

Inventory of black carbon and organic carbon emissions from China

Guoliang Cao^{a,b,*}, Xiaoye Zhang^a, Fangcheng Zheng^b

^aKey Laboratory of Atmospheric Chemistry, Centre for Atmosphere Watch & Services, CMA; Chinese Academy of Meteorological Sciences, Beijing 100081, PR China

^bSchool of environmental and municipal engineering, Xi'an University of Architecture & Technology, Xi'an, 710055, PR China

Received 28 October 2005; received in revised form 15 May 2006; accepted 25 May 2006

Abstract

We present detailed high-resolution emission inventories of black carbon (BC) and organic carbon (OC) from China in the year 2000. The latest fuel consumption data, including fossil and biomass fuels and socio-economic statistics were obtained from government agencies, mostly at the county level. Some new emission factors (EFs) from local measurements also were used. National and regional summaries of emissions are presented, and gridded emissions at $0.2^\circ \times 0.2^\circ$ resolution are shown. Our calculated emissions were 1500 Gg for BC and 4100 Gg for OC, mainly due to the burning of coal and biofuels. The carbonaceous aerosol emissions estimated here are higher than those in previous studies, mainly because coal burning by rural industries and residences were previously underestimated. More carbonaceous aerosols are emitted from eastern China than western China. A strong seasonal dependence is observed for emissions, with peaks in May and October and low emissions in April and July; this seasonality is mainly due to patterns in residential heating and agriculture waste open burning.

© 2006 Elsevier Ltd. All rights reserved.

Keywords: Carbonaceous aerosol; China; Emission inventory; Seasonality

1. Introduction

Aerosol particles containing black carbon (BC) and are important for climate because they strongly absorb solar radiation and their effects may extend over regional to global scales (Liousse et al., 1996; Cooke and Wilson, 1996; IPCC, 2001; Hansen and

Sato, 2001; Haywood and Boucher, 2000; Jacobson, 2001, 2002). BC can affect cloud albedo by changing the hygroscopicity of cloud condensation nuclei (Liousse et al., 1996), and BC also may lower crop yields (Chameides et al., 1999) and contaminate building materials (Ghedini et al., 2000). Organic carbon (OC), on the other hand, mainly scatters solar radiation without significant absorption, and OC-containing aerosol particles grow after absorbing water vapor, changing the particles optical properties. OC also can significantly affect the surface tension of cloud droplets (Shulman et al., 1996; Novakov and Penner, 1993). BC- and

*Corresponding author. Centre for Atmosphere Watch & Services, CMA; Chinese Academy of Meteorological Sciences 46#, ZhongGuanCunNandajie Road, Beijing 100081, PR China. Tel.: +86 10 58995247; fax: +86 10 62176414.

E-mail addresses: caogl@cams.cma.gov.cn, 82205975@163.com (G. Cao).

OC-containing aerosols often compose a major fraction of atmospheric particulate matter, especially in the polluted regions (He et al., 2001; Singh et al., 2002; Ye et al., 2003), and they can reduce atmospheric visibility (Reddy and Venkataraman, 2000; Qiu and Yang, 2000; Park et al., 2003) and have adverse health effects. Small ($< 1 \mu\text{m}$ diameter) carbonaceous particles can penetrate deep into the lungs, hence their potential adverse health effect can be much greater than that of larger particles.

Anthropogenic carbonaceous aerosols are released from incomplete combustion of carbonaceous fuels, such as fossil fuel and biomass. OC can either be emitted directly by anthropogenic and natural sources (primary OC), or it can be the product of condensed organic material formed in the atmosphere by oxidation of organic gases (secondary OC) (Turpin and Huntzicker, 1991, 1995). Submicron carbonaceous particles have an atmospheric residence time of 3–7 days (Liousse et al., 1996; Cooke and Wilson, 1996; Cooke et al., 2002) and therefore may spread over distances of hundreds to thousands of kilometers.

China burns large quantities of coal and biofuel (IEA, 2002), and it is generally recognized as a major global anthropogenic source for carbonaceous aerosols (Cooke and Wilson, 1996; Cooke et al., 1999; Streets et al., 2003; Bond et al., 2004). Emissions from China have been rapidly changing, however, and the lack of statistical data and technological information and limited availability of local measurements, make it difficult to establish the quantities of carbonaceous aerosol emitted from China. In particular, there is a general lack of understanding concerning emissions from rural China, and therefore the existing inventories have a high degree of uncertainty. The population density for the base period of concern (1980–1990) was used for spatially distributing the fuel use and emissions in previous studies, and an update of the base year (2000) is needed. In addition, some emission inventories have not considered seasonal variations, especially from biomass burning in rural China.

The emission factors (EFs) used to estimate emissions from fuel consumption have also been derived using various assumptions, and in some cases there is little experimental data to support these estimates. For example, previous inventories have arrived at a best approximation of biofuel combustion EFs, but these are based on limited measurements of wood burning in fire-places or space-heating stoves in developed countries

(Andreae and Merlet, 2001; Dasch, 1982) and from wind-tunnel experiments of agriculture waste burning (Turn et al., 1997; Jenkins et al., 1996; Liousse et al., 1996). The use of EFs measured for such non-representative combustion systems introduce uncertainties in the estimated emissions. A more accurate inventory for carbonaceous aerosol from China for a recent base year is needed as input to the regional-scale climate-modeling studies over China, including the China Meteorological Administration (CMA) aerosol projects. In this paper, we present carbonaceous aerosol emission inventories for China with spatial and seasonal variation.

2. Methodology

Emission inventories are calculated using fuel consumption data for the year 2000 and the detailed EFs derived herein. The underlying information is organized by source category or by region, and for a number of sources by season as well. Fig. 1 shows the general methodology for the development of emission estimates. The regional and national emission is the sum of each fuel sector. Each species emission is calculated using the following equation and it is similar to that described by Klimonta et al. (2002):

$$E_i = \sum_j \sum_k \sum_l A_{j,k,l} \text{EF}_{i,j,k,l} (1 - \eta_{i,j,k,l}),$$

where E is emissions, A is fuel consumption, η is removal efficiency, and EF is emission factor; the subscripts i, j, k , and l indicate the species, region, sector and fuel type, respectively.

Emissions from forest fire and grassland fire are calculated as follows:

$$E_{ij} = \sum_j A_j B_j C_j \text{EF}_{ij},$$

where E is emissions, A is burning area, B is fuel load, C is burning efficiency and EF is emission factor. i and j indicate the species and region.

2.1. Source type

The major sources included in the inventory are emissions from biomass burning (including agricultural waste open burning, forest and grassland fires), residences, power generating stations, industries, and the transportation sector. Emission sources are divided into two major categories: large point sources (LPSs) and area sources.

