



## Assessment of Global Emissions from Vegetation Fires

### Abstract

A large amount of information on emissions from the various types of vegetation fires has been accumulated over the past decade. However, because this information is scattered over a large number of publications, and has been reported in numerous different units and reference systems, it has so far not been readily accessible to the scientific and decision-making communities. This paper provides a critical evaluation of the presently available data, integrated into a consistent format. On the basis of this analysis, we present a set of emission factors for key species emitted from biomass fires. Using these emission factors, 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 modelling studies. Given the magnitude of the emissions from biomass burning, one must expect significant impacts on air quality, human health, climate and the water cycle.



**Figure 1.** A “pyro-cumulus” forming over a land-use fire in Amazonia. This photograph visualizes symbolically the effects of fire on the atmosphere by releasing gaseous and particle emissions. Pyrogenic emissions influence the composition and functioning of the atmosphere, including the formation of cloud condensation nuclei that change radiation and precipitation budgets. Photo: M.O. Andreae

### Introduction

Air pollution from the smoke of biomass fires has been humanity's constant companion for some two million years, ever since the origin of our species in the savannas and woodlands of Africa. The

impact of biomass smoke on human health is reflected, for example, in soot deposits in the lungs of mummies. In spite of the long history of biomass smoke as a pollutant, the first scientific papers on the impact of biomass burning on the chemistry of the atmosphere were only published in the 1970s and early 1980s (e.g., 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, especially in the tropics (Andreae 1983, Reichle et al. 1986, Fishman et al. 1990).

Satellite and airborne observations have shown elevated levels of O<sub>3</sub>, CO, and other trace gases over vast areas of Africa, South America, the tropical Atlantic, the Indian Ocean, and the Pacific. Smoke aerosols perturb regional and global radiation budgets by their light-scattering and absorbing effects and by their influence on cloud microphysical processes. 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.

To assess the atmospheric impact of biomass burning, and especially to represent it quantitatively in atmospheric models, accurate data on the emission of trace gases and aerosols from biomass fires are required. 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 IGAC (International Global Atmospheric Chemistry) Project of the IGBP (summarized in Scholes et al. 2002). 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.

This paper provides a synthesis of the currently available data on fire emission characteristics for a number of important chemical species in 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), separate emission factors for the different types of biomass burning, such as deforestation fires in the tropics, savanna fires, etc. are provided. The emission factor data are combined with activity estimates for the various fire categories to provide global estimates of emissions of biomass burning.

## **Methods**

Definitions: Emission information is usually represented in one of two basic forms, as emission ratios or emission factors. The emission ratio relates the emission of a particular species of interest to that of a reference species, such as CO<sub>2</sub> or CO, while the emission factor makes reference to the amount of fuel burned. Here, we will use emission factors only; a detailed discussion of the definitions and merits of the various emission units and their conversion can be found in Andreae and Merlet (2001). Briefly, the emission factor is defined as the amount of a compound released per amount of dry fuel consumed by combustion, expressed in units of g kg<sup>-1</sup>. 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. Emission factors from field experiments are therefore usually derived from field measurement of emission ratios, combined with estimates of the emission factor of the reference species.

## **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 combustion conditions. These, in turn, depend on parameters such as fuel structure, fuel and air moisture, temperature, wind speed, etc. 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. 1997), and will be reviewed here only briefly. Combustion of the individual fuel elements proceeds through a sequence of stages - ignition, pyrolysis, flaming+pyrolysis, glowing+pyrolysis (smouldering), glowing, and extinction - each with different chemical processes that result in different emissions. Plant biomass consists of cellulose and hemicelluloses (typically 50-70% d.m.), lignin (15-35%), proteins, aminoacids, 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 about 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<sub>2</sub>, H<sub>2</sub>O, NO, N<sub>2</sub>O, N<sub>2</sub>, and SO<sub>2</sub>. Depending on the interaction between chemical kinetics and physical dynamics in the flame, intermediate products of flame radical chemistry, like CO, CH<sub>4</sub>, H<sub>2</sub>, C<sub>2</sub>H<sub>4</sub>, C<sub>2</sub>H<sub>2</sub>, 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 smouldering begins, dominated by the gas-solid reaction between oxygen and carbon in the char layer at the fuel surface. This lower-temperature process emits large amounts of CO, as well as incompletely oxidized pyrolysis products that are similar to the products of the initial solid phase decomposition. It is thus this mode of combustion that is responsible for the vast diversity of emission products, some of which is shown in Table 1. Overall, several hundreds of organic compounds have been found in the gas and aerosol phases of biomass smoke. 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 smouldering combustion.

