A Technology-Based Global Inventory of Black and Organic Carbon Emissions from Combustion

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Abstract

We present a global tabulation of black carbon (BC) and organic carbon (OC) particles emitted from combustion processes. Previous "bottom-up" inventories of black and organic carbon have assigned emission factors to black carbon based on fuel type and economic sector alone. Because emission rates are highly dependent on combustion practice, we consider combinations of fuel, combustion type, and emission controls, and their prevalence on a regional basis. We have paid special attention to the residential and transportation sectors, which contribute about 50% of the global total by our estimates. The inventory is based on 1996 fuel-use data, updating previous estimates that have relied on consumption data from 1984. We include emissions from fossil fuels, biofuels, open biomass burning, and burning of urban waste. Central estimates of global annual emissions are 8.1 Tg for black carbon and 34.0 Tg for organic carbon. These estimates are 35% lower than those published previously. An offset between decreased emission factors and increased energy use since the base year of the previous inventory (1984) prevents this difference from being greater. The contributions of fossil fuel, biofuel, and open biomass burning are estimated as 40%, 20%, and 40% respectively for BC, and 7%, 19%, and 74% respectively for OC. While OC is dominated by open burning, we predict that energy-related combustion (fossil fuels and biofuels) generates about as much BC. As we have reported previously for the cases of China and Asia, low-technology combustion contributes greatly to both the emissions and to the uncertainties. We present a bottom-up estimate of uncertainties in source strength by combining uncertainties in particulate matter emission factors, emission characterization, and fuel use. The total uncertainties are about a factor of two for both BC and OC. Advances in emission characterization for small residential, industrial, and mobile sources, and top-down analysis combining measurements and transport modeling, will be required to reduce the uncertainties further.

1 The Carbonaceous Aerosol Challenge

1.1 Carbonaceous aerosols and the climate system

Anthropogenic contributions to the chemical composition of the atmosphere affect the balance of both visible and infrared radiation in the Earth-atmosphere system. The magnitude and details of the system's response to these changes have been debated, but the relationship between human activity, atmospheric composition, and changes in the radiative balance is less ambiguous. The radiative effect is typically expressed in terms of "forcing"—the change in net flux at the tropopause—which has units of W m^{-2}.

The first examinations of climate forcing addressed heat-trapping or "greenhouse" gases such as CO_2. The greenhouse effect is the best known of global atmospheric changes, the largest in terms of global averages (about +2.5 W m^{-2}, *IPCC*, 2002), and the most targeted for mitigation. An opposing effect, cooling the atmosphere, is provided by increases in "scattering" or reflective aerosols, primarily sulfates; in the past decade or so, this effect has been examined with three-dimensional models [e.g. *Charlson et al.*, 1991; *Kiehl and Briegleb*, 1993]. Current estimates of sulfate forcing are approximately –0.4 W m^{-2}, and are uncertain by about a factor of two [*IPCC*, 2002]. It was also recognized early on that the cooling effects of scattering particles could be opposed by light-absorbing aerosols, which exert a warming effect and increase the amount of energy retained by the earth-atmosphere system [*Charlson and Pilat*, 1969; *Schneider*, 1971]. The first estimates of forcing by light-absorbing black carbon particles were almost insignificant (+0.1 W m^{-2}, *Haywood and Shine* [1995], incorporated in *IPCC*, [1996]). Later, forcing estimates increased, ranging from +0.3 to +0.6 W m^{-2}. Considerations that raised the estimates included the location of absorbers over clouds [*Haywood and Ramaswamy*, 1998], the mixing state of black carbon [*Chylek et al.*, 1995; *Haywood and Ramaswamy*, 1998; *Jacobson*, 2001], and the inclusion of black carbon from biomass burning in addition to fossil-fuel burning [*Koch,* 2000; *Jacobson*, 2001; *Chung and Seinfeld*, 2002].

While particles of any composition reflect light back to space, only a few can absorb light. These include black carbon or “soot”, desert dust [*Sokolik and Toon*, 1996], and some organic carbon species [*Jacobson*, 1999; *Bond*, 2001]. Of these, black carbon is thought to dominate light absorption by aerosols in many regions [*Rosen et al.*, 1978], and it is the most efficient at absorbing visible light. On human time scales, light-absorbing carbon forms only at high temperatures [*Glassman et al.*, 1994], so its primary sources are the combustion of carbon-based fuels. Only a small fraction of the carbon in a fuel is transformed to black carbon, with the remainder being emitted as CO_2 or a variety of products of incomplete combustion. Black carbon remains in the atmosphere for about a week [*Ogren et al.*, 1984; *Parungo et al.*, 1994; *Müller*, 1984], while CO_2 lingers for several decades. Yet if even 0.2% of the fuel carbon were emitted as black carbon, and the remainder were emitted as CO_2, some models predict that the warming by black carbon would exceed the warming by CO_2 when integrated over a 100-year period. (This value was extrapolated from the discussion in *Jacobson* [2002]. The estimate of

BC emission for equivalence was rounded down to be conservative, was not rigorously derived and is not intended for purposes other than qualitative illustration.)

It has been proposed that reductions of light-absorbing particles may assist in slowing the rate of global warming [*Hansen et al.*, 2000]. Model results have also suggested that considering the climatic effects of BC emissions, as well as those of greenhouse gases, may alter conclusions about the technologies selected to reduce warming [*Jacobson*, 2002]. However, BC is emitted simultaneously with organic carbon (OC), which has a net negative climate forcing. Hence, the net climatic effect of reducing emissions of fine particles—those with diameters below 1.0 or 2.5 µm— is ambiguous until *at least* the relative amounts of BC and OC are known.

Along with the recognition of potentially important climatic effects, carbonaceous aerosols have received attention recently for other reasons. Primary particles from combustion— largely black and organic carbon—are suspected to have a host of other effects ranging from the mesoscale to the personal. Field studies have reported high atmospheric concentrations of black carbon [*Chowdhury et al.*, 2001] and organic carbon [*Novakov et al.*, 1997]. Large changes in regional radiative budgets may affect the hydrologic cycle [*Ramanathan et al.*, 2001]; an increase in particle number may reduce cloud droplet sizes, thereby altering cloud brightness [*Twomey et al.*, 1984] and persistence [*Rosenfeld*, 2000], while organic species affect the formation of cloud droplets [*Saxena et al.*, 1995; *Shulman et al.*, 1996]. Heating of the atmosphere by BC may reduce cloudiness [*Ackerman et al.*, 2000]; the addition of light-absorbing particles to the aerosol mix may cause circulation and rainfall shifts [*Menon et al.*, 2002]. Black carbon may play a role in atmospheric chemistry, including that of NO_y [*Lary et al.*, 1999] and sulfates [*Novakov et al.*, 1974]. Radiative interactions by both BC and OC reduce visibility [*Wolff et al.*, 1981; *Qiu and Yang*, 2000] and surface irradiance [*Luo et al.*, 2001], potentially affecting the sunlight available for agriculture [*Chameides et al.*, 1999].

There is a growing literature on the health effects of fine particles, of which *Dockery et al.* [1993] is one of the most widely cited. We will not detail these studies, other than to note that some information about the chemical nature of small particles may help in elucidating the relationship between respiratory problems and air pollution, and that indoor smoke exposures rank among the top public health hazards in developing countries [*Ezzati et al.*, 2002]. Public health considerations are driving reductions of fine particulate matter emissions; an assessment of current emissions is required to project the climatic impacts of these changes.

It is important to remember that *all* particles could be players in most of the detrimental effects just listed, and that focusing on a single aerosol type may oversimplify the knowledge required to understand and mitigate the impact of humans on the environment.

1.2 Estimates of BC and OC source strength

Previous estimates of the global annual source strength of black and organic carbon emissions include those of *Penner et al.* [1993] (13 and 24 Tg BC/year by two different methods); *Cooke and Wilson* [1996] (14 Tg BC/year); and *Cooke et al.* [1999] (5.1 Tg BC from fossil fuels only). These studies outlined methods for estimating global emissions of carbonaceous aerosols, and were invaluable for providing insight into the potential impacts of these particles on the climate system. Follow-up studies [*Kohler et al.*, 2001] have refined the inventory but have usually been based, at least in part, on these seminal papers. Most climate models use the inventories as they are distributed, while some account for economic growth by applying the published emission factors to other fuel-use data [e.g. *Chung and Seinfeld*, 2002].

Unlike emissions of CO_2, but like other trace species such as NOx and CO, the formation of black carbon is quite dependent on the combustion process. For this reason, emissions can vary even among apparently similar technologies. This paper reports a new global inventory of black and organic carbon that has placed special emphasis on the role of combustion practice in the determination of emissions. We also present a first attempt at quantifying the uncertainty in these emissions, which is known to be large [*IPCC*, 2002; *Cooke et al.*, 1999].

We view the inventory presented here as a preliminary step toward a more complete understanding of global sources of carbonaceous aerosols. While we have incorporated data from a variety of sources, it is impossible to be aware of the multitude of efforts on tabulating and characterizing emissions, which are housed in many regions and at many governmental levels. We expect the present inventory to evolve as other researchers share their knowledge and critiques, and we particularly welcome input from those with "on-the-ground" knowledge of source characteristics specific to each region.

In this paper, we have not discussed the use of our inventory in an atmospheric transport model, nor compared spatial distributions with atmospheric BC measurements. The comparison between model output and atmospheric measurements requires a more rigorous approach than simply comparing concentrations. Because energy use and technologies change over time, the year of measurement and inventory must be considered. Appropriate seasonality of emissions or averaging time of measurements affects the comparison as well. Predicted concentrations of atmospheric aerosols are affected not only by emission rates, but also by atmospheric transport, chemical reactions, and removal rates. Uncertainties in those transformations have significant effects in some regions [*Koch*, 2000; *Cooke et al.*, 2002]. Therefore, comparisons with available atmospheric measurements can suggest areas where the inventory might be inaccurate, but agreement does not "validate" it at present.

The required comparisons will be the subject of future studies. Here, we focus on assumptions used to generate the inventories and how they compare with observations made at combustion sources. If those assumptions are incorrect, observed agreement between modeled and measured concentrations is probably fortuitous, and the presence of atmospheric BC may be ascribed to the wrong sources.

1.3 Classifying carbonaceous aerosols

It is important to remember that the terms used to classify carbonaceous aerosols can vary based on the measurement method. A number of caveats are required for interpreting carbon measurements.

Since our focus is on radiative effects, we define BC in this work as the mass of combustion-generated, sp^2-bonded carbon that absorbs the same amount of light as the emitted particles. This carbon has been described as "aciniform" or grapelike rather than planar [*Medalia and Rivin*, 1982], and numerous electron-microscopy photographs confirm its morphology. Again for radiative-transfer purposes, we take the absorption efficiency of the *reference* particles as 7 m^2/g for 550-nm incident radiation. A discussion of the choice of absorption efficiency is beyond the scope of this paper; we refer to some of the few available measurements on *suspended* particles [*Mulholland and Choi*, 1998].

Most measurements of light-absorbing carbon are not well related to the precise definition just given. Ideally, methods used to measure BC, both at sources and in the atmosphere, should be interpreted in terms of their relationship with a "black carbon" standard agreed upon by the measurement community, which in turn should be related to the material's optical properties. Consensus on interpretation has not yet been reached for the current smorgasbord of available measurements. "Black" carbon is defined optically, by measuring the change in light transmittance or reflection caused by particles [*Gundel et al.*, 1984; *Hansen et al.*, 1984] or absorption [*Hitzenberger*, 1996]. However, the attenuation of light by particles collected on a filter is usually enhanced over that of suspended particles [*Horvath*, 1993; *Petzold et al.*, 1997; *Bond et al.*, 1999a] and cannot be used without adjustment.

Most source-characterization studies do not measure black carbon, but rather so-called “elemental” carbon (EC). Different from the analytical chemistry definition of pure, graphitic EC, the nomenclature commonly used in atmospheric chemistry refers to an operational definition based on the refractory behavior of carbon [*Huntzicker et al.*, 1982]. This type of measurement is widely used in air-quality and source-characterization applications. Even the results of similar methods, such as “thermal-optical” measurements, may differ based on the procedure used [*Schmid et al.*, 2001; *Chow et al.*, 2001], although the repeatability of identical procedures appears good [*Schauer et al.*, 2003]. Research on this method has identified artifacts associated with the adsorption of gaseous species [*Cadle et al.*, 1983; *Kirchstetter et al.*, 2001], the presence of metal oxides in the sample [*Novakov and Corrigan*, 1995; *Martins et al.*, 1998], and the charring of non-black carbon during analysis [*Yang and Yu*, 2002], but most reported measurements do not provide enough information to correct for these uncertainties. We have treated most measurements of EC as equal to BC, introducing an additional uncertainty to account for the measurement questions. Future work should include a treatment of the relationship between each measurement method and the quantity of interest—that is, light absorption. Recent measurement comparison efforts [*Schmid et al.*, 2001; *Weingartner et al.*, 2003] will assist in this regard. Further, carbon that absorbs light may not be black, and its molecular form may differ from that of BC [*Smith and Chughtai*, 1995; *Dobbins et al.*, 1995; *Bond*, 2001]; these differences are implicitly ignored in our treatment, but should also be addressed in future work.

2 Methodology

An overview of our calculation procedure is shown in Figure 1. Our approach to estimating emissions is based on combining fuel consumption data and application of combustion technologies and emission controls, and is similar to that described in *Klimont et al.* [2002]. The total emission for each country and species is given by the sum over all fuel/sector combinations. Emissions for a fuel/sector combination, in turn, are given by the contributions of all technologies within that sector. Total emissions for each species and country are:

$$Em_{j,k} = \sum_{l}\sum_{m} FC_{k,l,m}\left[\sum_{n} EF_{j,k,l.m,n} X_{k,l,m,n}\right] \tag{1}$$

where,

j,k,l,m,n = species, country, sector, fuel type, fuel/technology combination
Em = emissions;
FC = fuel consumption (kg/year);
EF = emission factor specific to fuel/technology, including the effects of control devices;
X = fraction of fuel for this sector consumed by a specific technology; ΣX=1.0 for each fuel and sector.

Our earlier papers have discussed some of the difficulties in developing BC and OC inventories from available data on particulate mass emissions [*Bond et al.* 1998, *Streets et al.* 2001]. Most reports on particulate matter (PM) emissions discuss total mass, because the important differences in the behavior of PM with different chemical compositions have only recently received attention. Where measured emission factors of black and organic carbon are not available, we have estimated them based on mass emission measurements combined with data on the submicron and carbonaceous fractions of the emissions as described in earlier papers. The net BC emission factor for submicrometer particles is given by:

$$EF_{BC} = EF_{PM}\, F_{1.0}\, F_{BC}\, F_{cont} \tag{2}$$

where EF_{PM} is the bulk particulate emission factor in g/kg; $F_{1.0}$ is the fraction of the emissions with diameters smaller than one micrometer, intended to separate BC from larger particles such as ash and char; F_{BC} is the fraction of the fine particulate matter that is black carbon, and F_{cont} is the fraction of *fine* PM that penetrates the control device. For combustion without emission controls, F_{cont}=1.0.

In this study, our values of EF_{PM} usually refer particulate matter with diameters below 10 μm (PM_{10}). When these are not available, we have used values of total particulate matter (TPM). Values of PM_{10} and TPM are similar for many types of combustion, but not all; coal combustion is a notable exception. In either case, the value of $F_{1.0}$ refers to the fraction of the *relevant* emissions that is submicron. Our values of F_{cont} are derived from size-resolved measurements and reflect the behavior of submicron particles, as most control devices capture fine particles less efficiently than larger particles. A calculation similar to Equation 2 applies for organic carbon. Here, we differentiate between the terms *organic carbon* (OC), or the purely carbonaceous mass of non-black carbon, and *organic matter* (OM), which includes the hydrogen and oxygen bound to this carbon. OC is the quantity resulting from thermal analysis of carbon aerosols, and the "BC-to-OC ratio" is sometimes reported. In order to estimate the mass of primary particles and the effects on radiative forcing, the mass of organic *matter* is needed; the ratio between organic matter and organic carbon is source-dependent. Defining this ratio as R_{org}, emissions of organic matter are:

$$EF_{OM} = EF_{PM}\, F_{fine}\, F_{OC}\, R_{org}\, F_{cont} \tag{3}$$

In this paper, we will present emissions of organic *carbon* (that is, without the factor R_{org} applied) for comparability with similar discussions in the community. In our model, each source has its own value of R_{org}, so that we have the ability to output distributions of either OC or OM.

Modeling and measurement studies related to climate have typically defined "fine" particles as those below 1.0 μm in diameter. Health-related studies usually consider particulate matter with diameter below 2.5 μm, or "$PM_{2.5}$". We have continued to tabulate particles below 1.0 μm for comparability with the climate community. Not only do larger particles have shorter atmospheric lifetimes, but it has been shown both theoretically [*Bergstrom*, 1973] and experimentally [*Dillner et al.*, 2001] that the mass extinction efficiency of light-absorbing carbon is lower for these larger particles. However, in order to compare with inventories developed by regulatory agencies (e.g., U.S. Environmental Protection Agency), tabulations of $PM_{2.5}$ will be desirable. We have also begun to compile values of $F_{2.5}$ in anticipation of these opportunities. For most combustion-generated carbonaceous particles, $PM_{2.5}$ is about 10-15% higher than $PM_{1.0}$ (e.g. *Durbin et al.* [1999a]). Inventories of BC and OC in the $PM_{2.5}$ range might be 10-15% higher than the values presented here. *Cooke et al.* [1999] estimated global "bulk" BC and OC emissions as 25% and 40% higher than submicron particles, respectively. Their "bulk" emissions, however, include some particles larger than 2.5 microns.

2.1 Technology divisions

As we discuss in Section 4, most of our fuel-consumption estimates are based on data from the International Energy Agency [*IEA*, 1998a, 1998b]. From the database of fuel consumption, described below, we extract over 50 combinations of fuel and usage. Within some of these fuel/usage combinations, there are practices that have distinctly different emissions. We further divide the fuel consumed in each of these sectors into different technologies (X in Equation 1) and apply appropriate emission factors for each. For example, in the industrial sector, we identify several combinations of combustion devices and emission controls (for example, "stoker with cyclone"). These latter divisions go beyond those already existing in the fuel-consumption data; they are specified by us for each world region and may be defined for each country if known.

This procedure provides several advantages over simply assigning emission factors to broad sectors such as “industrial” or “domestic” combustion:

- The choice of each emission factor is based on physical principles.
- Differences between regions can be represented as a function of technological choice. It is often assumed that emissions from a certain type of fuel in developing countries can be higher than those in industrialized nations; the mix of technologies in use provides a physical explanation.
- The change of emission *factors* over time can be represented physically, as a change in the relative fractions of technologies with different emissions. (The present paper, however, concentrates on current emissions.)
- High-emitting *technologies*, rather than just sectors, can be identified as targets for mitigation.

2.2 Inventory calculations

We developed a program (**S**peciated **P**articulate **E**missions **W**izard, or SPEW) to provide flexibility and transparency in the production of global inventories. In this document, we describe its application to submicron black and organic carbon. SPEW is developed in a relational database environment. It tabulates values of emission characteristics from the literature, including PM emission factors and BC and OC fractions. The journal reference and comments are recorded, as well as the mean, standard deviation and number of data points, and each value is keyed to a fuel/technology combination. The user interface provides statistical information about the measurement population, including estimates of the expected value and variance when normal and lognormal distributions are assumed. Based on this information, the user chooses an emission factor for each technology and region. There is the ability to enter regionally-specific emission factors, so that this emission inventory will evolve as more information is acquired from local scientists; country-specific emission factors can be also entered when available. However, we have preferred to represent regional differences by specifying regional technology divisions (*X*), rather than by varying emission factors.

One of the guiding principles of SPEW architecture is that no relationships are "hard-coded"; that is, definitions of activity disaggregation and emission factors (for example) are located in databases, not in the code itself. This approach allows simple alteration of the calculation parameters; it also provides data traceability, since we archive the supporting files for each successive version of the inventory. When emission data are disseminated, the data set is frozen, given a version number, and retained as permanent documentation. The data set presented in this paper is designated version 4.00.

2.3 Emission gridding

Although the fuel-use data (discussed in Section 4) contain only country-level estimates, the spatial distribution of emissions in large countries can be improved by first distributing the emissions on a state or provincial basis. We have accomplished this by using additional sources to provide fuel-use distributions for some of the largest fuel/usage breakdowns in the United States (diesel, jet fuel, and residential wood; *Energy Information Administration* [2001]); China (industrial, residential, and coking coal; *Fridley and Sinton* [2001]); and India (transport, power generation, and biofuels; *TERI* [1996]).

Emissions on the country level (or the state or provincial level for the United States, China and India) are distributed on a 1° x 1° grid according to proxies appropriate for each type of technology. Emissions associated with a fuel/technology combination can be gridded to any defined proxy for which data are available; for most technologies, the proxy is total population. For example, a grid cell with 2% of the population of a country would receive 2% of the emissions of that country. Technologies that use gridding proxies other than total population are listed in Table 1. The number of gridding proxies that can be defined is unlimited, to allow better spatial distribution of emissions in the future.

Open biomass burning is gridded according to the product of land cover and fire counts during the period 1999-2000. Fire counts appeared unreliable in some regions, particularly Central America, the Northern part of South America, and Eastern and Southern Africa. Low fire counts could not account for the moderate to high emission estimates. For these locations, biomass burning emissions were gridded according to land cover only.

Emissions from ocean-going ships are not concentrated around the countries to which the fuel consumption is assigned. We gridded international shipping emissions according to the sulfur distribution given by *Corbett et al.* [1999].

3 Uncertainties

While large uncertainties in the source strength of carbonaceous aerosols are widely acknowledged [e.g. *Cooke et al.*, 1999; *IPCC,* 2002], few attempts to quantify those uncertainties and their sources have appeared in the literature. As a direct result, estimates of climate forcing and other environmental effects have not accounted for these uncertainties. “Agreement” between measurements and models cannot be assessed without comparing confidence intervals. In fact, assimilation models that incorporate aerosol information [e.g. *Collins et al.*, 2001] require uncertainties to determine optimal adjustment of the model. Further, quantified uncertainty can both identify the most critical areas for further research and mark scientific progress when it is reduced. Therefore, we suggest that the identification of uncertainties and their propagation through general circulation models is just as important as calculations based on the central values.

We have made a preliminary assessment of uncertainties in source strengths that draws on previous work [*Cullen and Frey*, 1999; *IPCC*, 2000; *Suutari et al.*, 2001] and is applicable to a global calculation. The standard uncertainty propagation recommended by most statistical textbooks and by *IPCC* [2000] is valid only when the uncertainties are normally distributed, when the data are uncorrelated, and when the 95% confidence interval is less than 60% of the mean [*Olivier*, 2002]. None of these conditions is applicable to inventories of particulate matter. We expect to refine the uncertainty estimates presented here as a result of continuing work. Some of the methods we have used to combine uncertainties are not statistically rigorous. However, these procedures do assist in answering two questions: (1) How well are emissions of carbonaceous particles known? and (2) What and where are the major sources of uncertainty in these inventories?

