Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850–2000

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Received 13 September 2006; revised 23 January 2007; accepted 15 February 2007; published 30 May 2007.

[1] We present an emission inventory of primary black carbon (BC) and primary organic carbon (OC) aerosols from fossil fuel and biofuel combustion between 1850 and 2000. We reconstruct fossil fuel consumption and represent changes in technology on a national and sectoral basis. Our estimates rely on new estimates of biofuel consumption, and updated emission factors for old technologies. Emissions of black carbon increase almost linearly, totaling about 1000 Gg in 1850, 2200 Gg in 1900, 3000 Gg in 1950, and 4400 Gg in 2000. Primary organic carbon shows a similar pattern, with emissions of 4100 Gg, 5800 Gg, 6700 Gg, and 8700 Gg in 1850, 1900, 1950, and 2000, respectively. Biofuel is responsible for over half of BC emission until about 1890, and dominates energy-related primary OC emission throughout the entire period. Coal contributes the greatest fraction of BC emission between 1880 and 1975, and is overtaken by emissions from biofuel around 1975, and by diesel engines around 1990. Previous work suggests a rapid rise in BC emissions between 1950 and 2000. This work supports a more gradual increase between 1950 and 2000, similar to the increase between 1850 and 1925; implementation of clean technology is a primary reason.

Citation: Bond, T. C., E. Bhardwaj, R. Dong, R. Jogani, S. Jung, C. Roden, D. G. Streets, and N. M. Trautmann (2007), Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850–2000, *Global Biogeochem. Cycles*, *21*, GB2018, doi:10.1029/2006GB002840.

1. Importance of Carbonaceous Aerosols

[2] Recent work has suggested that climate forcing by carbonaceous aerosols is probably a significant component of anthropogenic forcing. *Schulz et al.* [2006] summarize modeled estimates of average forcing. Forcing by black carbon (BC) from fossil-fuel combustion range from about +0.1 to 0.3 W/m^2, and similar estimates for primary organic carbon (OC) particles are −0.01 to −0.06 W/m^2. As a global average, these values, especially BC forcing, are significant relative to the average CO_2 forcing of about +1.5 W/m^2. Regional aerosol forcings can be an order of magnitude greater than greenhouse gas forcings [e.g., *Satheesh and Ramanthan*, 2000]. In addition, early in the industrial era, aerosols may have contributed more to average forcing owing to low levels of accumulated anthropogenic CO_2 and comparatively high aerosol pollution.

[3] The "forcing-response" paradigm holds that the sensitivity of the climate system to anthropogenic forcing can be constrained by comparing calculated climate forcings with temperature records [*Hansen et al.*, 1997; *Andronova and Schlesinger*, 2001]. This comparison requires that climate forcings be derived from first principles, completely separate from the temperature record. Carbonaceous aerosols need to be included in models in order to estimate net anthropogenic forcing, and thereby assess the ability of models to simulate temperature records.

[4] There have been a few attempts to represent the time dependence of aerosol emissions. *Myhre et al.* [2001] applied emission factors of *Cooke et al.* [1999] to International Energy Agency data to infer a time trend since 1850. *Novakov et al.* [2003] estimated changes in black carbon between 1900 and 2000, accounting for changes in sources by applying a "technology factor" that decreased linearly between 1965 and 1985. *Lefohn et al.* [1999] considered changes in technology and activity to estimate sulfur emissions; their approach is most similar to the one we present here, but they did not estimate carbonaceous aerosol emissions. *Ito and Penner* [2005] applied detailed assumptions about emission factors for different end uses and economic sectors (residential, transport, industrial, and power generation) to estimate a historical trend since 1950. Prior to that time, they scaled BC and OC emissions to CO_2. Emissions of black carbon from fossil fuel predicted by both *Novakov et al.* [2003] and *Ito and Penner* [2005] show a sharp emission increase beginning around 1950. *Junker and Liousse* [2006] also calculated emissions for different sectors and used time-varying emission factors. Their time trends are similar to those of *Novakov et al.* [2003]. The Ito and Penner results are about half as large as

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0886-6236/07/2006GB002840$12.00

Table 1. Major Data Sources for Fossil Fuels[a]

Period	Data Source	Fuel Description	Division Between Sectors	Division Within Sector
1923–1943	League of Nations	some	little	no
1925–1965	RFF (Tabulation)	some	none	no
1850–1950	*Andres et al.* [1999]	basic	none	no
1950–1970	United Nations	full	very little	no
1949–2000	EIA, USA only	full	always	no
1970–1979	United Nations/International Energy Agency	full	some	some
1980–1985	UN/IEA	full	usually	some
1986–1990	UN/IEA	full	usually	many
1990 to present	IEA	full	always	usually

[a]"Basic" means that only solid/liquid/gas measurements were available. RFF, Resources for the Future [*Darmstadter et al.*, 1971]; ORNL, Oak Ridge National Laboratory; UN, United Nations; IEA, International Energy Agency; EIA, Energy Information Administration.

the Novakov estimates (2.5 Tg/yr versus 6 Tg/yr for 2000) owing to differences in emission factors.

[5] While many of these earlier works recognized the important contribution of technology in determining emissions, they did not explicitly account for such changes. The work presented here does reflect technology change, and demonstrates that it strongly affects the time history of carbonaceous aerosols. While we admit the existence of large uncertainties due to lack of data in the early industrial era, the potential importance of carbonaceous aerosol forcing demands the best possible estimate.

2. General Methodology

[6] We follow earlier work [*Bond et al.*, 2004; *Streets et al.*, 2004] by assuming emissions of particulate matter depend mainly on technology choice. The modified equation below represents time-dependent emissions for each pollutant species and country.

$$Em_{j,k}(t) = \sum_{l}\sum_{m} FC_{k,l,m}(t)\left[\sum_{n} EF_{j,k,l,m,n}X_{k,l,m,n}(t)\right], \qquad (1)$$

where

- j, k, l, m, n subscripts representing species, country, sector, fuel type, and fuel/technology combination, respectively;
- Em (t) emissions of species j from country k;
- FC fuel consumption (kg/year);
- EF emission factor specific to each fuel/technology, including the effects of postcombustion controls;
- X fraction of fuel for this sector consumed by a specific technology; $\Sigma X = 1$ for each fuel and sector.

[7] We assume that emission factors remain constant for individual technologies, except for improvements in combustion controls; generally, trends toward cleaner combustion can be represented as changes in technology splits. Thus we have focused on changes in technology fractions and sectoral divisions. Many of the emission factors are taken from *Bond et al.* [2004]. Additional emission factors for technologies that were widely used in earlier times are discussed in section 4.

[8] Data management was handled by the Speciated Pollutant Emission Wizard (SPEW). We have made two major additions to this program since its first description [*Bond et al.*, 2004]. First, technology fractions are now represented as time-dependent functions, $X(t)$. Second, a new module assembles sectoral fuel consumption from proxies, as described later.

[9] Like *Streets et al.* [2004], we often use the S-shaped curves discussed by *Grübler et al.* [1999] and *Geroski* [2000] to represent technology diffusion. These curves depict smooth transitions and cannot account for economic shocks. However, they fit historical trends better than linear or polynomial fits, so we often use them to represent time-dependent values of $X(t)$ throughout SPEW. The form we have found most useful is

$$X(t) = (X_0 - X_f)e^{\left(-(t-t_0)^2/2s^2\right)} + X_f, \qquad (2)$$

where X_0 and X_f are the initial and final values of the technology fractions, respectively; t_0 is the time at which the transition begins, and s is a rate. For polluting technologies that were phased out, X_f is often zero and X_0 is one. We represent time trends by obtaining values of s and t_0 from historical data, which are usually available for only a small number of countries. We then extrapolate these fits to other, similar countries. Parameters for some of these transitions are discussed throughout the paper, and are summarized in the auxiliary material[1], Table S1.

3. Sectoral Divisions for Fossil Fuels

[10] Total fossil-fuel consumption came from several major sources, listed in Table 1. *U. N. Statistics Division* [1995] and *International Energy Agency* [2004a, 2004b] data are used for recent years. These data originate from similar sources (country reports) and consumption totals are frequently identical. *Andres et al.* [1999] presented a database of fossil-fuel consumption before 1950 that has previously been used for historical emission estimates [*van Aardenne et al.*, 2001; *Smith et al.*, 2004]. The authors base their data on those compiled by *Etemad et al.* [1991]; they admit that the data contain uncertainties, particularly during World War II. Each data set provides production, imports and exports; some also give consumption data. We estimated consumption as the sum of production and imports less

[1]Auxiliary material data sets are available at ftp://ftp.agu.org/apend/gb/2006gb002840. Other auxiliary material files are in the HTML.