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 smouldering during the later part. Especially in forest fires, this late smouldering stage can continue for days or even weeks.

Each of the types of vegetation fires differentiated in Table 1 has characteristic fuel composition and structure, and typical ways in which these fires occur naturally or are managed by people. Therefore, they tend to have characteristic ratios of flaming and smouldering combustion, which to a large extent determines 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, on the other hand, is almost exclusively a smouldering 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 from a particular fire can be quite different from the average values presented in Table 1.

### **Emission factors for chemical species from fires in various vegetation types or burning practices**

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 140 publications. One problem 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<sub>2</sub>, CO, and CH<sub>4</sub>, sufficient data are available for all fire types. For many other

chemical species, data exist only from a few of the fire types. In order to represent the different levels of information upon which the estimates in Table 1 are placed, we have adopted the following convention: When 3 or more values (based on independent studies) are available for a given cell, the results are given as means and standard deviations ( $\bar{x} \pm s$ ). 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 three. For combinations of species and fire type without data available, we have supplied estimates (in *Italics*) using various techniques of extrapolation (Andreae and Merlet 2001).

Because of the fairly low combustion temperatures in biomass fires (compared to fossil fuel combustion) atmospheric  $N_2$  is not significantly converted to  $NO_x$  ( $NO + NO_2$ ), and the nitrogen species emissions are based only on the fuel nitrogen. As a consequence, linear relationships have been found between fuel nitrogen content and  $N_2O$  and  $NO_x$  emission (Lobert et al. 1991, Lacaux et al. 1993).  $NO$ ,  $NO_2$ ,  $N_2O$ , and molecular  $N_2$  are released predominantly during flaming combustion, whereas  $NH_3$ , amines and nitriles are associated with smouldering combustion. The most abundant  $NO_x$  species in the emissions is  $NO$ , but  $NO_2$  typically represents some 10-20% of  $NO_x$  (Griffith et al. 1991, Yokelson et al. 1996).  $NH_3$  emissions dominate in the smouldering stage, and can account for most of the N emissions in that stage.  $NH_3$  used to be considered a minor N species in smoke, but studies with improved analytical techniques, particularly FTIR, have shown that it is often emitted at roughly equimolar amounts to  $NO_x$  (Griffith et al. 1991, 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).

The methyl halides,  $CH_3Cl$ ,  $CH_3Br$ , and  $CH_3I$ , are formed predominantly in the smouldering stage, probably due to reaction between methanol and  $HCl$  etc. catalyzed at glowing char surfaces 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 smouldering combustion (Andreae et al. 1996), considerable variability in the emission ratios has been observed.

Fairly good information is now available on aerosol mass emission factors 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 particle number concentrations changes very rapidly in the initial phase of plume development. Just outside of the flames, particle concentrations are very high (ten thousands to millions per  $cm^3$ ) and rapid reduction of particle numbers by coagulation takes place. This is a highly non-linear process, which slows down rapidly when the particle concentration drops due to coagulation and dilution. At the same time, the particle size grows, both due to coagulation and condensation of vapours 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 and because of chemical transformation (oxidation) of the organic aerosol. 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  $\sim 3$  between total particle number and the size fraction  $>0.12 \mu m$  is consistent with the number median diameters of  $\sim 0.1 \mu m$  typically observed for slightly aged smoke (e.g., Hobbs et al. 1996, Reid and Hobbs 1998). The CN/CCN (1%) ratio of  $\sim 1.7$  agrees with the observation that biomass smoke particles tend to be good CCN after short aging (Warner and Twomey 1967, Rogers et al. 1991, Pham-Van-Dinh et al. 1994, Hobbs et al. 1996).

