Spatial distribution of black carbon emissions in China

ZHANG Nan, QIN Yue & XIE ShaoDong*

College of Environmental Sciences & Engineering, State Key Joint Laboratory of Environmental Simulation and Pollution Control, Peking University, Beijing 100871, China

Received November 2, 2012; accepted February 18, 2013; published online May 3, 2013

Based on the official statistics, locally measured emission factors, and the vehicular emission factor model most suitable for China, we developed a black carbon (BC) emission inventory for 2008 in China and at a spatial resolution of $0.5^{\circ} \times 0.5^{\circ}$. In 2008, the total BC emissions in China were 1604.94 Gg. Industry and the residential sector were the dominant contributors, estimated at 695.03 Gg and 636.02 Gg of BC, respectively. Together, these two source types contributed 82.9% of the total emissions. Emissions from transportation were 194.63 Gg, accounting for 12.1% of the total. Since emission contributions from different sectors showed significant spatial diversity among the 31 administrative districts, we divided the districts into four categories: industry contribution district, residential contribution district, industry and residential contribution district, and transportation contribution district. As for energy consumption, coal and biofuel contributed 51.0% and 32.2%, respectively, of the total emissions. Spatially, BC emissions in China were unevenly distributed, higher in the east and lower in the west, corresponding to regional economic development and rural population density. High emission districts, covering 5.7% of the territory, contributed 41.2% of the total emissions. Shanxi, Hebei, Shandong, Henan, and Sichuan were the largest contributors to national BC emissions.

black carbon, emission inventory, emission factor, spatial distribution, China

Citation: Zhang N, Qin Y, Xie S D. Spatial distribution of black carbon emissions in China. *Chin Sci Bull*, 2013, 58: 3830–3839, doi: 10.1007/s11434-013-5820-4

Black carbon (BC) is an important light-absorbing component of atmospheric aerosols, produced by the incomplete combustion of carbonaceous material [1,2]. Not only does BC affect regional and global climate change [3,4], but it reduces atmospheric visibility [5] and grain production [6], and leads to adverse human health outcomes [7] and the corrosion of buildings [8]. BC can absorb the light in the infrared to visible range [9,10]. Haywood and Ramaswamy [11] estimated its direct forcing as 0.400 W m^{-2} , while Jacobson [3] calculated the direct forcing of BC to be 0.55 W m^{-2} , just less than carbon dioxide. In addition, BC aerosols influence cloud albedo and cloud amount, thereby also indirectly affecting both regional and global climate [12,13]. Due to the short lifetime of BC aerosols in the atmosphere, a reduction in their emissions may have an immediate impact on climate change mitigation [14,15].

China is one of the world's largest contributors of BC, emitting approximately one-quarter of the global emissions [16]. Since pollutants are transported from the Eurasian continent to North America via midlatitude westerly winds, high BC emissions from China are likely to have a significant influence on global BC concentrations [17], which in turn leads to international pressure in terms of climate change negotiations and policy development. The Chinese government enacted the "China National Project to Respond to Climate Change" in 2007, which emphasized technological progress and innovation as means to mitigate global climate change. Because BC is an important contributor to climate forcing, controlling its emissions can help to control global warming. Emission inventories generally provide the emissions and spatial distribution of emission sources. Therefore, establishing a BC emissions inventory with a relevant spatial distribution is of great importance in formulating policies to control BC emissions and in negotiat-

*Corresponding author (email: sdxie@pku.edu.cn)

ing international treaties. Because of the 2008 Olympic Games, China has taken many actions to reduce pollutants emissions, including industrial restructuring and emission-control technologies updating. A detailed analysis of the 2008 BC emissions is now needed.

Many studies have investigated the Chinese BC emission inventory. Bond et al. [18] established a Chinese BC emission inventory for 1996 by considering fuel, combustion type, control technology, and regional features. Street et al. [19] estimated and predicted China's BC emissions in 1995 and 2020. Cao et al. [20,21] estimated the total Chinese BC emissions for 2000 and 2007 as 1499.4 Gg and 1399 Gg, respectively, using some locally derived emission factors. Zhang et al. [22] updated anthropogenic BC emission inventory for 2006 based on Street et al.'s study [23] by accounting for influences in technological progress and obtained a total BC emissions estimate of 1811 Gg. Some studies have estimated the multiyear BC emissions for China and presented their temporal variation [24–28]. However, due to the difficulty in collecting reliable activity data, and the lack of locally measured emission factors, these results still yielded large uncertainties. In addition, the updating of vehicular emission standards was often neglected in these studies [23,24], which influenced vehicular emission factors markedly. Zhang et al. [22] and Lei et al. [25] calculated particulate emission factors under different emission regulations using the MOBILE model. However, compared with MOBILE, the COPERT model is considered to be more suitable for countries with different emission standards and fewer traffic data [29]; therefore, emission factors from the COPERT model were used in our work to estimate BC emissions.