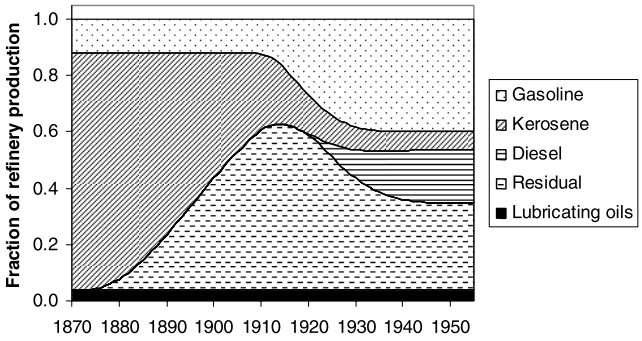

Figure 1. Estimated fractions of refinery output.

exports. Bunker fuels were assumed used in the year they were reported.

[11] Fuel-use data for recent years are divided into sectors: power generation, industry, transportation, and "other." The latter includes residential, commercial and agricultural consumption. One or more technologies characterize each sector, and may differ between sectors. For example, power generation generally uses larger coal burners with better combustion or controls than industrial applications. Therefore sectoral divisions of fuel consumption are critical in estimating emissions. Some specific end uses, such as consumption in the steel industry, also affect emissions. Table 1 shows that fuel consumption data give such divisions only for recent years, beginning around 1980.

[12] The first challenge in developing a historical emission record, then, was to reconstruct sectoral fuel-use consumption. This section describes how we divided fuel use into sectors based on indicators. The best indicator would be reported fuel consumption within a sector, but if it is not available, a second-best indicator is used, for example per-capita consumption multiplied by population. An unlimited number of indicators is possible, but in practice we used no more than three. Indicators can be represented as discrete data (for example, population) or as time-varying functions (for example, per-capita consumption) and can be country-specific, region-specific, or global defaults. The SPEW Historical Builder, developed for this project, chooses indicators in a hierarchical manner for each country.

[13] Many indicators were not available for all countries and years. When data such as per-capita consumption or vehicle ownership were not available for a country, we estimated values using a neighbor with data, or other similar countries. This process could introduce some uncertainty, because bordering countries may be drastically different owing to politics or resources. The availability of statistics in one country may indicate a level of development not present in neighboring countries without statistics. However, countries with missing data were usually not major fuel consumers.

3.1. Liquid Fuel Consumption

[14] The data set described by *Andres et al.* [1999] provides production, imports and exports of crude oil and total petroleum products between 1850 and 1949. The major products of crude-oil refineries have differing emission factors, and need to be separated, as demonstrated by *Marland and Rotty* [1984]. We divided crude oil into five refined products: kerosene, motor gasoline, middle distillates, residual fuel, and lubricating products.

[15] We used production, imports and exports of crude oil to estimate consumption, and assumed that all crude oil was used in refining. Between 1950 and 1960, refinery outputs rose from 92% to 95% of the input, and we use 92% as an efficiency estimate before 1950. The fraction of output appearing in each of the five products is not constant, but it is limited; Figure 1 shows the assumed fraction of each refinery product. For 1850–1929 and 1936–1950, we found refinery outputs for only the U.S. [*Schurr and Netschert*, 1960]. For 1929–1936, we used League of Nations data for the 21 largest refining countries. We use a constant lubricating oil fraction of 0.04; the remainder of the transitions are S-diffusion curves (auxiliary material Table S1).

[16] For each country, we calculated consumption of the five products according to the equation:

$$\begin{aligned} consumption_{prod} = {} & refinery\text{-}production_{prod} + imports_{tot\text{-}prod} \\ & \times import\text{-}fraction_{prod} - exports_{tot\text{-}prod} \\ & \times export\text{-}fraction_{prod}. \end{aligned} \quad (3)$$

[17] Here the subscript *prod* indicates product-specific data, and the subscript *tot-prod* indicates total refined petroleum products. *Andres et al.* [1999] give imports and exports of total refined petroleum products for each year. *Darmstadter et al.* [1971] give imports and exports of individual refinery products for 1925–1965. Missing years were obtained by fitting time-dependent curves.

3.2. Needed Sectoral Divisions

[18] We assumed that we would adequately represent emissions if we identified the fractions of the three major uses for diesel and residual oil, as summarized in Table 2 for 1970–1975. The percentages in the table indicate the sectors in which a fuel is commonly used, but they cannot be extrapolated to earlier years, because sectoral demand has changed substantially. Most light distillates (80%) were used in road transport, and we assume that all this fuel is used in transport for earlier years.

Table 2. Major Uses of Middle Distillate and Residual Fuel[a]

	Fraction of Consumption, %	EF_{PM}, g/kg
Residual fuel		
Power generation	38	1
Industry	33	1
Shipping	21	2
Middle distillates		
Domestic	50	0.2
Road and off-road vehicles	26	2 (road)
Railroad	5	3

[a]Percentage use is from United Nations data, 1970–1975, for countries in which sectoral divisions are given. Particulate matter emission factors are sectoral averages for Western Europe in 1996 from *Bond et al.* [2004], shown to one significant figure, to indicate when divisions are important.

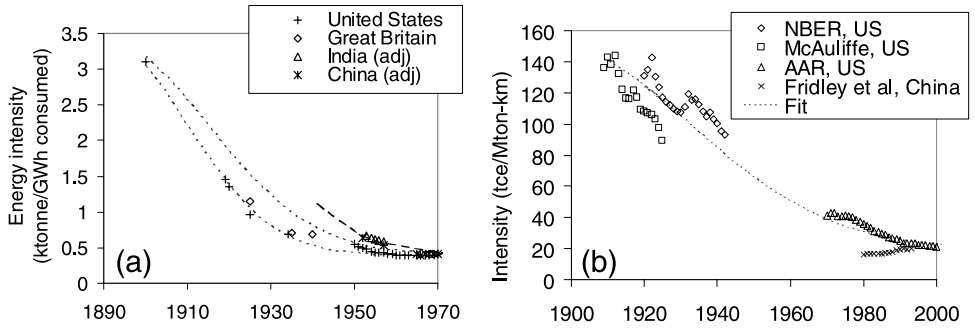

Figure 2. Energy intensity for (a) coal-fired electrical power plants and (b) rail freight. For power plants, the short-dashed lines show $s = 20$ and $s = 25$; we use $s = 22$. Intensities for India and China are corrected to heat content of 26 MJ/kg. For rail freight, data before 1940 and China data are coal-fired locomotives; AAR data are mostly diesel locomotives. Abbreviations: NBER, National Bureau of Economic Research; AAR, Association of American Railroads.

[19] Twelve countries accounted for 90% of total coal consumption between 1850 and 1970 (see auxiliary material Figure S1). We divided coal into power generation, industrial, transport, and domestic sectors. Industrial is further divided into coal used in coke ovens and other industrial uses, while transport includes railroads and steamships. Coke from coal-gas plants and coal gas made up less than 3% of the total [*Kiessling*, 1934–1936]. Annual records of sectoral consumption are available only for the United States [*U.S. Department of the Interior*, 1939; *Bituminous Coal Institute*, 1939–1941; *Energy Information Administration*, 2004]. Usage in other countries was assembled as discussed below.

3.3. Electric Utilities

[20] Progress in pulverized-coal burning made large-scale plants feasible in the 1930s [*Rosins*, 1947], and electrical generation grew rapidly. Thus the trend in electrification indicates a trend toward cleaner burning of coal. Coal used in electric utilities is available on an annual basis for the United States from 1918 onward. We did not find similarly detailed statistics for most other countries. To estimate coal used in power generation, we compiled statistics on electricity generated in thermal power plants in each country. The United Nations database provides this information for years after 1950. Before 1950, we used electrical consumption tabulated by *Mitchell* [1998a, 1998b, 1998c], and subtracted hydroelectric generation tabulated by *Darmstadter et al.* [1971], *Schurr and Netschert* [1960], and *Fridley and Sinton* [2004]. We assume that nuclear electric power was negligible prior to 1950. Coal consumption in power plants is given by

$$\begin{aligned} consump_{coal,power} = {} & thermal\text{-}electric(\mathrm{GWh}) \times fraction_{coal} \\ & \times energy\text{-}intensity(\mathrm{ktonne/Gwh}) \\ & \times heating\text{-}val_{ref}/heating\text{-}val, \end{aligned} \tag{4}$$

where $fraction_{coal}$ is the fraction generated with coal, *heating-val* is the 1980 heating value of the country's coal, and $heating\text{-}val_{ref}$ is the heating value of coal in countries used to determine energy intensity.

