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LONG-TERM STUDY OF ATMOSPHERIC AEROSOLS IN CUIABÁ, BRAZIL: MULTIELEMENTAL COMPOSITION, SOURCES AND SOURCE APPORTIONMENT

W. Maenhaut[1], M.-T. Fernández-Jiménez[1], and P. Artaxo[2]

[1]Institute for Nuclear Sciences, University of Gent, Proeftuinstraat 86, B-9000 Gent, Belgium
[2]Institute of Physics, Univ. Sao Paulo, P.O. Box 66318, CEP 05315-970, Sao Paulo, S.P., Brazil

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METHODS

Atmospheric aerosol samples were collected nearly continuously from July 1992 until December 1995 at Cuiabá (16°S, 58°W, 165 m asl), in the Brazilian savanna, south of the Amazon basin rain forest. The sampling device consisted of a stacked filter unit (SFU), which separates the aerosol into a coarse (2-10 μm equivalent aerodynamic diameter (EAD)) and a fine (<2 μm EAD) size fraction, and the collection time per sample was typically 3-5 days. The coarse and fine filters of all SFU samples (180 in total) were analyzed for the particulate mass (PM), black carbon (BC), and up to 47 elements (from Na upward). The multielemental analyses were done by instrumental neutron activation analysis and particle-induced X-ray emission (Maenhaut *et al.*, 1996). Absolute principal component analysis (APCA) (Maenhaut and Cafmeyer, 1987) and chemical mass balance (CMB) receptor modelling techniques were used to identify the major aerosol components (source types) in each of the two size fractions and to apportion the PM and the various aerosol constituents to these components.

RESULTS AND DISCUSSION

PM, BC and virtually all elements exhibited clearly higher atmospheric concentrations during the dry (biomass burning) season (July-September) than during the rest of the year. During the dry season, the levels of fine and coarse PM were in the range of 5-50 and 10-80 $\mu g/m^3$, respectively. The time trend for the fine PM is shown in Fig. 1. For the fine size fraction, 5 components were identified by APCA, i.e., mineral dust (with loadings of about 0.9 for the crustal elements Al, Si, Sc, Ti, Fe, La, Sm, and Th), a biomass burning (pyrogenic) component (with PM, BC, S, Cl, K, Zn, Br, Rb, and I having loadings in the range 0.7-0.9), two urban pollution components (one with V, Cu, As, and Sb; the other only highly loaded with Pb), and an almost pure Na component. As already indicated by Maenhaut *et al.* (1996), the pyrogenic component probably also contains some contribution from biogenic aerosols (plant emissions), as there are only few elements that enable one to discriminate between the pyrogenic and biogenic particle types (see also Echalar *et al.*, 1998). APCA attributed on average 55% of the fine PM to the pyrogenic component, 30% to the mineral dust, 10% to the first urban component (the one with V, Cu, As, and Sb), and less than 5% each to the Pb and Na components.

For the CMB calculations on the fine data set we used three source profiles, one for mineral dust, one for pyrogenic aerosol from savanna fires (Maenhaut *et al.*, 1996), and a pure $(NH_4)_2SO_4$ profile. The observed PM levels were well predicted by this 3-source CMB. The mean (total CMB-predicted)/observed concentration ratio and associated standard deviation were 0.95±0.30, and although there was thus some variability in the individual ratios, there was no systematic difference between dry and wet seasons. The CMB apportioned on average 70% of the fine PM to the pyrogenic aerosol type, 15% to the mineral aerosol, and 10% to ammonium sulphate. The average CMB attribution of 70% of the fine PM to the pyrogenic particles is in reasonable agreement with

the 55% attribution obtained in the APCA. A very remarkable finding was that the relative contribution from the pyrogenic aerosol to the fine PM varied only little over the entire sample set. It remained also around 60-70% during the wet season, but the absolute concentration levels of the pyrogenic particles were obviously much lower then. The time trend of the percentage contribution of the pyrogenic aerosol to the total CMB-modeled fine PM is displayed in Fig. 1. During the wet season, a substantial fraction of the natural biogenic aerosol most likely ended up in our pyrogenic aerosol contribution.

For the CMB on the coarse data set only two source profiles were used, i.e., one for mineral dust and a pure ammonium sulphate profile. The mineral dust contributed on average 75% to the experimental coarse PM and sulphate only 3%, so that the unexplained mass amounted to about 20%. This percentage varied with season, however. During the dry season, only 10% of the mass remained unexplained by our CMB, versus 30% during the wet season. The unexplained mass in the wet season is thought to be mainly attributable to natural biogenic emissions.

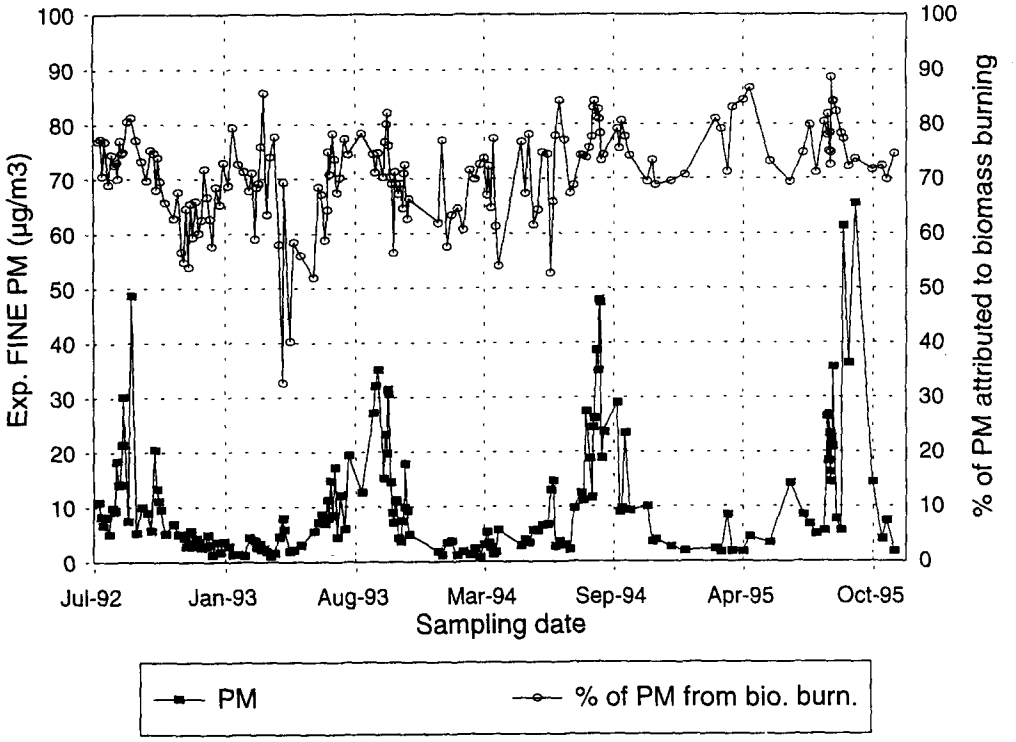

Fig. 1. Time trend of the fine (<2 µm EAD) PM at Cuiabá and percentage contribution of the pyrogenic aerosol to the total CMB-modeled fine PM.

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