A technology-based global inventory of black and organic carbon emissions from combustion

Tami C. Bond,[1,2,3] David G. Streets,[4] Kristen F. Yarber,[4] Sibyl M. Nelson,[4] Jung-Hun Woo,[5] and Zbigniew Klimont[6]

[1]   We present a global tabulation of black carbon (BC) and primary organic carbon (OC) particles emitted from combustion. We include emissions from fossil fuels, biofuels, open biomass burning, and burning of urban waste. Previous "bottom-up" inventories of black and organic carbon have assigned emission factors on the basis of 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. Central estimates of global annual emissions are 8.0 Tg for black carbon and 33.9 Tg for organic carbon. These estimates are lower than previously published estimates by 25–35%. The present inventory is based on 1996 fuel-use data, updating previous estimates that have relied on consumption data from 1984. An offset between decreased emission factors and increased energy use since the base year of the previous inventory prevents the difference between this work and previous inventories from being greater. The contributions of fossil fuel, biofuel, and open burning are estimated as 38%, 20%, and 42%, respectively, for BC, and 7%, 19%, and 74%, respectively, for OC. 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 2, with uncertainty ranges of 4.3–22 Tg/yr for BC and 17–77 Tg/yr for OC. Low-technology combustion contributes greatly to both the emissions and the uncertainties. Advances in emission characterization for small residential, industrial, and mobile sources and top-down analysis combining field measurements and transport modeling with iterative inventory development will be required to reduce the uncertainties further.   *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0360 Atmospheric Composition and Structure: Transmission and scattering of radiation; 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; *KEYWORDS:* emission, black carbon, organic carbon, fossil fuel, biofuel, biomass burning

1.   Challenge of Carbonaceous Aerosols

1.1.   Carbonaceous Aerosols and the Climate System

[2]   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}.

[3]   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}, *Intergovernmental Panel on Climate Change* (*IPCC*) [2001]), 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*

[1]Department of Atmospheric Sciences, University of Washington, Seattle, Washington, USA.
[2]National Center for Atmospheric Research, Boulder, Colorado, USA.
[3]Now at Department of Civil and Environmental Engineering, University of Illinois at Urbana-Champaign, Urbana, Illinois, USA.
[4]Argonne National Laboratory, Argonne, Illinois, USA.
[5]Center for Global and Regional Environmental Research, University of Iowa, Iowa City, Iowa, USA.
[6]International Institute for Applied Systems Analysis, Laxenburg, Austria.

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 2 [*IPCC*, 2001]. 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]. 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. Section 1.3 gives a more complete discussion of the meaning of "black" and "organic" carbon.

[4]   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 the study by *IPCC* [1996]). Shortly thereafter, three-dimensional models predicted forcing of about +0.15 to +0.2 W m^{-2} for black carbon from fossil fuel combustion [*Haywood and Ramaswamy*, 1998; *Penner et al.*, 1998; *Myhre et al.*, 1998]. Since that time, higher forcing estimates have resulted from considering 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*, 2001; *Jacobson*, 2001; *Chung and Seinfeld*, 2002].

[5]   On human timescales, 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 other 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. However, because BC is such a potent warmer, the immediate warming impacts are much greater than those of CO_2. 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]. 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.

[6]   Along with the recognition of potentially important climatic effects, carbonaceous aerosols have received attention recently for other reasons. Primary particles from combustion, which are 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 the number of particles 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 [*Satheesh and Ramanathan*, 2000; *Luo et al.*, 2001], potentially affecting the sunlight available for agriculture [*Chameides et al.*, 1999].

[7]   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.

[8]   It is important to remember that all particles, not only carbonaceous particles, could be players in most of the detrimental effects listed above, 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

[9]   Previous estimates of the global annual source strength of black and organic carbon emissions are summarized in Table 1. 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].

[10]   Unlike CO_2, but like other trace species such as NOx and CO, the emission 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*, 2001; *Cooke et al.*, 1999]. We view the inventory presented here as an important but preliminary step toward a more complete understanding of global sources of carbonaceous aerosols. A multitude of efforts on tabulating and characterizing emissions occurs in many regions and at many governmental levels, and may not be incorporated here. We expect that the present inventory will evolve as other researchers share

Table 1. Previous Estimates of BC and OC Emissions From Combustion[a]

Reference	Year[b]	Fossil Fuel		Biofuel/Biomass		Combined	
		BC	OC	BC	OC	BC	OC
Turco et al. [1983]	?	1.3–8.3	–	1.3–14	–	2.6–22	–
Penner et al. [1993]							
Based on fuel consumption	1980[c]	6.6	–	6.0	–	13	–
Based on ratios with sulfur	–	–	–	–	–	24	–
Cooke and Wilson [1996]	1984	8.0	–	6.0	–	14[d]	–
Liousse et al. [1996]	–	6.6[e]	28	5.6	45	12	73
Cooke et al. [1999]							
<1 μm	1984	5.1	10	–	–	–	–
"Bulk" (all sizes)	1984	6.4	7.0	–	–	–	–
Andreae and Merlet [2001][f]	–	–	–	4.8	36	–	–
This work	1996	3.0	2.4	5.0	31	8.0	33

[a]Units are Tg/yr.
[b]Year is that of energy consumption data.
[c]Other years appear to be included for some locations and fuels.
[d]Excludes biofuels.
[e]From *Penner et al.* [1993].
[f]The literature contains several estimates of emissions from biomass burning. As this document focuses on revision of fossil and biofuel emission factors, we summarize only the latest open-biomass estimate here.

their knowledge and critiques, and we particularly welcome input from those with "on-the-ground" knowledge of source characteristics specific to each region.

[11] 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*, 2001; *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. Such 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

[12] It is important to remember that the terms used to classify carbonaceous aerosols depend upon the measurement method. 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 m²/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].

[13] 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 assortment 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 [*Heintzenberg*, 1982; *Hitzenberger et al.*, 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.

[14] Most source characterization studies do not measure black carbon, but rather so-called "elemental" carbon (EC). The nomenclature commonly used in atmospheric chemistry differs from the analytical chemist's definition of pure, graphitic EC, and 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 on the basis of 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 nonblack 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: 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.

[15] "Organic carbon," as used in climate research, usually refers to the carbon fraction of the aerosol that is not black. This term is an oversimplification because organic carbon may contain hundreds or thousands of different organic compounds with varying atmospheric behavior. Here, we differentiate between organic carbon (OC), or the mass of carbon that is not black, and organic matter (OM), which includes the hydrogen and oxygen bound to this carbon. OC is the quantity that results from thermal analysis of carbon aerosols, and the "EC-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.

2. Methodology

[16] 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 by *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

$$Em_{j,k} = \sum_l \sum_m FC_{k,l,m} \left[\sum_n EF_{j,k,l,m,n} X_{k,l,m,n} \right], \quad (1)$$

where

j, k, l, m, n species, country, sector, fuel type, fuel/technology combination;
     Em emissions;
     FC fuel consumption, kg/yr;
     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, where $\Sigma X = 1$ for each fuel and sector.

[17] 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 on the basis of mass emission measurements combined with data on the submicron and carbonaceous fractions of the emis-
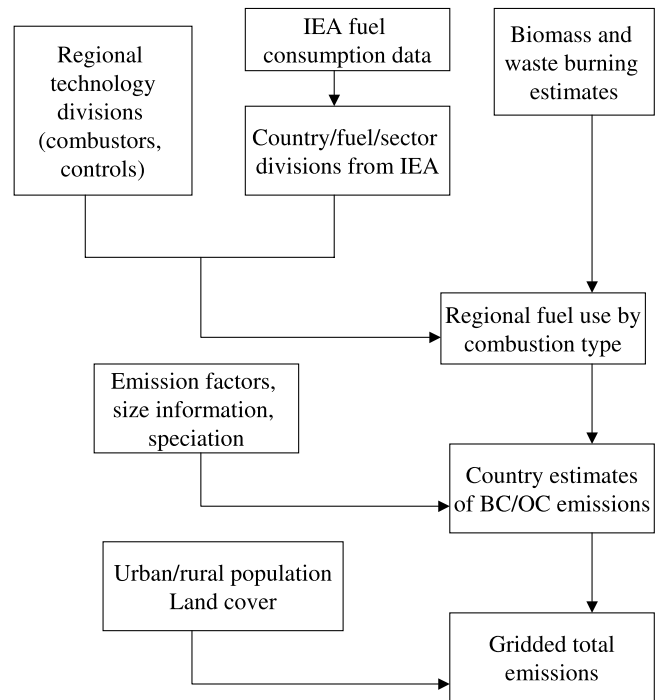

Figure 1. Schematic methodology for developing emission estimates.

sions as described in earlier papers. The net BC emission factor for submicrometer particles is given by

$$EF_{BC} = EF_{PM} \, F_{1.0} \, F_{BC} \, F_{cont}, \quad (2)$$

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, $F_{cont} = 1.0$.

[18] In this study, our values of EF_{PM} usually refer to 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. Defining the ratio between organic matter (OM) and organic carbon (OC) as R_{org}, emissions of organic matter are

$$EF_{OM} = EF_{PM} \, F_{1.0} \, F_{OC} R_{org} F_{cont}, \quad (3)$$

where F_{OC} is the fraction of fine particulate matter that is organic carbon, and the other terms are as defined

previously. In this paper, we will present emissions of organic carbon (that is, without the factor R_{org} applied) for comparability with previous inventories and with atmospheric measurements. 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. The OC tabulations do not account for atmospheric formation of particulate organic matter after emission.

[19] 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 compatibility with needs of climate researchers. 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 10 to 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 to 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

[20] 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" under the general category of industrial coal consumption). These latter divisions go beyond those in the fuel consumption data; they are specified by us for each world region and may be defined for each country if known.

[21] This procedure provides several advantages over simply assigning emission factors to broad sectors such as "industrial" or "domestic" combustion. (1) The choice of each emission factor is based on physical principles. (2) 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. (3) The change of emission factors over time can be represented physically, as a change in the relative fractions of technol-

ogies with different emissions. (The present paper, however, concentrates on current emissions.) (4) High-emitting technologies, rather than just sectors, can be identified as targets for mitigation.

2.2. Inventory Calculations

[22] We developed a program (Speciated Particulate Emissions Wizard, 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. Each value is keyed to a fuel/technology combination. The user interface provides statistical information about the measurements, including estimates of the expected value and variance when normal and lognormal distributions are assumed. On the basis of this information, the user chooses an emission factor for each technology and region. There is the ability to enter region-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. The region-specific technology mix represents the increased share of high-emitting devices, or "superemitters," that occurs when maintenance is less frequent. We assign emission characteristics for 12 regions: North America (excluding Mexico), Mexico, Latin America, Western Europe, Eastern Europe, Former USSR, Middle East, Africa, India, China, Other Asia, and Pacific (the latter includes Japan). When there is no information on technology divisions for a region, those parameters are estimated from practice in other regions.

[23] 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 4.5.

2.3. Spatial Distribution

[24] 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, Official energy statistics from the U.S. government, October 2001, available at http://www.eia.

Table 2. Technologies Gridded With Proxies Other Than Total Population

Gridding Proxy	Fuel/Technology Combination
Rural population	residential sector: biofuels/open fires and 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)[a]	forest burning
(Savanna cover) multiplied by (Fire counts 1999–2000)[a]	savanna burning
(Agricultural land cover) times (Fire counts 1999–2000)[a]	agricultural waste burning

[a]Gridded by appropriate land cover when fire counts appeared unreliable.

doe.gov); China (industrial, residential, and coking coal; *Fridley and Sinton* [2001]); and India (transport, power generation, and biofuels; *Tata Energy Research Institute* (*TERI*) [1996]).

[25] Emissions on the country level (or the state or provincial level for the United States, China and India) are distributed on a $1° \times 1°$ 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 many technologies, the proxy is total population. *Woo et al.* [2003a, 2003b] give further details on the data sets used for gridding our estimates. For example, a grid cell with 2% of the population of a country would receive 2% of the emissions of that country.

[26] The number of gridding proxies that can be defined is unlimited, so that future work can allow for more accurate spatial representation of emissions. Technologies that are gridded to proxies other than total population are listed in Table 2. We assume that only rural populations use firewood, agricultural waste, and coal in open fires and cooking stoves. While these rules are not strictly true, urban areas in all countries face more regulations and have more fuel alternatives than rural areas. Allocating solid fuel use to populous urban regions would greatly overestimate urban emissions.

[27] Open biomass burning is gridded according to the product of land cover and fire counts during the period 1999 to 2000 [*Woo et al.*, 2003b; *Streets et al.*, 2003a]. For interannual variability, these two years have been shown to be typical (not excessively dry or wet) for the amount of burning that occurred. In contrast to the ENSO-induced droughts of 1997–1998, which greatly enhanced Asian biomass burning, 1999 and 2000 were shown by *Duncan et al.* [2003] to be rather typical; the only unusual aspects were slightly enhanced burning in India and slightly reduced burning in Indonesia, both in 1999. Fire counts appeared unreliable in some regions, particularly Central America, 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 oceangoing 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

[28] While large uncertainties in the source strength of carbonaceous aerosols are widely acknowledged [e.g., *Cooke et al.*, 1999; *IPCC*, 2001], 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.

[29] 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 data are uncorrelated, and when the 95% confidence interval is less than 60% of the mean [*Olivier and Peters*, 2002]. Neither 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? (2) What and where are the major sources of uncertainty in these inventories?

[30] 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. Uncertainties inherent in the process of distributing the emissions spatially are not considered here; these may include the accuracy of the gridding proxies, the validity of those proxies to represent the spatial distribution of emissions, and assignment of border cells [*Olivier and Peters*, 2002].

[31] Often, true quantification of uncertainty is impossible, and expert judgment 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* [2001] estimate of forcing by black carbon from fossil fuel burning has a central value of +0.2, with a factor-of-2 uncertainty. However, the range presented by IPCC is largely due to the mixing state of black carbon. In this work, we derive a factor-of-2 uncertainty in the source strength alone, as

summarized in section 6.4. As the atmospheric lifetime and radiative properties also affect the forcing, the total uncertainty must be greater than a factor of 2, and the uncertainty in the IPCC estimate is probably underestimated.

3.1. Uncertainties in Emission Factors

[32] For this global approach, the smallest region considered is $1° \times 1°$ (approximately 100 km $\times$ 100 km). We assume that the number of sources in each grid box is large, so that the variability among individual sources does not affect the mean and standard deviation within the 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.

[33] When values of EF_{PM} are taken from *U.S. Environmental Protection Agency* (*U.S. EPA*)'s [1996] AP-42 document, we assign uncertainties on the basis of 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 distribution 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 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.

[34] 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 of particulate matter. Some 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.*, 2000] 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 is appropriate for 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.

[35] 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 (μ) 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

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

[36] 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 distribution 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 1 minus the desired confidence interval (95% in this case, or $\alpha/2 = 0.025$).

3.2. Composite Emission Characteristics

[37] 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.

[38] Since we assume lognormal distributions for some parameters and normal distributions for other 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 of the underlying distributions; this treatment is exact if the underlying distributions are lognormal, and exact for the expected value but not the variance if the underlying distributions are normal [*Craig*, 1936]. 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 emission factors

where available, if the combustion is thought to be representative.

3.3. Uncertainty in Technology Divisions

[39] 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 on the basis of available information, possibly engendering false confidence in the results.

[40] Therefore we include uncertainties in the technology divisions, such that our high-emission estimate has a greater contribution from higher-emitting technologies. This approach allows us to identify whether uncertainty results from limited knowledge about the technology mix 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 the technology divisions.

[41] The uncertainties in technology divisions are based entirely on estimation, as are the assumed mixes in many cases. In regions where we feel that our knowledge about sources is reasonable, we increase the fraction of the higher-emitting technologies so that they contribute an additional 10% of the total fraction (e.g., from 20% to 30%), and decrease the fraction of the lower emitting technologies by an equal amount. When we believe our understanding is more uncertain, we might alter the technology fractions by 30% of the total.

3.4. Summing Emissions

[42] 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 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.

[43] 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 emis-

sion factors, 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 emission factors 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.

[44] 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

[45] 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, such as 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 because of lack of information on the magnitude or even existence of emissions.

[46] 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 end use within sectors. In general, we have found that the IEA data have a greater number of end uses, 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 among 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.

[47] Although confidence intervals are not provided with official fuel-use statistics, 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 as follows: 10% for the industrial and power generation, and road transport 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, there is often no official reporting procedure for residential usage. 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 United States, 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.

[48] 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

[49] 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.

[50] 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. FAO data of earlier years were said to underestimate fuel consumption, but the agencies appear to be well aware of this problem, and the recent IEA data reflect higher estimates. Our estimates of total fuel consumption are higher than recent estimates by *Yevich and Logan* [2003], which are for an earlier year (1985).

[51] 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. *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. Biofuel production and consumption are not usually measured on an annual basis. Survey data from a single year may be extrapolated on the basis of population surveys, per capita consumption, and, sometimes, economic data (A. Koopmans, personal communication, 2001). *Kituyi and Kirubi* [2003] showed that a fuel-use measurement from a single day overpredicted the average by 50%. Per capita usage estimates are uncertain, varying by factors of 3 within the same country [*Smith*, 1987] and often remaining "frozen" from old studies that do not reflect economic changes [*Food and Agriculture Organization of the United Nations-Regional Wood Energy Development Programme in Asia* (*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 3 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 of a lack of independently developed estimates. 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, fuelwood comes not from felled trees, but from small branches from living trees [*deLucia*, 1983]. In Asia (at least), most fuel comes from nonforest land such as agricultural land and roadside trees, while periurban 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 (R. Overend, personal communication, 2001). The types of fuel available to a household may also vary on the basis of 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 to 30% [*Leach and Gowen*, 1987]. In addition to normal cooking

Table 3. Comparison of Biofuel Consumption Estimates for India[a]

Source	Year of Estimate	Rural	Urban	Total
National Council for Applied Economic Research (NCAER)[b,c]	1985	93		
Tata Energy Research Institute (TERI): rural energy database[b,d]	1991	252		
Tata Energy Research Institute: urban[b]	1988		11	
Planning Commission: Integrated Rural Energy[b]	1991	169		
International Energy Agency [1998b][e]	1991			265
Bhattacharya et al. [2000]	1991			125
Reddy and Venkataraman [2002b]	1996–1997	293	9	
Yevich and Logan [2003]	1985			220

[a]Units are Tg dry matter. Each of the sources also gave estimates for agricultural residue and dung, which are not included.

[b]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.

[c]The earlier year could contribute to the lower estimate; however, IEA estimates of total biomass energy are only 11% higher in 1985 and 1991.

[d]Central estimate; TERI also provided low and high estimates of 181 and 309 Tg.

[e]Converted from energy units assuming 15 MJ/kg. We created data for Jammu and Kashmir using values from surrounding states.

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 nonhousehold 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, kindling may include small wood, leftover charcoal, kitchen or urban waste, or plastic bags: in short, anything that burns readily. These uses may not be included in any or all of the country tabulations in the IEA database. Given the constraints just discussed, and the comparisons in Table 3, the uncertainty estimates may be optimistic.

