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Trend in Global Black Carbon Emissions from 1960 to 2007

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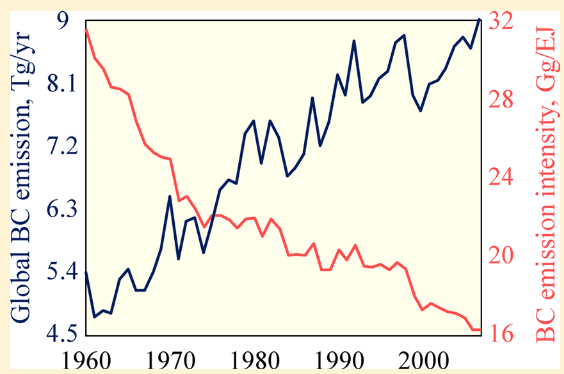
Supporting Information

ABSTRACT: Black carbon (BC) plays an important role in both climate change and health impact. Still, BC emissions as well as the historical trends are associated with high uncertainties in existing inventories. In the present study, global BC emissions from 1960 to 2007 were estimated for 64 sources, by using recompiled fuel consumption and emission factor data sets. Annual BC emissions had increased from 5.3 (3.4−8.5 as an interquartile range) to 9.1 (5.6−14.4) teragrams during this period. Our estimations are 11−16% higher than those in previous inventories. Over the period, we found that the BC emission intensity, defined as the amount of BC emitted per unit of energy production, had decreased for all the regions, especially China and India. Improvements in combustion technology and changes in fuel composition had led to an increase in energy use efficiency, and subsequently a decline of BC emission intensities in power plants, the residential sector, and transportation. On the other hand, the BC emission intensities had increased in the industrial and agricultural sectors, mainly due to an expansion of low-efficiency industry (coke and brick production) in developing countries and to an increasing usage of diesel in agriculture in developed countries.



INTRODUCTION

Black carbon (BC) warms the Earth system by absorbing sunlight and emitting infrared radiation, and by decreasing the ice/snow albedo.[1] With a direct radiative forcing of +0.4 (+0.05 to +0.8) W m^{-2} for BC from fossil fuel and biofuel suggested in the Fifth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC),[1] BC had become an important warming species in addition to carbon dioxide and methane. In addition to its effect on climate, BC is an important component in ultrafine particles in air and a strong absorbent for various toxic chemicals, and it is thus associated with a number of adverse health impacts.[2,3] Therefore, any efforts to reduce BC emission can lead to the dual benefit of slowing down global warming and of protecting human health.[4,5]

In the bottom-up approach to construct the emission inventories, BC emissions are calculated from the statistics of fuel consumption and BC emission factors (EF_{BC}) defined as BC emitted per fuel consumed or product produced by a given activity.[6,7] With a large number of data required, BC emission inventories are always associated with high uncertainties due to data limitation. For example, EF_{BC} values for a particular source could vary greatly under different conditions of combustion.[7,8] It has been recognized that EF_{BC} of many sources in the industrial, transportation, and residential sectors are significantly variable among countries, especially between developing and developed countries.[7,9,10] Unfortunately, most EF_{BC} used in the development of BC emission inventories are measured in developed countries, whereas developing countries contribute predominantly to the global total BC emissions.[6,7] In fact, the results in atmospheric transport models using existing BC inventories always underestimate atmospheric BC concentrations as well as light absorption compared with in situ measurements.[11−13] Therefore, continuous efforts have been made to improve the inventory by compiling more data and using new methods.[7−10,14−20] Turco et al.[14] derived the first estimate of global total BC emission (2.2−22 Tg/yr). Bond et al.[7] developed a global BC inventory taking into consideration the variation of EF_{BC} among different fuel types, combustion sources, and emission control technologies. They also provided an uncertainty range of 4.3−22 Tg/yr by using a method to combine uncertainties of different error sources.[7] Novakov et al.[15] made the first attempt to estimate BC emissions back to the 19th century. Bond et al.[9] studied the historic BC emissions from 1850 to 2000 by tracking the change of technology. The methodology used by Bond et al.[9] had been adopted to generate a historical gridded BC inventory, which was adopted

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to run the climate models for the Climate Model Intercomparison Program #5 in support of the IPCC Fifth Assessment report.[18−20] Recently, a high resolution (spatial and sectoral) BC emission inventory for the year 2007 (PKU-BC-2007)[21] was developed following the method by Bond et al.,[7,9] with a detailed fuel consumption data product (PKU-FUEL-2007)[22] and the compiling of recently published EF_{BC} data measured for developing countries. The inventory was used to model ambient air concentrations which were in better agreement with field observations.[21]

Emissions of BC change temporally because both fuel consumption and EF_{BC} change over time.[9,10] In the past 3 decades, the total annual fuel consumption in China has more than quadrupled,[23] making China the number one emitter of both CO_2 and BC.[9,24] In contrast, the coal consumption in Europe has declined since 1985 primarily due to the favor of oil/gas.[10] The values of EF_{BC} for many combustion sources have been decreasing in developed countries because of implementation of stricter standards and improvement of exhaust control technologies. For example, EF_{BC} for on-road vehicles in the United States decreased by more than 1 order of magnitude over the past 40 years.[25] Replacement of old industrial facilities can also substantially reduce BC emissions. A typical example is the phasing out of beehive coke ovens in China since 1996 when these ovens were banned.[26] Thanks to this new regulation, the average EF_{BC} of coke production in China has decreased by more than two fold.[27,28]

In this study, annual BC emissions from 64 sources in 222 countries/territories were estimated for the period from 1960 to 2007 and compared with those reported previously. As one of many efforts to reduce uncertainty of BC emission inventory, recently reported EF_{BC}, especially those measured in China for residential solid fuel stoves in simulated kitchens or on site,[29−42] were included in this study. Although these data have been used in some regional inventories,[28,43,44] they are not included in existing global inventories.[9,10,18−20] Fuel consumption and technology division data in China were updated by using the provincial, instead of national data. Finally, the emission uncertainties were characterized following a Monte Carlo approach for the first time in the global BC inventory.

■ METHODOLOGY

Emission Sources. A technology-based method developed by Bond et al.[7,9] and used by Wang et al.[21,27] was followed to calculate BC emissions from 1960 to 2007 in this study. Emission sources of BC belong to the six sectors that are power plants, industry, residential/commercial (residential hereafter), transportation, agriculture, and wildfires, and five fuel categories of coal, petroleum, natural gas, solid waste, and biomass. They were further divided into 64 types of combustion, as defined in a previous paper.[22] In addition, BC emissions from noncombustion industrial processes including coke production, petroleum refinery, and brick production were also considered.

