Sources of optically active aerosol particles over the Amazon forest

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Abstract

Size-fractionated ambient aerosol samples were collected at a pasture site and a primary rainforest site in the Brazilian Amazon Basin during two field campaigns (April–May and September–October 1999), as part of the European contribution to the Large-Scale Biosphere-Atmosphere Experiment in Amazonia (LBA-EUSTACH). The samples were analyzed for up to 19 trace elements by particle-induced X-ray emission analysis (PIXE), for equivalent black carbon (BC_e) by a light reflectance technique and for mass concentration by gravimetric analysis. Additionally, we made continuous measurements of absorption and light scattering by aerosol particles. The vertical chemical composition gradients at the forest site have been discussed in a companion article (Journal of Geophysical Research-Atmospheres 108 (D18), 4591 (doi:4510.1029/2003JD003465)). In this article, we present the results of a source identification and quantitative apportionment study of the wet and dry season aerosols, including an apportionment of the measured scattering and absorption properties of the total aerosol in terms of the identified aerosol sources. Source apportionments (obtained from absolute principal component analysis) revealed that the wet and dry season aerosols contained the same three main components, but in different (absolute and relative) amounts: the wet season aerosol consisted mainly of a natural biogenic component, whereas pyrogenic aerosols dominated the dry season aerosol mass. The third component identified was soil dust, which was often internally mixed with the biomass-burning aerosol. All three components contributed significantly to light extinction during both seasons. At the pasture site, up to 47% of the light absorption was attributed to biogenic particles during the wet season, and up to 35% at the tower site during the wet-to-dry transition period. The results from the present study suggest that, in addition to pyrogenic particles, biogenic and soil dust aerosols must be taken into account when modeling the physical and optical properties of aerosols in forested regions such the Amazon Basin.
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1. Introduction

Aerosol particles are of special climatic interest because they both act as cloud condensation nuclei (CCN) and because they scatter and absorb solar radiation, affecting the radiation budget. Amongst others, the Amazon Basin has become an area of increasing interest because it contains the world's largest rainforest and, from a global perspective, is probably the single largest continuous emitter of biogenic gases and particles from plants (Harriss et al., 1990; Andreae and Crutzen, 1997). It is also a region subject to intense deforestation, mostly through the use of fire (Gash et al., 1996). Aerosols and trace gases produced in tropical regions such as the Amazon Basin can be rapidly uplifted to high altitudes and become subject to long-range transport (Pickering et al., 1996; Andreae et al., 2001; Staudt et al., 2001), thereby impacting on health, climate, and atmospheric chemistry at regional as well as global scales. It is therefore important to understand the source types, source strengths, and the physical and chemical properties of aerosol particles from the Amazon Basin.

The composition and sources of Amazonian aerosols have previously been studied for both the "wet" and the "dry" seasons. The former period typically encompasses the months of November to April, and is characterized by intense rainfall and low loading of aerosol, mainly of biogenic and soil dust origins (Artaxo et al., 1990, 1994; Artaxo and Hansson, 1995; Echalar et al., 1998). Pyrogenic aerosols from anthropogenic burning activities dominate the aerosol loading during the dry season. Their characteristics and source apportionment have also been reported previously (see e.g., Andreae et al., 1988; Ward et al., 1992; Maenhaut et al., 1996, 2002; Artaxo et al., 1998; Echalar et al., 1998).

Natural biogenic aerosols emitted from the Amazon forest comprise large ($D > 2\,\mu m$) primary particles (for example, bacteria, fungi, spores, and plant and insects debris), and submicron particles originating from the gas-to-particle conversion of biogenic trace gases emitted by plants or microorganisms. Biogenic particles are characteristically composed of organic material and trace elements such as Na, Mg, P, S, K, Zn, and Rb (Artaxo et al., 1990, 1994). Other elements like N, Ca, and Fe are also macronutrients found in higher plants, whilst Mn, B, Cu, Mo, Cl, Co, Si, Se, and Ni are micronutrients; these elements are often present at very low concentrations, or are released in larger amounts by other sources, and are therefore generally not as useful as tracers for biogenic aerosols. Pyrogenic aerosol particles, which are primarily confined to the submicron mode, have a similar signature to that of biogenic particles, except that they are enriched in absorbing matter (typically elemental or "black" carbon) from combustion processes, and the elements K, S, Zn, and halogens (e.g., Maenhaut et al., 1996). Dust particles arise from wind action in dry areas, and typically contain Al, Si, Ca, Ti, Mn, and Fe. Such particles have also been reported to be transported over the Atlantic from the Sahara to the Amazon Basin (see, e.g., Swap et al., 1996; Formenti et al., 2001). Particles of other origins have also been reported in the Amazon Basin during the wet season, with sources including pyrogenic, urban pollution, and gold mining activities (Echalar et al., 1998), but these are usually confined to a local scale.

Andreae and Crutzen (1997) already reported that, during the wet season, when up to 90% of the Amazonian aerosol mass consists of organic matter, a significant fraction appears to be "black carbon", similar to the biomass burning influenced dry season. These authors suggested that the black carbon present in the wet season must be of biogenic rather than pyrogenic origin, but this hypothesis has not been validated to date by in situ measurements. In the present study, we first identified and quantified the sources of the wet and dry season Amazonian aerosols using a well-established absolute principal component analysis (ACPA) method. Then, we extended the use of the APCA technique by regressing aerosol absorption and scattering (integrated over the sampling time of the corresponding aerosol sample) on the absolute principal component scores (APCS) derived from the initial APCA. This technique allowed us to identify the major sources of optically active aerosol over the Amazon Basin under background and biomass burning conditions and to quantify for the first time their relative contributions to the total aerosol scattering and absorption. The results conclusively demonstrate the non-negligible contribution of biogenic and soil dust emissions to the total aerosol absorption, which previously has been thought to originate solely from combustion emissions.