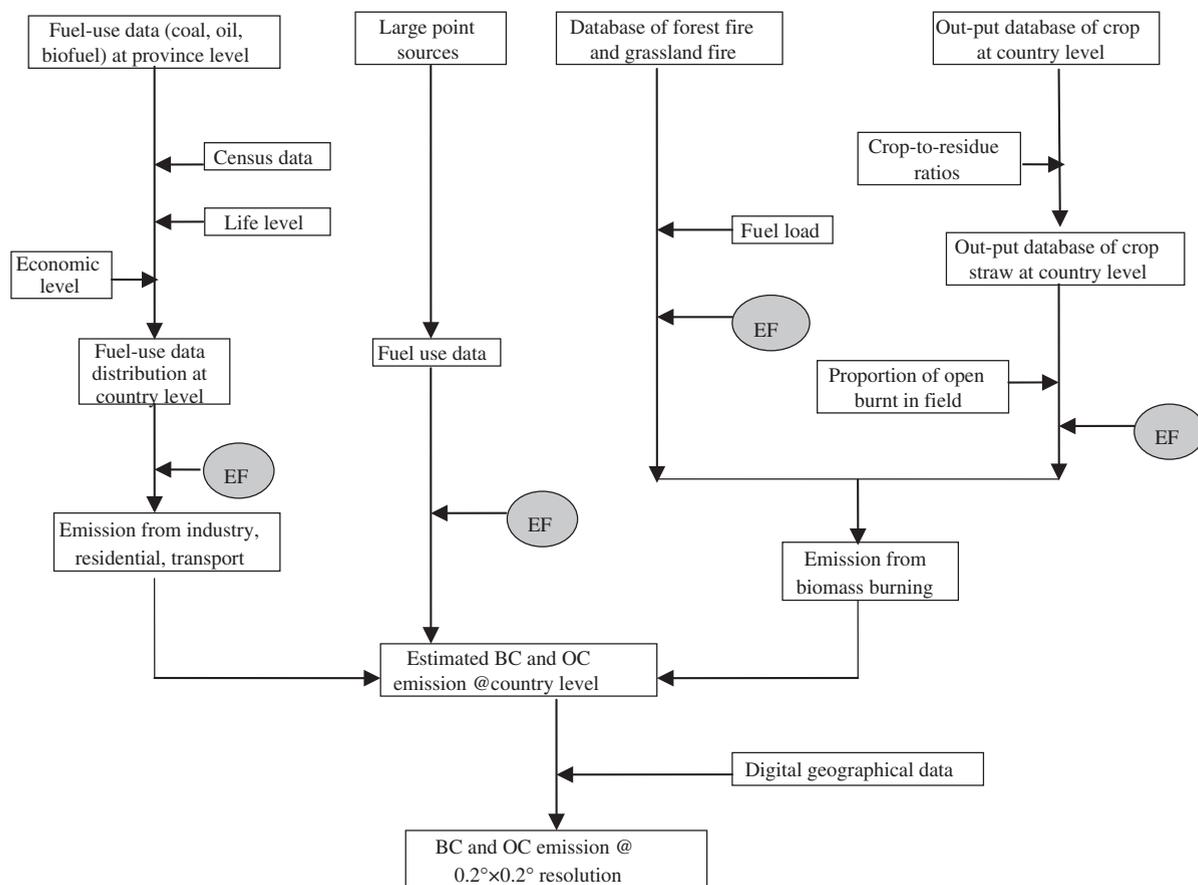


Fig. 1. Methodology for the development of BC and OC emission inventories in China.

We accounted for 363 LPSs, mainly power plants and iron and steel plants, in our emission estimates. Each LPS is located by latitude and longitude. There were 285 power plants considered in the assessment, with a combined installed capacity greater than 1.2 MW (State Power Corporation of China, 2001). The power plants are located mainly in eastern China and coal is the main fuel for the power generating stations. In 2000, China was the largest steel producer in the world, and the 78 large iron and steel plants, producing than 50,000 tons of steel per year; these are also located mainly in eastern China (Editorial Board of China Steel Yearbook, 2001).

Area sources include all stationary sources not included in the LPS category. The basic data used to estimate emissions from these sources, such as population, area, GDP (Gross domestic product), industrial output, fuel usage and vegetative cover, are taken from National Bureau of Statistics and

various government agencies, mainly at the county level. When faced with discrepant data, we use a method similar to that described by Wang et al. (2005) to resolve the differences.

2.2. Emission factors

Typically, carbonaceous aerosol emissions from fuel combustion are affected by the chemical composition of the fuel, combustion type and temperature, and efficiency of any emission control devices. There are few direct experimental measurements of BC and OC EFs from fuel combustion, especially in China (Jenkins et al., 1996; Kirchstetter et al., 1999). BC/OC emissions have been computed from particulate matter (PM) EFs and information on the carbonaceous fractions of PM (Cooke and Wilson, 1996; Cooke et al., 1999; Streets et al., 2001, 2003; Bond et al., 2004). These emission estimates have substantial uncertainties because a wide range

of EFs have been used in the models. These uncertainties can be reduced by better measurements of carbonaceous aerosol emissions and an improved understanding of the causes of emission variability.

We designed a combustion tower and installed it in a laboratory to simulate biofuel emissions from the kind cooking stoves used by peasants in rural China. The experimental design was described in detail (Cao et al., submitted manuscript, 2005). The straw used in the tests was collected from the representative parts of China, and BC/OC emissions were calculated for test materials that included wheat straw, rice straw, corn stover and cotton stalks. In related studies, Chen et al. (2005) estimated the EFs of carbonaceous aerosol from honeycomb briquettes, the most common type of coal burned in Chinese residences. Their calculated EF_{BC} was relatively low, but EF_{OC} was close to reported literatures. Wang (2002) estimated emis-

sions from light-duty vehicles in tunnel experiments and by heavy-duty diesel vehicles through dynamometer testing.

Table 1 presents EFs from literature and the experiments conducted here. We divided crop straw into five types, and the EFs determined through the experiments were found to be similar to literature values (Table 1). The inventories of Penner et al. (1993) and Cooke and Wilson (1996) often have been used as emission sources in modeling studies, but more recently Streets et al. (2004) argued that the EFs for fossil fuel combustion were not representative. We believe, however, on the basis of our studies and the EFs from various references (Bond et al., 2004; Chen et al., 2005), that at least some of the EFs in Cooke et al. (1999) are consistent with the low combustion efficiency and ineffective pollution controls of devices used in China. The EFs for transportation, which have been calculated from on-road testing, dynamometer testing and tunnel

Table 1
Emission factors used in this work ($g\ kg^{-1}$)

