkaline Earth elements compared to average world soils. Hence, the relatively high mass concentration of alkali and alkaline Earth elements in soil dust contained in the mixed layer over Amazonia suggests that this material has its source outside of the Amazon Basin.

The question of whether sea salt is intermixed with the soil dust enhancements over Amazonia is not easily answered. Both the aircraft and the ground-based aerosol data show virtually no detectable concentrations of Cl^- in the fine

particle fraction. Reactions of acidic gases with fine sea salt particles may have volatilized the Cl^- (as HCl) during transport into the Amazon Basin. The Cl/Na molar ratio in coarse particles associated with the late April soil dust enhancement averaged 0.75 from the aircraft observations and 0.80 from the ground-based aerosol collections. These values are 30–35% below the seawater ratio of 1.16 [Wilson, 1975]. Chloride might also have been depleted in coarse sea-salt particles by interactions with acidic atmospheric species. However, ratios of various alkali and alkaline Earth species are at least a factor of 10 higher than their corresponding ratios in seawater (Table 5). Since Na^+ is usually enriched in soil material relative to Cl^- , the intermixing of sea salt and soil materials would dilute the bulk Cl/Na ratio away from the sea-salt value of 1.16.

Based on the composition of average soil, the soil dust related aerosol Cl concentration should be less than 1 ppt. If we assume that all the coarse Cl is from sea salt, then about 35 ppt of the measured 50 ppt for Na (Table 4) must be from sea salt. Essentially the same conclusion is reached utilizing the Na/Si or Na/Al ratio in average soil and the corresponding Al or Si atmospheric concentration. That is, roughly 70% of the Na is attributed to sea salt and 30% to soil dust. Using

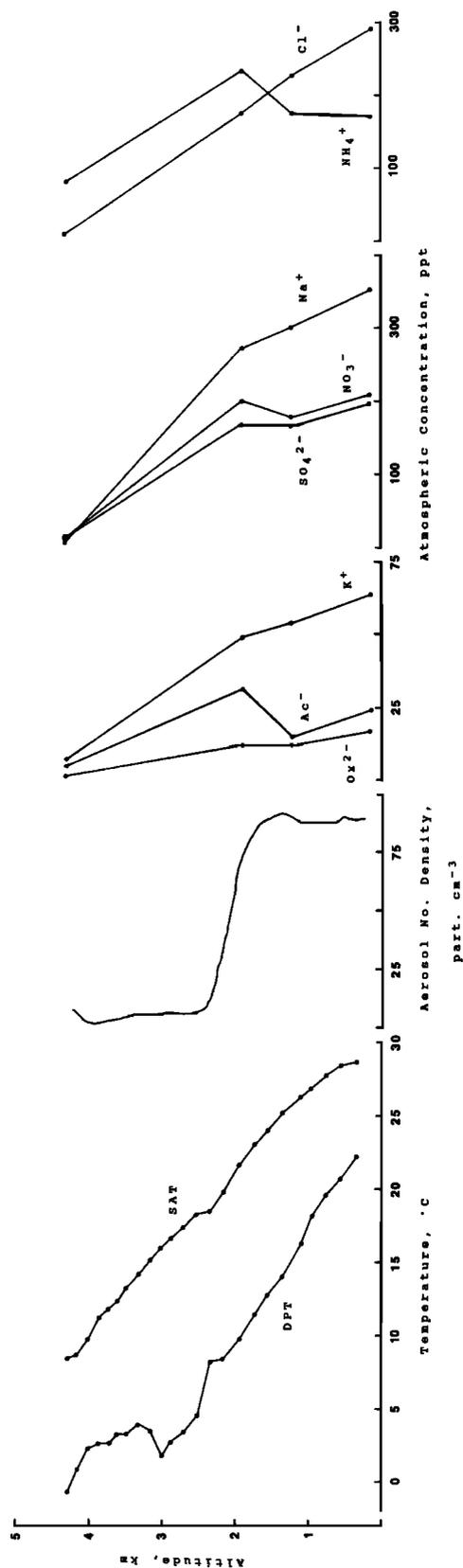


Fig. 9. Vertical distribution of aerosol constituents over pristine central Amazonian forest during mission 16 on April 29, 1987. Dew point (DPT) and static air temperature (SAT) vertical profiles are also shown. The aerosol number density distribution was obtained by Gregory *et al.* [this issue] using a Knollenberg probe sensitive to aerosols with aerodynamic diameters in the range of 0.12–15 μm .

35 ppt for sea salt related Na and a salinity to Na ratio of 3.252 in seawater [Savoie and Prospero, 1982], we estimate an average sea salt concentration of about $0.1 \mu\text{g m}^{-3}$ at ground level for the late April dust episode. An upper limit of $0.15 \mu\text{g m}^{-3}$ is obtained by assuming that all of the Na^+ was marine. For Mg and Ca sea salt can account for 10–15% of their atmospheric concentration. Estimates using values for average soil would put their soil dust contribution in the range of 40–50%. However, due to the variability in soil composition soil dust could easily account for all the nonsea-salt Mg and Ca. Biogenic debris transported with the soil dust also undoubtedly contributed to the measured concentrations of many species.

