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Quantifying the uncertainties of a bottom-up emission inventory of anthropogenic atmospheric pollutants in China

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Abstract

The uncertainties of a national, bottom-up inventory of Chinese emissions of anthropogenic SO_2, NO_x, and particulate matter (PM) of different size classes and carbonaceous species are comprehensively quantified, for the first time, using Monte Carlo simulation. The inventory is structured by seven dominant sectors: coal-fired electric power, cement, iron and steel, other industry (boiler combustion), other industry (non-combustion processes), transportation, and residential. For each parameter related to emission factors or activity-level calculations, the uncertainties, represented as probability distributions, are either statistically fitted using results of domestic field tests or, when these are lacking, estimated based on foreign or other domestic data. The uncertainties (i.e., 95% confidence intervals around the central estimates) of Chinese emissions of SO_2, NO_x, total PM, PM_{10}, $PM_{2.5}$, black carbon (BC), and organic carbon (OC) in 2005 are estimated to be −14%~12%, −10%~36%, −10%~36%, −12%~42% −16%~52%, −23%~130%, and −37%~117%, respectively. Variations at activity levels (e.g., energy consumption or industrial production) are not the main source of emission uncertainties. Due to narrow classification of source types, large sample sizes, and relatively high data quality, the coal-fired power sector is estimated to have the smallest emission uncertainties for all species except BC and OC. Due to poorer source classifications and a wider range of estimated emission factors, considerable uncertainties of NO_x and PM emissions from cement production and boiler combustion in other industries are found. The probability distributions of emission factors for biomass burning, the largest source of BC and OC, are fitted based on very limited domestic field measurements, and special caution should thus be taken interpreting these emission uncertainties. Although Monte Carlo simulation yields narrowed estimates of uncertainties compared to previous bottom-up emission studies, the results are not always consistent with those derived from satellite observations. The results thus represent an incremental research advance; while the analysis provides current estimates of uncertainty to researchers investigating Chinese and global atmospheric transport and

chemistry, it also identifies specific needs in data collection and analysis to improve on them. Strengthened quantification of emissions of the included species and other, closely associated ones – notably CO_2, generated largely by the same processes and thus subject to many of the same parameter uncertainties – is essential not only for science but for the design of policies to redress critical atmospheric environmental hazards at local, regional, and global scales.

1 Introduction

A series of studies have been conducted using bottom-up methods to explore Chinese emissions of anthropogenic atmospheric pollutants (Streets et al., 2001, 2003; Hao et al., 2002; Cao et al., 2006; Ohara et al., 2007; Zhang et al., 2009d; Klimont et al., 2009; Lei et al., 2010a). These studies indicated significant emission increases since 2000, attributed mainly to fast growth of the economy and energy consumption. Some of these results have been applied with different chemical transport models (CTMs) to simulate effects on regional air pollution and soil acidification in China (Carmichael et al., 2003a; Zhang et al., 2004; He et al., 2007; Wang et al., 2007; Saikawa et al., 2009; Chen et al., 2009; Zhao et al., 2009). Such CTM simulations, however, have often yielded modeled concentrations that are inconsistent with ground-, aircraft-, or space-based observations, raising questions about the uncertainties of bottom-up emission inventories (along with those of observational methods). For example, Carmichael et al. (2003b) compared modeled values and observations in Asia obtained in the Transport and Chemical Evolution over the Pacific (TRACE-P) program. They concluded that although TRACE-P emission inventories were of sufficient quality to support modeling studies of photochemistry, large discrepancies existed between modeled and observed behavior in central China and the Yellow Sea, probably due to underestimation of residential emissions. Ma et al. (2006) found that TRACE-P underestimated the tropospheric NO_2 column densities in China by more than 50% compared to satellite observations. Wang et al. (2004) applied the GEOS-Chem CTM in inverse mode to

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surface observations and TRACE-P aircraft measurements, indicating NO_x emissions 47% higher than those estimated bottom-up, with the largest discrepancy in central China. In another study, Wang et al. (2007) found a 33% underestimate of NO_x in east China, proposing biomass burning and microbial sources as additional sources. An inverse study by Zhao and Wang (2009), however, indicated a bottom-up overestimate of NO_x emissions in developed areas of east China. These studies, while sometimes inconsistent with each other, indicate a need for greater understanding of the uncertainties of bottom-up emission inventories.

Uncertainties introduced by energy statistics and applications of non-Chinese emission factors have been estimated in previous Chinese emission inventory studies. Streets et al. (2003) applied expert judgment, in which the coefficients of variation (i.e., standard deviation divided by the mean) of the activity levels were assumed based on measures of economic development and perceived statistical quality, and those of emission factors were based on the reliability ratings of US emission factors (USEPA, 2002). They estimated that the uncertainties of Chinese emissions varied from −12%~13% for SO_2 to −83%~495% for organic carbon (expressed as the lower and upper bounds of a 95% confidence interval, CI, around a central estimate). To investigate discrepancies in the trends of bottom-up NO_x emissions and of the NO_2 column by satellite, Zhang et al. (2007a) set alternative emission scenarios in which energy statistics from different sources and emission factors with different control levels were applied. They concluded that NO_x emissions in east-central China in 2004 could range from 7.9 to 9.3 Tg, and the annual growth rate of emissions from 1995 to 2004 could range from 5.5% to 7.1%. Other studies conducted preliminary uncertainty analyses of certain species or sectors in China using statistical methods. Bond et al. (2004) assumed that most emission factors of particulate matter (PM) followed lognormal distributions (with the exception of those of diesel vehicles, which had a gamma distribution) and evaluated the uncertainties of global black carbon (BC) and organic carbon (OC) emissions. The 95% CIs for Chinese emissions were estimated to be −37%~147% and −44%~104%, respectively. Wu et al. (2010) focused on mercury

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emissions from coal-fired power plants in China and estimated an 80% CI in 2003 of −37%∼71%.

To better understand the uncertainties of atmospheric pollutant emissions, Frey and Zheng (2002) developed a bootstrap simulation method and analyzed the uncertainties of NO_x emission factors and emissions of coal-fired power plants of different technology types. This method was then applied to other species and sectors (Frey and Li, 2003; Frey and Zhao, 2004). Based on bootstrap and Monte Carlo simulations, an emission factor database for Chinese coal-fired power plants has been established with detailed categories of combustion technologies and fuel qualities (Zhao et al., 2010). Such methods have proven difficult to apply to even one region of China due to lack of supporting data (Zheng et al., 2009), and they have never been previously used to evaluate the uncertainties of an integrated emission inventory for the entire country.

In this study, therefore, the uncertainties (expressed as the 95% CI around the central estimate) of a bottom-up emission inventory in China is evaluated with Monte Carlo simulations, combining comprehensive field measurements of domestic emission factors and investigations of activity levels. The species include SO_2, NO_x, total PM, PM_{10}, $PM_{2.5}$, BC, and OC. The target year is 2005 because it has the most available data. Determining the probability distributions of all of the related parameters is the main undertaking of this study. Section 2 reviews bottom-up emission inventory methods and analyzes the uncertainties of emission source fractions and corresponding activity levels. Section 3 is a thorough analysis of emission factors by sector and fuel type. Section 4 presents the results and related discussion of emission uncertainties by sector, sensitivity analyses, reliability and limitations of the analyses, and comparisons with other studies. Section 5 summarizes the present study.

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2 Uncertainties of activity levels and source categories

2.1 Review of emission inventory methodology and uncertainty analysis

The methodology of calculating bottom-up emission inventories has been detailed in previous studies, e.g., Streets et al. (2003), Ohara et al. (2007), and Zhang et al. (2009d). To set the context for analysis of the uncertainties of related parameters, we briefly review the methodology and make a small adjustment in the categorization of emission sources.

The emissions of each species are estimated by province and sector and then aggregated to the national level, as the product of activity levels (energy consumption or industrial/agricultural production), unabated emission factors (expressed as the mass of emitted pollutant per unit activity level), and one minus the removal efficiency. In total, seven sectors are included: coal-fired power plants (CPP), cement plants (CEM), iron and steel plants (ISP), other industries' boiler combustion (IND), other industries' non-combustion processes (PRO), transportation (TRA), and residential combustion (RES) (see Fig. S1 in the supplementary material for more details).

Emissions from stationary sources are calculated using Eq. (1):

$$E_{i,j} = \sum_k \sum_m \sum_n AL_{j,k,m,n} \times EF_{i,j,k,m} \times R_{i,j,k,m,n} \times (1-\eta_{i,n}) \tag{1}$$

where i, j, k, m, and n stand for species, province, sector, fuel type, and emission control technology, respectively; AL is the activity level; EF is the unabated emission factor; R is the penetration rate of emission control technology; and η is the removal efficiency. The application of emission control technologies is mainly considered for SO_2 and PM removal in power generation and industrial sectors.

