Emissions from miombo woodland and dambo grassland savanna fires

Parikhit Sinha,[1] Peter V. Hobbs,[1] Robert J. Yokelson,[2] Donald R. Blake,[3] Song Gao,[4,5] and Thomas W. Kirchstetter[6]

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[1] Airborne measurements of trace gases and particles over and downwind of two prescribed savanna fires in Zambia are described. The measurements include profiles through the smoke plumes of condensation nucleus concentrations and normalized excess mixing ratios of particles and gases, emission factors for 42 trace gases and seven particulate species, and vertical profiles of ambient conditions. The fires were ignited in plots of miombo woodland savanna, the most prevalent savanna type in southern Africa, and dambo grassland savanna, an important enclave of miombo woodland ecosystems. Emission factors for the two fires are combined with measurements of fuel loading, combustion factors, and burned area (derived from satellite burn scar retrievals) to estimate the emissions of trace gases and particles from woodland and grassland savanna fires in Zambia and southern Africa during the dry season (May–October) of 2000. It is estimated that the emissions of CO_2, CO, total hydrocarbons, nitrogen oxides (NO_x as NO), sulfur dioxide (SO_2), formaldehyde, methyl bromide, total particulate matter, and black carbon from woodland and grassland savanna fires during the dry season of 2000 in southern Africa contributed 12.3%, 12.6%, 5.9%, 10.3%, 7.5%, 24.2%, 2.8%, 17.5%, and 11.1%, respectively, of the average annual emissions from all types of savanna fires worldwide. In 2000 the average annual emissions of methane, ethane, ethene, acetylene, propene, formaldehyde, methanol, and acetic acid from the use of biofuels in Zambia were comparable to or exceeded dry season emissions of these species from woodland and grassland savanna fires in Zambia. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0312 Atmospheric Composition and Structure: Air/sea constituent fluxes (3339, 4504); 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); *KEYWORDS:* trace gases and particles, savanna fires, emissions and emission factors

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1. Introduction

[2] Biomass burning is the primary source of atmospheric pollution in the tropics [*Crutzen and Andreae*, 1990], and it affects large areas of the world through long-range transport [*Fishman et al.*, 1991]. Savanna fires are the largest source of biomass burning emissions worldwide, and tropical Africa contains about two thirds of the world's savanna [*Hao and Liu*, 1994]. Savanna burning is a source of a wide variety of compounds that are important in atmospheric chemistry, including carbon monoxide (CO), nitrogen oxides (NO_x), sulfur dioxide (SO_2), nonmethane organic compounds (NMOC), halocarbons, and particles [*Crutzen and Andreae*, 1990; *Sinha et al.*, 2003a]. Smoke aerosols perturb atmospheric radiation through their effects on light extinction and cloud properties [*Reid et al.*, 1998; *Haywood et al.*, 2003]. The photochemical oxidation of CO and NMOC in the presence of NO_x in smoke produces ozone (O_3) [*Radke et al.*, 1978; *Hobbs et al.*, 2003], a key precursor of the hydroxyl radical (OH), which is the primary oxidant in the troposphere.

[3] In August and September 2000 the University of Washington (UW) Cloud and Aerosol Research Group (CARG), with its Convair-580 research aircraft, participated in the Southern African Regional Science Initiative 2000 (SAFARI 2000) field project [*Swap et al.*, 2003]. One of the goals of SAFARI 2000 was to study the emissions and transformations of smoke from biomass burning in various parts of southern Africa. This paper describes measurements

[1]Department of Atmospheric Sciences, University of Washington, Seattle, Washington, USA.
[2]Department of Chemistry, University of Montana, Missoula, Montana, USA.
[3]Department of Chemistry, University of California, Irvine, California, USA.
[4]Department of Chemistry, University of Washington, Seattle, Washington, USA.
[5]Now at Department of Environmental Science and Engineering, California Institute of Technology, Pasadena, California, USA.
[6]Lawrence Berkeley National Laboratory, Berkeley, California, USA.

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of emissions from two prescribed savanna fires that occurred in miombo woodland and dambo grassland plots in Zambia on 1 September and 5 September 2000, respectively. The miombo woodland ecosystem is the most abundant type of savanna in southern Africa and the largest contiguous block of deciduous tropical woodlands and dry forests in the world, covering ~2.8 million km^2 including much of Africa from 4° to 17°S. Dambo grasslands are major enclaves within miombo woodlands, covering up to 40% of the landscape in some areas [*International Geosphere-Biosphere Programme*, 1997].

[4] Normalized excess mixing ratios are given at various locations over and downwind of the miombo and dambo fires for a number of gaseous species, including carbon dioxide (CO_2). CO, SO_2, dimethyl sulfide (DMS), NO_x, ammonia (NH_3), hydrogen cyanide (HCN), methane (CH_4), NMOC, halocarbons, and particulates. Emission factors are given for these species, and they are combined with measurements of fuel loading, combustion factors, and area burned to estimate their emissions from woodland and grassland savanna fires in Zambia and southern Africa during the dry season of 2000.

2. Sampling Techniques and Instrumentation

[5] Various trace gas and particle species were measured continuously or intermittently aboard the UW Convair-580 research aircraft. A complete list of the instruments aboard the Convair-580 in SAFARI 2000, and the methods of sampling are given in a technical appendix by P. V. Hobbs included by *Sinha et al.* [2003a]. Only the instruments and techniques that provided the measurements presented in this paper are described here.

[6] Continuous measurements of SO_2 were made using a Teco model 43S pulsed-fluorescence analyzer (precision of 7%, detection limit of 0.1 ppbv). The total concentration of particles (CN) in the size range ~0.003–3 μm diameter was measured continuously with a TSI 3025A ultrafine condensation particle counter (precision of 10%). The dry aerosol light-scattering coefficient was measured continuously with an MS Electron three-wavelength (0.45, 0.55, and 0.70 μm) nephelometer.

[7] Evacuated electropolished stainless steel canisters were used to sample ambient air using a stainless steel inlet that passed through the aircraft fuselage. For each canister sample, mixing ratios of selected C_2–C_9 nonmethane hydrocarbons (NMHC), methyl halides, DMS, and methyl nitrate (CH_3ONO_2) were determined by gas chromatography (GC) with flame ionization detection (FID), electron capture detection, and mass selective detection. The precision of the measurements was 3%, and the typical NMHC detection limit was 3 parts per trillion by volume. Mixing ratios of CO_2 (precision of 3%), CO (precision of 5%), and CH_4 (precision of 0.1%) in the canisters were determined using three separate GC/FID systems. A detailed description of the analytical procedure for the canister samples is given by *Colman et al.* [2001].

[8] An airborne Fourier transform infrared spectrometer (AFTIR) with a separate and specially coated inlet that directed ram air through a Pyrex multipass cell with an exchange time of 4–5 s was deployed aboard the UW Convair-580. The AFTIR measured CO_2, CO, CH_4, NO, nitrogen dioxide (NO_2), O_3, ethene, acetylene, formaldehyde, methanol, acetic acid, formic acid, NH_3, and HCN. The AFTIR technique is described by *Yokelson et al.* [2003].