In this study, official activity data were collected, an emission factor database based on local-measured data was established, and a high resolution ($0.5^\circ \times 0.5^\circ$) BC emission inventory was developed for China (excluding Hong Kong, Macao, and Taiwan).

1 Methodology

Calculations of BC emissions were aggregated into five categories: industry, power generation and heating, biomass burning, residences, and transportation. Emission sources were divided according to fuel type, including fossil fuels (coal, oil, and gases), biofuel (crop residues, firewood, and biogas), and biomass (open burning of agricultural waste, and forest and grassland fires).

1.1 Emissions estimation

The BC emissions for China in 2008 were calculated by combining emission factors and activity data for each sector. Detailed calculation methods for industry, power generation and heating, biomass burning, residential sector, and avia-

tion are referred to Qin and Xie [27,30]. Vehicular emissions were estimated by considering not only vehicle types and fuels, but also updates to vehicular emission regulations, as presented in eq. (1),

$$Q_v = \sum_i \sum_j \sum_k (VP_{i,j,k} VMT_i EF_{i,j,k}), \quad (1)$$

where Q_v is the BC emissions from vehicles; VP is the vehicle population; VMT is the annual average mileage; EF is the BC emission factor; and i , j , and k represent the vehicle type, the province, and the emission regulation, respectively.

1.2 Emission allocation

Using GIS technology, provincial BC emissions were allocated at the county level, based on various spatial proxy variables [30], as presented in eq. (2). The county-level BC emission inventory was further converted to grids at a resolution of $0.5^\circ \times 0.5^\circ$,

$$Q_{i,j} = Q_j \times \frac{FD_{i,j}}{FD_j}, \quad (2)$$

where Q is the BC emission, FD is the proxy variable, and i and j represent the county and the province, respectively. For industry, power generation, and transportation, gross domestic product (GDP) was chosen as the proxy variable; for heating and urban residential consumption, urban population was selected; for rural residential consumption and biomass burning, rural population and seeded crop area were used, respectively.

1.3 Activity data

The activity data were derived mainly from national or provincial statistical yearbooks, and missing data were calculated based on previous years' statistics (Table 1). In 2008, coal accounted for 70% of the total domestic energy consumption [31], and as the major fuel consumed for industry, power generation and heating sectors, 1.397, 1.365 and 0.218 billion tons were burned, respectively. Residential consumption was divided into urban residential consumption and rural consumption, with coal and biofuels being the dominant energy sources. Urban and rural consumption used 38 and 67 million tons coal, respectively, and for rural residents, crop residues and firewood consumption were 346 and 176 million tons, respectively. Chen et al. [32] estimated that bituminous coal accounted for roughly 80% of the residential coal consumption, and briquette coal was estimated as responsible for 40% of the consumption in 2000 and 80% by 2020. Assuming that the proportion of briquette consumption changed lineally from 2000 to 2020, the proportion of briquette consumption in 2008 was extrapolated to be 56%. Biomass burning included the open burning of agricultural waste, and forest and grassland fires,

for which emissions from the open burning of agricultural waste were calculated for each crop type. The production-to-crop ratio and open burning ratio were gained from previous studies [33].

Transportation BC emissions from vehicles and aviation were also estimated. Vehicles were classified into 10 categories: gasoline buses and coaches, diesel buses and coaches, gasoline passenger cars, diesel passenger cars, gasoline heavy-duty vans, diesel heavy-duty vans, gasoline other-duty vans, diesel other-duty vans, two-stroke motorcycles, and four-stroke motorcycles. In 2008, the total number of vehicles in China was about 140 million, with the largest vehicle population in Guangdong, Shandong, and Jiangsu. The annual average mileage of each vehicle type was obtained from Qin and Xie [27].

1.4 Emission factors

Emission factors were selected based on the following principles: locally measured emission factors were preferentially selected, and emission factors presented by foreign scholars were only used when local experimental results were either inaccurate or lacking [34].

Emission factors for residential coal, biofuels, and the open burning of agricultural waste were derived from local measurements. Coal combustion emission factors can be influenced by the maturity of coal, combustion mode, and stove type [32,35]. Chen et al. [32] measured the mean emission factors of anthracite raw coal, anthracite briquette, bituminous raw coal, and bituminous briquette based on 38 combinations of coal types/combustion modes/stove types. Compared with other locally measured results [35,36] and emission factors from foreign studies [19], Chen et al.'s results were adapted and used in this work. Li et al. [37] obtained emission factors for crop residues and firewood for typical rural areas in China, and Shen et al. [38] and Cao et al. [39] developed crop residue emission factors using actual and simulative stoves. The emission factors for biofuels in our work were deduced from the above researches; for example, the geometric mean of the three factors named above was used as the emission factor of crop residues. Emission factors for the open burning of agricultural waste were derived from laboratory simulation [40,41] and field

measurements [42].