Our analysis of the ground-based data suggests that soil dust comprises an important fraction of the aerosol mass concentration. The geochemical inputs to the Amazon atmosphere from sea salt are important for Cl^- and Na^+ . Also, the input of marine sulfur (i.e., sea salt plus biogenic) appears to be important to the sulfur budget during the wet season in Amazonia [Andreae *et al.*, this issue (a)].

During April 29 to May 1, sea salt derived SO_4^{2-} should have been insignificant at ground level, being only a few ppt at most. At altitudes from 0.15 to 2 km the Cl^- and Na^+ concentrations were about 5 times higher than their average concentrations determined at 45 m. We have no direct means of determining soil dust concentrations from the aircraft measurements. A simple model would be to scale up the ground-based observations linearly and assume the same relationships.

Selected aerosol samples were collected from the aircraft using a cascade impactor followed by bulk elemental analysis. Unfortunately, problems with the flow measurement instrumentation do not allow accurate atmospheric concentrations to be calculated from these data. The relative amounts of the various species was used, however, for chemical mass balance calculations (P. Artaxo, unpublished results, 1989). The results of this analysis indicated that soil dust accounted for 75–80% of the aerosol mass in the boundary layer during mission 16. Biogenic (plant) particles comprised 15–20%, while sulfates and sea salt contributed a few percent at most. The results of our calculations below are consistent with this predicted aerosol composition.

The average Cl^- concentration below 2 km was 210 ppt during mission 16 (after background 23 ppt subtracted), with 90% of this in the coarse fraction. Attributing all of this to sea salt would distribute the Na^+ as follows; 180 ppt to sea salt and 90 ppt to soil dust of an average Na^+ concentration of 270 ppt (after background 40 ppt subtracted) below 2 km. This distribution of 67% related to sea salt and 33% to soil dust is consistent with our ground-based estimates. The corresponding mass concentration of sea salt particles would be about $0.56 \mu\text{g m}^{-3}$. Assuming that all the Na^+ was derived from sea salt places the upper limit at $0.75 \mu\text{g m}^{-3}$. It appears that <25% of the SO_4^{2-} in the mixed layer was from sea salt. It is not possible to calculate the true concentration of sea salt over Amazonia due to confounding effects of significant Na^+ and Cl^- being derived from other materials such as biogenic debris, the potential loss of Cl^- from sea salt particles by volatilization, and the unknown total/soluble partitioning of Na^+ in soil dust.

A rough estimate of the average soil dust concentration between 0.15- and 2-km altitude can be made by comparing the fraction of the atmospheric Na^+ concentration attributed

TABLE 4. Mean Enhancement in Bulk Elemental Concentrations of Selected Aerosol Species at Ground Level Between April 29, and May 1, 1987

	Al	Si	Fe	Mg	Ca	K	Cl	Na
	<i>Parts per Trillion</i>							
Coarse	193	705	53	31	48	38	40	50
Fine	150	444	33	16	12	26	ND	13
	<i>Micrograms per Gram</i>							
Coarse	68,018	258,629	38,661	9,944	25,129	19,408	18,523	15,014
Fine	110,944	341,836	50,519	10,663	13,184	27,869	ND	8,193
Average soil*	71,000	330,000	38,000	5,000	13,700	14,000	100	6,300
Amazon soil†	106,000	243,000	111,000	200	70	800	—	300

The estimated mass concentrations in soil dust are reported for this same period.

ND, not detected.

*Bowen [1966].

†Kronberg *et al.* [1979].

to soil dust at the higher altitudes with that at 45 m. The ratio of these concentrations (90 ppt at 0.15–2 km/28 ppt at 45 m) is 3.2. The soil dust concentration at 45 m was $4.7 \mu\text{g m}^{-3}$, and assuming a linear increase with Na^+ we estimate a soil dust concentration of $15 \mu\text{g m}^{-3}$ at 0.15–2 km. If all the Na^+ measured at ground level was not soluble, the soil dust concentration estimated above is on the low side. It is likely that this is the case, but we have no means of correcting for this problem. Another estimate of the soil dust concentration can be made by scaling it to the sea-salt concentration estimated for the various altitudes. The sea-salt concentration appears to have increased by a factor of 5.6 from 45 m to the higher altitudes. This same factor would indicate an average soil dust concentration between 0.15 and 2 km of $26 \mu\text{g m}^{-3}$.

