# Emission of trace gases and aerosols from biomass burning

M. O. Andreae and P. Merlet

Biogeochemistry Department, Max Planck Institute for Chemistry, Mainz, Germany

**Abstract.** A large body of information on emissions from the various types of biomass burning has been accumulated over the past decade, to a large extent as a result of International Geosphere-Biosphere Programme/International Global Atmospheric Chemistry research activities. Yet this information has not been readily accessible to the atmospheric chemistry community because it was scattered over a large number of publications and reported in numerous different units and reference systems. We have critically evaluated the presently available data and integrated these into a consistent format. On the basis of this analysis we present a set of emission factors for a large variety of species emitted from biomass fires. Where data were not available, we have proposed estimates based on appropriate extrapolation techniques. We have derived global estimates of pyrogenic emissions for important species emitted by the various types of biomass burning and compared our estimates with results from inverse modeling studies.

## 1. Introduction

Human evolution and the use of fire have gone hand in hand ever since the origin of our species in the savannas and woodlands of Africa [*Schüle*, 1990]. As a result, air pollution from the smoke of biomass fires has been humanity's constant companion for some 2 million years, and its ancient impact on human health is reflected in soot deposits in the lungs of mummies. Emissions from fossil fuel burning gained notoriety as air pollutants in medieval England, but only in the last 2 centuries have they begun to play an important role worldwide. Yet the scientific exploration of air pollution has focused initially only on this much more recent threat, and the first pioneering papers on the impact of biomass burning on the chemistry of the atmosphere were only published in the 1970s and early 1980s [e.g., *Eagan et al.*, 1974; *Radke et al.*, 1978; *Crutzen et al.*, 1979]. Scientific interest in this topic grew when early estimates of pyrogenic emissions suggested that for some atmospheric pollutants biomass burning could rival fossil fuel use as a source of atmospheric pollution [*Seiler and Crutzen*, 1980; *Crutzen and Andreae*, 1990] and when it became evident that these emissions could affect large areas of the world as a consequence of long-range transport [*Andreae*, 1983; *Kirchhoff and Nobre*, 1986; *Reichle et al.*, 1986; *Fishman et al.*, 1990].

Satellite and airborne observations have shown elevated levels of $O_3$, CO, and other trace gases over vast areas of Africa, South America, the tropical Atlantic, and the Indian and Pacific Oceans [*Andreae et al.*, 1988; *Watson et al.*, 1990; *Fishman et al.*, 1991; *Andreae et al.*, 1994; *Cahoon et al.*, 1994; *Folkins et al.*, 1995; *Browell et al.*, 1996; *Connors et al.*, 1996; *Talbot et al.*, 1996; *Thompson et al.*, 1996; *Jonquières et al.*, 1998; *Novelli et al.*, 1998; *Blake et al.*, 1999; *Gregory et al.*, 1999; *Singh et al.*, 2000]. Smoke aerosols perturb regional, and probably global, radiation budgets by their light-scattering effects and by their influence on cloud microphysical processes [*Crutzen and Andreae*, 1990; *Penner et al.*, 1992; *Andreae*, 1995]. Following well-publicized large fire catastrophes in recent years and intensive scientific efforts over the last decade, the general public as well as the scientific community are now aware that emissions from biomass burning represent a large perturbation to global atmospheric chemistry, especially in the tropics.

To assess the atmospheric impact of biomass burning, and especially to represent it quantitatively in models of atmospheric transport and chemistry, accurate data on the emission of trace gases and aerosols from biomass fires are required. Emissions must typically be represented in the form of spatiotemporally resolved fields, where the emission per unit area and time is provided at a specified spatial and temporal resolution. These fields are obtained by multiplying an exposure term, for example, the amount of biomass burned within a grid cell during a time interval, with an emission factor, that is, the amount of the chemical species released per mass of biomass burned.

In the last couple of decades the efforts of individual groups to characterize fire emissions have been greatly extended by a number of coordinated biomass burning experiments in various ecosystems throughout the world, often under the auspices of the International Global Atmospheric Chemistry (IGAC) Project of the International Geosphere-Biosphere Programme (IGBP). Examples of these experiments are Dynamique et Chimie Atmosphérique en Forêt Equatoriale-Fire of Savannas (DECAFE-FOS) in West Africa [*Lacaux et al.*, 1995], Southern Africa Fire-Atmosphere Research Initiative (SAFARI 92) [*Andreae et al.*, 1996c; *Lindesay et al.*, 1996], Transport and Atmospheric Chemistry Near the Equator–Atlantic (TRACE-A) over Brazil, southern Africa, and the South Atlantic [*Fishman et al.*, 1996], Experiment for Regional Sources and Sinks of Oxidants in central Africa (EXPRESSO) [*Delmas et al.*, 1999], Smoke, Clouds, and Radiation-Brazil (SCAR-B) in Brazil [*Kaufman et al.*, 1998], and Fire Research Campaign Asia-North (FIRESCAN) in central Siberia [*FIRESCAN Science Team*, 1996]. These coordinated studies and numerous independent smaller investigations have resulted in a large body of information on the emission characteristics of biomass fires. These data describe qualitatively and quantitatively the pyrogenic emission of chemically and radiatively important gases and aerosol species, but, unfortunately, this information is scattered through the scientific literature and presented in a large variety of formats and units, making its use very difficult. An earlier synthesis of then available data was provided by *Delmas et al.* [1995], but it covers only a minor fraction of the information that has accumulated in recent years.

The objective of this paper is to synthesize the currently available data on fire emission characteristics for a large number of chemical species into a consistent set of units. In contrast to some previous summaries that gave only generic estimates independent of the type of fire [e.g., *Andreae*, 1993], here we provide separate emission factors for the different types of biomass burning, such as deforestation fires in the tropics, savanna fires, etc. We then combine the emission factor data with exposure estimates for the various fire categories to provide global estimates of emissions of biomass burning.

## 2. Methods

### 2.1. Definitions

Emission information is represented in two basic forms: emission ratios or emission factors. These parameters relate the emission of a particular species of interest to that of a reference species, such as $CO_2$ or $CO$ (emission ratio) or to the amount of fuel burned (emission factor); their definition and use will be discussed below.

Emission ratios are obtained by dividing the excess trace species concentrations measured in a fire plume by the excess concentration of a simultaneously measured reference gas, for example, $CO_2$ or $CO$. To obtain "excess" concentrations, the ambient background concentrations must be subtracted from the values measured in the smoke. For example, the emission ratio of methyl chloride, $CH_3Cl$, relative to $CO$ is

$$ER_{CH_3Cl/CO} = \frac{\Delta CH_3Cl}{\Delta CO} = \frac{(CH_3Cl)_{smoke} - (CH_3Cl)_{ambient}}{(CO)_{smoke} - (CO)_{ambient}}$$

Alternatively, the emission ratio can be determined as the regression slope of the species concentration versus the reference species. The various techniques for these calculations and the associated errors are discussed by *Le Canut et al.* [1996]. For gases, emission ratios are usually expressed in terms of molar ratios. For aerosols, emission ratios are most often given in units of mass aerosol per kilogram $CO_2$-carbon (g kg$^{-1}$ C($CO_2$)). The most commonly used reference species are $CO_2$ and $CO$, but other compounds such as $CH_4$ and acetylene ($C_2H_2$) have also been used.

Besides practical considerations, for example, the availability of a particular analyzer in a given study, the selection of $CO$ or $CO_2$ as reference gas is determined by the ultimate objective of the analysis and on the combustion type (flaming or smoldering) from which the species is preferentially released. $CO$ is emitted predominantly by smoldering combustion and is therefore a suitable reference gas for other species emitted mostly during smoldering. Close correlations between smoldering-derived gases and $CO$ can usually be observed, which allows fairly accurate estimation of trace gas emissions from fires for which the $CO$ emission is known. In contrast, correlations of smoldering-derived gases with $CO_2$ tend to be relatively poor, since the relative proportion of flaming versus smoldering combustion in different fires, or even in different parts of the same fire, results in variable trace gas to $CO_2$ ratios [*Lobert et al.*, 1991]. For smoldering-related species containing nitrogen or halogen elements, for example, $NH_3$ or the methyl halides, the emission ratio relative to $CO$ is also dependent on fuel composition, i.e., the nitrogen or halogen element content of the fuel.

Flaming-derived species, however, correlate well with $CO_2$, which makes this the obvious reference gas for these compounds. Because most of the biomass carbon is released from a fire as $CO_2$, the emission ratio of a gas relative to $CO_2$ permits the estimation of trace gas emission from fires based on the amount of biomass burned. Therefore this ratio is the most suitable parameter for regional or global estimations, and consequently, even smoldering-

related species are often related to $CO_2$ as a reference species. This can be done either by calculating an emission ratio directly from the regression slope of the species concentration to that of $CO_2$ or by first deriving an emission ratio to $CO$ and multiplying that with the average $CO/CO_2$ emission ratio for the fire.

Emission ratios have the advantage that they only require simultaneous measurements of the species of interest and the reference species in the smoke and appropriate measurements in the background air. They do not require knowledge of the fuel composition and the amounts of fuel burned and are therefore suitable for field studies, particularly airborne plume measurements where fuel information is usually not available.

In principle, it is possible to obtain an emission ratio from a single pair of plume and background measurements. In practice, however, several measurements are usually averaged, or information from continuous analyzers is used in order to derive data representative of a fire or fire phase. Since the emission ratio of smoldering-related species to $CO_2$ or the ratio of flaming-related species to $CO$ can vary widely through the course of a fire, it is essential that this averaging is done correctly. When the emission ratio for an entire fire is sought, two approaches are possible: (1) The entire amounts of trace and reference species emitted during the fire can be integrated and divided by one another, or (2) an average of instantaneous emission ratios can be calculated, in which the individual measurements are weighted according to the amount of fuel combusted in the time interval represented by the measurement. These approaches require information that is only available in experimental fires in the laboratory or to a limited extent in the field. For aircraft measurements one has to rely on the mixing processes in a plume to perform this averaging. This requires that both flaming and smoldering emissions are entrained into the plume and that the plume is sampled in a representative fashion. Ground-based sampling has a tendency to overrepresent smoldering emissions which tend to be emitted during less vigorous phases of a fire and therefore to remain closer to the ground, while airborne sampling may have a bias toward emissions from the flaming phase that rise to higher altitudes [*Andreae et al.*, 1996a]. For the calculation of average emission factors in Table 1 we have assumed that airborne measurements represent an unbiased average of the emissions. When smoldering and flaming emissions were given separately in ground-based studies, we have combined them to represent the complete fire. For this purpose we either used data on the fractions of fuel combusted in the smoldering and flaming stages provided in a given study or, when this information was not available, typical values from other studies on the same type of fire.

Another parameter frequently used to characterize emissions from fires is the emission factor, which is defined as the amount of a compound released ($M_x$) per amount of dry fuel consumed ($M_{biomass}$), expressed in units of g kg$^{-1}$. Calculation of this parameter requires knowledge of the carbon content of the biomass burned and the carbon budget of the fire; both parameters are difficult to establish in the field as opposed to laboratory experiments where they are readily determined. The total carbon released is usually estimated by adding the measured concentrations of $CO_2$, $CO$, hydrocarbons, and particulate carbon, when this information is available. The emission factor can then be expressed as

$$EF_x = \frac{M_x}{M_{biomass}} = \frac{M_x}{M_C} [C]_{biomass}$$

$$EF_x \cong \frac{[x]}{\sum([C_{CO_2}] + [C_{CO}] + [C_{CH_4}] + [C_{VOC}] + [C_{aeros}] + \ldots)} [C]_{biomass},$$

where $M_C$ is the mass of carbon emitted, $[C]_{biomass}$ is the carbon concentration in the biomass burned, $[x]$ is the concentration of

species $x$ in the smoke, and [$C_{CO2}$] etc. are the concentrations of the various carbon species in the smoke. Alternatively, the carbon budget of the fire can be estimated by multiplying the $CO_2$ in the smoke with an assumed factor to represent non-$CO_2$ carbon emissions (i.e., CO, hydrocarbons, and aerosol carbon). Where fuel and residue data at the ground are not available, a fuel carbon content of 45% is usually assumed in order to derive emission factors from emission ratios.

## 2.2. Conversion of Units

We adopted the use of emission factors, as defined in section 2.1, to express all emission data in this paper. The reason for this choice was that emission factors allow the most direct estimation of trace gas and aerosol release when the amount of biomass burned is given. This choice made it necessary to convert data given in other forms into emission factors. We proceeded as follows.

When emission factors were given following the same definition as specified in section 2.1, the data were used unchanged. Where emission factors were given relative to other fuel mass indicators (e.g., as mass species emitted per mass carbon burned or released), we multiplied the data with an appropriate conversion factor, typically, the carbon content of the fuel. When this information was not provided by the authors, we used a default carbon content of 45%.

