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Aerosol emissions by tropical forest and savanna biomass burning: characteristic trace elements and fluxes

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Abstract. Aerosol particles were collected during the dry season in the amazonian forest and in African and Brazilian savannas. Sampling was performed inside the plumes of vegetation fires and in the local background atmosphere. Samples were analysed for their elemental composition by Particle Induced X-Ray Emission (PIXE). Enrichment factors relative to the local background were calculated. Savanna fire aerosols are characterised by enrichments in elements like K, P, Cl, Zn, and Br, whereas forest fire emissions are enriched in Si and Ca. The unexpected low potassium enrichment factor for the forest fires could be due to the prevailing smoldering conditions, whereas the high values of potassium enrichment factor during savanna fires could be associated with the predominant flaming conditions. This result indicates that potassium may be considered a good tracer of the flaming phase of fires only. Emission factors of P, S, Cl, K, Ca, Mn, Zn, Br and total particulate matter were calculated for African savanna fires. Our estimates of the annual potassium and zinc emissions by tropical savanna fires indicate that the contribution of this source should be taken into account to understand the biogeochemical cycle of these elements.

Introduction

Biomass burning emission of gases and particles to the atmosphere attracts nowadays much attention due to their possible effects on the chemistry of the atmosphere and on climate [Crutzen and Andreae, 1990]. Biomass burning emissions are concentrated in the tropical belt, where is consumed more than 80% of biomass burnt in the world, a half of it during savanna fires [Andreae, 1991]. Biomass burning might represent also a significant source of trace elements that would have to be considered for the study of their global biogeochemical cycles. Indeed, the global extension of biomass burning activities results in a large amount of gases and particles injected into the atmosphere, and trace elements present in the bulk plant material or in particles previously deposited on the vegetation may be found in these products. On a local scale, the injection of a

significant amount of trace elements into the atmosphere may also be important as a pathway to exporting nutrients from the source region.

Previous works on trace element emission by fires in the tropical belt are scarce. Ward et al. [1991 and 1992] present the elemental content of particles emitted by rain forest and cerrado fires in Brazil. Artaxo et al. [1993] present the trace element content of particles obtained by airborne sampling of fire plumes in the Amazon basin. Quantitative estimates of the intensity of emissions in the tropical belt are still scarcer: besides the estimate of potassium emission by fires in the Amazon basin [Andreae, 1988], to our knowledge only one data set is available: that obtained from our ground-based measurements conducted in Ivory Coast [Gaudichet et al., 1992].

The main objective of this work is to characterise and compare trace element emission from fires of three different types of savannas and from the south-western amazonian rain forest. The aim of this study is to verify if there is a fingerprint that may characterise savanna fires or, in a more general scale, tropical biomass burning. We will also estimate the intensities of the trace element emissions and try to assess the strength of the biomass burning source.

Experimental Sites and Methods

Samples were collected at four ecologically different sites:

- (a) the guinean savanna: in Lamto, Ivory Coast. The vegetation at this site is representative of West Africa dense humid savannas (above ground biomass density of about 9 ton/ha [Lacaux et al., 1993]); these samples were collected in January 1991 during the FOS/DECAFE experiment;
- (b) the south African bush: in the Kruger National Park, located in the NE part of South Africa and constituted by very dry low density savannas (1.4 to 5 ton/ha [Trollope et al., 1993]);
- (c) the Brazilian savanna, or "cerrado": in the IBGE (Brazilian Institute of Geography and Statistics) Ecological Reserve, located 35 km south of Brasília; one grassland and two wooded savanna plots were burnt (typical densities of 7 to 10 ton/ha [Kauffman et al., 1994]);
- (d) in the south-western part of the Amazonian rain forest, about 100 km south of Porto Velho, in the State of Rondônia (about 360 ton/ha [Kauffman et al., personal communication]).