For the following calculations, we have chosen a soil dust concentration of $20 \mu\text{g m}^{-3}$ at 0.15- to 2-km altitude. In Table 6 we have estimated the mean water-soluble mass concentration of selected species in the soil dust for mission 16. We used the mean atmospheric concentration between 0.15 and 2 km and corrected it for contribution from sea salt and the average background level during nondust episodes. The estimated soluble mass concentrations appear to be quite similar to the chemical signature that we obtained for Saharan dust during the ABLE 1 experiment in the tropical North Atlantic. One significant difference in these data sets is the (molar) ratio $\text{NO}_3^-/\text{SO}_4^{2-}$. We observed an average value of 0.62 for this ratio in Saharan dust compared to 1.35 estimated here. However, Savoie *et al.* [1989] reported a marked seasonal variation in the $\text{NO}_3^-/\text{SO}_4^{2-}$ ratio associated with Saharan dust transport across the Atlantic. For the

period May 1984 through December 1987, the ratio had an average value of 0.56 during the summer compared to 2.2 during the winter. Our data for Saharan dust were obtained during June 1984 and are consistent with the summer ratio. Our ABLE 2B data for Amazonia fall in the transition period for the $\text{NO}_3^-/\text{SO}_4^{2-}$ ratio associated with Saharan dust. Our estimated soil dust ratio $\text{NO}_3^-/\text{SO}_4^{2-}$ of 1.35 during mission 16 is consistent with an African source region at this time of the year. However, we have no comparable data regarding a potential Brazilian source for the soil dust.

3.3.2. *Meteorology of mission 16.* The last 10 days of April and first 2 weeks of May 1987 were dominated by large mesosynoptic-scale systems of linear orientation which formed along the northeast coasts of the Guianas and Brazil. These Coastal Occurring Systems (COS in Greco *et al.* [this issue]) form in a strong NE trade regime as sea breeze instability lines along the coast. The instability lines propagate inland at $50\text{--}60 \text{ km h}^{-1}$ and grow in the NW-SE directions to over 2000 km in length and 100–200 km in width. On April 26, May 1, and May 3, 1987, three such instability lines moved through the ABLE 2B surface meteorological network NE of Manaus [Garstang *et al.*, 1990].

On April 26 the Intertropical Convergence Zone (ITCZ) was situated along the Amazon River extending from Manaus eastward to Belem on the coast. During the previous several days the ITCZ followed a trend of moving northward and then returning to the central Amazon basin every 24–36 hours. The northern hemisphere subtropical anticyclonic circulation at 850 mbar (Figure 10a) was well established with NE flow across the Guiana-Brazil coastline. The 500-mbar WASH as shown in Table 3 had been well established

TABLE 5. Ratios of Constituents in Soil Dust Associated With April 29 Episode and Comparison to Average World Soils and Seawater

	Cl/Na	K/Na	Mg/Na	Ca/Na	Ca/Mg	Cl/Mg
Coarse	0.80	0.76	1.24	1.92	1.55	0.65
Fine	—	0.50	2.46	1.85	0.75	—
Average soil*	0.010	1.31	1.50	2.49	1.66	0.0069
Seawater†	1.16	0.0218	0.227	0.0439	0.194	5.14

Quantities are given in $\mu\text{eq x}/\mu\text{eq Na}^+$ or Mg^{2+} .

*Calculated from values of Bowen [1966].

†Wilson [1975].

TABLE 6. Estimated Water-Soluble Mass Concentration of Selected Species in Soil Dust Over Amazonia and Comparison to Saharan Dust

Species	Amazon Basin Soil Dust	Tropical Atlantic Saharan Dust*
Na	4,300	3,704 (2,370)
K	2,750	1,663 (829)
NH ₄	4,950	1,796 (1,204)
NO ₃	20,650	5,132 (4,641)
SO ₄	23,600	12,887 (8,902)
Ox	1,150	658 (399)
Ac	2,450	843 (882)

Concentrations are stated in $\mu\text{g g}^{-1}$. Ox represents oxalate ion ($\text{C}_2\text{O}_4^{2-}$) and Ac acetate ion (CH_3COO^-).

*From Table 3 of Talbot *et al.* [1986]. Value in parentheses is one standard deviation from the mean ($N = 6$).

since April 21. On April 26 (Figure 10b) the WASH was linked across the Atlantic with two other anticyclones. The anticyclone often present over NE Brazil was displaced southwestward by the northern hemisphere inflow. By April 27 the ITCZ showed a strong southwest to northeast orientation across the Amazon Basin with its axis being near Manaus. Full disk images (GOES-east) illustrate the positioning of the ITCZ and the propagation of instability lines over the next 48 hours (Figure 11). Mission 16 took place just as the first of the next series of instability lines approached Manaus. Suppressed conditions ahead of the line produced a marked haze layer and inversion at 2.5-km altitude (Figure 9).

Mission 17, a night flight on April 30, encountered considerable weather as part of the line which produced large