Throughout this paper, the term "uncertainty" will refer to a 95% confidence interval about the mean. In this section, we discuss the development of uncertainties in emission factors and technology divisions. Uncertainties in fuel-use are also included and are discussed in Section 4.5. Often, true quantification of uncertainty is impossible, and "judicious guesswork" has been used liberally in the estimations we present here. Of course, a more scientifically justifiable approach is desirable. However, the lack of knowledge demands, not precludes, estimation of uncertainties before an emission estimate is used as the basis of modeling studies. Otherwise, the inherent uncertainty may be forgotten. For example, the current IPCC estimate of forcing by black carbon from fossil-fuel burning has a central value of +0.2, with a factor of two uncertainty. However, the range presented is largely due to the mixing state of black carbon. As we will show, the uncertainty in the IPCC estimate of forcing by black carbon is likely underestimated; it is similar to the uncertainty in the source strength alone, while the atmospheric lifetime and radiative properties also affect the radiative forcing and have comparable uncertainties.

3.1 Uncertainties in emission factors

For this global approach, the smallest region considered is 1° x 1° (approximately 100 km x 100 km). We assume that the number of sources is large, so that the mean and standard deviation within the smallest region of interest are nearly identical to the mean and standard deviation of the population. Therefore, the uncertainty results entirely from estimating the emission factors for the population, not from the limited population within a grid box. This assumption may underestimate the uncertainty for the case of large power plants; however, as we will discuss, emission factors of *carbonaceous* aerosols from these plants are low.

When values of EF_{PM} are taken from U.S. EPA's AP-42 document [1996], we assign uncertainties based on the emission factor rating in that document. The confidence intervals are not sanctioned by the U.S. EPA, which developed the ratings to provide qualitative uncertainty estimates only. Many of our emission factors are based on our own tabulation. For these, we use the available measurements to estimate population parameters, assuming that EF_{PM} is lognormally distributed. In many treatments, the methods of uncertainty propagation apply only to normal distributions for emission factors, even when the lognormal distribution is considered acceptable [e.g. *IPCC*, 2000]. We will argue the scientific case for the lognormal distribution in the next paragraph. Physically, the lognormal distribution implies that it is equally likely to find values that are twice the most common value and half the most common value. The normal distribution is better when values of zero are about as likely as values that are twice the most common value.

The lognormal distribution is preferred to the normal distribution for variables which must be positive and for which the coefficient of variation is greater than 30% [*Cullen and Frey*, 1999], as is the case for emission factors. Some populations of vehicle emission factors have been shown to follow the gamma distribution [*Zhang et al.*, 1994], and our analysis of a large database on diesel emissions [*Yanowitz et al.*, 2001] agrees with this assessment. Values of EF_{PM} from three other data sets are somewhat better represented by a lognormal distribution, as assessed by a two-sided Kolmogorov-Smirnov goodness-of-fit test [*Massey*, 1951]. These data sets include the fireplace emission factors given by *Fine et al.* [2001, 2002], the cookstove tests of *Oanh et al.* [1999, 2002] and the emission factors obtained by varying the operating parameters of three baby-taxis given by *Kojima and Khaliquzzaman* [2002]. Furthermore, the lognormal distribution provides a reasonable fit to the data, even when the gamma distribution yields a slightly better fit. On the other hand, the normal distribution yields a poor fit to particulate emission factors in all the cases we examined. We assume, then, that the lognormal distribution describes emission factors. For a lognormally distributed parameter x, we estimate the log-mean and log-standard deviation, μ and σ, with the average and standard deviation of $\ln(x)$, respectively. Better estimates of μ and σ could be determined by "bootstrapping" calculations, such as those described by *Cullen and Frey* [1999], although we have not attempted this more in-depth approach.

There is an important distinction between the *expected value* and the mean of the lognormal distribution. The expected value is the relevant quantity for determining average atmospheric concentrations or radiative forcing. For the normal distribution, the expected value is the same as the mean. For the lognormal distribution, the expected value is *not* the same as the exponentiated mean of the log-transformed data (i.e, $\exp(\mu)$ or the geometric average). The expected value depends on both the mean μ and the standard deviation of the log-transformed data, and is given by:

$$E(x) = \exp(\mu + 0.5\,\sigma^2) \tag{4}$$

The expected value is always greater than exp(μ), so that using the geometric average of emission factors will underestimate both emissions and atmospheric concentrations. The difference between E(x) and exp(μ) is small for a narrow distribution (10% if $\sigma_{\ln(x)}$=1) and large for a broad distribution (50% if $\sigma_{\ln(x)}$=3). We estimate the confidence interval for the population mean as:

$$\text{c.i.}_{(1-\alpha)}=\exp\left(\mu \pm \frac{\sigma}{\sqrt{n}} t_{\alpha/2,n-1}\right) \tag{5}$$

where *n* is the number of observations and α is one minus the desired confidence interval (95% in this case, or α/2=0.025).

3.2 Composite emission characteristics

As discussed previously, when emission factors of BC and OC are lacking (as is often the case), we assemble them by multiplying tabulated values of EF_{PM}, F_{BC} and F_{OC}, F_{fine} and F_{cont}. The lognormal distribution is appropriate for EF_{PM}, but not for some of the other values, because it can easily result in unphysical values for variables that are bounded above.

Since we assume lognormal distributions for some parameters and normal distributions for others parameters, the uncertainties cannot be combined analytically. The uncertainty in the product distribution could be estimated by combining parameters with a Monte Carlo or other sampling procedure, as described by *Cullen and Frey* [1999] and many other texts. However, the large number of individual emission factors precludes this approach. Instead, we approximate the expected value as the product of the expected values; this treatment is exact if the underlying distributions are lognormal. The upper confidence interval is created by treating the upper confidence intervals in the underlying distributions as uncertainties in a lognormal distribution and combining them in quadrature. The lower confidence interval is created in the same fashion, but separately from the upper confidence interval because it may not be symmetric about the mean.

The uncertainty involved in combining emission characteristics may be reduced by simply measuring the emission factors of interest: submicron BC and OC. We have used these EFs where available, if the combustion is thought to be representative.

3.3 Uncertainty in technology divisions

Identifying combustion practice and control devices as the factors that most affect BC and OC emissions, and accounting for these variables in the calculation procedure, implicitly sets the task of quantifying the prevalence of each technology. This information is difficult to obtain under the best of circumstances, and has not been tabulated for many regions. While this technology-based approach has the potential to represent emissions more accurately, it also has the potential to introduce more detail than is warranted based on available information, possibly engendering false confidence in the results.

We include uncertainties in the technology divisions, so that our high-emission scenario has a greater contribution from higher-emitting technologies. This approach allows us to identify whether uncertainty results from limited knowledge about *technology* or about the *emission factors* themselves. This distinction could suggest approaches to reducing uncertainty: more measurements are needed if most of the uncertainty results from the emission factor, but tabulating in-use technologies would yield a greater improvement if uncertainty in emission factors is low relative to uncertainty in practice.

The uncertainties in technology divisions are based entirely on estimation. In regions where we feel that our knowledge about the source population is reasonable, we increase the fraction of the higher-emitting technologies and decrease the fraction of the lower-emitting technologies by 10% of the total. When we believe our understanding is more uncertain, we might alter the technology fractions by 30% of the total.

3.4 Summing emissions

Uncertainty in the sum of two variables x_1 and x_2 that have uncertainties σ_1 and σ_2 respectively, is given by:

$$\sigma_{sum}^2 = \sigma_1^2 + 2\ \mathrm{cov}(x_1, x_2) + \sigma_2^2 \qquad (6)$$

where $\mathrm{cov}(x_1, x_2)$ is the covariance of x_1 and x_2. For two uncorrelated variables, it is appropriate to add the *variances*; that is, the uncertainties sum in quadrature. When the two variables are perfectly correlated, the uncertainty itself is summed. The uncertainty in the sum of uncorrelated variables is lower than the uncertainty in the sum of correlated variables, because of the reduced likelihood that two high or two low values occur simultaneously. If the two variables are neither uncorrelated nor perfectly correlated, combining confidence intervals is more complicated. As many other factors prevent the precise calculation of emission uncertainty, we do not use these more complex relationships. Whenever uncertainties must be combined, we determine whether the linear (correlated) or quadrature (uncorrelated) approach is more appropriate.

When one fuel/sector combination is divided into different technologies, the variations in the fuel-use assigned to each subset are necessarily correlated. An uncorrelated component is introduced if each subset has different emission factors for each subset, so that the resulting total emissions are partially correlated. Similar to the presentation by *Suutari et al.* [2001], we derived an analytical solution to the total uncertainty for the fuel-usage subset. However, there may also be some correlation for the EFs within each subset if they are drawn from similar measurement groups. We also believe that the detailed algebra is unwarranted given the level of confidence in the uncertainty itself. For that reason, we take a simplified and conservative approach. We treat emissions from each fuel/technology subdivision as perfectly correlated with the other subdivisions of the fuel/sector combination, so that the uncertainty for each fuel/sector combination is obtained by summing the *uncertainties* of the subsets. This approach somewhat overestimates the uncertainties, but the other extreme is implausible: summing the *variances* would cause the uncertainty to decrease as more fuel/technology subdivisions were added, even if there were substantial uncertainty in those subdivisions.

Emissions from separate fuel-usage categories should be uncorrelated with each other. Total emission uncertainty for each country is evaluated by summing the *variances* of the fuel-usage categories. Emissions for each grid cell are calculated similarly: we sum *uncertainties* in subsets of fuel-usage categories and *variances* to aggregate uncertainties.

4 Fuel usage

4.1 Fossil fuels

We begin with fuel-use data from the International Energy Agency (IEA), which include 38 fuels that are relevant to emissions and 138 countries. IEA provides information on the flows of fuels: imports, exports, and sectoral consumption; here, the term “sector” refers to broad classes of usage, such as power generation, industry, and residences. IEA data also divide consumption into various activities within each sector; when disaggregation may improve the representation of emissions, we separate these activities. For example, although both power generation plants and coke ovens fall under the “transformation” sector, their emissions are quite different. We aggregate some fuels when separate emission factors are not available and when the emissions are expected to be small. For example, we combine biogas with natural gas, since the end-use combustion may be similar. We have not tabulated emissions from some usage in the transformation sector, in particular the manufacture of “smokeless” fuels or coal gas. These activities are specified in IEA data and may be included in future inventories. They are currently excluded due to lack of information on the magnitude or even existence of emissions.

Previous emission inventories have used fuel-use data from the United Nations (UN) [*Cooke et al.*, 1999]. Our comparisons between the UN and IEA databases show few differences; values are often identical to the last decimal place. There are some dissimilarities in the divisions of fuels between sectors, and in the assignments of the usage breakdowns within sectors. In general, we have found that the IEA data have a greater number of usage breakdowns, and that production, imports, exports and consumption are balanced unless identified as “statistical differences.” We examined these statistical differences with the idea of apportioning them to various sectors as first described by *Cooke and Wilson* [1996]. In the IEA data, the magnitudes of these differences are small relative to total consumption, but not relative to smaller sectors such as residential consumption. Persistent statistical differences could be caused by misreporting in a single sector, especially if record-keeping is preferentially poor in that sector. For that reason, we have not apportioned the statistical differences among sectors, but include uncertainties in total fuel consumption.

Although confidence intervals are not provided for fuel-use data, we estimate uncertainties in consumption for each fuel-usage combination. The quantity of fuel used is generally well known if the fuel passes through official channels. Again, we are presenting uncertainties as 95% confidence intervals relative to the mean. Our assumptions of these uncertainties are: 10% for the industrial and power-generation sectors, 20% for liquid fossil fuels in the residential sector, and 33% for coal in the residential sector.

We estimate a higher uncertainty for coal consumption in the residential sector because this fuel can be mined directly and does not have to undergo a distillation process. For that reason, compulsory quantification is less likely. In the residential sector, consumption estimates are not constrained by electrical output (as in power-generation) or output of goods (as in industry). As consumption in this sector is also smaller, a small absolute uncertainty can lead to a large relative uncertainty. For example, "statistical differences" in coal use (i.e. unexplained consumption) compared with power-generation consumption are 6% and 0.2% in China and the U.S., respectively. The statistical differences are 30% and 9% of the residential consumption for the same two countries. We have not attempted to account for the use of other fuels that are misreported or that pass through illicit channels. While it is quite possible that such usage exists, it is difficult to quantify because it may be sensitive to both governments and consumers.

IEA data have the disadvantage of lumping smaller countries into categories such as "Other Africa" or "Other Asia". As these divisions represent a small fraction of the fuel-use in a region, we accept that inconvenience, calculate the emissions for the lumped countries, and distribute them according to population as described in Section 2.3.

4.2 Biofuels

We have used IEA estimates for usage of biofuels, including wood, agricultural waste, animal waste, and charcoal. While IEA has traditionally focused on tabulating energy from fossil-fuel use, the agency began greater efforts to include renewable fuels during the 1990s. We used IEA statistics on consumption of charcoal to determine emissions at end-use, and data on fuelwood used for charcoal production to determine emissions from that activity.

IEA data are assembled by examining reports from countries and organizations, including the United Nations, the Food and Agriculture Organization (FAO) and its subsidiary, the now-defunct Regional Wood Energy Development Programme (RWEDP). Other data sources consulted by IEA, are given in the notes to the IEA statistics volumes. IEA data match tabulations in the RWEDP database (available at www.rwedp.org), as well as those in a detailed report on biofuel energy in Africa sponsored by FAO [*Amous*, 1999]. For Africa, we have used the detailed report to divide fuel use in IEA's "Other Africa" category into the component countries. These organizations have expended great effort in understanding biofuel usage through working with local and national energy and forestry offices over many years. While FAO data of earlier years tended to underpredict fuel consumption, recent IEA data appear to have corrected that problem and are more in line with higher estimates. We have not yet had the opportunity to compare the IEA database with recent work by *Yevich and Logan* [2003], which became available after much of the present work was complete.

Estimates of biofuel combustion are approximate at best, and we assume uncertainties of 100% for the residential sector, 50% for industrial usage, and 20% for power-generation. Biofuel production and consumption are not usually measured on an annual basis. They may be estimated from population surveys, per-capita consumption, and, sometimes, economic data [*Auke Koopmans, FAO, personal communication*, 2001]. Measurement surveys typically occur in only one year (or even season) and subsequent years are extrapolated based on population data. *Smith* [1987] summarized several per-capita estimates for different countries; some of these differed by factors of 3-4 within the same country. *Kituyi and Kirubi* [2003] showed that extrapolating a single day's fuel-use measurement could overpredict fuel use by 50%. *Yevich and Logan* [2003], considering published values of per-capita usage, estimated uncertainties of –30% to +40% in Africa and –40% to +95% in Asia. These per-capita consumption figures can be "frozen" from studies more than a decade old [*FAO-RWEDP*, 1997]. Agreement between official values (e.g. United Nations, International Energy Agency, and official country statistics) does not imply confidence, because these reports often draw on the same sources. Table 2 summarizes a number of estimates for India, one of the best-studied countries. This variation is not observed in reports for other countries, probably because there is a lack of independently developed estimates, not because of greater understanding.

The uncertainty in quantifying biofuel use is inherent in the nature of the system. Wood and other biofuels are usually part of a complex system that meets a multiplicity of needs, including animal fodder and building materials, in addition to energy requirements [*Reddy*, 1983; *Barnard and Kristoferson*, 1985]. Often, people obtain fuelwood not by felling trees, but by removing small branches from living trees [*deLucia*, 1983]. In Asia (at least), most fuel comes from non-forest land such as agricultural land and roadside trees, while peri-urban dwellers may scavenge sawdust, construction debris, or mill waste [*FAO-RWEDP*, 1996]. These sorts of activities are obviously difficult to quantify. Types of fuel used vary seasonally according to availability, and surveys taken at one time of year may misrepresent the average situation [*Ralph Overend, NREL, personal communication,* 2001]. The types of fuel available to a household may also vary based on constraints such as land tenure, animal ownership, or storage space [*Barnard and Kristoferson*, 1985; *Leach and Gowen*, 1987], so that it is difficult to draw inferences about the general population without detailed observations. Inferring fuel use from delivered-energy needs requires assumptions about combustion and heat-transfer efficiencies of simple stoves, for which estimates range from 5-30% [*Leach and Gowen*, 1987]. In addition to normal cooking and heating for a household, biofuels may be used for cooking animal feed [*Wang and Fend*, 1996] and for celebrations or rituals [*Babu and Moorthy*, 2001]. There is also a range of non-household uses such as brick or pottery kilns, restaurant cooking [*Westhoff and Germann*, 1995], food drying, distilleries, and tire retreading [*Balla et al.*, 1991; *FAO-RWEDP*, 1998]. Finally, large pieces of solid fuel usually require kindling to start, and that may include small wood, leftover charcoal, kitchen or urban waste, or plastic bags—in short, anything that burns readily. We do not know whether these uses have been included in any or all of the country tabulations in the IEA database. Given the constraints just discussed, and the comparisons in Table 2, we believe that our uncertainty estimates are actually optimistic.

IEA data are given in energy units. We converted the data to a mass of "as-burned" fuel by calculating the lower heating value of burned matter from average values in the *PHYLLIS* database [*ECN*, no date]. The values used are: wood, agricultural residue, and municipal solid waste, 15 MJ/kg; charcoal, 24 MJ/kg; and dung, 12.5 MJ/kg. For dung, we assumed a lower water content and a higher heating value than that in *PHYLLIS* to account for the drying that takes place before combustion.

4.3 Waste combustion

When waste collection is unavailable, inconvenient or expensive, garbage may be burned in open piles or makeshift combustors such as metal cans. Common practices include combustion of waste at streetside and burning at landfills, either because of spontaneous combustion or deliberately to reduce waste volume [*FAO*, 2000]. An extensive characterization of waste generation for the United States is available [*U.S. EPA*, 1998a], but this work may not be representative of most of the world. We assembled per-capita waste generation rates from *Hoornweg and Thomas* [1999], *Vermenicheva et al.* [1999], and *Ministério da Ciéncia e Tecnologia Brasil* [1991] and applied these to the urban populations. The *United Nations Human Settlements Programme* [2001] reports the fraction of waste burned for a few cities, and we estimated the fraction burned in all urban areas based on these data points. These values are quite uncertain, as waste-burning practice could be highly localized depending on the available alternatives. Our estimates of open waste burning are about 24 Mtonne/year, including 12 Mtonne in Asia (including China and India) and 5 Mtonne in Africa. We assign uncertainties of 200% to these estimates.

Waste generation in rural areas is even more difficult to estimate, and we have not included it in the inventory. In developing countries, availability of packaged goods is usually limited, but the packages (including plastic) are burned when on hand. We have not included rural waste in the inventory. Neither have we accounted for combustion of industrial quantities of waste, such as used tires or electronic components, which may contribute to air pollution on a localized basis.

4.4 Open biomass burning

Unlike the data for fossil-fuel and biofuel burning, our biomass burning estimates are not developed specifically for 1996, but for a "typical" year in the mid-1990s. While there is large interannual variability in burning, the estimates provided here are intended for use in studies that are not specific to a given year. In particular, these estimates do not represent the unusual activity of the 1996 fire season [*Lavoué et al.*, 1996].

Quantifying global biomass burning involved a comprehensive search of available data including scientific papers, individual country communications to the IPCC, Global Fire Monitoring Center publications, and FAO statistics. A detailed description of the general methodology followed can be found in *Streets et al.* [2003a]. However, that paper discusses Asian estimates only; the additional sources of data used to create a global inventory are discussed here. Any deviations from the procedure given by *Streets et al.* [2003a] are also discussed in this section. This tabulation represents a new global estimate of biomass burning and is summarized in Table 3. Earlier work [*Streets et al.*, 2003b] used regionally-dependent uncertainties of biomass burning ranging from 10-200%. Those uncertainties tended to be lower when absolute quantities were lower, so that the higher values were more important in determining total uncertainty. For simplicity, we have used uncertainties of 150% here.

4.4.1 Africa

For Africa, forest burning values were derived from FAO [1997], and crop burning was calculated based on FAOSTAT following the method outlined in *Streets et al.* [2003a]. For some areas where data was unavailable, burning was assumed to be zero based on expert opinion. Savanna burning estimates were based on a combination of papers [*Delmas,* 1996; *Menaut,* 1996; *Hao and Liu*, 1994]. Burning estimates given in *Delmas* [1996] and *Menaut* [1996] were disaggregated and assigned to countries based on spatial distribution of emissions data in *Hao and Liu* [1994]. For tropical African countries in regions not included in either *Delmas* [1996] or *Menaut* [1996], *Hao and Liu* [1994] values were used. However, *Hao and Liu* [1994] values for Africa were adjusted upward (by 44%) to reflect more recent changes in burning practices (as reflected in *UN-ECE* papers). Other sources used included a number of country communications to the IPCC, *UN-ECE* [2000, 2001], and *Akeredolu and Isichei* [1996].

4.4.2 The Americas

Data concerning South and Central America (including the Caribbean) came from *Hao and Liu* [1994], country communications to the IPCC, and *FAO* [1997]. Crop burning was calculated based on *FAOSTAT* [*FAO,* 2001] except for select Central and South American countries for which data were available in the country communications for the IPCC, and for the United States and Mexico. Forest burning and crop burning were derived for Mexico from *SEMARNAP* [2001] and for the United States from *EPA* [2001]. Forest burning in Canada came from *Environment Canada* [1995] and *CIFFC* [2001]. Grassland burning in Canada and the United States were assumed to be zero because no reliable sources of data could be found. Other references used for the Americas were *UNECE* [1993, 2002], *Fearnside* [1996], and *Sanhueza* [1996].

4.4.3 Europe and the Middle East

International Forest Fire News [UNECE, 1990, 1992, 1993, 1995a, 1995b, 1999a, 1999b, 2000] papers were the most common data source used for Europe and the Middle East. Country communications for the IPCC were also used for many countries, as was CORINAIR '94 [*EEA*, 2000a]. *FAO* [1997] was used as the source for savanna burning in Afghanistan. FAOSTAT was used as a basis of calculation for crop residue burning in the Middle East, but not for Europe because burning practices are largely defined by government regulations that vary greatly by country. The majority of crop burning data for Europe came from country communications for the IPCC, and from CORINAIR '94 [*EEA*, 2000a]. For some European countries for which statistics could not be found, burning was assumed to be zero.

4.4.4 Oceania

Many areas in Oceania were considered to be too small to have a noticeable contribution to global biomass burning, and so were assigned burning estimates of zero. The obvious exception is Australia for which burning estimates were based on *Hao and Liu* [1994]. For a few of the larger islands, data detailing burning were available in country communications to the IPCC or in *FAO* [1997]. When that was the case, those values were used in this inventory. Likewise, for the larger islands for which crop statistics were available from the FAO, the *FAOSTAT* method of calculation was used to derive crop-burning estimates.