Regarding SO_2 emissions from combustion sources, the emission levels are closely related to the sulfur content of fuels, and thus can be calculated using Eq. (2):

$$E_{SO_2,j} = \sum_k \sum_m \sum_n AL_{j,k,m,n} \times SC_{j,m} \times SR_{k,m} \times R_{j,k,m,n} \times (1-\eta_n) \times 2 \tag{2}$$

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where SC is the sulfur content of fuel; and SR is the sulfur release ratio (%).

Similarly, PM emissions from coal combustion sources, including size distribution, can be calculated using Eq. (3):

$$EF_{\mathrm{PM},y,j,m=\mathrm{coal}} = \sum_k \sum_n AL_{j,k,m=\mathrm{coal},n} \times AC_{j,m=\mathrm{coal}} \times AR_{k,m=\mathrm{coal}} \times R_{j,k,m=\mathrm{coal},n} \times f_{k,m=\mathrm{coal},y} \times (1-\eta_{n,y}) \quad (3)$$

where y stands for the particulate size; AC is the ash content of the fuel; AR is the ash release ratio (%); and f is the particulate mass fraction by size.

Emissions from mobile sources are calculated as the product of fuel consumption and the emission factor expressed as emitted pollutant per unit fuel consumption.

For carbonaceous aerosols, the emission factors are drawn directly from field measurements of small coal stoves and biomass burning, and are obtained by applying the mass fractions of BC (F_{BC}) and OC (F_{OC}) to $PM_{2.5}$ for other sectors or fuels.

Monte Carlo simulation is used to analyze the uncertainties of the emission inventory in this study. For parameters with adequate domestic measurement data, a probability distribution is fitted using Crystal Ball, a statistical software package, and the Kolmogorov-Smirnov test for the goodness-of-fit ($p = 0.05$). For parameters with limited observation data, and those that fail to pass the goodness-of-fit test, probability distributions must be assumed by the authors. Finally, all of the input parameters of activity levels and emission factors, with corresponding statistical distributions, are placed in a Monte Carlo framework, and 10 000 simulations are performed to analyze the emission uncertainties by sector and species, as well as which parameters significantly contribute to the uncertainties.

2.2 Uncertainties of activity levels

The accuracy of activity levels (usually from energy and economy statistics) has been discussed in previous studies. Akimoto et al. (2006) argued that Chinese energy

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consumption during 1996–2003 was probably underestimated and thereby not recommended for use in the study of emission inventories during this period. However, Zhang et al. (2007a) and Wu et al. (2010) found that the discrepancies in energy data from different sources were relatively small (e.g., less than ±5% for coal consumption by the power sector) and did not dominate the emission uncertainties. That conclusion is supported by comparison of energy statistics and the compiled coal consumption at unit level by the authors (Zhao et al., 2008). Therefore, we generally assume normal distributions with coefficients of variation (CV, the standard deviation divided by the mean) of 5% of the mean values for fossil fuel consumption by power, industry, and residential sectors. The same distributions are also applied for the production of cement, iron and steel, and other industrial processes including manufacturing of lime, brick, nonferrous metal, glass, sulfuric acid, nitric acid, fertilizer, and the refining of oil. These assumptions of distributions are thus based on expert judgment.

For on-road vehicles, the fuel consumption is calculated with Eq. (4):

$$AL_j = \sum_t VP_{j,t} \times VMT_{j,t} \times FE_{j,t} \tag{4}$$

where t is the vehicle type; VP is the vehicle population; VMT is annual average vehicle mileage traveled, and FE is the average value of on-road fuel economy (fuel consumed per kilometer traveled).

Seven vehicle types are assessed: light duty gasoline vehicles (LDGV), light duty diesel vehicles (LDDV), light duty gasoline trucks (LDGT), light duty diesel trucks (LDDT), heavy duty gasoline vehicles (HDGV), heavy duty diesel vehicles (HDDV), and motorcycles (MC). As with fossil energy use in non-transportation sectors and with industrial production, we assume a normal distribution of the vehicle population by type, with CV of 5%. The mean values of VMT in this study are obtained from He et al. (2005) and Wang et al. (2006). A global study determined that the uncertainties of VMT had a normal distribution with CV of 10% (Kioutsioukis et al., 2004). A Chinese domestic study in Shanghai, however, implied that the discrepancies of VMT calculated by different methods were less than 10% (Wang et al., 2008). In this study we thus assume

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a normal distribution with CV of 5% for the *VMT*. Regarding fuel economy (*FE*), the China Association of Automobile Manufacturers and China Automotive Technology & Research Center conducted national surveys covering 984 vehicles from 38 manufacturers (CAAM and CATRA, 2009), and the resulting *FE* data fitted a normal distribution with CV of 14%. Based on these assumptions, the CV of fuel consumption by on-road vehicles in China is estimated to be 16% in the Monte Carlo simulations, comparable to an estimate used in international research (Karvosenoja et al., 2008). Regarding non-road transportation, since there is little information available for uncertainty analysis, we tentatively assume that the probability distribution is same as on-road vehicles, i.e., normal distribution with CV of 16%.

Burning of biomass is another important emission activity. Estimates of consumption of firewood and agricultural wastes for biofuel are obtained from official statistics on non-commercial energy consumption. Given larger presumed uncertainties of statistics for such informal energy use compared to that of commercial energy, the probability of biofuel consumption is assumed to have a normal distribution with CV of 20%. Open burning of agricultural wastes, i.e. not as biofuels but to clear fields, is calculated using Eq. (5):

$$AL_j = \sum_g GP_{j,g} \times RSG_g \times RB_j \tag{5}$$

where g is the grain type; *GP* is the grain production; *RSG* is the waste-to-grain ratio of the plant; and *RB* is the percentage of residual material that is burned in the fields.

The grain production is taken from official agricultural statistics, with an assumed normal distribution and CV of 5%. The mean values and ranges of *RSG* are taken from a global study of different grain types (Lal, 2005). Since the data are not specific for China and thus have high uncertainty, a uniform distribution is assumed, in which the probability is constant within the range. The mean values and CVs of *RB* by province are obtained from a questionnaire-based investigation (Wang and Zhang, 2008), with a normal distribution assumed.

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Table S1 in the Supplement summarizes the uncertainties of the activity levels.

2.3 Uncertainties of emission source fractions

This section discusses the uncertainties of fractions or penetrations of different technologies for each source category by sector, i.e., the *R* values in Eqs. (1)–(3), which are also summarized in Table S2 in the supplementary material.

A thorough investigation by the authors has produced a detailed database for the coal-fired power sector, in which all information related to emissions were compiled at the generating unit level (Zhao et al., 2008). Thus we believe that the penetration rates of different boilers, burners, fuels, and emission control devices are fully known and assume no uncertainty for them.

Cement kilns mainly includes shaft, precalciner, and other rotary kilns. Although shaft kilns with poor operations and low efficiency dominated the cement industry in the 1990s, deployment of precalciner kilns has been dramatically increasing since 2002 (Lei et al., 2010b). In this study, the penetration of precalciner kilns is estimated to be 44.3% in 2005, based on investigation of actual operations of precalciner manufacturing lines by province. Regarding the uncertainty, we calculated the lower and upper bounds to be 38.4% and 50.3% by assuming that none of the newly built manufacturing lines were operated in 2005 or that all were fully operated, respectively. With this likeliest value and these bounds, a triangular distribution is given to this parameter.

As shown in Fig. S1, the iron and steel industry is mainly comprised of five processes. (Although part of coke production occurs outside of the iron and steel industry in China, we include it in this sector for classification simplicity.) In the steel-making process, the penetration rates of open hearth, converter, and electric arc furnace technologies were 0.2%, 88.1%, and 11.7%, respectively, according to statistics of the China Iron and Steel Association, and no uncertainty is assumed for this parameter.

Coal-fired industrial boilers include grate and circulating fluidized bed (CFB) combustion types. According to different sources, the penetration rate of CFB ranged from 8–10% in recent years (Huang and Xia, 2004; Qu, 2008), and its probability distribution

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is assumed to be uniform. Regarding coal consumption by residential boilers, there are currently no statistical data for the shares of different technologies and large variations may exist. Zhang et al. (2007b) estimated the fractions of grate boilers, hot water systems, and small stoves to be 28%, 19%, and 53% respectively, while Zhang et al. (2009a) suggested that the fraction of stoves reaches 61%. In this study we take the results of those two studies and assume uniform distributions given the high uncertainties of the parameters.

