Atmospheric Environment 165 (2017) 207–221

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Quantifying the uncertainties of China's emission inventory for industrial sources: From national to provincial and city scales

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HIGHLIGHTS

- Uncertainty of industrial emission inventories was analyzed at varied spatial scales.
- Uncertainty of total emissions was not largely reduced at small scale except for SO_2.
- Plant-specific data reduced uncertainty of emissions for power and industry boilers.
- PM emissions at provincial level were estimated larger than those at national level.

ARTICLE INFO

Article history:
Received 10 May 2017
Received in revised form
20 June 2017
Accepted 27 June 2017
Available online 29 June 2017

Keywords:
Uncertainty
Multi-scale emission inventories
Emission factor
Air quality
China

ABSTRACT

A comprehensive uncertainty analysis was conducted on emission inventories for industrial sources at national (China), provincial (Jiangsu), and city (Nanjing) scales for 2012. Based on various methods and data sources, Monte-Carlo simulation was applied at sector level for national inventory, and at plant level (whenever possible) for provincial and city inventories. The uncertainties of national inventory were estimated at −17–37% (expressed as 95% confidence intervals, CIs), −21–35%, −19–34%, −29–40%, −22–47%, −21–54%, −33–84%, and −32–92% for SO_2, NO_X, CO, TSP (total suspended particles), PM_{10}, $PM_{2.5}$, black carbon (BC), and organic carbon (OC) emissions respectively for the whole country. At provincial and city levels, the uncertainties of corresponding pollutant emissions were estimated at −15–18%, −18–33%, −16–37%, −20–30%, −23–45%, −26–50%, −33–79%, and −33–71% for Jiangsu, and −17–22%, −10–33%, −23–75%, −19–36%, −23–41%, −28–48%, −45–82%, and −34–96% for Nanjing, respectively. Emission factors (or associated parameters) were identified as the biggest contributors to the uncertainties of emissions for most source categories except iron & steel production in the national inventory. Compared to national one, uncertainties of total emissions in the provincial and city-scale inventories were not significantly reduced for most species with an exception of SO_2. For power and other industrial boilers, the uncertainties were reduced, and the plant-specific parameters played more important roles to the uncertainties. Much larger PM_{10} and $PM_{2.5}$ emissions for Jiangsu were estimated in this provincial inventory than other studies, implying the big discrepancies on data sources of emission factors and activity data between local and national inventories. Although the uncertainty analysis of bottom-up emission inventories at national and local scales partly supported the "top-down" estimates using observation and/or chemistry transport models, detailed investigations and field measurements were recommended for further improving the emission estimates and reducing the uncertainty of inventories at local and regional scales, for both industrial and other sectors.

1. Introduction

Emission inventories are fundamental for atmospheric science research with chemical transport modeling (CTM) and for policy

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http://dx.doi.org/10.1016/j.atmosenv.2017.06.045
1352-2310/

making in pollution mitigation. Recently the focus on China's emissions is in particular rising, as the country contains a wide variety of emissions sources, relatively large emission intensities, and fast changes in temporal and spatial patterns of emissions, attributed to the swift growth of economy and to the multiple measures of emission controls. At continental/national scale, various inventories have been developed including the one for Intercontinental Chemical Transport Experiment-Phase B (INTEX-B, Zhang et al., 2009), the Emissions Database for Global Atmospheric Research (EDGAR, JRC/PBL, 2011), the Regional Emission inventory in Asia (REAS, Ohara et al., 2007; Kurokawa et al., 2013), and the Multi-resolution Emission Inventory for China (MEIC, http://www.meicmodel.org/). Resulting from different methods and data sources, discrepancies existed between inventories and the model performances varied significantly when they were applied in CTM (Saikawa et al., 2017), motivating the uncertainty analysis of current inventories. Compared to earlier studies based mainly on expert judgment (Streets et al., 2003; Zhang et al., 2009; Lei et al., 2011a), Monte-Carlo simulation have been applied to quantify the uncertainties of emissions more carefully, and reduced uncertainty could be expected (Zhao et al., 2011, 2013; Lu et al., 2011). In those studies, emission factors (emissions per unit of energy consumption or industrial/agricultural production) were the main sources of uncertainties attributed mainly to lack of domestic measurements on various sources. However, recent studies suggested that the uncertainty of China's energy statistics should be larger than expected, enhancing the uncertainty of emission estimation for the country (Guan et al., 2012; Hong et al., 2017).

Besides national ones, increasing attentions have been paid on regional/local inventories, motivated mainly by the urgent needs for haze pollution mitigation in specific regions across the country (Zheng et al., 2009; Wang et al., 2010; Fu et al., 2013; Zhao et al., 2015; Zhou et al., 2017). Zhao et al. (2015) incorporated detailed information on individual plants and developed a city scale emission inventory for Nanjing in eastern China. The emission estimates, compared to the downscaled national inventory, were more consistent with the "top-down" constraints obtained from ground/satellite observation. Better model performances could also be achieved when the local inventory was used in high-resolution CTM (Zhou et al., 2017). To further understand the discrepancies between observation and CTM simulation, quantifying the uncertainties of emissions at small spatial scales gets essential. Nevertheless, very limited studies have quantified the uncertainty of emission inventory at local scale, except for given species (e.g., biogenic volatile organic compounds and NH_3 in Pearl River Delta by Zheng et al. (2010) and (2012), respectively) or single source type (e.g., biomass burning by He et al. (2011), on-road vehicles by Wang et al. (2008), and ships by Li et al. (2016)). Moreover, the methods and results of uncertainty analyses could potentially vary a lot for national and local inventories based on different data sources and depth of details. Current available studies, however, simply followed the method for national inventory and seldom conducted careful analysis on individual emission sources. To current knowledge of data and methods for multi-scale inventory development, the uncertainty of emission estimation at local scale, and its difference between local and national inventories remain unclear.

In this work, therefore, a comprehensive uncertainty analysis was conducted on emission inventories for industrial sectors at different spatial scales. We focused on industrial sectors because the methods and data in local inventory could be largely improved in contrast to other sectors (e.g., residential and commercial). We selected Jiangsu province and its capital city Nanjing in eastern China as examples of provincial- and city-level inventories. The processes and data of the inventory development were fully tracked and a detailed Monte-Carlo simulation framework was established, evaluating the uncertainties of emissions from individual plants. At national scale, the uncertainties of emissions of China and Jiangsu were also quantified following the method of our previous work (Zhao et al., 2011). The impacts of various data and methods between multi-scale inventories on the uncertainties of emission estimation are further analyzed.

2. Data and methods

2.1. Multi-scale emission inventories

Annual emissions of eight species (SO_2, NO_X, CO, TSP (total suspended particles), PM_{10}, $PM_{2.5}$, black carbon (BC), and organic carbon (OC)) from industrial sources in 2012 were obtained at three spatial scales, i.e., national, provincial and city-levels. The year 2012 is the base year for China's National Action Plan of Air Pollution Prevention and Control, and the accuracy of its emissions is of great concerns in both scientific and policy-making communities. At national level, China's emissions were estimated by province based mainly on the provincial energy and industrial production statistics (Xia et al., 2016). The industrial sources were classified into thermal power plants, industrial boilers, cement production, iron & steel production, and other industrial processes (see detailed source category in Table S1 in the supplement). The annual emissions for given province were calculated using Eq. (1):

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

where i, p, k, m, and n stand for species, province, sector, fuel type, and emission control technology, respectively; AL is the activity level, either energy consumption or industrial production; EF is the unabated emission factor; R is the penetration rate of the relevant emission control technology; and η is the removal efficiency of that technology.

For SO_2 and PM with specific particle size (i.e., $PM_{2.5}$ and PM_{10}) from coal combustion, emission factors were calculated using Eqs. (2) and (3), respectively:

$$EF_{SO_2,p,k,m} = SC_{p,k,m} \times SR_{k,m} \times \left(1 - \eta_{SO2,p,n}\right) \times 2 \tag{2}$$

$$EF_{PM,y,p,k,m} = AC_{p,k,m} \times AR_{k,m} \times f_{y,k,m} \times \left(1 - \eta_{PM,y,p,n}\right) \tag{3}$$

where y stands for the particulate size; SC and AC are the sulfur and ash content of the fuel, respectively; SR and AR are the release ratio (%) of sulfur and ash, respectively; and f is the particle mass fraction by size. For carbonaceous aerosols, emission factors are obtained by applying the mass fractions of BC (F_{BC}) and OC (F_{OC}) to $PM_{2.5}$.

We select Jiangsu province and its capital city Nanjing for the uncertainty analysis of provincial and city-level inventories, respectively. The geographic locations of Jiangsu and Nanjing with major emission sources are illustrated in Figure S1 in the supplement. Jiangsu is located in the Yangtze-River Delta with developed economy, large fossil fuel consumption and anthropogenic emissions. Incorporating the detailed information of individual sources compiled by local environmental protection agency (i.e., the officially published Environmental Statistics and Pollution Source Census data), a bottom-up methodology was developed to estimate the emissions from industrial point sources. It should be noted that data from Environmental Statistics and Pollution Source Census, and those from provincial energy and economy statistics were compiled by different official departments, and gaps existed between them. The differences between the aggregated activity data from point sources and the provincial energy/economic statistics were calculated and used to estimate the emissions from area sources by sector (Zhou et al., 2017). The aggregated activity data compiled plant by plant, including coal consumption of power generation, production of cement, clinker, coke, pig iron, and crude steel, were estimated at 108%, 95%, 120%, 109%, 104%, and 98% of the provincial statistics, respectively. The data indicates, on one hand, that larger activity levels were obtained based on detailed investigation of individual emission sources than official statistics for power and most processes of iron & steel sectors. On the other hand, almost complete investigation on point sources was conducted for those sectors, and very small fractions of activities had to be estimated as area sources. Through similar method, emissions for Nanjing were calculated from expanded databases including data from unconventional investigations and measurements on specific emitters besides routinely published Environmental Statistics (Zhao et al., 2015). In general the emissions of industrial sources at provincial/city level were calculated by aggregating the emissions of point and area sources:

$$E_i = \sum_j AL_j \times EF_{i,j} \times \left(1 - \eta_{i,j}\right) + \sum_k \sum_m \sum_n AL_{a,k,m,n} \times EF_{i,a,k,m} \times \left(1 - \eta_{i,a,n}\right) \tag{4}$$

where j represented the individual plant, and a presented the area sources.

2.2. Uncertainty analysis of multi-scale inventories

Following our previous work (Zhao et al., 2011), Monte Carlo simulation was applied to quantify the uncertainties of national inventory by source category for the whole country and Jiangsu province. In the simulation framework, uncertainty of each parameter was characterized by probability distribution function (PDF), and the framework randomly generated values for uncertain variables over and over to simulate a model. A simulation calculated numerous scenarios of a model by repeatedly picking values from the PDFs for all the uncertain variables, and the uncertainty of target (emissions in this case) was determined according to the ranges of calculated scenarios. The PDF was fitted for parameters with adequate measurement data, using the Kolmogorov-Smirnov test (K-A test) for the goodness-of-fit ($p = 0.05$). For parameters with limited measurements, and those that fail to pass the goodness-of-fit test, probability distributions must be assumed. In particular, the uncertainties of activity levels for power sector were taken from Zhao et al. (2011). Small difference was found between the annual coal consumption officially reported for the entire country and that from the sum of provincial consumption of power plants, implying good reliability of statistics for the sector (Zhao et al., 2013). Following the guideline by IPCC (2006), normal distribution was assumed and the coefficients of variation (CV, the standard deviation divided by the mean) was set at 5% for activity data of power sector. For other industrial sources, enhanced uncertainty of coal consumption was found through comparisons between the old and updated national energy statistics, and it would lead to an apparent uncertainty of 38% in SO_2 emissions at most (Hong et al., 2017). Accordingly, CV of activity data for industrial sources other than power sector was elevated to 20% in this work (i.e., the 95% confidence interval (CI) was 39.2% around the central estimate). Determination of PDFs for emission factors will be discussed in details in the following section. All of the input activity levels and emission factors with corresponding statistical distributions are then placed in the Monte Carlo framework, and 10,000 simulations are performed to analyze the uncertainties of emissions by sector and species. Parameters that were most significant in determining uncertainties were identified according to their contributions to the variance of emissions.

To quantify the uncertainties of regional (provincial and city) inventories, Monte Carlo framework was improved by incorporating plant-specific information and then applied at each plant for point sources. As the energy consumption data of power sector were compiled and crosschecked based on multiple data sources (Zhou et al., 2017; Zhao et al., 2015), we assumed that the uncertainty would be smaller than other sources and we tentatively set the CV at 10% for each individual plant. For point sources of other categories and area sources, in contrast, the CVs of activity data were set at 20%, the same as the national inventory. For emission factors, we divided the parameters into two types, i.e., the common ones and plant-specific ones. The common parameters indicated those that keep stable or change slightly for certain technology/fuel type, e.g., the sulfur and ash release ratio of bituminous pulverized combustion. The PDFs were determined following Zhao et al. (2011, 2013), with the most recent measurement data incorporated. The plant-specific parameters indicated those that are independent and might vary largely between different plants, e.g., the SO_2 and NO_X removal efficiencies of flue gas desulfurization (FGD) and selective catalytic reduction (SCR) systems, respectively, and the sulfur and ash contents of fossil fuels. Available information was collected and comprehensively compiled to generate the PDFs for those parameters. Details are discussed in the following section.