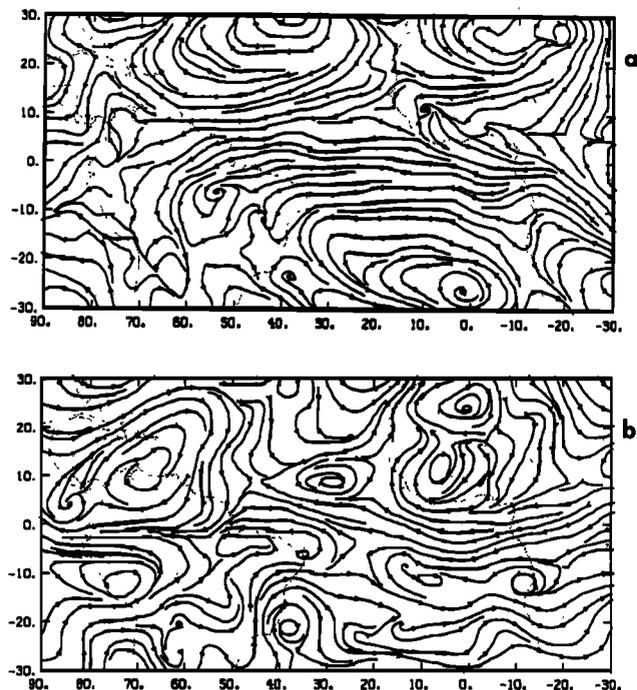


Fig. 10. Streamlines at 850 mbar from the same source as Figure 7: (a) April 26, 1987, showing strong inflow onto the NE Guyana coast and (b) amplification of the E-W wind field (u) components and intensification of convergence over the eastern Amazon Basin (ITCZ).

rainfall on May 1 in the Manaus area. During mission 17, severe alterations to the flight paths were made to avoid precipitating systems. The aerosol data showed no clear evidence of the soil dust enhancements that we had observed the previous day. Apparently, precipitation and air mass changes had effectively removed the soil dust from the atmospheric column in our sampling region.

The meteorological conditions for the period April 29 to May 2 were consistent with an input of Saharan dust at the 500 mb level. Strong large-scale subsidence over the Amazon Basin appears to have effectively trapped the dust below 3-km altitude. The aerosol vertical distribution on April 29, 1987, as depicted by the UV differential absorption lidar (DIAL) [Browell *et al.*, this issue], showed very low concentrations above 3-km altitude as is characteristic of subsidence conditions. Significant inputs of marine aerosol in the boundary layer (1–2 km) can be expected in the pulses of strong onshore NE trades and instability lines occurring near the end of April. However, precipitation and recycling of water in transit from the coast to the central Amazon Basin would rapidly deplete the marine aerosols particularly in the region above the mixed layer.

3.3.3. *Additional evidence for an African soil dust source.* We have utilized images obtained from the European Space Agency satellite Meteosat to look for geographic regions influenced by large-scale aerosol enhancements in the troposphere. We searched the visible aerosol channel images for the period of the ABLE 2B experiment. Periodic outbreaks of Saharan dust over the equatorial Atlantic are clearly evident during the April–May 1987 time frame. One of the largest outbreaks occurred around April 23. The Meteosat image obtained between 1430 and 1500 UT on April 23 is shown in Figure 12. Essentially the entire tropical North Atlantic troposphere from Africa to northern South America appears to be affected by inputs of Saharan dust. A large intense plume appears to be moving off the west coast of Africa on April 23. The transit time for the dust across the ≈ 4000 -km Atlantic fetch to South America is 4–5 days [Prospero *et al.*, 1981]. This would indicate that the dust plume leaving Africa on April 23 would be near the South American coast around April 27–28. The advection of northern hemispheric air into the Amazon Basin the last few days of April would probably bring Saharan dust into the central Amazon region. Prominent elevations in the concentration of Saharan dust were observed at Barbados, West Indies, 24–48 hours prior to the April–May 1987 dust episodes in central Amazonia (D. L. Savoie, personal communication, 1989). Coincident with the dust at Barbados were elevated concentrations of NO_3^- and SO_4^{2-} in proportions similar to those that we observed during the Amazonia dust episodes. The Meteosat images showed no aerosol plumes associated with the arid region of southeastern Brazil. Together these data strongly suggest that the soil dust over Amazonia was of west African origin.

3.4. Hypothesized Wet Season Aerosol Regime

We hypothesize that basin-scale changes in aerosol composition during the wet season are controlled by large scale tropospheric circulation patterns. Artaxo *et al.*'s [this issue] data clearly show the signature of natural biogenic particles released by Amazonian vegetation. This biogenic source