Sector	Fuel type	BC	OC
Industry	Coal	0.32 ^a , 1.1–1.58 ^d	2.1 ^d
	Oil	0.07–0.36 ^a , 0.15 ^j	0.19–0.27 ^b , 0.25 ^d , 0.04 ⁱ
	Biofuel	1.0 ^a , 0.59 ^c	4.0 ^c
Power generation	Coal	0.003–0.32 ^a , 0.0095 ^g	0.25 ^f
	Oil	0.25–0.36 ^a , 0.15 ^j	0.27 ^b , 0.04 ⁱ
Biomass burning	Agriculture waste	0.73 ^a , 0.69 ^c , 0.47 ^f	3.3 ^c , 0.7 ^f
	Forest fire	0.56 ^c , 0.98 ^f	8.6–9.7 ^c , 6.1 ^f
	Grassland fire	0.48 ^c	3.4 ^c
Transportation	Diesel	1.0 ^a , 2 ^d , 1.3 ^c , 0.29 ^g , 0.11–0.87 ^l	0.03–0.18 ^b , 0.5 ^c , 0.22 ^g , 0.96–6.7 ^l
	Gasoline	0.08 ^a , 0.03 ^d , 0.035 ^c , 0.07 ^g , 0.006–0.03 ^l	0.01–0.13 ^b , 0.053 ^c , 0.5 ^g , 0.042–0.24 ^l
Residential	Biofuel-rice straw	0.86 ^h , 0.5 ⁱ , 1.0 ^j , 0.52 ^m	0.94 ⁱ , 0.32 ^j , 1.96 ^m
	Biofuel-wheat straw	1.2 ^h , 0.8 ⁱ , 0.52 ^m	2.2 ⁱ , 3.83 ^m
	Biofuel-corn stover	0.96 ^h , 0.75 ⁱ , 0.78 ^m	1.8 ⁱ , 2.21 ^m
	Biofuel-cotton stalk	0.82 ^m	1.83 ^m
	Biofuel-others	1.0 ^a , 0.59 ^c , 0.78 ⁱ	5.0 ^b , 4.0 ^c , 1.9 ⁱ
	Firewood	1.0 ^a , 0.59 ^c , 0.41 ^f	0.27 ^f
	Coal	0.12–3.7 ^a , 1.58–4.1 ^d , 1.83 ^g , 5 ^j , 0.28 ^k	0.12–3.0 ^b , 4.77 ^d , 7.8 ^g , 4 ^j , 7.82 ^k
	Oil	0.36–0.7 ^a	0.1 ^d

^aStreets et al., 2001.

^bStreets et al., 2003.

^cAndreae and Merlet, 2001.

^dCooke et al., 1999.

^eKirchstetter et al., 1999.

^fReddy and Venkataraman, 2002a; OM/OC ratio assumed as 1.3.

^gReddy and Venkataraman, 2002b; OM/OC ratio assumed as 1.3.

^hLioussé et al., 1996.

ⁱJenkins et al., 1996; Turn et al., 1997.

^jBond et al., 2004.

^kChen et al., 2005; Bituminous honeycomb briquette.

^lWang et al., 2002.

^mExperiment testing, emission factors.

experiments closely match literature values (Kirchstetter et al., 1999; Streets et al., 2001, 2003; Bond et al., 2004), and the results of Wang (2002). For open biomass burning, the EFs are taken from Andreae and Merlet (2001).

3. Fuel usage

3.1. Biofuels

The biofuels data used here include agriculture residues that are used as fuels for rural domestic cooking, and firewood used for cooking and heating and as an industrial energy source in rural China. Agricultural residues and firewood consumption data were collected from China government statistic data (NBSC, 2001a,b; Editorial Board of China Rural Energy Yearbook, 2001) that are at province level. The total amount of agricultural residues burned was estimated to be 291.43 Tg for 2000, and the largest contributions were from Shandong, Jiangsu, Heilongjiang and Anhui Provinces, whose residents mainly live in rural areas and where there is limited energy as available in the form of coal, natural gas or firewood. Total firewood consumption was calculated to be 136.44 Tg for 2000, the largest contributions were from mountainous regions in south-China, including Hunan, Guangxi, Sichuan and Hubei Provinces. In comparison, northwestern China has lower population densities and fewer forests, and therefore a lower consumption of firewood.

3.2. Fossil fuels

Fossil fuel sources considered include oil, liquefied petroleum gas (LPG) and coal. Coal is the major energy source used in China, accounting for 63% of the total energy consumed. China is also the largest consumer and producer of coal in the world (NBSC, 2001b). Coal consumption in 2000 was 1.24 billion short tons in China, accounting for 24% of the world total (NBSC, 2001a, b). The largest coal-consuming sectors are electric power generation (546.1 Mt) and industry (333.1 Mt). In the residential sector, the total consumption was 213.4 Mt, of which 40% was burned as honeycomb briquettes (Chen et al., 2005). Spatially, the largest quantities of coal burned are in Shanxi, Hebei, Shandong, Jiangsu, and Henan Provinces.

For emissions from oil combustion, only motor vehicles, domestic and industrial sources were

considered, that is, shipping and aviation were not included. Diesel oil (diesel) and petrol (gasoline) are the major fuels used by motor vehicles in China. Total consumption of diesel, petrol, kerosene and fuel oil is 67.70, 35.00, 8.70 and 38.70 Mt, respectively. Transportation was the largest user of diesel, petrol and kerosene, about 25.40, 13.90 and 5.40 Mt, respectively. The largest quantities of oil are burned in Guangdong, Shandong, Shanghai and Heilongjiang Provinces.

3.3. Biomass burning

The open burning of agriculture waste is a common practice in China, and this mainly occurs in three kinds of regions: (1) grain-producing regions with a low population densities such as Jilin and Heilongjiang Provinces; (2) developed regions including Shanghai, Zhejiang and Jiangsu Provinces; and (3) energy producing regions including Shanxi and Shaanxi Provinces. The emissions from the burning of agriculture waste are estimated with consideration given to climatic conditions, urban vs. rural population densities, vegetative cover, and the main types of crops cultivated; these calculations are done on a province-by-province basis. Unfortunately, only limited literature data related to open biomass burning in China exist, but the available studies indicate that approximately 27.2% of crop straw was burned in 1995 (Hu, 2000) and 15.2% in 1998 (Weng et al., 2004). More detailed studies indicate that in Jiangsu Province, 33% of the crop straw burned during 1998 (Wu et al., 2001) while the corresponding figures were 32.4% for Guangdong Province (Lin and Song, 2002), 40% for Fuzhou city in 2001 (Yu, 2003), 39.6% for Shanghai in 2000 (Yao, 2001), respectively.

We deduced that the amount of crop straw openly burnt is proportional to peasants' income level, and this hypothesis had been confirmed by investigations in Henan Province (Chen, 2001). The correlation coefficient we estimated between income of peasant and fraction burnt is 0.81. The per capita average peasant income for 2000 at county level came from Chinese government sources (NBSC, 2001a, b; Ministry of Agriculture of China, 2001) and we used that information to estimate the proportion of crop straw burnt openly in agricultural fields. Crop production statistics (Ministry of Agriculture of China, 2001), also available at the county level, are combined with crop-to-residue

ratios (CAREI, 2000, p. 24) to calculate the regional crop straw quantities. Total production of crop straw in China for 2000 was 606.4 Tg, and the rice straw, wheat straw and corn stover accounted for 76% of the total. Total open burning consumption is estimated to be 157.5 Tg for 2000, and the largest contributions are from Shandong, Jiangsu, Hebei, Heilongjiang and Henan Provinces, where the income of peasants and the rural population density are above the average in China.

