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Major components of China's anthropogenic primary particulate emissions

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Abstract
This paper presents the first comprehensive estimates of particulate emissions in China by size distribution and major components. Using a technology-based emission inventory approach, we are able to classify particulate emissions into three size ranges, TSP, PM_{10} and $PM_{2.5}$, and identify the contributions of black carbon (BC), organic carbon (OC), Ca and Mg. Total particulate emissions are estimated to be 27.4 Tg for the year 2001, of which 17.8 Tg are PM_{10} and 12.7 Tg are $PM_{2.5}$. Industrial processes are the major sources of particles over all three size ranges, but residential biofuel use and transportation sources become increasingly important for PM_{10} and $PM_{2.5}$. The industrialized coastal provinces, such as Shandong, Jiangsu and Hebei, are the major sources of particulate emissions. The industrialized and developing regions show different characteristic emission ratios of $PM_{2.5}$/TSP, (BC + OC)/$PM_{2.5}$ and (Ca + Mg)/TSP. In the future, we can expect significant reductions in primary particulate emissions and major changes in the patterns of size and species.

Keywords: China, particulate matter, emissions

1. Introduction

Particulate matter (PM) impacts local, regional and global environments in many ways. Fine PM with diameters less than or equal to 2.5 μm ($PM_{2.5}$) has negative effects on human health (Pope *et al* 1995). Particles are able to travel across national boundaries and impact regional air quality and hemispheric backgrounds (Heald *et al* 2006). Carbonaceous particles have also been linked with global and regional climate change (Andreae 2001, Jacobson 2001, 2002, Menon *et al* 2002, Hansen and Nazarenko 2004). In China, PM_{10} is the dominant air pollutant in cities, and PM_{10} concentrations often exceed China's national air quality standard in major urban areas (He *et al* 2002).

Although there are several inventories that address China's carbonaceous aerosol emissions as national inventories or as part of regional/global inventories (Streets *et al* 2001, 2003a, Bond *et al* 2004, Cao *et al* 2006), a detailed approach that considers the main components of aerosols and their size distribution with a consistent methodology is missing. Recently, we revisited China's inventory and updated CO and NO_x emissions with an improved methodology (Streets *et al* 2006, Zhang *et al* 2007). The purpose of this paper is to present China's emissions of total suspended particulates (TSP), PM_{10}, $PM_{2.5}$, BC, OC, Ca and Mg in a consistent way, based on our improved methodology.

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2. Methodology

We have implemented an improved, technology-based methodology to estimate China's anthropogenic primary aerosol emissions. Emissions are calculated from the combination of activity rate, technology distributions, emission factors in raw gas and the penetration of emission control equipment. The general approach used in this work is similar to that of Klimont *et al* (2002); that is, emissions are estimated for three diameter ranges: large particles ($>PM_{10}$), coarse fraction ($PM_{2.5-10}$) and fine fraction ($PM_{2.5}$). Thereby, PM_{10} is calculated as the sum of fine and coarse fractions and TSP as the sum of fine, coarse and $>PM_{10}$ fractions. The size-fractioned PM emissions are calculated by the following equation:

$$E_{i,y} = \sum_{j,k,m} E_{i,j,k,m,y} = \sum_{j,k,m} A_{i,j,k,m} \mathrm{ef}_{i,j,k,m,y}. \quad (1)$$

For a given j, k and m, the final emission factor of diameter range y is estimated by the following equation:

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

where i represents province; y represents diameter range of PM; j represents the sector; k represents fuel or product type; m represents the combustion or production technology; n represents PM control technology or equipment; $E_{i,y}$ means the emission amount of PM in diameter y in province i; A means activity level, such as fuel consumption, production, etc; ef means emission factor; $\mathrm{EF}_{\mathrm{TSP}}$ means raw emission factor of TSP; F_y means mass proportion of PM in diameter y out of total PM; C_n means the percentage of control technology n; $\eta_{n,y}$ means the removal efficiency of control technology n for PM in diameter y.

