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The Atmospheric Component of Biogeochemical Cycles in the Amazon Basin

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Tropical forests, with their high biological activity, have the potential to emit large amounts of trace gases and aerosol particles to the atmosphere. The accelerated development and land clearing that is occurring in large areas of the Amazon basin suggest that anthropogenic effects on natural biogeochemical cycles are already occurring (Gash et al. 1996). The atmosphere plays a key role in this process. The tropics are the part of the globe with the most rapidly growing population, the most dramatic industrial expansion and the most rapid and pervasive change in land use and land cover. Also the tropics contain the largest standing stocks of terrestrial vegetation and have the highest rates of photosynthesis and respiration. It is likely that changes in tropical land use will have a profound impact on the global atmosphere (Andreae 1998, Andreae and Crutzen 1997). A significant fraction of nutrients are transported or dislocated through the atmosphere in the form of trace gases, aerosol particles, and rainwater (Keller et al. 1991). Also the global effects of carbon dioxide, methane, nitrous oxide, and other trace gases have in the forest ecosystems a key partner. The large emissions of isoprene, terpenes, and many other volatile organic compounds could impact carbon cycling and the production of secondary aerosol particles over the Amazon region. Vegetation is a natural source of many types of aerosol particles that play an important role in the radiation budget over large areas (Artaxo et al. 1998).

There are 5 major reservoirs in the Earth system: atmosphere, biosphere (vegetation, animals), soils, hydrosphere (oceans, lakes, rivers, groundwater), and the lithosphere (Earth crust). Elemental cycles of carbon, oxygen, nitrogen, sulfur, phosphorus, and other elements interact with the different reservoirs of the Earth system. The carbon cycle has important aspects in tropical forests due to the large amount of carbon stored in the tropical forests and the high rate of tropical deforestation (Jacob 1999).

In Amazonia there are two very different atmospheric conditions: the wet season (mostly from November to June) and the dry season (July-October) (see Marengo and Nobre, this volume). Biomass burning emissions dominate completely the atmospheric concentrations over large areas of the Amazon basin during the dry season (Artaxo et al. 1988). In the wet season, a very clean atmosphere shows very low concentrations for most of trace gases and aerosol concentration (Artaxo et al. 1990).

Trace Gases Relevant to the Biogeochemical Cycles in Amazonia

Nitrogen and oxygen account for 99% of the Earth's atmosphere. Their concentrations have stayed nearly constant over the

last several hundred million years. Anthropogenic influences are altering the concentration of most of the other relevant gases, which occur in only trace amounts. Carbon dioxide (CO_2) is chemically inert in the troposphere, with a lifetime of about 10^5 years. Methane (CH_4) has an atmospheric lifetime of about 9.6 years and is chemically active and emitted in large amounts from the flooded forest, from swamps, and through biomass burning. Carbon monoxide (CO) is emitted through incomplete combustion and in soil processes, whereas oxides of nitrogen (NO, NO_2, N_2O) are also emitted as a consequence of biogenic processes in soils and through biomass burning (Warneck and Zellner 1999). The tropics present the highest concentrations of the radical OH, because of the high water vapor and UV radiation (Andrea and Crutzen 1997). As OH is responsible for most of the removal mechanism of O_3, CH_4, CO, and other trace gases from the atmosphere, it influences significantly tropical trace gas concentrations (Jacob 1999, Brasseur et al. 1999). Tropical savannah soils are important sources of many trace gases such as CO_2, CH_4, N_2O, and NO (Sanhueza et al. 1994). After emissions some of these gases are chemically transformed under the very active tropical atmosphere.

Carbon dioxide (CO_2) is the most important trace gas in the forest ecosystem, and it is subject to strong and complex biogeochemical interactions (Houghton et al. 1998). CO_2 participates in respiration and photosynthesis and it is the driving force behind most of the biological processes in the soil and vegetation. Several short-term recent experiments show an accumulation of carbon in the forest ecosystem. More long-term measurements and better integration of carbon flux measurements with other ecosystem and climatic measurements are needed to better address the issue of whether tropical forest is a sink or a source of CO_2 to the atmosphere. Soil respiration is also an important source of CO_2, and the average soil respiration rate depends on soil moisture and temperature. Biomass burning influences significantly soil respiration rates for the cerrado and savanna ecosystems (Matson and Harriss 1995).