As already alluded to above, 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, e.g. biofuel use, including charcoal making and burning. 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, e.g. 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 the least due to the poor state of knowledge concerning emission factors (Mayol-Bracero et al. 2002). 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<sub>3</sub>, SO<sub>2</sub>, and CH<sub>3</sub>Cl. 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.

### **Emissions from global biomass burning**

While the average emission factors for many important species, such as CO and CH<sub>4</sub>, 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 enough from the remote sensing tools becoming available at this time. Where comparisons between different approaches (e.g., inventories vs. remote sensing) to combustion estimates have been made, they have shown large differences for specific regions. In southern Africa, for example, a difference of almost an order of magnitude has been found between regional estimates based on average 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 Logan and Yevich (R. Yevich, personal communication, 2001). 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 inventory-based estimates for biomass burned have changed little over the last decade (Scholes et al. 2002), but this is more due 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  $\pm 50\%$ .

Some support for the accuracy of the estimates in Table 2 comes from the application of inverse models to the analysis of the budgets of CO and CH<sub>4</sub>. Our estimate of pyrogenic CH<sub>4</sub> (41 Tg a<sup>-1</sup>) agrees very well with the inverse-modeling estimate of  $40 \pm 12$  Tg a<sup>-1</sup> by Hein et al. (1997). In the case of CO, an inverse model considering both concentration and isotopic composition data predicted CO emissions of 483-633 Tg a<sup>-1</sup>, 140-245 Tg a<sup>-1</sup>, and 0-87 Tg a<sup>-1</sup> for tropical forest burning, savanna burning, and burning at latitudes  $>30^\circ\text{N}$ , respectively (Bergamaschi et al. 2000). The total pyrogenic CO emission was estimated between 663 and 807 Tg a<sup>-1</sup>. Comparison with Table 2 shows very good agreement between our a priori estimate and the inversion results for the total CO source, but poorer agreement for the individual contributions, especially that of forest burning. This is probably due to the fact, that Table 2 includes emission from fire types not included in the model (biofuel burning, agricultural burning, and charcoal making). While the agreement between our inventory-based estimates and the results of inverse modelling does not provide very narrow constraints on the accuracy of our estimates, it suggests that the pyrogenic emissions of CO and CH<sub>4</sub> lie within  $\pm 50\%$  of the values in Table 2. Since the emission factors for CO and CH<sub>4</sub> are fairly well known, this implies that the total amount of biomass burned annually is also within an uncertainty range of about  $\pm 50\%$  around the value of 9200 Tg dry matter used in Table 2.

### **Environmental impact of biomass burning**

While in this brief assessment there is not adequate space to discuss the environmental impacts in extenso, an indication can be gleaned from a comparison of the emissions of key pollutants from biomass burning and from fossil fuel burning. In the last column of Table 2 we list data on emissions from fossil fuel burning (and some industrial activities) taken from the Third Assessment Report of the IPCC (Houghton et al. 2001).

Biomass burning releases about two-thirds as much CO<sub>2</sub> as fossil fuel burning. It can be argued that a substantial fraction of the CO<sub>2</sub> released from vegetation burning is taken up into the biosphere again after a short time. This only applies, however, as long as burning is done in a sustainable manner, which is not the case for deforestation fires and much of domestic biofuel use. For two other

greenhouse gases, methane and nitrous oxide, pyrogenic emissions are very significant as well. In the case of methane, fires emit about one-third as much as fossil-fuel related activities (including pipeline losses, etc.). For N<sub>2</sub>O, pyrogenic sources rival the sum of all industrial emissions.