Fuel Consumptions. Annual consumptions of various fuels from 1960 to 2007 were taken from the International Energy Agency (IEA) data set,[23] except for natural gas flaring,[45] brick production,[46] aluminum production,[47] nonorganized waste incineration,[48] firewood consumptions,[49] agriculture waste combustion in the field (see below), and shipping (global total).[50] National consumptions of coal, oil, natural gas, and solid biofuels in China were taken from the provincial data in a local energy statistics[51] for a period from 1980 to 2007, which differ from the IEA data set.[27] For biomass burning in wildfires, we used the Global Fire Emissions Database version 3 (GFED3) data set for the period from 1997 to 2007,[52] and the RETRO (reanalysis of the tropospheric chemical composition over the past 40 years) biomass burning emission reconstruction data set for the period from 1960 to 1996.[53] The amount of agricultural residues burned in the field were calculated from the crop production by species (wheat, rice, cotton, beans, maize, and others) in China[54] and other countries,[49] region-specified production-to-residue ratios,[55] and the percentage of field burned residues.[7]

Technology Divisions. Bond et al.[7,9] developed a method to consider the *technology division*, which was used in our previous studies[21,27] and was followed in this study to account for the technology splits in different countries during the period from 1960 to 2007. Accordingly, 17 of the 64 emission sources with a large spread of EF_{BC} were divided into subcategories, so that the adequate EF_{BC} could be applied to each individual subcategory. These emission subcategories are coal combustion in power plants and industrial boilers, brick kilns, coke production, on-road motor vehicles (diesel and gasoline), residential firewood, residential coal, and agriculture residue burned in the field. Detailed information on these technology divisions are listed in Table S1 (Supporting Information). In brief, coal consumptions in power plants and industrial boilers were divided into technologies of two types of combustion boilers (pulverized-coal or stoker/cyclone boilers) and three types of control facilities (cyclone, scrubber, electrostatic precipitators). The share of each technology was determined by S-shaped curves with the associated parameters taken from Bond et al.[9] The data of briquette used in the residential sector were compiled for China by province for the period from 1980 to 2007 and by the nation from 1960 to 1979.[51] Since there are only data for four countries excluding China and the average fraction of briquette in residential coal is less than 3%,[46] the briquette used in countries other than China was ignored. For brick kilns, 10% and 100% tunnel kilns were used for developing and developed countries, respectively.[29] For coke ovens, the ratios of beehive to recovery battery coking in China were derived for each year from local statistics,[51,56] and constant ratios were applied for other developing countries (20%) and all developed countries (0). For on-road emissions of diesel and gasoline motor vehicles, a fraction of 10% and 5% of *superemitters*[7,9] (vehicles with high EF_{BC} due to poor maintenance), were adopted for developing and developed countries, respectively. For normal gasoline powered motor vehicles, the fractions of four-stroke vehicles were derived by region from the International Road Federation data set.[57] For firewood, we assumed that 75% of this fuel was burned in fireplaces in Europe and North America, and the other 25% was burned in stoves.[7] In other countries, it was assumed that all the firewood was burned in stoves. Agriculture residues were classified into six different types: crop straws of wheat, rice, cotton, beans, maize, and others, based on the production data in China and other countries.[49,54]

Emission Factors for BC (EF_{BC}). An EF_{BC} data set was compiled from a literature survey of 121 references (57 of them are listed in Supporting Information, Table S2 and the rest are given in a previous study[58]). Many of these EF_{BC}[29−35,38−42,55,59] added were measured in developing countries, which have not been used in existing global BC inventories. Since the direct measurements of EF_{BC} are very limited, the reported emission factors of element carbon measured using the thermal method were also adopted as estimation of EF_{BC}. It should be noted

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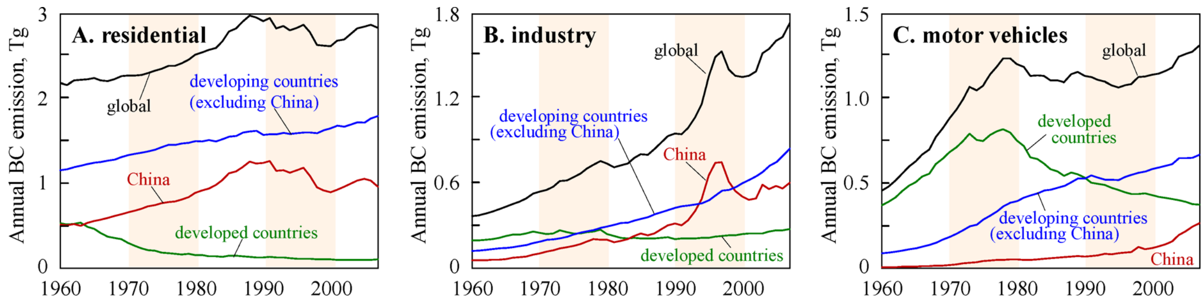

Figure 1. Historical annual BC emissions in the residential sector (A), industrial sector (B), and motor vehicles (C) from 1960 to 2007. The emissions are shown for China (red), other developing countries (blue), developed countries (green), and global total (black).

that uncertainty thus introduced cannot be avoided. After a review of data collected based on the same principle described in previous studies,[27,58] we included a total of 706 EF_{BC} measured in 13 countries (four developing countries and nine developed countries) during a period from 1985 to 2011. We accounted for the spatial and temporal variations of EF_{BC} for motor vehicles and coke production: for normal gasoline (four-stroke) and diesel vehicles, regression models were developed based on per capita gross domestic product (GDP) to predict EF_{BC} for individual countries in different years;[58] the emission factors of fine particles from battery coking ovens were also predicted by a regression model based on per capita GDP,[27] and then converted into EF_{BC} using a constant BC-to-fine particles ratio (0.9).[8] For the remainder of the sources, since the reported EF_{BC} are generally log-normally distributed,[27,58] means and standard deviations of $\log_{10}$-transformed EF_{BC} were derived (see Supporting Information, Table S2) and used in calculating the BC emissions and estimating the uncertainties (see below).

