Historical emissions of carbonaceous aerosols from biomass and fossil fuel burning for the period 1870–2000

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[1] Historical changes of black carbon (BC) and particulate organic matter (POM) emissions from biomass burning (BB) and fossil fuel (FF) burning are estimated from 1870 to 2000. A bottom-up inventory for open vegetation (OV) burning is scaled by a top-down estimate for the year 2000. Monthly and interannual variations are derived over the time period from 1979 to 2000 based on the TOMS satellite aerosol index (AI) and this global map. Prior to 1979, emissions are scaled to a CH_4 emissions inventory based on land-use change. Biofuel (BF) emissions from a recent inventory for developing countries are scaled forward and backward in time using population statistics and crop production statistics. In developed countries, wood consumption data together with emission factors for cooking and heating practices are used for biofuel estimates. For fossil fuel use, we use fuel consumption data and specific emission factors for different fuel use categories to develop an inventory over 1950–2000, and emissions are scaled to a CO_2 inventory prior to that time. Technology changes for emissions from the diesel transport sector are included. During the last decade of this time period, the BC and POM emissions from biomass burning (i.e., OV + BF) contribute a significant amount to the primary sources of BC and POM and are larger than those from FF. Thus 59% of the NH BC emissions and 90% of the NH POM emissions are from BB in 2000. Fossil fuel consumption technologies are needed prior to 1990 in order to improve estimates of fossil fuel emissions during the twentieth century. These results suggest that the aerosol emissions from biomass burning need to be represented realistically in climate change assessments. The estimated emissions are available on a $1^\circ \times 1^\circ$ grid for global climate modeling studies of climate changes.

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1. Introduction

[2] Aerosol emission changes may have contributed to global temperature changes in the past century [*Novakov et al.*, 2003]. Through their direct effects, some aerosols (sulfate, organics, nitrates) cause a net cooling of the Earths' surface, while others (e.g., black carbon (BC) and dust) may cause a net warming with changes in the vertical temperature profile and the hydrologic cycle of particular concern [*Ramanathan et al.*, 2001; *Menon et al.*, 2002; *Penner et al.*, 2003]. Carbonaceous aerosols are composed of BC and particulate organic matter (POM) and are especially important in determining the amount of warming since BC is the primary aerosol component that absorbs short wave radiation, while POM has light-scattering properties similar to sulfate aerosols [*Sloane*, 1983; *Liousse et al.*, 1995, 1996; *Cachier*, 1998]. These aerosols are emitted into the atmosphere through incomplete combustion of biomass and fossil fuels. Thus the amounts of BC and POM depend on the combustion efficiency of the fuel utilization as well as the fuel amount [*Novakov and Hansen*, 2004].

[3] Accurate estimates of historical emissions are hampered by high uncertainties in biomass burning emissions [*van Aardenne et al.*, 2001] (see also A. Ito and J. E. Penner, Estimates of CO emissions from open biomass burning in Southern Africa for the year 2000, submitted to *Journal of Geophysical Research*, 2005) (hereinafter referred to as Ito and Penner, submitted manuscript, 2005) and significant interannual variability [*Langenfelds et al.*, 2002; *van der Werf et al.*, 2004]. *Schultz* [2002], *Duncan et al.* [2003], and *Generoso et al.* [2003] used fire products from the Along Track Scanning Radiometer (ATSR) and the Advanced Very High Resolution Radiometer (AVHRR) satellites to analyze spatial and temporal variations in emissions from biomass burning. However, studies have shown that active fire products do not represent an unbiased sample of fire activity [*Eva and Lambin*, 1998; *Boles and Verbyla*, 2000; *Kasischke et al.*, 2003]. *Duncan et al.* [2003] also made use of the Total Ozone Mapping Spectrometer (TOMS)

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Aerosol Index (AI) data product as a surrogate to estimate interannual variability in biomass burning. The TOMS AI has also been used to constrain the emissions from biomass burning, using an inverse model in conjunction with a chemical transport model (S. Zhang et al., Inverse modeling of biomass burning emissions using Toms AI for 1997, submitted to *Journal of Geophysical Research*, 2004) (hereinafter referred to as Zhang et al., submitted manuscript, 2004).

[4] This paper presents estimates of the historical changes of BC and POM emissions from open vegetation (OV), biofuel (BF), and fossil fuel (FF) burning. We estimate the direct primary emissions of fine particles (those with diameters below 1.0 or 2.5 μm). Secondary organic aerosols (SOAs), which are formed from certain volatile organic compounds (VOCs) in the atmosphere, are not included. Our estimates in each case are based on present-day emissions estimates from *Ito and Penner* [2004] and *Arellano et al.* [2004] for OV, the *Food and Agricultural Organization* [2004] statistical database (hereinafter referred to as FAOSTAT) for BF in developed countries, *Yevich and Logan* [2003] for BF in developing countries, and *Bond et al.* [2004] for FF and are extrapolated back in time using available data. These data include both satellite data from the TOMS instrument [*Herman et al.*, 1997; *Torres et al.*, 1998] and the anthropogenic CH_4 emissions from *Stern and Kaufmann* [1996] for OV emissions, wood consumption, population statistics, and crop production data from the FAOSTAT, *U.S. Energy Information Administration* [2004] (hereinafter referred to as EIA), and the *United Nations* (*U.N.*) [1973] for BF emissions, and *U.N.* [1976, 1984] and *International Energy Agency* [2002a, 2002b] (hereinafter referred to as IEA) data for fossil fuel consumption together with a previous inventory for CO_2 emissions [*Andres et al.*, 1999] for fossil fuel emissions. We also show comparisons of our results with other estimates.

2. Biomass Burning Emissions

[5] Biomass burning consists of open vegetation fires and biomass used for fuels (biofuels). Below, we describe how we estimated the present-day and historical emissions for each of these components.

2.1. Open Vegetation Burning

[6] To develop our historical inventory for open vegetation burning, we first focused on developing an inventory of the total biomass burned for the year 2000 which was based on the regional estimates of *Arellano et al.* [2004] and the $1^\circ \times 1^\circ$ spatially differentiated global map of open biomass burning developed by *Ito and Penner* [2004]. *Arellano et al.* [2004] estimated the emissions of CO in 2000 from open biomass burning using a chemical transport model and measurements of CO mixing ratios derived from the Measurements of Pollution in the Troposphere (MOPITT) instrument. This top-down estimate of CO emissions for the sum of July and September 2000 was in good agreement with the best estimate from regional bottom-up approaches in southern Africa if the Moderate Resolution Imaging Spectroradiometer (MODIS) burned area data set [*Roy et al.*, 2002] was used (Ito and Penner, submitted manuscript, 2005). Because *Arellano et al.* [2004] only derived the annual emissions for the year 2000 for eight different regions, a monthly and spatially resolved emission map for 2000 was derived using the regionally constrained annual results from the *Arellano et al.* [2004] estimates and the spatially resolved monthly emissions of *Ito and Penner* [2004]. Thus we calculated the emissions for BC and POM, $E_{s,t}$, for each region, s, and month, t, by scaling the regional CO emissions, C_s, from *Arellano et al.* [2004] to the BC and POM emissions, $A_{s,t}$, from *Ito and Penner* [2004],

$$[E_{s,t}] = [A_{s,t}] \times \left([C_s]/[C'_s]\right), \quad (1)$$

where C'_s are the total annual regional CO emissions from *Ito and Penner* [2004]. The BC and POM emissions were calculated from the emissions of particulate matter less than 2.5 μm in diameter ($PM_{2.5}$) developed by *Ito and Penner* [2004] and the emission factors of BC from *Andreae and Merlet* [2001] for each land cover type. POM is calculated from $PM_{2.5} - BC$.

[7] Scaling factors ($([C_s]/[C'_s])$ in equation (1)) are shown in Table 1. The scaling factors in central and northern South America, southern South America, and Asia are very large, which may reflect underestimates of the burned areas in those regions [*Ito and Penner*, 2004]. Underestimates are expected partly because the remote sensing instruments on satellites cannot detect small burn scars that are below the instrument pixel resolution [*Grégoire et al.*, 2003]. In addition, burn scars are not detected in forested areas with understory fires because the surface albedo of the forested scene does not necessarily change [*Simon et al.*, 2004]. These underestimates may be detected and corrected using inverse modeling, but there are potential errors in the derived emissions from inverse modeling as well.