3. Uncertainties of emission factors

3.1. Common parameters

Applied for uncertainty analyses of both national and regional (provincial and city level) inventories, common parameters related to emission factors were updated from existing studies by species and source category (Zhao et al., 2011, 2012; 2013; Cui et al., 2015), and summarized in Table S2. The PDFs of particle mass fraction by size were taken from Zhao et al. (2011). Combining bootstrap and Monte-Carlo simulation, uncertainties of the unabated emission factors and the particle removal efficiencies of dust collectors for power sector have been thoroughly analyzed by boiler types, fuel quality, and emission control device in our previous study (Zhao et al., 2010, 2012). We assumed there were few changes on those parameters and applied them in this work. For national inventory analysis, triangle distributions were applied for the removal efficiencies of SO_2 and NO_X of FGD and SCR systems, respectively, based on domestic studies (Lu et al., 2011; Tian et al., 2013).

For boilers in other industrial sectors, Zhao et al. (2011) relied most on a relatively old database (SEPA, 1996) to quantify the uncertainty of emission factors. In this work, updated databases with recent measurements were included in the analysis (MEP, 2010; He, 2015), and the PDFs of emission factors were correspondingly revised. For example, the sulfur release ratio of coal combustion (*SR* in Eq. (2)) was elevated from 85% to 90% (i.e., the same as that of power plant), as the combustion technology was improved during past years. In contrast, the ash release ratio (*AR* in Eq. (3)) for grate stokers was kept unchanged at 13% with a logistic distribution.

For cement and iron & steel production, domestic investigation and measurements have been conducted recently (Lei et al., 2011a,b; Huo et al., 2012), and the results were obtained to update the uncertainty analysis of relevant emission factors. For example, the average NO_X emission factor for coking was re-estimated at 1.0 kg/t-production with a uniform distribution

(0.2–1.7). Uncertainties of emission factors of other industrial processes were mainly obtained from different databases that contain most existing domestic measurement results (SEPA, 1996; MEP, 2010; He, 2015). For gaseous pollutants and particles, there was little change in the PDFs of unabated emission factors for most source types compared to previous studies (Zhao et al., 2011, 2012; 2013). The reduced emissions from newer plants were assumed to result from the enhanced penetrations of emission control technologies (Xia et al., 2016). Normal distributions with CV at 50% were tentatively assumed for source categories without sufficient data support of PDF calculation (e.g., refinery, fertilizer production and glass production). For carbonaceous aerosols, although recent field tests implied that OC fractions in $PM_{2.5}$ started to decline in residential combustion sources (Cui et al., 2015), few tests were supplemented for industrial processes and thus PDFs of OC and BC fractions were assumed unchanged from our previous analysis (Zhao et al., 2011).

3.2. Plant-specific parameters

Plant-specific parameters were applied mainly in uncertainty analysis of point sources in regional inventories (for area sources and point sources without detailed information, common parameters were applied instead). For sources related to fossil fuel combustion (e.g., power plants, industrial boilers/kilns, and industrial processes that contain coal combustion), the PDFs of sulfur content, ash content, and SO_2 and NO_X removal efficiencies of corresponding air pollutant control devices (APCDs) were determined for each plant. Fig. 1a and b shows the frequencies of sulfur and ash contents for Jiangsu's power plants and other industrial boilers. The average sulfur content was lower for coals used for power generation than those for other boilers. Normal distributions were assumed for sulfur and ash contents of coal burned for individual plant. Standard deviations of those distributions were determined for each plant with following steps. First, standard deviation for the whole sector was determined through normal distribution fitting with all the data samples within the sector. The difference between sulfur/ash content of single plant and average of the whole sector was then calculated for each plant. The larger value obtained from above two steps was selected as the standard deviation for single plant.

Fig. 1c and d shows the frequencies of removal efficiencies of SO_2 and NO_X for Jiangsu's power and other industrial plants. For both source categories, SO_2 control systems were more widely applied than NO_X control, while the average NO_X removal efficiency based on limited data samples was higher than that of SO_2 for other industrial boilers. Triangular distribution was assumed for the SO_2 removal efficiency for each plant. Clear difference was found between sectors. In 2012, SO_2 control technologies (including wet-FGD and other simple technologies) were applied at 90% of Jiangsu's coal-fired power units, with the average removal efficiency calculated at 68%. For industrial boilers and kilns, in contrast, SO_2 control was applied only 18% of the plants and the average removal rate was 42%. Thus the ranges of triangular distributions differed between source categories. For power sector, the ranges of SO_2 removal rates for FGD systems and other technologies (e.g., circulating fluidized bed, CFB) were 50–98% and 10–85%, respectively, according to the investigated data. For other industrial boilers and kilns, similarly, the ranges were determined at 50–98%, 30–85%, 10–60%, and 10–85% for wet-FGD, dry-FGD, CFB and other technologies, respectively. Besides coal combustion, FGD systems were also applied for other industrial processes. Relatively high removal efficiencies were found for sintering in iron & steel production, and a triangular distribution ranged 50–98% was assumed for individual plant. For other processes including production of ammonia, sulphuric acid, and fertilizer, non ferrous metal smelting, and refinery, the SO_2 removal rates were lower and varied significantly between plants. A triangular distribution ranged 10–85% was tentatively assumed. There were fewer plants applying NO_X control technologies in 2012. For SCR systems, triangular distributions ranged 22–84% were assumed for the removal efficiency, based on the minimum and maximum values of the investigated plants. For selective non-catalytic reduction (SNCR) technology, there were insufficient data to determine PDF, and uniform distribution ranged 25–50% was applied as indicated by Tian et al. (2013).

The removal efficiencies of dust collector by particle size are crucial for estimating the size-fractioned PM emissions of single plant. Nevertheless, there was little information on removal rates of $PM_{2.5}$ or PM_{10} but TSP for individual plants in current databases, and limited field measurements across the country had to be relied on (Zhao et al., 2010). In this study, the investigated removal rates for TSP by plant and the PM size fractions by dust collector type from previous work were incorporated to calculate the removal rates for size-fractioned PM by plant. For data samples that passed K–S test, bootstrap simulations were applied to quantify the mean values and 95% CIs of the removal rates by dust collector type and particle size. Fig. 2 provides bootstrap simulation examples for particle removal efficiencies of wet scrubber for cement production and those of fabric filter for sintering. For those that failed to pass K–S test, normal and uniform distributions were subjectively applied for the parameters with sample size ≥ 10 and < 10, respectively. The 95% CIs were calculated based on the relative ranges provided in our previous work by dust collector type (Zhao et al., 2010). Therefore, the PDFs of PM removal rates were obtained based on the data of individual plants but were applied by dust collector type at sector level, as summarized in Table S3 in the supplement.

For the case of city level inventory, more local information was collected from investigation and tests on single plants and the uncertainties of relevant parameters were further revised compared to the provincial inventory. For example, field measurements on PM removal efficiencies by particle size were conducted for individual power units (unpublished data by local environmental protection administration), which were assumed to reduce the uncertainties of emission estimation for power sector. The PDFs of the parameters were thus determined at plant level according to the dust collector type, following the rules and relative ranges around the central estimates by Zhao et al. (2010). There were only two iron & steel plants in the city, and the release ratios of coke gas and flue gas of blast furnaces were tested to be clearly smaller than the national average (Zhao et al., 2012). The CO emission factors of coking and pig iron production for the plants were thus 73–76% and 39–98% lower than the national average levels, respectively, while relative ranges around the central estimates were assumed the same as those of national levels.

4. Results and discussions

4.1. Uncertainties of multi-scale emission inventories

The uncertainties of industrial emission estimations for China and Jiangsu 2012 in the national inventory are shown in Table 1 by source category, expressed as 95% CIs. The uncertainties were estimated at −17–37%, −21–35%, −19–34%, −29–40%, −22–47%, −21–54%, −33–84%, and −32–92% for the emissions of SO_2, NO_X, CO, TSP, PM_{10}, $PM_{2.5}$, BC, and OC respectively for the whole country. The analogue numbers for Jiangsu were −54–53%, −24–30%, −21–36%, −28–46%, −27–57%, −30–60%, −34–89%, and −36–74%, respectively.

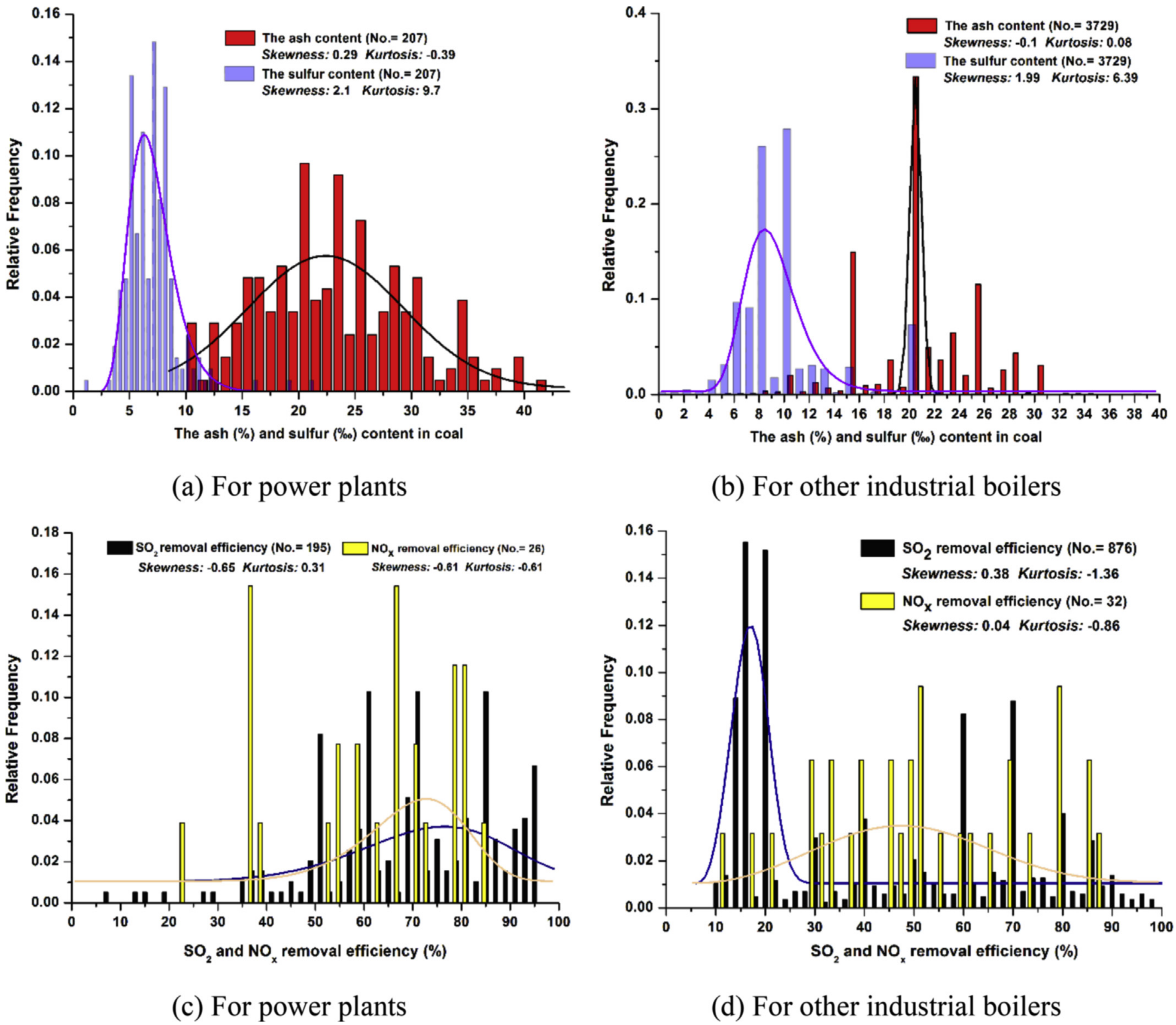


Fig. 1. The frequency of sulfur and ash contents in coal and that of removal efficiencies of SO_2 and NO_X for Jiangsu's power plants and other industrial boilers, based on plant-by-plant investigation. The plant numbers are indicated in the parentheses.

For SO_2, power sector was the largest source category, contributing 46% and 67% to China's and Jiangsu's industrial emissions, respectively. In contrast to previous studies (Zhao et al., 2011; Chen et al., 2014), elevated uncertainties of SO_2 emissions from power plants were found in the national inventory (−33−71% and −77−81% for the whole country and Jiangsu, respectively), leading to bigger 95% CIs of total industrial emissions than other national inventories (e.g., the inventories for TRACE-P and INTEX-B). The reasons will be discussed later in Section 4.2. For NO_X and CO, the largest uncertainties were found in industrial boiler combustion, ranging from −47−151% and −53−86% for the whole country, and −35−99% and −49−84% for Jiangsu, respectively. Due to the poor information on technology at national scale, uniform emission factor had to be applied, and it ignored differences of combustion efficiencies across the sector, and yielded considerable uncertainty.

