Quantifying the Uncertainties of a Bottom-Up Emission Inventory of Anthropogenic Atmospheric Pollutants in China

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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 $-14\%\sim13\%$, $-13\%\sim37\%$, $-11\%\sim38\%$, $-14\%\sim45\%$, $-17\%\sim54\%$, $-25\%\sim136\%$, and $-40\%\sim121\%$, 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., 2011a). 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 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 U.S. emission factors (USEPA, 2002). They estimated that the uncertainties of Chinese emissions varied from $-12\% \sim 13\%$ for SO_2 to $-83\% \sim 495\%$ 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 analysis 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 $-37\% \sim 147\%$ and $-44\% \sim 104\%$, respectively. Wu et al. (2010) focused on

mercury emissions from coal-fired power plants in China and estimated an 80% CI in 2003 of $-37\% \sim 71\%$.

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 analysis, reliability and limitations of the analysis, and comparisons with other studies. Section 5 summarizes the present study.

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 described in details 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 Supplement for more details).

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

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

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):

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

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):

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

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 consumption during 1996–2003 was probably underestimated and thereby not recommended for use in the study of emission inventories during this period. However, Wu et al. (2010) found that the discrepancies in energy data from power sector were relatively small (e.g., less than ±5% for coal consumption) 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 coal consumption by power sector. For industrial energy consumption, larger inconsistencies (~20%) have been found between official statistics at provincial and national levels (Zhang et al., 2007a). Similarly, an uncertainty of 15–20% has been suggested for industrial fuel use in countries with less developed statistical systems (IPCC, 2006). Therefore, we assume normal distributions with CV of 10% for activity levels of industrial combustion (i.e., the 95% CI is ±19.6% around the central estimate). 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. Even larger uncertainties are believed to exist regarding residential fossil fuel use, attributed to poor statistics on fuel combustion by household stoves in rural areas, and normal distributions with CV of 20% are assumed, as suggested by IPCC (2006). All of these assumptions of distributions are thus based on expert judgment.

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

$$AL_j = \sum_t VP_{j,t} \times VMT_{j,t} \times FE_{j,t} \quad (4)$$

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). 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 a normal distribution with CV of 5% for the VMT. Regarding fuel economy (FE), the China Association of Automobile Manufacturers and China Automotive Technology and 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%.

In principle it would be valuable to compare the official statistics and the calculated fuel consumption in this study. However, Chinese official energy statistics classify fuel consumption for a sector that combines transportation, warehousing, and postal service, and there is no basis available to disaggregate these activities to match activity levels regarding vehicles in the emission inventory framework. Moreover, the statistics for the sector only reflects fuel used in commercial activities. Therefore direct comparisons cannot be made between official and estimated fuel consumption by transportation. Wang et al. (2006) estimated that the oil consumption by vehicles (including rural vehicles) in 2005 is 108.6 million metric tons (Mt), close to our estimate of 101.1 Mt. The discrepancy is within the uncertainty range estimated in this study.

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 30%, in accordance with the suggestion by IPCC (2006). 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 30%. 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.

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 Supplement.

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. Such information at provincial level is summarized in Table S3.

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., 2011b). 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 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. (2011a, 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 1999 and 2003, respectively. Specifically Beijing implemented those standards in advance of other provinces. The fleet compositions of different control levels by vehicle type are calculated based on an average vehicle age of 15 years and are shown in Table S2. 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.

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 S4 of the Supplement.

