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Toward a better practice for estimating the CO_2 emission factors of cement production: An experience from China

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ABSTRACT

CO_2 emission estimation of cement production is an underlying tool to identify the CO_2 sources, to evaluate the reduction efforts and to set prospective reduction targets for the Chinese cement industry. However, lack of domestic data has limited the reliability and accuracy of CO_2 emission estimation for Chinese cement industry. To develop an accurate and comprehensive CO_2 emission factor for Chinese cement industry, this study established a factory-level database of 197 cement production lines from 21 provinces covering various capacity scales. On the basis of this database, process, fuel, electricity and synthesized emission factor were computed. Furthermore, bootstrap simulation and Monte Carlo simulation were applied to evaluate the uncertainty of these factors. After corrections for cement kiln dust (CKD), incomplete decomposition, organic carbon and inorganic carbon, the medians of process, fuel and direct emission factors are 525, 369, and 919 kg CO_2/t clinker, respectively. Electricity emission factor is 74.9 kg CO_2/t clinker. The final synthesized emission factor for cement product is 761 kg CO_2/t cement with uncertainties of [−34.8%, +31.69%]. In this study, two revised calculation methods for the process emission factor are applied. Two calculation methods for the fuel emission factor are adopted as well. These practices seek to improve the reliability and accuracy of cement CO_2 emission factor. The simulated results indicate that the revised output method produces more accurate estimation for the process emission factor than the revised input method and unrevised output method. For fuel emission factor, the CC (carbon content) method is more accurate than the NCV (net calorific value) method. Simulated results are also compared with other authoritative estimation to validate the reliability of the CO_2 emission factors calculated in the present study.

1. Introduction

Cement production is one of the principal anthropogenic sources for CO_2 emissions (Gregg et al., 2008). It was estimated that CO_2 emissions from fossil fuel combustion and industrial processes contributed about 78% to the total GHG emission increase between 1970 and 2010 (IPCC, 2014), of which the cement sector is roughly responsible for 5–8% (Kajaste and Hurme, 2016). This means that the cement industry alone has a significant impact on environment (Mikulčić et al., 2016). Given large amounts of CO_2 released from cement production (Lei et al., 2011) and the foreseeable growth of cement demand (Cao et al., 2016), cement manufacture is inescapably identified as one of the key categories in a national CO_2 emission inventory, especially in emerging countries.

The cement industry plays a pivotal role in Chinese CO_2 emissions (Xi et al., 2013). China is by far the world's leading cement producer and consumer (Huisingh et al., 2015), due to unprecedented construction in China since the late 1970s (Liu et al., 1995). Recent past has seen a huge growth in Chinese cement production. According to the USGS cement statistics report, the cement output of China has soared from 1040 Mt in 2005–2500 Mt in 2014, accounting for more than half of global output (Van Oss, 2007; Van Oss, 2015). Cement production is both energy-intensive and emission intensive (Shen et al., 2014) and contributes 7% of total energy use (Zhang et al., 2015) and 15% of national emissions (Wen

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et al., 2015). Thus, dramatic growth of cement yield has led Chinese cement industry to be the focal industry for national low carbon development. Based on estimation of the initial and second 'The People's Republic of China: National Communication on Climate Change', cement production is the largest industrial process emitter (NDRC, 2004, 2013b). The cement industry has been pushed to implement low-CO_2 economic restructuring policy by the Chinese government. Through industry plans and energy consumption norm, Chinese cement industry has achieved remarkable progress in promotion of energy efficiency and CO_2 emission intensity (Cai et al., 2009). Nevertheless, the average CO_2 intensity of Chinese cement industry still remains behind the international advanced level (Xi et al., 2013). To provide a baseline for CO_2 emission control targets and crediting of carbon reduction in the cement industry, an accurate and comprehensive CO_2 emission accounting is a precondition.