For particles, iron & steel industry and other industrial processes were the largest contributors, while emissions from cement production were constrained compared to earlier years (Lei et al., 2011b; Zhao et al., 2011; Xia et al., 2016) attributed mainly to the increased penetration of fabric filters. Relatively big uncertainties were found for cement production and industrial boilers, and larger uncertainties were estimated for smaller particles for almost all the sectors. For carbonaceous aerosols, bigger uncertainties were found for power and industrial boilers, but the uncertainties of total industry were dominated by iron & steel production, the main industrial source of BC and OC. Although uncertainties of emissions from other industrial processes were not the largest for most species, it does not imply that emissions of the source category were clearly known. The results in Table 1 aggregate the uncertainties of all processes and thus cannot reveal the potentially larger uncertainties for individual processes.

Table 2 summarized the uncertainties of provincial (Jiangsu) and city-level (Nanjing) emission inventory by source category. The uncertainties for Jiangsu were estimated at −15−18%, −18−33%, −16−37%, −20−30%, −23−45%, −26−50%, −33−79%,

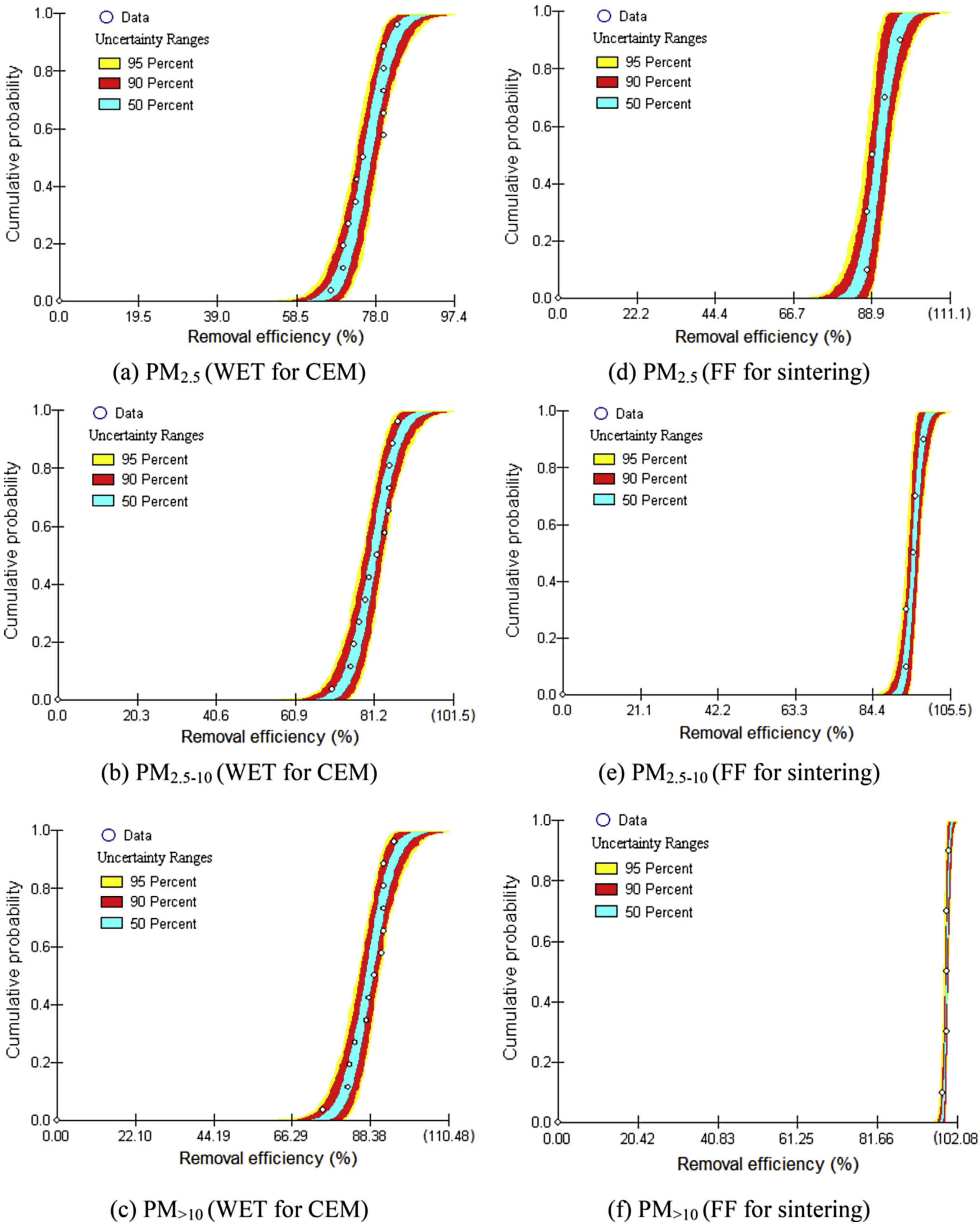


Fig. 2. The probability bands of particle removal efficiencies of wet scrubbers for cement production (a–c) and of fabric filters for sintering (d–f), through bootstrap simulation with individual plant data.

Table 1
Uncertainties of China's and Jiangsu's emissions (unit: Gg) in the national inventory by industrial source category for 2012. The percentages in the parentheses indicate the 95% CI around the central estimate.

	SO_2	NO_X	CO	TSP	PM_{10}	$PM_{2.5}$	BC	OC
China								
PP	10,531 (−33%, 71%)	10,355 (−21%, 28%)	1783 (−24%, 35%)	1830 (−25%, 38%)	1,462 (−27%, 45%)	889 (−37%, 60%)	7 (−59%, 562%)	1 (−29%, 271%)
CEM	1702 (−53%, 55%)	3,613 (−62%, 74%)	6,390 (−41%, 57%)	2,698 (−57%, 168%)	2,137 (−63%, 177%)	1,243 (−68%, 197%)	10 (−73%, 181%)	25 (−74%, 221%)
ISP	2,112 (−56%, 90%)	1,503 (−57%, 23%)	33,940 (−44%, 52%)	4,797 (−40%, 71%)	2,897 (−45%, 65%)	2,283 (−49%, 63%)	325 (−48%, 108%)	296 (−49%, 100%)
PRO	4,409 (−28%, 36%)	1,182 (−37%, 43%)	32,504 (−25%, 62%)	8,397 (−56%, 55%)	2,458 (−36%, 38%)	1,373 (−29%, 55%)	108 (−53%, 62%)	111 (−60%, 34%)
OIC	4,287 (−40%, 59%)	2,797 (−47%, 151%)	6,634 (−53%, 86%)	1750 (−58%, 137%)	1,064 (−56%, 171%)	644 (−59%, 256%)	74 (−71%, 319%)	26 (−54%, 1,054%)
Total	23,041 (−17%, 37%)	19,430 (−21%, 35%)	81,252 (−19%, 34%)	19,471 (−29%, 40%)	10,019 (−22%, 47%)	6,432 (−21%, 54%)	524 (−33%, 84%)	460 (−32%, 92%)
Jiangsu								
PP	882 (−77%, 81%)	834 (−24%, 36%)	171 (−25%, 37%)	185 (−37%, 63%)	145 (−38%, 70%)	86 (−44%, 87%)	1 (−64%, 547%)	0 (−37%, 746%)
CEM	85 (−61%, 69%)	297 (−63%, 74%)	272 (−46%, 54%)	192 (−65%, 183%)	154 (−70%, 190%)	91 (−75%, 207%)	1 (−79%, 202%)	2 (−79%, 240%)
ISP	78 (−55%, 54%)	61 (−59%, 23%)	2,175 (−39%, 49%)	324 (−40%, 73%)	192 (−47%, 57%)	155 (−49%, 57%)	13 (−58%, 131%)	13 (−55%, 124%)
PRO	224 (−32%, 40%)	63 (−34%, 51%)	1942 (−29%, 56%)	386 (−53%, 57%)	103 (−38%, 42%)	52 (−31%, 63%)	7 (−54%, 61%)	8 (−61%, 36%)
OIC	49 (−58%, 68%)	61 (−35%, 99%)	60 (−49%, 84%)	15 (−52%, 111%)	10 (−45%, 124%)	7 (−40%, 144%)	1 (−49%, 179%)	1 (−32%, 382%)
Total	1,318 (−54%, 53%)	1,317 (−24%, 30%)	4,621 (−21%, 36%)	1,103 (−28%, 46%)	604 (−27%,57%)	392 (−30%, 60%)	23 (−34%, 89%)	23 (−36%, 74%)

Table 2
Uncertainties of Jiangsu's and Nanjing's emissions (unit: Gg) by industrial source category for 2012 in the provincial and city-level inventories, respectively. The percentages in the parentheses indicate the 95% CI around the central estimate.

	SO_2	NO_X	CO	TSP	PM_{10}	$PM_{2.5}$	BC	OC
Jiangsu								
PP	612 (−12%, 16%)	685 (−25%, 28%)	219 (−36%, 64%)	366 (−23%, 35%)	164 (−25%, 35%)	71 (−33%, 54%)	1 (−73%, 337%)	0 (−49%, 421%)
CEM	36 (−34%, 44%)	122 (−58%, 58%)	113 (−26%, 31%)	931 (−46%, 75%)	478 (−58%, 108%)	233 (−76%, 131%)	2 (−71%, 108%)	5 (−74%, 140%)
ISP	118 (−32%, 116%)	90 (−36%, 23%)	2,965 (−23%, 37%)	605 (−21%, 44%)	356 (−30%, 42%)	278 (−37%, 41%)	14 (−54%, 101%)	28 (−47%, 68%)
LIM	3 (−48%, 38%)	6 (−35%, 38%)	78 (−29%, 42%)	267 (−65%, 12%)	32 (−66%, 19%)	5 (−66%, 26%)	0 (−68%, 29%)	0 (−83%, 69%)
BRI	22 (−37%, 43%)	5 (−42%, 59%)	120 (−40%, 36%)	143 (−40%, 45%)	29 (−41%, 51%)	10 (−44%, 51%)	4 (−35%, 74%)	4 (−47%, 42%)
NFS	69 (−65%, 84%)	0 (−43%, 135%)	0 (−86%, 365%)	45 (−75%, 285%)	42 (−75%, 283%)	38 (−75%, 280%)	–	–
AP	1 (−50%, 401%)	7 (−38%, 74%)	165 (−78%, 53%)	2 (−79%, 85%)	1 (−66%, 186%)	0 (−52%, 268%)	0 (−77%, 307%)	0 (−44%, 388%)
GLA	6 (−30%, 58%)	6 (−43%, 90%)	1 (−73%, 449%)	35 (−22%, 18%)	33 (−31%, 15%)	32 (−32%, 15%)	–	–
FER	2 (−55%, 178%)	1 (−71%, 232%)	1 (−95%, 393%)	8 (−48%, 54%)	7 (−51%, 48%)	6 (−53%, 47%)	–	–
REF	5 (−28%, 85%)	4 (−44%, 48%)	137 (−84%, 415%)	2 (−41%, 47%)	2 (−41%, 47%)	1 (−40%, 52%)	–	–
SAP	5(−40%, 51%)	1 (−69%, 172%)	1 (−93%, 507%)	2 (−78%, 103%)	1 (−71%, 239%)	0 (−69%, 450%)	0 (−98%, 84%)	0 (−82%, 607%)
NAP	0 (−70%, 134%)	2 (−81%, 811%)	0 (−95%, 427%)	3 (−88%, 132%)	1 (−86%, 230%)	0 (−82%, 459%)	0 (−97%, 545%)	0 (−79%, 2056%)
OIC	252 (−13%, 47%)	202 (−34%, 62%)	130 (−50%, 124%)	123 (−72%, 96%)	31 (−66%, 187%)	13 (−51%, 280%)	2 (−79%, 339%)	1 (−46%, 426%)
Area	235 (−73%, 79%)	77 (−70%, 216%)	466 (−52%, 102%)	109 (−48%, 97%)	48 (−41%, 134%)	24 (−38%, 188%)	1 (−93%, 367%)	1 (−61%, 632%)
Total	1,366 (−15%, 18%)	1,207 (−18%, 33%)	4,396 (−16%, 37%)	2,640 (−20%, 30%)	1,223 (−23%, 45%)	712 (−26%, 50%)	24 (−33%, 79%)	39 (−33%, 71%)
Nanjing								
PP	52 (−18%, 22%)	81 (−12%, 33%)	32 (−30%, 41%)	16 (−30%, 39%)	11 (−34%, 47%)	7 (−39%, 54%)	0 (−82%, 128%)	0 (−63%, 76%)
CEM	6 (−46%, 47%)	20 (−56%, 68%)	18 (−26%, 39%)	17 (−64%, 162%)	12 (−72%, 179%)	8 (−83%, 212%)	0 (−84%, 193%)	0 (−83%, 253%)
ISP	24 (−27%, 177%)	20 (−28%, 49%)	550 (−33%, 47%)	71 (−26%, 58%)	47 (−36%, 46%)	38 (−42%, 49%)	2 (−67%, 104%)	3 (−49%, 116%)
LIM	0 (−41%, 46%)	0 (−39%, 41%)	4 (−28%, 47%)	16 (−66%, 15%)	2 (−67%, 21%)	0 (−66%, 25%)	0 (−68%, 35%)	0 (−83%, 65%)
BRI	2 (−37%, 48%)	1 (−42%, 66%)	8 (−38%, 39%)	15 (−39%, 42%)	3 (−40%, 47%)	1 (−40%, 43%)	0 (−33%, 65%)	0 (−44%, 35%)
NFS	0 (−69%, 70%)	0 (−44%, 113%)	0 (−67%, 216%)	0 (−66%, 267%)	0 (−72%, 261%)	0 (−74%, 270%)	0 (−93%, 436%)	0 (−46%, 1,484%)
OIC	10 (−28%, 33%)	14 (−48%, 110%)	8 (−80%, 307%)	5 (−83%, 124%)	3 (−83%, 169%)	2 (−84%, 196%)	0 (−88%, 392%)	0 (−56%, 1,126%)
Area	19 (−36%, 42%)	7 (−43%, 154%)	147 (−69%, 341%)	5 (−39%, 77%)	3 (−31%, 87%)	3 (−32%, 98%)	0 (−85%, 406%)	0 (−48%, 1,241%)
Total	114 (−17%, 22%)	144 (−10%, 33%)	768 (−23%, 75%)	145 (−19%, 36%)	82 (−23%, 41%)	59 (−28%, 48%)	3 (−45%, 82%)	4 (−34%, 96%)

and −33–71% for emissions of SO_2, NO_X, CO, TSP, PM_{10}, $PM_{2.5}$, BC, and OC respectively. Power plants and industrial boilers were the main contributors to SO_2 and NO_X emissions. Compared to the results of the national inventory, the uncertainty of SO_2 emissions for the two source categories were reduced to −12–16% and −13–47%, and that of NO_X to −25–28% and −37–62%, respectively. Reduced uncertainties in the provincial inventory indicated the benefits of detailed investigations on the activity data and emission factors for individual plants instead of entire sectors or sub-sectors. Cement and iron & steel production were the most important sources of primary particles, and iron & steel production dominated the CO, BC and OC emissions. Similarly, the uncertainties of those source categories for given species were smaller than those in the national inventory. For other industrial processes, although information was collected at plant level by source category, relatively large uncertainties were still estimated for most source types, indicating the poor understanding of the emission characteristics for this sector. In addition, the 95% CIs of emissions for area sources were calculated as one combined group, thus they could not reveal the uncertainties for any single source category.