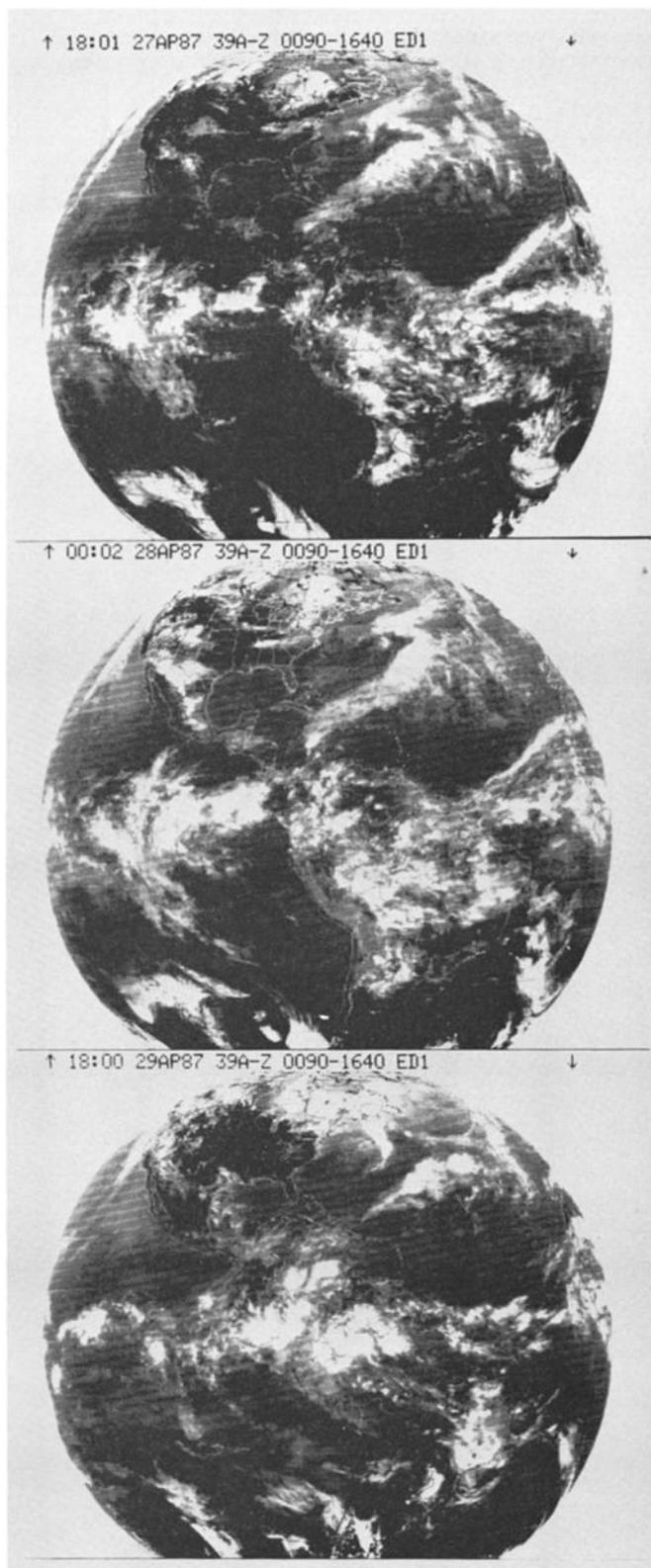


Fig. 11. GOES images depicting large-scale meteorological regime and configuration of the ITCZ leading up to the soil dust intrusion into central Amazonia in late April 1987.

appears to provide an aerosol of consistent composition both in the dry and wet seasons. Superimposed upon this aerosol in the wet season atmosphere is a varying mixture of soil dust and marine aerosol. This mixture appears to infiltrate

the lower troposphere over the Amazon Basin when planetary scale circulation fields allow the northern hemisphere subtropical anticyclone to pump Saharan dust laden air into the region. Marine aerosols may be intermixed with the soil dust during transit across the Atlantic or within the sea breeze regime along the northeast coast of South America and Brazil. *Artaxo et al.* [this issue] estimate that the soil dust concentration over Amazonia was 5.7 times higher in the fine mode and 2.4 times higher in the coarse fraction during the ABLE 2B wet season experiment than during the ABLE 2A dry season study period. The Si/Al ratio in soil dust over Amazonia was essentially constant during the entire ABLE 2B study period (Figure 5), suggesting that the soil dust over Amazonia was derived primarily from a single dominant source. The wet season aerosol scenario appears to be characterized as a natural biogenic aerosol with small amounts of soil dust intermixed. Periodically, this regime receives a new pulse of soil dust and marine aerosol. The soil dust concentration is quickly reduced in 1–3 days to 5–10% of its peak value by large-scale changes in the circulation field with subsequent decoupling from the source region, frequent precipitation, and possible intermixing of northern and southern hemispheric air masses. This scenario would explain the anionic-cationic perturbations that we observed. Indeed, *Savoie and Prospero* [1982] reported that significant amounts of Ca^{2+} are soluble from Saharan dust. During the dry season we observed a remarkably constant aerosol composition on a basinwide scale. At this time of the year the southern hemisphere subtropical anticyclone dominates and the ITCZ is too far north to permit Saharan dust intrusions into the Amazon Basin. If the soil dust originated from Brazil, we would have expected a strong influence from this source during the dry season.

The source of the NO_3^- and SO_4^{2-} associated with the soil dust is uncertain, but *Savoie et al.* [1989] hypothesized that biomass burning in Africa contributes significantly during this time of the year. Most of the NO_3^- was transported into Amazonia in association with coarse particles, and may reflect surface uptake of HNO_3 by alkaline soil dust material. African biomass burning emissions appear to be nitrogen rich [*Delmas, 1982*]. Our measurements of biomass burning emissions in Brazil also support the idea that these combustion derived aerosols should be a nitrogen rich source [*Andreae et al., 1988a*]. Such a source input might also explain the enhanced CO levels in the basin coincident with the soil dust episodes [*Harriss et al., this issue (b)*]. Detailed trajectory analysis along with additional field measurements are needed to resolve these issues.