[52] IEA biofuel 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 (Netherlands Energy Research Foundation (ECN), Phyllis: Database for biomass and waste, January 2003, available at http://www.ecn.nl/). The values used are as follows: wood, agricultural residue, and municipal solid waste, 15 MJ/kg; charcoal, 24 MJ/kg; and animal waste (dung), 12.5 MJ/kg. For animal waste, 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

[53] 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] (available at http://www.un.org.kh/fao/environment/index. html). 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] and other Internet sources and applied these to the urban populations. The United Nations Human Settlements Programme (http://www. unhabitat.org, 2000) reports the fraction of waste burned for a few cities, and we estimated the fraction burned in

all urban areas on the basis of these data points. These values are quite uncertain, as waste-burning practice could be highly localized depending on the available alternatives. Waste generation in rural areas is even more difficult to estimate. In developing countries, waste generation is low because goods are inherently recycled. Availability of packaged goods is usually limited, but the packages (including plastic) are burned when available, often for energy use. We have assumed that waste burning in rural areas of developing countries is zero. In the United States and Europe, we assume that vegetative waste is burned as estimated by the *U.S. EPA* [2002]. We have not accounted for combustion of industrial quantities of waste, such as used tires or electronic components, which may contribute to local air pollution. Our estimates of open waste burning are about 33 Tg/yr worldwide, including 14 Tg in Asia (including China and India) and 5 Tg in Africa. We assign uncertainties of 200% to these estimates.

4.4. Open Biomass Burning

[54] 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.*, 2000].

[55] Quantifying global biomass burning involved a comprehensive search of available data including scientific papers, individual country communications to the UNFCCC Secretariat, Global Fire Monitoring Center publications, and FAO statistics. A detailed description of the general methodology followed is given by *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 4. The estimate of total global vegetation burned (excluding biofuels) in this new inventory, 5900 Tg, is in good agreement

Table 4. Tabulation of Open Biomass Burning by World Region[a]

	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

[a]Units are Tg dry matter.

with the estimate of 5700 Tg reported by *Andreae and Merlet* [2001], which is based on unpublished data of J. A. Logan and R. Yevich. It also agrees with the estimate by *van Aardenne et al.* [2001] of 6000 Tg, back-calculated from CO emissions from biomass burning. Our estimate is lower than that of *Galanter et al.* [2000], 6900 Tg. In all cases there are some differences among vegetation types and countries/regions. We have used uncertainties of 150% for activity data.

[56] For Africa, forest burning values were derived from *FAO* [1997], and crop burning was calculated on the basis of FAOSTAT following the method outlined by *Streets et al.* [2003a]. For some areas where data was unavailable, burning was assumed to be zero on the basis of expert opinion. Savanna burning estimates were based on a combination of papers [*Delmas et al.*, 1996; *Menaut et al.*, 1996; *Hao and Liu*, 1994]. Burning estimates given by *Delmas et al.* [1996] and *Menaut et al.* [1996] were disaggregated and assigned to countries on the basis of spatial distribution of emissions data by *Hao and Liu* [1994]. For tropical African countries in regions not included by either *Delmas et al.* [1996] or *Menaut et al.* [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 *United Nations Economic Commission for Europe* (*UN-ECE*) [2000a, 2001a] papers. Other sources used included a number of country communications to the UNFCCC Secretariat, *UN-ECE* [2000a, 2001a], and *Akeredolu and Isichei* [1996].

[57] Data on South and Central America (including the Caribbean) came from *Hao and Liu* [1994], country communications to the UNFCCC Secretariat, and *FAO* [1997]. Crop burning was calculated on the basis of FAOSTAT (FAO, FAOSTAT agricultural database, 2001, available at http://apps.fao.org/page/collections?subset = agriculture) 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 *Secretaria del Medio Ambiente*, Recursos Naturales y Pesca [SEMARNAP] [2001] (available at http://www.semarnat.gob.mx/wps/portal/) and for the United States from *U.S. EPA* [2001]. Forest burning in Canada came from Environment Canada (CAC emission summaries, 1995, available at http://www.ec.gc.ca/pdb/ape/cape_home_e.cfm) and *Johnston* [2001] (available at http://www.ciffc.ca/news/canreport01.pdf). 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 *UN-ECE* [1993a, 2003], *Fearnside* [1996], and *Sanhueza* [1996].

[58] International Forest Fire News [*UN-ECE*, 1990a, 1990b, 1990c, 1992, 1993b, 1995a, 1995b, 1999a, 1999b, 2000b, 2000c, 2001b, 2002] papers were the most common data source used for Europe and the Middle East. Country communications to the UNFCCC Secretariat were also used for many countries, as was CORINAIR '94 [*European Environment Agency* (*EEA*), 2000a] (2002 update available at http://reports.eea.eu.int/EMEPCORINAIR3/en). *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 to the UNFCCC Secretariat, and from CORINAIR '94 [*EEA*, 2000a]. Burning was assumed to be zero in some European countries for which statistics could not be found.

[59] 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 UNFCCC Secretariat 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

[60] In this section, we discuss the development of emission factors. The discussion contains detailed reviews of emission measurements and underlying generation mechanisms. Readers who are more interested in the final results may skip to section 6 after this cautionary paragraph, in which we provide three warnings on the estimation of BC emission factors. (Similar cautions apply to OC as well.) (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 unresolved 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 large amounts of CO and relatively small amounts of 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.

[61] For the reasons just stated, BC and OC emission factors must be based on direct measurements, which are discussed in this section. It is not possible to provide a full justification for the large number of choices we have made for each emission characteristic and technology division. We discuss only the choices that affect emission estimates the most. We have undertaken an extensive review of the literature in order to assign particulate emission factors 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 emission factors 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. For that reason, this tabulation has eliminated some references that appear in other literature.

[62] There are some exceptions to the rule of accepting only primary references. One of those is the U.S. EPA's [1996] "Compilation of Air Pollutant Emission Factors" (hereinafter referred to by its report number, AP-42). We have examined the supporting information and background documents, but not the original reports if a recommended emission factor 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 3.1, 1999, available at http://www.epa.gov/ttn/chief/software/speciate/index.html], and cite some documents that are difficult to obtain. When other information is not available, we use the values from these tabulations.

[63] Values of EF_{PM} for stationary combustion are given in Tables 5 and 6. Table 5 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 6 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. In Table 6, the uncertainty ranges indicate the half-width of the 95% confidence interval. As discussed in section 3, these confidence intervals may be asymmetric about the mean; an uncertainty greater than the mean does not imply a possibility of negative emission factors.

[64] Table 7 summarizes mobile sources, with the investigation required to choose EF_{PM} for these sources discussed in section 5.4. Table 8 shows technology divisions for fuel/sector combinations with the largest fuel uses or emissions, for three representative regions. One of these regions is primarily industrialized (North America), one an economy in transition (former USSR), and one is considered a developing country (India). In our model, technology divisions may be different among countries of the same economic status, so that Table 8 allows only a partial reconstruction of the inventory. The BC and OC emission factors listed in Tables 9 and 10 result from combining the emission characteristics with technology divisions.

[65] 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 emission factors when those choices have resulted in significant revision of global inventories. Differences in opinion, such as 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 and variations in the technology mix to choose appropriate emission characteristics, and from drawing on data that has recently become available. The present work, combined with the background given by 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

[66] 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.

[67] 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 postflame 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

Table 5. Central Values of Particulate Matter Emission Characteristics for Stationary Combustion, Excluding Residential Solid Fuels[a]

Fuel	Technology	EF_{PM}, g/kg	$F_{1.0}$	F_{BC}	F_{OC}	F_{cont}
Biofuel	stoker	2.2[b]	0.86	0.05[c]	0.2[c]	0.4–1.0[d]
Biofuel	traditional[e]	10	0.85	0.1	0.6	1.0
Briquettes	stoker[f]	2.5	0.25	0.01	0.02	0.3–1.0
Brown coal	cyclone	33	0.17	0.006[g]	0[g]	0.03–1.0
Brown coal	pulverized	29	0.09	0.006[g]	0[g]	0.05–0.9
Brown coal	stoker	17	0.11	0.05[h]	0.66[i]	0.06–1.0
Hard coal	cyclone	1.3	0.15	0.006[g]	0[g]	0.03–1.0
Hard coal	pulverized	12[j]	0.09	0.006[k]	0[l]	0.05–0.9
Hard coal	stoker	4.2[m]	0.33	0.20[n]	0.04[n]	0.05–1.0
Hard coal	traditional brick kiln[o]	10	0.9	0.5	0.4	1.0
Coking coal[p]	coke oven[q]	5.8	0.35	0.48[r]	0.34[r]	0.33
Coking coal	coke oven (uncaptured)[r]	20	0.5	0.48[r]	0.34[r]	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 combustion	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]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.

[b]Average of AP-42 emission factors (2001 revision), including upward revision and increased uncertainty to account for higher emission factors from bagasse.

[c]Average of several wood-fired boilers in SPECIATE (U.S. EPA, SPECIATE 3.1, 1999).

[d]Advanced boiler control based on prevalence of controls on wood-fired industrial boilers in the United States (J. Eddinger, U.S. EPA, personal communication, 2003) combined with submicron escape efficiency.

[e]No data; see section 5.6.2.

[f]All values except F_{cont} taken from *Ge et al.* [2001]; assumed that $F_{1.0} = 0.85\ F_{2.5}$.

[g]No data: assumed same as hard coal in pulverized combustor.

[h]Based on *Bond et al.* [1999b].

[i]Based on *Pinto et al.* [1998] for residential combustion, but with a high uncertainty.

[j]Average of various types, with uncertainty accounting for variation.

[k]*Fisher et al.* [1978], *Mamane et al.* [1986], *Olmez et al.* [1988], *Querol et al.* [1995], and *Smith et al.* [1979].

[l]*Olmez et al.* [1988].

[m]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.

[n]See discussion in section 5.2.2.

[o]No information; used value intermediate between home heating stove and stoker with high uncertainty. See discussion in section 5.3.

[p]Emission factors are per fuel input.

[q]Sum of several processes in coking, primarily door leaks.

[r]See discussion in section 5.2.3.

[s]SPECIATE (U.S. EPA, SPECIATE 3.1, 1999).

[t]*Hildemann et al.* [1991], B. Wehner (manuscript in preparation, 2003), and *Goldstein and Siegmund* [1976].

[u]*U.S. EPA* [1991] NEVES.

[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, also personal communication, 2002], and *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] and *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 data; see section 5.7.

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.

[68] 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.

Table 6. Compilation of Particulate Matter Emission Factors for Residential Solid-Fuel Combustion

Fuel/Technology	References[a]	EF_{PM},[b] g/kg
Fossil Fuels		
Bituminous coal/apartment building stoker	2.0–2.4 [*Beijing Environmental Protection Bureau*, 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, also personal communication, 2002]; 2.4 ± 0.7 [*Smith et al.*, 2000]	4.1 ± 4.8
Wood/apartment 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 [*U.S. EPA*, 1996]; 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) [*U.S. EPA*, 1996], 2.3–7.2 [*McDonald et al.*, 2000]	15 ± 8
Wood/traditional cookstove	6.4–8.9 [*Smith*, 1987],[c] 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],[c] 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, also personal communication, 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, also personal communication, 2002]	3.8 ± 2.1

[a]Ranges indicate multiple sources measured, while "±" indicates standard deviation of same source.
[b]Under "EF_{PM}," "±" indicates half-width of 95% confidence interval, not necessarily centered about the mean. See text for discussion of other emission characteristics.
[c]Citing conference proceedings by S. S. Butcher et al.

[69] Coarse particles are not built during the combustion process; 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. Emissions of PM consist mostly of small particles if (a) large particles are not provided to the combustion, as is the case with all gaseous and most liquid fuels, or (b) air velocities are low, as is the case in most residential combustion.

[70] 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 particles of different composition result from quite separate mechanisms and are governed by disparate aspects of the combustion process. 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, when both fine and coarse particles are emitted from the same process, they result from different formation mechanisms. The composition of total particulate matter is dominated by the coarse particles, and cannot be used to represent that of fine particulate matter.

5.2. Coal: Industrial Combustion
5.2.1. Pulverized Coal

[71] Technologies for burning coal vary widely (for examples, see *Babcock and Wilcox Company* [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 burn suspended par-

Table 7. Central Values of Particulate Matter Emission Factors for Mobile Sources[a]

Fuel	Conditions	EF_{PM}, g/kg	$F_{1.0}$	F_{BC}	F_{OC}
Aviation fuel, aircraft	all	0.14[b]	–	0.7[b]	0.2[b]
Diesel, on-road general	standards in place	1.5[c]	0.86[d]	0.66[e]	0.21[e]
Diesel, on-road general	standards beginning	3.5[f]	0.86[d]	0.66[e]	0.21[e]
Diesel, superemitter	all	12[g]	0.86[d]	0.66[e]	0.21[e]
Diesel, farm vehicles	all	4.0[h]	0.86[d]	0.66[e]	0.21[e]
Diesel, nonfarm off-road vehicles	all	5.5[h]	0.86[d]	0.66[e]	0.21[e]
Diesel and heavy oil, ships	all	1.8[i]	0.86[d]	0.66[e]	0.21[e]
Diesel, railroad	all	2.7[j]	0.86[d]	0.66[e]	0.21[e]
Gasoline, all vehicles	standards in place	0.15[k]	0.85[d]	0.34[e]	0.36[e]
Gasoline, all vehicles	standards beginning	0.50[l]	0.85[d]	0.34[e]	0.36[e]
Gasoline, superemitter	–	2.0[m]	0.85[d]	0.34[e]	0.36[e]
Gasoline, two-stroke	standard	15[n]	0.95	0.05[o]	0.79[o]
Gasoline, two-stroke	high-emission practice[p]	30[n]	0.95	0.05[o]	0.79[o]

[a]References in footnotes describe primary sources of data, but see section 5.4 for full discussion of analysis underlying these choices. We typically do not rely on traffic tunnel measurements for reasons discussed in the text.

[b]EF_{PM} calculated from BC emission rates given by *Penner et al.* [1999]; *Petzold and Dopelheuer* [1998]; *Petzold et al.* [1999], increased by OC/BC ratio given by *Hopke* [1985].

[c]Based primarily on data from *Clark et al.* [2000] and over 400 dynamometer tests summarized by *Yanowitz et al.* [2000], using vehicle age to create a composite.

[d]*Durbin et al.* [1999a].

[e]Based on review by *Gillies and Gertler* [2000].

[f]Estimated from *Faiz et al.* [1996], *Mäkelä* [1995], and older vehicles in the study by *Yanowitz et al.* [2000].

[g]*Faiz et al.* [1996]; *Shah and Nagpal* [1996a]; *Karim* [1999]; *McCormick et al.* [2003], as well as inferences from references under following note m.

[h]*U.S. EPA* [1991]; *Samaras and Zierock* [1995]; *BUWAL* [1996]; *Kean et al.* [2000]. For these and all other off-road uses, measured values of $F_{1.0}$, F_{BC} and F_{OC} were not available and were assumed equal to on-road engines.

[i]*U.S. EPA* [1991, 2000].

[j]*U.S. EPA* [1997].

[k]*Kirchstetter et al.* [1999]; *Durbin et al.* [1999a]; *Chase et al.* [2000].

[l]*Williams et al.* [1989]; *Durbin et al.* [1999a].

[m]*Hansen and Rosen* [1990]; *Lawson* [1993]; *Sagabiel et al.* [1997]; *Durbin et al.* [1999b]; *Lawson and Smith* [1998]; *Bishop et al.* [2000]. Note that some of these studies measure CO only.

[n]*U.S. EPA* [1991]; *Patschull and Roth* [1995]; *Faiz et al.* [1996]; *Kojima et al.* [2000]; *Kojima and Khaliquzzaman* [2002].

[o]*Sakai et al.* [1999]; *Kojima et al.* [2000].

[p]For definition, see discussion in section 5.4.2.

ticles. In those combustors, soot forms in a cloud around the coal particle as volatile matter is ejected [*Seeker et al.*, 1981], but encounters a high-temperature environment shortly after it is emitted and is rapidly oxidized to CO_2. 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. This type of combustion is the preferred technology for power generation.

[72] Although the literature contains several detailed reports of speciated 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 lower than those in the previous work. However, volatile organic material is released during the combustion process, and either staged combustion [*Veranth et al.*, 1998] or poor mixing could result in higher carbonaceous emissions.

[73] 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 in earlier work [*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.*, 2000; *Winiwarter et al.*, 2001], expert estimates and communication with national experts participating in the UNECE 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

[74] 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 Company*, 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

Table 8. Technology Divisions for Sample Regions and Fuel/Usage Combinations, for Those Fuel/Usages With Either Large Emissions or Large Fuel Use[a]

Fuel	Sector or Usage	Technology	North America	Former USSR	India
Diesel fuel	transport	vehicle improved[b]	0.95	0.6	–
Diesel fuel	transport	vehicle unimproved[b]	–	0.3	0.8
Diesel fuel	transport	superemitter	0.05	0.1	0.2
Hard coal	power generation	cyclone/cyclone	–	–	0.05
Hard coal	power generation	cyclone/ESP	–	–	0.05
Hard coal	power generation	pulverized coal/ESP	0.96	0.8	0.7
Hard coal	power generation	pulverized	–	0.1	0.1
Hard coal	power generation	coal/scrubber	0.02	0.08	0.05
Hard coal	power generation	stoker/cyclone	0.02	0.02	0.05
Hard coal	power generation	stoker/ESP			
Hard coal	industry	brick kiln	–	–	0.08
Hard coal	industry	cyclone/cyclone	–	–	0.05
Hard coal	industry	cyclone/ESP	–	–	0.11
Hard coal	industry	pulverized coal/cyclone	0.1	0.1	–
Hard coal	industry	pulverized coal/ESP	0.6	0.4	0.16
Hard coal	industry	stoker/cyclone	0.1	0.3	0.38
Hard coal	industry	stoker/ESP	0.3	0.1	0.11
Hard coal	industry	stoker/uncontrolled	–	0.1	0.11
Middle dist. oil	industry	boiler	0.24	0.225	0.2
Middle dist. oil	industry	boiler/superemitter	0.01	0.025	0.05
Middle dist. oil	industry	off-road engine	0.71	0.68	0.6
Middle dist. oil	industry	off-road superemitter	0.04	0.07	0.15
Coking coal	coking	captured	0.999	0.95	0.95
Coking coal	coking	uncaptured	0.001	0.05	0.05
Hard coal	residential	heating stove	1.0	0.07	–
Hard coal	residential	open fire	–	0.01	–
Hard coal	residential	stoker/cyclone	–	0.2	–
Hard coal	residential	stoker/uncontrolled	–	0.72	0.5
Hard coal	residential	traditional cookstove	–	–	0.5
Gasoline	transportation	two-stroke	0.01	0.025	–
Gasoline	transportation	two-stroke high emissions	–	0.025	0.35
Gasoline	transportation	vehicle improved[b]	0.94	0.25	–
Gasoline	transportation	vehicle unimproved[b]	–	0.6	0.52
Gasoline	transportation	superemitter	0.05	0.1	0.13
Wood	industry	stoker	0.5	0.5	0.3
Wood	industry	stoker/cyclone	0.5	0.5	0.2
Wood	industry	traditional industry	–	–	0.5
Wood	residential	fireplace	0.25	0.02	–
Wood	residential	heating stove	0.72	0.88	–
Wood	residential	improved cookstove	–	–	0.04
Wood	residential	open fire	–	0.1	0.30
Wood	residential	traditional cookstove	–	–	0.66

[a]Central values only are listed here.
[b]In this table, "improved" vehicles are those existing in regions where many years of regulation have led to decreasing emissions. "Unimproved" vehicles are those where implementation is just beginning.

not used for modern power plants, but are still employed in smaller applications, as well as in countries with less-advanced technology.

[75] Stokers, and coal beds in general, can have high emissions of carbonaceous particles and especially of black carbon, because the volatile matter ejected during coal heating is not well mixed with air. Furthermore, 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.

