

Particulate Content of Savanna Fire Emissions

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Abstract. As part of the FOS-DECAFE experiment at Lamto (Ivory Coast) in January 1991, various aerosol samples were collected at ground level near prescribed fires or under local background conditions, to characterize the emissions of particulate matter from the burning of savanna vegetation. This paper deals with total aerosol (TPM) and carbon measurements. Detailed trace element and polycyclic hydrocarbon data are discussed in other papers presented in this issue.

Near the fire plumes, the aerosols from biomass burning are primarily of a carbonaceous nature ($C\% \sim 70\%$ of the aerosol mass) and consist predominantly of submicron particles (more than 90% in mass.) They are characterized by their organic nature (black to total carbon ratio C_b/C_t in the range 3–20%) and their high potassium content ($K/C_b \sim 0.6$). These aerosols undergo aging during their first minutes in the atmosphere causing slight alterations in their size distribution and chemical composition. However, they remain enriched in potassium ($K/C_b = 0.21$) and pyrene, a polycyclic aromatic hydrocarbon, such that both of these species may be used as tracers of savanna burning aerosols. We show that during this period of the year, the background atmosphere experiences severe pollution from both terrigenous sources and regional biomass burning (44% of the aerosol). Day-night variations of the background carbon concentrations suggest that fire ignition and spreading occur primarily during the day. Simultaneous TPM and CO_2 real-time measurements point to a temporal and spatial heterogeneity of the burning so that the ratio of the above background concentrations ($\Delta TPM/\Delta CO_2$) varies from 2 to 400 g/kg C. Smoldering processes are intense sources of particles but particulate emissions may also be important during the rapidly spreading heading fires in connection with the generation of heavy brown smoke. We propose emission factor values (EF) for aerosols from the savanna biomass burning aerosols: $EF(TPM) = 11.4 \pm 4.6$ and 69 ± 25 g/kg $C_{dry\ plant}$ and $EF(C_t) = 7.4 \pm 3.4$ and 56 ± 16 g C/kg $C_{dry\ plant}$ for flaming and smoldering processes respectively. In these estimates, the range of uncertainty is mostly due to the intra-fire variability. These values are significantly lower than those reported in the literature for the combustion of other types of vegetation. But due to the large amounts of vegetation biomass being burnt in African savannas, the annual flux of particulate carbon into the atmosphere is estimated to be of the order of 8 Tg C, which rivals particulate carbon emissions from anthropogenic activities in temperate regions.

Key words: particulate carbon, total particulate matter, savanna burning, tropical aerosols, emission factors.

1. Introduction

The growing concern about the potential effects on the radiative balance of the atmosphere by tropospheric aerosols has increased interest in their natural and anthropogenic source strengths and the factors governing their variability. Up to now, the focus has been placed on the important atmospheric loading of sulphate aerosols (Charlson *et al.*, 1987, 1992) with controversial theses on the origin of these particles and their role in climate control (Schwartz, 1988). But in spite of our patchy knowledge of their spatial and temporal distributions, aerosols emitted by combustion processes are also likely to contribute to the global burden of tropospheric fine aerosols (Heintzenberg, 1989; Cachier *et al.*, 1990) and they have recently been proposed to counteract the anthropogenic greenhouse warming, either by a direct reflection of incoming solar radiation or a modification of the cloud cover (Ghan and Penner, 1992).

A comparison of estimates of worldwide fuel consumption shows that the burning of fossil fuels (Boden *et al.*, 1990) and that of vegetation fuels (Levine, 1990; Houghton, 1991) roughly consume the same amount of material annually, about 5.10^{15} g C each. Both types of combustion produce primary and secondary fine aerosols that are potentially radiatively effective. It might be assumed here that due to the less favorable physical conditions of the combustions, biomass burnings are more effective producers of particles, on average, than industrial combustion processes.

More than 80% of the biomass burning occurs in the tropics where among the various type of burns, the savanna and bushfires are really the most important. Due to the importance of savanna areas in Africa and the increasing occurrence of the burns (Cahoon *et al.*, 1992), it appears that now more than 30% of the tropical biomass is burned during savanna fires on this continent alone (Andreae, 1991). Thus, the large-scale issue of an influence of particulate emissions by African savanna fires on the regional and global atmospheric environment has to be addressed.

In light of few previous data and in an effort to improve our knowledge on particulate emissions by savanna biomass burning, we set up ground experiments at Lamto, a savanna site in the Ivory Coast, as part of the FOS-DECAFE/91 campaign during which various prescribed savanna fires were explored. For the first time, qualitative and quantitative data were obtained in the field that document the physico-chemical aspects of the aerosol phase, in order to provide tracers of this type of biomass combustion and to propose emission factors for the main elements found in the particles. The various ground-based measurements which were performed with regard to the aerosol phase are listed in Table I, with references to the main papers appearing in this issue. The present study focusses on the bulk aerosol (TPM: total suspended matter) and its main component, the carbonaceous fraction.

TABLE I. Aerosol phase samplings and measurements performed during the DECAFE savanna fire experiment at Lamto (Ivory Coast) in January 1991. Main papers of this issue are indicated in the last file

Total Suspended Matter (TPM) ^{a, c}	Optical analysis (RAM, MIE INC.) or weighing nucleopore filters	Continuous sampling	This work
Total carbon (Ct) ^{a-c}	Coulometry (Strohlein analyser)	Glass-fiber filter (GF/F Whatman)	This work
Black carbon (Cb or "soot") ^{a-c}	Optical (MAGEE aethalometer) or thermal analysis and coulometry	Glass-fiber filter (GF/F Whatman)	This work
Individual observation of particles ^{a, c}	Electron transmission microscopy (Jeol 100 CX II microscope)	Nucleopore filter (0.4 μm)	Gaudichet <i>et al.</i>
Trace element content ^{a, c} (S, Cl, K, Mg, Na, Ca, P, Si, Zn...)	XRF, PIXE	Nucleopore filter (0.4 μm)	Gaudichet <i>et al.</i>
Molecular tracers: PAH ^{b, c} (polycyclic aromatic hydrocarbons)	HPLC Spectra-Physics 4270	Glass-fiber filter (GF/A Whatman) and Amberlite XAD ₂ adsorbant	Masclet <i>et al.</i>
Particle size-distribution ^{b, c}	Sierra model 235 cascade impactor	Glass-fiber filter (GF/F Whatman)	This work
Smoke optical properties ^{a-c} (optical depth, size-distribution)	Portable sunphotometer (449-648-845-940-937 nm)	Discontinuous measurements	Lioussé <i>et al.</i>
Polonium-210 ^{a-c} (normalization)	Alpha-spectrometry	Glass-fiber filter (GF/F Whatman)	LeClouarec <i>et al.</i>
Carbon dioxide CO ₂ ^{a, c} (normalization)	Infra-red Binocs analyser	Continuous sampling	This work and Gaudichet <i>et al.</i>

^a in the fire plume; ^b 20 m from the plume; ^c in the background atmosphere.

2. Experimental

2.1. AEROSOL COLLECLTION STRATEGY

Prescribed savanna fires were sampled at Lamto in January 1991 during the FOS-DECAFE experiment. An exhaustive description of the experiment is given in this issue by Lacaux *et al.* (1995). Briefly, the sampling site of Lamto (6° N, 5° W) is favorably situated in a moderately inhabited area of the Guinean savanna of the Ivory Coast. During the dry season (from November to March), the whole region experiences severe savanna bush fires whose frequency and intensity generally peak in January. Four experimental plots and 2 large fires were ignited to allow the sampling of gases and aerosols emitted during the various combustion phases and types of burning.

Sampling of the fire plume aerosols was performed with power-autonomous mobile pumping units each consisting of a 12v pump, battery and gasmeter. Three filter holders were mounted together at the head of the same pole in order to avoid artefacts due to the spatial heterogeneity of the fire plumes. The sampling height was about 4 meters above ground level and the average sampling duration was 3 minutes. Aerosols were simultaneously collected by low volume pumping (1 to 2.5 m³/hr) on a glass fiber filter for particulate carbon and polonium-210 analyses, and on 2 nuclepore filters for the determination of trace element contents and particle morphology investigations. Two additional sampling lines were provided for the continuous monitoring of total suspended matter (TPM) and CO₂ in order to allow emission factor (EF) assessments. The combustion gas flow transit time from the inlet to the sensors was kept to a few seconds by minimizing volumes in the connecting tubings. In the field, this low time constant proved to be valuable in finding a favorable position for the samplers in the fire plumes. Because of the temporal variability of each burn and the maximum amount of aerosol material which can be accumulated on the filters, a sampling time of 3 to 5 minutes intervals was chosen. This sampling strategy allowed the collection of 2 to 5 samples per prescribed fire and a description of the different phases of each burn (Table II).