5 Emission characteristics

In this section, we discuss the development of emission factors. The discussion is fairly lengthy and contains detailed reviews of emission measurements and underlying generation mechanisms. Readers who are more interested in the final results are encouraged to skip to Section 6 after this cautionary paragraph, in which we provide three warnings on the estimation of BC emission factors. (1) BC is *not* predictable from overall stoichiometry (the balance of fuel and air provided to combustion). Its formation and destruction are limited by kinetics, not equilibrium states, and are governed by small-scale mixing, not average composition. The study of "soot" formation is an area with many outstanding questions, even for the simplest configurations and the purest fuels. (2) General correlations between BC and another product of incomplete combustion, carbon monoxide (CO), depend greatly on the source. This is evidenced by the fact that gasoline engines emit high concentrations of CO concentrations and low particulate matter, while diesel engines do the opposite. (3) General correlations between BC and sulfur are also source-dependent; these correlations are affected by the local consumption of sulfur-containing fuels, usually coal but also including diesel. Apparent correlations of these species in ambient air result from the collocation of sources, not necessarily from simultaneous generation. Predictions of BC and OC based on measurements of other species are likely to be erroneous as the regional mix of sources changes.

BC and OC emission factors must be based on direct measurements, and these measurements are discussed in this section. It is not possible to provide a full justification for the large number of choices for each emission characteristic and technology division. We have attempted to discuss only the choices that affect emission estimates the most. Readers are encouraged to contact the authors if specific questions are not addressed here. We have undertaken an extensive review of the literature in order to assign particulate emission factors (EFs) that are appropriate to each type of combustion. Our review has included examining each reference cited in previous inventories [*Penner et al.*, 1993; *Cooke and Wilson*, 1996; *Liousse et al.*, 1996; *Cooke et al.*, 1999], unless we were unable to obtain the reports, since measured EFs depend on both the combustion technology and the measurement method. In general, we have included only primary references, tracing the measurements back to their original presentation, to avoid the appearance of consensus that occurs when references cite each other. For that reason, this tabulation has eliminated some references that appear in other literature.

There are some exceptions to the rule of accepting only primary references. The U.S. Environmental Protection Agency's "Compilation of Emission Factors" (hereinafter referred by its report number, "AP-42") combines a number of measurements. We have examined the supporting information, but not the background reports if a recommended EF represents the compilation of tens of measurements. Other reviews (e.g. *Yanowitz et al.*, 2000; *Houck and Tiegs*, 1998) have examined the literature on hundreds of source tests with close attention to measurement methods and combustion types. We generally accept the results of those reviews. Finally, two useful tabulations report chemical speciation of particulate matter [*Hopke*, 1985; *U.S. EPA, SPECIATE,* 1999], and these cite some documents that are difficult to obtain. When other information is not available, we use the values from these tabulations even if we did not obtain the original reports.

Values of EF_{PM} for stationary combustion are given in Table 4 and Table 5. Mobile sources do not appear in the tables because they require a more extended discussion, which appears in Section 5.4.

Table 4 summarizes all sources except residential combustion of solid fuels. Particulate matter emission factors, as well as $F_{1.0}$, are largely drawn from AP-42. F_{cont} refers to penetration of *submicron* aerosol (typically larger than F_{cont} for total particulate matter), and are inferred from size-resolved data given in AP-42. Values of F_{BC} and F_{OC} are also given in the table, and most of the discussion in the text centers around these.

Many small, residential combustion sources are not prevalent in the U.S. EPA's jurisdiction and thus are not covered by AP-42. Table 5 tabulates values of EF_{PM} for residential combustion, and values of the other parameters required to determine BC and OC emissions (F_{fine}, F_{BC}, F_{OC}, and F_{cont}), as well as technology divisions (X in equation 1) are discussed in the text. The result of combining the emission characteristics is the BC and OC emission factors listed in Tables 6 and 7.

We will discuss differences between our work and the most detailed global BC inventory that was previously available [*Cooke et al.*, 1999]. We will also provide an expanded discussion of our chosen EFs when those choices have resulted in significant revision of global inventories. Differences in opinion, for example selecting one measurement and disregarding another, are not major contributors to differences between this inventory and previous work. Our choices result from applying an understanding of combustion technology to choose appropriate emission characteristics, and from drawing on data that has recently become available. This work, combined with the background in *Streets et al.* [2001], represents a complete re-examination of carbonaceous aerosol emission factors and global emissions of BC and OC.

5.1 Particulate matter from combustion

We begin by reviewing the ways in which particulate matter can be emitted from combustion processes. A detailed review is also given by *Lighty et al.* [2000]. The following discussion is confined to those aspects of combustion that affect the inference of BC and OC fractions from mass emission measurements. Fine particles in the atmosphere (those with aerodynamic diameters lower than some nominal size such as 1 μm or 2.5 μm) have different sources than do coarse (larger) particles. The same is true in combustion. The discussion here focuses mainly on fine particles, which have both longer atmospheric lifetimes and greater scattering and absorption efficiencies than coarse particles.

Black carbon (often called "soot" by the combustion community) is usually formed under conditions in which insufficient oxygen is present for complete oxidation of carbonaceous fuel to CO_2 (fuel-rich). Fuel-rich zones always exist when flame reactions are limited by mixing of fuel and air, so that all diffusion flames offer the possibility of soot formation, as the luminosity of candles attests. A minimum temperature is required to begin formation of black carbon [*Glassman et al.*, 1994]. In the post-flame zone, the soot may burn out if oxygen is present, and this process is enhanced at higher temperatures. The formation and emission of BC is therefore governed by the time-temperature history of the fuel and combustion products. Some of the extensive literature on soot formation has been summarized by *Haynes and Wagner* [1981] and *Smith* [1981]. Formation of BC from liquid droplets or coal tar introduces another level of complexity [e.g. *Solum et al.*, 2001]. Combustion processes can also emit *organic carbon*, which absorbs very little light. Organic vapors can condense onto existing particles, and they may form particles by nucleation if the concentration is high enough. These vapors may be products of incomplete combustion, or they may not have passed through a combustion zone at all; for example, pyrolysis of wood at low temperatures releases organic material that condenses quickly after it is emitted.

Rounding out the picture of fine particle formation, *inorganic compounds* in the submicrometer size range can form from vaporization of minerals and subsequent condensation, or from bursting of mineral inclusions in the fuel [*Flagan and Friedlander*, 1978; *Smith*, 1980]. Unlike carbonaceous aerosols, most mineral matter cannot be eliminated from the flue gas by oxidation.

Coarse particles are not formed within combustion; the residence times involved are insufficient for either building these particles or coagulating them from smaller ones. Rather, these particles are left over from large particles present at the start of combustion, although the initial particles may divide during the combustion process. Coarse particles may include both mineral matter and char, with the latter referring to the portion of carbon that never leaves the original fuel particle. They escape from the combustion zone into the exhaust when aided by higher air velocities, so their emission is enhanced by forced draft. Therefore, emissions of PM consist mostly of small particles if (a) large particles are not provided to the combustion, as is the case with most liquid fuels, or (b) air velocities are low, as is the case in most residential combustion.

The discussion above has presented a relatively simplistic picture of particulate emissions from combustion. Other confounding factors may exist: for example, interactions between different types of aerosols during formation, and elimination of fine particles through coagulation with coarse particles. The salient point is that *separate mechanisms*, governed by disparate aspects of the combustion process, are responsible for different types of particles. For example, black carbon emissions may scale with flame length, while emissions of mineral matter may scale with fuel mineral content, and one should not expect proportionality between emissions of carbonaceous particles and total particulate matter. Furthermore, the composition of total particulate matter cannot be used to represent that of fine particulate matter.

5.2 Coal: industrial processes

5.2.1 Pulverized coal

Technologies for burning coal vary widely (for examples, see *Babcock and Wilcox*, 1992). There is one notable distinction among these technologies: whether coal particles are suspended in the oxidizer or piled in a bed. Pulverized-coal or cyclone furnaces are examples of the former technology. There, soot forms in a cloud around the coal particle as volatile matter is ejected [*Seeker et al.*, 1981], but encounters a hot oxidizing environment shortly after it is emitted. The incombustible component (ash) remains suspended in the exhaust stream, so the primary emission from combustion of coal particles consists of mineral matter and not carbonaceous material. This dependence is obvious from the fact that AP-42 emission factors for pulverized coal are based on ash content.

Although the literature contains several detailed chemical speciations of pulverized coal burning emissions, many studies do not measure the carbonaceous fraction because it is expected to be so low. In the absence of other information, *Cooke et al.* [1999] assumed that 25% of the particulate matter emitted from pulverized coal burning is BC, and another 25% is OC. A review of the literature found no BC fractions of this magnitude for pulverized coal burning; total carbon fractions are typically below 1%. For this reason, our emission factors are substantially less than those in the previous work. However, volatile carbonaceous material is released during the combustion process, and either staged combustion [*Veranth et al.*, 1998] or poor mixing could result in higher carbonaceous emissions.

As most of the particulate emissions from pulverized coal are thought to be mineral matter, the division into various classes of particulate removal does not greatly affect the predicted emissions of *carbonaceous* particles. We have pursued these classifications to allow the later usage of this model for estimating emissions of *total* particulate matter. Divisions for power generation and industry in China were estimated as part of an earlier paper [*Streets et al.*, 2001]. Technology divisions for these sectors in India are taken from *Reddy and Venkataraman* [2002a]. For Europe and the Former USSR, technology divisions were derived from the RAINS model (www.iiasa.ac.at/rains) developed at the International Institute for Applied Systems Analysis (IIASA). These, in turn, are based on European studies [e.g., *Pfeiffer et al.* 2002; *Winiwarter et al.*, 2001], expert estimates and communication with national experts participating in the UN/ECE Task Force on Emission Inventories and Projections. No such data were found for North America (rather surprisingly), Africa, Latin America, and the Pacific region. Therefore, technology divisions for these regions were developed from the data for the four regions where this information was available. In future work, we will attempt to improve technology divisions for all world regions.

5.2.2 Stokers and heating stoves

Before pulverized-coal burners were developed, coal was burned on grates, with various feeding and airflow mechanisms designed to increase efficiency and decrease smoke [e.g. *Randall and Weeks*, 1909; *Babcock and Wilcox*, 1992]. Coal stokers, especially hand-fed units, were the predominant technology contributing to the "smoke nuisance" in the United States in the early-to-mid 1900s [*Davidson*, 1979] and in Europe for the previous centuries [*Brimblecombe and Bowler*, 1992]. Stokers are not used for modern power plants, but are still employed in smaller applications, as well as in countries with less-advanced technology.

Stokers, and coal beds in general, can have very high emissions of carbonaceous particles and especially of black carbon, because tar from some coals is eminently suitable for forming soot. Studies on liquid or gaseous fuels usually find that a minimum temperature of 1600 K is required for soot formation [*Glassman et al.*, 1994]; this temperature dependence may result from kinetic limitations, with the formation of the first aromatic ring being the rate-limiting step [*Frenklach et al.*, 1984]. However, coal tar contains large polyaromatic hydrocarbons that are favored as soot nuclei, [*Vanderwal*, 1996], so that soot formation begins at much lower temperatures—about 900 K [*Ma et al.*, 1996]. This means that low-temperature combustion *of coal* can produce black carbon, and this fact is relevant not only to stokers, but also to coal used in residential combustion and other industrial processes.

In stokers, the air supplied to the coal bed—the "primary" air—is usually insufficient to complete combustion, and a high concentration of products of incomplete combustion (PICs) exists downstream of the coal bed. The velocity of the primary air stream may be kept low in order to avoid lofting particles from the coal bed. After additional ("secondary") air is introduced, oxidation proceeds, but complete combustion relies on good mixing of the incomplete combustion products and the secondary air. Contrasting with the combustion of suspended particles, the hot, oxygen-starved conditions within the coal bed are ideal for producing carbonaceous material. The vaporization/condensation reactions that produce fine mineral particles can also occur in coal-bed combustion. *Ge et al.* [2001] gave measurements on a chain-grate boiler burning raw coal; F_{BC} was 0.10 and F_{OC} about 0.02. Recent research in U.S. stokers gave a similar BC/OC ratio [*James Schauer, University of Wisconsin, personal communication*, 2003].

Although lignite is burned in the same types of combustors as hard coal, its emissions can be quite different. It has a lower heating value than hard coal, resulting in less-efficient combustion. As discussed previously, stokers and heating stoves are thought to produce more BC than pulverized coal. However, the volatile matter in lignite is composed of lighter hydrocarbons [*Suuberg et al.*, 1978], rather than the tar in bituminous coal that is highly correlated with BC formation. Other studies have found that tar devolatilized from lignite [*Lucas and Wall,* 1994] and pulverized lignite particles [*Seeker et al.*, 1981] forms little or no "soot" compared with higher-ranked coals. Measurements of lignite in both industrial plants [*Bond et al.*, 1999b] and home heating applications [*Pinto et al.*, 1998; *Bond et al.*, 2002] show that the emitted particles absorb little light. For this reason, the values of F_{BC} given in Tables 4 and 5 are lower than those for hard coal.

Emission factors for anthracite, which is classified as hard coal, are not given in Table 5. Emission factors from anthracite are much lower than those of bituminous coal [*Butcher and Ellenbecker*, 1982; *Mitra et al.*, 1987]. We have ignored the lower emissions of anthracite because bituminous coal is most prevalent in the residential sector (over 90% of consumption in the United States) and because no global apportionment among the various grades of hard coal is available.Most of the emission factors for residential heating stoves in Table 5 were measured in the early 1980s; the lone recent exception shows much lower emission factors. The EF_{PM} of 12 g/kg is lower than the average of the measurements, in an attempt to account for recent developments in heating stoves.

5.2.3 Iron and steel industry

Particulate matter is emitted from many sources in the iron and steel industry, many of which release mostly metallic particles. We have estimated BC and OC from cokemaking (coke ovens) and ironmaking (blast furnaces) only. According to IEA consumption data, these uses constituted 16% of world hard coal use in 1996. We treat this usage separately because it could be a large contributor to emissions of carbonaceous aerosols. Coke is the residue of a particular class of coal, created by driving off the volatile matter by heating the coal under specified conditions. In many countries, coke ovens are subject to strict emission regulations regarding the capture of the lost carbon. We have not found values of F_{BC} or F_{OC} especially for cokemaking. Although the emissions have been studied extensively for health effects [e.g. *Chen et al.*, 1999], the focus has been on specific organic compounds or compound types and not on speciating the whole aerosol. *Mamuro et al.* [1979] reported that trace elements made up only 3% of PM emissions from a coking still. We have assumed that 95% of the emissions are carbonaceous, half BC and half OM.

In the absence of regulation, emissions could be vented directly to the atmosphere, as was common during the days of "beehive" coke ovens in the United States (early 1900s). The *World Bank* [1998] reports that uncaptured exhaust gas may produce 1-11 g/kg of PM; background documents for AP-42 (chapter 12.2) state that emission factors for vented exhaust may be around 40 g/kg (although this value is not a measurement and thus does not appear in the summary tables). The optimum temperature for making coke is around 900-1100° C [*U.S. Steel*, 1985]. Based on our experience with heating coal at this temperature, values in the range of 10-40 g/kg are quite plausible. We have chosen EF_{PM}=20 g/kg for uncaptured coke emissions, with a 100% uncertainty, and we use the same values of F_{BC} and F_{OC} as for captured emissions.

We also estimate carbonaceous emissions from blast furnaces. When the gas is captured, it is usually burned for heat after being filtered, and we assume that particulate emissions are zero. Uncontrolled emissions can reach 50 g/kg, but the particles are relatively large [*U.S. Steel*, 1985]. We assume that submicron emissions are 0.05 g/kg as given in AP-42; here, we do not account for blast furnace "slip", an intermittent event with high emissions. We have not found measurements of the carbonaceous fraction of these particles; *Mamuro et al.* [1979] measured trace metals comprising about 55% of the PM; with the associated oxides, they would compose about 70% of the PM. We assume that the remaining 30% is BC, not OC, because of the reducing atmosphere in the blast furnace, with a high uncertainty as usual for this speculative value.

Venting the exhaust from coking ovens and blast furnaces is not considered desirable, and estimates of the prevalence of these practices are scarce. Theoretically, one could manipulate energy statistics to estimate venting fractions, by comparing fuel usage in coke ovens with reported usage of coke oven gas. However, chemicals emitted from coke ovens are often recovered, not consumed for energy. Also, official fuel-use data are not of sufficient quality to support this derivation. For example, India reports no usage of blast-furnace or coke oven gas, although some must surely be recaptured. AP-42 (chapter 12.2) suggests that "bypass" emissions occur 4 hours per year, and we use an "uncaptured" fraction of 0.1% for the United States. We assume the same fraction for Europe and the Pacific, including Japan, another large producer of coke.

In 1996, China was the largest coke producer in the world. Based on a 1998 field survey in Shanxi Province, *Polenske and McMichael* [2002] reported that 7% of the facilities were indigenous or "beehive" coke processes; the survey was done after the official closure of these units. The *U. S. Embassy* [2001] reported that prior to cleanup in that province, 80% of the coke was made in informal coke ovens. The *China Energy Databook* [*Fridley and Sinton*, 2001] reported that in 1996, half of the coke production was "old coke" made in clay or dirt kilns, but it does not indicate what fraction of these were vented. The fraction of "old coke" increased sharply in the mid-1990s and appears to be a direct result of China's efforts to increase coke production. After the closure of the polluting units in 1998, Chinese coke production fell by about 15%, according to IEA data. Since 7% of the units were still "indigenous" after that time, we will assume that 20% of the coke was produced by beehive ovens, with a large uncertainty. Cokemaking contributes both a large absolute amount and a large uncertainty to our estimate of Chinese emissions; we note that the mid-1990s were somewhat anomalous in this regard, and that predictions even two or three years after our base year of 1996 should probably be lower.

We have found no information regarding the prevalence of “beehive” coke production in other countries (although one could argue that values ranging from 7% to 80% are little better than no information). According to *Reddy and Venkataraman* [2002a], most of the steel in India is produced by integrated steel companies. We have assumed that 20% of the coke required for steel outside these large companies, or 5% of the total, is produced without capturing the exhaust. For blast furnaces, we assume that 5% is uncaptured in the U.S. and Europe, and 10% is uncaptured in the rest of the world.

5.2.4 Brick kilns

Emission inventories from both India [*Reddy and Venkataraman*, 2002a] and Kathmandu [*Shah and Nagpal*, 1996a] identified brick kilns as a polluting industry, especially those fired on coal. In these kilns, raw fuel and bricks are heated together in different configurations. There are a variety of configurations for heating fuel and bricks. “Bull’s trench” kilns are said to have prolific smokestacks, smaller clamp kilns are sealed and may produce less smoke, and newer technology such as the vertical kilns manufactured in China may have lower emissions [*Tuladhar and Raut*, 2002]. While AP-42 gives emission factors for modern tunnel kilns, we have not found emission factors for other kilns such as Hoffman kilns, Bull’s trench kilns, or clamp kilns. In these devices, the combustion is similar to that in stokers or heating stoves, although without proper control of airflow and with a high chimney that may enhance burnout. We have used an average of the EF for heating stoves and stokers, with a 200% uncertainty, and the values of $F_{1.0}$ and F_{BC} from these heating stoves.

Reddy and Venkataraman [2002a] assumed that polluting brick kilns consumed 4% of industrial coal in India. Another estimate gives 1991 consumption in brick kilns as 14 Mtonne for India, or 20% of industrial coal use [*FAO-RWEDP*, 1993a]; the same report estimates kiln consumption in Bangladesh and Pakistan, and a comparison of data suggests that nearly all the industrial coal use in these countries is due to the brick industry. As not all kilns are of the worst type, we assume that 25% of the fuel-use in the industrial “non-metallic minerals” sector is used in polluting kilns.

5.3 Coal: residential combustion

We reported in an earlier paper [*Streets et al.*, 2001] that domestic burning of solid fuels results in both a large fraction of, and a large uncertainty in, total emissions. In our preliminary tabulation of EFs for residential coal, we combined all types of burning and applied a general emission factor to this sector. Because this sector is so important, have revised the analysis to separate cooking use, small heating stoves, and stokers such as those used in apartment buildings. Combustion in each region is a mix of these fuels; for example, in China, we assume that all rural fuel-use takes place in cookstoves or open fires, and all urban fuel-use takes place in stokers. Table 5 summarizes emission factors from the literature, as well as central values and uncertainties in emission factors.

Larger stokers are often designed so that both fuel addition and exhaust characteristics minimize emissions. Smaller heating stoves may not have these attributes, as shown by their much higher emission factors (12±11 g/kg, as opposed to 2.5±3.0 g/kg for residential stokers). In determining the expected value for heating stoves, we have excluded the highest point reported by *Jaasma and Macumber* [1982], where the coal was deliberately chosen for its smokiness. If that point were included, the expected value of EF_{PM} would increase to 20 g/kg; variability due to coal composition leads to large uncertainties. Emissions for coal-burning cookstoves or open cooking fires average lower than heating stoves; we have increased the uncertainty because other measurements, both in our laboratory [*Bond et al.*, *unpublished data*] and elsewhere [*Linwei Tian, UC Berkeley*, *personal communication*, 2002] have suggested even higher emission factors.

Streets et al. [2001] reviewed *total* carbon fractions for this type of combustion as 0.45-0.93. As we have found no published measurements of chemical speciation for submicron aerosol from heating stoves, we briefly discuss our own (as yet unpublished) measurements of emissions from bituminous coal. We burned five samples of coal from the U.S. and China at two different temperatures (800 K and 1100 K just outside the coal bed). BC fractions averaged about 0.5 and 0.6 at the two temperatures, respectively; the particles were more than 90% carbon and associated material. We use the values from the cooler temperature with high uncertainties.

5.4 Mobile sources

We expect regional differences in vehicle emissions for several reasons. First, the vehicle population in many locations has not been required to respond to increasingly stringent emission regulations, as it has in the United States, Europe, and other industrialized nations. Comparing control technologies, *Omursal and Gautam* [1997] found that Mexican vehicles lagged U.S. vehicles by about 15 years. Second, lower fuel quality in some countries leads to higher emissions. In some countries, the use of high-sulfur fuels precludes some emission-abatement technologies, such as catalysts. Diesel fuel itself may include heavier fractions in developing countries [*Karim*, 1999]. Finally, emissions are thought to have an inverse relationship with income [*Bradley et al.*, 1999], because people with fewer resources might have older vehicles and less-frequent maintenance. Even when vehicles are well-maintained on the average, a large fraction of emissions comes from poorly tuned or defective vehicles [*e.g. Lawson,* 1993], sometimes called "smokers". We prefer the term "superemitters," as high emission and obvious smoke are not always concurrent [*Sagabiel et al.*, 1997].

Regional inventories of vehicle emissions typically account for a range of variables, especially characterization of the vehicle population by weight class and age [*U.S. EPA*, 1995]. This degree of detail is not possible for a global inventory. Furthermore, the results of these calculations are too low in some cases because of “malperformance”, driver behavior, and altitude, so that the average EF can be dominated by factors not accounted for in these detailed studies. Ongoing work on the topic of “real world” emissions is summarized by *Cadle et al.* [2000]. We do not differentiate between light-duty and heavy-duty vehicles; emission factors per mass of fuel (not per distance) are very similar among vehicle classes [*Yanowitz et al.*, 2000]. As we will show, the major uncertainties are in the fraction and magnitude of high-emitting vehicles. Additional subdivisions, such as model years, do affect the totals but would not reduce most of the uncertainties in the present work.