In addition to total aerosol emissions, we also estimate the emissions of several key chemical components in aerosols: BC, OC, Ca and Mg. Emission factors of BC and OC are determined by the mass ratio of BC and OC to $PM_{2.5}$ emission factors, which is similar to the approach described in Bond *et al* (2004) and Kupiainen and Klimont (2007); similarly, emission factors of Ca and Mg are determined by their fraction in TSP emissions, following Zhu *et al* (2004).

We classify emission sources into four groups: stationary combustion, industrial process, mobile sources and biomass open burning. The stationary combustion sources involve three sectors (power plants, industry and residential) and eight types of fuel (coal, diesel, kerosene, fuel oil, liquid petroleum gas, coal gas, natural gas and biomass). The industrial process sources include 11 products in metallurgical industries, mineral products industries and chemical industries. For each industrial process, end-of-pipe and fugitive PM emission are estimated separately. The mobile emission sources include seven types of on-road mobile sources: light-duty gasoline vehicles, light-duty gasoline trucks, mid-duty gasoline trucks, light-duty diesel vehicles, heavy-duty gasoline vehicles, heavy-duty diesel vehicles and motorcycles; and six types of off-road mobile sources: rural vehicles, tractors, building equipment, farm equipment, locomotives and vessels. Biomass burning emissions include forest fires, grassland fires and the open burning of crop residues.

According to equations (1) and (2), the necessary data for emissions calculations include: (a) fuel consumption and industrial production; (b) distribution of emission control technologies; (c) PM emission factor for each type of emission source; (d) share of PM emissions in each diameter range out of total PM emissions; (e) mass ratio of emissions of specific components (BC, OC, Ca and Mg) in total PM emissions; and (f) removal efficiency of each PM control technology in each diameter range. The activity data (a) and (b) and emission factors ((c)–(f)) were developed from a wide variety of sources.

The general approach used in this work to develop detailed activity data has been described in Streets *et al* (2006) and Zhang *et al* (2007), in connection with the development of improved CO and NO_x emission inventories for China. The activity data at sector level rely mainly on statistics published by different government agencies of China. Fuel consumption by sector and by province is derived from the China Energy Statistical Yearbook 2001 (NBS 2004). Industrial production by sector and by province and the vehicle population data are derived from a variety of government statistics (ACT 2002, AISIC 2002, NBS 2002a, 2002b). Technology distributions within each sector are obtained from a wide variety of Chinese technology reports and other publications. Vehicle population data and fuel consumption by vehicle type are taken from He *et al* (2005), using a transportation fuel consumption model. Table 1 summarizes the main activity data used in this study. Readers are referred to Streets *et al* (2006) and Zhang *et al* (2007) for further details of sources of activity data. We have collected appropriate emission factors based on available measurements in China or used estimations based on the actual technology level and practice, e.g. Zhang *et al* (2000), Chen *et al* (2005, 2006) and Yi *et al* (2006). SEPA (1996) was widely consulted for TSP emission factors. In some cases, where local information is lacking, we use emission factors for similar activities from the US AP-42 database (USEPA 1996) and the RAINS-PM model (Klimont *et al* 2002).

3. Results and discussion

3.1. Size distribution

Table 2 summarizes China's anthropogenic primary PM emissions in 2001 by size and by sector. We estimate that anthropogenic TSP emissions in China in 2001 were 27.4 Tg, and PM_{10} emissions were 17.8 Tg, accounting for 65% of TSP emissions. $PM_{2.5}$ emissions, which are the most harmful part of the aerosols, comprise a large proportion of primary aerosol emissions. National $PM_{2.5}$ emissions were 12.7 Tg in 2001, accounting for 45% of TSP emissions and 70% of PM_{10} emissions. Industrial processes are the largest contributor to TSP emissions, with a contribution of 58% to total TSP emissions, followed by residential biofuel and power plants, which contributed 12% and 9% of total TSP emissions, respectively. For $PM_{2.5}$, industrial processes are still the largest contributor, but their contribution drops to 46%, while the contribution of residential biofuel emissions to national total $PM_{2.5}$ emissions increases to 24%. The sector differences in

Table 1. Activity levels for PM emission estimates in China in 2001.