To convert data given as molar emission ratios, we used the following equation:

$$EF_x = ER_{(X/Y)} \frac{MW_X}{MW_Y} EF_Y,$$

where $ER_{(X/Y)}$ is the emission ratio of species $x$ relative to the reference species $Y$, $MW_X$ and $MW_Y$ are the molecular weights of the species $x$ and the reference species $Y$, and $EF_Y$ is the emission factor of the reference species. If $EF_Y$ was provided in the data source, it was used directly; otherwise, the mean $EF_Y$ for the appropriate type of fire (forest, savanna, etc.) was applied.

## 3. Results and Discussion

### 3.1. Pyrogenic Emissions and the Combustion Process

The emission factors of the various smoke constituents are determined by the composition of the fuel and by the physical and chemical processes during combustion. Since the carbon content of fuels varies only over a fairly limited range (typically 37–54%), the emission factors for compounds that contain only carbon, oxygen, and hydrogen are predominantly a function of the combustion process. Charcoal burning is an exception to this rule because of the high carbon content of this fuel (>80%). The emission of substances containing minor elements, such as nitrogen, sulfur, and the halogens, is determined both by the concentrations of those elements in the fuel and by the combustion conditions.

The processes during the combustion of biomass have been described in detail [Lobert and Warnatz, 1993; Yokelson et al., 1996, 1997] and will be reviewed here only briefly. Combustion of the individual fuel elements proceeds through a sequence of stages (ignition, flaming plus glowing plus pyrolysis, glowing plus pyrolysis (smoldering), glowing, and extinction), each with different chemical processes that result in different emissions. Plant biomass consists of cellulose and hemicelluloses (typically 50–70% dry matter), lignin (15–35%), proteins, amino acids, and other metabolites, including volatile substances (alcohols, aldehydes, terpenes, etc.). In addition, it contains minerals (up to 10%) and water (up to 60%). Thermal degradation begins with a drying/distillation step, in which water and volatiles are released, followed by pyrolysis, during which thermal cracking of the fuel molecules occurs. This results in the formation of char (less volatile solids of high C content), tar (molecules of intermediate molecular weight), and volatile compounds in the form of a flammable white smoke. When temperatures in the fuel bed exceed 450 K, the process becomes exothermic, and at ~800 K, glowing combustion begins. At this point also, a complex mixture of tar and gas products are released, which, when diluted with air, form a flammable mixture. When this mixture ignites, flaming combustion occurs, which converts the complex mixture of relatively reduced substances emitted during pyrolysis to simple molecules, particularly $CO_2$, $H_2O$, NO, $N_2O$, $N_2$, and $SO_2$. Depending on the interaction between chemical kinetics and physical dynamics in the flame, intermediate products of flame radical chemistry, like CO, $CH_4$, $H_2$, $C_2H_4$, $C_2H_2$, PAH, and soot particles, are also released during this stage.

When most volatiles have been released from the near-surface region of the fuel, flaming combustion ceases, and smoldering begins. The energy-producing reaction in this phase is the gas-solid reaction between oxygen and carbon in the char layer at the fuel surface, yielding predominantly CO at temperatures usually <850 K. This lower-temperature process emits large amounts of incompletely oxidized pyrolysis products that are similar to the products of the initial solid phase decomposition [Lobert et al., 1991; Yokelson et al., 1997]. It is thus this mode of combustion that is responsible for the vast diversity of emission products that is shown in Table 1. The amount of substances emitted from a given fire and their relative proportions are thus determined to a large extent by the ratio of flaming to smoldering combustion. It is this proportion, often expressed as "combustion efficiency" (i.e., the ratio between $CO_2$ and the sum of all carbon species emitted), which has been used as an effective predictor for the emission of smoke gas composition from biomass fires [Ward et al., 1996].

Open vegetation fires are typically dynamic fires, in which a moving fire front passes through a fuel bed, such as a savanna or forest. Consequently, all combustion types are present at any given time, and their combined emissions are released into the smoke plume. Their proportions vary over time, typically dominated by flaming in the earlier part of the fire and smoldering during the later part. Especially in forest fires, this late smoldering stage can continue for days or even weeks. This temporal separation of combustion types may introduce a bias toward flaming combustion products into emission measurements, as these late emissions may not always be sampled, especially in airborne campaigns. Time course studies on boreal and tropical forest fires show, however, that the vast majority of emissions (>99%) take place within ~4–8 hours after ignition, so that this bias is likely to be minor [Ward and Hardy, 1991; Ward et al., 1992].

It may seem desirable to provide separate emission factors for flaming and smoldering combustion in Table 1, but this is not possible given the data available. Airborne measurements sample an integrated mixture of the emissions from both combustion types, and even at the ground the separation is incomplete because both processes occur simultaneously in a given patch at most times. This is not a serious problem, however, since each of the types of vegetation fires differentiated in Table 1 has a characteristic fuel composition and structure and typical ways in which these fires occur naturally or are managed by people; they tend to have characteristic ratios of flaming and smoldering combustion, which to a large extent determine their pattern of emission factors. In savanna fires, for example, flaming combustion dominates, and the emission factors for reduced species are fairly low. Charcoal making, however, is almost exclusively a smoldering and glowing process. Even in a given fire type, however, the proportion of flaming combustion can vary considerably as a function of internal parameters, particularly fuel moisture and structure, and external parameters, such as the movement of the flame front relative to wind direction or terrain slope. As a result, the emission pattern

**Table 1.** Emission Factors for Pyrogenic Species Emitted From Various Types of Biomass Burning[a]

| Species | Savanna and Grassland[b] | Tropical Forest[c] | Extratropical Forest[d] | Biofuel Burning[e] | Charcoal Making[f] | Charcoal Burning[f] | Agricultural Residues[i] |
|---|---|---|---|---|---|---|---|
| $CO_2$ | 1613 ± 95 | 1580 ± 90 | 1569 ± 131 | 1550 ± 95 | 440 | 2611 ± 241 | 1515 ± 177 |
| CO | 65 ± 20 | 104 ± 20 | 107 ± 37 | 78 ± 31 | 70 | 200 ± 38 | 92 ± 84 |
| $CH_4$ | 2.3 ± 0.9 | 6.8 ± 2.0 | 4.7 ± 1.9 | 6.1 ± 2.2 | 10.7 | 6.2 ± 3.3 | 2.7 |
| Total nonmethane hydrocarbons | 3.4 ± 1.0 | 8.1 ± 3.0 | 5.7 ± 4.6 | 7.3 ± 4.7 | 2.0 | 2.7 ± 1.9 | (7.0)[h] |
| $C_2H_2$ | 0.29 ± 0.27 | 0.21–0.59 | 0.27 ± 0.09 | 0.51–0.90 | 0.04 | 0.05–0.13 | (0.36)[h] |
| $C_2H_4$ | 0.79 ± 0.56 | 1.0–2.9 | 1.12 ± 0.55 | 1.8 ± 0.6 | 0.10 | 0.46 ± 0.33 | (1.4)[h] |
| $C_2H_6$ | 0.32 ± 0.16 | 0.5–1.9 | 0.60 ± 0.15 | 1.2 ± 0.6 | 0.10 | 0.53 ± 0.48 | (0.97)[h] |
| $C_3H_4$ | 0.022 ± 0.014 | 0.013 | 0.04–0.06 | (0.024)[b] | – | (0.06)[h] | (0.032)[h] |
| $C_3H_6$ | 0.26 ± 0.14 | 0.55 | 0.59 ± 0.16 | 0.5–1.9 | 0.06 | 0.13–0.56 | (1.0)[h] |
| $C_3H_8$ | 0.09 ± 0.03 | 0.15 | 0.25 ± 0.11 | 0.2–0.8 | 0.04 | 0.07–0.30 | (0.52)[h] |
| 1-butene | 0.09 ± 0.06 | 0.13 | 0.09–0.16 | 0.1–0.5 | – | 0.02–0.20 | (0.13)[h] |
| i-butene | 0.030 ± 0.012 | 0.11 | 0.05–0.11 | 0.1–0.5 | – | 0.01–0.16 | (0.08)[h] |
| trans-2-butene | 0.024 ± 0.014 | 0.05 | 0.01–0.05 | 0.05–0.3 | – | 0.01–0.06 | (0.04)[h] |
| cis-2-butene | 0.021 ± 0.011 | 0.042 | 0.008–0.13 | 0.05–0.18 | – | 0.01–0.03 | (0.05)[h] |
| Butadiene | 0.07 ± 0.05 | | 0.06–0.08 | 0.11–0.36 | – | 0.01–0.10 | (0.09)[h] |
| n-butane | 0.019 ± 0.09 | 0.041 | 0.069 ± 0.038 | 0.03–0.13 | – | 0.02–0.10 | (0.06)[h] |
| i-butane | 0.006 ± 0.003 | 0.015 | 0.022 ± 0.009 | 0.01–0.05 | – | 0.006–0.01 | (0.015)[h] |
| 1-pentene | 0.022 ± 0.010 | 0.056 | 0.04–0.07 | 0.5 | – | 0.028 | 0.008 |
| n-pentane | 0.005 ± 0.004 | 0.014 | 0.05–0.06 | 0.07 | – | 0.10 | (0.025)[h] |
| 2-methyl-butenes | 0.008 ± 0.004 | 0.074 | 0.033 | 0.16 | – | 0.015 | 0.007 |
| 2-methyl-butane | 0.011 ± 0.012 | 0.008 | 0.026–0.029 | 0.08 | – | 0.07 | (0.018)[h] |
| Isoprene | 0.020 ± 0.012 | 0.016 | 0.10 | 0.15–0.42 | – | 0.017 | (0.05)[h] |
| Cyclopentene | 0.012 ± 0.008 | (0.02)[b] | 0.019 | 0.61 | – | 0.035 | (0.02)[h] |
| 4-methyl-1-pentene | 0.048 | 0.048 | (0.05)[h] | 0.015 | – | (0.09)[h] | 0.016 |
| 1-hexene | 0.037 ± 0.016 | 0.063 | 0.07–0.11 | (0.05)[h] | – | (0.13)[h] | 0.013 |
| n-hexane | 0.039 ± 0.045 | (0.05)[h] | 0.03–0.06 | (0.04)[h] | – | 0.063 | (0.05)[h] |
| Isohexanes | 0.05 | (0.08)[h] | (0.08)[h] | (0.06)[h] | – | (0.15)[h] | (0.08)[h] |
| Heptane | 0.05 | (0.08)[h] | (0.08)[h] | (0.06)[h] | – | (0.15)[h] | (0.08)[h] |
| Octenes | 0.003–0.008 | 0.012 | 0.005 | (0.007)[h] | – | (0.017)[h] | 0.004 |
| Terpenes | 0.015 | (0.15)[i] | 0.22 | (0.15)[i] | – | 0.0 | (0.015)[h] |
| Benzene | 0.23 ± 0.11 | 0.39–0.41 | 0.49 ± 0.08 | 1.9 ± 1.0 | – | 0.3–1.7 | 0.14 |
| Toluene | 0.13 ± 0.06 | 0.21–0.29 | 0.40 ± 0.10 | 1.1 ± 0.7 | – | 0.08–0.61 | 0.026 |
| Xylenes | 0.045 ± 0.025 | 0.04–0.08 | 0.20 | 0.55 ± 0.44 | – | 0.04–0.22 | 0.01 |
| Ethylbenzene | 0.013 ± 0.003 | 0.013–0.035 | 0.048 | 0.17 ± 0.12 | – | 0.01–0.07 | 0.03 |
| Styrene | 0.024 | (0.03)[h] | 0.13 | 0.04–0.5 | – | 0.03–0.22 | (0.03)[h] |
| PAH | 0.0024 | (0.025)[j] | (0.025)[j] | (0.025)[j] | – | (0.025)[j] | (0.025)[j] |
| Methanol | (1.3)[h] | (2.0)[b] | 2.0 ± 1.4 | (1.5)[h] | 0.16 | (3.8)[h] | (2.0)[h] |
| Ethanol | (0.011)[h] | (0.018)[h] | 0.018 | (0.013)[h] | – | (0.03)[h] | (0.018)[h] |
| 1-propanol | 0.025 | (0.04)[h] | (0.04)[h] | (0.03)[h] | – | (0.08)[h] | (0.04)[h] |
| Butanols | 0.008 | 0.009 | (0.011)[h] | (0.008)[h] | – | (0.02)[h] | 0.012 |
| Cyclopentanol | 0.032 | 0.031 | (0.04)[h] | (0.03)[h] | – | (0.08)[h] | 0.017 |
| Phenol | 0.003 | 0.006 | (0.005)[h] | (0.004)[h] | – | (0.01)[h] | 0.001 |
| Formaldehyde | 0.26–0.44 | (1.4)[b] | 2.2 ± 0.5 | 0.13 ± 0.05 | – | (2.6)[h] | (1.4)[b] |
| Acetaldehyde | 0.50 ± 0.39 | (0.65)[b] | 0.48–0.52 | 0.14 ± 0.05 | – | (1.2)[h] | (0.65)[b] |
| Acrolein, propenal | 0.08 | (0.18)[h] | 0.13–0.35 | 0.01–0.1 | – | (0.35)[h] | (0.18)[h] |
| Propanal | 0.009 | (0.08)[h] | 0.03–0.25 | 0.02–0.03 | – | (0.15)[h] | (0.08)[h] |
| Butanals | 0.053 | 0.071 | 0.21 | 0.04–0.05 | – | (0.20)[h] | 0.021 |
| Hexanals | 0.002–0.024 | 0.031 | 0.02 | 0.004–0.009 | – | (0.04)[h] | 0.012 |
| Heptanals | 0.003 | 0.003 | (0.004)[b] | (0.003)[h] | – | (0.008)[h] | 0.001 |
| Acetone | 0.25–0.62 | (0.62)[b] | 0.52–0.59 | 0.01–0.04 | 0.02 | (1.2)[h] | (0.63)[b] |
| 2-butanone | 0.26 | (0.43)[h] | 0.17–0.74 | 0.03–0.06 | – | (0.83)[h] | (0.44)[h] |
| 2,3-butanedione | (0.57)[h] | (0.92)[h] | 0.35–1.5 | (0.68)[h] | – | (1.8)[h] | (0.9)[h] |
| Pentanones | 0.01–0.02 | 0.028 | 0.09 | (0.04)[h] | – | (0.09)[h] | 0.007 |
| Heptanones | 0.006 | 0.002 | (0.005)[h] | (0.004)[h] | – | (0.01)[h] | 0.002 |
| Octanones | 0.015 | 0.019 | (0.02)[h] | (0.016)[h] | – | (0.04)[h] | (0.02)[h] |
| Benzaldehyde | 0.029 | 0.027 | (0.036)[h] | 0.02–0.03 | – | (0.07)[h] | 0.009 |
| Furan | 0.095 | (0.48)[h] | 0.40–0.45 | 0.65 | – | (0.9)[h] | (0.5)[h] |
| 2-methyl-furan | 0.044–0.048 | 0.17 | 0.47 | (0.18)[h] | – | (0.46)[h] | 0.012 |
| 3-methyl-furan | 0.006–0.011 | 0.029 | 0.05 | (0.023)[h] | – | (0.06)[h] | 0.003 |
| 2-ethylfuran | 0.001 | 0.003 | 0.006 | (0.003)[h] | – | (0.007)[h] | 0.001 |
| 2,4-dimethyl-furan | 0.008 | 0.024 | (0.019)[h] | (0.014)[h] | – | (0.04)[h] | 0.002 |
| 2,5-dimethyl-furan | 0.002 | (0.03)[h] | 0.05 | (0.021)[h] | – | (0.05)[h] | (0.03)[h] |
| Tetrahydrofuran | 0.016 | 0.016 | (0.02)[h] | (0.015)[h] | – | (0.04)[h] | 0.006 |
| 2,3-dihydrofuran | 0.012 | 0.013 | (0.017)[h] | (0.012)[h] | – | (0.031)[h] | 0.005 |
| Benzofuran | 0.014 | 0.015 | 0.026 | (0.016)[h] | – | (0.04)[h] | 0.004 |
| Furfural | (0.23)[h] | (0.37)[h] | 0.29–0.63 | 0.22 | 0.12 | (0.72)[h] | (0.37)[h] |
| Methyl formate | (0.015)[h] | (0.025)[h] | 0.025 | (0.018)[h] | – | (0.05)[h] | (0.025)[h] |