Our approach consists of: (1) determining baseline emission factors for vehicles in regions where the more stringent United States or “Euro” standards have been implemented; (2) estimating a similar baseline for other regions; and (3) estimating the fraction of superemitters in each region. We also assume that in some regions, such as Eastern Europe, the baseline is mixed, with about 30% of fuel consumed by on-road vehicles built to more stringent standards. Many countries, from Australia to Thailand, have scheduled implementation of more stringent standards from the mid-1990s onwards [*Faiz et al.*, 1996]. A more detailed approach will be required to estimate BC emission trends in subsequent years.

Because of the difficulty of ensuring a representative vehicle sample and driving conditions, even the appropriate EF for vehicles in the United States is not well known, despite extensive studies. Dynamometer tests measure a few vehicles over a range of conditions; measurements in traffic tunnels and at inspection stations average emissions from many vehicles under a single condition. No procedure measures both the full spectrum of both vehicles and conditions. There is also some evidence for a systematic low bias in many of these studies. Dynamometer tests can be biased toward lower *emissions* by excluding high-emission conditions and driving cycles [*Faiz et al.*, 1996; *Clark et al.* 1999a], driver behavior [*Clark et al.*, 1999b], loaded vehicles [*Durbin et al.*, 2000] and lower *emitters*; the last occurs because people with poorly-functioning vehicles are understandably reluctant to volunteer for emission studies [*Wenzel et al.*, 2000]. Inspection stations usually use simple measurements, such as opacity, for which interpretation in terms of either mass or absorption coefficient is uncertain. Opacity and particulate matter are not well correlated [*Yanowitz et al.*, 1999], because light extinction depends on many particle characteristics, including size and chemical composition. Furthermore, the tests used (e.g. "snap-acceleration") may not represent actual operating conditions or correlate well with emissions based on more representative driving cycles [*Yanowitz et al.*, 1999]. Some procedures report the highest opacity experienced during the test [*Faiz et al.*, 1996], which obviously cannot represent overall emissions. With these caveats in mind, we discuss our choice of emission factors.

5.4.1 Gasoline vehicles

Most of the extensive measurement sets for gasoline vehicles examine only carbon monoxide (CO), nitric oxide (NO) and hydrocarbons (HC). However, *Chase et al.* [2000] measured 22 current-technology vehicles; PM emissions ranged from 0.005-0.2 g/kg with only a slight difference between low- and high-mileage vehicles. (For this calculation, we assumed fuel consumption of 0.14 liter/km.) *Durbin et al.* [1999a] measured 129 vehicles, including some that were out of compliance with emission regulations, and showed that PM emissions decreased with model year. For these data, a weighted average of model-year bins based on estimated road miles for each model year gives 0.15 g/kg; pre-1985 models average about 0.24 g/kg. Tunnel measurements by *Kirchstetter et al.* [1999] estimated EF_{PM} for light-duty vehicles as 0.11 g/kg. An older study of 22 vehicles reports PM emissions as 0.75 g/kg [*Williams et al.*, 1989], higher than the oldest models measured by *Durbin et al.* [1999a]. For regions where emission standards have been progressively tightened, we choose a value of 0.15±0.10 g/kg, assuming that the *Williams et al.* [1989] data are no longer representative of the vehicle population. For other regions, we use EF_{PM}=0.5±0.4 g/kg.

How much could superemitting vehicles contribute? *Sagabiel et al.* [1997] reported that smoking vehicles emitted about 10 times the PM of non-smokers for a sample of 23 light-duty vehicles, or 2 g/kg. *Durbin et al.* [1999b] estimated that 1.1-1.7% of the light-duty fleet consisted of smokers, which emitted about 15 times more PM than normal vehicles. *Hansen and Rosen* [1990] found that 10% of gasoline vehicles emitted 50% of light-absorbing particles, implying that superemitter EFs are about 10 times higher. Another study indicates that the emission rates of superemitters are "more than 100 times greater than new-technology vehicles" [*Lawson and Smith*, 1998]. We emphasize that the frequency of superemitters is not independent of the enhancement by these vehicles; it depends upon the baseline chosen, and whether that baseline averages "new" technology or the "normal" fleet.

Measurements of CO do not parallel enhancement of PM, but might suggest the fraction of vehicles that are poorly maintained. *Lawson* [1993] reported that 10% of the light-duty vehicle fleet in California emitted 60 percent of the carbon monoxide (CO), implying that the high-emitting fraction averaged 13 times greater than the rest. *Bishop et al.* [2000] reported over 3 million individual-vehicle remote-sensing measurements from the Denver area. From those data, we infer that "superemitters" are about 6% of the population and, on average, emit about 17 times the CO of the remainder.

Supposing a superemitting population that emits 10-15 times the U. S. average, the emission factor for these high-emitting vehicles is about 2 g/kg, in accordance with *Durbin et al.* [1999b]. Based on the preceding discussion, we choose a central value of 5% superemitters for the U.S. and similar regions, with a high uncertainty (1-10%). We have found no estimates of superemitter fractions for other regions; *Omursal and Gautam* [1997] report that 2% of light-duty vehicles inspected in Mexico City were barred from driving indefinitely, while another 25% did not meet standards at the time. Based on the discussion in the diesel section below, we assume a superemitter fraction of 20% for Asia and Latin America based on these reports and, somewhat arbitrarily, 20% for Africa and 10% for Eastern Europe and the former USSR.

Fractions of BC and OC are taken from data presented by *Gillies and Gertler* [2000], who reviewed source profiles in the SPECIATE database as well as transportation profiles developed for three other studies. The review found that most of the particulate matter (>85%) emitted from both gasoline and diesel vehicles is carbonaceous. Based on a reanalysis of data presented in that paper, we estimate F_{BC} as 34% ± 12% and F_{OC} as 21 ± 6%. We use F_{fine} of about 0.85 from *Durbin et al.* [1999a].

5.4.2 Two-stroke gasoline engines

In some regions, two-stroke gasoline engines are popular because of their high power at low speeds and ease of maintenance. These engines have markedly different emission characteristics than the four-stroke engines used in most passenger cars. Because intake of fresh air and fuel is simultaneous with exhaust of combustion products, unburned fuel and oil can escape from the cylinder easily.

Kojima et al. [2000] summarized measurements on five U.S. motorcycles averaging 6 g/kg. These authors also measured an uncontrolled two-stroke motorcycle under several different conditions, with average emissions ranging from 3-5 g/kg depending on the measurement method. The same motorcycle on a European transient driving cycle emitted about 19 g/kg. The U.S. Environmental Protection Agency recently tested five two-stroke motorcycles, with emissions ranging from 11-31 g/kg; larger motorcycles had lower PM emissions [*Matt Spears, U.S. Environmental Protection*

Agency, personal communication, 2002]. *Patschull and Roth* [1995] inferred mass emission rates from size distributions of particles. For an oil fraction of 2%, they reported emission factors from 15-90 g/kg depending on engine load and speed (with the highest value occurring at only one of eighteen load/speed combinations). *Kojima and Khaliquzzaman* [2002] reported opacity measurements for 700 "baby-taxis" (commercial three-wheeled vehicles) that could be interpreted as about 32 g/kg. *Faiz et al.* [1996] cited a study suggesting that the average emission from 167 Thai motorcycles was about 28 g/kg, also based on opacity measurements. In a different application of uncontrolled two-stroke gasoline engines, the *U.S. EPA* [1991] suggests values of 16-32 g/kg for non-road engines.

Emissions are affected by both the type of lubricating oil used and the fraction of oil in the fuel-oil mixture. *Patschull and Roth* [1995] showed an increase from 45 g/kg to 130 g/kg as the oil fraction increased from 1% to 4%, using the load and speed where maximum emission occurred. *Kojima et al.* [2000] reported measurements showing an increase of EF_{PM} from 8 to 35 g/kg as oil content increased from 1 to 7%. *Kojima and Khaliquzzaman* [2002] measured three "baby-taxis" under a range of conditions. High oil fraction, and mineral oil instead of the more appropriate "2T" oil, both increased emissions. We estimate EF_{PM} as 15±10 g/kg in most regions of the world. In Asia, where observed practices contribute to high emissions, we use EF_{PM}=30±20 g/kg. We do not have observations to support this high level of emissions in other world regions, although it is possible.

As *Patschull and Roth* [1995] measured most particles with diameters below 0.5 µm, we use F_{fine}=0.95. We have found no measurements of BC and OC from two-stroke engines. Based on solvent extraction and sample color, *Kojima et al.* [2000] suggested that more than 95% of the particulate matter from a two-stroke engine was lubricating oil. The samples were brown, implying that the particles absorb some light; so a small fraction of lubricating oil may meet our definition of "black" carbon although it is not elemental or graphitic carbon (and might not be measured as such with a thermal carbon analysis). If the unburned fuel and oil come directly from the intake, they have not passed through the temperatures required to produce black carbon, and the BC fraction is probably low. Emissions from these engines usually appear white or bluish, not black. We have estimated that BC is 5% of the particulate matter, with a factor of 4 uncertainty, and that the remainder is OC.

Similar to our earlier work [*Streets et al.*, 2003b], we estimated fractions of gasoline used in two-stroke and four-stroke engines by using populations of cars, motorcycles and other vehicles given by *World Road Statistics* [*IRF*, 2000]. These were combined with estimates of kilometers driven and fuel efficiencies for each vehicle type. We determined the fraction of fuel consumed in two-stroke vehicles by using the divisions given by *Kojima et al.* [2000] for five Asian countries, including India, and other information for China [*Tai Chan, General Motors, personal communication,* 2001]. *World Road Statistics* does not tabulate three-wheeled vehicles; while not as numerous as two-wheeled vehicles, they drive longer distances because they are typically used as taxis. Our estimate of two-stroke fractions include an upward adjustment to account for the number of three-wheeled vehicles tabulated by *Kojima et al.* [2000]. In India and China, motorcycles are numerous; about one-third of gasoline is consumed in two-stroke vehicles in India, and 5% in China. In the United States and Western Europe, automobiles are more numerous, and the two-stroke fraction is low (2%).

5.4.3 On-road diesel vehicles

Yanowitz et al. [2000], hereinafter *Yanowitz*, tabulated over 400 dynamometer studies on diesel vehicles. They reported a decrease in United States PM emissions after 1988 that follows government regulations, but at about half the specified rate. The PM emission factors from low-altitude tests in the *Yanowitz* database fall into three rough categories based on model year: pre-1988 (average 3.0±1.4

g/kg), 1988-1993 (1.6±0.8 g/kg), and 1994 onward (0.9±1.0 g/kg). The averages increase by about 50% for high-altitude tests. For North America and Europe, we take EF_{PM} for normal vehicles as 1.5±0.75 g/kg. This value is an average of the *Yanowitz* emission factors just given, weighted by the mileage driven of trucks with different ages given by the *U.S. Department of Commerce* [1997]. A weighted average using the emission factors summarized by *Clark et al.* [2000] gives a similar answer.

For countries that have not implemented the more stringent U.S./European diesel standards, we assume that normal vehicles emit about 3.5 g/kg. In the *Yanowitz* database, EF_{PM} averaged 3.3 g/kg for the 49 vehicles with model years prior to 1982 (those that were over 15 years old in 1996). *Mäkelä* [1995] used a value of 3.8 g/kg for Russia and the Baltic States. Furthermore, many countries have regulations regarding diesel smoke, and one common standard specifies a maximum of 65 "Hartridge smoke units" (HSU) at free acceleration. Assuming an extinction efficiency of 5 m^2/g for this smoke, we interpret the standard as an instantaneous emission rate of approximately 7.5 g/kg. Graphs in *Faiz et al.* [1996] suggest that emissions are approximately uniformly distributed for "normal" vehicles, so the average would be about 3.7 g/kg. (We remember that the free-acceleration test is unlikely to represent driving conditions, but more representative tests are not available for most regions.) Lastly, recent, independent estimates suggest a similar value for the baseline in Eastern Europe [*Klimont et al.*, 2002].

There are even fewer measurements of superemitting diesel vehicles than there are for gasoline vehicles. Measurements of "smoking" diesel vehicles that would fail California opacity tests averaged 8 g/kg [*McCormick et al.*, 2003]. Measurements of the highest 20% of emitters of diesel buses in Santiago de Chile suggest an average emission factor of 18 g/kg [*Faiz et al.*, 1996; assuming a fuel consumption of 0.5 liter/km, a typical value for older vehicles]. Data from Kathmandu, Nepal, suggest a high fraction of superemitters, with only 5-15% of vehicles passing the 65 HSU standard; the average emission factor for diesel vehicles above the standard is about 17 g/kg [*Shah and Nagpal*, 1996a]. Opacity measurements in Bangladesh suggest an average of 20-25 g/kg or greater [*Karim*, 1999]. (The emission factors in that study could be even higher because some vehicles saturated the smokemeter.) We choose an emission factor of 12g/kg for superemitters in all locations with a factor of two uncertainty.

The fraction of high-emitting vehicles obviously has a large effect on total emissions, and we have not found rigorous reports of this fraction for heavy-duty diesel vehicles. *McCormick et al.* [2003] cite recent results showing that 4% of newer vehicles and 25% of older vehicles fail opacity tests. Based on those measurements and the preceding discussion, we extrapolate light-duty measurements to the heavy-duty population and choose a central value of 5% superemitters for countries "similar" to the U.S., with a high uncertainty (1-10%). For Southeast Asian countries, *Shah and Nagpal* [1996b] assumed that 20% of diesel vehicles were "smoke belchers". A distribution of opacity measurements from buses in Santiago de Chile [*Faiz et al.*, 1996] also shows that 20% of the diesel vehicles are above our cutoff level of 7.5 g/kg. However, there are reports of much higher superemitter populations. Measurements in Kathmandu showed that over 90% of vehicles failed to meet the 65 HSU standard. A study in Dhaka found that only 14% of the 908 vehicles measured *passed* the 65 HSU standard; another similar study covered four cities, including Dhaka, and reported that only 36% were "black smoke emitters" [*Karim*, 1999]. We assume a superemitter fraction of 20% for Asia and Latin America, but with a high uncertainty.

Our fleet average EF_{PM} is: 2.3 g/kg for the U.S. and Western Europe; 4.4 g/kg for Eastern Europe/former USSR; and 6.4 g/kg for Asia/Latin America. For the U.S., the average is about equal to that for 1988-1990 vehicles in the U.S. EPA's *PART5* program [1995], and lower than those resulting from the analogous EU *COPERT* program [*EEA*, 2000b]. The rate for Asia is higher than that assumed by *Shah and Nagpal* [1996b], mainly because we selected a higher base emission factor. The available evidence suggests that the fleet-average EF in most locations is lower than that used for "underdeveloped" countries in earlier inventories [15 g/kg in *Cooke et al.*, 1999]. Data from Kathmandu [*Shah and Nagpal*, 1996a] are one exception.

We summarize traffic tunnel measurements here for comparison only, as these measurements do not encompass all driving conditions.1.4g/kg (1992 U.S., reviewed by *Cadle et al.*, 1997); 1.1g/kg (1993 Europe, *Weingartner et al.*, 1997); 2.5 g/kg (1997 U.S., *Kirchstetter et al.*, 1999); 0.65 g/kg (1999 U.S., *Gertler et al.*, 2001). With the exception of the *Kirchstetter* study, the results are converted to a mass basis by assuming a fuel efficiency of 0.35 liter/km. While our average EF_{PM} for the U.S. and Europe is lower than official European estimates, it is much higher than most of the tunnel results.

As PM is difficult to measure, it would be desirable to find a more easily-measured proxy to increase the size of the global dataset. We investigated the use of carbon monoxide (CO) for this purpose, because both CO and PM result from incomplete combustion, but rejected this possibility for two reasons. First, the results for 400 vehicles in the supplementary information for *Yanowitz*, as well as other analyses [*EEA Inc.,* 2000] show that CO emissions are not good predictors for PM, with poor correlations even when the data are binned by age ($R^2<0.4$). Although the ratio between CO and PM is consistent for the same vehicle under different driving conditions [*Clark et al.*, 1999c], the same relationship among different vehicles is poor. The two species do not even have similar statistical distributions, according to the *Yanowitz* database. Secondly, there is a lack of relevant data on CO that could be used as a proxy. A large database of CO emission factors in various world regions is developing from remote-sensing studies [*Bradley et al.*, 1999; *Bishop et al.*, 2000], but so far these data have been designed to address gasoline vehicles. Further analysis of these data sets may allow a more expanded treatment of diesel emission factors.

Fraser et al. [2002] have shown that BC fractions can range from 0.19-0.94, depending on operating conditions. Since a general profile of the variation of F_{BC} with driving conditions has not yet been developed, and driving cycles for each world region are not available, we use the general measurements provided by *Gillies and Gertler* [2000]. From their data, we estimated BC and OC fractions as 66%±16% and OC as 21±6%, respectively. The different particulate emission mechanisms for high-emitting and normal vehicles might result in a varied chemical composition. However, while high-emitting vehicles tend to have a slightly larger fraction of OC, that finding is not statistically significant [*Gillies and Gertler*, 2000]. Roadside measurements of EC and OC composition on both sides of the U.S.-Mexico border, which might have different vehicle mixes, were not significantly different [*Watson and Chow*, 2001]. We have assigned the same BC and OC fractions to normal vehicles and high emitters.

As mentioned previously, inferences of mass from opacity measurements are prone to error. However, we are using an *optical* definition of BC (7 m^2 absorption/g BC) because our application is radiative-transfer. As opacity is also an optical measurement, it could serve to bound BC emissions (although OC emissions are not similarly bounded). Theoretical calculations over a range of particle sizes and refractive indices suggest that absorption is rarely more than 75% of extinction at combustion aerosol sizes [*Bond*, 2000]. Combining this fact with a mass balance on the combustion process, we find that 85 HSU—approximately the fleet average reported for Kathmandu by *Shah and Nagpal* [1996a]— translates to a maximum of 7 g BC/kg fuel. Thus, our fleet average of 3.6 g BC/kg fuel could be low by a factor of two in this area where superemitters are prevalent, and this is within the uncertainty of our estimates.

5.4.4 Off-road engines

Several studies have examined higher emission factors from off-road diesel vehicles, which are subject to both fewer regulations and higher fuel sulfur content than on-road vehicles. Based on IEA statistics, we tabulate the following categories of off-road usage separately: ships, railroads, agriculture, industry, and residential use. Industry usage of diesel includes construction, mining, and logging, but this disaggregation is not available for all countries, so we use only the lumped "industrial" category. At this time, we do not consider recreational usage such as snowmobiles and pleasure boats, which may also be high emitters [*Bishop et al.*, 1999].

Emission factors for ships differ between two EPA reports [*U.S. EPA*, 1991, 2000], and we use an intermediate value of EF_{PM}=1.8±1.5 g/kg for both distillate and heavy fuel oil. (Note that the later reference includes the frequently-cited emission measurements reported by Lloyd's Register, and that emission factors *by fuel mass* are not highly variable with power.) For railroad locomotives, our value of EF_{PM}=2.7±2.1 g/kg is also taken from a *U.S. EPA* report [1997], and the high uncertainty accounts for the range of observed values in various operating modes.

The Nonroad Engine and Vehicle Emission study (NEVES, U.S. EPA [1991]) gave in-depth estimates of the contribution of non-road vehicles, assuming an EF_{PM} value of about 12 g/kg for tractors and 6-10 g/kg for construction equipment. The values exceeded measurements because actual use cycles were thought to have more transients, and hence greater emissions, than the test cycles used during measurements. A subsequent report [*U.S. EPA,* 1998b] found that emission tests simulating agriculture actually emit less PM (by about one-third) than the measurement test cycle, but that more

PM is emitted when simulating construction activities (factor of 1.2-2.0). Also, most of the PM emission estimates in NEVES were based on a study of vehicles done in 1973, and tests on 1991 vehicles show that their emissions have improved by a factor of 2-4 [*U.S. EPA*, 1991]. *Kean et al.* [2000] recommended EF_{PM} of 3.8 g/kg for farm equipment and 5-6 g/kg for other vehicles, based on U.S. EPA emission factors. *BUWAL* [1996] summarized the results of European studies ranging from 3.5-8.6 g/kg. *Samaras and Zierock* [1995] recommended emission factors in the range of 4.3-8.2 g/kg, where higher emission factors were associated with smaller engines. Following *Kean et al.* [2000], we use 4 g/kg for farm vehicles and 5.5 g/kg for other vehicles. We also assume the same fraction of superemitters in agricultural and construction sectors as in the on-road diesel population.

In the residential sector, stationary diesel generators may be used for power generation, especially when connections to electricity grids are unavailable or irregular. For this application, EF_{PM}=6±8 g/kg comes from AP-42 (Section 3.3). The high uncertainty accounts for increased emissions that may result from intermittent operation, poor maintenance, and possibly fuel adulteration in these engines. We have not found measurements of speciation particular to off-road usage, and use the same values of F_{fine}, F_{BC}, and F_{OC} as for diesel vehicles.

Finally, coal is used in railroad locomotives. In the absence of measured data, we accept the assumption of *Marsh* [1947] that 3%, or 30 g/kg, is transformed to smoke, but assume that only 50% of that mass is PM_{10}. As this combustion is similar to that in a stoker, we use the remainder of the parameters as described for stokers in Table 4.

5.4.5 Aviation fuel

We separate aviation fuel used for aircraft only. While turbine and piston engines presumably have different emission factors, we do not have enough measured data to represent this distinction. *Penner* *et al.* [1999] recommended a BC emission factor of 0.04 g/kg; *Petzold and Dopelheuer* [1998] and *Petzold et al.* [1999] reported 0.12-0.15 g/kg at medium thrust, with higher values at full thrust. The latter body of work used an engine that was thought to be higher-emitting than the average. We use an emission factor of 0.1 g/kg for EF_{BC}. These emissions are thought to be largely black carbon *[Petzold et al.*, 1999] and we use a BC:OC ratio of 4 to determine the OC emission factor.

5.5 Other fossil fuels

5.5.1 Distillate oil: external combustion

Middle distillates and residual fuel oil, termed "gas/diesel oil" and "heavy fuel oil" respectively by IEA, are burned in external-combustion devices such as furnaces and boilers. Middle distillates can also be used for small-scale heat or electricity production in stationary internal-combustion generators, as discussed previously. Emissions from internal and external combustion are quite different. In an engine, the combustion is intermittent and its quality is greatly affected by the timing of ignition, which in turn depends on fuel composition. The steady-state burning in a boiler may be less sensitive, although still affected by burner characteristics that can alter fuel-air ratio and mixing. Values of EF_{PM} for external combustion are given in Table 4. Emission factors for No. 5 oil are applied to heavy fuel oil for both industrial and power generation sectors, and those for No. 2 oil to gas/diesel oil for power generation.

According to the *SPECIATE* database [*U.S. EPA*, 1999], carbonaceous fractions for heavy fuel oil emissions are low (0.08 for BC and 0.03 for OC). For middle-distillate oil, both *Hildemann et al.* [1991] and *Wehner et al.* [2003] report higher fractions for BC, and we use $F_{BC} = 0.30$ and $F_{OC} = 0.09$.

The contribution of high-emitting vehicles to emissions from transportation is beginning to receive attention, as discussed previously. High-emitting events such as "puffing" or "upsets" may also increase emissions from boilers [*Hart*, 2001]. We have observed enhanced emission factors due to isolated events in our measurements on an oil boiler [*Wehner et al.*, 2003], but these are even less well characterized than superemitting vehicles. We have increased the high bound of our emission factors to account for this enhancement; however, the total contribution of oil boilers to BC and OC emissions is small, so that this increase makes only a small difference in the total.