Methane (CH_4) has important soil, water, and vegetation emissions, and it is the second most important greenhouse gas after CO_2. Biomass burning of cerrado and tropical forest is an important global source of methane. The CH_4 emission rates for soils depend strongly on soil moisture. Large positive and negative fluxes were observed for savannas in Africa, depending on flooding condition and ecosystem type. Termite and enteric fermentation in large herbivores are also a significant source of methane (Jacob 1999).

Globally, the oxides of nitrogen, NO (nitric oxide), NO_2 (nitrogen oxide), and N_2O (nitrous oxide), are key species involved in the chemistry of the troposphere and stratosphere. NO and N_2O are produced mostly by microbial soil activity, whereas biomass burning is also an important source of NO. Nitric oxide is a species involved in the photochemical production of ozone in the troposphere, is involved in the chemical production of nitric acid, and is an important component of acid precipitation. Nitrous oxide plays a key role in stratospheric ozone depletion and is an important greenhouse gas, with a global warming potential more than 200 times that of CO_2.

Carbon monoxide (CO) strongly influences the concentration of the radical OH in the tropical atmosphere. CO oxidation can lead to either production or destruction of ozone, depending on the NO_x mixing ratio. Tropical soils are either a sink or a weak source of CO, where photochemical oxidation of methane and other hydrocarbons and biomass burning emissions are the predominant CO sources.

Tropospheric ozone (O_3) is a gas that has no direct emission sources and is the third most important greenhouse gas after CO_2 and CH_4. The lifetime of O_3 in the

atmosphere is on the order of days to weeks, leading to a highly variable temporal and spatial distribution. The chemical precursors of O_3 are hydrocarbons (including CH_4 and nonmethane hydrocarbons-NMHCs), CO, and the oxides of nitrogen (NO_x). NO_x play a particularly important role in the ozone budget, since their abundance determines if the photochemical oxidation of hydrocarbons and CO results in net O_3 production or destruction. During the biomass burning period in Amazonia, ozone concentrations reach 60-90 ppb and may actually become toxic to vegetation. Concentrations of ozone in Amazonia during the wet season are very low, in the range of 8-12 ppb at midday.

NMHCs are an important emission category for tropical forests. Guenther et al. (1995) estimate that vegetation is the source of over 90% of all NMHC in the global atmosphere and Amazonia is the largest contributor. The reason for this large natural biogenic emission is still unknown. This category includes isoprene, terpenes, and hundreds of volatile organic compounds. There are large variations in emission rates for different plant species and different landscapes. Some recent studies suggest that a plant emits isoprene as part of the leaf temperature regulation mechanism. Terpenes are know to form aerosol particles via efficient mechanisms and are thus responsible for a significant part of the total organic aerosol mass. Several NMHCs have mass fractions in the aerosol and gas phases (Hewitt 1998, Andreae and Crutzen 1997).

Biomass burning is an important global source of hydrogenated species (e.g. CH_4, NMHCs, CH_3Cl, and CH_3Br), as well as other trace gases such as CO (Crutzen and Andreae 1990). These species are produced predominantly in smoldering conditions characterized by insufficient O_2 supply. Methyl bromide and methyl chloride are important in stratospheric ozone depletion. Ozone secondarily produced by biomass burning emissions is observed in regions very far from the emissions, such as tropical Pacific and southern Atlantic oceans.

The Role of Aerosols in Biogeochemical Cycles in Amazonia

Aerosol particles are important to the Amazon ecosystem because of several roles they play in the atmosphere and in biogeochemical cycles. Aerosol particles are responsible for the airborne transport of phosphorus, calcium, sulfur, nitrogen compounds, and other essential nutrients. Aerosols also act as cloud condensation nuclei affecting the cloud formation mechanisms (Rogers et al. 1992). Aerosols affect regional and global climate through their radiative properties, reducing the amount of solar radiation that reaches the ecosystem. The forests emit large amounts of aerosol particles to the atmosphere as part of their natural metabolism. During the wet season, when no biomass burning occurs, aerosol concentrations for particles less than 10 μm are about 10-20 $\mu g/m^3$. Aerosol particles in forest regions are also formed as secondary products from natural emissions of biogenic hydrocarbons, such as terpenes. A large fraction of the aerosol mass (70-85%) is in the form of organic matter, transporting significant amounts of carbon in the form of many different organic compounds.