Penetration rates of emission control devices are important to emission estimation. As noted above, the penetrations of emission control devices in the power sector, including flue gas desulphurization systems (FGD), low-NO_x burners (LNB), fabric filter systems (FF), electrostatic precipitators (ESP), wet scrubbers (WET), and cyclones (CYC) are known at the unit level for 2005. Unfortunately, there is very little available statistical evidence on emission control deployments in other sectors. In this study we compile the penetration rates of dust collectors in industrial and residential sectors from investigations by Zhang et al. (2007b) and Lei et al. (2010a, b), and must assume uniform distributions for those parameters without further data support.

For the transportation sector, new on-road vehicles have ideally been required to meet China's stage I and II standards (equivalent to Euro I and II) since 2000 and 2004, respectively. The fleet compositions of different control levels by vehicle type are calculated based on an average vehicle age of 15 years. It is difficult to evaluate the uncertainties for this parameter, not only because of the large variation of vehicle ages but also because emission requirements are not necessarily met by corresponding vehicles. Therefore, normal distributions with CV of 20% are tentatively assumed for the vehicle fractions meeting the requirements of the stage I or II standards.

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3 Uncertainty analysis of emission factors

3.1 Unabated emission factors

The uncertainties of unabated emission factors for stationary sources are summarized in detail in Table S3 of the supplementary material.

Uncertainties of emission factors for coal-fired power plants have been analyzed in our previous study combining bootstrap and Monte Carlo simulation with consideration of boiler type, fuel quality, and emission control device (Zhao et al., 2010). Those results are used in this work. Shown in Fig. 1 are the probability distributions of NO_x emission factors for nine different categories of coal-fired power units, obtained from 309 measurement data points (Zhao et al., 2010). The release ratios of sulfur and ash are estimated to be 90% and 69%, respectively, both with beta distributions. Regarding the sulfur and ash contents of coal, the probability distributions are fitted at the provincial level using the plant-by-plant data compiled by the authors (Zhao et al., 2008).

For boilers in the industrial sector, the sulfur release ratio of coal combustion (*SR* in Eq. 2) is estimated from domestic measurements (SEPA, 1996) to be 85% with a beta distribution, lower than the value of 90% for power plants (Zhao et al., 2010). Similarly, the ash release ratio (*AR* in Eq. 3) for grate stokers, the dominant industrial boiler type, is estimated to be 13% with a logistic distribution. The ratio for CFB boilers ranges from 48–60% based on field measurements by the authors (Zhao et al., 2010), with a uniform distribution. The sulfur and ash contents of coal by province are calculated based on the mined coal quality and the inter-provincial coal flows. With little information on uncertainty, normal distributions with CVs of 20% are assumed for those parameters. The NO_x emission factor for grate stokers is estimated to be 4.2 kg per metric ton of coal burned (kg/t), and the probability is fitted to a lognormal distribution with geometric standard deviation (GSD) of 1.8 kg/t based on 93 data points ranging 1.1–24.5 kg/t in tests by SEPA (1996). The mean value is comparable to the result from a US study, 2.8–5.5 kg/t (USEPA, 2002), but the quite long tail of the distribution

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implies large uncertainty for the parameter, as shown in Fig. 1j. The NO_x emission factor for CFB boilers is assumed to be 20–40% lower than regular boilers based on measurements (Zhao et al., 2010), and a uniform distribution is assumed.

For grate boilers used in the residential sector, we apply the same sulfur and ash release ratios and NO_x emission factor as those for the industrial sector. For hot-water systems, the sulfur release ratio and emission factors for NO_x and PM are estimated to be 80%, 1.8 kg/t, and 1.9 kg/t, respectively, with beta, gamma, and lognormal distributions, based on measurements (SEPA, 1996). For small coal-combustion stoves, the sulfur release ratio is assumed to be the same as that of hot water systems. Zhang and Smith (2000) conducted field measurements of 28 fuel/stove combinations (including other fuels like biomass) in China. According to their results, the NO_x emission factors for coal stoves range from 0.1–3.9 kg/t, with a mean of 0.9 kg/t. A triangular distribution is thus assumed in this study. Chen et al. (2005, 2006) measured PM emission characteristics from residential stoves burning bituminous and anthracite coals. Based on their results and the application rates of those coal types, PM emission factors are estimated to be 11 kg/t, with a beta distribution.

Regarding combustion of biofuels (agricultural waste and firewood), the uncertainties of emission factors for different species are fitted and summarized in Table S3, based on the measurements by Zhang and Smith (2000) and Li et al. (2007a). Li et al. (2007b) conducted emission tests of open biomass burning, and the results are applied in this study. Since the samplings were not enough for probability fitting, uniform distributions are assumed for all the emission factors of open burning. Emission factors of stationary sources burning other fuels like oil and natural gas are mainly taken from Zhang and Smith (2000) and Hao et al. (2002), as well as foreign studies (USEPA, 2002; Klimont et al., 2002). Uniform distributions are assumed due to the limited data availability.

For cement kilns, the sulfur release ratio and sulfur contents of coal are assumed to be the same as those of industrial boilers. To reevaluate the emission standards of cement making, the Chinese Research Academy of Environmental Sciences (CRAES) conducted national surveys and measured NO_x emission factors of 20 kilns with

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different technologies (CRAES, 2003). With these published mean values and ranges, the uncertainties of NO_x emission factors by technology are assumed to have triangular distributions, as shown in Table S3. The distributions of PM emission factors are fitted by technology based on field tests (SEPA, 1996) and thorough investigation by Lei et al. (2010b).

The SO_2 and NO_x emissions of iron and steel production are mainly from the sintering process. The emission factor for SO_2 is estimated to be 2.7 kg/t of product with lognormal distribution (GSD: 1.5) (SEPA, 1996), and for NO_x it is 0.64 kg/t, with triangular distribution from 0.5–0.76 (AISGC, 2007). The uncertainties of PM emission factors are fitted based on domestic tests of sintering, pig iron production, and steel making by technology type (SEPA, 1996). For other processes like coking and casting, the emission factors from Lei et al. (2010a) are applied, and normal distributions with CV of 20% are assumed on a tentative basis.

Uncertainties of emission factors of other industrial processes (e.g., lime and non-ferrous metal production) are mainly fitted based on the database of SEPA (1996), as summarized in Table S3.

For the transportation sector, an emission factor database of on-road vehicles in China by Zhang et al. (2008a, 2009a) is used in this study, determined by the regulatory standards that they were required to meet at the time of manufacture: pre-stage I, stage I, or stage II (see details in Table S4 in the Supplement). Due to a lack of domestic data, however, the emission factors for non-road sources must be taken from foreign studies (Kean et al., 2000; Klimont et al., 2002). Liu et al. (2009) conducted emission factor tests of on-road, heavy-duty diesel vehicles, and found that the CVs of NO_x emission factors were 36% and 17% for stage I and II vehicles, respectively, and the analogous values for PM were 59% and 34%. Without further information, we apply those values for all vehicle types and assume that CVs of pre-stage I vehicles and non-road sources are as large as those of stage I. With those CVs, lognormal distributions are assumed for all the emission factors of transportation sources (Karvosenoja et al., 2008).

3.2 Size distribution and carbonaceous fractions of PM

The uncertainties of PM of different size categories (i.e., *f* in Eq. 3) from coal-fired power plants were analyzed by the authors using field measurements (Zhao et al., 2010) and are directly incorporated into this study. Although a few similar tests have been conducted of industrial boilers, most of the results cannot be directly applied due to lack of data on coarse fractions. Therefore, results of foreign studies are used for sectors other than power generation (USEPA, 2002; Klimont et al., 2002), and uniform distributions are assumed given the potentially large uncertainties. Additionally, PM emitted from gasoline or diesel combustion in the transportation sector is assumed to be entirely $PM_{2.5}$ (see details in Table S5 in the Supplement).

Regarding the carbonaceous species, field studies found that the ratios of BC and OC to $PM_{2.5}$ for grate boilers ranged from 0–22% and 1–23%, respectively (Zhang et al., 2008b; Wang et al., 2009; Li et al., 2009a). The BC and OC emission factors were tested for coal (Chen et al., 2005, 2006; Zhang et al., 2008b) and biomass open burning (Li et al., 2007b). The irregularity of the resulting data, due to large variations of combustion technologies and conditions, cannot be easily fitted to distributions for coal and open biomass combustion, and uniform distributions are thus conservatively assumed. For biofuel burned in residential applications, lognormal distributions are fitted based on the field measurement results by Li et al. (2007a, 2009b). Regarding transportation, Zhang et al. (2009b, c) measured emissions of heavy-duty diesel engines and obtained 23 datasets of carbonaceous emissions. The ratios of BC and OC to $PM_{2.5}$, respectively, are estimated to be 43% with a gamma distribution and 37% with a logistic distribution. For OC and BC from other sectors, the ranges of non-Chinese studies are accepted (Streets et al., 2001; Bond et al., 2004; Cao et al., 2006) due to lack of domestic research, with uniform distributions assumed.