**Table 1.** (continued)

| Species | Savanna and Grassland[b] | Tropical Forest[c] | Extratropical Forest[d] | Biofuel Burning[e] | Charcoal Making[f] | Charcoal Burning[f] | Agricultural Residues[i] |
|---|---|---|---|---|---|---|---|
| Methyl acetate | 0.055 | (0.10)[h] | 0.09–0.12 | (0.07)[h] | – | (0.19)[h] | (0.10)[h] |
| Acetonitrile | 0.11 | (0.18)[i] | 0.19 | (0.18)[i] | – | (0.18)[h] | (0.18)[h] |
| Formic acid | (0.7)[h] | (1.1)[h] | 2.9 ± 2.4 | 0.13 | 0.20 | (2.0)[h] | 0.22 |
| Acetic acid | (1.3)[h] | (2.1)[h] | 3.8 ± 1.8 | 0.4–1.4 | 0.98 | (4.1)[h] | 0.8 |
| $H_2$ | 0.97 ± 0.38 | 3.6–4.0 | 1.8 ± 0.5 | (1.8)[h] | – | (4.6)[h] | (2.4)[h] |
| $NO_x$ (as NO) | 3.9 ± 2.4 | 1.6 ± 0.7 | 3.0 ± 1.4 | 1.1 ± 0.6 | 0.04 | 3.9 | 2.5 ± 1.0 |
| $N_2O$ | 0.21 ± 0.10 | (0.20)[i] | 0.26 ± 0.07 | 0.06 | 0.03 | (0.20)[i] | 0.07 |
| $NH_3$ | 0.6–1.5 | (1.30)[i] | 1.4 ± 0.8 | (1.30)[i] | 0.09 | (1.30)[i] | (1.30)[i] |
| HCN | 0.025–0.031 | (0.15)[i] | (0.15)[i] | (0.15)[i] | (0.15)[i] | (0.15)[i] | (0.15)[i] |
| $N_2$ | (3.1)[j] | (3.1)[j] | (3.1)[j] | (3.1)[j] | – | (3.1)[j] | (3.1)[j] |
| $SO_2$ | 0.35 ± 0.16 | 0.57 ± 0.23 | 1.0 | 0.27 ± 0.30 | – | (0.40)[i] | (0.40)[i] |
| COS | 0.015 ± 0.009 | (0.04)[i] | 0.030–0.036 | (0.04)[i] | (0.04)[i] | (0.04)[i] | 0.065 ± 0.077 |
| $CH_3Cl$ | 0.075 ± 0.029 | 0.02–0.18 | 0.050 ± 0.032 | 0.04–0.07 | (0.01)[i] | 0.012 | 0.24 ± 0.14 |
| $CH_3Br$ | 0.0021 ± 0.0010 | 0.0078 ± 0.0035 | 0.0032 ± 0.0012 | (0.003)[i] | (0.003)[i] | (0.003)[i] | (0.003)[i] |
| $CH_3I$ | 0.0005 ± 0.0002 | 0.0068 | 0.0006 | (0.001)[i] | – | (0.001)[i] | (0.001)[i] |
| $Hg^0$ | 0.0001 | (0.0001)[i] | (0.0001)[i] | (0.0001)[i] | – | (0.0001)[i] | (0.0001)[i] |
| $PM_{2.5}$ | 5.4 ± 1.5 | 9.1 ± 1.5 | 13.0 ± 7.0 | 7.2 ± 2.3 | – | (9)[i] | 3.9 |
| TPM | 8.3 ± 3.2 | 6.5–10.5 | 17.6 ± 6.4 | 9.4 ± 6.0 | 4.0 | (12)[i] | 13 |
| TC | 3.7 ± 1.3 | 6.6 ± 1.5 | 6.1–10.4 | 5.2 ± 1.1 | – | 6.3 | 4.0 |
| OC | 3.4 ± 1.4 | 5.2 ± 1.5 | 8.6–9.7 | 4.0 ± 1.2 | – | 4.8 | 3.3 |
| BC | 0.48 ± 0.18 | 0.66 ± 0.31 | 0.56 ± 0.19 | 0.59 ± 0.37 | – | 1.5 | 0.69 ± 0.13 |
| Levoglucosan | (0.28)[i] | 0.42 | (0.75)[i] | (0.32)[i] | – | – | (0.27)[i] |
| K | 0.34 ± 0.15 | 0.29 ± 0.22 | 0.08–0.41 | 0.05 ± 0.01 | – | 0.40 | 0.13–0.43 |
| CN | $(3.4 \times 10^{15})^j$ | $(3.4 \times 10^{15})^j$ | $(3.4 \times 10^{15})^j$ | $(3.4 \times 10^{15})^j$ | – | $(3.4 \times 10^{15})^j$ | $(3.4 \times 10^{15})^j$ |
| CCN [at 1% SS] | $(2 \times 10^{15})^i$ | $(2 \times 10^{15})^i$ | $[2.6 \pm 4.2] \times 10^{15}$ | $(2 \times 10^{15})^i$ | – | $(2 \times 10^{15})^i$ | $(2 \times 10^{15})^i$ |
| $N_{(>0.12\ \mu m\ diam)}$ | $1.2 \times 10^{15}$ | $(1 \times 10^{15})^i$ | $(1 \times 10^{15})^i$ | $(1 \times 10^{15})^i$ | – | $(1 \times 10^{15})^i$ | $(1 \times 10^{15})^i$ |

[a] Emission factors are given in gram species per kilogram dry matter burned. See text for the conventions used for reporting uncertainties. Abbreviations are as follows: $PM_{2.5}$, particulate matter <2.5 μm diameter; TPM, total particulate matter; TC, total carbon; BC, black carbon; CN, condensation nuclei; CCN, cloud condensation nuclei at 1% supersaturation; and $N_{(>0.12\ \mu m\ diam)}$, particles > 0.12 μm diameter. Values in parentheses represent estimates for emission factors that have not been measured directly. Estimation methods are indicated by superscripts.

[b] Data sources are *Delmas and Servant* [1982], *Brunke et al.* [2001], *Cofer et al.* [1988, 1989], *Ward and Hardy* [1989], *Cofer et al.* [1990a], *Bonsang et al.* [1991], *Delmas et al.* [1991], *Ward et al.* [1991], *Laursen et al.* [1992], *Ward et al.* [1992], *Hao and Ward* [1993], *Lacaux et al.* [1993], *Hurst et al.* [1994a, 1994b], *Manö and Andreae* [1994], *Singh et al.* [1994], *Bonsang et al.* [1995], *Cachier et al.* [1995], *Echalar et al.* [1995], *Gaudichet et al.* [1995], *Helas et al.* [1995], *Lacaux et al.* [1995], *Masclet et al.* [1995], *Nguyen et al.* [1995], *Rudolph et al.* [1995], *Scholes* [1995], *Anderson et al.* [1996], *Andreae et al.* [1996a, 1996b], *Blake et al.* [1996], *Cachier et al.* [1996], *Cofer et al.* [1996a], *Hao et al.* [1996a, 1996b], *Koppmann et al.* [1996], *Lacaux et al.* [1996], *Le Canut et al.* [1996], *Ward et al.* [1996], *Lee et al.* [1997], *Andreae et al.* [1998], *Ferek et al.* [1998], *Friedli et al.* [2001], and *Yamasoe et al.* [2000].

[c] Data sources are *Greenberg et al.* [1984], *Andreae et al.* [1988], *Ward et al.* [1991, 1992], *Hao and Ward* [1993], *Delmas et al.* [1995], *Andreae et al.* [1996b], *Blake et al.* [1996], *Koppmann et al.* [1996], *Andreae et al.* [1998], *Ferek et al.* [1998], *Yamasoe et al.* [2000], and *Graham et al.* [2001].

[d] Data sources are *Miner* [1969], *Eagan et al.* [1974], *Crutzen et al.* [1979], *Stith et al.* [1981], *Ward and Hardy* [1986], *Radke et al.* [1988], *Cofer et al.* [1989, 1990b], *Hegg et al.* [1990], *Radke et al.* [1990], *Susott et al.* [1990], *Ward et al.* [1990], *Cofer et al.* [1991], *Einfeld et al.* [1991], *Radke et al.* [1991], *Ward and Hardy* [1991], *Laursen et al.* [1992], *Wofsy et al.* [1992], *Hao and Ward* [1993], *Nance et al.* [1993], *Blake et al.* [1994], *Lefer et al.* [1994], *Manö and Andreae* [1994], *Singh et al.* [1994], *Manö* [1995], *Cofer et al.* [1996a, 1996b], *FIRESCAN Science Team* [1996], *Hobbs et al.* [1996], *Hurst et al.* [1996], *Martins et al.* [1996], *Vose et al.* [1996], *Worden et al.* [1997], *Andreae et al.* [1998], *Cofer et al.* [1998], *Yokelson et al.* [1999], *Friedli et al.* [2001], and *Goode et al.* [2000].