As the EFs for residential boilers are much lower than those for generators (discussed in the previous section), emissions from the residential sector are quite dependent on the choice of division between boilers/furnaces and generators. We assumed that residential diesel use occurs in generators in regions where per-capita electricity use is below 1000 kWh/year, and in external-combustion devices above that level. Our assumption is obviously an oversimplification, as the real situation more likely depends on the fraction and quality of rural electrification, as well as the availability of diesel engines and mechanics.

5.5.2 Kerosene and LPG

Both kerosene and liquefied petroleum gas (LPG) are used for cooking and lighting in the domestic sector. (Where these fuels are used in non-residential sectors, we have combined them with other light distillates.) The few measured emission factors for these fuels are given in Table 4. Particulate emissions from kerosene heaters have been found to be primarily sulfates [*Cheng et al.*, 2001]. Kerosene used for lighting and cooking is likely to perform differently, especially the wick stoves; observers have reported that these can be quite sooty. We use F_{BC}=0.13 and F_{OC}=0.09 after *Cheng et al.* [2001], but assume that F_{BC} could be as high as 0.5. *Muhlbaier* [1981] measured the chemical composition of emissions from LPG furnaces, but the PM emission factors are much lower than those given by *Smith et al.* [2000] for cooking devices. We assume that this combustion is different and use the same speciation as for kerosene.

5.5.3 Natural gas

For natural gas, our emission factors of 0.002 g/kg for BC and 0.004 g/kg for OC are taken from AP-42, assuming that all filterable material is BC and all condensable material is organic matter. These assumptions are likely to overestimate the amounts of emitted PM, but the overestimation has negligible impact on the totals because of the low emission factors. Measurements of light absorption at a natural-gas boiler confirm that emissions of light-absorbing material are quite low [*Wehner et al.*, 2003]. Reported emissions from residential combustion [*Muhlbaier*, 1981; *Hildemann et al.*, 1991] are similar to those in the industrial sector.

5.6 Biofuels

In combustion of biofuels, as in other types of burning, particles of different chemistry have separate generation mechanisms. Some of the reasons can be understood by watching a piece of burning wood, and this demonstration is recommended to the reader if sufficient ventilation is available. Black smoke emanates from flame regions, while whitish smoke, consisting of unburned material generated from pyrolysis, is emitted from regions where there is no flame. The white smoke is particularly profuse when the wood is hot enough to release volatile material, but not hot enough to maintain the chain-branching chemical reactions that result in a flame.

The escape of pyrolysis products from the combustion bed also depends upon the exhaust's flow path. If the smoke is drawn through the flame, the organic material can be consumed; if the exhaust is kept at high temperature, the smoke may be eliminated by oxidation. Immediate dilution of the exhaust, as in an open fire, may quench the oxidation process and result in higher emissions, as observed by *Timothy et al.* [1986] for coal combustion.

Several macroscopic variables affect the emissions from wood combustion. Burning rate has been shown to have a particularly strong effect on both quantity and composition of particulate emissions [e. g. *Rau*, 1989]; probably because of the restrictions in air flow used to slow the combustion. Hardwoods and softwoods may have different emission characteristics, and moisture content also has an effect [*McDonald et al.*, 2000]. Fuel size affects both emissions and chemical composition. In a larger piece of wood, heat can be conducted away from the flame zone during initial heating of the fuel, preventing combustion and allowing escape of more white smoke. At the size used in fireplaces and heating stoves (~10 cm diameter), larger wood results in higher PM emissions, with the increase occurring mainly in the purely organic (condensable) fraction [*Dasch*, 1982]. The relationship between wood size and emissions is likely not monotonic. In cooking stoves, measured emissions for brushwood are higher than those for other wood [*Zhang et al.*, 2000], possibly because of the higher mineral fraction in the smaller wood.

Sampling problems contribute to uncertainty in all emission studies, but they are especially prevalent in wood-burning emissions, which are rich in semi-volatile organics. The dilution and temperature history of the sample affect the amount of OC that condenses from the gas to the particulate phase. Greater mass and higher OC fractions are measured at the cooler temperatures that promote condensation. In order of decreasing temperature and increasing mass measured, general methods include: (1) *direct*: sampling directly from the hot exhaust stack, and collecting on filters maintained at a certain temperature; (2) *dilution*: immediately diluting the sample with fresh air, often collecting on filters at ambient temperature; (3) *condensation*: filtering the directly-sampled warm exhaust, forcing condensation by passing the sample through an ice bath, and counting both filtered and condensed matter. The second method was detailed by *Hildemann et al.* [1989] and has been used in many speciation studies. The third method is used extensively by the U.S. EPA for source testing; the "filterable" component probably underestimates the emitted PM, while the "filterable" plus "condensable" component overestimates it. These effects are *in addition* to the uncertainties in the method of measuring "elemental" or "organic" carbon once the sample has been taken.

5.6.1 Fireplaces and heating stoves

When electricity or natural gas is available for subsistence tasks such as cooking, wood is burned only in heating stoves for space-heating, and in fireplaces for space-heating and aesthetic reasons. The wood burned in this manner is often large pieces split from logs, and combustion is typically untended. Total emissions and chemical composition from wood combustion are affected by the type of wood [e.g. *Fine et al.*, 2002], fuel loading and heat release rate [*Butcher and Ellenbecker*, 1982], and sap, ash, and moisture content. In general, higher PM is associated with the emission of organic material, with low-emission units producing largely black smoke and high-emission cases producing tarry, yellowish material [*Norbert Senf, Masonry Heater Association, personal communication*, 2003].

Emission factors for both fireplaces and heating stoves are listed in Table 4. The literature review by *Houck and Tiegs* [1998] was completed after the last revision of EPA's AP-42. It examined hundreds of measurements and included most of the other references listed in the table. We accept the results of *Houck and Tiegs* [1998] as central values: 12 g/kg for fireplaces and 18 g/kg for heating stoves, recognizing that these tests primarily used the condensation method of sampling. Results from recent European studies [*Spitzer et al.*, 1998] suggest much lower emission factors, which is probably due to both sampling (direct from the stack) and recent improvements in wood stoves. Emissions from wood combustion are 90% below $PM_{2.5}$ [*Baumbach et al.*, 1999], and we assume this value for the submicron fraction.

Measured fractions of "elemental" and organic carbon for fireplaces and heating stoves are summarized in Table 8. . The values we chose lie in the middle of the measurements: for fireplaces and heating stoves, respectively, F_{BC} was 0.15 and 0.10 and F_{OC} was 0.6 and 0.65. These values also lie between the "hot" and "cool" burning measurements given by *Rau* [1989]. It is of concern that the mass emission factors in most of the speciation measurements (4-5 g/kg) are quite different from the tabulated EFs of the nearly 400 measurements reported by *Houck and Tiegs* [1998]. Measurements by both *Fine et al*. [2001, 2002] and *McDonald et al.* [2000] fall in the lowest 20% of the larger database. The differences probably result from measurement methods, fuel loading procedures, or both. Only the mass emission factors from *Dasch et al.* [1982], which reported a higher BC fraction, are similar to those in the larger review. Therefore, the speciation measurements may be inappropriate for the general population of fireplaces, and we have increased the uncertainties to account for this possibility. Fireplaces do not represent most of the worldwide wood combustion, so the uncertainty may affect only a few regions. However, this issue is probably not unique to fireplace combustion; it is quite possible that the problem appears here only because sufficient measurement data bring it to light.

Finally, *McDonald et al.* [2000] reported high black carbon fractions for emissions from wax logs, which are not included in our database. Applying their measurement of 6.7 g BC per kg fuel to the approximately 300 ktonnes of logs burned in the United States each year [*Houck and Tiegs*, 1998], we estimate that wax logs result in 1.8 Gg of BC emissions and a smaller amount of OC. As this value is less than 3% of the emissions from residential wood burning in the United States, we have not included it in the final emission database.

Fireplaces have a comparatively higher fuel share in North America than elsewhere. We assume that 25% of wood in the United States is burned in fireplaces, and the remainder in heating stoves [*Houck and Tiegs,* 1998]. In Europe, we assign about 5% to fireplace combustion, 25% to heating stoves and the remainder to boilers, discussed in the next section. For Eastern Europe and the USSR, the usage is mostly heating stoves. In Asia, Africa and Central America, we assume that the usage is largely cookstoves, which are discussed later.

5.6.2 Boilers

Wood is consumed in larger stoker boilers for building heat. These boilers are scarce in the United States, but are common in Europe; measurements of EF_{PM} are given in Table 5. Values of $F_{1.0}$ come from *Baumbach* [1999]. We have not found measurements of F_{BC} and F_{OC}, so we use those from other wood combustion of 0.12 and 0.65. However, the lower emission factor suggests that efficiencies could be higher and BC fractions greater, and we have included this possibility in the uncertainties.

Wood and other vegetal waste are also burned for process heat and power in industry. Table 4 gives emission factors for bark and wood boilers, taken from AP-42. As discussed previously, wood fuels many industries in developing countries, including large ovens used for drying and processing food [*FAO-RWEDP*, 1998]. We have not found emission factors for this type of combustion, and we use values of EF_{PM}, and other emission characteristics, intermediate between heating stoves and wood boilers. For each region, we estimated the fraction of wood use in traditional ovens versus more advanced boilers based on estimates of use in the food processing industry (assumed to contain some fraction of traditional ovens), the pulp and paper industry (assumed to be advanced) and a country's development level.

5.6.3 Cooking: wood

In developing countries, biofuels are used for cooking and heating as well as a range of other applications. Cooking takes place over open or "three-stone" fires, in traditional cookstoves, or in a range of "improved" stoves [for a compendium, see *Westhoff and Germann*, 1995]. The combustion in these devices is not ideal, and overall efficiencies of 5-30% have been reported [*Leach and Gowen*, 1987; *Gill*, 1987]. However, efficiencies of fires made by novices are lower than those of experienced fire-builders, so that laboratory measurements may not be representative of actual practice [*Dean Still, Aprovecho Research, personal communication*, 2001].

For many reasons, the combustion and emissions of cooking fires might differ from those of heating stoves or fireplaces. The flow of the hot exhaust may be designed to maximize heat transfer to the cooking pot, so that smoke from pyrolysis may be guided through the flame. A cookstove with an enclosed combustion chamber differs from an open fire if the chimney or combustion chamber induces a draft through the fire [*FAO-RWEDP*, 1993b]. On the other hand, as evidenced by the blackness of cooking pots, the exhaust can be quenched on the pot so that the ability to burn out particles is reduced. A wide range of wood sizes is used, from brush to split logs; the wood is frequently smaller than that burned in fireplaces and heating stoves. As we discussed previously for the case of fireplaces, smaller wood has been found to yield lower PM emissions and a higher BC fraction.

If wood is scarce and its acquisition is time-consuming, it is advantageous to optimize the efficiency. (In fact, the most inefficient "cooking" fires in the world may well occur in United States campgrounds, where several kilograms of wood are consumed in order to roast a few grams of marshmallows.) Methods of increasing efficiency include using smaller wood [*Kituyi and Kirubi*, 2003], drying fuel, careful fire tending, and extinguishing the fire immediately after the cooking task is completed [*Leach and Gowen*, 1987]. A fire optimized for heat transfer may remain in the flaming- or glowing-combustion mode more than fireplaces or open field combustion. As efficient combustion produces higher BC fractions [*Cachier et al.*, 1996], the BC fraction for particles emitted from cooking fires could be higher than those of other biofuel applications. Therefore, attempts to increase efficiency could reduce particulate emissions and increase BC fractions. On the other hand, both *Kituyi et al.* [2001] and *Ludwig et al.* [2003] report that domestic fires in Africa have slightly lower combustion efficiencies than open fires.

Total PM emissions from open cooking fires and cookstoves are lower than those from heating stoves fireplaces, as shown in Table 5. It is possible that sampling methodologies explains some of the difference in emission factors; the cookstove tests used the dilution method while heating stoves are tested with the condensation method. However, the difference in emission factors is great enough that we emphasize the following point: for wood fuel, *emission factors in "industrialized" areas may not be used to represent those for "developing" areas*. Some studies give EF_{PM} of 10 g/kg or greater in countries where cooking stoves dominate usage. In our experience, these values *always* originate with the early-1980s literature on fireplaces and heating stoves, not with cooking-stove measurements. Care should be taken to identify the primary references before using any such high emission factors for cooking stoves.

While some studies find that improved stoves have higher particulate emissions than traditional stoves [e.g *Zhang et al.*, 2000], others show the opposite [*Venkataraman and Rao*, 2001]. The results depend on the nature of the "improvements"; for example, high-mass earth stoves may actually have poorer combustion characteristics than the traditional stoves they replace [*FAO-RWEDP*, 1993b; *Barnes et al.*, 1994; *Still et al.*, 2000]. Nearly 30 types of improved stoves exist in India alone [*FAO-RWEDP*, 1993c]. We have made no attempt to account for the regional differences in improvements; we obtain the results in Table 5 by weighting by the number of different stoves measured in each study. The uncertainties are lower than those presented in our earlier work [*Streets et al.*, 2001] because we have treated cookstoves separately.

About 90% of the particulate matter from cooking fires is in the submicron range [*Ballard-Tremeer and Jawurek*, 1996]. Measurements of BC and OC fractions from open fires appear in the work of *Brocard et al.* [1996] and *Cachier et al*. [1996], who reported a BC fraction of 0.08 from several open cooking fires in the Ivory Coast. Here, F_{BC} and F_{OC} are derived from the measurements by assuming that all PM is carbonaceous and that OM:OC ratio is 1.4. *Brocard et al.* [1996] observed that these fires remained in the flaming phase 85% of the time but did not describe the wood size or tending practices.

No BC emission factors have yet been published for cookstoves, although at least one measurement project is in progress (Chandra Venkataraman, Indian Institute of Technology, Mumbai, India). We have made measurements in our laboratory of boiling water with carefully tended, small, dry wood [*Bond, unpublished results*]. This experiment yielded F_{BC} of about 0.25 from an open fire and as high as 0.5 from improved, low-mass cookstoves provided by Aprovecho Research (Cottage Grove, Oregon). Our open fires were probably far more careful than the average, as evidenced by overall cooking efficiencies of 25%, so BC fractions of 0.5 are probably an upper limit of that possible from wood combustion. Assuming that “hot” combustion and smaller wood are characteristic of cookstoves, and avoiding the “typical” fireplace/heating stove measurements summarized in Table 8 for that reason, we estimate F_{BC} as 0.2. We also assume that OM makes up the remainder of the particulate matter.

For Asia, we have used estimates of fuel burned in improved cookstoves (ICS) from the work of *Bhattacharya et al.* [2000]. These include 50% for China, 4% for India, and 1% for other Asian countries. China had the greatest success with ICS distribution; even so, the estimated ICS fraction is lower than the number of disseminated cookstoves discussed by *Smith et al.* [1993]. For Africa and Central/South America, we have assumed that the penetration (or at least the persistence) of ICS is small, with a central value of 5%. A highly successful African ICS, the Kenyan Ceramic Jiko, is used for charcoal burning and does not relate to firewood.

5.6.4 Cooking: other fuels

EF_{PM} for agricultural waste is highly variable, as shown in Table 5. We distinguish between domestic use of agricultural waste, which provides for household energy needs, and *open* burning of agricultural residue. The term "agricultural waste" encompasses a wide variety of material, usually stalks, husks, or shells, whose composition is just as varied as the food it bears. For example, ash contents range from less than 1% (for coconut waste) to 19% (for rice straw), and this variation in content affects the pyrolysis of the material [*Raveendran et al.*, 1995]. Rice and wheat stalks, which have high ash contents, produce higher emissions. The effect on residential combustion characteristics has not been studied, however.

The type of waste chosen for burning depends upon not only production, but the suitability of the material for other purposes (such as fodder). Because we do not have these estimates, we have not divided agricultural waste into different types as did *Liousse et al.* [1996]. This results in a high uncertainty and our estimate of EF_{PM} is 7.5±6.3. As agricultural waste is typically burned in cooking stoves, but burns at lower temperatures, we chose a slightly lower value of F_{BC} and higher value of F_{OC} than for cooking stoves (0.15 and 0.57, respectively). It is unlikely that the combustion of agricultural waste is similar to that of wood, so these estimates are quite uncertain.

Table 5 lists measurements of emissions on animal waste (dung), and we choose a central value of EF_{PM}=3.9±1.7 g/kg from the measurements in Table 5. We assume the same $F_{1.0}$ as for other solid biomass. We assume that the BC and OC fractions are the same as those assumed for agricultural wastes, for similar reasons. Again, the value is quite uncertain due to lack of measurements.

5.6.5 Charcoal

The charcoal fuel-cycle emits particulate matter at two points in the fuel cycle: during its manufacture and during end-use. We have used IEA statistics on fuelwood consumed for charcoal production as a basis for determining emissions from producing charcoal, and data on charcoal consumption in the residential sector to estimate emissions at end-use. The totals are far lower than the 50% of fuelwood assumed by *Liousse et al.* [1996] to be transformed to charcoal.

A wide range of emission factors is measured from charcoal production kilns. We assume that 90% of these emissions are submicron. The expected value of the available measurements of EF_{PM} is 2.6 g/kg, as shown in Table 5. Values of F_{BC}=0.08 and F_{OC}=0.55 come from *Cachier et al.* [1996]. EF_{PM} for charcoal end-use is 4.1±4.8 g/kg. As charcoal is often used in urban areas because it is a cleaner fuel, it is surprising that the end-use measurements average higher than either the production of charcoal or the burning of wood in cookstoves. Clearly, more measurements are needed to address this question. We have found no speciated measurements of end-use charcoal combustion and assume that the emissions are entirely carbonaceous, half BC and half OC.

5.7 Other fuels

Table 4 also lists emission factors for municipal waste and open waste combustion. Waste incineration in industrial applications has been extensively studied because of public concerns about health effects. Less information is available for residential incineration and open burning of waste combustion. The assumptions used to generate emission factors are listed in the footnotes; for open burning, F_{BC} and F_{OC} were chosen so that they were and so that BC+OM accounted for all particulate matter. As our estimates of urban waste burning are small compared to fuel consumption, the high uncertainties do not contribute greatly to global totals, although they may be important on an urban scale.

5.8 *Open burning*

The selection of emission factors for biomass burning requires a separate study. Fortunately, such a review has recently been published by *Andreae and Merlet* [2001], and we use the emission factors from that work. Emission factors for various types of open burning are: savanna, 0.48±0.20 g/kg for BC and 3.4±1.3 g/kg for OC; tropical forest, 0.66±0.31 g/kg for BC and 5.2±1.5 g/kg for OC; extratropical forest, 0.56±0.20 g/kg for BC and 8.0±2.0 g/kg for OC, and agricultural waste, 0.69±0.13 g/kg for BC and 3.3±1.2 g/kg for OC. Emissions depend on other factors, such as fuel moisture content, which changes throughout the season [e.g. *Hoffa et al.*, 1999]. We have not attempted to account for these seasonal changes.

In the review by *Andreae and Merlet* [2001], emission factors for BC from forest and savanna burning are significantly lower than those used in previous work [e.g. *Liousse et al.*, 1996]. In fact, the earlier emission factors lie outside the uncertainty range given by *Andreae and Merlet* [2001], even though the review purportedly includes a tabulation of all previous studies. For the present study, our intention was to draw upon the substantial body of work being conducted on the topic of open biomass burning emissions, and to provide new analysis in those areas that had previously received less attention—that is, emission factors from fossil fuels and biofuels, and the amounts of burned matter in open biomass burning. The discrepancy between the recent review and the previous studies requires further investigation and justification, which is beyond the scope of this study. Since no justification for the inconsistencies has appeared in the literature, we have increased the uncertainties beyond those of *Andreae and Merlet* [2001] to encompass the earlier emission factors.

6 Results

6.1 BC/OC emission overview

The emission estimates discussed here are based on fuel-use statistics for the year 1996. Calculated emissions of black and primary organic carbon by world region are summarized in Tables 9 and 10. The contribution of fossil-fuel combustion is 3.3 Tg for BC and 2.4 Tg for primary OC, 35% below the previously published estimates of 5.1 Tg for 1984 data. The reasons for these decreases will be discussed below.

In discussing emission distributions, we separate emissions from "contained" combustion—that is, combustion excluding open burning of forests and fields, but including all fuels used to meet energy requirements, regardless of origin. Previous summaries [*Liousse et al.*, 1996; *IPCC*, 2002] have tabulated fossil-fuel separately from biofuel and open or “biomass” combustion. We combine fossil-fuel and biofuel emissions into “contained” combustion for three reasons. First, the emission characteristics of biofuel and biomass emissions are different, since the former results from tended fires. Second, the reason for combustion of both fossil fuels and biofuels is the energy produced, while open burning occurs for other purposes (land clearing, land management, or accident). Finally, mitigation strategies for fossil fuel and biofuels are more similar to each other than they are to approaches to reduce emissions from open burning; fossil fuels may replace biofuels as people move up the “energy ladder”, although the choice of fuel use is more complex than simple substitution [*Leach*, 1992; *Smith et al.*, 1994; *Masera et al.*, 2000]. Figure 2 shows the global distribution of BC emissions from contained and open burning separately, and Figure 3 shows primary OC emissions. The color map in the two figures is approximately logarithmic and the scale is the same for all four plots. For BC from contained combustion, large surface fluxes are evident over areas of high population density, particularly Asia (about half of both global BC and OC emissions from contained burning). BC and OC distributions from contained combustion are similar. BC from contained combustion is generally greater than BC from open combustion in the Northern Hemisphere; the opposite is true in the Southern Hemisphere. Figure 3, as well as Tables 9 and 10, clearly show that open burning dominates OC emissions on an annual-average basis in most of the world. This is true even for regions where contained combustion figures heavily, such as North America and Europe. The exceptions are China and India, where large population densities and consumption of solid fuels result in high OC emissions.

6.2 Major sources of carbonaceous aerosol

Regional differences in the sources of BC and OC are of interest, both for interpreting measurement data and for assisting policy decisions. Here, we discuss the major contributors to BC and OC emissions in different regions. The discussion here is based on the central estimates, and inferences about fractional contributions are, of course, affected by the uncertainties in the absolute magnitudes.

Open vegetative burning is the largest contribution to both BC and OC emissions. Its components are summarized in Table 11. The emission factors given by *Andreae and Merlet* [2001], used in this work, are lower than those assumed by other studies [e.g. *Liousse et al.*, 1996; *Chin et al.*, 2002]. The effect of applying different choices for emission factors may be estimated from Table 11. Savanna burning in Africa contributes the greatest amount to BC in this sector, and burning of crop residues the least. Burning of forests and savannas in South America are also large contributors to the budget.

The remainder of global BC emissions comes from "contained" combustion, as does a large fraction of OC. (In this work, we do not include the contribution of natural emissions to the organic aerosol.) Figure 4 summarizes the major contributors from contained combustion. The background gray bars show the fractional contribution of contained combustion. Also indicated on the graph is the relative contribution of each region. Transport is the dominant contributor to BC in many regions, such as North America, Latin America, and Europe. On-road and off-road diesels contribute about equally. Industrial processes and the residential sector are also important in these regions. In less-developed regions like Africa, the dominance of the residential sector is obvious. In most of Asia, including China and India, there are large contributions from industry, residential and transport sectors, and the residential sector contributes the most. Industrial BC emissions in those regions are heavily influenced by the high-emitting technologies identified earlier (cokemaking and brick kilns). On a global basis, transport, industry, and the residential sector have similar contributions to BC (20%, 13% and 27% respectively), with open burning providing most of the rest.