[9] Aerosol samples collected on quartz filters (Pallflex 2500 QAT-UP) were used to determine the concentration of particulate carbon. The quartz filters were baked before use at 800°C for at least 6 hours to remove carbonaceous impurities. They were then analyzed for total carbon content using the evolved gas analysis method described by *Novakov* [1981, 1982] and *Kirchstetter et al.* [2003]. Black carbon (BC) concentrations were measured with an optical transmission technique similar to that described by *Rosen and Novakov* [1983]. This method compares the attenuation of white light through a loaded filter relative to that of a blank filter. The relationship between optical attenuation (ATN) and the BC concentration (in μg cm^{-2}) is given by ATN = σBC, where ATN = 100 × ln(I_o/I), I_o and I are the transmitted light intensities through the blank and loaded filters, respectively, and σ is the mass absorption cross section for BC deposited on the quartz filters (in m^2 g^{-1}). A value of 20 m^2 g^{-1} was used for the mass absorption cross section [*Gundel et al.*, 1984], which is consistent with the calibration factor of a commercial aethalometer that employs the same optical transmission method for measuring BC concentrations [*Bodhaine*, 1995]. Further discussion of this optical transmission method, including uncertainties and comparison with other methods, is given by *Kirchstetter et al.* [2003] and *Sinha et al.* [2003b].

[10] Particles were also collected on Teflon filters (Gelman Sciences Teflon membrane, 2.0 μm pore size). The Teflon filters were weighed before and after particle sampling in a relative humidity (RH)– and temperature (T)-controlled chamber (RH = 40%, T = 293 K) to determine the masses of dry particulate matter (PM) collected on the filters. From control and field blank filters the uncertainty of the total dry PM measured with these filters was estimated to be ±6 μg. After gravimetric analysis the Teflon filters were extracted in deionized water (high-performance liquid chromatography grade) and analyzed using a standard ion chromatography system (Dionex DX 500). This analysis yielded mass concentrations of three inorganic ions (nitrate, sulfate, and chloride) with a precision of 5%. An inductively coupled plasma-atomic emission spectrometer (Jarrell Ash 955) was used to measure the mass concentration of the potassium ion to a precision of 4%. Further details on the Teflon filter analyses of aerosol compounds are given by *Gao et al.* [2003].

[11] When sampling smoke plumes we employed a "grab bag" technique to obtain samples for the filters and SO_2. The grab bag consisted of a 2.5 m^3 electrically conducting plastic (Velostat) bag that could be filled with a sample of smoke in 12 s when exposed to ram air. The grab bag system had an aerosol cutoff diameter of about 4 μm; larger particles were lost in the inlet and on the walls of the grab bag.

3. Definitions and Calculations of Normalized Excess Mixing Ratios, Emission Factors, and Combustion Efficiencies

[12] A normalized excess mixing ratio of species *X* is obtained by dividing the excess (i.e., above background) mixing ratio of *X* by the excess mixing ratio of a simulta-

neously measured reference gas (such as CO_2 or CO). For example, a normalized excess mixing ratio (NEMR) of species X relative to CO is

$$\mathrm{NEMR}(X)_{\mathrm{CO}} = \frac{[\Delta X]}{[\Delta \mathrm{CO}]}. \quad (1)$$

[13] Emission factors for gases and particles were calculated using the carbon balance mass method [*Radke et al.*, 1988]. The underlying premise of this method is that all of the carbon combusted in a fire and released to the atmosphere is emitted into the smoke plume as CO_2, CO, CH_4, NMOC, and particulate carbon (PC). The emission factor (EF) of a species X is the ratio of the excess mass concentration $[\Delta X]$ of X to the excess total carbon concentration $[\Delta \mathrm{C}]$ in the plume:

$$\mathrm{EF}(X) = \frac{[\Delta X]}{[\Delta \mathrm{C}]_{\mathrm{CO_2}} + [\Delta \mathrm{C}]_{\mathrm{CO}} + [\Delta \mathrm{C}]_{\mathrm{CH_4}} + [\Delta \mathrm{C}]_{\mathrm{NMOC}} + [\Delta \mathrm{C}]_{\mathrm{PC}}}. \quad (2)$$

The emission factor is expressed in units of grams of X emitted per gram of carbon burned. To convert this emission factor to units of grams of X emitted per gram of fuel burned, EF is multiplied by the mass fraction of carbon in the fuel. Typically, the carbon content of biomass fuels varies from 45 to 55% [*Susott et al.*, 1996]; in this paper we assume that it is 50%.

[14] The combustion efficiency (CE) is the ratio of carbon emitted as CO_2 to the total carbon emitted [*Ward and Hardy*, 1991],

$$\mathrm{CE} = \frac{[\Delta \mathrm{C}]_{\mathrm{CO_2}}}{[\Delta \mathrm{C}]_{\mathrm{CO_2}} + [\Delta \mathrm{C}]_{\mathrm{CO}} + [\Delta \mathrm{C}]_{\mathrm{OC}} + [\Delta \mathrm{C}]_{\mathrm{PC}}}, \quad (3)$$

where the subscript OC indicates total organic compounds (i.e., methane and NMOC). Thus CE is the fraction of fuel carbon emitted that is completely oxidized to CO_2. Combustion efficiency is a useful way to quantify the relative amounts of flaming and smoldering combustion: When CE < 1, the fire emissions were produced by a mixture of flaming and smoldering combustion; when CE $>$ 90%, more than 50% of the emissions were produced by flaming combustion; and when CE $<$ 90%, more than 50% of the emissions were produced by smoldering combustion [*Ward and Hardy*, 1991].

[15] Although CE is a useful quantity for fire models, it is often difficult to measure all of the individual carbon species in the emissions from a fire. Therefore, in this study we use the modified combustion efficiency (MCE) to describe the relative amounts of flaming and smoldering combustion [*Ward and Hao*, 1992], which is defined as

$$\mathrm{MCE} = \frac{[\Delta \mathrm{C}]_{\mathrm{CO_2}}}{[\Delta \mathrm{C}]_{\mathrm{CO_2}} + [\Delta \mathrm{C}]_{\mathrm{CO}}}. \quad (4)$$

Since OC and PC are emitted in relatively small quantities relative to CO_2 and CO, the difference between CE and MCE is typically only a few percent.

4. Vegetation and Fires

[16] The prescribed fire on 1 September 2000 was ignited at 0846 UTC (local time is UTC plus 2 hours) at 14.82°S, 24.48°E near Kaoma, Zambia. The fire burned for ~90 min over 95 ha (J. M. C. Pereira et al., unpublished manuscript, 2002) The vegetation type, as classified by *White* [1981, 1983], was "wetter Zambezian miombo woodland." This ecosystem, which we will refer to as "miombo woodland," is dominated by the tree genera *Brachystegia*, *Julbernardia*, and *Isoberlinia* [*White*, 1981, 1983]. The vegetation combusted in the miombo woodland fire considered here consisted of standing grass, leaf and twig litter, large fallen dead wood, and live leaves. Fuel loading, combustion completeness, fire intensity, and rate of spread of this fire are given by J. M. C. Pereira et al. (unpublished manuscript, 2002).

Figure 1. Photograph of a dambo grassland fire in Zambia taken at 1157 UTC (8 min after ignition) on 5 September 2000 from the University of Washington Convair-580 research aircraft. (Photograph courtesy of P. V. Hobbs.)

[17] The prescribed fire on 5 September 2000 was ignited at 1149 UTC at 14.81°S, 24.46°E, also near Kaoma, Zambia. The fire burned for ~50 min over 70 ha (J. M. C. Pereira et al., unpublished manuscript, 2002). The vegetation type, as classified by *White* [1981, 1983], was "edaphic grassland mosaics with semiaquatic vegetation." This ecosystem, which we will refer to as "dambo grassland," occupies seasonally waterlogged shallow valley depressions with a medium-dense, uniform grass mat dominated by the grass species *Loudetia simplex* [*White*, 1981, 1983]. The vegetation combusted in the dambo grassland fire considered here consisted exclusively of standing grass. Other biomass parameters for this fire are given by J. M. C. Pereira et al. (unpublished manuscript, 2002). Figure 1 shows an image of the dambo fire taken at 1157 UTC, 8 min after ignition.