In light of the importance of the CO_2 emission accounting, the estimation of Chinese cement CO_2 emission has garnered attention from numerous organizations and scholars. It is noted that diverse emission factors and accounting methods are used in various researches, through preliminary comparative study of different calculating methods (Ke et al., 2013). In the earlier researches, a default emission factor (i.e., 0.5 kg/kg clinker) of cement clinker was applied to make a crude approximation of total CO_2 emission from cement production (Worrell et al., 2001; Van Oss and Padovani, 2003). Due to complex material and energy flow in cement production (Wang et al., 2013), a three-tier methodology to estimate the cement CO_2 emission is developed by Intergovernmental Panel on Climate Change (IPCC) in 1996 and revised step by step (IPCC, 1996, 2000; 2006). Thereafter, prevailing emission factors for estimating the CO_2 emission of Chinese cement production is derived from the IPCC guideline (e.g., Hasanbeigi et al., 2013; Kim and Worrell, 2002; Lei et al., 2011; Li et al., 2014; Shen et al., 2015; Wang et al., 2013; Xu et al., 2012; Zhao et al., 2012). A default emission factor is usually adopted in the preceding studies. Several researchers (e.g., Gao et al., 2015; Ke et al., 2012; Shen et al., 2014) opted to adopt the CSI calculation standard instead of IPCC. Gao et al. (2015) and Shen et al. (2014) have established the calculation methodology based on the CSI calculation standard. Ke et al. (2012) adopted the default value from CSI to calculate the process emission of Chinese cement production. The Cement Sustainability Initiative (CSI), which is the authoritative cement organization under auspice by the World Business Council for Sustainable Development (WBCSD), has established a CO_2 Accounting Protocol for cement industry (Rehan and Nehdi, 2005). This method, essentially, is conformable with the calculation principle in IPCC (CSI, 2011). Cai et al. (2015) has investigated the process-related and fuel-related CO_2 emission factor of Chinese cement industry from an enterprise perspective. However, the indirect emission caused by electricity consumption is not included in Cai et al.'s study and in-depth statistical analysis for uncertainty within various CO_2 emission factors is still lacking.

Reliance on global default emission factors is the most significant limitation in previous studies of Chinese cement CO_2 emissions. The cement CO_2 emission accounting methodologies proposed by IPCC and CSI are the benchmarking in cement industry. On the ground of the IPCC and CSI benchmarking, a draft accounting guideline has been developed by China National Development and Reform Commission (NDRC, 2013a). However, these accounting methodologies have not provided a specific CO_2 emission factor for Chinese cement industry. Domestic data are lacking and the uncertainties of the preceding three methodologies should be comprehensively evaluated. According to the definition of 'good practice' in Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories, the accounting methodologies of CO_2 emissions should be applicable while the data quality and uncertainty should be controlled as far as possible (Penman et al., 2000). To this end, the present work first prepares a factory-level CO_2 emission factor database of 197 cement production lines from 21 provinces covering detailed parameters which provide comprehensive and transparent information of the emission factors. On the basis of the domestic database, the inherent uncertainties of direct (including process and fuel), indirect and synthesized CO_2 emission factors of Chinese cement production are to be evaluated by bootstrap simulation and Monte Carlo simulation. The process CO_2 emission factors are cross-checked through forward and reverse calculation methods. For fuel CO_2 emission factors, the calculation results of Carbon Content (CC) method and Net Calorific Value (NCV) method are crosschecked. In addition, calculation results are compared with other data sources.

2. Scopes for cement CO_2 emission accounting

Cement production usually can be divided into three basic stages: raw material preparation (stage 1), clinker calcination (stage 2) and cement grinding (stage 3) (Fig. 1). According to the definition of different emission sources, CO_2 emissions from thermal decomposition of limestone and burning of fuel in a cement kiln can be classified as direct emissions (WBCSD/WRI, 2014). Direct emissions are largely produced in the preheater, precalcinator and pyro-processing kiln within the stage 2 (CSI, 2011). Indirect CO_2 emissions refer to electricity purchased from outside and consumed throughout the whole procedure of cement production. Furthermore, direct emissions can be classified as two components: process emissions and fuel emissions.

Consistent accounting and evaluation for CO_2 emissions depends on a clear understanding of system boundaries (Marland, 2008). Knowing the system boundary of CO_2 emissions sources is the foundation for accurate accounting, evaluation and comparison. As shown in Table 1, system boundaries of the two representative accounting systems (i.e., IPCC and CSI) are inconsistent. Obviously, unclear definition of the system boundary of CO_2 emissions might lead to overestimation or underestimation, further, unreliability in comparative analysis (Josa et al., 2004, 2007). Without consistent definition of the system boundary, it is not easy to completely evaluate and compare the CO_2 emissions from different entities (Gartner, 2004). In terms of this point, the system boundary of cement production should be specified ahead of the CO_2 emission accounting.

CO_2 emissions for cement production result not only from kiln operations, but also from upstream and downstream processes (CSI, 2011). Neglect of several types of indirect emissions might lead to deviation in mitigation policy (Liu et al., 2015a). A broader system boundary should be set up to estimate the CO_2 emissions in cement production, thus, an inclusive practice is to expand the boundary to include other indirect activities (Matthews et al., 2008). Hence, the CO_2 emissions of cement production not only include the following installations: raw material quarrying and preparation, clinker production, grinding of clinker and cement substitutes, but also clinkers purchased from other factories and electricity purchased from the grid and on-site power generation[1] should be incorporated into the CO_2 emission. This practice also complies with the criterion put forward by National Development and Reform Commission (NDRC, 2013a). In the present study, the accounting boundaries of various cement plants are explicitly

[1] The on-site power generation largely comes from the kiln waste heat, thus the CO_2 emission is to be subtracted when accounting the total CO_2 emissions of a plant.