2. Sampling location and experimental method

2.1. Sampling location

Aerosol particles were sampled over two field campaigns at a pasture site (Fazenda Nossa Senhora Aparecida, FNS, located at $10°45'44''$ S, $61°21'27''$ W, 315 m above sea level) and on a 54 m tower situated in a primary rainforest with a mean canopy height of about 32 m (Reserva Biologica Jarú, RBJ, $10°04'55''$S, $61°55'48''$W, 110 m above sea level). Both sites are located in the state of Rondônia, Brazil, and separated by a distance of ca. 80 km. The first campaign (LBA-EUSTACH 1) took place in January–May 1999 for the pasture site, covering most of the wet season, and in April–May 1999 for the forest site, covering the end of the "wet season" period and the transition period

toward the biomass-burning-influenced "dry season". The second campaign (LBA-EUSTACH 2) was conducted in September–October 1999 throughout the end of the "dry season" and the transition period toward the "wet season". For a more complete description of the measurement locations, meteorological, and overall sampling conditions, refer to Andreae et al. (2002) and Silva Dias et al. (2002).

2.2. Filter sampling and chemical analysis

At the pasture site, two-stage "Gent" stacked filter units (SFUs) were positioned ca. 3 m above ground level on a scaffolding tower. SFUs were also positioned on the tower at the forest site, at heights of 48 m (ca. 16 m above maximum canopy height), 22 m (at the average canopy height), and 3 m above ground level. The SFUs, which were fitted with a PM_{10} impactor inlet (Hopke et al., 1997), were operated at a flow rate of 16 l min^{-1}, with two filters in series being used to collect the "coarse" ($2.0 < D_p < 10 \mu m$; with D_p the aerodynamic diameter) and "fine" ($D_p < 2.0 \mu m$) particulate matter (CPM and FPM, respectively). No attempt was made to dry the particles prior to sampling.

Concentrations of up to 19 elements from Mg to Pb could be determined by particle-induced X-ray emission analysis (PIXE) for both coarse and fine SFU aerosol fractions (Johansson and Campbell, 1988). Detection limits were typically 5 ng m^{-3} for elements in the range $13 < Z < 22$, and 0.4 ng m^{-3} for elements with $Z > 23$. Precision was typically better than 10%, and up to 20% for elements with concentrations close to the detection limit. Mass concentrations were obtained by gravimetric analysis. SFU filters were weighed (at 20°C and 50% relative humidity (RH)) before and after sampling using a Mettler M3 electronic balance with 1-μg sensitivity, and an accuracy of $\pm 5 \mu g$. So-called "black carbon equivalent" (BC_e) concentrations were obtained for the fine fraction of the SFU filters by a light reflectance technique (Andreae, 1983). Comparing several techniques, Reid et al. (1998) concluded that the light reflectance technique provided the best agreement with the extinction cell plus nephelometer reference method (Weiss and Hobbs, 1992), resulting in uncertainties of ca. 17%. For more details concerning the SFU filter sampling and analysis, see Artaxo et al. (2002) and Guyon et al. (2003c).

2.3. Real-time aerosol measurements

Light-scattering measurements were performed at the forest tower using a single-wavelength ($\lambda = 545$ nm) nephelometer (model M903, Radiance Research, Seattle, USA), positioned at a height of 49 m above the ground. Aerosols were sampled continuously and the data averaged and collected on a 1-min time resolution. The instrument was operated without cut-off device at the inlet except a rain cap with insect screen and no attempt was made to dry the particles prior to sampling. Nevertheless, the internal heat produced by the instrument itself may have partially dried out the sampled particles. On average, the RH inside the instrument was 15% (± 6%) lower than ambient RH. An ambient RH of 92% was found to be the threshold value above which the scattering coefficient showed an erratic response to increasing RH, and only data collected below 92% RH were included in the analysis (because of the persistently high RH at night, almost all the nighttime scattering data was discarded). The data were corrected for the angular truncation of the instrument (Guyon et al., 2003b); therefore, the errors associated with the scattering values presented herein are considered to be due only to measurement error of the instrument itself (ca. 5%).

Continuous absorption measurements were performed at the RBJ site using a Radiance Research particle soot absorption photometer (PSAP) 52 m above ground. During the dry season, absorption was also measured using two aethalometers (Magee Scientific Co.) at 3 and 50 m above the ground. Both the PSAP and the aethalometer operate using the principle of an integrating plate, measuring transmittance through a glass fiber filter at a wavelength of 565 and 880 nm, respectively. PSAP absorption values were collected every 5 min during the wet season campaign and every minute during the dry season, corrected according to Bond et al. (1999), and uncertainties were computed following the work of Anderson et al. (1999). Because the Bond correction requires the use of light scattering data, as obtained from a nephelometer, corrected PSAP data could only be obtained when the nephelometer was functioning and ambient RH was <92%. For the purpose of integrating the absorption values over the filter sampling time, ca. 30% of the dry season Bond-corrected PSAP absorption coefficient values (σ_a) were estimated from the linear regression of the available σ_a data on the uncorrected absorption coefficient data ($\sigma_{a,raw}$), for periods with ambient RH <92% when the nephelometer data were not available. The linear equation used in these cases was $\sigma_a = \sigma_{a,raw} \times 0.61 - 0.03$ M m^{-1} (obtained from the regression of 3170 10-min averaged measurements, $r^2 = 0.98$). Similarly, for the wet season campaign, ca. 70% of the Bond-corrected PSAP data used here were estimated from the regression $\sigma_a = \sigma_{a,raw} \times 0.68 - 0.05$ M m^{-1} (obtained from the regression of 1188 10-min averaged measurements, $r^2 = 1.00$) because the majority of the nephelometer data collected during this campaign was unusable due to the consistently high RH. It is noted here that these equations account for all three corrections that have to be applied to the PSAP data—the deposit area, the over-response to scattering, and the adjusted calibration response (Bond et al., 1999).

However, once integrated over the sampling time of the corresponding filter sample, the estimated σ_a values compared well with other absorption data, as shown later (Section 3.1).

The aethalometers were operated with a 5–min time resolution, and the internal calibration absorption efficiency of $19 \, m^2 \, g^{-1}$ was used to convert the concentration output of the instruments ($\mu g \, m^{-3}$) into absorption coefficients (Mm^{-1}). The largest possible absolute error associated with the PSAP and the aethalometer is ca. 20–40% (Reid et al., 1998).

For the purpose of the regression analysis, the values obtained from these instruments (when available) were averaged over the sampling time of the corresponding filters. For a complete description of the instruments, data handling, and uncertainty calculations refer to Guyon (2002) and Guyon et al. (2003a).