Because of the limited measurements of emission factors for industry, and power generation and heating, emission factors of these categories were mainly obtained from Streets et al. [19] and Bond et al. [18]. Emission factors from coking processes were gained from Bond et al. [18,43]. Control devices were installed to reduce pollutant emissions from coal combustion in large industrial boilers, power plants and heating boilers. The use ratio of various control devices and the removal efficiency were deduced from Streets et al. [19], the same method used by Qin and Xie [27]. Emission factors for industry, power generation and heating, residential, and biomass burning are presented in Table 2.

Since 1999, the vehicle emission regulations in China have been implemented using those specified in Euro I–Euro IV. Corresponding emission factors were required for vehicles under different regulations. Cai and Xie [45] calculated the emission factors of particulate matter (PM) for each vehicle type under different emission regulations in 2008 based on the COPERT IV model. BC emission factors for vehicles were deduced from the PM emission factors and the BC fraction of PM [27].

2 Results and discussion

2.1 Emission estimate

The total emissions of black carbon in mainland China in 2008 were 1604.94 Gg, including 695.03 Gg from industrial and 636.02 Gg from residential sources. These two source-types were the major contributors, accounting for 43.3% and 39.6% of the total emissions, respectively. Transportation and biomass burning emissions were approximately 194.63 Gg and 67.68 Gg, responsible for 12.1% and 4.2% of national total emissions, respectively, while emissions from the power generation and heating sector were relatively small, as shown in Figure 1(a). Compared with anthropogenic sources, BC emissions of natural sources were negligible at only 0.64 Gg. Lu et al. [26] estimated BC emissions of China in 2008 to be 1787 Gg, which is in agreement with our work roughly, although the source contribution was somewhat different between studies. The emission contributions of different fuels are presented in

Table 1 Activity data for emission sources

Emission sources	Activity data
Industry, Power generation and heating	<i>China Energy Statistical Yearbook</i>
Residential–Fossil fuel	<i>China Energy Statistical Yearbook</i>
Residential–Biogas	<i>China Rural Statistical Yearbook</i>
Residential–Crop residues and firewood	<i>China Energy Statistical Yearbook</i> ^{a)}
Transportation–Vehicles	<i>China Automotive Industry Yearbook</i> <i>China Auto-market Yearbook</i>
Transportation–Aviation	<i>China Civil Aviation Statistical Yearbook</i>
Open burning of agricultural waste	<i>China Agriculture Statistics</i>
Forest fire	<i>China Agriculture Yearbook</i>
Grassland fire	<i>China Animal Husbandry Yearbook</i> ^{b)}

a) Calculated by linear or nonlinear regression based on historical data; b) substituted with the statistics of 2007.

Table 2 Black carbon emission factors from stationary sources (g/kg)

Source	Fuel type	Related emission factor	Our emission factor
Industry	coal	0.003 [19], 0.32 [19], 0.000062 [35]	0.044 ^{a)}
	oil	0.00012–0.117 [18]	0.00012–0.117
	gases	0.00012 [18]	0.00012
	coke	0.03136 [18]	0.03136
	indigenous coking	3.8 [43], 4.8 [18]	3.8
	mechanical coking	0.97 [18]	0.97
Power generation ^{b)}	coal	0.003 [19], 0.32 [19]	0.0013 ^{a)}
Heating ^{b)}	coal	0.003 [19], 0.32 [19]	0.044 ^{a)}
Residential–fossil fuel	bituminous raw coal	3.05 [32], 3.7 [19], 2.750 [35], 3.81 [36]	3.05
	anthracite raw coal	0.007 [32], 0.12 [19], 0.028 [35], 0.007 [36]	0.007
	bituminous briquette	0.09 [32], 0.12 [19], 0.082 [36]	0.09
	anthracite briquette	0.004 [32], 0.004 [36]	0.004
	oil	0.0676–0.117 [18]	0.07, 0.117, 0.25, 0.0676
	natural gas	0.00012 [18] ,	0.00012
Residential–biofuel	crop residues	0.67 [39], 0.43 [37], 1.38 [38]	0.74 ^{c)}
	firewood	1.49 [37]	1.49
	biogas	0.0001 [18]	0.0001
Biomass burning	wheat straw	0.52 [40], 0.49 [42]	0.50 ^{c)}
	paddy straw	0.52 [40], 0.43 [41]	0.47 ^{c)}
	corn straw	0.78 [40], 0.35 [42]	0.52 ^{c)}
	cotton straw	0.82 [40]	0.82
	other straw	0.69 [44]	0.69
	forest fire	0.56 [44], 0.99 [27]	0.99
	grassland fire	0.48 [44]	0.48

a) Calculated based on unabated emission factors (EFs), the use ratio, and the removal efficiencies of different control devices; b) EF_{BC} of oil and gases were the same as that of the industry sector; c) the geometric mean of related EFs.

Figure 1 Contribution of BC emissions by sector (a) and fuel type (b).