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3.3 Effects of emission control devices

The emission control devices evaluated include FGD systems and different types of dust collectors. Regarding NO_x control, the primary technologies of selective catalytic reduction and selective non-catalytic reduction were rarely in use in 2005 and thus are not included in the analysis.

Although the SO_2 removal efficiency of wet-FGD can ideally reach 95% (Zhao et al., 2010), actual operations very rarely achieve this (Xu et al., 2009). The current study uses results of an unpublished government survey concluding that the removal efficiency of FGD in practice is 75%, with a triangular distribution (55–95%). The control effects of other FGD systems are poorer, with removal efficiency of only 20% (Zhao et al., 2010) and a triangular distribution (10–60%).

Electrostatic precipitators (ESP) are the most widely applied dust collectors in power plants. Thorough field measurements conducted by the authors estimated the removal efficiencies for $PM_{2.5}$, $PM_{2.5-10}$, and $PM_{>10}$ to be 92.31%, 96.97%, and 99.46%, with lognormal, lognormal, and normal distributions, respectively (Zhao et al., 2010). Besides ESP, a few tests have been conducted in China on other dust collectors including fabric filter systems (FF), wet scrubbers (WET), and cyclones (CYC) (Yi et al., 2008; Wang et al., 2009; Li et al., 2009a; Zhao et al., 2010), but the sampling data are insufficient for probability fitting. In this study we accept the results of those studies and assume triangular distributions (see details in Table S6 in the supplementary materials). The side effects of PM control by wet-FGD systems were also measured by the authors (Zhao et al., 2010) and the removal efficiencies of $PM_{2.5}$, $PM_{2.5-10}$, and $PM_{>10}$ were respectively estimated to be 53.74%, 81.21%, and 92.63%, with normal distributions.

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4 Results and discussion

4.1 Uncertainties of national emission inventory

Chinese emissions in 2005 with 95% CIs are shown in Table 1 by sector. To be consistent with previous studies, emissions from industrial boilers, cement plants, iron and steel plants, and other industrial processes are aggregated into a single category of industry emissions (see Table S7 in the Supplement for emissions and uncertainties in these sub-sectors). The uncertainties (expressed as the 95% CI around the central value) are estimated to be −14%~12%, −10%~36%, −10%~36%, −12%~42%, −16%~52%, −23%~130%, and −37%~117% for total emissions of SO_2, NO_x, PM, PM_{10}, $PM_{2.5}$, BC, and OC respectively (see Fig. S2 for the output distributions of those species by Monte Carlo simulation).

As the largest contributor of national SO_2 and NO_x emissions (52% and 34% of the totals in this study, respectively), the emissions from coal-fired power generation are the least uncertain among the four sectors for these gaseous pollutants, as well as for total PM and PM_{10}. Regarding carbonaceous aerosols, despite very large uncertainties, emissions from power plants have little impact on the total variations due to tiny shares of emissions. Among all species, the 95% CI for NO_x is the smallest, ranging from −19%~16%, as it is derived from thorough data collection and detailed categories of power units.

In contrast to power plants, the largest uncertainty for NO_x is found in the total industry sector, ranging from −29%~93%, with values reaching −48%~169% for industrial boilers. Due to poor resolution of the data with respect to technology and fuel types, the activity levels of industrial boilers are largely undifferentiated and one emission factor is applied for almost the entire sub-sector. This ignores differences of combustion efficiencies and fuel qualities across industrial boilers, and yields considerable uncertainty. This is also supported by a region-specific study in China (Zheng et al., 2009). Large uncertainty is also found for SO_2 emissions from iron and steel plants (ISP), of which the 95% CI reaches -51%~102% due to variations of emission factors of

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the sintering process. For sub-sectors other than ISP, the emission uncertainties of PM (particularly $PM_{2.5}$ and carbonaceous aerosols) are generally larger than those of other species. The estimated uncertainties of emissions from industrial processes (PRO in Table S7 in the supplementary materials) are relatively small. Those results, however, do not imply that the emission characteristics of PRO are well understood, because: (1) very few field measurements for PRO emissions can be found and the calculations rely mainly on one source (SEPA, 1996); (2) the results shown in Table S7 aggregate the uncertainties of all industrial processes and thus do not reflect larger uncertainties for individual processes (e.g., the 95% CI of PM emissions from lime production is −55%∼127%, much larger than that of total PRO, −39%∼26%).

The residential sector is the main contributor of carbonaceous aerosols (50% and 79% of the BC and OC totals, respectively, in this study), and the largest uncertainties in this sector are found for these species. Regarding the emissions by fuel type, the estimated uncertainties for PM and carbonaceous aerosols from fossil fuel combustion are generally larger than those from biomass combustion (see Table S7 in the supplementary for details). Some explanations of the result follow. The PM emission characteristics of residential coal combustion in China were tested by different researchers (Zhang and Smith, 2000; Chen et al., 2005; Zhang et al., 2008b), yielding varied emission factors. Very few estimates of the emission factors of Chinese combustion of biomass can be found, however, except for a series of measurements by Li et al. (2007a, 2007b, 2009b). Those measurements were made in four provinces with different types of biomass. As no other domestic tests are available for direct comparisons, systematic biases in these available measurements cannot be ruled out. In this study, we adopt the results from Li et al. (2007a, 2007b, 2009b) fully, lacking further domestic evidence. Given potential high uncertainties, however, a comparison test is conducted for OC emitted by its leading source, biomass combustion, in which the uncertainties of emission factors obtained from Li et al. (2007a, 2007b, 2009b), are replaced with those of foreign studies (Andreae and Merlet, 2001; Streets et al., 2001; Bond et al., 2004). In this case the 95% CI of OC emission from biomass combustion

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changes slightly from −57∼226% to −28%∼292%, suggesting caution in interpretation of results based on limited domestic field measurements (see also the discussion in Sect. 4.3).

4.2 Sensitivity analyses

In addition to estimating uncertainty, sensitivity analyses are conducted using Monte Carlo simulation. The results are expressed as contributions of parameter uncertainties to the variance of emissions by sector and species, as shown in Table 2.

In the power sector, SO_2 emissions are estimated to be most sensitive to the sulfur content of coal, at least in the several provinces examined. These include Guizhou, where high-sulfur coal is dominant, and Shandong, where the largest installed capacity of power units is found. NO_x emissions are most sensitive to the emission factors of bituminous-burning units without LNB. It should be noted, however, that such small and inefficient units have largely been shut down since 2005 under national policies to save energy and reduce emissions, and thus this contribution to emission uncertainty should have largely diminished. The emissions of PM of different size categories are generally sensitive to the $PM_{2.5}$ mass fraction of unabated total PM for pulverized boilers and the removal efficiency of $PM_{2.5}$ by ESP, suggesting a need for more research on fine particles.

In the industrial sector, emissions of gaseous pollutants and carbonaceous aerosols are most sensitive to estimates of combustion by industrial grate boilers. This is particularly true for NO_x, in which the uncertainty of the emission factor for grate boilers is estimated to contribute 86% to the variance of emissions. In cement processing (i.e., not combustion but cooling, grinding, and crushing), emissions of PM of different sizes are most sensitive to the unabated PM emission factor and $PM_{2.5}$ mass fraction. This result spotlights the complexity of the emission characteristics of the cement industry. Even after the most thorough evaluation to date (Lei et al., 2010b), there are still large uncertainties in the PM emissions from cement, which contribute significantly to the variation of total industry PM emissions.

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The emissions of most species in the transportation and residential sectors are most sensitive to emission factors of non-road sources and of biomass burning, respectively. Due to scarce data, the uncertainties of these emission factors are assumed based either on other related sources (in the case of non-road transportation) or limited field tests (biomass burning). Thus the actual variations of these parameters might even be larger than those assumed in this study.

Due to low penetration rates of advanced emission control devices like wet-FGD, the contributions of their removal efficiencies to emission uncertainties were not significant in 2005. Given sharply expanded deployment in subsequent years, however, they likely play an increasingly important role. For example, the penetration rate of wet-FGD systems will increase from 14% of the installed capacity in 2005 to over 70% in 2010, based on the updated power sector database of the authors (Zhao et al., 2008). Applying the same method and assumptions used for 2005, the uncertainty of SO_2 removal efficiency by wet-FGD is estimated to contribute 70% of the variance of SO_2 emissions from power plants, in contrast to only 2% in 2005. For this reason, understanding the actual operational effectiveness of wet-FGD as well as other emission control devices like SCR and FF will be critical to reducing the relevant emission uncertainty in near future.