2.4. Absolute principal component analysis

Aerosol sources were computed using absolute principal component analysis (APCA) (Thurston and Spengler, 1985; Keiding et al., 1986). With this technique, the sources are first identified using principal component analysis (PCA), which involves constructing a model of trace element concentration variability (in our case only variables that were below the detection limit $<5\%$ of the time were allowed in the model). During the analysis, a set of inter-correlated variables is transformed into a set of independent variables by finding the eigenvalues and eigenvectors (factors/components) of the correlation matrix. The most prominent eigenvectors are retained and subjected to VARIMAX rotation, to yield a "component-loading" matrix that represents the correlations between the elements and each component. In APCA, absolute principal component scores (APCS) are also obtained, which indicate the importance of the identified components in individual samples. Mass/elemental component apportionments are then obtained from the regressions of the measured mass/elemental concentrations on the calculated APCS (Artaxo et al., 1988, 1990, 1998; Maenhaut et al., 1996; Echalar et al., 1998). The stability of this procedure depends on the number of degrees of freedom in the analysis (i.e., the number of samples) (Ito et al., 1986). It has been suggested that at least 30 are required to obtain statistically robust results (Henry, 1991).

In the present study, in order to investigate the origin of the light absorbing and scattering material, we performed APCA to retrieve the aerosol sources, and then regressed the in situ measured aerosol optical properties against the obtained absolute principal component scores (APCS), as detailed in the following sections.

3. Results and discussion

3.1. Wet season: the sources of the LBA-EUSTACH 1 aerosols

The left-hand side of Table 1 shows a summary of the PCA results obtained for the wet season aerosol sampled above the canopy at the tower site in Rondônia. In this analysis, fine and coarse aerosol data were combined together, in order to facilitate further correlation of the identified sources with aerosol optical properties. The values reported for each element (or variable) correspond to the "component loadings" of the element in the different components ("sources"). Only loadings larger than three times their associated standard deviation are statistically significant (Heidam, 1982). In our case standard deviations were typically lower than 0.1, so that loadings larger than ca. 0.3 can be considered as being statistically significant.

It is evident from Table 1 that most of the variance in the element concentrations was explained by a four-component model (as indicated by communality values approaching a value of 1), in which all four components have eigenvalues larger than unity. Prior to performing the PCA, we computed the Kaiser–Meyer–Olkin measure-of-sampling-adequacy test, which yielded a value of 0.71, indicating that the variance in the data may be explained by the underlying components, and that the PCA was appropriate. However, it should be noted that these results were obtained using data derived from the analysis of only 28 SFU samples, which is slightly less than the minimum required to achieve statistically robust results (see Section 2.4).

During the wet season, aerosol samples (76) were also collected at a pasture site situated ca. 80 km from the RBJ site, and analyzed in an identical fashion to our samples (Artaxo et al., 2002). We, therefore, performed a separate PCA using this data set in order to check the robustness of our results for the forest site. The results, also reported in Table 1, indicate that the aerosols measured at the forest and pasture sites had the same origin—the first and second components for each site show high correlations with the same elements, and the fourth component of the forest data set is similar to the third for the pasture site. These three components were responsible for about the same percentage of the variance at both sites. The main difference between the two data sets was the identification of a third component for the forest site aerosol, mainly dominated by Cu in both the fine and coarse fractions. Copper is often identified as a single component in the analysis of Amazonian aerosol data and is suspected to be of biogenic origin. A Cu-containing component was not identified for the pasture site simply because this element was not measurable frequently enough at this site to be included in the APCA model. Therefore, despite the fact

Table 1
VARIMAX-rotated principal component analysis loading matrices for the fine and coarse aerosol fractions collected at the Rondônian forest (top of the tower) and pasture sites during the 1999 wet season

Var.[a]	Wet season								
	Forest site					Pasture site			
	Comp. 1	Comp. 2	Comp. 3	Comp. 4	Comm.	Comp. 1	Comp. 2	Comp. 3	Comm.
FPM	**0.94**	−0.06	0.01	−0.17	0.91	**0.87**	−0.17	−0.05	0.78
CPM	0.27	**0.88**	−0.08	0.10	0.87	0.39	**0.86**	0.02	0.89
BC_e	**0.93**	0.18	−0.02	−0.18	0.93	**0.86**	0.23	−0.10	0.80
Si_f	**0.91**	−0.20	0.05	0.11	0.88	**0.60**	−0.40	**0.54**	0.82
P_f	**0.57**	−0.47	0.47	0.08	0.76	0.18	−0.27	**0.68**	0.57
S_f	**0.92**	0.24	−0.02	−0.21	0.95	**0.93**	−0.13	−0.05	0.89
K_f	**0.93**	0.15	0.00	−0.22	0.94	**0.94**	−0.10	−0.07	0.91
Ca_f	—	—	—	—	—	0.42	−0.38	**0.57**	0.64
Mn_f	**0.89**	−0.21	−0.06	0.06	0.83	**0.83**	−0.39	0.13	0.85
Fe_f	**0.94**	−0.07	−0.00	−0.17	0.91	**0.86**	−0.37	−0.06	0.88
Cu_f	−0.00	−0.24	**0.86**	−0.09	0.81	—	—	—	—
Zn_f	—	—	—	—	—	**0.93**	−0.06	−0.17	0.90
Si_c	—	—	—	—	—	**0.90**	−0.13	−0.16	0.85
P_c	−0.41	**0.84**	0.01	0.21	0.92	−0.04	**0.95**	0.13	0.92
S_c	0.45	**0.74**	−0.07	0.03	0.76	0.62	**0.76**	−0.03	0.92
Cl_c	−0.37	0.29	−0.09	**0.77**	0.83	−0.02	**0.75**	0.52	0.83
K_c	0.01	**0.84**	−0.04	**0.44**	0.89	0.23	0.94	0.05	0.93
Ca_c	**0.78**	0.13	−0.08	**0.53**	0.92	**0.62**	0.26	**0.48**	0.68
Mn_c	**0.88**	0.05	−0.07	0.19	0.82	**0.92**	−0.06	−0.19	0.88
Fe_c	**0.94**	0.19	−0.06	−0.16	0.95	**0.94**	−0.05	−0.27	0.86
Cu_c	−0.23	**0.51**	**0.72**	−0.12	0.83	—	—	—	—
Zn_c	0.08	**0.81**	0.02	−0.12	0.68	0.54	**0.76**	−0.11	0.87
λ[b]	9.2	4.5	1.6	1.2		9.9	5.0	1.8	
% variance	48.5	23.4	8.2	6.1		49.6	25.1	9.1	