The regional source of reduced N and S gases from soils and wetlands in central Amazonia appears to be significantly lower in the wet season than in the dry season [*Andreae et al., this issue (a)*; *Bakwin et al., this issue*]. In contrast, the wet deposition fluxes of N and S in central Amazonia appear to be similar for the two seasons [*Andreae et al., this issue (b)*]. Our results support the speculation made by *Reichhoff* [1986] that long-range atmospheric transport of aerosol constituents into the Amazon Basin may have important biogeochemical implications.

4. CONCLUSIONS

We have hypothesized that aerosol chemistry in Amazonia during the wet season is strongly influenced by long-range transport of soil dust, marine aerosol, and possibly

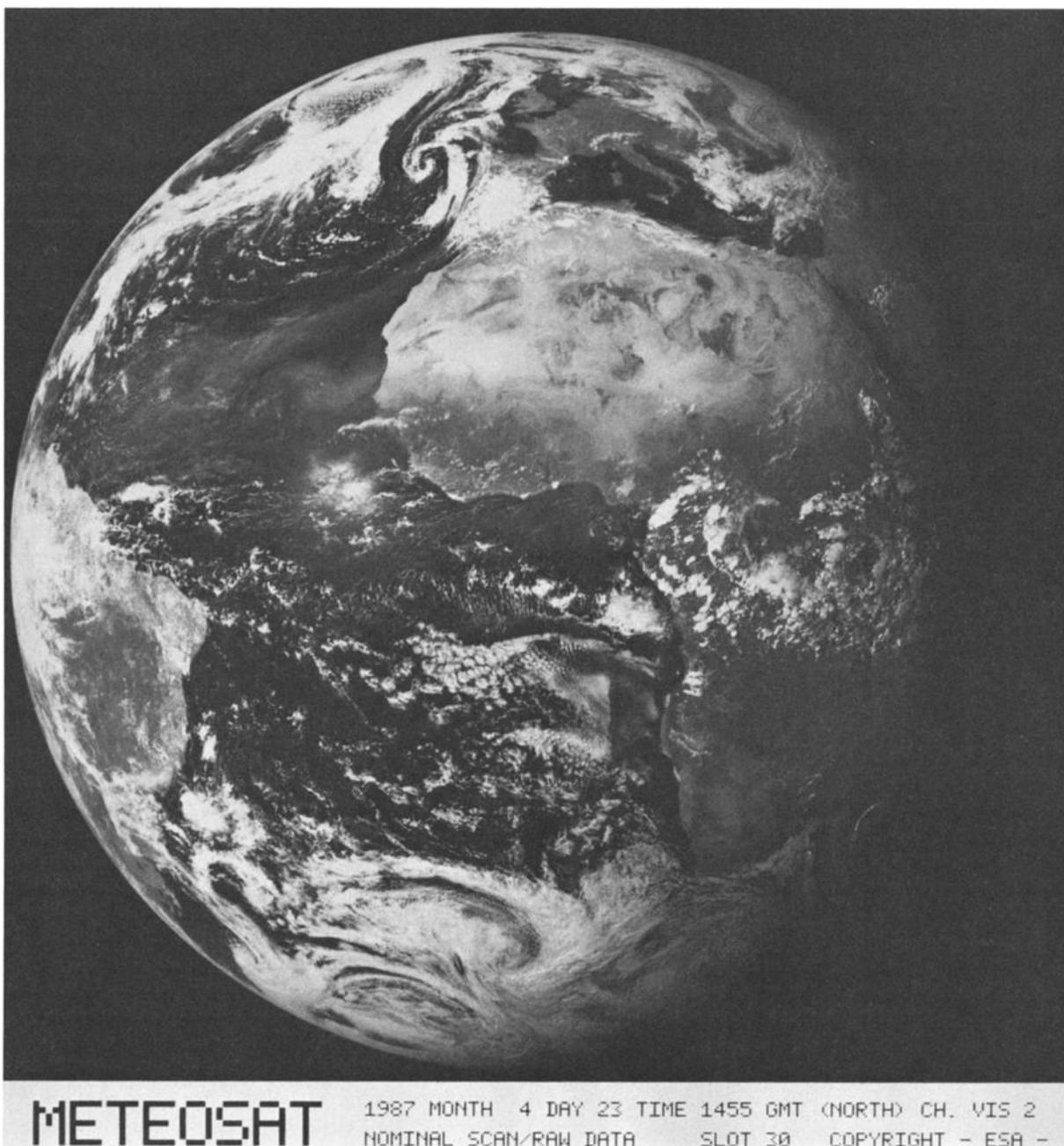


Fig. 12. Meteosat image obtained with the visible channel on April 23, 1987, showing large-scale intrusion of Saharan dust over the tropical Atlantic between west Africa and South America. An intense plume of Saharan dust can be seen leaving the African coast heading west out over the Atlantic. The Meteosat image was supplied by the European Space Agency.

biomass combustion products advected into the central Basin by large scale tropospheric circulation. This scenario produces periodic pulses of material input to boundary layer air over Amazonia. The resultant wet season aerosol regime is a dynamical one, and is very much in contrast to the remarkably uniform distribution and composition of natural biogenic aerosols during the dry season. Although the source of the NO_3^- , SO_4^{2-} , and NH_4^+ associated with the dust episodes is uncertain, they may originate from biomass burning on the African continent nearly 5000 km away.

