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Seasonal Variation and Ecosystem Dependence of Emission Factors for Selected Trace Gases and PM2.5 for Southern African Savanna Fires

S. Korontzi
University of Maryland - College Park

Darold E. Ward
USDA Forest Service - Rocky Mountain Research Station

Ronald A. Susott
USDA Forest Service - Rocky Mountain Research Station

Robert J. Yokelson
University of Montana - Missoula, bob.yokelson@umontana.edu

C. O. Justice
University of Maryland - College Park

See next page for additional authors

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Authors
S. Korontzi, Darold E. Ward, Ronald A. Susott, Robert J. Yokelson, C. O. Justice, Peter V. Hobbs, E.A. H. Smithwick, and Wei Min Hao

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Seasonal variation and ecosystem dependence of emission factors for selected trace gases and $PM_{2.5}$ for southern African savanna fires

S. Korontzi,[1] D. E. Ward,[2,3] R. A. Susott,[2] R. J. Yokelson,[4] C. O. Justice,[1] P. V. Hobbs,[5] E. A. H. Smithwick,[6] and W. M. Hao[2]

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[1] In this paper we present the first early dry season (early June-early August) emission factor measurements for carbon dioxide (CO_2), carbon monoxide (CO), methane (CH_4), nonmethane hydrocarbons (NMHC), and particulates with a diameter less than 2.5 μm ($PM_{2.5}$) for southern African grassland and woodland fires. Seasonal emission factors for grassland fires correlate linearly with the proportion of green grass, used as a surrogate for the fuel moisture content, and are higher for products of incomplete combustion in the early part of the dry season compared with later in the dry season. Models of emission factors for NMHC and $PM_{2.5}$ versus modified combustion efficiency (MCE) are statistically different in grassland compared with woodland ecosystems. We compare predictions based on the integration of emissions factors from this study, from the Southern African Fire-Atmosphere Research Initiative 1992 (SAFARI-92), and from SAFARI-2000 with those based on the smaller set of ecosystem-specific emission factors to estimate the effects of using regional-average rather than ecosystem-specific emission factors. We also test the validity of using the SAFARI-92 models for emission factors versus MCE to predict the early dry season emission factors measured in this study. The comparison indicates that the largest discrepancies occur at the low end (0.907) and high end (0.972) of MCE values measured in this study. Finally, we combine our models of MCE versus proportion of green grass for grassland fires with emission factors versus MCE for selected oxygenated volatile organic compounds measured in the SAFARI-2000 campaign to derive the first seasonal emission factors for these compounds. The results of this study demonstrate that seasonal variations in savanna fire emissions are important and should be considered in modeling emissions at regional to continental scales. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 1610 Global Change: Atmosphere (0315, 0325); *KEYWORDS:* seasonal fire emissions, savannas, southern Africa, emission factors

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1. Introduction

[2] Savanna fires are an important ecosystem process in southern Africa, with significant implications for regional and global atmospheric chemistry and biogeochemical cycles [*Scholes et al.*, 1996; *Frost*, 1996]. The majority of fires in southern Africa occur typically during the dry season, from May to October. There are significant interannual variations in the magnitude and location of biomass burning emissions at the regional scale, in response to the seasonal variability that occurs at a different rate from year to year [*Barbosa et al.*, 1999]. However, only a few studies have looked at the seasonality of fire emissions [*Hoffa et al.*, 1999; *Justice et al.*, 2002; *Korontzi et al.*, 2003]. Emission factors for pyrogenically produced atmospheric species are among the information required for emissions modeling. Thus far, regional fire emissions calculations in southern Africa have been mainly based on late dry season (August–October) ground-based and airborne measurements of emission factors [*Ward et al.*, 1996; *Hao et al.*, 1996; *Cofer et*

[1]Department of Geography, University of Maryland, College Park, Maryland, USA.
[2]USDA Forest Service, Rocky Mountain Research Station, Fire Sciences Laboratory, Missoula, Montana, USA.
[3]Now at Enviropyronics, White Salmon, Washington, USA.
[4]Department of Chemistry, University of Montana, Missoula, Montana, USA.
[5]Department of Atmospheric Sciences, University of Washington, Seattle, Washington, USA.
[6]Department of Zoology, University of Wisconsin, Madison, Wisconsin, USA.

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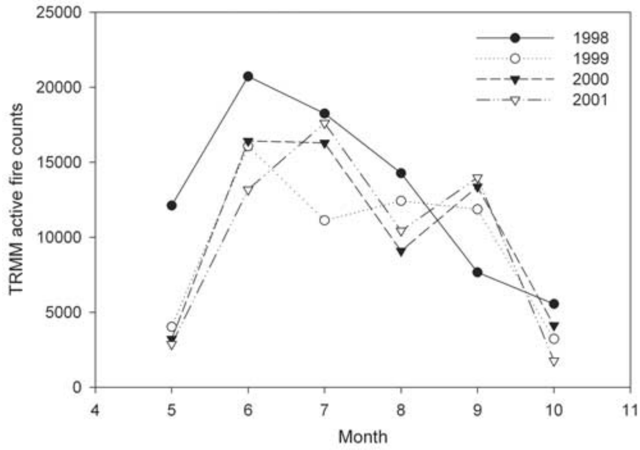

Figure 1. Seasonal and interannual TRMM active fire distribution in southern Africa in the main dry season (May–October).

al., 1996; *Scholes et al.*, 1996; *Yokelson et al.*, 2003; *Sinha et al.*, 2003] and/or average values for a particular ecosystem type [*Andreae and Merlet*, 2001].

[3] During the early dry season, from May to late July, fires transition from a condition where they will barely burn to one where they burn with higher intensity. There is a need to determine the effects of this transition on emissions. In the early part of the fire season the ground fuels typically have higher moisture content, which in addition to other important factors such as fuel loading variations, leaf fall, and weather conditions, may affect the type and the amount of the combustion products and play an important role in the overall budgets of pyrogenically produced trace gases and aerosols [*Hoffa et al.*, 1999; *Justice et al.*, 2002; *Korontzi et al.*, 2003]. Prescribed burning in the early part of the dry season is commonly advocated as a land management tool in tropical savannas [*Frost*, 1996; *Williams et al.*, 1998]. Wetter burns produce lower fire intensities and result in less vegetation consumed and damage to the soil. Pastoralists burn extensively in the early dry season to stimulate regrowth of palatable grasses for their cattle; fire is used for rapid nutrient release prior to the new growing season by farmers; and early burning is used in national parks as a preventive measure against late dry season fires which tend to have higher intensities and be presumably more destructive. Fire is also used to maintain the competitive balance between trees and grasses.

[4] Currently, the majority of the fires occurring in southern Africa are of anthropogenic origin. Fire regimes are likely to change with changing human population and land use practices, making early burning more widespread [*Russell-Smith et al.*, 1997; *Bucini and Lambin*, 2002]. Figure 1 illustrates 4 years of Tropical Rainfall Mapping Mission (TRMM) active fire distribution in southern Africa in the main dry season [*Giglio et al.*, 2000]. Despite the limitations of using active fire as a surrogate for burned area, these satellite data provide evidence for the seasonal variability of fires and the important contribution of early dry season burning [*Justice and Korontzi*, 2001].

[5] In this paper, explicit early dry season measurements of emission factors for selected carbonaceous (i.e., carbon containing) trace gases and aerosols in southern African savanna fires made from early June to early August 1996, are presented. In addition, the dependency of emission factors on ecosystems with distinct fuel types, grasslands or woodlands, is explored. More specifically, the following questions are posed: (1) What are the seasonal trends in emission factors and how do they relate to the fuel moisture condition?; (2) Is the relationship between modified combustion efficiency (an index of the completeness of emission oxidation) and emission factors for each of the atmospheric species analyzed here different for grasslands and woodlands, or can a single model be used to describe the data?; and (3) How well do results from the early dry season of 1996 compare with results from the late dry season Southern African Fire-Atmosphere Research Initiative 1992 (SAFARI-92) [*Lindesay et al.*, 1996] and SAFARI-2000 [*Swap et al.*, 2002] campaigns in southern Africa?

2. Methods

2.1. Site Description

[6] The field site and fires used for the 1996 study of early dry season fire emission measurements are reported in detail by *Hoffa et al.* [1999]. The field site was located about 7.5 km southeast of Kaoma, Western Province, Zambia in the Kaoma Local Forest 310 (14°52′S, 24°49′E at approximately 1170 m). Thirteen 2-ha plots (100 m × 200 m) were burned between 5 June and 6 August 1996. Six plots were in a semideciduous, open canopy, semiarid woodland (miombo) and seven in a seasonally flooded grassland (dambo). The ecosystem sites were separated by approximately 500 m and a dirt road. Three distinct sampling clusters were equally spaced along the long axis of each 2-ha plot, as described by *Hoffa et al.* [1999].