The species which constitute a component are highlighted.
    [a] "Var." is the variable included in the model, with the subscripts "f" and "c" denoting the fine and coarse fraction, respectively. "Comp." is the extracted principal component, and "Comm." the communality associated with each variable in the model.
    [b] λ is the eigenvalue associated with the principal component.

that the pasture site was suspected to be more influenced by anthropogenic activities, and showed slightly higher concentrations of dust particles (Artaxo et al., 2002; Guyon et al., 2003b, c), the results of the component analysis for the pasture site support those obtained for the forest site PCA.

The four components extracted from the PCA explain 86% of the total sample variance. As the combined uncertainties associated with sampling and analysis are estimated to have been about 10%, the model satisfactorily explains all the variability associated with the data set. The communalities of FPM and CPM are larger than 0.87, indicating that most of the variability in each of these two mass concentrations could be explained.

The four components were identified as follows. The first component was responsible for 49% of the variance in the data. It is associated with biomass burning and contains elements of the fine fraction typically considered to be of biogenic origin and enriched in pyrogenic aerosols (S, K, and Zn), as well as BC_e from combustion

(Artaxo et al., 1988; Maenhaut et al., 1996; Echalar et al., 1998). However, this component also correlated well with some soil dust-associated elements (Si, Mn, and Fe in the fine fraction, and Ca, Mn, and Fe in the coarse), which may have been due to uplift of significant quantities of dust during fires. Both biomass-burning aerosol and soil dust cannot originate from the Amazonian rain forest during the wet season, and the sources of these aerosols were most likely the drier neighboring regions. It is worth noting that backward trajectory analyses showed that biomass-burning sources were originating some 2–3 days south from the sampling sites during the wet season (Guyon et al., 2003b). The correlation between biomass burning and dust-associated elements may, therefore, also have been due to mixing of independently emitted smoke and dust aerosol during long-range transport, essentially forming one indivisible component (Artaxo and Hansson, 1995). This component, which we further associate with a mixed soil dust/biomass burning aerosol, also displayed

a high degree of correlation with FPM, whereas all the other components showed low correlation with this mass fraction.

The second component could be readily identified as coarse fraction biogenic aerosol (high P, S, K, Cu, and Zn loadings), and was responsible for 23% of the variance in the data, and for most of the variance in the CPM values. This component is also slightly anti-correlated with fine fraction P, in the same manner that the first component is with coarse fraction P. The third and fourth components displayed very low correlation values with PM, which suggests that they are of minor importance for the aerosol mass burden. The third component showed high loadings with coarse and fine fraction Cu, and, to a lesser extent, fine fraction P, and is probably associated with biogenic material. The fourth component is mainly loaded with the elements Cl, K, and Ca of the coarse fraction, also probably of biogenic origin.

It is somewhat unfortunate that soil dust (present in the coarse and fine fraction) and biomass burning particles (most abundant in the fine fraction) formed one inseparable component, as it seemed reasonable to suspect that these two aerosol types were the most important in terms of light extinction, and we were interested to apportion the optical effects of the total aerosol between the various contributing sources (see below). Performing separate PCA/APCA for the fine and coarse aerosol fractions could have apparently separated the two aerosol types. However, this separation would not have allowed us to perform further multivariate regressions of the aerosol optical properties on the aerosol sources because of the covariance of these two components. Therefore, we chose to continue our analysis with fine and coarse fraction data kept together in one APCA. Care should be taken when conducting APCA with the coarse and fine aerosol fractions separated and regressing an ''independent dataset'' (e.g., absorption or scattering coefficients) on them. Although more information (components) can be obtained when analyzing coarse and fine aerosol fractions separately, the apparent ''better solution'' may mask some covariance in the data between the

fractions, and lead to errors in the quantification of the sources identified by APCA for the independent variable of interest.

The average mass source apportionment for the total aerosol, as estimated from the APCA results was obtained from multivariate regression of the measured PM on the APCS. The r^2 value for the regression (28 samples) was 0.99. As expected from their low correlation with FPM and CPM in the PCA, components 3 and 4 did not significantly contribute to PM, and each contributed to only $4 \pm 4\%$ of the total PM for the wet season aerosols. Therefore, the wet season aerosols were mainly constituted of soil dust/biomass burning and biogenic material, which contributed 42% and 50% of the PM, respectively (Fig. 1a). The main aerosol component during the wet season was therefore biogenic material (e.g., pollen grains, bacteria, spores, plant debris), confirming the results of previous studies in the region. Artaxo et al. (1990) already reported that only two components could explain ca. 90% of the variance in their data for fine and coarse fraction Amazonian aerosols of the wet season at a forest site near Manaus. These two components were soil dust (in our case mixed with some biomass burning) and natural biogenic aerosols, the latter accounting for 55–95% of PM.

In contrast to the findings of Artaxo et al. (1990) from Manaus, our data indicate that even during the wet season biomass burning may have contributed substantially to the PM in Rondônia, highlighting the dramatic influence that anthropogenic activities are having on total aerosol loading in parts of the Amazon Basin year-round. Maenhaut et al. (2002) also found a biomass burning contribution to the wet season aerosols at the Amazonian site of Alta Floresta (16% of the FPM concentration on average), whereas Echalar et al. (1998) did not identify a pyrogenic component for that season at the same site, and attributed 65% of FPM to biogenic aerosols.