Sector	Fuel/product/vehicle type	Activity level	Reference
Power	Coal	675 Tg coal burned	NBS (2004)
Industry	Coal	187 Tg coal burned[a]	NBS (2004)
Industry	Coke	131 Tg coke produced	NBS (2002b)
Industry	Sinter	191 Tg sinter produced	AISIC (2002)
Industry	Pig iron	145 Tg pig iron produced	AISIC (2002)
Industry	Steel	153 Tg steel produced	AISIC (2002)
Industry	Cement	661 Tg cement produced	NBS (2002a)
Industry	Brick	560 billion bricks produced	NBS (2002b), Zhou *et al* (2003)
Industry	Lime	117 Tg lime produced	NBS (2002b), Zhou *et al* (2003)
Residential	Coal	170 Tg coal burned	NBS (2004)
Residential	Wood	171 Tg wood burned	NBS (2004)
Residential	Crop residue	305 Tg crop residue burned	NBS (2004)
Transportation	Passenger car	5.9 million vehicles	ACT (2002), He *et al* (2005)
Transportation	Light-duty gasoline truck	4.9 million vehicles	ACT (2002), He *et al* (2005)
Transportation	Mid-duty gasoline truck	0.6 million vehicles	ACT (2002). He *et al* (2005)
Transportation	Light-duty diesel truck	1.6 million vehicles	ACT (2002), He *et al* (2005)
Transportation	Heavy-duty gasoline truck	1.5 million vehicles	ACT (2002), He *et al* (2005)
Transportation	Heavy-duty diesel truck	3.5 million vehicles	ACT (2002), He *et al* (2005)
Transportation	Motorcycle	43.4 million vehicles	ACT (2002), He *et al* (2005)

[a] Coal consumption in cement kilns, brick kilns and lime kilns was subtracted from total industry coal combustion to avoid double counting.

Table 2. Primary PM emissions by sector in China, 2001 (unit: Tg).

	Power plants	Industrial combustion	Industrial processes	Residential fossil fuel	Residential biofuel	Transportation	Biomass burning	Total
TSP	2.39	1.65	15.88	1.58	3.31	0.30	2.24	27.35
PM_{10}	1.85	0.87	8.77	1.08	3.18	0.29	1.74	17.79
$PM_{2.5}$	1.14	0.53	5.87	0.77	3.08	0.28	1.02	12.69
$PM_{2.5}$/TSP	0.48	0.32	0.37	0.49	0.93	0.94	0.45	0.46
BC	0.04	0.09	0.46	0.36	0.51	0.14	0.11	1.71
OC	0.01	0.03	0.47	0.28	1.98	0.07	0.75	3.58
(BC + OC)/$PM_{2.5}$	0.04	0.22	0.16	0.82	0.81	0.75	0.85	0.42
Ca	0.10	0.07	4.27	0.08	N/A	N/A	N/A	4.52
Mg	0.02	0.02	0.17	0.02	N/A	N/A	N/A	0.23
(Ca + Mg)/TSP	0.05	0.05	0.28	0.06	N/A	N/A	N/A	0.17

coarse and fine particle distribution partly explain the high emission ratio of $PM_{2.5}$/TSP in China: significant emissions come from inefficient combustion of coal and biofuels in the residential sector, where most of the emitted aerosols are fine particles. Another factor contributing to the high $PM_{2.5}$/TSP emission ratio is the low penetration of effective PM control technology in China: many small power plants and industrial plants still use wet particle scrubbers and cyclones, which have high removal efficiency for the coarse components of PM, but low efficiency for smaller particles.

China's power plants emitted 2.39 Tg TSP in 2001, of which 1.14 Tg were $PM_{2.5}$. Electrostatic precipitators (ESP) have been widely used in power plants in China, but there were still 20% of power plants only equipped with low-efficiency PM control technologies in 2001. Those 20% of power plants emitted 62% of the total $PM_{2.5}$ emissions from power plants. Therefore, improving the PM control technologies in these plants could effectively reduce the $PM_{2.5}$ emissions in the power sector. Industrial combustion devices emitted 1.65 Tg TSP and 0.53 Tg $PM_{2.5}$ in 2001. Industrial boilers in China are installed with a variety of different types of PM removal equipment, but most are wet scrubbers and cyclones, which have low PM removal efficiencies, especially for fine particles. Residential combustion is another important source of PM emissions, especially for fine particles. In 2001, $PM_{2.5}$ emissions from residential combustion were 3.85 Tg, with 80% of them from biofuel combustion. Residential combustion is also identified as a major contributor of carbonaceous aerosol emissions in China (Streets *et al* 2001, Streets and Aunan 2005), which will be discussed later.