The sources of OC are somewhat different that those of BC. "Contained" combustion is a smaller fraction, because of the high OC emission factors from open biomass burning. As a consequence of the poorer combustion in small devices, residential solid fuels (biofuel and coal) dominate "contained" OC emissions in all regions but the Middle East and the Pacific. Although open burning is the most important sector by far, we estimate that emissions from residential solid fuels contribute nearly 20% to the global budget of OC. Transport comprises only 4% of OC emissions.

6.3 Comparison with previous work

6.3.1 Regional inventories

For the most part, emission inventories developed by local and national air quality agencies focus on total particulate matter, usually PM_{10} (particles with diameters below 10 microns). PM_{10}includes a number of emissions not comprised of carbonaceous particles (e.g. road or construction dust), so it is not possible to compare this work with regional inventories of PM_{10}. Total $PM_{2.5}$ emissions are more similar to carbonaceous aerosols, except that they include mineral matter from combustion.

A black carbon inventory for the United States and the world was commissioned by the U.S. EPA [*Battye et al.*, no date]. The United States central estimate was 443 Gg, in good agreement with our value of 451 Gg, although the sectoral totals are different.

Regional emission inventories of BC and OC are also available for Asia. For India, *Reddy and Venkataraman* [2002a, 2002b] estimated BC and OM emissions as: fossil fuels, 100 Gg and 300 Gg;

biofuel, 207 Gg and 629 Gg; open burning, 39 Gg and 313 Gg. Our totals are: fossil fuels, 180 Gg and 154 Gg; biofuel, 330 Gg and 1963 Gg; open burning, 87 Gg and 700 Gg. Our fossil-fuel BC totals for India are higher because we use a higher EF for diesels and also consider the contribution of two-stroke engines. Biofuel estimates are higher because our wood-use estimates are slightly higher and both EF_{BC} and EF_{OM} are higher. Our estimates of biomass burned are also higher than those of the other study.

Bhattacharya et al. [2000] provided estimates of TSP emissions from biofuels for several Asian countries, for years ranging from 1991 to 1995. As biofuel emissions are largely submicron and carbonaceous, we compare our BC+OM totals with these estimates. For countries where our biofuel totals are similar (within 5% for Nepal, Pakistan, Philippines, and Sri Lanka), our emission estimates are about 3.5 times lower because we chose lower emission factors representative of cookstoves. In India and China, our biofuel usage totals—primarily for wood—are higher; and our database includes animal waste in Vietnam. These additions compensate for our lower EF, and our emission estimates are about 1.5 times lower.

Our emission inventories for the TRACE-P field experiment [*Streets et al.*, 2003b] differ from the present inventory in two respects. First, the inventory presented here is based on the latest year for which *global* energy consumption data were available at the time of calculation (1996). The underlying energy consumption data for TRACE-P were based on estimates for the year 2000 and account for changes in both population and technology in the intervening years. Second, emission factors for TRACE-P were based on those of *Streets et al.* [2001].. Some of the analysis completed since that work includes: (1) the separation of biofuel EFs for cooking and fireplaces, reducing our estimate of cooking emissions; (2) the re-analysis of mobile-source EFs in regions where emission regulations were not implemented until the late 1990s or later, increasing our EFs for both diesel and gasoline consumption; and (3) examination of small but polluting sources, such as cokemaking and brick kilns. Our emission estimates in the earlier paper for China, India, and the rest of Asia were: BC, 1050 Gg, 600 Gg, and 890 Gg respectively; and OC, 3390 Gg, 2840 Gg, and 4200 Gg, respectively. In the current estimate, BC emission estimates for India remain about the same; the introduction of polluting sources balances our reduction in woodfuel emission factor. BC emissions for China and the rest of Asia are greater than those in the TRACE-P inventory, due to increases in some emission factors and decreases in polluting technologies represented in the later year. The current OC emission estimates are all lower than the TRACE-P estimates due to revisions of emission factors. Our bottom-up estimate for India indicates that only about 10% of carbonaceous aerosol (BC+OC) comes from fossil-fuel combustion, unlike the much higher estimates given by *Novakov et al.* [2000]. Assuming the highest fossil-fuel contribution and the lowest biofuel contribution (presumably, a probability of less than 1%), 70% could be attributed to fossil fuels.

6.3.2 Global inventories

Modeling studies of the climatic effects of carbonaceous aerosols have used inventories developed by *Cooke et al.* [1999], *Liousse et al.* [1996], and *Penner et al.* [1993]. Here, we compare our results with one of the most widely used data sets, which combines *Cooke et al.* [*1999*] for fossil fuels and *Liousse et al.*[1996] for biofuel and biomass burning. We contrast with two sets of values, which we call *Previous84* and *Previous96*. *Previous84* is the inventory of BC emissions that has been distributed to the modeling community; it contains the fossil-fuel inventory of *Cooke et al.* [1999][1] and the biomass/biofuel inventory of *Liousse et al.* [1996]. Although the work was published in 1999, we call it *Previous84* because 1984 is the base year of the energy use data. Only the total emissions are available for *Previous84*, so we cannot diagnose the sources of differences. Changes in energy use between 1984 and 1996 contribute to these differences; for example, global consumption of hard coal increased by 50%, and usage of diesel fuel increased by 70%. Differences in emission factors and gridding choices also affect the comparison.

Our second comparison, *Previous96*, is calculated by combining the emission factors published by *Cooke et al.* [1999] with the same fuel-use data used for the current inventory (IEA 1996). The purpose this estimate is to assess the effect of our revised emission factors without the confounding effect of changes in energy use. The authors of the *Cooke et al.* [1999] paper selected emission factors for each country according to one of three development levels: “developed”, “semi-developed” and “developing”. We have attempted to replicate this process, but there may be some disparities between our assignations and those the authors would have chosen. In general, if *Previous84* and *Previous96* estimates agree, the differences between our inventory and the others is due to emission factors; if they do not agree, the differences may be due to activity estimates.

Tables 9 and 10 summarize the comparison. The net effect of our emission estimates on *Previous96* is a reduction of about 45% in BC and 70% in OC. This relatively greater reduction in OC is true in nearly every world region; we are predicting a slightly darker aerosol in general, and a much

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[1] The inventory available from the Global Emission Inventory Activity at http://weather.engin.umich.edu/geia, is that of *Cooke and Wilson* [1996], not *Cooke et al.* [1999].

darker aerosol in some regions, than the previous study. Table 12 lists the sectors with the largest changes between our inventory and *Previous96*, in order of their effect on the BC inventory. The largest change is in open vegetative burning of all types, because the emission factors summarized by *Andreae and Merlet* [2001] are lower than those used in the earlier work.

For contained combustion, three other sectors have large reductions: coal used in power generation, diesel fuel used in transport, and biofuel used in domestic applications. These sectors make up more than 95% of the reduction in BC emission from contained combustion. The reasons for these differences were discussed in the section on emission characterization. Briefly, emission factors from coal in power generation are reduced because the literature suggests that particles from pulverized coal plants are primarily mineral matter, with very little carbonaceous material. Diesel emission factors are lower because we were not able to support the previous emission factor of 10 g/kg for BC, despite considering superemitting vehicles and the off-road sector. Emission factors for residential biofuel have also been reduced because we have tabulated many studies with lower emission factors. For OC, our reduction in residential biofuel emissions is quite significant—almost as great as that in open biomass burning. Estimated gasoline emissions are higher than those of the previous work, because of our distinction of two-stroke engines. Residential coal emissions have decreased because we use a slightly lower emission factor and a higher black carbon fraction. Most of these studies were not available at the time the previous inventory was developed; the considerable change due to the advent of new information points to the importance of obtaining measurements on emission factors and characteristics for a variety of these small sources.

We compare the present work with *Previous84* and *Previous96* in Figures 5 and 6. We have presented results for BC only. Unlike our earlier analysis, we divide the BC inventory into *fossil-fuel* combustion and *biofuel plus open biomass* combustion, in order to compare with the *Previous84* inventory widely used by the modeling community. The figure also shows our uncertainty estimates, which we will discuss later. In each figure, the upper left graph shows zonal averages of emissions, with the three remaining graphs showing the longitudinal distribution for different latitude bands to identify continents. Our modified emission factors have changed the emissions significantly in some regions.

For fossil-fuel BC, our estimate is remarkably similar to that of *Previous84* for latitudes below 30° N (except Australia) and for China. This agreement is fortuitous, resulting from the combination of lower emission factors and higher fuel-use in our inventory. *Previous96* shows the effect of applying the earlier emission factors to 1996 energy-use data, and is much higher than our prediction in each of these regions. Both *Previous84* and *Previous96* are higher than our prediction for latitudes above 30° N, although they are within the substantial uncertainty estimates. For Europe, our *Central* estimates are lower than either *Previous84* or *Previous96*, largely due to our assumptions about the use of diesel fuel in the residential sector and the decrease in emission factors for coal-burning power plants. In India, our *Central* estimate is also lower than either of the other two because of our lower EF for diesel vehicles and coal-burning power plants. Many of the same observations would be found for OC emissions.

Figure 6 shows comparison graphs for the combination of biofuel combustion and open vegetative burning. Again, we have included biofuel with biomass burning so that we can compare with the available inventories. We still refer to the inventory as distributed as *Previous84* for consistency; however, it is not based on 1984 burning data. The zonal averages (upper left) show that the emission distribution is quite different. In *Previous84*, the sources in the Southern Hemisphere are nearly absent.

As the *Liousse et al.* [1996] emission factors are much higher than those we use, the lower emissions must result from the estimate of burned matter. *Penner et al.* [2002] reported that models generally underpredict aerosol optical depth from 10°-30° S; this is a possible explanation. In contrast, both *Previous84* and *Previous96* are much higher in the Northern Hemisphere tropics, and our estimates of activity (burned matter) are similar. However, the longitudinal distribution (lower right) shows differences in spatial distribution. Our inventory has higher activity estimates in Eastern South America and India; the other inventory has higher activity estimates in Western South America, Africa, and Southeast Asia. As for fossil-fuel combustion, agreement between *Central* and *Previous84* estimates in North America and Europe results from compensation between increased activities and decreased emission factors in our inventory. Estimates of biofuel activity in India and China appear to be far lower than those in *Previous84*.

6.4 Uncertainties

With the uncertainties that we considered, we provide 95% confidence intervals for total global emissions. For BC, these are 3400-10300 Gg/year for contained combustion, and 1680-7600 Gg/year for open biomass burning. Compared to the mean, the confidence intervals are –32 to +110% for contained burning, and –50% to +130% for open burning. For OC, the confidence intervals are: contained combustion, 5200-17200 Gg/year (-42% to +95%), and open burning, 12800-25300 Gg/year (-49% to 130%). The asymmetric confidence intervals are a consequence of our lognormal treatment of emissions.

Higher fuel usage and greater uncertainties about emission factors from particular types of burning cause regionally-distinct uncertainties in emissions. Figure 5 shows that the magnitude of uncertainty is largest in Asia, where uncertainties in emission factors and activity data are combined with high population and energy-use densities. However, the upper confidence limit is about 100% in most regions. Biomass/biofuel uncertainties have the largest magnitudes in Africa, India, and Asia.

Open biomass burning contributes the most to uncertainties, because it comprises a large fraction of BC emissions and dominates OC emissions. This study has estimated that “contained” combustion is a somewhat greater contributor to BC emissions, whereas previous work found that the contributions of “contained” and open burning were about equal. Confirming this result requires a thorough assessment of the BC emission factors presented by *Andreae and Merlet* [2001] as compared with those used by other studies [e.g. *Liousse et al.*, 1996], as mentioned earlier. While this sector is obviously important in the global budget, recent literature and ongoing studies are providing inventories [*Lavoué et al.*, 2000], overview studies [*Andreae and Merlet*, 2001], and field experiments (e. g. SCAR-B in Brazil [*Kaufman et al.*, 1998], and the recent SAFARI-2000 in Africa). We have concentrated our efforts on the remainder of fuel usage, which is less thoroughly studied in regard to its global climatic effects.

For BC from “contained” combustion, Table 13 lists the fuel/sector combinations that contribute the most to variance. For each of these combinations, we estimated the effect of each underlying parameter by setting its uncertainty to zero and recalculating the total variance. The major contributors to variance, in order of their contributions, are given in the table.

For BC, the largest variance by far is due to the cokemaking process, particularly in China where the fraction of “beehives” is not known. As discussed previously, this contribution could have been a transient phenomenon in the mid-1990s and may no longer exist due to Chinese regulation of the coking sector. The second greatest variance is due to residential wood combustion. Uncertainties in emission characteristics contribute, but uncertainties in the amount of fuel consumed contribute the most to variance. For the third and fourth greatest contributors, industrial coal combustion and on-road diesel emissions, the prevalence of and appropriate emission factors for small, highly-polluting sources (e.g. brick kilns and superemitters) have a large impact on variance.

Table 14 repeats the analysis for organic carbon. Here, emissions from residential wood combustion dominate uncertainties, with activity estimates again leading the contributors to variance, and EF_{PM} for many types of burning also playing a role. Gasoline consumption in transportation is the second highest contributor, with fractions and magnitudes of two-stroke engines being of concern. Agricultural waste, industrial use of biofuels, and cokemaking are also contributors.

Of course, each world region is subject to different uncertainties, and Table 15 qualitatively indicates the sectors that have the largest contribution to variance in each region. The table first shows the contribution of open vegetative burning to the uncertainties. That dominates OC uncertainties in all regions. It contributes varying fractions to the variance of BC, ranging from 6% in Europe to 98% in Africa.

The other lines of the table summarize the sectors that cause moderate and large uncertainties in the remaining, “contained” combustion. The sectors that contributed most to global uncertainties—cokemaking and industrial coal—are mainly important in Asia and the former USSR, where some technologies are less advanced and coal is widely available. In other regions, cokemaking is less important and industrial coal plays very little role. Burning of wood in the residential sector is a major contributor to uncertainty in both BC and OC inventories in nearly all regions, while animal waste and agricultural residues affect mainly Asia and Africa. On-road and off-road diesels are important in the Americas and Europe. Gasoline used for transport in the Americas is also important.

6.4.1 Surprise

Although we have attempted a careful accounting of uncertainty based on available data, emissions may lie outside our confidence intervals if the measurements used to infer them do not represent general practice. Technologies that are prone to high emissions may also provide surprises. Off-road vehicles and cokemaking together comprise over 10% of our BC inventory. Their potential importance became apparent only in the last few months of our work on this project, and was not identified by previous global inventories. Even when a sector is thought to be well understood, surprises have surfaced. For example, source-apportionment results discussed by *Lawson and Smith* [1998] found that gasoline vehicles emitted three times more $PM_{2.5}$ than diesel vehicles in the Denver area, although inventories predicted that they should emit three times less. We do not discount the possibility that some sectors, regions, or technologies may hold additional surprises that will cause the magnitude of emissions to lie outside the uncertainty bounds presented here.

Examining both physical and social phenomena, *Shlyakhter* [1994] presented a method of accounting for "surprise", proposing that widening confidence intervals according to an exponential distribution could account for unexpected sources of variance. For physical phenomena, he suggested increasing the confidence intervals by a factor of 3.8 to achieve a 95% uncertainty. For social predictions, the analogous increase in confidence interval was much higher (greater than 7). The emission phenomenon is partly physical and partly social, currently relying on sparse information in both disciplines to characterize the system; using the distribution given by *Shlyakhter*, we should increase our confidence intervals by an intermediate value (say, 5, resulting in a factor-of-10 uncertainty). The uncertainties presented throughout this paper do not account for this more conservative treatment of variance, although our assignation of uncertainties does include some judgment about the depth of knowledge in each sector. Interestingly, *Shlyakhter* [1994] found that aggregate economic data were just as uncertain as sectoral data, implying that compensating errors might not prevent our global totals from being just as uncertain as any individual sector. However, emission work has one advantage: inverse estimates of errors in the source term, based on modeling and atmospheric measurements, can bound the uncertainties, as long as model errors are carefully characterized.

6.4.2 Reducing uncertainties

The variance analysis provides a clear picture that our knowledge of BC and OC emissions is most limited by the following, not in order of importance:

(1) Particulate matter emission factors for technologies that have not been well studied (residential combustion, traditional industry, superemitters);

(2) Speciation of PM from high-emitting technologies into BC and OC; this especially affects BC emissions, and involves measurement uncertainties as well as population variability;

(3) *How* fuel is burned (technology divisions in sectors that contain even a small fraction of highly-polluting devices); and

(4) *Amounts* of fuel burned in informal sectors, where fuel and output do not pass through official channels and are not tabulated by official means.

The first two are currently uncertain because relevant work has not been funded until recent years. Ongoing work on residential combustion, such as that of *Venkataraman and Rao* [2001], *Oanh et al.* [1999, 2002], and *Smith et al.* [2000] will provide more information on domestic combustion. More field studies like those of *Brocard et al.* [1996] are needed to confirm that laboratory measurements are representative of practice.

Sampling bias makes the third item difficult to quantify. However, even some simple measurements could assist in reducing the uncertainties. For example, remote-sensing measurements such as those described by *Bishop et al.* [2000] could assist in estimating the fraction of superemitters in a population. However, the interpretation of these measurements needs refinement in order to attribute particulate matter (not just gaseous pollutants) to different vehicle types. Tabulations like those of *Polenske et al.* [2002] can assist in understanding industrial contributions.

Despite the continuing advances in emission characterization just listed, the emission inventories contain fundamental uncertainties due to the use of fuels in informal sectors. The amount of biofuel used remains uncertain despite a legacy of studies spanning at least 20 years [e.g. *deLucia*, 1983; *FAO-RWEDP*, 1997]. It is unlikely that studies focused on climate or air-quality issues alone will be able to improve consumption estimates beyond previous work supporting rural energy policies, in which the study focus was presumably of greater interest to the examined population. At best, the climate, air-quality, health-effects, and energy communities must begin to share information, as demonstrated in the recent work of *Yevich and Logan* [2003].

As just discussed, it is likely that “bottom-up” inventory estimates will fail to provide the required accuracy in estimating anthropogenic effects on global and regional climate. Elucidating the contribution of biofuel emissions to regional and global loadings of carbonaceous particles will require parallel work using recently-developed techniques in source apportionment, such as organic tracers [e.g. *Schauer et al.*, 1996]. It would be particularly useful to identify the differences in source profiles for different combustion types and the same fuels (e.g. biofuels vs open biomass; diesel superemitters vs normal engines).

6.5 Previous Measurements

As mentioned previously, a rigorous comparison of modeled results using the inventory with atmospheric measurements will be the focus of future studies. Here, we discuss only a few current issues regarding the comparison between modeled and measured BC and OC concentrations and comment on the effect of our inventory revisions.

The relative amounts of BC and OC, along with other co-emitted species such as sulfate precursors, may determine the sign of climate forcing. Ratios of primary OC to BC ratios in *Streets et al.* [2003b] were 3.2 for China and 4.7 for other Asian countries; the current ratios are 1.9 for China, 3.0 for India, and 2.2 for the rest of Asia. A comparison of OC/BC ratios between inventory and measurements might provide a reality check, but the available measurements exhibit a wide range of ratios and include secondary OC, which is formed from gaseous organic compounds in the atmosphere. OC/BC ratios measured near India include 0.8-2.5 measured by *Chowdhury et al.* [2001] and 1.3 reported by *Mayol-Bracero et al.* [2002]. Different ratios may be expected from measurements in urban and rural areas, with diesel- or coal-dominated pollution resulting in lower ratios. In urban China, measured OC/BC ratios range from about 2 [*Ye et al.*, 2003] to as high as 12 [*Bergin et al.*, 2001]. Thus, we do not consider our "better" agreement with the INDOEX OC/BC ratios as "validation" of this inventory. Determination of regionally representative ratios, as well as community-wide agreement on the measurement techniques that separate OC from BC, is needed before measured OC/BC ratios can be compared with inventory.

Using backtrajectory calculations combined with measurements at a coastal station downwind of Europe, *Derwent et al.* [2001] estimated emissions from the United Kingdom and Europe, respectively, as 46±13 and 480±140 Gg/year during 1995-1998. This is in quite good agreement with our central estimates of 46 and 550 Gg/year, even though the uncertainties in the inventory are a factor of 2 and 3, respectively. However, the *Derwent et al.* [2001] estimates are highly dependent on the appropriate choice of conversion between optical measurements and BC mass.

Several studies have compared results of models using previous inventories with atmospheric measurements. The results of earlier comparisons might suggest whether the revisions we prescribe will provide better agreement, although more rigorous comparisons accounting for model uncertainties are needed to draw firm conclusions about whether the emissions are the source of the discrepancy. *Cooke et al.* [2002] found that modeled concentrations were usually lower than measurements at a mid-continental site in North America. Our fossil-fuel emissions in Eastern North America are about 30% lower than those of the modeled inventory (*Previous84*), but with our inclusion of biofuel and biomass burning, the two inventories are about the same. Thus, the new inventory neither improves nor worsens the comparison.

Chin et al. [2002] used the fossil-fuel inventory of *Cooke et al.* [1999] and a biomass inventory based on that of *Yevich and Logan* [2003]. Using emission factors for biomass burning that were much higher than those presented here, they obtained optical depths that were similar to those measured by the AErosol RObotic NETwork (AERONET), a ground-based remote-sensing network, in many regions including those of biomass burning. This result suggests that higher emission factors for biomass burning might be more appropriate, and this issue should be investigated, as we have mentioned.

Sato et al. [2003] compared modeling results with atmospheric light absorption measured by AERONET data. Using data assembled from previous inventories, they suggested that BC needed to increase by a factor of 2-4 and that OC needed to increase by a factor of 1.6-1.8 to match measurements. Since our estimates have decreased compared with previous inventories, they will not provide better model-measurement agreement. One exception is Europe, where modeled concentrations using the inventory of *Cooke et al.* [1999] are too high and where our estimates have decreased.

Dickerson et al. [2002] examined BC measurements made off the South Asian coast during the Indian Ocean Experiment (INDOEX), inferred the likely magnitude of the BC source, and compared it with bottom-up emission inventory estimates in the range of 0.6 - 1.0 Tg for the year 2000 from the TRACE-P data set [*Streets et al.*, 2003b]. By developing BC/CO correlations, *Dickerson et al.* [2002] speculated that the BC source strength on the South Asian continent may be 2-3 Tg yr^{-1}, significantly higher than can presently be explained by any bottom-up inventories. Such an analysis can be confounded by poor understanding of BC removal from the atmosphere, but it nevertheless reinforces the need for a much more detailed investigation of the sources of carbonaceous aerosols in South Asia.

In our experience, modelers who have used previous inventories suggest that higher emissions of BC are needed to account for the measured BC in the atmosphere. As discussed throughout the present paper, we have frequently *decreased* the recommended BC emission factors because investigation showed that some of the previous estimates were unwarranted and often too high, leaving even more atmospheric BC unexplained. Despite the possibility of larger measurement-model discrepancies resulting from use of this inventory, the reliance on *measured* emission factors is a step in a necessary direction.