Vegetation has long been recognized as an important source of both primary and secondary aerosol particles. Forest vegetation is the principal global source of atmospheric organic particles, and in a tropical forest natural vegetation plays a major role in airborne particle concentrations. Only a few studies of natural biogenic aerosols from vegetation in tropical rain forests have been undertaken (Artaxo et al. 1988, 1990, 1994, 1998, Echalar et al. 1998). Natural biogenic aerosols consist of many different types of particles, including pollen, spores, bacteria,

algae, protozoa, fungi, fragments of leaves, excrement, and fragments of insects. This aerosol component is mainly in the coarse-size fraction (dp > 2 μm). The mechanisms of particle emission are still not well understood, but probably include mechanical abrasion by wind, biological activity of microorganisms on plant surfaces and forest litter, and plant physiological processes such as transpiration and guttation. These process-

Table 3.1 Elemental composition of natural biogenic particles in the Amazon basin*.

	Alta Floresta Wet Season				Cuiabá Wet Season			
	Fine Mode		Coarse Mode		Fine Mode		Coarse Mode	
Variable	Mean	Standard	Mean	Standard	Mean	Standard	Mean	Standard
Mass**	5.5	3.5	16.4	9.4	3.9	1.8	11.7	5.7
BC**	0.66	0.41	-	-	0.72	0.33	-	-
Mg	103	110	103	105	-	-	-	-
Al	27	23	230	240	33	30	407	270
Si	74	57	270	230	96	67	970	590
P	4.6	7.4	86	56	2.3	1.4	18	13
S	170	130	140	103	170	110	65	39
Cl	2.3	1.3	41	24	1.03	0.93	14.3	9.1
K	94	74	270	180	80	44	200	100
Ca	8.0	7.2	44	28	12.0	6.5	180	120
Ti	2.1	1.7	17	15	3.4	2.6	36	26
V	-	-	1.05	0.76	0.49	0.43	1.6	1.4
Cr	5.5	3.2	2.21	-	1.2	2.3	1.16	0.88
Mn	0.53	0.46	3.0	3.1	0.57	0.37	3.4	2.5
Fe	24	23	190	190	82	60	790	470
Ni	1.2	1.7	0.49	0.48	0.19	0.10	0.55	0.40
Cu	0.69	0.99	1.2	1.7	0.73	0.72	2.4	3.5
Zn	1.19	0.72	2.3	1.6	2.0	1.1	4.5	8.3
Ga	0.10	0.12	0.59	0.76	-	-	-	-
Ge	0.12	0.10	0.233	0.015	-	-	-	-
As	0.19	0.08	0.24	0.17	-	-	-	-
Se	0.12	0.16	0.21	0.30	0.104	0.062	-	-
Br	1.37	0.42	0.68	0.46	0.64	0.30	0.68	0.51
Rb	0.53	0.43	0.80	0.54	0.32	0.21	0.91	0.68
Sr	0.21	0.14	0.67	0.54	0.16	0.14	0.96	0.68
Zr	0.81	.68	0.90	0.29	0.104	0.055	1.44	0.92
Au	0.18	0.13	0.29	0.18	-	-	-	-
Pb	0.52	0.30	1.0	1.4	1.24	0.86	3.8	3.0

* Averages and standard deviations are presented for the atmospheric concentration (in ng m^{-3}) of the particulate mass, black carbon, and aerosol trace elements in samples collected in Alta Floresta from August 92 to March 95, and in Cuiabá from July 90 to August 95.

** Aerosol mass and black carbon (BC) are expressed in μg m^{-3}.

es may generate particles containing biogenic-related elements such as Na, Mg, P, S, K, Ca, Zn, and Rb. Also, a significant fraction of aerosols is comprised of secondary aerosols, formed by gas-to-particle conversion of organic and sulfur-related natural biogenic gases. These biogenic particles are mostly submicrometer in size. Bacteria in forested areas were found in the size range of 0.5 to 2.5 μm. Biological activity of microorganisms on leaf surfaces and forest litter results in airborne particles. Windblown pollens certainly contribute to coarse particles in forested areas. The transpiration of plants can lead to migration of Ca, SO_4, Cl, K, Mg and Na to the atmosphere. The biogenic-related elements (K, P, S, Zn, Rb, and others) are essential to superior plants. They are present in the fluids circulating in the plant and are released from the leaves to the atmosphere.