4.3 Reliability of uncertainty analysis

As shown in Tables S1–S6 in the supplementary material, each probability distribution included in the uncertainty analysis is categorized as A, B, C, or D for ease of discussion, based on the following criteria: A indicates distributions obtained by data fitting of domestic field measurements; B also represents distributions based on domestic field measurements, but when data fitting is infeasible; C indicates that the distribution is determined from foreign studies; and D represents distributions that must be assumed by expert judgment, lacking relevant data. The ratings from A to D thus represent a diminishing statistical basis for determining parameter uncertainties, and for the most

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part thereby reflect decreasing reliability of the uncertainty analyses. (In one case discussed later in this section, this criterion of reliability may not hold.)

The parameters related to activity levels and penetrations of technologies and emission control devices are mostly rated D, except for those of the data-rich power sector, implying relatively poor reliability of the uncertainty analyses of those parameters. The reasons include: (1) a lack of published independent research to compare to official energy and industrial data, making it difficult to systematically quantify the uncertainties of official statistics; and (2) investigations of technology penetration are still far from adequate to supply trustworthy values or ranges, particularly for secondary industrial processes. Despite such D-level reliability, however, most of these parameters (particularly in commercial energy uses) contribute insignificantly to the total emission variations and thus are not critical determinants of the final emission inventory uncertainties, as discussed in Sect. 4.2. Similar conclusions are drawn by Zhang et al. (2007a) and Wu et al. (2010).

The ratings for emission factors vary considerably by sector and species. Among those classified C and D, the PM emission factors and the mass fraction of $PM_{2.5}$ for some industrial sources like cement and brick production are important to the uncertainties of industrial PM emissions. Similarly, the emission factors of non-road transportation sources including rural tractors and construction equipment are important to the uncertainties of transportation emissions. With few domestic field tests to date, these parameters are based largely on results of foreign studies. To improve the reliability of uncertainty analyses and reduce the uncertainties of Chinese emission inventories, new research on such parameters with relatively low reliability but high contributions to emission variations is particularly required.

The situations for emission factors with higher reliability (i.e., A and B) are more complicated. For coal-fired power plants, the data on which the probability distributions for most parameters are fitted are obtained from both thorough field measurements by the authors and published studies by others (see details in Zhao et al., 2010). These support different emission factors across burner types, fuel qualities, and emission

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control levels. The uncertainty analyses for this sector are thus the most reliable in this study. Such confidence, however, does not apply to the residential sector, particularly biomass burning, even though the emission factors meet A or B criteria. As discussed in Sect. 4.1, the data on which the distributions are based are limited in number and systematic bias may result in this sector. In other words, the reliability of uncertainty analyses of biomass burning is in fact poor despite the statistical strength of data fitting. Given its high contributions to uncertainties in emission estimates, more field measurements of emissions from biomass burning in China are urgently needed.

For industrial combustion, the reliability of emission factors and sulfur/ash release ratios is relatively strong because there are more sampling data to draw from. The high emission uncertainties in these cases result mainly from poor source classification, not inadequate test data. To further reduce the uncertainties, more detailed technological categorization in the accounting of industrial boilers is thus suggested.

4.4 Emission uncertainty of power plants: comparison between sector and unit level

As described above, the uncertainty analyses in this study are based on sector (or subsector) estimates, i.e., the emission factors for all plants in a given emission source category are assumed to be identical for the Monte Carlo simulations. That assumption, however, is hardly realistic, as the complexity and diversity of emission characteristics for a given source category will lead to differences in practice. This approach might therefore overestimate somewhat the uncertainties of emissions.

Exploiting the database of coal-fired power plants established by the authors (Zhao et al., 2008), a unit-based approach to estimating the emission uncertainties of the power sector is developed, in which the uncertainties of activity levels and emission factors can be calculated unit-by-unit. In other words, independent and identical distributions are assumed for the same parameters of different units within a given category, and errors can then be efficiently compensated. It should be stressed that the unit-based method might underestimate the uncertainties of emissions because correlations of

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emission factors between the units of a given category should not be neglected. By comparing sector- and unit-based results, however, the differences between the two methods can be approximated.

In this study, such a test is conducted for one province in China as an example, Guizhou, where coal with extremely high sulfur content is used in power generation. As shown in Fig. 2, the 95% CIs using the sector-based method is largest for $PM_{2.5}$ (−30%∼108%) and smallest for NO_x (−25%∼20%). Compared to NO_x, the 95% CI of SO_2 emission is relatively large, reaching −40%∼42%, and the variation of sulfur contents is estimated to contribute almost 90% to that uncertainty. Employing a unit-based method, the 95% CIs for all species are significantly reduced, with the largest ranging −9%∼18%, for $PM_{2.5}$ and the smallest −8%∼8%, for SO_2. Figure 3 shows the difference in distributions of SO_2 emissions applying the two methods. The test confirms that the uncertainties of provincial and national emissions from power plants may be larger using a sector-based method compared to a unit-based one. Lacking detailed source information, however, such judgment cannot be easily extrapolated to other sectors without similar tests.

4.5 Comparisons with other studies

Figure 4 compares the resulting emission inventory uncertainties with those of other studies, none of which included Monte-Carlo simulation. The estimated uncertainty ranges for SO_2 and NO_x emissions are very similar among the available studies, while those of PM fractions of different sizes or carbonaceous species are significantly reduced. For example, the 95% CIs of $PM_{2.5}$, BC, and OC emissions are estimated to be −16∼52%, −23%∼130%, and −37%∼117% in this study, respectively, much lower than −57%∼130%, −68%∼208%, and −72%∼258% recently derived by Zhang et al. (2009d). The main reasons include: (1) the source categories in this study are more detailed, and random errors are thus significantly reduced due to the "compensation-of-error" mechanism (the same statistical mechanism described for the power sector comparison in Sect. 4.4); and (2) emission factors are mostly taken from the latest

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domestic measurements, eliminating variations introduced by emission characteristics derived from studies outside of China. It should be acknowledged that the former method can have its own weakness, when domestic field tests have limited sample sizes and/or source types compared to foreign studies, as discussed above regarding biomass burning.

Although inverse studies that evaluate emission inventories using observations and CTMs have indicated that Chinese NO_x emissions may be underestimated by around 50% (Wang et al., 2004; Ma et al., 2006), the uncertainty estimated in this study is relatively small, ranging only -10%~36%. The difference may result in part from omission in bottom-up inventories of anthropogenic NO_x sources of entirely different character, such as microbial decomposition of organic wastes associated with the human-animal food chain and applications of chemical fertilizer (Wang et al., 2004). Moreover, emissions vary considerably across regions of China (Wang et al., 2007; Zhao and Wang, 2009) and seasons (Zhang et al., 2007a), and emission uncertainties differentiated accordingly may be larger than national estimates. In order to compare better emission estimates derived from satellite, aircraft, or surface observations, therefore, further investigation of the uncertainties of bottom-up inventories by regions and seasons is needed. These would require region-specific uncertainty assumptions for emission factors and activity levels, and seasonal distribution parameters.

5 Conclusions

This study applies Monte Carlo simulation to quantify, for the first time, the uncertainties of a bottom-up emission inventory of China that is comprehensive in terms of key air pollutants, emitting sectors of the economy, and national scope. While providing current estimates of uncertainty to researchers investigating Chinese and global atmospheric transport and chemistry, this advance is nevertheless an incremental step, as it also illustrates the need for new investigations in diverse fields to narrow these uncertainties. In particular, the many parameter assumptions described in the study, par-

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ticularly outside of the coal-fired power sector, identify specific limitations of available literature and data concerning emission factors, activity levels, and even technology distributions in China. Improved quantification of emissions of the included species and other, closely associated ones – notably CO_2, generated largely by the same processes and thus subject to many of the same parameter uncertainties – is essential not only for science but for design of policies to redress critical atmospheric environmental hazards at local, regional, and global scales. Among these are photochemical smog, ecosystem acidification, and global climate change. Data collection and analyses of relevant parameters to narrow emission uncertainties, moreover, must be sustained, as these parameters are inevitably evolving as China's rapid growth restructures its economy, transforms its industries, and urbanizes its population.

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References

Akimoto, H., Ohara, T., Kurokawa, J., and Horii, N.: Verification of energy consumption in China during 1996–2003, Atmos. Environ., 40, 7664–7667, 2006.

Andreae, M. O. and Merlet, P.: Emission of trace gases and aerosols from biomass burning, Global Biogeochem. Cy., 15, 955–966, 2001.

Anshan Iron and Steel Group Corporation (AISGC): Description on developing emission standard of air pollutants for iron and steel industry-sintering, internal report (in Chinese), 2007.

Bond, T. C., Streets, D. G., Yarber, K. F., Nelson, S. M., Woo, J-H., and Klimont, Z.: A technology-based global inventory of black and organic carbon emissions from combustion, J. Geo. Res., 109, D14203, doi:10.1029/2003jd003697, 2004.