Two high volume samplers were operated at ground level with a power generator at a distance of 10 to 100 meters from the fire hearth. The mean sampling duration was 20–30 minutes in this case, leading to samples representing an integration over the spatial and temporal variability of the burns. A Sierra bulk aerosol sampler operating at 20 m³/hr was fitted with a cartridge holder to provide samples for the analysis of gaseous and particulate polycyclic hydrocarbons (PAH). The second sampler run in parallel to the first was a Sierra model 235 cascade impactor dedicated to measurements of the size-distribution of the carbonaceous particles. This impactor was modified by reducing the number of stages from 5 to 3; as the instrument was operated at the flow rate (68 m³/hr) recommended by the manufacturer, it may be assumed that the aerodynamic diameter cutoffs for the particles collected on each stage were maintained.

TABLE II. Total Particulate Matter (TPM) and Carbon results for prescribed savanna fire samples

Plot	Sample n°	Type of burn	ΔCO_2 (ppm)	ΔTPM (mg/m ³) optical data	ΔTPM (mg/m ³) weight	% Fine particles in TPM (<2 μm)	Cb ($\mu\text{gC}/\text{m}^3$)	Ct ($\mu\text{gC}/\text{m}^3$)
<i>Plot 1</i> (<i>Ludetia</i>) Fire efficiency 83%	F ₁₁	Backing fire	163	0.62			164	1336
	F ₁₂	Heading fire	274	1.12			218	2057
<i>Plot 2</i> (<i>Ludetia</i>) Fire efficiency 91%	F ₂₁	Backing fire	467	1.62			81	987
	F ₂₂	Backing fire	281	1.74			137	999
	F ₂₃	Heading fire	1099	3.65	2.62	86%	489	6224
<i>Plot 3</i> (<i>Hypparhenia</i>) Fire efficiency 84%	F ₃₁	Backing fire	910	1.13			117	929
	F ₃₂	Backing fire	1305	2.40	2.32	82%	338	3342
	F ₃₃	Heading fire	1169	12.98			283	2497
	F ₃₄	Smoldering fire	97	2.66			266	6045
<i>Plot 4</i> (<i>Hypparhenia</i>) Fire efficiency 65%	F ₄₁	Backing fire					433	4386
	F ₄₂	Backing fire	972	11.64	18.25	66%	621	5343
	F ₄₃	Backing fire	1210	2.31			190	1829
	F ₄₄	Heading fire	1466	5.90	6.09	46%	613	4285
	F ₄₅	Smoldering fire	346	57.9			930	13121
<i>Big fire 1</i> (<i>Ludetia</i>)	BIF ₁₁	Heading fire	1789	12.77			68	644
	BIF ₁₂	Smoldering fire	42	1.65	0.52	36%	20	711
<i>Big fire 2</i> (mixed)	BIF ₂₁	Backing fire					242	1745
	BIF ₂₂	Backing fire					97	501
	BIF ₂₃	Heading fire			0.60	86%	307	1822
	BIF ₂₄	Mixed fire					274	2340

In addition, several day and night aerosol samples were collected at a site two kilometers apart from the low anthropogenic activity of the Lamto station. In these conditions and at this period of the year when savanna fires are important, it may be assumed that results obtained from these samples will be representative of the regional atmospheric background of biomass burning particles. The average filtration volume was 20 m^3 . Atmospheric black carbon concentrations were also recorded in parallel in real-time using an aethalometer (Magee Scientific Inc.).

Finally, sunphotometer measurements at 5 different wavelengths (from 450 to 940 nm) were made and used to retrieve atmospheric water vapor contents, aerosol optical depths and the particle size distribution of savanna fire smokes (Lioussé *et al.*, 1995, this issue).

2.2. TOTAL PARTICULATE MATTER DATA (TPM)

TPM data were obtained in real time by optical measurements with a GCA Corporation real-time aerosol monitor (RAM-1, MIE Inc.). The principle of this instrument is based on the detection of near forward scattered light in the near infra-red region. The different ranges covered the important TPM variability observed on field (from 0.5 to 200 mg/m^3).

The instrument is operated at a flow rate of 2.2 l/min and is precalibrated with a standard powder (Arizona road dust of $1.5 \mu\text{m}$ mean diameter and 2.61 density). Controlled burns conducted in a combustion chamber at Lannemezan (France) with a number of different temperate and tropical grasses have been used to calibrate the instrument. The production of carbonaceous aerosols was reproducible with a ratio of black to total carbon (Cb/Ct) of $8.5 \pm 2\%$ and a very consistent correlation between the TPM recorded with the RAM and the carbon content of the aerosols: $\text{Ct/TPM} = 0.26 \pm 0.03$ (16 samples).

The average carbon content usually quoted for biomass burning smoke is $\sim 66\%$ (Crutzen and Andreae, 1990). This experiment thus suggests that the calibration of the instrument gives TPM overestimates by at least a factor of 2. In the field the agreement between TPM and total carbon measurements is less evident, and some discrepancies are seen especially during extreme burning conditions which may be characterized either by hot flaming burns with low particulate emissions or smoldering burns with particularly high particle content. From these results, it is suggested that the RAM optical sensor response shows a significant variability which may be related to the proper variability in size and chemical content of the particles emitted during the various phases of the combustion. Consequently, further improvement of the use of such sensors during biomass burning experiments should be based on calibrations for various combustion phases; these calibrations might be obtained in the combustion chamber.

TPM was also determined in a clean laboratory under controlled atmosphere by weighing the Nuclepore membranes devoted to trace element analyses. These membranes (0.4 and $8 \mu\text{m}$ pore size) mounted on stack unit filter holders (SFU)

could allow the separation of fine ($< 2 \mu\text{m}$) and coarse particles (Andreae, 1983; Artaxo *et al.*, 1988). A comparison of 5 samples (Table II) gave agreement between the 2 TPM data sets generally better than 40%. For one of the sample collected during smoldering conditions (BIF₁₂) the disagreement is more important, confirming the tendency of the RAM instrument to overestimate the carbon content.

In the present study, therefore, TPM optical data will be used primarily for qualitative assessments and an interpretation of the whole data set. Quantitative assessments mostly rely on gravimetric measurements, the optical data used were corrected for smoldering combustion or combustion chamber experiments only.

2.3. CARBON MEASUREMENTS

Precleaned Whatman GF/F glass-fiber filters were used as collection surfaces for the purpose of carbon analyses. Among other available fiber filters, these filters have a more favorable collection efficiency ($\Phi < 0.7 \mu\text{m}$) and show a satisfactory resistance to humidity. In our analytical protocol routinely applied in the laboratory (Cachier *et al.*, 1989a), the aerosol samples are decarbonated prior to analysis. Then, their black carbon (Cb, also referred to as soot carbon) and total carbon (Ct = organic carbon + black carbon) contents are determined in aliquots of the same filter deposits by coulometric titration with a Ströhlein Coulomat 702C analyser. Carbon contents are obtained with a precision better than 10%.

For the background atmosphere, the continuous aerosol sampling mode and the number of samples (17 samples) has allowed the calculation of the average carbon concentration as the arithmetic mean with its standard deviation. In the fire plumes however, as quoted by Lobert *et al.* (1991), due to the patchiness of the sampling, average concentrations are better obtained by their median values and presented with their quartile range.

Black carbon real-time data were also obtained in parallel with an aethalometer. In this instrument, the aerosols are collected on a quartz-fiber filter and analysed by the attenuation of a transmitted visible light beam. Due to high ambient black carbon concentrations, the adopted time-base was 2 minutes. As previously shown by Lioussé *et al.* (1993), the calibration of the aethalometer (i.e., the conversion of the optical attenuation toward a mass of deposited black carbon into the help of σ , the specific attenuation cross-section) had to be scaled to the aerosols generated by the biomass burning aerosols, with a value of $25 \text{ m}^2/\text{g}$ instead of $19 \text{ m}^2/\text{g}$ generally used in the instrument runs.