[7] Miombo is used to describe the central, southern and eastern African woodlands, dominated by the genera *Brachystegia*, *Julbernardia* and/or *Isoberlinia* [*Frost*, 1996]. It covers more than 2.7 million km^2 of Africa and 80% of Zambia. Miombo woodlands receiving less than 1100 mm rain annually are considered semiarid [*Chidumayo*, 1987]. Fire spread in the miombo ecosystem is largely dependent on the amount of grass cover, coupled with meteorological parameters (i.e., wind speed, relative humidity and temperature). Grass production is high in areas of low woodland cover or where the land cover has been disturbed by, for example, gardening or charcoal making. Leaf litter and downed wood are likely the major components of the fuel in the undisturbed miombo. Fires in the humid miombo ecosystem tend to be more frequent and burn with higher fire intensities, presumably due to higher fuel loads [*Frost*, 1996]. Dambos are distinctive areas of African grassland produced by seasonal flooding; they occupy about 10% of Zambia [*Hoffa et al.*, 1999]. Dambos play an important role in traditional land use systems in Africa. They are mainly used for grazing, cultivation of food and cash crops, and as a water supply for domestic use and livestock [*Acres et al.*, 1985].

2.2. Measurement of Emissions

[8] SAFARI-2000 results showed that the composition of smoke from savanna fires changes rapidly as the smoke

ages [*Hobbs et al.*, 2003]. In the 1996 study we measured the initial emissions from grassland savanna and miombo woodland fires for carbon dioxide (CO_2), carbon monoxide (CO), methane (CH_4), nonmethane hydrocarbons (NMHC) and particulate matter with diameter less than 2.5 μm ($PM_{2.5}$). The sampling design at each plot and the emissions analyses are described by *Shea et al.* [1996], *Ward et al.* [1996], and *Hao et al.* [1996]. A Fire-Atmosphere Sampling System (FASS) tower was placed at the center of each cluster (three towers per plot) to collect smoke samples for emissions measurements. Each FASS system collected a background sample before the fire was ignited and two canisters from each burn approximately timed to sample separately the flaming and smoldering combustion. The plots were successively burned at approximately 1–2 week intervals throughout the study period. *Hoffa et al.* [1999] give descriptions of the vegetation fuel types, loads, environmental conditions and fire behavior at these plots. CO_2, CO, CH_4, and NMHC (C_2–C_3 aliphatic compounds and some aromatic compounds) were analyzed with gas chromatography (GC) as described by *Hao et al.* [1996]. The $PM_{2.5}$ concentration was determined from the increase in weight of Teflon filters exposed to the smoke divided by the volume of air sampled [*Ward et al.*, 1996].

[9] The quantification of different compounds emitted from fires is commonly expressed using the emission factor (EF). The EF is the mass of a specific gas or particulate matter emitted by the combustion per unit mass of dry fuel consumed (g kg^{-1}). To calculate the EF, the carbon content of the fuel is needed. To make our results comparable with those from previous studies we used a standard carbon fuel content of 50% [*Ward et al.*, 1996; *Yokelson et al.*, 2003; *Sinha et al.*, 2003]. The EFs for carbon-containing species are often linearly correlated to the modified combustion efficiency (MCE), which is the molar ratio of emitted CO_2 to the sum of CO and CO_2 [*Ward et al.*, 1996; *Sinha et al.*, 2003; *Yokelson et al.*, 2003]. The MCE is an indicator of the relative contribution of flaming and smoldering combustion in a fire. Laboratory fire experiments have shown that MCE ranges from 0.98 ± 0.01 for flaming combustion to near 0.80 ± 0.08 for smoldering combustion [*Yokelson et al.*, 1996]. A value of 1.00 suggests a complete oxidation of the carbon fuel (i.e., full conversion to CO_2).

[10] Table 1 provides the net concentrations of emitted CO_2, CO, CH_4, NMHC and $PM_{2.5}$ and the proportion of fuel consumed during the flaming and smoldering combustion in the fire at each plot. EFs were calculated from the net concentrations using the carbon mass balance technique described by *Ward et al.* [1996]. The fuel consumption ratios were determined using the FASS carbon flux technique and they were used in calculating the fire-weighted emissions factors. The measured background range for CO_2 was 340 ppm to 360 ppm. Many of the smoldering samples and two flaming samples in the grassland fires were close to natural background with very low net concentrations and within the error range of the canister analysis. While background might change with season, we believe there should not be much change across the plot on the same day. EFs for non-CO_2 compounds cannot be calculated without CO_2, in the carbon mass balance method used. Therefore all samples that had net CO_2 concentrations less than 20 ppm difference from background were rendered as highly uncertain and were not included in the calculations of the MCE or the EFs for all atmospheric species in the grassland fires. The low concentration non-CO_2 EFs were excluded since MCE is a linear function of the CO_2 and any uncertainty in MCE will propagate in the regressions of EFs versus MCE. The 26 July $PM_{2.5}$ collections were also below our limit for accurate emission factor data.

[11] In the case of the miombo woodland samples, due to the unavailability of reliable FASS fuel consumption data, EFs were weighted by assuming a 85/15 ratio for flaming and smoldering, respectively [*Ward et al.*, 1996; *Hoffa et al.*, 1999]. *Hoffa et al.* [1999] weighted the MCEs by assuming the 85/15 ratio in both dambo grasslands and miombo woodlands. Since we used a different weighting procedure for the grassland fires, the grassland MCEs presented here are slightly different than those reported by *Hoffa et al.* [1999] A single MCE and EF value was calculated for each FASS tower. The MCE and EF values from the FASS towers at each plot were then averaged to obtain a plot value used in the analysis (Table 2).

2.3. Statistical Analyses

[12] The EF data versus MCE were analyzed using simple linear regression. Models were developed for each set of woodland and grassland EF data using a linear least squares residual fitting technique. The separate regression lines were then compared to a single regression model, derived from the combined grassland and woodland data. The purpose of this analysis was to determine possible statistically significant differences (hereafter referred to as significant) in EFs between ecosystems in the unique, but limited amount of the 1996 data. However, in our interpretation of the results, we did not rely solely on the accept/reject logic of statistical hypothesis testing because, in some cases, small statistical differences are meaningless to prospective fire information users (e.g., in regional and global emissions modeling).

[13] To measure the overall variability around the regression lines, the pooled estimate of the variance about the two regression lines, $s^2_{\mathrm{EF.MCE}_p}$ was computed as

$$s^2_{\mathrm{EF.MCE}_p} = \frac{(n_g - 2)s^2_{\mathrm{EF.MCE}_g} + (n_w - 2)s^2_{\mathrm{EF.MCE}_w}}{(n_g + n_w - 4)},$$

where $s^2_{\mathrm{EF.MCE}}$ is the standard error of the estimate, and $n_g + n_w - 4 = \nu$ are the degrees of freedom [*Glantz*, 1997]. Subscripts *g* and *w* refer to the grassland and woodland data, respectively. The improvement in the fit obtained by fitting the data sets with separate regression lines, compared to a single regression line was computed using

$$s^2_{\mathrm{EF.MCE}_{\mathrm{imp}}} = \frac{\mathrm{SS}_{\mathrm{res}_c} - \mathrm{SS}_{\mathrm{res}_p}}{2},$$

where $\mathrm{SS}_{\mathrm{res}_c}$ is the sum of squared residuals around the common regression line and $\mathrm{SS}_{\mathrm{res}_p}$ is the sum of squared residuals about the separate regression lines.

[14] The relative improvement in the fit obtained by fitting the two data sets separately was quantified using the *F* test statistics. This value was then compared with the critical value of the *F* test statistic for $\nu_n = 2$ numerator

Table 1. Concentrations of Emitted CO_2, CO, CH_4, NMHC, and $PM_{2.5}$ and the Proportion of Total Fuel Consumed by the Grassland and Woodland Fires