[21] Figure 2a shows the development of energy intensity for coal-fired electrical plants. These data came from *Largo-Afonso* [1959–1961], *Hoar* [1930], *Kiessling* [1935], *Rosins* [1947], *Bituminous Coal Institute* [1948], *Energy Information Administration* [2004], and *Fridley and Sinton* [2004]. The short-dashed lines shown in the figure bound the data for the United States and United Kingdom; we use an average for North America and Europe. Values of 1.2 and 0.39 ktonne/GWh correspond to overall thermal efficiencies of 11% and 33%, respectively. Fuel intensity in India and China is higher; the former has been adjusted to account for the lower heating value of its coal. We assume that efficiency improvements follow a similar path to that in Western countries ($s = 22$ years), but begin 17 years later, resulting in the long-dashed curve.

[22] Thermal electricity is generated with coal, residual oil, and natural gas. United Nations data for 1970–1985 show that the share of power generated by residual oil is approximately linear with the share in total consumption (auxiliary material Figure S2). This is also true of natural gas, except for a few countries that preferentially avoid natural gas for power generation. The fraction generated by coal is the remainder. These relationships were extrapolated backward in time for each country.

3.4. Shipping

[23] The United Nations database tabulates fuels used in international shipping ("bunker fuels") after 1950. *Darmstadter et al.* [1971] list coal and liquid bunker fuels for selected countries and years between 1925 and 1965. We interpolated fuel use for missing years and inferred it for some countries by scaling to numbers of steam and motor ships from *Mitchell* [1998a, 1998b, 1998c] if those were available, and to population if they were not.

[24] Fuel is also consumed for shipping within national boundaries, and this usage is not included in bunker fuel totals. We compared residual fuel used for internal navigation with that used in international shipping for 1970–1994. Countries differ; in general, countries with coastlines use comparatively more fuel for internal shipping. We applied

regionally specific ratios to bunker fuel estimates to estimate total fuel used by ships.

3.5. Railroads

[25] Railroads used large quantities of coal in the early 1900s; in 1930, they accounted for about 30% of coal consumption in the United States. Coal burned by railroads, especially for some of the largest countries, came from the works of *Hopwood* [1904], *Jevons* [1920], *McAuliffe* [1927], *Canada Dominion Bureau of Statistics* [1929], *Dominion Coal Board* [1949–1959], *Wright* [1984], and the same references used for electric utilities.

[26] Because limited data were available for most countries, we estimated railroad consumption using statistics on freight (in ton-kilometers) and passengers (in passenger-kilometers). Freight and passenger movements were multiplied by fuel intensity to obtain total fuel consumed. Finally, we estimated the transition from coal-fired steam engines to diesel engines by comparing the numbers of steam and diesel locomotives. Many of the ton-kilometer and passenger-kilometer figures came from the *League of Nations* [1926–1944], *International Union of Railways* [1957–1965], *Mitchell* [1998a, 1998b, 1998c], and the World Bank Railways Database (http://www.worldbank.org/transport/rail/rdb.htm, 2004). Data for individual countries on activity and locomotive types came from resources listed in auxiliary material.

[27] Fuel intensity for freight transport, shown in Figure 2b for locomotives in the United States and China, has improved greatly during the last century. During the early 20th century, railroads were working to improve efficiency by changing operations, such as by altering how coal was added to the firebox. These actions would affect emissions in addition to efficiency. Fuel intensity for passenger transport (not shown) also improved with time. There were some exceptions: United States passengers in the 1920s demanded more comfort, and intensity rose about 20%. We did not represent this increase, assuming that it was country-specific and temporary.

[28] Railways in different countries began the transition between coal and oil at very different times; for example, the change in the U.S. began around 1940 [*Association of American Railroads*, 2003], while India still had 70% steam locomotives in 1980. Regardless of the difference in transition time, the data are consistent with transition times of about 9 years. For all countries but China, which appeared to have a longer transition time of 12 years, we assumed a value of $s = 9$ years and inferred the value of $t0$ from the few observations of steam and diesel engines (see auxiliary material Table S1). We assume less than 100% coal in early years because some locomotives were powered by fuel oil or electricity.

3.6. Steel Industry

[29] The steel industry has been a major consumer of solid fuel for both blast furnaces and process heat, beginning before the industrial revolution. Volatile matter is eliminated from coal or wood to produce coke or charcoal for use in blast furnaces, which produce pig iron. Because coking has a different goal (driving off volatile matter) than other combustion processes, emission factors for coking are much different than for other thermal processes. While some countries provide data on coal usage in the steel industry, most do not. We used pig iron production to estimate coal used for coking as the product

$$pig\text{-}iron \times coke\text{-}per\text{-}pig\text{-}iron \times coal\text{-}per\text{-}coke. \quad (5)$$

[30] In 1850, the United Kingdom and the United States accounted for 55% and 14% of world production, respectively [*Bodsworth*, 2001; *Temin*, 1964]. For other European countries, we obtained data from *British Iron and Steel Federation* [1938] for 1866–1937, and from *American Metal Market* [1911–1998] for 1938 to 2000. The references overlapped between 1910 and 1937, and generally agreed within 2%. Data given by *Mitchell* [1998a, 1998b, 1998c] were used for other countries.

[31] Both coke and its biofuel analogue, charcoal, were used to manufacture pig iron, although by 1900 the fraction produced by charcoal had decreased to 1% [*Pounds and Parker*, 1957; *Temin*, 1964; *Bodsworth*, 2001]. We separated the fraction of pig iron produced with charcoal. By 1850, that fraction was close to zero in the United Kingdom [*Bodsworth*, 2001], but about 60% in France [*Pounds and Parker*, 1957]. In the United States before 1840, nearly all steel was from charcoal [*Swank*, 1892], but that fraction had decreased to 47% by 1854 [*Temin*, 1964]. Availability of forest resources played a large role in the choice between coal or wood. We assumed that Western Europe used charcoal similarly to France, and that the rest of the world, primarily Canada and Russia before 1909, used charcoal similar to the United States.

[32] The efficiency of pig iron production has also increased as practices improved. Producing one ton of pig iron required 3.5 tons of coke in 1830 and 0.53 tons in the midtwentieth century [*Gray*, 1969; *Gladman and Pickering*, 2001; *American Metal Market*, 1911–1998]. Likewise, charcoal needed to produce one ton of pig iron decreased from 1.8 tons in 1840 to 1.4 tons in 1880 [*Temin*, 1964; *Ayres and Simonis*, 1994]. Estimates of coke produced in the Soviet Union agreed with individual annual reports [*Shimkin*, 1965] within about 30%.

[33] The amount of coal needed to produce coke depended on the process, varying between 1.5 and 1.8 tons of coal per ton of coke [*British Iron and Steel Federation*, 1938]. For simplicity, we use a constant ratio of 1.67 recommended by *Gray* [1969]. Finally, *Ayres and Simonis* [1994] estimate the quantity of wood needed to produce charcoal.

3.7. Domestic Fuel

[34] Before the industrial era, fuel for domestic use was usually wood or other biofuels, except in a few locations. Coal sometimes replaced wood, and eventually electricity or cleaner fossil fuels such as natural gas, liquefied petroleum gas or middle distillate oil became the fuels of choice. This transition is still ongoing in some regions. Driving forces include cost, convenience (coal burns more steadily than wood), resource availability (including loss of forest cover), and government policies that favor specific fuels.

[35] To estimate domestic coal use, we represent transitions in per-capita consumption. Such data are available for the United States and Great Britain. In the United States, coal consumption decays slowly, beginning around 1915, with $s = 25$ years. The diffusion-type curve fits well except for a decrease of about 40% during the Depression in the 1930s; this "Depression factor" is applied to the consumption estimates. In Great Britain, on the other hand, the transition time is about 9 years. Although some cities had smokeless zones as early as 1939 [*Douglas et al.*, 2002], broad regulation of and monetary support for fuel switching occurred only after the United Kingdom's Clean Air Act of 1956 [*Scarrow*, 1972]. The transition in per-capita consumption began immediately after that time.

[36] When coal and biofuel substituted directly for each other, a single transition affects multiple fuels. Our approach to Europe and the former Soviet Union accounts for substitution, and is described by *Fernandes et al.* [2007]. In countries with coal resources, per-capita coal consumption usually rose after 1850, and dropped in the 1960s. Exceptions were Eastern European countries which implemented coal-fired central heating between 1960 and 1970 [*Shoup*, 1981] so that coal consumption remained high. We assumed that the transition to central heating, expressed in our technology splits, occurs at the same time, and we make the same assumption for the former USSR.