Figure 1(b). Emissions from coal and biofuels were 818.14 Gg and 516.49 Gg, responsible for 51.0% and 32.3%, respectively, of the total emissions. The contribution of emissions from oil combustion was estimated to be 12.6%, mostly from transportation. Estimated BC emissions of gases and

from forest and grassland fires were very small.

According to the contributions of sectors in different areas, the 31 administrative districts in mainland China were divided into four categories: industry contribution district, residential contribution district, industry and residential jointly

contribution district, and transportation contribution district, as demonstrated in Table 3. Industry contribution districts included seven areas such as Shanxi, Hebei, and Shandong, where industry was the main contributor to local emissions, especially from heavy industry such as the coking, iron, and steel industries. As municipalities, Shanghai and Tianjin's industrial emissions were responsible for the majority of total emissions because of their small residential emissions. Twelve areas were classified into residential contribution districts, in which most areas had high residential emissions with high rural population density and relatively low-level industrial development, such as Sichuan, Guangxi, and Guizhou. Even without a high population density, the residential sector in Hainan was still the major contributor of local BC emissions because emissions from other sources were lower than those from the residential sector. Eight ar-

eas (industry and residential jointly contribution districts) had roughly equal BC contributions from the industrial and residential sectors including Henan, Liaoning, and Inner Mongolia. Guangdong, Zhejiang, Beijing, and Xizang were transportation contribution districts, and their transportation emissions accounted for more than 30% of their total emissions. Due to the 2008 Olympic Games, a large number of industrial enterprises with heavy pollution were relocated to implement industrial restructuring in Beijing, resulting in transportation becoming the major contributor of BC emissions. In Xizang, industry and residential emissions were relatively small, so transportation was responsible for most emissions. Emission contributions from industry (21.7%) and residential consumption (32.1%) in Zhejiang were also exceeded by transportation (31.7%).

Shanxi had the largest emissions among all of China in

Table 3 Summary of Chinese black carbon emissions by sector and province in 2008 (Gg)

Area	Industry	Residential	Power generation and heating	Biomass burning	Transportation	Total
Shanxi	164.14	19.62	0.36	1.42	6.95	192.49
Hebei	76.69	41.28	0.59	3.88	12.03	134.46
Shandong	64.54	31.83	1.70	5.67	13.56	117.30
Shaanxi	31.61	18.93	0.14	1.36	3.61	55.66
Shanghai	15.88	0.84	0.21	0.16	3.77	20.87
Tianjin	6.19	1.56	0.34	0.21	2.92	11.22
Ningxia	4.91	1.90	0.11	0.39	1.17	8.49
Sichuan	20.10	58.20	0.11	2.86	8.59	89.85
Anhui	17.27	35.11	0.21	4.65	6.89	64.13
Hubei	17.89	32.20	0.21	2.05	6.19	58.54
Guangxi	7.12	42.61	0.13	2.65	4.57	57.07
Guizhou	16.67	29.27	0.13	1.24	3.36	50.67
Hunan	10.82	26.67	0.18	4.27	7.34	49.27
Jilin	7.36	15.43	0.58	3.87	4.82	32.06
Chongqing	6.37	18.22	0.08	0.98	2.83	28.48
Jiangxi	7.00	14.55	0.09	1.22	5.18	28.04
Gansu	6.64	15.45	0.20	1.05	2.82	26.15
Fujian	3.16	11.17	0.12	0.98	4.07	19.51
Hainan	0.07	2.70	0	0.30	1.13	4.21
Henan	45.49	44.95	0.50	6.43	11.04	108.42
Liaoning	35.33	26.77	1.05	2.38	9.53	75.05
Inner Mongolia	30.45	20.48	0.76	3.10	7.99	62.78
Yunnan	26.86	24.74	0.06	1.66	6.87	60.19
Jiangsu	23.96	19.19	0.87	4.64	9.60	58.27
Heilongjiang	17.54	24.56	0.77	4.65	6.96	54.47
Xinjiang	13.81	14.57	0.39	2.17	5.88	36.82
Qinghai	3.00	2.64	0.01	0.15	1.35	7.16
Guangdong	5.54	28.85	0.37	1.89	18.99	55.65
Zhejiang	5.13	7.57	1.01	1.12	8.73	23.56
Beijing	3.49	3.67	0.27	0.17	4.98	12.58
Xizang	0	0.49	0	0.10	0.93	1.52
Total	695.03	11.58	636.02	67.68	194.63	1604.94

2008, followed by Hebei, Shandong, Henan, and Sichuan. These five provinces cover 11.5% of China's landmass but generated over 40.0% of the total BC emissions. The major contributor of BC emissions in Shanxi was industry, accounting for 85.3% of the total. As a famous coal-producing province, Shanxi has a well-developed coking industry, leading to high BC emissions. Industry was responsible for 55% of the corresponding total emissions in Hebei and Shandong, being their major contributor, but residential emissions were relatively high compared with other areas. Sichuan with a large population had a significant consumption of crop residues and firewood, leading to high residential emissions. The industrial and residential sectors jointly contributed to Henan's BC emissions, each accounting for approximately 40% of its total emissions.