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 ($kg\,t^{-1}$), and the probability is fitted to a lognormal distribution with geometric standard deviation (GSD) of $1.8\,kg\,t^{-1}$ based on 93 data points ranging 1.1–$24.5\,kg\,t^{-1}$ in tests by SEPA (1996). The mean value is comparable to the result from a US study, 2.8–$5.5\,kg\,t^{-1}$ (USEPA, 2002), but the quite long tail of the distribution 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%, $1.8\,kg\,t^{-1}$, and $1.9\,kg\,t^{-1}$, 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–$3.9\,kg\,t^{-1}$, with a mean of $0.9\,kg\,t^{-1}$. 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 $11\,kg\,t^{-1}$, 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 S4, 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. Median values of those tests
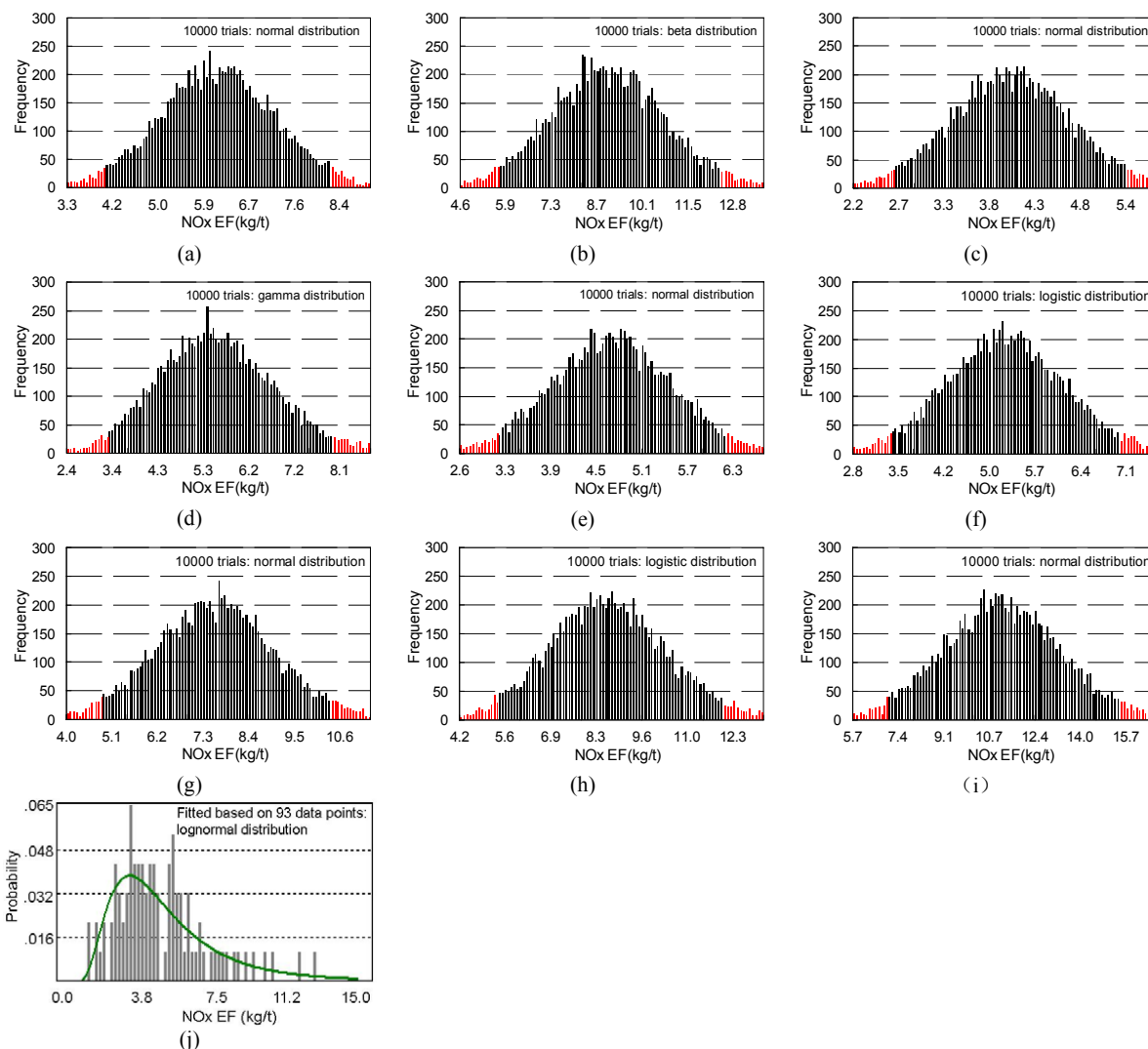
Fig. 1. The distributions of NO_x emission factors for coal-fired power plants and industrial boilers. The red bars are beyond the 95% CIs. The figures represent power generating units: **(a)** without LNB and bituminous combustion (<300 MW); **(b)** without LNB and anthracite combustion (<300MW); **(c)** 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 (≥300 MW). The last figure **(j)** represents industrial grate boilers.

are used as the central values of the distributions. 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 Environm. Sci.s (CRAES) conducted national surveys and measured NO_x emission factors of 20 kilns with different technolo-

gies (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 S4. The distributions of PM emission factors are fitted by technology based on field tests (SEPA, 1996) and thorough investigation by Lei et al. (2011b).