Site[a]	CO_2, ppm	CO, ppm	CH_4, ppm	NMHC, ppm	$PM_{2.5}$, mg m^{-3}	FASS Fuel Ratio
G1AF	201.8	19.58	1.061	0.821	1.430	1.00
G1AS						
G1BF	15.8	1.89	0.103	0.120	0.200	0.86
G1BS	1.2	0.26	0.027	0.008	0.200	0.14
G2AF	376.8	30.93	1.608	1.311	2.180	0.94
G2AS	93.3	4.87	0.172	0.190	0.350	0.06
G2BF	198.1	22.33	1.216	1.041	1.620	1.00
G2BS	3.8	0.61		0.019	0.240	0.00
G3AF	246.5	10.55	0.393	0.350	0.770	0.99
G3AS	1.7	0.10			0.150	0.01
G3BF	233.9	12.15	0.503	0.488	0.645	0.98
G3BS	7.1	1.19	0.055	0.032	0.290	0.02
G4AF	423.7	18.94	0.761	0.569	1.040	0.99
G4AS	0.0	0.31	0.009	0.000	0.320	0.01
G4BF	840.7	24.44	0.913	0.787	1.290	1.00
G4BS	9.2	0.64	0.027	0.037	0.215	0.00
G4CF	336.1	14.07	0.519	0.423	0.770	1.00
G4CS	3.2	0.52	0.026	0.012	0.405	0.00
G5AF	413.6	13.39	0.435	0.401	0.940	0.99
G5AS	0.6	0.58	0.011	0.021	0.040	0.01
G5BF	501.4	12.73	0.382	0.381	1.185	0.99
G5BS	2.0	0.18			0.075	0.01
G6AF	10.7	1.32	0.029	0.040		0.80
G6AS	6.0	1.25	0.054	0.052		0.20
G6BF	94.1	4.70	0.150	0.140		0.96
G6BS	6.1	1.75	0.066	0.016		0.04
G6CF	49.0	2.44	0.082	0.056		1.00
G6CS	7.9	1.51	0.082	0.031		0.00
G7AF	1042.7	61.01	3.823	2.335	4.580	1.00
G7AS	19.6	2.42	0.149	0.142	0.950	0.00
G7BF	471.0	36.40	2.287	1.274	2.575	0.98
G7BS	5.8	0.79	0.045	0.026	0.580	0.02
G7CF	612.2	25.95	1.558	1.052	2.695	1.00
G7CS	3.6	0.72	0.056	0.047	0.955	0.00
W1AF	345.0	23.91			3.130	1.00
W1AS	46.6	2.55	0.118	0.090	0.080	0.00
W1BF	109.6	7.08	0.314	0.228	0.495	1.00
W1BS	13.5	0.80	0.036	0.018	0.215	0.00
W2AF	974.2	75.22	4.517	1.954	8.770	
W2AS	77.5	9.94	0.722	0.210	0.560	
W2BF	222.0	8.22	0.243	0.198		
W2BS	46.1	3.11	0.112	0.063		
W2CF	702.8	27.29	0.997	0.518	2.990	1.00
W2CS	26.9	3.57	0.145	0.058	0.195	0.00
W3AF	545.5	38.00	2.080	1.001	3.790	0.97
W3AS	24.5	2.54	0.175	0.077	0.340	0.03
W3BF	255.0	12.52	0.538	0.339	1.010	0.99
W3BS	156.0	1.67	0.084	0.024	0.145	0.01
W3CF	64.3	3.58	0.151	0.113	0.680	
W3CS	38.9	2.86	0.140	0.083	0.295	
W4AF	457.4	29.94	1.646	0.648	1.820	0.99
W4AS	59.6	8.59	0.564	0.185	0.320	0.01
W4BF	761.2	49.89	2.665	1.158	5.120	1.00
W4BS	92.9	8.49	0.538	0.227	0.885	0.00
W4CF	135.4	6.29	0.235	0.157	1.075	0.94
W4CS	48.7	3.87	0.191	0.133	0.480	0.06
W5AF	132.8	8.13	0.380	0.245	0.715	0.03
W5AS	289.2	17.26	0.948	0.365	1.965	0.97
W5BF	804.9	43.78	2.190	1.079	5.480	0.67
W5BS	265.3	22.99	1.405	0.516	1.730	0.33
W5CF	646.1	48.56	2.659	1.088	5.310	1.00
W5CS	58.0	7.30	0.456	0.162	0.450	0.00
W6AF	370.9	41.17	2.774	0.928	5.590	0.82
W6AI	89.3	12.65	0.895	0.261	1.010	0.11
W6AS	25.7	4.18	0.282	0.099	0.895	0.07
W6BF	982.4	91.85	5.810	2.131	18.410	0.86
W6BI	185.5	25.59	1.837	0.497	1.850	0.09
W6BS	65.2	8.29	0.571	0.170	0.88	0.05
W6CF	483.1	41.78	2.522	0.921	6.765	
W6CI	117.2	13.04	0.822	0.277	0.600	
W6CS	46.9	6.04	0.423	0.158	0.900	

Table 2. Early Dry Season Modified Combustion Efficiency and Weighted Average Emission Factors for CO_2, CO, CH_4, NMHC, and $PM_{2.5}$ for Grassland and Woodland Fires[a]

Site	Date	MCE	$EFCO_2$, g kg^{-1}	EFCO, g kg^{-1}	$EFCH_4$, g kg^{-1}	EFNMHC, g kg^{-1}	$EFPM_{2.5}$, g kg^{-1}
G1	5 June 1996	0.912	1637.4	101.12	3.132	4.734	6.461
G2	14 June 1996	0.913	1638.5	100.35	3.045	5.036	6.293
G3	26 June 1996	0.955	1735.3	52.27	1.181	2.142	2.842
G4	9 July 1996	0.963	1754.4	42.98	0.940	1.449	2.042
G5	18 July 1996	0.972	1772.3	32.56	0.584	1.074	2.288
G6	26 July 1996	0.953	1706.8	54.16	1.011	1.554	
G7	6 Aug. 1996	0.944	1707.8	64.31	2.282	2.747	4.514
W1	6 June 1996	0.940	1700.0	68.99	1.754	2.363	5.889
W2	18 June 1996	0.941	1704.4	68.03	1.971	1.861	4.997
W3	5 July 1996	0.952	1722.9	55.44	1.374	1.737	6.493
W4	16 July 1996	0.932	1685.8	78.19	2.529	2.014	5.310
W5	24 July 1996	0.937	1692.9	72.60	2.185	2.053	6.436
W6	29 July 1996	0.907	1614.6	105.79	3.921	2.786	15.145

[a]MCE, modified combustion efficiency; EF, emission factor.

degrees of freedom and $v_d = n_g + n_w - 4$ denominator degrees of freedom. The F test statistic is defined as

$$F = \frac{s^2_{\mathrm{EF.MCE_{imp}}}}{s^2_{\mathrm{EF.MCE}_p}}.$$

If the observed value of F exceeds the critical value of F_{crit}, it indicates that a significantly better fit to the data (measured by the residual variation about the regression line) was obtained by fitting the two data sets with separate regression lines than by fitting all of the data to a single line.

[15] Finally, we combined all of the EFs from this study with results from the SAFARI-92 and SAFARI-2000 late dry season field campaigns to derive a synthetic regression predictive model for regional EFs from MCEs. The conventional significance level of 95% ($P \leq 0.05$) was used for all hypotheses tested. Throughout the analyses, checks were performed to test for the assumptions of normality of the residuals and homogeneity of the variances. In some cases, one or both of the assumptions were violated, mostly when all the data were fitted with the common regression line. Other investigators encountered similar problems [e.g., *Ward et al.*, 1996; *Hao et al.*, 1996; *Yokelson et al.*, 2003]. Despite these statistical problems, the empirically derived regression models combined with our conceptual models provide a useful tool to estimate the natural variation of the data and for comparison with previous results.

3. Results and Discussion

3.1. Seasonal Trends

3.1.1. Modified Combustion Efficiency

[16] In the 1996 data there is a lower limit for MCE of 0.907 and 0.912 and an upper limit of 0.952 and 0.972 for the woodland and the grassland fires, respectively (Table 2). There is a more pronounced seasonal change in the grassland MCE than in the woodland MCE (Figure 2). In grasslands, it appears that MCE varies inversely to the moisture content of the grass fuel [*Ward et al.*, 1996; *Hoffa et al.*, 1999; *Saarnak*, 1999]. Generally, for this region, as the season progresses and the grasses achieve lower moisture content, the combustion process becomes more efficient and the MCE increases.

[17] *Hoffa et al.* [1999] found that the MCE of the 1996 grassland fires was correlated with the proportion of green grass (PGREEN) in the fuel, with higher moisture content than dead grass. The correlation between MCE and PGREEN is recalculated here since we used a different weighting procedure to derive the grassland MCEs (Figure 3) than *Hoffa et al.* [1999]:

$$\mathrm{MCE} = 1.010 - 0.217(\mathrm{PGREEN}), \quad R^2 = 0.73. \qquad (1)$$

[18] It should be pointed out, that despite the different weighting factors used for flaming and smoldering in the 1996 study for the grassland fires compared with *Hoffa et al.* [1999] the seasonal trends in MCE are similar for both methods of data analysis.

[19] *Ward et al.* [1996] found, that in woodlands, where grass was a larger fraction of the fuel, the MCE relates to the proportion of the grass in the fuel. In other woodlands, where the grass fuel component is minor, as was the case for the specific 1996 Zambian site (between 7% and 14%), it appears that other fuel types than grass, that increasingly contribute to burning as the dry season progresses, control the MCE. Litter fall occurs as the dry season progresses, so that the amount of leaf litter increases seasonally [*Hoffa et al.*, 1999]. The litter and woody fuels dry slower than the grasses and tend to burn by smoldering, which can lower MCE [*Bertschi et al.*, 2003]. Each fuel type makes a different contribution to the MCE, with litter and woody fuels having the opposite effect compared to the grasses. The combustion factors (the percentage of fuel consumed by the fire) for the burning of all fuel types and the fire intensity generally increase as the dry season progresses [*Hoffa et al.*, 1999]. Whereas though, the grasses tend to involve more flaming combustion which seems to increase the MCE, the litter and woody fuels tend to involve more

Note to Table 1

[a]Italics denote samples that were not included in the analysis on the basis of marginal net concentrations (<20 ppm CO_2). A, B, and C refer to the three sampling clusters centered around each FASS tower that were used to calculate the average at each plot. G, grassland; W, woodland; F, flaming combustion; S, smoldering combustion; I, intermediate combustion.