7 Summary and Final Remarks

Our intent in this work was to clarify the role of combustion practice in determining emission rates of primary carbonaceous species to the atmosphere. We have identified some of the major variations in practice that affect emission rates, and present a new global inventory of black and organic carbon accounting for regional variations in technology. In detailing these regional practices, we attempt to strike a balance between accuracy and the feasibility of representing the existing variability The emission factors we suggest here contain uncertainty, but represent best estimates based on reviewing the literature on combustion processes and source characterization. We report large differences—often decreases—in appropriate emission factors when compared with previous inventories [*Cooke et al.*, 1999]. In some cases, we were not able to find measurements supporting the emission factors used earlier.

The reductions in emission factors are partially offset by an increase in energy consumption since the base year of the previous study. The net effect is an overall decrease in emission estimates, especially those from fossil fuels. A redistribution of the emissions also results, with a reduction of fossil-fuel emissions at mid- to high- Northern latitudes and biomass/biofuel emissions in the Northern tropics, and an increase in biomass/biofuel emissions in the Southern Hemisphere and higher Northern latitudes.

We have constructed a representation of emission uncertainty, accounting for many potential contributors. Emission rates, speciation of particulate matter from small sources, measurements of BC and OC fractions, prevalence of different technologies, and activity rates in informal sectors are all important factors contributing to uncertainty. The overall uncertainty in emissions from "contained" combustion is a factor of two, with greater uncertainties in some regions. This uncertainty in *emissions* *alone* is comparable to the range of estimates of climate forcing by black carbon [*IPCC*, 2001]. Since removal processes and radiative effects are also uncertain, we conclude that the error bars given by *IPCC* [2001] underestimate the true uncertainty.

Except for a thorough investigation of the discrepancies in emission factors from biomass burning, further examination of the literature is unlikely to yield reductions in uncertainty. More needed are characterization of small, polluting sources or those with high emission factors; agreement on consistent measurement methods; engagement of international scientists and regulatory agencies in collaborations of technology assessment; and creative methods of assessing regional emission rates and source apportionment.

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Table 1. Technologies gridded with proxies other than total population

Gridding proxy	**Fuel/Technology combination**
Rural population	Residential sector: biofuels/open fires & cooking stoves; coal/open fires and cooking stoves
Urban population	Open waste burning (estimated from urban populations)
Agricultural land cover	Agricultural diesel use
Gridded sulfur emissions	International shipping
(Forest cover) multiplied by (Fire counts 1999-2000)*	Forest burning
(Savanna cover) multiplied by (Fire counts 1999-2000)*	Savanna burning
(Agricultural land cover) times (Fire counts 1999-2000)*	Agricultural waste burning

Gridded by appropriate land cover when fire counts appeared unreliable.

Table 2. Comparison of biofuel consumption estimates for India, in Mtonne. Each of the sources also gave estimates for agricultural residue and dung, which are not included.

Source	**Year of estimate**	**Rural**	**Urban**	**Total**
National Council for Applied Economic Research (NCAER) [a, b]	1985	93		
Tata Energy Research Institute (TERI): rural energy database [a, c]	1991	252		
Tata Energy Research Institute: urban [a]	1988		11	
Planning Commission: Integrated Rural Energy [a]	1991	169		
International Energy Agency [d]	1991			265
Bhattacharya et al. [2000]	1991			125
Reddy and Venkataraman [2002b]	1996-97	293	9	
Yevich and Logan [2003]	1985			220

(a) As cited by *TERI* [1996]. That reference provides regional breakdowns of the NCAER and TERI estimates; the relationship between the two estimates is not constant in different agro-climatic zones. (b) The earlier year could contribute to the lower estimate; however, IEA estimates of total biomass energy are only 11% higher in 1985 and 1991. (c) Central estimate; TERI also provided low and high estimates of 181 and 309 Mtonne. (d) Converted from energy units assuming 15 MJ/kg. We created data for Jammu and Kashmir using values from surrounding states.

Table 3. Tabulation of open biomass burning by world region (Tg).

	Savanna	**Forest**	**Agricultural Residue**	**Total**
Africa	2337	444	73	2855
Asia	147	491	271	908
Australia/Oceania	270	22	30	321
Central America/Caribbean	8	93	6	107
Europe	2	81	29	111
North America	0	224	26	249
South America	808	585	39	1433
Total	*3572*	*1939*	*475*	*5896*

Table 4. Central values of particulate matter (PM) emission factors for stationary combustion, excluding residential solid fuels. The classification “all” means “all stationary sources”. Uncertainties are not given here but are used in calculating uncertainty. Values of EF_{PM} and $F_{1.0}$ are from AP-42 [*U.S. EPA*, 1996] unless noted and refer to PM_{10} unless only total PM is given. Values of F_{cont} are inferred from AP-42 for submicron particles. F_{BC} and F_{OC} are both for submicron particles. Ranges in F_{cont} cover further subdivisions that account for different emission controls.

Fuel	**Technology**	**EF_{PM} (g/kg)**	**$F_{1.0}$**	**F_{BC}**	**F_{OC}**	**F_{cont}**
Biofuel	Stoker	2.2[a]	0.86	0.05[b]	0.2[b]	0.4-1.0[c]
Biofuel	Traditional[d]	10	0.85	0.1	0.6	1.0
Briquettes	Stoker[e]	2.5	0.25	0.01	0.02	0.3-1.0
Brown coal	Cyclone	33	0.17	0.006[f]	0[f]	0.03-1.0
Brown coal	Pulverized	29	0.09	0.006[f]	0[f]	0.05-0.9
Brown coal	Stoker	17	0.11	0.05[g]	0.66[h]	0.06-1.0
Hard coal	Cyclone	1.3	0.15	0.006[f]	0[f]	0.03-1.0
Hard coal	Pulverized	12[i]	0.09	0.006[j]	0[k]	0.05-0.9
Hard coal	Stoker	4.2[l]	0.33	0.10[m]	0.02[m]	0.05-1.0
Hard coal	Trad. brick kiln[n]	10	0.9	0.5	0.4	1.0
Coking coal	Coke oven[o]	5.8	0.35	0.48[p]	0.34[p]	0.33
Coking coal	“ (uncaptured)[q]	20	0.5	0.48[p]	0.34[p]	1.0
Coke	Blast furnace[r]	0.4	0.28	0.28	0.05	0.05-1.0
Heavy fuel oil	All	1.1	0.45	0.08[s]	0.03[s]	0.13-1.0
Middle dist. oil	Industry/Power	0.49	0.18	0.30[t]	0.09[t]	0.13-1.0
Middle dist. oil	Generator	6.0[u]	0.86[v]	0.66[v]	0.21[v]	1.0
Mid/light dist	External comb.	0.25[w]	0.90	0.29[x]	0.13[x]	1.0
Kerosene	Residential	0.9[y]	1.0	0.13[z]	0.10[z]	1.0
LPG	Residential	0.52[aa]	1.0	0.13[bb]	0.10[bb]	1.0
Natural gas	All	0.002[cc]	1.0	0.06[dd]	0.5[dd]	1.0
Solid waste	All	12.6	0.1[ee]	0.035[s]	0.001[s]	0.05
Solid waste	Open burning	30[ff]	0.5[gg]	0.37[gg]	0.37[gg]	1.0

(a) Average of AP-42 emission factors (2001 revision), including upward revision and increased uncertainty to account for higher emission factors from bagasse. (b) Average of several wood-fired boilers in SPECIATE [*U.S. EPA*, 1999]. (c) Advanced boiler control based on prevalence of controls on wood-fired industrial boilers in the United States [*James Eddinger, U.S. EPA, personal communication*] combined with submicron escape efficiency. (d) No data; see text. (e) All values except F_{cont} taken from *Ge et al.* [2001]; assumed that $F_{1.0}$=0.85 $F_{2.5}$. (f) No data—assume same as hard coal in pulverized combustor. (g) Based on *Bond et al.* [1999b]. (h) Based on *Pinto et al.* [1998] for residential combustion, but with a high uncertainty. (i) Average of various types, with uncertainty accounting for variation. (j) *Fisher et al.* [1978]; *Mamane et al.* [1986]; *Olmez et al.* [1988]; *Querol et al.* [1995]. (k) *Olmez et al.* [1988]. (l) Average of various types in AP-42; the higher values given by *Hangebrauck et al.* [1964] probably refer to total PM, which is higher than PM_{10} by 30-60% for stokers. (m) *Ge et al.* [2001]. (n) No information; use value intermediate between home heating stove and stoker with high uncertainty. See discussion in Section 5.3. (o) Sum of several processes in coking, primarily door leaks. (p) No information; we assume that emissions are comprised of thermally-processed, devolatilized coal tar and are half BC and half OM. (q) See discussion in Section 5.2.3. (r) See discussion in Section 5.2.3. (s) SPECIATE [*U. S. EPA*, 1999]. (t) *Hildemann et al.* [1991]; *Wehner et al.* [2003]. (u) *U.S. EPA*, NEVES [1991]. (v) No information; use fine fraction and speciation data from diesel automobiles. (w) AP-42 gives 0.05 g/kg, but also 0.40 for pre-1971 units. (x) *Hildemann et al.* [1991]. (y) *Smith et al.* [2000]; *Oanh et al.* [2002]; *Reddy and Venkataraman* [2000], citing an unpublished study by *TERI*. (z) *Cheng et al.* [2001]. (aa) *Smith et al.* [2000]. (bb) Assumed same as kerosene. (cc) Filterable PM only. (dd) *Muhlbaier* [1981]; *Hildemann et al.* [1991] (average of very different results for OC). (ee) No size-resolved data; used fraction of *total* filterable PM from stokers, which is the most similar combustion. (ff) U.S. EPA gives 8 g/kg for open burning and 50 g/kg for automobile components such as tires. (gg) No information.

Table 5. Compilation of particulate matter emission factors for residential solid-fuel combustion. Under "References", ranges indicate multiple sources measured, while "±" indicates standard deviation of same source. Under "EF_{PM}", "±" indicates half-width of 95% confidence interval, not necessarily centered about the mean. See text for discussion of other emission characteristics.

Fuel/Technology	**References**	**EF_{PM} (g/kg)**
	Fossil fuels	
Bituminous coal/ Apt. building stoker	2.0-2.4 [*Beijing EPB*, 1996], 6-18 [*Hangebrauck et al.*, 1964], 1.3-4.4 [*Spitzer et al.*, 1998]	2.5±3.0
Bituminous coal/ Heating stove	10.4 [*Butcher and Ellenbecker*, 1982]; 10-22 (hot air furnace) [*Hughes and DeAngelis*, 1982]; 17-79 [*Jaasma and Macumber*, 1982]; 0.6-65 [*Sanborn*, 1982]; 7.6 [*Truesdale and Cleland*, 1982]; 4.6±2.1 [*Spitzer et al.*, 1998]	12±8
Bituminous coal/ Cooking	8.2 (open pit) [*Mumford et al.*, 1987], 12±17 (clay stove) [*Bond et al.*, 2002], 0.13-14.5 (improved stove) [*Zhang et al.*, 2000]	7.7±6.5
Lignite/all	2.7-6.5 [*Bond et al.*, 2002]	4.6±4.6
	Biofuels	
Agricultural waste/ Domestic use	2.4-9.4 [*Joshi et al.*, 1989], 1.7-4.0 (maize stalks) 4.7-17.8 (wheat stalks) [*Zhang et al.*, 2000], 0.63-4.3 (mustard stalks) and 0.8-16 (rice stalks) [*Smith et al.*, 2000]	6.5±3.0
Animal waste/ Domestic use	4.9-5.6 [*Joshi et al.*, 1989], 0.55-2.2 [*Smith et al.*, 2000]; 3.9-4.9 [*Venkataraman and Rao*, 2001]	3.7±2.0
Charcoal/ Production	4.0±1.5 [*Brocard et al.*, 1996]; 0.7-4.2 [*Smith et al.*, 1999]; 8.4 [*Pennise et al.*, 2001] (all in g/kg wood, not charcoal)	2.6±2.2
Charcoal/ Domestic use	3.9-7.5 [*Oanh et al.*, 1999]; 2.4±0.7 [*Smith et al.*, 2000]	4.1±4.8
Wood/ Apt. building stoker	1.0-1.7 [*Spitzer et al.*, 1998], 1.4-3.9 (hot water boiler) [*Hughes and DeAngelis*, 1982]	1.4±1.0
Wood/ Fireplace	11.8±11.6 [*Houck and Tiegs*, 1998]; 17.3 [*EPA, AP-42*]; 5-17 [*Dasch*, 1982]; 2.9-9.0 [*McDonald et al.*, 2000]; 2.7-11.4 [*Fine et al.*, 2001]; 1.6-6.8 [*Fine et al.*, 2002]	12±6
Wood/ Heating stove	0.66 [*Truesdale and Cleland*, 1982]; 1.2-3.3 [*Spitzer et al.*, 1998]; 6.1 (improved), 18.5 (conventional) [*Houck and Tiegs*, 1998]; 15.3 [*U. S. EPA*, 1996]; 1.6-6.4 [*Butcher and Ellenbecker*, 1982], 3.1 [*Bond*, 2000], 3.3-28 [*Sanborn and Blanchet*, 1982], 10.2-15.3 (cordwood), 2.1-4.4 (pellet stoves) [*EPA AP-42*], 2.3-7.2 [*McDonald et al*, 2000]	15±8
Wood/ Traditional cookstove	6.4-8.9 [*Smith et al.*, 1987[b]], 1.9±0.7 [*Joshi et al.*, 1989], 1.0 [*Smith et al.*, 2000], 2.8 [*Venkataraman and Rao*, 2001]	3.9±3.0
Wood/ Improved cookstove	4.5 [*Smith*, 1987[a]], 2.0-2.8 [*Joshi et al.*, 1989], 0.67-1.5 [*Ballard-Tremeer and Jawurek*, 1996], 1.5-4.6 [*Zhang et al.*, 2000], 1.2-4.0 [*Smith et al.*, 2000], 0.9-1.2 [*Venkataraman and Rao,* 2001], 3.7 [*Oanh et al.*, 2002]	2.3±0.8
Wood/ Open cooking fire	5±3 [*Brocard et al.*, 1996], 0.8-1.1 [*Ballard-Tremeer and Jawurek*, 1996], 0.94-2.0 [*Smith et al.*, 2000], 8.5 (eucalyptus chips) [*Oanh et al.*, 1999]	3.8±2.1

[a] Citing conference proceedings by *Butcher et al.*

Table 6. Range of emission factors (EFs) for black carbon (BC) in g/kg. The ranges bracket central values, with differences being due to regional variations in technology mix; categories with only one value do not have a range of technologies. Emission factors for each technology also have associated uncertainties (not shown here) resulting from imperfect knowledge of emission factors and other characteristics. Although technologies used in power generation and industry have the same EFs, the fuel use is apportioned to different technologies within the two sectors.

Fuel	**Power**	**Industry**	**Residential**	**Transport**	**Other end uses**
Fossil fuels					
Briquettes, coke	0.011	0.005	0.15	--	Blast furnaces (0.002-0.004)
Brown coal, peat	0.000-0.002	0.001-0.15	0.18	--	none
Charcoal	--	w/coke	1.0	--	none
Diesel fuel	0.25	3.4-4.4	0.06-4.0	1.3-3.6	Rail (0.51), ships (0.34), tractors (2.6-3.7)
Hard coal, coking coal	0.008-1.2	0.007-1.2	0.89-5.4	3.0 (rail)	Coking (0.32-1.2)
Heavy fuel oil	0.04	0.04	0.07	0.34 (ships)	none
Gasoline, aviation fuel, kerosene	--	0.14	0.9	0.08-0.43	Aircraft (0.10)
LPG	--	--	0.20	--	none
Natural gas	0	0	0	0	none
Waste	0.013	0.013	4.2	--	open (5.5)
Biofuels					
Animal wastes	--	--	0.53	--	none
Agricultural residues	--	w/wood	1.0	--	none
Wood	0.044	0.08-0.55	0.3-1.4	--	charcoal production (0.2)
Biomass burning	*Savanna*	*Crop residues*	*Forest*		
	0.48	0.69	0.56-0.61		

Table 7. Range of emission factors for primary organic carbon (OC) in g/kg. (See notes for Table 6.)

Fuel	**Power**	**Industry**	**Residential**	**Transport**	**Other end uses**
Fossil fuels					
Briquettes, coke	0.004	0.008	0.14	--	none
Brown coal, peat	0.003-0.04	0.03-0.68	2.7	--	none
Charcoal	--	w/coke	1.3	--	none
Diesel fuel	0.001	1.1-1.4	0.03-1.1	0.4-1.1	Rail (1.6), ships (1.1), agriculture (0.84-1.2)
Hard coal, coking coal, coke	0-0.001	0.001-0.9	0.44-4.3	0.015	Rail (0.1), blast furnaces (0-0.001), coking (0.23-0.86)
Heavy fuel oil	0.015	0.015	0.015	1.1	none
Gasoline, aviation fuel, kerosene	--	0.04	0.09	0.19-5.4	Aircraft (0.03)
LPG	--	--	--	0.05	none
Natural gas	0.001	0.001	0.001	0.001	none
Waste	0.002	0.002	0.4	--	open (5.5)
Biofuels					
Animal wastes	--	--	1.8	--	none
Agricultural residues	--	w/wood	3.3	--	none
Wood	0.18	0.31-3.2	1.7-7.8		charcoal production (1.3)
Biomass burning	*Savanna*	*Crop residues*	*Forest*		
	3.4	3.3	5.2-8.0	--	

Table 8. Summary of measurements on speciation of residential wood-burning emissions. All measurements except for those of *Dasch* [1982] used the dilution method of sampling.

Reference	**Comments**	**EC fraction**	**OC fraction**
Fireplaces			
Dasch, 1982	lower EC frac for hardwood	0.08-0.33	0.38-0.46
Rau, 1989	hard/soft wood	0.17	0.52
Hildemann et al., 1991	hard/soft wood	0.03-0.05	0.48
McDonald et al., 2000	hard/soft wood	0.14-0.15	0.59-0.63
Fine et al., 2001	Northeastern U.S. wood	0.04-0.31	0.75-1.0
Fine et al., 2002	Southeastern U.S. wood	0.01-0.18	0.74-1.0
Heating stoves			
Rau, 1989 [a]	hot burning, hard/soft wd	0.15-0.29	0.13-0.28
	cool burning, hard/soft wd	0.04-0.03	0.56-0.59
McDonald et al., 2000	hardwood	0.08	0.60

[a] Particles below 0.3 μm diameter. Composition was similar for larger particles. Ranges indicate means of different types of wood or testing and not standard deviations of the data.

Table 9. Black and organic carbon emissions from contained combustion (that is, all excluding open vegetative burning). Emissions are based on 1996 fuel-use data; units are Gg/year. *Prev96* applies emission factors from *Cooke et al.* [1999] to 1996 fuel-use data. Ratio of *Prev96* to the present work is also given.

	Black carbon			**Organic carbon**		
Region	**Central**	**(Low-High)**	**Prev96 (Ratio)**	**Central**	**(Low-High)**	**Prev96 (Ratio)**
North America	497	(375-815)	670 (1.3)	589	(338-1122)	935 (1.6)
Cent/S America	346	(246-711)	681 (2)	658	(379-1422)	1272 (1.9)
Europe	494	(367-887)	1218 (2.5)	686	(400-1317)	2249 (3.3)
Former USSR	193	(142-547)	633 (3.3)	220	(143-502)	944 (4.3)
China	1374	(873-3419)	2528 (1.8)	2115	(1171-4264)	5409 (2.6)
India	490	(314-1042)	1212 (2.5)	1366	(803-2474)	4576 (3.4)
Other Asia	742	(543-1469)	1255 (1.7)	1376	(873-2452)	4137 (3)
Pacific	35	(26-55)	50 (1.4)	29	(20-51)	144 (5)
Africa	500	(319-1028)	1019 (2)	1519	(921-2689)	6225 (4.1)
Middle East	103	(67-265)	88 (0.9)	171	(55-578)	84 (0.5)
Total	*4775*	*(3272-10237)*	*9355 (2)*	*8729*	*(5104-16872)*	*25974 (3)*

Table 10. Emissions of black and organic carbon from open vegetative burning, in Gg/year. *Prev96* applies emission factors from *Cooke et al.* [1999] to our estimates of dry matter burned. Ratio of *Prev96* to the present work is also given.

	Black carbon			**Organic carbon**		
Region	**Central**	**(Low-High)**	**Prev96 (Ratio)**	**Central**	**(Low-High)**	**Prev96 (Ratio)**
North America	113	(46-491)	283 (2.5)	1466	(593-3738)	2126 (1.4)
Central/So Amer	843	(424-2496)	1672 (2)	6238	(3464-13473)	12016 (1.9)
Europe	59	(28-230)	137 (2.3)	700	(307-1722)	997 (1.4)
Former USSR	98	(42-413)	240 (2.5)	1238	(510-3133)	1787 (1.4)
China	113	(62-255)	160 (1.4)	702	(399-1468)	909 (1.3)
India	87	(44-214)	127 (1.5)	500	(255-1120)	755 (1.5)
Other Asia	287	(133-931)	579 (2)	2086	(1087-4686)	4207 (2)
Pacific	164	(74-467)	273 (1.7)	1125	(518-2709)	1773 (1.6)
Africa	1466	(669-4217)	2629 (1.8)	10503	(5114-24567)	17871 (1.7)
Middle East	5	(3-15)	9 (1.7)	33	(17-75)	60 (1.8)
Total	*3234*	*(1524-9729)*	*6108 (1.9)*	*24592*	*(12263-56692)*	*42500 (1.7)*

Table 11. Breakdown of open-burning emissions (Gg/year).

	Black carbon				Organic carbon			
	Forest	Savanna	Ag res	Total	Forest	Savanna	Ag res	Total
North America	98	0	15	113	1393	0	73	1466
Central Am/Mexico	56	0	4	60	440	0	18	458
South America	425	388	30	843	3345	2749	144	6238
Europe	44	1	15	59	622	6	72	700
Former USSR	81	0	16	98	1160	0	78	1238
Middle East	3	0	2	5	21	0	12	33
Pacific	14	130	21	164	109	918	99	1125
Africa	293	1122	51	1466	2312	7947	244	10503
China	15	25	73	113	177	177	348	702
India	25	4	58	87	195	29	276	500
Other Asia	192	52	42	287	1513	372	201	2086
Total	*1244*	*1722*	*327*	*3293*	*11287*	*12198*	*1565*	*25050*

Table 12. Major differences in BC and OC predicted by current and previous inventories (in order of greatest difference in BC inventory). Units are Gg/year based on 1996 fuel-use data (for contained combustion) or annual averages (for open burning).