3. Results

3.1. ELEMENTAL COMPOSITION OF THE SAVANNA BIOMASS BURNING AEROSOLS

As shown in Figure 1a, the aerosol particles in the fire plume contain primarily carbonaceous material (ratio $\text{Ct}_{\text{mean}}/\text{TPM}_{\text{mean}} = 73\%$) with a relatively small

black carbon component ($Cb_{\text{mean}}/TPM_{\text{mean}} = 7\%$). These aerosols also contain several trace elements. If the vegetation matrix is the origin of these elements in the smoke particles, some of them such as Cl or trace metals show an enrichment in the aerosols indicating a preferential or sole release in the particulate phase during the combustion process. This is particularly evident for K which is present to 0.30% by mass in the savanna grass (Cachier *et al.*, 1991a). Potassium is the most abundant trace element in the savanna biomass burning aerosols with 5.5% of the particle mass. Conversely, Si, a major constituent of the grass is not particularly abundant in the smoke particles and should be predominantly found in ashes.

The average aerosol mass concentration (TPM) in the background atmosphere is high: $98 \mu\text{g}/\text{m}^3$. The distribution of elements in the background aerosols is shown in Figure 1b. Carbon is still the main element (14.5% of the TPM) with a black component representing 3.5% of the TPM, which indicates unambiguously an overwhelming contribution from combustion sources. But the presence of other components such as Si, Al, Ca or Fe traces a significant contribution of terrigenous matter as expected during the dry season in this region. Based on saharan dust composition with an average Al abundance of 7.1% (Bowen, 1966), this contribution is estimated to be 56%.

With the exception of O, H and N, our data include the main elements of the aerosol phase. The average N content of the aerosols is estimated to be of the order of 4% (Cachier *et al.*, 1991). Taking into account a minor terrigenous contribution ($\sim 4\%$ of the TPM) in the fresh smoke aerosols (Gaudichet *et al.*, 1995, this issue), it can be roughly estimated that the C/O ratio in fresh smoke carbonaceous particles is of the order of 9. Although much less abundant than in the vegetation where the C/O ratio is of the order of 1, the presence of oxygen suggests the image of carbonaceous particles comprising numerous organic functions. These oxygenated functions might partially account for the hydrophilic behavior of the biomass burning particles in the atmosphere (Ducret and Cachier, 1992; Rogers *et al.*, 1991).

3.2. FINE COMPONENT OF THE AEROSOLS

Savanna fires are likely to produce copious amounts of fine particles (Cros *et al.*, 1981). Table II shows the mass percentage of small particles ($\leq 1 \mu\text{m}$) in the TPM collected in the fire plumes, on the second filter of the stack filter units (SFU). Under flaming conditions these small particles always form the predominant fraction of the aerosols, $66 \pm 10\%$ on average. One sample (BIF₁₂) obtained during smoldering combustion has fewer small particles (36%), confirming previous results which indicated the tendency of low temperature burning processes to produce a coarser aerosol (Radke *et al.*, 1988).

In the background atmosphere, the fine mass fraction of the aerosols represents $27 \pm 6\%$ of the TPM mass only. This result may be easily explained by the important amounts of dust particles (24%) found in the atmosphere during the

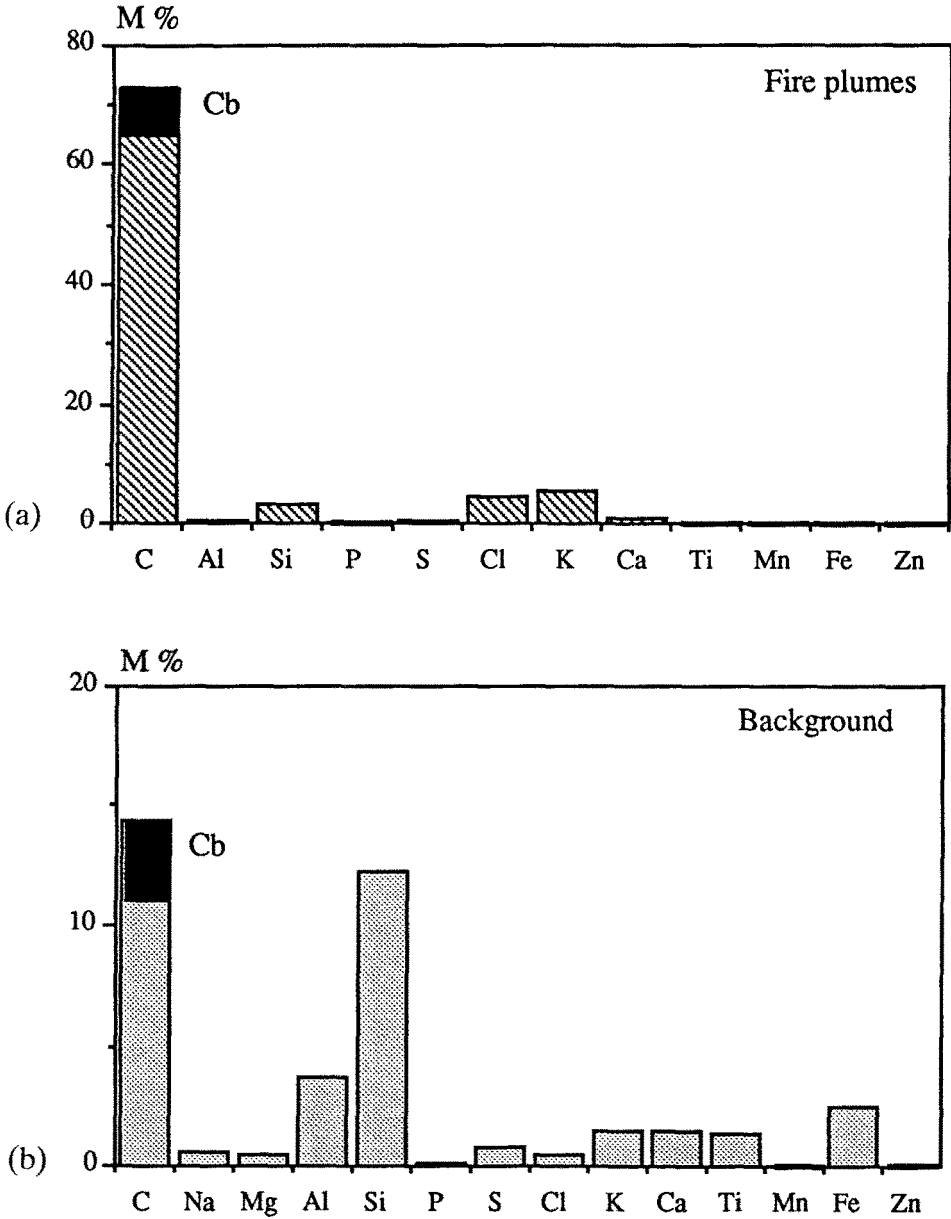


Fig. 1. Elemental composition of particles in (a) the savanna biomass burning plumes and (b) the background atmosphere.

period of our experiment (Gaudichet *et al.*, 1995, this issue) and recalling the average size distribution of terrigenous aerosols ($d_{\text{mean}} > 2 \mu\text{m}$, Ackerman and Noon, 1981). The presence of additional dust accounting for a shift in the particle size distribution toward coarser particles could also explain the exceptionally low

mass content of small particles (44%) in the fire plume sample $n^{\circ}F_{44}$ (Table II). Indeed during the sampling, the heading fire was particularly violent and may have caused a reentrainment of dust particles previously deposited on leaves or even at soil surface.

The data obtained with the high volume impactors confirm the overwhelming production of fine carbonaceous particles ($\leq 1 \mu\text{m}$) in the savanna fire plumes. In this case, the fine carbonaceous fraction was $95 \pm 2\%$ in mass. A slight shift towards coarser particles is observed in the background aerosol samples where the fine carbonaceous particles represent $83 \pm 3\%$ of total carbon. This may be accounted by additional terrigenous dust inputs into the atmosphere; the accretion of fresh carbonaceous smoke may be also envisaged.