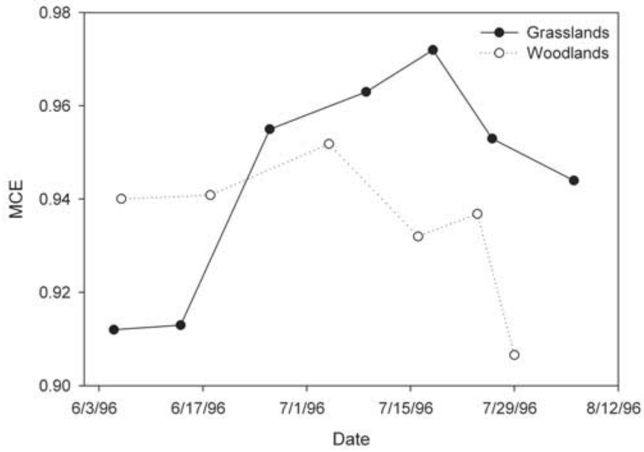

Figure 2. Seasonal progression of the modified combustion efficiency (MCE) for grassland and woodland fires.

smoldering combustion and may decrease the MCE. This might explain the lower MCE in the 29 July 1996 woodland burn. Given the vast area and diversity of African woodlands there could be seasonal trends in miombo woodlands, which are not apparent from the limited measurements made in this 1996 study.

3.1.2. Emission Factors

[20] A distinct seasonal trend was observed in the EFs for all measured species in smoke from the dambo grassland fires. The $EFCO_2$ increased as the season progressed due to the higher degree of oxidation from the combustion of the drier fuels, but the variability was small with a maximum difference of about 8.2% (Figure 4a). On the other hand, the EFs of the products of incomplete combustion varied substantially during the fire season (Figures 4b and 5a–5c). On average, they were highest in the first part of the early dry season relative to later in the early dry season by maximum factors of 3.1 for CO, 5.4 for CH_4, 4.7 for NMHC and 3.2 for $PM_{2.5}$.

[21] EFs are directly related to PGREEN in grasslands, supporting the hypothesis that as the fuels dry out a higher degree of oxidation is achieved, resulting in more CO_2 and less products of incomplete combustion compared with earlier in the dry season when the grasses have a higher moisture content. The regression models of EFs versus PGREEN in grasslands (Figures 6a–6b and 7a–7c) are

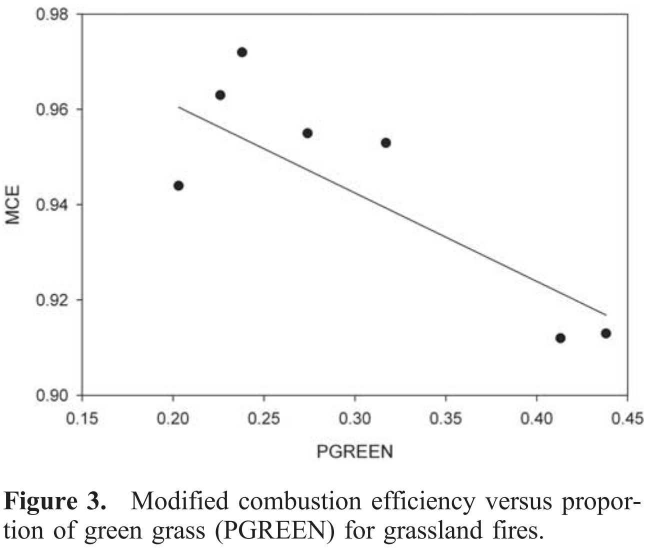

Figure 3. Modified combustion efficiency versus proportion of green grass (PGREEN) for grassland fires.

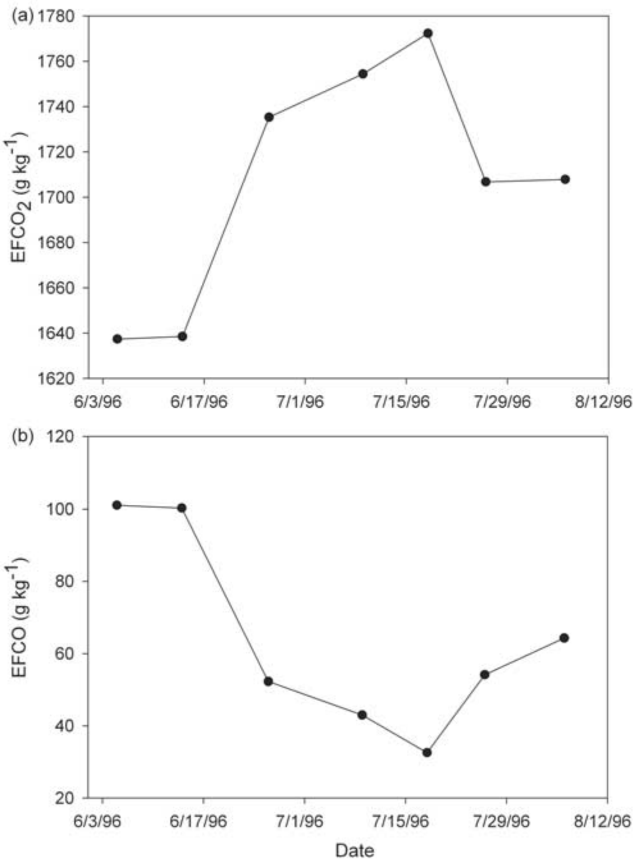

Figure 4. Seasonal emission factors for (a) CO_2 and (b) CO for grassland fires.

$$EFCO_2 = 1857.8 - 499.0(PGREEN), \quad R^2 = 0.76, \tag{2}$$

$$EFCO = -11.06 + 249.02(PGREEN), \quad R^2 = 0.73, \tag{3}$$

$$EFCH_4 = -0.705 + 8.114(PGREEN), \quad R^2 = 0.50, \tag{4}$$

$$EFNMHC = -1.631 + 14.298(PGREEN), \quad R^2 = 0.68, \tag{5}$$

$$EFPM_{2.5} = -0.747 + 16.138(PGREEN), \quad R^2 = 0.68. \tag{6}$$

[22] Linking PGREEN to a remotely sensed vegetation condition index, such as the Normalized Difference Vegetation Index (NDVI), which is sensitive to the presence of green vegetation, may be useful for regional applications of the above relationships to estimate emissions from grassland fires.

[23] In the woodland site, the lower $EFCO_2$ (Figure 8a) and the higher EFs for products of incomplete combustion

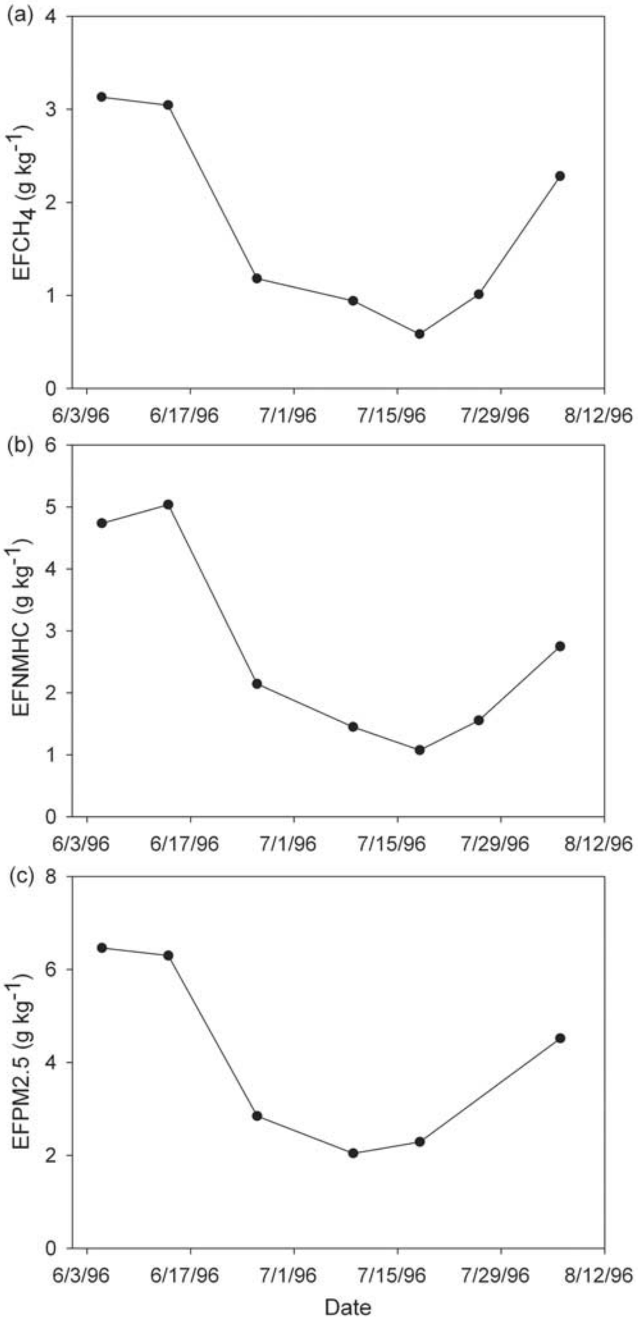

Figure 5. Seasonal emission factors for (a) CH_4, (b) NMHC, and (c) $PM_{2.5}$ for grassland fires.

(Figures 8b and 9a–9c) on the last day of burning suggest a higher contribution of smoldering combustion. This could be due to drier litter and woody fuels becoming more involved in the combustion and lowering the MCE. Additional early dry season studies are needed to evaluate seasonal emissions from diverse types of miombo woodlands, with different canopy covers, fuel loadings, land uses, vegetation structure and moisture conditions.