China is not rich in forest resources. In recent years, forest fires in China have been reduced to 0.4‰ (the ratio of forest fires affected area and the total area of forest), far less than world average of 1‰ (FAO, 2001). There were only 5934 forest fires in China during 2000, with total burned area of 1670.98 km². Heilongjiang, NeiMongol and Fujian Provinces account for 67% of the total forest fire (State Forestry Administration, 2001). The fuel load (fuel weight per unit area) of different forest biomes in China ranged from 55 to 348 t ha⁻¹ (Tian et al., 2003). Tropical zones have the highest forest biomass fuel load, while warm temperate zones have the lowest. The burning efficiency of different forest biomes were 0.09–0.3 (Tian et al., 2003).

There were 652 cases of grassland fire during 2000 for China. The total burned area was 2891.13 km², and Heilongjiang, Nei Mongol, Sichuan and Xinjiang Provinces accounted for 95.5% of total (Ministry of Agriculture of China, 2001). The fuel load of grasslands in different climate zones were 2.2 t ha⁻¹ (Dry matter) for cold temperate, 2.4 t ha⁻¹ (Dry matter) for warm temperate (IPCC, 2003), respectively. The average burning efficiency of grassland biomass is 0.99 (Shea et al., 1996).

4. Results and discussion

4.1. National and regional emission estimates

National and regional emissions of BC and OC aerosols in China for 2000 are estimated in Table 2. Emissions are derived for 31 Provinces, excluding Hong Kong, Macao and Taiwan. Total emission were 1499.2 Gg BC and 4241.1 Gg OC (Table 2). The five largest contributions by province are from Hebei, Shandong, Henan, Shanxi and Sichuan; these are the regions with high rural population densities.

Spatial allocation (gridding) is important for model application and analysis. We use a Geographical Information System (GIS) to support the

research needs of the CMA aerosol project. Emissions from all LPS are assigned to the grid cell in which the source is found. Fig. 2 shows gridded BC and OC aerosols emissions for the year 2000 at 0.2° × 0.2° resolution. These figures are presented at the same scale, to facilitate direct comparisons. As most of the basic data are at the county level, in many cases we can better localize the emissions on higher resolution maps. Where the requisite county level data are not available, we prorate the provincial data for each category using appropriate socio-economic statistics such as population, area, industrial output or GDP. The distribution method was similar to that used by Wang et al. (2005).

The BC emission fluxes are highest (> 10 kg km⁻²) in the eastern China and central China (Fig. 2a), where the rural population density and economic level are high. The lowest BC emission areas (< 10 kg km⁻²) are in western China and Nei Mongol, with lower rural population densities and lower economic level. A similar spatial distribution of OC emissions is also seen in Fig. 2b.

4.2. Industrial

Industrial carbonaceous aerosol emissions estimated in this work include coal, oil and biofuel combustion for both urban and rural environments. The complexity of the industrial structure in China, with respect to the wide variety of fuels, boilers, combustion conditions and control devices in different types of combustors, and the fact that some statistical data were difficult to obtain, makes accurate estimations of carbonaceous aerosol emissions very difficult. One would expect that because of the greater attention to atmospheric impacts and emission controls, industrial combustion sources would tend to have lower emissions than domestic ones. Moreover, pollution control efficiency obviously differs between urban and rural regions. Average PM removal efficiency for urban industry was relatively high, ~80% in this work (SEPA, 2000) compared with only 26% for rural industry (SEPA, 1997).

Total emission from this sector was 543.9 Gg BC and 1116.1 Gg OC (Table 3), these composed 36.3% and 26.3% of total emissions, respectively. The majority of these emissions were from uncontrolled or poorly controlled coal-fired boilers, kilns and furnaces in rural industry; these amounted to 374.7 Gg BC and 830.1 Gg OC. In general, pollution from rural industries has received greater attention

Table 2
Provincial inventories of BC and OC emissions in 2000 (Gg)

Province	BC						OC					
	Biomass burning	Power plant	Transportation	Industry	Residential	Total	Biomass burning	Power plant	Transportation	Industry	Residential	Total
Beijing	1.2	0.1	0.9	7.1	7.6	16.9	5.0	0.2	1.4	11.7	25.8	44.1
Tianjin	0.8	0.2	0.7	5.5	7.2	14.4	3.5	0.2	1.1	9.8	22.8	37.4
Hebei	8.6	0.7	2.1	42.4	71.6	125.3	35.4	0.8	2.9	83.1	193.7	315.9
Shanxi	2.4	0.4	1.0	37.8	74.1	115.7	9.4	0.5	1.4	81.1	153.3	245.7
Nei Mongol	4.3	0.4	0.7	5.0	15.5	25.8	19.1	0.5	0.9	8.6	54.0	83.1
Liaoning	3.9	0.4	1.2	21.4	29.8	56.8	12.1	0.5	1.7	38.2	101.9	154.5
Jilin	7.1	0.2	0.7	11.5	22.4	41.9	20.8	0.2	1.0	23.2	72.5	117.8
Heilongjiang	8.9	0.3	0.8	9.6	31.4	51.0	34.2	0.4	1.1	18.0	110.9	164.5
Shanghai	0.5	0.3	0.6	14.5	0.4	16.3	2.1	0.4	0.8	18.9	11.9	34.0
Jiangsu	7.6	0.8	1.2	36.7	31.1	77.4	35.6	1.0	1.8	72.1	143.7	254.2
Zhejiang	2.9	0.4	1.3	37.3	15.5	57.3	12.2	0.5	1.8	76.1	66.1	156.7
Anhui	3.7	0.2	0.7	26.9	33.0	64.5	17.1	0.3	0.9	58.3	127.6	204.3
Fujian	2.0	0.1	0.6	8.1	4.7	15.5	12.1	0.1	0.8	18.3	18.1	49.4
Jiangxi	1.9	0.1	0.5	13.8	21.8	38.1	8.0	0.2	0.7	32.5	72.1	113.4
Shandong	13.6	0.8	2.0	43.9	57.4	117.7	57.8	1.0	2.9	87.8	204.4	353.9
Henan	8.1	0.5	1.7	42.3	66.3	118.9	37.5	0.6	2.4	88.3	188.1	316.9
Hubei	3.8	0.2	0.8	27.1	33.9	65.7	16.4	0.2	1.1	62.3	129.2	209.3
Hunan	2.7	0.2	0.8	30.3	45.5	79.6	10.4	0.2	1.2	66.2	146.8	224.8
Guangdong	3.2	0.5	3.0	22.8	18.4	47.9	12.6	0.6	4.3	43.6	88.6	149.7
Guangxi	1.7	0.1	0.5	17.7	18.8	38.8	6.5	0.1	0.8	42.4	81.8	131.6
Hainan	0.3	0.0	0.1	0.9	7.6	9.0	1.3	0.0	0.2	1.8	35.0	38.3
Chongqing	1.5	0.1	0.3	7.6	21.7	31.3	5.9	0.1	0.5	15.2	67.0	88.8
Sichuan	4.5	0.1	1.1	35.2	44.3	85.3	19.0	0.2	1.6	74.9	148.1	243.7
Guizhou	1.1	0.2	0.5	5.6	41.7	49.2	4.5	0.2	0.6	11.2	105.6	122.2
Yunnan	1.2	0.1	1.1	8.4	23.0	33.7	4.4	0.1	1.6	20.3	77.0	103.4
Xizang	0.0	0.0	0.1	0.0	0.0	0.1	0.2	0.0	0.1	0.0	0.0	0.3
Shannxi	1.8	0.2	0.6	16.5	24.3	43.3	7.5	0.2	0.8	35.2	73.4	117.2
Gansu	1.2	0.1	0.4	6.1	20.0	27.8	5.4	0.1	0.6	14.1	59.0	79.2
Qinghai	0.1	0.0	0.2	0.3	4.7	5.3	0.6	0.0	0.2	0.4	13.8	15.0
Ningxia	0.5	0.1	0.2	0.4	4.4	5.6	1.9	0.1	0.3	0.5	11.4	14.3
Xinjiang	1.8	0.1	0.7	1.4	19.3	23.3	7.5	0.1	0.9	1.8	47.1	57.5
Total	103.0	7.9	26.8	543.9	817.6	1499.2	425.9	9.8	38.4	1116.1	2650.8	4241.1