Uncertainty Analysis. A Monte Carlo simulation consisting of 1000 calculations of the BC emission inventory was run to characterize the uncertainty based on the variations of both fuel consumptions and EF_{BC}. The uncertainty in fuel was assessed by assuming uniform distributions with fixed coefficients of variation (30% for nonorganized waste incineration,[7] 20% for residential biofuel,[7] 20% for open biomass burning,[52] 15% for shipping,[50] and 10% for other sources[60,61]). Log-normal uncertainty distributions were applied for EF_{BC} with standard deviations (see above). The Monte Carlo simulation generated estimates of the emissions and their uncertainty through the medians and the interquartile ranges (R_{50}).

RESULTS AND DISCUSSION

Historical Trends of BC Emissions from Various Sectors. On the basis of the fuel consumptions and EF_{BC}, annual BC emissions from residential, industrial, and on-road motor vehicles from 1960 to 2007 were calculated for all countries. Figure 1 shows the temporal trends of global total BC emissions, together with emissions in China, other developing countries, and developed countries. Annual BC emissions for all 222 countries/territories are listed in the Supporting Information Excel file.

The residential sector is the most important anthropogenic BC emission source (Figure 1A). From 1960 to 1988, global BC emissions from the residential sector had increased by 37% from 2226 to 3021 gigagram (Gg). After 1988, the total emissions leveled off but seem to increase again since the mid-1990s to reach another peak of 2956 Gg in 2006. The global upward trend of residential BC emissions before 1988 was primarily controlled by rising emissions from developing countries, due to rapid population growth.[62] After 1988, even though the population of developing countries continued to increase (reaching 1.32 and 5.62 billion in China and all developing countries, respectively, in 2007),[62] annual BC emissions showed a decrease in most developing countries due to fuel shifts in urban areas. In China, for instance, residential coal in cities had been gradually replaced by gas/oil after 1988.[63] Meantime, in rural areas, the consumption of firewood, straw, and coal also decreased since the mid-2000s, largely due to a decrease in rural population caused by urbanization.[62] As a result, the percentage of traditional solid fuels in the residential sector of developing countries had decreased from 82% in 1960 to 70% in 2007 (see Supporting Information, Figure S1). These trends should be accounted for when predicting the future trend of BC emissions.

For the industrial sector (including diesels used in agriculture), the global total emissions were increasing gradually before 1990, even though the emissions from developed countries began to decrease slowly by the end of the 1970s (Figure 1B). This is consistent with that reported by Bond et al.[9] and Junker and Liousse.[10] Thereafter, the emissions surged around 1991, due to rapid economic development and fast growth of energy demand in China. Among all activities, rapid expansion of beehive coke ovens due to increased market demand contributed a large share to this jump (see Supporting Information, Figure S2), which was not found in previous global inventories without using local statistics for this source. Because of the extremely high EF_{BC}, beehive coke ovens contributed 25% of the total BC emissions in China in 1996, even though only 1.2% of the energy was consumed by this activity.[27] As a result, global BC emissions in the industrial sector peaked (1516 Gg) in 1997. In 1996, the Coal Law was enforced in China and beehive coke ovens were banned.[26] Although full compliance with this regulation took several years, and not all the beehive ovens in China had disappeared until the beginning of this century,[56] a sharp decline in the emissions was observed during this period (Figure 1B). After 2000, BC emissions from this sector in China remained roughly stable, but the increase in other developing countries was accelerated.

For motor vehicles (Figure 1C), BC emissions in developed countries showed a broad hump peaking around 1978, driven by two opposite forces of an increasing motor vehicles number but decreasing EF_{BC} per vehicle.[58] For developing countries, the emissions were increasing continuously over the period, reflecting an increase in the number of vehicles (0.86 and 172 million in 1960 and 2010, respectively[64]). Although the EF_{BC}

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for motor vehicles in developing countries also decreased since the 1990s,[65] the increase in vehicle number has so far overridden the reduction of EF_{BC}. In China, for example, the number of motor vehicles increased from 13 million in 2000 to 78 million in 2010.[66] In addition, a variation in the share of gasoline vs diesel vehicles also influenced the trend. For example, due to policy favoring diesel vehicles and the availability of small diesel engines to engines in cars, the ratio of diesel to gasoline vehicles increased from 0.48 to 1.5 in Europe and from 0.06 to 0.38 in North America during the 1960–2007. This shift leads to 4.3% fewer CO_2 emissions, but 59% greater BC emissions, which should be considered for assessing the short versus long-term impacts on climate.[67]

Relationship of Trends in BC Emission and Energy Production. Figure 2 shows the temporal trends of the energy

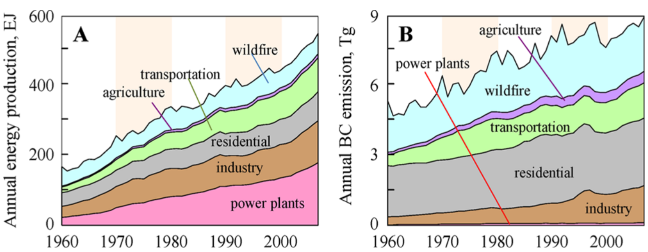

Figure 2. Global temporal trends of annual energy productions (A) and BC emissions (B) in various sectors from 1960 to 2007.

consumptions and associated BC emissions from 1960 to 2007 from six sectors. Global BC emissions from all sources were 5386 (3434–8488 as R_{50}) and 9052 (5598–14437) Gg in 1960 and 2007, respectively. Anthropogenic BC emissions amounted to 3196 Gg in 1960 and increased by 110% to an all time high of 6708 Gg by 2007. Meanwhile, annual anthropogenic energy production increased from 112 EJ in 1960 to 492 EJ in 2007, that is, a 339% increase. Previous studies found that the emissions were increasing before 1990, which leveled off thereafter.[9,10,15] Our study confirmed this fact, but moreover, noticed some unrevealed fluctuations due to technology changes in China. For example, the transfer of coal to oil/gas in Chinese cities in the late 1980s led to an emission peak in 1987 (7835 Gg). Similarly, the increase of beehive coking ovens in early 1990s followed by a control since 1996 resulted in a first-increasing and later-decreasing trend in the global total emissions in that decade.