Natural biogenic aerosol particles emitted by plants play an important role in nutrient cycling in tropical ecosystems. Tropical ecosystems maintain a delicate nutrient balance characterized by intense internal recycling and depend on atmospheric input of certain mineral nutrients to fulfill certain nutrient requirements. It has been shown that biogenic particles may influence cloud properties (Schnell 1982) and can act as cloud condensation nuclei, potentially affecting cloud formation mechanisms and cloud dynamics. These particles can travel for long distances (Talbot et al. 1990, Artaxo et al. 1998). Table 3.1 shows the average elemental composition of natural biogenic aerosol particles collected in two background sampling stations in Amazonia (Artaxo et al. 1998, Echalar et al. 1998). The first site (Alta Floresta) is a forest site, and the second (Cuiabá) is a cerrado vegetation site. The average aerosol mass and black carbon concentration is also presented in Table 3.1. It is possible to observe that most of the elements occur predominantly in the coarse mode, with sulfur being the only exception. The aerosol mass for particles less than 10 μm is 15-20 $\mu g/m^3$, a very significant aerosol mass. Phosphorus is strongly present in the coarse mode with more than 90% of P concentrations in particles larger than 2 μm. This means that the cycling of phosphorus is mostly local or regional.

Figure 3.1 shows the time series of aerosol mass concentrations for Alta Floresta from 1991 to 1998. Alta Floresta is in the northern part of Mato Grosso state and is a place where heavy biomass burning occurs from July to October. The large increase in aerosol concentrations during the biomass burning period is remarkable. Background aerosol concentrations of about 10-20 $\mu g/m^3$ of particles less than 10 μm in size during the wet season can increase to 400-500 $\mu g/m^3$ during the dry season. Even if nutrients comprise only a small fraction of this aerosol fraction, it can represent a significant mass fraction of the elements that are lost from the ecosystem.

The natural biogenic aerosol component is comprised of particles that can travel for thousands of kilometers and has an atmospheric residence time of 2-10 days. The composition of these natural biogenic aerosol particles can be observed in Table 3.1, for particles collected in Alta Floresta and Cuiabá and for the fine and coarse mode fractions. We observe that phosphorus, for example, shows concentrations of about 90 ng/m^3 in Alta Floresta, with 80% of this concentration in the coarse mode fraction. The coarse fraction has mostly local or regional impact. In contrast, Table 3.2 shows the average elemental concentration for aerosols during the biomass burning period. Concentrations presented in Table 3.2 come from samples collected in the INPE Bandeirante aircraft during the SCAR-B experiment and also ground-based concentrations for Alta Floresta and Cuiabá. Sulfur can reach an average of 1000 ng/m^3 during