[8] For example, errors in inverse modeling may derive from the use of poorly constrained meteorology in different chemical transport models. The study by *Arellano et al.* [2004] was based on data for a limited period of time from April to December 2000, and so may produce errors in regions where substantial open biomass burning occurs from January to March (e.g., northern Africa). On the other hand, *Pétron et al.* [2004] used an inverse method to derive monthly average CO emissions for an entire year of MOPPIT data from April 2000 to March 2001. Although the time period for these two studies differs, a comparison between these two estimates may provide us with an estimate of the potential magnitudes of the uncertainties in inverse model studies. The annual a posteriori open biomass burning emission from *Arellano et al.* [2004] is 26% larger than that from *Pétron et al.* [2004] (i.e., 552 Tg CO yr^{-1} from *Arellano et al.* [2004] versus 408 Tg CO yr^{-1} from *Pétron et al.* [2004]). The disagreement may partly derive from the use of different time periods, but it may also be associated with the use of different chemical transport models and inversion methods. Because the emissions from the prolonged, smoldering fires associated with coarse woody debris and organic matter in soils are injected near the surface, the transport of these emissions from the surface to the altitude where the MOPITT instrument has enough

Table 1. Annual Amounts of CO Emissions (Tg CO yr^{-1}) From Open Vegetation Fires and Scaling Factors

Regions[a]	*Ito and Penner et al.* [2004]	*Arellano et al.* [2004]	Scaling Factors
Central and northern South America	6	105	18.2
Southern South America	12	109	9.2
Northern Africa	74	98	1.3
Southern Africa	94	60	0.6
Southeast Asia	11	102	9.4
Boreal regions	46	10	0.2
Others	22	69	3.1
Total	266	552	2.1

[a]Regions are defined by *Arellano et al.* [2004].

sensitivity to measure the CO associated with these emissions is particularly important. The estimates of CO from different inverse studies can therefore differ because different chemical transport models may differ in their transport of surface-emitted air pollutants.

[9] The sum of the total area burned in July and September 2000 in Southern Africa from MODIS (831×10^3 km^2) [*Roy et al.*, 2002] is substantially larger than that from the Global Burned Area data set for the year 2000 (GBA2000) (475×10^3 km^2) [*Tansey et al.*, 2004], which was used to develop a global map of emissions from open vegetation in *Ito and Penner* [2004]. As a result, the estimated emissions of *Ito and Penner* [2004] might be significantly smaller (i.e., 263–421 Tg CO yr^{-1}) than the emissions that would be calculated with an improved burned area data set. Unfortunately, a global map of burned area data for the year 2000 is not available from MODIS. However, *Arellano et al.* [2004] estimated global CO emissions that were significantly larger that the estimates from *Ito and Penner* [2004], i.e., from 486 to 633 Tg CO yr^{-1} (−12% to 15%). Therefore we scaled the bottom-up estimate of *Ito and Penner* [2004] by the top-down estimate of *Arellano et al.* [2004].

[10] The estimated errors associated with ranges in the OV estimates for BC and POM can be estimated using the formula

$$[\mathrm{Error_x}] = \left(\mathrm{Error_{co}^2} + \left(\mathrm{Error_{EFx}}/\mathrm{Error_{EFco}}\right)^2\right)^{1/2}, \qquad (2)$$

where x is either BC or POM, and EF represents the emission factor. The error in the CO emission estimate is not carried over from the 2-sigma root mean square error estimate of approximately ±3% for the four error scenarios and three retrieval data sets (column, 700 mb, and 500 mb CO) considered by *Arellano et al.* [2004], because they noted that the true uncertainties from the inverse model were likely larger than the ranges estimated from their limited exploration of the sensitivity of the results to methodological assumptions. Alternatively, the global 2-sigma error of 28% deduced in the inverse study by Zhang et al. (submitted manuscript, 2004) (see also *Zhang* [2004]) for 1997 is adopted. The errors associated with the average emission factors for BC, organic carbon (OC), and CO are available from the data summarized by *Andreae and Merlet* [2001], but the error in the ratio of BC and OC to CO is much smaller than the 28% error adopted in the CO emissions since the emission factors for BC or OC and CO are correlated. As a result, we obtain an error estimate that is similar to the error estimate associated with that determined by Zhang et al. (submitted manuscript, 2004), i.e., approximately 28%.

[11] Next, we developed a monthly emission map for 1979–2000 using the TOMS AI data product. The TOMS AI is a measure of the change of spectral contrast in the near ultraviolet due to the radiative transfer effects of aerosols in a Rayleigh scattering atmosphere [*Herman et al.*, 1997; *Torres et al.*, 1998]. By definition, AI is positive for absorbing aerosols, near zero (±0.2) in the presence of clouds or large size non-absorbing aerosols, and negative for small size non-absorbing aerosols [*Torres et al.*, 2002].

[12] Following the suggestions of O. Torres (2000) from *Duncan et al.* [2003], we multiplied the AI from Nimbus 7 TOMS by 0.75 to normalize it to the data from the Earth-Probe TOMS. Also, following the study of *Duncan et al.* [2003], 2 months of data, July and August 1990, were removed because of apparent data quality problems. We processed only positive values of AI data ($0.2 < \mathrm{AI}$), corresponding to absorbing aerosols and created a monthly mean value of AI (MAI) for each $1^\circ \times 1.25^\circ$ grid box. For each month, we used the total number of days of available data. In order to estimate the interannual variability of open biomass burning, first we removed the background MAI ($\mathrm{MAI_{min}}$) for each region by subtracting the value of MAI when no open biomass burning occurs (i.e., the annual minimum) for each region, as defined by *Duncan et al.* [2003]. Data gaps for the 2 months in 1990 and from mid-1993 to mid-1996 were filled by the averaged MAI for each region. We calculated the monthly emissions of BC and POM by adjusting the annual emissions for 2000 described above by the calculated MAI for each region. Thus our historical emissions for each region, s', and month, t, and year, u, were calculated from

$$\left[E_{s',t,u}\right] = \left[E_{s',2000}\right] \times \left(\left[\mathrm{MAI}_{s',t,u}\right] - \left[\mathrm{MAI}_{\mathrm{min},s',u}\right]\right) / \left(\left[\mathrm{MAI}_{s',2000}\right] - \left[\mathrm{MAI}_{\mathrm{min},s',2000}\right]\right). \qquad (3)$$

Emissions within each region were distributed according to the $1^\circ \times 1^\circ$ distribution developed in the bottom up inventory of *Ito and Penner* [2004]. This extrapolation may cause additional uncertainties in the seasonal distribution at the $1^\circ \times 1^\circ$ resolution, because *Pétron et al.* [2004] noted that there was a 1- to 2-month delay between the peak in the

MODIS fire counts and the peak in the MOPITT CO retrievals for most regions in the Southern Hemisphere. Further studies are needed to quantify the seasonal variations of OV emissions accurately.

[13] The errors associated with scaling the 2000 emissions by the MAI in each region are difficult to assess. Nevertheless, the global average emissions that we find by our scaling method (6.1 Tg BC) are within 10% of those (6.8 Tg BC) estimated by Zhang et al. (submitted manuscript, 2004) for 1997. Taking the difference between our emissions and those from Zhang et al. (submitted manuscript, 2004) by region as indicative of the error results in an overall global average error of 24%, which is similar to the global 2-sigma error (28%) deduced in that inverse study. This suggests that the errors in our regional emissions might be of similar accuracy to those deduced in this inverse study which ranged from 25% to 35% in most regions but was as large as 55% in Southeast Asia and was close to 200% in Indonesia (owing to the unusual fires in Indonesia in 1997). Thus we estimate that our global average emissions over the time period from 1979 to 1999 might have an overall 2-sigma error of order 28%. Of course larger (and generally unquantified) errors are expected within some of the regions and within each grid cell.

[14] During the period from 1870 to 1978, we used the averaged seasonal variations of BC and POM for 1979–2000 and the global historical anthropogenic CH_4 emissions from biomass burning developed by *Stern and Kaufmann* [1996] (available from the Carbon Dioxide Information Analysis Center (CDIAC)). *Stern and Kaufmann* [1996] based their emissions estimates on anthropogenic CO_2 emissions from the land-use change estimated by *Houghton et al.* [1983]. Our BC and POM emissions were scaled from the CH_4 emissions by assuming that the ratio of the mean BC and POM emissions per unit CH_4 emission was constant and equal to the ratio in 1979. For 1979, the global annual average BC and POM emissions were summed from the estimates in all grids. Thus the annually averaged emissions P (Tg species yr^{-1}) for each year, t, were calculated from

$$[P_{t,x}] = [P_{1979,x}]/[Q_{1979}] \times [Q_t], \quad (4)$$

where x is either BC or POM and Q is emissions of methane. These annual emissions were redistributed back to the $1^\circ \times 1^\circ$ degree grids by using the monthly average and spatially resolved emissions patterns from the 1979–2000 time period.