While in the savanna plots fire was always set to the

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standing vegetation, in rain forest plots most of trees had been cut down 40 to 60 days before the fire, otherwise they would not have been dry enough to burn efficiently.

An important difference that must be pointed out is that most of the savanna vegetation burns quickly during the flaming phase, while in the forest trunks may remain on field and keep smoldering or glowing several days after the fire. According to Ward et al. [1992], more than 97 % of carbon in cerrado fires is released during the flaming phase, while in forest fires most of biomass is consumed during the smoldering phase. In the following discussion all results concerning savanna fires refer to samples collected under flaming conditions, while results concerning tropical forest fires refer only to samples collected in the individual plumes of smoldering or glowing trunks.

The sampling and analyses protocols are described in detail in Gaudichet et al. [1992]. In short, aerosol sampling was carried out in biomass burning plumes using an autonomous and mobile pumping unit. This sampling set-up allowed the simultaneous collection of different types of samples in parallel, in the fire plumes or in background conditions.

Parallel to the aerosol sampling, CO₂, in Ivory Coast, and CO₂ and CO, in South Africa, were measured in real time using a portable infrared spectrometer (BINOS analyser), in order to obtain a quantitative estimate of the intensity of aerosol emissions.

Trace element concentrations were obtained using the PIXE method [Johansson and Campbell, 1990]. Filters were weighed twice, with a Mettler M3 electronic balance, before and after the sampling to obtain the particulate mass of the samples.

Parallel to some of our trace elements samples, particles were collected on Whatman GF/F glass fiber filters for black carbon analysis by coulometric titration [Cachier et al., 1993].

Similar measurements were carried out in the background atmosphere of the tree savanna sites.

Results and Discussions

Characteristic Trace Elements

Particles sampled in plumes of savanna and forest fires may have two different sources: the combustion of vegetation or the remobilisation of particles that were previously deposited on the vegetation. Trace elements that may characterise biomass burning should be those for which the combustion of the vegetation is the main source.

To determine which are these elements our first step was to assess the characteristics of the particles deposited on the vegetation. The trace element content measured in the background aerosol is supposed to reflect these characteristics. In the Ivory Coast, the situation was simple: the atmospheric aerosol is composed of particles emitted by savanna fires and by soil dust [Gaudichet et al., 1992]. But in the other sites source apportionment was more complex. Our approach then was to use enrichment factors of each element in the plume with reference to its concentration in the local background. Iron, which is assumed not to be predominantly produced by the combustion of the vegetation, was used here as the reference element. Median values of these enrichment factors are shown in Figure 1.

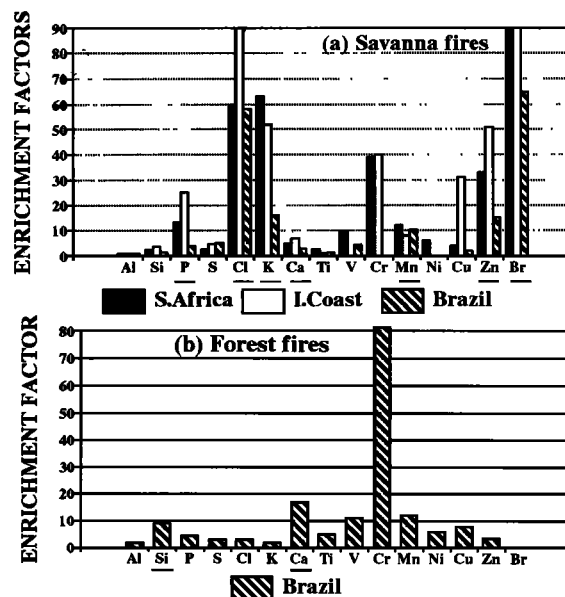


Figure 1. Median trace element enrichment factors for (a) savanna and (b) forest fire aerosols: $(X/Fe)_{\text{plume}} / (X/Fe)_{\text{background}}$. Background aerosol data for the amazonian forest obtained from Artaxo et al. [1988]. Forest samples correspond to smoldering conditions, while savanna samples correspond to flaming conditions. Elements underlined are enriched and have ratios to Fe in the plume statistically different from the same elemental ratios in the background atmosphere according to Student's t-test at the 95% confidence level.