[76] 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

Table 9. Range of Emission Factors for Black Carbon, Showing Variations in Regional Emission Factors[a]

Fuel	Power[b]	Industry[b]	Residential	Transport and 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
Diesel fuel	0.25	3.4–4.4	0.06–4.0	vehicles (1.3–3.6), rail (0.51), ships (0.34), tractors (2.6–3.7)
Hard coal, coking coal	0.002–0.009	0.013–1.2	0.76–5.4	rail (3.0), coking (0.32–1.2)
Heavy fuel oil	0.04	0.04	0.07	ships (0.34)
Gasoline, aviation fuel, kerosene	–	0.14	0.9	vehicles (0.08–0.43), aircraft (0.10)
LPG	–	–	0.20	none
Natural gas	0	0	0	transport (0)
Waste	0.013	0.013	4.2	open (5.5)
Biofuels				
Animal wastes	–	–	0.53	none
Charcoal	–	w/coke	1.0	none
Agricultural residues	–	w/wood	1.0	none
Wood	0.044	0.08–0.55	0.3–1.4	charcoal production (0.2)
	Savanna	Crop Residues	Forest	
Biomass burning	0.48	0.69	0.56–0.61	

[a]Units are g/kg dry matter. The ranges show the variability in central values used for each region, with differences being due to regional variations in technology mix. Categories with only one value do not have a range of technologies. The ranges shown here do not encompass uncertainties resulting from imperfect knowledge of emission factors and other emission characteristics.
[b]Although technologies used in power generation and industry have the same EFs, the fuel use is apportioned to different technologies within the two sectors.

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 on U.S. stokers gave a similar BC/OC ratio (J. Schauer, personal communication, 2003). However, the emission factor measured by *Ge et al.* [2001] was much lower than those tabulated by the *U.S. EPA* [1996], suggesting better soot burnout in the stoker in their study. We use F_{BC} of 0.20 and F_{OC} of 0.04, with a high uncertainty.

[77] Although lignite is burned in the same types of combustors as hard coal, its emissions are quite different. It has a lower heating value than hard coal, resulting in cooler 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. Available measurements of lignite emissions, both in industrial plants [*Bond et al.*, 1999b] and home heating applications [*Pinto et al.*, 1998; *Bond et al.*, 2002] show that the emitted particles

Table 10. Range of Emission Factors for Primary Organic Carbon[a]

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

[a]Units are g/kg dry matter. Footnotes for Table 9 apply.

absorb little light. For this reason, the value of F_{BC} given in Table 5 is lower than those for hard coal.

[78] Table 6 does not give emission factors for anthracite, which is classified as hard coal. 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 between anthracite and bituminous coal is available. Most of the emission factors for residential heating stoves in Table 6 were measured in the early 1980s; the lone recent exception shows 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

[79] Particulate matter is emitted from sources in the iron and steel industry, many of which release mostly metallic particles. We have estimated BC and OC from coke making (coke ovens) and iron making (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 the process and emission factors are quite different from other types of combustion, and because it could be a large contributor to emissions of carbonaceous aerosols.

[80] Coke is the residue of a particular class of coal, created by heating the coal under specified conditions and driving off the volatile matter. In many countries, coke ovens are subject to strict emission regulations regarding the capture of the lost carbon. 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 to 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 900 to 1100°C [*U.S. Steel*, 1985]. On the basis of 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.

[81] We have not found values of F_{BC} or F_{OC} especially for coke making. 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. Assuming that these elements and associated oxygen would account for 5% of the emissions, we have assumed that 95% of the emissions are carbonaceous. We also assume that the emissions are half BC and half OM for both captured and uncaptured emissions, again based on experience with heating coal to the relevant temperature range.

[82] 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 of PM 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.

[83] 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 may be recovered for other uses, in addition to the fraction that is 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.

[84] In 1996, China was the largest coke producer in the world. On the basis of 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. Prior to cleanup in that province, it was said that 80% of the coke was made in informal coke ovens (U.S. Embassy, www.usembassy-china.org.cn/sandt/Shanxi.htm, June 2001). 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. Coke making 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.

[85] We have found no information regarding the prevalence of "beehive" coke production in countries other than China (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; these large companies are likely to have more advanced practices. 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 United States and Europe, and 10% is uncaptured in the rest of the world.

5.2.4. Brick Kilns

[86] 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 emission factor for heating stoves and stokers, with a 200% uncertainty, and the values of $F_{1.0}$ and F_{BC} from these heating stoves.

[87] *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 "nonmetallic minerals" end use is used in polluting kilns.

5.3. Coal: Residential Combustion

[88] 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 emission factors for residential coal, we combined all types of burning and applied a general emission factor to this sector. Because this sector is so important, we 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 6 summarizes emission factors from the literature, as well as central values and uncertainties in emission factors.

[89] 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 PM, 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 (T. C. Bond, unpublished data, 2002) and elsewhere (L. Tian, personal communication, 2002) have suggested higher emission factors.

[90] *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 use our own (as yet unpublished) measurements of emissions from bituminous coal. We burned five samples of coal from the United States 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

[91] We expect regional differences in vehicle emissions for several reasons. First, vehicles in many locations have not been required to respond to increasingly stringent emission regulations, as they have in the United States, Europe, Japan, 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].

[92] Regional inventories of vehicle emissions typically account for a range of variables, especially characterization of vehicles 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 they do not include "malperformance," driver behavior, and altitude, so that the average emission factor 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.

[93] Our approach consists of the following: (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 super-

emitters 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 onward [*Faiz et al.*, 1996]. A more detailed approach will be required to estimate BC emission trends in subsequent years.

[94] Because of the difficulty of ensuring a representative vehicle sample and driving conditions, even the appropriate emission factor 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].

[95] 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. However, because opacity is an optical measurement, and an optical definition of BC is most appropriate for radiative transfer estimates, opacity measurements could serve to bound BC emissions. Theoretical calculations over a range of particle sizes and refractive indices [*Bond*, 2000] and measurements [*Wu et al.*, 1997; *Bond*, 2000] suggest that absorption is rarely more than 75% of extinction at combustion aerosol sizes. Opacity measurements can set an upper limit on BC emission factors by combining this fact with a mass balance on the combustion process, and assuming a value of 7 m^2 absorption/g BC, although OC emissions are not similarly bounded. With these caveats in mind, we discuss our choice of emission factors.

5.4.1. Gasoline Vehicles

[96] 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 to 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 existing vehicles. For other regions, we use $EF_{PM} = 0.5 ± 0.4$ g/kg.

[97] How much could superemitting vehicles contribute? *Sagabiel et al.* [1997] reported that smoking vehicles emitted about 10 times the PM of nonsmokers 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] (available at http://www.nfraqs.colostate.edu/nfraqs/). 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.

[98] 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, 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 vehicles and, on average, emit about 17 times the CO of the remainder.

[99] Supposing that superemitters emit 10 to 15 times the U.S. average, the value of EF_{PM} for these high-emitting vehicles would be about 2 g/kg, in accordance with *Durbin et al.* [1999b]. On the basis of the preceding discussion, we choose a central value of 5% superemitters for the United States 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. On the basis of the discussion in the diesel section below, we assume a superemitter fraction of 20% for Asia and Latin America on the basis of these reports and, somewhat arbitrarily, 20% for Africa and 10% for Eastern Europe and the former USSR. The high uncertainty is represented in the ranges of superemitter fractions: 1−10%, 5−20%, and 5−60% for regions whose central values were 5%, 10% and 20%, respectively. 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. On the basis of a reanalysis of data presented in that paper, we estimate F_{BC} as 34% ± 12% and F_{OC} as 21 ± 6%.

5.4.2. Two-Stroke Gasoline Engines

[100] 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.

[101] *Kojima et al.* [2000] summarized measurements on five U.S. motorcycles with EF_{PM} averaging 6 g/kg. These authors also measured an uncontrolled two-stroke motorcycle under several different conditions, with average emissions ranging from 3 to 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 to 31 g/kg; larger motorcycles had lower PM emissions (M. Spears, 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 to 90 g/kg depending on engine load and speed (with the highest value occurring at only 1 of 18 load/speed combinations). *Kojima and Khaliquzzaman* [2002] reported opacity measurements for 700 "baby taxis" (commercial three-wheeled vehicles) that could be interpreted as EF_{PM} 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 nonroad engines.

[102] 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. While we do not have observations to support this high level of emissions in other world regions we assume that these practices may also occur in South America and Africa. Uncertainty in both technology divisions and emission factors accounts for the possibility that emissions are lower. 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. On the basis of solvent extraction and sample color, *Sakai et al.* [1999] found that soluble organic fraction made up over 99% of the smoke. *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 OM.

[103] Similar to our earlier work [*Streets et al.*, 2003b], we estimated fractions of gasoline used in two-stroke and four-stroke engines by using numbers of cars, motorcycles and other vehicles given by *International Road Federation* (*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 (T. Chan, personal communication, 2001). IRF 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 includes 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

[104] *Yanowitz et al.* [2000] (hereinafter referred to as 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, as does a calculation using Western European values of mileage driven versus age.

[105] 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 given by *Faiz et al.* [1996] suggest that emissions are approximately uniformly distributed for ''normal'' vehicles, so the average would be about 3.7 g/kg. (Although the free-acceleration test is unlikely to represent driving conditions, more representative tests are not available for most regions.) Last, recent, independent estimates suggest a similar value for the baseline in Eastern Europe [*Klimont et al.*, 2002].

[106] 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 to 25 g/kg or greater [*Karim*, 1999]. (The emission factors in that study could be even higher because some vehicles saturated the smoke-meter.) We choose an emission factor of 12 g/kg for superemitters in all locations with a factor-of-2 uncertainty.

[107] 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. On the basis of those measurements and the preceding discussion, we extrapolate light-duty measurements to heavy-duty vehicles and choose a central value of 5% superemitters for countries ''similar'' to the United States, with a high uncertainty (1 to 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 fractions. 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.

[108] Our fleet average EF_{PM} for some sample regions is as follows: 2.3 g/kg for the United States, Western Europe, and Japan; 4.4 g/kg for Eastern Europe/former USSR; and 6.4 g/kg for Asia/Latin America. For the United States, the average is about equal to that for 1988–1990 vehicles in the *U.S. EPA*'s [1995] PART5 program, 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 emission factor in most locations

is lower than that used for ''underdeveloped'' countries in earlier inventories (15 g/kg in the study by *Cooke et al.* [1999]). Data from Kathmandu [*Shah and Nagpal*, 1996a] are one exception.

[109] As traffic tunnel measurements do not encompass all driving conditions, we summarize them here for comparison only: 1.4 g/kg (1992 United States, reviewed by *Cadle et al.* [1997]); 1.1g/kg (1993 Europe, *Weingartner et al.* [1997]); 2.5 g/kg (1997 United States, *Kirchstetter et al.* [1999]); 0.65 g/kg (1999 United States, *Gertler et al.* [2001]). With the exception of the Kirchstetter et al. 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 United States and Europe is lower than official European estimates, it is much higher than most of the tunnel results.

[110] As PM is difficult to measure, it would be desirable to find a more easily measured proxy to increase the size of the global data set. 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 [*Energy and Environmental Analysis, Inc. (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. Second, 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.

[111] *Fraser et al.* [2002] have shown that BC fractions can range from 0.19 to 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 yet available, we use the general measurements provided by *Gillies and Gertler* [2000]. From their data, we estimated BC and OC fractions as $66 \pm 16\%$ and OC as $21 \pm 6\%$, respectively. The different particulate emission mechanisms for high-emitting and normal vehicles might result in a varied chemical composition. However, while high-emitting vehicles often 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. Our fleet average is 3.6 g BC/kg fuel in countries with high superemitter fractions. Opacity measurements reported by *Shah and Nagpal* [1996a] in Kathmandu, which had a very high superemitter fraction, translate 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 2 in this area where superemitters are prevalent. This uncertainty is accounted for by the uncertainty in superemitter fraction.

5.4.4. Off-Road Vehicles

[112] Several studies have reported higher emission factors from off-road diesel vehicles, which are subject to both fewer regulations and higher fuel sulfur content than on-road vehicles. On the basis of IEA statistics, we tabulate the following categories of off-road usage separately: ships, railroads, agriculture, and industry. Industry usage of diesel includes construction, mining, and logging, but this disaggregation is not available for all countries. Further, fuel usage in each industry may include usage in both boilers and off-road engines, such as logging vehicles and process heat in the wood products industry. When countries do provide disaggregation into specific industries, off-road usage appears to account for about 75% of industrial diesel fuel use, a fraction that we assume for all countries. 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].

[113] Emission factors for ships differ between two EPA reports [*U.S. EPA*, 1991, 2000], and we use an intermediate value of $EF_{PM} = 1.8 \pm 1.5$ 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, rather than by power output, are not highly variable with power.) For diesel railroad locomotives, our value of $EF_{PM} = 2.7 \pm 2.1$ g/kg is also taken from an EPA report [*U.S. EPA*, 1997], and the high uncertainty accounts for the range of observed values in various operating modes.

[114] The Nonroad Engine and Vehicle Emission Study (NEVES) [*U.S. EPA*, 1991] gave in-depth estimates of the contribution of nonroad 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. *Bundesamt für Umwelt, Wald und Landschaft* (*BUWAL*) [1996] summarized the results of European studies ranging from 3.5 to 8.6 g/kg. *Samaras and Zierock* [1995] recommended emission factors in the range of 4.3 to 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 agriculture and construction as in on-road diesels.

[115] Finally, railroad locomotives also use coal. In the absence of measured data, we accept the assumption of *Marsh* [1947] that 3%, or 30 g/kg, is transformed to particulate matter, 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 5.

5.4.5. Aviation Fuel

[116] 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 [*Hopke*, 1985].

5.5. Other Fossil Fuels

[117] 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 5. 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. Environmental Protection Agency, SPECIATE 3.1, 1999), carbonaceous fractions for heavy fuel oil emissions are low (0.08 for BC and 0.03 for OC). For middle-distillate oil, *Hildemann et al.* [1991] and data from B. Wehner (manuscript in preparation, 2003) report higher fractions for BC, and we use $F_{BC} = 0.30$ and $F_{OC} = 0.09$.

[118] 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 (B. Wehner, manuscript in preparation, 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.

[119] 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, $EF_{PM} = 6 \pm 8$ 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.

[120] As the emission factors for residential boilers are much lower than those for generators, 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/yr, 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. To account for the uncertainty, the "low" emission case assumes 25% consumption in boilers in countries with low per capita usage, and the "high" emission case assumes 20% generator consumption where per capita usage is high.

[121] Both kerosene and liquefied petroleum gas (LPG) are used for cooking and lighting in the domestic sector. (Where these fuels are used in nonresidential sectors, we have combined them with other distillates.) The few measured emission factors for these fuels are given in Table 5. 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 combustion for heating and cooking has the same speciation as for kerosene.

[122] 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 (B. Wehner, manuscript in preparation, 2003). Reported emissions from residential combustion [Muhlbaier, 1981; Hildemann et al., 1991] are similar to those in the industrial sector.

5.6. Biofuels

[123] 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.

[124] 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.

[125] 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 airflow 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.

[126] Sampling problems contribute to uncertainty in all emission studies, but they are especially prevalent in wood-burning emissions, which are rich in semivolatile 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 the following: (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 separate from and additional to uncertainties in the method of measuring "elemental" or "organic" carbon once the sample has been taken.

5.6.1. Fireplaces and Heating Stoves

[127] 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

Table 11. Summary of Measurements on Speciation of Residential Wood-Burning Emissions

Reference	Comments	EC Fraction	OC Fraction
Fireplaces			
Dasch [1982][a]	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][b]	hot burning, hard/soft wood	0.15–0.29	0.13–0.28
Rau [1989][b]	cool burning, hard/soft wood	0.04–0.03	0.56–0.59
McDonald et al. [2000]	hardwood	0.08	0.60

[a]Used direct sampling (all others used dilution method).
[b]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.

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 (N. Senf, personal communication, 2003). 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.

[128] Emission factors for both fireplaces and heating stoves are listed in Table 6. 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 for EF_{PM}: 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.

[129] Measured fractions of "elemental" and organic carbon for fireplaces and heating stoves are summarized in Table 11. The values we chose lie in the middle of the range of 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 emission factors 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 may result from measurement methods, fuel loading procedures, or both. Only the mass emission factors from *Dasch* [1982], which reported a higher BC fraction, are similar to those in the *Houck and Tiegs* [1998] review. Therefore the speciation measurements may be inappropriate for the fireplaces in general, 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.

[130] 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 Gg 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.

5.6.2. Boilers

[131] Wood is consumed in larger stoker boilers for building heat. These boilers are common in Europe, but much less so in the United States; measurements of EF_{PM} are given in Table 6. Values of $F_{1.0}$ come from *Baumbach et al.* [1999]. We have not found measurements of F_{BC} and F_{OC}, so we use those from other wood combustion (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.

[132] Wood and other vegetal waste are also burned for process heat and power in industry. Table 5 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 on the

basis of estimates of use in the food processing industry (assumed to contain about 50% traditional ovens) versus the pulp and paper industry (assumed to be 100% stoker combustion). Not all nations provide these divisions, and information from individual countries was often used to develop regional profiles.

5.6.3. Cooking: Wood

[133] 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 to 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 (D. Still, personal communication, 2001).

[134] 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.

[135] If wood is scarce and its acquisition is time-consuming, it is advantageous to optimize the efficiency of combustion and heat transfer. (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.

[136] Total PM emissions from open cooking fires and cookstoves are lower than those from heating stoves fireplaces, as shown in Table 6. It is possible that differences in sampling methodologies explain 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: 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. The original source of data should be identified before assuming that high emission factors are applicable to cooking stoves.

[137] 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 earthen 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 between improved stoves; we obtain the results in Table 6 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.

[138] 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.

[139] No BC emission factors have yet been published for cookstoves, although at least one measurement project is in progress (C. Venkataraman, Indian Institute of Technology, Mumbai, India). We have made measurements in our laboratory of boiling water with carefully tended, small, dry wood (T. C. Bond, unpublished results, 2002). 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 11 for that reason, we estimate F_{BC} as 0.2. We also assume that OM makes up the remainder of the particulate matter.

[140] 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 cook-stoves 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

[141] EF_{PM} for agricultural waste is highly variable, as shown in Table 6. 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.

[142] 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 6 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 6. 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 because of lack of measurements.

5.6.5. Charcoal

[143] The charcoal fuel cycle emits particulate matter at two points: 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.

[144] 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 6. 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

[145] Table 5 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 equal and 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

[146] The selection of emission factors for large-scale, open 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 as follows: 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 many factors that are specific to location and season, such as fuel moisture content [e.g., *Hoffa et al.*, 1999]. We have not attempted to account for these changes.

