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Primary aerosol emission trends for China, 1990–2005

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Abstract

An inventory of anthropogenic primary aerosol emissions in China was developed for 1990–2005 using a technology-based approach. Taking into account changes in the technology penetration within industry sectors and improvements in emission controls driven by stricter emission standards, a dynamic methodology was derived and implemented to estimate inter-annual emission factors. Emission factors of $PM_{2.5}$ decreased by 7%–69% from 1990 to 2005 in different industry sectors of China, and emission factors of TSP decreased by 18%–80% as well. Emissions of $PM_{2.5}$, PM_{10} and TSP presented similar trends: increased in the first six years of 1990s and decreased until 2000, then increased again in the following years. Emissions of TSP reached a historical high (35.5 Tg) in 1996, while the peak of PM_{10} (18.8 Tg) and $PM_{2.5}$ (12.7 Tg) emissions occurred in 2005. Although various emission trends were identified across sectors, the cement industry and biofuel combustion in the residential sector were consistently the largest sources of $PM_{2.5}$ emissions, accounting for 53%–62% of emission over the study period. The non-metallic mineral product industry, including the cement, lime and brick industries, accounted for 54%–63% of national TSP emissions. There were no significant trends of BC and OC emissions until 2000, but the increase after 2000 brought the historical high of BC (1.51 Tg) and OC (3.19 Tg) emissions in 2005. Although significant improvements in the estimation of primary aerosols are presented, there still exist large uncertainties. More accurate and detailed activity information and emission factors based on local tests are essential to further improve emission estimates, this especially being so for the brick and coke industries, as well as for coal-burning stoves and biofuel usage in the residential sector.

1 Introduction

Understanding China's anthropogenic aerosol emission trends has considerable scientific importance due to the broad impact of aerosols on climate and air quality. Human-

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made aerosols impact the climate system directly by enhancing the scattering and absorption of solar radiation and indirectly by providing the condensation nuclei for cloud drops and ice crystals (Ramanathan et al., 2001; Ramanathan and Carmichael, 2008). Atmospheric aerosol trends in China have been suggested as possible causes for many of the fundamental changes in regional climate that have been observed. These include the decrease of surface temperature (Qian and Giorgi, 2000; Giorgi et al., 2002, 2003; Menon et al., 2002; Qian et al., 2003; Huang et al., 2006), changes in surface solar radiation trends (Kaiser and Qian, 2002; Che et al., 2005; Qian et al., 2006; Streets et al., 2006a, 2008, 2009; Xia et al., 2007), changes in cloud properties (Kawamoto et al., 2006; Qian et al., 2006), the reduction of precipitation (Giorgi et al., 2003; Zhao et al., 2006; Huang et al., 2007; Rosenfeld et al., 2007), increased summer floods in South China and drought in North China (Menon et al., 2002), and even intensification of Pacific storm events (Zhang et al., 2007c).

Aerosols downgrade air quality and visibility, and damage human health (Pope et al., 1995). Heavy aerosol loadings have been reported throughout China, from the coast to the interior (e.g., He et al., 2001; Ho et al., 2003; Wang et al., 2006; Cao et al., 2007; Li et al., 2007; Zhang et al., 2008a). Satellite observations have also indicated the possibility of significant health hazards due to aerosol pollution throughout the country (Carmichael et al., 2009). In recent Atmospheric Brown Cloud (ABC) observations, a number of Chinese mega-cities were identified as "aerosol hot spots" from satellite observations (Ramanathan et al., 2007). To date, particulate matter less than 10 μm in diameter (PM_{10}) has been the main atmospheric pollutant exceeding the National Ambient Air Quality Standard (NAAQS) in major Chinese cities, and has been the focus of local and national government control efforts (He et al., 2002; Hao and Wang, 2005; Chan and Yao, 2008). Aerosols can also impact regional air quality through their long-range transport. Modeling studies have indicated that Beijing's PM concentrations have been significantly enhanced by anthropogenic emissions from surrounding provinces (Chen et al., 2007; Streets et al., 2007). It is even argued that aerosol concentrations found within the United States are enhanced by Asia's emissions through

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trans-Pacific transport (Heald et al., 2006; Dunlea et al., 2009). In addition to effects on atmosphere, Calcium and Magnesium in aerosols also play important roles in soil acidification process in China (Zhao et al., 2007).

A primary aerosol emission inventory for China with inter-annual trends is essential for both the atmospheric science community and China's stakeholders. Primary aerosol emission inventories that include data on particulate size ranges and inter-annual trends are available for certain developed countries through their national emission inventory systems; e.g., USA (USEPA, 2007), Canada (EC, 2007), and most Europe countries (UNECE, 2003; Vestreng, 2006). But this is not the case for developing countries like China. China's Ministry of Environmental Protection (MEP) reports annually the national total suspended particulate (TSP) emissions in the two categories of "smoke" (generated from combustion) and "dust" (generated from mechanical impact and grinding during industrial processes), but these statistics only include emissions from large industries (ECCEY, 1992–2006). Furthermore, sectoral information and the spatial distribution of emissions are not provided, and therefore these reported statistics are insufficient for comprehensive scientific study.

China's carbonaceous aerosol emissions have previously been estimated within a national inventory (Streets et al., 2001, 2008; Streets and Aunan, 2005; Cao et al., 2006) or as part of regional (Streets et al., 2003; Ohara et al., 2007) and global (Cooke et al., 1999; Bond et al., 2004) inventories, and emission trends have also been reported by some of these studies (e.g., Streets et al., 2008; Ohara et al., 2007). A few studies on emissions of base cations indicated that China's anthropogenic emissions of Ca and Mg might be larger than natural sources (Zhu et al., 2004), although significant emissions of mineral dusts come with sand storms. In our previous study, using a technology-based approach, we presented the first comprehensive estimates of primary aerosol emissions in China for the year 2001 based on three particulate size fractions, i.e., TSP, PM_{10} and fine particulate matter less than 2.5 μm in diameter ($PM_{2.5}$), and four major components, i.e., black carbon (BC), organic carbon (OC), Ca and Mg (Zhang et al., 2006, 2007b). Using the same methodology, and as part of

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INTEX-B Asian emission inventory, we then updated the estimates and reported for the year 2006 (Zhang et al., 2009). However, the temporal coverage of the above work has been limited and as yet bottom-up inventory studies have not been used to gain insights into China's anthropogenic aerosol emission trends.

The purpose of this paper is to rectify this situation by developing a comprehensive view of China's anthropogenic aerosol emission trends using bottom-up methodology. In this work, we apply model frameworks similar to those described in Zhang et al. (2006, 2007b) and we use a dynamic methodology similar to that of Zhang et al. (2007a) to reflect the dramatic change in China's aerosol emissions driven by energy growth and technology renewal. The dynamic methodology used in this study is detailed in Sect. 2. The inter-annual variations of net aerosol emission factors (EFs) derived from the dynamic methodology are then given in Sect. 3. The results, including inter-annual emissions of TSP, PM_{10}, $PM_{2.5}$, BC, OC, Ca and Mg, and gridded emissions are reported in Sect. 4. We compare our estimates with other bottom-up and top-down studies in Sect. 5, and also discuss the uncertainties associated with our analysis in that section.

2 Methodology

To date, estimating primary aerosol emissions for China remains a challenge and is much more difficult than for other gaseous pollutants. Firstly, in addition to emissions from energy consumption, primary aerosols are widely emitted from various industrial processes and construction activities, some of which are fugitive and therefore make accurate quantification of emissions from these sources very difficult. Secondly, the net aerosol emission rate from a specific sector is closely related to the degree of penetration of control technologies within that sector. Therefore an understanding of the utilization of various control technologies is necessary to allow meaningful EF estimates. Finally, but most importantly for emission trends, net EFs can change dramatically in only a few years in China because new technologies are continually coming into the

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market. For example, the building of new, large coal-fired power plants to replace or augment older, smaller plants has dramatically altered the balance of power plant technologies in use, and has reduced the average NO_x EF of the whole power sector by 16% in just 10 years (Zhang et al., 2007a). This situation could also be true for aerosol emissions.

Here we develop a dynamic, technology-based methodology to estimate the primary aerosol emissions in China. A spreadsheet model was established to calculate the emissions. The geographical extent covers 31 provinces of mainland China, and the temporal scope is 1990–2005. The key innovation of this method is the estimation of EFs on a year-by-year basis using careful examination of the utilization of new control technologies during the period, instead of using fixed EFs for all years.

2.1 Model structure and calculation method

Emissions were calculated from the combination of activity rate, technology distribution, unabated EFs, the penetration of emission control technologies and the removal efficiency of those technologies, using an approach similar to that of Klimont et al. (2002) and Zhang et al. (2007b). The emissions were estimated for three size fractions: $PM_{2.5}$, $PM_{2.5-10}$ (PM with diameter more than 2.5 μm but less than 10 μm, coarse particles), and $PM_{>10}$ (PM with diameter more than 10 μm). The basic equations are:

$$E_{i,y,z} = \sum_j \sum_k A_{i,j,k,z} \left[\sum_m X_{i,j,k,m,z} \mathrm{EF}_{j,k,m,y,z} \right] \quad (1)$$

For a given technology m, the final EF of diameter range y was estimated by the following equation:

$$\mathrm{EF}_{y,z} = \mathrm{EF}_{\mathrm{TSP}} F_y \sum_n C_{n,z}(1-\eta_{n,y}) \quad (2)$$

Where i represents the province (municipality, autonomous region); j represents the economic sector; k represents the fuel or product type; y represents the diameter

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range of PM; z represents the year; m represents the type of combustion and process technology; n represents the PM control technology; $E_{y,z}$ is the emissions of PM in diameter y in year z; A is the activity rate, such as fuel consumption or material production; X_m is the fraction of fuel or production for a sector consumed by a specific technology m, and $\sum_m X_m=1$; EF is the net EF after abatement by control devices; EF_{TSP} is the unabated EF of TSP before emission control; F_y is the mass proportion of PM in diameter y relative to total PM; $C_{n,z}$ is the penetration of PM control technology n in year z, and $\sum_n C_n=1$; $\eta_{n,y}$ is the removal efficiency of control technology n for PM in diameter y.

In addition to total aerosol emissions, we also estimated the emissions of several chemical components in aerosols: BC, OC, Ca and Mg. EFs for BC and OC were calculated as the mass ratio of BC and OC to $PM_{2.5}$ EFs, with the assumption that control technologies have the same removal efficiency for $PM_{2.5}$, BC and OC. Similarly, EFs for Ca and Mg were determined by their fraction in TSP emissions.