The changes in EF_{BC} for various sectors are reflected in trends of BC emission intensities, defined as the amount of BC emitted per unit of energy production. Changes of BC emission intensities since 1960 are shown in Figure 3. Among various sectors, BC emission intensities decreased dramatically in power plants (−44%), residential sector (−38%), and transportation (−48%) during this period, owing to the fuel shift as well as technology improvements. For instance, the percentage of residential energy produced by oil and gas had increased from 62% to 90% in developed countries and from 18% to 30% in developing countries over the period. Meantime, improvement in combustion technology is also important, especially for transportation. For example, the EF_{BC} of motor vehicles had decreased because of the introduction of the exhaust catalytic converter,[68] the enhancement of transmission configuration of diesel engines,[69] and the application of injection timing retardation technology.[70] By contrast, the increase of some high-emission activities brought the emission intensity up in the

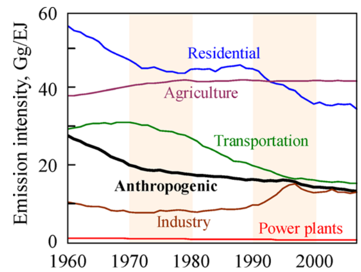

Figure 3. Global trends in BC emission intensities (defined as the amount of BC emitted per unit of energy production) in various sectors and on average for all anthropogenic sources from 1960 to 2007.

industrial and agricultural sectors. For instance, during 1960–2007, the production of coke and brick in China, with relatively high EF_{BC}, had increased by 35 and 8 folds.[51] Similarly, the global consumption of diesel in agriculture had increased by 5 folds during this period.[23] However, the overall BC emission intensity for all anthropogenic sectors had decreased by 52%, more than those of all the individual sectors, due to the increase in the fraction of fuel consumed in power plants.

Comparison with Previous BC Emission Inventories. The historical trend in global anthropogenic BC emissions from this study (PKU-BC) is compared with previous estimates over the period in Figure 4. The best estimations as medians and

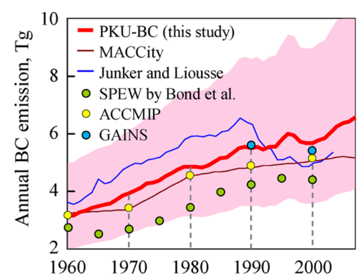

Figure 4. Comparison of annual anthropogenic BC emissions from 1960 to 2007 between this study (PKU-BC) and those previously reported, including ACCMIP,[18] MACCity,[19,20] SPEW by Bond et al.,[7,9,11] GAINS,[17] and Junker and Liousse.[10] The annual BC emissions and uncertainties are shown as median values (red line) and R_{50} (shaded area) from a Monte Carlo simulation. Annual emission data are available for MACCity (purple line) and from Junker and Liousse (blue line), while SPEW (green solid circles) and ACCMIP (yellow solid circles) provide emissions with a 5-year and 10-year resolution, respectively. GAINS (blue solid circles) provides estimates for 2 years (1990 and 2000).

uncertainties as R_{50} derived from a Monte Carlo simulation in PKU-BC are compared with those in ACCMIP[18] and MACCity[19,20] prepared for the Atmospheric Chemistry and Climate Model Intercomparison Project, those in SPEW (Speciated Pollutant Emissions Wizard) by Bond et al.,[7,9,11] those reported by Junker and Liousse,[10] and those in GAINS (for 1990 and 2000).[17]

The overall temporal trend derived in this study is similar to those of ACCMIP,[18] MACCity,[19,20] and SPEW,[7,9,11] all of which show a general increase from 1960 to 2007. The only exception is that reported by Junker and Liousse.[10] Based on their results, the emissions had increased from 1960 to 1988, followed by a sharp decrease up to 1999.[10] As a result, the annual emissions by Junker and Liousse are 10–24% higher than our estimates for the period from 1960 to 1988 and 11–

16% lower than our estimates for the period from 1995 to 2003. Lamarque et al.[18] had compared different inventories, and noticed that EF_{BC} of coal in power plants and industrial boilers used by Junker and Liousse are higher than other inventories, which is likely the reason for their high-bias prior to 1988.[10]

Although the general trends in global BC emissions are similar between this study and previous work, we found annual BC emissions in this study are higher than those estimated by ACCMIP,[18] MACCity,[19,20] and SPEW,[7,9,11] and close to estimates by GAINS.[17] On average, our annual BC emissions (median estimate) are 11% and 16% higher than those of MACCity and SPEW, respectively, mainly due to the differences in EF_{BC} for residential sources and diesel vehicles in developing countries. For example, a constant fraction (20%) of BC in fine particles was used to derive the EF_{BC} for wood stoves because of the lack of direct measurements for this source in the inventory by Bond et al.[7] However, on the basis of data published recently,[33] a relatively higher value (40−70%) was reported. In addition to EF_{BC}, a difference can also be found in some energy statistics used between our study and previous inventories. For instance, fuel consumptions in the United States and China were from the national statistics in the IEA[23] or United Nations[71] database in previous inventories,[12,16,18] while provincial data from local energy statistics were used in our study. It should be noted that the national fuel consumptions in China could, to a certain extent, be underestimated by the IEA database.[60,72]

We have similar estimations and the trends of BC emissions from wildfires with other inventories[18−20,52,53] (Supporting Information, Figure S3) because the same published GFED3 data and similar data of EF_{BC} were applied. In a recent study, Bond et al. constrained the BC emissions based on the observed BC light absorption, and scaled the bottom-up estimated BC emissions by factors ranging from 1.1 to 4.1 in different regions.[11] The results are compared with ours in Table 1. Although our estimations in East Asia, South Asia, Middle East, and the global total are higher than those from SPEW, large differences among those compared can be seen, suggesting that there is still large room for further improvements in the inventory.

Historical Shifts of BC Emissions and Their Drivers between Groups of Countries. Temporal trends in the produced energy, BC emissions, and BC emission intensities in North America, South America, Africa, Europe, and Asia (China, India, and other Asian countries) are shown in Figure 5. Bond et al.[9] and Novakov et al.[15] reported that North America, Former USSR, and Europe dominated the BC emissions in the early industrial era (before 1960), and the emissions in China and other Asian countries increased rapidly in the latter half of the 20th century. In general, we observed similar trends (Figure 5B). To isolate the driving forces from increasing fuel consumption and technology changes, we further studied the evolution of BC emission intensities by region (Figure 5C).