[147] 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: emission factors from fossil fuels and biofuels, and the amounts of burned matter in open biomass burning. The discrepancy between the *Andreae and Merlet* [2001] 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

[148] 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 12 and 13. The contribution of fossil

Table 12. Black and Organic Carbon Emissions From Contained Combustion in 1996[a]

Region	Black Carbon			Organic Carbon		
	Central	(Low-High)	Previous96 (Ratio)[b]	Central	(Low-High)	Previous96 (Ratio)[b]
North America[c]	382	(278–618)	576 (1.5)	575	(325–1106)	962 (1.7)
Central/South America	360	(254–791)	730 (2)	893	(518–2022)	1609 (1.8)
Europe	466	(339–841)	1092 (2.3)	572	(319–1137)	2027 (3.5)
Former USSR	178	(135–524)	454 (2.6)	225	(147–509)	954 (4.2)
China	1365	(863–3395)	2432 (1.8)	2111	(1164–4283)	5432 (2.6)
India	483	(307–1035)	1213 (2.5)	1362	(799–2470)	4581 (3.4)
Other Asia	727	(528–1457)	1185 (1.6)	1339	(836–2424)	4304 (3.2)
Pacific	32	(23–52)	48 (1.5)	27	(18–50)	152 (5.6)
Africa	477	(297–1000)	1002 (2.1)	1494	(898–2662)	6258 (4.2)
Middle East	156	(107–372)	390 (2.5)	258	(116–755)	658 (2.6)
Total	4626	(3132–10,084)	9122 (2)	8856	(5141–17,419)	26,936 (3)

[a]Units are Gg/yr. "Contained" combustion includes fossil and biofuel combustion and excludes open burning.
[b]Previous96 applies emission factors from *Cooke et al.* [1999] to 1996 fuel-use data.
[c]North America includes Canada, the United States, and Mexico.

fuel combustion is 3.0 Tg for BC, with an uncertainty range of 2.0–7.4 Tg, and 2.4 Tg for primary OC (1.2–6.9 Tg). Compared with previously published estimates of 5.1 Tg and 10 Tg for 1984 data, these estimates are 40% and 75% lower. The reasons for these decreases will be discussed below. Central values and uncertainty ranges for biofuel emissions are 1.6 (0.7–4.4) Tg BC and 6.5 (3.1–13) Tg OC. For open burning, emissions are 3.3 (1.6–9.8) Tg BC and 25 (13–57) Tg OC.

[149] In discussing emission distributions, we separate emissions from "contained" combustion: that used to meet energy requirements, regardless of origin. Previous summaries [*Liousse et al.*, 1996; *IPCC*, 2001] have tabulated fossil fuel in one category, and biofuel and large-scale, open "biomass" combustion in another. We combine fossil fuel and biofuel emissions into "contained" combustion for three reasons. First, the 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]. "Open" combustion includes burning of urban and rural waste, which is small in comparison with burning of forests, fields and savannas.

[150] Figure 2 shows the global distribution of BC emissions from contained and open burning separately, and Figure 3 shows the same for primary OC emissions. 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 12 and 13, 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

[151] 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

Table 13. Emissions of Black and Organic Carbon From Open Burning[a]

Region	Black Carbon			Organic Carbon		
	Central	(Low-High)	Previous96 (Ratio)[b]	Central	(Low-High)	Previous96 (Ratio)[b]
North America	116	(49–494)	283 (2.4)	1473	(599–3745)	2126 (1.4)
Central/South America	910	(487–2581)	1812 (2)	6727	(3929–14,026)	13,074 (1.9)
Europe	59	(29–229)	132 (2.2)	691	(300–1705)	976 (1.4)
Former USSR	100	(44–416)	240 (2.4)	1245	(516–3139)	1787 (1.4)
China	124	(72–282)	160 (1.3)	713	(410–1483)	909 (1.3)
India	92	(49–224)	127 (1.4)	505	(260–1126)	755 (1.5)
Other Asia	275	(127–896)	546 (2)	1962	(1006–4453)	3970 (2)
Pacific	165	(75–469)	275 (1.7)	1132	(523–2720)	1786 (1.6)
Africa	1472	(675–4223)	2628 (1.8)	10,504	(5116–24,567)	17,862 (1.7)
Middle East	11	(6–29)	15 (1.3)	60	(31–134)	90 (1.5)
Total	3325	(1614–9842)	6217 (1.9)	25,013	(12,691–57,099)	43,334 (1.7)

[a]Units are Gg/yr.
[b]Previous96 applies emission factors from *Liousse et al.* [1996] to our estimates of dry matter burned.
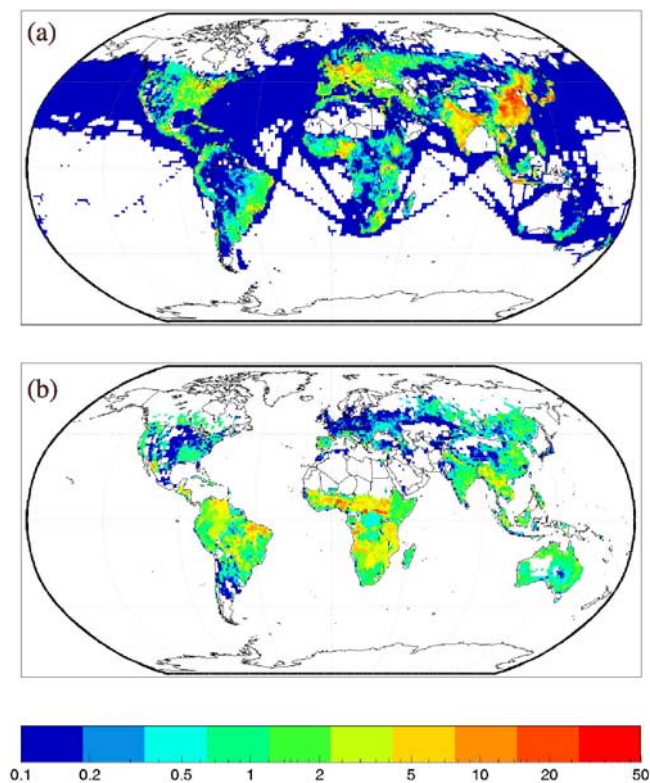
Figure 2. Emissions of black carbon aerosol. (a) "Contained" combustion, based on 1996 activity data. (b) Open burning, annual average. The color coding is an approximately logarithmic scale. Units are ng/m^2/s (1 ng/m^2/s $\sim$ 32 kg/km^2/yr).

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 burning is the largest contribution to both BC and OC emissions. Its components are summarized in Table 14. Savanna burning in Africa contributes the greatest amount to BC for open burning, and burning of forests and savannas in South America are also large contributors to the budget. Burning of wastes and crop residues contribute the least. 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 14.

[152] The remainder of global BC emissions comes from "contained" combustion, as does a large fraction of OC. (Again, biogenic emissions are not included in these totals.) Figure 4 summarizes the major contributors from contained combustion. Both the fractional contribution of contained combustion and the relative contribution of each region are indicated on the graph. 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 combustion 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 (coke making and brick kilns). On a global basis, transport, industry, and the residential sector have notable contributions to BC (20%, 10% and 25%, respectively), with open burning providing most of the rest.

[153] The sources of OC are somewhat different than 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 contributor 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

[154] For the most part, emission inventories developed by local and national air quality agencies focus on total particulate matter, usually PM$_{10}$. PM$_{10}$ includes a number of emissions other than 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
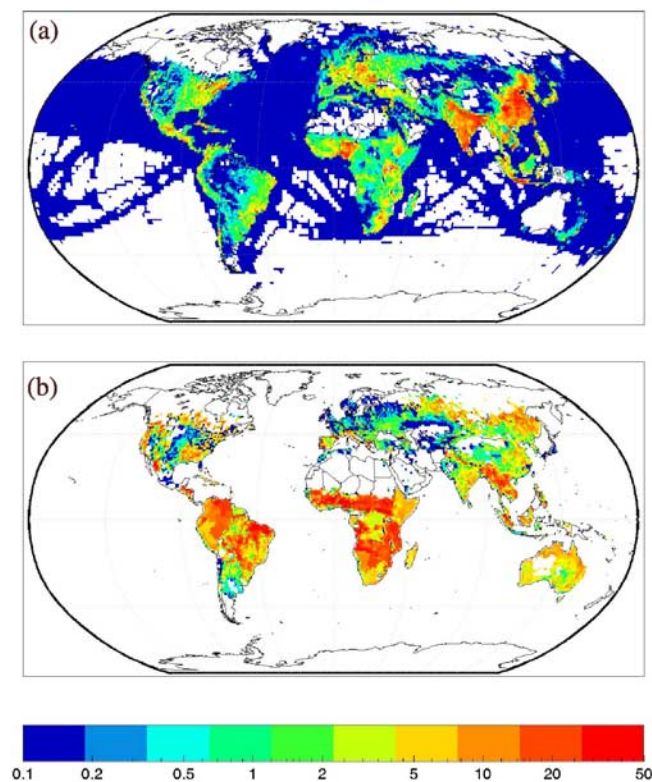

Figure 3. Emissions of primary organic carbon aerosol based on 1996 activity data. (a) "Contained" combustion, based on 1996 activity data. (b) Open burning, annual average. The color variation is approximately logarithmic and is the same as in Figure 2. Units are ng/m^2/s.

Table 14. Breakdown of Open-Burning Emissions[a]

	Black Carbon					Organic Carbon				
	Forest	Savanna	Agricultural Residue	Waste	Total	Forest	Savanna	Agricultural Residue	Waste	Total
North America	98	0	15	3	116	1393	0	73	7	1473
Central America/Mexico	53	4	2	0	59	415	28	11	0	454
South America	428	388	32	3	851	3369	2749	152	3	6273
Europe	43	1	11	4	59	620	6	54	11	691
Former USSR	81	0	16	3	100	1160	0	78	7	1245
Middle East	3	0	7	1	11	27	0	32	1	60
Pacific	14	130	21	0	165	113	920	99	0	1132
Africa	293	1122	51	7	1472	2309	7947	242	7	10,504
China	15	25	73	11	124	177	177	348	11	713
India	25	4	58	5	92	195	29	276	5	505
Other Asia	186	41	42	6	275	1462	292	202	6	1962
Total	1238	1715	328	44	3325	11,241	12,147	1567	58	25,013

[a]Units are Gg/yr.

are more similar to carbonaceous aerosols, except that they include mineral matter from combustion. A preliminary estimate of black carbon for the United States was commissioned by the U.S. EPA (W. Battye et al., unpublished report, 2002); 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 follows: fossil fuels, 100 Gg and 300 Gg; biofuel, 207 Gg and 629 Gg; open burning, 39 Gg and 313 Gg. Our totals are as follows: 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 emission factor 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.

[155] *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 are higher, primarily for wood; and our database includes animal waste in Vietnam. These additions compensate for our lower emission factor, and our emission estimates are about 1.5 times lower.

[156] 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 (1996) at the time of calculation. 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 the following: (1) the separation of biofuel emission factors for cooking and fireplaces, reducing our

estimate of cooking emissions; (2) the reanalysis of mobile source emission factors in regions where emission regulations were not implemented until the late 1990s or later, increasing our emission factors for both diesel and gasoline consumption; and (3) examination of small
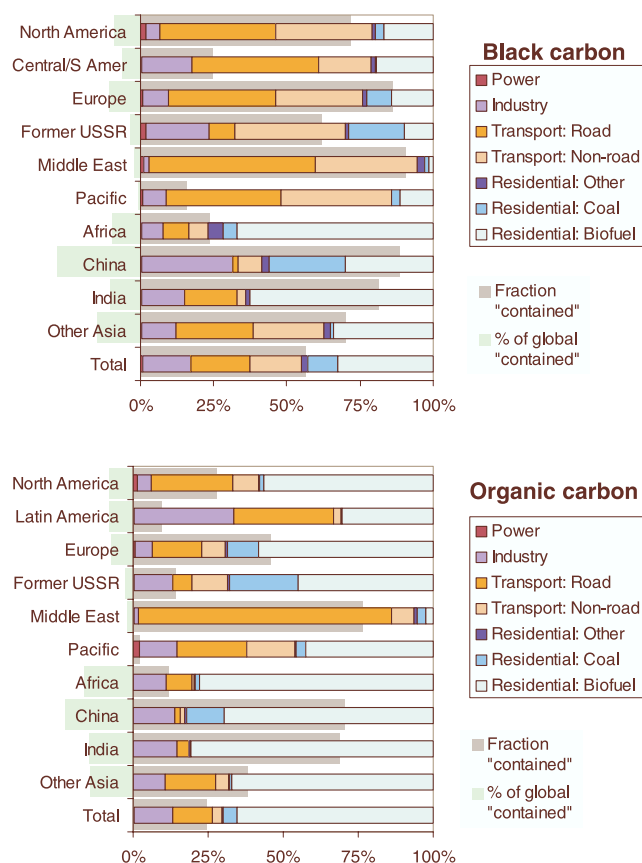

Figure 4. Sectoral contributions to emissions of black and organic carbon emissions. The gray bars behind the colored bars represent the fraction of emissions from "contained" combustion (that undertaken for energy use, excluding open burning) in each region. The green bars to the left indicate the relative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

Table 15. Major Differences in BC and OC Predicted by Current and Previous Emission Factors (in Order of Greatest Absolute Difference in BC Inventory)[a]

Fuel/Sector	Black Carbon			Organic Carbon		
	This Work	Previous96	Difference	This Work	Previous96	Difference
Open burning						
Open burning/forest	1238	2966	−1728	11,239	22,875	−11,636
Open burning/savanna	1715	2894	−1179	12,147	18,935	−6788
Open burning/crop resid	328	356	−28	1567	1519	48
Contained combustion						
Coal/power generation	7	1606	−1599	5	2359	−2354
Diesel fuel/on-road	792	2179	−1387	292	1278	−986
Wood/residential	880	1921	−1041	3506	11,537	−8031
Agricultural waste/residential	393	90	303	1492	1231	261
Animal waste/residential	208	417	−209	750	5465	−4715
Coal/industry[b]	642	1181	−539	450	1253	−803
Diesel fuel/residential	85	369	−284	28	185	−157
Coal/residential	480	761	−281	422	1716	−1294
Diesel fuel/off-road	579	691	−112	288	469	−181
Gasoline/transport	125	50	75	904	201	703
Other	478	524	−46	776	1242	−466
Total	7950	16,005	−8055	33,866	70,265	−36,399

[a]Units are Gg/yr based on 1996 fuel-use data (for contained combustion) or annual averages (for open burning).
[b]Includes coking, which accounts for 380 Tg BC, 270 Tg OC in this work; 320 Tg BC, 320 Tg OC in Previous96.

but polluting sources, such as coke making and brick kilns. Our emission estimates in the TRACE-P paper for China, India, and the rest of Asia were as follows: 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, because of increases in some emission factors estimated in this work and decreases in polluting technologies in the later year of the TRACE-P estimates (2000). The current OC emission estimates are lower than the TRACE-P estimates because of revisions of emission factors.

6.3.2. Global Inventories

[157] 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 the *Cooke et al.* [1999] inventory for fossil fuels and the *Liousse et al.* [1996] inventory 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] and the biomass/biofuel inventory of *Liousse et al.* [1996]. (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].) 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.

[158] Our second comparison, Previous96, is calculated by combining the emission factors published by *Cooke et al.* [1999] with the fuel-use data used for the current inventory (IEA 1996). The purpose of this estimate is to assess the effect of our revised emission factors without the confounding effect of changes in energy use. The *Cooke et al.* [1999] data selected emission factors according to one of three development levels, and there may be some disparities between our choices and those of the authors. However, if Previous84 and Previous96 estimates agree, the differences between our inventory and the others is usually due to emission factors; if they do not agree, the differences may be due to activity estimates.

[159] Tables 12 and 13 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; compared with the previous study, we estimate a slightly higher BC fraction, and therefore an aerosol that absorbs light more efficiently. Table 15 lists the fuels and uses with the largest changes between our inventory and Previous96, in order of their effect on the BC inventory. Two large changes result from the lower emission factors summarized by *Andreae and Merlet* [2001], which decrease estimates from forest and savanna burning.

[160] For contained combustion, three other fuel/sector combinations have large reductions: coal used in power generation, diesel fuel used in transport, and biofuel used in domestic applications. These uses 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 and off-road vehicles. Emission factors for residential biofuel have also been reduced because we have tabulated many studies with
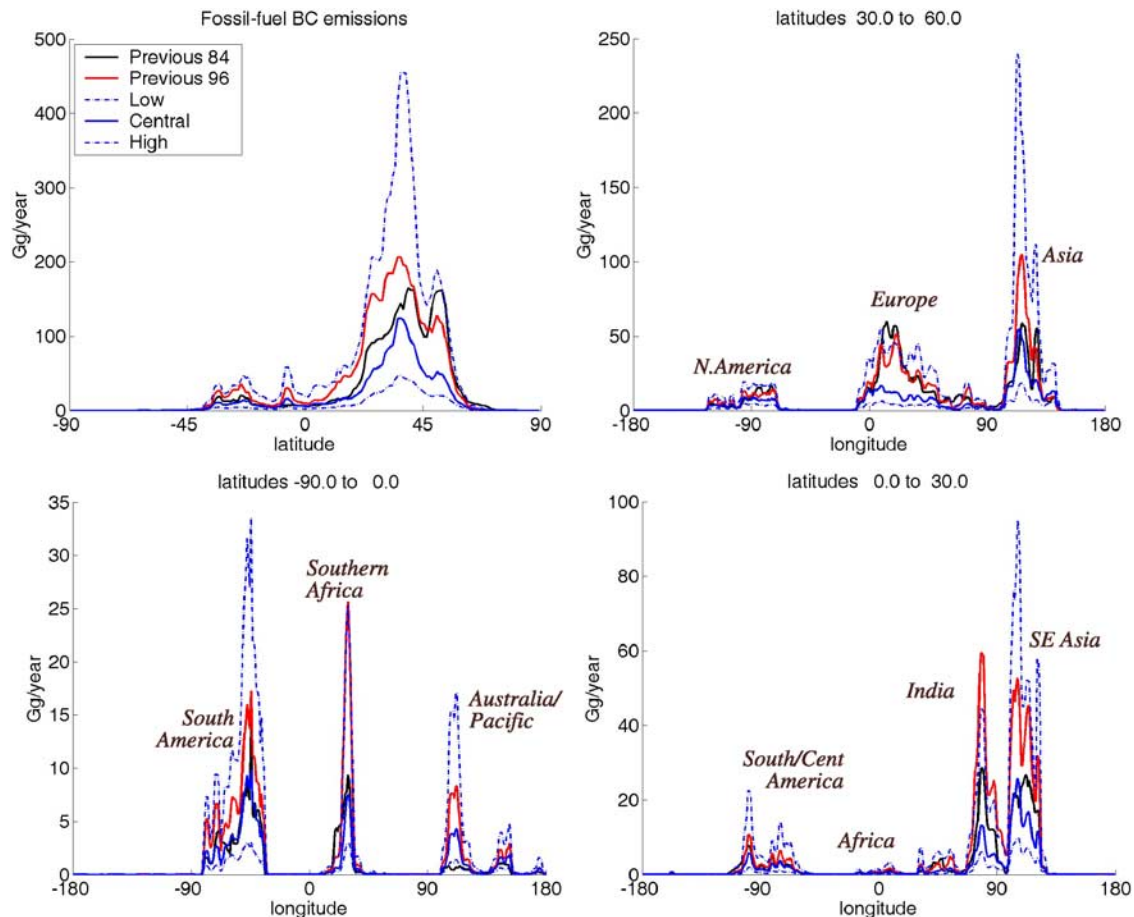
Figure 5. Regional differences among inventories of fossil fuel BC emissions. "Low," "Central," and "High" are from this work and are based on 1996 fuel-use data. Previous84 is the previous inventory developed by *Cooke et al.* [1999], which is based on 1984 fuel-use data, from the emission grids distributed to the community. Previous96 is our calculation of emissions combining fuel-use data from 1996 and the emission factors given by *Cooke et al.* [1999]. The plot 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.

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, although we use a higher black carbon fraction. Many of these studies were not available at the time the previous inventory was developed; the considerable change resulting from new information points to the importance of obtaining measurements on emission factors and characteristics for a variety of these small sources.

[161] 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.