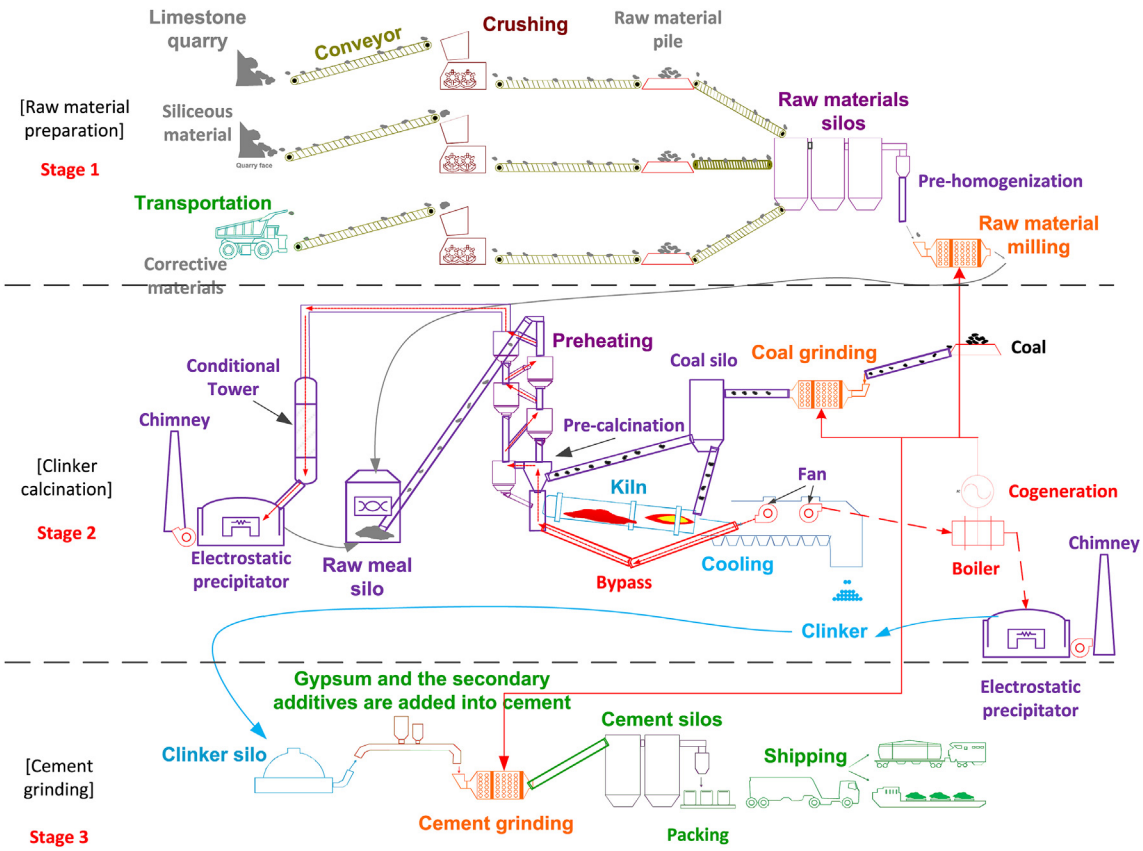

Fig. 1. Cement production procedure in a typical cement plant (New suspension preheater, NSP).

Table 1
Extension of system boundary for cement CO_2 accounting systems.

Emission sources	Emission components	IPCC	CSI	Extension in China
Direct emissions	Decomposition of carbonates in raw meal	✓	✓	✓
	Decomposition of cement kiln dust	✓	✓	✓
	Decomposition of inorganic carbon in fuels		✓	✓
	Burning of organic carbon in raw meal		✓	✓
	Burning of fossil fuels		✓	✓
Indirect emissions	Electricity consumption		✓	✓
	Waste heat recovery			✓
	Clinker purchased from outside		✓	✓

defined to keep the consistency of estimation results.

3. Accounting methods for emission factors

There exist two methods to capture CO_2 emissions factors of cement production: measurement versus calculation. The calculation-based method stems from conservation of matter (CSI, 2011) while the measurement-based method is based on continuous monitoring of the rate of gas discharge and the concentration of CO_2 in the discharged gas (Marland et al., 2009). The high labor and financial costs of flux measurement is the primary limiting factors for popularity of measurement-based method (CSI, 2011). Furthermore, this method can't distinguish CO_2 emissions from different sources (i.e., process emission and fuel emission). Therefore, the calculation-based method is a better practice to estimate the CO_2 emissions of the cement industry than measurement-based method. To adapt to the data availability in Chinese cement industry, the calculation-based methods proposed by IPCC and CSI are revised in this section.