At city scale, the uncertainties of Nanjing's industrial sources were estimated at −17–22%, −10–33%, −23–75%, −19–36%, −23–41%, −28–48%, −45–82%, and −34–96% for emissions of SO_2, NO_X, CO, TSP, PM_{10}, $PM_{2.5}$, BC, and OC respectively. The 95% CIs were similar to the relative ranges of provincial inventory for most species, with an exception that enhanced uncertainty was found for CO in the city-scale inventory. Power sector and iron & steel industry were identified as the most important sources for gaseous pollutants (except CO) and particles, respectively, and small changes in 95% CIs (relative ranges) were found for corresponding sector/species compared to the provincial inventory.

4.2. Identification of key parameters

Table 3 summarized the 1st and 2nd ranking parameters contributing most to the uncertainties of China's and Jiangsu's emissions in the national inventory by industrial source category. As can be seen, the emission factors (or associated parameters) were identified as the biggest contributors to the uncertainties of emissions for most source categories except iron & steel production.

For SO_2 from power generation, the uncertainty of emissions for the whole country was dominated by the average removal efficiency of wet FGD, while the main source of uncertainties for Jiangsu was the average sulfur content of coals in the province. For iron & steel production, the unabated EF of sintering process was the most significant parameter to the emission uncertainty for both China and Jiangsu. For NO_X, EF of grate boilers was the key parameter for uncertainties of emissions from power generation, although the penetration of the technology was very limited in 2012. The uncertainty of cement production emissions was mainly influenced by the EF of precalciner kilns (the most widely applied technology in the sector). Difference existed in the analysis for other industrial combustion: EF of grate boiler (coal combustion) was crucial for the uncertainty of the whole country's emissions while that of oil combustion for the uncertainty of the province. For CO, iron & steel production and other processes were the biggest sources for both China and Jiangsu, and the activity data of pig iron and crude steel production, and EFs of refinery and brick production were respectively identified as the most crucial parameters to emission uncertainties of the two source categories.

In most cases, the same parameters were identified as the crucial parameters for emission uncertainties of particles with different sizes (i.e., TSP, PM_{10}, and $PM_{2.5}$), except for the category of other industrial process. Unabated EF of lime production were the most important parameter for the uncertainty of coarse particles, as the mass fraction of particles above 10 μm was 88%. With a mass fraction of 82% for $PM_{2.5}$, in contrast, EF of copper smelting was estimated to contribute most to uncertainty of $PM_{2.5}$ for industrial process. For power sector, mass fraction of $PM_{2.5}$ for pulverized combustion was the most crucial for uncertainty of PM emissions for the whole country, while the average ash content for that of Jiangsu province. For carbonaceous aerosols, the mass fractions of BC and OC to PM EFs were identified as the most significant parameters for most source categories, while uncertainties of total industrial emissions were largely influenced by the activity data of coke and pig iron production, as iron & steel production was the main source of industrial carbonaceous aerosol emissions.

To evaluate the effects of activity levels on emission uncertainty, a sensitivity analysis was further conducted, in which the CVs of

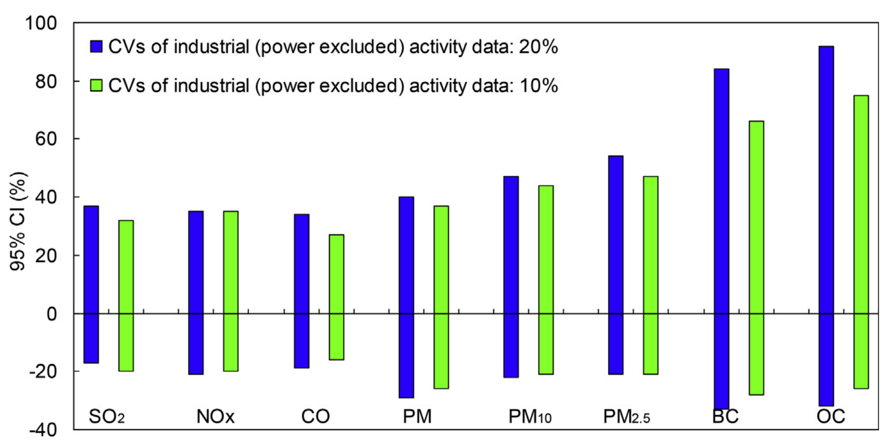
Table 3
The parameters contributing most to emission uncertainties for China and Jiangsu in the national inventory, by industrial source category and species. The percentages in the parentheses indicate the contributions of the parameters to the variance of corresponding emissions (see Section 2.1 for the abbreviations of parameters).

	SO_2	NO_X	CO	PM	PM_{10}	$PM_{2.5}$	BC	OC
China								
PP	$\eta_{SO2\ FGD}$ (−78%)	EF_{grate} (37%)	$EF_{pulverized\ (\geq 200MW)}$ (44%)	$f_{PM2.5\ pulverized}$ (33%)	$f_{PM2.5\ pulverized}$ (41%)	$f_{PM2.5\ pulverized}$ (58%)	$F_{BC\ pulverized}$ (79%)	$F_{OC\ grate}$ (34%)
	AL_{coal} (3%)	$EF_{tangential\ bituminous}$ (18%)	$EF_{pulverized\ (<200MW)}$ (26%)	$\eta_{PM2.5\ ESP}$ (−12%)	$\eta_{PM2.5\ ESP}$ (−14%)	$\eta_{PM2.5\ ESP}$ (−19%)	$f_{PM2.5\ pulverized}$ (5%)	$f_{PM2.5\ grate}$ (23%)
CEM	AL_{cement} (49%)	$EF_{precalciner}$ (62%)	AL_{cement} (66%)	$EF_{PM\ precalciner}$ (38%)	$EF_{PM\ precalciner}$ (41%)	$EF_{PM\ precalciner}$ (42%)	$EF_{PM\ precalciner}$ (40%)	$EF_{PM\ precalciner}$ (33%)
	$\eta_{SO2\ precalciner}$ (−23%)	AL_{cement} (26%)	EF_{shaft_kiln} (14%)	$R_{precalciner\ ESP}$ (20%)	$R_{precalciner\ ESP}$ (18%)	$R_{precalciner\ ESP}$ (16%)	$R_{precalciner\ ESP}$ (15%)	F_{OC} (21%)
ISP	$EF_{sintering}$ (26%)	AL_{pig_iron} (15%)	$AL_{sintering}$ (15%)	AL_{pig_iron} (15%)	AL_{pig_iron} (15%)	AL_{pig_iron} (15%)	AL_{coke} (13%)	AL_{coke} (13%)
	$AL_{sintering}$ (12%)	AL_{coke} (15%)	AL_{steel} (15%)	AL_{coke} (15%)	AL_{coke} (15%)	AL_{coke} (15%)	AL_{pig_iron} (13%)	AL_{pig_iron} (13%)
PRO	$EF_{Cu\_smelting}$ (55%)	AL_{lime} (34%)	$EF_{refinery}$ (34%)	$R_{lime\ nocontrol}$ (45%)	$EF_{PM\ lime}$ (17%)	$EF_{PM\ Cu\_smelting}$ (18%)	$EF_{PM\ brick}$ (38%)	$EF_{PM\ brick}$ (38%)
	AL_{brick} (13%)	EF_{lime} (33%)	EF_{brick} (17%)	$EF_{PM\ lime}$ (22%)	$R_{lime\ nocontrol}$ (16%)	$R_{Cu\_smelting\ nocontrol}$ (14%)	$AL_{solid\_clay\_brick}$ (36%)	$AL_{solid\_clay\_brick}$ (37%)
OIC	AL_{coal} (48%)	EF_{grate} (68%)	$EF_{grate\_handfeed}$ (41%)	AR_{grate} (52%)	AR_{grate} (37%)	$f_{PM2.5\ grate}$ (48%)	$F_{BC\ grate}$ (45%)	$F_{OC\ grate}$ (43%)
	SR_{grate} (14%)	$EF_{oil\_combustion}$ (10%)	AL_{coal} (30%)	AL_{coal} (20%)	$f_{PM2.5\ grate}$ (29%)	AR_{grate} (28%)	$f_{PM2.5\ grate}$ (20%)	$f_{PM2.5\ grate}$ (26%)
Total	$\eta_{SO2\ FGD}$ (−56%)	EF_{grate} (44%)	$AL_{pig\ iron}$ (13%)	$R_{lime\ nocontrol}$ (16%)	$EF_{PM\ precalciner}$ (14%)	$EF_{PM\ precalciner}$ (11%)	AL_{coke} (11%)	$AL_{pig\ iron}$ (11%)
	AL_{coal} (5%)	CEM_B56 (17%)	AL_{steel} (13%)	$EF_{PM\ lime}$ (8%)	AL_{coke} (7.4%)	AL_{coke} (8%)	$AL_{pig\ iron}$ (11%)	AL_{coke} (11%)
Jiangsu								
PP	$SC_{Jiangsu\ coal}$ (58%)	EF_{grate} (42%)	$EF_{pulverized\ (<200MW)}$ (43%)	$AC_{Jiangsu\ coal}$ (65%)	$AC_{Jiangsu\ coal}$ (58%)	$AC_{Jiangsu\ coal}$ (42%)	$F_{BC\ pulverized}$ (64%)	$F_{OC\ grate}$ (37%)
	$\eta_{SO2\ FGD}$ (−28%)	$EF_{tangential\ bituminous}$ (29%)	$EF_{pulverized\ (\geq 200MW)}$ (26%)	$f_{PM2.5\ pulverized}$ (10%)	$f_{PM2.5\ pulverized}$ (16%)	$f_{PM2.5\ pulverized}$ (32%)	$AC_{Jiangsu\ coal}$ (9%)	$f_{PM2.5,\ grate}$ (27%)
CEM	$SC_{Jiangsu\ coal}$ (31%)	$EF_{precalciner}$ (64%)	AL_{cement} (56%)	$EF_{PM\ precalciner}$ (43%)	$EF_{PM\ precalciner}$ (46%)	$EF_{PM\ precalciner}$ (46%)	$EF_{PM\ precalciner}$ (44%)	$EF_{PM\ precalciner}$ (38%)
	AL_{cement} (29%)	AL_{cement} (26%)	$EF_{precalciner}$ (33%)	$R_{precalciner\ ESP}$ (22%)	$R_{precalciner\ ESP}$ (20%)	$R_{precalciner\ ESP}$ (18%)	$R_{precalciner\ ESP}$ (18%)	F_{OC} (17%)
ISP	$EF_{sintering}$ (14%)	AL_{coke} (15%)	AL_{pig_iron} (16%)	AL_{pig_iron} (14%)	AL_{pig_iron} (14%)	AL_{pig_iron} (14%)	AL_{coke} (11%)	AL_{steel} (12%)
	$AL_{sintering}$ (13%)	AL_{pig_iron} (14%)	AL_{steel} (16%)	AL_{steel} (14%)	AL_{coke} (14%)	AL_{coke} (14%)	AL_{steel} (11%)	AL_{steel} (12%)
PRO	$EF_{Cu\ smelting}$ (49%)	EF_{brick} (24%)	EF_{brick} (28%)	$R_{lime\ nocontrol}$ (40%)	$EF_{PM\ lime}$ (16%)	$EF_{PM\ Cu\_smelting}$ (30%)	$EF_{PM\ brick}$ (40%)	$EF_{PM\ brick}$ (40%)
	AL_{brick} (22%)	EF_{lime} (20%)	$EF_{refinery}$ (24%)	$EF_{PM\ lime}$ (21%)	$R_{lime\ nocontrol}$ (13%)	$H_{PM2.5\ CYC}$ (−9%)	$AL_{solid\_clay\_brick}$ (36%)	$AL_{solid\_clay\_brick}$ (37%)
OIC	SR_{oil} (66%)	$EF_{oil\_combustion}$ (50%)	$EF_{grate\_handfeed}$ (40%)	AR_{grate} (47%)	AR_{grate} (34%)	$f_{PM2.5\ grate}$ (43%)	$F_{BC\ grate}$ (32%)	$F_{OC\ grate}$ (36%)
	AL_{oil} (7%)	AL_{oil} (20%)	AL_{coal} (32%)	AL_{coal} (17%)	$f_{PM2.5\ grate}$ (28%)	AR_{grate} (27%)	$f_{PM2.5\ grate}$ (17%)	$f_{PM2.5\ grate}$ (24%)
Total	$SC_{Jiangsu\ coal}$ (56%)	CEM_B56 (26%)	$AL_{sintering}$ (13%)	$EF_{PM\ precalciner}$ (9%)	$EF_{PM\ precalciner}$ (19%)	$EF_{PM\ precalciner}$ (15%)	AL_{steel} (10%)	AL_{steel} (11%)
	$\eta_{SO2\ FGD}$ (−26%)	EF_{grate} (26%)	AL_{pig_iron} (13%)	AL_{steel} (8%)	$R_{precalciner\ ESP}$ (8%)	AL_{steel} (8%)	AL_{coke} (10%)	AL_{pig_iron} (11%)