Emission sources are classified into three groups: stationary combustion, industrial process, and mobile sources. The stationary combustion sources involve three sectors (power plants, industry, and residential) and seven types of fuel (coal, diesel, kerosene, fuel oil, gas, wood and crop residues). The industrial process sources cover 22 products/processes in metallurgical industries, non-metallic mineral production industries and chemical industries, where cement production, coke production and iron and steel production were the most important. The mobile emission sources include seven types of on-road mobile sources: light-duty gasoline vehicles (LDGV), light-duty gasoline trucks (LDGT1), mid-duty gasoline trucks (LDGT2), light-duty diesel vehicles (LDDV), heavy-duty gasoline trucks (HDGV), heavy-duty diesel trucks (HDDV), and motorcycles (MC); and six types of off-road mobile sources: rural vehicles, tractors, construction equipment, farming equipment, locomotives and vessels.

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2.2 Combination of activity rates and EFs

2.2.1 Activity rates (*A*)

We followed our previous approach to derive activity data from a wide variety of sources, with a critical examination of the data quality (Streets et al., 2006b; Zhang et al., 2007a). Generally, fuel consumption by sector and industrial production by product can be accessed from various statistics at the provincial level. In this study, fuel consumption in stationary combustion by sector and by province was derived from the China Energy Statistical Yearbook (except diesel, see below) (CESY, National Bureau of Statistics, 1992–2007). Industrial production by product and by province was obtained from the China Statistical Yearbook (CSY, National Bureau of Statistics, 1991–2006a) and many unofficial statistics from industry associations (China Iron and Steel Industry Association, 1994–2006; China Brick Industry Association, unpublished data; China Lime Industry Association, unpublished data).

Diesel consumption was broken down into three categories: industrial boilers, on-road vehicles, and off-road vehicles and machinery, following the method described in Zhang et al., (2007a) (see Sect. 3.3 of that paper for details). For on-road vehicles, the calculation of gasoline and diesel consumption by vehicle type was further refined using a fuel consumption model developed by He et al. (2005). For off-road vehicles and machinery, fuel consumption by tractors and rural vehicles was estimated from their population, fuel economy and annual travel mileage; diesel consumption by farming and construction machinery was estimated from their total power (National Bureau of Statistics, 1991–2006b) and their average number of working hours (Nian, 2004); diesel consumption of trains and vessels was estimated based on passenger and freight turnover for railways and inland waterways, respectively, fuel economy, and the distribution of the modes of transport (YHCTC, 1991–2006).

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2.2.2 Technology distributions (X)

Unabated PM emissions are always determined by the technology used for combustion or in the industry process. Over recent decades, the balance of technologies used has changed considerably in China. For instance, the percentage of cement produced by precalciner kilns increased from 20% in the mid-1990s to 65% in 2008 (Lei et al., 2010). The distribution of the combustion technology in each sector and the processing technology for each industrial product were generally not available from national government statistics. We therefore collected these data from a wide range of published and unpublished statistics provided by various industrial associations and technology reports. The detailed data sources for the main sectors are listed in Table 1.

2.2.3 Unabated EFs (EF_{TSP} and F_y)

According to Eq. (2), net EFs for PM were determined by unabated EFs for TSP, the size distribution of PM, the penetration of PM control technologies and their removal efficiency. Unabated EFs for TSP and the size distribution were considered constant for each specific technology in stationary emission sources, as listed in Table 2. Most of the information was derived from available measurements in China or from estimates based on the actual technology level and practice (SEPA, 1996a; Zhang et al., 2000, 2006; Lei et al., 2010). EFs for similar activities from the USA AP-42 database (USEPA, 1995) and the RAINS-PM model (Klimont et al., 2002) were used where local information was lacking. The control measures for PM emissions from on-road vehicle emissions were different from stationary sources. The EFs of each type of on-road vehicle under each emission standard were derived from Zhang et al. (2006), and are listed in Table 3.

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2.2.4 Penetrations of PM control technology (C)

There is little statistical information on the penetration of PM control technologies in China's emission sources. Where data were lacking we used an alternative method to estimate the penetration of PM control technologies. We considered the Chinese government's new emission standards to be the driving force for the implementation of advanced control technologies. The typical penetration of PM control technologies in relevant emission sources was estimated according to the threshold value of each new emission standard; the progress of replacement of old technologies with advanced PM control technologies was then estimated based on the lifespan of existing control devices, as well as on the increase in industrial capacity and vehicle density. The emission standards considered in this work are listed in Table 4.

2.2.5 Removal efficiencies (η)

Four types of end-of-pipe emission control technologies were considered: cyclones (CYC), wet scrubbers (WET), electrostatic precipitators (ESP), and fabric filters (FAB). Fugitive dust control technologies were categorized into "normal practice" and "good practice". The International Institute for Applied Systems Analysis (IIASA) has summarized the removal efficiencies of these technologies based on practices in Europe (Klimont et al., 2002), but any sub-optimum operation of the control devices would lead to lower removal efficiencies. The removal efficiencies that we used are listed in Table 5; they are mostly taken from IIASA's estimation, but some changes were made based on local emission source tests made in China (Yi et al., 2006a).

2.2.6 EFs for BC, OC, Mg and Ca

BC and OC, formed during incomplete combustion, are mainly concentrated in the fine fractions. The United States Environmental Protection Agency (USEPA) has compiled the mass ratio of BC and OC in $PM_{2.5}$ for major sources in SPECIATE, a source profile

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database. But there is little systematic research on source profiles of $PM_{2.5}$, especially from boilers and kilns in China. In this study, for most emission sources we cite the mass ratio of BC and OC in $PM_{2.5}$ given by Bond et al. (2004) and Kupiainen and Klimont (2004). The exception was for residential coal stoves, because emission tests for fine PM, BC and OC have been conducted by Chinese researchers in recent years (Chen et al., 2005, 2006, 2009; Zhang et al., 2008b; Zhi et al., 2008, 2009). As such, we used the average EFs derived from the latest BC and OC emission test results (Chen et al., 2009). Although Li et al. (2009) calculated EFs for BC and OC from biofuel combustion based on local tests in China, their calculated ratio of BC/OC is much higher than the published results from other research. They attributed the high ratio both to the tested stoves having a better oxidization atmosphere and hence improved combustion efficiency and to the protocol used in BC and OC analysis. Since there is no evidence to show that stoves typically used in China will have the relatively high combustion efficiency of Li et al.'s (2009) study, we did not use their BC and OC emission factors. The mass ratios of BC and OC to $PM_{2.5}$ are listed in Table 6.

Emissions of Ca and Mg in PM come from coal burning and the raw materials used in industrial processes. Zhu et al. (2004) investigated the mass percentage of Ca and Mg in fly ash from coal combustion and the raw materials used in non-metallic mineral product industries by province. Here we use the mass ratios of Ca and Mg derived from their study, as listed in Table 7.

3 Trends in net emission factors

Net EFs for PM are not only affected by the penetration of PM control technologies, but also by the balance of technologies employed within the emission sources. In this section, we focus on some emission sources (including power plants, the cement industry, the iron and steel industry, the coke industry, residential coal stoves and on-road vehicles) which may make a significant contribution to China's PM emissions, or which may show a significant change through time.

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3.1 Power plant boilers

The power sector is the largest consumer of coal in China. China's thermal power generation increased from 0.49 trillion kWh in 1990 to 2.05 trillion kWh in 2005 (NBS, 1992–2007). Accordingly, coal consumption by China's power plants increased from 270 Tg to 1050 Tg (NBS, 1992–2007), with an annual rate of increase of 9.4% and a percentage share of total coal consumption increasing from 30% to 50%.

Pulverized boilers are the dominant combustion technology used in power plants, accounting for 92% of capacity in the power sector (SEPA, 1996a). Grate furnaces account for the remaining 8%, mostly used in small electricity generation units within industry self-supplying power plants. ESP, WET and CYC were widely used in power plants to mitigate PM emissions. In recent years, FAB has increasingly been installed, but we do not consider it in our model as its share of the power sector before 2005 is negligible. There were three emission standards for thermal power plants published from 1990 to 2005. The first release gave various standard values for new power plants using coals with different ash contents (SEPA, 1991); the second release gave a unique standard value for all new power plants (SEPA, 1996b), resulting in a phasing out of inefficient PM removal technologies such as CYC; and the third release gave a stricter standard value (SEPA, 2003), which not all power plants could meet without the use of ESP or FAB. In this work, based on the penetration rate in China of the three types of PM control technologies in the early 1990s (SEPA, 1996a) and after 2000 (China Electricity Council, unpublished data), we estimated the PM EFs from 1990 to 2005 by interpolating penetration rates of the PM control technologies based on the three versions of emission standards, as shown in Fig. 1. The estimated net EF of $PM_{2.5}$, $PM_{2.5-10}$ and $PM_{>10}$ were found to have decreased by 67%, 65%, and 54% from 1990 to 2005, respectively.

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3.2 Cement industry

China's cement industry is a typical emission source that utilizes both new, advanced technologies and older, increasingly out-moded ones. Shaft kilns, which have been replaced in many industrially more advanced countries, have played a major role in China's cement industry for a long period, and in the mid-1990s accounted for over 80% of cement production. Precalciner kilns (generally known as "new-dry process kilns" in China) increased their cement production 11 times over between 2000 and 2008, and in 2006 exceeded production from shaft kilns (Lei et al., 2010). Unabated PM EFs are different among cement-producing processes, but what greatly increased the difference in net EFs is the quite different PM control technologies utilized within cement plants.

There have been three emission standards for the cement industry in China (SEPA, 1985, 1996c, 2004). CYC was applied to recycle the raw material before publication of the first standard. After that, WET, ESP and FAB were gradually developed and introduced into the market place, enabling cement plants to reduce PM emissions. SEPA (1996a) calculated the net TSP EF to be 23.2 g/kg in the early 1990s by testing 264 cement production lines. The Chinese Research Academy of Environmental Sciences (CRAES, 2003) tested emissions from 90 cement plants utilizing advanced PM control devices, and found the average net TSP EF to be approximately 2 g/kg. Based on this information, we estimated PM EFs for different types of cement kilns in China for the period from 1990 to 2008 (Lei et al., 2010). The penetration of PM control technologies as well as the net PM EFs from 1990 to 2005 is shown in Fig. 1. The net EF of $PM_{2.5}$, $PM_{2.5-10}$ and $PM_{>10}$ decreased by 69%, 72% and 75% from 1990 to 2005, respectively.