As for residential consumption, 43.88 Gg of BC emissions were from the urban residential consumption of fossil fuel, 75.66 Gg were from the rural residential consumption of fossil fuel, and 526.49 Gg were emitted from the rural residential consumption of biofuels, with emissions from coal combustion reaching 119.53 Gg. Residential emissions were high in Sichuan, Henan, Hebei, and Guangxi, and were closely related to local population and agriculture. BC emissions from residential consumption were emitted mainly from the burning of untreated coal, crop residues, and firewood in household stoves [19,20]. Compared with large-scale industrial boilers, burning in household stoves is more likely to produce larger fine-particle emissions due to low combustion temperature and a lack of both oxygen and effective emission control devices. Li et al. [37] estimated BC emissions from residential biofuel consumption to be 334 Gg in 2000 and 430 Gg in 2005. Annual variations in biofuel consumption, and the use of lower emission factors by Li

et al. [37], resulted in the discrepancies between our results. Figure 2 shows the contributions of BC emissions from the various subsectors of residential sources. A large amount of BC aerosols were emitted from crop residues and firewood, fuel sources for cooking and heating energy in most rural areas of the country, which contributed the majority of residential emissions. The combustion of urban and rural fossil fuel was also responsible for some emissions. Compared with rural areas, more oil and gas were used in urban areas, reducing the emissions of urban residential consumption. Emissions from biogas were extremely small and can be neglected, differing by four to six orders of magnitude from the emissions of crop residues as fuel.

With regard to transportation, BC emissions from China in 2008 were 194.63 Gg, accounting for 12.1% of the total emissions, with vehicular emissions representing 193.54 Gg. The transportation emissions were largest in Guangdong, followed by Shandong, Hebei, and Henan, consistent with their vehicle populations. Vehicles were divided into diesel vehicle and gasoline vehicle according to fuel consumption, emitting 185.43 and 8.11 Gg, respectively, in which diesel vehicles were responsible for the majority (96%) of total transportation emissions. The contributions of transportation emissions from various subsectors are presented in Figure 3, illustrating that diesel vans contributed the highest level of BC emissions, followed by diesel passenger cars. In most areas, vans were the dominant contributors, accounting for 60%–85% of the emissions, while passenger cars, buses and coaches were responsible for 65% of the local transportation emissions in Beijing. Contributions from motorcycle emissions were small in all areas. Aviation occupied a notable proportion of transportation emissions only in Beijing, Shanghai, and Hainan where aviation is well developed.

Figure 2 Contribution of residential BC emissions from various subsectors.

Figure 3 Contribution of transportation BC emissions from various subsectors.

2.2 Spatial distribution of BC emissions

Figure 4 shows the spatial distribution of Chinese BC emissions at a resolution of $0.5^\circ \times 0.5^\circ$. Districts with emissions of more than 1000 t/a cover 5.7% of the national territory, but were responsible for 41.2% of the total BC emissions. As shown in Figure 4, BC emission intensity in eastern China was much higher than in western China. In eastern China, emissions in Liaoning, Shandong, Hebei, Henan, Shanxi, Jiangsu, Anhui, and Hubei were high because of the considerable industrial emissions in Shanxi and Liaoning and the large emissions from both industrial and residential sectors in Shandong, Hebei, Henan, Jiangsu, Anhui, and Hubei. While emissions in western areas were overall low, the emission intensity in eastern Sichuan, Chongqing, Guizhou, and southern Guangxi was high—primarily from elevated residential emissions. For example, emissions from the residential sector in Sichuan were the highest in the country. The low-emitting districts, which include Xizang, Xinjiang, and Qinghai, cover 37.9% of the national territory, but represent only 2.9% of China's total BC emissions due to their underdeveloped industry and low population densities. Overall, the spatial distribution of BC emissions was related to local industry and rural population density, with industry and residential consumption contributing the majority of total BC emissions.

The spatial distribution characteristics of BC emissions were compared with those of SO_2 , NO_2 , and non-methane volatile organic compounds (NMVOCs). Emissions of both BC and SO_2 were higher in the southeast than the northwest; however, high BC emitting areas were located regionally, while the high SO_2 emitting areas were scattered and concentrated around power plants or urban areas [26]. Spatial

Figure 4 Spatial distribution of Chinese BC emissions at a resolution of $0.5^\circ \times 0.5^\circ$ in 2008 (t/a).

differences in the distribution of pollutants were mainly due to differences in dominant sources: industry and residential consumption for BC, and industry and power generation for SO_2 . Districts with high NO_2 emissions were generally located in eastern coastal areas where power plants, industry, and transportation contributed as major emission sources [46]. In contrast, BC emissions were widely distributed over eastern and central China, resulting from high residential BC emissions. NMVOCs emissions in northern coastal areas, eastern coastal areas, southern coastal areas, northeastern provinces, and the Sichuan Basin were estimated to be high and were closely related with local economic development [47], in contrast to the spatial distribution of BC emissions,

influenced by industry and population density.