[e] Data sources are *Rasmussen et al.* [1980], *Smith et al.* [1993], *Brocard et al.* [1996], *Liousse et al.* [1996], *Zhang and Smith* [1996], *Zhang et al.* [1999], *Kituyi et al.* [2001], and Veldt (unpublished manuscript, 1992).

[f] Data sources are *Crutzen et al.* [1979], *De Angelis et al.* [1980], *Myers* [1980], *Rasmussen et al.* [1980], *Dasch* [1982], *Edgerton et al.* [1986], *Smith* [1988], *Delmas et al.* [1991], *Joshi* [1991], *Veldt* [1992], *Hao and Ward* [1993], *Smith et al.* [1993], *Brocard et al.* [1996], *Cachier et al.* [1996], *Liousse et al.* [1996], *Piccot et al.* [1996], *Zhang and Smith* [1996], *Schauer* [1998], *Zhang and Smith* [1999], *Zhang et al.* [1999], *Kituyi et al.* [2001], *Ludwig et al.* [2001], *Zhang et al.* [2000], and Veldt (unpublished manuscript, 1992).

[g] Data sources are *Rasmussen et al.* [1980], *Lobert et al.* [1991], *Hao and Ward* [1993], *Nguyen* [1994], *Nguyen et al.* [1995], *Andreae et al.* [1996a], *Koppmann et al.* [1996], *Liousse et al.* [1996], *Zhuang et al.* [1996], *Andreae et al.* [1998], *de Zarate et al.* [2000], *Kituyi et al.* [2001], and *Ludwig et al.* [2001].

[h] Extrapolation is based on emission ratios to CO.

[i] Value is best guess.

[j] Estimate is based on laboratory studies.

from a particular fire can be quite different from the average values presented in Table 1.

## 3.2. Emission Factors for Chemical Species From Fires in Various Vegetation Types or Burning Practices

During the numerous fire emission studies over the last few decades, including the various Biomass Burning Experiment (BIBEX) field experiments, a large number of emission ratios or factors have been obtained. However, there is no coherent, critical compilation of these data on which a table of "recommended" emission values could be based. Previous reviews have either only provided average values for "biomass burning" in general, ignoring differences between the different types of fires [e.g., Andreae, 1993], or given values only for specific fire types or regions, such as savanna fires [e.g., Andreae, 1997]. In Table 1 we present emission data for the most important types of fire regimes (savannas and grasslands, tropical forest, extratropical forest, domestic biofuel burning, charcoal production and combustion, and agricultural waste burning) for a wide variety of gaseous and particulate emission products. These emission factors are based on an analysis of some 130 publications.

One problem that we encountered in compiling Table 1 was that the amount of information available for each data cell was quite different between chemical species and fire types. For the major carbon species emitted from fires, such as $CO_2$, CO, and $CH_4$, sufficient data are available for all fire types. Conversely, adequate data exist for most species listed in Table 1 in the case of savanna and grassland fires, but for other fire types only the emissions of some key species have been determined satisfactorily. In order to represent the different levels of information upon which the estimates in Table 1 are founded, we have adopted the following convention: When three or more values (based on independent references) are available for a given cell, the results are given as means and standard deviations ($x \pm s$). In the case of two available measurements they are given as a range, and where only a single measurement is available, it is given without an uncertainty estimate. For single measurements it can usually be assumed that the uncertainty is no less than a factor of 3.

For combinations of species and fire type without data available, we have supplied estimates (in parentheses in Table 1) using the following three techniques of extrapolation. (1) For species not closely tied to the smoldering stage and where information is available for most but not all fire types, a weighted estimate was calculated from the averages given for those fire types where data are available. The weighting was based on the global amount of carbon burned in each fire type (see Table 2). (2) The emission factors of species that are predominantly emitted in the smoldering phase can be estimated using the fact that they tend to be closely correlated to the emission factor for CO. For this purpose we calculated a mean ratio $EF_X/EF_{CO}$ from the available data and multiplied it by the $EF_{CO}$ characteristic for the fire types for which $EF_X$ was not measured. (3) For the remaining cases, where there was inadequate information to derive an objective estimate, we provide a "best guess," based on the subjective evaluation of the available information, including the results from laboratory burning studies. For different ways of estimation see footnotes in Table 1.

As mentioned in section 3.1, the level of information available on the different fire types and chemical species varies dramatically across Table 1. While savanna fire emissions are reasonably well characterized, glaring deficiencies prevail for other fire types. An example is charcoal making: Here all the information available is based on the study of Lacaux et al. [1994], which did not include a large set of organic species. Similarly, the organic emissions from charcoal burning are characterized in only one study [Zhang and

Smith, 1996]. The remaining information on volatile organic compound emissions from charcoal making and burning is based on extrapolations provided by C. Veldt (GEIA note on residential biomass burning: A short communication about emission factors for the residential combustion of biomass fuels, unpublished manuscript, 1992) (hereinafter referred to as Veldt, unpublished manuscript, 1992).

While the paucity of data on charcoal-related emissions may not be a major problem, given the relatively small contribution of charcoal burning to the global budget, the limited data on emissions from wood and dung burning have proven to cause serious problems in the interpretation of the measurements of aerosol and trace gas composition downwind of regions where domestic biomass burning is a major contributor, for example, India. Here the relative contributions of biomass and fossil fuel burning have proven difficult to assess, and emission inventories and ambient measurements have proven difficult to reconcile, not in the least because of the poor state of knowledge concerning emission factors [Novakov et al., 2000; Mayol-Bracero et al., 2001]. In India, particularly, dung cakes contribute significantly to the biofuel budget [Streets and Waldhoff, 1999]. Because of their high N, S, and Cl content they have high emission factors for species such as NO, $NH_3$, $SO_2$, and $CH_3Cl$. They have, however, not been included in the average given for biofuels in Table 1 because of the limited availability of emission data and because of the difficulty of assessing a broadly representative dung cake contribution to the fuel mix. Supplementary data on these fire types can be obtained from the authors.

We cannot discuss here all the deficiencies in the presently available data on pyrogenic emissions, and we invite the reader to examine Table 1 in detail. However, we would like to point out some key compounds of global interest for which more data are urgently needed. In this context the small number of field data on emissions of acetonitrile and other nitrogen species, as well as the organic acids and other oxygenized organic compounds, are particularly problematic. With the development of novel in situ techniques that allow the determination of these compounds without the problems associated with sample collection and storage, we can hope for improved data sets for these species in the near future [Holzinger et al., 1999; Crutzen et al., 2000; Reiner et al., 2001]. There are several compounds and compound classes that have not been included in Table 1, particularly the amines and pyrroles. These compounds have been identified in laboratory studies of biomass burning but have not yet been measured in the field. Because of the difficulty of extrapolating from laboratory to field conditions, we have decided to exclude these compounds from the present analysis.

Because of the fairly low combustion temperatures in biomass fires (compared to fossil fuel combustion), atmospheric $N_2$ is not converted to fixed nitrogen to a significant extent, and the nitrogen species emissions are based only on the fuel nitrogen. For this reason, linear relationships have been found between $N_2O$ [Lobert et al., 1991] and $NO_x$ [Lacaux et al., 1993] emission and fuel nitrogen content. NO, $NO_2$, $N_2O$, and molecular $N_2$ are released predominantly during flaming combustion, whereas $NH_3$, amines, and nitriles are associated with smoldering combustion. The most abundant N species in the emissions is typically NO, and it has been frequently assumed that all $NO_x$ is released in this form. Where $NO_2$ has been determined selectively, however, it has been found that it represents some 10% of $NO_x$ already directly above the fire [Griffith et al., 1991; Yokelson et al., 1996]. Our emission factor estimate for $N_2O$ is based only on data that have been collected without any or with minimal sample storage to avoid the positive artifact that results from prolonged storage in containers. $NH_3$ emissions dominate in the smoldering stage and can account for most of the N emissions in that phase [Griffith et al., 1991]. This trace gas used to be considered a minor N species in smoke,

**Table 2.** Global Emission of Selected Pyrogenic Species in the Late 1990s, Based on the Emission Factors in Table 1 and the Biomass Burning Estimates of J. A. Logan and R. Yevich[a]

| | Savanna and Grassland | Tropical Forest | Extratropical Forests | Biofuel Burning | Charcoal Making | Charcoal Burning | Agricultural Residues[b] | Total |
|---|---|---|---|---|---|---|---|---|
| Dry matter burned, Tg | 3,160 | 1,330 | 640 | 2,701 | 158 | 38 | 540 | 8,600 |
| $CO_2$ | 5,096 | 2,101 | 1,004 | 4,187 | 70 | 99 | 818 | 13,400 |
| CO | 206 | 139 | 68 | 209 | 11 | 7.6 | 50 | 690 |
| $CH_4$ | 7.4 | 9.0 | 3.0 | 16.5 | 1.7 | 0.24 | 1.5 | 39 |
| NMHC | 10.7 | 10.8 | 3.6 | 19.6 | 0.3 | 0.10 | 3.5 | 49 |
| $C_2H_2$ | 0.92 | 0.53 | 0.17 | 1.90 | 0.01 | 0.004 | 0.20 | 3.7 |
| Methanol | 3.8 | 2.6 | 1.3 | 3.9 | 0.02 | 0.14 | 0.9 | 12.7 |
| Formaldehyde | 1.1 | 1.8 | 1.4 | 0.4 | – | 0.10 | 0.7 | 5.5 |
| Acetaldehyde | 1.6 | 0.86 | 0.32 | 0.37 | – | 0.05 | 0.31 | 3.5 |
| Acetone | 1.4 | 0.83 | 0.35 | 0.06 | 0.00 | 0.05 | 0.30 | 3.0 |
| Acetonitrile | 0.33 | 0.24 | 0.12 | 0.49 | – | 0.007 | 0.10 | 1.3 |
| Formic acid | 2.1 | 1.4 | 1.8 | 0.36 | 0.03 | 0.08 | 0.1 | 5.9 |
| Acetic acid | 4.2 | 2.8 | 2.5 | 2.4 | 0.15 | 0.15 | 0.4 | 12.6 |
| $H_2$ | 3.1 | 5.1 | 1.2 | 4.8 | – | 0.17 | 1.1 | 15.3 |
| $NO_x$ | 12.2 | 2.2 | 1.9 | 2.9 | 0.01 | 0.15 | 1.3 | 20.7 |
| $N_2O$ | 0.67 | 0.27 | 0.17 | 0.16 | 0.00 | 0.008 | 0.04 | 1.31 |
| $NH_3$ | 3.4 | 1.7 | 0.88 | 3.5 | 0.01 | 0.05 | 0.7 | 10.3 |
| HCN | 0.09 | 0.20 | 0.10 | 0.41 | 0.02 | 0.006 | 0.08 | 0.90 |
| $N_2$ | 9.8 | 4.1 | 2.0 | 8.4 | – | 0.12 | 1.7 | 26.0 |
| $SO_2$ | 1.1 | 0.76 | 0.64 | 0.74 | – | 0.015 | 0.22 | 3.5 |
| COS | 0.05 | 0.05 | 0.02 | 0.11 | 0.01 | 0.002 | 0.03 | 0.27 |
| $CH_3Cl$ | 0.24 | 0.10 | 0.03 | 0.14 | – | 0.0005 | 0.13 | 0.65 |
| $CH_3Br$ | 0.006 | 0.010 | 0.002 | 0.008 | – | 0.00011 | 0.002 | 0.029 |
| $CH_3I$ | 0.0016 | 0.0090 | 0.0004 | 0.0027 | – | 0.00004 | 0.0005 | 0.014 |
| $Hg^0$ | 0.0003 | 0.0001 | 0.00006 | 0.0003 | – | – | 0.0001 | 0.0008 |
| $PM_{2.5}$ | 16.1 | 12.0 | 8.3 | 19.4 | – | 0.34 | 2.1 | 58.3 |
| TPM | 26.2 | 11.3 | 11.3 | 25.5 | 0.63 | 0.46 | 7.0 | 82.4 |
| TC | 11.7 | 8.7 | 5.3 | 14.0 | – | 0.24 | 2.2 | 42.2 |
| OC | 10.6 | 7.0 | 5.8 | 10.7 | – | 0.18 | 1.8 | 36.1 |
| BC | 1.5 | 0.88 | 0.36 | 1.6 | – | 0.06 | 0.37 | 4.8 |
| K | 1.09 | 0.39 | 0.16 | 0.14 | – | 0.02 | 0.15 | 1.9 |
| CN | 1.1E + 28[c] | 4.5E + 27 | 2.2E + 27 | 9.2E + 27 | – | 1.3E + 26 | 1.8E + 27 | 2.9E + 28 |
| CCN (1% SS) | 6.3E + 27 | 2.7E + 27 | 1.7E + 27 | 5.4E + 27 | – | 7.6E + 25 | 1.1E + 27 | 1.7E + 28 |
| $N_{(>0.12\ \mu m\ diam)}$ | 3.7E + 27 | 1.3E + 27 | 6.4E + 26 | 2.7E + 27 | – | 3.8E + 25 | 5.4E + 26 | 9.0E + 27 |

[a] Values are given in mass of species per year ($Tg\ yr^{-1}$). Biomass burning estimates are from R. Yevich (personal communication, 2001).
[b] Value excludes agricultural waste used as biofuel.
[c] Read, for example, 1.1E + 28 as $1.1 \times 10^{28}$.

but studies with improved analytical techniques, particularly Fourier transform infrared spectroscopy, have shown that it is often emitted at roughly equimolar amounts to $NO_x$ [*Griffith et al.*, 1991; *Yokelson et al.*, 1996; *Worden et al.*, 1997; *Yokelson et al.*, 1997; *Goode et al.*, 1999]. Some 30–40% of the fuel nitrogen is released in the form of molecular $N_2$ [*Kuhlbusch et al.*, 1991]. Because it is impossible to detect the small enhancement of atmospheric $N_2$ that would be present in a smoke plume, only laboratory data are available for the emission factor of this species, and the value obtained in the single study published on $N_2$ emission has been used in Table 1. Hydrogen cyanide (HCN) has been proposed as a potentially valuable tracer of biomass burning since it can be determined by remote sensing from space [*Li et al.*, 2000]. Unfortunately, there are only two field studies during which HCN was measured, both in Australian savanna fires [*Hurst et al.*, 1994a, 1994b], and the emission factors from these studies are an order of magnitude lower than the results of laboratory emission measurements (0.31 ± 0.25 mg $kg^{-1}$) [*Lobert et al.*, 1991; *Holzinger et al.*, 1999]. In view of the large uncertainty regarding this interesting species, additional field measurements would be highly desirable.