[37] Although domestic use of coal is now high in China, it began relatively late in comparison with other countries. *Wright* [1984] reports that the cost of coal precluded use in rural areas, except when people lived very close to mines or railways. He explains that most rural population growth in the early 1900s occurred in the south, away from coal-producing regions, although urban coal-using populations in the north grew rapidly. We assumed that coal consumption in China made up the domestic energy requirements not supplied by biofuel.

3.8. Diesel Vehicles

[38] To estimate consumption of diesel fuel by road vehicles, we developed a database of vehicles in each country, tabulating passenger cars, buses and coaches, goods vehicles and vans, and motorcycles. Vehicles which could have been diesel (trucks and buses) were the most important part of this tabulation. We obtained vehicle numbers from the *International Road Federation* [2003] and *League of Nations* [1926 to 1944]; we also used country-specific information, as listed in the auxiliary material. For missing years, data were scaled by population.

[39] We found average annual distance traveled for only a few countries, and there was very little data on fuel intensity [e.g., *Davis and Diegel*, 2004]. When these sparse values were combined to estimate consumption of diesel fuel by road vehicles, they overpredicted reported fuel consumption. Instead, we calculated diesel consumed per truck between 1970 and 1985 by comparing recorded diesel consumption with numbers of trucks. We extrapolated country-specific values to earlier years, using regional averages for countries that did not have data. In Africa, we assumed that all diesel fuel was used in vehicles or generators rather than in external combustion boilers [*Bond et al.*, 2004].

[40] The method we used accounts for both actual differences in fleet composition and reporting differences between countries. We recognize that the number of trucks is only a proxy for diesel-consuming activity, and may be affected by trucks with gasoline engines, or other diesel-consuming vehicles such as buses. Including buses in the total number of diesel-consuming vehicles did not reduce variation in per-vehicle consumption. Per-vehicle consumption did not show a statistically significant dependence on time. Data from the United States [*Schurr and Netschert*, 1960] and China [*Fridley and Sinton*, 2004] show that, over time, vehicles become more efficient and trucks carry progressively heavier loads; these changes offset each other.

[41] We did not find sufficient data on tractors or other off-road equipment to estimate off-road consumption. In countries that reported both on-road and agricultural consumption (farm machinery) between 1970 and 1985, the agricultural fuel use was about 25% of the on-road use. Other heavy equipment, such as construction, would also consume middle-distillate fuel. We therefore assumed that off-road transport was half of the on-road transport consumption.

3.9. Biofuel

[42] Previous work [e.g., *van Aardenne et al.*, 2001] has estimated historical emissions from biofuels by assuming constant per-capita consumption for rural population. The actual situation is somewhat more complex; per-capita usage changes as people gain access to more convenient fossil fuels, both for home consumption and for industrial applications, and as they can afford these alternatives. *Ito and Penner* [2005] estimated wood fuel consumption by considering trends in urban and rural population and in per-capita consumption. Their estimates, however, assumed that per-capita changes occurred at the same time as in the United States. We rely on new work which accounts for different transition times in individual countries, considering introduction of fossil fuels and forest depletion. These estimates are the subject of a companion paper [*Fernandes et al.*, 2007].

4. Emission Factor Updates

[43] Most of the emission characteristics we used, including particulate matter emission factor and BC fraction, were adopted from *Bond et al.* [2004]. Many of the technologies that contributed most to emissions in early years have fallen out of favor, and their emission factors were either not needed or were very uncertain in our earlier paper. Here we describe updates to the hard-coal emission factors for some of these important uses; Table 3 summarizes central values. We did not revisit emission factors for brown coal, which produces very little BC or OC regardless of combustion. The detailed study by *Goss* [1916] provides additional mass emission data.

4.1. Inferences From Opacity Data

[44] Many smoke observations were made in the 1900s using the Ringelmann scale [e.g., *Popplewell*, 1901], an

Table 3. Emission Characteristics for Coal Combustion Updated in the Present Work[a]

Technology	EF_{PM}, g/kg	$F_{1.0}$	F_{BC}	F_{OC}
Chain-grate stoker	4.7	0.2	0.2	0.05
Hand-fired stoker	12	0.2	0.2	0.05
Brick kiln	80	0.15	0.2	0.05
Rail locomotive	6	0.35	0.2	0.05
Ship	14	0.2	0.2	0.05
Coking ovens	25	0.5	0.3	0.6

[a]Limited data are available for F_{BC} and F_{OC}; see text for implications.

observer-based opacity measurement similar to those presently used for compliance measurements [*U.S. Environmental Protection Agency*, 1990]. These measurements are not well suited to determining BC or PM emission factors, because there is not a perfect correlation between opacity and mass [*Conner and Hodkinson*, 1972; *Uekoetter*, 2005]. However, we have not found any other measurements of coal stoker emissions, and we use this method to estimate PM emission factors. *Conner and Hodkinson* [1972] found that the Ringelmann number recorded by trained observers underpredicted extinction by about 10% on average. Opacity, or extinction, is related to the mass concentration of particulate matter [*PM*] through the Beer-Lambert Law:

$$-\frac{1}{L}\ln(opac) = [PM]\sum_i f_i(MAC_i + MSC_i), \qquad (6)$$

where *opac* is unitless, [*PM*] is in g m^{-3}, *L* is the visual path length across the stack, f_i is the fraction of mass in species i, and MAC_i and MSC_i are the mass-normalized absorption and scattering cross sections, respectively, for species *i* in m^2 g^{-1}.

[45] Mass emission factors are determined from the equation

$$EF = \frac{[PM]}{[C]}cfrac, \qquad (7)$$

where [*C*] is the concentration of all carbon in exhaust (kg/m^3) and *cfrac* is the fraction of carbon in the fuel; to calculate [*C*], we use reported carbon dioxide concentrations and temperatures from *Popplewell* [1901].

[46] In order to estimate emission factors, we first assume a fine (submicrometer) fraction, fine aerosol composition, MAC_i, and MSC_i. These estimates draw on those of *Bond et al.* [2004], *Bond and Bergstrom* [2006], *U.S. Environmental Protection Agency* [2001] and *Wyatt* [1980], and are described in the auxiliary material. We assume that only black carbon absorbs light; absorption by organic carbon is small at observed visible wavelengths [e.g., *Kirchstetter et al.*, 2004]. Next, we use the data of *Goss* [1916], who gave both average smoke opacity and particulate matter emission factors for different sources in Chicago. We estimate *L* and iterate the submicrometer fraction until opacity values match the mass emission factors. Finally, we apply the composition determined to observations which provide opacity (*opac*) and chimney width (*L*), to obtain a greater number of observations.

[47] Because BC has the highest extinction per mass, the assumed fraction has a large effect on the estimated emission of mass. We used the estimates from *Bond et al.* [2004] for fine particulate matter. There is little information on the chemical composition of emissions for early combustors. *Ashworth* [1933] found that particles deposited in a factory town were about 26% carbonaceous, while *Ge et al.* [2001] reported 20%. Values around 60% BC have been measured in a laboratory furnace [*Sarofim and Lighty*, 2003]. We increased the BC fraction of fine aerosol to 60% and propagated the result through our procedure. The PM emission factor increased by 20%, and the BC emission factor increased by 40%. This factor is included in our uncertainties. If a time-averaged value of opacity is used when chemical composition is changing, equation (6) also introduces a reciprocal-averaging bias. We accounted for this bias when possible, and included it in our uncertainty.

4.2. Coal Stokers

[48] For stokers, our present-day PM emission factors were based on *U.S. Environmental Protection Agency* [2001] data (3.1−6.6 g/kg for uncontrolled stokers). Figure 3 shows emission factors for two types of stokers determined with the method described in the previous section, using data from the *University of Pittsburgh* [1912]. Hand-fired stokers show the greatest variability. They average 12 ± 12 g PM/kg fuel, similar to the

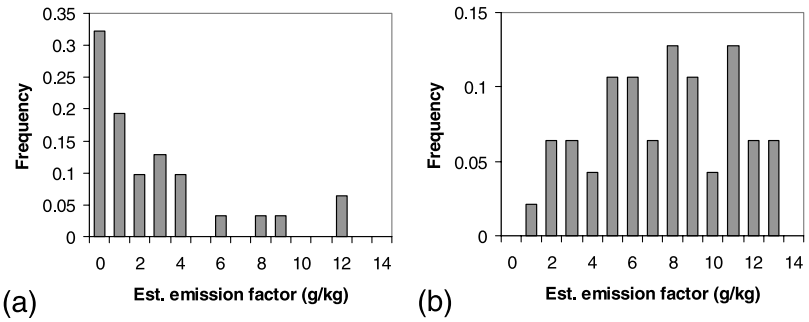

Figure 3. Total particulate matter emission factors for (a) chain-grate and (b) hand-fired stokers, derived from opacity and operation data as discussed in section 4.1. For chain-grate stokers, N = 36, and for hand-fired stokers, N = 61.