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 $2.7\,\mathrm{kg\,t^{-1}}$ of product with lognormal distribution (GSD: 1.5) (SEPA, 1996), and for NO_x it is $0.64\,\mathrm{kg\,t^{-1}}$, 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. (2011a) 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 from the database of SEPA (1996), as summarized in Table S4.

The unabated emission factors from SEPA (1996) are applied to certain industrial sub-sectors, for which more recent field measurements are lacking. It is acknowledged that this assumption may introduce additional uncertainties, and thus relatively large uncertainty ranges are applied in the analysis. Absent further evidence on emission factors from new field tests, however, we believe that this assumption is acceptable for two reasons. First, our assessment year is 2005 and China's major "energy saving and emission reduction" policies began in 2006 as part of the 11th Five Year Plan. Prior to the initiation of these policies, we think that the unabated emission factors for these industrial sources improved only slightly. New plants with advanced technologies successfully reduced emissions prior to 2006, but this was mainly due to improved removal efficiencies in pollution control, not to advances in the combustion processes of given technology types. Second, in any case, the removal efficiencies of control technologies are stronger determinants of emission levels than the unabated emission factors. As described in Sect. 3.3, those removal efficiencies are mostly taken from recent measurements by the authors and other researchers, and thus are assumed to be more up-to-date and reliable.

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 S5 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 on-road vehicle types and assume that CVs of pre-stage I vehicles are as large as those of stage I. For non-road sources, the uncertainties of emission factors are expected to be larger, with few direct measurements in China to date. The lifetimes of non-road sources are usually longer than those of on-road ones, and emission factors subsequently increase as vehicle performance deteriorates over time. In this study, a CV of 100% is tentatively applied for emission factors of non-road sources. 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 S6 in 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. Median values of those tests are use as the central values of the distribution. 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.

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%).

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	16 258 (−16%, 21%)	11 522 (−24%, 21%)	241 (−21%, 43%)	3064 (−53%, 43%)	31 085 (−14%, 13%)
NO_x	6730 (−19%, 16%)	6296 (−31%, 96%)	4724 (−29%, 70%)	2035 (−34%, 96%)	19 785 (−13%, 37%)
PM	2768 (−18%, 38%)	25 816 (−14%, 45%)	590 (−35%, 59%)	5498 (−48%, 83%)	34 672 (−11%, 38%)
PM_{10}	1859 (−19%, 49%)	11 845 (−18%, 57%)	577 (−35%, 61%)	4958 (−50%, 89%)	19 239 (−14%, 45%)
$PM_{2.5}$	912 (−26%, 80%)	6936 (−21%, 72%)	552 (−36%, 63%)	4711 (−50%, 91%)	13 111 (−17%, 54%)
BC	16 (−68%, 379%)	602 (−37%, 86%)	238 (−75%, 89%)	841 (−47%, 259%)	1697 (−25%, 136%)
OC	2 (−72%, 2307%)	571 (−31%, 95%)	102 (−68%, 98%)	2528 (−54%, 148%)	3203 (−40%, 121%)

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 S7 in the Supplement). 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.

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 S8 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%~13%, −13%~37%, −11%~38%, −14%~45%, −17%~54%, −25%~136%, and −40%~121% 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 carbona-

ceous 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 −31%~96%, with values reaching −50%~159% for industrial boilers. Due to poor resolution of the data with respect to technology and fuel type, 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%~103% due to variations of emission factors of 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 S8 in the Supplement) 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 S8 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 −56%~132%, much larger than that of total PRO, −40%~28%).

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 S8 in the Supplement for details). Some explanations of