Carmichael, G. R., Tang, Y., Kurata, G., Uno, I., Streets, D., Woo, J.-H., Huang, H., Yienger, J., Lefer, B., Shetter, R., Blake, D., Atlas, E., Fried, A., Apel, E., Eisele, F., Cantrell, C., Avery, M., Barrick, J., Sachse, G., Brune, W., Sandholm, S., Kondo, Y., Singh, H., Talbot, R., Bandy, A., Thorton, D., Clarke, A., and Heikes, B.: Regional-scale chemical transport modeling in support of the analysis of observations obtained during the TRACE-P experiment, J. Geo. Res., 108(D21), 8823, doi:10.1029/2002jd003117, 2003a.

Carmichael, G. R., Tang, Y., Kurata, G., Uno, I., Streets, D. G., Thongboonchoo, N., Woo, J.-H., Guttikunda, S., White, A., Wang, T., Blake, D. R., Atlas, E., Fried, A., Potter, B., Avery, M. A., Sachse, G. W., Sandholm, S. T., Kondo, Y., Talbot, R. W., Bandy, A., Thorton, D., and Clarke, A. D.: Evaluating regional emission estimates using the TRACE-P observations, J. Geo. Res., 108(D21), 8810, doi:10.1029/2002jd003116, 2003b.

Cao, G. L., Zhang, X. Y., and Zheng, F. C.: Inventory of black carbon and organic carbon emissions from China, Atmos. Environ., 40, 6516–6527, 2006.

Chen, D., Wang, Y., McElroy, M. B., He, K., Yantosca, R. M., and Sager, P. L.: Regional CO pollution and export in China simulated by the high-resolution nested-grid GEOS-Chem model, Atmos. Chem. Phys., 9, 3825–3839, doi:10.5194/acp-9-3825-2009, 2009.

Chen, Y. J., Zhi, G. R., Feng, Y. L., Fu, J. M., Feng, J. L., Sheng, G. Y., and Simoneit, B. R. T.: Measurements of emission factors for primary carbonaceous particles from residential raw-coal combustion in China, Geophys. Res. Lett., 33, L20815, doi:10.1029/2006gl026966, 2006.

Chen, Y. J., Sheng, G. Y., Bi, X. H., Feng, Y. L., Mai, B. X., and Fu, J. M.: Emission factors for carbonaceous particles and polycyclic aromatic hydrocarbons from residential coal

combustion in China, Environ. Sci. Technol., 39, 1861–1867, 2005.
China Association of Automobile Manufacturers (CAAM) and China Automotive Technology & Research Center (CATRA): Evaluation report of automobile energy saving in China (in Chinese), available online at: http://caam.org.cn/files/file/0906/zhyh.pdf, 2009.
Chinese Research Academy of Environmental Sciences (CRAES), Description on developing emission standard of air pollutants for cement industry (in Chinese), internal report, Beijing, 2003.
Frey, H. C. and Zhao, Y. C.: Quantification of variability and uncertainty for air toxic emission inventories with censored emission factor data, Environ. Sci. Technol., 38, 6094–6100, 2004.
Frey, H. C. and Li, S.: Methods for quantifying variability and uncertainty in AP-42 emission factors: case studies for natural gas-fueled engines, J. Air Waste Manage. Assoc., 53, 1436–1447, 2003.
Frey, H. C. and Zheng, J. Y.: Quantification of variability and uncertainty in air pollutant emission inventories: method and case study for utility NO_x emissions, J. Air Waste Manage. Assoc., 52, 1083–1095, 2002.
Hao, J. M., Tian, H. Z., and Lu, Y. Q.: Emission inventories of NO_x from commercial energy consumption in China, 1995–1998, Environ. Sci. Technol., 36, 552–560, 2002.
He, K. B., Huo, H., Zhang, Q., He, D. Q., An, F., Wang, M., and Walsh, M. P.: Oil consumption and CO_2 emissions in China's road transport: current status, future trends, and policy implications, Energy Policy, 33, 1499–1507, 2005.
He, Y. J., Uno, I., Wang, Z. F., Ohara, T., Sugimoto, N., Shimizu, A., Richter, A., and Burrows, J. P.: Variations of the increasing trend of tropospheric NO_2 over central east China during the past decade, Atmos. Environ., 41, 4865–4876, 2007.
Huang, J. Y., and Xia, J. X.: General situation of industrial boiler industry in China and product trend (in Chinese), Energy Environ., 1, 21–23, 2004.
Karvosenoja, N., Tainio, M., Kupiainen, K., Tuomisto, J. T., Kukkonen, J., and Johansson, M.: Evaluation of the emissions and uncertainties of $PM_{2.5}$ originated from vehicular traffic and domestic wood combustion in Finland, Boreal Environ. Res., 13, 465–474, 2008.
Kean, A. J., Sawyer, R. F., and Harley, R. A.: A fuel-based assessment of off-road diesel engine emissions, J. Air Waste Manage. Assoc., 50, 1929–1939, 2000.
Kioutsioukis, I., Tarantola, S., Saltelli, A., and Debora, G.: Uncertainty and global sensitivity analysis of road transport emission estimates, Atmos. Environ., 38, 6609–6620, 2004.
Klimont, Z., Cofala, J., Bertok, I., Amann, M., Heyes, C., and Gyarfas, F.: Modeling particulate

ACPD
10, 29075–29111, 2010

emissions in Europe: A framework to estimate reduction potential and control costs, Interim report, IR-02-076, Institute for Applied Systems Analysis, Laxenburg, Austria, 2002.

Klimont, Z., Cofala, J., Xing, J., Wei, W., Zhang, C. Y., Wang, S. X., Jiang, K.J., Bhandari, P., Mathur, R., Purohit, P., Rafaj, P., Chambers, A., and Amann, M: Projections of SO_2, NO_x, and carbonaceous aerosols emissions in Asia, Tellus B., 61B, 602–617, 2009.

Lal, R.: World crop residues production and implications of its use as a biofuel, Environ. Int., 31, 575–584, 2005.

Lei, Y., Zhang, Q., He, K. B., and Streets, D. G.: Primary aerosol emission trends for China, 1990–2005, Atmos. Chem. Phys. Discuss., 10, 17153–17212, doi:10.5194/acpd-10-17153-2010, 2010a.

Lei, Y., Zhang, Q., Nielsen, C. P., and He, K. B.: An inventory of primary air pollutants and CO_2 emissions from cement industry in China, 1990–2020, Atmos. Environ., in preparation, 2010b.

Li, C., Li, X. H., Duan, L., Zhao, M., Duan, J. C., and Hao, J. M.: Emission characteristics of PM_{10} from coal-fired industrial boiler (in Chinese), Environ. Sci., 30, 650–655, 2009a.

Li, X. H., Wang, S. X., Duan, L., Hao, J. M., and Nie, Y. F.: Carbonaceous aerosol emissions from household biofuel combustion, Environ. Sci. Technol., 43, 6076–6081, 2009b.

Li, X. H., Duan, L., Wang, S. X., Duan, J. C., Guo, X. M., Yi, H. H., Hu, J. N., Li, C., and Hao, J. M.: Emission characteristics of particulate matter from rural household biofuel combustion in China, Energ. Fuel, 21, 845–851, 2007a.

Li, X. H., Wang, S. X., Duan, L., Hao, J. M., Li C., Chen, Y. S., and Yang, L.: Particulate and trace gas emissions from open burning of wheat straw and corn stover in China, Environ. Sci. Technol., 41, 6052-6058, 2007b.

Liu, H., He, K. B., Lents, J. M., Wang, Q. D., and Tolvett, S.: Characteristics of diesel truck emission in China based on portable emissions measurement systems, Environ. Sci. Technol., 43, 9507–9511, 2009.

Ma, J. Z., Richter, A., Burrows, J. P., Nuss, H., and van Aardenne, J. A.: Comparison of model-simulated tropospheric NO_2 over China with GOME-satellite data, Atmos. Environ., 40, 593–604, 2006.

Ohara, T., Akimoto, H., Kurokawa, K., Horii, N., Yamaji, K., Yan, X., and Hayasaka, T.: An Asian emission inventory of anthropogenic emission sources for the period 1980–2020, Atmos. Chem. Phys., 7, 4419–4444, doi:10.5194/acp-7-4419-2007, 2007.