3.2. Ecosystem Differences

3.2.1. Methane

[24] Table 3 shows the regression lines and coefficients using the ecosystem-specific data and those using the combined grassland and woodland data set. A comparison of the $EFCH_4$ versus MCE regression models for the woodland and grassland ecosystems (Figure 10a) shows that the mean residual variation for the two separate models is not significantly different from the mean residual variation about a single regression model (i.e., for grassland and woodland EFs taken together) ($F = 1.90 < F_{crit} = 4.26$). This indicates that for the 1996 data the EFs for CH_4 are essentially the same for grassland and woodland savanna fires. No ecosystem difference was found in the $EFCH_4$ for controlled burns conducted in different southern African savanna ecosystems during SAFARI-92, as well [*Hao et al.*, 1996].

3.2.2. Nonmethane Hydrocarbons

[25] For NMHC, the mean residual variation about the two ecosystem-specific regression lines is significantly different from that of the common regression line ($F = 36.77 > F_{crit} = 4.26$), indicating an ecosystem dependence for the EFNMHC in the 1996 data (Table 3). Figure 10b illustrates the relationship between EFs and MCE for the two ecosystem types. There is a much greater increase in NMHC emissions with decreasing MCE in grassland than in woodland savannas. At the lowest MCE (0.907) found in this 1996 study, the predicted grassland EFNMHC is 86% higher than the measured woodland EFNMHC at this MCE. Thus it appears that using an ecosystem-specific model improves the fit for the 1996 NMHC data. This is in contrast with *Hao et al.* [1996] who found that the emission ratios of

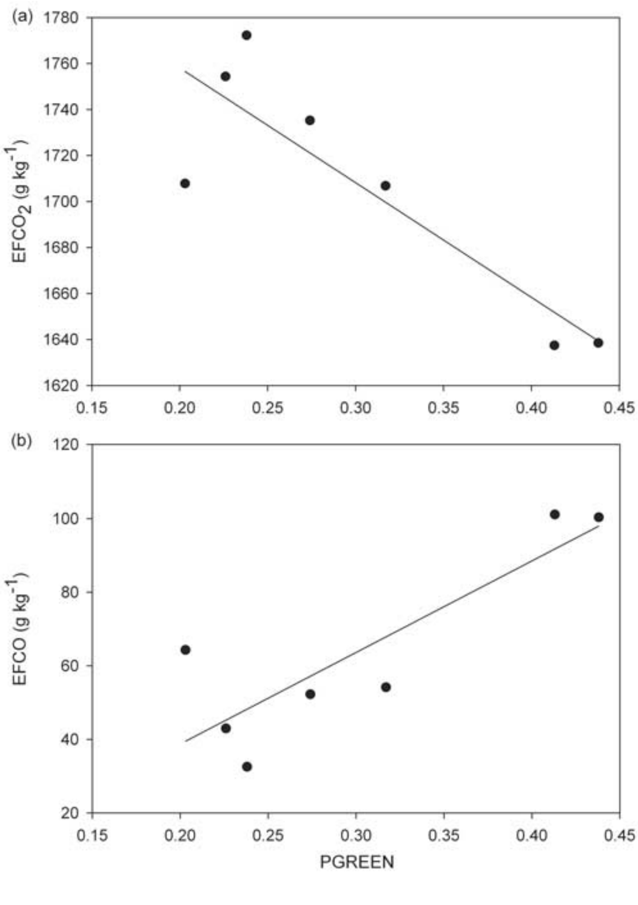

Figure 6. Emission factors for (a) CO_2 and (b) CO versus proportion of green grass for grassland fires.

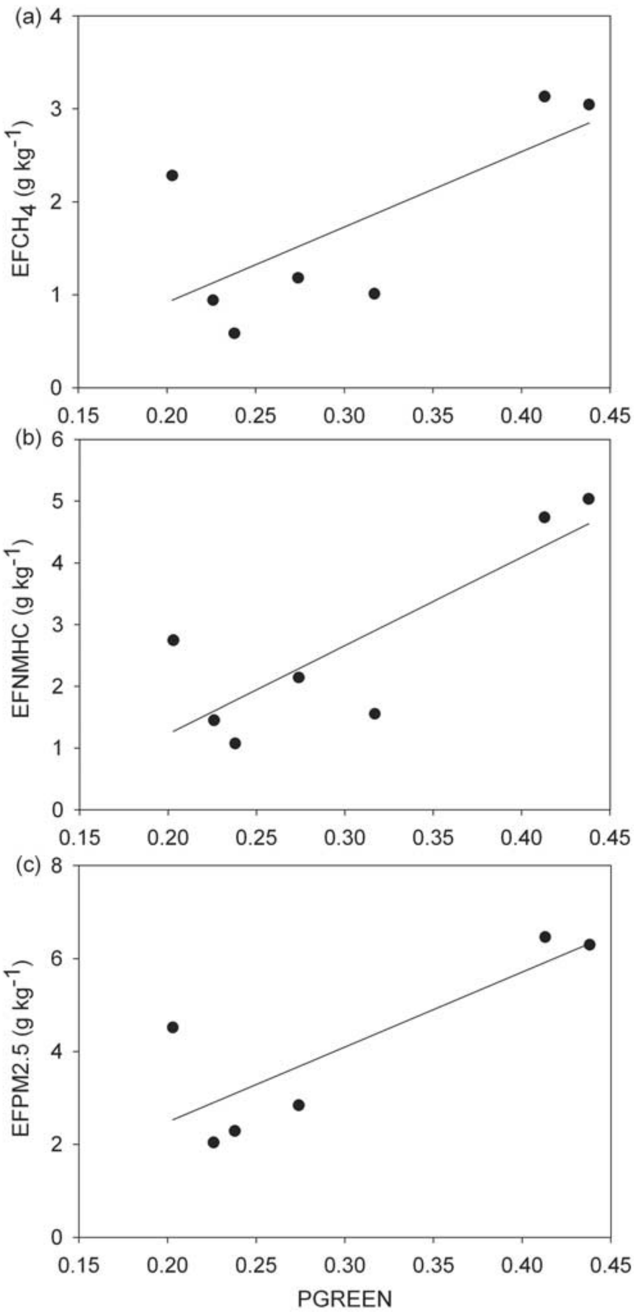

Figure 7. Emission factors for (a) CH_4, (b) NMHC, and (c) $PM_{2.5}$ versus proportion of green grass for grassland fires.

NMHC over CH_4 were independent of savanna type and fuel amount in the SAFARI-92 measurements.

3.2.3. Particulate Matter

[26] An ecosystem dependence exists also for $PM_{2.5}$ ($F = 6.44 > F_{crit} = 4.46$) (Table 3). There is approximately a difference of a factor of two between the two ecosystems at the lowest MCE in $EFPM_{2.5}$ (Figure 10c). The emissions are higher from woodland savanna than from grassland savanna fires, which is the opposite of what was observed for the NMHC emissions.

[27] The NMHC and $PM_{2.5}$ data indicate that there may be more of an ecosystem dependence early in the dry season than later in the dry season. The ecosystem-specific models for EF versus MCE hinge on a small number of low-MCE samples (especially for woodlands) and they need to be verified by more study. However, if the trends suggested from this unique set of early dry season measurements are valid, this has important implications for estimates of smoke emissions from southern African savanna fires.

3.3. Regional Synthesis of Emission Factors

[28] Figures 11a–11c and Table 3 integrate the EFs from the 1996 study with those from the SAFARI-92 and SAFARI-2000 dry season field campaigns [*Ward et al.*, 1996; *Hobbs et al.*, 2003; *Yokelson et al.*, 2003] to develop regional-average models of EFs versus MCE. Specifically, the woodland and grassland ecosystem-specific regression models from 1996 (Figures 10a–10c) are compared with the regional-average EF models to determine their maximum differences over the corresponding range of MCE values measured in this 1996 study. The regional-average models described in this section are considered to be more robust because they are based on measurements that were conducted in a variety of savanna regions, including humid woodland, semiarid woodland and moist grassland sites, and combine both late and early dry season measurements. In the case of NMHC and $PM_{2.5}$ (Figures 10b–10c and 11b–11c, respectively), the integration of the data sets significantly decreases the regression

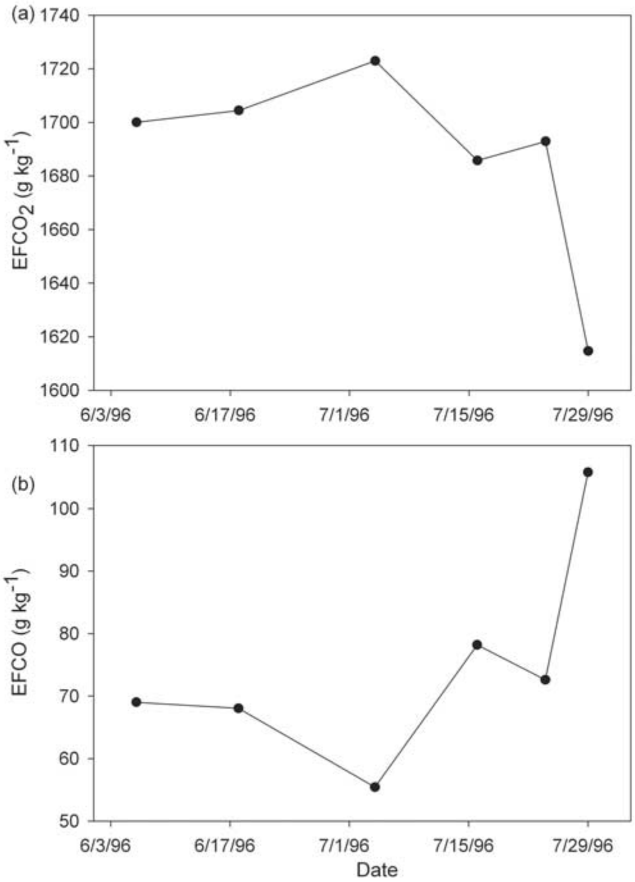

Figure 8. Seasonal emission factors for (a) CO_2 and (b) CO for woodland fires.