The large releases of the photochemically active hydrocarbons and NO<sub>x</sub> lead to the formation of ozone and photochemical smog. Ozone concentrations in regions affected by biomass smoke are comparable to those in industrial regions. Together with the vast amounts of smoke aerosol particles emitted from fires, these smog gases constitute a serious health hazard. Beyond their health effects, smoke particles influence the Earth's climate and hydrological cycles in ways that are still inadequately understood. Light-absorbing ("soot") particles absorb solar radiation, and thereby warm the atmosphere, cool the Earth's surface, and reduce the evaporation of water from oceans and land. On the other hand, smoke particles scatter sunlight back into space and change the properties of clouds, including their ability to produce rain. Given the uneven distribution of aerosols in space and time, we must expect substantial regional impacts on climate and water availability.

## **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 acetonitrile. Some combustion types also need further study, e.g., 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 designed to test the agreement between emission inventories and transport and chemistry models. The emissions from biomass burning have significant impacts on air quality, human health, climate and the water cycle.

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**Table 1.** Emission factors (in g species per kg dry matter burned) for pyrogenic species emitted from various types of biomass burning. See text for the conventions used for reporting uncertainties.

| Species                         | Savanna and grassland <sup>1</sup> | Tropical forest <sup>2</sup> | Extratropical forest <sup>3</sup> | Biofuel burning <sup>4</sup> | Charcoal making <sup>5</sup> | Charcoal burning <sup>5</sup> | Agricultural residues <sup>6</sup> |
|---------------------------------|------------------------------------|------------------------------|-----------------------------------|------------------------------|------------------------------|-------------------------------|------------------------------------|
| CO <sub>2</sub>                 | 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 <sub>4</sub>                 | 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 <sup>c</sup>                   |
| 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                              |
| methanol                        | 1.3 <sup>c</sup>                   | 2.0 <sup>c</sup>             | 2.0±1.4                           | 1.5 <sup>c</sup>             | 0.16                         | 3.8 <sup>c</sup>              | 2.0 <sup>c</sup>                   |
| formaldehyde                    | 0.26-0.44                          | 1.4 <sup>c</sup>             | 2.2±0.5                           | 0.13±0.05                    | ---                          | 2.6 <sup>c</sup>              | 1.4 <sup>c</sup>                   |
| acetone                         | 0.25-0.62                          | 0.62 <sup>c</sup>            | 0.52-0.59                         | 0.01-0.04                    | 0.02                         | 1.2 <sup>c</sup>              | 0.63 <sup>c</sup>                  |
| acetonitrile                    | 0.11                               | 0.18 <sup>g</sup>            | 0.19                              | 0.18 <sup>g</sup>            | ---                          | 0.18 <sup>c</sup>             | 0.18 <sup>c</sup>                  |
| formic acid                     | 0.7 <sup>c</sup>                   | 1.1 <sup>c</sup>             | 2.9±2.4                           | 0.13                         | 0.20                         | 2.0 <sup>c</sup>              | 0.22                               |