[15] The errors associated with this scaling from 1870 to 1978 are difficult if not impossible to quantify. *Van Aardenne et al.* [2001] found it impossible to be quantitative when they used a similar type of scaling, but noted that this type of scaling provides the lowest quality and that the use of a global aggregated estimate further decreases the quality of the data. Significant assumptions have been made in determining the land-use change emissions of *Houghton et al.* [1983], and further assumptions are made in using the global average ratio of emissions of CH_4 from land-use change in 1979 to scale the total open biomass burning sources prior to 1979. It is quite likely that the errors become larger as one goes back farther in time.

2.2. Biofuel Burning

[16] Biofuel burning (BF) includes biofuel wood burning, charcoal burning, burning of animal waste (dung) and crop residues, and charcoal making. The global distributions from biofuel burning were estimated for the period 1870–2000, based on methods used by *Ito and Penner* [2004]. The BC and POM emission factors are based on the compilation by *Bond et al.* [2004].

[17] As pointed out by *Bond et al.* [2004], the emission factors for wood fuels used for heating (i.e., $1.4 \pm 1.0 - 15 \pm 8$ g PM/kg wood) differ significantly from those used for cooking (i.e., $2.3 \pm 0.8 - 3.9 \pm 3.0$ g PM/kg wood) so that the relative magnitude of wood burning in these practices must be differentiated. Following the study of *Bond et al.* [2004], we represent regional differences in EF for biofuel by changing the fuel use technology, rather than by varying emission factors. Thus the same emission factors for each fuel usage are applied to both developed and developing countries. However, the differences in emission factors among the different usages are great enough so that the averaged emission factors for wood fuels in "industrialized" countries differ significantly from those for "developing" countries. This is because the fraction of wood burned in cooking or heating in developed countries differs from that burned in these practices in developing countries. Hence the following description of our methods is divided along these lines.

2.2.1. Developed Countries

[18] The total consumption of wood fuel for heating and cooking and the consumption and production of charcoal for developed countries for the time period from 1961 to 2000 were derived from FAOSTAT on a country and an annual basis. Since IEA has traditionally focused on tabulating energy from fossil fuel use, the agency just began to include renewable fuels during the 1990s. Therefore the statistics for biofuel consumption are not complete over the 1961–2000 time period. For example, the data for the primary biomass consumed in the residential sector started in 1993 in the United States. While the FAOSTAT database is available over the time period from 1961 to 2000, it does not separate the consumption of wood fuel into categories associated with the industrial or residential sector, as does the IEA database. According to the IEA database for 1996 in the Organization for Economic Co-operation and Development (OECD) countries, most of primary solid biomass is consumed in the residential (e.g., 55%) and industrial sectors (e.g., 40%). More than half of the primary solid biomass consumption in the industrial sector can be ascribed to the paper, pulp, and printing industry (e.g., 53%). *Bond et al.* [2004] estimated the emissions from wood used for the industrial sector in 1996 and found that they were only 2% of global BC emissions from "contained" combustion, which includes FF and BF. Moreover, the largest fraction of this industrial wood fuel use is in developing countries. Therefore, in the developed countries, we assumed that the consumption of wood fuel from the FAOSTAT database was entirely in the residential sector,

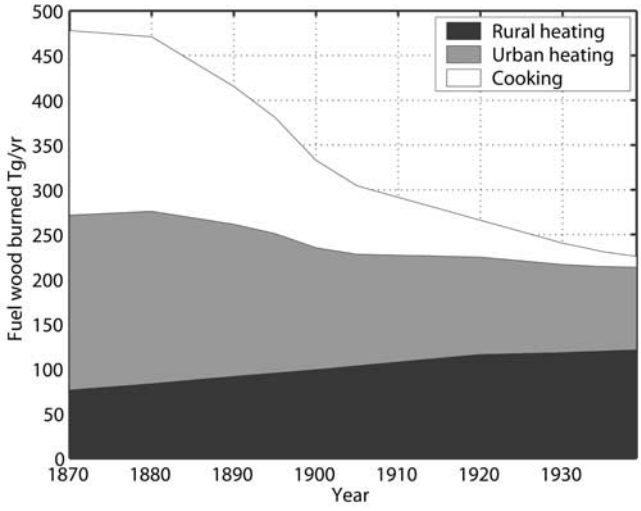

Figure 1. Estimated fuel wood burned (Tg wood yr^{-1}) in developed countries from 1870 to 1939.

and we used the residential wood fuel emission factors outlined below over all time periods.

[19] For 1949 to 1960, we could not find a time series of country-specific data for annual wood fuel consumption except in the United States, so we based our estimates of residential wood fuel consumption in developed countries on data for residential wood consumption data in the United States. The U.S. IEA provides data for wood fuel use in residential, commercial, and industrial sectors. Thus the residential fuel consumption R (Tg yr^{-1}) for each year, t, over the period 1949–1960 in each developed country was calculated from

$$[R_{t,y}] = [R_{t,\text{u.s.}}]/[R_{1961,\text{u.s.}}] \times [R_{1961,y}], \quad (5)$$

where y is country. For 1870 to 1948, only total wood consumption data in the United States are available, so we assumed that the decrease in the residential wood fuel consumption for each developed country was proportional to the change in the total wood consumption in the United States. On the basis of the U.S. data in 1949, the residential sector was dominant (68%) in the total wood consumption, so the assumption that the decrease in total wood fuel used was mainly determined by the change in the residential sector is reasonable.

[20] For charcoal consumption and production, we used the FAOSTAT data from 1961–2000, and assumed the same trends as the total wood consumption prior to 1961.

[21] The trend in the use of wood fuel for heating could be different between urban and rural areas because fossil fuels replaced wood fuels in urban areas prior to their widespread use in rural areas. Therefore the total wood consumption for the time period from 1870 to 1940 was divided into rural and urban fractions by using population data for 1940. In the United States, urban and rural populations were taken from the *United States Census Bureau Population Division* [1993]. For Canada, Europe, Oceania, and the U.S.S.R., rural population data were taken from the *United Nations Statistical Office* [1969]. On the basis of these data, we assumed that rural heating increased as the rural population increased from 1870 to 1940, while urban heating decreased as the wood consumption decreased during this period. Thus wood consumption for rural heating, *WRH*, and urban heating, *WUH*, for North America, Europe, the Pacific islands, and the U.S.S.R. was calculated from

$$[WRH](t) = [WRH](1940) \times [RP](t)/[RP](1940) \quad (6)$$

$$[WUH](t) = [WUH](1940) \times [TWC](t)/[TWC](1940), \quad (7)$$

where *RP* is rural population and *TWC* is total wood consumption. Thus the total consumption of wood was used to estimate the interannual variability of wood fuel burning from 1870 to 1940. The wood used in cooking was estimated from the difference between total wood consumption and that used in heating,

$$[WCC](t) = [TWC](t) - [WRH](t) - [WUH](t). \quad (8)$$

Although the population in urban regions is growing during this time period, the fraction of wood fuel used in urban heating does not grow in order to account for the replacement of BF with FF in urban areas.

[22] Figure 1 shows the estimated fuel wood burned (Tg yr^{-1}) in developed countries from 1870 to 1939. The overall fuel wood burned in developed countries has decreased from 478 in 1870 to 226 Tg yr^{-1} in 2000. Heating (57%) and cooking (43%) contribute approximately equally to the total fuel wood burned in 1870, while heating contributes a large fraction to the total amount of wood burned after fossil fuels displace wood as a fuel in developed countries.

[23] Since emission factors differ for wood burned in heating and wood burned in cooking, we accounted for the fraction of wood burned in cooking and in different heating technologies. Following *United States Environmental Protection Agency* (*U.S. EPA*) [1998] assumptions, we assumed that the fraction of wood burned in different heating technologies and in cooking was constant for each country from 1940 to 2000. For the United States and Canada, we assumed that 25% of wood used for heating in the United States and Canada is burned in fireplaces, and the remainder (75%) in heating stoves [*Houck and Tiegs*, 1998; *Bond et al.*, 2004]. As in the work of *Bond et al.* [2004], in Western Europe, we assigned 5% to fireplace combustion and 25% to heating stoves and the remainder to boilers, while in eastern Europe and the U.S.S.R, we assigned 88% to heating stoves, 10% to open cooking fires, and 2% to fireplaces. Following the Diffuse Emissions Manuals from *Department of the Environment and Heritage* [1999] in Australia, we allocated to the following uses in the Pacific islands (Japan, New Zealand, and Australia): 30% in fireplaces, 52.5% in conventional heaters, and 17.5% in controlled combustion heaters.