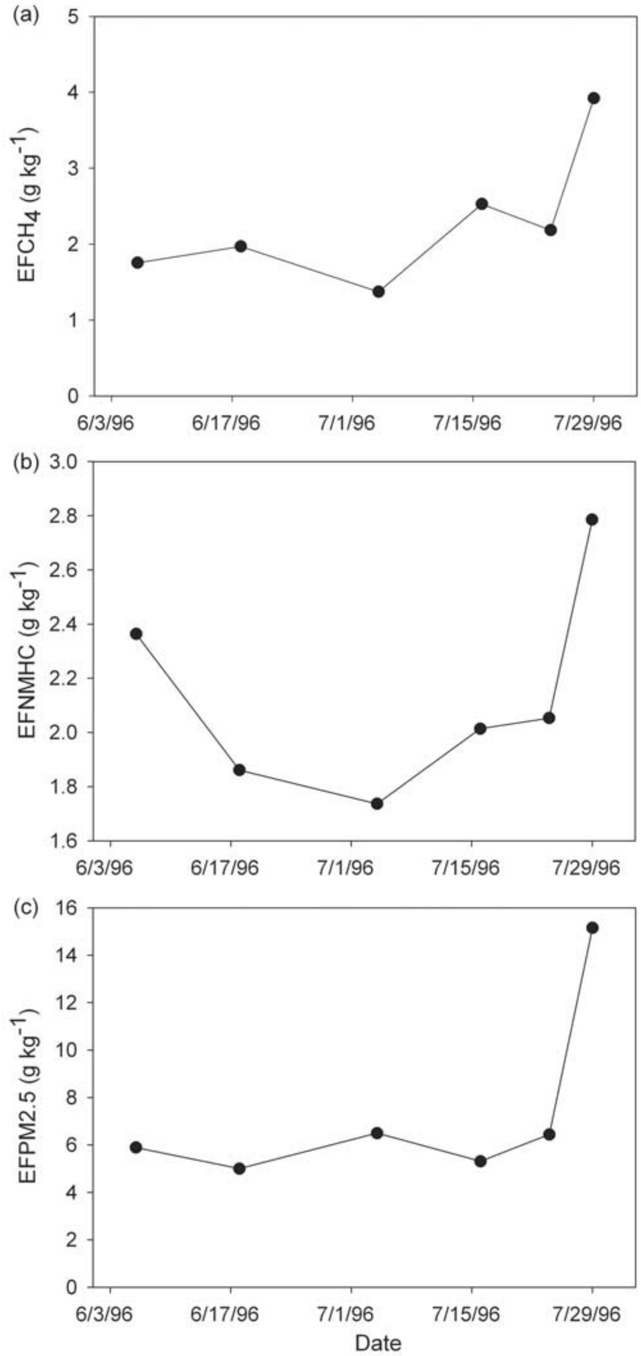

Figure 9. Seasonal emission factors for (a) CH_4, (b) NMHC, and (c) $PM_{2.5}$ for woodland fires.

coefficients (Table 3). For woodlands, the regional average model predicts an EFNMHC that is 38.9% larger at the lowest MCE of 0.907. The difference decreases with increasing MCE and becomes zero at an MCE value of 0.984. At the mean of all woodland MCE values observed here (0.935) the regional-average approach predicts an EFNMHC that is 32.0% larger compared with the woodland model. On the other hand, the regional-average model predicts an EFNMHC that is lower by 25.1% at the lowest grassland MCE of 0.912 and by 7.2% at the average grassland MCE of 0.945 compared with the grassland model. There is no difference in the grassland EFNMHC when using the two models at the MCE of 0.951 (where the regression lines cross). For MCE values greater than this, the regional average model predicts EFNMHC that are higher than the grassland model. For example, at the highest grassland MCE of 0.972, measured in this 1996 study, the regional average model predicts an EFNMHC that is higher by 77.5% compared with the grassland model.

[29] In the case of $EFPM_{2.5}$, the maximum difference between the regional average and the grassland models of 57.0% occurs at the highest grassland MCE value of 0.972. As the MCE decreases, the difference between the two models decreases but the two models never coincide over the entire range of grassland MCE values measured here. At the lowest grassland MCE of 0.912 the regional-average model predicts an $EFPM_{2.5}$ that is higher by 34.6% compared with the grassland model. Theoretically, the regional-average model will always over predict the grassland MCE values compared with the grassland model, since the calculated concurrence between the two models occurs at an MCE value of greater than 1.000. The regional-average model predicts an $EFPM_{2.5}$ for woodland fires that is higher by 33.6% at the highest woodland MCE value of 0.952, smaller by 31.9% at the lowest woodland MCE value of 0.907, and smaller by 11.7% at the average woodland MCE of 0.935, compared with the woodland model. More measurements are needed in the early dry season to determine if the 1996 data are outliers, or if an ecosystem dependence can be documented more strongly. In the case of CH_4 (Figures 10a and 11a), the integration of the data sets produces a small decrease in the correlation coefficient (Table 3) and little difference compared with the ecosystem-specific algorithms.

Table 3. Average Values of Regression Slopes, Intercepts, and Correlation Coefficients for Emission Factors for CO_2, CO, CH_4, NMHC, and $PM_{2.5}$ Versus the Modified Combustion Efficiency[a]

	Grasslands	Woodlands	Combined	Regional
		$EFCO_2$		
Intercept	−388.1	−613.6	−436.9	−288.4
Slope	2218.6	2460.7	2270.9	2118.1
R^2	0.97	0.99	0.98	0.90
		EFCO		
Intercept	1145.30	1119.07	1137.23	1158.08
Slope	−1144.79	−1117.02	−1136.34	−1157.63
R^2	0.99	0.99	0.99	0.98
		$EFCH_4$		
Intercept	42.951	56.710	47.068	46.929
Slope	−43.630	−58.214	−47.948	−47.737
R^2	0.94	0.98	0.94	0.81
		EFNMHC		
Intercept	65.982	22.757	47.916	36.367
Slope	−67.021	−22.059	−48.389	−35.885
R^2	0.97	0.76	0.65	0.44
		$EFPM_{2.5}$		
Intercept	75.924	211.108	124.050	95.762
Slope	−76.180	−217.932	−126.011	−95.488
R^2	0.96	0.73	0.58	0.32

[a] R^2, correlation coefficient.

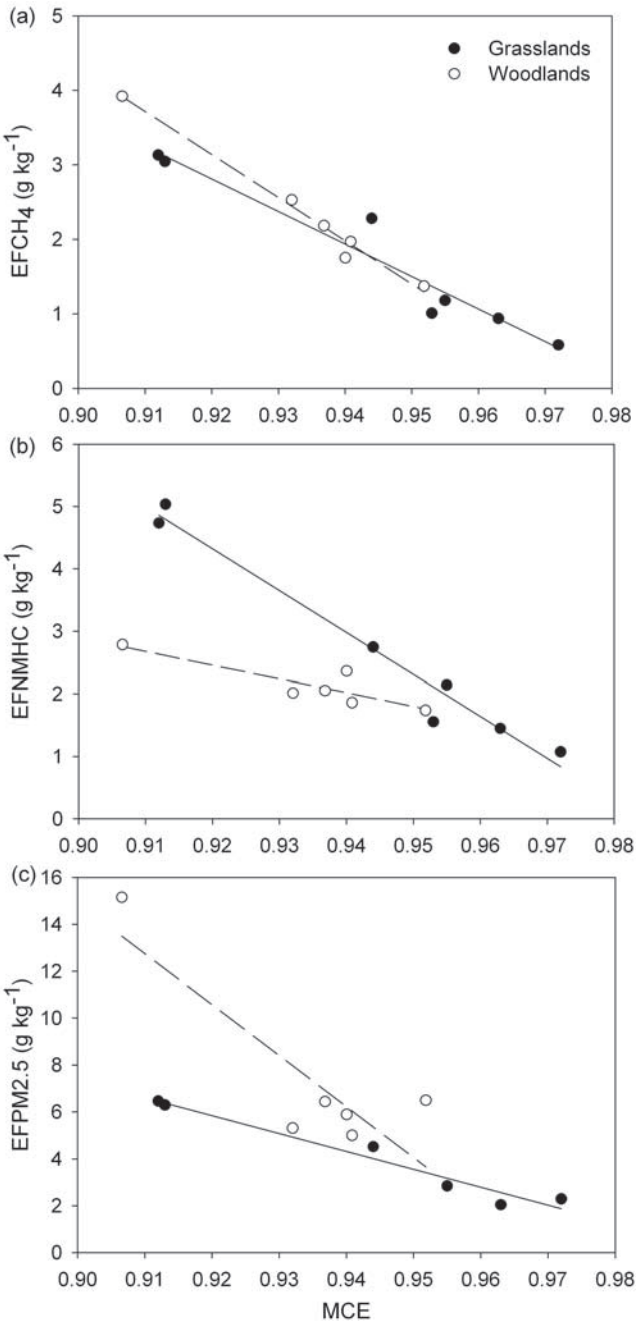

Figure 10. Emission factors for (a) CH_4, (b) NMHC, and (c) $PM_{2.5}$ versus modified combustion efficiency for grassland and woodland fires.