3.3 Coke industry

China is the largest coke producer in the world. Production of coke increased 3.5 times during the period 1990–2005, driven by a tremendous demand from the domestic iron

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and steel industries and its high price on international markets. In industrially more advanced countries, coke plants are usually located within iron and steel plants, and supply coke for iron smelting. However in China, two thirds of total coke production comes from individual coke companies, many of which are equipped with small-scale indigenous ("beehive") coke production facilities.

PM is emitted not only from coke ovens, but also by several processes such as coal crushing, coal feeding and coke quenching (USEPA, 1995). However, China has no emission standard for these processes, only for the direct emissions from coke ovens (SEPA, 1996d). PM control devices are installed in most large coking plants with mechanized coking facilities, however few are installed in small plants with indigenous coking facilities. Through a similar approach described in the previous two sections, the penetration of PM control technologies and the net PM EFs were calculated from the annual production from mechanized and indigenous coking ovens, as shown in Fig. 1. Our estimations indicate that EFs increased in the first half of the 1990s as the share of coke produced from indigenous ovens increased. However, this share decreased from 49% in 1995 to 18% in 2005, resulting in a decrease in PM EFs as well.

3.4 Iron and steel industry

The iron and steel industries involve a series of interrelated processes. Besides coke production (see above), the major release points of PM include sinter production, pig iron production, steel production and casting. There are three type of technology in steel production: Open Hearth Furnace (OHF), Basic Oxygen Furnace (BOF) and Electric Arc Furnace (EAF). These processes/technologies were considered separately in our estimation of PM emissions from the iron and steel industry.

Prior to 2005, in China there have been two emission standards for the iron and steel industry (SEPA, 1988, 1996e). We assume that more efficient control technologies were promoted in most processes after the release of the 1996 standard, except for casting and OHF, which were gradually replaced by other processes after the mid-1990s. The penetration of PM control technologies before 1996 was derived from

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source test results (SEPA, 1996a), and the penetration after 1996 was calculated based on investigation of key iron and steel companies (Sino-Steel TianCheng Environmental Protection Science and Technology Co., Ltd, 2007). The trends in TSP EFs in the iron and steel industry were then estimated for different processes/technologies, as shown in Fig. 2. The EFs of TSP from sinter production, iron production, BOF and EAF decreased by 18% to 27% from 1996 to 2005, and EFs of $PM_{2.5}$ decreased by 7% to 21%.

3.5 Residential coal stove combustion

It is believed that residential coal stoves are a major source of BC emissions in China (Streets et al., 2001; Bond et al., 2004). Recent experimental research conducted in China indicated that the following three factors could lead to one or two orders of magnitude difference in EFs for BC and OC: (1) the type of coal (e.g. bituminous or anthracite), (2) the shape of the coal when it is burned (e.g. chunk or briquette), and (3) the type of stove (e.g. traditional stoves or improved stoves) (Chen et al., 2009; Zhi et al., 2009). Chen et al. (2009) estimated the BC and OC emissions from China's residential coal stoves with the assumption that the share of briquettes increase from 40% in 2000 to 80% to 2020. Assuming the mix of chunk and briquette coal changed lineally from 1990, we followed Chen et al.'s (2009) approach and estimated EFs for BC and OC for the period 1990–2005, as shown in Fig. 3. As the share of briquettes in coal consumption in residential coal stoves increased from 20% to 50%, average net EFs for BC and OC dropped by 34% and 10%, respectively.

3.6 On-road vehicles

Net annual EFs of on-road vehicles were estimated from the population of new-sale vehicles and raw EFs using similar methodology to that described by Zhang et al. (2007a). The raw EF of new-sale vehicles was estimated from the current emission standard in force at the time of manufacture. China began to implement emission control standards

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for on-road vehicles in 1999. As listed in Table 8, Beijing and Shanghai implemented the standards in advance of the other provinces of China, resulting in a greater reduction within those provinces of the average net PM EFs as the proportion of new vehicles increased through time (Fig. 4). Our estimates show that from 1999 to 2005, the national average EFs of $PM_{2.5}$ from gasoline vehicles, diesel vehicles and motorcycles decreased by 36%, 38% and 19%, respectively.

4 Estimates of PM emissions

4.1 Inter-annual emissions

4.1.1 TSP, PM_{10} and $PM_{2.5}$

Figure 5 shows an overview of inter-annual trends of PM emissions by particle size, as well as the contribution of PM emissions by sector from 1990 to 2005. The breakdown of emissions of $PM_{2.5}$, PM_{10} and TSP by sector in 1990, 1995, 2000 and 2005 is listed in Table 9. PM emissions increased rapidly in the six years after 1990 and reached a high of 35.5 Tg for TSP in 1996. Rapid development of the economy and the rise in energy consumption were the major driving forces of this trend in emissions. From 1996 to 2000, the decrease in PM emissions can be attributed to a much reduced increase of energy consumption and industrial production, coupled with the implementation of several new emission standards. After 2000, industries with high PM emissions developed at an enormous speed. Production of steel, cement and aluminium increased by 179%, 79% and 157% in 5 years, respectively, while additionally coal consumption for power generation increased by 88%. These dramatic increases in the macro-economy and in energy consumption offset the effects of utilizing more efficient PM control technologies, and led to increases of PM emissions, especially for fine PM, after 2000. As a result, emissions of $PM_{2.5}$ and PM_{10} reached historical highs of 12.9 Tg and 18.8 Tg, respectively, in 2005.

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The cement industry and biofuel combustion in the residential sector were the largest emitters of $PM_{2.5}$ in China, accounting for 54%–62% of emissions during the period 1990–2005. Power plants contributed about 10% of total $PM_{2.5}$ emissions, a value similar to the total emissions from other coal combustion sources. $PM_{2.5}$ emissions from mobile sources were minor relative to other sources; however their proportion in total $PM_{2.5}$ emissions more than doubled in the 15-year study window (from 1.3% to 2.9%).

The lime and brick industries are more important in terms of emissions of larger particles. The non-metallic mineral product industry, including the cement, lime and brick industries, accounted for 55%–65% of national TSP emissions. This estimate is larger than the official statistical data (ECCEY, 1992–2006). We attribute the difference to the absence from the official data of emission estimates from small plants. These small plants commonly lack emission control devices and moreover are generally not included in official emission statistics because of their diffused distribution over rural China, away from cities.

Figure 6 shows the $PM_{2.5}$ emissions by province in 1990, 1995, 2000 and 2005. Shandong, Hebei, Jiangsu, Henan, Guangdong and Sichuan combined accounted for about 40% of total $PM_{2.5}$ emissions in China. Emissions of PM_{10} and TSP have a similar distribution across provinces to that of $PM_{2.5}$. PM emissions from provinces that have more advanced economies, such as Beijing, Shanghai, Guangdong, Jiangsu and Zhejiang, showed a reduction after 1995. This trend is due to the requirements of local government for greater environmental protection and the transition of the economy from heavy industry to high-tech and commercial sectors. However, emissions from Shandong, Hebei and Henan increased, especially after 2000, a trend consistent with the construction of many new power, cement, and iron and steel plants in these provinces. Emissions from all western provinces increased after 2000, reflecting the impact of the government's "West China Development" policies.

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4.1.2 Carbonaceous aerosols

Emissions of BC increased from 1.1 Tg in 1990 to 1.5 Tg in 2005, and emissions of OC varied between 2.5 and 3.2 Tg for the same period, as shown in Table 9 and Fig. 7. Significant increase was presented for both BC and OC emissions during 2000–2005. Most of the increase (0.13 Tg of BC and 0.51 Tg of OC) were due to bio fuel combustion, followed by Coke industry (0.09 Tg of BC and 0.11 Tg of OC) and mobile sources (0.04 Tg of BC and 0.02 Tg of OC). The residential sector is the largest contributor of carbonaceous aerosol emissions, accounting for 47%–69% of China's total BC emissions and 81%–92% of total OC emissions.

Compared with developed countries such as the United States and OECD Europe, where the transportation sector contributes most to anthropogenic BC emissions (203 of 354 Gg in the USA, and 226 of 343 Gg in OECD Europe), the transportation sector is much less important in China in terms of national BC emissions (Bond et al., 2004). Total BC emissions from China's mobile sources, including on-road transportation and off-road mobile sources, was 187 Gg in 2005, much less than that of the industrial (609 Gg) or residential (701 Gg) sectors. Figure 8 illustrates the large differences in BC emissions among sectors and provinces that our analysis identified. Industries such as coke and brick-making plants are the most significant contributors in northern China (Hebei, Shanxi, Shandong and Henan), while the residential sector is the dominant source of emissions in the south, and especially in the southwest (e.g., Guangxi, Chongqing and Sichuan) since much more coal and biofuel are used there. This indicates that strategies to control BC emissions should be tailored to the local emission source profile.

4.1.3 Ca and Mg

Figure 9 shows the emission trends of Ca and Mg in China. The cement and lime industries contribute 90% of total Ca emissions, while production of cement, iron, steel, lime and brick contribute 75% of total Mg emissions. Ca and Mg showed similar emis-

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sion trends in the 1990s: an increase in the first 6 years followed by a decrease. After 2000, emissions of Ca were relatively stable, although they show a decrease in 2005. However emissions of Mg showed a further increase from 2000 to 2005, a trend that can mainly be attributed to increased emissions from the iron and steel industries.

Our estimates of emissions in 2001 (6.11 Tg Ca and 0.29 Tg Mg) are higher than those of Zhang et al. (2007b), who estimated emissions of 4.52 Tg and 0.23 Tg, respectively. Further examination reveals that the discrepancy is due to the different data sources used for brick and lime production. There were more than 80 000 small brick workshop and about 5000 small lime plants in China (Zhou, 2003), but there is no statistical data on production of brick and lime in recent years. This situation therefore increases the uncertainty of any estimation of Ca and Mg emissions.

4.2 Trends in several key sectors

Trends of PM emissions were found to be different for each sector. Here we discuss six key sectors that either emitted large amounts of PM or showed a sharp change in emissions.