[162] 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 estimate in each of these regions. Both Previous84 and Previous96 are higher than our estimate 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 because of 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 emission factor for diesel vehicles and coal-burning power plants.
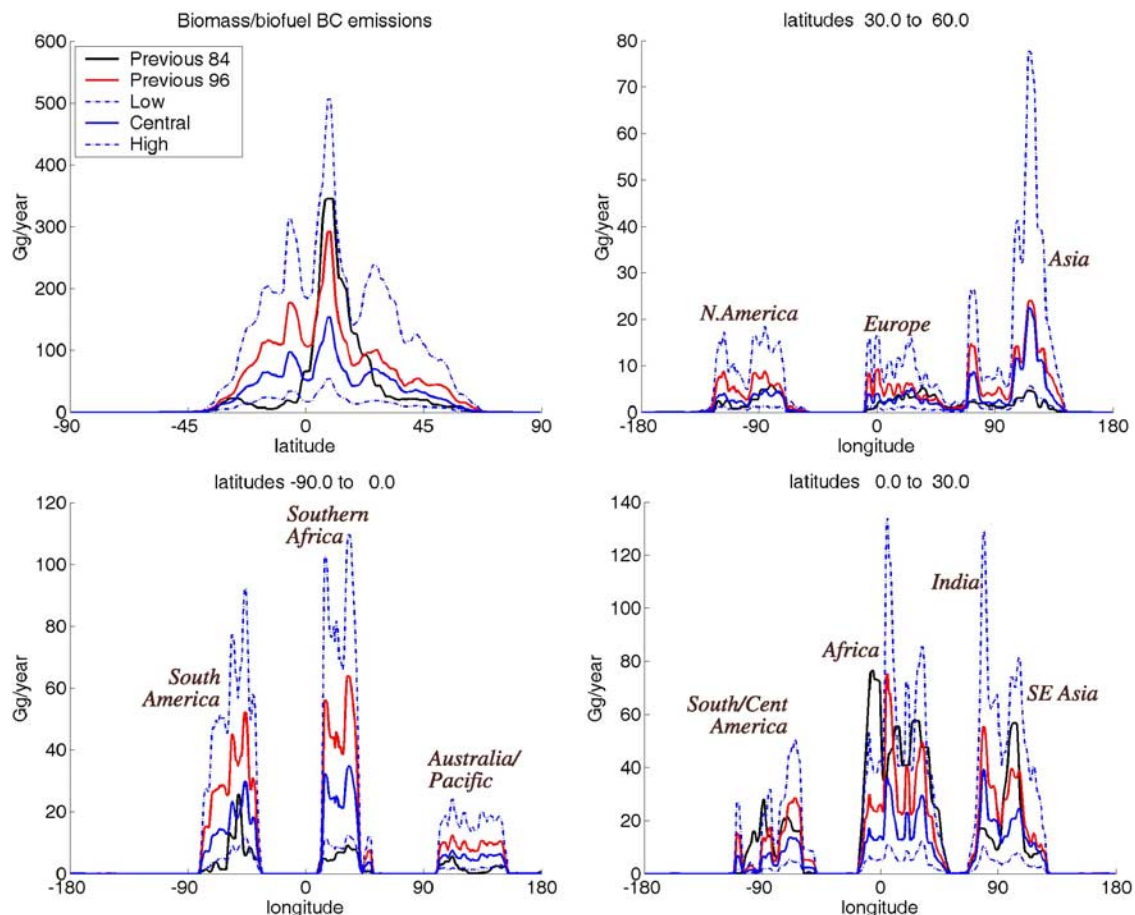
Figure 6. Regional differences among BC emission inventories of biofuel and open biomass. "Low," "Central," and "High" are from this work and are based on 1996 fuel-use data (for biofuel) and an average year (for biomass). Previous84 is the previous inventory developed by *Liousse et al.* [1996], from the emission grids distributed to the community. Previous96 applies the emission factors given by *Cooke et al.* [1999] to our activity data. The plot 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.

Many of the same observations would be found for OC emissions.

[163] 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 underestimate aerosol optical depth from 10° to 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 earlier 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. Our estimates of biofuel activity in India and China appear to be far lower than those in Previous84.

6.4. Uncertainties

[164] With the uncertainties that we considered, we provide 95% confidence intervals for total global emissions. For BC, these are 3.1 to 10.0 Tg/yr (−30% to +120%) for contained combustion, and 1.6 to 9.8 Tg/yr (−50% to +200%) for open biomass burning. For OC, the confidence intervals are as follows: contained combustion, 5.1 to 14 Tg/yr (−40% to +100%), and open burning, 13 to 58 Tg/yr (−50% to 130%). The asymmetric confidence intervals are a consequence of our lognormal treatment of emissions.

[165] Higher fuel usage and greater uncertainties about emission factors from particular types of burning cause regionally distinct uncertainties in emissions. Figure 5

Table 16. Largest Contributors to Variance in Global BC Emissions From Contained Combustion

Sector	Percent Variance	Contributors
Coke making	31%	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)
Agricultural waste/residential	8%	F_{BC}, fuel use, EF_{PM}
Diesel/off-road	5%	tech divisions, all characteristics (open fire and cookstove), fuel use
Coal/residential	4%	EF_{PM} (all techs), tech divisions
Animal 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} (two-strokes), EF_{PM} (two-strokes, superemitters)

shows that the magnitude of uncertainty in "contained" combustion 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.

[166] Open biomass burning contributes the most to uncertainties, because it comprises a large fraction of BC emissions and dominates OC emissions. The present study has estimated that "contained" combustion is a slightly greater contributor to BC emissions than open burning, 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, for which detailed reviews of global emissions are not available.

[167] Table 16 lists the fuel/usage combinations that contribute the most to variance in "contained" combustion. 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. The contribution to variance is not the same as the contribution to uncertainty; if the two largest contributors to variance were eliminated, the total variance would decrease by about 50%, but the uncertainty would decrease by only 30%.

[168] For BC, the largest variance by far is due to the coke making process, particularly in China where the fraction of "beehives" is not known. Although we estimated BC emissions from this source as 380 Gg/yr, the lack of information was such that upper emission estimates were over 7 times higher. As discussed previously, this contribution could have been a transient phenomenon in the mid-1990s and may be greatly reduced because of Chinese regulation of coking. The second greatest variance is due to residential wood combustion. While uncertainties in emission characteristics play a role, 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. As predicted by *Olivier and Peters* [2002], small sources have received less attention when emission data are tabulated.

[169] Table 17 repeats the analysis for organic carbon. Here, emissions from residential wood combustion dominate uncertainties, with activity estimates again leading the contribution to variance, and EF_{PM} for many types of burning also contributing to uncertainty. 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 coke making are also contributors.

[170] Of course, each world region is subject to different uncertainties, and Table 18 qualitatively indicates the largest contributors to variance in each region. The table first shows the contribution of open vegetative burning to the uncertainties in total emissions. That source dominates OC uncertainties in all regions. It contributes varying fractions to the variance of BC, ranging from 6% in Europe to 98% in Africa.

[171] The other lines of the table summarize the sources that cause moderate and large uncertainties in "contained"

Table 17. Largest Contributors to Variance in OC Emissions From Contained Combustion

Sector	Percent Variance	Contributors
Wood/residential	57%	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} (two-strokes), F_{OC} (two-strokes)
Agricultural waste/residential	9%	fuel use, F_{OC}, EF_{PM}
Wood/industrial	8%	EF_{PM} (traditional), tech divisions, F_{OC} (traditional)
Coke making	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
Animal waste/residential	2%	fuel use, EF_{PM}, F_{OC}
Diesel/on-road	1%	tech divisions, EF_{PM} and F_{OC} (superemitter), EF_{PM} (normal)

Table 18. Regional Contributors to Uncertainty in BC and OC Emissions[a]

Sector	North America BC	North America OC	South/Central America BC	South/Central America OC	Europe BC	Europe OC	Former USSR BC	Former USSR OC	Asia BC	Asia OC	Africa BC	Africa OC
Open burning	70%	95%	97%	99%	19%	81%	49%	98%	22%	79%	98%	100%
Contained combustion												
Agricultural waste/residential	–	–	–	–	–	–	–	–	1	1	2	–
Animal waste/residential	–	–	–	–	–	–	–	–	2	2	1	1
Coal/industrial	–	–	–	–	2	2	1	1	1	2	2	–
Coal/residential	–	–	–	–	2	2	1	1	1	2	2	–
Coal/coke making	2	–	1	2	1	2	1	1	1	2	2	–
Diesel/off-road	1	2	1	–	1	2	1	2	2	–	2	–
Diesel/on-road	1	2	1	2	1	2	2	–	2	–	2	–
Diesel/residential	2	–	–	–	1	2	2	–	–	–	2	–
Gasoline/transport	2	1	1	1	2	2	–	2	–	2	2	2
Wood/charcoal production	–	–	–	2	–	–	–	–	–	–	2	2
Wood/industrial	–	2	1	1	–	–	–	–	2	2	1	1
Wood/residential	1	1	1	1	1	1	2	1	1	1	1	1

[a]The first line of the table shows the fraction of total variance attributable to open burning. The other lines indicate combustion processes that are primary (1), secondary (2), or minimal (–) contributors to uncertainty in "contained" combustion. The classification of primary/secondary/minimal was accomplished by comparing the difference between high and central estimates from each process (here termed "uncertainty") with the central estimate of "contained" combustion emissions. Primary (1) indicates that the uncertainty is greater than 25% of the central estimate; secondary (2) indicates that uncertainty is 5–25%; and minimal (–) indicates that uncertainty is less than 10%.

combustion. The sources that contributed most to global uncertainties (coke making 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, coke making 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, and contribute a lesser amount to uncertainty in Asia and Africa. Gasoline used for transport in the Americas is also important.

6.4.1. Surprise

[172] 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 coke making together comprise about 8% of our BC inventory. Their potential importance was not identified by previous global inventories. 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.

[173] 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. The recommended increase in confidence intervals ranges from a factor of 4–7. 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.

6.4.2. Reducing Uncertainties

[174] For "contained" combustion, 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 source 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. There is also a large uncertainty in amounts, emissions and timing of open vegetative burning, which contributes greatly to the uncertainty in atmospheric concentrations in some locations. As the present work has focused on emissions from energy use, we discuss only those uncertainties here.

[175] Emission characteristics 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 technology divisions 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, contributing to understanding of gaseous emissions in addition to PM. However, the interpretation of these measurements needs refinement in order to attribute particulate matter to different vehicle types. Tabulations like those of *Polenske and McMichael* [2002] can assist in understanding industrial contributions.

[176] Despite the continuing advances in emission characterization just listed, the emission inventories contain fundamental uncertainties due to the use of fuels in informal sectors, including biofuel and coal. 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. The climate, air-quality, health effects, and energy communities must begin to share information, as demonstrated nearly a decade ago by the work of *Hall et al.* [1994] and the recent work of *Yevich and Logan* [2003].

[177] It is unlikely that "bottom-up" inventories alone can provide the required accuracy in estimating anthropogenic effects on global and regional climate, and a combination of emission inventories, atmospheric chemistry and transport modeling, and field observations is needed to help refine inventories. This technique has been particularly effective in the recent ACE-Asia and TRACE-P field campaigns in Asia. Field observations of individual species coupled with back-trajectory analysis [*Carmichael et al.*, 2003], long-term data sets combined with inverse modeling [*Kasibhatla et al.*, 2002], or satellite data sets combined with inverse modeling can provide insight into source strengths. Parallel work using recently developed techniques in source apportionment, such as organic tracers [e.g., *Schauer et al.*, 1996], will also be useful, especially if it can identify source profiles for different combustion types and the same fuels (e.g., biofuels versus open biomass; diesel superemitters versus normal engines). Results of these combined studies must be iterated with bottom-up inventories to provide reliable data that supports modeling efforts.

6.5. Measurements of Atmospheric Concentration

[178] 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.

[179] The relative amounts of BC and OC, along with other coemitted species such as sulfate precursors, may determine the sign of climate forcing. Concentration ratios of primary OC to BC ratios given by *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 during the INDOEX campaign 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, community-wide agreement on the measurement techniques that separate OC from BC, and methods of excluding

secondary OC from comparisons are needed before measured OC/BC ratios can be compared with inventory data.

[180] Using backtrajectory calculations combined with measurements at a coastal station downwind of Europe, *Derwent et al.* [2001] estimated BC emissions from the United Kingdom and Europe, respectively, as 46 ± 13 and 480 ± 140 Gg/yr during 1995–1998. This is in quite good agreement with our central estimates of 46 and 550 Gg/yr, even though the uncertainties in our 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.

[181] Several studies have compared results of models using previous inventories with atmospheric measurements. The results of earlier comparisons might suggest whether the revisions recommended here 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 midcontinental 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.

[182] *Chin et al.* [2002] used the fossil fuel inventory of *Cooke et al.* [1999] and a biomass inventory based on the work 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 in section 5.8. The high interannual variability of biomass burning should also be considered in such comparisons.

[183] *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. The needed increase in BC could also be provided by an increase in its absorption efficiency. 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.

[184] *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 to 1.0 Tg for the year 2000 from the TRACE-P data set [*Streets et al.*, 2003b]. From BC/CO correlations, *Dickerson et al.* [2002] speculated that the BC source strength on the South Asian continent may be 2–3 Tg/yr, significantly higher than 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. 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 of 80% given by *Novakov et al.* [2000].

[185] 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, not increased, the recommended BC emission factors because investigation showed that some of the previous estimates were unwarranted and usually too high. This alteration leaves even more atmospheric BC unexplained. Despite the possibility of larger measurement-model discrepancies resulting from use of this inventory, the reliance on measured, rather than assumed, emission factors is a step in a necessary direction. If current atmospheric concentrations can be simulated only when models rely on unsupportable emission data, then the ability to predict past and future emissions, and hence past and future impacts of atmospheric aerosols, is in serious doubt. Achieving a predictive capability for atmospheric concentrations will require iterative convergence of bottom-up inventories, modeling, and measurements.

7. Summary and Final Remarks

[186] Our intent in this work is 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 and other combustion practices. In detailing these regional practices, we attempted to strike a balance between accuracy and the feasibility of representing the existing variability. The emission factors we suggest here often contain high 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.

[187] 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 middle 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.

[188] We have constructed a representation of emission uncertainty, accounting for many potential contributors. For contained combustion, 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 2, 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.

[189] Except for investigations directly targeted at the largest uncertainties, and a rigorous explanation 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.

References

Ackerman, A. S., O. B. Toon, D. E. Stevens, A. J. Heymsfield, V. Ramanathan, and E. J. Welton (2000), Reduction of tropical cloudiness by soot, *Science*, *288*, 1042–1047.

Akeredolu, F., and A. O. Isichei (1996), Emissions of carbon, nitrogen, and sulfur from biomass burning in Nigeria, in *Biomass Burning and Global Change*, vol. 2, edited by J. S. Levine, pp. 162–166, MIT Press, Cambridge, Mass.

Amous, S. (1999), *The Role of Wood Energy in Africa*, Food and Agric. Organ., U. N., Rome.

Andreae, M. O., and P. Merlet (2001), Emission of trace gases and aerosols from biomass burning, *Global Biogeochem. Cycles*, *15*, 955–966.

Babcock and Wilcox Company (1992), *Steam: Its Generation and Use*, 40th ed., Barberton, Ohio.

Babu, S. S., and K. K. Moorthy (2001), Anthropogenic impact on aerosol black carbon mass concentration at a tropical coastal station: A case study, *Current Sci.*, *81*(9), 1208–1214.

Balla, M. K., S. Chaudhary, and T. B. Karkee (1991), Wood energy flows: RRA study in Pokhara, Nepal, *Field Doc. 26*, Reg. Wood Dev. Programme in Asia, Food and Agric. Organ., Bangkok.

Ballard-Tremeer, G., and H. H. Jawurek (1996), Comparison of five rural, wood-burning cooking devices: Efficiencies and emissions, *Biomass Bioenergy*, *11*(5), 419–430.

Barnard, G., and L. Kristoferson (1985), *Agricultural Residues as Fuel in the Third World*, Earthscan, London.

Barnes, D. F., K. Openshaw, K. R. Smith, and R. vanderPlas (1994), What makes people cook with improved biomass stoves?, *Tech. Rep. 242*, World Bank, Washington, D. C.

Baumbach, G., U. Zuberbühler, M. Struschka, D. Straub, and K. R. G. Hein (1999), *Feinstaubuntersuchungen an Holzfeuerungen*, Univ. Stuttgart, Stuttgart, Germany.

Beijing Environmental Protection Bureau (1996), *Study of Total Plan of Beijing Environment* (in Chinese), vol. 2, Beijing.

Bergin, M. H., et al. (2001), Aerosol radiative, physical and chemical properties in Beijing during June 1999, *J. Geophys. Res.*, *106*(D16), 17,969–17,980.

Bergstrom, R. W. (1973), Extinction and absorption coefficients of the atmospheric aerosol as a function of particle size, *Contrib. Atmos. Phys.*, *46*, 223–234.

Bhattacharya, S. C., P. A. Salam, and M. Sharma (2000), Emissions from biomass energy use in some selected Asian countries, *Energy*, *25*, 169–188.

Bishop, G. A., D. H. Stedman, M. Hektner, and J. D. Ray (1999), An in-use snowmobile emission survey in Yellowstone National Park, *Environ. Sci. Technol.*, *33*, 3924–3926.

Bishop, G. A., D. H. Stedman, R. B. Hutton, L. Bohren, and N. Lacey (2000), Drive-by motor vehicle emissions: Immediate feedback in reducing air pollution, *Environ. Sci. Technol.*, *34*, 1110–1116.

Bond, T. C. (2000), Light absorption by primary particles from fossil-fuel combustion: Implications for radiative forcing, Ph.D. dissertation, 383 pp., Univ. of Wash., Seattle.

Bond, T. C. (2001), Spectral dependence of visible light absorption by carbonaceous particles emitted from coal combustion, *Geophys. Res. Lett.*, *28*(21), 4075–4078.

Bond, T. C., R. J. Charlson, and J. Heintzenberg (1998), Quantifying the emission of light-absorbing particles: Measurements tailored to climate studies, *Geophys. Res. Lett.*, *25*(3), 337–340.

Bond, T. C., T. L. Anderson, and D. Campbell (1999a), Calibration and intercomparison of filter-based measurements of visible light absorption by aerosols, *Aerosol. Sci. Technol.*, *30*, 582–600.

Bond, T. C., M. Bussemer, B. Wehner, S. Keller, R. J. Charlson, and J. Heintzenberg (1999b), Light absorption by primary particle emissions from a lignite burning plant, *Environ. Sci. Technol.*, *33*, 3887–3891.

Bond, T. C., D. S. Covert, J. C. Kramlich, T. V. Larson, and R. J. Charlson (2002), Primary particle emissions from residential coal burning: Optical properties and size distributions, *J. Geophys. Res.*, *107*(D21), 8347, doi:10.1029/2001JD000571.

Bradley, K., D. H. Stedman, and G. A. Bishop (1999), A global inventory of carbon monoxide emissions from motor vehicles, *Chemosphere*, *1*, 65–72.

Brimblecombe, P., and C. Bowler (1992), The history of air pollution in York, England, *J. Air Waste Manage. Assoc.*, *42*, 1562–1566.

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

Bundesamt für Umwelt, Wald und Landschaft (BUWAL) (1996), Schadstoffemissionen und Treibstoffverbrauch des offroad-sektors, *Umweltmater. 49*, Bern.

Butcher, S. S., and M. J. Ellenbecker (1982), Particulate emission factors for small wood and coal stoves, *J. Air Pollut. Control Assoc.*, *32*, 380–384.

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

Cadle, S. H., P. J. Groblicki, and P. A. Mulawa (1983), Problems in the sampling and analysis of carbon particulate, *Atmos. Environ.*, *17*, 593–600.