5. Results

5.1. Background Conditions

[18] The miombo and dambo fires took place in different background meteorological conditions. In the first week of September 2000 the passage of a cold front originating in the subtropics induced northwesterly flow throughout southern Africa. Heavy smoke from intense biomass burning regions in Angola was transported over Zambia and other southern African nations toward the Indian Ocean in an event that has been termed the "river of smoke" [*Annegarn et al.*, 2002]. On 1 September the river of smoke was just beginning, but by 5 September it was well

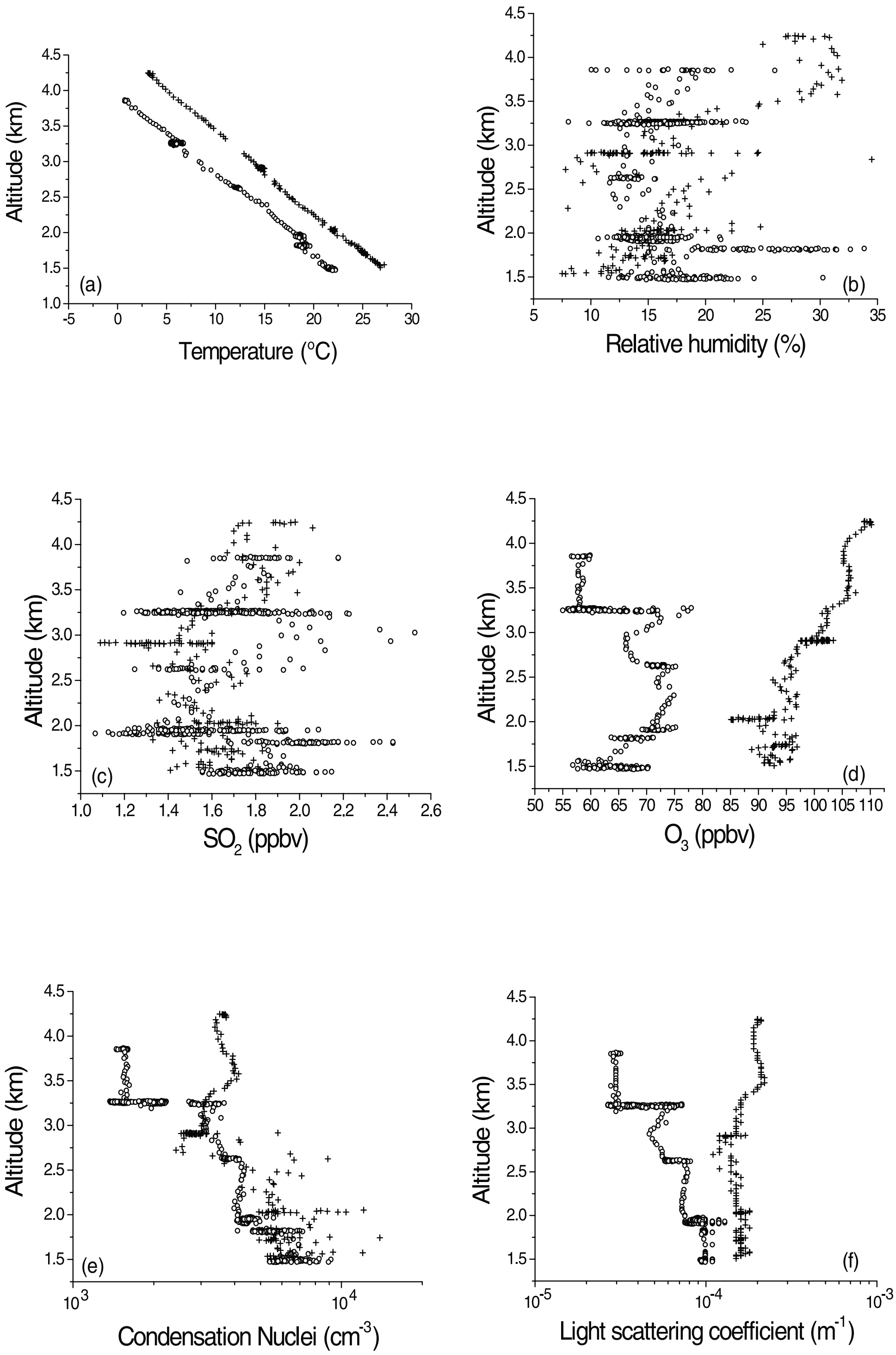

Figure 2. Vertical profiles of temperature, relative humidity, SO_2, O_3, CN, and the light-scattering coefficient (at λ = 0.55 μm) measured upwind of the miombo woodland fire (circles) and dambo grassland fire (crosses) from 0650 to 0856 UTC on 1 September 2000 and from 1054 to 1150 UTC on 5 September 2000, respectively.

underway. Consequently, background conditions for the miombo fire on 1 September were generally less polluted than for the dambo grassland fire on 5 September.

[19] Background vertical profiles of temperature, relative humidity, SO_2, O_3, CN, and light-scattering coefficient (at λ = 0.55 μm) for the miombo and dambo fires are shown in Figure 2. In both cases, temperature lapse rates were generally 8°–9°C km^{-1}, resulting in conditionally unstable atmospheres. For the miombo fire, stable layers were present at ~1.75 and ~3.25 km, which inhibited vertical transport of the smoke. For the dambo fire the atmosphere was well mixed up to at least 4 km.

[20] Ozone concentrations upwind of the dambo fire were ~20–30 ppbv higher than those upwind of the miombo fire, reflecting the transport of aged biomass burning emissions within the river of smoke. This is supported by background CO concentrations measured by AFTIR for the miombo (~300 ppbv) and dambo (~500 ppbv) fires, which indicate a more polluted background for the dambo fire than the miombo. Also, upwind of the fires, the light-scattering coefficient ranged from 4×10^{-5} to 2×10^{-4}, with higher values upwind of the dambo fire than upwind of the miombo fire. On the other hand, background CN and SO_2 concentrations were similar for both cases, ranging from $\sim 5 \times 10^3$ to 1×10^4 cm^{-3} and from 1 to 2.5 ppbv, respectively, near the surface. Despite similar CN profiles the difference in the light-scattering coefficient between the two cases suggests different aerosol size distributions with more combustion-generated submicron particles upwind of the dambo grassland fire than the miombo woodland fire. The relative humidity was low (<35%) upwind of both fires. Therefore the effect of RH on the aerosol light-scattering coefficient was small [*Magi and Hobbs*, 2003]. The average surface wind speed upwind of the miombo and dambo fires was 5.6 ± 2.1 m s^{-1} from the north and 6.3 ± 2.2 m s^{-1} from the northeast, respectively.

5.2. Plume Dimensions

[21] Horizontal cross sections of the plumes of smoke from the miombo and dambo fires are shown in Figure 3. The width and length of the miombo plume were ~3–6 and ~14 km, respectively, and the width and length of the dambo plume were ~2–3 and ~5 km, respectively. The smaller plume dimensions of the dambo fire reflect in part the smaller area burned (70 ha) compared to the area burned by the miombo fire (95 ha). Also, the duration of the dambo fire (~50 min) was less than that of the miombo fire (~90 min). Surface wind speeds were similar in both cases (~6 m s^{-1}). Since the combusted vegetation in the dambo fire consisted exclusively of standing grass, it burned more rapidly than the miombo fire, which consumed both standing grass and less flammable fuel such as large, fallen dead wood. In both fires, CN concentrations peaked at $\sim 10^6$ cm^{-3} over the fire and decreased to nearly background concentrations of $\sim 10^4$ cm^{-3} several kilometers downwind of the fires. The high background concentrations of CN limited the distances downwind that the smoke plumes could be detected.