As a further step, we attempted an apportionment of the aerosol light absorption between the various sources identified above by performing multivariate regression of absorption on the APCS, which is a technique based
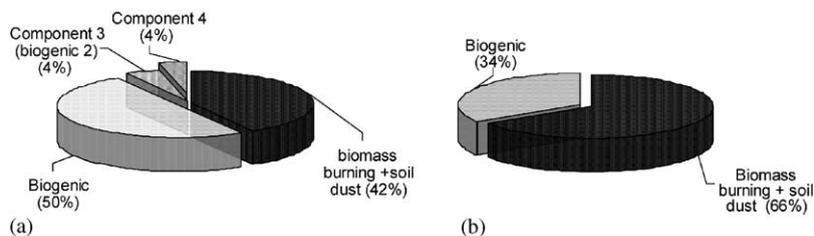

Fig. 1. Relative source apportionment of the total aerosol mass (a) and the light absorption as measured using a reflectance photometer technique (BC$_e$) (b), for the wet season aerosol sampled near the top of the Jarú meteorological tower in Rondônia, Brazil, between 8 April and 21 May 1999.

on a linear least-squares fitting. Light absorption data, which were obtained from a PSAP instrument for the tower site, were available and integrated for 22 of the 28 samples. Multivariate regressions were also performed using the BC_e data obtained for 28 samples of the fine fraction. The r^2 of the regressions were 0.98 in both cases. Fig. 1b shows the apportionment based on the BC_e data. This indicates that only the soil dust/biomass burning and biogenic aerosol components contributed to the absorption. The results obtained from the regression using the PSAP data differed from those presented in Fig. 1b by only 1%, with the soil dust/ biomass burning component contributing 65% of the absorption, and the biogenic component the remaining 35%. As expected, the soil dust/biomass-burning component contributed more to the absorption than the biogenic component, indicating a substantially larger absorption efficiency (given its lower mass concentration). Nonetheless, biogenic aerosols were found to contribute significantly to the absorption (ca. 35%) during the wet season.

The apportionment of the scattering coefficient to the identified sources was performed for 13 samples. The results indicate that only the soil dust/biomass burning component contributed significantly to scattering, in agreement with the results of Guyon et al. (2003c), who found that CPM did not contribute significantly to light scattering during the wet season.

A similar analysis to that presented above was performed on the data obtained from 76 samples collected at the pasture site (Artaxo et al., 2002), allowing better statistics than those presented for the forest site data. The two first components listed in Table 1 were associated with soil dust/biomass burning and natural coarse biogenic particles, respectively. The third component was less straightforward to identify, and displayed high loadings with: (1) some fine fraction elements typical for biogenic material (P and Cl (the latter not attributed to sea-salt)), (2) fine fraction Ca and Mn, and coarse fraction Ca, which can be found in both plant material and soil dust (Artaxo et al., 1994), and (3) fine fraction Si, usually more typical of soil dust

particles, but which can also be emitted from plants (Artaxo and Hansson, 1995). This component is, therefore, most likely associated with fine fraction biogenic particles, which may have mixed with soil-dust (Artaxo et al., 1990; Artaxo and Hansson, 1995).

The APCA of the pasture site data revealed that all three components contributed significantly to PM. It can be seen from the results (Fig. 2a) that the main difference between these data and those from the forest site, apart from the now significant fine biogenic/soil dust component, is the dramatic decrease in the relative contribution of the soil dust/biomass burning component. In absolute amounts, the two models applied to the forest and the pasture site data showed comparable amounts of the soil dust/biomass burning component, but a larger biogenic component was found for the pasture site. Although the data obtained at the pasture site encompassed more of the wet season period (only 6 of the 74 samples were taken after 12 May 1999) than the RBJ data, which were more representative of the wet/dry transition (6 of the 28 samples were taken after 12 May 1999), these results remain surprising because pasture site aerosols were more influenced by anthropogenic activity (Artaxo et al., 2002).

The regression of the wet season fine mode BC_e on the APCS for the pasture site yielded relative contributions to the absorption of ca. 27%, 47%, and 26% for soil dust/biomass burning, biogenic, and fine biogenic/soil dust aerosols, respectively (Fig. 2b). This suggests that all three kinds of aerosols contributed significantly to the absorption in the Amazon Basin, and that it may be important to consider the optical properties of all three in aerosol models.

3.2. Dry season: the sources of the LBA-EUSTACH 2 aerosols

Table 2 presents a summary of the PCA results obtained from 81 SFU samples collected near the top of the forest tower during the dry season campaign. The three components extracted from the PCA (with eigenvalues larger than unity) explain 80% of the total
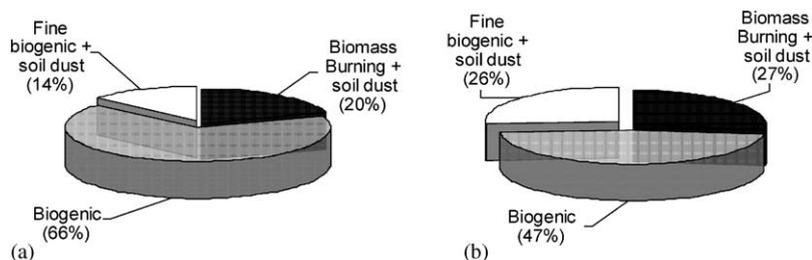

Fig. 2. Relative source apportionment of the total aerosol mass (a) and the light absorption as measured using a reflectance photometer technique (BC_e) (b), for the wet season aerosol sampled at the Fazenda Nossa Senhora Aparecida pasture site in Rondônia, Brazil, between 28 January and 22 May 1999.