Qu, L. F.: Economical operation analysis of industrial boilers in China 2007 (in Chinese), China

Electrical Equipment Industry, 6, 24–29, 2008.
Saikawa, E., Naik, V., Horowitz, L. W., Liu, J. F., and Mauzerall, D. L.: Present and potential future contributions of sulfate, black and organic carbon aerosols from China to global air quality, premature mortality and radiative forcing, Atmos. Environ., 43, 2814–2822, 2009.
State Environmental Protection Administration (SEPA): Handbook of Industrial Pollution Emission Factors (in Chinese), China Environmental Science Press, Beijing, 1996.
Streets, D. G., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, Q., He, D., Klimont, Z., Nelson, S. M., Tsai, N. Y., Wang, M. Q., Woo, J. H., and Yarber, K. F.: An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, J. Geosci. Res., 108(D21), 8809, doi:10.1029/2002jd003093, 2003.
Streets, D. G., Gupta, S., Waldhoff, S. T., Wang, M. Q., Bond, T. C., and Bo, Y. Y.: Black carbon emissions in China, Atmos. Environ., 35, 4281–4296, 2001.
US Environmental Protection Agency (USEPA): Compilation of Air Pollutant Emission Factors, available online at: http://www.epa.gov/ttn/chief/ap42/index.html, 2002. .
Wang, S. X. and Zhang, C. Y.: Spatial and temporal distribution of air pollutant emission from open burning of crop residues in China (in Chinese), Sciencepaper online, 3, 329–333, 2008.
Wang, Y. X., McElroy, M. B., Wang, T., and Palmer, P. I.: Asian emissions of CO and NO_x: Constraints from aircraft and Chinese station data, J. Geo. Res., 109, D24304, doi:10.1029/2004jd005250, 2004.
Wang, M., Huo, H., Johnson, L., and He, D. Q.: Projection of Chinese motor vehicle growth, oil demand, and CO_2 emissions through 2050, Argon National Laboratory, ANL/ESD/06-6, 2006.
Wang, Y. X., McElroy, M. B., Martin, R. V., Streets, D. G., Zhang, Q., and Fu, T. M.: Seasonal variability of NO_x emissions over east China constrained by satellite observations: Implications for combustion and microbial sources, J. Geo. Res., 112, D06301, doi:10.1029/2006jd007538, 2007.
Wang, H. K., Chen, C. H., Huang, C., and Fu, L. X.: On-road vehicle emission inventory and its uncertainty analysis for Shanghai, China, Sci. Total. Environ., 398, 60–67, 2008.
Wang, S. X., Zhao, X. J., Li, X. H., Wei, W., and Hao, J. M.: Emission characteristics of fine particles from grate firing boilers (in Chinese), Environ. Sci., 30, 963–968, 2009..
Wu, Y., Streets, D. G., Wang, S. X., and Hao, J. M.: Uncertainties in estimating mercury emissions from coal-fired power plants in China, Atmos. Chem. Phys., 10, 2937–2947,

ACPD
10, 29075–29111, 2010

doi:10.5194/acp-10-2937-2010, 2010.
Xu, Y., Williams, R. H., and Socolow, R. H.: China's rapid deployment of SO_2 scrubbers, Energy Environ. Sci., 2, 459–465, 2009.
Zhang, J. F. and Smith, K. R.: Greenhouse gases and other airborne pollutants from household stoves in China: a database for emission factors, Atmos. Environ., 34, 4537-4549, 2000.
Zhang, M. G., Uno, I., Yoshida, Y., Xu, Y. F., Wang, Z. F., Akimoto, H., Bates, T., Quinn, T., Bandy, A., and Blomquist, B.: Transport and transformation of sulfur compounds over East Asia during the TRACE-P and ACE-Asia campaigns, Atmos. Environ., 38, 6947–6959, 2004.
Zhang, Q., Streets, D. G., He, K., Wang, Y., Richter, A., Burrows, J. P., Uno, I., Jang, C. J., Chen, D., Yao, Z., and Lei, Y.: NOx emission trends for China, 1995-2004: The view from the ground and the view from space, J. Geo. Res., 112, D22306, doi:10.1029/2007jd008684, 2007a.
Zhang, Q., Streets, D. G., He, K. B., and Klimont, Z.: Major components of China's anthropogenic primary particulate emissions, Environ. Res. Lett., 2, 045027, doi:10.1088/1748-9326/2/4/045027, 2007b.
Zhang, C. Y., Wang, S. X., Xing, J., Zhao, Y., and Hao, J. M.: Current status and future projections of NO_x emissions from energy related industries in China (in Chinese), Acta Scientiae Circumstantiae, 28, 2470–2479, 2008a.
Zhang, Y. X., Schauer, J. J., Zhang, Y. H., Zeng, L. M., Wei, Y. J., Liu, Y., and Shao, M.: Characteristics of particulate carbon emissions from real-world Chinese coal combustion, Environ. Sci. Technol., 42, 5068-5073, 2008b.
Zhang, C. Y., Wang, S. X., Zhao, Y., and Hao, J. M.: Current status and future prospects of anthropogenic particulate matter emissions in China (in Chinese), Environ. Sci., 30, 1881–1887, 2009a.
Zhang, J., He, K. B., Shi, X. Y., and Zhao, Y.: Effect of SME biodiesel blends on $PM_{2.5}$ emission from a heavy-duty engine, Atmos. Environ., 43, 2442–2448, 2009b.
Zhang, J., He, K. B., Ge, Y. S., Shi, X. Y.: Influence of fuel sulfur on the characterization of PM_{10} from a diesel engine, Fuel, 88, 504–510, 2009c.
Zhang, Q., Streets, D. G., Carmichael, G. R., He, K., Huo, H., Kannari, A., Klimont, Z., Park, I., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, L., and Yao, Z.: Asian emissions in 2006 for the NASA INTEX-B mission, Atmos. Chem. Phys., 9, 5131–5153, doi:10.5194/acp-9-5131-2009, 2009d.
Zhao, C. and Wang, Y. H.: Assimilated inversion of NO_x emissions over east Asia using OMI

ACPD
10, 29075–29111, 2010

NO$_2$ column measurements, Geophys. Res. Lett., 36, L06805, doi:10.1029/2008gl037123, 2009.
Zhao, Y., Wang, S. X., Duan, L., Lei, Y., Cao, P. F., and Hao, J. M.: Primary air pollutant emissions of coal-fired power plants in China: current status and future prediction. Atmos. Environ., 42, 8442–8452, 2008.
Zhao, Y., Duan, L., Xing, J., Larssen, T., Nielsen, C. P., and Hao, J. M.: Soil acidification in China: Is controlling SO$_2$ emissions enough, Environ. Sci. Technol., 43, 8021-8026, 2009.
Zhao, Y., Wang, S. X., Nielsen, C. P., Li, X. H., and Hao, J. M.: Establishment of a database of emission factors for atmospheric pollutants from Chinese coal-fired power plants, Atmos. Environ., 44, 1515–1523, 2010.
Zheng, J. Y., Zhang, L. J., Che, W. W., Zheng, Z, Y., Yin, S. S.: A highly resolved temporal and spatial air pollutant emission inventory for the Pearl River Delta region, China and its uncertainty assessment, Atmos. Environ., 43, 5112–5122, 2009.

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Table 1. Uncertainties of Chinese emissions by sector in 2005. The estimated emissions are expressed as kilo metric tons (kt). The percentages in the parentheses indicate the 95% CI around the central estimate.

	Power plants	Total industry	Transportation	Residential	Total
SO_2	16258 (−16%, 20%)	11522 (−24%, 17%)	241 (−21%, 42%)	3064 (−47%, 24%)	31085 (−14%, 12%)
NO_x	6730 (−19%, 16%)	6296 (−29%, 93%)	4724 (−20%, 47%)	2035 (−31%, 91%)	19785 (−10%, 36%)
PM	2768 (−19%, 39%)	25803 (−14%, 42%)	590 (−31%, 45%)	5498 (−44%, 77%)	34659 (−10%, 36%)
PM_{10}	1859 (−19%, 49%)	11833 (−17%, 54%)	577 (−31%, 46%)	4945 (−45%, 84%)	19214 (−12%, 42%)
$PM_{2.5}$	912 (−27%, 80%)	6925 (−20%, 69%)	552 (−32%, 48%)	4711 (−46%, 86%)	13100 (−16%, 52%)
BC	16 (−69%, 378%)	602 (−29%, 79%)	238 (−74%, 74%)	841 (−44%, 238%)	1698 (−23%, 130%)
OC	2 (−73%, 2367%)	571 (−23%, 87%)	102 (−68%, 86%)	2528 (−51%, 138%)	3203 (−37%, 117%)

Table 2. The parameters contributing most to emission uncertainties, by sector and species. The percentages in the parentheses indicate the contributions of the parameters to the variance of corresponding emissions (see Sect. 2.1 for the abbreviations of parameters).