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) (28%) $\eta_{SO_2,other-FGD}$ (9%) EF_{SO_2} (sintering) (9%)	SR (non-road coal) (47%) AL (non-road coal) (32%) SC (gasoline) (4%)	EF_{SO_2}(hot water system) (46%) AL (coal) (41%) EF_{SO_2} (biomass open burning) (2%)
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) (84%) AL (coal) (3%) EF_{NO_x} (precalciner) (2%)	EF_{NO_x} (shipping) (45%) EF_{NO_x} (tractor) (11%) EF_{NO_x} (rural machine) (8%)	EF_{NO_x} (biofuel-waste) (32%) EF_{NO_x} (biomass open burning) (24%) AL (biofuel-waste) (8%)
PM	$f_{PM_{2.5}}$ (pulverized) (19%) AR (pulverized) (14%) $\eta_{PM_{2.5},ESP}$ (10%)	EF_{PM} (cement process) (14%) AR (grate) (9%) EF_{PM} (brick) (8%)	$EF_{PM_{2.5}}$ (tractor) (36%) $EF_{PM_{2.5}}$ (rural machine) (23%) $EF_{PM_{2.5}}$ (construction machine) (9%)	$EF_{PM_{2.5}}$ (biofuel-waste) (33%) EF_{PM}(small coal stove) (24%) $EF_{PM_{2.5}}$ (biomass open burning) (11%)
PM_{10}	$f_{PM_{2.5}}$ (pulverized) (33%) $\eta_{PM2.5-10,ESP}$ (10%) AR (pulverized) (9%)	$f_{PM_{2.5}}$ (cement process) (19%) EF_{PM} (cement process) (16%) AR (grate) (9%)	$EF_{PM_{2.5}}$ (tractor) (36%) $EF_{PM_{2.5}}$ (rural machine) (23%) $EF_{PM_{2.5}}$ (construction machine) (9%)	$EF_{PM_{2.5}}$ (biofuel-waste) (33%) EF_{PM} (small coal stove) (26%) $EF_{PM_{2.5}}$ (biomass open burning) (11%)
$PM_{2.5}$	$f_{PM_{2.5}}$ (pulverized) (55%) $\eta_{PM_{2.5},WET}$ (12%) $\eta_{PM_{2.5},ESP}$ (10%)	$f_{PM_{2.5}}$ (cement process) (36%) $f_{PM_{2.5}}$ (grate) (13%) AR (grate) (9)	$EF_{PM_{2.5}}$ (tractor) (37%) $EF_{PM_{2.5}}$ (rural machine) (24%) $EF_{PM_{2.5}}$ (construction machine) (10%)	$EF_{PM_{2.5}}$ (biofuel-waste) (33%) EF_{PM}(small coal stove) (26%) $EF_{PM_{2.5}}$ (biomass open burning) (11%)
BC	F_{BC} (pulverized) (46%) F_{BC} (grate) (22%) $f_{PM_{2.5}}$ (grate) (12%)	AL (coke) (20%) F_{BC} (grate) (17%) $f_{PM_{2.5}}$ (grate) (9%)	F_{BC}(non-road) (67%) $EF_{PM_{2.5}}$ (tractor) (9%) $EF_{PM_{2.5}}$ (rural machine) (5%)	EF_{BC}(small coal stove) (53%) EF_{BC} (biofuel-wood) (22%) EF_{BC} (biofuel-waste) (8%)
OC	F_{OC} (grate) (46%) $f_{PM_{2.5}}$ (grate) (28%) AR (grate) (20%)	F_{OC} (grate) (19%) AL (coke) (17%) $f_{PM_{2.5}}$ (grate) (16%)	F_{OC}(non-road) (55%) F_{OC}(on-road, diesel) (16%) $EF_{PM_{2.5}}$ (tractor) (7%)	EF_{OC} (biofuel-waste) (39%) EF_{OC}(small coal stove) (30%) EF_{OC} (biomass open burning) (8%)

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, b, 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, b, 2009b) fully, lacking further domestic evidence. Given potential high uncertaities, 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, b, 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 changes from $-58 \sim 216\%$ to $-31\% \sim 311\%$, suggesting caution in interpretation of results based on limited domestic field measurements (see also the discussion in Sect. 4.3).

4.2 Sensitivity analysis

In addition to estimating uncertainty, sensitivity analysis 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 2006 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 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 84% to the variance of emissions. Emissions of PM of different sizes are most sensitive to the unabated PM emission factor and $PM_{2.5}$ mass fraction of cement processing (i.e., not combustion but cooling, grinding, and crushing). This result spotlights the complexity of the emission characteristics of the cement industry. Even after the most thorough evaluation to date (Lei et al., 2011b), there are still large uncertainties in the PM emissions from cement, which contribute significantly to the variation of total industry PM emissions.