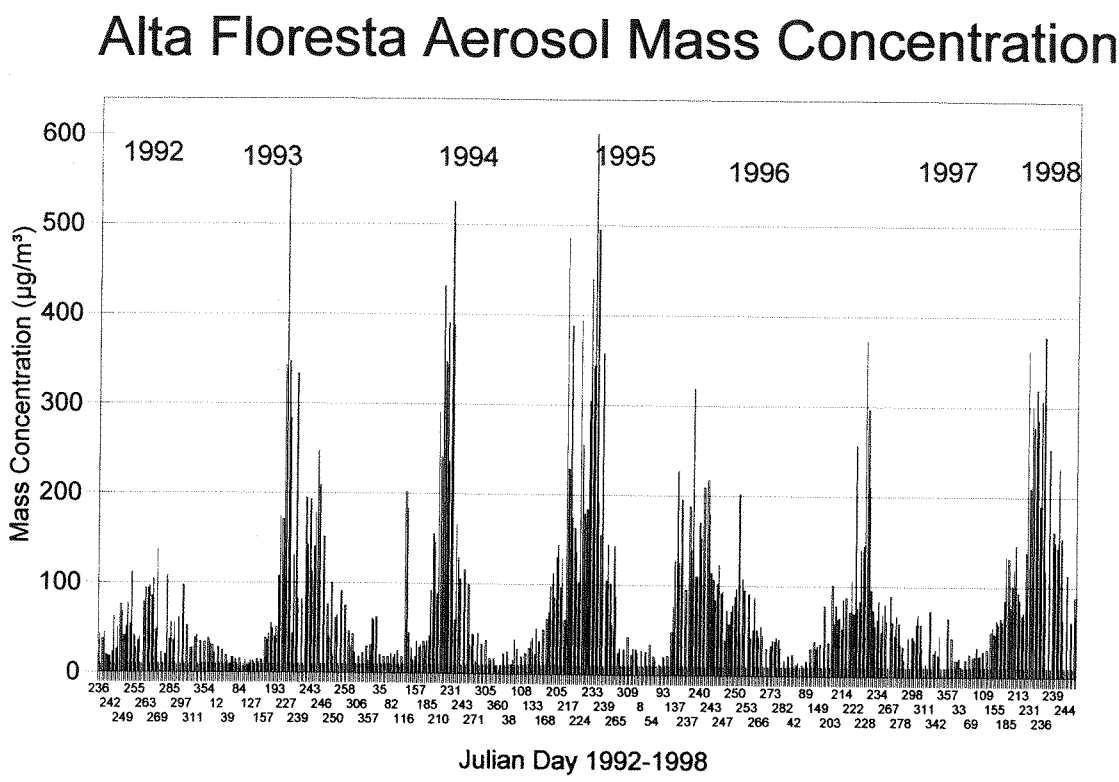

Fig. 3.1 Time series of PM_{10} aerosol mass concentrations in µg/m for Alta Floresta from 1992 to 1998. Aerosol mass concentration are for particles less than 10µm.

the dry season, compared with 300 ng/m^3 during the wet season. For the past 20 years most of the tropical forest areas have been under strong pressure by rapid change of land cover, with forest and adjacent savannas being cleared, most of the time through the use of fire, and converted to pasture and agricultural fields at a substantial rate (Artaxo et al. 1998).

Pyrogenic and natural biogenic emissions to the atmosphere in the Amazon basin may have an impact on the global tropospheric chemistry, because this region exhibits intense convective activity that injects gases and aerosols to high altitudes where they can be transported over long distances (Andreae et al. 1996, 1998, Echalar et al. 1998). Elemental concentrations in aerosols from Amazonia and Africa are very similar (Artaxo et al. 1994, Echalar et al. 1995, Andreae et al. 1998). Trace gas emissions in the burning of African savannah and the Brazilian cerrado are also similar (Scholes et al. 1996, Scholes and Andreae 2000). The pyrogenic particles are efficient cloud condensation nuclei (CCN) (Rogers et al 1992). The abundance of CCN in the dry season over Amazonia and Africa certainly has a strong effect on cloud formation and precipitation patterns, even for regions far from biomass burning, but this issue has not yet been properly addressed.

The aerosols emitted during biomass burning can travel long distances. Figure 3.2 shows a three-dimensional trajectory analysis for aerosol emissions during the dry season (Freitas et al. 1997). From each biomass burning spot, the air masses that transport the aerosol particles and trace gases stay at

Table 3.2 Elemental composition of biomass burning aerosols in the Amazon basin measured during the SCAR-B experiment and comparison with long-term ground-based measurements at Alta Floresta and Cuiabá.

Element	SCAR-B Mean (ng m^{-3})	Standard Deviation	SCAR-B Minimum (ng m^{-3})	SCAR-B Maximum (ng m^{-3})	Alta Floresta Dry Season (ng m^{-3})	Cuiabá Dry Season (ng m^{-3})
Na	95.4	59.9	30.0	270	-	-
Mg	659	385	196	2356	1130	-
Al	2292	1808	350	7297	2540	1342
Si	3126	2104	713	8673	2620	2930
P	140	136	3.50	559	122	21.7
S	1198	900	59.0	4333	1130	560
Cl	222	229	27.6	1291	125	36
K	1581	1076	137.3	5581	1590	1020
Ca	1251	1134	36.2	5722	270	489
Ti	121	123	9.29	533	162	130.8
V	3.67	3.43	0.54	14.2	7.1	6.37
Cr	5.14	1.86	2.29	8.16	41.6	4.6
Mn	67.7	64.4	10.8	322	18.8	16.5
Fe	1132	985	76.9	4327	1623	1990
Cu	2.90	1.71	0.64	8.77	7.9	5.91
Zn	10.7	6.3	1.92	27.7	13.1	11.85
Rb	8.82	5.32	1.55	25.9	10	-
Sr	16.4	14.2	1.44	59.5	5.4	3.34
Br	13.1	8.7	2.40	36.9	19.0	6.1
Ga	0.49	0.34	0.01	1.46	2.36	-
As	0.21	0.16	0.041	0.85	2.7	-
Zr	6.10	3.69	2.08	15.7	8.2	5.86
Mass*	107	60.7	8.05	297	81.2	49.1
BC*	5.49	3.92	0.23	17.5	5.73	2.6