in recent years. In order to control pollution, the Chinese government closed 84,000 small factories with serious pollution problems in 2000 (SEPA, 2000). With better pollution controls, emissions from urban industries are lower than those in rural areas, 137.4 Gg for BC and 174.2 Gg for OC.

4.3. Power generation

In China, approximately 67% of the electric energy is derived from coal combustion and 22% from oil combustion (State Power Corporation of China, 2001). Because the combustion temperatures in power plants, which are predominantly pulverized-coal burners, are high, the amounts of carbonaceous materials formed are relatively low. Furthermore, all new power plants and many of

the large, old power plants use electrostatic precipitators to collect particulate matter. The average removal efficiency of these devices was reported to be 95% (State Power Corporation of China, 2001), and so the BC and OC emission are relative low, only 7.9 and 9.8 Gg, contributing about 0.5% and 0.2% of total emissions, respectively.

4.4. Transportation

Emissions from vehicles fueled with gasoline are gaseous pollutants like carbon monoxide, nitrogen oxides and hydrocarbons. Diesel vehicles emit more PM compared with gasoline vehicles, and diesel-generated carbonaceous aerosols account for 76–80% of total PM (Durbin and Norbeck, 2002). There were 16.1 million total vehicles used in China

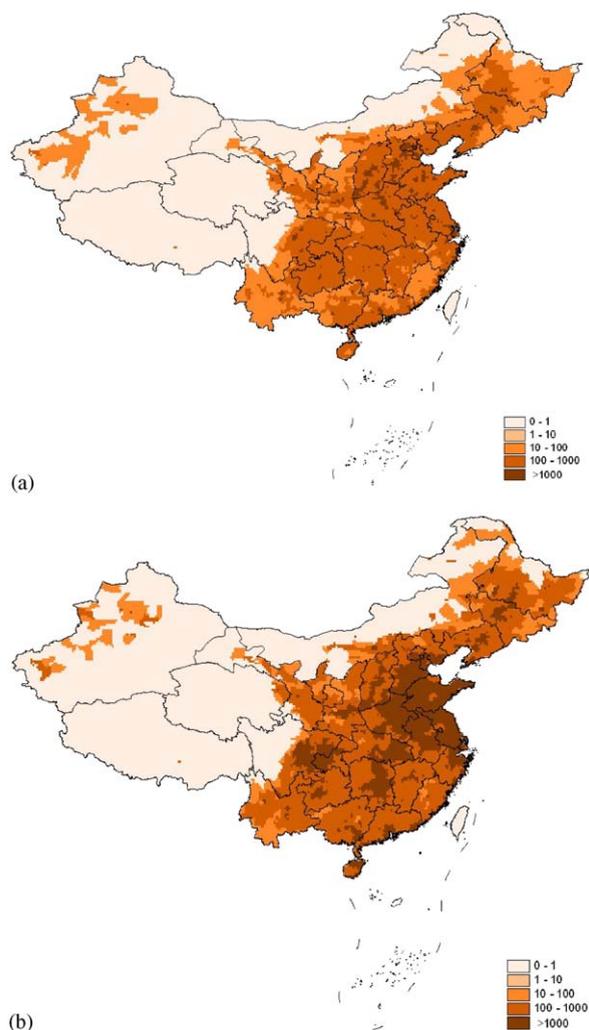


Fig. 2. Gridded carbonaceous aerosol emissions for the year 2000 in China: (a) BC, (b) OC. ($0.2^\circ \times 0.2^\circ$, unit: kg km^{-2}).

during 2000, of which about 25% were fueled by diesel (CATARC, 2001). Since EFs per mass of fuel are very similar among vehicles (Yanowitz et al., 2000; Bond et al., 2004) and lack of regulation and level of maintenance data in China, we do not differentiate between light- and heavy-duty vehicles, only by the fuel used. Emissions from transportation were not high in 2000, 26.8 Gg for BC and 38.4 Gg for OC (1.8% and 0.9% of total emissions, respectively).

4.5. Residential

The largest contribution to carbonaceous aerosol emissions in China comes from the use of raw coal, coal briquettes, and biofuels in the residential

Table 3
Summary of BC and OC emission estimates by sectors and fuel types (Gg)

Sector	Fuel	BC emissions	OC emissions
Industry	Urban industry-coal	122.4	162.7
	Urban industry-oil	15.0	11.5
	Rural industry-coal	374.7	830.1
	Rural industry-oil	10.5	5.5
	Biofuel	21.3	106.3
	Subtotal	543.9	1116.1
Power generation	Coal	7.8	9.8
	Oil	0.1	0.1
	Subtotal	7.9	9.8
Biomass burning	Agriculture waste	100.0	395.8
	Forest fire	2.7	27.4
	Grassland fire	0.4	2.7
	Subtotal	103.0	425.9
Transportation	Diesel	25.5	34.6
	Gasoline	1.3	3.8
	Subtotal	26.8	38.4
Residential	Agriculture residues	186.6	771.2
	Firewood	109.2	545.8
	Rural residential-coal	487.1	899.0
	Rural residential-oil	0.4	0.6
	Urban residential-coal	33.7	433.8
	Urban residential-oil	0.6	0.4
	Subtotal	817.6	2650.8
Total		1499.2	4241.1

sector. Since these fuels are burned in small domestic stoves, cookers, and heaters without any emission controls, emission of fine particles are greatly enhanced under these conditions, EFs are highly than industrial or power production processes. Household stoves, even if individually small, are numerous and thus have the potential to contribute significantly to inventories of aerosol (Zhang et al., 2000; He et al., 2004).