Acknowledgments. We thank the government of Brazil for permission to conduct research in Amazonia and acknowledge the collaboration of the Brazilian science team under the leadership of J. R. B. Coelho. The cooperation of the flight crew of the NASA Electra research aircraft and the staff of the NASA Wallops Flight Center is gratefully acknowledged. We thank D. L. Savoie for providing data on the daily concentration of Saharan dust at Barbados during April–May 1987. This research was supported by the NASA Tropospheric Chemistry Program. P. Artaxo acknowledges W. Maenhaut for the PIXE analysis and useful discussions, and grant 85/3566-3 from “Fundacao de Amparo a Pesquisa do Estado de Sao Paulo.”

REFERENCES

- Andreae, M. O., Soot carbon and excess fine potassium: Long-range transport of combustion-derived aerosols, *Science*, **220**, 1148–1151, 1983.
- Andreae, M. O., et al., Biomass-burning emissions and associated haze layers over Amazonia, *J. Geophys. Res.*, **93**, 1509–1527, 1988a.
- Andreae, M. O., H. Berresheim, T. W. Andreae, M. A. Kritz, T. S. Bates, and J. T. Merrill, Vertical distribution of dimethylsulfide, sulfur dioxide, aerosol ions, and radon over the northeast Pacific Ocean, *J. Atmos. Chem.*, **6**, 149–173, 1988b.
- Andreae, M. O., H. Berresheim, H. Bingemer, D. Jacob, and R. Talbot, The atmospheric sulfur cycle over the Amazon Basin, 2, Wet season, *J. Geophys. Res.*, this issue (a).
- Andreae, M. O., R. W. Talbot, H. Berresheim, and K. M. Beecher, Precipitation chemistry in central Amazonia, *J. Geophys. Res.*, this issue (b).
- 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.*, this issue.
- Bakwin, P., S. C. Wofsy, S.-M. Fan, M. Keller, S. Trumbore, and J. M. da Costa, Emission of nitric oxide (NO) from tropical forest soils and exchange of NO between the forest canopy and atmospheric boundary layers, *J. Geophys. Res.*, this issue.
- Bowen, H. J. M., *Trace Elements in Biochemistry*, 241 pp., Academic, San Diego, Calif., 1966.
- Browell, E. V., G. L. Gregory, R. C. Harriss, and V. W. J. H. Kirchoff, Ozone and aerosol distributions over the Amazon Basin during the wet season, *J. Geophys. Res.*, this issue.
- Cachier, H., P. Buat-Menard, 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., P. Buat-Menard, M. Fontugne, and R. Chesselet, Long-range transport of continentally-derived particulate carbon in the marine atmosphere: Evidence from stable carbon isotope studies, *Tellus*, **38B**, 161–177, 1986.
- Crozat, G., Sur l'émission d'un aerosol riche en potassium par la forêt tropicale, *Tellus*, **31**, 52–57, 1979.
- Crozat, G., J. L. Domergue, J. Baudet, and V. Bogui, Influence des feux de brousse sur la composition chimique des aerosols atmosphériques en Afrique de l'Ouest, *Atmos. Environ.*, **12**, 1917–1920, 1978.
- Delmas, R., On the emission of carbon, nitrogen and sulfur in the atmosphere during bushfires in the intertropical savannah zones, *Geophys. Res. Lett.*, **9**, 761–764, 1982.
- Delmas, R., J. Baudet, J. Servant, and Y. Baziard, Mise en évidence des sources naturelles de sulfate en milieu tropical humide, *Tellus*, **30**, 158–168, 1978.
- Desalmand, F., J. Baudet, and R. Serpolay, Influence of rainfall on the seasonal variations of cloud condensation nuclei concentrations in a sub-equatorial climate, *J. Atmos. Sci.*, **39**, 2076–2082, 1982.
- Duce, R. A., Speculations on the budget of particulate and vapor phase non-methane organic carbon in the global troposphere, *Pure Appl. Geophys.*, **116**, 244–273, 1978.
- Duce, R. A., et al., Organic material in the global troposphere, *Rev. Geophys.*, **21**, 921–952, 1983.
- Garstang, M., et al., The Amazon Boundary Layer Experiment (ABLE 2B): A meteorological perspective, *Bull. Am. Meteorol. Soc.*, **71**, 19–32, 1990.
- Greco, S., R. Swap, M. Garstang, S. Ulanski, M. Shipham, R. C. Harriss, R. Talbot, M. O. Andreae, and P. Artaxo, Rainfall and surface kinematic conditions over central Amazonia during ABLE 2B, *J. Geophys. Res.*, this issue.
- Gregory, G. L., et al., Air chemistry over the tropical forest of Guyana, *J. Geophys. Res.*, **91**, 8603–8612, 1986.
- Gregory, G. L., E. V. Browell, L. S. Warren, and C. H. Hudgins, Amazon Basin ozone and aerosol: Wet season observations, *J. Geophys. Res.*, this issue.