3.1. Process emission factors

3.1.1. Fundamentals for calculation of process emission

Process CO_2 emission of cement production is mainly originated from the decomposition of carbonate. The core principle to calculate this CO_2 emission is to establish the mass equilibrium of input feedstock and output product. The elementary chemical reaction in cement production is decarbonation of $CaCO_3$ and $MgCO_3$ (Benhelal et al., 2013):

$$CaCO_3 \rightarrow CaO + CO_2\uparrow$$

$$MgCO_3 \rightarrow MgO + CO_2\uparrow$$

There are two types of often-used calculation-based methods to account the industrial CO_2 emission: input approach and output approach (Table 2). Units of the emission factors, employed by diverse methods, should match the types of activity data used in these methods. Selection of calculation methods depends on the

Table 2
Categorization of industrial CO_2 emission calculation methods from IPCC and CSI.

Organization	Method	Types of calculation methods		Types of activity data		
		Input	Output	Carbonate-based	Clinker-based	Cement-based
IPCC	Tier 1		✓			✓
	Tier 2		✓		✓	
	Tier 3	✓		✓		
CSI	Method A1	✓		✓		
	Method A2	✓		✓		
	Method B1		✓		✓	
	Method B2		✓		✓	

availability of activity data.

Input methods directly calculate process emissions based on the carbonates consumed and the corresponding emission factors of carbonates. Conversely, output method aims to deduce process emissions from the clinker output and the content of decomposed oxides in clinker, e.g., calcium oxide (CaO) and magnesium oxide (MgO). If lack of clinker data, a rough calculation-based on cement might be an expedient. In terms of the chemical balance, the output method and input method can be expressed by Eq. (1) or Eq. (2):

$$E_{out} = \left(Clinker_{CaO} \times \frac{44}{56} + Clinker_{MgO} \times \frac{44}{40}\right) \times 1000 \times Output_{clinker} \quad (1)$$

$$E_{in} = \left(Carbonate_{CaO} \times \frac{44}{56} + Carbonate_{MgO} \times \frac{44}{40}\right) \times 1000 \times Consumption_{carbonate} \quad (2)$$

where E_{in} refers to industrial CO_2 emissions with the input method, kg CO_2; E_{out} refers to industrial CO_2 emissions with the output method, kg CO_2; $Clinker_{CaO}$ and $Clinker_{MgO}$ refer to the share of CaO and MgO in clinker, %; $Output_{clinker}$ refers to clinker output, tonnes; $Carbonate_{CaO}$ and $Carbonate_{MgO}$ refer to the share of CaO and MgO in carbonate, %; $Consumption_{carbonate}$ is the carbonate consumed in cement production,[2] tonnes; 44/56 and 44/40 refer to the mole ratios of CO_2 to CaO and CO_2 to MgO, respectively.

Theoretically, the calculation results of input and output are equivalent (CSI, 2011), whereas the starting points of these two methods are different. The former is forward-deducing and the latter is reverse-deducing. The output method is based on an emission factor of clinker and clinker output while the input method generates a carbonate-based emission factor. For output method, the statistical data of clinker output is available, which provides the best activity data for industrial emission accounting (Cai et al., 2015). However, unlike the output method, it is deemed impractical to compute the quantity of carbonates decomposed in input method due to its extensive data requirements.

[2] Sometimes, a certain amount of carbonate is blended into cement without decomposition. This should not be considered into the industrial emission.

3.1.2. Revised input method from carbonate-based to clinker-based

The input method requires taking full consideration of all species of carbonates (IPCC, 2006), of which the data in China are not available. Therefore, the input method proposed by IPCC and CSI should be amended into a clinker-based one for better practice as in Eq. (3) and Eq. (4):

$$EF_{in} = \frac{\left(Carbonate_{CaO} \times \frac{44}{56} + Carbonate_{MgO} \times \frac{44}{40}\right) \times RM_{carbonate} \times (1 - Ash_{coal})}{1 - LOI_{RM}} \times 1000 \quad (3)$$

$$Ash_{coal} = Ash_{content} \times EI \quad (4)$$

where EF_{in} refers to process emission factor with the input method, kg CO_2/t clinker; $RM_{carbonate}$ refers to the share of carbonate in raw materials, %; LOI_{RM} refers to loss on ignition (LOI) of raw meal, %; Ash_{coal} refers to the share of coal ash added into clinker blends since burned coal leaves the coal ash into the clinker, %; $Ash_{content}$ refers to ash content in coal, %; EI refers to fuel consumption per unit of clinker product, t/t clinker.