The methyl halides, $CH_3Cl$, $CH_3Br$, and $CH_3I$, are formed predominantly in the smoldering stage, probably because of reaction between methanol and HCl etc. catalyzed at glowing char surfaces [*Reinhardt and Ward*, 1995] or by radical reactions in flames. Since the emission factors depend both on the halogen content of the fuel and on the proportion of flaming to smoldering combustion [*Andreae et al.*, 1996b], considerable variability in the emission ratios has been observed.

Fairly good information is now available on aerosol emissions from most types of burning, with the exception of agricultural fires and charcoal use. This does not apply, however, to measurements of the number of particles emitted per amount of biomass burned. The determination of this parameter is problematic, since it changes very rapidly in the initial phase of plume development [*Liousse et al.*, 1995; *Hobbs et al.*, 1996; *Reid et al.*, 1998]. Just outside of the flames, particle concentrations are very high (ten thousands to millions per cubic centimenter), and rapid reduction of particle numbers by coagulation takes place. This is a highly nonlinear process and slows down rapidly when the particle concentration drops because of coagulation and dilution. At the same time the particle size grows, both because of coagulation and condensation of vapors as the plume cools down. The ability of the particles to act as cloud condensation nuclei (CCN) also increases sharply during plume aging, presumably because of condensational uptake of water-soluble material on the particles [*Hobbs et al.*, 1996]. For these reasons the values in Table 1 given for particle number emission factors must be considered as fairly rough estimates. They are meant to apply to a slightly aged (1–2 hours) smoke plume to avoid the large temporal variations shortly after emission. The results for the different particle classes appear reasonably internally consistent, even though they come from a

variety of sources. The ratio of ~3 between total particle number and the size fraction >0.12 μm is reasonably consistent with the number median diameters of ~0.1 μm typically observed for slightly aged smoke [e.g., *Hobbs et al.*, 1996; *Reid et al.*, 1998]. Also, the CN/CCN (1%) ratio of ~1.7 agrees with the observation that biomass smoke particles tend to be good CCN after short aging [*Warner and Twomey*, 1967; *Rogers et al.*, 1991; *Hobbs et al.*, 1996; *Pham-Van-Dinh et al.*, 1996].

### 3.3. Emissions From Global Biomass Burning

While, as we have shown in section 3.2, the average emission factors for many important species, such as CO and $CH_4$, are now known with an uncertainty of about 20–30%, large uncertainties persist for regional and global fire emissions because of the difficulties inherent in estimating the amount of biomass burned. The estimation of the amounts of biomass combusted per unit area and time is still based on rather crude assessments and has not yet benefited significantly from the remote sensing tools becoming available at this time. Where comparisons between different approaches (e.g., inventories versus remote sensing) and combustion estimates have been made, they have shown large differences for specific regions. One example comes from the savanna regions of southern Africa, where a difference of almost an order of magnitude has been found between regional estimates based on estimates of fire frequencies in the various vegetation types and those based on fire counts obtained from remote sensing [*Scholes et al.*, 1996; *Scholes and Andreae*, 2000].

Table 2 provides a set of global emission estimates for the late 1990s, based on the emissions factors in Table 1 and the biomass burning estimates of J. A. Logan and R. Yevich (R. Yevich, personal communication, 2001). The data on charcoal use were updated using the Food and Agriculture Organization's FAOSTAT database at http://apps.fao.org/). Uncertainties are not explicitly stated in Table 2, in part because there is not enough information to estimate them quantitatively. For each entry in Table 2 the appropriate error would result from error propagation from the emission factor data in Table 1 and the estimates of biomass burned. The uncertainties related to the emission factors have been discussed in section 3.2, where it was pointed out that in many cases there are not enough data available for a statistical error analysis. Given the methodological problems associated with estimating the amount of biomass burned, there is again no basis for a statistically valid error estimate. The inventory-based estimates for biomass burned have changed little over the last decade [*Scholes et al.*, 2001], but this is due more to the use of a relatively constant underlying information base and methodology than to actual accuracy of the data. Until tools become available to perform independent validation of these estimates, we must assume that they are uncertain to at least ±50%.

Some support for the accuracy of our estimates comes from the application of inverse models to the analysis of the budgets of CO and $CH_4$. Our estimate of pyrogenic $CH_4$ (39 Tg yr$^{-1}$) agrees very well with the inverse modeling estimate of 40 ± 12 Tg yr$^{-1}$ by *Hein et al.* [1997]. For a variety of model scenarios an inverse model considering both CO concentration and isotopic composition data predicted CO emissions of 483–633 Tg yr$^{-1}$, 140–245 Tg yr$^{-1}$, and 0–87 Tg yr$^{-1}$ for tropical forest burning, savanna burning, and burning at latitudes >30°N, respectively [*Bergamaschi et al.*, 2000]. The total pyrogenic CO emission was estimated to be between 663 and 807 Tg yr$^{-1}$. Comparison with Table 2 shows very good agreement between our a priori estimate and the inversion results for the total CO source but shows poorer agreement for the individual contributions, especially that of forest burning. This is probably because Table 2 includes emission from fire types not included (biofuel burning, agricultural burning, and charcoal making) in the categories that are represented in the

model. While the agreement between our inventory-based estimates and the results of inverse modeling do not provide very narrow constraints on the accuracy of our estimates, they suggest that the pyrogenic emissions of CO and $CH_4$ lie within ±50% of the values in Table 2. On the basis of a comparison between modeled and observed CO concentrations, *Galanter et al.* [2000] also conclude that global estimates of pyrogenic emissions for these species are considerably more accurate than a factor of 2. Since the emission factors for CO and $CH_4$ are fairly well known, this implies that the total amount of biomass burned annually is also within an uncertainty range of about ±50% around the value of 8600 Tg dry matter used in Table 2.

## 4. Conclusions

Considerable progress has been made over the last decade with regard to the determination of emission factors from biomass burning. A critical evaluation of the available data shows that a vast number of chemical species have been identified in biomass burning smoke and that reliable emission information exists for most of the key species, at least for savanna fires. There remain, however, serious gaps for important species, including ones that could be valuable atmospheric tracers, such as HCN and acetonitrile. Some combustion types also need further study, for example, the various types of biofuel use, including charcoal making. The global emission estimates from biomass burning have been refined but require further validation. This applies particularly to the estimates of biomass burned as a function of space, time, and type of combustion. The agreement between the results from inverse models and the inventory-based estimates presented here is encouraging, but more rigorous constraints of emission estimates could come from regional experiments (similar to that described by *Cautenet et al.* [1999]) that are designed to test the agreement between emission inventories and transport and chemistry models.

## References

Anderson, B. E., W. B. Grant, G. L. Gregory, E. V. Browell, J. E. Collins, Jr., G. W. Sachse, D. R. Bagwell, C. H. Hudgins, D. R. Blake, and N. J. Blake, Aerosols from biomass burning over the tropical South Atlantic region: Distributions and impacts, *J. Geophys. Res.*, 101, 24,117–24,138, 1996.

Andreae, M. O., Soot carbon and excess fine potassium: Long-range transport of combustion-derived aerosols, *Science*, 220, 1148–1151, 1983.

Andreae, M. O., The influence of tropical biomass burning on climate and the atmospheric environment, in *Biogeochemistry of Global Change: Radiatively Active Trace Gases*, edited by R. S. Oremland, pp. 113–150, Chapman and Hall, New York, 1993.

Andreae, M. O., Climatic effects of changing atmospheric aerosol levels, in *World Survey of Climatology*, vol. 16, *Future Climates of the World*, edited by A. Henderson-Sellers, pp. 341–392, Elsevier Sci., New York, 1995.

Andreae, M. O., Emissions of trace gases and aerosols from southern African savanna fires, in *Fire in the Southern African Savannas: Ecological and Environmental Perspectives*, edited by B. W. van Wilgen et al., pp. 161–183, Witwatersrand Univ. Press, Johannesburg, South Africa, 1997.

Andreae, M. O., et al., Biomass-burning emissions and associated haze layers over Amazonia, *J. Geophys. Res.*, 93, 1509–1527, 1988.

Andreae, M. O., B. E. Anderson, D. R. Blake, J. D. Bradshaw, J. E. Collins, G. L. Gregory, G. W. Sachse, and M. C. Shipham, Influence of plumes from biomass burning on atmospheric chemistry over the equatorial Atlantic during CITE-3, *J. Geophys. Res.*, 99, 12,793–12,808, 1994.

Andreae, M. O., E. Atlas, H. Cachier, W. R. Cofer, III, G. W. Harris, G. Helas, R. Koppmann, J.-P. Lacaux, and D. E. Ward, Trace gas and aerosol emissions from savanna fires, in *Biomass Burning and Global Change*,

edited by J. S. Levine, pp. 278–295, MIT Press, Cambridge, Mass., 1996a.

Andreae, M. O., et al., Methyl halide emissions from savanna fires in southern Africa, *J. Geophys. Res.*, *101*, 23,603–23,613, 1996b.

Andreae, M. O., J. Fishman, and J. Lindesay, The Southern Tropical Atlantic Region Experiment (STARE): Transport and Atmospheric Chemistry near the Equator— Atlantic (TRACE A) and Southern African Fire-Atmosphere Research Initiative (SAFARI): An introduction, *J. Geophys. Res.*, *101*, 23,519–23,520, 1996c.

Andreae, M. O., et al., Airborne studies of aerosol emissions from savanna fires in southern Africa, 2, Aerosol chemical composition, *J. Geophys. Res.*, *103*, 32,119–32,128, 1998.

Bergamaschi, P., R. Hein, C. A. M. Brenninkmeijer, and P. J. Crutzen, Inverse modeling of the global CO cycle, 2, Inversion of $^{13}C/^{12}C$ and $^{18}O/^{16}O$ isotope ratios, *J. Geophys. Res.*, *105*, 1929–1945, 2000.

Blake, D. R., T. W. Smith, T.-Y. Chen, W. J. Whipple, and F. S. Rowland, Effects of biomass burning on summertime nonmethane hydrocarbon concentrations in the Canadian wetlands, *J. Geophys. Res.*, *99*, 1699–1719, 1994.

Blake, N. J., D. R. Blake, B. C. Sive, T. Y. Chen, F. S. Rowland, J. E. Collins, G. W. Sachse, and B. E. Anderson, Biomass burning emissions and vertical distribution of atmospheric methyl halides and other reduced carbon gases in the South Atlantic region, *J. Geophys. Res.*, *101*, 24,151–24,164, 1996.

Blake, N. J., et al., Influence of southern hemispheric biomass burning on midtropospheric distributions of nonmethane hydrocarbons and selected halocarbons over the remote South Pacific, *J. Geophys. Res.*, *104*, 16,213–16,232, 1999.

Bonsang, B., G. Lambert, and C. C. Boissard, Light hydrocarbons emissions from African savanna burnings, in *Global Biomass Burning: Atmospheric, Climatic and Biospheric Implications*, edited by J. S. Levine, pp. 155–161, MIT Press, Cambridge, Mass., 1991.