3.3. AGING OF THE AEROSOL

In the smoke plume, the aerosol particles may interact with water vapor and numerous other gaseous organic and mineral species emitted concurrently during the combustion of the vegetation. From these interactions the carbonaceous particles probably acquire a coating of condensation products leading rapidly to an internally mixed aerosol phase. Additional transformations by the accretion of particles and photochemical reactions also are likely to occur.

All the physical or chemical measurements carried out on aerosols from savanna burnings strikingly demonstrate an aging of the aerosol which appears to occur rapidly. An example of the occurrence of both physical and chemical evolution of the carbonaceous particles is presented in Figure 2 which shows a plot of the particulate carbon size distribution versus its black carbon content; data are obtained from different savanna burning aerosols: very young aerosols sampled in the fire plume, 2–5 minute old aerosols sampled 10–100 meters from the burn and hour old aerosols sampled in the regional background atmosphere. The coagulation of carbonaceous particles alters their size distribution, while the growth in their black carbon fraction (reported by the increase of the Cb/Ct concentration ratio) may be explained by a partial disappearance of the organic component probably by evaporation or oxidation processes.

Particle growth due to aging is confirmed by the size distribution retrieved from the photometric data, which shows an increase of the effective particle radius toward the accumulation mode, from 0.15 to 0.5 μm (Lioussé *et al.*, 1995, this issue). Gas-particle interactions in biomass burning smoke plumes are also evidenced by the evolution of the trace element content of the aerosols. From the relative amounts of sulphur and chlorine in the particles, it is observed that young aerosols have a predominant Cl content whereas in older aerosols sulphur becomes more important (Gaudichet *et al.*, 1995, this issue). The attachment of sulphur to the old particles may be due to the oxidative conversion of SO_2 to particulate sulphate.

These modifications of the particles are also reflected in the optical properties of the aerosols. Namely, it has been shown that the black carbon specific attenuation

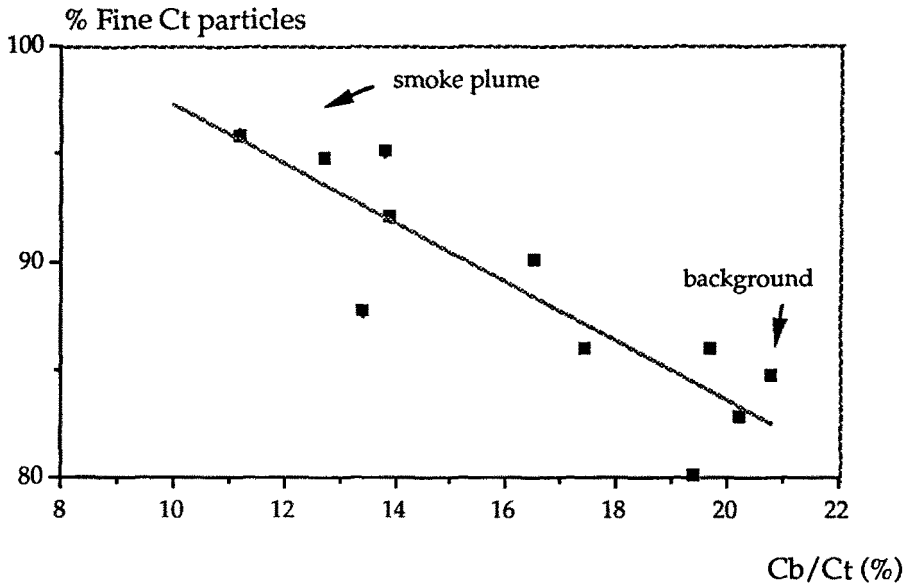


Fig. 2. Evolution of the carbonaceous biomass burning aerosols: parallel measurements of the Cb/Ct ratio and the abundance of submicron particles (cascade impactor data).

cross-section (σ) which is an indicator of its light absorbing capacity covers a large range of values (from 5 to 25 m^2/g) depending on the particle coating and size and thus the age of the aerosol (Liousse *et al.*, 1993, 1995).

3.4. CHARACTERIZATION OF THE BACKGROUND ATMOSPHERE

The high average total and black carbon concentrations ($\text{Ct} = 14.3 \pm 3.7 \mu\text{g C}/\text{m}^3$ and $\text{Cb} = 3.4 \pm 1.1 \mu\text{g C}/\text{m}^3$) compare well with those obtained in temperate suburban areas (Brémond *et al.*, 1989). Although Harmattan winds cause important dust inputs, the terrigenous carbon contribution is expected to be small. Biomass burning appears to be the major contributor to the atmospheric carbonaceous particle burden (Cachier *et al.*, 1985). During a small experiment conducted 2 years ago at the same site and during the same period of the year (January 1989), atmospheric concentrations were very similar ($\text{Ct} = 13.6 \mu\text{g C}/\text{m}^3$), confirming the yearly repetitive pollution pattern experienced in this region (Cachier *et al.*, 1991). IncurSIONS of vicinal fires would have been visibly recorded in the data (cf. Figure 3a). Accordingly, the elevated carbon concentrations found at Lamto in the background aerosols most likely reflect a regional atmospheric pollution by large-scale fires ranging in Western Africa in this season. This is supported both by reports from people living in the region and by satellite data (Brustet *et al.*, 1991).

A noticeable day and night pattern is observed in individual samples with an average increase of concentrations by about 50% during the day (Figure 3a).

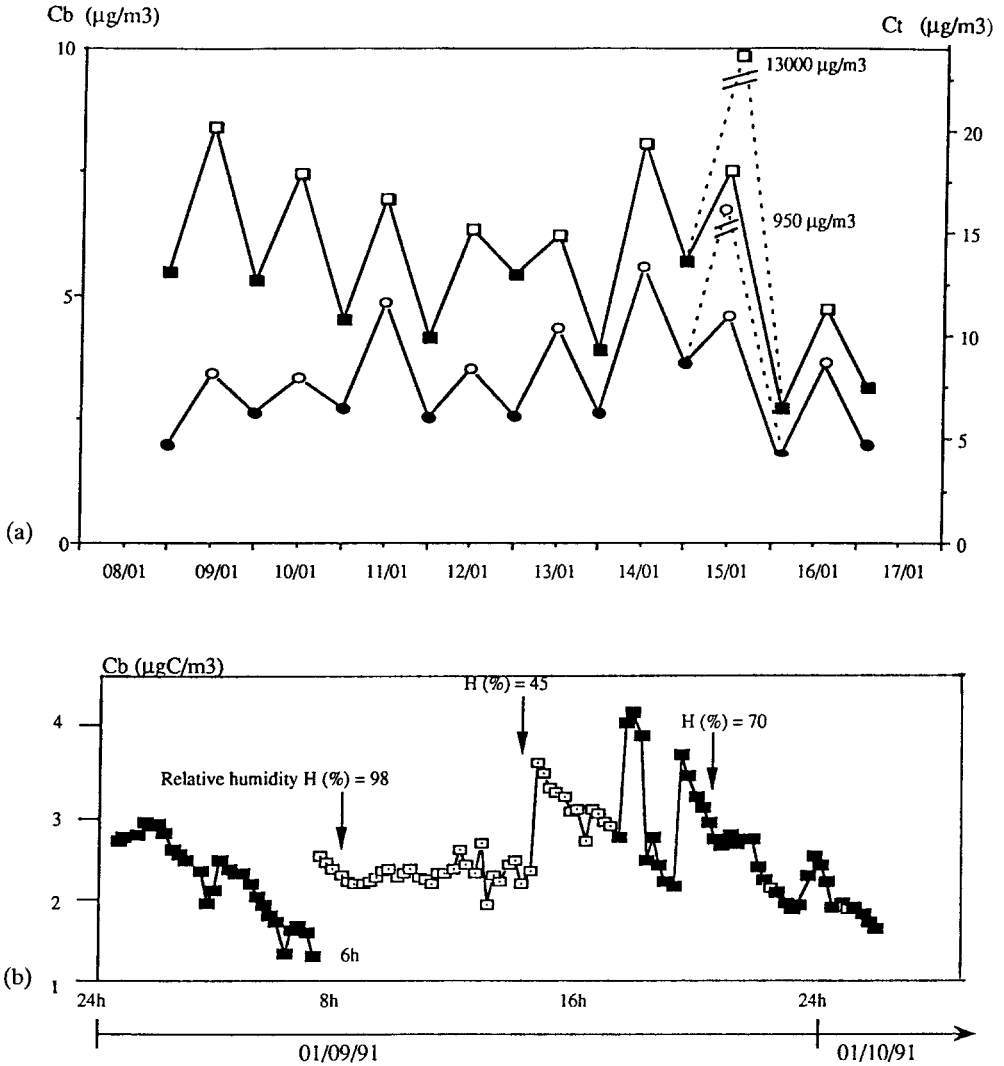


Fig. 3. Day-night pattern of the carbonaceous aerosol concentrations in Lamto background atmosphere (January 1991); (a) 12 h samples (total and black carbon). The dashed lines indicates excursions from vicinal fires; (b) real-time black carbon data.