5.3. Plume Samples

[22] Smoke samples from both fires were obtained in a series of passes perpendicular to the plume axes at various distances and altitudes downwind of the fires. Table 1 summarizes the time, location, age, and various state parameters for these samples. Ground level was ~1100 m, and most smoke samples were obtained <300 m above ground level. The age of each smoke sample was estimated by dividing the downwind distance of the sample by the average wind speed. The MCE of each sample indicates the stage of combustion during which the sample was released, with values >0.90 indicating predominantly flaming combustion and values <0.90 indicating predominantly smoldering combustion.

5.4. Normalized Excess Mixing Ratios

[23] Table 2 shows the excess mixing ratios of 49 trace gas and particle species normalized with respect to excess CO for samples 1–8 from the miombo woodland fire and samples a–f from the dambo grassland fire described in Table 1. The data set is unique in providing plume measurements of recently emitted smoke of known ages (~1–40 min and ~1–4 min for the miombo woodland and dambo grassland savanna fire, respectively (Table 1)) for a large number of trace gas and particle species, including carbon dioxide, carbon monoxide, sulfur dioxide, nitrogen oxides, methane, ammonia, dimethyl sulfide, nonmethane organic compounds, halocarbons, gaseous organic acids, aerosol ionic components, carbonaceous aerosols, and condensation nuclei. Additionally, because the fires in this study were prescribed, the combustion processes at the ground were well characterized (section 4). A discussion of the variations in the measured emissions from southern African savanna fires is given by *Sinha et al.* [2003a] and *Yokelson et al.* [2003], and the subsequent evolution of the emissions is discussed by *Hobbs et al.* [2003].

[24] The downwind transformations of chemical species in the plume of a 1000 ha savanna fire in South Africa

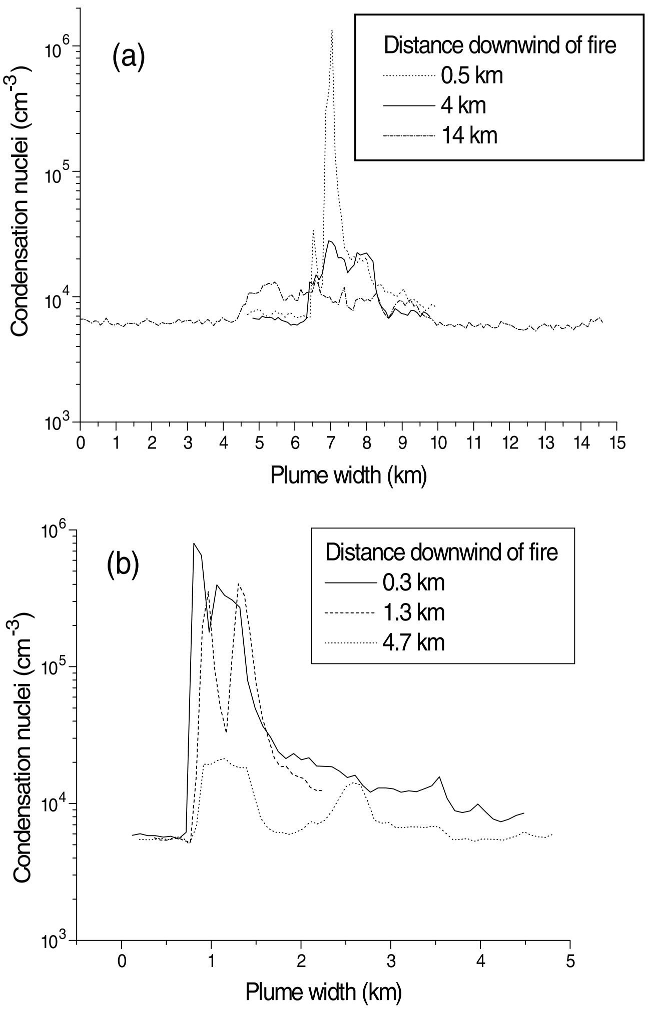

Figure 3. Condensation nucleus concentrations across the widths of (a) miombo woodland and (b) dambo grassland smoke plumes at three distances downwind of each of the fires.

Table 1. Samples of Smoke From the Miombo Woodland Fire on 1 September 2000 and the Dambo Grassland Fire on 5 September 2000

Sample[a]	Time, UTC	Latitude, °S	Longitude, °E	Distance From Fire, m	Altitude, m	Age of Smoke, min	Temperature, °C	Pressure, hPa	Relative Humidity, %	Modified Combustion Efficiency
1	0857	14.798	24.487	2,549	1,340	7.5	23	870	17.5	0.935
2	0902	14.816	24.478	517	1,325	1.5	24	871	17.0	0.917
3	0909	14.826	24.477	744	1,257	2.2	24.8	878	17.0	0.944
4	0919	14.829	24.482	1,019	1,330	3.0	23.8	871	16.7	0.942
5	0927	14.817	24.461	2,073	1,905	6.1	18.3	814	25.3	0.952
6	0939	14.822	24.364	12,495	1,976	36.8	18.1	810	25.1	0.913
7	0947	14.806	24.349	14,150	1,959	41.7	18.8	811	18.9	—
8	1006	14.814	24.444	4,009	2,055	11.8	18.1	803	21.0	0.985
a	1155	14.812	24.457	374	1,292	1.0	30.1	871	10.9	0.923
b	1157	14.814	24.455	750	1,339	2.0	29.7	867	10.3	0.960
c	1202	14.815	24.454	823	1,340	2.2	30.4	865	8.8	0.977
d	1205	14.814	24.453	912	1,323	2.4	32.2	868	9.7	0.976
e	1209	14.818	24.451	1,259	1,343	3.3	29.4	865	8.7	0.966
f	1233	14.814	24.448	1,346	1,347	3.6	28.8	863	7.4	0.961

[a]Samples 1–8 are from the miombo woodland fire; samples a–f are from the dambo grassland fire.

(the Timbavati fire) have been described by *Hobbs et al.* [2003]. The Timbavati fire occurred in the semiarid south Zambezian undifferentiated woodland savanna of the South African lowveld. Various alkenes and NO_2 were observed to be depleted in smoke aged about 40–45 min downwind of the Timbavati fire, while other species such as gaseous acetic acid, ozone, and particulate nitrate were enhanced in the aged smoke.

[25] Since the areal extents of the two Zambian fires were an order of magnitude less than those of the Timbavati fire, fewer downwind measurements were obtained in these fires. Nevertheless, downwind enhancements of gaseous acetic acid and particulate nitrate were observed in the Zambian fires (Table 2). Combining the normalized excess mixing ratios from the miombo woodland and dambo grassland fires with those from the Timbavati fire provides linear estimates of gaseous acetic acid and particulate nitrate production in aged smoke downwind of southern African savanna fires (Figure 4).

[26] In addition to nitrate particles the normalized excess mixing ratios of total particulate matter, organic carbon, chloride, and sulfate were enhanced by 60–300% in smoke aged $\sim$37 min relative to smoke aged $\sim$1.5 min for the miombo woodland fire (Table 2). Oxidation of NMOC, NO_x, and SO_2 and subsequent gas-particle conversion has been shown to enhance particle distributions in aged smoke downwind of savanna fires [*Gao et al.*, 2003; *Hobbs et al.*, 2003; *Kirchstetter et al.*, 2003].