	Black carbon			**Organic carbon**		
Fuel/sector	**This work**	**Previous96**	**Difference**	**This work**	**Previous96**	**Difference**
Open burning/forest	1244	2980	-1736	11287	22984	-11697
Coal/power gen	7	1594	-1587	4	2324	-2320
Open burning/savanna	1726	2912	-1186	12225	19057	-6832
Diesel fuel/on-road	907	1917	-1010	289	1147	-858
Wood/residential	877	1921	-1044	3523	11538	-8015
Dung/residential	208	417	-209	750	5465	-4715
Coal/industrial	610	1181	-571	440	1251	-811
Diesel fuel/residential	98	369	-271	32	0	32
Coal/residential	517	761	-244	425	1683	-1258
Open burning/crop res	327	356	-29	1565	1518	47
Diesel fuel/off-road	682	696	-14	217	360	-143
Gasoline/transport	108	41	67	823	156	667
Ag waste/residential	393	90	303	1492	1231	261
Other	524	447	77	880	871	9
Total	8228	15682	-7454	33952	69585	-35633

Table 13. Largest contributors to variance in global BC emissions from contained combustion. The contribution to *variance* somewhat overstates the importance of the largest sectors in reducing *uncertainty*. If the variance in the two largest sectors were eliminated, the total variance would decrease by about 50%, but the uncertainty would decrease by only 30%.

Sector	**%Variance**	**Contributors**
Cokemaking	30%	Tech divisions; EF_{PM} and F_{BC}/uncaptured coking
Wood/residential	18%	Fuel use, F_{BC} and EF_{PM} (traditional cookstove), F_{BC} (open fire)
Coal/industrial	14%	EF_{PM} (brick kilns), tech divisions, F_{BC} (kilns)
Diesel/on-road	13%	Tech divisions, EF_{PM} and F_{BC} (superemitter), EF_{PM} (normal)
Ag waste/residential	8%	F_{BC}, fuel use, EF_{PM}
Coal/residential	5%	Tech divisions, all characteristics (open fire and cookstove), fuel use
Diesel/off-road	5%	EF_{PM} (all techs), tech divisions
An. waste/residential	2%	F_{BC}, fuel use, EF_{PM}
Wood/industrial	2%	F_{BC} (traditional), tech divisions, EF_{PM} (traditional)
Gasoline/transport	1%	Tech divisions, F_{BC} (2-strokes), EF_{PM} (2-strokes, superemitters)

Table 14. Largest contributors to variance in OC emissions from contained combustion.

Sector	**%Variance**	**Contributors**
Wood/residential	58%	Fuel use, EF_{PM} (trad cookstove), tech divisions, EF_{PM} (heating stove), F_{OC} (all), EF_{PM} (open fire, fireplace)
Gasoline/transport	10%	Tech divisions, EF_{PM} (2-strokes), F_{OC} (2-strokes)
Ag waste/residential	9%	Fuel use, F_{OC}, EF_{PM}
Wood/industrial	8%	EF_{PM} (traditional), tech divisions, F_{OC} (traditional)
Cokemaking	7%	Tech divisions; EF_{PM} and F_{OC}/uncaptured coking
Coal/industrial	3%	EF_{PM} (brick kilns), tech divisions, F_{OC} (kilns)
Coal/residential	2%	Tech divisions, all characteristics (open fire and cookstove), fuel use
Anim waste/residential	2%	Fuel use, EF_{PM}, F_{OC}
Diesel/on-road	1%	Tech divisions, EF_{PM} and F_{OC} (superemitter), EF_{PM} (normal)
Diesel/off-road	0.2%	EF_{PM} (all techs), tech divisions
Wood/charcoal prod	0.2%	Fuel use, EF_{PM}, FOC

Table 15. Largest contributors to variance in BC and OC emissions. The first line of the table shows the contribution of open vegetative burning to variance. The remaining lines give the relative importance of uncertainty in each sector. (1)=uncertainty is more than 25% of the central estimate of “contained” combustion; (2)=uncertainty 10-25% of the central estimate.

	N.Amer.		**S/C Amer.**		**Europe**		**Fr. USSR**		**Asia**		**Africa**	
Sector	**BC**	**OC**	**BC**	**OC**	**BC**	**OC**	**BC**	**OC**	**BC**	**OC**	**BC**	**OC**
Open veg burning	*36%*	*95%*	*93%*	*99%*	*6%*	*77%*	*17%*	*98%*	*23%*	*80%*	*98%*	*100%*
Agri waste/residential									1	1		
Anim waste/residential									2		1	1
Coal/industrial									1	2		
Coal/residential					2		2		1	2		
Cokemaking			2		2	2	1	1	1	1	2	
Diesel/off-road	1		2		2		2		2		2	
Diesel/on-road	1		1		1				2		2	
Diesel/residential	2				2							
Gasoline/transport	2	2	2	1							2	2
Wood/charcoal prod												2
Wood/industrial			2	1						2	1	1
Wood/residential	2	1	2	5	2	1		2	1	1	1	1

Figure 1. Schematic methodology for developing emission estimates.

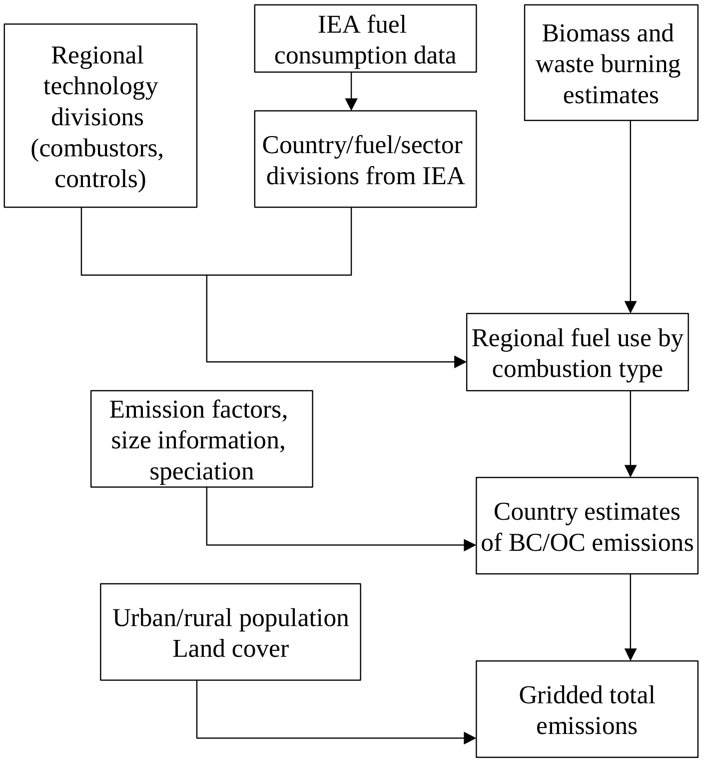


Figure 2. Emissions of black carbon aerosol. Top: “Contained” combustion, based on 1996 activity data; bottom: open vegetative burning, annual average. The color coding is an approximately logarithmic scale. Units are ng/m^2/sec.

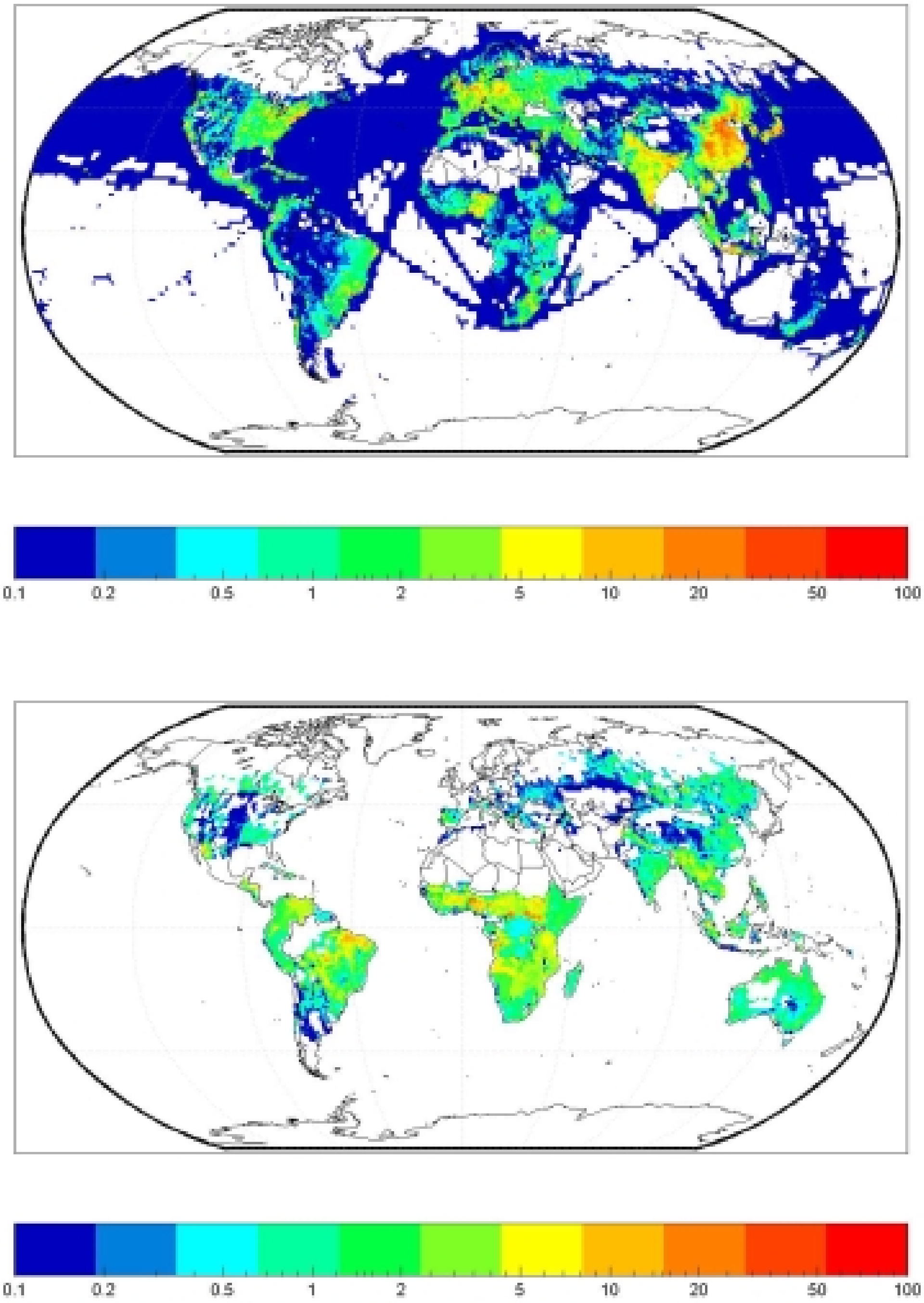


Figure 3. Emissions of primary organic carbon aerosol based on 1996 activity data. Top: “Contained” combustion, based on 1996 activity data; bottom: open vegetative burning, annual average. The color variation is approximately logarithmic and is the same as in Figure 2. Units are ng/m^2/sec.

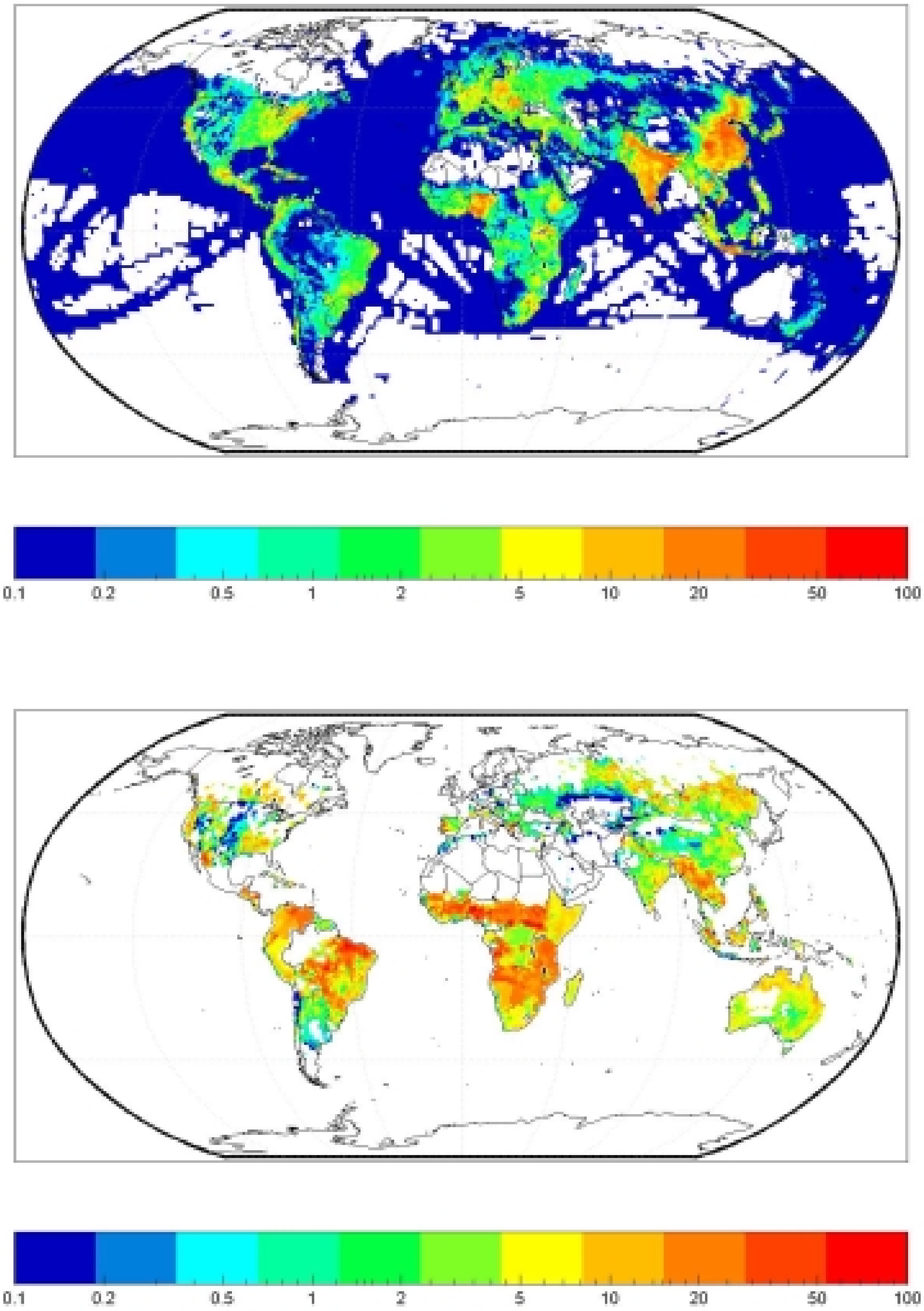
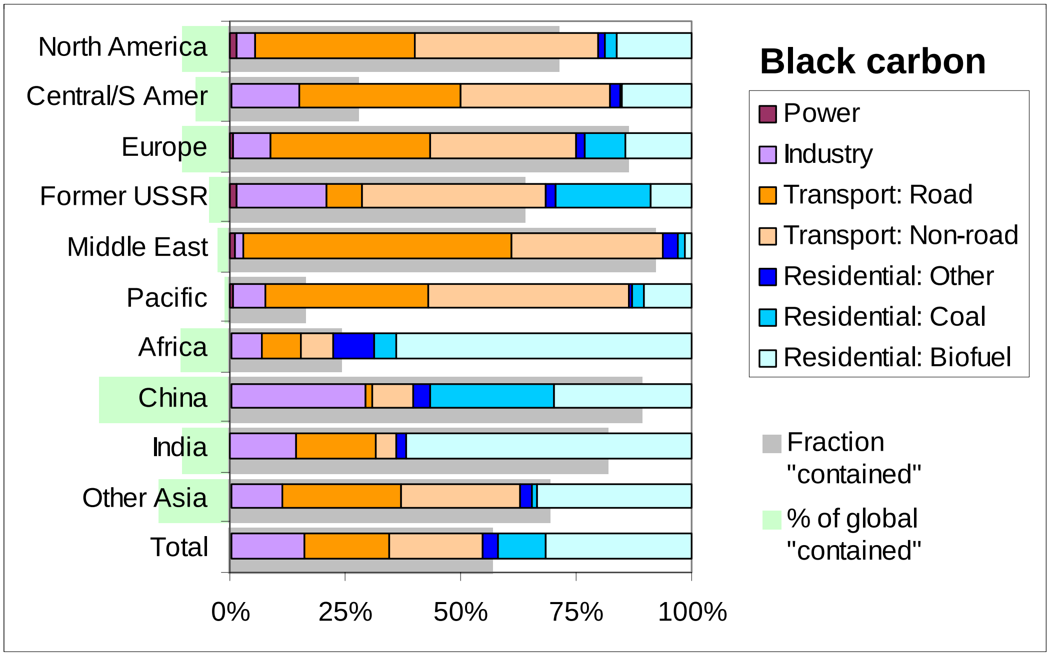


Figure 4. Sectoral contributions to emissions of black and organic carbon emissions. The gray bars behind the colored bars represent the fraction of emissions from “contained” combustion (that undertaken for energy use, excluding open burning) in each region. The green bars to the left indicate the relative contribution of each region to the total. The contributions are based on our central values, and carry all the caveats outlined in the text.



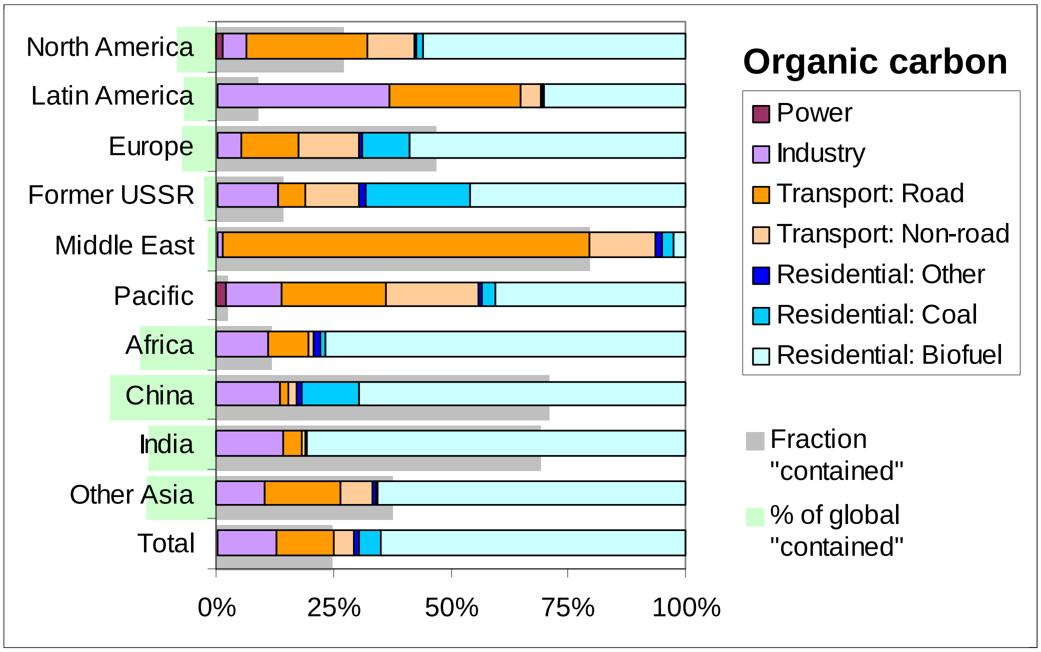


Figure 5. Regional differences among inventories of fossil-fuel BC emissions. *Low*, *Central*, and *High* are from this work and are based on 1996 fuel-use data. *Previous 84* is the previous inventory developed by *Cooke et al.* [1999], which is based on 1984 fuel-use data, from the emission grids distributed to the community. *Previous 96* is our calculation of emissions combining fuel-use data from 1996 and the emission factors given by *Cooke et al.* [1999]. The figure in the upper left shows zonal averages for all emissions. Clockwise from upper right are longitudinal averages over designated latitude bands to isolate contributions from major regions. All curves have been smoothed with a 3-degree running median filter.

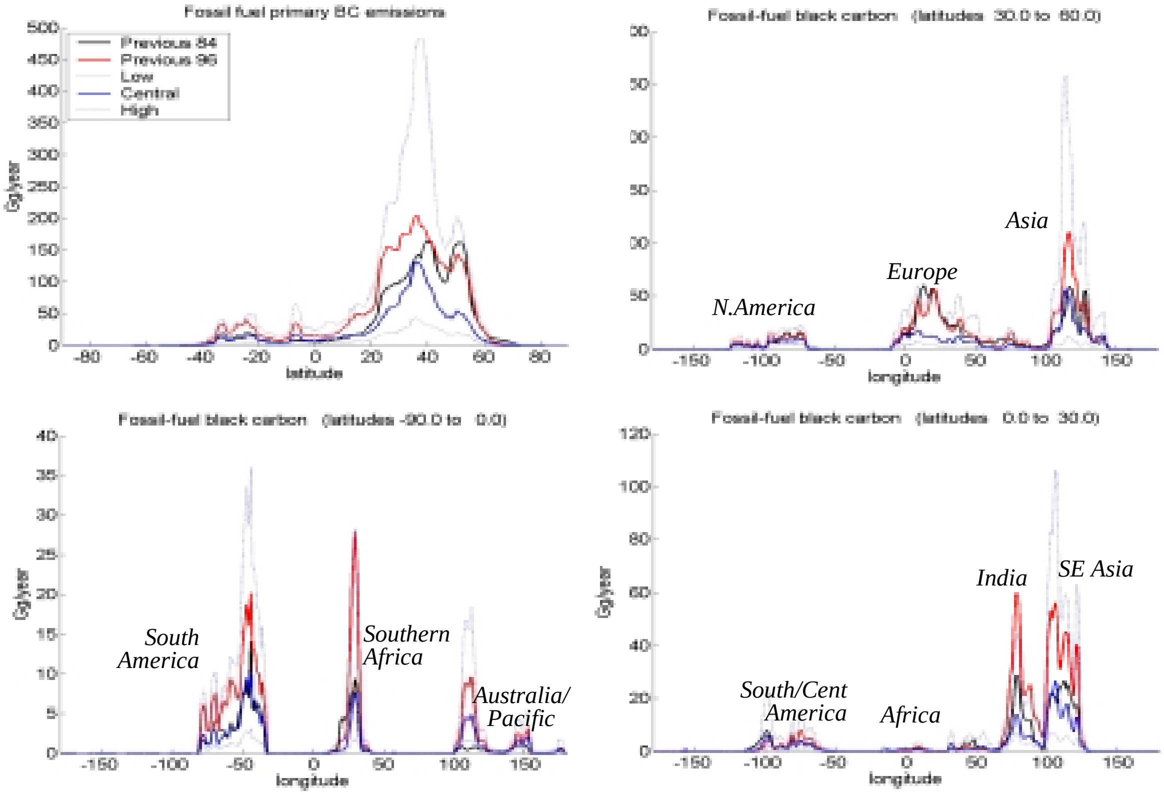


Figure 6. Regional differences among BC emission inventories of biofuel and open biomass. *Low*, *Central*, and *High* are from this work and are based on 1996 fuel-use data (for biofuel) and an average year (for biomass). *Previous 84* is the previous inventory developed by *Liousse et al.* [1996], from the emission grids distributed to the community. *Previous 96* applies the emission factors given by *Liousse et al.* [1999] to our activity data. The figure in the upper left shows zonal averages for all emissions. Clockwise from upper right are longitudinal averages over designated latitude bands. The curves have been smoothed with a 3-degree running median filter.