Total emissions from this sector were 817.6 Gg BC and 2650.8 Gg OC (Table 3), composed 54.5% and 62.5% of total emissions, respectively. The majority of these emissions were from uncontrolled or poorly controlled use of raw coal and coal briquettes for cooking or heating in rural China (487.1 Gg BC and 899.0 Gg OC). Emissions from

agriculture residues and firewood in rural China were also large, especially the region with high rural population density (Table 2). Emissions from urban residential is lower, 34.3 Gg BC and 433.8 Gg OC, respectively.

4.6. Biomass burning

The estimated emission from biomass open burning were 103.0 Gg BC and 425.9 Gg OC, contributing 6.9% and 10.0% of total emissions, respectively. The majority of these emissions were from agriculture waste open burning in field after harvest, amounted to 100.0 Gg BC and 395.8 Gg OC. Agriculture waste open burning has been a common practice in China, getting more serious recent years, and causing regional environment pollution (Duan et al., 2004; Wu et al., 2001). Since forest fire and grassland fire occurred in China was maintained at a low level for 2000, emissions from these biomass burning sources are low (Table 2).

4.7. Comparison with other emission inventories

Our emission inventories differ from previous inventory in several aspects. First, our inventories are on the basis of the latest year data that is just available—China government statistic data of fuel consumption, population and technology in 2000. Second, EFs of biofuel combustion, residential coal combustion and vehicle were derived from local measurements. The comparisons of different inventories of China are shown in Fig. 3. The BC emission inventories of Cooke et al. (1999) were only from fossil fuel combustion. Penner et al. (1993) derived the BC emission estimated by using the ratios of BC to “smoke” and SO₂ measurements, proposed that BC emissions could be inferred from observed BC/S ratios in different parts of the world, and got the value of 2680 Gg for

China, which is not directly comparable with our work.

Our estimated BC/OC emissions are higher than previous inventories, mainly due to our inclusion of emissions from coal combustion in rural industry and rural resident, which were significant but often underestimated (Table 3). The same result of underestimated has been described by Wang et al. (2005). The OC/BC ratio in this work is 2.8, larger than the value of 1.9 in Bond et al. (2004), but lower than the value of 3.2 in Streets et al. (2003). Since BC emissions in 1996 by Bond et al. (2004) are larger than Streets et al. (2003) and close to our value, and that of OC emission is the smallest, resulting in the smallest OC/BC ratio in Bond et al. (2004).

Estimated BC emission from biomass burning (103.0 Gg) matches the value of Streets et al. (2003) for 112.4 Gg and Bond et al. (2004) for 130 Gg, but OC emission (425.9 Gg) is lower than their values of 728.1 and 717 Gg. BC emission from industry (543.9 Gg) is significantly higher than the value of Streets et al. (2003) for 88.94 Gg, but agreement with Bond et al. (2004) of 430 Gg, however, OC emission (1116.1 Gg) is significantly higher than their values for 27.61 and 282 Gg, respectively. The poorer control devices for coal combustion in rural industry and rural residences may gave a big contribution to these types of fuel use. Emissions from residential sector and power plants somewhat agrees with the values of Streets et al. (2003) and Bond et al. (2004) well. Estimated emissions from transportation are lower than the value of Streets et al. (2003) and Bond et al. (2004), due to the relative lower EFs of local testing were used.

5. Seasonality of emission

One can expect seasonal variations of BC/OC emissions because (1) the heating energy used for cooking in rural China will depend on outdoor temperature; (2) peoples in northern China may use coal or biofuel combustion for heating, but people in southern China may not; (3) agriculture waste open burning in field occurred mainly after the harvest period; (4) peasant workers in China has a strong mobility. Taking into account heating in resident, mobility of rural resident population and crop harvest time for each province of China into consideration, monthly fraction of emissions at national level were illustrated in Fig. 4. Two peaks were found for BC/OC emissions in May and

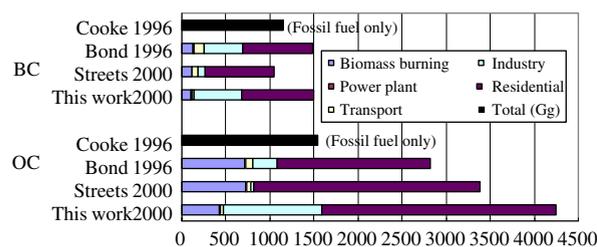


Fig. 3. Comparison of BC and OC emission inventories (Gg).

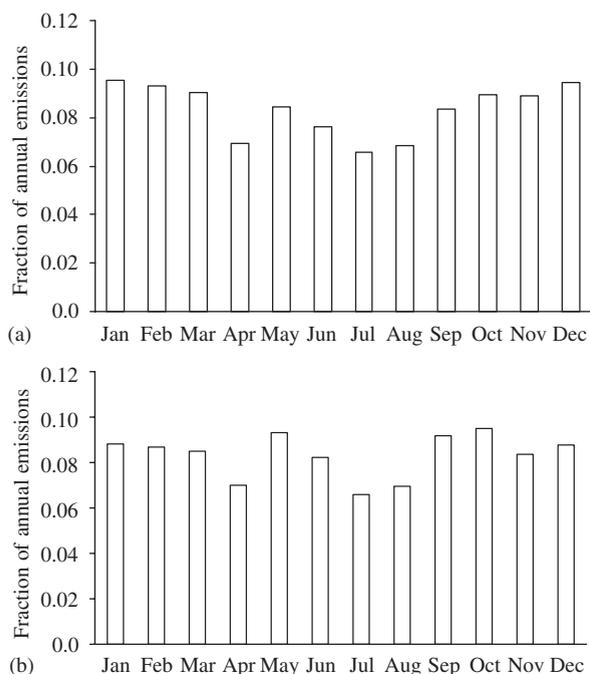


Fig. 4. Seasonality of carbonaceous aerosol emission for the year 2000 in China: (a) BC, (b) OC.

October, with significant lower emissions in April and July. The factors for highest value to lowest ones are all 1.4 for BC and OC. This significant monthly difference in BC/OC emissions are mainly due to strong seasonality of residential heating and agriculture waste open burning.