4.2.1 Power plant boilers

PM emissions from power plants rose from 1990 to 1996, and then dropped until 2000. With significant increases in power generation since 2000, PM emissions increased again after 2000, and reached their historical high in 2005 (1.4 Tg $PM_{2.5}$, 2.3 Tg PM_{10}, and 3.1 Tg TSP).

Estimates of PM emissions were compared with China's governmental statistical data (ECCEY, 1992–2006) in Fig. 10a. Our estimates are about 25% lower than the statistical data, but show a similar inter-annual trend. Since the government's statistics are mostly based on calculated emissions, not derived from monitored data, we attribute the difference between the government's estimates and our own to the different parameter values used in the calculations. We also compared our PM emissions in

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2001 and 2003 with Zhang et al. (2007b) and Yi (2006b), and the differences are much less (approximately 2%).

4.2.2 Cement industry

As a major contributor of PM emissions, the cement industry accounts for about 30% of total emissions in China. Historically there have been two periods where cement production increased very rapidly: 1990–1995, when the average annual rate of increase was 17.8%, and 2002–2005, when the average annual rate of increase was 12.4%. However, the emissions of PM show a different trend in these two periods, as shown in Fig. 10b. In the first period, PM emissions increased rapidly and reached their historical high in 1997, with 4.4 Tg $PM_{2.5}$, 7.2 Tg PM_{10} and 10.4 Tg TSP. With the implementation of a new emission standard that was released in 1996, and the slowing down in the expansion of the cement industry, PM emissions dropped in the late 1990s. In spite of a rapid increase in cement production after 2000, PM emissions remained at around 8 Tg, because the widespread replacement of older shaft kilns by newer precalciner kilns offset any potential increase in PM emissions. From 2004 to 2005, cement production from shaft kilns decreased by 9% while that from precalciner kilns increased by 50%. This structural change within the cement industry led to a 5.4% decrease in PM emissions in just one year.

4.2.3 Coke industry

The historical trend of PM emissions from the coke industry is shown in Fig. 10c. Annual PM emissions from the coke industry have been about 1 Tg since 1995, of which $PM_{2.5}$ accounts for more than half of the mass. Two emission peaks are identified, in 1995 and 2005, which are in accordance with the historical changes in coke production.

Thirty-six percent of national coke production was from Shanxi province for the period 1990–2005. Indigenous coke ovens were dominant in Shanxi in 1990s, accounting for more than 80% of coke production (Polenske, 2006). Although the indigenous coke

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ovens were largely replaced by automatic, mechanized coke ovens after 2000, the rapid increase in coke production offset any decrease in PM emissions from the use of cleaner technologies. Our result shows that the annual emissions of $PM_{2.5}$ from the coke industry in Shanxi have been above 200 Gg since 1994, accounting for more than one-third of total emissions in this province.

4.2.4 Iron and steel industry

PM emissions from the iron and steel industry show a continuous increase over the period 1990–2005, as shown in Fig. 10d. Although EFs levelled off after 1996, production of steel increased from 130 Tg in 2000 to 360 Tg in 2005 and, as a result, PM emissions from the industry doubled in the five years, from 1.2 Tg TSP to 2.3 Tg TSP.

$PM_{>10}$ accounts for about 60% of total PM emissions by mass. Our results show that 86% of $PM_{>10}$ are fugitive dust from the processes of sinter production and pig iron production. $PM_{2.5}$ emissions are dominated by three points of emission: the beginning and end processes of the sinter machine, the cast house in iron production, and the EAF in steel production, which combined account for more than 75% of total emissions.

4.2.5 Residential sector

As the largest contributor of $PM_{2.5}$ emissions, the residential sector emitted about 4 Tg of $PM_{2.5}$ annually from 1990 to 2005, as shown in Fig. 10e. Eighty percent of $PM_{2.5}$ emissions in this sector come from the combustion of biofuel (firewood and stalks) in rural households. As fuel for cooking and heating, firewood and stalks are usually combusted in indigenous stoves that have low thermal efficiency and high emissions. Biofuel will continue to play an important role in supplying energy to rural China in the near future (Zhou, 2003). Promotion of cleaner biomass stoves could be one way to reduce PM emissions from the residential sector.

Coal boilers and stoves contribute the remaining 20% of $PM_{2.5}$ emissions from this sector. On the one hand, coal as a fuel for cooking is being gradually replaced by gas

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and electricity with the process of urbanization and with the general improvement in the quality of life across China; however, on the other hand, coal consumption for heating has shown a very rapid increased. As the result, coal consumption in the residential sector has increased by 25% over the 1990–2005 study period, and correspondingly we calculate that $PM_{2.5}$ emissions have remained more or less constant at around 0.8 Tg.

As shown in Fig. 7, the residential sector is dominant in terms of BC and OC emissions. Although BC and OC emissions from residential coal combustion decreased by 41% and 19%, respectively, from 1990 to 2005, emissions from the sector as a whole did not change greatly because the emissions from biofuel combustion are relatively constant.

4.2.6 On-road vehicles

PM emissions from on-road vehicles were much less than from stationary sources; however our findings show that they increased more than any other sector. $PM_{2.5}$, accounting for 90% of total PM emissions from on-road vehicles, increased from 27.7 Gg in 1990 to 132.15 Gg in 2005, with an average annual increase rate of 11%, as shown in Fig. 10f.

As discussed in Sect. 3.6, emission standards for vehicles began to be implemented in China in 1999. However, these standards only applied to new vehicles; thus PM emissions continued to increase for several years, as many more vehicles came onto the market than were taken off the road. PM emissions decreased a little in 2005, and this decrease or levelling off may be a feature of the near future as stricter emission standards come into effect.

4.3 Gridded emissions and data availability

Using a similar approach to that of Streets et al. (2003) and Woo et al. (2003), we mapped PM emissions onto 30 min×30 min grids using various spatial proxies. Fig-

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ure 11 shows the mapped emissions of PM_{10}, $PM_{2.5}$, BC and OC in 1990 and 2005. A significant increase of $PM_{2.5}$ and PM_{10} emissions can be seen in Northern China, especially over Shandong, Hebei, Henan and Jiangsu, mainly due to the intensive development of heavy industries. The trend of BC emissions was similar to $PM_{2.5}$ and PM_{10}, while OC emissions showed a little different trend. The most significant increase of OC emissions took place in Sichuan when biofuels were more and more used by rural residents; however, OC emissions in more developed provinces, such as Jiangsu and Zhejiang, decreased, possibly due to the gradual replacement of biofuel by cleaner fuels such as gas.

All regional and gridded emission data sets can be downloaded from our web site (http://mic.greenresource.cn/China-aerosol-trends). Users can examine emissions by province and by sector from the summary tables. Gridded data include the emissions of $PM_{2.5}$, PM_{10}, BC and OC by sector (power, industry, residential, and transportation) at 30 min×30 min resolution.

5 Discussion

5.1 Comparison with other emission estimates

5.1.1 Differences from our previous studies

Our previous work estimated the emissions of PM, BC, OC, Ca and Mg in 2001 and 2006 (Zhang et al., 2007b, 2009). By taking more technology information into account, both emission factors and activity data were updated in this study. As a result, although PM_{10} emissions were similar (16.1 Tg), higher TSP emissions (30.3 Tg vs. 25.1 Tg) and lower $PM_{2.5}$ emissions (10.9 Tg vs. 11.7 Tg) were calculated in our updated estimations for 2001, and consequently our new results show higher emissions of Ca and Mg but lower emissions of BC and OC (Fig. 12).

Different emission estimates for the industrial sector are the main reasons for the

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differences in total emissions. With updated information from various industry associations, emission factors of some industrial processes were adjusted in this study. Firstly, our previous study used unabated EFs from Europe (Klimont et al., 2002) for several industry processes, while here we have been able to update them based on operational practices in China. Updated EFs for $PM_{2.5}$ are usually smaller, but those for TSP are usually larger, compared to the European EFs; for example, average unabated EFs of $PM_{2.5}$, PM_{10} and TSP for the cement industry changed from 23.4 g/kg, 54.6 g/kg and 130.0 g/kg to 16.6 g/kg, 51.3 g/kg and 191.5 g/kg, respectively. Secondly, updated penetration rates of PM removal technologies within the industrial sector also contribute to the differences in emission estimates.

The other big difference between this study and Zhang et al. (2007b) is the estimation of BC emissions from coal combustion in the residential sector. In our previous studies, the ratio of BC to $PM_{2.5}$ was assumed to be 0.50; however, recent local tests (Chen et al., 2005, 2006, 2009; Zhi et al., 2008, 2009) indicate that this ratio could in fact be much lower. Indeed, in this study the ratio was determined to be 0.17 in 2001, following the approach described in Sect 3.5. Consequently, the estimate of BC emissions for this sub-sector was reduced by 66.5% to 127 Gg.

5.1.2 PM emissions from power sector

Emissions from power sector have been a hot topic because power plants account for more than half coal consumptions in China in recent years. The estimates in this study are 38%, 24% and 12% higher than those of Zhao et al. (2008) who estimated power sector emissions in 2005 to be 994 Gg, 1842 Gg and 2774 Gg for $PM_{2.5}$, PM_{10} and TSP, respectively. The latest database of EFs for China's power plants incorporates the results from recent test results, and includes an analysis of the sources of uncertainties in determining the EFs (Zhao et al., 2010). The database assumed lower removal efficiency of ESP (92% for $PM_{2.5}$, 97% for $PM_{2.5-10}$ and 99.5% for $PM_{>10}$) and resulted in higher final EFs. Consequently, the estimates of $PM_{2.5}$, PM_{10} and TSP emissions would be 11%, 25% and 20% higher than this study if the same activity data were used.

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5.1.3 TSP emissions

The estimation of PM emissions is little studied in China. China's emission statistics for national TSP emissions are based on calculations using a bottom-up approach, while information about PM_{10} and $PM_{2.5}$ emissions is unavailable. TSP emissions from our estimates as well as the government's statistical data are shown in Fig. 13. The statistical data are systematically lower than our estimates because two important emission sources (small industries and the rural residential sector) are not taken into account in the government data. Statistical TSP emissions changed significantly in 1993–1994 and 1996–1997; the main reason for this is the change in statistical approach over this time period. The two sets of data show similar trends in the late 1990s, when China's energy consumption decreased. However, according to our estimates, emissions increased after 2000, while the statistical data suggest that annual emissions remain at around 20 Tg. Our estimates may be more accurate because most sectors grew rapidly during this period of time, as discussed in previous sections. Our previous studies on CO (Streets et al., 2006b) and NO_x (Zhang et al., 2007a) show similar increases in emissions.