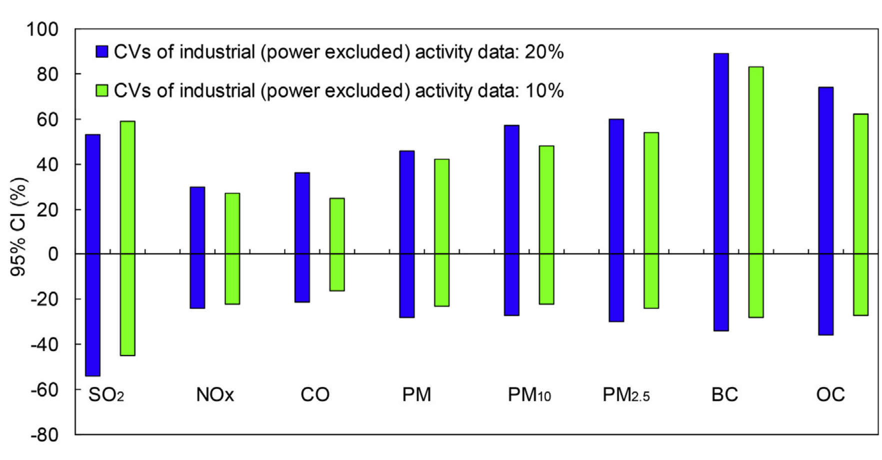
industrial production and fossil fuel consumption other than power sector were reduced to 10%, the same as Zhao et al. (2011). The uncertainties for certain species/source category combinations (e.g., CO and particles from cement and iron & steel production) decreased with the declined CVs of activity data, as summarized in Table S4 in the supplement. The changes in uncertainties of total industrial emissions of most species, however, were small for both the whole country and Jiangsu, as shown in Fig. 3a and b, respectively. The result indicated that the limitation of current systems on China's energy and economic statistics was not the main source of uncertainties of the total emissions of air pollutants.

Table 4 summarized the 1st and 2nd ranking parameters contributing most to the uncertainties of Jiangsu's and Nanjing's emissions in the provincial and city-level inventory by industrial source category, respectively. There were differences in the key parameter identification for the province in the national and provincial inventory. For SO_2 from power sector, as an example, the removal efficiencies of FGD for given individual plants were identified as the most significant parameters to the uncertainty, with very limited contribution to variance. Similarly, the uncertainty of emissions from other industrial combustions was estimated to be mostly affected by the sulfur content and FGD removal rate of one single plant. The results thus indicated the importance of better understanding on emission characteristics for certain individual plants when emission inventory was developed using a "plant-based" method. For total SO_2 emissions, sulfur content and coal consumption of area sources were identified as the most significant parameters, implying the necessity to improve the studies on area sources in the future. For NO_X, as unabated EFs could not be provided for individual plants but for source/technology types, the EF of bituminous combustion with tangentially-fired burners (mainly for power sector) and that of coal combustion with grate boilers were identified as the main sources of emission uncertainty. In contrast to SO_2 control, SCR technology for NO_X reduction was not widely applied in 2012, even in power sector, and the removal rates of SCR for very limited plants were not significant for the emission uncertainties. Iron & steel production was the main source of CO, BC and OC. In contrast to the national inventory in which activity data dominated the uncertainty for the species, EF-related parameters (e.g., EF of CO from sintering) played more important roles in the provincial inventory. For particles, EF of cement production were the main source of uncertainty for the provincial inventory, similar to the national one.

At city level, the EF-related parameters dominated the uncertainties of emissions for all the sectors in Nanjing. Large power and iron & steel plants were responsible for most of energy consumption and industrial production in the city, and the activity data of those big sources were carefully studied, leading to small



(a) Whole country



(b) Jiangsu

Fig. 3. The 95% CIs of emissions in the national inventory for the whole country (a) and Jiangsu (b). Blue and green bars represent the results with the CVs of industrial (power excluded) activity data set at 20% and 10%, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 4
The parameters contributing most to emission uncertainties for Jiangsu and Nanjing in the provincial and city-level inventories, respectively, by industrial source category and species. The percentages in the parentheses indicate the contributions of the parameters to the variance of corresponding emissions.