Globally, the increase of energy productions was faster than that of BC emissions. As discussed above, this occurs because most of the increase in energy is in power plants with lower EF_{BC} than other sources. In India for instance, energy production increased steadily over the period 1960−2007 (Figure 5A), driving an increase of BC emissions. However, the growth of the energy (4% per year) was faster than that of BC emissions (2% per year). The trends in other Asian countries (except China and India) were similar to India. In China, even though both energy and BC emissions increased, the emission intensity decreased faster than that in India during 1960−2007. In fact, a decrease of BC emission intensity in China is the largest among all countries/groups of countries shown in Figure 5C. In 1960, the Chinese BC emission intensity was as high as 83 Gg/EJ, and was divided by a factor of 4 to a value of 21 Gg/EJ in 2007. This decrease has even accelerated since 1996 due to fuel shifts and technology changes (see above). In contrast, the trend of BC emission intensity in Africa was similar to that of India for the period from 1960 to the later 1980s. By 2007, the BC emission intensity in Africa was the highest due to a large amount of biofuel consumption. In North America and Europe, the BC emission intensities decreased slowly after 1970. By 2007, the emission intensity of North America (4.3 Gg/EJ) and Europe (6.8 Gg/EJ) were similar to each other and were approximately 20−30% of those in China and India. The regional difference in the trend of BC emission intensity among regions, as shown in Figure 5C, reflects the shifts in the share of each region to global energy as well as to BC emissions. For example, the contribution to global energy had decreased from 37% to 21% in North America, but increased from 21% to 44% in Asia. Meantime, the contribution to BC emissions decreased from 14% to 6.8% in North America, but increased from 38% to 60% in Asia.

Figure 6 panels A and B show the geographical distributions of annual anthropogenic BC emission density (emission per unit area over grid cells with population density >1 cap per km^2 using the 2007 gridded population data set[73]) between 1960 and 2007. Since the emission density over only populated areas are shown, high values are found not only in countries with large emissions, but also in countries with population highly concentrated in small cities, such as Egypt and Libya (Figure 6B). The differences by sector are shown in Figure S4. The difference between 2007 and 1960 is shown in Figure 6C. Junker and Liousse[10] reported that there was a fast increase in the BC emission density in semideveloped countries, such as China and India, and a decrease over North America and Europe. This trend were also found in our study. Globally, a large increase of BC emission densities occurred in developing countries, such as Cambodia (+0.37 $g/(m^2/yr)$), China (+0.30 $g/(m^2/yr)$), and India (+0.13 $g/(m^2/yr)$), while a large decrease was found in developed countries like UK (−1.1 g/

Table 1. Comparison of BC Emissions ($Gg\ yr^{-1}$) by Region among Those Estimated by Bond et al. (SPEW),[9,11] Constrained by Observations,[11] and Derived from This Study (PKU-BC)

	SPEW[9,11]	from observations[11]	PKU-BC
	2000	2000−2010	2000−2007
North America	420	500	420
Latin America	990	2000	1018
Middle East	80	140	156
Africa	2020	4400	1973
Europe	460	510	506
EECCA[a]	550	600	451
South Asia	630	2580	857
East Asia	1450	2820	2076
Southeast Asia	780	2200	703
Pacific	320	1200	202
global	**7700**	**16950**	**8363**

[a]EECCA for Eastern Europe, Caucasus and Central Asia.

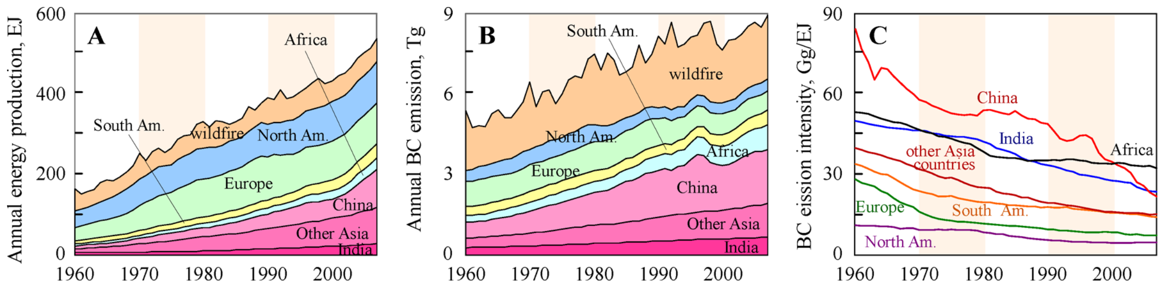

Figure 5. Trends in annual energy production (A), annual BC emission (B), and anthropogenic BC emission intensity (C) in various regions/countries (North America, South America, Africa, Europe, China, India, and other Asian countries) from 1960 to 2007.

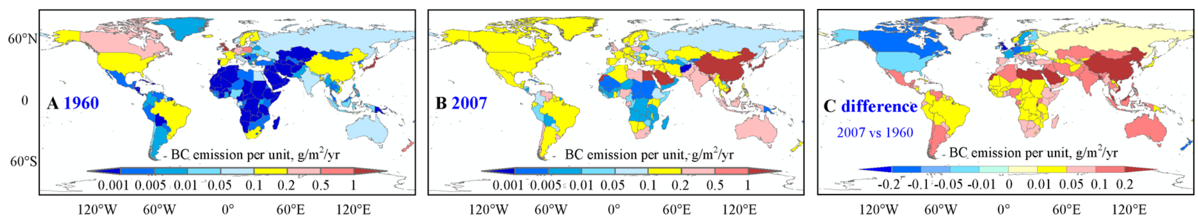

Figure 6. Comparison of geographical distributions of annual anthropogenic BC emission density (emission per unit area over grid cells with population density >1 cap per km^2 using the 2007 gridded population data set[73]) between 1960 and 2007. (A) Country-average BC emission density in 1960, (B) country-average BC emission density in 2007, (C) differences in country-average BC emission densities between 2007 and 1960.

(m^2/yr)), Denmark (−0.27 g/(m^2/yr)), and Netherlands (−0.21 g/m^2/yr) due to the reasons discussed above.

Implications. With the newly compiled fuel consumption and EF_{BC} data set, time series of global BC emissions were generated in this study, which provide information for climate forcing study, human exposure assessment, and abatement strategy development. Similar to previous studies, we confirmed that BC emissions in developed countries have decreased significantly in the past. In developing countries, even though total BC emissions have increased continuously, the BC emission intensities have started to decrease due to the improvements in technology and the change in fuel composition. Such a trend will eventually lead to a decrease in total BC emissions in the near future. In addition, rapid urbanization rates in China and also other developing countries in economic transitions are favorable for BC emission reduction. It is also noted that fuel consumptions in residential and agricultural sectors have the highest BC emission intensities, and subsequently the highest reduction potential.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Technology splits in PKU-BC; EF_{BC} data set used in PKU-BC; temporal trends of energy production and fuel composition in residential sector; temporal trends of coke production by beehive and nonbeehive coking plants; comparison of BC emissions from wildfires among different emission inventories; difference in country-average BC emission density between 2007 and 1996 in three major sectors. Annual BC emissions for 222 countries/territories from 1960 to 2007 are listed in an Excel file. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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■ REFERENCES

(1) Boucher, O.; Randall, D.; Artaxo, P.; Bretherton, C.; Feingold, G.; Forster, P.; Kerminen, V.-M.; Kondo, Y.; Liao, H.; Lohmann, U.; et al. Clouds and Aerosols. In *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. Stocker, T. F., et al. Eds.; Cambridge Univ. Press: Cambridge and New York, 2013.