* Aerosol mass and black carbon (BC) are expressed in μg m^{-3}.

low altitude (below 3000 meters) in the Amazon region. When an air mass reaches the Andes, it will rise to higher altitudes because of orographical effects and will reach the Pacific Ocean in only a few days. In the south of Brazil, when Amazon-derived air masses encounter cold fronts they can rise to very high altitudes of about 10 kilometers where atmospheric transport is fast and very efficient. High ozone and aerosols in the South Atlantic were observed during August-September as a result of biomass burning in Amazonia and Africa (Andreae et al. 1996).

Talbot et al. (1990) and Swap et al. (1995) have shown the chemistry changes associated with long-range transport of aerosols in the Amazon basin. Also, the Amazon region could receive a large influx of trace elements from Sahara dust and tropical Africa (Swap et al. 1995). The magnitude of this flux that can carry essential nutrients to the Amazon basin is still unknown.

The Role of Wet Deposition in Amazonia

The Amazon basin is characterized by high rainfall rates, and wet deposition could be significant in the biogeochemical cycles of several nutrients (Andreae et al. 1990). The biogeochemistry of trace elements in the atmosphere is influenced by different sources of gases and aerosols. During a rain event, these gases and aerosols are incorporated into raindrops, and a significant amount of these aerosols are cloud processed, and incorporated into cloud droplets from the very beginning.

During the Amazonian wet season, rainwater chemistry is dominated by natural biogenic emissions, consisting mostly of organic acids, potassium, phosphorus, and other components (Lesack and Melack 1996). In the wet season, average pH is about 5.0 (Williams et al. 1997, Andreae et al. 1990). During the biomass burning period, rainwater pH decreases to about 4.5, mostly because of enhanced concentrations of organic acids, ammonia, and nitrates.

Williams et al. also measured DOC (Dissolved Organic Carbon) at average values of 138 and 238 μM for the dry and wet seasons, respectively. The annual volume weighted mean of 159 μM implies an annual deposition of about 0.4 moles C m^{-2}. The acidity was basically determined by the concentrations of organic acids. The concentration of acetate was 9.3 μeq l^{-1} and formate was 2.9 μeq l^{-1}. The average conductivity was

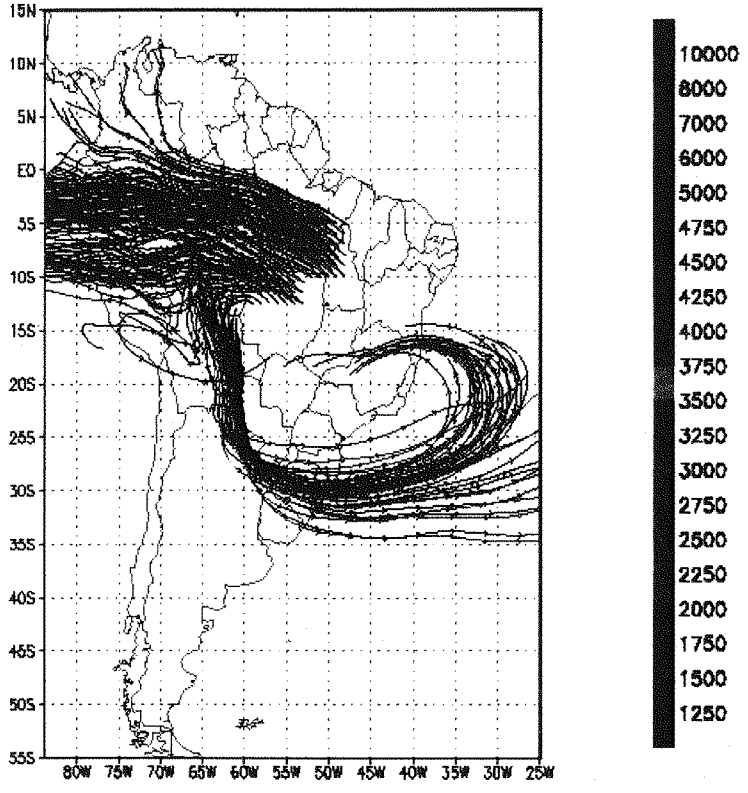

Fig. 3.2 Three dimensional forward air mass trajectories starting from biomass burning spots on August 22, 1985, during the SCAR-B experiment. Shades of gray indicate elevation in meters.