	Power plants	Total industry	Transportation	Residential
SO_2	*SC*: Guizhou (14%) *SC*: Shandong (11%) *SC*: Shanxi (11%)	*SR* (grate) (37%) $\eta_{SO_2 other-FGD}$ (13%) EF_{SO_2} (sintering) (11%)	*SR* (non-road coal) (47%) *AL* (non-road coal) (31%) *SC* (gasoline) (5%)	EF_{SO_2} (hot water system) (77%) *AL* (coal) (4%) EF_{SO_2} (biomass open burning) (3%)
NO_x	EF_{NO_x} (non-LNB, bituminous) (53%) EF_{NO_x} (tangential, bituminous) (14%) EF_{NO_x} (non-LNB, anthracite) (12%)	EF_{NO_x} (grate) (86%) EF_{NO_x} (precalciner) (3%) EF_{NO_x} (oil) (1%)	EF_{NO_x} (shipping) (39%) *AL* (on-road diesel) (10%) EF_{NO_x} (tractor) (7%)	EF_{NO_x} (biofuel-waste) (38%) EF_{NOx} (biomass open burning) (28%) EF_{NO_x} (biofuel-wood) (7%)
PM	$f_{PM_{2.5}}$ (pulverized) (19%) *AR* (pulverized) (14%) $\eta_{PM_{2.5},ESP}$ (10%)	EF_{PM} (cement process) (16%) *AR* (grate) (9%) EF_{PM} (brick) (9%)	$EF_{PM_{2.5}}$ (tractor) (34%) $EF_{PM_{2.5}}$ (rural machine) (22%) $EF_{PM_{2.5}}$ (construction machine) (9%)	$EF_{PM_{2.5}}$ (biofuel-waste) (37%) EF_{PM} (small coal stove) (26%) $EF_{PM_{2.5}}$ (biomass open burning) (14%)
PM_{10}	$f_{PM_{2.5}}$ (pulverized) (33%) $\eta_{PM2.5-10,ESP}$ (10%) *AR* (pulverized) (9%)	$f_{PM_{2.5}}$ (cement process) (21%) EF_{PM} (cement process) (18%) *AR* (grate) (9%)	$EF_{PM_{2.5}}$ (tractor) (34%) $EF_{PM_{2.5}}$ (rural machine) (23%) $EF_{PM_{2.5}}$ (construction machine) (9%)	$EF_{PM_{2.5}}$ (biofuel-waste) (37%) EF_{PM} (small coal stove) (27%) $EF_{PM_{2.5}}$ (biomass open burning) (14%)
$PM_{2.5}$	$f_{PM_{2.5}}$ (pulverized) (55%) $\eta_{PM2.5,WET}$ (12%) $\eta_{PM2.5,ESP}$ (10%)	$f_{PM_{2.5}}$ (cement process) (39%) $f_{PM_{2.5}}$ (grate) (13%) *AR* (grate) (9%)	$EF_{PM_{2.5}}$ (tractor) (35%) $EF_{PM_{2.5}}$ (rural machine) (22%) $EF_{PM_{2.5}}$ (construction machine) (9%)	$EF_{PM_{2.5}}$ (biofuel-waste) (37%) EF_{PM} (small coal stove) (27%) $EF_{PM_{2.5}}$ (biomass open burning) (14%)
BC	F_{BC} (pulverized) (46%) F_{BC} (grate) (22%) $f_{PM_{2.5}}$ (grate) (12%)	F_{BC} (grate) (25%) $f_{PM_{2.5}}$ (grate) (15%) *EF* (brick) (10%)	F_{BC} (non-road) (74%) $EF_{PM_{2.5}}$ (tractor) (7%) $EF_{PM_{2.5}}$ (rural machine) (4%)	EF_{BC} (small coal stove) (58%) EF_{BC} (biofuel-wood) (23%) EF_{BC} (biofuel-waste) (8%)
OC	F_{OC} (grate) (46%) $f_{PM_{2.5}}$ (grate) (28%) *AR* (grate) (20%)	F_{OC} (grate) (26%) $f_{PM_{2.5}}$ (grate) (18%) *AR* (grate) (11%)	F_{OC} (non-road) (58%) F_{OC} (on-road, diesel) (16%) $EF_{PM_{2.5}}$ (tractor) (6%)	EF_{OC} (biofuel-waste) (47%) EF_{OC} (small coal stove) (34%) EF_{OC} (biomass open burning) (8%)

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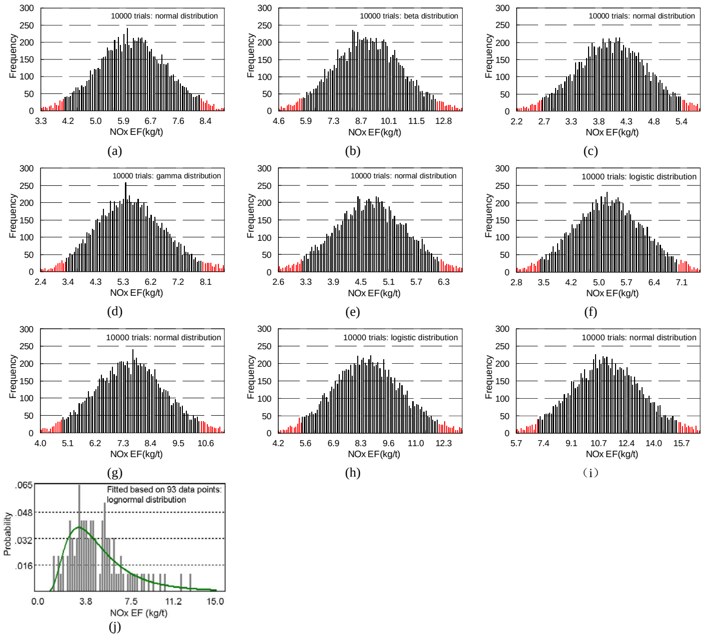

Fig. 1. The distributions of NO_x emission factors for coal-fired power plants and industrial boilers. The red bars are beyond the 95% CIs. The figures represent power generating units: **(a)** without LNB and bituminous combustion (<300 MW); **(b)** without LNB and anthracite combustion (<300MW); **(c)** with LNB and bituminous combustion (<300 MW); **(d)** with LNB and anthracite combustion (<300MW); **(e)** with tangential burner and bituminous combustion (≥300 MW); **(f)** with wall-fired burner and bituminous combustion (≥300 MW); **(g)** with tangential burner and anthracite combustion (≥300 MW); **(h)** with wall-fired burner and anthracite combustion (≥300 MW); **(i)** with W-flame burner and anthracite combustion (≥300MW). The last figure **(j)** represents industrial grate boilers.

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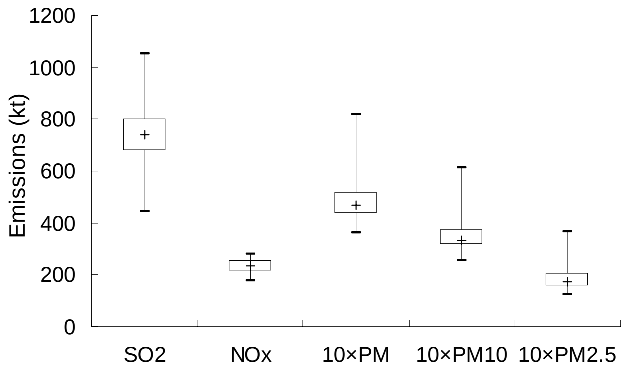

Fig. 2. Comparisons of emission uncertainties of coal-fired power plants in Guizhou in 2005 using sector-based and unit-based methods. The "+" represents the mean value and the box represents the 95% CI using the unit-based method, while the larger range line represents the 95% CI using the sector-based method.

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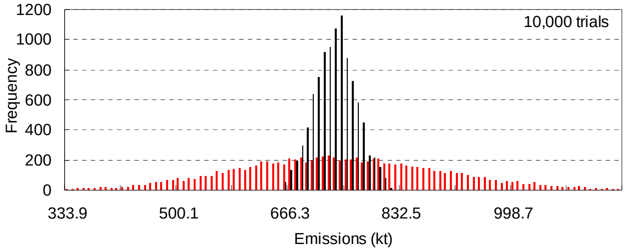

Fig. 3. The distributions of estimated SO_2 emissions from coal-fired power plants in Guizhou in 2005, using Monte-Carlo simulation. The black and red bars represent the results from unit-based and sector-based methods, respectively.

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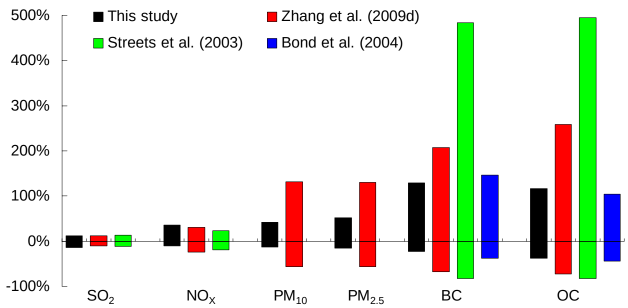

Fig. 4. Comparisons of emission uncertainties for different species in China from different studies, expressed as 95% CIs around the central estimates.

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