3.1.3. Revision on output method for no-carbonate substitution in raw meal

The non-carbonate CaO and MgO should be deducted from the share of CaO and MgO in the clinker. Thus, a revision of the output method is presented in Eq. (5), Eq. (6) and Eq. (7):

$$EF_{out} = \left(RClinker_{CaO} \times \frac{44}{56} + RClinker_{MgO} \times \frac{44}{40}\right) \times 1000 \quad (5)$$

$$RClinker_{CaO} = Clinker_{CaO} - Ash_{coal} \times Ash_{CaO} - \frac{NCarbonate_{CaO}}{1 - LOI_{RM}} \times (1 - Ash_{coal}) \quad (6)$$

$$RClinker_{MgO} = Clinker_{MgO} - Ash_{coal} \times Ash_{MgO} - \frac{NCarbonate_{MgO}}{1 - LOI_{RM}} \times (1 - Ash_{coal}) \quad (7)$$

where EF_{out} refers to process emission factor with the output method, kg CO_2/t clinker; $RClinker_{CaO}$ and $RClinker_{MgO}$ represent the

revised CaO and MgO content in clinker that comes from carbonates, respectively, %; Ash_{CaO} and Ash_{MgO} refer to the CaO and MgO content of coal ash, %; $NCarbonate_{CaO}$ and $NCarbonate_{MgO}$ refer to non-carbonate CaO content and MgO content in raw meal, respectively, %.

3.2. Fuel emission factors

Conceptually, the calculation of CO_2 emission from fossil fuels is calculated by the carbon content (CC) method (Marland and Rotty, 1984) as presented in Eq. (8):

$$EF_{CC} = EI \times C \times \frac{44}{12} \times FO \times 1000 \tag{8}$$

where EF_{CC} refers to the fuel emission factor with the CC method, kg CO_2/t clinker; C represents carbon content per unit mass of fuel, %;44/12 refers to the mole ratio of CO_2 to C; FO refers to oxidation fraction of fuel, %.

To accurately estimate CO_2 emissions from the fuel it is necessary to acquire the carbon content of the fuel consumed (Marland, 2012). Since carbon content of fuels might differ for different regions, domestic field tests are given preference to estimate Chinese fuel CO_2 emission (Zhao et al., 2012). The carbon content is not routinely tested for commercial coals (Quick and Glick, 2000). Therefore, it is rather prevailing to apply the net calorific value (NCV) method for Chinese cement industry than the CC method. It is a convention for a cement plant to control the net calorific value standard of fuel used in the kiln (Zhao and Wei, 2013). The CC-based calculation can be converted into a NCV-based version (Eq. (9)):

$$EF_{NCV} = EI \times NCV_f \times F_f \times FO \times 1000 \tag{9}$$

where EF_{NCV} refers to the fuel emission factor with the NCV method, kg CO_2/t clinker; NCV_f refers to net calorific value of per unit mass of fuel, TJ/kg (1 TJ = 10^6 MJ); F_f refers to CO_2 emission on a per unit calorific value of fuel, t CO_2/TJ.

3.3. The corrections for emission factors

In any case, the dust leaving the kiln, the organic carbon in raw materials and incomplete calcination should not be neglected. Corrections for these three aspects should be incorporated into the scope of accounting as the dust leaving the kiln, the organic carbon in raw materials and incomplete calcination will have impact on the direct emissions.

3.3.1. Cement kiln dust (CKD)

The non-recycled CKD is treated as a 'loss' in CO_2 emission accounting. The loss can be traced by chemical balance as shown in Eq. (10):

$$\lambda = 1 - \frac{Clinker_{CaO}}{\frac{RM_{CaO}}{1-LOI_{RM}} + Ash_{coal} \times Ash_{CaO}} \tag{10}$$

where λ refers to the proportion of non-recycled CKD, %; RM_{CaO} refers to CaO content of the raw meal, %.

In terms of different accounting methods, corrections for CKD are adverse. As presented in Eq. (11) and Eq. (12), the emissions from CKD are a subtraction in input method, but an addition in output method. The opposite computation reflects the mass flow in cement production. If neglecting the non-recycled CKD, in another word, the initial input method has overestimated the industrial CO_2 emission. In contrast, the original output method has underestimated the industrial CO_2 emission.

$$CKD_{in} = 1 - \lambda \times (1 - F_{CKD}) \tag{11}$$

$$CKD_{out} = 1 + \lambda \times F_{CKD} \tag{12}$$

where CKD_{in} refers to CKD correction ratio for input-based industrial emission factor, %; CKD_{out} refers to CKD correction ratio for output-based industrial emission factor, %; F_{CKD} represents the calcination fraction of non-recycled CKD, the default value is set to 85% since approximately 85% decomposition of carbonate takes place before entering into the kiln (Gao et al., 2016).