(2) United States Environmental Protection Agency. *Black Carbon Effects on Public Health and the Environment*. Report to Congress on Black Carbon. 2012; www.epa.gov/blackcarbon/2012report/Chapter3.pdf (accessed August 22, 2013).

(3) Janssen, N. A. H.; Gerlofs-NijlandM. E.; Lanki, T.; Salonen, R. O.; Cassee, F.; Hoek, G.; Fischer, P.; Brunekreef, B.; Krzyzanowski, M. *Health Effects of Black Carbon*; WHO Report; World Health Organization: Copenhagen, Denmark, 2012

(4) Anenberg, S. C.; Schwartz, J.; Shindell, D.; Amann, M.; Faluvegi, G.; Klimont, Z.; Janssens-Maenhout, G.; Pozzoli, L.; van Dingenen, R.; vignati, E.; et al. Global air quality and health co-benefits of mitigating near-term climate change through methane and black carbon emission controls. *Environ. Health. Persp.* **2012**, *120* (6), 831−839.

(5) Shindell, D.; Kuylenstierna, J. C. I.; Vignati, E.; van Dingenen, R.; Armann, M.; Kilmont, Z.; Anenberg, S. C.; Muller, N.; Janssens-Maenhout, G.; Raes, F.; et al. Simultaneously mitigating near-term climate change and improving human health and food security. *Science* **2012**, *335*, 183−189.

(6) Penner, J. E.; Eddleman, H.; Novakov, T. Towards the development of a global inventory for black carbon emissions. *Atmos. Environ.* **1993**, *27A* (8), 1277−1295.

(7) Bond, T. C.; Streets, D. G.; Yarber, K. F.; Nelson, S. M.; Woo, J. H.; Klimont, Z. A technology-based global inventory of black and organic carbon emissions from combustion. *J. Geophys. Res.* **2004**, *109*, D14203.

(8) Kupiainen, K.; Klimont, Z. *Primary Emissions of Submicron and Carbonaceous Particles in Europe and the Potential for their Control*;