Table 2
VARIMAX-rotated principal component analysis loading matrix for the aerosol (fine and coarse combined) collected at the Rondônian forest (top of the tower) site during the 1999 dry season

Var.	Dry season			
	Forest site, fine and coarse aerosol fractions			
	Comp. 1 Biomass burning + soil dust	Comp. 2 Biogenic	Comp. 3 Soil dust	Comm.
FPM	**0.85**	0.18	0.39	0.91
CPM	0.36	**0.68**	0.01	0.59
BC_e	**0.84**	0.15	0.39	0.88
Si_f	**0.76**	0.29	−0.12	0.68
P_f	**0.78**	0.45	0.03	0.81
S_f	**0.81**	−0.14	0.34	0.79
K_f	**0.87**	0.24	0.32	0.92
Mn_f	**0.69**	0.42	0.32	0.76
Fe_f	**0.52**	−0.05	**0.64**	0.75
Zn_f	**0.73**	0.42	0.36	0.84
P_c	0.15	**0.91**	0.04	0.85
S_c	0.21	**0.63**	0.50	0.69
K_c	0.06	**0.84**	0.48	0.94
Mn_c	0.27	0.50	**0.64**	0.73
Fe_c	0.20	0.23	**0.89**	0.88
λ	8.62	2.04	1.35	
% variance	57.5	13.6	9.0	

The species which constitute a component are highlighted.

sample variance. The lowest of all computed communalities is associated with CPM, indicating that the model does not explain the variability of coarse aerosol for this season very well. When the model was implemented with a fourth component, with an associated eigenvalue of only 0.75 (explaining 5% of the variance in the sample), the communality associated with CPM increased to 0.83. This fourth component was mainly associated with biogenic material (the highest loadings were for coarse fraction Mn, K, and P, and fine fraction Mn and Zn). However, close examination of the time series of the APCA computed-components (Fig. 3) revealed that the fourth component was mainly due to only a few samples in the data set. Therefore, despite the low communality associated with CPM, we decided to only include the three components with eigenvalues > 1 in further analysis (Table 2).

The first component, which explains 58% of the variance, showed high loadings for FPM, typical vegetation-related elements (P, S, K, and Zn) and BC_e in the fine fraction, and to a lesser extent, fine fraction Si, Mn, and Fe. This suggests that this component was associated with biomass burning and some soil dust. Scanning electron microscopy (using a high-resolution field emission scanning electron microscope (FESEM 1530, Oxford Instruments) with a 1.50 kV electron beam) of a fine aerosol sample from the dry season revealed that a large number of aerosol particles were soil dust/organic agglomerates (i.e., dust particles covered with an organic layer, which probably
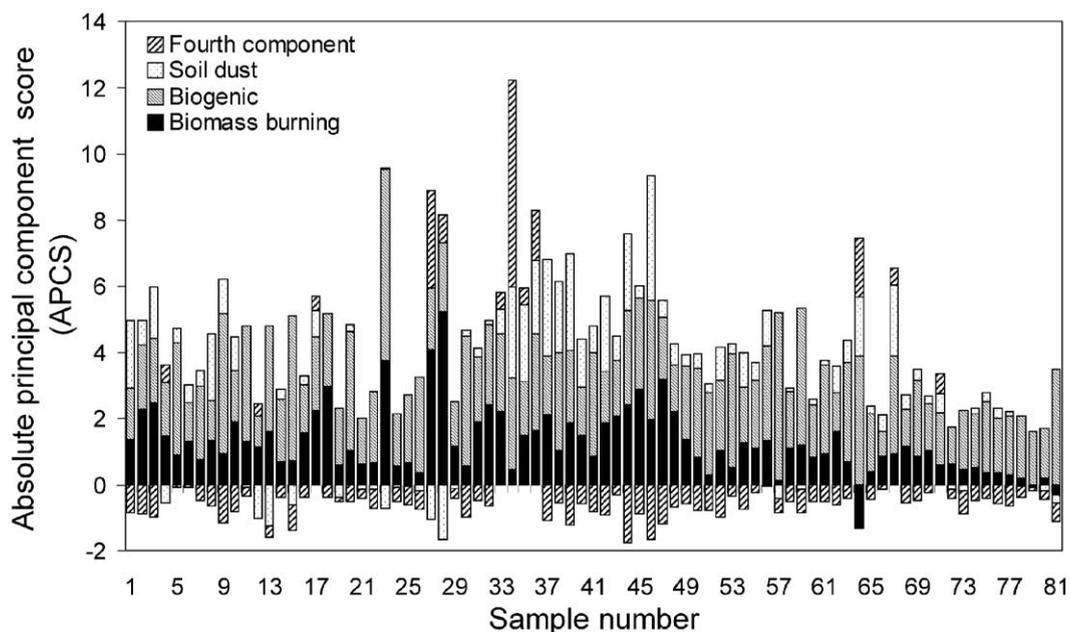

Fig. 3. Absolute principal component scores (APCS), as obtained from a four-component APCA model, for each of the aerosol samples collected near the top of the Jarú meteorological tower in Rondônia, Brazil, between 6 September and 31 October 1999.

originated from biomass burning) (Guyon, 2002). The second component is mainly loaded with CPM, and coarse fraction elements associated with biogenic aerosol (P, S, and K). The third component showed high loadings for fine and coarse fraction Fe, and with coarse fraction Mn. We performed regression analysis of coarse and fine fraction Fe against other typical tracer elements for soil dust (Al, Si, and Ti) that were present in our samples more than 5% of the time below detection limit, and therefore not included in the PCA. Aside from fine fraction Si (which we found to correlate with biomass burning), Fe correlated well with all other soil dust markers, especially across the two size fractions. This supports the contention that the third component was associated with soil dust. The fact that fine and coarse fraction soil elements were relatively well correlated indicates that the soil dust particles in both size fractions were probably derived from the same source. This source was most likely the resuspension of soil dust by biomass burning, as suggested by the correlation of the soil dust elements of the fine fraction with the biomass-burning component. The influence of the pyrogenic

component decreased toward the end of the campaign (Fig. 3), which coincided with the transition to the wet season. In contrast, the biogenic component remained relatively constant, indicating that it is part of the continuously emitted natural background aerosol.