For the three types of savanna a similar pattern may be observed in this figure: biomass burning particles are very enriched in K, Cl, Br, and Zn (enrichment factor > 10) and moderately enriched in P, Ca, and Mn ($4 < \text{enrichment factor} < 10$). All these elements are present in plants [Hewitt, 1983] and may be associated to the combustion of vegetation. By contrast, biomass burning particles present low enrichment factors for Al and Si, indicating that the contribution of the combustion of vegetation to the measured concentration is small. It must be pointed out that low enrichment factors do not mean low concentrations. In guinean savanna samples, for example, Si, Al and Fe concentration levels are very high, the low enrichment factors indicating that the fire induced an intense remobilisation of terrigenous particles deposited on the vegetation [Gaudichet et al., 1992]. There are some differences regarding some minor elements: V is enriched in cerrado and south-African savannas plumes, while Cr and Ni are enriched mostly in African savanna plumes. These enrichments are somewhat surprising, as they are elements usually associated with industrial pollution. The re-entrainment of industrial particles can not be excluded, but, as these elements may be present in the plants [Hewitt, 1983], a "biomass burning" origin must also be considered. In the forest the pattern is different: aerosols are enriched in Ca, Si. The unexpected low enrichment in K, Cl, Zn and P of forest fire aerosols may be explained by the intensity of natural emissions from the vegetation. These emissions dominate the background aerosol and are themselves rich in such elements [Artaxo et al., 1988]. This result suggest

Table 1. Excess potassium to soot carbon ratios in the fire plumes and in the background atmosphere. Excess potassium corresponds to the measured concentration corrected for the crustal contribution.

Type of fire	K/Cs	Reference
guinean savanna fires	0.64±0.28	Cachier et al. [1993]
S. African savanna fires	1.14±0.96	this work
cerrado fires	0.52±0.61	this work
forest fires	0.055±0.039	this work
wood smoke	0.007 ± 0.002	Currie [1991]
aged plumes of forest fires in Amazonia	0.11	Andreae et al.[1988]

that Si or Ca might be considered better markers for wood fire than potassium, which is commonly presented as an ubiquitous tracer of biomass burning aerosol. However, due to the wide variety of Si and Ca sources, their role as tracers is questionable.

In order to check our puzzling K results, we introduce the potassium to black carbon ratio (Table 1), which is often used as a tracer of biomass burning aerosols [Cachier et al., 1993]. This ratio varies in the range 0.007 - 1.14, showing lowest values for wood burning. Our forest results are then in agreement with previous measurements.

The K content in particulate emissions from savanna fires (3.2 to 4.4 %) is about 10 times higher than in our samples from forest fires (0.3 ± 0.1 %). It is necessary to verify if this difference is related to a different vegetation composition or to differences in the prevailing fire stage (flaming for savannas versus smoldering for the forest). Potassium contents of the vegetation matrix present a smaller variation (minimum of 0.16±0.30% in the forest [Kauffman et al.; personal communication] to about 0.60 % in cerrado [Kauffman et al.; 1994]), so the observed differences in K aerosol contents may then be attributed to the different combustion processes: flaming x smoldering. Ward et al. [1992] in another amazonian forest site observed also that potassium content in the smoldering phase is lower than in the flaming phase. We suggest that the thermodynamic conditions of the smoldering phase hinders the volatilisation of potassium; it may be supposed also that most of the potassium has been used up during the flaming phase of the forest fires. However, smoldering results from Ward et al. [1992] are nearly one order of magnitude higher than those observed in our samples. This difference might be explained by the fact that Ward et al.'s smoldering samples have been collected just after the flaming phase, and could have integrated the plumes of smoldering vegetation and aged products from the flaming phase, whereas our samples have been collected later, in the individual plumes of smoldering.