Bonsang, B., C. Boissard, M. F. Le Cloarec, J. Rudolph, and J. P. Lacaux, Methane, carbon monoxide and light non methane hydrocarbon emissions from African savanna burnings during the FOS/DECAFE experiment, *J. Atmos. Chem.*, *22*, 149–162, 1995.

Brocard, D., C. Lacaux, J. P. Lacaux, K. Kouadio, and V. Yoboué, Emissions from the combustion of biofuels in western Africa, in *Biomass Burning and Global Change*, edited by J. S. Levine, pp. 350–360, MIT Press, Cambridge, Mass., 1996.

Browell, E. V., M. A. Fenn, et al., Ozone and aerosol distributions and air mass characteristics over the South Atlantic basin during the burning season, *J. Geophys. Res.*, *101*, 24,043–24,068, 1996.

Brunke, E. G., C. Labuschagne, and F. Slemr, Gaseous mercury emissions from a fire in the Cape Peninsula, South Africa, during January 2000, *Geophys. Res. Lett.*, *28*, 1483–1486, 2001.

Cachier, H., C. Liousse, P. Buat-Menard, and A. Gaudichet, Particulate content of savanna fire emissions, *J. Atmos. Chem.*, *22*, 123–148, 1995.

Cachier, H., C. Liousse, M.-H. Pertuisot, A. Gaudichet, F. Echalar, and J.-P. Lacaux, African fire particulate emission and atmospheric influence, in *Biomass Burning and Global Change*, edited by J. S. Levine, pp. 428–440, MIT Press, Cambridge, Mass., 1996.

Cahoon, D. R., Jr., B. J. Stocks, J. S. Levine, W. R. Cofer, III, and J. M. Pierson, Satellite analysis of the severe 1987 forest fires in northern China and southeastern Siberia, *J. Geophys. Res.*, *99*, 18,627–18,638, 1994.

Cautenet, S., D. Poulet, C. Delon, R. Delmas, J. M. Gregoire, J. M. Pereira, S. Cherchali, O. Amram, and G. Flouzat, Simulation of carbon monoxide redistribution over central Africa during biomass burning events (Experiment for Regional Sources and Sinks of Oxidants (EXPRESSO)), *J. Geophys. Res.*, *104*, 30,641–30,657, 1999.

Cofer, W. R., III, J. S. Levine, D. I. Sebacher, E. L. Winstead, P. J. Riggan, J. A. Brass, and V. G. Ambrosia, Particulate emissions from a midlatitude prescribed chaparral fire, *J. Geophys. Res.*, *93*, 5207–5212, 1988.

Cofer, W. R., III, J.S. Levine, D. I. Sebacher, E. L. Winstead, P. J. Riggan, B. J. Stocks, J. A. Brass, V. G. Ambrosia, and P. J. Boston, Trace gas emissions from chaparral and boreal forest fires, *J. Geophys. Res.*, *94*, 2255–2259, 1989.

Cofer, W. R., III, J. S. Levine, E. L. Winstead, P. J. LeBel, A. M. Koller, and C. R. Hinkle, Trace gas emissions from burning Florida wetlands, *J. Geophys. Res.*, *95*, 1865–1870, 1990a.

Cofer, W. R., III, J. S. Levine, E. L. Winstead, and B. J. Stocks, Gaseous emissions from Canadian boreal forest fires, *Atmos. Environ., Part A*, *24*, 1653–1659, 1990b.

Cofer, W. R., III, J. S. Levine, E. L. Winstead, and B. J. Stocks, New estimates of nitrous oxide emissions from biomass burning, *Nature*, *349*, 689–691, 1991.

Cofer, W. R., III, J. S. Levine, E. L. Winstead, D. R. Cahoon, D. I. Sebacher, J. P. Pinto, and B. J. Stocks, Source composition of trace gases

released during African savanna fires, *J. Geophys. Res.*, *101*, 23,597–23,602, 1996a.

Cofer, W. R., III, E. L. Winstead, B. J. Stocks, L. W. Overbay, J. G. Goldammer, D. R. Cahoon, and J. S. Levine, Emissions from boreal forest fires: Are the atmospheric impacts underestimated?, in *Biomass Burning and Global Change*, edited by J. S. Levine, pp. 834–839, MIT Press, Cambridge, Mass., 1996b.

Cofer, W. R., III, E. L. Winstead, B. J. Stocks, J. G. Goldammer, and D. R. Cahoon, Crown fire emissions of $CO_2$, CO, $H_2$, $CH_4$, and TNMHC from a dense jack pine boreal forest fire, *Geophys. Res. Lett.*, *25*, 3919–3922, 1998.

Connors, V. S., M. Flood, T. Jones, B. Gormsen, S. Nolf, and H. G. Reichle, Jr., Global distribution of biomass burning and carbon monoxide in the middle troposphere during early April and October 1994, in *Biomass Burning and Global Change*, edited by J. S. Levine, pp. 99–106, MIT Press, Cambridge, Mass., 1996.

Crutzen, P. J., and M. O. Andreae, Biomass burning in the tropics: Impact on atmospheric chemistry and biogeochemical cycles, *Science*, *250*, 1669–1678, 1990.

Crutzen, P. J., L. E. Heidt, J. P. Krasnec, W. H. Pollock, and W. Seiler, Biomass burning as a source of atmospheric gases CO, $H_2$, $N_2O$, NO, $CH_3Cl$, and COS, *Nature*, *282*, 253–256, 1979.

Crutzen, P. J., et al., High spatial and temporal resolution measurements of primary organics and their oxidation products over the tropical forests of Surinam, *Atmos. Environ.*, *34*, 1161–1165, 2000.

Dasch, J. M., Particulate and gaseous emissions from wood-burning fireplaces, *Environ. Sci. Techol.*, *16*, 639–645, 1982.

De Angelis, D. G., D. S. Ruffin, and R. B. Rezink, Preliminary characterization of emissions from wood-fired residential combustion equipment, *Rep. EPA-600/2-80-042b*, U.S. Environ. Prot. Agency, Washington, D. C., 1980.

Delmas, R., and J. Servant, The origins of sulfur compounds in the atmosphere of a zone of high productivity (Gulf of Guinea), *J. Geophys. Res.*, *87*, 11,019–11,026, 1982.

Delmas, R. A., A. Marenco, J. P. Tathy, B. Cros, and J. G. R. Baudet, Sources and sinks of methane in the African savanna: $CH_4$ emissions from biomass burning, *J. Geophys. Res.*, *96*, 7287–7299, 1991.

Delmas, R., J. P. Lacaux, and D. Brocard, Determination of biomass burning emission factors: Methods and results, *Environ. Monit. Assess.*, *38*, 181–204, 1995.

Delmas, R. A., et al., Experiment for Regional Sources and Sinks of Oxidants (EXPRESSO): An overview, *J. Geophys. Res.*, *104*, 30,609–30,624, 1999.

de Zarate, I. O., A. Ezcurra, J. P. Lacaux, and P. Van Dinh, Emission factor estimates of cereal waste burning in Spain, *Atmos. Environ.*, *34*, 3183–3193, 2000.

Eagan, R. C., P. V. Hobbs, and L. F. Radke, Measurements of cloud condensation nuclei and cloud droplet size distributions in the vicinity of forest fires, *J. Appl. Meteorol.*, *13*, 553–557, 1974.

Echalar, F., A. Gaudichet, H. Cachier, and P. Artaxo, Aerosol emissions by tropical forest and savanna biomass burning: Characteristic trace elements and fluxes, *Geophys. Res. Lett.*, *22*, 3039–3042, 1995.

Edgerton, S. A., M. A. K. Khalil, and R. A. Rasmussen, Source emission characterization of residential wood-burning stoves and fireplaces: Fine particle/methyl chloride ratios for use in chemical mass balance modeling, *Environ. Sci. Technol.*, *20*, 803–807, 1986.

Einfeld, W., D. E. Ward, and C. C. Hardy, Effects of fire behavior on prescribed fire smoke characteristics: A case study, in *Global Biomass Burning: Atmospheric, Climatic, and Biospheric Implications*, edited by J. S. Levine, pp. 412–419, MIT Press, Cambridge, Mass., 1991.

Ferek, R. J., J. S. Reid, P. V. Hobbs, D. R. Blake, and C. Liousse, Emission factors of hydrocarbons, halocarbons, trace gases, and particles from biomass burning in Brazil, *J. Geophys. Res.*, *103*, 32,107–32,118, 1998.

FIRESCAN Science Team, Fire in Ecosystems of Boreal Eurasia: The Bor Forest Island Fire Experiment Fire Research Campaign Asia-North (FIRESCAN), in *Biomass Burning and Global Change*, edited by J. S. Levine et al., pp. 848–873, MIT Press, Cambridge, Mass., 1996.

Fishman, J., C. E. Watson, J. C. Larsen, and J. A. Logan, Distribution of tropospheric ozone determined from satellite data, *J. Geophys. Res.*, *95*, 3599–3617, 1990.

Fishman, J., K. Fakhruzzaman, B. Cros, and D. Nganga, Identification of widespread pollution in the southern hemisphere deduced from satellite analyses, *Science*, *252*, 1693–1696, 1991.

Fishman, J., J. M. Hoell, R. D. Bendura, R. J. McNeil, and V. W. J. H. Kirchhoff, NASA GTE TRACE A Experiment (September–October 1992): Overview, *J. Geophys. Res.*, *101*, 23,865–23,879, 1996.

Folkins, I. A., A. J. Weinheimer, B. A. Ridley, J. G. Walega, B. Anderson, J. E. Collins, G. Sachse, R. F. Pueschel, and D. R. Blake, $O_3$, $NO_y$, and

$NO_x/NO_y$ in the upper troposphere of the equatorial Pacific, *J. Geophys. Res.*, *100*, 20,913–20,926, 1995.

Friedli, H. R., E. Atlas, V. R. Stroud, L. Giovanni, T. Campos, and L. F. Radke, Volatile organic trace gases emitted from North American wildfires, *Global Biogeochem. Cycles*, *15*, 435–452, 2001.

Galanter, M., H. Levy, and G. R. Carmichael, Impacts of biomass burning on tropospheric CO, NOx, and $O_3$, *J. Geophys. Res.*, *105*, 6633–6653, 2000.

Gaudichet, A., F. Echalar, B. Chatenet, J. P. Quisefit, G. Malingre, H. Cachier, P. Buat-Ménard, P. Artaxo, and W. Maenhaut, Trace elements in tropical African savanna biomass burning aerosols, *J. Atmos. Chem.*, *22*, 19–39, 1995.

Goode, J. G., R. J. Yokelson, R. A. Susott, and D. E. Ward, Trace gas emissions from laboratory biomass fires measured by open-path Fourier transform infrared spectroscopy: Fires in grass and surface fuels, *J. Geophys. Res.*, *104*, 21,237–21,245, 1999.

Goode, J. G., R. J. Yokelson, D. E. Ward, R. A. Susott, R. E. Babbitt, M. A. Davies, and W.-M. Hao, Measurements of excess $O_3$, $CO_2$, CO, $CH_4$, $C_2H_4$, $C_2H_2$, HCN, NO, $NH_3$, HCOOH, $CH_3COOH$, HCHO, and $CH_3OH$ in 1997 Alaskan biomass burning plumes by airborne Fourier transform infrared spectroscopy (AFTIR), *J. Geophys. Res.*, *105*, 22,147–22,166, 2000.

Graham, B., O. L. Mayol-Bracero, P. Guyon, G. C. Roberts, S. Decesari, M. C. Facchini, P. Artaxo, W. Maenhaut, P. Köll, and M. O. Andreae, Water-soluble organic compounds in biomass burning aerosols over Amazonia, 1, Characterization by NMR and GC-MS, *J. Geophys. Res.*, in press, 2001.

Greenberg, J. P., P. R. Zimmerman, L. Heidt, and W. Pollock, Hydrocarbon and carbon monoxide emissions from biomass burning in Brazil, *J. Geophys. Res.*, *89*, 1350–1354, 1984.

Gregory, G. L., et al., Chemical characteristics of Pacific tropospheric air in the region of the Intertropical Convergence Zone and South Pacific Convergence Zone, *J. Geophys. Res.*, *104*, 5677–5696, 1999.

Griffith, D. W. T., W. G. Mankin, M. T. Coffey, D. E. Ward, and A. Riebau, FTIR remote sensing of biomass burning emissions of $CO_2$, CO, $CH_4$, $CH_2O$, NO, $NO_2$, $NH_3$, and $N_2O$, in *Global Biomass Burning: Atmospheric, Climatic and Biospheric Implications*, edited by J. S. Levine, pp. 230–239, MIT Press, Cambridge, Mass., 1991.