Fig. 4a presents the average source apportionment for the total dry season aerosol sampled near the top of the Jarú tower, as estimated from the APCA results. As expected, biomass burning was found to be the major contributor to aerosol mass (65%), but biogenic organics (30%) and soil dust (5%) were also present in non-negligible amounts. It could be that the contribution of the soil dust component was underestimated in our study, because parts of the soil dust elements of the fine fraction were included in the biomass-burning component. Artaxo et al. (1998) reported that biomass burning, natural biogenic, and soil dust components accounted for 54%, 19%, and 16%, respectively, of the total aerosol mass during the SCAR-B (Smoke, Clouds, and Radiation-Brazil) experiment in August–September 1995. Additionally, they reported a second soil source and a sea-salt (NaCl) component, each making up ca.
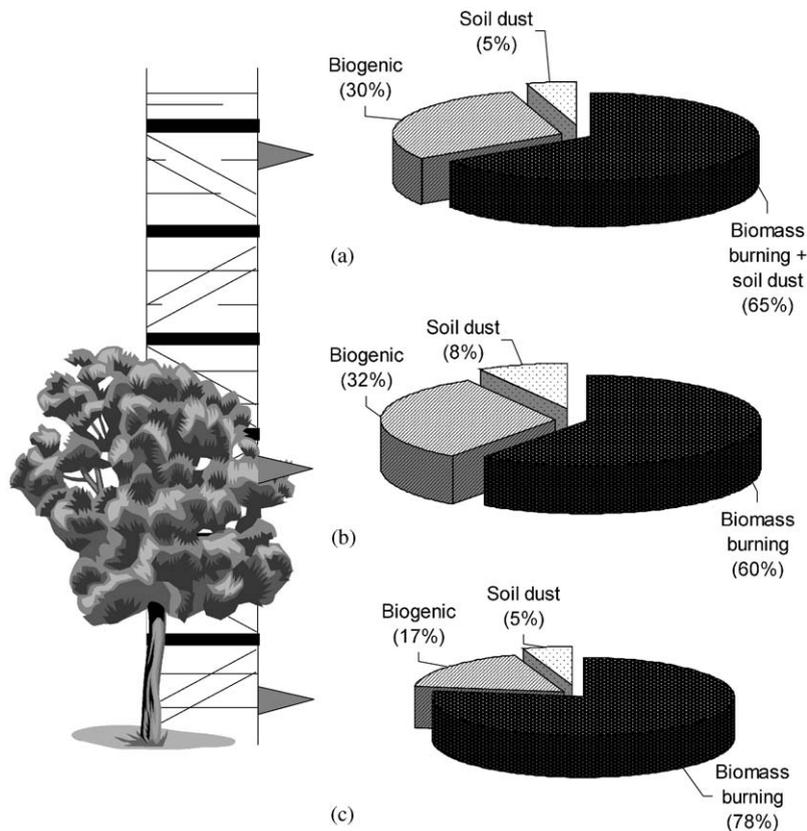

Fig. 4. Total aerosol mass source apportionment for the dry season aerosol sampled near the top (a), at canopy level (b), and at ground level (c) on the Jarú meteorological tower in Rondônia, Brazil, between 6 September and 31 October 1999. The gray triangles on the side of the tower represent the sampling heights, whereas the tree represents the mean canopy height. The size of the pie charts is proportional to the mean mass concentration measured for each level.

6% the total particulate mass. It is possible that the soil dust component that could not be differentiated from biomass burning in our analysis corresponds to one of the two soil dust components identified by the latter authors. It was not possible for us to identify a NaCl component, because Na was not measured in our study, and Cl was not detected frequently enough to be incorporated in the analysis. However, our measurement site was situated ca. 2000 km from the coast, and it is likely that NaCl was present in fairly negligible amounts in our samples.

Our relative mass apportionment for the biogenic and the pyrogenic components are both ca. 11% larger than the values reported by Artaxo et al. (1998). This may be partially explained by the fact that we could only explain ca. 80% of the variance in our data, whereas Artaxo et al. (1998) could explain ca. 91% of theirs. Also, the latter authors sampled aboard an airplane, while our equipment was positioned much closer to the sources of the biogenic aerosol, which is composed predominantly of coarse particles and might be expected to have a limited dispersal range.

It may appear astonishing that biogenic aerosol would make up such a high fraction of the aerosol mass during the dry season. Artaxo et al. (1998) already mentioned that the two main sources for biogenic particles are primary biogenic aerosol particles, and gas-to-particle conversion from biogenic volatile organic compounds (VOCs), but noted that the nature and mechanism of emission of these particles is not fully understood. Biomass burning could have contributed to the additional release of coarse fraction biogenic particles into the atmosphere via convective updrafts (Guyon et al., 2003c).

The relative contribution of the sources identified above for the dry season aerosol to the light absorption measured with a PSAP and an aethalometer (both situated near the top of the tower), and to BC_e measured on the filter samples, was calculated by multivariate regression of the integrated absorption values on the corresponding APCS. The regressions were performed for 49, 62, and 81 samples, yielding r^2 values of 0.95, 0.84, and 0.96 for the PSAP, aethalometer, and BC_e data, respectively. The results obtained using the BC_e data are shown in Fig. 5a. The source apportionment of the absorption derived from the PSAP and the aethalometer instruments were very similar despite different sampling dates and number of data points. The biomass burning, biogenic, and soil dust components were found to contribute 58%, 31%, and 11% of the absorption (PSAP), respectively, and to 66%, 27%, and 7% of the absorption (aethalometer), respectively. The contribution of the biomass burning/soil dust component is slightly greater according to the regression parameters obtained using the aethalometer data, which could be due to the fact that PSAP data were not

available between 9 and 20 October (corresponding to samples 44–67), a period which included a number of intense biomass burning events (Fig. 3).

Overall, the results indicate the major contribution of the biomass burning/soil dust component to the light absorption. Dust particles, which contributed up to 11% of the total absorption, are known optical absorbers (Andreae, 1996; Tegen et al., 1996); however, the surprising result to emerge from our analysis is the relatively large contribution of biogenic aerosols to absorption (ca. 30% according to the analysis performed using the PSAP and aethalometer data, and 17% when using the BC_e data), confirming the findings of the wet season data analysis. An explanation could be that this absorption is due primarily to large biogenic particles, since the source apportionment of BC_e, which was measured in the fine particle fraction only, indicated that a much larger fraction of the absorption in the fine aerosol (ca. 77%) was attributable to biomass burning particles (neither the PSAP or aethalometer were equipped with an upper cut-off diameter inlet, so the absorption measured by these instruments relates to the total aerosol).

The relative source apportionment of the light scattering measured with a nephelometer near the top of the forest tower was obtained from the regression of the scattering coefficients on the dry season APCS, with the scattering coefficients averaged over the sampling time of the corresponding filter. The regression was calculated on the basis of 62 data points, and yielded an r^2 value of 0.89. The results suggest that biomass burning was the major aerosol source responsible for light scatter (ca. 80%), with soil dust and biogenic aerosol contributing roughly equally to the remaining 20%. It should be noted, however, that the regression analysis was not significant for the biogenic component (*p*-value of 0.18), whereas it was highly significant for the other components, and the contribution of this component to scattering might be overestimated in our analysis. The results, therefore, indicate that biogenic aerosols contributed less to scattering than to absorption.