Cadle, S. H., P. A. Mulawa, J. Ball, C. Donase, A. Weibel, J. C. Sagebiel, K. T. Knapp, and R. Snow (1997), Particulate emission rates from in-use high-emitting vehicles recruited in Orange County, California, *Environ. Sci. Technol.*, *31*, 3405–3412.

Cadle, S. H., R. A. Gorse, B. K. Bailey, and D. R. Lawson (2000), Real-world vehicle emissions: A summary of the ninth coordinating research council on-road vehicle emissions workshop, *J. Air Waste Manage. Assoc.*, *50*, 278–291.

Carmichael, G. R., et al. (2003), Regional-scale chemical transport modeling in support of intensive field experiments: Overview and analysis of the TRACE-P observations, *J. Geophys. Res.*, *108*(D21), 8823, doi:10.1029/2002JD003117.

Chameides, W. L., et al. (1999), Case study of the effects of atmospheric aerosols and regional haze on agriculture: An opportunity to enhance crop yields in China through emission controls?, *Proc. Natl. Acad. Sci. U. S. A.*, *96*(24), 13,626–13,633.

Charlson, R. J., and M. J. Pilat (1969), Climate: The influence of aerosols, *J. Appl. Meteorol.*, *8*(6), 1001–1002.

Charlson, R. J., J. Langner, H. Rodhe, C. B. Leovy, and S. G. Warren (1991), Perturbation of the Northern Hemisphere radiative balance by backscattering from anthropogenic sulfate aerosols, *Tellus, Ser. B*, *43*, 152–163.

Chase, R. E., G. J. Duszkiewicz, T. E. Jensen, D. Lewis, E. J. Schlaps, A. T. Weibel, S. Cadle, and P. Mulawa (2000), Particle mass emission rates from current-technology, light-duty gasoline vehicles, *J. Air Waste Manage. Assoc.*, *50*, 930–935.

Chen, M.-L., I.-F. Mao, M.-T. Wu, J.-R. Chen, C.-K. Ho, T. J. Smith, D. Wypij, and D. C. Christian (1999), Assessment of coke oven emissions exposure among coking workers, *Am. Ind. Hyg. J.*, *60*(1), 105–110.

Cheng, Y.-S., Y. Zhou, J. Chow, J. Watson, and C. Frazier (2001), Chemical composition of aerosols from kerosene heaters burning jet fuels, *Aerosol Sci. Technol.*, *35*, 949–957.

Chin, M., P. Ginoux, S. Kinne, O. Torres, B. N. Holben, B. N. Duncan, R. V. Martin, J. A. Logan, A. Higurashi, and T. Nakajima (2002), Tropospheric optical thickness from the GOCART model and comparisons with satellite and Sun photometer measurements, *J. Atmos. Sci.*, *59*, 461–483.

Chow, J. C., J. G. Watson, D. Crow, D. H. Lowenthal, and T. Merrifield (2001), Comparison of IMPROVE and NIOSH carbon measurements, *Aerosol Sci. Technol.*, *34*, 23–34.

Chowdhury, Z., L. S. Hughes, L. G. Salmon, and G. R. Cass (2001), Atmospheric particle size and composition measurements to support light extinction calculations over the Indian Ocean, *J. Geophys. Res.*, *106*(D22), 28,597–28,605.

Chung, S. H., and J. H. Seinfeld (2002), Global distribution and climate forcing of carbonaceous aerosols, *J. Geophys. Res.*, *107*(D19), 4407, doi:10.1029/2001JD001397.

Chylek, P., G. Videen, D. Ngo, R. G. Pinnick, and J. D. Klett (1995), Effect of black carbon on the optical properties and climate forcing of sulfate aerosols, *J. Geophys. Res.*, *100*(D8), 16,325–16,332.

Clark, N. N., J. J. Daley, R. D. Nine, and C. M. Atkinson (1999a), Application of the New City-Suburban Heavy Vehicle Route (CSHVR) to truck emissions characterization, *SAE Pap. 1999-01-1467*, Soc. of Automotive Eng., Warrendale, Pa.

Clark, N. N., M. Gautam, B. L. Rapp, D. W. Lyons, M. S. Graboski, R. L. McCormick, T. L. Alleman, and P. Norton (1999b), Diesel and CNG transit bus emissions characterization by two chassis dynamometer laboratories: Results and issues, *SAE Pap. 1999-01-1469*, Soc. of Automotive Eng., Warrendale, Pa.

Clark, N. N., R. P. Jarrett, and C. M. Atkinson (1999c), Field measurements of particulate matter emissions, carbon monoxide, and exhaust opacity from heavy-duty diesel vehicles, *J. Air Waste Manage. Assoc.*, *49*, 76–84.

Clark, N. N., J. C. Prucz, M. Gautam, and D. W. Lyons (2000), The West Virginia heavy duty vehicle emissions database as a resource for inventory and comparative studies, *SAE Pap. 2000-01-2854*, Soc. of Automotive Eng., Warrendale, Pa.

Collins, W. D., P. J. Rasch, B. E. Eaton, B. V. Khattatov, J. F. Lamarque, and C. S. Zender (2001), Simulating aerosols using a chemical transport model with assimilation of satellite aerosol retrievals: Methodology for INDOEX, *J. Geophys. Res.*, *106*(D7), 7313–7336.

Cooke, W. F., and J. J. N. Wilson (1996), A global black carbon aerosol model, *J. Geophys. Res.*, *101*(D14), 19,395–19,409.

Cooke, W. F., C. Liousse, H. Cachier, and J. Feichter (1999), Construction of a 1° × 1° fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model, *J. Geophys. Res.*, *104*(D18), 2137–2162.

Cooke, W. F., V. Ramaswamy, and P. Kasibhatla (2002), A general circulation model study of the global carbonaceous aerosol distribution, *J. Geophys. Res.*, *107*(D16), 4279, doi:10.1029/2001JD001274.

Corbett, J. J., P. S. Fischbeck, and S. N. Pandis (1999), Global nitrogen and sulfur inventories for oceangoing ships, *J. Geophys. Res.*, *104*(D3), 3457–3470.

Craig, C. C. (1936), On the frequency function of xy, *Ann. Math. Stat.*, *7*(1), 1–15.

Cullen, A. C., and H. C. Frey (1999), *Probabilistic Techniques in Exposure Assessment*, Plenum, New York.

Dasch, J. M. (1982), Particulate and gaseous emissions from wood-burning fireplaces, *Environ. Sci. Technol.*, *16*, 639–644.

Davidson, C. I. (1979), Air pollution in Pittsburgh: A historical perspective, *J. Air Pollut. Control Assoc.*, *29*, 1035–1041.

Delmas, R. A., P. Loudjani, A. Podaire, and J. C. Menaut (1996), Biomass burning in Africa: An assessment of annually burned biomass, in *Biomass Burning and Global Change*, vol. 2, edited by J. S. Levine, pp. 126–132, MIT Press, Cambridge, Mass.

deLucia, R. (1983), Defining the scope of wood fuel surveys, in *Wood Fuel Surveys*, pp. 5–28, Food and Agric. Organ., U. N., Rome.

Derwent, R. G., D. B. Ryall, S. G. Jennings, T. G. Spain, and P. G. Simmonds (2001), Black carbon aerosol and carbon monoxide in European regionally polluted air masses at Mace Head, Ireland during 1995–1998, *Atmos. Environ.*, 35, 6371–6378.

Dickerson, R. R., M. O. Andreae, T. Campos, O. L. Mayol-Bracero, C. Neusuess, and D. G. Streets (2002), Analysis of black carbon and carbon monoxide observed over the Indian Ocean: Implications for emissions and photochemistry, *J. Geophys. Res.*, 107(D19), 8017, doi:10.1029/2001JD000501.

Dillner, A. M., C. Stein, S. M. Larson, and R. Hitzenberger (2001), Measuring the mass extinction efficiency of elemental carbon in rural aerosol, *Aerosol Sci. Technol.*, 35, 1009–1021.

Dobbins, R. A., R. A. Fletcher, and W. Lu (1995), Laser microprobe analysis of soot precursor particles and carbonaceous soot, *Combust. Flame*, 100, 301–309.

Dockery, D. W., C. A. Pope, X. P. Xu, J. D. Spengler, J. H. Ware, M. E. Fay, B. G. Ferris, and F. E. Speizer (1993), An association between air pollution and mortality in six United States cities, *N. Engl. J. Medicine*, 329(24), 1753–1759.

Duncan, B. N., R. V. Martin, A. C. Staudt, R. Yevich, and J. A. Logan (2003), Interannual and seasonal variability of biomass burning emissions constrained by satellite observations, *J. Geophys. Res.*, 108(D2), 4100, doi:10.1029/2002JD002378.

Durbin, T. D., J. M. Norbeck, M. R. Smith, and T. J. Truex (1999a), Particulate emission rates from light-duty vehicles in the South Coast Air Quality Management District, *Environ. Sci. Technol.*, 33, 4401–4406.

Durbin, T. D., M. R. Smith, J. M. Norbeck, and T. J. Truex (1999b), Population density, particulate emission characterization, and impact on the particulate inventory of smoking vehicles in the South Coast Air Quality Management District, *J. Air Waste Manage. Assoc.*, 49, 28–38.

Durbin, T. D., J. M. Nobeck, R. D. Wilson, and H. A. Galdamez (2000), Effect of payload on exhaust emissions from light heavy-duty diesel and gasoline trucks, *Environ. Sci. Technol.*, 34, 4708–4713.

Energy and Environmental Analysis, Inc. (EEA, Inc.) (2000), Documentation and analysis of heavy-duty diesel emission test data, report to New York Department of Environmental Conservation, Arlington, Va.

European Environment Agency (EEA) (2000a), EMEP/CORINAIR emissions inventory guidebook, *Tech. Rep. 30*, Copenhagen.

European Environment Agency (EEA) (2000b), COPERT III: Computer Programme to Calculate Emissions from Road Transport, *Tech. Rep. 49*, Copenhagen.

Ezzati, M., A. D. Lopez, S. VanderHoorn, and C. J. L. Murray (2002), Selected major risk factors and global and regional burden of disease, *Lancet*, 360(9343), 1347–1360.

Faiz, A., C. S. Weaver, and M. P. Walsh (1996), *Air Pollution From Motor Vehicles: Standards and Technologies for Controlling Emissions*, World Bank, Washington, D. C.

Fearnside, P. M. (1996), Greenhouse gas contributions from deforestation in Brazilian Amazonia, in *Biomass Burning and Global Change*, vol. 2, edited by J. S. Levine, pp. 92–105, MIT Press, Cambridge, Mass.

Fine, P. M., G. R. Cass, and B. R. T. Simoneit (2001), Chemical characterization of fine particle emissions from fireplace combustion of woods grown in the northeastern United States, *Environ. Sci. Technol.*, 35, 2665–2675.

Fine, P. M., G. R. Cass, and B. R. T. Simoneit (2002), Chemical characterization of fine particle emissions from fireplace combustion of woods grown in the southern United States, *Environ. Sci. Technol.*, 36, 1442–1451.

Fisher, G. L., B. A. Prentice, D. Silberman, J. M. Ondov, A. H. Biermann, R. C. Ragaini, and A. R. McFarland (1978), Physical and morphological studies of size-classified coal fly ash, *Environ. Sci. Technol.*, 12, 447–451.

Flagan, R. C., and S. K. Friedlander (1978), Particle formation in pulverized coal combustion: A review, in *Recent Developments in Aerosol Science*, edited by D. T. Shaw, pp. 25–59, John Wiley, Hoboken, N. J.

Food and Agriculture Organization of the United Nations (FAO) (1997), *State of the World's Forests 1997*, Rome.

Food and Agriculture Organization of the United Nations (FAO) (2000), *ETAP Reference Guidebook*, Rome.

Food and Agriculture Organization of the United Nations–Regional Wood Energy Development Programme in Asia (FAO-RWEDP) (1993a), Status and development issues of the brick industry in Asia, *Field Doc. 35*, Bangkok.

Food and Agriculture Organization of the United Nations–Regional Wood Energy Development Programme in Asia (FAO-RWEDP) (1993b), Improved solid biomass burning cookstoves, *Field Doc. 44*, Bangkok, Sept.

Food and Agriculture Organization of the United Nations–Regional Wood Energy Development Programme in Asia (FAO-RWEDP) (1993c), Indian improved cookstoves: A compendium, *Field Doc. 41*, Bangkok, July.

Food and Agriculture Organization of the United Nations–Regional Wood Energy Development Programme in Asia (FAO-RWEDP) (1996), Wood-fuel flows, *RWEDP Rep. 30*, Bangkok.

Food and Agriculture Organization of the United Nations–Regional Wood Energy Development Programme in Asia (FAO-RWEDP) (1997), Regional study on wood energy today and tomorrow in Asia, *Field Doc. 50*, Bangkok, Oct.

Food and Agriculture Organization of the United Nations–Regional Wood Energy Development Programme in Asia (FAO-RWEDP) (1998), Images of wood and biomass energy in industries in Thailand, *Field Doc. 52*, Bangkok, Aug.

Fraser, M. P., K. Lakshmanan, S. G. Fritz, and B. Ubanwa (2002), Variation in composition of fine particulate emissions from heavy-duty diesel vehicles, *J. Geophys. Res.*, 107(D21), 8346, doi:10.1029/2001JD000558.

Frenklach, M., D. W. Clary, J. William, C. Gardiner, and S. E. Stein (1984), Detailed kinetic modeling of soot formation in shock-tube pyrolysis of acetylene, in *Twentieth Symposium (International) on Combustion*, pp. 887–901, Combust. Inst., Pittsburgh, Pa.

Fridley, D., and J. Sinton (2001), China energy databook, version 5.0, Lawrence Berkeley Lab. and Energy Res. Inst., Berkeley, Calif.

Galanter, M., H. Levy, and G. R. Carmichael (2000), Impacts of biomass burning on tropospheric CO, NOx, and O$_3$, *J. Geophys. Res.*, 105, 6633–6653.

Ge, S., Z. Bai, W. Liu, T. Zhu, T. Wang, S. Qing, and J. Zhang (2001), Boiler briquette coal versus raw coal: Part I. Stack gas emissions, *J. Air Waste Manage. Assoc.*, 51, 524–533.

Gertler, A. W., M. Abu-Allaban, W. Coulombe, J. A. Gillies, W. R. Pierson, C. F. Rogers, J. C. Sagabiel, L. Tarnay, and T. A. Cahill (2001), Measurements of mobile source particulate emissions in a highway tunnel, *Int. J. Vehicle Design*, 27(1–4), 86–93.

Gill, J. (1987), Improved stoves in developing countries: A critique, *Energy Policy*, 15, 135–144.

Gillies, J. A., and A. W. Gertler (2000), Comparison and evaluation of chemically speciated mobile source PM2.5 particulate matter profiles, *J. Air Waste Manage. Assoc.*, 50, 1459–1480.

Glassman, I., O. Nishida, and G. Sidebotham (1994), Critical temperatures of soot formation, in *Soot Formation in Combustion: Mechanisms and Models*, edited by H. Bockhorn, pp. 316–324, Springer-Verlag, New York.

Goldstein, H. L., and C. W. Siegmund (1976), Influence of heavy fuel oil composition and boiler combustion conditions on particulate emissions, *Environ. Sci. Technol.*, 10, 1109–1114.

Gundel, L. A., R. L. Dod, H. Rosen, and T. Novakov (1984), The relationship between optical attenuation and black carbon concentration for ambient and source particles, *Sci. Total Environ.*, 26, 197–202.

Hall, D. O., F. Rosillocalle, and J. Woods (1994), Biomass utilization in households and industry: Energy use and development, *Chemosphere*, 29, 1099–1119.

Hangebrauck, R. P., D. J. van Lehmden, and J. E. Meeker (1964), Emissions of polynuclear hydrocarbons and other pollutants from heat-generation and incineration processes, *J. Air Pollut. Control Assoc.*, 14, 267–278.

Hansen, A. D. A., and H. Rosen (1990), Individual measurements of the emission factor of aerosol black carbon in automobile plumes, *J. Air Waste Manage. Assoc.*, 40, 1654–1657.

Hansen, A. D. A., H. Rosen, and T. Novakov (1984), The aethalometer: An instrument for the real-time measurement of optical absorption by aerosol particles, *Sci. Total Environ.*, 36, 191–196.

Hansen, J. E., M. Sato, R. Ruedy, A. Lacis, and V. Oinas (2000), Global warming in the twenty-first century: An alternative scenario, *Proc. Natl. Acad. Sci. U. S. A.*, 97(18), 9875–9880.

Hao, W. M., and M. Liu (1994), Spatial and temporal distribution of biomass burning, *Global Biogeochem. Cycles*, 8, 495–503.

Hart, J. R. (2001), Transient puffs of trace organic emissions from a batch-fed waste propellant incinerator, *Chemosphere*, 42, 559–569.

Haynes, B. S., and H. G. Wagner (1981), Soot formation, *Prog. Energy Combust. Sci.*, 7, 229–273.

Haywood, J. M., and V. Ramaswamy (1998), Global sensitivity studies of the direct radiative forcing due to anthropogenic sulfate and black carbon aerosols, *J. Geophys. Res.*, 103(D6), 6043–6058.

Haywood, J. M., and K. P. Shine (1995), The effect of anthropogenic sulfate and soot aerosol on the clear sky planetary radiation budget, *Geophys. Res. Lett.*, 22(5), 603–606.

Heintzenberg, J. (1982), Size-segregated measurements of particulate elemental carbon and aerosol light absorption at remote arctic locations, *Atmos. Environ.*, 16, 2461–2469.

Hildemann, L. M., G. R. Cass, and G. R. Markowski (1989), A dilution stack sampler for collection of organic aerosol emissions: Design, characterization and field-tests, *Aerosol Sci. Technol.*, 10, 193–204.

Hildemann, L. M., G. R. Markowski, and G. R. Cass (1991), Chemical composition of emissions from urban sources of fine organic aerosol, *Environ. Sci. Technol.*, 25, 744–759.

Hitzenberger, R., U. Dusek, and A. Berner (1996), Black carbon measurements using an integrating sphere, *J. Geophys. Res.*, 101(D14), 19,601–19,606.

Hoffa, E. A., D. E. Ward, W. M. Hao, R. A. Susott, and R. H. Wakimoto (1999), Seasonality of carbon emissions from biomass burning in a Zambian savanna, *J. Geophys. Res.*, 104(D11), 13,841–13,853.

Hoornweg, D., and L. Thomas (1999), *What a Waste: Solid Waste Management in Asia*, World Bank, Washington, D. C.

Hopke, P. K. (1985), *Receptor Modeling in Environmental Chemistry*, appendix, pp. 267–314, John Wiley, Hoboken, N. J.

Horvath, H. (1993), Comparison of measurements of aerosol optical absorption by filter collection and a transmissometric method, *Atmos. Environ., Part A*, 27, 319–325.

Houck, J. E., and P. E. Tiegs (1998), Residential wood combustion technology review, *Rep. EPA-600/R-98-174a*, U.S. Environ. Prot. Agency, Washington, D. C.

Hughes, T. W., and D. G. DeAngelis (1982), Emissions from coal-fired residential combustion equipment, in *1981 International Conference on Residential Solid Fuels*, pp. 333–348, Oreg. Grad. Cent., Portland.

Huntzicker, J. J., R. L. Johnson, J. J. Shah, and R. A. Cary (1982), Analysis of organic and elemental carbon in ambient aerosols by a thermal-optical method, in *Particulate Carbon: Atmospheric Life Cycle*, edited by G. T. Wolff and R. L. Klimisch, pp. 79–88, Plenum, New York.

Intergovernmental Panel on Climate Change (IPCC) (1996), *Climate Change 1995: The Science of Climate Change*, edited by J. T. Houghton et al., chap. 2, 572 pp., Cambridge Univ. Press, New York.