3.3.2. Incomplete decomposition

Commonly, complete decomposition is one of the assumptions in CO_2 emission calculation since carbonates are decomposed at high temperature and for a long time (Li et al., 2014). Nevertheless, the decomposition rate of carbonate material can't reach 100%. It is found that the loss on ignition of clinker is commonly at the rate below 1%. Likewise, loss on ignition of clinker is one of the routine indicators tested by the cement plant in China. Thus, a calcination fraction modification for the industrial emission based on the loss on ignition in clinker is recommended.

3.3.3. Organic carbon in raw materials

The raw materials may contain a proportion of organic carbon, which would yield additional CO_2 emission (IPCC, 2006). The corresponding CO_2 emission from burned organic carbon is computed with Eq. (13):

$$EF_{organic} = \frac{1 - Ash_{coal}}{1 - LOI_{RM}} \times R_{OC} \times \frac{44}{12} \times 1000 \tag{13}$$

where $EF_{organic}$ refers to CO_2 emission factor from burned organic carbon, kg CO_2/t clinker; $1-Ash_{coal}/1-LOI_{RM}$ refers to raw material clinker mass ratio, %; R_{OC} refers to the content of organic carbon in raw material, %; 44/12 refers to the mole ratio of CO_2 to C.

3.3.4. Inorganic carbon in fuels

There exists a certain amount of inorganic carbon (TIC) in fuel consumed in Chinese cement industry (Geng et al., 2015). Normally, to maximize the profit, coal suppliers adulterate washed coal with coal gangues which encompass a portion of carbonate. The decomposition of inorganic carbon in fuel should be categorized as industrial emission. The computation of TIC is presented in Eq. (14).

$$EF_{TIC} = EI \times R_{TIC} \times \frac{44}{60} \times 1000 \tag{14}$$

where EF_{TIC} refers to the emission factor of TIC, kg CO_2/t clinker; R_{TIC} refers to the content of inorganic carbon in fuel, %; 44/60 refers to the mole ratio of CO_2 to CO_3^{2-}.

3.4. Electricity emission factor

CO_2 emission of electricity consumed in cement production is defined as an indirect source. CO_2 emission factors of electricity are different because of various fuel mix and technologies adopted in the generation of electricity (Zhang et al., 2009). Since the provincial electricity emission factor of 2011–2013 is not available, the electricity emission factor from Baseline Emission Factor for Regional Power Grids in China is adopted. In addition, the waste heat recovery (WHR) power generation should be taken into consideration when accounting for electricity consumption.

3.5. Synthesized emission factor for cement production

With a clinker fraction in cement, the process, fuel and electricity emission factors can be integrated into a synthesized emission factor for cement product. Accounting for import or export of clinker in a production line is crucial to determine an accurate clinker/cement ratio. The actual clinker/cement ratio is calculated as Eq. (15).

$$EF_{cement} = \frac{Pro_{clinker} - Exp + Im}{Pro_{cement}} \times \left(EF_{pro} \times F_{CKD} \times F_{IC} + EF_{organic} + EF_{TIC} + EF_{fuel}\right) + EF_{ele} \quad (15)$$

where EF_{cement} refers to emission factor of cement product, kg CO_2/t cement; $Pro_{clinker}$ refers to clinker production, tonnes; Exp refers to export of clinker, tonnes; Im refers to import of clinker, tonnes; Pro_{cement} refers to cement production, tonnes; EF_{pro} refers to process emission factor, kg CO_2/t clinker; F_{CKD} refers to CKD correction fraction, %; F_{IC} refers to incomplete decomposition correction fraction, %; $EF_{organic}$ refers to organic carbon emission factor, kg CO_2/t clinker; EF_{TIC} refers to inorganic carbon emission factor, kg CO_2/t clinker; EF_{fuel} refers to fuel emission factor, kg CO_2/t clinker; EF_{ele} refers to electricity emission factor, kg CO_2/t cement.

3.6. Data collection and uncertainty

With the deployment of new technology and retrofitting of the outdate production lines in Chinese cement industry, the portion of cement clinker produced by new suspension preheater (NSP) cement production lines has by far reached 96.2% (CCA, 2015). During 2011–2013, the number of cement production lines under operating is 1428, 1537 and 1587, respectively. Supposing that 1587 is the population sample size, the required minimum sample size is 96 under a desired relative margin of error at 10%, a desired relative population standard deviation at 50% and a confidence level of 95%. Considering the data recovery, the required minimum sample size should be magnified 10%. Therefore, the required minimum sample size is 106. Fortunately, 197 valid samples of NSP cement production lines have been collected, which are far more than the required minimum sample size.