	SO_2	NO_X	CO	PM	PM_{10}	$PM_{2.5}$	BC	OC
Jiangsu								
PP	$\eta_{SO2\ FGD\ j}$ (−1%)	$EF_{tangential\ bituminous}$ (18%)	EF_{CFBC} (11%)	$\eta_{PM>10\ ESP}$ (−14%)	$\eta_{PM2.5\_10\ ESP}$ (−7%)	$f_{PM2.5\ pulverized}$ (14%)	$F_{BC\ pulverized}$ (20%)	$F_{OC\ grate}$ (9%)
	$\eta_{SO2\ FGD\ j}$ (−1%)	$EF_{no\_LNB\ bituminous}$ (4%)	$EF_{grate\_autofeed}$ (6%)	$AR_{\ pulverized}$ (2%)	$f_{PM2.5\ pulverized}$ (5%)	$\eta_{PM2.5\ ESP}$ (−7%)	$f_{PM2.5\ pulverized}$ (1%)	$f_{PM2.5\ grate}$ (6%)
CEM	$\eta_{SO2\ precalciner}$ (−17%)	$EF_{precalciner}$ (25%)	$EF_{precalciner}$ (23%)	$EF_{PM\ precalciner}$ (13%)	$EF_{PM\ precalciner}$ (14%)	$EF_{PM\ precalciner}$ (13%)	$EF_{PM\ precalciner}$ (12%)	$EF_{PM\ precalciner}$ (9%)
	$SC_{Jiangsu\ coal}$ (3%)	N/A	$AL_{coal,\ j}$ (1%)	$EF_{PM\ CEM\_process}$ (8%)	$EF_{PM\ CEM\_process}$ (4%)	$f_{PM2.5\ CEM\_process}$ (7%)	$f_{PM2.5\ CEM\_process}$ (7%)	F_{OC} (6%)
ISP	$EF_{sintering}$ (14%)	EF_{coke} (13%)	$EF_{sintering}$ (11%)	$EF_{PM\ pig\_iron}$ (7%)	$f_{PM2.5\ steel}$ (7%)	$f_{PM2.5\ steel}$ (10%)	$F_{BC\ pig\_iron}$ (6%)	$f_{PM2.5\ steel}$ (15%)
	EF_{pig_iron} (9%)	$EF_{sintering}$ (10%)	EF_{steel} (6%)	$\eta_{PM>10\ CYC}$ (−4%)	$EF_{PM\ pig\_iron}$ (4%)	$EF_{PM\ steel}$ (3%)	AL_{pig_iron} (6%)	$EF_{PM\ steel}$ (4%)
BRI	EF_{brick} (25%)	EF_{brick} (24%)	EF_{brick} (24%)	$EF_{PM\ brick}$ (24%)	$EF_{PM\ brick}$ (20%)	$EF_{PM\ brick}$ (22%)	$EF_{PM\ brick}$ (20%)	$EF_{PM\ brick}$ (21%)
	N/A	N/A	N/A	N/A	$f_{PM2.5\_10\ brick}$ (4%)	$f_{PM2.5\ brick}$ (2%)	$f_{PM2.5\ brick}$ (2%)	$f_{PM2.5\ brick}$ (2%)
LIM	EF_{lime} (24%)	EF_{lime} (24%)	EF_{lime} (23%)	$EF_{PM\ lime}$ (24%)	$EF_{PM\ lime}$ (22%)	$EF_{PM\ lime}$ (21%)	$EF_{PM\ lime}$ (19%)	$F_{OC\ lime}$ (15%)
	$\eta_{SO2\ FGD\ j}$ (−1%)	$AL_{lime,j}$ (1%)	$AL_{coal,j}$ (1%)	N/A	$f_{PM2.5\_10\ lime}$ (2%)	$f_{PM2.5\ lime}$ (3%)	$F_{BC\ lime}$ (3%)	$EF_{PM\ lime}$ (8%)
NFS	$EF_{Cu\_smelting}$ (21%)	EF_{grate} (18%)	$EF_{grate\_autofeed}$ (25%)	$EF_{PM\ Cu\_smelting}$ (24%)	$EF_{PM\ Cu\_smelting}$ (24%)	$EF_{PM\ Cu\_smelting}$ (23%)	–	–
	$\eta_{SO2\ FGD\ j}$ (−1%)	$EF_{oil\_combustion}$ (4%)	N/A	$AL_{Cu\_smelting,\ j}$ (1%)	$AL_{Cu\_smelting,\ j}$ (1%)	$AL_{Cu\_smelting,\ j}$ (1%)	–	–
AP	EF_{AP} (23%)	EF_{grate} (16%)	EF_{AP} (23%)	$AR_{\ grate}$ (18%)	$AR_{\ grate}$ (12%)	$f_{PM2.5\ grate}$ (12%)	$F_{BC\ grate}$ (12%)	$F_{OC\ grate}$ (8%)
	N/A	$EF_{gas\_combustion}$ (6%)	$AL_{AP,\ j}$ (1%)	$\eta_{AP\ PM>10\ WET}$ (−2%)	$f_{PM2.5\ grate}$ (6%)	$AR_{\ grate}$ (8%)	$f_{PM2.5\ grate}$ (5%)	$f_{PM2.5\ grate}$ (5%)
GLA	$SC_{Jiangsu\ oil}$ (8%)	$EF_{oil\_combustion}$ (16%)	$EF_{grate\_autofeed}$ (24%)	$EF_{PM\ glassproduct}$ (15%)	$EF_{PM\ glassproduct}$ (10%)	$EF_{PM\ glassproduct}$ (10%)	–	–
	$SC_{coal,j}$ (8%)	EF_{grate} (6%)	$EF_{oil\_combustion}$ (1%)	$EF_{PM\ glass}$ (7%)	$f_{PM2.5\ glass}$ (6%)	$f_{PM2.5\ glass}$ (7%)	–	–
FER	$SC_{coal,j}$ (17%)	EF_{grate} (22%)	$EF_{grate\_autofeed}$ (25%)	$EF_{PM\ FER}$ (18%)	$EF_{PM\ FER}$ (16%)	$EF_{PM\ FER}$ (15%)	–	–
	$\eta_{SO2\ FGD\ j}$ (−1%)	$AL_{coal,j}$ (2%)	$AL_{coal,j}$ (1%)	$AL_{PM\ fertilizer}$ (6%)	$AL_{PM\ fertilizer}$ (6%)	$AL_{PM\ fertilizer}$ (5%)	–	–
REF	EF_{REF} (16%)	EF_{REF} (18%)	EF_{REF} (24%)	$EF_{PM\ REF}$ (14%)	$EF_{PM\ REF}$ (14%)	$EF_{PM\ REF}$ (11%)	–	–
	$\eta_{SO2\ FGD\ j}$ (−5%)	$AL_{oil\_production,\ j}$ (6%)	$AL_{oil\_production,\ j}$ (1%)	$AL_{oil\_production,\ j}$ (8%)	$AL_{oil\_production,\ j}$ (8%)	$f_{PM2.5\ REF}$ (7%)	–	–
SAP	EF_{SAP} (21%)	EF_{grate} (22%)	$EF_{grate\_autofeed}$ (24%)	$AR_{\ grate}$ (18%)	$AR_{\ grate}$ (11%)	$f_{PM2.5\ grate}$ (12%)	$F_{BC\ grate}$ (12%)	$F_{OC\ grate}$ (9%)
	$AL_{SAP,\ j}$ (1%)	$AL_{coal,\ j}$ (1%)	N/A	$AL_{coal,\ j}$ (2%)	$f_{PM2.5\ grate}$ (6%)	$AR_{\ grate}$ (7%)	$f_{PM2.5\ grate}$ (5%)	$f_{PM2.5\ grate}$ (6%)
NAP	$SC_{coal,\ j}$ (17%)	EF_{NAP} (24%)	$EF_{grate\_autofeed}$ (24%)	$AR_{\ grate}$ (11%)	$AR_{\ grate}$ (9%)	$f_{PM2.5\ grate}$ (9%)	$F_{BC\ grate}$ (10%)	$F_{OC\ grate}$ (8%)
	$AL_{coal,\ j}$ (3%)	$AL_{NAP,\ j}$ (1%)	$AL_{coal,\ j}$ (1%)	$AC_{coal,\ j}$ (10%)	$f_{PM2.5\ grate}$ (3%)	$AR_{\ grate}$ (7%)	$f_{PM2.5\ grate}$ (4%)	$f_{PM2.5\ grate}$ (5%)
OIC	$SC_{coal,\ j}$ (6%)	EF_{grate} (9%)	$EF_{grate\_autofeed}$ (11%)	AR_{grate} (22%)	AR_{grate} (13%)	$f_{PM2.5\ grate}$ (13%)	$F_{BC\ grate}$ (12%)	$F_{OC\ grate}$ (9%)
	$\eta_{SO2\ FGD\ j}$ (−6%)	$EF_{gas\_combustion}$ (8%)	$EF_{gas\_combustion}$ (7%)	$AC_{Jiangsu\ coal}$ (1%)	$f_{PM2.5\ grate}$ (5%)	AR_{grate} (9%)	$f_{PM2.5\ grate}$ (6%)	$f_{PM2.5\ grate}$ (6%)
Area	$SC_{Jiangsu\ coal}$ (18%)	EF_{grate} (21%)	EF_{AP} (23%)	$AR_{\ grate}$ (12%)	$AR_{\ grate}$ (8%)	$f_{PM2.5\ grate}$ (9%)	$F_{BC\ grate}$ (12%)	$F_{OC\ grate}$ (8%)
	AL_{area_coal} (6%)	AL_{coal} (3%)	EF_{REF} (13%)	$AC_{Jiangsu\ coal}$ (4%)	$f_{PM2.5\ grate}$ (4%)	$AR_{\ grate}$ (6%)	$f_{PM2.5\ grate}$ (5%)	$f_{PM2.5\ grate}$ (5%)
Total	$SC_{Jiangsu\ coal}$ (10%)	$EF_{tangential\ bituminous}$ (7%)	$EF_{sintering}$ (6%)	$EF_{PM\ precalciner}$ (10%)	$EF_{PM\ precalciner}$ (12%)	$EF_{PM\ precalciner}$ (10%)	$F_{BC\ pig\_iron}$ (5%)	$f_{PM2.5\ steel}$ (13%)
	AL_{area_coal} (3%)	EF_{grate} (4%)	EF_{REF} (5%)	$EF_{PM\ CEM\_\ process}$ (6%)	$EF_{PM\ CEM\_process}$ (3%)	$f_{PM2.5\ CEM\_process}$ (5%)	$EF_{PM\ pig\_iron}$ (4%)	$EF_{PM\ steel}$ (3%)
Nanjing								
PP	$\eta_{SO2\ FGD\ j}$ (−18%)	$EF_{no\_LNB\ bituminous}$ (27%)	$EF_{pulverized\ (<200MW)}$ (48%)	$f_{PM2.5\ pulverized}$ (22%)	$f_{PM2.5\ pulverized}$ (31%)	$f_{PM2.5\ pulverized}$ (44%)	$F_{BC\ pulverized}$ (51%)	$F_{OC\ gas}$ (54%)
	$\eta_{SO2\ FGD\ j}$ (−6%)	$EF_{tangential\ bituminous}$ (12%)	$EF_{gas\_combustion}$ (5%)	$AR_{\ pulverized}$ (8%)	$\eta_{PM2.5\_10\ ESP,\ j}$ (−6%)	$\eta_{PM2.5\ ESP,\ j}$ (−6%)	$f_{PM2.5\ pulverized}$ (4%)	$EF_{PM\ gas}$ (4%)
CEM	$\eta_{SO2\ precalciner}$ (−36%)	$EF_{precalciner}$ (55%)	$EF_{precalciner}$ (49%)	$EF_{PM\ precalciner}$ (40%)	$EF_{PM\ precalciner}$ (39%)	$EF_{PM\ precalciner}$ (35%)	$EF_{PM\ precalciner}$ (34%)	$EF_{PM\ precalciner}$ (26%)
	$SR_{precalciner,coal}$ (10%)	$AL_{coal,\ j}$ (1%)	$AL_{coal,\ j}$ (2%)	$\eta_{PM2.5\ CEM\ FF}$ (−4%)	$\eta_{PM2.5\ CEM\ FF}$ (−6%)	$\eta_{PM2.5\ CEM\ FF}$ (−13%)	$\eta_{PM2.5\ CEM\ FF}$ (−12%)	F_{OC} (14%)
ISP	$EF_{sintering}$ (33%)	EF_{coke} (45%)	$EF_{sintering}$ (46%)	$\eta_{PM>10\ CYC}$ (−19%)	$f_{fugitive\_PM2.5\ iron}$ (11%)	$f_{fugitive\_PM2.5\ iron}$ (13%)	$F_{fugitive\_BC\ iron}$ (28%)	$f_{PM2.5\ steel}$ (21%)
	EF_{pig_iron} (15%)	$EF_{sintering}$ (6%)	EF_{steel} (6%)	$EF_{fugitive\_PM\ iron}$ (14%)	$f_{PM2.5\ steel}$ (7%)	$f_{PM2.5\ steel}$ (10%)	$EF_{PM\ pig\_\ iron}$ (8%)	$EF_{PM\ steel}$ (11%)
BRI	EF_{brick} (52%)	EF_{brick} (54%)	EF_{brick} (54%)	$EF_{PM\ brick}$ (52%)	$EF_{PM\ brick}$ (45%)	$EF_{PM\ brick}$ (49%)	$EF_{PM\ brick}$ (45%)	$EF_{PM\ brick}$ (47%)
	$AL_{brick,\ j}$ (3%)	$AL_{brick,\ j}$ (2%)	$AL_{coal,\ j}$ (1%)	$AL_{brick,\ j}$ (3%)	$f_{PM2.5\_10\ brick}$ (8%)	$f_{PM2.5\ brick}$ (3%)	$F_{BC\ brick}$ (5%)	$AL_{brick,\ j}$ (3%)
LIM	EF_{lime} (50%)	EF_{lime} (49%)	EF_{lime} (47%)	$EF_{PM\ lime}$ (55%)	$EF_{PM\ lime}$ (50%)	$EF_{PM\ lime}$ (49%)	$EF_{PM\ lime}$ (46%)	$F_{OC\ lime}$ (34%)
	$AL_{Lime,\ j}$ (2%)	$AL_{Lime,\ j}$ (3%)	$AL_{coal,\ j}$ (10%)	$AL_{Lime,\ j}$ (1%)	$f_{PM2.5\_10\ lime}$ (4%)	$f_{PM2.5\ lime}$ (6%)	$F_{BC\ lime}$ (6%)	$EF_{PM\ lime}$ (17%)
NFS	$EF_{Cu\_smelting}$ (52%)	EF_{grate} (54%)	$EF_{grate\_autofeed}$ (46%)	$EF_{PM\ Cu\_smelting}$ (55%)	$EF_{PM\ Cu\_smelting}$ (54%)	$EF_{PM\ Cu\_smelting}$ (54%)	$F_{BC\ grate}$ (32%)	$F_{OC\ grate}$ (28%)
	$AL_{Cu\_smelting,\ j}$ (1%)	$AL_{gas,j}$ (2%)	EF_{gas} (6%)	$AL_{Cu\_smelting,\ j}$ (1%)	$AL_{Cu\_smelting,\ j}$ (1%)	$AL_{Cu\_smelting,\ j}$ (1%)	$f_{PM2.5\ grate}$ (13%)	$f_{PM2.5\ grate}$ (16%)
OIC	SR_{grate} (25%)	EF_{grate} (54%)	$EF_{grate\_autofeed}$ (55%)	AR_{grate} (41%)	$f_{PM2.5\ grate}$ (24%)	$f_{PM2.5\ grate}$ (32%)	$F_{BC\ grate}$ (29%)	$F_{OC\ grate}$ (25%)
	$\eta_{SO2\ FGD\ j}$ (−14%)	$AL_{coal,\ j}$ (1%)	EF_{gas} (1%)	$f_{PM2.5\ grate}$ (10%)	AR_{grate} (23%)	AR_{grate} (18%)	$f_{PM2.5\ grate}$ (12%)	$f_{PM2.5\ grate}$ (15%)
Area	$SC_{Jiangsu\ coal}$ (18%)	EF_{NAP} (33%)	EF_{REF} (52%)	AR_{grate} (29%)	AR_{grate} (23%)	$f_{PM2.5\ grate}$ (22%)	$F_{BC\ grate}$ (29%)	$F_{OC\ grate}$ (25%)
	AL_{area_oil} (13%)	EF_{grate} (14%)	AL_{area_oil} (13%)	AL_{area_coal} (9%)	$f_{PM2.5\ grate}$ (15%)	AR_{grate} (14%)	$f_{PM2.5\ grate}$ (12%)	$f_{PM2.5\ grate}$ (15%)
Total	$EF_{sintering}$ (19%)	EF_{grate} (14%)	EF_{REF} (38%)	$\eta_{PM>10\ CYC}$ (−14%)	$EF_{PM\ precalciner}$ (13%)	$f_{fugitive\_PM2.5\ iron}$ (9%)	$F_{fugitive\_BC\ iron}$ (18%)	$f_{PM2.5\ steel}$ (17%)
	EF_{pig_iron} (9%)	$EF_{no\_LNB\ bituminous}$ (12%)	$EF_{sintering}$ (11%)	$EF_{PM\ precalciner}$ (8%)	$f_{fugitive\_PM2.5\ iron}$ (6%)	$EF_{PM\ precalciner}$ (7%)	$F_{BC\ grate}$ (6%)	$EF_{PM\ steel}$ (10%)

uncertainties. For SO_2, removal efficiencies of FGD for individual plants were the key parameters for the uncertainties of emissions from power sector, while EF of sintering and pig iron production for the total emissions. Similar to Jiangsu province, EF of bituminous combustion without low-NO_X burner (LNB) and that of coal combustion with grate boilers were identified as the crucial parameters of NO_X uncertainty, and the EF-related parameters of cement and iron & steel production dominated the uncertainties of particle emissions with different sizes and carbonaceous components.

4.3. Comparisons between multi-scale inventories

As can be seen in Table 1, there were no significant differences in the uncertainties (expressed as 95% CIs) for China and Jiangsu in the national emission inventory. Notably, larger uncertainty of SO_2 emissions from power sector was found for Jiangsu province (−77%, 81%) than that for the whole country (−33%, 71%). As shown in Table 3, the sulfur content of coals in the province was assumed to vary considerably, and it contributed more to the uncertainty of SO_2 emissions than the average removal rate of FGD did. As power sector was the biggest source of SO_2 in Jiangsu, the enhanced uncertainty of SO_2 emissions from power led to elevated uncertainty of total industrial emissions. For iron & steel production, less uncertainty was found for Jiangsu than that for China. In the sector, sintering was the main source of SO_2 emissions, and EF of sintering with relatively big uncertainty (lognormal distribution with 95% CI: 1.2−6.1 kg/t-product) dominated the uncertainty of emissions for the whole country. In Jiangsu, however, the sintering production was relatively less compared to its pig iron and steel production. As a result, the effects of sintering EF on emission uncertainty were reduced, while effects of activity data (e.g., steel production) were elevated. Enhanced uncertainties were found for particle emissions of Jiangsu province. Similar to SO_2, the ash content of coals was more influential on the uncertainty of emissions from power generation, resulting in the elevated emission uncertainty for the sector.

The discrepancies in Jiangsu's emissions between national and provincial inventories varied for species. For gaseous pollutants, the discrepancies were small, i.e., 4%, −9% and −5% (calculated as the differences between the provincial and national estimates relative to the provincial ones) for SO_2, NO_X and CO, respectively. In contrast, the particle emissions in the provincial inventory were doubled compared to those in the national one, attributed mainly to the poorer removal rates of dust collectors investigated and applied in the provincial inventory (Zhou et al., 2017). Besides total emissions, the contribution of certain sectors also varied, and it resulted partly from the different methods in activity data estimation in the two inventories. For example, the coal combustion of Jiangsu's industrial boilers other than power was estimated at 3.6 million tons (Mt) using a “downscaled method” for industrial activity levels (Zhao et al., 2015; Xia et al., 2016). Through plant-by-plant survey, however, the compiled coal consumption in the provincial inventory reached 32.6 Mt, i.e., 9 times of that in the national inventory. The big discrepancy in activity data resulted in different sector contributions to emissions in the two inventories, particularly for SO_2 and NO_X that were mainly from coal combustion. The fractions of emissions from OIC to Jiangsu's total industrial emissions were estimated at 18% and 4% for SO_2, and 17% and 5% for NO_X, in the provincial and national inventory, respectively.