[24] Prior to 1940, in order to account for the fraction of wood burned in heating and cooking, we derived the

amount of fuel used in these activities as a function of time. In the U.S. energy history [*U.S. EIA*, 2004], wood was a major source of energy for heating and cooking in 1860. As the population continued to increase, most rural homes were still heated with wood. In towns, coal was displacing wood in homes, which resulted in a decrease of the use of wood fuel. The decline can be attributed to the fact that fossil fuels were abundant, cheap, and more convenient than wood fuel during this period. Thus, 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 [*Bond et al.*, 2004].

[25] In most areas, we used the emission factors (EF) for wood fuel used in heating compiled by *Bond et al.* [2004] which were 0.15 g BC/kg and 1.1 g POM/kg for building stoker boilers, 0.27 g BC/kg and 3.2 g POM/kg for open cooking fires, 1.6 g BC/kg and 9.2 g POM/kg for fireplaces, and 1.4 g BC/kg and 12 g POM/kg for heating stoves. For Australia, New Zealand, and Japan, we used 1.1 g BC/kg and 9.7 g POM/kg for conventional heaters and 0.5 g BC/kg and 4.5 g POM/kg for controlled combustion heaters, following the Diffuse Emissions Manuals from *Department of the Environment and Heritage* [1999] in Australia. For each developed country, we applied the weighted average emission factors from *Bond et al.* [2004] to the wood used by country. For cooking, we used the arithmetic mean of emission factors for cooking fires from traditional and improved cooking stoves and open cooking fires (e.g., 0.46 g BC/kg wood and 2.5 g POM/kg wood). For the time period from 1870 to 1940, these same weighted average emission factors were applied for the heating part of the wood fuel used and the cooking emission factor was applied to the portion of wood used for cooking. The emission factors used for the consumption (e.g., 4.4 g BC/kg charcoal and 4.4 g POM/kg charcoal) and production (e.g., 1.2 g BC/kg charcoal and 14 g POM/kg charcoal) of charcoal were calculated from the measurements compiled by *Bond et al.* [2004].

[26] The emissions from these fuels are distributed within each country using population density maps for the years of 1990 and 1995 from *Center for International Earth Science Information Network* (*CIESIN*) [2000], and the emissions from heating are assigned to winter [*Liousse et al.*, 1996]. The 1990 population map was applied to 1870–1992, and the 1995 map was applied to 1993–2000. Since we used 1990 maps for population in the past, changes in population distributions within countries are not taken into account in this study. This extrapolation causes additional uncertainties in the spatial distribution at a $1^\circ \times 1^\circ$ resolution. A similar problem was discussed by *van Aardenne et al.* [2001]. We expect larger uncertainties in our fuel distribution within large countries with substantial internal migration (e.g., for the United States and U.S.S.R.) and larger uncertainties during the earliest time periods.

2.2.2. Developing Countries

[27] Biofuel use in developing countries was based on the data for 1985 provided by *Yevich and Logan* [2003]. *Yevich and Logan* [2003] estimated the biofuel burned in developing countries from government statistics, energy assessments from the World Bank, and many technical reports. To determine biofuel use in 1995 from the data which they summarized for 1985, they assumed that household wood fuel consumption correlates with population size. Therefore the total household biofuel use for a given country was estimated by multiplying the per capita usage by the population for that country. Population was also used to extrapolate the amount of emissions associated with charcoal burning, charcoal production, crop residue used for domestic fuels, and animal waste. We also used these assumptions to calculate estimates for residential biofuel burned in developing countries from 1961 to 2000. The population changes from FAOSTAT during 1961–2000 were used for this purpose.

[28] To determine the 1995 biofuel used as agro-industrial fuel (i.e., the use of crop wastes for an energy source in factories), *Yevich and Logan* [2003] scaled the amount in agro-industrial use for 1985 by the ratio of total crop production for 1995 to that for 1985 in each country. Again, we followed these methods for the time period 1961–2000, and used crop production changes from FAOSTAT during 1961–2000.

[29] The emissions for biofuel burning from 1870 to 1960 were extrapolated from those for 1961, using the same assumptions as those outlined above for residential biofuel. Thus these emissions were scaled by the population data from the *United Nations* [1973]. We assumed the same ratios of fuels burned in residential use and in agro-industrial use as for 1961.

[30] We used the same emission factors for cooking fires, and for consumption and production of charcoal, as those mentioned above for developed countries. The emission factors used for animal waste (e.g., 0.50 g BC/kg and 2.8 g POM/kg) and agricultural waste (e.g., 0.88 g BC/kg and 5.0 g POM/kg) were taken from the compilation of *Bond et al.* [2004]. As in the work of *Yevich and Logan* [2003], we applied emission factors for domestic biofuels to industrial use of biofuel, since we lack data on the emission factors for the later for developing countries. Measurements of the emission factors are needed to improve the estimates for industrial use of biofuel in developing countries.

[31] *Yevich and Logan* [2003] estimated a 1-sigma uncertainty for woodfuel consumption in the developing world of ±40% and a caution that their estimates for agricultural residue could be uncertain to ±50%. One would need to add the uncertainty associated with the emission factors for different biofuels to these estimates in order to estimate an overall uncertainty in the emissions. *Bond et al.* [2004] estimated the 95% confidence interval for "contained" combustion (i.e., biofuels plus fossil fuels) to be −30% to +120% for BC and −40% to 100% for OC. Wood used in the residential sector contributed 18% to the overall variance in the BC emissions estimates and 57% to the variance in the OC emissions estimates while agricultural waste in the residential sector contributed 8% of the variance in both the BC and OC emissions estimates and animal waste in the residential sector contributed 2% of the variance in both the BC and OC emissions estimates. If we use these variances to determine the overall uncertainties associated with biofuels, we find an estimated 95% confidence interval for BC

emissions from biofuels of −50% to 180% and an estimated 95% confidence interval for OC emissions from biofuels of −55% to 125%. This global average uncertainty is smaller than the uncertainty in individual regions [*Bond et al.*, 2004]. Here we adopt these error estimates for BC and POM from biofuels, since our emissions estimates used methods that are similar to those of *Bond et al.* [2004] and since we obtained estimates for contained combustion (5 Tg BC and 14 Tg POM) that are in good agreement with those of *Bond et al.* [2004] (5 Tg BC and 12 Tg POM = 1.3 × OC). We note, however, that the total uncertainties for some of the individual sectors and for the developing world, in particular, are probably larger than these numbers.

[32] Estimating the errors for time periods prior to and after 1985, which was the specific year for which *Yevich and Logan* [2003] determined their specific fuel use for developing countries, depends on whether scaling by crop production between 1961 and 2000 for agro/industrial use and scaling by population for biofuel and agro/industrial use over the entire time period is an adequate measure of any change. Technology changes (i.e., changes in the types of fuels burned and the specific burning practices for each fuel) introduce larger uncertainty during the earlier periods of our estimates. These comments also apply to our methods for developing biofuel use over time in the developed countries. As in the work of *Van Aardenne et al.* [2001], we cannot quantify these errors, but certainly these methods of determining emissions are of lower quality compared to methods that might use country-specific data on fuel use with country-specific information on the technologies used for burning each fuel.

3. Fossil Fuel Burning Emissions

[33] Sources of fuel consumption data for each fuel sector from 1960 to 2000 for OECD countries and from 1971 to 2000 for non-OECD countries were taken from *IEA* [2002a, 2002b] and emission factors of BC and POM for the base year of 1996 were obtained from *Bond et al.* [2004]. Here the term ''fuel sector'' refers to broad classes of fuel usage, such as transformation (i.e., the conversion of primary sources of energy to secondary, as in the conversion of heavy fuel oil to electricity), transport, industry, residential, the commercial and public sector, and agriculture. IEA also provides data for fuel use within certain subcategories for each sector. When discontinuities for the data for each fuel usage sector or each subcategory were found, we used the ratios of fuels by category during the most recent time period prior to the discontinuity to determine the fuel used in each category. From 1950 to 1970, data available from statistics developed by the *U.N.* [1976, 1984] were used at 5-year intervals, and data gaps between these 5-year intervals from 1950 to 1970 were filled by a linear interpolation.

[34] We adopted the emission factors and the technology breakdown (i.e., the types of equipment used for combustion) for each category within the fuel use sectors from *Bond et al.* [2004]. The technology breakdown for each country was not always provided by *Bond et al.* [2004]. In these cases, we assigned the technology breakdown for China given by *Streets et al.* [2001] to developing countries, and the most advanced technology to developed countries. Semi-developed countries wer eassigned a net emission factor for each category that was the square root of the product of emission factors for developed and developing countries. Tables 2a and 2b show the net submicron emission factor for each category of fuel use included in our inventory, while Table 3 summarizes the technology breakdown for these categories that we used for developing and developed countries. Except for diesel transport emissions, the technology breakdown was assumed constant with time.