6. Conclusions

This paper presents detailed inventories of anthropogenic carbonaceous aerosol emissions for China during 2000; these are mainly based on the available information concerning fuel sources and emission factors. The latest fuel consumption data, which included fossil fuel and biomass fuel and socio-economic statistics were obtained from governmental agencies. Some new EFs from local measurements were developed and used in the assessment. National and regional summaries of emissions of carbonaceous aerosols in China were calculated, as were gridded emissions at $0.2^\circ \times 0.2^\circ$ resolution. We estimate 1499.2 Gg of BC and 4121.1 Gg of OC were emitted from sources in China, making it a significant global source of carbonaceous aerosol.

Our estimated emissions are higher than those from previous studies, mainly because the inclusion

of coal combustion by rural industries and residences proved to be significant. The residential sector and industrial emissions were the major contributors to the total carbonaceous aerosol emissions. Emission densities were higher in eastern China and lower in western China. Strongly seasonality was observed for the emissions. We find peaks in May and October, with significantly lower emissions in April and July. The monthly differences in BC/OC emissions are mainly due to the strong seasonality of residential heating and the open burning of agriculture wastes.

Since there are few locally measured EFs in China, we were forced to use some data from other countries or in some cases global average EFs. Future studies should be designed to take into account local knowledge and expand the measurements made in China.

Acknowledgments

We thank Dr. R. Arimoto for the revision of the manuscript. This work was supported by China Meteorological Administration Climate Change Research Foundation (CCRF 2006-2). We wish to thank members of CMA Aerosol Project team, members of CAWAS and Dr. Y.Q. Wang at Chinese Academy of Meteorological Sciences who helped us with GIS work.

References

- Andreae, M.O., Merlet, P., 2001. Emissions of trace gases and aerosols from biomass burning. *Global Biogeochemical Cycles* 15 (4), 955–966.
- Bond, T.C., Streets, D.G., Yarber, K.F., Nelson, S.M., Woo, J., 2004. A technology-based global inventory of black and organic carbon emissions from combustion. *Journal of Geophysical Research* 109, D14203.
- Cao, G.L., Zhang, X.Y., Wang, D., Zheng, F.C., Qu, W.J., 2005. Inventory of pollutants emission from biofuel burning in rural China, Part I—experiment. *Environmental Management*, in revision.
- CAREI, Strategic considerations for development and utilisation of biological energy in China, Chinese Association for Rural Energy Industries (in Chinese). July, 2000. Beijing, 61pp.
- Chameides, W.L., et al., 1999. Case study of the effects of atmospheric aerosols and regional haze on agriculture: an opportunity to enhance crop yields in China through emission controls? *Proceedings of the National Academy of Sciences* 96, 13,626–13,633.
- Chen, X.F., 2001. Economics analysis on pollution from straw burning and managing in rural China. *China Rural Economy* 2001 (2), 47–52 (in Chinese).

- Chen, Y.J., et al., 2005. Emission factors of carbonaceous particles and polycyclic aromatic hydrocarbons from residential coal combustion in China. *Environmental Science and Technology* 39, 1861–1867.
- China Automotive Technology & Research Center (CATARC), 2001. *China Automotive Industry Yearbook 2000*. CATARC press, Tianjin, China.
- Cooke, W.F., Liousse, C., Cachier, H., Feichter, J., 1999. Construction of a $1^\circ \times 1^\circ$ fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model. *Journal of Geophysical Research* 104, 22,137–22,162.
- Cooke, W.F., Ramaswamy, V., Kasibhatla, P., 2002. A general circulation model study of the global carbonaceous aerosol distribution. *Journal of Geophysical Research* 107 (D16).
- Cooke, W.F., Wilson, J.J.N., 1996. A black carbon aerosol model. *Journal of Geophysical Research* 101, 19395–19409.
- Dasch, J.M., 1982. Particulate and gaseous emissions from wood-burning fire places. *Environmental Science and Technology* 16, 639–645.
- Duan, F.K., Liu, X.D., Yu, T., 2004. Identification and estimate of biomass burning contribution to the urban aerosol organic carbon concentrations in Beijing. *Atmospheric Environment* 38 (9), 1275–1282.
- Durbin, T.D., Norbeck, J.M., 2002. Comparison of Emissions for Medium-Duty Diesel Trucks Operated on California In-Use Diesel, ARCO's EC-Diesel, and ARCO C-Diesel with a Diesel Particulate Filter. National Renewable Energy Laboratory Under Contract # ACL-1-30110-01 And The Ford Motor Company, US, July 2002.
- Editorial Board of China Rural Energy Yearbook, 2001. *China Rural Energy Yearbook 2000*. China Agriculture Press, Beijing, China.
- Editorial Board of China Steel Yearbook, 2001. *China Steel Yearbook 2000*. Metallurgical Industry Press, Beijing, China.
- Food and Agriculture Organization (FAO), 2001. Global forest fire assessment, 1990–2000. Rome.
- Ghedini, N., Gobbi, G., Sabbioni, C., Zappia, G., 2000. Determination of elemental and organic carbon on damaged stone monuments. *Atmospheric Environment* 34, 4383–4391.
- Haywood, J., Boucher, O., 2000. Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: a review. *Reviews of Geophysics* 38 (4), 513–543.
- Hansen, J., Sato, M., 2001. Trends of measured climate forcing agents, *Proceedings of the National Academy of Sciences* 98 (26), 14778–14783.
- He, K.B., et al., 2001. The characteristics of PM_{2.5} in Beijing, China. *Atmospheric Environment* 35, 4959–4970.
- He, L.Y., Hua, M., Huang, X.F., Yu, B.D., 2004. Measurement of emissions of fine particulate organic matter from Chinese cooking. *Atmospheric Environment* 38, 6557–6564.
- Hu, D.Z., 2000. The Utilizing Status and Prospects of the Crop Straw Resources in China. *Resource Development & Market* 16 (1), 19–20 (in Chinese).
- International Energy Agency (IEA), 2002. *Energy Statistics of Non-OECD Countries*, Paris.
- IPCC (Inter-governmental Panel on Climate Change), 2001. *Third Assessment Report Climate Change 2001: The Scientific Basis*. Cambridge University Press, UK.
- IPCC (Inter-governmental Panel on Climate Change), 2003. *Good Practice Guidance for Land Use, Land-Use Change and Forestry*, Chapter 3: LUCF Sector Good Practice Guidance(Task 1). IGES,2003.
- Jacobson, M.Z., 2001. Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols. *Nature* 409, 695–697.
- Jacobson, M.Z., 2002. Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming. *Journal of Geophysical Research* 107 (D19), 4410.
- Jenkins, B.M., Turn, S.Q., Williams, R.B., Goronea, M., Abd-el-Fattah, H., 1996. Atmospheric pollutant emission factors from open burning of agricultural and forest biomass by wind tunnel simulations, Volume 1. California State Air Resources Board (NTIS PB97-133037).
- Kirchstetter, T.W., et al., 1999. On-road measurement of fine particle and nitrogen oxide emissions from light- and heavy-duty motor vehicles. *Atmospheric Environment* 33 (18), 2955–2968.
- Klimonta, Z., Streets, D.G., Gupta, S., Cofala, J., Fu, L.X., Ichikawa, Y., 2002. Anthropogenic emissions of non-methane volatile organic compounds in China. *Atmospheric Environment* 36 (8), 1309–1322.
- Lin, R.Q., Song, D.L., 2002. Utilizing status and problems of crop straw on Guangdong province. *Soil and Environmental sciences* 11 (1), 110 (in Chinese).
- Liousse, C., Penner, J.E., Chuang, C., Walton, J.J., Eddleman, H., Cachier, H., 1996. A global three-dimensional model study of carbonaceous aerosols. *Journal of Geophysical Research* 101, 19411–19422.
- Ministry of Agriculture of China, 2001. *China Agriculture Yearbook 2001*. China Agriculture Press, Beijing, China.
- National Bureau of Statistics of China (NBSC), 2001a. *China Energy Statistical Yearbook 2000*. China Statistical Press, Beijing, China.
- National Bureau of Statistics of China (NBSC), 2001b. *China Statistical Yearbook 2001*. China Statistics Press, Beijing, China.
- Novakov, T., Penner, J.E., 1993. Large contribution of organic aerosols to cloud-condensation-nuclei concentrations. *Nature* 365, 823–826.
- Park, R.J., Jacob, D.J., Chin, M., Martin, R.V., 2003. Sources of carbonaceous aerosols over the United States and implications for natural visibility. *Journal of Geophysical Research* 108 (D12), 4355.
- Penner, J.E., Eddleman, H., Novakov, T., 1993. Towards the development of a global inventory for black carbon emissions. *Atmospheric Environment* 27A, 1277–1295.
- Qiu, J., Yang, L., 2000. Variation characteristics of atmospheric aerosol optical depths and visibility in North China during 1980–1994. *Atmospheric Environment* 34, 603–609.
- Reddy, M.S., Venkataraman, C., 2000. Atmospheric optical and radiative effects of anthropogenic aerosol constituents from India. *Atmospheric Environment* 34, 4511–4523.
- Reddy, M.S., Venkataraman, C., 2002a. Inventory of aerosol and sulphur dioxide emissions from India: I—fossil fuel combustion. *Atmospheric Environment* 36, 677–697.
- Reddy, M.S., Venkataraman, C., 2002b. Inventory of aerosol and sulphur dioxide emissions from India: Part II—biomass combustion. *Atmospheric Environment* 36, 699–712.