Interim Report IR-040079; International Institute for Applied Systems Analysis: Laxenburg, Austria, 2004.
(9) Bond, T. C.; Bhardwaj, E.; Dong, R.; Jogani, R.; Jung, S. K.; Roden, C.; Streets, D. G.; Trautmann, N. M. Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850−2000. *Global Biogeochem. Cycles* **2007**, *21*, 2006GB002840.
(10) Junker, C.; Liousse, C. A global emission inventory of carbonaceous aerosol from historic records of fossil fuel and biofuel consumption for the period 1860−1997. *Atmos. Chem. Phys.* **2008**, *8*, 1195−1207.
(11) Bond, T. C.; et al. Bounding the role of black carbon in the climate system: A scientific assessment. *J. Geophys. Res.* **2013**, *118* (11), 5380−5552.
(12) Myhre, G.; et al. Radiative forcing of the direct aerosol effect from AeroCom Phase II simulations. *Atmos. Chem. Phys.* **2013**, *13*, 1853−1877.
(13) Chung, C. E.; Ramanathan, V.; Decremer, D. Observationally constrained estimates of carbonaceous aerosol radiative forcing. *Proc. Natl. Acad. Sci. U.S.A.* **2012**, *109*, 11624−11629.
(14) Turco, R. P.; Toon, O. B.; Whitten, R. C.; Hamill, P. The global cycle of particulate elemental carbon. In *Precipitation Scavenging, Dry Deposition, and Resuspension*; Pruppacher, H. R., Semonin, R. G., Slinn, W. G. N., Eds.; Elsevier Sci.: New York, 1983.
(15) Novakov, T.; Ramanathan, V.; Hansen, J. E.; Kirchstetter, T. W.; Sato, M.; Sinton, J. E.; Satahye, J. A. Large historical changes of fossil-fuel black carbon aerosols. *Geophys. Res. Lett.* **2003**, *30* (6), 1324.
(16) Ito, A.; Penner, J. Historical emissions of carbonaceous aerosols from biomass and fossil fuel burning for the period 1870−2000. *Global Biogeochem. Cycles* **2005**, *19*, doi:10.1029/2004GB002374.
(17) Cofala, J.; Amann, M.; Klimont, Z.; Kupiainen, K.; Hoglund-Isaksson, L. Scenarios of global anthropogenic emissions of air pollutants and methane until 2030. *Atmos. Environ.* **2007**, *41* (38), 8486−8499.
(18) Lamarque, J.-F.; Bond, T. C.; Eyring, V.; Granier, C.; Heil, A.; Klimont, Z.; Lee, D.; Liousse, C.; Mieville, A.; Owen, B.; et al. Historical (1850−2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application. *Atmos. Chem. Phys.* **2010**, *10*, 7017−7039.
(19) Granier, C.; Bessagnet, B.; Bond, T.; D'Angiola, A.; Denier van der Gon, H.; Frost, G. J.; Heil, A.; Kaiser, J.; Kinne, S.; Klimont, Z.; et al. Evolution of anthropogenic and biomass burning emissions of air pollutants at global and regional scales during the 1980−2010 period. *Climatic Change* **2011**, *109*, 163−190.
(20) Diehl, T.; Heil, A.; Chin, M.; Pan, X.; Streets, D.; Schultz, M.; Kinne, S. Anthropogenic, biomass burning, and volcanic emissions of black carbon, organic carbon, and SO_2 from 1980 to 2010 for hindcast model experiments. *Atmos. Chem. Phys. Discuss.* **2012**, *12* (9), 24895−24954.
(21) Wang, R.; Tao, S.; Balkanski, Y.; Ciais, P.; Boucher, O.; Liu, J.; Piao, S.; Shen, H.; Vuolo, M. R.; Valari, M.; et al. Exposure to ambient black carbon derived from a unique inventory and high resolution model. *Proc. Natl. Acad. Sci. U.S.A.* **2014**, *111* (7), 2459−2463.
(22) Wang, R.; Tao, S.; Ciais, P.; Shen, H. Z.; Huang, Y.; Chen, H.; Shen, G. F.; Wang, B.; Li, W.; Zhang, Y. Y.; et al. High resolution mapping of combustion processes and implications for CO_2 emissions. *Atmos. Chem. Phys.* **2013**, *13*, 5189−5203.
(23) International Energy Agency. IEA World Energy Statistics and Balances. http://www.oecd-ilibrary.org/statistics (accessed August 22, 2013).
(24) Guan, D.; Peters, G. P.; Weber, C. L.; Hubacek, K. Journey to world top emitter: An analysis of the driving forces of China's recent CO_2 emissions surge. *Geophys. Res. Lett.* **2009**, *36*, doi: 10.1029/2008GL036540.
(25) Kirchstetter, T. W.; Aguiar, J.; Tonse, S.; Fairley, D.; Novakov, T. Black carbon concentrations and diesel vehicle emission factors derived from coefficient of haze measurements in California: 1967−2003. *Atmos. Environ.* **2008**, *42* (3), 480−491.
(26) China Law Press. *Law of the People's Republic of China on the Coal Industry*; China Law Press: Beijing, 2001.
(27) Wang, R.; Tao, S.; Wang, W.; Liu, J.; Shen, H.; Shen, G.; Wang, B.; Liu, X.; Li, W.; Huang, Y.; et al. Black carbon emissions in China from 1949 to 2050. *Environ. Sci. Technol.* **2012**, *46*, 7595−7603.
(28) Zhang, Q.; Streets, D. G.; Carmichael, G. R.; He, K. B.; Huo, H.; Kannari, A.; Klimont, Z.; Park, I. S.; Reddy, S.; Fu, J. S.; et al. Asian emissions in 2006 for the NASA INTEX-B mission. *Atmos. Chem. Phys.* **2009**, *9*, 4081−4139.
(29) Bi, X. H.; Simoneit, B. R. T.; Sheng, G. Y.; Fu, J. M. Characterization of molecular markers in smoke from residential coal combustion in China. *Fuel* **2008**, *87*, 112−119.
(30) Cao, G. L.; Zhang, X. Y.; Gong, S. L.; Zheng, F. C. Investigation on emission factors of particulate matter and gaseous pollutants from crop residue burning. *J. Environ. Sci.* **2008**, *20*, 50−55.
(31) Chen, Y. J.; Zhi, G. R.; Feng, Y. L.; Liu, D. Y.; Zhang, G.; Li, J.; Sheng, G. Y.; Fu, J. M. Measurements of black and organic carbon emission factors for household coal combustion in China: Implication for emission reduction. *Environ. Sci. Technol.* **2009**, *43*, 9495−9500.
(32) Li, X. H.; Wang, S. X.; Duan, L.; Hao, J. M.; Li, C.; Chen, Y. S.; Liu, Y. Particulate and trace gas emissions from open burning of wheat straw and corn stover in China. *Environ. Sci. Technol.* **2007**, *41*, 6052−6058.
(33) Li, X. H.; Wang, S. X.; Duan, L.; Hao, J. M.; Nie, Y. F. Carbonaceous aerosol emissions from household biofuel combustion in China. *Environ. Sci. Technol.* **2009**, *43*, 6076−6081.
(34) Liu, Y.; Zhang, Y. X.; Wei, Y. J.; Dou, H.; Gu, D. S.; Zeng, L. M.; Shao, M. Measurement of emission factors of carbonaceous aerosols from residential coal combustion. *Acta Sci. Circumst.* **2007**, *27* (9), 1409−1415.
(35) Olivares, G.; Strom, J.; Johansson, C.; Gidhagen, L. Estimates of black carbon and size-resolved particle number emission factors from residential wood burning based on ambient monitoring and model simulations. *J. Air Waste Manage. Assoc.* **2008**, *58*, 838−848.
(36) Roden, C. A.; Bond, T. C.; Conway, S.; Pinel, A. B. O.; MacCarty, N.; Still, D. Laboratory and field investigations of particulate and carbon monoxide emissions from traditional and improved cookstoves. *Atmos. Environ.* **2009**, *43* (6), 1170−1181.
(37) Johnson, M.; Edwards, R.; Frenk, C. A.; Masera, O. In-field greenhouse gas emissions from cookstoves in rural Mexican households. *Atmos. Environ.* **2008**, *42*, 1206−1222.
(38) Saud, T.; Gautam, R.; Mandal, T. K.; Gadi, R.; Singh, D. P.; Sharma, S. K.; Dahiya, M.; Saxena, M. Emission estimates of organic and elemental carbon from household biomass fuel used over the Indo-Gangetic-plain (IGP), India. *Atmos. Environ.* **2012**, *61*, 212−220.
(39) Shen, G. F.; Yang, Y.; Wang, W.; Tao, S.; Zhu, C.; Min, Y.; Xue, M.; Ding, J.; Wang, B.; Wang, R.; et al. Emission factors of particulate matter and elemental carbon for crop residues and coals burned in typical household stoves in China. *Environ. Sci. Technol.* **2010**, *44*, 7157−7162.
(40) Shen, G. F.; Wei, S. Y.; Wang, W.; Zhang, Y. Y.; Min, Y. J.; Wang, B.; Wang, R.; Li, W.; Shen, H. Z.; Huang, Y.; et al. Emission Factors, size distributions, and emission inventories of carbonaceous particulate matter from residential wood combustion in Rural China. *Environ. Sci. Technol.* **2012**, *46*, 4207−4214.
(41) Shen, G. F.; Tao, S.; Wei, S.; Chen, Y.; Zhang, Y; Shen, H.; Huang, Y.; Zhu, D.; Yuan, C.; Wang, H.; et al. Field measurement of emission factors of PM, EC, OC, Parent, Nitro-, and Oxy- Polycyclic Aromatic Hydrocarbons for residential briquette, coal cake, and wood in rural Shanxi, China. *Environ. Sci. Technol.* **2013**, *47* (6), 2998−3005.
(42) Zhang, Y. X.; Schauer, J. J.; Zhang, Y. H.; Zeng, L. M.; Wei, Y. J.; Liu, Y.; Shao, M. Characteristics of particulate carbon emissions from real-world Chinese coal combustion. *Environ. Sci. Technol.* **2008**, *42*, 5068−5073.
(43) Lu, Z.; Zhang, Q.; Streets, D. G. Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996−2010. *Atmos. Chem. Phys.* **2011**, *11*, 9839−9864.
(44) Qin, Y.; Xie, S. D. Spatial and temporal variation of anthropogenic black carbon emissions in China for the period 1980−2009. *Atmos. Chem. Phys.* **2012**, *12* (11), 4825−4841.