Intergovernmental Panel on Climate Change (IPCC) (2000), Good practice guidance and uncertainty management in national greenhouse gas inventories, Tech. Support Unit, Natl. Greenhouse Gas Invent. Programme, Hayama, Japan.

Intergovernmental Panel on Climate Change (IPCC) (2001), *Climate Change 2001: The Scientific Basis*, edited by J. T. Houghton et al., 880 pp., Cambridge Univ. Press, New York.

International Energy Agency (IEA) (1998a), *Energy Statistics of OECD Countries*, Paris.

International Energy Agency (IEA) (1998b), *Energy Statistics of Non-OECD Countries*, Paris.

International Road Federation (IRF) (2000), *World Road Statistics 2000*, Geneva.

Jaasma, D. R., and D. W. Macumber (1982), Measurement techniques and emission factors for hand-fired coal stoves, in *Residential Wood and Coal Combustion*, pp. 129–150, Air Pollut. Control Assoc., Louisville, Ky.

Jacobson, M. Z. (1999), Isolating nitrated and aromatic aerosols and nitrated aromatic gases as sources of ultraviolet light absorption, *J. Geophys. Res.*, 104(D3), 3527–3542.

Jacobson, M. Z. (2001), Global direct radiative forcing due to multicomponent anthropogenic and natural aerosols, *J. Geophys. Res.*, 106(D2), 1551–1568.

Jacobson, M. Z. (2002), Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming, *J. Geophys. Res.*, 107(D19), 4410, doi:10.1029/2001JD001376.

Johnston, T. (2001), Canada report 2001, Can. Interagency For. Fire Cent., Winnipeg, Manit., Canada.

Joshi, V., C. Venkataraman, and D. R. Ahuja (1989), Emissions from burning biofuels in metal cookstoves, *Environ. Manage. N. Y.*, 13(6), 763–772.

Karim, M. M. (1999), Traffic pollution inventories and modeling in metropolitan Dhaka, Bangladesh, *Transp. Res., Part D: Transp. Environ.*, 4, 291–312.

Kasibhatla, P., A. Arellano, J. A. Logan, P. I. Palmer, and P. Novelli (2002), Top-down estimate of a large source of atmospheric carbon monoxide associated with fuel combustion in Asia, *Geophys. Res. Lett.*, 29(19), 1900, doi:10.1029/2002GL015581.

Kaufman, Y. J., et al. (1998), Smoke, Clouds, and Radiation–Brazil (SCAR-B) experiment, *J. Geophys. Res.*, 103(D24), 31,783–31,808.

Kean, A. J., R. F. Sawyer, and R. A. Harley (2000), A fuel-based assessment of off-road diesel engine emissions, *J. Air Waste Manage. Assoc.*, 50, 1929–1939.

Kiehl, J. T., and B. P. Briegleb (1993), The relative roles of sulfate aerosols and greenhouse gases in climate forcing, *Science*, 260, 311–314.

Kirchstetter, T. W., R. A. Harley, N. M. Kreisberg, M. R. Stolzenburg, and S. V. Hering (1999), On-road measurement of fine particle and nitrogen oxide emissions from light- and heavy-duty motor vehicles, *Atmos. Environ.*, 33, 2955–2968.

Kirchstetter, T. W., C. E. Corrigan, and T. Novakov (2001), Laboratory and field investigation of the adsorption of gaseous organic compounds onto quartz filters, *Atmos. Environ.*, 35, 1663–1671.

Kituyi, E., and C. Kirubi (2003), Influence of diet patterns on fuelwood consumption in Kenyan boarding schools and implications for data and energy policies, *Energy Convers. Manage.*, 44, 1099–1109.

Kituyi, E., L. Marufu, S. O. Wandiga, I. O. Jumba, M. O. Andreae, and G. Helas (2001), Carbon monoxide and nitric oxide from biofuel fires in Kenya, *Energy Convers. Manage.*, 42, 1517–1542.

Klimont, Z., J. Cofala, I. Bertok, M. Amann, C. Heyes, and F. Gyarfas (2002), Modelling particulate reductions in Europe: A framework to estimate reduction potential and control costs, *Interim Rep. IR-02-076*, Int. Inst. for Appl. Syst. Anal., Laxenburg, Austria.

Koch, D. (2001), Transport and direct radiative forcing of carbonaceous and sulfate aerosols in the GISS GCM, *J. Geophys. Res.*, 106(D17), 20,311–20,332.

Kohler, I., M. Dameris, I. Ackermann, and H. Hass (2001), Contribution of road traffic emissions to the atmospheric black carbon burden in the mid-1990s, *J. Geophys. Res.*, 106(D16), 17,997–18,014.

Kojima, M., and M. Khaliquzzaman (2002), Bangladesh: Reducing emissions from baby-taxis in Dhaka, Joint UNDP/World Bank Energy Sector Manage. Assist. Programme, Washington, D. C.

Kojima, M., C. Brandon, and J. Shah (2000), *Improving Urban Air Quality in South Asia by Reducing Emissions From Two-Stroke Engine Vehicles*, World Bank, Washington, D. C.

Lary, D. J., D. E. Shallcross, and R. Toumi (1999), Carbonaceous aerosols and their potential role in atmospheric chemistry, *J. Geophys. Res.*, 104(D7), 15,929–15,940.

Lavoué, D., C. Liousse, H. Cachier, B. J. Stocks, and J. G. Goldammer (2000), Modeling of carbonaceous particles emitted by boreal and temperate wildfires at northern latitudes, *J. Geophys. Res.*, 105(D22), 26,871–26,890.

Lawson, D. R. (1993), "Passing the test": Human behavior and California's smog check program, *J. Air Waste Manage. Assoc.*, 43, 1567–1575.

Lawson, D. R., and R. E. Smith (1998), The Northern Front Range Air Quality Study: A report to the Governor and General Assembly, Colo. State Univ., Fort Collins, Dec.

Leach, G. (1992), The energy transition, *Energy Policy*, 20, 116–123.

Leach, G., and M. Gowen (1987), Household energy handbook: An interim guide and reference manual, *Tech. Pap. 67*, World Bank, Washington, D. C.

Lighty, J. S., J. M. Veranth, and A. F. Sarofim (2000), Combustion aerosols: Factors governing their size and composition and implications to human health, *J. Air Waste Manage. Assoc.*, 50, 1565–1618.

Liousse, C., J. E. Penner, C. Chuang, J. J. Walton, and H. Eddleman (1996), A global three-dimensional study of carbonaceous aerosols, *J. Geophys. Res.*, 101(D14), 19,411–19,432.

Lucas, J. A., and T. F. Wall (1994), Volatile matter release, particle/cloud ignition, and combustion of near-stoichiometric suspensions of pulverized coal, in *Twenty-Fifth Symposium (International) on Combustion*, pp. 485–491, Combust. Inst., Pittsburgh, Pa.

Ludwig, J., L. T. Marufu, B. Huber, M. O. Andreae, and G. Helas (2003), Domestic combustion of biomass fuels in developing countries: A major source of atmospheric pollutants, *J. Atmos. Chem.*, 44, 23–37.

Luo, Y. F., D. R. Lu, X. J. Zhou, W. L. Li, and Q. He (2001), Characteristics of the spatial distribution and yearly variation of aerosol optical depth over China in last 30 years, *J. Geophys. Res.*, 106(D13), 14,501–14,513.

Ma, J., T. H. Fletcher, and B. W. Webb (1996), Conversion of coal tar to soot during coal pyrolysis in a post-flame environment, in *Twenty-Sixth Symposium (International) on Combustion*, pp. 3161–3167, Combust. Inst., Pittsburgh, Pa.

Mäkelä, K. (1995), Traffic emissions in Russia and the Baltic states, *Sci. Total Environ.*, 169, 219–229.

Mamane, Y., J. L. Miller, and T. G. Dzubay (1986), Characterization of individual fly-ash particles emitted from coal- and oil-fired power plants, *Atmos. Environ.*, 20, 2125–2135.

Mamuro, T., A. Mizohata, and T. Kubota (1979), Elemental compositions of suspended particles released from iron and steel works, *Annu. Rep. Radiat. Cent. Osaka Prefect.*, 20, 19–28.

Marsh, A. (1947), *Smoke: The Problem of Coal and the Atmosphere*, Faber and Faber, New York.

Martins, J. V., P. Artaxo, C. Liousse, J. S. Reid, P. V. Hobbs, and Y. Kaufman (1998), Effects of black carbon content, particle size, and mixing on light absorption by aerosol from biomass burning in Brazil, *J. Geophys. Res.*, 103(D24), 32,041–32,050.

Masera, O. R., B. D. Saatkamp, and D. M. Kammen (2000), From linear fuel switching to multiple cooking strategies: A critique and alternative to the energy ladder model, *World Dev.*, 28(12), 2083–2103.

Massey, F. J. (1951), The Kolmogorov-Smirnov test for goodness of fit, *J. Am. Stat. Assoc.*, 46(253), 68–78.

Mayol-Bracero, O. L., R. Gabriel, M. O. Andreae, T. W. Kirchstetter, T. Novakov, J. Ogren, P. Sheridan, and D. G. Streets (2002), Carbonaceous aerosols over the Indian Ocean during the Indian Ocean Experiment

(INDOEX): Chemical characterization, optical properties, and probable sources, *J. Geophys. Res.*, *107*(D19), 8030, doi:10.1029/2000JD000039.

McCormick, R. L., M. S. Graboski, T. L. Alleman, J. R. Alvarez, and K. G. Duleep (2003), Quantifying the emission benefits of opacity testing and repair of heavy-duty emission vehicles, *Environ. Sci. Technol.*, *37*, 630–637.

McDonald, J. D., B. Zielinska, E. M. Fujita, J. C. Sagebiel, J. C. Chow, and J. G. Watson (2000), Fine particle and gaseous emission rates from residential wood combustion, *Environ. Sci. Technol.*, *34*, 2080–2091.

Medalia, A. I., and D. Rivin (1982), Particulate carbon and other components of soot and carbon black, *Carbon*, *20*(6), 481–492.

Menaut, J. C., L. Abbadie, F. Lavenu, P. Loudjani, and A. Podaire (1996), Biomass burning in West African savannas, in *Biomass Burning and Global Change*, vol. 2, edited by J. S. Levine, pp. 133–142, MIT Press, Cambridge, Mass.

Menon, S., J. Hansen, and L. Nazarenko (2002), Climate effects of black carbon aerosols in China and India, *Science*, *297*, 2250–2253.

Mitra, A., A. F. Sarofim, and E. Bar-Ziv (1987), The influence of coal type on the evolution of polycyclic aromatic hydrocarbons during coal devolatilization, *Aerosol Sci. Technol.*, *6*, 261–271.

Muhlbaier, J. L. (1981), Particulate and gaseous emissions from natural gas furnaces and water heaters, *J. Air Pollut. Control Assoc.*, *31*, 1268–1273.

Mulholland, G. W., and M. Y. Choi (1998), Measurement of the mass-specific extinction coefficient for acetylene and ethene smoke using the Large Agglomerate Optics Facility, in *Twenty-Seventh Symposium (International) on Combustion*, pp. 1515–1522, Combust. Inst., Pittsburgh, Pa.

Müller, J. (1984), Atmospheric residence time of carbonaceous particles and particulate PAH-compounds, *Sci. Total Environ.*, *36*, 339–346.

Mumford, J. L., D. B. Harris, K. Williams, J. C. Chuang, and M. Cooke (1987), Indoor air sampling and mutagenicity studies of emissions from unvented coal combustion, *Environ. Sci. Technol.*, *21*, 308–311.

Myhre, G., F. Stordal, K. Restad, and I. S. A. Isaksen (1998), Estimation of the direct radiative forcing due to sulfate and soot aerosols, *Tellus, Ser. B*, *50*, 463–477.

Novakov, T., and C. E. Corrigan (1995), Thermal characterization of biomass smoke particles, *Microchim. Acta*, *119*, 157–166.

Novakov, T., S. G. Chang, and A. B. Harker (1974), Sulfates as pollution particulates: Catalytic formation on carbon (soot) particles, *Science*, *186*, 259–261.

Novakov, T., D. A. Hegg, and P. V. Hobbs (1997), Airborne measurements of carbonaceous aerosols on the east coast of the United States, *J. Geophys. Res.*, *102*(D25), 30,023–30,330.

Novakov, T., M. O. Andreae, R. Gabriel, T. W. Kirchstetter, O. L. Mayol-Bracero, and V. Ramanathan (2000), Origin of carbonaceous aerosols over the tropical Indian Ocean: Biomass burning or fossil fuels?, *Geophys. Res. Lett.*, *27*(24), 4061–4064.

Oanh, N. T. K., L. B. Reutergardh, and N. T. Dung (1999), Emission of polycyclic aromatic hydrocarbons and particulate matter from domestic combustion of selected fuels, *Environ. Sci. Technol.*, *33*, 2703–2709.

Oanh, N. T. K., L. H. Nghiem, and Y. L. Phyu (2002), Emission of polycyclic aromatic hydrocarbons, toxicity, and mutagenicity from domestic cooking using sawdust briquettes, wood, and kerosene, *Environ. Sci. Technol.*, *36*, 833–839.

Ogren, J. A., P. J. Groblicki, and R. J. Charlson (1984), Measurement of the removal rate of elemental carbon from the atmosphere, *Sci. Total Environ.*, *36*, 329–338.

Olivier, J. G. J., and J. A. H. W. Peters (2002), Uncertainties in global, regional and national emission inventories, in *Non-CO₂ Greenhouse Gases: Scientific Understanding, Control Options and Policy Aspects, Proceedings of the Third International Symposium, Maatstricht, Netherlands, 21–23 January 2002*, edited by J. Van Ham et al., pp. 525–540, Millpress Sci., Rotterdam.

Olmez, I., A. E. Sheffield, G. E. Gordon, J. E. Houck, L. C. Pritchett, J. A. Cooper, T. G. Dzubay, and R. L. Bennett (1988), Compositions of particles from selected sources in Philadelphia for receptor modeling applications, *J. Air Pollut. Control Assoc.*, *38*, 1392–1402.

Omursal, B., and S. P. Gautam (1997), Vehicular air pollution: Experiences from seven Latin American urban centers, *Tech. Pap. 373*, World Bank, Washington, D. C.

Parungo, F., C. Nagamoto, M.-Y. Zhou, A. D. A. Hansen, and J. Harris (1994), Aeolian transport of aerosol black carbon from China to the ocean, *Atmos. Environ.*, *27*, 3251–3260.

Patschull, J., and P. Roth (1995), Measurement and reduction of particles emitted from a two-stroke engine, *J. Aerosol Sci.*, *26*(6), 979–987.

Penner, J. E., H. Eddleman, and T. Novakov (1993), Towards the development of a global inventory for black carbon emissions, *Atmos. Environ., Part A*, *27*, 1277–1295.

Penner, J. E., C. C. Chuang, and K. Grant (1998), Climate forcing by carbonaceous and sulfate aerosols, *Clim. Dyn.*, *14*(12), 839–851.

Penner, J. E., D. H. Lister, D. G. Griggs, D. J. Dokken, and M. McFarland (Eds.) (1999), *Aviation and the Global Atmosphere: A Special Report of IPCC Working Groups I and III*, 373 pp., Cambridge Univ. Press, New York.

Penner, J. E., et al. (2002), A comparison of model- and satellite-derived aerosol optical depth and reflectivity, *J. Atmos. Sci.*, *59*, 441–460.

Pennise, D. M., K. R. Smith, J. P. Kithinji, M. E. Rezende, T. J. Raad, J. F. Zhang, and C. W. Fan (2001), Emissions of greenhouse gases and other airborne pollutants from charcoal making in Kenya and Brazil, *J. Geophys. Res.*, *106*(D20), 24,143–24,155.

Petzold, A., and A. Dopelheuer (1998), Reexamination of black carbon mass emission indices of a jet engine, *Aerosol Sci. Technol.*, *29*, 355–356.

Petzold, A., C. Kopp, and R. Niessner (1997), The dependence of the specific attenuation cross-section on black carbon mass fraction and particle size, *Atmos. Environ.*, *31*, 661–672.

Petzold, A., A. Dopelheuer, C. A. Brock, and F. Schroder (1999), In situ observations and model calculations of black carbon emission by aircraft at cruise altitude, *J. Geophys. Res.*, *104*(D18), 2171–2181.

Pfeiffer, F., M. Struschka, and G. Baumbach (2000), Ermittlung der mittleren Emissionsfaktoren zur Darstellung aus Feuerungsanslagen im Bereich der Haushalte und Kleinverbraucher, *Texte 14/00*, Umweltbundesamt, Berlin, May.

Pinto, J. P., R. K. Stevens, R. D. Willis, R. Kellogg, Y. Mamane, J. Novak, J. Santroch, I. Benes, J. Lenicek, and V. Bures (1998), Czech air quality monitoring and receptor modeling study, *Environ. Sci. Technol.*, *32*, 843–854.

Polenske, K. R., and F. C. McMichael (2002), A Chinese cokemaking process-flow model for energy and environmental analysis, *Energy Policy*, *30*, 865–883.

Qiu, J., and L. Yang (2000), Variation characteristics of atmospheric aerosol optical depths and visibility in North China during 1980–1994, *Atmos. Environ.*, *34*, 603–609.

Querol, X., J. L. Fernández-Turiel, and A. López-Soler (1995), Trace elements in coal and their behaviour during combustion in a large power station, *Fuel*, *74*(3), 331–343.

Ramanathan, V., P. J. Crutzen, J. T. Kiehl, and D. Rosenfeld (2001), Aerosols, climate and the hydrological cycle, *Science*, *294*, 2119–2124.

Randall, D. T., and H. W. Weeks (1909), *The Smokeless Combustion of Coal in Boiler Plants*, U.S. Govt. Print. Off., Washington, D. C.

Rau, J. A. (1989), Composition and size distribution of residential wood smoke particles, *Aerosol Sci. Technol.*, *10*, 181–192.

Raveendran, K., A. Ganesh, and K. C. Khilar (1995), Influence of mineral matter on biomass pyrolysis characteristics, *Fuel*, *74*(12), 1812–1822.

Reddy, A. K. N. (1983), Rural fuelwood: Significant relationships, in *Wood Fuel Surveys*, pp. 29–52, Food and Agric. Organ., U. N., Rome.

Reddy, M. S., and C. Venkataraman (2000), Atmospheric optical and radiative effects of anthropogenic aerosol constituents from India, *Atmos. Environ.*, *34*, 4511–4523.

Reddy, M. S., and C. Venkataraman (2002a), Inventory of aerosol and sulphur dioxide emissions from India: I. Fossil fuel combustion, *Atmos. Environ.*, *36*, 677–697.

Reddy, M. S., and C. Venkataraman (2002b), Inventory of aerosol and sulphur dioxide emissions from India: II. Biomass combustion, *Atmos. Environ.*, *36*, 699–712.

Rosen, H., A. D. A. Hansen, L. Gundel, and T. Novakov (1978), Identification of the optically absorbing component in urban aerosols, *Appl. Opt.*, *17*(24), 3859–3861.

Rosenfeld, D. (2000), Suppression of rain and snow by urban air pollution, *Science*, *287*, 1793–1796.

Sagabiel, J. C., B. Zielinska, P. A. Walsh, J. C. Chow, S. H. Cadle, P. A. Mulawa, K. T. Knapp, R. B. Zweidinger, and R. Snow (1997), PM-10 exhaust samples collected during IM-240 dynamometer tests of in-service vehicles in Nevada, *Environ. Sci. Technol.*, *31*, 75–83.