Hao, W.-M., and D. E. Ward, Methane production from global biomass burning, *J. Geophys. Res.*, *98*, 20,657–20,661, 1993.

Hao, W. M., D. E. Ward, G. Olbu, S. P. Baker, and J. R. Plummer, Emissions of trace gases from fallow forests and woodland savannas in Zambia, in *Biomass Burning and Global Change*, edited by J. S. Levine, pp. 361–369, MIT Press, Cambridge, Mass., 1996a.

Hao, W.-M., D. E. Ward, G. Olbu, and S. P. Baker, Emissions of $CO_2$, CO and hydrocarbons from fires in diverse African savanna ecosystems, *J. Geophys. Res.*, *101*, 23,577–23,584, 1996b.

Hegg, D. A., L. F. Radke, P. V. Hobbs, R. A. Rasmussen, and P. J. Riggan, Emissions of some trace gases from biomass fires, *J. Geophys. Res.*, *95*, 5669–5675, 1990.

Hein, R., P. J. Crutzen, and M. Heimann, An inverse modeling approach to investigate the global atmospheric methane cycle, *Global Biogeochem. Cycles*, *11*, 43–76, 1997.

Helas, G., et al., Airborne measurements of savanna fire emissions and the regional distribution of pyrogenic pollutants over western Africa, *J. Atmos. Chem.*, *22*, 217–239, 1995.

Hobbs, P. V., J. S. Reid, J. A. Herring, J. D. Nance, R. E. Weiss, J. L. Ross, D. A. Hegg, R. D. Ottmar, and C. Liousse, Particle and trace-gas measurements in the smoke from prescribed burns of forest products in the Pacific Northwest, in *Biomass Burning and Global Change*, edited by J. S. Levine, pp. 697–715, MIT Press, Cambridge, Mass., 1996.

Holzinger, R., C. Warneke, A. Hansel, A. Jordan, W. Lindinger, D. H. Scharffe, G. Schade, and P. J. Crutzen, Biomass burning as a source of formaldehyde, acetaldehyde, methanol, acetone, acetonitrile, and hydrogen cyanide, *Geophys. Res. Lett.*, *26*, 1161–1164, 1999.

Hurst, D. F., D. W. T. Griffith, J. N. Carras, D. J. Williams, and P. J. Fraser, Measurements of trace gases emitted by Australian savanna fires during the 1990 dry season, *J. Atmos. Chem.*, *18*, 33–56, 1994a.

Hurst, D. F., D. W. T. Griffith, and G. D. Cook, Trace gas emissions from biomass burning in tropical Australian savannas, *J. Geophys. Res.*, *99*, 16,441–16,456, 1994b.

Hurst, D. F., D. W. T. Griffith, and G. D. Cook, Trace-gas emissions from biomass burning in Australia, in *Biomass Burning and Global Change*, edited by J. S. Levine, pp. 787–792, MIT Press, Cambridge, Mass., 1996.

Jonquières, I., A. Marenco, A. Maalej, and F. Rohrer, Study of ozone formation and transatlantic transport from biomass burning emissions over West Africa during the airborne Tropospheric Ozone Campaigns TROPOZ I and TROPOZ II, *J. Geophys. Res.*, *103*, 19,059–19,073, 1998.

Joshi, V., Biomass burning in India, in *Global Biomass Burning: Atmospheric, Climatic, and Biospheric Implications*, edited by J. S. Levine, pp. 185–193, MIT Press, Cambridge, Mass., 1991.

Kaufman, Y. J. P., et al., Smoke, Clouds, and Radiation - Brazil (SCAR-B) experiment, *J. Geophys. Res.*, *103*, 31,783–31,808, 1998.

Kirchhoff, V. W. J. H., and C. A. Nobre, Atmospheric chemistry research in Brazil: Ozone measurements at Natal, Manaus, and Cuiabá, *Rev. Geofis.*, *24*, 95–108, 1986.

Kituyi, E., L. Marufu, S. O. Wandiga, I. O. Jumba, M. O. Andreae, and G. Helas, Carbon monoxide and nitric oxide from biofuel fires in Kenya, *Energy Convers. Manage.*, in press, 2001.

Koppmann, R., A. Khedim, J. Rudolph, G. Helas, M. Welling, and T. Zenker, Airborne measurements of organic trace gases from savanna fires in southern Africa during SAFARI-92, in *Biomass Burning and Global Change*, edited by J. S. Levine, pp. 309–319, MIT Press, Cambridge, Mass., 1996.

Kuhlbusch, T. A., J. M. Lobert, P. J. Crutzen, and P. Warneck, Molecular nitrogen emissions from denitrification during biomass burning, *Nature*, *351*, 135–137, 1991.

Lacaux, J.-P., H. Cachier, and R. Delmas, Biomass burning in Africa: An overview of its impact on atmospheric chemistry, in *Fire in the Environment: The Ecological, Atmospheric, and Climatic Importance of Vegetation Fires*, edited by P. J. Crutzen and J. G. Goldammer, pp. 159–191, John Wiley, New York, 1993.

Lacaux, J. P., D. Brocard, C. Lacaux, R. Delmas, B. Ahoua, V. Yoboué, and M. Koffi, Traditional charcoal making: An important source of atmospheric pollution in the African Tropics, *Atmos. Res.*, *35*, 71–76, 1994.

Lacaux, J. P., J. M. Brustet, R. Delmas, J. C. Menaut, L. Abbadie, B. Bonsang, H. Cachier, J. Baudet, M. O. Andreae, and G. Helas, Biomass burning in the tropical savannas of Ivory Coast: An overview of the field experiment Fire Of Savannas (FOS/DECAFE '91), *J. Atmos. Chem.*, *22*, 195–216, 1995.

Lacaux, J.-P., R. Delmas, C. Jambert, and T. A. J. Kuhlbusch, $NO_x$ emissions from African savanna fires, *J. Geophys. Res.*, *101*, 23,585–23,596, 1996.

Laursen, K. K., P. V. Hobbs, L. F. Radke, and R. A. Rasmussen, Some trace gas emissions from North American biomass fires with an assessment of regional and global fluxes from biomass burning, *J. Geophys. Res.*, *97*, 20,687–20,701, 1992.

Le Canut, P., M. O. Andreae, G. W. Harris, F. G. Wienhold, and T. Zenker, Airborne studies of emissions from savanna fires in southern Africa, 1, Aerosol emissions measured with a laser optical particle counter, *J. Geophys. Res.*, *101*, 23,615–23,630, 1996.

Lee, M., B. G. Heikes, D. J. Jacob, G. Sachse, and B. Anderson, Hydrogen peroxide, organic hydroperoxides, and formaldehyde as primary pollutants from biomass burning, *J. Geophys. Res.*, *102*, 1301–1309, 1997.

Lefer, B. L., et al., Enhancement of acidic gases in biomass burning impacted air masses over Canada, *J. Geophys. Res.*, *99*, 1721–1737, 1994.

Li, Q. B., D. J. Jacob, I. Bey, R. M. Yantosca, Y. J. Zhao, Y. Kondo, and J. Notholt, Atmospheric hydrogen cyanide (HCN): Biomass burning source, ocean sink?, *Geophys. Res. Lett.*, *27*, 357–360, 2000.

Lindesay, J. A., M. O. Andreae, J. G. Goldammer, G. Harris, H. J. Annegarn, M. Garstang, R. J. Scholes, and B. W. van Wilgen, International Geosphere-Biosphere Programme/International Global Atmospheric Chemistry SAFARI 92 field experiment: Background and overview, *J. Geophys. Res.*, *101*, 23,521–23,530, 1996.

Liousse, C., C. Devaux, F. Dulac, and H. Cachier, Aging of savanna biomass burning aerosols: Consequences on their optical properties, *J. Atmos. Chem.*, *22*, 1–17, 1995.

Liousse, C., J. E. Penner, J. J. Walton, H. Eddleman, C. Chuang, and H. Cachier, Modeling biomass burning aerosols, in *Biomass Burning and Global Change*, edited by J. S. Levine, pp. 492–508, MIT Press, Cambridge, Mass., 1996.

Lobert, J. M., and J. Warnatz, Emissions from the combustion process in vegetation, in *Fire in the Environment: The Ecological, Atmospheric, and Climatic Importance of Vegetation Fires*, edited by P. J. Crutzen and J. G. Goldammer, pp. 15–37, John Wiley, New York, 1993.

Lobert, J. M., D. H. Scharffe, W.-M. Hao, T. A. Kuhlbusch, R. Seuwen, P. Warneck, and P. J. Crutzen, Experimental evaluation of biomass burning emissions: Nitrogen and carbon containing compounds, in *Global Biomass Burning: Atmospheric, Climatic and Biospheric Implications*, edited by J. S. Levine, pp. 289–304, MIT Press, Cambridge, Mass., 1991.

Lobert, J. M., W. C. Keene, J. A. Logan, and R. Yevich, Global chlorine emissions from biomass burning: Reactive Chlorine Emissions Inventory, *J. Geophys. Res.*, *104*, 8373–8389, 1999.

Ludwig, J., L. T. Marufu, B. Huber, M. O. Andreae, and G. Helas, Com-

bustion of biomass fuels in developing countries—A major source of atmospheric pollutants, *J. Atmos. Chem.*, in press, 2001.

Manö, S., Messung von partiell oxidierten Kohlenwasserstoffen in Emissionen von Biomasseverbrennung, Ph.D. thesis, 120 pp., Goethe-Univ., Frankfurt, Germany, 1995.

Manö, S., and M. O. Andreae, Emission of methyl bromide from biomass burning, *Science*, 263, 1255–1257, 1994.

Martins, J. V., P. Artaxo, P. V. Hobbs, C. Liousse, H. Cachier, Y. Kaufman, and A. Plana-Fattori, Particle size distributions, elemental compositions, carbon measurements, and optical properties of smoke from biomass burning in the Pacific Northwest of the United States, in *Biomass Burning and Global Change*, edited by J. S. Levine, pp. 716–732, MIT Press, Cambridge, Mass., 1996.

Masclet, P., H. Cachier, C. Liousse, and H. Wortham, Emissions of polycyclic aromatic hydrocarbons by savanna fires, *J. Atmos. Chem.*, 22, 41–54, 1995.

Mayol-Bracero, O. L., R. Gabriel, M. O. Andreae, T. W. Kirchstetter, T. Novakov, J. A. Ogren, P. J. Sheridan, and D. Streets, Carbonaceous aerosols over the Indian Ocean during INDOEX: Chemical characterization, optical properties, and probable sources, *J. Geophys. Res.*, in press, 2001.

Miner, S., Preliminary air pollution survey of ammonia, *Rep. APTD-69-25*, 39 pp., Natl. Air Pollut. Control Admin., Raleigh, N. C., 1969.

Myers, N., *Conversion of Tropical Moist Forests*, Natl. Acad. Press, Washington, D. C., 1980.

Nance, J. D., P. V. Hobbs, L. F. Radke, and D. E. Ward, Airborne measurements of gases and particles from an Alaskan wildfire, *J. Geophys. Res.*, 98, 14,873–14,882, 1993.

Nguyen, B. C., Rice straw burning in southeast Asia as a source of CO and COS to the atmosphere, *J. Geophys. Res.*, 99, 16,435–16,439, 1994.

Nguyen, B. C., N. Mihalopoulos, J. P. Putaud, and B. Bonsang, Carbonyl sulfide emissions from biomass burning in the tropics, *J. Atmos. Chem.*, 22, 55–65, 1995.

Novakov, T., M. O. Andreae, R. Gabriel, T. Kirchstetter, O. Mayol-Bracero, and V. Ramanathan, Origin of carbonaceous aerosols over the tropical Indian Ocean: Biomass burning or fossil fuels?, *Geophys. Res. Lett.*, 27, 4061–4064, 2000.

Novelli, P. C., K. A. Masarie, and P. M. Lang, Distributions and recent changes of carbon monoxide in the lower troposphere, *J. Geophys. Res.*, 103, 19,015–19,033, 1998.

Penner, J. E., R. E. Dickinson, and C. A. O'Neill, Effects of aerosol from biomass burning on the global radiation budget, *Science*, 256, 1432–1434, 1992.

Pham-Van-Dinh, R. Serpolay, and J.-P. Lacaux, Laboratory investigation on aerosols from the combustion of savanna grass and cereal straw, in *Biomass Burning and Global Change*, edited by J. S. Levine, pp. 472–484, MIT Press, Cambridge, Mass., 1996.

Piccot, S. D., L. Beck, S. Srinivasan, and S. L. Kersteter, Global methane emissions from minor anthropogenic sources and biofuel combustion in residential stoves, *J. Geophys. Res.*, 101, 22,757–22,766, 1996.