Table 3.3 Rainwater composition at Lake Calado, West of Manaus.

	Volume Weighted Mean ((eql^{-1})			Deposition (meq m^{-2})		
Solute	Annual	Dry Season	Wet Season	Annual	Dry Season	Wet Season
H	17.0	31.7	11.2	46.8	24.7	22.1
pH	4.7	4.5	5.0	-	-	-
NH_4	3.0	7.4	1.2	8.2	5.8	2.4
Na	2.4	3.4	2.1	6.7	2.6	4.1
K	0.8	1.2	0.7	2.3	0.9	1.4
Ca	2.4	2.4	2.4	6.6	1.9	4.7
Mg	0.9	0.6	1.0	2.5	0.5	2.0
Cl	4.6	4.5	4.6	12.6	3.5	9.1
SO_4	2.0	2.8	1.6	5.4	2.2	3.2
NO_3	4.2	6.4	3.3	11.5	5.0	6.5
PO_4	0.03	0.06	0.02	0.09	0.05	0.04
TDP (μM)*	0.25	0.47	0.15	0.7	0.4	0.3

Source: Adapted from Williams et al. 1997.
* TDP is the total dissolved phosphorus.

6.5 μScm^{-1}, with larger conductivity during the dry season. Williams et al. (1997) observed that solute deposition occurred in proportion to total rainfall, suggesting that there was a rather constant source of aerosols in the atmosphere throughout the year. Andreae et al. (1990), during two campaigns in the 1987 wet season and the 1985 dry season during the ABLE experiments, measured similar rainwater concentrations. Significant marine influences were observed in the rainwater collected near Manaus, indicating the long-range transport of marine aerosols within the Amazon basin. A large fraction of the sulfate content of wet season rain appears to be of marine origin. The emissions of biogenic sulfur and nitrogen species from the rain forest ecosystem and their subsequent oxidation in the atmosphere appear to make only a small contribution to the deposition flux of sulfate and nitrate in central Amazonia. Forti et al. (1991) analyzed samples of rainwater and throughfall during ABLE-2B for a site near Manaus (Reserva Ducke). The throughfall was enriched 40-90% compared with the rainwater for Mg, K, Na, Ca, Cl, and SO_4. Absorption of NH_4^+ by the vegetation was also observed during the dry period. These values are similar to rainwater composition in Africa (Freydier et al. 1998), where a pH value of 5.0 was observed. In the tropical forest of Costa Rica, Eklund et al. (1997) also identified organic acids as the major determinant of precipitation pH, but they observed a higher marine aerosol component than that in Central Amazonia.

Measurement of total nutrient deposition requires studies integrating rainfall, throughfall, and stemflow in the wet component, as well as aerosol dry deposition. No studies in Amazonia have yet integrated all these components. Throughfall studies indicate a large enrichment of potassium, on the order of 2.7 to 10.2, leached from plant leaves (Holscher et al. 1998). This large variation in potassium enrichment could be caused by different floristic composition and precipitation patterns.

Conclusions

The atmosphere in tropical forests is a very active part of biogeochemical cycles that

include the soil, vegetation, and hydrological pathways. Trace gases, aerosol particles, and rainwater interact with the system in a variety of ways. Atmospheric transport extends local or regional processes to far larger scales, reaching even to global scales. Integrated studies involving wet and dry deposition fluxes for key nutrients in Amazonia and other tropical areas are needed. Also the issue of how CCN affects cloud formation mechanisms in the dry season must be more thoroughly investigated to assess the impact of anthropogenic influences in tropical forests. A flux of trace elements and nutrients from Africa to Amazonia and from Africa to the Indian Ocean and Australia has been well documented qualitatively for many years. Efforts in quantifying these fluxes are very important for improving our understanding of how tropical ecosystems use and exchange key nutrients.

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