5.1.4 BC and OC emissions

BC and OC emissions of this study were compared with the previous studies of Bond et al. (2004), Cao et al. (2006), Ohara et al. (2007), Streets et al. (2003) and Zhang et al. (2009) in Fig. 14. All these studies show that the residential sector is the dominant source of BC and OC. Our estimation of BC emissions from the residential sector is 30% lower than that of others because a much lower EF for briquette combustion was incorporated in the current study. As with our previous studies (Bond et al., 2004, and Zhang et al., 2009), this study estimates higher BC emissions from industry, because we consider small coke plants and brick plants to be potentially important sources, although there are large uncertainties in the estimates. Our estimation of OC emissions is close to that of Zhang et al. (2009) and Streets et al. (2003); with any differences

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mainly being due to the different parameters used to calculate emissions from biofuel.

Ohara et al. (2007) estimated the emission trends of BC and OC with activity data for 1995, 2000 and 2003, assuming constant EFs. Our study does not indicate the same trends, with the differences mainly being attributable to different sources of activity data, especially the biofuel used within the residential sector. Biofuel usage in this study is 14%, 13%, and 41% higher than that of Ohara et al. (2007) for 1995, 2000 and 2003, respectively. This indicates that in addition to EFs, uncertainty about biofuel consumption data could be another important source of error in the estimation of BC and OC emissions.

A comparison by Carmichael et al. (2003) of model calculations using the emission inventories of Streets et al. (2003) and using TRACE-P measurement data led to the conclusion that Streets et al.'s estimates of BC emissions are qualitatively correct. However, it is likely that BC emissions over Southeast China were overestimated while those in northeast China were underestimated (Hakami et al., 2005). Since our estimation is similar to Streets et al. (2003)'s results, it could also be true that there are some uncertainties in the spatial distribution of BC emissions.

5.2 Uncertainties

A detailed uncertainty analysis was conducted by combining uncertainties of both EFs and activity levels, following the approach described by Streets et al. (2003). As listed in Table 10, the uncertainties measured as 95% confidence intervals of PM_{10}, $PM_{2.5}$, BC and OC are similar to the results of Zhang et al. (2009). For most sectors, uncertainties of emissions in 2005 are lower than those in 1990 because for the later date we are more confident about both the penetration of PM control technologies and the accuracy of activity level data. Industry is the only exception, and what increases the level of uncertainty is the fact that the contribution from industries whose emissions are less easily quantified (e.g. lime and brick production) is getting larger while emissions from the cement industry are significantly reduced.

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6 Conclusions

We use a technology-based methodology to estimate historical PM emissions in China in recent years. With this methodology, we derive a 15-year trend of PM emission factors in China from 1990 to 2005, taking into account the change in technology structure within sectors and improvements in emission controls driven by emission standards. Our results show that emission factors of $PM_{2.5}$ and TSP from several industry sectors decreased by 7% to 69% and 18% to 80% in China during the 15 years, respectively.

Emissions of TSP, PM_{10}, $PM_{2.5}$, BC, OC, Ca and Mg during the 15-year period are estimated. The trends of emissions of PM are similar with which of energy consumption in China during 1990–2005, that is, increased in the first six years of 1990s and decreased until 2000, then increased again in the following years. Emissions of TSP reached a historical high (35.5 Tg) in 1996, while emissions of PM_{10} and $PM_{2.5}$ reached historical highs in 2005 (18.5 Tg PM_{10} and 12.7 Tg $PM_{2.5}$). With significant increase of BC and OC emissions presented during 2000–2005, BC and OC emissions reached historical high in 2005 (15.1 Tg and 3.19 Tg, respectively). The cement industry and biofuel combustion in the residential sector were consistently the dominant sources of $PM_{2.5}$ emissions in China, accounting for 53% to 62% of emissions from 1990 to 2005. The non-metallic mineral production industry, including the cement, lime and brick industries, accounted for 54% to 63% of national TSP emissions.

The careful consideration of technology details significantly improves the accuracy of emission inventories; however there still remain large uncertainties in the estimation of primary aerosol emissions in China. More accurate and detailed activity information coupled with the measurement of emission factors from local tests are essential to further improve the quality of emission estimates, this especially being so for the brick and coke industries, as well as for coal-burning stoves and biofuel usage within the residential sector.

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Table 1. Source of activity data for main PM emitting sectors in China.

Emitting Sector	Data Sources
Power plants	China State Electricity Regulatory Commission, unpublished data
Industry boilers	China Mechanical Industry Yearbook
Residential combustion, rural cooking	China Statistical Yearbook
Residential combustion, rural heating	Chinese provincial statistical yearbooks
Coke production	National Bureau of Statistics, unpublished data
Cement production	Chinese Cement Association, unpublished data
Iron & Steel production	China Iron and Steel Statistics
On-road vehicles	Using a modeling approach documented in He et al. (2005)

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Table 2. Unabated EFs for PM from stationary sources.

Sector	Fuel/product	Technology	$PM_{2.5}$	$PM_{2.5-10}$	$PM_{>10}$	TSP	Reference
Stationary combustion (k/kg fuel)							
Power plants	Coal	Pulverized	12.00	34.00	154.00	200.00	Zhang et al., 2006
	Coal	Grate furnace	5.25	8.63	23.63	37.50	Zhang et al., 2006
	Fuel oil		0.62	0.23	0.35	1.20	USEPA, 1995
Industry	Coal	Circulating fluidized bed	5.40	22.68	79.92	108.00	Zhang et al., 2006
	Coal	Grate furnace	1.89	3.51	21.60	27.00	Zhang et al., 2006
	Fuel oil		0.67	0.36	0.17	1.20	USEPA, 1995
Residential	Coal	Grate furnace	1.89	3.51	21.60	27.00	Zhang et al., 2006
	Coal	Hand-feed grate furnace	2.00	1.50	1.50	5.00	Zhang et al., 2000
	Coal	Stove	6.86	1.96	0.98	9.80	Zhang et al., 2000
	Fuel oil		0.28	0.47	0.46	1.20	USEPA, 1995
	Firewood	Stove	5.58	0.18	0.24	6.00	Zhang et al., 2000
	Stalks	Stove	6.98	0.23	0.30	7.50	Zhang et al., 2000
All	Diesel oil		0.50	0.00	0.00	0.50	USEPA, 1995
	Kerosene		0.90	0.00	0.00	0.90	USEPA, 1995
	Gas		0.17	0.00	0.00	0.17	USEPA, 1995
Industry process (g/kg product)*							
Metallurgical	Sinter		2.62	3.43	34.25	40.30	SEPA, 1996a
	Pig iron		6.00	3.65	54.55	64.20	SEPA, 1996a
	Steel	Open hearth furnace	13.80	5.30	3.90	23.00	SEPA, 1996a
		Basic oxygen furnace	10.45	4.18	6.27	20.90	Klimont et al., 2002
		Electric arc furnace	6.02	2.10	5.88	14.00	SEPA, 1996a
	Casting		8.48	3.35	3.93	15.76	SEPA, 1996a
	Aluminum	Primary	18.28	8.23	19.20	45.71	SEPA, 1996a
		Secondary	5.20	1.78	4.93	11.91	SEPA, 1996a
	Alumina		297.13	99.04	1254.53	1650.70	SEPA, 1996a
	Other non-ferrous metal		246.00	30.00	24.00	300.00	SEPA, 1996a
Mineral products	Cement	Pre-calcination kiln	28.46	48.97	168.57	246.00	Lei et al., 2010
		Other rotary kiln	23.51	44.97	170.71	239.20	Lei et al, 2010
		Shaft kiln	12.86	29.77	128.37	171.00	Lei et al, 2010
	Glass	Float glass	7.92	0.35	0.43	8.70	SEPA, 1996a
		Sheet glass	10.69	0.47	0.58	11.74	SEPA, 1996a
		Other glass	2.94	0.13	0.16	3.23	SEPA, 1996a
	Bricks		0.27	0.44	2.99	3.70	SEPA, 1996a
	Lime		1.40	10.60	88.00	100.00	Klimont et al., 2002
Chemical	Coke	Mechanical oven	5.22	3.57	4.22	13.00	SEPA, 1996a
		Indigenous oven	5.22	3.57	4.22	13.00	SEPA, 1996a
	Refined oil		0.10	0.02	0.00	0.12	Klimont et al., 2002
	Fertilizer		1.86	0.26	0.24	2.36	SEPA, 1996a
	Carbon black		1.44	0.16	0.18	1.78	Klimont et al., 2002

* Size distribution of PM emissions from industrial processes is based on Klimont et al., 2002.

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Table 3. PM EFs for on-road vehicles under different emission standards (g/kg fuel).

	LDGV	LDDV	LDGT1	LDGT2	LDDT	HDGV	HDDV	MC
Uncontroll	0.25	5.12	0.25	0.40	5.50	0.40	3.00	4.00
EUROI	0.15	2.01	0.16	0.25	2.20	0.25	1.60	2.80
EUROII	0.08	1.30	0.07	0.10	1.40	0.10	0.70	1.20

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Table 4. Emission standards for industry and on-road vehicles before 2005.

Industry sector/vehicle type	Standard code	Year published/revised
Power plants	GB13223	1991, 1996, 2003
Cement plants	GB4915	1985, 1996, 2004
Coking oven	GB16171	1996
Other industry*	GB9078	1988, 1996
LDGV	GB18352	1999, 2001
LDGT, HDGV	GB14762	2002
LDDV, HDDV	GB17691	1999, 2001
MC	GB14622	2000, 2002

* Emission standards for some individual industries were replaced by this standard.

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Table 5. Removal efficiency of different PM control technologies.

	Control technology	$PM_{>10}$	$PM_{2.5-10}$	$PM_{2.5}$
End-of-pipe	FAB	99.9	99.5	99
	ESP	99.5	98	93
	WET	99	90	50
	CYC	90	70	10
Fugitive	Normal practice	20	15	10
	Good practice	70	50	30

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Table 6. Mass ratio of BC and OC to $PM_{2.5}$ from different emission sources.