[27] Normalized excess mixing ratios of four species (CO_2, CH_4, ethene, and acetylene) were obtained by two techniques (GC/C and AFTIR) in the smoke from the two Zambian fires. Normalized excess mixing ratios from these techniques for samples obtained at approximately the same time are compared in Figure 5. The slope of the correlation is 1.03 ± 0.12 with a correlation coefficient (r^2) of 0.88, indicating reasonable agreement between these two independent measurement techniques, except for ethene for which emission factors obtained by AFTIR were about a factor of 2 greater than those obtained by GC/C. It should be noted, however, that these measurements were not made on exactly the same smoke samples since the two measurement techniques had different sampling inlets and slightly different sampling times.

5.5. Emission Factors and Estimates of Regional Emissions

[28] Estimates of fuel consumed by woodland and grassland savanna fires in southern Africa are presented in Table 3. Combining these fuel consumption estimates with emission factors for 42 trace gas and seven particulate species from the miombo woodland and dambo grassland fires (Table 4), we can obtain estimates of the emissions of these trace gases and particles from woodland and grassland savanna fires in southern Africa during the dry season (May–October) of 2000 (Table 4).

[29] The fuel consumption values in Table 3 were calculated by multiplying burned area by the amount of fuel biomass above ground and by a combustion factor (CF), defined as the fraction of the available fuel biomass above ground combusted in a fire. Burned areas by month and country in southern Africa in 2000 for two broad vegetation types, woodland savanna and grassland savanna, were obtained from SPOT satellite burned area retrievals (Global Burnt Area 2000 Project, Joint Research Centre, Ispra, Italy, available at www.grid.unep.ch/activities/earlywarning/preview/ims/gba). The burned-area estimates used in our study differ from those reported by *Silva et al.* [2003] because of updates in the SPOT burned-area algorithm. The amount of biomass fuel loading above ground in plots of miombo woodland and dambo grassland in Zambia was measured by *Hoffa et al.* [1999], *Shea et al.* [1996] and J. M. C. Pereira et al. (unpublished manuscript, 2002) with average values of 0.87 ± 0.30 kg m^{-2} and 0.34 ± 0.10 kg m^{-2} for miombo woodland and dambo grassland, respectively. Since the moisture content of vegetation decreases as the dry season progresses, the CF increases from May to October in southern Africa [*Hoffa et al.*, 1999]. The fuel consumption measurements indicate that for miombo woodland fires the CF increases fairly linearly from a value of $\sim$0.01 at the beginning of the dry season to $\sim$0.89 near the end of the dry season. For dambo grassland fires the CF remains at 0.7 ± 0.2 throughout the dry season.

Table 2. Normalized Excess Mixing Ratios of 49 Trace Gas and Particle Species for Samples 1–8 and a–f from Table 1[a]

Species	Measurement Technique[b]	Sample 1	2	3	4	5	6	7	8	a	b	c	d	e	f
CO_2	GC/C		14,700	15,100					64,800		24,900			28,800	
	AFTIR	14,500	8,700	18,700	16,100	20,000	10,500			12,100	22,800	43,200	40,200		24,600
CH_4	GC/C		36.4	29.0					74.1		23.0			22.5	
	AFTIR	42.1	44.0	32.1	30.6	29.2	55.6			26.3	27.8	44.2	40.9		17.5
NO	AFTIR	10.1	12.2	19.4	15.1					6.4	7.8	30.1	41.4		22.9
NO_2	AFTIR	35.5	19.7	32.7	35.4	39.8				28.4	40.9	48.1	40.0		51.3
NH_3	AFTIR	17.8	16.2	5.5	4.6	2.7	44.5								
HCN	AFTIR	8.1	5.3	6.5	4.1					5.0	5.8	13.6	12.6		
$MeONO_2$	GC/C		0.0008	0.0020					0.0019		0.0027			0.0012	
SO_2	Teco 43S		2.3		1.0			4.4	11.1		1.2			6.1	6.9
DMS	GC/C		0.77	0.36							0.01				
O_3	AFTIR	−18.8	−12.6	−15.5	−16.5	−25.2	9.0			−5.0	−13.4	−22.3	−13.0		−37.8
CH_3Br	GC/C		0.0016	0.0023					0.0046		0.0004			0.0011	
CH_3Cl	GC/C		0.19	0.25					0.41		0.23			0.31	
CH_3I	GC/C		0.0002	0.0002					0.0012		0.0001			0.0004	
Ethane	GC/C		2.75	2.00					3.98		1.66			1.83	
Ethene	GC/C		5.66	6.23					3.41		8.91			9.35	
Ethene	AFTIR	16.24	10.35	12.14	12.17	17.26				16.82	19.48	26.06	24.89		14.84
Propane	GC/C		0.50	0.33					0.73		0.24			0.25	
Propene	GC/C		1.66	1.44					0.56		1.53			1.72	
Acetylene	GC/C		2.13	2.70					2.76		4.19			3.85	
Acetylene	AFTIR	3.55	2.84	4.31	4.06					4.26	6.12	8.69	7.82		
i-Butane	GC/C		0.03	0.02					0.02		0.01			0.01	
n-Butane	GC/C		0.11	0.07					0.11		0.05			0.06	
t-2-Butene	GC/C		0.85	0.41							0.04			0.05	
1-Butene	GC/C		0.25	0.21					0.08		0.22			0.25	
c-2-Butene	GC/C		0.83	0.40							0.03			0.04	
i-Pentane	GC/C		0.006	0.005							0.007			0.008	
n-Pentane	GC/C		0.03	0.02					0.03		0.02			0.02	
1,3-Butadiene	GC/C		1.06	0.63							0.32			0.34	
3-Methyl-1-butene	GC/C		0.79	0.37							0.02			0.02	
t-2-Pentene	GC/C		0.79	0.37							0.01			0.02	
2-Methyl-2-butene	GC/C		0.79	0.37							0.01			0.01	
2-Methyl-1-butene	GC/C		0.80	0.37							0.01			0.02	
c-2-pentene	GC/C		0.78	0.36							0.01			0.01	
n-Hexane	GC/C		0.05	0.04					0.02		0.06			0.05	
Isoprene	GC/C		0.35	0.25					1.13		0.32			0.39	
2-Methyl-1-pentene	GC/C		0.78	0.36											
Heptane	GC/C		0.02	0.02							0.03			0.02	
Benzene	GC/C		0.64	0.65					0.66		0.76			0.77	
Toluene	GC/C		0.49	0.47					0.34		0.55			0.41	
Formaldehyde	AFTIR	14.7	10.5	10.2	7.8	12.0				6.2	19.1	6.2	14.6		
Methanol	AFTIR	13.7	15.0	12.1	11.6	13.9				10.4	10.0	9.1	8.7		17.5
Acetic acid	AFTIR	22.8	17.0	13.9	8.4	23.9	63.6			18.2	18.1	10.9	10.0		
Formic acid	AFTIR		3.7	5.3	6.8	4.6				7.3	10.3	8.5	8.9		13.5

Table 2. (continued)

Species	Measurement Technique[b]	Sample 1	2	3	4	5	6	7	8	a	b	c	d	e	f
Total particulates	F/GB		172.6					438.8			96.1				
Organic carbon	F/GB		41.0					59.2			14.5				
Black carbon	F/GB		6.3					7.0			5.9				
Chloride	F/GB		3.5					5.1			0.4				
Nitrate	F/GB		2.6					7.6			2.0				
Sulfate	F/GB		1.8					5.6			2.8				
Potassium	F/GB		5.1					3.0			3.0				
CN[c]	TSI 3025A	8.2×10^5	1.2×10^6	4.4×10^5	8.3×10^4	1.3×10^5	6.5×10^4	2.3×10^4	1.9×10^5	1.9×10^5	2.0×10^5	1.7×10^5	3.0×10^5	3.2×10^5	3.3×10^5

[a]Trace gas ratios are given in ppbv $ppmv^{-1}$ of CO; particle species ratios are given in $\mu g\ m^{-3}\ ppmv^{-1}$ of CO.
[b]Abbreviations are GC/C, gas chromatography via canisters; AFTIR, airborne Fourier transform infrared spectroscopy; and F/GB, filters via grab bag.
[c]Units are number $ppmv^{-1}$ of CO.