It is surprising that PM emissions from industrial processes are higher than the sum of emissions from all other stationary combustion sources. In 2001, industrial processes emitted 15.9 Tg TSP and 5.87 Tg $PM_{2.5}$. Production of cement, brick, lime, coke, and iron and steel are identified as the main contributors of PM emissions. China's cement industry produced 660 million tons of cement in 2001 and released 7.5 Tg TSP and 4.35 Tg $PM_{2.5}$, representing the largest individual source group. TSP and $PM_{2.5}$ emissions from China's brick production were 3.63 Tg and 0.37 Tg, respectively. This estimate is thought to be quite uncertain, because specific PM emission factors for brick production are not available, and we assumed that brick industrial PM emissions are similar to those of stokers in the industrial sector. We estimate that TSP

emissions from China's coke production and iron and steel production in 2001 were 0.98 Tg and 0.84 Tg, respectively, and their $PM_{2.5}$ emissions were 0.50 Tg and 0.21 Tg, respectively. This estimation is also quite uncertain, partly due to the lack of local emission factors and partly due to the fact that a significant part of the PM emission is fugitive, which is very difficult to measure. We also observe that most PM emissions in coke production come from fugitive emissions in indigenous coking processes, which emitted 0.36 Tg. China's recent regulation on closing indigenous coking plants could be very positive for PM emission reduction.

Most particles emitted from vehicles are fine particles. $PM_{2.5}$ emissions in the transportation sector were 0.28 Tg in China in 2001, about 76% of $PM_{2.5}$ emissions of the transportation sector in the USA in 2002 (USEPA 2007), but only accounting for 2% of China's total anthropogenic $PM_{2.5}$ emissions. We also calculated aerosol emissions from open biomass burning by using the methodology described in Streets *et al* (2003b) and the emission factors from Andreae and Merlet (2001). We estimate that TSP emissions from open biomass burning in China were 2.24 Tg in 2001, where 1.02 Tg were $PM_{2.5}$.

3.2. Carbonaceous aerosols

In previous studies, we estimated that China's BC emissions were 1.34 Tg in 1995 and 1.05 Tg in 2000, respectively (Streets *et al* 2001, 2003a). Our previous estimate of OC emissions for 2000 was 3.38 Tg (Streets *et al* 2003a). The decreasing trend in BC emissions is due to the decrease in solid fuel consumption in the residential sector. Bond *et al* (2004) estimated that China's BC and OC emissions in 1996 were 1.49 Tg and 2.82 Tg, respectively, as a part of their global inventory. They obtained higher estimates of BC emissions by identifying coke production and brick production as two important BC sources for China, while their OC estimates were lower than previous studies due to the lower emission factors used. Cao *et al* (2006) presented 1.50 Tg BC for China for 2000 and attributed the increase from previous studies to coal combustion in rural industry and rural residences. However, they did not report which industries were responsible for the increase.

We estimate that BC and OC emissions from anthropogenic sources in China in 2001 were 1.71 Tg and 3.58 Tg, respectively. The residential sector is the largest contributor to carbonaceous aerosols, emitting 0.87 Tg BC and 2.25 Tg OC in 2001, which is quite comparable with previous studies. We restate our previous estimates (Streets *et al* 2003b) for open biomass burning: 0.11 Tg BC and 0.75 Tg. By using the same emission factors as in Bond *et al* (2004), we estimate that industrial processes contributed 0.46 Tg BC and 0.47 Tg OC in 2001, mostly from coke production and brick production. However, we are not very confident about these numbers, because the emission factors are determined by very few measurements and local measurements are not available.