(45) National Oceanic and Atmospheric Administration. Global Gas Flaring Estimates. http://www.ngdc.noaa.gov/dmsp/interest/gas_flares.html (accessed August 22, 2013).
(46) United Nations Industrial Development Organization. *International Yearbook of Industrial Statistics 2008*. Edward Elgar Publishing: Cheltenham, U.K., 2008.
(47) U.S. Geological Survey. Commodity Statistics and Information. http://minerals.usgs.gov/minerals/pubs/commodity (accessed August 22, 2013).
(48) United Nations Statistics Division. *Environmental Indicators: Waste*; http://unstats.un.org/unsd/environment/qindicators.htm. (accessed August 22, 2013).
(49) Food and Agriculture Organization of the United Nations. Statistics at FAO; http://faostat.fao.org/site/291/default.aspx (accessed August 22, 2013).
(50) Endresen, Ø. E.; Sørgård, H. L.; Behrens, P. O.; Brett, P. O.; Isaksen, I. S. A. A historical reconstruction of ships' fuel consumption and emissions. *J. Geophys. Res.* **2007**, *112*, D12301.
(51) National Bureau of Statistics and National Energy Administration. *China Energy Statistical Yearbook, 1986, 1989−2008*; China Statistics Press: Beijing, 2009.
(52) van der Werf, G. R.; Randerson, J. T.; Giglio, L.; Collatz, G. J.; Mu, M.; Kasibhatla, P. S.; Morton, D. C.; DeFries, R. S.; Jin, Y.; van Leeuwen, T. T. Global fire emissions and the contribution of deforestation, savanna, forest, agriculture, and peat fires. *Atmos. Chem. Phys.* **2010**, *10*, 16153−16230.
(53) Schultz, M. G.; Heil, A.; Hoelzemann, J. J.; Spessa, A.; Thonicke, K.; Goldammer, J. G.; Held, A. C.; Pereira, J. M. C.; van het Bolscher, M. Global wildland fire emissions from 1960 to 2000. *Global Biogeochem. Cycles* **2008**, *22*, GB2002 DOI: 10.1029/2007GB003031.
(54) National Bureau of Statistics Rural Social Economic Survey. *Compilation and Agricultural Statistics in the Three Decades of Reform and Opening-up*; China Statistics Press: Beijing, 2007.
(55) Cao, G. L.; Zhang, X. Y.; Zheng, F. C. Inventory of black carbon and organic carbon emissions from China. *Atmos. Environ.* **2006**, *40*, 6516−6527.
(56) . *Coke Production Database in China*; Association of Iron and Steel Industry: Beijing, 2010.
(57) *World Road Statistics 2009*; International Road Federation: Geneva, 2009.
(58) Wang, R.; Tao, s.; Shen, H.; Wang, X.; Li, B.; Shen, G.; Wang, B.; Li, W.; Liu, X.; Huang, Y.; et al. Global emission of black carbon from motor vehicles from 1960 to 2006. *Environ. Sci. Technol.* **2012**, *46*, 1278−1284.
(59) Kleeman, M. J.; Robert, M. A.; Riddle, S. G.; Fine, P. M.; Hays, M. D.; Schauer, J. J.; Hannigan, M. P. Size distribution of trace organic species emitted from biomass combustion and meat charbroiling. *Atmos. Environ.* **2008**, *42*, 3059−3075.
(60) Akimoto, H.; Ohara, T.; Kurokawa, J.; Horii, N. Verification of energy consumption in China during 1996−2003 by using satellite observational data. *Atmos. Environ.* **2006**, *40*, 7663−7667.
(61) Ciais, P.; Paris, J. D.; Marland, G.; Peylin, P.; Piao, S. L.; Levin, I.; Pregger, T.; Scholz, Y.; Friedrich, R.; Rivier, L.; et al. The European carbon balance, Part 1: fossil fuel emissions. *Global Change Biol.* **2010**, *16*, 1395−1408.
(62) The World Bank. World Development Indicators. http://ddpext.worldbank.org/ext/DDPQQ (accessed August 22, 2013).
(63) Ministry of Environmental Protection of the People's Republic of China. *China Environment Yearbook 2008*; China Environment Yearbook, Inc.: Beijing, 2008.
(64) Davis, S. C.; Diegel, S. W.; Boundy, R. G. *Transportation Energy Data Book*, 31st ed. ORNL-6986. Oak Ridge National Laboratory: Oak Ridge, TN, July 2012.
(65) Subramanian, R.; Winijkul, E.; Bond, T. C.; Thiansathit, W.; Oanh, N. T. K.; Paw-armart, I.; Duleep, K. G. Climate-relevant properties of diesel particulate emissions: Results from a piggyback study in Bangkok, Thailand. *Environ. Sci. Technol.* **2009**, *43*, 4213−4218.
(66) China Automobile Industry Association. *China Automotive Industry Yearbook, 1985−2008*; China Machine Press: Beijing, 2008.
(67) Olivier, B.; Reddy, M. S. Climate trade-off between black carbon and carbon dioxide emissions. *Energy Policy* **2008**, *36*, 193−200.
(68) Hildemann, L. M.; Markowskl, G. R.; Cass, G. R. Chemical composition of emissions from urban sources of fine organic aerosol. *Environ. Sci. Technol.* **1991**, *25*, 744−759.
(69) Prucz, J. C.; Clark, N. N.; Gautam, M.; Lyons, D. W. Exhaust emissions from engines of the Detroit Diesel Corporation in transit buses: A decade of trends. *Environ. Sci. Technol.* **2001**, *35*, 1755−1764.
(70) Lloyd, A. C.; Cackette, T. A. Diesel engines: Environmental impact and control. *J. Air Waste Manage. Assoc.* **2001**, *51*, 809−847.
(71) United Nations Statistics Division. *United Nations Energy Statistics, 1950−1995*. New York, 1995.
(72) Guan, D.; Liu, Z.; Geng, Y.; Lindner, S.; Hubacek, K. The gigatonne gap in china's carbon dioxide inventories. *Nat. Clim. Change* **2012**, *2*, 672−675.
(73) Oak Ridge National Laboratory (ORNL). *LandScan Global Population 2007 Database*. ORNL, 2008; http://www.ornl.gov/sci/landscan/ (accessed October 1, 2010).