It can be found that the SO_2 uncertainties for Jiangsu were largely reduced in the provincial inventory. Fig. 4a shows the difference in distributions of SO_2 emissions from power plants in the national and provincial inventories. Based on the information collected at plant level, improved estimations on emissions from big plants were anticipated for power and industrial boilers. As independent PDFs were assumed for the key parameters (e.g., sulfur content of coal and removal rate of FGD) of each plant, the emission uncertainties at sector level could further decrease attributed to the “compensation-of-error” mechanisms. Moreover, the contribution of individual plants to the emission uncertainty could also be enhanced in the provincial inventory, compared to the variables at sector level in the national inventory. For NO_X, CO, BC and OC, the uncertainties of the provincial inventory were not significantly reduced compared to the national inventory. As shown in Fig. 4b, the shapes of probability distributions of NOx from power plants in the provincial and national inventories did not vary much. Although large fraction of activity data was compiled at plant-level in the provincial inventory, limited progress was made to improve the estimation of the sector/technology-based emission factors that played more important roles on emission uncertainty for those species. In some cases, even bigger uncertainty was estimated for provincial inventory, e.g., CO emissions from power and industrial boilers. According to on-site investigations, the penetration of grate boilers was larger than it was assumed in the national inventory, and EF of grate boilers with relatively big variation (lognormal distribution with 95% CI: 0.1−14.6) enhanced the uncertainty of emissions. The uncertainties of particles with different sizes were reduced to some extent in the provincial inventory. With the improved estimation on coal quality at plant level, the effects of average ash content on uncertainty of emissions from power sector decreased in the provincial inventory. Similarly, the penetrations of various dust collector types on cement production were much clearer in the provincial inventory, resulting in less uncertainty of particle emissions for the sector. The uncertainties of Nanjing's emissions were not considerably reduced compared to the provincial emissions for most cases. The uncertainty of industrial CO emissions was even enhanced, resulting mainly from the poor quantification of emission factors for the intensively distributed refinery plants in the city.

Fig. 5 compares the central estimates of Jiangsu's emissions (at provincial level) by this work with those of other studies. As limited information was available for the same year as this work (2012), the results for 2010 and 2014 were included for comparisons as well. Those inventories were developed either at national scale (MEIC; Xia et al., 2016) or regional scale (Fu et al., 2013; Shanghai Research Academy of Environmental Science, SRAES). It can be seen that most estimates are within the 95% CIs calculated in the current study for gaseous pollutants, BC and OC. Inter-annual variability could partly explain the lower estimates by SRAES and Fu et al. (2013). Different data sources (in particular the penetrations and removal rates of emission control technologies) in multiple-scale inventories also contributed to the discrepancies in emission estimates. For PM_{10} and $PM_{2.5}$, emissions from other studies were much smaller than this work at provincial level, even out of the 95% CIs around the central estimates. This large discrepancy came mainly from the difference in the removal rates of dust collectors used in various inventories. While all other studies applied the national/regional averages of dust removal rates based on limited test samples to calculate the emissions, the plant-specific data obtained from on-site investigations generally implied much lower removal rates for the province, leading to larger emission estimates in the provincial inventory. The comparison revealed the importance of developing and applying local emission factors on establishment of high-resolution emission inventory whenever possible.

4.4. Implication of air quality research

Besides the bottom-up methodology, “top-down” estimations in emissions of gaseous pollutants have also been made through inverse methods that apply satellite observation and/or chemistry

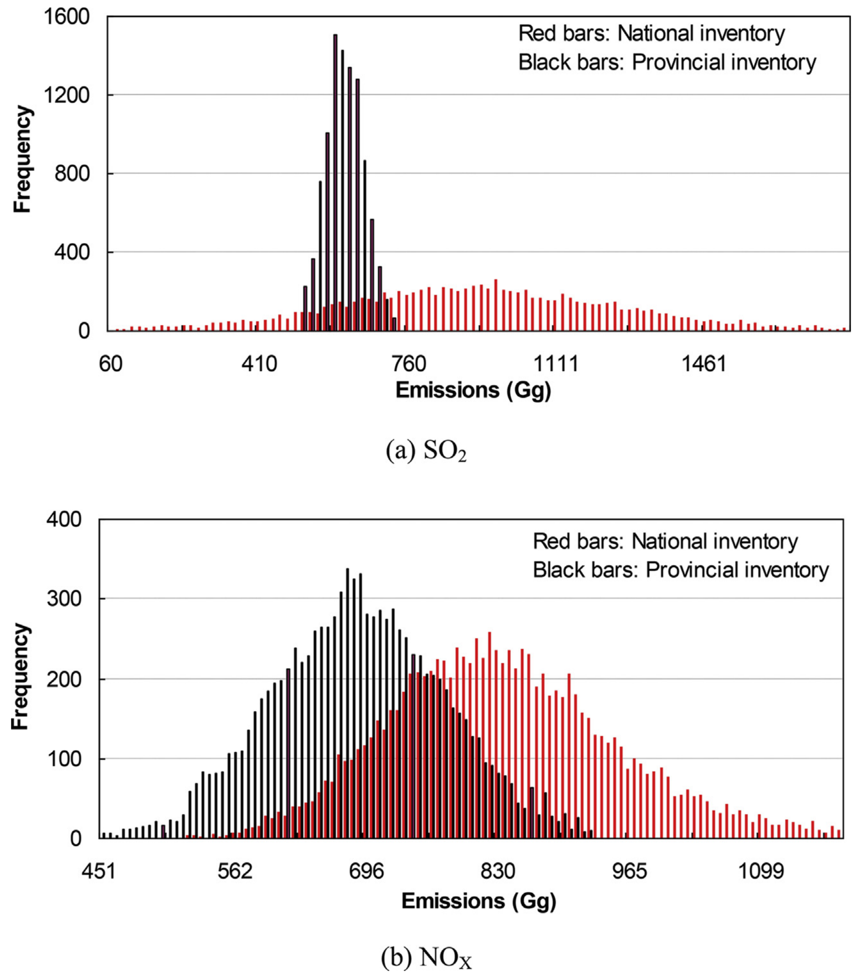


Fig. 4. The distributions of SO_2 (a) and NO_X (b) emissions from power plants in Jiangsu in 2012, using Monte-Carlo simulation. The red and black bars represent the results from the national and provincial inventories, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

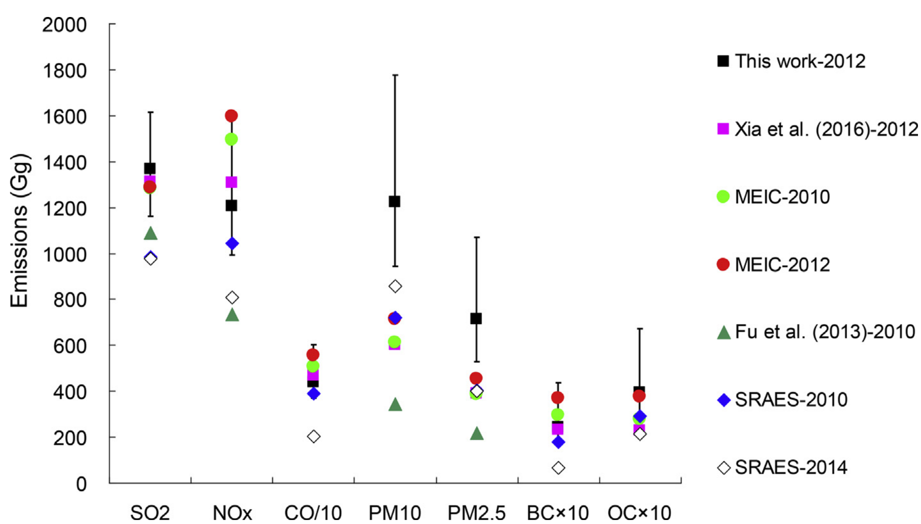


Fig. 5. Comparisons of Jiangsu's annual emissions from different studies. The range line represents the 95% CIs around the central estimates of the provincial inventory by this work.

transport modeling. Difference between results from the two methods thus implied the uncertainty of current emission inventories to some extent. While earlier studies provided relatively big gaps between the bottom-up and top-down NO_X estimates for China (e.g., Wang et al., 2004; Ma et al., 2006), the difference tended to be smaller in later studies for east China. Based on the observed tropospheric column densities of NO_2 from Global Ozone Monitoring Experiment (GOME), NO_X emissions from fuel combustion in east China were expected to be underestimated by 15% through GEOS-Chem modeling (Wang et al., 2007). With the observed NO_2 from Ozone Measuring Instrument (OMI), NOx emissions from fossil fuel combustion were expected to be over-estimated by 13% using a regional chemical transport model (REAM) by Zhao and Wang (2009). Lin et al. (2010) combined OMI and GOME data and estimated NO_X emissions at 5.5 TgN/yr for east China through GEOS-Chem, very close to those from bottom-up method (5.7 TgN/yr). For CO, GEOS-Chem and Measurement of Pollution in the Troposphere (MOPITT) were applied to estimate the emissions for the whole country (Tanimoto et al., 2008; Yumimoto et al., 2014), and the discrepancies were ranged 2–25% relative to the bottom-up estimates for multiple years between 2005 and 2010. All those results were within the uncertainty ranges (expressed as 95% CI) provided in this study. It should be noted that discrepancies between top-down and bottom-up methods might vary considerably across regions, and that the uncertainty analysis of sectors other than industry were not effectively improved at provincial and city levels in this study compared to previous one (Zhao et al., 2011). In addition, uncertainty in top-down estimations would result from systematic and random errors in the retrievals of satellite data and the uncertainties of chemistry transport model related to the assumptions on diurnal profiles of emissions, natural emissions (soil and lightning, etc), and planetary boundary layer mixture scheme. For example, differences in systematic errors between individual retrievals might lead to underestimation in top-down emissions of NO_X by at most 17%, and the standard deviation was estimated at 13% from the best top-down estimate, attributed to the impacts of factors associated with errors and uncertainties in GEOS-Chem (Lin et al., 2010). Such limitations prevented better comparisons between the emission estimates from different approaches. Besides the regional emissions, emission burdens from top-down method have also been estimated for individual power plants recently (Wang et al., 2015; Liu et al., 2016). Liu et al. (2016) found the difference relative to bottom-up estimates for NO_X was within 30% for most concerned plants, close to 95% CIs of power sector emissions in this work. Based on OMI observation, Wang et al. (2015) estimated the average removal equivalence of SO_2 was 56.0%, substantially lower than the official report (74.6%) for selected 26 power plants across China. The result suggested a 42% underestimation on SO_2 emissions from power sector applying the national average of SO_2 removal rate, within the 95% CIs of power sector emissions estimated at national level in this work.

At city scale, “top-down” constraints expressed as correlations of certain pairs of pollutants were derived from observed ambient concentrations and then applied for evaluating the “bottom-up” estimates of primary emissions for Nanjing (Zhao et al., 2015). In this work, we analyzed the uncertainties of primary emission ratios of BC to CO and OC to BC, combining current available information on uncertainties of Nanjing emissions by sector. As shown in Table 5, emissions and their uncertainties of industrial sector were taken from current work, and emissions from residential and transportation sectors were kept unchanged from Zhao et al. (2015). As little progress was made to improve the uncertainty analysis of city-scale emissions from the latter two sectors, the relative ranges around the central estimates for national inventory were applied (Zhao et al., 2011). For fugitive dust, lognormal distributions with a CV at 100% was tentatively assumed to reflect the highly uncertain estimates. With 10,000 Monte-Carlo simulations, the 95% CIs of BC to CO and OC to BC emission ratios were calculated at 0.0033–0.0133 and 0.79–2.92, with central estimates at 0.0074 and 1.39, respectively. In contrast, the “top-down ”constraints from ground observation were 0.0084 and 1.59, respectively, i.e., they were 14% larger than the bottom-up ratios, and within the 95% CIs. It should be noted that uncertainty also existed in the method of “top-down” constraint estimation. For example, the uncertainty of BC to CO emission ratio derived from observation was estimated at 20%, as the wet, dry deposition and chemistry of the species could not be fully understood (Wang et al., 2011; Zhao et al., 2015). Moreover, the primary OC to BC emission ratio was estimated with a semi-quantitative method, EC-tracer method. The determination of the ratio was arbitrary and unable to obtain single OC to BC ratio that represented a mixture of primary sources varying in time and space. Errors could be caused by the occasional irregular contributions from sources with a primary OC to EC ratio vastly different from the usual mix of sources (Cui et al., 2015). Therefore, the comparison between the bottom-up and top-down estimates needed to be interpreted with caution. Judged by the contribution to the variance of emission ratio, industrial CO and BC emissions were identified to be most significant to the uncertainty of primary BC to CO ratio, with the contribution to variance calculated at 36% and 25%, respectively. Industrial OC, fugitive OC, and industrial BC emissions were identified to be most significant to uncertainty of primary OC to BC, with the contribution to variance at 25%, 23%, and 20%, respectively. The result highlighted the importance of industrial emissions on the “bottom-up” emission ratio estimates.

5. Conclusion

Uncertainties of multi-scale emission inventories based on different methods and data sources were quantified with a modified Monte-Carlo simulation framework for mainland China, Jiangsu, and Nanjing. Although the completeness of data were improved from national to provincial and city scales, the uncertainties of total emissions were not significantly reduced for most species, with an exception of SO_2. Nevertheless, the emission uncertainties of certain species for given sources (e.g., SO_2 and NO_X from power plants and other industrial boilers) were reduced at local scales compared to those at national scale. Plant-specific

Table 5
The emissions of BC, OC and CO by source category and the ratios of BC to CO and OC to BC emissions for Nanjing 2012.