	BC	OC	Reference
Sinter	1	5	Kupiainen and Klimont, 2004
Pig Iron	10	2	Kupiainen and Klimont, 2004
Bof	0	20	Kupiainen and Klimont, 2004
Eaf	0	2	Kupiainen and Klimont, 2004
Casting	0	3	Kupiainen and Klimont, 2004
Cement	0.6	1	Kupiainen and Klimont, 2004
Lime	2	1	Kupiainen and Klimont, 2004
Brick	40	35	Kupiainen and Klimont, 2004
Coking	30	35	Kupiainen and Klimont, 2004
Diesel Vehicle	57	18	Bond et al., 2004
Gasoline Vehicle	29	31	Bond et al., 2004
Power Plants	0.6	0	Bond et al., 2004
Grate Furnace	20	4	Bond et al., 2004
Stove/Coal*	14.6–22.8	43.1–48.0	Chen et al., 2009
Stove/Firewood	20	80	Bond et al., 2004
Stove/Stalks	15	57	Bond et al., 2004
Diesel (Boiler)	30	9	Bond et al., 2004
Kerosene	13	10	Bond et al., 2004
Fuel Oil	8	3	Bond et al., 2004
Gas	10	30	Bond et al., 2004
Motorcycle	5	75	Bond et al., 2004
Off-Road Mobile	57	18	Bond et al., 2004

* Average mass ratio of BC and OC to $PM_{2.5}$ from coal stove dropped while share of briquettes in coal consumption increased.

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Table 7. Mass ratio of Ca and Mg to TSP from different emission sources.

	Ca	Mg
Power plants	4.3	1.0
Industry boilers	4.2	1.0
Domestic boilers	4.4	1.1
Domestic stoves	5.0	1.0
Coking	3.6	0.8
Iron and steel	7.1	3.5
Cement	39.9	1.0
Lime	39.9	1.0
Brick	4.2	1.0
Other industry processes	4.9	1.1

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Table 8. Starting date of implementation of China's Stage I and Stage II emission standards for vehicles.

Region	Stage I				Stage II	
	LDGV	LDDV, HDDV	LDGT, HDGV	MC	Vehicle	MC
Beijing	Jan 1999	Jan 2000	Jan 2000	Jan 2001	Jan 2003	Jan 2004
Shanghai	Jul 1999	Oct 2001	Jul 2003	Jul 2003	Mar 2003	Jan 2005
Rest of China	Jul 2000	Oct 2001	Jul 2003	Jul 2003	Sep 2004	Jan 2005

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Table 9. Sector breakdown emissions of $PM_{2.5}$, PM_{10}, TSP, BC and OC in 1990, 1995, 2000 and 2005. (Tg).

	$PM_{2.5}$				PM_{10}				TSP				BC				OC			
	1990	1995	2000	2005	1990	1995	2000	2005	1990	1995	2000	2005	1990	1995	2000	2005	1990	1995	2000	2005
Power	1.09	1.43	1.12	1.37	1.76	2.29	1.81	2.28	2.22	2.87	2.32	3.09	0.01	0.01	0.01	0.01	0.00	0.00	0.00	0.00
Industry boiler	0.45	0.52	0.48	0.86	0.86	0.96	0.85	1.40	2.37	2.45	1.93	2.37	0.08	0.09	0.08	0.15	0.00	0.00	0.00	0.00
Residential coal	0.83	0.83	0.76	0.79	1.08	1.08	1.00	1.07	1.27	1.28	1.21	1.40	0.19	0.16	0.13	0.11	0.39	0.38	0.33	0.32
Residential biofuel	3.49	3.15	2.80	3.60	3.60	3.25	2.89	3.72	3.75	3.38	3.01	3.87	0.57	0.52	0.46	0.59	2.22	2.02	1.77	2.29
Iron and steel	0.29	0.42	0.38	0.67	0.35	0.54	0.49	0.88	0.72	1.16	1.19	2.33	0.00	0.00	0.00	0.01	0.01	0.01	0.02	0.03
Cement	2.23	4.21	3.68	3.48	3.79	6.97	5.90	5.47	5.86	10.28	8.25	7.33	0.01	0.03	0.02	0.02	0.02	0.04	0.04	0.03
Brick	0.31	0.53	0.51	0.54	0.70	1.19	1.14	1.20	3.06	5.18	4.95	5.25	0.13	0.21	0.20	0.21	0.11	0.19	0.18	0.19
Lime	0.14	0.18	0.17	0.20	0.75	1.00	0.94	1.07	4.73	6.27	5.88	6.57	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Coke	0.23	0.50	0.41	0.72	0.33	0.76	0.59	0.96	0.43	1.05	0.77	1.16	0.07	0.15	0.12	0.22	0.08	0.18	0.14	0.25
Other process	0.10	0.16	0.22	0.36	0.11	0.18	0.25	0.40	0.12	0.20	0.28	0.47	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00
Mobile	0.12	0.17	0.28	0.37	0.15	0.20	0.30	0.38	0.34	0.37	0.37	0.42	0.06	0.09	0.14	0.19	0.02	0.04	0.07	0.09
Total	9.28	12.11	10.79	12.95	13.50	18.43	16.14	18.83	24.86	34.49	30.16	34.26	1.13	1.27	1.18	1.51	2.87	2.86	2.54	3.19

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Table 10. Uncertainty in emissions estimates of TSP, PM_{10}, $PM_{2.5}$, BC, and OC in 1990 and 2005 (±95% confidence intervals), numbers shown in table as percentage.

	1990					2005				
	TSP	PM_{10}	$PM_{2.5}$	BC	OC	TSP	PM_{10}	$PM_{2.5}$	BC	OC
Power	98	98	98	92	44	60	60	60	58	44
Industry	146	119	111	304	346	156	129	124	305	355
Residential	353	357	365	323	377	341	353	363	321	378
Transportation	148	105	105	112	93	80	82	82	91	70
All sectors	124	138	175	244	343	127	121	143	204	312
Zhang et al. 2009*							132	130	208	258

* Uncertainties in emission estimates for 2006.

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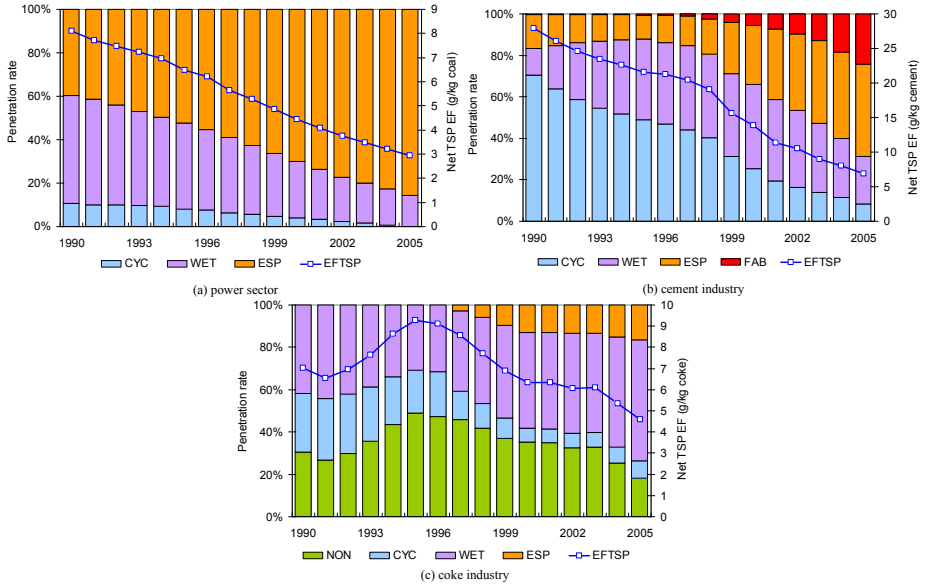

Fig. 1. As high efficient PM control technologies were gradually promoted during 1990–2005, EFs of TSP from **(a)** power sector, **(b)** cement industry, and **(c)** coke industry decreased. Bars represent the penetration rate of PM control technologies within the industries, and line represents the net emission factor of TSP.

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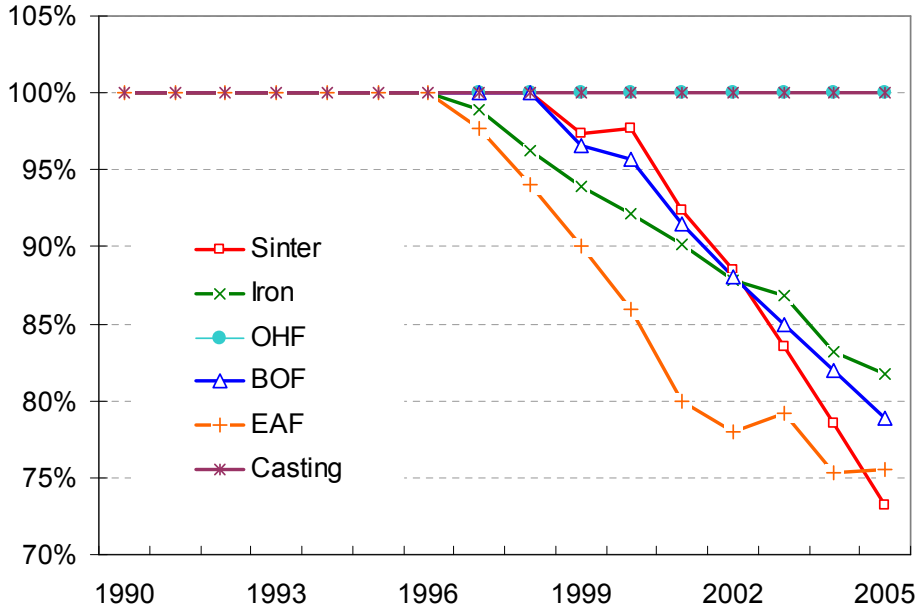

Fig. 2. Trends of net EF of TSP from processes/technologies in iron and steel industry. All data are normalized to the year 1996.