[30] Considering that the data used here were collected in different rainfall years (1992 was dry, 1996 was average, and 2000 was wet), in different locations, and were collected (ground and airborne) and analyzed using different methods (GC and airborne Fourier transform infrared spectroscopy), it is not surprising to find these variations between regional-average and ecosystem-specific EFs. It should be noted, that these differences have different meanings for various users of fire information. For regional and global emissions estimation, the differences in these EFs are likely of lesser importance relative to the larger uncertainties in some of the other modeling variables, such as fuel load and burned area, which may result in emission estimates that vary by an order of magnitude (compare *Scholes et al.* [1996] with *Hao et al.* [1990]).

[31] At the same time, it is important to know and consider the differences in EFs discussed here when reporting the overall error of a regional emissions model. For example, *Scholes et al.* [1996] estimated that their emissions model was accurate to within ±60%. Compared to that level

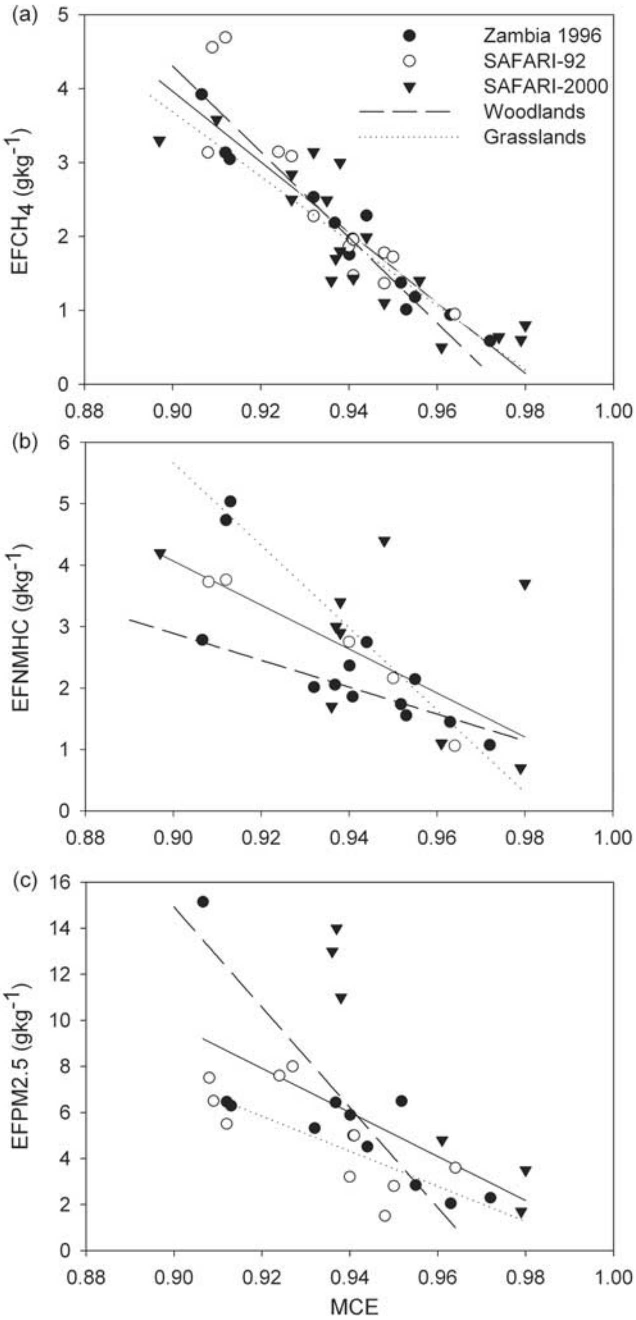

Figure 11. Regional integration of emission factors from this study, SAFARI-92, and SAFARI-2000 for (a) CH_4, (b) NMHC, and (c) $PM_{2.5}$ versus modified combustion efficiency. The corresponding grassland and woodland models are also shown.

of claimed accuracy, the differences between regional-average and ecosystem-specific EFs presented here appear significant and suggest that an ecosystem-specific approach could be more appropriate. The mixture of grassland and woodland fires, which likely changes seasonally and from year to year, will determine the importance of these differences and the resulting implications for regional emissions estimation.

[32] On the other hand, for landscape-level emission studies, for which accurate fuel loading databases are in place (e.g., national parks), EFs might prove to be a larger source of uncertainty than burned area and fuel consumption. Comprehensive ground-based measurements of burned area and fuel consumption are possible at this scale and the availability of high-resolution satellite information (e.g., Landsat, SPOT) permits a reasonably accurate estimation of area burned [e.g., *Korontzi et al.*, 2003]. Field data combined with satellite information can also provide reliable modeling of fuel consumption (T. Landmann, unpublished data, 2000). Unless there are explicit EF measurements over a specific fire event, EFs have to be modeled [*Ward et al.*, 1996; *Hoffa et al.*, 1999]. Depending on whether an ecosystem-specific model is used or not, the resulting emissions quantification outcome may differ significantly.

3.4. Prediction of Early Dry Season Emission Factors From the SAFARI-92 Models

[33] The objective of this section is to test the validity of applying the SAFARI-92 late dry season EFs versus MCE models to predict the range of early dry season EF values measured here. *Korontzi et al.* [2003] and *Justice et al.* [2002] compared seasonal non-CO_2 emissions, using Landsat-derived monthly time series of burned area and calculated seasonal EFs, with emissions using the annual area burned and late dry season values of EFs. It was found that considerable underestimation of products of incomplete combustion occurred when average late dry season EF values were used as representative of early dry season burning.

[34] Owing to the lack of early dry season data in the literature, *Korontzi et al.* [2003] and *Justice et al.* [2002] used the SAFARI-92 late dry season modeled relationships [*Ward et al.*, 1996] and the early dry season MCE values from *Hoffa et al.* [1999] to derive the seasonal EF values. Here, we compare the EF values predicted from the SAFARI-92 late dry season models with the EF values predicted from the combined grassland-woodland models, for the range of grassland and woodland MCE values measured in this early dry season 1996 study. The lowest MCE value (0.907) was measured in a woodland burn, whereas the highest MCE value (0.972) was measured in a grassland fire (Table 2). In the comparison we use the combined models (Table 3), rather than the ecosystem-specific grassland and woodland models, since the SAFARI-92 models were developed from a number of measurements at sites with variable fuel composition. The comparison indicates that the EF differences between the SAFARI-92 models and the combined models from this study are highest either at the low or the high end of MCE values, depending on the atmospheric species, and that the level of agreement improves for values of MCE that are in between (Table 4). The large difference, by a factor of 4.7, in late dry season $EFCH_4$ raises some questions about using the SAFARI-92 model to estimate CH_4 emissions at higher MCEs, such as the ones of grassland fires in the late dry season. The highest grassland MCE measured here (0.972) is greater than any of the MCEs measured during the SAFARI-92 campaign. This might be explained by the fact that only one pure grassland fire was studied in SAFARI-92.

Table 4. Comparison of Seasonal Emission Factors for CH_4, NMHC, and $PM_{2.5}$ Predicted From the Combined Grassland-Woodland Models of this 1996 Study With the Corresponding Seasonal Emission Factors Calculated Using the SAFARI-92 Models Over the Range of Modified Combustion Efficiency Values Measured in this Study

MCE	% Difference in $EFCH_4$	% Difference in EFNMHC	% Difference in $EFPM_{2.5}$
0.907	−13.9	1.6	32.6
0.912	−13.2	1.3	32.0
0.952	5.5	−2.9	30.0
0.972	369.0	−10.7	−2.7

3.5. Implications of Grassland Fires for Regional Emissions

[35] As shown above, the seasonality in grassland fire emissions is more apparent compared with woodland fires. To evaluate the potential importance of grassland fire emissions to southern African regional emissions budgets we analyzed the satellite-derived Global Burned Area Product 2000 (GBA-2000) for southern Africa [*Silva et al.*, 2003]. The most recent version of GBA-2000, released in December 2002 (J. M. N. Silva, personal communication, 2003) shows that a total area of approximately 1,071,100 km^2 burned in southern Africa in 2000, from which about 264,000 km^2 was in grasslands. The MODIS percent tree cover (PTC) remote sensing product [*Hansen et al.*, 2002] was used to distinguish between ecosystem types. Land areas with PTC less than or equal to 10% were classified as grasslands, whereas areas with PTC greater than 10% and smaller than 80% were classified as woodlands. The threshold PTC value of 10% was derived from the Food and Agricultural Organization of the United Nations (FAO) definition of forest [*Food and Agriculture Organization of the United Nations*, 2001]. Therefore it appears that at the regional level, grassland fires are important.