2.3 Comparison with other emission inventories

The comparison of our work with previous inventories is illustrated in Table 4. However, biomass burning was not taken into account by Ohara et al. [24], Lei et al. [25], or Zhang et al. [22]. As for total emissions, our results were roughly consistent with the inventories of recent years and higher than the emissions reported for the year 2000. Discrepancies arose not only from increases in energy consumption such as the explosive growth of motor vehicles, but also from improvements in estimation methodology.

The differences between our study and other emissions inventories arose mainly from the industrial and residential sectors. Industrial emissions reported here were much higher than those of previous studies. Coking in our work was divided into indigenous coking and mechanical coking, with the latter having lower emission factor due to technological advancements. Zhang et al. [22] considered the influences of technological improvement and estimated emissions from coke production and brick kilns separately using lower emission factors for coking, which led to reduced industrial emissions. Compared with the results of Lu et al. [26], discrepancies from industry were due to differences in the emission factors selected and the sources of fuel consumption data. For the residential sector, the emissions in our work were lower than other studies; for example, emissions from both residential fossil fuel and biofuel consumption were lower than those of Lu et al.'s study [26]. Compared with results from Zhang et al. [22], locally measured emission factors for the residential sector were updated and different ratios of residential coal types were used in our work, leading to discrepancies in residential emissions. Wang et al. [28] divided residential coal into chunk coal and briquette, without considering emissions differences between bituminous and anthracite coals. Wang et al. [28] also selected a higher BC emission factor and a larger consumption ratio of chunk coal, causing their estimates of residential emissions to be higher than ours. In addition, Lu et al. [26] calculated transportation emissions using oil consumption data and

updated emission factors on the basis of previous studies [18], which resulted in a higher estimation of vehicular emissions than our work.

2.4 Uncertainty analysis

Although we used emission factors from local measurements and reliable official statistical data, the accuracy of emission factors for diverse source areas and the random errors associated with measurement and statistics still led to uncertainties in our inventory. A Monte Carlo simulation was conducted to quantify the uncertainty of the BC emission inventory. Emission factors and activity data were assumed to follow lognormal distributions [18] and normal distributions [48], respectively. Referring to Qin and Xie [30], we assumed the coefficient of variability (CV) for emission factors from local experiments to be 50%. Emission factors from foreign studies had CVs increase to 150% and 250% for coal and oil, respectively, and the CVs of emission factors for coking and vehicles were 350% and 150%, respectively. Activity data CVs were assumed to be 10% for industry, 5% for coal consumption by power plants and heating, 20% for residential consumption, 30% for crop production, and 5% for vehicle population, based on related studies [30,48].

The Monte Carlo simulation was repeated for 100000 iterations, obtaining an uncertainty level for the Chinese BC emission inventory at the 95% confidence interval of (−53.51%, 179.92%), as illustrated in Figure 5. The emission inventory approximately followed lognormal distribution, and the mean value, 2.5th percentile value and 97.5th percentile value were 1603.95, 745.65, and 4489.73 Gg, respectively. Industry was the major contributor to inventory uncertainty, with an uncertainty at the 95% confidence interval of (−91.16%, 409.67%). Streets et al. [23] and Zhang et al. [22] determined the uncertainty of BC emissions at $\pm 484\%$ and $\pm 208\%$ based on an error propagation methodology, achieving higher values than our work. Compared with our study, the uncertainty calculated by Lu et al. [26] based on the same simulation method was lower (−43%, 93%). Differences in inventory uncertainty resulted

Table 4 Comparison of BC emission inventories for China (Gg)

Year	Industry	Power	Residential	Biomass burning	Transportation	Total	Reference
2008	695.0	11.6	636.0	67.7	194.6	1604.9	this work
2000	88.9	6.8	780.7	112.4	59.8	1048.6	[23]
2000	99	18	938		38	1093	[24]
2005	610	10	700		190	1510	[25]
2006	575	36	1002		198	1811	[22]
2007	529	13	651	104	145	1399	[21]
2007	646	50.7	988	77.7	188	1957	[28]
2008	510	19	888	110	259	1787	[26]

Figure 5 Uncertainty analysis for China's BC emission inventory in 2008 based on 100000 Monte Carlo simulations.

mainly from estimation methods and CVs.

3 Conclusion

An emission factor database was established based on local-measured data. Combined with activity data, the inventory and spatial distribution of Chinese BC emissions were established. Inventory uncertainty was reduced by updating emission factors, including the use of emission factors from local experiments and from the COPERT model.