Hangebrauck et al. [1964] average of 9.7 g/kg. Observations for chain-grate stokers are similar to present-day values, at 2.7 ± 3.9 g/kg. Smoke observations reported by *Randall and Weeks* [1909] translate to even lower emission factors, averaging 1.0 ± 1.4 g/kg. Although this data set is extensive, it appears inconsistent with other measured values. We retained the values for stokers from *Bond et al.* [2004] and added hand-fired stokers in earlier years.

[49] Although there are significant uncertainties in this approach, reviewing opacity data has not caused a large revision in stoker emission factors. If anything, this investigation has suggested that emission factors might need to be decreased, with the exception of those for hand-fired stokers. It calls into question high values used in other inventories. For example, the black carbon emission factor of 1.8 g/kg used by *Novakov et al.* [2003] would have resulted in continuous 50% opacity for a 1-m stack, even if no other particles were emitted. This is about 10 times greater than observations.

[50] We account for only a few of the factors that affect emissions from coal boilers. Emission rates depend partly on coal composition, but systematic studies gave no clear relationship between emission factor and coal volatile content [*Higgins*, 1941]. Coal smoke has long been considered a nuisance, but there are limited data on measures taken against it. Control of coal smoke in London was attempted as early as the 13th century [*Brimblecombe*, 1987]. Pittsburgh, USA took action to improve over 400 boilers early in the 1900s [*City of Pittsburgh*, 1915]. About 30% of the measures in Pittsburgh would have been captured by energy statistics, because they involved fuel-switching. Another 30% included changes in firing practice, which could decrease smoke by 50–75% [*Higgins*, 1941]. However, this method was not considered a reliable mitigation measure because it depended on individual operator behavior [*Holmes*, 1908].

[51] There are also anecdotal reports of high-emitting uses. For example, the earliest high-pressure steam engines were very smoky [*Brimblecombe*, 1987]. *Nicholson* [1927] reports that brick kilns produced "dense black smoke" for 3–4 hours after lighting. According to *Goss* [1916], such kilns had very high emission factors. We increased the value of EF_{PM} for these kilns to 80 g/kg, but also decreased the submicrometer fraction (0.15) for consistency with opacity values.

4.3. Rail Locomotives

[52] Like those of other mobile sources, emission factors for coal-fired railroad locomotives depend on operation. The highest-emitting conditions were thought to be immediately after lighting the firebox and climbing hills [*Poethke*, 1909; *Skeat*, 1950]. Hill-climbing or accelerating locomotives are often featured in photographs, because the belching smoke was viewed positively as a sign of industrial progress. Such photographs would overestimate emission factors. Observations of over 15,000 locomotives [*Deutch and Radner*, 1941] suggest an emission factor of about 6 g/kg, using the opacity method described above. *Deutch and Radner* [1941] also indicate that emission factors decreased by 50% between 1910 and 1939, which is consistent with earlier, higher data given by *Goss* [1916]. We applied this change in the United States and Europe with a transition time of s = 10 years.

4.4. Coking Ovens

[53] *Bond et al.* [2004] acknowledged a lack of measured data for coke ovens where offgas is not captured. Their assumptions gave emission factors of 4.8 and 3.4 g/kg for BC and OC under these conditions. We updated emission factors and BC and OC ratios on the basis of work by *Lunge* [1909], *Weitkamp et al.* [2005] and information from China (Lei Yu, personal communication, 2006). Net BC and OC emission factors for uncaptured ovens are now 3.8 and 6.2 g/kg coal, respectively. The auxiliary material contains more extensive discussion.

5. Technology Splits

[54] We have relied on fuel consumption by sector to account for most of the changes in technology. For example, the power-generation sector is dominated by pulverized coal plants, while industrial boilers are often smaller and more likely to be stokers. Technology shifts within sectors did occur as new options became available. Our general procedure for estimating these changes is: (1) assess characteristic transition times and starting years from countries where data are available (frequently the United States); (2) assume that transition times, but not starting years or final technology splits, are similar between regions; (3) estimate different starting years ($t0$) for all regions on the basis of the implementation of regulations; and (4) estimate final splits on the basis of present-day technology in the region. Within each region, we assume that the present-day situation represents the final splits only if sufficient time has elapsed since $t0$.

5.1. Coal Boilers: Pulverized Coal and Stokers

[55] Data on firing and control technology for coal burned in United States industrial sector were given by *Moore* [1966], *U.S. Environmental Protection Agency* [1979], and the boiler database developed by the U.S. Environmental Protection Agency (http://www.epa.gov/ttn/atw/combust/iccrarch/bo.html, known as ICCR or Industrial Combustion Coordinated Rulemaking). Early boilers were stokers; some pulverized coal burners came into use when the technology became available in the 1930s. Another transition occurred around the time of national regulations requiring emission controls, such as the Clean Air Act in the United States. We used the onset of national regulation in each region (see auxiliary material Table S3) to fix the beginning of this second transition in some countries. More details can be found in the auxiliary material.

[56] Auxiliary material Table S1 provides illustrative parameters for transitions in North America, the former USSR, and India. To obtain the fraction of coal burned using a particular firing and control technology, the division for firing technology (e.g., pulverized coal) is multiplied by the division for emission control technology (e.g., cyclone). This procedure results in the firing and control combinations discussed by *Bond et al.* [2004].

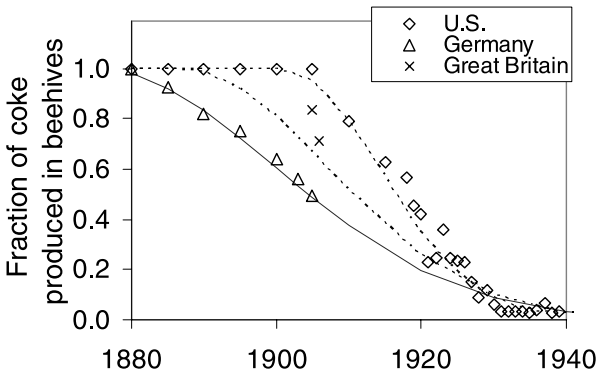

Figure 4. Fraction of coke produced in beehives.

[57] We treated the power-generation sector similarly, except that it was more dominated by pulverized coal combustion. These facilities provided 10–20% of the electricity in Great Britain around 1930 [*Rosins*, 1947]. By 1971, they accounted for nearly 90% of power generation in the United States, and the fraction continued to grow [*Vandergrift et al.*, 1971] also U.S. Environmental Protection Agency ICCR database). The Energy Information Administration (Annual steam-electric plant operation and design data 2002, Form EIA-767 database, http://www.eia.doe.gov/cneaf/electricity/page/eia767.html) provided a time trend for end-of-pipe control devices in the United States between 1950 and 2000. We assumed that this trend applied to industrialized countries. As with industrial boilers, we used the start of regulation to indicate introduction of some control devices. Lacking better information, we assumed that the fraction of industrial coal used in brick kilns was the same as present day. We also assumed that chain-grate or other, somewhat cleaner stokers were used in the industrial sector, and hand-fired boilers were used in the residential sector.

5.2. Iron and Steel Industry

[58] The major transition in coking has been capturing and using the volatile matter driven off. No volatile matter was captured in the early days of coking, when the ovens were called "beehives" owing to their shape. Capture of byproducts in recovery ovens, particularly in Germany, began in the late 1800s in Europe [*Lunge*, 1909]. The United States began recovery of byproducts around 1910 [*Tarr*, 1996], although new beehives were constructed until 1931 [*Kiessling*, 1934]. Figure 4 shows changes in beehive fractions for three major coke producers in the early 1900s (see also auxiliary material Table S1), using data from *Lunge* [1909], *Tarr* [1996], *Warren* [2001], and *Bituminous Coal Institute* [1948–1951]. Along with France, these countries accounted for 83% of coke consumption in 1900 and 66% in 1950 (see auxiliary material Figure S3).