[36] *Korontzi et al.* [2003] demonstrated for grassland fires at the landscape level that due to seasonal effects, burned area is nonlinearly related to emission. The same amount of burned area may produce several times higher emissions of products of incomplete combustion early in the dry season compared with the late dry season. The analysis of GBA-2000 for the main dry season (May–October) in southern Africa shows that 57% of the burning occurs from May to July, 18% in August, 17% in September and 8% in October. The temporal distribution the GBA-2000 is in good general agreement with the TRMM active fire data in Figure 1. These results demonstrate that early dry season burning is wide spread, despite the common belief that August and September are the most intensive biomass

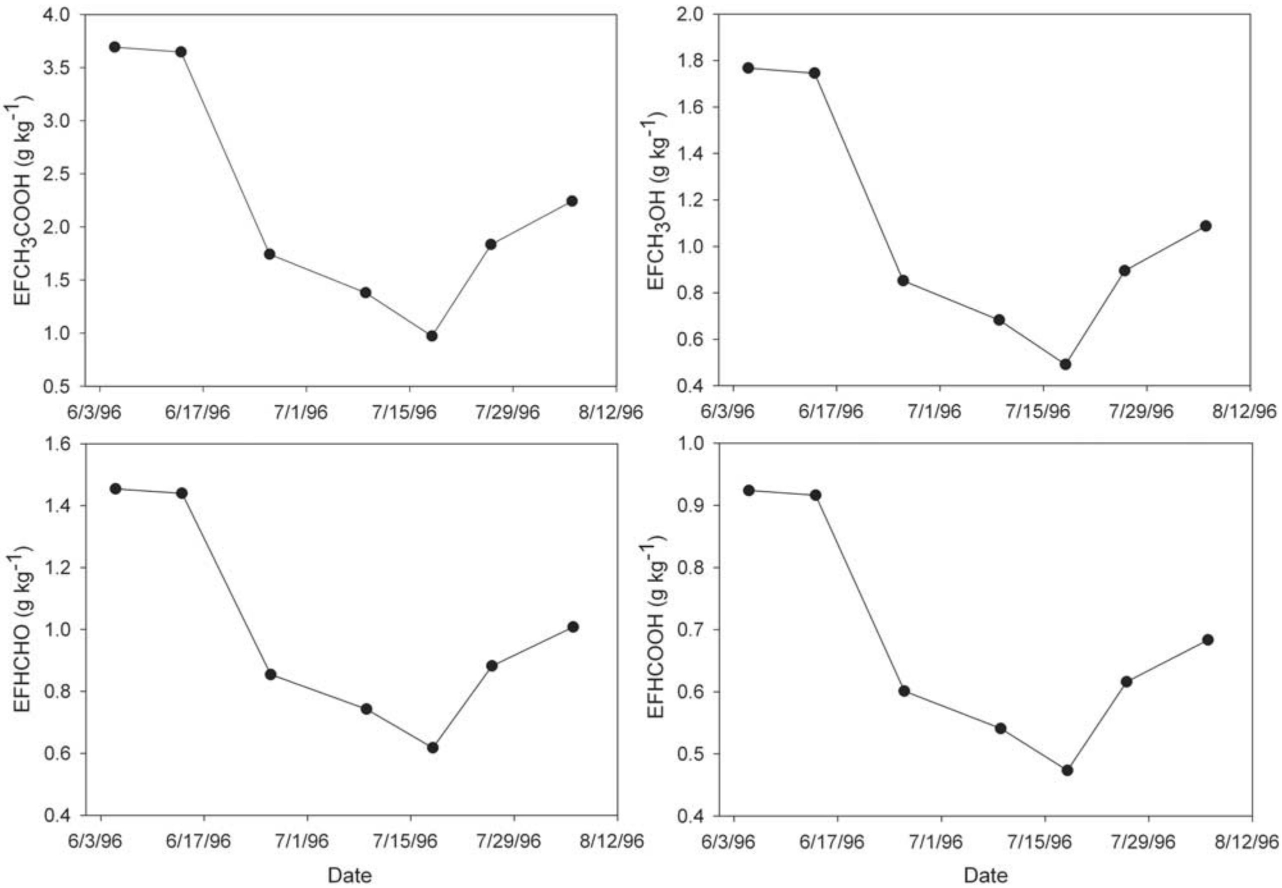

Figure 12. Calculated seasonal emission factors for selected oxygenated volatile organic compounds for grassland fires.

burning months in southern Africa, and that temporal patterns of biomass burning need to be integrated in the emissions modeling framework.

3.6. Seasonal Emission Factors for Oxygenated Volatile Organic Compounds

[37] One of the major gaps in our knowledge of the chemistry of the emissions from African savanna fires has been addressed recently by the first quantitative measurements of the EFs for oxygenated volatile organic compounds (OVOC) during the SAFARI-2000 dry season field campaign [*Yokelson et al.*, 2003]. The OVOC are about 5 times more abundant than NMHC in the southern hemisphere and they are more reactive (e.g., acetic acid (CH_3COOH), formic acid (HCOOH), and formaldehyde (HCHO), reported here) [*Singh et al.*, 2001]. Methanol (CH_3OH), which is fairly long lived, is the second most abundant organic compound in the atmosphere after CH_4. Here, we combine our seasonal grassland MCE values and the relationship of MCE versus PGREEN with the relationships of EFOVOC vs. MCE reported by *Yokelson et al.* [2003] to calculate the first seasonal trends in EF of these compounds for southern African grassland fires (Figure 12) and relate them to PGREEN. In the absence of early dry season EFOVOC versus MCE models, we applied the late dry season relationships to predict the early dry season EFOVOC. The calculated values of the EFOVOC in the early dry season are a maximum of 3.8, 2.4, 3.6, and 2.0 times higher for CH_3COOH, HCHO, CH_3OH and HCOOH, respectively, than the corresponding values in the late dry season. The OVOC emissions are related to PGREEN as following:

$$EFCH_3COOH = 9.836(PGREEN) - 0.749, \quad (7)$$

$$EFHCHO = 3.025(PGREEN) + 0.088, \quad (8)$$

$$EFCH_3OH = 4.618(PGREEN) - 0.318, \quad (9)$$

$$EFHCOOH = 1.630(PGREEN) + 0.188. \quad (10)$$

Note that the sum of the EFOVOC for the four OVOC that were most abundant in the SAFARI-2000 measurements is greater than the EFNMHC.

4. Conclusions

[38] Savanna fires are believed to produce zero net emissions of CO_2 due to its sequestration by subsequent vegetation growth [*Scholes et al.*, 1996]. At the same time

products of incomplete combustion may exhibit significant seasonal variations in their emissions [*Hoffa et al.*, 1999; *Justice et al.*, 2002; *Korontzi et al.*, 2003]. The seasonal budgets of these non-CO_2 trace gases and aerosols, and the implications for regional and global atmospheric chemistry, are largely unknown. The contribution of the early dry season emissions to the total annual emissions needs to be quantified.

[39] Information on EFs is required to improve the accuracy of emissions models. We have presented here the first early dry season EF measurements in southern African and they indicate some important and interesting seasonal trends in fire emissions and correlations to fuel characteristics. We have also derived the first seasonal EFOVOC for grassland fires and their relation to the proportion of green grass, which due to the importance of OVOC in tropospheric chemistry need to be included in future emissions modeling studies. The results from the integration of the different EF data sets enables estimates of the effects of using regional-average rather than ecosystem-specific EF models. The results presented here indicate that the seasonal trends of fire emissions require further attention. Clearly, a more intensive sampling is required to create a larger database that will allow the development of more robust seasonal EF models as a function of fuel condition. Fires exhibit high variability and the degree to which the seasonal measurements in this paper are representative of African savanna fires should not be overestimated. Through the development of seasonally sensitive emission estimates, it should be possible though, to do a better job of assessing emissions for Intergovernmental Panel on Climate Change (IPCC) national reporting.

[40] **Acknowledgments.** We thank Louis Giglio for providing the TRMM active fire data and Stephen Baker for the chemical analysis. We also thank the two anonymous reviewers for their helpful suggestions. This study was carried out as part of SAFARI-2000. Funding in partial support of this research was provided by NASA's MODIS grant NAS-531365. Stefania Korontzi is also supported by NASA's Earth System Graduate Research Fellowship.

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W. M. Hao and R. A. Susott, USDA Forest Service, Rocky Mountain Research Station, Fire Sciences Laboratory, P.O. Box 8089, Missoula, MT 59807, USA. (whao@fs.fed.us; rsusott@fs.fed.us)

P. V. Hobbs, Department of Atmospheric Sciences, University of Washington, Box 351640, 502 ATG Building, Seattle, WA 98195-1640, USA. (phobbs@atmos.washington.edu)

C. O. Justice and S. Korontzi (corresponding author), Department of Geography, 2181 LeFrak Hall, University of Maryland, College Park, MD 20742, USA. (justice@hermes.geog.umd.edu; stef@hermes.geog.umd.edu)

E. A. H. Smithwick, Department of Zoology, Birge Hall, University of Wisconsin, 430 Lincoln Drive, Madison, WI 53713, USA. (easmithwick@facstaff.wisc.edu)

D. E. Ward, Enviropyronics, P.O. Box 968, White Salmon, WA 98672, USA. (pyroward@aol.com)

R. J. Yokelson, Department of Chemistry, University of Montana, Missoula, MT 59812, USA. (byok@selway.umt.edu)