- Harriss, R. C., et al., The Amazon Boundary Layer Experiment: Wet season 1987, *J. Geophys. Res.*, this issue (a).
- Harriss, R. C., G. W. Sachse, G. F. Hill, L. O. Wade, and G. L. Gregory, Carbon monoxide over the Amazon Basin during the wet season, *J. Geophys. Res.*, this issue (b).
- John, W., S. Hering, G. Reischl, G. Sasaki, and S. Goren, Characteristics of Nuclepore filters with large pore size, II, Filtration problems, *Atmos. Environ.*, **17**, 373–382, 1983.
- Kronberg, B. I., W. S. Fyfe, O. H. Leonardos, and A. M. Santos, The chemistry of some Brazilian soils: Elemental mobility during intense weathering, *Chem. Geol.*, **24**, 211–229, 1979.
- Lawson, D. R., and J. W. Winchester, Sulfur and trace element concentration relationships in aerosols from the South American continent, *Geophys. Res. Lett.*, **5**, 195–198, 1978.
- Lawson, D. R., and J. W. Winchester, Sulfur, potassium, and phosphorus associations in aerosols from South American tropical rain forests, *J. Geophys. Res.*, **84**, 3723–3727, 1979a.
- Lawson, D. R., and J. W. Winchester, Atmospheric sulfur concentrations and characteristics from the South American continent, *Science*, **205**, 1267–1269, 1979b.
- 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.
- Parkin, D. W., D. R. Phillips, R. A. L. Sullivan, and L. R. Johnson, Airborne dust collections down the Atlantic, *Q. J. R. Meteorol. Soc.*, **98**, 798–808, 1972.
- Prospero, J. M., Eolian transport to the world ocean, in *The Oceanic Lithosphere*, vol. 7, *The Sea*, edited by C. Emiliani, pp. 801–874, John Wiley, New York, 1981.
- Prospero, J. M., and T. N. Carlson, Saharan air outbreaks over the tropical North Atlantic, *Pure Appl. Geophys.*, **119**, 677–691, 1981.
- Prospero, J. M., R. A. Glaccum, and R. T. Nees, Atmospheric transport of soil dust from Africa to South America, *Nature*, **289**, 570–572, 1981.
- Reichhoff, J. H., Is Saharan dust a major source of nutrients for the Amazonian rain forest?, *Stud. Neotropical Fauna Environ.*, **21**, 251–255, 1986.
- Savoie, D. L., and J. M. Prospero, Water-soluble potassium, calcium, and magnesium in the aerosols over the tropical North Atlantic, *J. Geophys. Res.*, **85**, 385–392, 1982.
- Savoie, D. L., J. M. Prospero, and E. S. Saltzman, Non-sea-salt sulfate and nitrate in trade wind aerosols at Barbados: Evidence for long-range transport, *J. Geophys. Res.*, **94**, 5069–5080, 1989.
- Schnell, R. C., and G. Vali, Atmospheric ice nuclei from decomposing vegetation, *Nature*, **236**, 163–165, 1973.
- Servant, J., R. Delmas, J. Rancher, and M. Rodriguez, Aspects of the cycle of inorganic nitrogen compounds in the tropical rain forest of the Ivory Coast, *J. Atmos. Chem.*, **1**, 391–401, 1984.
- Talbot, R. W., R. C. Harriss, E. V. Browell, G. L. Gregory, D. I. Sebach, and S. M. Beck, Distribution and geochemistry of aerosols in the tropical North Atlantic troposphere: Relationship to Saharan dust, *J. Geophys. Res.*, **91**, 5173–5182, 1986.
- Talbot, R. W., M. O. Andreae, T. W. Andreae, and R. C. Harriss, Regional aerosol chemistry of the Amazon Basin during the dry season, *J. Geophys. Res.*, **93**, 1499–1508, 1988.
- Wilson, T. R. S., Salinity and the major elements of sea water, in *Chemical Oceanography*, vol. 1, 2nd ed., edited by J. P. Riley and G. Skirrow, pp. 365–413, Academic, San Diego, Calif., 1975.
- Zenchelsky, S., and M. Youssefi, Natural organic atmospheric aerosols of terrestrial origin, *Rev. Geophys.*, **17**, 459–463, 1979.

M. O. Andreae, Max-Planck-Institute for Chemistry, Postfach 3060, D-6500 Mainz, Federal Republic of Germany.

P. Artaxo, Instituto de Fisica, Universidade de Sao Paulo, Sao Paulo, Brazil.

K. M. Beecher, Atmospheric Sciences Division, NASA Langley Research Center, Hampton, VA 23665.

H. Berresheim, School of Geophysical Sciences, Georgia Institute of Technology, Atlanta, GA 30332.

M. Garstang, Department of Environmental Sciences, University of Virginia, Charlottesville, VA 22903.

R. C. Harriss and R. W. Talbot, Institute for the Study of Earth, Oceans and Space, University of New Hampshire, Durham, NH 03824.

S. M. Li, Department of Oceanography, Florida State University, Tallahassee, FL 32306.

(Received July 18, 1989;
revised March 9, 1990;
accepted March 9, 1990.)