[59] Germany, where much of the recovery technology was developed, moved to byproduct recovery earlier than the United States and Great Britain, which had longer histories of coke production. We used the transition midway between Great Britain and Germany for the rest of Europe, reasoning that neighboring European countries might obtain advanced technology from Germany, and that they did not have extensive capital investment in old technologies. We found no data regarding coke production in the former USSR, and also used the intermediate transition there. The assumption that the former USSR adopted recovery technology during its period of rapid growth in the 1950s could be a major uncertainty.

[60] *Fridley and Sinton* [2004] give the fraction of beehive coke for China from 1949 to 1996. This fraction was applied directly for that country. The data do not follow a smooth trend, in particular showing a sharp peak during the Great Leap Forward. Because technology was driven by changing government policies, we did not use the beehive fractions in China for any other country. However, the China data demonstrate that recovery technology was used worldwide as early as 1950, and we assume that recovery began then in the rest of the world.

5.3. Biofuel

[61] Large differences in biofuel emissions are caused by the combustor type, for example open fires versus fireplaces. *Ito and Penner* [2005] apportion biofuel between cooking and heating to account for this emission factor difference, and this method would be appropriate if all cooking had occurred in stoves that are similar to today's developing-country cookstoves. However, in North America, many cooking stoves were similar to today's heating stoves, metal combustion chambers designed to accept large pieces of wood. This practice is not representative of countries where fuel is scarce and large wood is frequently not available.

[62] Lacking better information, we used the technology splits from *Bond et al.* [2004] throughout the entire period. We reasoned that the predominant wood size and burning practice within a region has been relatively constant during the past 150 years; North Americans did not begin cooking on small, careful fires, nor did Chinese build large fireplaces as their consumption increased. One exception is improved cookstoves, which were introduced around 1980.

5.4. Vehicles

[63] As regulations are introduced, vehicle technology becomes better and emissions decrease. This improvement may appear as a trend with vehicle model year, because newer vehicles are designed to meet regulations. Not all emission reductions are directly driven by regulations. Figure 5 summarizes dynamometer test data for heavy-duty vehicles in the United States, assembled by *Yanowitz et al.* [2000]. The figure also shows a diffusion-curve fit with $s = 9$ years and $t0 = 1972$. Particulate matter from heavy-duty vehicles, however, was not regulated until around the late 1980s [*Faiz et al.*, 1996]. The earlier onset of clean technology could have been driven by regulations that targeted efficiency, other pollutants, or other vehicles. We therefore assumed that improvements began when vehicle regulations were introduced, even if they did not target particulate matter. *Faiz et al.* [1996] discussed the introduction of regulations in different countries.

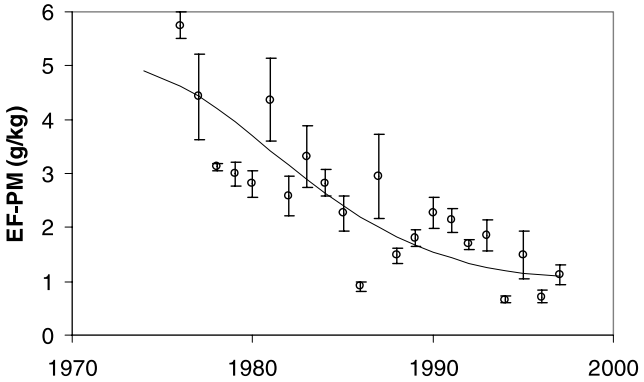

Figure 5. Diesel emission factors from *Yanowitz et al.* [2000]. Not shown is a single high test from a 1974 vehicle. Error bars are standard deviation of dynamometer tests divided by $\sqrt{N}$. Dashed curve represents best fit.

[64] Regulations affect only new vehicles, and the fleet-averaged emission factor responds more slowly than the new-vehicle emission factor. A simple dynamic model [*Streets et al.*, 2004] showed that the fleet-average value lags introduction by about four years (see auxiliary material Table S1 for parameters). Superemitting vehicles were assumed to result from lack of maintenance and to occur at the same rates given by *Bond et al.* [2004] for all years.

6. Uncertainties

[65] Uncertainties in activity data, technology splits, and emission factors were propagated to determine uncertainties in total emissions as described by *Bond et al.* [2004]. For activity data, we estimated uncertainties on the basis of the indicator used. For example, an IEA record of coal used to generate power would be assigned an uncertainty of 5%, while the calculation based on electric consumption and energy intensity had an uncertainty of 20%. This meant that relative uncertainties decreased from the beginning to the end of the period, as better reporting became available.

[66] Uncertainties in technology splits were also estimated individually. The low-emission case was calculated by assuming relatively lower shares of high-emission technologies.

7. Results

7.1. Emission Trends

[67] Black and organic carbon aerosol emitted from different fuel types are shown in the upper portion Figure 6. Emissions of black carbon increase almost linearly, totaling about 1000 Gg in 1850, 2200 Gg in 1900, 3000 Gg in 1950, and 4400 Gg in 2000. The slower growth between 1900 and 1950 is partly due to economic circumstances, but also results from implementing cleaner technology. Primary organic carbon shows a similar pattern of linear growth that is slightly slower in the mid-1900s. Emissions are 4100 Gg, 5800 Gg, 6700 Gg, and 8700 Gg in 1850, 1900, 1950, and 2000 respectively. Present-day emissions are slightly smaller than those in our earlier paper [*Bond et al.*, 2004] because we use a new biofuel inventory [*Fernandes et al.*, 2007].

[68] These estimates show that black carbon has been dominated by emissions from coal since early in the industrial revolution; coal emissions overtook those from biofuel after 1880. BC from biofuel remained relatively constant until about 1960, because replacement of some of the biofuel with fossil fuels offset population growth. After that time, population became the determining factor and emissions began to increase. For BC, emissions from biofuel again became greater than coal emissions around

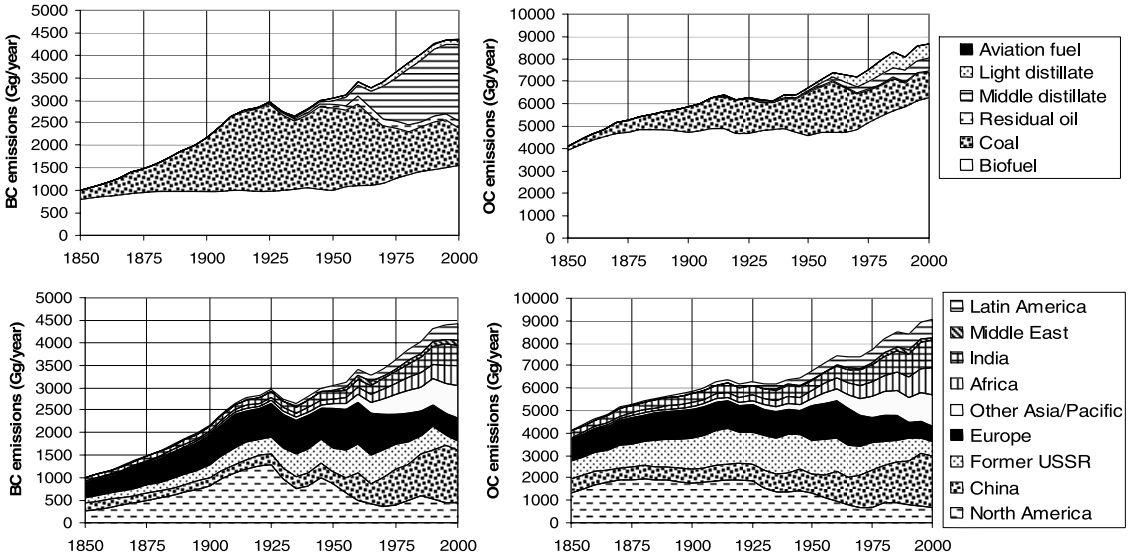

Figure 6. Emissions of (left) black carbon and (right) organic carbon. Emissions are segregated by (top) fuel and (bottom) region.

Table 4. Causes of Changes in Coal Emissions Between 1950 and 2000[a]

	1950 Emission, Gg	2000 Emission, Gg	Change		Consumption Increase, %	Effect of Sectoral Shift, %	Emission Improvement	
			Absolute, Gg	Percent			Industry, %	Power, %
North America	680	30	−650	−96	110	−96	−28	−47
Europe	900	46	−850	−95	19	−82	−12	−24
Former USSR	320	36	−280	−89	53	−66	−19	−7
China	26	690	670	2600	3100	−23	12	−1
India	28	55	27	94	1100	−76	−110	−5
Other Asia/Pacific	13	90	77	600	610	−40	55	−4

[a]"Effect of sectoral shift" indicates the difference from expected emissions if emission factors and sectoral percentages remained constant. "Emission improvement" refers to change in all coal emissions, not just the indicated sectors.