To ensure the completeness of all samples in the database, year-round of chemical data and production data of a selected year were collected from the historic accounting documents of these production lines during 2011–2013. The chemical data is collected from the assay laboratory of cement plants while the production data is collected from the production department of cement plants. The year-round data covers 12 months in a selected year. In the selected year, cement production lines were operating under normal conditions. The database contains detail parameters for accounting CO_2 emissions in cement production. The chemical data include chemical composition of raw materials, raw meal and clinker. The production parameters include geographic locations, addresses, capacity, clinker output, cement output, fuel consumption, and electricity consumption of each production stage (consisting of raw material preparation, clinker calcination, cement grinding and other processing). Sampled cement production lines have covered 21 provinces (Table 3) in which the cement output account for about 90% of national output in 2011 (NBSC, 2012).

Monte Carlo simulation is usually used to analyze the uncertainties of the emission factors (Zhao et al., 2011). The input parameters with corresponding probability density functions (PDFs) are placed into the Monte Carlo framework. The PDF of input parameters is estimated from the collected data. The estimation of PDF of input parameters follows three steps:

(1) The initial PDFs for an input parameter are obtained by maximum likelihood estimation and bootstrap simulation (Frey and Bammi, 2002).
(2) According to three classical statistics (i.e., Cramer-von Mises, Kolmogorov-Smirnov and Anderson-Darling statistics) and two fitting evaluation criteria (Aikake's Information Criteria and Bayesian Information Criteria), the best-fitting PDF of the input parameter is selected (Delignette-Muller and Dutang, 2014).
(3) With 1000 times of bootstrap simulation, the PDF of the input parameter is inferred with the median of bootstrap simulation results.

The normal distribution is the most appropriate distribution for many categories of emission inventories (IPCC, 2006). However, input parameters in cement CO_2 emissions factor are usually non-negative variables and known to be positively skewed. Skewed distributions, such as lognormal, Weibull and gamma can better fit data than symmetrical distribution. Owing to the length limitation of this paper, the net calorific value of coal is selected as the instance to explain how to select a distribution function that can better fit data. As presented in Table 4, goodness-of-fit indicators of this input parameter give preference to the Weibull distribution since the goodness-of-fit statistics and criteria are relatively lower than other distributions.

Once a particular parametric distribution is selected, random sample sets of the same sample size as the original observed data are drawn from the assumed distribution (Frey and Zheng, 2002). The random sample sets are simulated by bootstrap sampling technology and the median of bootstrap samples is adopted. For parameters that lack domestic data, the PDFs of these parameters are obtained from existing literatures. Table 5 summarizes the PDFs of all input parameters in the present study. It is shown that asymmetric distribution is more suitable when the ratio of standard deviation to mean of original samples is rather large.

4. Results and discussion

In this section, 100,000 times Monte Carlo simulation are performed to obtain emission factors of cement production. Furthermore, the simulated results are compared with other data sources.

4.1. Process emission factor

Simulation results of the revised input method (Eq. (3)) and the revised output method (Eq. (5)) are shown in Fig. 2. To validate whether CO_2 emissions overestimated by CaO and MgO content in non-carbonate have been eliminated, the simulation results of unrevised output method are presented as well.

The solid lines trace the median of simulation results. The medians of revised input and revised output methods are 519 and 525 kg CO_2/t clinker while that of unrevised output method is 535 kg CO_2/t clinker. The two revised approaches yield almost identical estimates of process emission factors with a gap of 6 kg CO_2/t clinker. Compared to unrevised output method, the two revised approaches could rectify the deviation of 16 and 10 kg CO_2/t clinker. This suggests that revisions for process emission accounting methods could avoid overestimation of from non-carbonate.

The dashed lines represent the 95% confidence intervals (CIs) of the simulation results. The relative uncertainties (95% CIs) of revised input, revised output and unrevised output methods are

Table 3
Distribution of sampled cement production lines.

Provinces	Sample numbers	Provinces	Sample numbers	Provinces	Sample numbers
Beijing	1	Henan	10	Guangdong	9
Xinjiang	3	Shanxi	8	Yunnan	10
Liaoning	14	Hebei	10	Fujian	8
Sichuan	13	Shaanxi	9	Jiangxi	7
Shandong	12	Hubei	8	Hunan	11
Zhejiang	8	Anhui	14	Inner Mongolia	14
Jiangsu	6	Guangxi	10	Guizhou	12

Table 4
Goodness-of-fit statistics and criteria for net calorific value of coal.