Sakai, T., T. Nakajima, and H. Yamazaki (1999), O-PM/emitted matters caused by two-stroke engine oil and its reduction, *SAE Pap. 1999-91-3260*, Soc. of Automotive Eng., Warrendale, Pa.

Samaras, Z., and K.-H. Zierock (1995), Off-road vehicles: A comparison with those from road transport, *Sci. Total Environ.*, *169*, 249–255.

Sanborn, C. R. (1982), Characterization of emissions from residential coal stoves, in *Proceedings of the Residential Wood and Coal Combustion Specialty Conference*, pp. 151–160, Air Pollut. Control Assoc., Louisville, Ky.

Sanborn, C. R., and M. A. Blanchet (1982), Particulate emissions from residential wood combustion in Vermont, in *1981 International Conference on Residential Solid Fuels*, pp. 188–198, Oreg. Grad. Cent., Portland.

Sanhueza, E. (1996), Effects of vegetation burning on the atmospheric chemistry of the Venezuelan Savanna, in *Biomass Burning and Global Change*, vol. 2, edited by J. S. Levine, pp. 122–125, MIT Press, Cambridge, Mass.

Satheesh, S. K., and V. Ramanthan (2000), Large differences in tropical aerosol forcing at the top of the atmosphere and Earth's surface, *Nature*, *405*, 60–63.

Sato, M., J. Hansen, D. Koch, A. Lacis, R. Ruedy, O. Dubovik, B. Holben, M. Chin, and T. Novakov (2003), Global atmospheric black carbon inferred from AERONET, *Proc. Natl. Acad. Sci. U. S. A.*, *100*(11), 6319–6324.

Saxena, P., L. M. Hildemann, P. H. McMurry, and J. H. Seinfeld (1995), Organics alter hygroscopic behavior of atmospheric particles, *J. Geophys. Res.*, *100*(D9), 18,755–18,770.

Schauer, J. J., W. F. Rogge, L. M. Hildemann, M. A. Mazurek, and G. R. Cass (1996), Source apportionment of airborne particulate matter using organic compounds as tracers, *Atmos. Environ.*, *30*, 3837–3855.

Schauer, J. J., et al. (2003), ACE-Asia intercomparison of a thermal-optical method for the determination of particle-phase organic and elemental carbon, *Environ. Sci. Technol.*, *37*, 993–1001.

Schmid, H. L., et al. (2001), Results of the "carbon conference" international aerosol carbon round robin test stage I, *Atmos. Environ.*, *35*, 2111–2121.

Schneider, S. H. (1971), A comment on "Climate: The influence of aerosols," *J. Appl. Meteorol.*, *10*(4), 840–841.

Secretaria del Medio Ambiente, Recursos Naturales y Pesca (SEMARNAP) (2001), Programa forestal y de suelo 1995–2000, Mexicali, Mexico.

Seeker, W. R., G. S. Samuelsen, M. P. Heap, and J. D. Trolinger (1981), The thermal decomposition of pulverized coal particles, in *Eighteenth Symposium (International) on Combustion*, pp. 1213–1226, Combust. Inst., Pittsburgh, Pa.

Shah, J., and T. Nagpal (Eds.) (1996a), *URBAIR: Urban Air Quality Management Strategy in Asia, Kathmandu Valley Report*, World Bank, Washington, D. C.

Shah, J., and T. Nagpal (Eds.) (1996b), *URBAIR: Urban Air Quality Management Strategy in Asia, Jakarta Report*, World Bank, Washington, D. C.

Shlyakhter, A. I. (1994), An improved framework for uncertainty analysis: Accounting for unsuspected errors, *Risk Anal.*, *14*(4), 441–447.

Shulman, M. L., M. C. Jacobson, R. J. Charlson, R. E. Synovec, and T. E. Young (1996), Dissolution behavior and surface tension effects of organic compounds in nucleating cloud droplets, *Geophys. Res. Lett.*, *23*(3), 277–280.

Smith, D. M., and A. R. Chughtai (1995), The surface structure and reactivity of black carbon, *Colloids Surf. A*, *105*, 47–77.

Smith, K. R. (1987), *Biofuels, Air Pollution, and Health: A Global Review*, Plenum, New York.

Smith, K. R., S. Gu, K. Huang, and D. Qiu (1993), One hundred million improved cookstoves in China: How was it done?, *World Dev.*, *21*(6), 941–961.

Smith, K. R., M. G. Apte, Y. Ma, W. Wongsekiarttirat, and A. Kulkarni (1994), Air pollution and the energy ladder in Asian cities, *Energy*, *9*, 587–600.

Smith, K. R., D. M. Pennise, P. Khummongkol, V. Chaiwong, K. Ritgeen, J. Zhang, W. Panyathanya, R. A. Rasmussen, and K. A. Khalil (1999), Greenhouse gases from small-scale combustion devices in developing countries: Charcoal-making kilns in Thailand, *EPA-600/R-99-109*, U.S. Environ. Prot. Agency, Research Triangle Park, N. C.

Smith, K. R., R. Uma, V. V. N. Kishore, K. Lata, V. Joshi, J. Zhang, R. A. Rasmussen, and K. A. Khalil (2000), Greenhouse gases from small-scale combustion devices in developing countries: Household stoves in India, *Rep. EPA-600/R-00-052*, U.S. Environ. Prot. Agency, Research Triangle Park, N. C.

Smith, O. I. (1981), Fundamentals of soot formation in flames with application to diesel engine particulate emissions, *Prog. Energy Combust. Sci.*, *7*, 275–291.

Smith, R. D. (1980), The trace element chemistry of coal during combustion and the emissions from coal-fired plants, *Prog. Energy Combust. Sci.*, *6*, 53–119.

Smith, R. D., J. A. Campbell, and K. K. Nielson (1979), Characterization and formation of submicron particles in coal-fired plants, *Atmos. Environ.*, *13*, 607–617.

Sokolik, I. N., and O. B. Toon (1996), Direct radiative forcing by anthropogenic airborne mineral aerosols, *Nature*, *381*, 681–683.

Solum, M. S., A. F. Sarofim, R. J. Pugmire, T. H. Fletcher, and H. Zhang (2001), ^{13}C NMR analysis of soot produced from model compounds and a coal, *Energy Fuels*, *15*, 961–971.

Spitzer, J., P. Enzinger, G. Fankhauser, W. Fritz, F. Golja, and R. Stiglbrunner (1998), *Emissionsfaktoren für Feste Brennstoffe*, Joanneum Res., Graz, Germany.

Still, D., M. Hatfield, and P. Scott (2000), *Capturing Heat Two*, Aprovecho Res. Cent., Cottage Grove, Oreg.

Streets, D. G., S. Gupta, S. T. Waldhoff, M. Q. Wang, T. C. Bond, and Y. Bo (2001), Black carbon emissions in China, *Atmos. Environ.*, *35*, 4281–4296.

Streets, D. G., K. F. Yarber, J.-H. Woo, and G. R. Carmichael (2003a), Biomass burning in Asia: Annual and seasonal estimates and atmospheric emissions, *Global Biogeochem. Cycles*, *17*(4), 1099, doi:10.1029/2003GB002040.

Streets, D. G., et al. (2003b), An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, *J. Geophys. Res.*, *108*(D21), 8809, doi:10.1029/2002JD003093.

Suuberg, E. M., W. A. Peters, and J. B. Howard (1978), Product compositions and formation kinetics in rapid pyrolysis of pulverized coal: Implications for combustion, in *Seventeenth Symposium (International) on Combustion*, pp. 117–130, Combust. Inst., Pittsburgh, Pa.

Suutari, R., M. Amann, J. Cofala, Z. Klimont, M. Posch, and W. Schöpp (2001), From economic activities to ecosystem protection in Europe: An uncertainty analysis of two scenarios of the RAINS integrated assessment model, *CIAM/CCE Rep. 1/2001*, Int. Inst. for Appl. Syst. Anal., Laxenburg, Austria.

Tata Energy Research Institute (TERI) (1996), *TEDDY: TERI Energy Data Directory and Yearbook*, New Delhi, India.

Timothy, L. D., D. Froelich, A. F. Sarofim, and J. M. Beér (1986), Soot formation and burnout during the combustion of dispersed pulverized coal particles, *Twenty-First Symposium (International) on Combustion*, pp. 1141–1148, Combust. Inst., Pittsburgh, Pa.

Truesdale, R. S., and J. G. Cleland (1982), Residential stove emissions from coal and other alternative fuels combustion, in *Residential Wood and Coal Combustion*, pp. 115–128, Air Pollut. Control Assoc., Louisville, Ky.

Tuladhar, B., and A. K. Raut (2002), Final report on environment and health impacts of Kathmandu's brick kilns, Clean Energy Nepal, Kathmandu, June.

Turco, R. P., et al. (1983), The global cycle of particulate elemental carbon, in *Precipitation Scavenging, Dry Deposition, and Resuspension*, edited by H. R. Pruppacher, R. G. Semonin, and W. G. N. Slinn, pp. 1337–1351, Elsevier Sci., New York.

Twomey, S. A., M. Piepgrass, and T. L. Wolfe (1984), An assessment of the impact of pollution on global cloud albedo, *Tellus, Ser. B*, *36*, 356–366.

United Nations Economic Commission for Europe (UN-ECE) (1990a), The 1989 forest fire season (Austria), *Int. For. Fire News*, no. 4, 3.

United Nations Economic Commission for Europe (UN-ECE) (1990b), Forest fires in Greece, 1990, *Int. For. Fire News*, no. 4, 7.

United Nations Economic Commission for Europe (UN-ECE) (1990c), A synopsis of the Greek wildland fire problem, *Int. For. Fire News*, no. 4, 6–7.

United Nations Economic Commission for Europe (UN-ECE) (1992), Long-term fire statistics update (Germany), *Int. For. Fire News*, no. 7, 17, 21.

United Nations Economic Commission for Europe (UN-ECE) (1993a), Improving the national wildland fire statistics of Argentina, *Int. For. Fire News*, no. 9, 6–7.

United Nations Economic Commission for Europe (UN-ECE) (1993b), Forest fires in Croatia: A permanent danger, *Int. For. Fire News*, no. 9, 13–15.

United Nations Economic Commission for Europe (UN-ECE) (1995a), Wildfires in Albania, *Int. For. Fire News*, no. 12, 2–4.

United Nations Economic Commission for Europe (UN-ECE) (1995b), Forest fire profile (France), *Int. For. Fire News*, no. 13, 12–15.

United Nations Economic Commission for Europe (UN-ECE) (1999a), Forest fires in 1998: An average year (Spain), *Int. For. Fire News*, no. 20, 78–79.

United Nations Economic Commission for Europe (UN-ECE) (1999b), Forest fires in Italy 1998, *Int. For. Fire News*, no. 21, 60–70.

United Nations Economic Commission for Europe (UN-ECE) (2000a), Forest fires in Algeria and the case of the Domanial forest of Bou-Taleb, Setif, *Int. For. Fire News*, no. 22, 9–15.

United Nations Economic Commission for Europe (UN-ECE) (2000b), The 1998 forest fire season (Bulgaria), *Int. For. Fire News*, no. 22, 19–20.

United Nations Economic Commission for Europe (UN-ECE) (2000c), The 1999 forest fire season (Greece), *Int. For. Fire News*, no. 22, 25–28.

United Nations Economic Commission for Europe (UN-ECE) (2001a), Fire situation in Kenya, *Int. For. Fire News*, no. 25, 12–14.

United Nations Economic Commission for Europe (UN-ECE) (2001b), Baltic fire special, and forest fire situation in Kazakhstan, *Int. For. Fire News*, no. 24, 2–59.

United Nations Economic Commission for Europe (UN-ECE) (2002), Fire situation in Portugal, *Int. For. Fire News*, no. 27, 65–67.

United Nations Economic Commission for Europe (UN-ECE) (2003), Forest fire situation in Chile, *Int. For. Fire News*, no. 28, 57–65.

U.S. Department of Commerce (1997), 1997 economic census: Vehicle inventory and use survey, Bureau of the Census, Washington, D. C.

U.S. Environmental Protection Agency (U.S. EPA) (1991), Nonroad engine and vehicle emission study (NEVES), *Rep. EPA-21A-2001*, Research Triangle Park, N. C.

U.S. Environmental Protection Agency (U.S. EPA) (1995), Draft user's guide to PART5: A program for calculating particle emissions from motor vehicles, *Rep. EPA-AA-AQAB-94-2*, Ann Arbor, Mich.

U.S. Environmental Protection Agency (U.S. EPA) (1996), Compilation of air pollutant emission factors (and subsequent updates), *Rep. AP-42*, Research Triangle Park, N. C.

U.S. Environmental Protection Agency (U.S. EPA) (1997), Emission factors for locomotives, *Rep. EPA420-F-97-051*, Research Triangle Park, N. C.

U.S. Environmental Protection Agency (U.S. EPA) (1998a), Characterization of municipal solid waste in the United States: 1997 update, *Rep. EPA530-R-98-007*, Research Triangle Park, N. C.

U.S. Environmental Protection Agency (U.S. EPA) (1998b), Exhaust emission factors for nonroad engine modeling: Compression-ignition, *Rep. NR-009-A*, Research Triangle Park, N. C.

U.S. Environmental Protection Agency (U.S. EPA) (2000), Analysis of commercial marine vessels emissions and fuel consumption data, *Rep. 420-R-00-002*, Research Triangle Park, N. C.

U.S. Environmental Protection Agency (U.S. EPA) (2001), National air quality and emissions trends report 1999, *Rep. EPA 454/R-01-004*, Research Triangle Park, N. C.

U.S. Environmental Protection Agency (U.S. EPA) (2002), Documentation for version 2 of the 1999 NEI for criteria air pollutants: Area sources, Research Triangle Park, N. C.

U.S. Steel (1985), *The Making, Shaping, and Treating of Steel*, 10th ed., Assoc. of Iron and Steel Eng., Pittsburgh, Pa.

van Aardenne, J. A., F. J. Dentener, J. G. J. Olivier, C. G. M. Klein Goldewijk, and J. Lelieveld (2001), A 1° × 1° resolution data set of historical anthropogenic trace gas emissions for the period 1890–1990, *Global Biogeochem. Cycles*, 15, 909–928.

Vanderwal, R. L. (1996), Soot precursor material: Visualization via simultaneous LIF-LII and characterization via TEM, in *Twenty-Sixth Symposium (International) on Combustion*, pp. 2269–2275, Combust. Inst., Pittsburgh, Pa.

Venkataraman, C., and G. U. M. Rao (2001), Emission factors of carbon monoxide and size-resolved aerosols from biofuel combustion, *Environ. Sci. Technol.*, 35, 2100–2107.

Veranth, J. M., D. W. Pershing, A. F. Sarofim, and J. E. Shield (1998), Sources of unburned carbon in the fly ash produced from low-NOx pulverized coal combustion, in *Twenty-Seventh Symposium (International) on Combustion*, pp. 1737–1744, Combust. Inst., Pittsburgh, Pa.

Wang, X., and Z. Fend (1996), Survey of rural household energy consumption in China, *Energy*, 21, 703–705.

Watson, J. G., and J. C. Chow (2001), Source characterization of major emission sources in the Imperial and Mexicali Valleys along the U. S./Mexico border, *Sci. Total Environ.*, 276, 33–47.

Weingartner, E., C. Keller, W. A. Stahel, H. Burtscher, and U. Baltensperger (1997), Aerosol emission in a road tunnel, *Atmos. Environ.*, 31, 451–462.

Weingartner, E., H. Saathof, M. Schnaiter, N. Streit, B. Bitnar, and U. Baltensperger (2003), Absorption of light by soot particles: Determination of the absorption coefficient by means of aethalometers, *J. Aerosol Sci.*, 13, 1445–1463.

Wenzel, T., B. C. Singer, and R. Slott (2000), Some issues in the statistical analysis of vehicle emissions, *J. Transp. Stat.*, 3(2), 1–14.

Westhoff, B., and D. Germann (1995), Stove images: A documentation of improved and traditional stoves in Africa, Asia and Latin America, Comm. of the Eur. Communities, Brussels.

Williams, D. J., J. W. Milne, and D. B. Roberts (1989), Particulate emissions from 'in-use' motor vehicles: I. Spark ignition engines, *Atmos. Environ.*, 23, 2639–2645.

Winiwarter, W., C. Trenker, and W. Höflinger (2001), Österreichische Emissionsinventur für Staub, *ARCS-0151*, Austrian Res. Cent., Seibersdorf.

Wolff, G. T., R. J. Countess, P. J. Groblicki, M. A. Ferman, S. H. Cadle, and J. L. Muhlbaier (1981), Visibility-reducing species in the Denver "brown cloud": II. Sources and temporal patterns, *Atmos. Environ.*, 15, 2485–2502.

Woo, J.-H., J. M. Baek, J.-W. Kim, G. R. Carmichael, N. Tongboonchoo, S. T. Kim, and J. H. An (2003a), Development of a multi-resolution emission inventory system and its impact on sulfur distribution for northeast Asia, *Water Air Soil Pollut.*, 148, 259–278.

Woo, J. H., et al. (2003b), The contribution of biomass and biofuel emissions to trace gas distributions in Asia during the TRACE-P experiment, *J. Geophys. Res.*, 108(D21), 8812, doi:10.1029/2002JD003200.

World Bank (1998), *Pollution Prevention and Abatement Handbook 1998: Toward Cleaner Production*, Washington, D. C.

Wu, J.-S., S. S. Krishnan, and G. M. Faeth (1997), Refractive indices at visible wavelengths of soot emitted from buoyant turbulent diffusion flames, *J. Heat Transfer*, 119, 230–237.

Yang, H., and J. Yu (2002), Uncertainties in charring correction in the analysis of elemental and organic carbon in atmospheric particles by thermal/optical methods, *Environ. Sci. Technol.*, 36, 5199–5204.

Yanowitz, J., M. S. Graboski, L. B. A. Ryan, T. L. Alleman, and R. L. McCormick (1999), Chassis dynamometer study of emissions from 21 in-use heavy-duty diesel vehicles, *Environ. Sci. Technol.*, 33, 209–216.

Yanowitz, J., R. L. McCormick, and M. S. Graboski (2000), In-use emissions from heavy-duty diesel vehicles, *Environ. Sci. Technol.*, 34, 729–740.

Ye, B., X. Ji, H. Yang, X. Yao, C. K. Chan, S. H. Cadle, T. Chan, and P. A. Mulawa (2003), Concentration and chemical composition of $PM_{2.5}$ in Shanghai for a 1-year period, *Atmos. Environ.*, 37, 499–510.

Yevich, R., and J. A. Logan (2003), An assessment of biofuel use and burning of agricultural waste in the developing world, *Global Biogeochem. Cycles*, 17(4), 1095, doi:10.1029/2002GB001952.

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

Zhang, Y., G. A. Bishop, and D. H. Stedman (1994), Automobile emissions are statistically gamma-distributed, *Environ. Sci. Technol.*, 28, 1370–1374.

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T. C. Bond, Department of Civil and Environmental Engineering, University of Illinois, MC-250, 205 N. Mathews Avenue, Urbana, IL 61801, USA. (yark@uiuc.edu)

Z. Klimont, International Institute for Applied Systems Analysis, Laxenburg, A-2361 Austria. (klimont@iiasa.ac.at)

S. M. Nelson, D. G. Streets, and K. F. Yarber, Argonne National Laboratory, DIS/900, 9700 South Cass Avenue, Argonne, IL 60439, USA. (dstreets@anl.gov)

J.-H. Woo, Center for Global and Regional Environmental Research, University of Iowa, Iowa City, IA 52242, USA. (woojh21@cgrer.uiowa.edu)