[35] *Novakov et al.* [2003] assumed a reduction of the diesel transport emission factor in western countries by a factor of 5 from 1965 to 1985 to account for the development of new technologies and their implementation. In the United States, exhaust emissions from heavy-duty vehicle engines were first regulated in 1974. *Yanowitz et al.* [2000] reviewed over 400 dynamometer studies on diesel vehicles. They reported a decrease in emission factors for particulate matter (PM) from the model year 1974 through 1997 of -0.17 g kg^{-1} yr^{-1} for low-altitude tests. Here, in developed and semi-developed countries, we assumed that the emission factors for BC and POM decreased linearly with time proportional to the slope in the PM emission factor estimated by *Yanowitz et al.* [2000]. We fixed the EF for 1996 to that given by *Bond et al.* [2004]. This linear trend then suggests that the decrease began in 1977 in order to fit the value suggested by *Bond et al.* [2004] for developing countries in 1996. Starting the decrease in 1977 rather than 1974 is consistent with the fact that older cars are only gradually replaced with newer cars, though a number of variables may affect the trend in EF [e.g., *Cadle et al.*, 2000; *Ito et al.*, 2002; *U.S. EPA*, 2003]. For semi-developed countries, we began the decrease in 1984 to be consistent with the emission factors for semi-developed countries for 1996 in Tables 2a and 2b. Although the trend of EF for semi-developed countries may be different from that for developed countries, our EF of 3.8 g PM kg^{-1} estimated for 1992 is identical to that for 1992 in Russia and the Baltic states used by *Mäkelä* [1995].

[36] For other categories, the introduction of time-varying net emission factors would require specific information about when and how new technologies were introduced, and such a study was beyond the scope of our current project. Since the net emission factors for a given sector or category of fuel have probably decreased with time as more efficient technologies for fuel utilization were developed, the emissions estimated using constant technology may be too small in the past.

[37] In fact, if we used the assumption of *van Aardenne et al.* [2001] that the emission factors for a given category in developing countries applies to those in semi-developed and developed countries prior to 1970, we would find an abrupt decrease in the total global emissions of BC in from 3.1 Tg yr^{-1} in 1970 to 1.7 Tg yr^{-1} in 1971. The major contributors to this decrease can be ascribed to the changes in the emission factors for residential diesel fuels (65%) and coal transformation (28%). As in the work of *Bond et al.* [2004], we assumed that residential diesel use occurs in generators (i.e., 6 ± 8 g PM/kg) in regions where the per capita

Table 2a. Net Submicron Emission Factors for BC[a]

Fuel Category	Developing	Semi-Developed	Developed
Coal transformation			
Coke ovens	1.2172	0.6256	0.3216
Blast furnaces	0.0045	0.0027	0.0016
Rest of transformation	0.0017	0.0005	0.0001
Coal industry			
Iron and steel	1.2172	0.6256	0.3216
Nonmetallic minerals	1.1468	0.0729	0.0046
Rest of industry[b]	0.0293	0.0117	0.0046
Coal transport	0.4950	0.4950	0.4950
Coal residential	2.2950	1.6068	1.1250
Lignite residential	1.0350	1.0350	1.0350
Lignite transformation			
Coke ovens	2.4635	1.2661	0.6508
Blast furnaces	0.0092	0.0054	0.0032
Rest of transformation	0.0023	0.0008	0.0003
Lignite industry			
Iron and steel	2.4635	0.2271	0.0209
Nonmetallic minerals	2.2919	0.1087	0.0052
Rest of industry[c]	0.0202	0.0102	0.0052
Diesel transport			
Internal navigation	1.1286	1.1286	1.1286
Rails	1.5325	1.5325	1.5325
Road[d]	2.9515	2.4691[e]	1.1494
Diesel, other sectors			
Agriculture	3.1786	2.7245[e]	2.4974
Rest of "other" sectors[f]	3.4056	0.4714	0.0653
Gasoline transport	0.2909[g]	0.2141[e]	0.0829
Transport aviation	0.0980	0.0980	0.0980
Heavy fuel oil	0.0396	0.0143	0.0051

[a]Units are g BC/kg fuel.
[b]These values were also used for the energy sector for coal.
[c]These values were also used for the energy sector for lignite.
[d]This sector includes the "nonspecified transport" category from the IEA database. These are net emission factors for 1996. See text for the emission factors for earlier time periods.
[e]The technology breakdown was specified explicitly for eastern Europe by *Bond et al.* [2004].
[f]This sector includes residential and commercial/public use.
[g]Special treatment for two-stroke engines in India yields a net EF of 0.6371.

electricity use is below 1000 k Wh/yr, and in external combustion devices in regions where the per capita use is above that level (i.e., 0.25 g PM/kg). In reality, the choice of the technology division between external combustion devices and generators is not so simple. For example, diesel generators equipped with particulate matter filters are used to some extent in developed counties. In any case, we did not try to estimate the historical changes of residential generators and external combustion devices nor the improvement of technologies for the emissions from coke ovens when emissions are captured (5.8 g PM/kg) compared to those from uncaptured coke emissions (20 g PM/kg) in developed and semi-developed countries. This discussion points out the difficulty in determining uncertainties in emissions associated with changes in the methods used to burn the various fuels over the time period when emissions from fossil fuels are growing rapidly (1950–2000). As noted above, *Bond et al.* [2004] estimated the 95% confidence interval for "contained" combustion (BF plus FF) to be −30% to +120% for BC and −40% to 100% for OC. The major fossil fuel based burning activities that contribute to the variance associated with these uncertainty estimates for BC were coke making (31%), industrial burning of coal (14%), on-road and off-road diesel use (18%), residential coal (4%), and gasoline use in transport (1%). For OC the major contributors to the variance in emissions estimates were gasoline use in transport (10%), coke making (7%), coal in the industrial (3%) and residential (2%) sectors, and on-road diesel use (1%). As above, if we use these estimates of the contribution of fossil fuel burning uncertainties to the total variance for contained combustion, we estimate an overall 95% confidence interval for BC emissions from fossil fuels of −40% to 150% and for OC emissions from fossil fuels of −50% to 125% (for 1996). However, on the basis of the above discussion, we caution that the uncertainty in fossil fuel emissions of BC and POM might be almost twice as large during the timeframe before 1970 and might be even larger prior to 1950. Nevertheless, there are

Table 2b. Net Submicron Emission Factors for POM[a]

Fuel Category	Developing	Semi-Developed	Developed
Coal transformation			
Coke ovens	1.3187	0.6778	0.3483
Blast furnaces	0.0117	0.0069	0.0040
Rest of transformation	0.1160	0.0499	0.0215
Coal industry			
Iron and steel	1.3187	0.6778	0.3483
Nonmetallic minerals	1.3505	0.5505	0.2244
Rest of industry[b]	0.3006	0.2597	0.2244
Coal transport	4.4550	4.4550	4.4550
Coal residential	2.2950	1.6068	1.1250
Lignite residential	3.1050	3.1050	3.1050
Lignite transformation			
Coke ovens	7.8010	4.0095	2.0607
Blast furnaces	0.0565	0.0332	0.0195
Rest of transformation	0.2707	0.1185	0.0519
Lignite industry			
Iron and steel	7.8010	4.2651	2.3319
Nonmetallic minerals	7.1802	1.9162	0.5114
Rest of industry[c]	0.4665	0.4884	0.5114
Diesel transport			
Internal navigation	0.5263	0.5263	0.5263
Rails	0.7895	0.7895	0.7895
Road[d]	0.1642	1.5205[e]	0.0592
Diesel, other sectors			
Agriculture	1.6374	1.4035[e]	1.2866
Rest of "other" sectors[f]	1.7544	0.5294	0.1598
Gasoline transport	1.7801[g]	1.0233[e]	0.4041
Transport aviation	0.0420	0.0420	0.0420
Heavy fuel oil	0.4554	0.1642	0.0592

[a]Units are g POM/kg fuel.
[b]These values were also used for the energy sector for coal.
[c]These values were also used for the energy sector for lignite.
[d]This sector includes the "nonspecified transport" category from the IEA database. These are net emission factors for 1996. See text for the emission factors for earlier time periods.
[e]The technology breakdown was specified explicitly for eastern Europe of *Bond et al.* [2004].
[f]This sector includes residential and commercial/public use.
[g]Special treatment for two-stroke engines in India yields a net EF of 9.5017.