Nocturnal inversions would have favored the opposite tendency. Consequently the concentration increase during the day is likely to be caused by enhanced biomass activity. Similar conclusions have been obtained previously from Lamto aerosol data derived from biomass burning ionic tracers, K^+ and $\text{C}_2\text{O}_4^{2-}$ (Cachier *et al.*, 1991a).

Black carbon real-time data obtained continuously throughout the experiment has allowed the reconstruction of the regional burning activities which are primarily related to the population activity and the atmospheric humidity (Liousse and

Cachier, 1992). A typical 24 hr record of black carbon concentrations is given in Figure 3b: domestic fires and new savanna burns ignited in the early morning contribute to an increase of black carbon concentrations but the high atmospheric humidity (more than 90%) does not allow the fires to extend or persist; after 12 h however, atmospheric water content is generally lower and black carbon concentration increases sharply with extensive fires during the next 6 hours; finally at night, the fires progressively reduce their intensity or vanish. This striking day-night pattern is likely to be found in other savanna regions of Africa and is somewhat in contradiction with the recent assessments of Cahoon *et al.* (1992) based on night IR satellite images.

3.5. AEROSOLS TRACERS FOR BIOMASS BURNING

From our studies, it appears possible to propose chemical indicators for the aerosols associated with biomass burning. This would allow the differentiation from industrial combustion particles.

The enrichment of potassium (K) in the fire plume particles is maintained in the background aerosols as demonstrated by their high potassium to carbon ratio (Table III). As the organic carbon component of the aerosols may further undergo chemical reactions or have additional natural inputs, the characterization of the particles arising from biomass burning should rely on the black carbon component. In calculations of the potassium to black carbon ratios, the terrigenous fraction in the fire plume and in the background particles were taken into account. The relative abundance of potassium and black carbon appears to be of the same order in the fire plume aerosols (ratio $K/Cb \sim 0.64$). But the slight decrease observed in the K/Cb ratio of the background aerosols ($K/Cb \sim 0.21$) cannot be explained easily. However, recalling our previous studies at the same site where a very similar Ks/Cb ratio value of 0.26 was obtained with soluble potassium measurements (Cachier *et al.*, 1991a), it may be considered that the K/Cb value of 0.21 is representative of aged savanna biomass burning aerosols in this region. Such a value stresses the importance of the potassium component in particles from biomass burning. Potassium is far more abundant in this case than in industrial aerosols (Cooper *et al.*, 1981). Since black carbon rapidly acquires a hydrophilic behavior, even in source regions (Ducret and Cachier, 1992)), the two Cb and K components are expected to follow similar atmospheric pathways and to be found with the same relative abundance at remote sites impacted by biomass burning effluents. This result is consistent with aerosol data obtained by Andreae *et al.* (1988) in the marine tropical atmosphere, giving a K_{excess}/Cb ratio of the order of 0.11.

Microscopic investigations have shown that K is also an absolute tracer of individual soot particles; indeed, the carbonaceous particle X-ray records systematically display the potassium signal, a feature which is never encountered in fossil-fuel derived micro-soots (Cachier *et al.*, 1991; Gaudichet *et al.*, 1995, this issue).

TABLE III. Potassium abundance in the savanna biomass burning aerosols (K/C in grass is 0.007). K concentration values are corrected for the soil-dust re-entrainment as follows: in Saharan dust the average K/Al ratio is 7.1 (Bücher and Lucas, 1975). The K dust component is calculated using the average Al concentration values in the fire plume ($14 \mu\text{g}/\text{m}^3$) and in the background atmosphere ($3.9 \mu\text{g}/\text{m}^3$); see Gaudichet *et al.*, 1995). In these two situations, the K biomass burning component represents 99% and 43% of the total K content, respectively

	Fire plume	Background atmosphere	References
Savanna smoke			
K/Ct	0.06 ± 0.03	0.047 ± 0.013	This work
K/Cb	0.64 ± 0.28	0.21 ± 0.06	
Wood smoke			
K/Ct	0.007 ± 0.002		Currie (1992)
Remote tropical			
K/Cb		0.095	Andreae (1983)
Amazonia haze layer			
K/Cb		0.11	Andreae <i>et al.</i> (1988)

Finally, some polycyclic aromatic hydrocarbons (PAH) may be other tracers for aerosols generated by savanna biomass burning. In either fire plume or background particles, the concentration distribution pattern of the various individual PAH molecules is found to be strikingly constant and very different from that observed in urban aerosols. This work is detailed in this issue by Masclet *et al.* The noticeable importance of pyrene in both sets of gaseous and particulate PAH could lead to the proposal of this component as a tracer of savanna burning smokes. Retene, another PAH was found abundant in forest fire plumes (Ramdahl, 1983). So, to what extent pyrene may be used to trace any other type of vegetation burning deserves further investigations.

3.6. PRESCRIBED FIRE PLUME AEROSOLS

There is considerable spatial and temporal intra-fire heterogeneity during the spreading of a fire. For the small plots, the fire-setting scheme was adopted to establish both backing and heading conditions. At the onsets of the fire the vegetation loses its water content rapidly, as seen by the white palls behind the fire front. The combustion itself is often, but not always, accompanied by the formation of light brownish to black plumes, illustrating the importance and the variability of the particulate emissions. An interesting observation is that during the flaming processes, especially when the fire is spreading rapidly, savanna burns may to some extent be oxygen limited or occur at low temperature and consequently may be prolific sources of particles. This phenomenon was also observed for other types of burn (Radke *et al.*, 1988; Stith *et al.*, 1981; Ward *et al.*, 1991). For forest fires Cofer *et al.* (1989) hypothesized that, during turbulent intense flaming, both oxy-

TABLE IV. Relative abundance of black carbon in biomass burning plume aerosols related as the Cb/Ct ratio (%)

Vegetation burn type	Cb/Ct (%)	References
<i>Field measurements</i>		
Boreal forest flaming	7	Mazurek <i>et al.</i> (1991)
smoldering	2	Mazurek <i>et al.</i> (1991)
Cerrado mixture	5.5	Ward <i>et al.</i> (1991)
Tropical forest flaming	15	Susott <i>et al.</i> (1991)
Amazonia haze layer mixed	11	Andreae <i>et al.</i> (1991)
Savanna fires flaming	12	This work
smoldering	5	This work
<i>Combustion chamber</i>		
Savanna grass mixed	9	This work
Wood debris flaming	31	Patterson <i>et al.</i> (1984)
smoldering	3	Patterson <i>et al.</i> (1984)

gen and thermal quenching could occur due to the rapid formation of pockets of oxygen-depleted air or the entrainment of cold air seeking to fill the void left by the strong fire updrafts.

Aerosols emitted by biomass burning are rich in organic components but contain little black carbon. The mean Cb/Ct ratio of the prescribed savanna fire carbonaceous aerosols is $11.1 \pm 3.8\%$. The variability of this ratio may be an indicator of the varying combustion conditions (Cachier *et al.*, 1989b). Pyrolysis conditions which are likely to produce primary organic aerosols, may be created either during smoldering phases with low temperature or during intense heading fires with oxygen or thermal quenching. Individual sample results are displayed in Figure 4 to show that a clear cut exists between the flaming ($12.1 \pm 2.8\%$) and the smoldering ($4.9 \pm 2.0\%$) samples. This is similar to other vegetation fires (Table IV). On the average, samples from heading phases cannot be distinguished from the other samples from flaming fires on the basis of their Cb/Ct ratio alone. But samples collected under backing fire conditions during well established burning regimes from larger fires (Fire BiF2), seem to produce aerosols significantly enriched in black carbon ($Cb/Ct = 15.5 \pm 3.3\%$). For the combustion chamber experiment, the mean Cb/Ct ratio of $9.0 \pm 2.0\%$ was observed, indicating that the burning conditions were intermediate between flaming and smoldering.