Estimates of carbonaceous aerosol emissions are thought to be of high uncertainty, especially due to the lack of local measurements in China. In recent studies, Chen *et al* (2005, 2006) measured small coal combustion devices in China and then calculated carbonaceous aerosol emissions for residential coal combustion. Their BC estimates were lower than other previous studies by a factor of two or three, indicating that there are still huge uncertainties in carbonaceous aerosol inventories and additional comprehensive investigations are needed.

3.3. Ca and Mg emissions

Alkaline Ca and Mg are important components of particulate emissions, because of their ability to neutralize acidic species in the atmosphere such as sulfuric and nitric acids. Ca and Mg emissions come mainly from large combustion devices and industrial processes. China's anthropogenic Ca and Mg emissions from anthropogenic sources in 2001 were 4.75 Tg combined. About 94% of Ca and 76% of Mg emissions were emitted from industrial processes, e.g. cement production, lime production, and iron and steel production. The remainder was mainly from combustion, especially from large combustion devices such as power plants and large industrial boilers. Ca and Mg emissions from residential stoves are very small.

Cement production and lime production are the largest contributors of Ca and Mg emissions. Cement production emitted 3.0 Tg Ca and 0.07 Tg Mg in China in 2001, accounting for 66% of total Ca emissions and 30% of total Mg emissions. Ca and Mg emissions from lime production were 1.0 Tg and 0.02 Tg, respectively. The huge Ca and Mg emissions from cement production can be attributed to the following reasons. First, the proportion of Ca in TSP from the process is very high. The unabated TSP emission factor for cement production is about 100–150 g kg^{-1} cement, where about 40% is Ca. Second, under the lenient PM emission standard of the cement industry in 2001, many old cement plants were only equipped with wet scrubbers and cyclones, which had low removal efficiencies and resulted in high final PM emissions. However, it should be noted that China implemented a new strict emission standard for the cement industry in 2005, which required that TSP emissions from all cement plants could not exceed 100 mg per Nm^3 flow gas, corresponding to 1.5 g kg^{-1} cement (SEPA 2004). If all factories are able to meet this requirement, the final TSP emission factor of China's cement industry will be reduced by 70%. This will reduce China's TSP emissions markedly in the future and also reduce Ca and Mg emissions that affect the chemical composition of the atmosphere. This could have important implications for acid/base reactions in the atmosphere, e.g. acid rain formation.

3.4. Regional distribution of emissions

Figure 1 shows the provincial distribution of anthropogenic PM emissions in China in 2001. PM emissions show significant regional differences. The five provinces with the largest emissions are Shandong, Hebei, Jiangsu, Henan and Guangdong, all of which are located in China's coastal regions. TSP emissions in each of the above provinces exceeded 1.5 Tg in 2001 and $PM_{2.5}$ emissions exceeded 0.7 Tg. In some provinces in Western China, TSP emissions were less than 0.2 Tg per year, such as Qinghai, Ningxia and Tibet. Figure 2 presents the provincial differences in aerosol components. We

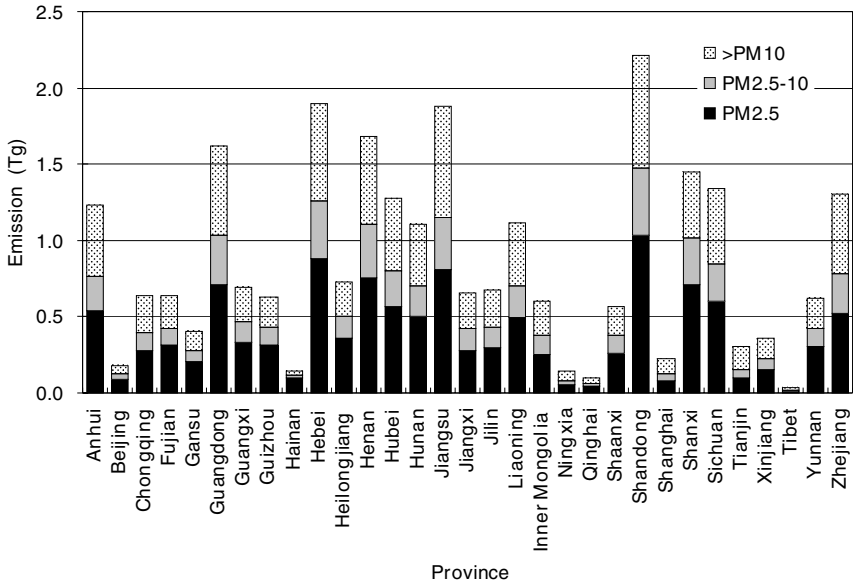

Figure 1. Primary PM emissions by province in China, 2001.