| acetic acid                     | 1.3 <sup>c</sup>                   | 2.1 <sup>c</sup>             | 3.8±1.8                           | 0.4-1.4                      | 0.98                         | 4.1 <sup>c</sup>              | 0.8                                |
| NO <sub>x</sub> (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 <sub>2</sub> O                | 0.21±0.10                          | 0.20 <sup>g</sup>            | 0.26±0.07                         | 0.06                         | 0.03                         | 0.20 <sup>g</sup>             | 0.07                               |
| NH <sub>3</sub>                 | 0.6-1.5                            | 1.30 <sup>g</sup>            | 1.4±0.8                           | 1.30 <sup>g</sup>            | 0.09                         | 1.30 <sup>g</sup>             | 1.30 <sup>g</sup>                  |
| SO <sub>2</sub>                 | 0.35±0.16                          | 0.57±0.23                    | 1.0                               | 0.27±0.30                    | ---                          | 0.40 <sup>g</sup>             | 0.40 <sup>g</sup>                  |
| COS                             | 0.015±0.009                        | 0.04 <sup>g</sup>            | 0.030-0.036                       | 0.04 <sup>g</sup>            | 0.04 <sup>g</sup>            | 0.04 <sup>g</sup>             | 0.065±0.077                        |
| CH <sub>3</sub> Cl              | 0.075±0.029                        | 0.02-0.18                    | 0.050±0.032                       | 0.04-0.07                    | 0.01 <sup>g</sup>            | 0.012                         | 0.24±0.14                          |
| CH <sub>3</sub> Br              | 0.0021±0.0010                      | 0.0078±0.0035                | 0.0032±0.0012                     | 0.003 <sup>g</sup>           | 0.003 <sup>g</sup>           | 0.003 <sup>g</sup>            | 0.003 <sup>g</sup>                 |
| Hg <sup>0</sup>                 | 0.0001                             | 0.0001 <sup>g</sup>          | 0.0001 <sup>g</sup>               | 0.0001 <sup>g</sup>          | ---                          | 0.0001 <sup>g</sup>           | 0.0001 <sup>g</sup>                |
| PM <sub>2.5</sub>               | 5.4±1.5                            | 9.1±1.5                      | 13.0±7.0                          | 7.2±2.3                      | ---                          | 9 <sup>g</sup>                | 3.9                                |
| TPM                             | 8.3±3.2                            | 6.5-10.5                     | 17.6±6.4                          | 9.4±6.0                      | 4.0                          | 12 <sup>g</sup>               | 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                          |
| K                               | 0.34±0.15                          | 0.29±0.22                    | 0.08-0.41                         | 0.05±0.01                    | ---                          | 0.40                          | 0.13-0.43                          |
| CN                              | 2.6·10 <sup>15</sup>               | 3.4·10 <sup>15</sup> (l)     | 3.4·10 <sup>15</sup> (l)          | 3.4·10 <sup>15</sup> (l)     | ---                          | 3.4·10 <sup>15</sup> (l)      | 3.4·10 <sup>15</sup> (l)           |
| CCN (at 1% SS)                  | 2·10 <sup>15</sup> (g)             | 2·10 <sup>15</sup> (g)       | (2.6±4.2)·10 <sup>15</sup>        | 2·10 <sup>15</sup> (g)       | ---                          | 2·10 <sup>15</sup> (g)        | 2·10 <sup>15</sup> (g)             |
| N <sub>(&gt;0.12 μm dia.)</sub> | 1.2·10 <sup>15</sup>               | 1·10 <sup>15</sup> (g)       | 1·10 <sup>15</sup> (g)            | 1·10 <sup>15</sup> (g)       | ---                          | 1·10 <sup>15</sup> (g)        | 1·10 <sup>15</sup> (g)             |