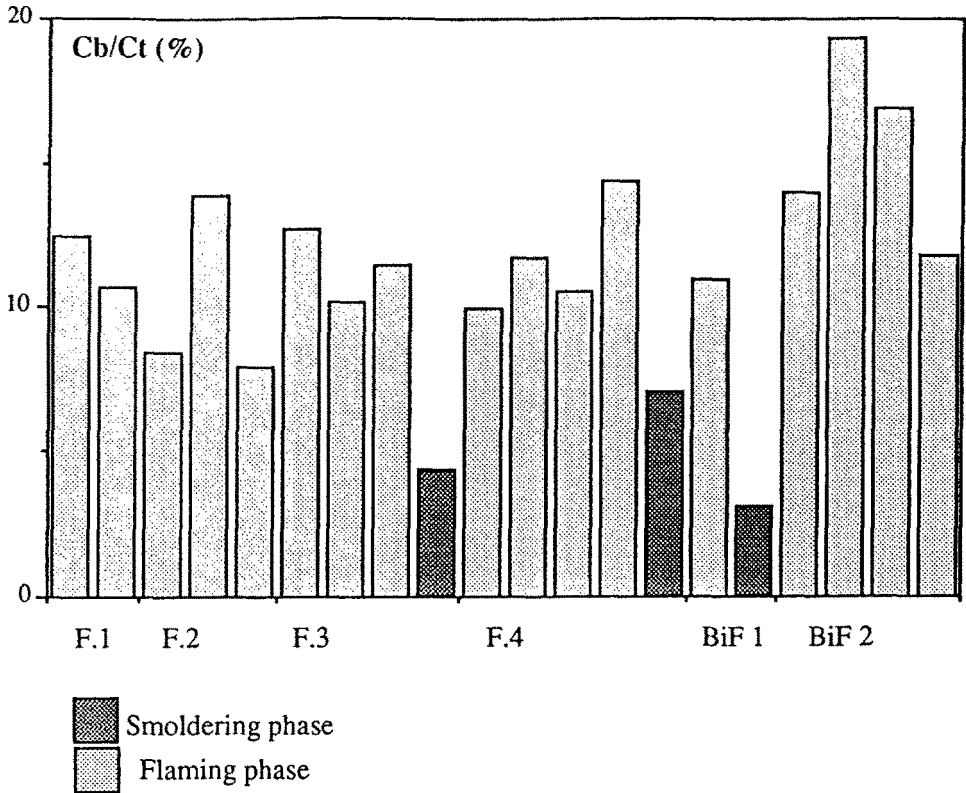


Fig. 4. Black carbon to total carbon ratio (Cb/Ct) as an indicator of the vegetation combustion process conditions: inter-sample variability.

Quantitative assessments of particulate emissions during the different flaming phases may be obtained from the ratio $\Delta\text{TPM}/\Delta\text{CO}_2$ calculated with the background corrected TPM and CO_2 concentrations recorded in real-time. For a given burn and a given fire regime, the particulate emission rates parallel those for CO_2 as shown by the linear correlation obtained between concentrations (Figure 5a). Intra and inter-fire variability is demonstrated in Figure 5b and c, using heading and backing data for the 2 different types of grassy plots. Our results are clearly different for emissions during the flaming phase from different grasses (ludetia versus hyparrhenia). These differences are however, within a factor of 2, and likely to be obscured by the burning regime variability which may be responsible of a 10 fold enhancement of the particle emissions from backing to heading conditions.

The production of particulates during the smoldering phases is more than one order of magnitude higher than during flaming phases, which points out the importance of low-temperature burning stages to the production of particles. This is evidenced by comparison of the mean $\Delta\text{TPM}/\Delta\text{CO}_2$ ratio values of $3 \times 10^{-1} \text{ mg/m}^3/\text{ppmv}^{-1}$

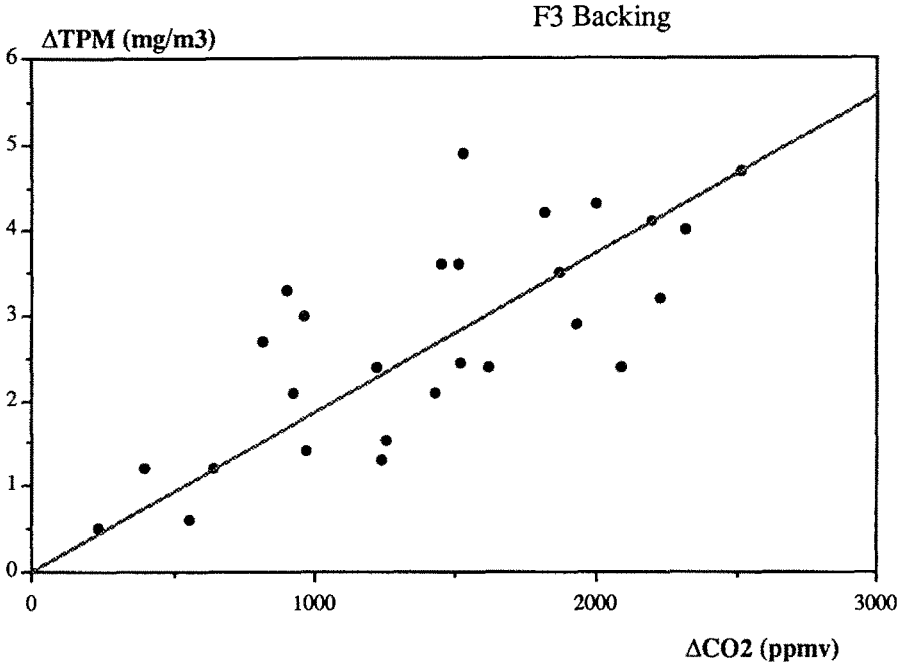


Fig. 5a.

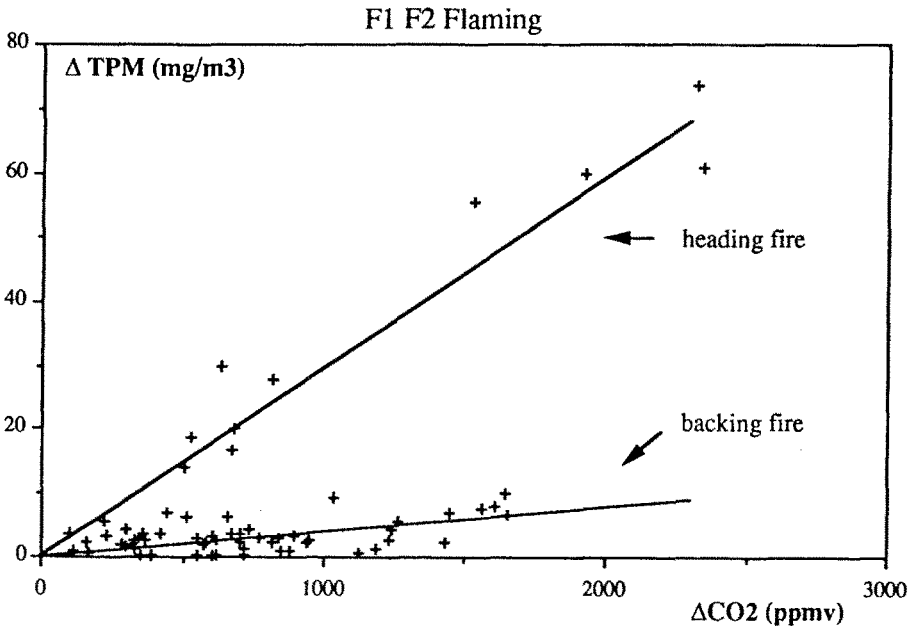


Fig. 5b.