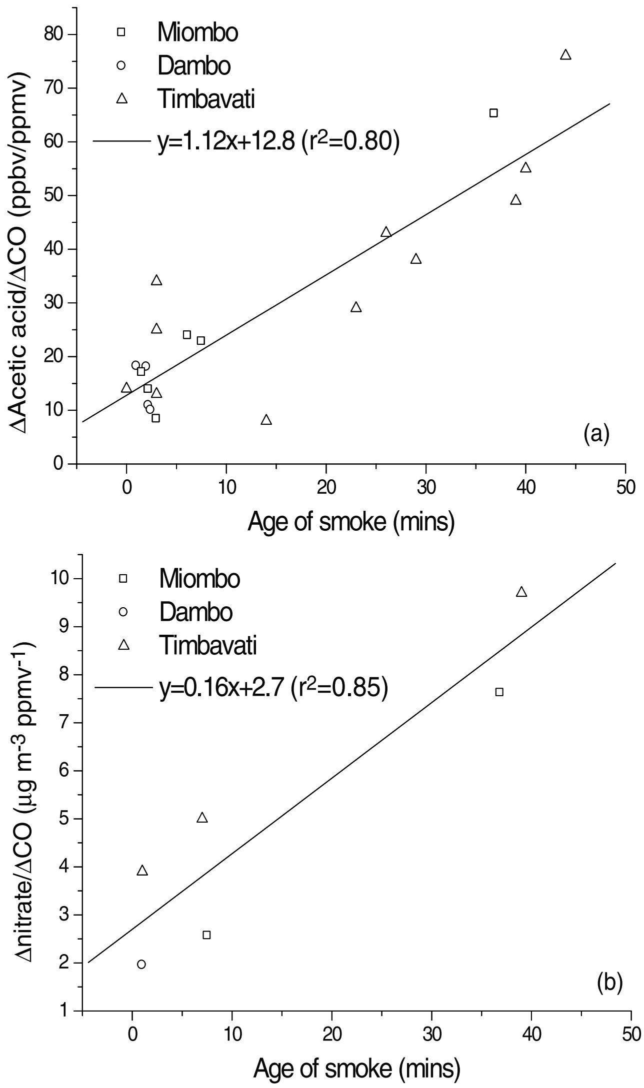

Figure 4. Excess mixing ratios of (a) gaseous acetic acid and (b) particulate nitrate normalized with respect to CO as a function of age of the smoke from the miombo woodland, dambo grassland, and the Timbavati fires.

[30] In Figure 6 the fuel consumption values in Table 3 are summed and plotted by month for the dry season of 2000. These results suggest that the biomass combusted in woodland and grassland savanna fires in southern Africa in 2000 peaked in July and September at a value of ~90 Tg. The July peak was primarily due to burning in Angola and the Democratic Republic of Congo, whereas the September peak was largely attributable to burning in Angola, Mozambique, and Zambia (Table 3). *Eck et al.* [2003] report monthly means of aerosol optical depth (at a wavelength of 500 nm) from 1995 to 2000 at Mongu, Zambia. This study showed that as the dry season progresses, aerosol optical depth increases and peaks at a value of ~0.65 in September, the month of highest biomass combustion in Zambia in 2000 (18,372 Gg, Table 3). We also note that *Swap et al.* [2003] proposed that biomass combustion in southern Africa during the 2000 dry season was above average because of above-average rainfall in the preceding wet season. Above-average rainfall would enhance fuel loading, but changes in com-

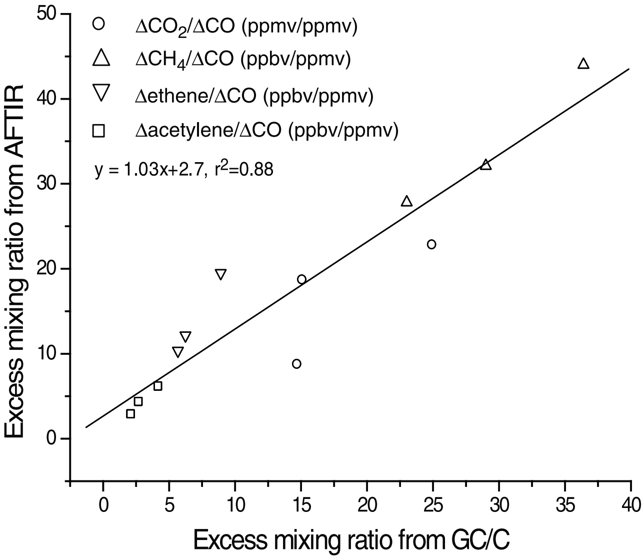

Figure 5. Comparisons of excess mixing ratios of CO_2, CH_4, ethene, and acetylene, normalized with respect to CO, for the dambo grassland and miombo woodland fire plumes determined from gas chromatography measurements on canister samples (GC/C) and from airborne Fourier transform infrared spectrometer measurements (AFTIR).

bustion factors and burned area would also have to be considered to determine interannual variability in biomass combustion. A study of interannual variability of biomass burning emissions constrained by satellite observations [*Duncan et al.*, 2003] found that emissions of CO from biomass burning in southern Africa in 2000 were not above average with respect to the 1979–2000 study period.

[31] In Table 4 we give estimates of emissions from all woodland and grassland savanna fires in southern Africa from May–October of 2000, the bulk of the southern African dry season. These estimates can be compared with the average annual global emissions from savanna burning worldwide estimated by *Andreae and Merlet* [2001] (Table 4) to obtain rough comparisons of the contributions of emissions from woodland and grassland savanna fires in the southern African dry season to the annual emissions from savanna burning worldwide. Andreae and Merlet estimated emissions assuming that a total of 3160 Tg of biomass is burned annually in savanna fires worldwide. However, the uncertainty in this total was large enough to make a defensible error analysis impossible. In Table 3 we estimate that ~367 Tg of biomass were burned in woodland and grassland savanna fires in southern Africa during the dry season (May–October) of 2000. This is within the (large) range of previous estimates for this region (90–2719 Tg [*Scholes et al.*, 1996]) and would account for ~12% of the biomass that *Andreae and Merlet* [2001] estimated is burned annually by savanna fires worldwide.