Table 3. Technology Assumptions for Different Fuel Categories[a]

Fuel Category	Fraction of Given Technology	Fcont[b]
Coal Transformation		
Rest of transformation[c]		
PC ESP	0.50	0.02
PC/scrubber	0.35	0.24
PC/cyclone	0.10	0.05
Stoker/scrubber	0.02	0.24
Stoker/cyclone	0.03	0.10
Coke oven[d]		
Coke oven	0.80	0.33
Beehive polluting	0.20	1.00
Iron (blast)[e]		
Uncaptured	0.10	1.00
Captured	0.90	0.05
Coal Industry		
Other industry[f]		
Pulverized	0.15	0.24
Scrubber + heating stove/stoker	0.25	0.42
Cyclone + heating stove/stoker	0.55	0.10
None + heating stove/stoker	0.05	1.00
Nonmetallic[g]		
Brick kilns	0.25	1.00
Average of industrial	0.75	N.A.
Iron and steel[d]		
Coke oven	0.80	0.33
Beehive polluting	0.20	1.00
Coal residential[h]		
Residential stokers	0.50	1.00
Cookstoves/open fires	0.50	1.00
Diesel Transport		
Road[i]		
Regular fleet	0.80	1.00
Super-emitters	0.20	1.00
Diesel, Other Sectors		
Rest of "other" sectors[j]		
Stationary generators	1.00	1.00
External combustion device[k]	0.00	1.00
Agriculture[i]		
Regular fleet	0.80	1.00
Super-emitters	0.20	1.00
Gasoline transport[l]		
Regular fleet	0.76	1.00
Super-emitters	0.19	1.00
2-stroke	0.05	1.00

[a]Technology factors for lignite are assumed to be the same as those for coal.
[b]Fcont represents the fraction of fine PM that penetrates the control device. One refers to combustion without emission controls.
[c]For developed countries, the technology assumption is assigned into the 100% PC/scrubber.
[d]For developed countries, the technology assumption is assigned into the 100% coke oven.
[e]For developed countries, the technology assumption is assigned into the 100% captured category.
[f]For developed countries, the technology is split as 75% pulverized and 25% cyclone + heating stove/stoker.
[g]For developed countries, the technology assumption is assigned into 100% of the average of industrial emissions.
[h]For developed countries, the technology assumption is assigned into the 100% residential stokers.
[i]The fraction of "regular fleet" is 95% in developing countries and 90% in semi-developed countries.
[j]This sector includes residential and commercial/public use.
[k]This category is used for developed countries.
[l]The fraction of regular fleet, super-emitters, and two-stroke combustors are assumed to be 93%, 5%, and 2% of the total fleet in developed countries and 85.5%, 9.5%, and 5.0% of the total fleet in semi-developed and developing countries except in India, where they are 53%, 13%, and 33%.

large changes during 1870 (0.01–0.24 Tg BC) through 2000 (1.7–7.0 Tg BC) just associated with changes in the types of fuels burned.

[38] Thus, we followed *Novakov et al.* [2003], and, except for diesel fuel used in the on-road transport category, made the simple choice to keep the net emission factors in each category constant with time. The changes we define may represent the bulk of the emission changes, but there is clearly a need to better define the technology introduced in different countries as a function of time.

[39] The emissions from the fossil fuels are distributed within each country using population distribution procedures developed by Logan et al. (1990) as briefly summarized by *Dignon* [1992]. First, the area associated with each country or its political subdivisions was identified and placed on the 1×1 degree cells. Then the populations of all urban areas with population greater than 50,000 were assigned to the proper $1^\circ \times 1^\circ$ geographic cell. Finally, the remaining rural population for each country or its political subdivisions was distributed with a uniform density among all populated cells. Populated cells were those cells identified with some human use, according to *Matthews and Fung* [1987]. This strategy avoids putting emissions into regions that are largely unpopulated, such as forested areas.

[40] The historical emissions of BC and POM from 1870 to 1950 were extrapolated, using the CO_2 emission data for each country for fossil fuel burning [*Andres et al.*, 1999] and assuming that the mean BC and POM emission per unit CO_2 emission remained constant for each country during this period. Country statistics from the $1^\circ \times 1^\circ$ inventory developed by *Andres et al.* [1999] (available from CDIAC) were calculated by summing the estimates in all grids associated with each country. Then our historical emissions S (Tg species yr^{-1}) for each year, t, and country, y, were calculated from

$$[S_{t,x,y}] = [S_{1950,x,y}]/[T_{1950}] \times [T_t], \quad (9)$$

where x is either BC or POM and T is emissions of CO_2.

4. Results and Discussion

[41] Figures 2a–2d show the estimated emissions from open vegetation (OV), biofuel (BF), and fossil fuel (FF) burning for BC emissions in the Northern Hemisphere (Figure 2a) and the Southern Hemisphere (Figure 2b) and POM emissions in the Northern Hemisphere (Figure 2c) and the Southern Hemisphere (Figure 2d). The overall global BC emissions have increased from 2.1 in 1870 to 8.2 Tg BC yr^{-1} in 2000, while the POM emissions have increased from 15 to 41 Tg POM yr^{-1}. Note that because the BC emissions increased more rapidly than those of POM, the ratio of POM to BC has decreased from 7.2 in 1870 to 5.0 in 2000. The rapid rise in FF emissions after 1950 is mainly due to the substantial increase in fossil fuel consumption, and is consistent with the rapid rise in emissions of *Novakov et al.* [2003]. Apparently, the BC and POM emissions in the Northern Hemisphere have been the major contributor to the total emissions throughout the century (e.g., 73% for BC in 2000 and 68% for POM in 2000).

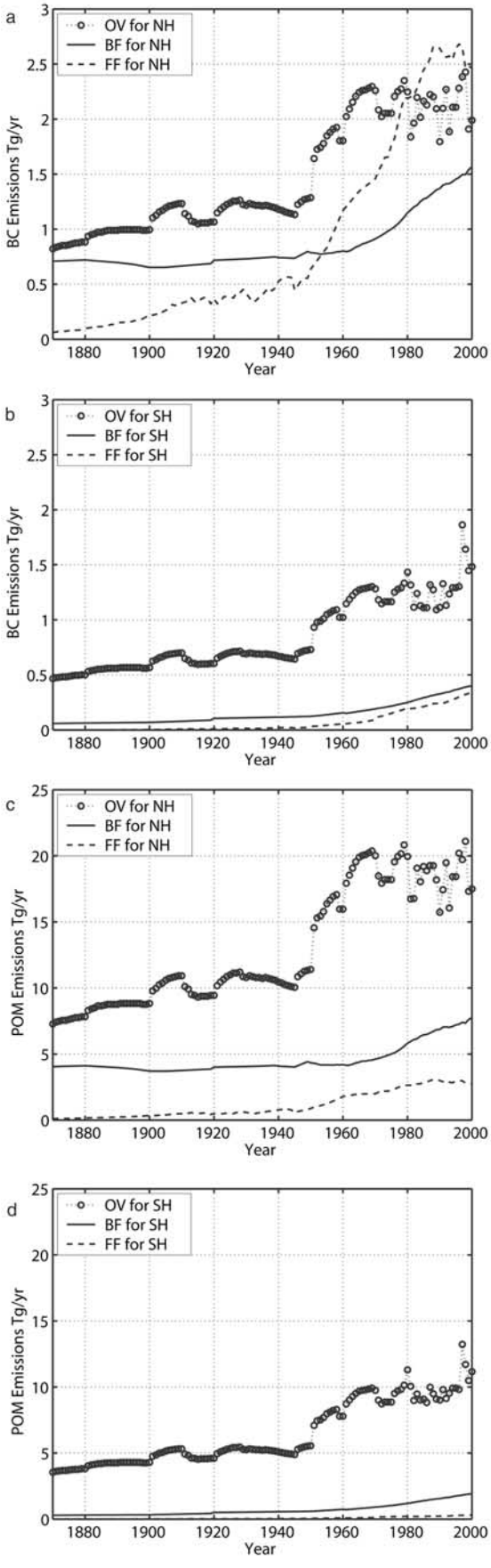

Figure 2. Estimated emissions from the open vegetation, biofuel, and fossil fuel burning. (a) BC emissions (Tg BC yr^{-1}) for the Northern Hemisphere. (b) BC emissions (Tg BC yr^{-1}) for the Southern Hemisphere. (c) POM emissions (Tg POM yr^{-1}) for the Northern Hemisphere. (d) POM emissions (Tg POM yr^{-1}) for the Southern Hemisphere.