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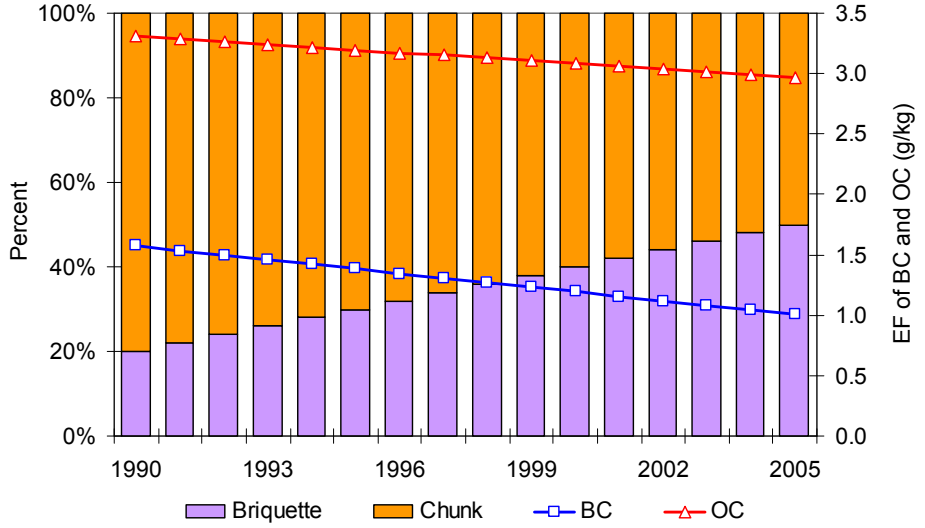

Fig. 3. Emission factors of BC (blue line) and OC (red line) drops as share of briquettes in residential stoves increased.

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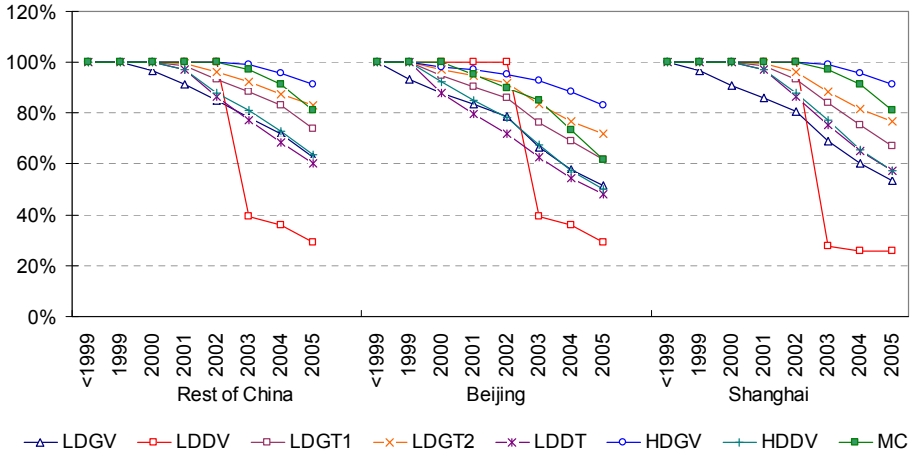

Fig. 4. Trends of net EF of $PM_{2.5}$ from on-road vehicles. All data are normalized to the year 1999.

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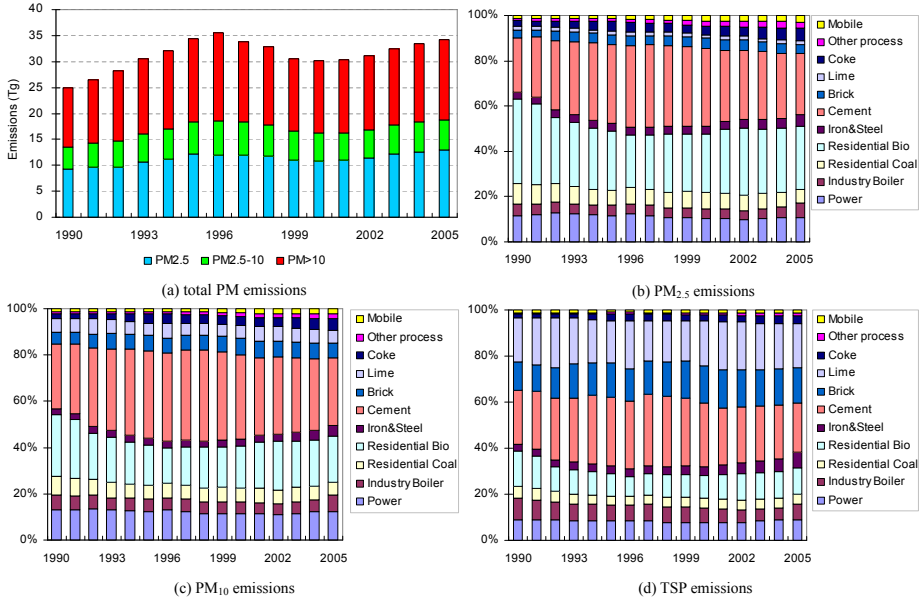

Fig. 5. (a) PM emissions from 1990 to 2005 and the breakdown of emissions of **(b)** $PM_{2.5}$, **(c)** PM_{10} and **(d)** TSP by different sectors.

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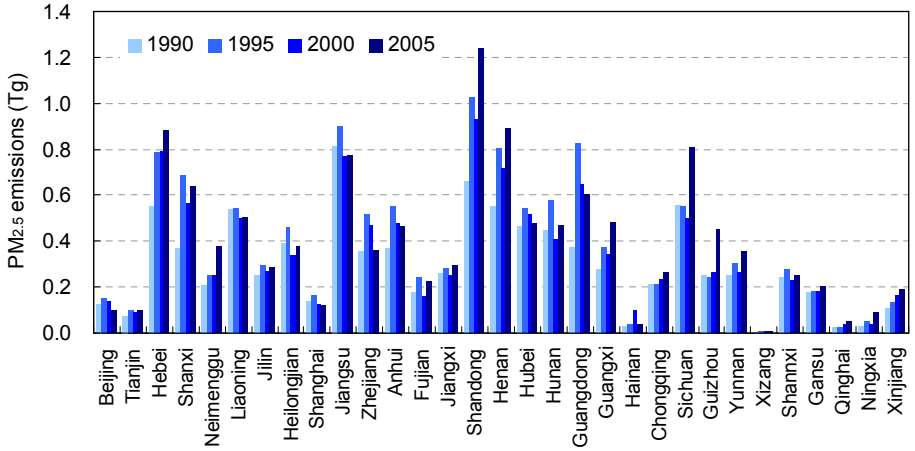

Fig. 6. Emissions of $PM_{2.5}$ by province for 1990, 1995, 2000 and 2005.

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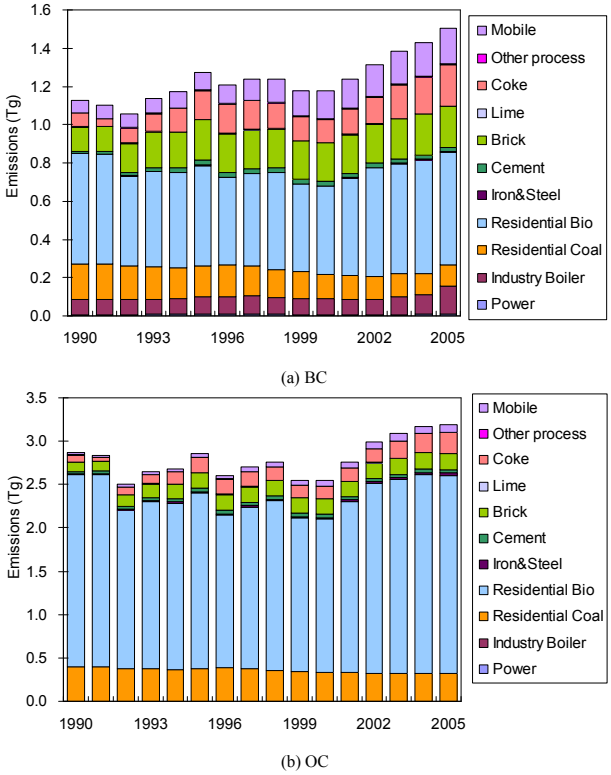

Fig. 7. Emissions of **(a)** BC and **(b)** OC from 1990 to 2005.

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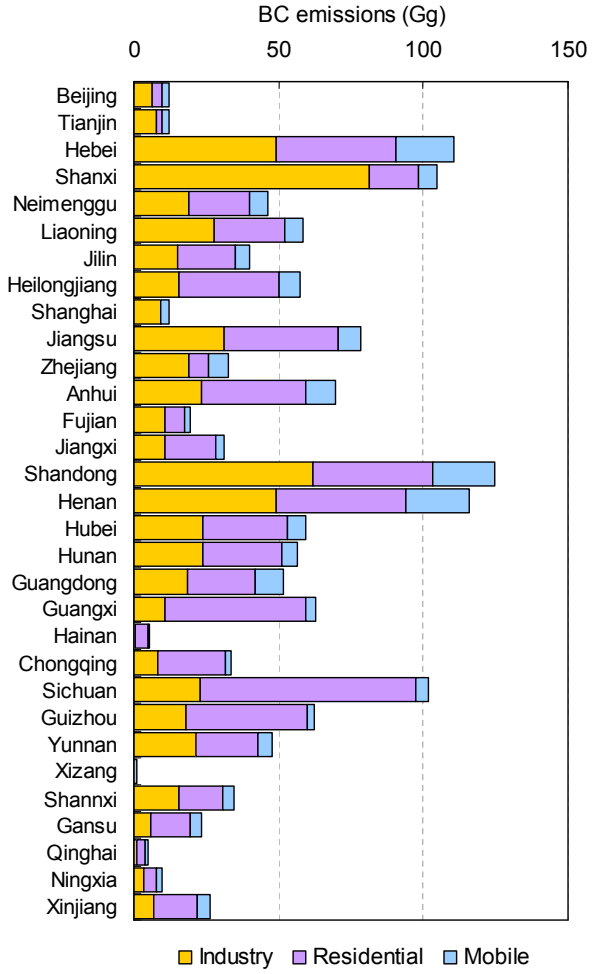

Fig. 8. Provincial BC emissions in 2005.