[32] Comparing species emissions from woodland and grassland savanna fires in southern Africa in the dry season of 2000 with those from global annual savanna burning [*Andreae and Merlet*, 2001], we find that the woodland and grassland savanna fires in southern Africa accounted for ~12.3%, 12.6%, 5.9%, 10.3%, 4.0%, and 7.5% of the annual emissions of CO_2, CO, hydrocarbons (CH_4 + NMHC), NO_x (as NO), NH_3, and SO_2, respectively, from all types of savanna fires worldwide. For total particulate matter, black carbon, organic carbon, and potassium the corresponding percentages are ~17.5%, 11.1%, 9.9%, and 12.2%, respectively. For oxygenated species, such as formaldehyde, acetic acid, formic acid, and methanol, the per-

Table 3. Biomass Combusted by Woodland and Grassland Savanna Fires in Southern Africa During the Dry Season of 2000[a]

Country	Ecosystem	May	June	July	August	September	October	May–October
Angola	grassland	89	1,260	2,015	1,182	594	93	5,232
Angola	woodland	309	12,904	34,092	28165	18,182	3,460	97,112
Botswana	grassland	24	174	10	29	104	205	546
Botswana	woodland	89	1,878	171	733	3,334	8,052	14,257
Congo[b]	grassland	671	2,314	2,244	621	93	12	5,956
Congo[b]	woodland	1,581	16,188	25,935	10,098	1,953	311	56,066
Lesotho	grassland	14	15	10	32	13	1	86
Lesotho	woodland	11	36	40	177	91	6	362
Malawi	grassland	2	2	34	15	43	16	111
Malawi	woodland	5	13	458	279	1,047	460	2,263
Mozambique	grassland	2	6	145	301	294	181	928
Mozambique	woodland	12	125	4,975	14,530	18,252	13,743	51,637
Namibia	grassland	45	17	18	38	120	34	272
Namibia	woodland	309	344	613	1,788	7,326	2,547	12,927
South Africa	grassland	189	1,275	709	753	1,471	332	4,729
South Africa	woodland	118	2,356	2,164	3,236	8,126	2,241	18,242
Swaziland	grassland	<1	<1	8	2	6	<1	17
Swaziland	woodland	<1	2	59	26	88	3	178
Tanzania	grassland	166	983	834	493	348	366	3,189
Tanzania	woodland	342	5,989	8,390	6,982	6,328	8,139	36,170
Zambia	grassland	39	259	561	294	609	222	1,985
Zambia	woodland	134	2,612	9,344	6,899	18,372	8,181	45,542
Zimbabwe	grassland	2	3	29	71	220	136	461
Zimbabwe	woodland	4	19	306	1,050	4,159	3,131	8,668

[a]See section 5.5 for details of how the quantity of biomass combusted was derived in this study. "Southern Africa" is defined as the African continent below 4°S, excluding Madagascar. Values are given in Gg.
[b]Democratic Republic of Congo.

Table 4. Emission Factors for 49 Trace Gas and Particle Species for Miombo Woodland and Dambo Grassland Fires in Zambia and Emissions of These Species for Woodland and Grassland Savanna Fires in Zambia and Southern Africa[a]

Species	Emission Factor for the Miombo Fire[b]	Emission Factor for the Dambo Fire[b]	Emissions From Woodland Savanna Fires in Southern Africa May–October 2000	Emissions From Grassland Savanna Fires in Southern Africa May–October 2000	Annual Emissions From Savanna Fires Worldwide[c]	Emissions From Woodland and Grassland Savanna Fires in Zambia May–October 2000	Annual Emissions From Use of Biofuels in Zambia[d]
CO_2	1,705	1,759	585,500	41,400	5,096,000	81,100	13,100
CO	73	42	25,100	988	206,000	3,410	970
CH_4	1.4	0.5	481	12	7,400	65	109
NMHC	1.7	1.1	584	26	10,700	80	
NO_x (as NO)	3.5	2.4	1,202	56	12,200	164	14
NH_3	0.4		137		3,400	18	10
HCN	0.37	0.31	127	7.3	90	17	
$MeONO_2$	0.00028	0.00023	0.096	0.005		0.01	
SO_2	0.22	0.3	75.6	7.1	1,100	10.6	
DMS	0.00049	0.00016	0.17	0.004		0.02	
CH_3Br	0.00048	0.0001	0.16	0.002	6	0.02	
CH_3Cl	0.029	0.02	9.96	0.47	240	1.36	
CH_3I	0.000084	0.000055	0.03	0.001	6	0.004	
Ethane	0.19	0.078	65.3	1.8		8.8	20
Ethene	0.43	0.38	148	8.9		20.3	19
Propane	0.048	0.016	16.5	0.38		2.2	
Propene	0.17	0.1	58.4	2.4		7.9	10
Acetylene	0.16	0.16	54.9	3.8		7.6	11.4
i-Butane	0.004	0.001	1.37	0.02		0.18	
n-Butane	0.013	0.0048	4.46	0.11		0.60	
t-2-Butene	0.01	0.0038	3.43	0.09		0.46	
1-Butene	0.034	0.019	11.67	0.45		1.59	
c-2-Butene	0.0072	0.0028	2.47	0.07		0.33	
i-Pentane	0.0011	0.00081	0.38	0.02		0.05	
n-Pentane	0.0041	0.0019	1.41	0.04		0.19	
1,3-Butadiene	0.039	0.027	13.4	0.63		1.83	
3-Methyl-1-butene	0.0038	0.0022	1.31	0.05		0.18	
t-2-Pentene	0.0033	0.0015	1.13	0.04		0.15	
2-Methyl-2-butene	0.004	0.0013	1.37	0.03		0.18	
2-Methyl-1-butene	0.0044	0.0016	1.51	0.04		0.20	
c-2-Pentene	0.0019	0.00094	0.65	0.02		0.09	
n-Hexane	0.0101	0.0071	3.47	0.17		0.47	
Isoprene	0.053	0.036	18.2	0.85		2.49	
2-Methyl-1-pentene	0.0028	0.0003	0.96	0.01		0.13	
Heptane	0.0056	0.004	1.92	0.09		0.26	
Benzene	0.13	0.089	44.6	2.1		6.1	
Toluene	0.15	0.11	51.5	2.6		7.0	
Formaldehyde	0.75	0.39	258	9.2	1,100	35	26
Methanol	1	0.32	343	7.5	3,800	46	57
Acetic acid	2.2	0.8	756	18.8	4,200	102	78
Formic acid	0.57	0.43	196	10.1	2,100	27	5
Total particulate matter	13	4.8	4,464	113	26,200	602	
Total particulate carbon	3.5	0.94	1,202	22	11,700	161	
Organic carbon	3	0.7	1,030	16.5	10,600	138	
Black carbon	0.47	0.24	161	5.6	1,500	22	
Chloride	0.26	0.0093	89.3	0.22		11.9	
Nitrate	0.19	0.05	65.3	1.2		8.8	
Sulfate	0.14	0.09	48.1	2.1		6.6	
Potassium	0.38	0.12	131	2.8	1,090	17.5	
CN[e]	5.9×10^{16}	1.7×10^{16}	2.0×10^{28}	4.0×10^{26}	1.1×10^{31}	2.7×10^{27}	

[a]Emission factors are given in g kg^{-1} fuel burned; emissions are given in Gg. See section 5.5 for discussion.
[b]Source is this paper.
[c]Source is *Andreae and Merlet* [2001].
[d]Source is *Bertschi et al.* [2003a].
[e]Units of CN emission factors and CN emissions are number of particles kg^{-1} fuel burned and number of particles, respectively.

centages are ~24.2%, 18.4%, 9.8%, and 9.2%, respectively. Percentages for the methyl halides CH_3Br, CH_3Cl, and CH_3I are ~2.8%, 4.3%, and 0.5%, respectively.