3.3. Dry season, the tower profile

For the dry season, 80 and 81 samples were available for the within-canopy and below-canopy levels, respectively; however, the relatively large amount of "missing" data (due to elemental concentrations lying below detection limit) prevented us from obtaining statistically robust models of aerosol variability at these levels. Nevertheless, we present a brief description of the APCA results for these latter data sets here because canopy profile data for Amazon forest aerosols is rare in the literature. It must be cautioned, however, that the
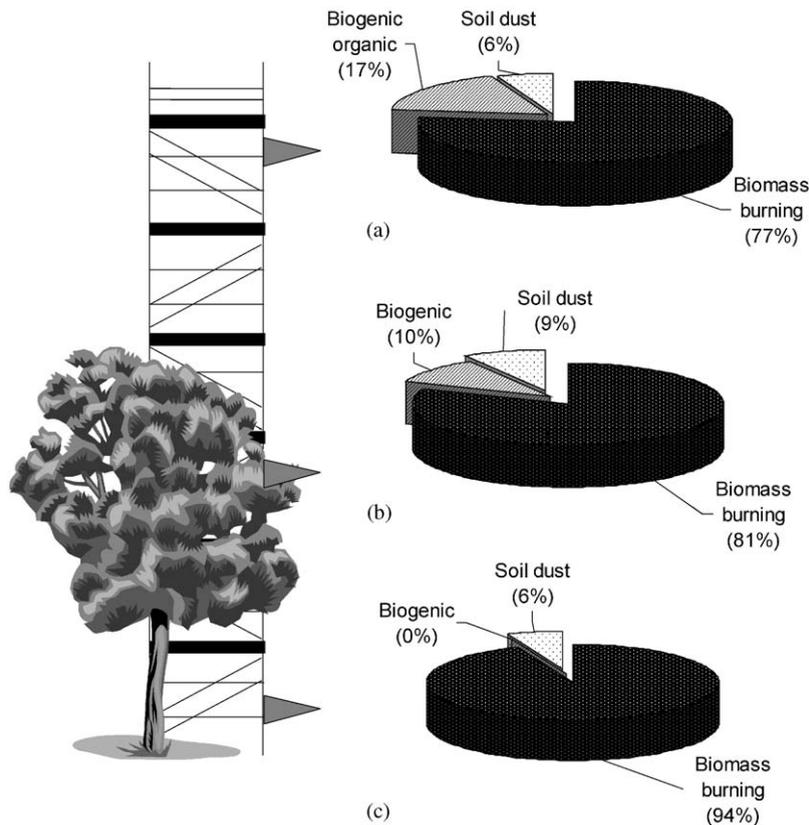
Fig. 5. Relative source apportionment of the light absorption, as measured using a reflectance photometer technique (BC_e), for the dry season aerosols sampled near the top (a), at canopy level (b), and at ground level (c) on the Jarú meteorological tower in Rondônia, Brazil, between 6 September and 31 October 1999. The gray triangles on the side of the tower represent the sampling heights, whereas the tree represents the mean canopy height. The size of the pie charts is proportional to the mean BC_e concentration measured for each level.

conclusions drawn from these results are tentative at this stage.

Overall, the same three components that were extracted from the analysis of the samples collected near the top of the tower were also observed at canopy and below-canopy heights. Fig. 4b and c show the relative mass source apportionments, obtained from APCA, for the dry season aerosol at canopy and ground level, respectively. Comparison of these apportionments with that for the above-canopy level (Fig. 4a) suggests that pyrogenic particles were responsible for the bulk of the particulate mass at all measurement heights during the dry season. Overall, the relative composition of the aerosol at all levels was very similar; however, the below-canopy level aerosol featured a larger contribution of pyrogenic and a smaller contribution of biogenic aerosol. This indicates that the above-canopy and canopy levels were probably fairly well decoupled from the below-canopy level and that they were subject to different emission/sink processes (Guyon et al., 2003c).

Fig. 5b and c present the relative apportionment of the fine fraction BC_e measured at canopy and ground

levels, respectively, to the sources identified above. The relative contribution of biomass burning aerosol to the absorption was found to increase from above to below the canopy, indicating efficient downward mixing of the fine aerosol, whilst soil dust particles probably were deposited in the forest. The source apportionment for the light absorption measured at ground level with an aethalometer agreed to within 2% with the results obtained using the BC_e at this level, indicating that biomass-burning aerosol was responsible for 92–94% of absorption at this level, with soil dust accounting for the remaining minor fraction. At ground level, particles of biogenic origin did not contribute to absorption, whereas these particles accounted for 10% of the absorption at the canopy level and 17% at the above-canopy level (ca. 30% when using the PSAP or aethalometer data). Due to the large amount of fine pyrogenic particles present in the atmosphere during this season, it is not surprising that the natural biogenic particles emitted at ground level (Guyon et al., 2003c) did not contribute significantly to absorption, whereas particles such as fine ash (which may not have been

differentiated from the biogenic component in our analyses for the upper two levels) may well have.

4. Summary and conclusions

In this study we have shown that APCA may be a useful tool for identifying and quantifying the contribution of different aerosol types to aerosol light absorption and scattering. Overall, three aerosol types were found to make up the bulk of the total aerosol mass in the Amazon Basin, namely biomass burning smoke, natural biogenic aerosol, and soil dust. Even in the wet season, the contribution of (mostly aged) biomass burning smoke to the fine aerosol was found to be substantial. All three identified components were found to contribute significantly to the optical properties of the aerosol. This suggests that along with biomass-burning emissions, biogenic and soil dust particles must be included in aerosol models for the Amazon region to account for their optical properties and climatic impacts. Our data also reveal that these components cannot be assumed to be completely independent from one another; in particular, our PCA provides evidence that pyrogenic aerosols are strongly associated with soil dust particles, a conclusion supported by single-particle analyses made using electron microscopy, which showed the two aerosol constituents to be internally mixed.

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