1980. Around 1985, diesel emissions took the lead as the largest worldwide contributor.

[69] While coal use has increased nearly continuously, emissions per unit of coal have improved greatly. To a large extent, this progress has occurred because of shifts to sectors that are cleaner by nature. For example, in North America in 1950, about 20% of the coal was used for power generation, and 16% was used in the residential sector. In 2000, those percentages were 90% and less than 1%, respectively. Despite uncertainty in technology splits within each sector, the higher emission intensity of the residential sector is indisputable. Because each installation is small, it is difficult to employ advanced technology such as pulverized-coal boilers or electrostatic precipitators. Another reason for the decline in emission intensity for both coal and diesel fuel is the implementation of cleaner technology. This change is also important, although it is less so than the sectoral shifts. Table 4 summarizes the causes of transformation in emissions from coal.

[70] Emissions of organic carbon appear even more constant than those of BC until about 1950. Biofuel produces the greatest fraction of emissions by far, and accounts for the increase in OC after 1950. Total fossil-fuel emission has remained relatively constant; decreases in emissions from coal offset those from liquid fuels.

[71] Figure 6 also shows our estimates divided by region. Our work agrees with previous findings [e.g., *Novakov et al.*, 2003] that North America and Europe dominated black carbon emissions in the early industrial era. The former USSR was also a contributor, but emissions declined in the late twentieth century along with the economy of that region. Meanwhile, emissions from China and other Asian countries grew rapidly in the latter half of the twentieth century.

7.2. Comparison With Previous Work

[72] Figure 7 compares our estimates with those from previous work. *Ito and Penner* [2005] show a similar trend

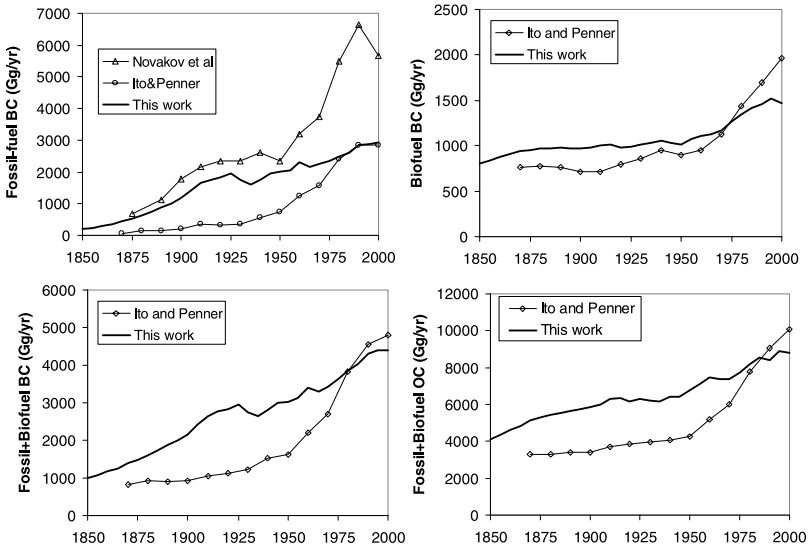

Figure 7. Comparison with previous studies. (top) BC emissions from fossil fuel and biofuel separately; *Novakov et al.* [2003] estimated fossil-fuel emissions only. (bottom) Combined fossil-fuel and biofuel emissions for both BC and OC and are directly comparable with Figure 6. Data from *Novakov et al.* [2003] and *Ito and Penner* [2005] were read every 10 years, and do not capture annual fluctuations. Primary organic matter given by *Ito and Penner* was divided by 1.3 to obtain OC for comparison.

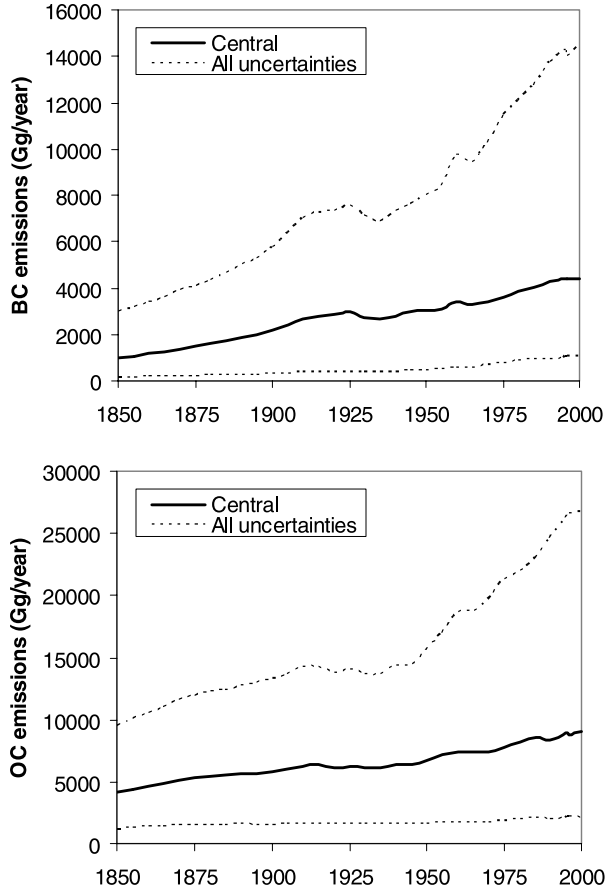

Figure 8. Uncertainties in black and organic carbon emissions.

and magnitude in BC from biofuel, but the magnitude of their fossil-fuel emissions is far lower. In the late twentieth century, our biofuel emissions increase less (30% between 1960 and 2000, compared to 100% for *Ito and Penner* [2005]). *Novakov et al.* [2003] estimate slightly higher fossil-fuel emissions than our work in the early 1900s. However, their work estimated total black carbon aerosol; if they had estimated submicron fraction only, their 1900–1940 estimates would have been lower than ours by about one third. The *Novakov et al.* [2003] estimates, like ours, have flat emissions between about 1910 and 1950, although our estimate is 15% lower.

[73] The most apparent difference between the present work and earlier estimates is the more gradual transition in the latter half of the twentieth century. Our data suggest a smaller increase after 1950, a factor of less than two, as opposed to a factor of three for the other studies. Both *Novakov et al.* [2003] and *Ito and Penner* [2005] considered the introduction of cleaner diesels and some changes in sectoral divisions. We model shifts to cleaner burning, through increases in consumption in cleaner sectors. Our study represents the shift to cleaner burning of coal explicitly for the first time. The difference between the three studies is largely attributable to choice of emission factors, which implicitly represents some assumptions about technology choice. Because our emission factors are similar to those of *Ito and Penner* [2005], it is unsurprising that our estimates in the late twentieth century are comparable to theirs. They admit that their estimates for the early 1900s could be too low, because they did not consider changes in combustion except for diesels. Our fossil-fuel emissions are comparable to those of *Novakov et al.* [2003] before 1950, suggesting that the average emission factor chosen for developed countries was similar between the two studies. (Because our average emission factor includes beehive coke, it might not be appropriate for other times or locations.) *Novakov et al.* [2003] used the emission factors of *Cooke et al.* [1999], which assumed that emission factors in developing countries were five times higher than those in developed countries, without regard to technology. While we do represent the use of less-efficient burning in developing countries, we also allow for use of cleaner facilities. Combined with the decrease in emission intensity in developed countries, this ameliorates the transition in the latter half of the twentieth century.

[74] Like the BC trend, the increase in OC between 1950 and 2000 is more gradual than that predicted by *Ito and Penner* [2005]. (*Novakov et al.* [2003] did not discuss OC emissions.) For most of the period, our OC emissions are higher than those of *Ito and Penner* [2005], probably owing to greater estimates of biofuel consumption [*Fernandes et al.*, 2007]. OC emissions from biofuel depend greatly on assumptions about wood-burning technology, and could appear different if there were a significant shift in burning type.

[75] Finally, Figure 8 shows our calculated uncertainties. For black carbon, the high-emission case exhibits a rapid rise in the latter half of the twentieth century, similar to other estimates [*Novakov et al.*, 2003; *Ito and Penner*, 2005]. This is partly due to our choice of technology splits; the rapid rise would occur if low-emitting technologies were adopted less than we estimate.