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 subjective judgments (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 increases from 12% 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–S7 in the Supplement, 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 part thereby reflect decreasing reliability of the uncertainty analysis. (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 analysis 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 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 certain industrial sources like cement 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 analysis and reduce the uncertainties of Chinese emission inventories, new research on such parameters with relatively low reliability but high contributions to emission uncertainties 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 control levels. The uncertainty analysis 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 analysis 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 analysis in this study are based on sector (or sub-sector) 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
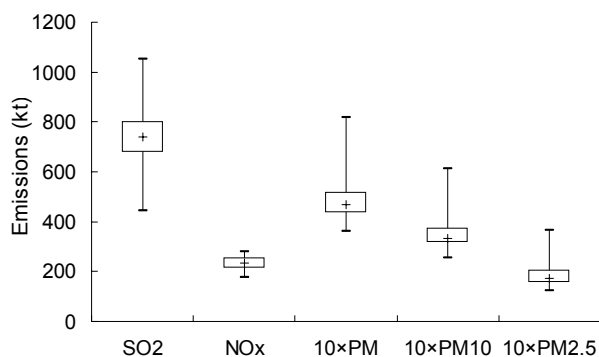
Fig. 2. Comparisons of emission uncertainties of coal-fired power plants in Guizhou in 2005 using sector-based and unit-based methods. The "+" represents the central value and the box represents the 95% CI using the unit-based method, while the larger range line represents the 95% CI using the sector-based method.

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 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 $PM_{2.5}$ ($-30\% \sim 108\%$) and smallest for NO_x ($-25\% \sim 20\%$). Compared to NO_x, the 95% CI of SO_2 emission is relatively large, reaching $-40\% \sim 42\%$, 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 $-9\% \sim 18\%$, for $PM_{2.5}$ and the smallest $-8\% \sim 8\%$, 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 central estimates of Chinese national emissions by this work with those of other studies. It can be seen that most estimates are within the 95% CIs calculated in the current study, except for SO_2, NO_x and BC emissions by Ohara et al. (2007). Interannual variability can partly explain the lower estimates of NO_x and BC by Ohara et al. (2007), which provided an emission inventory
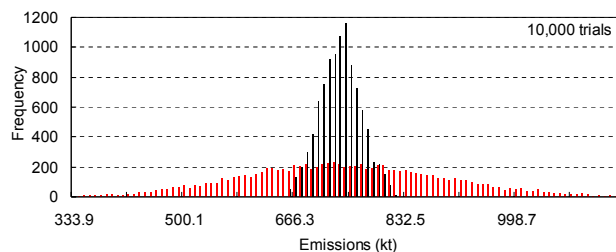

Fig. 3. The distributions of estimated SO_2 emissions from coal-fired power plants in Guizhou in 2005, using Monte Carlo simulation. The black and red bars represent the results from unit-based and sector-based methods, respectively.
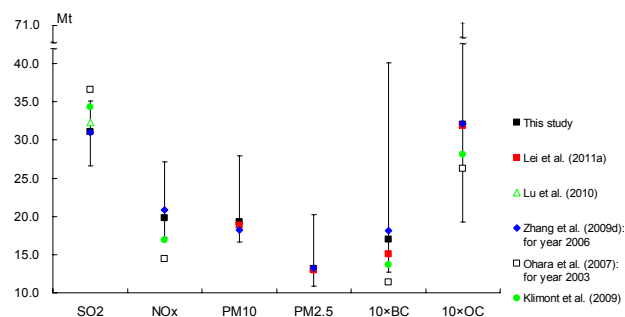
Fig. 4. Comparisons of annual emissions for China from different studies. The emissions are for year 2005 unless noted. The range line represents the 95% CIs around the central estimates of this study.

for 2003 instead of 2005. Moreover, open burning of biomass was not included in Ohara et al. (2007). Regarding SO_2, Ohara et al. (2007) estimated a total emission of 36.6 Mt for China in 2003, much higher than all other estimates for 2005 and later. This large discrepancy may come mainly from the difference between the updated domestic emission factors used in this study, and the foreign emission factor database that Ohara et al. (2007) relied more upon. Figure 5 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 $-17 \sim 54\%$, $-25\% \sim 136\%$, and $-40\% \sim 121\%$ in this study, respectively, much lower than $-57\% \sim 130\%$, $-68\% \sim 208\%$, and $-72\% \sim 258\%$ 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
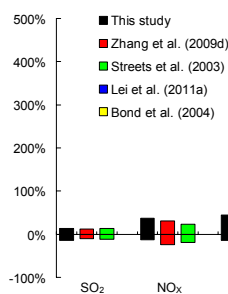
Fig. 5. Comparisons of emission uncertainties for different species in China from different studies, expressed as 95% CIs around the central estimates.

mostly taken from the latest 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 $-13\%\sim37\%$. 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, particularly 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. Future regional differentiation of those input parameters will further reduce uncertainties. 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 analysis 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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