Chinese BC emissions were 1604.94 Gg in 2008, mainly contributed by industry (695.03 Gg) and residential consumption (636.02 Gg), together accounting for 82.9% of the total emissions. Emissions from transportation reached 194.63 Gg and were responsible for 12.1% of the national emissions. Based on the contributions of different sources, the 31 administrative districts were aggregated into four districts: industry contribution district, residential contribution district, industry and residential jointly contribution district, and transportation contribution district. As for the BC emissions from various fuels, coal and biofuel contributed the majority of emission at 51.0% and 32.2% respectively, with crop residues and firewood being the major contributors to biofuel emissions. The highest emissions were concentrated in Shanxi, Hebei, Shandong, Henan, and Sichuan, covering only 11.5% of China's territory, but generating 40.0% of the country's total emissions. Spatially, BC emissions were high in the east and low in the west; however, Sichuan, Chongqing, and Guizhou, which are located in the west, also had large emissions. Their emission levels were related to the local industrial structure, the level of industrial development, and rural population density. According to this inventory, BC emissions from various sources, including industry, residential consumption, and transportation, should all be controlled to reduce total emissions. In the interim, BC high-emitting districts should be paid more attention to

and control measures should be implemented in different districts based on their emission profiles.

This work was supported by the National Basic Research Program of China (2010CB955608).

- Seinfeld J. Atmosphere science: Black carbon and brown cloud. *Nat Geosci*, 2008, 1: 15–16
- Qin S G, Tang J, Shi G Y, et al. Observational study of black carbon at Wenjiang, Sichuan Province (in Chinese). *Acta Sci Circum*, 2007, 27: 1370–1376
- Jacobson M Z. Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols. *Nature*, 2001, 409: 695–697
- Menon S, Hansen J, Nazarenko L, et al. Climate effects of black carbon aerosols in China and India. *Science*, 2000, 297: 2250–2253
- Qiu J, Yang L. Variation characteristics of atmospheric aerosol optical depths and visibility in North China during 1980–1994. *Atmos Environ*, 2000, 34: 603–609
- Chameides W L, Yu H, Liu S C, et al. Case study of the effects of atmospheric aerosols and regional haze on agriculture: An opportunity to enhance crop yields in China through emission controls? *Proc Natl Acad Sci USA*, 1999, 96: 13626–13633
- Saikawa E, Naik V, Horowitz L W, et al. Present and potential future contributions of sulfate, black and organic carbon aerosols from China to global air quality, premature mortality and, radiative forcing. *Atmos Environ*, 2009, 43: 2814–2822
- Hamilton R S, Mansfield T A. Airborne particulate elemental carbon: Its sources, transport and contribution to dark smoke and soiling. *Atmos Environ*, 1991, 25: 715–723
- IPCC. *Climate Change 2001: The Scientific Basis*. New York: Cambridge University Press, 2001
- Sato M, Hansen J, Koch D, et al. Global atmospheric black carbon inferred from AERONET. *Proc Natl Acad Sci USA*, 2003, 100: 6319–6324
- Haywood J M, Ramaswamy V. Global sensitivity studies of the direct radiative forcing due to anthropogenic sulfate and black carbon aerosols. *J Geophys Res*, 1998, 103: 6043–6058
- Ackerman A S, Toon O B, Stevens D E, et al. Reduction of tropical cloudiness by soot. *Science*, 2000, 288: 1042–1047
- Wang C. A modeling study on climate impacts of black carbon aerosols. *J Geophys Res*, 2004, 109: D03106
- Andreae M O. The dark side of aerosols. *Nature*, 2001, 409: 671–672
- Jacobson M Z. Control of fossil-fuel particulate black carbon and or-