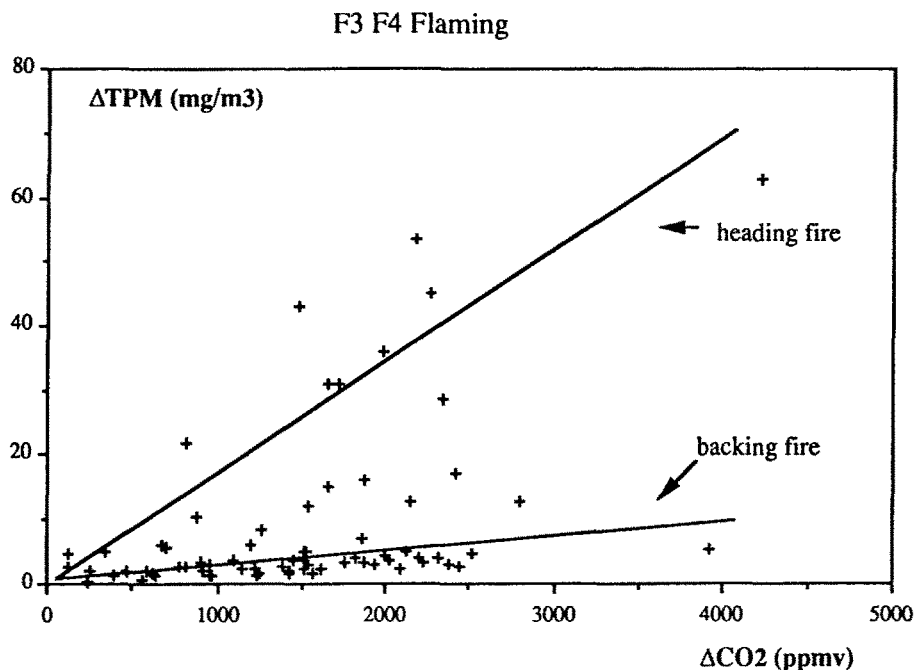


Fig. 5c.

Fig. 5a-c. Particle emission ratio as the ratio $\Delta\text{TPM}/\Delta\text{CO}_2$; (a) F3 backing fire ($\text{ER}_b = 1.9 \pm 1.0 \times 10^{-3} \text{ mg/m}^{-3}/\text{ppmv}^{-1}$); (b) F1-F2 backing and heading fires ($\text{ER}_b = 4.0 \times 10^{-3} \text{ mg/m}^{-3}/\text{ppmv}$), $\text{ER}_h = 2.0 \times 10^{-2} \text{ mg/m}^{-3}/\text{ppmv}^{-1}$); (c) F3-F4 backing and heading fires ($\text{ER}_b = 3.0 \times 10^{-3} \text{ mg/m}^{-3}/\text{ppmv}$; $\text{ER}_h = 1.7 \times 10^{-2} \text{ mg/m}^{-3}/\text{ppmv}^{-1}$).

obtained under smoldering conditions and $2 \times 10^{-2} \text{ mg/m}^3/\text{ppmv}^{-1}$ under heading conditions.

We present now the *particle emission factors* for the savanna biomass burning aerosols. Emission factors (EF_x) derived here, refer to the ratio of the mass of the emitted species X to the mass of dry fuel consumed. They are expressed in units of gram of X released per kilogram of dry vegetation ($\text{g X/kg}_{\text{dry plant}}$). Previous efforts to determine this quantity have concentrated mainly on forest fires (Table V) the validity of which for 'average biomass burns' could be questionable.

Emission factors (EF_x) may also refer to the amount of carbon in the vegetation fuel expressed as $\text{g X/kg C}_{\text{dry plant}}$. Based on the abundance of carbon in the dry plant, both types of emission factors are related to each other by a ratio of 0.42. The second type of emission factor will be used later in this paper in order to discuss easily the carbon budget during the combustion processes.

EF values have been computed using the equation

$$\text{EF} = \Delta\text{X}(\mu\text{gX/m}^3) \times k / \Delta\text{CO}_2(\mu\text{gC/m}^3), \quad (1)$$

TABLE V. Biomass burning particle emission factor (EF): literature data. EF are expressed in gram per kilogram dry matter

Fire type	Fire phase	EF (TPM)	EF (Cb)	EF (Ct)	References
Wild fires	Mean	20–50			Seiler and Crutzen (1980)
Conifer slash	Airborne flaming	2–20			Stith <i>et al.</i> (1981)
Wild fires	Mean	32	0.98 ± 0.6		Turco (1983)
Wild fires	Mean	33 ± 27	3.2 ± 0.6		Crutzen <i>et al.</i> (1984)
Test fires (Pine logging and slash)	(5) Flaming	16.9 ± 6.3	0.99 ± 0.12		Patterson <i>et al.</i> (1986)
	(8) Smoldering	14.7 ± 3.3	0.60 ± 0.13		Patterson <i>et al.</i> (1986)
Laboratory	(14) Flaming	14.7 ± 17.8	2.40 ± 2.55		Patterson <i>et al.</i> (1984)
	Smoldering	92.5 ± 20.3	0.71 ± 0.34		Patterson <i>et al.</i> (1984)
Test fires	(41) Mean	16.8 ± 10.1			Radke <i>et al.</i> (1988)
Wild fires	(3) Mean	20.2 ± 12.7			Radke <i>et al.</i> (1988)
Wheat stubble	(1)	44			Radke <i>et al.</i> (1988)
Haze layer (Amazonia)	Airborne	4.5 ^a	0.8 ^a	2.9 ^a	Andreae <i>et al.</i> (1988)
Agricultural wild	Mean	11 ± 6 ^a	2 ± 1 ^a	7.4 ± 3.7 ^a	Crutzen and Andreae (1990)
Fir logging slash and timber	Airborne flaming	2–4	0.8		Einfeld <i>et al.</i> (1991)
	Airborne smoldering	6–13	0.5		Einfeld <i>et al.</i> (1991)
Rice straw (wind tunnel)	Flaming	3.3			Jenkins <i>et al.</i> (1991)
Prescribed fires (birch, pine)	Flaming	14			Susott <i>et al.</i> (1991)
	Intermediate	100			Susott <i>et al.</i> (1991)
	Instrumented tower	Smoldering	1648		
Cerrado (< 2.5 μm)	Airborne	4.4			Ward <i>et al.</i> (1991)
Forest (> 2.5 μm)	Airborne	10.5 ± 5			Ward <i>et al.</i> (1991)
Prescribed fires	Airborne flaming	1.5 ^a			Woods <i>et al.</i> (1991)
(Sub-tropical grassy wetland)	Airborne smoldering	7 ^a			Woods <i>et al.</i> (1991)
Savanna grass Combustion chamber	(7) mixed	15 ± 4	0.92 ± 0.26	9.9 ± 1.4	This work
Prescribes fires	(15) flaming	5.0 ± 2.0	0.39 ± 0.18	3.2 ± 1.5	This work
	(3) smoldering	30 ± 11	1.00 ± 0.40	24 ± 7	This work

^a Adapted values from emission ratios (ER = $\Delta X / \Delta CO_2$) with a conversion factor of 0.37

TABLE VI. Carbon budget in savanna burning emissions: determination of the relative amount of C emitted as CO₂ (*k*); (a) Carbon budget in combustion chamber experiments; (b) Carbon budget for savanna burning emissions; (c) Carbon in the particulate phase of savanna burning emissions

(a)	C in ashes		C in identified gaseous species (mainly CO ₂ , CO, CH ₄ , NMHC)		C in unidentified species (including aerosols)	
Data from Lobert <i>et al.</i> (1991):	5%	+	90%	+	5%	
Corrected data (with ash carbon results)	1%	+	93.8%	+	5.2%	

The 'unidentified' fraction (ΔX) of the carbonaceous emissions is mostly emitted during reducing conditions and is likely to follow the CO emissions as seen for other minor species, CH₄ (Bonsang *et al.*, this issue) or OCS (N'Guyen *et al.*, this issue), as: $\Delta X = x \cdot \Delta CO$, with *x* value found as 0.91 from the previous combustion chamber data.

(b)	Ashes%	$\Delta CO\%$	$\Delta X\%$	$\Delta CH_4\%$	$\Delta NMHC\%$	$\Sigma \Delta\%$	<i>k</i>
Flaming	1.2	7.06	6.42	0.42	0.92	16.0	0.86
Smoldering	1.7	19.1	17.38	1.07	1.61	40.9	0.71

Carbonaceous species concentrations ($\Delta\%$) are given in (C/C in CO₂)%; $\Delta CO_2\%$ is $100\% - \Sigma \Delta\%$; *k* may be calculated as $\Delta CO_2 / (\Sigma \Delta + \Delta CO_2)$ or: $1 / (\Sigma \Delta / CO_2 + 1)$.