[42] The BC and POM emissions from biomass burning (i.e., OV + BF) contribute a large fraction to the primary sources of carbonaceous particles even after fossil fuels became extensively used in the Northern Hemisphere (59% for BC and 90% for POM in 2000). We note, however, that the fossil fuel emissions could have been significantly larger prior to the decade of the 1990s had we accounted for emissions changes associated with different fossil fuel use technologies within all sectors. It is fairly apparent that there are discontinuous changes in the OV data set, because the original data sets for deforestation rates compiled by *Houghton et al.* [1983] are discontinuous. In particular, the rapid rise of deforestation rates in 1950 in tropical Africa in the estimate of *Houghton et al.* [1983] is apparent in both Figures 2a and 2b. (We note that the methods and data sources used to derive the time series of flux estimates of *Houghton et al.* [1983] were updated by *Houghton* [2003] and include regional estimates, but these CO_2 emissions cannot directly be used to estimate particle emissions because they represent the net CO_2 flux, which can be less than zero.) The global emissions of CH_4 from *Stern and Kaufmann* [1996] do not provide regional emissions trends, so that our regional emissions from 1950 to 1979 are merely an extension of the average regional estimates in 1979 and, as such, include emissions from savannahs as well as emissions from land use change. Past emissions from open biomass burning are clearly highly uncertain, particularly for the period before any satellite data pertaining to fire detection or fire emissions were available.

[43] To investigate the uncertainties using the global emissions from *Stern and Kaufmann* [1996], Figure 3 shows the estimated BC emissions (Tg BC yr^{-1}) from scaling the CH_4 emissions given by *Stern and Kaufmann* [1996] and those from scaling the TOMS AI based on work by *Duncan et al.* [2003] during the period of overlap when both estimates are available (from 1979 to 1994). The overall emission derived from the extrapolation of the *Stern and Kaufmann* [1996] data has increased from 3.8 in 1979 to 4.7 Tg yr^{-1} in 1988 and subsequently declines slightly, while that based on the TOMS data shows a slight decrease to 3.5 Tg yr^{-1} in 1994 with slight interannual variations on a global scale. The emissions from scaling the *Stern and Kaufmann* [1996] data are generally larger than those based on the TOMS data over the time period from 1980 to 1994 by 32% on an average. We conclude, therefore, that errors of at least 30% over any 15-year time period are possible using this procedure.

[44] Because the changes in CH_4 emissions produced by *Stern and Kaufmann* [1996] using the CO_2 emissions developed by *Houghton et al.* [1983] only refer to emissions from land-use change, they do not represent emissions from savanna fires initiated by humans. The latter might be

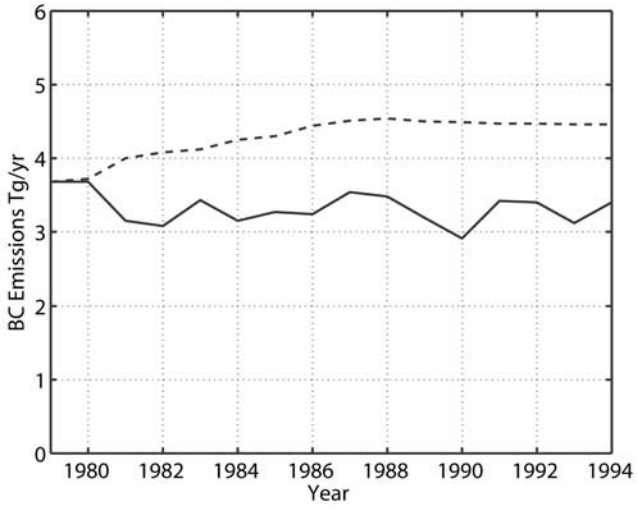

Figure 3. Estimated BC emissions (Tg BC yr^{-1}) based on different scaling methods from 1979 to 1994. The dashed line represents the scaling by CH_4 emissions from *Stern and Kaufmann* [1996]. The solid line denotes the scaling by TOMS AI.

assumed to scale with population, but since these fires are often uncontrolled, their spatial extent depends not only on their being started initially by humans but also on more complicated factors such as wind speed, humidity, and the local density of biomass. While several simplifying assumptions might have been used to develop this portion of the emissions back in time prior to 1979 [e.g., *van Aardenne et al.*, 2001], we simply relied on the trend developed by *Stern and Kaufmann* [1996] as being indicative of the possible overall trend. Spatially disaggregated simulation models of fire and vegetation dynamics together with regional estimates of burned areas may provide better space-and-time-resolved data on a global scale in the future [e.g., *Keane et al.*, 2004].

[45] Figure 4 shows the estimated CO emissions (Tg CO yr^{-1}) from open vegetation burning in this work and those of *van Aardenne et al.* [2001] from 1890 to 1990. Our open vegetation burning emissions are systematically higher by a factor of 2.1 on average. These higher emissions are tied to the higher emissions which we developed for the year 2000 based on the top-down estimate of OV CO emissions by *Arellano et al.* [2004]. Although our emissions are on average higher, the trend prior to 1950 is similar to that of *van Aardenne et al.* [2001], partly because of the use of the same types of proxies for extending emissions back in time. For instance, *van Aardenne et al.* [2001] scaled their early emissions for deforestation fires in tropical regions by rural population, while *Houghton et al.* [1983] used population data for their estimate of agricultural expansion in the tropics.

[46] The annual emissions of BC (Tg BC yr^{-1}) and POM (Tg POM yr^{-1}) are compared with previous estimates in Table 4. The estimate by *Liousse et al.* [1996] includes anthropogenic SOA, because their POM emissions were related to the amount of BC produced from anthropogenic sources using measured ratios of POM to BC in urban areas. SOA is not included in the other estimates in Table 4.

[47] Our POM estimate for the open biomass burning (46 ± 13 Tg POM) in 1996 is significantly larger than that for the sum of biofuels and fossil fuel (8–28 Tg POM). Our emissions for fossil fuel burning are in good agreement with those of *Bond et al.* [2004] for 1996. However, they are significantly smaller than previous estimates by *Liousse et al.* [1996] and *Cooke et al.* [1999], mainly owing to our use of the revised net emission factors developed by *Bond et al.* [2004] for specific fuel use sectors. The *Liousse et al.* [1996] emissions generally agreed well with observations when used in chemical transport models [*Penner et al.*, 2001; Zhang et al., submitted manuscript, 2004]. The FF POM estimate of *Liousse et al.* [1996] is larger than our estimate of FF primary organic aerosols by 25 Tg POM yr^{-1}. Estimates of annual global production of SOA from FF anthropogenic VOC range from 0.05 to 2.62 Tg yr^{-1} [*Penner et al.*, 2001; *Tsigaridis and Kanakidou*, 2003; *Lack et al.*, 2004]. These results suggest that our new POM inventory will lead to significantly smaller atmospheric POM burdens over industrialized areas than those in previous modeling studies, even if SOA production is included. Because use of the older inventories gave results that were in reasonable agreement with measured values, it is likely that this new inventory will result in POM burdens that are far too small. Achieving a predictive capability for atmospheric POM concentrations will require further work involving laboratory experiments, modeling, and observations.

[48] Our emissions for biomass burning are generally in good agreement with previous estimates. *Generoso et al.* [2003] used the ATSR fire counts to estimate interannual variations. The emissions derived from the fire counts show larger interannual variations than those derived from TOMS AI. Use of the former method does not account for inter-

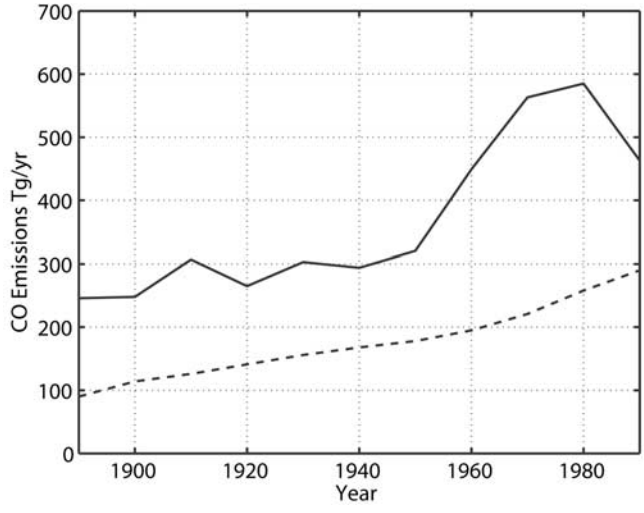

Figure 4. Estimated CO emissions (Tg CO yr^{-1}) from the open vegetation burning from 1890 to 1990. The dashed line represents the estimates from *van Aardenne et al.* [2001]. The solid line denotes those in this work.