- Shea, R., et al., 1996. Fuel biomass and combustion factors associated with fires in savanna ecosystems of South Africa and Zambia. *Journal of Geophysical Research* 101 (D19), 23,551–23,568.
- Shulman, M.L., Jacobson, M.C., Carlson, R.J., Synovec, R.E., Young, T.E., 1996. Dissolution behavior and surface tension effects of organic compounds in nucleating cloud droplets. *Geophysical Research Letters* 23 (3), 277–280.
- Singh, M., Jaques, P.A., Sioutasa, C., 2002. Size distribution and diurnal characteristics of particle-bound metals in source and receptor sites of the Los Angeles Basin. *Atmospheric Environment* 36, 1675–1689.
- State Environmental Protection Administration (SEPA), 2000. Report on the State of the Environment in China. State Environmental Protection Administration of China, Beijing.
- State Environmental Protection Administration (SEPA), 1997. Investigation Report on Sources of Township Industrial Pollution (in 1997). State Environmental Protection Administration of China, Beijing.
- State Forestry Administration, 2001. China Forestry Statistics Year book 2001. China Forestry Publishing House, Beijing, China.
- State Power Corporation of China, 2001. China Electricity Yearbook 2000. China Electricity Press, Beijing, China.
- Streets, D.G., et al., 2001. Black carbon emissions in China. *Atmospheric Environment* 35, 4281–4296.
- Streets, D.G., et al., 2003. An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. *Journal of Geophysical Research* 108 (D21), 8809.
- Streets, D.G., Bond, T.C., Lee, T., Jang, C., 2004. On the future of carbonaceous aerosol emissions. *Journal of Geophysical Research* 109 (D24212).
- Tian, X.R., Shu, L.F., Wang, M.Y., 2003. Direct Carbon Emissions from Chinese Forest Fires, 1991–2000. *Fire Safety Science* 12 (1), 6–10 (in Chinese).
- Turn, S.Q., Jenkins, B.M., Chow, J.C., Pritchett, L.C., 1997. Element characterization of particulate matter emitted from biomass burning: wind tunnel derived source profiles for herbaceous and wood fuels. *Journal of Geophysical Research* 102 (D3), 3683–3699.
- Turpin, B.J., Huntzicker, J.J., 1991. Secondary formation of organic aerosol in the Los Angeles Basin: a descriptive analysis of organic and elemental carbon concentrations. *Atmospheric Environment* 25A, 207–215.
- Turpin, B.J., Huntzicker, J.J., 1995. Identification of secondary aerosol episodes and quantization of primary and secondary organic aerosol concentrations during SCAQS. *Atmospheric Environment* 29, 3527–3544.
- Wang, X.P., et al., 2005. A high-resolution emission inventory for eastern China in 2000 and three scenarios for 2020. *Atmospheric Environment* 39, 5917–5933.
- Wang, Y., 2002. The studies on the characteristic of motor vehicle particle emissions in Beijing district. Ph.D. dissertation. Jilin University, China, Jilin.
- Weng, W., Yang, J.T., Zhao, Q.L., Zhang, B.L., 2004. Current situation and developing direction of straw utilization technology in China. *China Resources Comprehensive Utilization* 2004 (7), 19–21 (in Chinese).
- Wu, L., Chen, J., Zhu, X.D., Xu, Y.P., Feng, B., Yang, L., 2001. Straw-burning in rural areas of China: caused and controlling strategy. *China Population, Resources and Environment* 11, 110–112 (in Chinese).
- Yanowitz, J., McCormick, R.L., Graboski, M.S., 2000. In-use emissions from heavy-duty diesel vehicles. *Environmental Science & Technology* 34 (5), 729–740.
- Yao, Z., Wang, S.H., Jiang, X.H., 2001. The current situation and approach of return straw to field in suburb of Shanghai. *Agro-Environment and Development* 2001 (3), 40–41 (in Chinese).
- Ye, B.M., et al., 2003. Concentration and chemical composition of PM_{2.5} in Shanghai for a 1-year period. *Atmospheric Environment* 37, 499–510.
- Yu, Z., 2003. The developing trend of resources treatment of crop stalk in Fuzhou city (in Chinese). *Fujian Environment* 20 (5), 31–32.
- Zhang, J., et al., 2000. Greenhouse gases and other airborne pollutants from household stoves in China: a database for emission factors. *Atmospheric Environment* 34 (26), 4537–4549.