(c)	$\Delta X\%$	$\Delta Ct\%$	$\Delta Ct / \Delta X$
Flaming	6.42	0.72	11%
Smoldering	17.48	7.48	43%

where Δ are the above background concentrations (ΔTPM obtained with the weighted filters, ΔCt or ΔCb obtained by the filter carbon analysis and ΔCO_2 by real-time record), and *k* is a numerical factor indicating the proportion of fuel carbon released as CO₂.

The basic idea for our approach is the hypothesis that it is possible to obtain a mean characteristic *k* value for flaming conditions (0.86) and for smoldering conditions (0.71). The *k* calculations which are based on the carbon budget during a burn are derived from both field results and combustion chamber experiments reported by Lobert *et al.* (1991). They are shown in Table VI. Although this first attempt has to be considered as preliminary, the low *k* values obtained here reinforce the idea that reducing conditions prevail during most stages of biomass burning combustions.

In the field, for the experimental plots the mean EF values are: 11.4 ± 4.6 g TPM/g C_{dry plant}, 7.9 ± 3.4 g Ct/g C_{dry plant} and 0.98 ± 0.37 g Cb/g C_{dry plant} during the flaming phase and one order of magnitude higher: 69 ± 25 g TPM/g C_{dry plant}, 56 ± 16 g Ct/g C_{dry plant} and 2.3 ± 0.8 g Cb/g C_{dry plant} during the smoldering stage. A comparison with EF literature data shows that our values are lower than those

EF Ct (%)

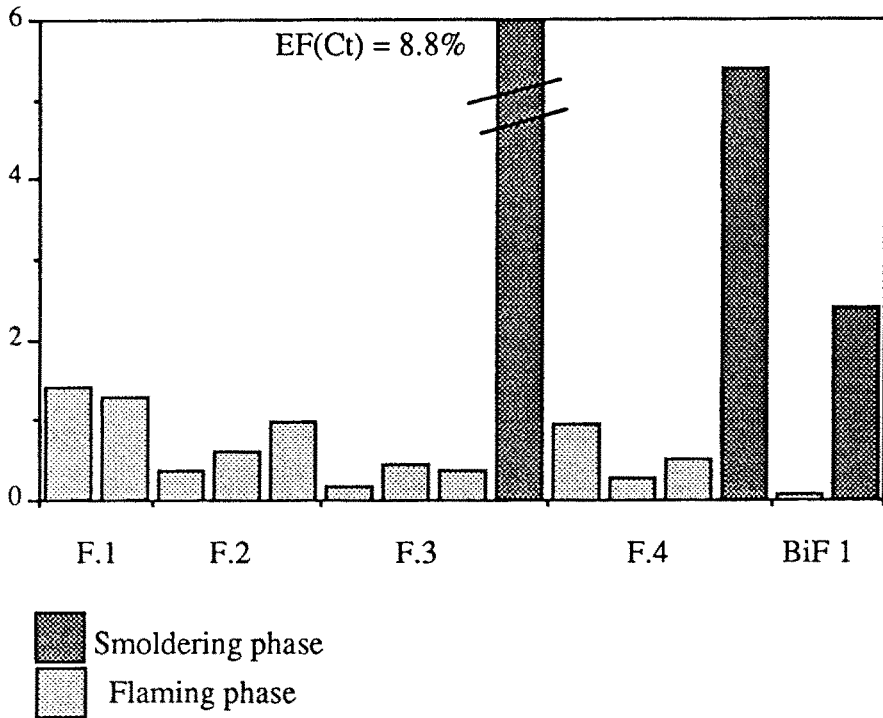


Fig. 6. Total carbon emission factor inter-sample variability (EF in g C/kg C dry plant).

generally assumed for a mean ‘vegetation combustion’ (Table V). This literature survey shows however a tendency of EF estimates to decrease according to recent studies. Moreover, the lowest values are found for ‘grass type’ vegetation fuel and an explanation may be the importance of smoldering stages in wild forest or logging fires during which up to 50% of the vegetation fuel is consumed (Susott *et al.*, 1991). This situation is never encountered with grass as fuel.

Individual sample carbon EF results are compared in Figure 6. The EF variability is primarily due to the differences between flaming and smoldering processes. In contrast to the TPM real time data, the integrated sample results do not exhibit clear intra-fire variability for backing or heading fires. However, there is a significant inter-fire variability which appears to be related to the fire efficiency which in turn is linked to the type of grass burnt: the emission factor for both TPM and carbonaceous particles increase with the fire efficiency (Table VII). It must be underlined here that fire efficiency is defined here as the mass ratio of burnt biomass to biomass exposed to the fire. Our results thus suggest that one important parameter for the intensity of particulate emissions is the amount of biomass burnt per unit area.

TABLE VII. Particle emission factors for the experimental savanna plots in relation with the fire efficiency: TPM (real-time optical data) and carbon (filter samples) results in g/kg C_{dryplant}

	Fire efficiency	EF _{TPM}	EF _{Ct}	EF _{Cb}
F ₁ + F ₂ plots (<i>ludetia</i>)	87%	20.6	8.9 ± 2.6	1.3 ± 0.2
F ₃ + F ₄ plots (<i>hypparhenia</i>)	75%	17.1	5.6 ± 1.9	0.66 ± 0.22

With the help of the carbon budget presented in Table VI, it is now possible to calculate from the aerosol EF_c results, the contribution of particulate carbon to the unidentified fraction ΔX of the budget. The surprisingly low value obtained for flaming conditions ($\sim 11\%$) suggests either that a significant fraction of gaseous carbonaceous species are not yet identified, or that the combustion chamber experiments are not representative of mean flaming conditions in the field. Both of these alternative conclusions deserve further investigations.

4. Conclusions

This study focuses on particulate emissions from savanna burning and allows to characterize the aerosol fluxes from this type of combustion.

Emission factors EF are found lower than in previous assessments which were mostly based on studies of forest fire studies. Global flux estimates should be thus significantly lower than those presented previously (Levine, 1990; Crutzen and Andreae, 1990) and used in models (Penner *et al.*, 1992). But, due to the global importance of the vegetation mass burnt each year in savanna regions, the amount of particles emitted into the atmosphere remains important: in the African savanna alone (Delmas *et al.*, 1991), 2.5 Pg of vegetation biomass are burnt each year (1/4 of the biomass burnt world-wide) which could lead to an annual flux of 12 ± 5 Tg of TPM, 8 ± 4 Tg of carbon including 1 ± 0.5 Tg of black carbon. These carbonaceous emissions are of the same order of magnitude as those attributed to fossil fuel combustions in industrial countries (22 Tg and 4 Tg for total and black carbon, respectively).

The main uncertainty in the estimates is the amount of burnt vegetation. However, further investigations in the field regarding the relative importance of the different stages of combustions are still necessary: if most of the biomass is burnt during flaming fires and only a little amount during smoldering fires, the heading flaming fires which may be high emitters of particles definitely deserve further studies.

Aerosols from savanna biomass burning are primarily in the submicrometer-sized range and are composed of organic compounds. The amount of black carbon

(Cb/Ct \sim 11%) is not high enough to envisage a significant absorption radiative effect of the particles. However, an important potential regional climatic impact of these aerosols may rely mainly on their properties as cloud condensation nuclei (CCN), as the particles are likely to be rapidly coated with soluble substances present in the fire plume and thus to obtain a hydrophilic character.

From the study of savanna biomass burning particles presented in this work and joined papers in this issue, it appears that the export outside source regions of combustion derived aerosols is possible as free atmospheric particles or in cloud droplets. This export could be traced by the characteristic abundance of some species such as pyrene, a condensed polycyclic aromatic hydrocarbon, or potassium (K). The export of carbonaceous material is accompanied by that of several mineral species attached to the particles. An important consequence of the long-range atmospheric transport of such particles could be a translocation of nutrients from the savanna regions to more remote places, the results of which is presently unknown.

In West Africa, the background atmosphere is shown to be highly polluted by components from savanna fires raging on a continental scale. Particulate carbon concentrations are as high as in suburban regions (Ct = 14 $\mu\text{g C/m}^3$). A clear day-night pattern shows that the fires occur mainly during the day, a result which points to the importance of photochemical reactions in the fresh plumes.

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