	Total	Industry[a]	Transportation[b]	Residential[b]	Fugitive dust[b]
	Emissions in Gg	Emissions in Gg (95% CIs)			
BC	5,436	2,514 (−45%, 82%)	1910 (−75%, 89%)	296 (−47%, 259%)	716 (−86%, 261%)
OC	7,567	4,253 (−34%, 96%)	719 (−68%, 98%)	685 (−54%, 148%)	1909 (−86%, 261%)
CO	916,004	767,811 (−23%, 75%)	101,038 (−34%, 56%)	47,155 (−50%, 102%)	–
	Ratio (95% CIs)	The contribution to the uncertainty of emission ratio (BC, CO for BC/CO; OC, BC for OC/BC)			
BC/CO	0.0074 (0.0033, 0.0113)	25%, 36%	24%, 1%	2%, 1%	11%, 0%
OC/BC	1.39 (0.79, 2.92)	25%, 20%	2%, 18%	1%, 2%	23%, 9%

[a] This work.
[b] Taken from Zhao et al. (2015).

information on emission factors was identified to be crucial for the uncertainties of emissions for those source categories. The results indicated the importance of better understanding on emission characteristics of individual plants when emission inventory was developed using a "plant-based" method. Such experience could be extended to other regions/cities with similar economic and industrial structures. Due to lack of sufficient field measurements, however, the plant-specific emission factors for all species were still unavailable at local spatial scale for other industrial sources, and technology-based emission factors had to be assigned for all the plants of the same manufacturing technology types, without independent uncertainty assumptions. Moreover, the uncertainty analysis for sectors other than industry at provincial and city scales could hardly be improved, as data from current available statistics or investigations could not better differentiate the emission characteristics by source type or provide detailed information on errors of activity data and emission factors. Those limitations motivated more detailed on-site investigations and field measurements on typical emission sources for further improving the emission estimates at local scales.

Acknowledgements

This work was sponsored by the National Key Research and Development Program of China (2017YFC0210106), Natural Science Foundation of China (91644220 and 41575142), the National Key Research and Development Program of China (2016YFC0201507), Natural Science Foundation of Jiangsu (BK20140020), Jiangsu Science and Technology Support Program (SBE2014070918), and Special Research Program of Environmental Protection for Commonweal (201509004). We thank Qiang Zhang and Xiao Fu from Tsinghua University, and Cheng Huang from SRAES for providing the national and regional emission inventories. Thanks should also go to the two anonymous reviewers for their very valuable comments to improve this work.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2017.06.045.

References

Chen, L., Sun, Y., Wu, X., Zhang, Y., Zheng, C., Gao, X., Cen, K., 2014. Unit-based emission inventory and uncertainty assessment of coal-fired power plants. Atmos. Environ. 99, 527–535.
Cui, H., Mao, P., Zhao, Y., Nielsen, C.P., Zhang, J., 2015. Patterns in atmospheric carbonaceous aerosols in China: emission estimates and observed concentrations. Atmos. Chem. Phys. 15, 8657–8678.
Fu, X., Wang, S., Zhao, B., Xing, J., Cheng, Z., Liu, H., Hao, J., 2013. Emission inventory of primary pollutants and chemical speciation in 2010 for the Yangtze River Delta region, China. Atmos. Environ. 70, 39–50.
Guan, D.B., Liu, Z., Geng, Y., Lindner, S., Hubacek, K., 2012. The gigatonne gap in China's carbon dioxide inventories. Nat. Clim. Change 2, 672–675.
He, K. (Ed.), 2015. Technique Handbook of City-scale Emission Inventory Development for Atmospheric Pollutants, Internal Report. Beijing (in Chinese).
He, M., Zheng, J., Yin, S., Zhang, Y., 2011. Trends, temporal and spatial characteristics, and uncertainties in biomass burning emissions in the Pearl River Delta, China. Atmos. Environ. 45, 4051–4059.
Hong, C., Zhang, Q., He, K.B., Guan, D., Li, M., Liu, F., Zheng, B., 2017. Variations of China's emission estimates: response to uncertainties in energy statistics. Atmos. Chem. Phys. 17, 1227–1239.
Huo, H., Lei, Y., Zhang, Q., Zhao, L., He, K., 2012. China's coke industry: recent policies, technology shift, and implication for energy and the environment. Energy Policy 51, 397–404.
Intergovernmental Panel on Climate Change (IPCC), 2006. 2006 IPCC Guidelines for National Greenhouse Gas Inventories, IPCC National Greenhouse Gas Inventories Programme.
Joint Research Centre (JRC)/Netherlands Environmental Assessment Agency (PBL), 2011. Emission Database for Global Atmospheric Research (EDGAR), release version 4.2. Available online at: http://edgar.jrc.ec.europa.eu.
Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T., Kawashima, K., Akimoto, H., 2013. Emissions of air pollutants and greenhouse gases over Asian regions during 2000-2008: regional Emission inventory in ASia (REAS) version 2. Atmos. Chem. Phys. 13, 11019–11058.
Lei, Y., Zhang, Q., He, K.B., Streets, D.G., 2011a. Primary anthropogenic aerosol emission trends for China, 1990-2005. Atmos. Chem. Phys. 11, 931–954.
Lei, Y., Zhang, Q., Nielsen, C.P., He, K.B., 2011b. An inventory of primary air pollutants and CO_2 emissions from cement industry in China, 1990–2020. Atmos. Environ. 45, 147–154.
Li, C., Yuan, Z., Ou, J., Fan, X., Ye, S., Xiao, T., Shi, Y., Huang, Z., Ng, S.K.W., Zhong, Z., Zheng, J., 2016. An AIS-based high-resolution ship emission inventory and its uncertainty in Pearl River Delta region, China. Sci. Total Environ. 573, 1–10.
Lin, J., McElroy, M.B., Boersma, K.F., 2010. Constraint of anthropogenic NO_X emissions in China from different sectors: a new methodology using multiple satellite retrievals. Atmos. Chem. Phys. 10, 63–78.
Liu, F., Beirle, S., Zhang, Q., Dorner, S., He, K., Wagner, T., 2016. NOX lifetimes and emissions of cities and power plants in polluted background estimated by satellite observation. Atmos. Chem. Phys. 16, 5283–5298.
Lu, Z., Zhang, Q., Streets, D.G., 2011. Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996-2010. Atmos. Chem. Phys. 11, 9839–9864.
Ma, J., Richter, A., Burrows, J.P., Nuss, H., van Aardenne, J.A., 2006. Comparison of model-simulated tropospheric NO_2 over China with GOME-satellite data. Atmos. Environ. 40, 593–604.
Ministry of Environmental Protection of China (MEP), 2010. Handbook of Emission Factors of Industrial Pollution Sources for the First National Pollution Source Survey, Internal Report. Beijing (in Chinese).
Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., Hayasaka, T., 2007. An Asian emission inventory of anthropogenic emission sources for the period 1980-2020. Atmos. Chem. Phys. 7, 4419–4444.
Saikawa, E., Kim, H., Zhong, M., Zhao, Y., Janssens-Manehout, G., Kurokawa, J., Klimont, Z., Wagner, F., Naik, V., Horowitz, L., Zhang, Q., 2017. Comparison of emissions inventories of anthropogenic air pollutants in China. Atmos. Chem. Phys. 17 (in press).
State Environmental Protection Administration (SEPA), 1996. Handbook of Industrial Pollution Emission Factors. China Environmental Science Press, Beijing (in Chinese).
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., Yarber, K.F., 2003. An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. J. Geophys. Res. 108 (D21), 8809,. http://dx.doi.org/10.1029/2002jd003093.
Tanimoto, H., Sawa, Y., Yonemura, S., Yumimoto, K., Matsueda, H., Uno, I., et al., 2008. Diagnosing recent CO emissions and ozone evolution in East Asia using coordinated surface observations, adjoint inverse modeling, and MOPITT satellite data. Atmos. Chem. Phys. 8, 3867–3880.
Tian, H., Liu, K., Hao, J., Wang, Y., Gao, J., Qiu, P., Zhu, C., 2013. Nitrogen oxides emissions from thermal power plants in China: current Status and Future Predictions. Environ. Sci. Technol. 47, 11350–11357.
Wang, Y.X., McElroy, M.B., Wang, T., Palmer, P.I., 2004. Asian emissions of CO and NOX: constraints from aircraft and Chinese station data. J. Geophys. Res. 109, D24304. http://dx.doi.org/10.1029/2004jd005250.
Wang, Y.X., McElroy, M.B., Martin, R.V., Streets, D.G., Zhang, Q., Fu, T.M., 2007. Seasonal variability of NOX emissions over east China constrained by satellite observations: implications for combustion and microbial sources. J. Geophys. Res. 112, D06301. http://dx.doi.org/10.1029/2006jd007538.
Wang, H., Chen, C., Huang, C., Fu, L., 2008. On-road vehicle emission inventory and its uncertainty analysis for Shanghai, China. Sci. Total Environ. 398, 60–67.
Wang, S., Zhao, M., Xing, J., Wu, Y., Zhou, Y., Lei, Y., He, K., Fu, L., Hao, J.M., 2010. Quantifying the air pollutants emission reduction during the 2008 Olympic Games in Beijing. Environ. Sci. Technol. 44, 2490–2496.
Wang, Y.X., Wang, X., Kondo, Y., Kajino, M., Munger, J.W., Hao, J.M., 2011. Black carbon and its correlation with trace gases at a rural site in Beijing: top-down constraints from ambient measurements on bottom-up emissions. J. Geophys. Res. 116, D24304,. http://dx.doi.org/10.1029/2011JD016575.
Wang, S., Zhang, Q., Martin, R.V., Philip, S., Liu, F., Li, M., Jiang, X., He, K., 2015. Satellite measurements oversee China's sulfur dioxide emission reductions from coal-fired power plants. Environ. Res. Lett. 10, 114015,. http://dx.doi.org/10.1088/1748-9326/10/11/114015.
Xia, Y., Zhao, Y., Nielsen, C., 2016. Benefits of China's efforts in gaseous pollutant control indicated by the bottom-up emissions and satellite observations 2000–2014. Atmos. Environ. 136, 43–53.
Yumimoto, K., Uno, I., Itahashi, S., 2014. Long-term inverse modeling of Chinese CO emission from satellite observations. Environ. Pollut. 195, 308–318.
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., Yao, Z., 2009. Asian emissions in 2006 for the NASA INTEX-B mission. Atmos. Chem. Phys. 9, 5131–5153.
Zhao, C., Wang, Y.H., 2009. Assimilated inversion of NO_X emissions over east Asia using OMI NO2 column measurements. Geophys. Res. Lett. 36, L06805. http://dx.doi.org/10.1029/2008gl037123.
Zhao, Y., Wang, S.X., Nielsen, C.P., Li, X.H., Hao, J.M., 2010. Establishment of a database of emission factors for atmospheric pollutants from Chinese coal-fired power plants. Atmos. Environ. 44, 1515–1523.
Zhao, Y., Nielsen, C.P., Lei, Y., McElroy, M.B., Hao, J., 2011. Quantifying the uncertainties of a bottom-up emission inventory of anthropogenic atmospheric

pollutants in China. Atmos. Chem. Phys. 11, 2295–2308.
Zhao, Y., Nielsen, C.P., McElroy, M.B., Zhang, L., Zhang, J., 2012. CO emissions in China: uncertainties and implications of improved energy efficiency and emission control. Atmos. Environ. 49, 103–113.
Zhao, Y., Zhang, J., Nielsen, C.P., 2013. The effects of recent control policies on trends in emissions of anthropogenic atmospheric pollutants and CO_2 in China. Atmos. Chem. Phys. 13, 487–508.
Zhao, Y., Qiu, L.P., Xu, R.Y., Xie, F.J., Zhang, Q., Yu, Y.Y., Nielsen, C.P., Qin, H.X., Wang, H.K., Wu, X.C., Li, W.Q., Zhang, J., 2015. Advantages of a city-scale emission inventory for urban air quality research and policy: the case of Nanjing, a typical industrial city in the Yangtze River Delta, China. Atmos. Chem. Phys. 15, 12523–12644.
Zheng, J., Zhang, L., Che, W., Zheng, Z., Yin, S., 2009. A highly resolved temporal and spatial air pollutant emission inventory for the Pearl River Delta region, China and its uncertainty assessment. Atmos. Environ. 17, 1227–1239.
Zheng, J., Zheng, Z., Yu, Y., Zhong, L., 2010. Temporal, spatial characteristics and uncertainty of biogenic VOC emissions in the Pearl River Delta region, China. Atmos. Environ. 44, 1960–1969.
Zheng, J., Yin, S., Kang, D., Che, W., Zhong, L., 2012. Development and uncertainty analysis of a high-resolution NH_3 emissions inventory and its implications with precipitation over the Pearl River Delta region, China. Atmos. Chem. Phys. 12, 7041–7058.
Zhou, Y., Zhao, Y., Mao, P., Zhang, Q., Zhang, J., Qiu, L., Yang, Y., 2017. Development of a high-resolution emission inventory and its evaluation and application through air quality modeling for Jiangsu Province, China. Atmos. Chem. Phys. 17, 211–233.