<sup>c</sup>) Extrapolation based on emission ratios to CO

<sup>g</sup>) Best guess

<sup>l</sup>) Estimate based on laboratory studies

Abbreviations: PM<sub>2.5</sub>: 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, N<sub>(>0.12 μm dia.)</sub>: particles > 0.12 μm diameter.

**Table 2.** Global emission of selected pyrogenic species in the late 1990s (in mass of species per year; Tg a<sup>-1</sup>), based on the emission factors in Table 1 and the biomass burning estimates of Logan and Yevich (R. Yevich, personal communication, 2001).

|                                 | Savanna and grassland | Tropical forest | Extra-tropical forests | Biofuel burning | Charcoal making and burning | Agricultural residues | Total pyrogenic | Fossil fuel burning |
|---------------------------------|-----------------------|-----------------|------------------------|-----------------|-----------------------------|-----------------------|-----------------|---------------------|
| Tg dm burned                    | 3160                  | 1330            | 640                    | 2663            | 196                         | 1190                  | 9200            | ---                 |
| CO <sub>2</sub>                 | 5096                  | 2101            | 1004                   | 4128            | 169                         | 1802                  | 14,300          | 23,100              |
| CO                              | 206                   | 139             | 68                     | 206             | 19                          | 110                   | 750             | 650                 |
| CH <sub>4</sub>                 | 7.4                   | 9.0             | 3.0                    | 16.2            | 1.9                         | 3.2                   | 41              | 110                 |
| NMHC                            | 10.7                  | 10.8            | 3.6                    | 19.3            | 0.4                         | 7.6                   | 53              | 200                 |
| Methanol                        | 3.8                   | 2.6             | 1.3                    | 3.9             | 0.16                        | 2.1                   | 13.8            | ---                 |
| Formaldehyde                    | 1.1                   | 1.8             | 1.4                    | 0.4             | 0.10                        | 1.4                   | 6.3             | ---                 |
| Acetaldehyde                    | 1.6                   | 0.86            | 0.32                   | 0.36            | 0.05                        | 0.68                  | 3.9             | ---                 |
| Acetone                         | 1.4                   | 0.83            | 0.35                   | 0.06            | 0.05                        | 0.65                  | 3.3             | ---                 |
| Acetonitrile                    | 0.33                  | 0.24            | 0.12                   | 0.48            | 0.01                        | 0.21                  | 1.4             | ---                 |
| Formic acid                     | 2.1                   | 1.4             | 1.8                    | 0.35            | 0.11                        | 0.3                   | 6.0             | ---                 |
| Acetic acid                     | 4.2                   | 2.8             | 2.5                    | 2.4             | 0.30                        | 1.0                   | 13.1            | ---                 |
| NO <sub>x</sub> (as NO)         | 12.2                  | 2.2             | 1.9                    | 2.9             | 0.16                        | 3.0                   | 22.3            | 45                  |
| N <sub>2</sub> O                | 0.67                  | 0.27            | 0.17                   | 0.16            | 0.01                        | 0.08                  | 1.4             | 2.0                 |
| NH <sub>3</sub>                 | 3.4                   | 1.7             | 0.88                   | 3.5             | 0.06                        | 1.5                   | 11.0            | 0.4                 |
| SO <sub>2</sub>                 | 1.1                   | 0.76            | 0.64                   | 0.73            | 0.015                       | 0.48                  | 3.7             | 228                 |
| COS                             | 0.05                  | 0.05            | 0.02                   | 0.11            | 0.01                        | 0.07                  | 0.31            | ---                 |
| CH <sub>3</sub> Cl              | 0.24                  | 0.10            | 0.03                   | 0.14            | 0.0005                      | 0.28                  | 0.80            | ---                 |
| CH <sub>3</sub> Br              | 0.006                 | 0.010           | 0.002                  | 0.008           | 0.00011                     | 0.004                 | 0.031           | ---                 |
| PM <sub>2.5</sub>               | 16.1                  | 12.0            | 8.3                    | 19.1            | 0.34                        | 4.6                   | 60              | ---                 |
| TPM                             | 26.2                  | 11.3            | 11.3                   | 25.1            | 1.1                         | 15.5                  | 91              | ---                 |
| TC                              | 11.7                  | 8.7             | 5.3                    | 13.8            | 0.24                        | 4.8                   | 45              | 27                  |
| OC                              | 10.6                  | 7.0             | 5.8                    | 10.5            | 0.18                        | 3.9                   | 38              | 20                  |
| BC                              | 1.5                   | 0.88            | 0.36                   | 1.6             | 0.06                        | 0.82                  | 5.2             | 6.6                 |
| K                               | 1.09                  | 0.39            | 0.16                   | 0.14            | 0.02                        | 0.33                  | 2.1             | ---                 |
| CN                              | 1.1E+28               | 4.5E+27         | 2.2E+27                | 9.1E+27         | 1.3E+26                     | 4.0E+27               | 3.1E+28         | ---                 |
| CCN (1% SS)                     | 6.3E+27               | 2.7E+27         | 1.7E+27                | 5.3E+27         | 7.6E+25                     | 2.4E+27               | 1.8E+28         | ---                 |
| N <sub>(&gt;0.12 μm dia.)</sub> | 3.7E+27               | 1.3E+27         | 6.4E+26                | 2.7E+27         | 3.8E+25                     | 1.2E+27               | 9.6E+27         | ---                 |