Radke, L. F., J. L. Stith, D. A. Hegg, and P. V. Hobbs, Airborne studies of particles and gases from forest fires, *J. Air Pollut. Control. Assoc.*, 28, 30–34, 1978.

Radke, L. F., D. A. Hegg, J. H. Lyons, C. A. Brock, P. V. Hobbs, R. Weiss, and R. Rasmussen, Airborne measurements on smokes from biomass burning, in *Aerosols and Climate*, edited by P. V. Hobbs and M. P. McCormick, pp. 411–422, A. Deepak Publishing, Hampton, Va., 1988.

Radke, L. F., J. H. Lyons, P. V. Hobbs, D. A. Hegg, D. V. Sandberg, and D. E. Ward, Airborne monitoring and smoke characterization of prescribed fires on forest lands in western Washington and Oregon, *Tech. Rep. PNW-GTR-251*, 81 pp., For. Serv., U.S. Dep. of Agric., Portland, Ore., 1990.

Radke, L. F., D. A. Hegg, P. V. Hobbs, J. D. Nance, J. H. Lyons, K. K. Laursen, R. E. Weiss, P. J. Riggan, and D. E. Ward, Particulate and trace gas emissions from large biomass fires in North America, in *Global Biomass Burning: Atmospheric, Climatic and Biospheric Implications*, edited by J. S. Levine, pp. 209–224, MIT Press, Cambridge, Mass., 1991.

Rasmussen, R. A., L. E. Rasmussen, M. A. K. Khalil, and R. W. Dalluge, Concentration distribution of methyl chloride in the atmosphere, *J. Geophys. Res.*, 85, 7350–7356, 1980.

Reichle, H. G., V. S. Conners, J. A. Holland, W. D. Hypes, H. A. Wallio, J. C. Casas, B. B. Gormsen, M. S. Saylor, and W. D. Hasketh, Middle and upper tropospheric carbon monoxide mixing ratios as measured by a satellite-borne remote sensor during November 1981, *J. Geophys. Res.*, 91, 10,865–10,888, 1986.

Reid, J. S., P. V. Hobbs, R. J. Ferek, D. R. Blake, J. V. Martins, M. R. Dunlap, and C. Liousse, Physical, chemical, and optical properties of regional hazes dominated by smoke in Brazil, *J. Geophys. Res.*, 103, 32,059–32,080, 1998.

Reiner, T., D. Sprung, C. Jost, R. Gabriel, O. Mayol-Bracero, M. O. Andreae, T. Campos, and R. E. Shetter, Chemical characterization of pollution layers over the tropical Indian Ocean: Signatures of emissions from biomass and fossil fuel burning, *J. Geophys. Res.*, in press, 2001.

Reinhardt, T. E., and D. E. Ward, Factors affecting methyl chloride emissions from forest biomass combustion, *Environ. Sci. Techol.*, 29, 825–832, 1995.

Rogers, C. F., J. G. Hudson, B. Zielinska, R. L. Tanner, J. Hallett, and J. G. Watson, Cloud condensation nuclei from biomass burning, in *Global Biomass Burning: Atmospheric, Climatic and Biospheric Implications*, edited by J. S. Levine, pp. 431–438, MIT Press, Cambridge, Mass., 1991.

Rudolph, J., A. Khedim, R. Koppmann, and B. Bonsang, Field study of the emissions of methyl chloride and other halocarbons from biomass burning in western Africa, *J. Atmos. Chem.*, 22, 67–80, 1995.

Schauer, J. J., Source contributions to atmospheric organic compound concentrations: Emission measurements and model predictions, Ph.D. thesis, Calif. Inst. of Technol., Pasadena, Calif., 1988.

Scholes, M., and M. O. Andreae, Biogenic and pyrogenic emissions from Africa and their impact on the global atmosphere, *Ambio*, 29, 23–29, 2000.

Scholes, M., P. Matrai, K. Smith, M. O. Andreae, and A. Guenther, Biosphere–atmosphere interactions, in *Atmospheric Chemistry in a Changing World*, edited by G. Brasseur, R. Prinn, and A. Pszenny, Springer-Verlag, New York, in press, 2001.

Scholes, R. J., Greenhouse gas emissions from vegetation fires in southern Africa, *Environ.l Monit. Assess.*, 38, 169–179, 1995.

Scholes, R. J., J. Kendall, and C. O. Justice, The quantity of biomass burned in southern Africa, *J. Geophys. Res.*, 101, 23,667–23,676, 1996.

Schüle, W., Landscapes and climate in prehistory: Interactions of wildlife, man and fire, in *Fire in the Tropical Biota: Ecosystem Processes and Global Challenges*, edited by J. Goldammer, pp. 273–318, Springer-Verlag, New York, 1990.

Seiler, W., and P. J. Crutzen, Estimates of gross and net fluxes of carbon between the biosphere and the atmosphere from biomass burning, *Clim. Change*, 2, 207–247, 1980.

Singh, H. B., D. O'Hara, D. Herlth, G. W. Sachse, D. R. Blake, J. D. Bradshaw, M. Kanakidou, and P. J. Crutzen, Acetone in the atmosphere: Distribution, sources, and sinks, *J. Geophys. Res.*, 99, 1805–1819, 1994.

Singh, H. B., et al., Biomass burning influences on the composition of the remote South Pacific troposphere: Analysis based on observations from PEM-Tropics-A, *Atmos. Environ.*, 34, 635–644, 2000.

Smith, K. R., Air pollution: Assessing total exposure in developing countries, *Environment*, 30, 16–35, 1988.

Smith, K. R., M. A. K. Khalil, R. A. Rasmussen, S. A. Thorneloe, F. Manegdeg, and M. Apte, Greenhouse gases from biomass and fossil fuel stoves in developing countries: A Manila pilot study, *Chemosphere*, 26, 479–505, 1993.

Stith, J. L., L. F. Radke, and P. V. Hobbs, Particle emissions and the production of ozone and nitrogen oxides from the burning of forest slash, *Atmos. Environ.*, 15, 73–82, 1981.

Streets, D. G., and S. T. Waldhoff, Greenhouse-gas emissions from biofuel combustion in Asia, *Energy*, 24, 841–855, 1999.

Susott, R. A., D. E. Ward, R. E. Babbitt, D. J. Latham, L. G. Weger, and P. M. Boyd, Fire dynamics and chemistry of large fires, final report, 39 pp., For. Serv., U.S. Dep. of Aric., Missoula, Mont., 1990.

Talbot, R. W., J. D. Bradshaw, et al., Chemical characteristics of continental outflow over the tropical South Atlantic Ocean from Brazil and Africa, *J. Geophys. Res.*, 101, 24,187–24,202, 1996.

Thompson, A. M., K. E. Pickering, D. P. McNamara, M. R. Schoeberl, R. D. Hudson, J.-H. Kim, E. V. Browell, V. W. J. H. Kirchhoff, and D. Nganga, Where did tropospheric ozone over southern Africa and the tropical Atlantic come from in October 1992? Insights from TOMS, GTE TRACE-A, and SAFARI 1992, *J. Geophys. Res.*, 101, 24,251–24,278, 1996.

Vose, J. M., W. T. Swank, C. D. Geron, and A. E. Major, Emissions from forest burning in the southeastern United States: Application of a model determining spatial and temporal fire variation, in *Biomass Burning and Global Change*, edited by J. S. Levine, pp. 733–749, MIT Press, Cambridge, Mass., 1996.

Ward, D., and C. Hardy, Advances in the characterization and control of emissions from prescribed broadcast fires of coniferous species logging slash on clearcut units, final report, U.S. Environ. Prot. Agency, Missoula, Mont., 1986.

Ward, D. E., and C. C. Hardy, Emissions from prescribed chaparral burning, paper presented at Annual Meeting, Air and Waste Manage. Assoc., Anaheim, Calif., 1989.

Ward, D. E., and C. C. Hardy, Smoke emissions from wildland fires, *Environ. Int.*, *17*, 117–134, 1991.

Ward, D. E., R. A. Susott, R. E. Babbitt, and C. C. Hardy, Properties and concentration of smoke near the ground from biomass field tests, paper presented at the Symposium on Smoke/Obscurants XIV, Laurel, Md., April 17–19, 1990.

Ward, D. E., A. W. Setzer, Y. J. Kaufman, and R. A. Rasmussen, Characteristics of smoke emissions from biomass fires of the Amazon region—BASE-A experiment, in *Global Biomass Burning: Atmospheric, Climatic, and Biospheric Implications*, edited by J. S. Levine, pp. 394–402, MIT Press, Cambridge, Mass., 1991.

Ward, D. E., W.-M. Hao, R. A. Susott, R. A. Babbitt, R. W. Shea, J. B. Kauffman, and C. O. Justice, Effect of fuel composition on combustion efficiency and emission factors for African savanna ecosystems, *J. Geophys. Res.*, *101*, 23,569–23,576, 1996.

Ward, D. E., R. A. Susott, J. B. Kauffman, R. E. Babbitt, D. L. Cummings, B. Dias, B. N. Holben, Y. J. Kaufman, R. A. Rasmussen, and A. W. Setzer, Smoke and fire characteristics for cerrado and deforestation burns in Brazil: BASE-B experiment, *J. Geophys. Res.*, *97*, 14,601–14,619, 1992.

Warner, J., and S. Twomey, The production of cloud nuclei by cane fires and the effect on cloud drop concentrations, *J. Atmos. Sci.*, *24*, 704–706, 1967.

Watson, C. E., J. Fishman, and H. G. Reichle, The significance of biomass burning as a source of carbon monoxide and ozone in the Southern Hemisphere tropics: A satellite analysis, *J. Geophys. Res.*, *95*, 14,443–14,450, 1990.

Wofsy, S. C., et al., Atmospheric chemistry in the Arctic and subarctic: Influence of natural fires, industrial emissions, and stratospheric inputs, *J. Geophys. Res.*, *97*, 16,731–16,746, 1992.

Worden, H., R. Beer, and C. P. Rinsland, Airborne infrared spectroscopy of 1994 western wildfires, *J. Geophys. Res.*, *102*, 1287–1299, 1997.

Yamasoe, M. A., P. Artaxo, A. H. Miguel, and A.G. Allen, Chemical composition of aerosol particles from direct emissions of vegetation fires in the Amazon Basin: Water-soluble species and trace elements, *Atmos. Environ.*, *34*, 1641–1653, 2000.

Yokelson, R. J., D. W. T. Griffith, and D. E. Ward, Open-path Fourier transform infrared studies of large-scale laboratory biomass fires, *J. Geophys. Res.*, *101*, 21,067–21,080, 1996.

Yokelson, R. J., R. Susott, D. E. Ward, J. Reardon, and D. W. T. Griffith, Emissions from smoldering combustion of biomass measured by open-path Fourier transform infrared spectroscopy, *J. Geophys. Res.*, *102*, 18,865–18,877, 1997.

Yokelson, R. J., J. G. Goode, D. E. Ward, R. A. Susott, R. E. Babbitt, D. D. Wade, I. Bertschi, D. W. T. Griffith, and W. M. Hao, Emissions of formaldehyde, acetic acid, methanol, and other trace gases from biomass fires in North Carolina measured by airborne Fourier transform infrared spectroscopy, *J. Geophys. Res.*, *104*, 30,109–30,125, 1999.

Zhang, J., and K. R. Smith, Hydrocarbon emissions and health risks from cookstoves in developing countries, *J. Expo. Anal. Environ. Epidemiol.*, *6*, 147–161, 1996.

Zhang, J., and K. R. Smith, Emissions of carbonyl compounds from various cookstoves in China, *Environ. Sci. Technol.*, *33*, 2311–2320, 1999.

Zhang, J., K. R. Smith, R. Uma, Y. Ma, V. V. N. Kishore, K. Lata, M. A. K. Khalil, R. A. Rasmussen, and S. T. Thorneloe, Carbon monoxide from cookstoves in developing countries, 1, Emission factors, *Chemosphere Global Change Sci.*, *1*, 353–366, 1999.

Zhang, J., K. R. Smith, Y. Ma, S. Ye, F. Jiang, W. Qi, P. Liu, M. A. K. Khalil, R. A. Rasmussen, and S. A. Thorneloe, Greenhouse gases and other airborne pollutants from household stoves in China: A database for emission factors, *Atmos. Environ.*, *34*, 4537–4549, 2000.

Zhuang, Y.-H., M. Cao, X. Wang, and H. Yao, Spatial distribution of trace gas emissions from burning crop residue in China, in *Biomass Burning and Global Change*, edited by J. S. Levine, pp. 764–770, MIT Press, Cambridge, Mass., 1996.

M. O. Andreae and P. Merlet, Biogeochemistry Department, Max Planck Institute for Chemistry, P.O. Box 3060, Mainz D-55020, Germany. (moa@mpch-mainz.mpg.de)