[33] In Table 4 our estimate of HCN emissions from woodland and grassland savanna fires in southern Africa (~134 Gg) exceeds *Andreae and Merlet*'s [2001] estimate of 90 Gg from global annual savanna burning. However, Andreae and Merlet calculated global annual emissions of HCN from savanna burning using an HCN emission factor of 0.025–0.031 g kg^{-1} of fuel burned. If the global annual emissions of HCN from savanna burning are recalculated using the HCN emission factor derived from our measure-

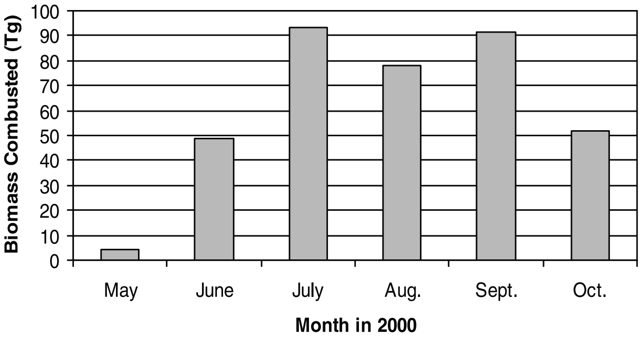

Figure 6. Monthly biomass combusted by woodland and grassland savanna fires in southern Africa during the 2000 dry season.

ments (0.31–0.37 g kg^{-1} fuel burned), the total HCN emitted by global annual savanna fires is ~1.1 Tg. In a study of global sources and sinks of HCN, *Li et al.* [2000] estimated an annual source of 2.7–5.6 Tg from all types of biomass burning worldwide.

[34] In Table 4 we give estimates of emissions from woodland and grassland savanna fires in Zambia alone from May–October of 2000. *Bertschi et al.* [2003a] estimated annual savanna fire emissions from Zambia using emission factors from *Yokelson et al.* [2003] and model estimates of annual biomass burned from *Hao and Liu* [1994]. *Bertschi et al.* [2003a] also estimated annual emissions due to the use of biofuels in Zambia; they found that biofuel emissions are comparable to or exceed those from savanna burning for a number of species, including CH_4, CO, ethane, acetylene, acetic acid, formaldehyde, methanol, and NH_3. Annual biofuel emissions from Zambia reported by Bertschi et al. are shown in Table 4. When we compare them with our estimates of dry season emissions from woodland and grassland savanna fires in Zambia (Table 4), we find that the annual emissions of CH_4, ethane, ethene, acetylene, propene, formaldehyde, methanol, and acetic acid from biofuels are comparable to or exceed dry season emissions from woodland and grassland savanna fires in Zambia.

[35] A similar comparison can be made for the southern African region. Averaging two recent estimates of annual biofuel use in southern Africa [*Yevich and Logan*, 2003; *Food and Agriculture Organization*, 1999], we estimate that each year ~108 Tg of wood fuel is combusted along with ~4 Tg of charcoal. Combining these estimates with biofuel emission factors measured by *Bertschi et al.* [2003a], we estimate annual biofuel emissions (in Gg) in southern Africa of CO_2, CO, CH_4, NO_x, ethane, ethene, acetylene, acetic acid, formic acid, formaldehyde, and methanol to be ~182,000, 12,300, 1360, 230, 214, 275, 182, 1020, 81, 398, and 569, respectively. The fourth and fifth columns of Table 4 show emissions of these species from woodland and grassland savanna fires, respectively, during the dry season of 2000 in southern Africa. Nearly three times as much CH_4 is produced by annual biofuel use in southern Africa than by dry season woodland and grassland savanna fires in southern Africa. The relative importance of biofuel emissions of the other species is smaller although still substantial.

[36] Our emission estimates (Table 4) are subject to considerable uncertainty, most of which is due to uncertainty in the total area burned by woodland and grassland savanna fires in southern Africa during the 2000 dry season. In addition to the SPOT satellite burned-area retrievals used in this paper, preliminary burned area retrievals have been derived from measurements made by the Moderate-Resolution Imaging Spectroradiometer (MODIS) on the Terra satellite. In a comparison for September 2000 the burned area derived from MODIS measurements for southern Africa by S. Korontzi et al. (Modeling and sensitivity analysis of fire emissions in southern Africa during SAFARI 2000, submitted to *Remote Sensing of Environment*, 2003) is 55% larger than the SPOT burned area used in the present paper. The values used in the present paper for the available aboveground biomass fuel for miombo woodland and dambo grassland plots are uncertain to ~30%. The combustion factors used in the present paper are uncertain to ~30%, and the emission factors are uncertain to ~10%. Therefore the emission estimates in Table 4 for woodland and grassland savanna fires in southern Africa during the 2000 dry season are uncertain to ~70%.

[37] An additional source of uncertainty is emissions from residual smoldering combustion (RSC), which is biomass combustion that produces smoke that is not lofted by strong fire-induced convection. Emissions from RSC are generally not measured from aircraft, and they are not included in the emission factors reported in this paper. Residual smoldering combustion can occur in miombo woodland fires because of the presence of dead/downed logs in the fuel complex. *Bertschi et al.* [2003b] made the first measurements of emission factors from RSC in a miombo woodland. They found that the inclusion of RSC emissions in estimating total emissions from miombo fires had little effect on many compounds but had a significant effect on some. Little data exist on the amount and composition of RSC smoke. Therefore we will not explore the topic further here except to note that some additional uncertainty due to RSC emissions may occur for some compounds, particularly methane and methanol.

6. Conclusions

[38] In this paper we have described airborne measurements of the emissions and initial evolution of trace gases and particles from a miombo woodland and a dambo grassland fire in Zambia. The principal conclusions from analyses of these measurements are as follows.

[39] 1. Woodland and grassland savanna fires in southern Africa during the 2000 dry season were estimated to account for ~12.3%, 12.6%, 5.9%, 10.3%, 4.0%, and 7.5% of annual emissions of CO_2, CO, total hydrocarbons, NO_x (as NO), NH_3, and SO_2, respectively, from all types of savanna fires worldwide. For total particulate matter, black carbon, organic carbon, and potassium the corresponding percentages are ~17.5%, 11.1%, 9.9%, and 12.2%, respectively. For oxygenated species such as formaldehyde, acetic acid, formic acid, and methanol the percentages are ~24.2%, 18.4%, 9.8%, and 9.2%, respectively. For CH_3Br, CH_3Cl, and CH_3I they are ~2.8%, 4.3%, and 0.5%, respectively. Our emissions estimates for woodland and

grassland savanna fires during the 2000 dry season in southern Africa are uncertain to ~70%.

[40] 2. Average annual emissions of CH_4, ethane, ethene, acetylene, propene, formaldehyde, methanol, and acetic acid from the use of biofuels in Zambia are comparable to or exceed dry season emissions from miombo woodland and dambo grassland fires in Zambia. Average annual emissions of methane from the use of biofuels in southern Africa are nearly 3 times higher than those from woodland and grassland savanna fires in southern Africa during the dry season of 2000.

[41] **Acknowledgments.** We thank all members of the UW-CARG and the pilots of the UW Convair-580 for their valuable help in obtaining measurements. We thank Dan Jaffe for help in calibrating the gas instruments. This study was carried out as part of the SAFARI 2000 Southern African Regional Science Initiative. Research support from grants NAG5-9022 and NAG5-7675 from NASA's Radiation Science Program and grants ATM-9901624 and ATM-9900494 from NSF's Division of Atmospheric Sciences is gratefully acknowledged.

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D. R. Blake, Department of Chemistry, 516 Rowland Hall, University of California, Irvine, Irvine, CA 92697-2025, USA. (drblake@uci.edu)

S. Gao, Department of Environmental Science and Engineering, California Institute of Technology, Pasadena, CA 91125, USA. (sgao@its.caltech.edu)

P. V. Hobbs and P. Sinha, Department of Atmospheric Sciences, University of Washington, Box 351640, Seattle, WA 98195-1640, USA. (phobbs@atmos.washington.edu; psinha@atmos.washington.edu)

T. W. Kirchstetter, Environmental Energy Technologies Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA. (twkirchstetter@lbl.gov)

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