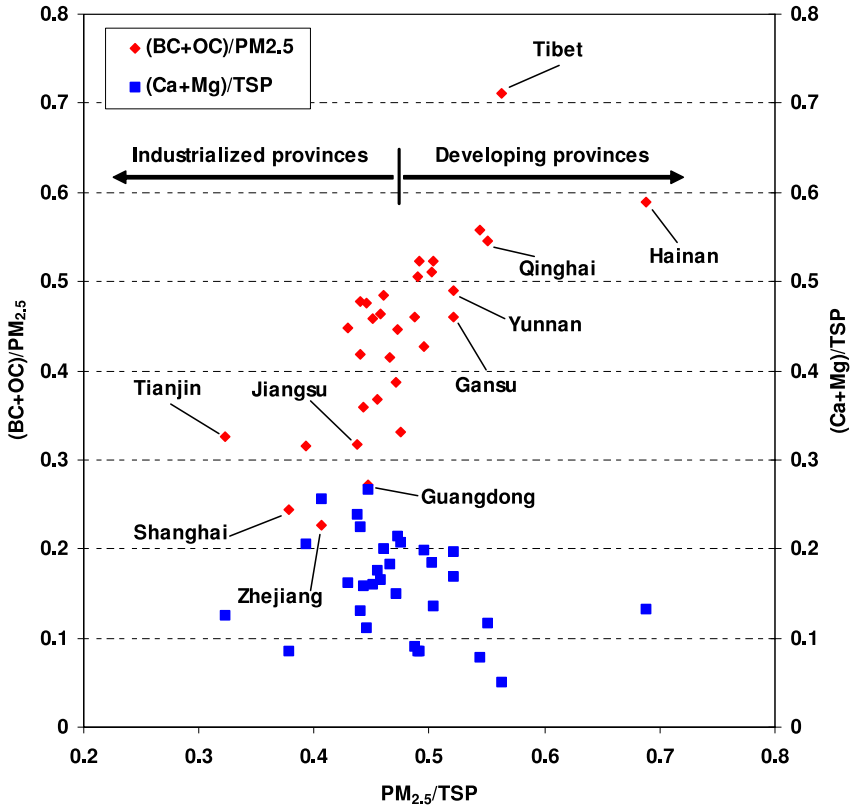

Figure 2. Components of primary aerosol emissions by province.

use carbonaceous aerosols and base cation aerosols as tracers for residential emissions and industrial emissions, respectively. In remote and developing regions, residential emissions are the dominant fraction of PM emissions, with the effect that $PM_{2.5}/TSP$ and $(BC + OC)/PM_{2.5}$ ratios in those provinces are higher, but the $(Ca + Mg)/TSP$ ratio is low. In the industrialized provinces, the industrial sector contributes a large share of PM emissions, where the $(Ca + Mg)/TSP$ ratio is high but the $PM_{2.5}/TSP$ and $(BC + OC)/PM_{2.5}$ ratios are lower. These regional differences in PM emissions in China

are mainly caused by the differences in economic development, industry structure and population.

The magnitudes and components of primary particulate emissions are expected to change dramatically with rapid economic development. In recent years, China has implemented several new emission standards for power plants, cement plants and industrial boilers. The strengthened emission standards will result in a major reduction of PM emissions in key emitting sectors. Industrialized regions, especially megacities such as Beijing and Shanghai, are taking more active measures to prevent the release of health-damaging pollutants. Energy consumption and industrial production are still growing; however, the increased penetration of effective PM control equipment will remove most of the coarse fraction of PM emissions, leading to an increasing trend in the $PM_{2.5}$/TSP ratio. The (Ca + Mg)/TSP ratio is expected to decrease in industrialized regions, due to the abatement of emissions from industrial processes. Developing regions will tend to move toward the characteristics of industrialized regions.

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