Table 4. Annual Amounts of Submicron BC and POM From Combustion[a]

		Biomass		Fossil Fuel	
Reference	Year	BC	POM	BC	POM
Liousse et al. [1996]	1980	5.6	45	6.6	28[b]
This work	1980	5.9	59	2.4	2.8
Cooke and Wilson [1996]	1984	6.0	...	8.0	...
Cooke et al. [1999]	1984	...	...	5.1	9.1[c]
This work	1984	5.5	52	2.6	2.9
Andreae and Merlet [2001]	1985	4.8	53[d]	...	...
This work	1985	5.7	54	2.7	3.1
Bond et al. [2004]	1996	5.0	40[c]	3.0	3.1[c]
This work	1996	6.5	60	3.0	3.3
Generoso et al. [2003]	1997	7.1	61	...	...
This work	1997	6.1	50	3.0	3.2
Generoso et al. [2003]	1998	6.5	59	...	...
This work	1998	6.0	49	2.8	3.1
Generoso et al. [2003]	1999	3.9	34	...	...
This work	1999	5.3	44	2.8	3.1
Generoso et al. [2003]	2000	3.4	29	...	...
This work	2000	5.4	45	2.8	3.1

[a]Units are Tg BC yr^{-1} for BC and Tg POM yr^{-1} for POM.
[b]The estimate includes secondary organic aerosols.
[c]A conversion factor of 1.3 for POM/OC is used [*Liousse et al.*, 1996].
[d]POM is calculated from $PM_{2.5}$ − BC.

annual variations in the spatial distribution of fuel consumption and fire characteristics [*Generoso et al.*, 2003]. On the other hand, use of the latter method includes the interannual and regional variation of the absorbing aerosols themselves. However, TOMS AI may not detect smoke plumes from low-intensity fires, such as smoldering leaf-litter, savanna grass, and peat fires [*Duncan et al.*, 2003]. Therefore it is possible that large amounts of emissions from low intensity fires are underestimated for any particular event. For instance, if the BC emissions in Indonesia for 1997 are calculated from the intermediate and upper estimates for above ground vegetation and below ground peat burning from *Page et al.* [2002] and the emission factor for tropical forest burning from *Andreae and Merlet* [2001], the BC emissions from open biomass burning in Indonesia range from 1.0 to 3.1 Tg BC yr^{-1}. The component of these emissions due to aboveground biomass burning is estimated as 0.2–0.6 Tg BC yr^{-1}. Our estimate for 1997, based on scaling the year 2000 emissions by the TOMS AI, results in a total estimate for Indonesia which is only 0.3 Tg BC yr^{-1}, significantly smaller than the total emissions based on the work of *Page et al.* [2002] but similar to the range estimated for aboveground biomass burning. If we estimate the emissions for Indonesia for 1997 based on scaling the ATSR fire counts by calculating the ratio of CO emissions between 1997 and 2000 from the estimate of *Schultz* [2002] and multiplying by our 2000 estimated BC emissions for Indonesia, the estimate is about a factor of 2 larger (0.7 Tg BC yr^{-1}) than the estimate based on the TOMS AI scaling, but is still smaller than that based on the work of *Page et al.* [2002].

[49] Figure 5 shows our BC emissions from FF burning and those derived using methods outlined by *Novakov et al.* [2003]. We note that our emissions in this comparison are bulk emissions, which include larger particles. The results of the comparison are shown from 1950 through 2000, because both our estimates and those of *Novakov et al.* [2003] prior to 1950 are primarily based on extending the emissions in 1950 by the trend in fossil-fuel CO_2 emissions. Our trend in the BC emissions from fossil fuel burning is in fairly good agreement with that from *Novakov et al.* [2003], but the emissions disagree in their absolute magnitudes. The agreement is due in part to a substantial overlap of the original data sources for both series (i.e., the fuel consumption data), while the disagreement is mainly caused by differences in the net emission factors used by *Novakov et al.* [2003] and *Bond et al.* [2004]. We adopted the emission factors of *Bond et al.* [2004] because they summarized emission factors from a large set of measurements with consideration of the technology used in each fuel category, whereas the emission factors adopted by *Novakov et al.* [2003] were mainly applied to the broader fuel use sectors described in the IEA database. Moreover, the emission factors used by *Novakov et al.* [2003] were based on the review of *Cooke et al.* [1999], who did not review as extensive a literature to develop their emission factors as did *Bond et al.* [2004].

5. Summary and Conclusions

[50] We constructed a data set of historical carbonaceous aerosol emissions for the period 1870–2000, which can be used in trend studies of tropospheric aerosols and in environmental assessments. First we developed a present-day inventory of emissions of aerosols from OV burning. Because of the large uncertainties in open biomass burning emissions from bottom-up estimates of burning [*Ito and Penner*, 2004], the estimates were regionally scaled by previous results from an inverse modeling study for CO for the year 2000 [*Arellano et al.*, 2004]. Then we used data

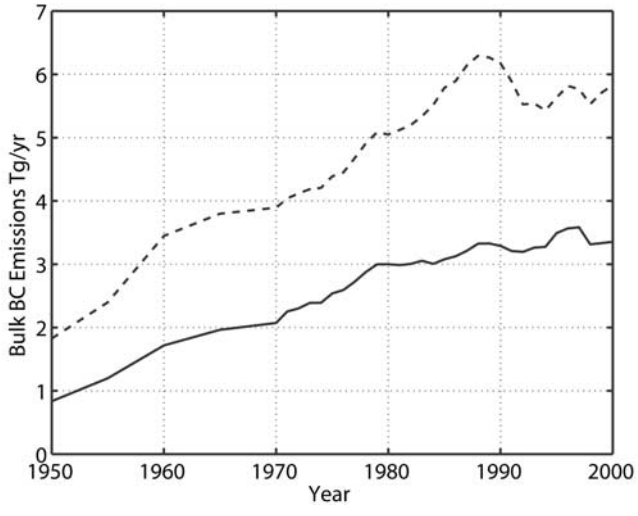

Figure 5. Estimated bulk BC emissions (Tg bulk BC yr^{-1}) from the fossil fuel burning from 1950 to 2000. The dashed line represents estimates derived using methods outlined by *Novakov et al.* [2003]. The solid line represents those of this work. Our emissions in this figure are bulk emissions, which include larger particles.

from the TOMS satellite [*Herman et al.*, 1997; *Torres et al.*, 1998] to extend the inventory to the period from 1979 through 2000. The global emissions are in good agreement with previous studies, although there are large differences in the estimated interannual variability. Emissions are extrapolated to the pre-satellite era using historical anthropogenic CH_4 emissions generated by *Stern and Kaufmann* [1996], which were based on estimates of anthropogenic CO_2 emissions from land-use change developed by *Houghton et al.* [1983].

[51] Next we used data sets provided by *Yevich and Logan* [2003] and data sets from FAOSTAT together with emission factors from *Bond et al.* [2004] to separately determine the sources of emissions from biofuel burning in developing and developed countries. Emissions in developed countries are derived from the country-specific FAOSTAT data for wood consumption for residential use from 1961 to 2000. Estimates of wood used in these activities prior to 1961 were extrapolated from these data using data for the residential (or total) wood consumption, and rural and urban population data. In developing countries, residential biofuel emissions are extrapolated in time using the per capita household usage developed by *Yevich and Logan* [2003] and population statistics. The agro-industrial uses in each country together with crop production changes for 1961–2000 were used to the estimate the agro-industrial combustion of these fuels. Prior to 1961, this usage was extrapolated using population statistics.

[52] The development of historical emissions from fossil fuel combustion relies mainly on IEA and U.N. data for fuel use consumption over the time period from 1950 to 2000 together with the technology divisions in each sector and emission factors for each technology that are summarized by *Bond et al.* [2004]. Changes in technology and the resulting net emission factors have occurred over this time period, but we only estimated these changes for emissions from diesel vehicles. Improvements are needed, especially for residential diesel fuels, where the changes in technologies and the division between boiler/furnaces and generators may cause large changes to our historical inventory. Emissions from fossil fuels prior to 1950 were extrapolated based on an available inventory for CO_2 emissions.

[53] The trends of fossil fuel burning emissions are in good agreement with those of *Novakov et al.* [2003], but there are large differences in the magnitude of the total fossil fuel emissions. An accurate representation of the magnitudes and temporal changes due to the introduction of different technologies with time is needed to improve these estimates. Our inventory for FF POM will lead to significantly smaller atmospheric POM burdens over industrialized areas in chemical transport models. Further laboratory experiments, modeling, and observations are needed to accurately predict atmospheric concentrations of POM using a global chemical transport model.

[54] During the last decade of this time period, the BC and POM emissions from biomass burning (i.e., OV + BF) contribute a significant amount to the primary sources of BC and POM and are larger than those from FF. Thus 59% of the NH BC emissions and 90% of the NH POM emissions are from BB in 2000.

[55] The emission model developed here is specific to the time period analyzed and has the advantage of accounting explicitly for much of the expected spatial and temporal variations. The BC data set has been tested in a global chemical transport model in conjunction with an inverse model of biomass burning emissions [*Penner et al.*, 2004]. Global climate models will be used to study the relative contribution of BC and POM emissions to climate change and to estimate the radiative forcing associated with anthropogenic emissions of carbonaceous aerosols.

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