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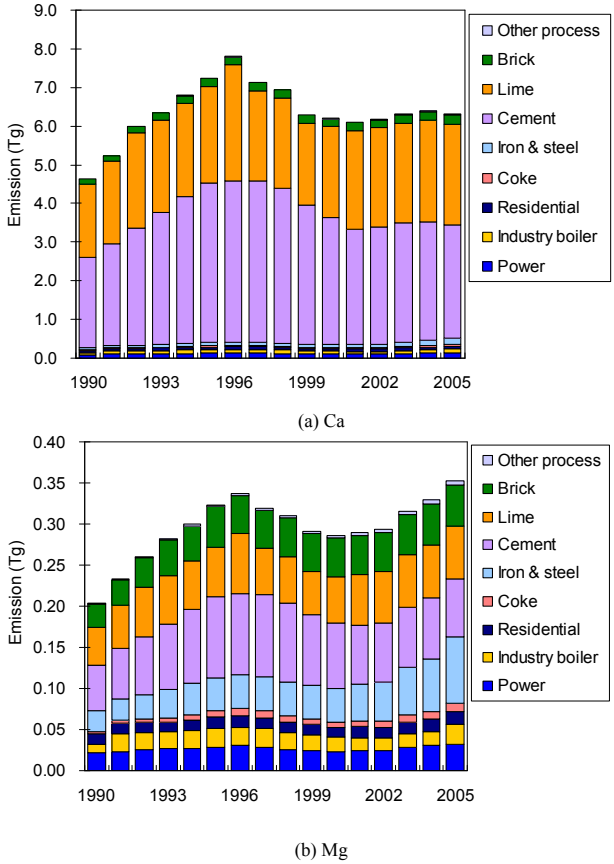

Fig. 9. Emissions of **(a)** Ca and **(b)** Mg from 1990 to 2005.

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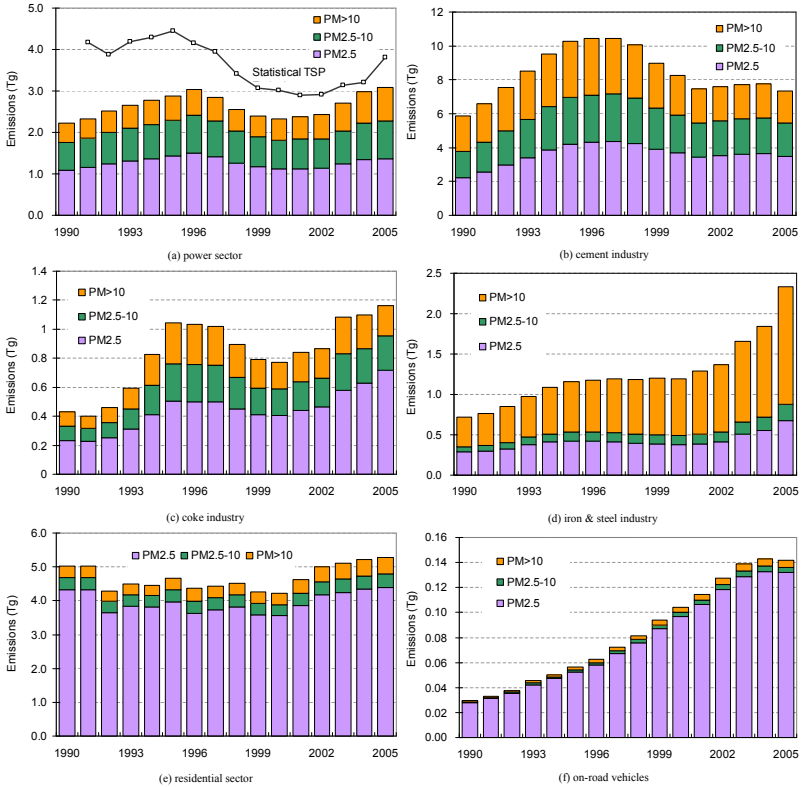

Fig. 10. PM emissions during 1990–2005 from **(a)** power sector, **(b)** cement industry, **(c)** coke industry, **(d)** iron and steel industry, **(e)** residential sector and **(f)** on-road vehicles.

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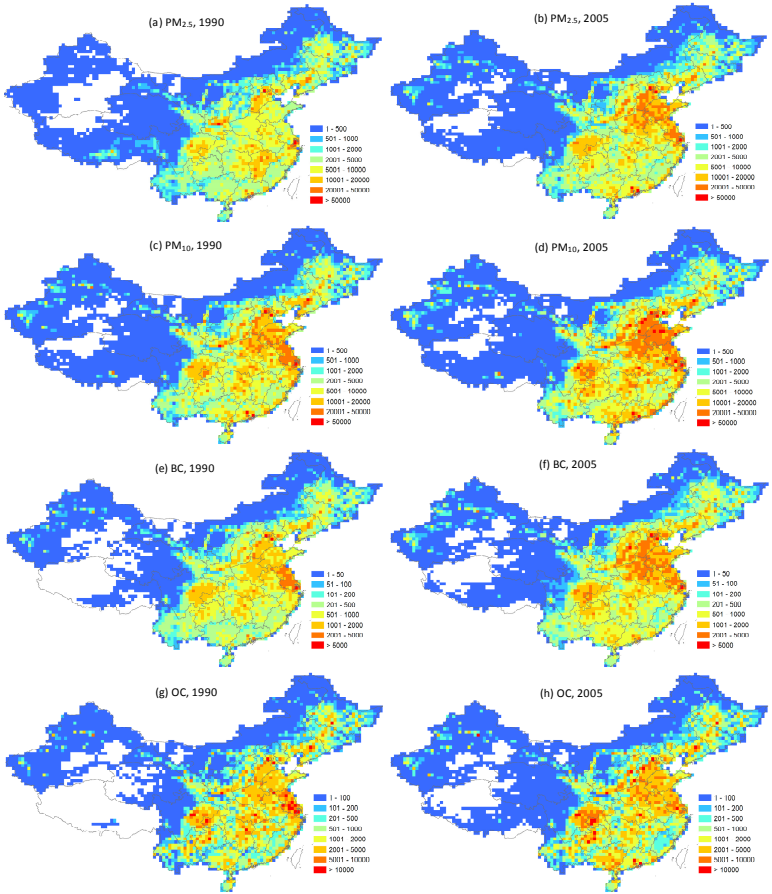

Fig. 11. Emissions of $PM_{2.5}$, PM_{10}, BC and OC in 1990 and 2005 by $30' \times 30'$ grids. Unit: Mg/grid cell.

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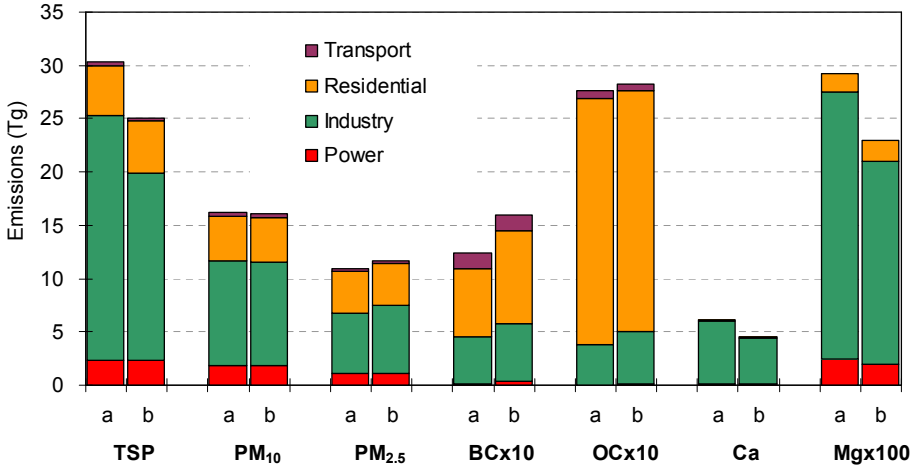

Fig. 12. Comparison of emission estimates for 2001 in **(a)** this study and **(b)** our previous results from Zhang et al., 2007b.

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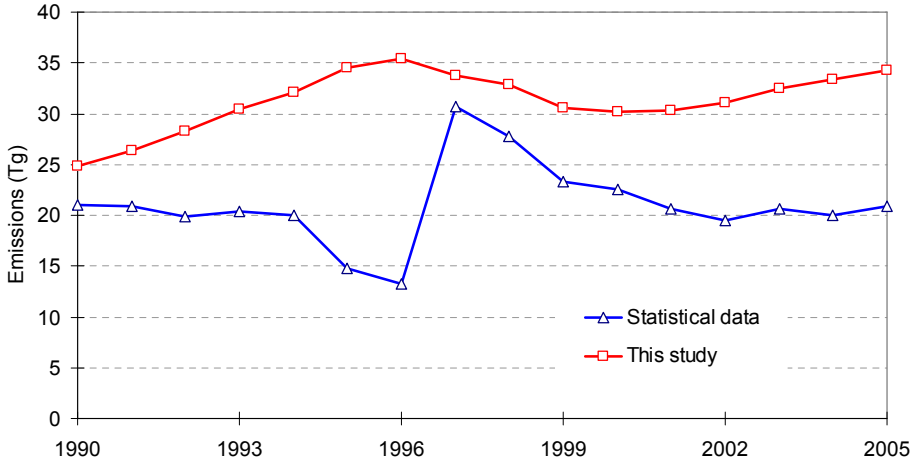

Fig. 13. Comparison of TSP emissions between estimates by this study and China's government statistical data.

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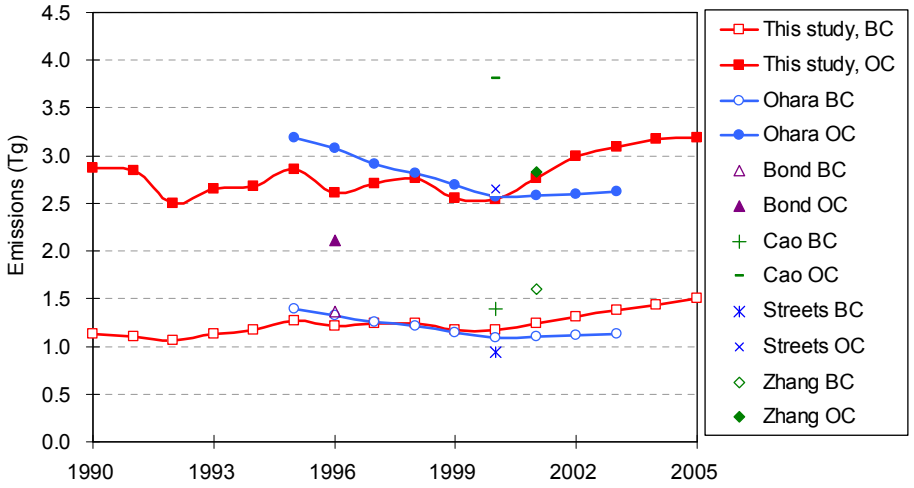

Fig. 14. Comparison of BC and OC emission estimation among recent studies: Bond et al. 2004; Cao et al., 2006; Ohara et al., 2007; Streets et al., 2003; Zhang et al., 2009.

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