[76] Emission estimates by both *Novakov et al.* [2003] and *Ito and Penner* [2005] lie within our large uncertainties. However, some of the uncertainties result from lack of knowledge of emission factors or properties; uncertainties in the present-day inventory were about a factor of two [*Bond et al.*, 2004]. Uncertainty in the rate of change may itself be important for climate-change attribution studies, and is governed by changes in fuel consumption and in technology. Without a detailed analysis, which is beyond the scope of this work, it is not possible to determine whether earlier estimates of the rates of change lie within the uncertainties presented here.

[77] Emissions were gridded as described by *Bond et al.* [2004]. Figure 9 shows results for 1850, 1900 and 1950; others are available from the authors. *Klein Goldewijk* [2005] produced temporally varying distributions of urban and rural population, which consider changes within countries due to migration. We used these data to distribute emissions.

7.3. Caveats

[78] In addition to the significant uncertainties associated with historical reconstruction, the methods we use do not

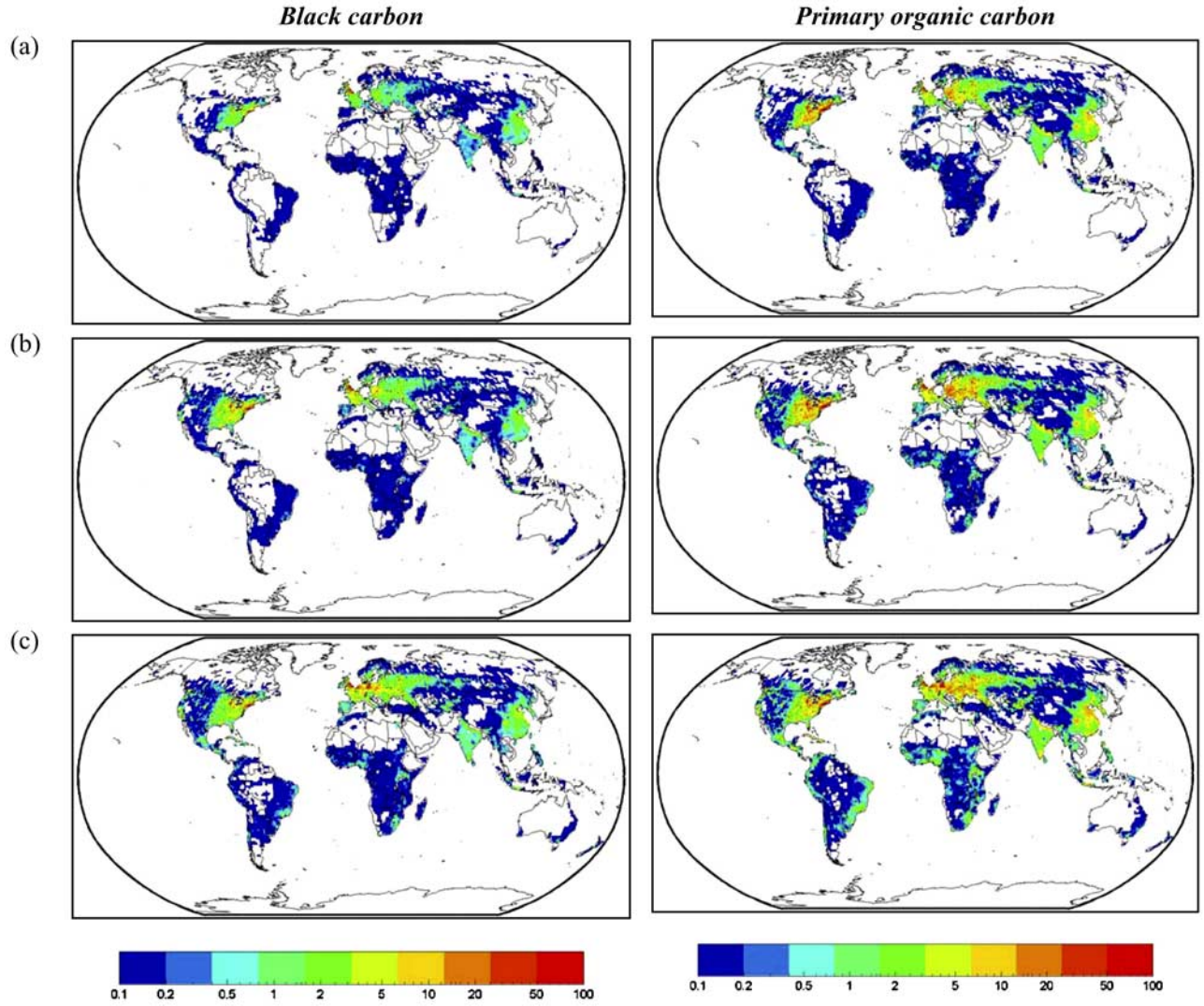

Figure 9. Spatially distributed emissions of black and primary organic carbon from fossil-fuel and biofuel combustion in (a) 1850, (b) 1900, and (c) 1950 (units: $ng/m^2/s$).

account for rapid transitions. Long-term growth is often punctuated by shocks such as wars or depressions. During the period covered by this inventory, major upheavals took place in important emitting regions. For example, after World War I, the Austro-Hungarian empire was divided, coal fields were reallocated, and the country presently known as Austria underwent a major change in energy consumption. We could not account for these changes in any but the broadest sense.

[79] The assumption of similar transition times between regions, which we used heavily to determine technology transitions, is questionable for at least two reasons. First, most of our transition times are determined from United States or European data. In these regions, technology is often developed to meet standards; this situation is known as *technology forcing* [*Faiz et al.*, 1996]. After new technologies are developed, they are available for other regions to adopt, so that standards are *technology following*. The latter transition may occur more quickly [*Marcotullio et al.*, 2005], depending on the region. Transition times may also depend on enforceability of regulations. If penalties for emission are light, then fewer people may adopt clean but expensive technology. These two effects are of opposing magnitude and may partially counteract each other. In addition, air quality regulations are only now being implemented in many world regions, so that our assumptions about how technology responds to regulation affect only the last few years of emission history.

[80] To produce an inventory at global scale, we have had to extrapolate a general characterization of technology and emissions, such as the industrial boiler mix, to several countries and regions. Detailed, relevant data are probably available at national level, but the scope of this project did not allow examination at this level. As countries develop

their own historical estimates, the understanding of global historical emissions should improve, and we encourage efforts to share this information. In particular, information from regions such as China and the former Soviet Union, which are important contributors to total emissions, may be available in the native languages of these countries. More accurate global emissions will require global participation.

[81] Finally, comparison with atmospheric measurements has suggested that the present-day inventory of *Bond et al.* [2004] is underestimated in some regions [*Park et al.*, 2005], or that the spatial distribution of emissions is incorrect [*Hakami et al.*, 2005]. These findings have identified the need to revisit technology mixes and emission factors. We acknowledge that these factors could produce a low bias in this historical inventory as well, and we encourage emission observations which provide greater accuracy. However, we have resisted implementing arbitrary emission increases which would produce a better match with atmospheric concentrations. Until the causal factors of these mismatches are identified, as for example by *Streets et al.* [2006], we cannot confidently represent the relationship between socioeconomic changes and emissions.

8. Summary

[82] We have calculated emission trends of black and primary organic carbon emissions between 1850 and 2000. This work relies on a sectoral, fuel-specific reconstruction of fossil fuel consumption and on new historical estimates of biofuel consumption. Because we account for technology transitions, largely through sectoral divisions, our trends are quite different than those found in previous work, especially during the latter half of the twentieth century.

[83] Biofuel is responsible for over half of BC emission until about 1890, and dominates energy-related primary OC emission throughout the entire period. Coal contributes the greatest fraction of BC emission between 1880 and 1975; although its usage continues to increase, cleaner technology results in overall emission reductions. BC emissions from coal are overtaken by biofuel around 1975, and by diesel fuel around 1990; these latter two sectors represent future challenges in reducing BC emissions. Technology improvements gradually counteract the sharp rise in fuel consumption in the latter half of the 20th century. Previous work suggests a rapid rise in BC emissions between 1950 and 2000; this work supports a more gradual, smooth increase between 1950 and 2000.

[84] **Acknowledgments.** The team at UIUC was supported by the Climate Dynamics and Atmospheric Chemistry Programs at the National Science Foundation under grant ATM-0349292. The Argonne team is funded by the U.S. Department of Energy, Office of Fossil Energy, Office of Planning and Environmental Analysis. Argonne National Laboratory is operated by the University of Chicago under contract W-31-109-ENG-38 with the U.S. Department of Energy. We thank Joel Darmstadter for gracious responses to questions on his valuable resource book, and James Hansen, Dorothy Koch, and V. Ramanathan for their encouragement.

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