- ganic matter, possibly the most effective method of slowing global warming. *J Geophys Res*, 2002, 107: 4410
- 16 Cooke W F, Lioussé C, Cachier H, et al. Construction of a 1 degrees ×1 degrees fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model. *J Geophys Res*, 1999, 104: 22137–22162
 - 17 Wilkening K E, Barrie L A, Engle M. Trans-Pacific air pollution. *Science*, 2000, 290: 65–67
 - 18 Bond T C, Streets D G, Yarber K F, et al. A technology-based global inventory of black and organic carbon emissions from combustion. *J Geophys Res*, 2004, 109: D14203
 - 19 Streets D G, Gupta S, Waldhoff S T, et al. Black carbon emissions in China. *Atmos Environ*, 2001, 35: 4281–4296
 - 20 Cao G L, Zhang X Y, Wang Y Q, et al. Inventory of black carbon emission from China (in Chinese). *Adv Clim Change Res*, 2006, 2: 259–264
 - 21 Cao G L, Zhang X Y, Gong S L, et al. Emission inventories of primary particles and pollutant gases for China. *Chin Sci Bull*, 2011, 56: 781–788
 - 22 Zhang Q, Streets D G, Carmichael G R, et al. Asian emission in 2006 for the NASA INTEX-B mission. *Atmos Chem Phys*, 2009, 9: 5131–5153
 - 23 Streets D G, Bond T C, Carmichael G R, et al. An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. *J Geophys Res*, 2003, 108: 8809
 - 24 Ohara T, Akimoto H, Kurokawa J, et al. An Asian emission inventory of anthropogenic emission sources for period 1980–2020. *Atmos Chem Phys*, 2007, 7: 4419–4444
 - 25 Lei Y, Zhang Q, He B K, et al. Primary anthropogenic aerosol emission trends for China, 1990–2005. *Atmos Chem Phys*, 2011, 11: 931–954
 - 26 Lu Z, Zhang Q, Streets D G. Sulfur dioxide and primary carbonaceous aerosol emission in China and India, 1996–2010. *Atmos Chem Phys*, 2011, 11: 9839–9864
 - 27 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: 4825–4841
 - 28 Wang R, Tao S, Wang W T, et al. Black carbon emissions in China from 1949 to 2050. *Environ Sci Technol*, 2012, 46: 7595–7603
 - 29 Xie S D, Song X Y, Shen X H. Calculating vehicular emission factors with COPERTIII mode in China (in Chinese). *Environ Sci*, 2006, 27: 415–419
 - 30 Qin Y, Xie S D. Estimation of county-level black carbon emissions and its spatial distribution in China in 2000. *Atmos Environ*, 2011, 45: 6995–7004
 - 31 National Bureau of Statistics. *China Energy Statistical Yearbook 2009* (in Chinese). Beijing: China Statistics Press, 2010
 - 32 Chen Y J, Jiang X H, Zhi G R, et al. Black carbon emissions from residential coal combustion and reduction strategy (in Chinese). *Sci China Ser D-Earth Sci*, 2009, 39: 1554–1559
 - 33 Wang S X, Zhang C Y. Spatial and temporal distribution of air pollutant emissions from open burning of crop residues in China (in Chinese). *Sci Pap Online*, 2008, 3: 329–333
 - 34 Liu Y, Shao M. Estimation and prediction of black carbon emissions in Beijing city. *Chin Sci Bull*, 2007, 52: 1274–1281
 - 35 Zhang Y X, Schauer J J, Zhang Y H, et al. Characteristics of particulate carbon emissions from real-world Chinese coal combustion. *Environ Sci Technol*, 2008, 42: 5068–5073
 - 36 Zhi G R, Chen Y G, Feng Y L, et al. Emission characteristics of carbonaceous particles from various residential coal-stoves in China. *Environ Sci Technol*, 2008, 42: 3310–3315
 - 37 Li X H, Wang S X, Duan L, et al. Carbonaceous aerosol emissions from household biofuel combustion in China. *Environ Sci Technol*, 2009, 43: 6076–6081
 - 38 Shen G F, Yang Y F, Wang W, 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
 - 39 Cao G L, Zhang X Y, Gong S L, et al. Investigation on emission factors of particulate matter and gaseous pollutants from crop residue burning. *J Environ Sci*, 2008, 20: 50–55
 - 40 Cao G L, Zhang X Y, Wang Y Q, et al. Estimation of emissions from field burning of crop straw in China. *Chin Sci Bull*, 2008, 53: 784–790
 - 41 Zhang Y S, Shao M, Lin Y, et al. Emission inventory of carbonaceous pollutants from biomass burning in the Pearl River Delta Region, China. *Atmos Environ*, doi:10.1016/j.atmosenv.2012.05.005
 - 42 Li X H, Wang S X, Duan L, et al. Particulate and trace gas emissions from open burning of wheat straw and corn stover in China. *Environ Sci Technol*, 2007, 41: 6052–6058
 - 43 Bond T C, Bhardwaj E, Dong R, et al. Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850–2000. *Glob Biogeochem Cycle*, 2007, 21: GB2018
 - 44 Andreae M O, Merlet P. Emission of trace gases and aerosols from biomass burning. *Glob Biogeochem Cycle*, 2001, 15: 955–966
 - 45 Cai H, Xie S D. Determination of emission factors from motor vehicles under different emission standards in China (in Chinese). *Acta Sci Nat Univ Peking*, 2010, 46: 319–326
 - 46 Zhang Q, Streets D G, He B K, et al. NO_x emission trends for China, 1995–2004: The view from the ground and the view from space. *J Geophys Res*, 2007, 112: 22306–22323
 - 47 Bo Y, Cai H, Xie S D. Spatial and temporal variation of historical anthropogenic NMVOCs emission inventories in China. *Atmos Chem Phys*, 2008, 8: 7297–7316
 - 48 Zhao Y, Nielsen C P, Lei Y, et al. Quantifying the uncertainties of a bottom-up emission inventory of anthropogenic atmospheric pollutants in China. *Atmos Chem Phys*, 2011, 11: 2295–2308