Source Strength and Flux Estimates

Quantitative assessments rely on estimates of emission factor. The emission factor of a given element represents the amount of this element emitted to the atmosphere per unit mass of vegetation consumed by the fire. The computation of emission factors was carried out using the following equation:

$$EF_x = k \times \Delta X / \Delta CO_2$$

where Δ are the above background concentrations and k is a numerical factor indicating the proportion of fuel carbon released as CO_2 .

In the case of African savanna fires it was possible to calculate emission factors for every sample (Table 2), using our CO and CO_2 data, carbon content in ashes and partially burnt material [T. Kuhlbusch et al., unpublished manuscript], and literature data on the emission of other carbonaceous products [Lobert et al., 1991; Bonsang et al. 1993]. Taking account of the emission factors variability at each sampling site, it may be considered that there is little difference between emission factors obtained at two different ecosystems. Consequently they may be characteristic of tropical savanna fires and may be used in global estimates.

To get an idea of the importance of the atmospheric transfer of trace elements by biomass burning, some comparisons with other local or regional sources may be proposed. The flux of a given element may be estimated as the product of its emission factor, the biomass density exposed to fire, the surface burnt, and the fraction of biomass exposed to the fire that is actually burnt, i.e., the burning efficiency.

Global fluxes from tropical savanna fires were computed assuming that the amount of biomass consumed each year is 3700 tons of dry matter [Andreae, 1991]. These results (Table 2) indicate that pyrogenic emissions might have an impact on the global cycle of some trace elements. For Zn, the main source is anthropogenic: savanna fire emissions could correspond to about 6 % of all anthropogenic emissions and about 20% of the total natural emissions (131×10^3 ton/year [Nriagu and Pacyna, 1988]; and $45 \times$

Table 2. Median and ranges of emission factors from african savannas fires, and the annual flux emitted to the atmosphere by the fires in all tropical savannas. Fluxes (in Tg/year) were estimated using the mean of the two median emission factors, and the amount of biomass burnt each year in the tropical savannas (3690 Tg/year [Andreae, 1991]). In parentheses the number of points for which the emission factors were calculated.

	South Africa (g/kg dry matter)	Ivory Coast (g/kg dry matter)	FLUX (Tg/year)
P	0.0047 (14) 0.00052 - 0.017	0.011 ^t (12) -0.0020 - 0.050	0.029
S	0.052 (17) 0.028 - 0.12	0.020 ^t (12) 0.0030 - 0.050	0.13
Cl	0.28 (17) 0.11 - 0.50	0.22 ^t (12) 0.057 - 1.2	0.92
K	0.30 (17) 0.16 - 0.61	0.34 ^t (12) 0.067 - 1.0	1.2
Ca	0.013 (17) 0.0056 - 0.062	0.030 ^t (12) 0.0030 - 0.15	0.080
Mn	0.0013 (16) 0.00043 - 0.0039	0.0015 (4) 0.00018 - 0.0033	0.005
Zn	0.0012 (16) 0.00055 - 0.0047	0.0032 ^t (11) 0.00036 - 0.0089	0.008
Br	0.0026 (12) 0.0015 - 0.0057	0.0045 (4) 0.0019 - 0.017	0.013
TPM	6.5 (17) 2.9 - 16	7.9 ^t (4) 1.2 - 16	26

^t from Gaudichet et al. [1992].

10³ ton/year [Nriagu, 1989] respectively).

On a regional scale the emission of trace elements to the atmosphere might be comparable to loss of trace elements by run-off. This is the case of West Africa, where we estimated the release of K to the atmosphere by savanna fires to be about 2.25 kg/ha/year, which is comparable to the loss of potassium by run off (1.4 kg/ha/year [Lesack et al., 1984]). Thus, in this region the emission to the atmosphere by the fires may play an important role and must be considered for the K cycle.

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