	Weibull	Gamma	Lognormal	Normal
(a) Goodness-of-fit statistics				
Kolmogorov-Smirnov statistic	0.032	0.071	0.078	0.058
Cramer-von Mises statistic	0.049	0.455	0.608	0.229
Anderson-Darling statistic	0.349	0.929	3.889	1.496
(b) Goodness-of-fit criteria				
Aikake's Information Criterion	1766	1802	1815	1783
Bayesian Information Criterion	1773	1809	1823	1790

[-18.8%, 22.4%], [-3.58%, 1.85%] and [-4.70%, 8.12%], respectively. Uncertainties of revised output method are relatively small. This is because chemical compositions of clinker are required to fall into the range of national standard. The distribution of process emission factor obtained by the revised output is asymmetric because asymmetric properties of non-carbonate CaO content and MgO content in raw meal have been propagated to process emission factor. The logarithmic standard deviations of these two parameters are 0.93 and 0.67, resulting in skewed distributions. Given the relative narrower confidence intervals of revised output method, revised output method is given preference to process emission accounting and should be served as the parameter for calculation of synthesized emission factors.

4.2. Fuel emission factor

Analogous comparison is performed for fuel emission factor which is shown in Fig. 3. The simulation results present consistent estimates for the fuel emission factor. The medians of CC and NCV methods are 369 and 364 kg CO_2/t clinker, respectively. The relative uncertainties (95% CIs) of these two methods are [-35.7%, 39.8%]

Table 5
PDFs of input parameters for Chinese cement CO_2 emission factor.

Parameters	Distribution	Characteristics for distribution functions		Data sources	Descriptive statistics of original samples	
		Characteristic A	Characteristic B		Mean	Standard deviation
(a) Parameters for input method						
CaO content in carbonate (%)	Normal	49.79	2.12	Simulated by authors	49.75	2.18
MgO content in carbonate (%)	Gamma	2.42	3.48	Simulated by authors	1.44	0.75
Carbonate in raw meal (%)	Lognormal	4.44	0.05	Simulated by authors	84.92	4.54
(b) Parameters for output method						
CaO content in clinker (%)	Normal	65.27	0.79	Simulated by authors	65.26	0.81
MgO content in clinker (%)	Lognormal	0.70	0.45	Simulated by authors	2.22	0.96
No-carbonate CaO content in raw meal (%)	Lognormal	−0.83	0.93	Simulated by authors	1.02	1.54
No-carbonate MgO content in raw meal (%)	Lognormal	−1.71	0.67	Simulated by authors	0.25	0.19
CaO content in coal ash (%)	Gamma	0.50	1.86	Simulated by authors	4.59	4.41
MgO content in coal ash (%)	Gamma	2.99	2.85	Simulated by authors	1.27	1.35
(c) Parameters for both of input and output methods						
Ash content of coal (%)	Gamma	0.25	5.10	Simulated by authors	20.89	9.93
Loss on ignition (LOI) of raw meal (%)	Normal	35.55	0.68	Simulated by authors	35.48	0.86
(d) Parameters for corrections						
CaO content in raw meal (%)	Normal	43.46	0.84	Simulated by authors	43.46	0.91
Loss on ignition (LOI) of clinker (%)	Gamma	18.53	5.67	Simulated by authors	0.31	0.14
Organic carbon content in raw material (%)	Weibull	0.18	1.49	Simulated by authors	0.19	0.15
Inorganic carbon in fuel (%)	Uniform	0.39	3.12	Geng et al., 2015		
(e) Parameters for fuel emission factors						
Fuel consumption per unit clinker (kg/t clinker)	Lognormal	5.02	0.12	Simulated by authors	153.97	20.62
Carbon content in coal (%)	Weibull	71.62	8.70	Simulated by authors	66.94	10.39
Net calorific value of coal (MJ/kg)	Weibull	27.34	7.82	Simulated by authors	25.36	4.45
Net carbon content (t C/TJ)	Lognormal	3.26	0.04	Simulated by authors	26.61	2.45
Oxidation rate (%)	Normal	98	1	CSI, 2011		
(f) Parameters for electricity emission factors						
Electricity emission factor of cement product (kg CO_2/t cement)	Weibull	84.37	3.14	Simulated by authors	82.17	38.53
Clinker/cement ratio	Weibull	0.78	9.54	Simulated by authors	0.74	0.09

Note:
a. For normal distribution, characteristic A and characteristic B refer to mean and standard deviation, respectively.
b. For uniform distribution, characteristic A and characteristic B refer to lower and upper boundary, respectively.
c. For lognormal distribution, characteristic A and characteristic B refer to mean and standard deviation, respectively.
d. For Weibull distribution, characteristic A and characteristic B refer to scale and shape, respectively.
e. For gamma distribution, characteristic A and characteristic B refer to rate and shape, respectively.
f. The details of distribution functions are presented in Appendix.

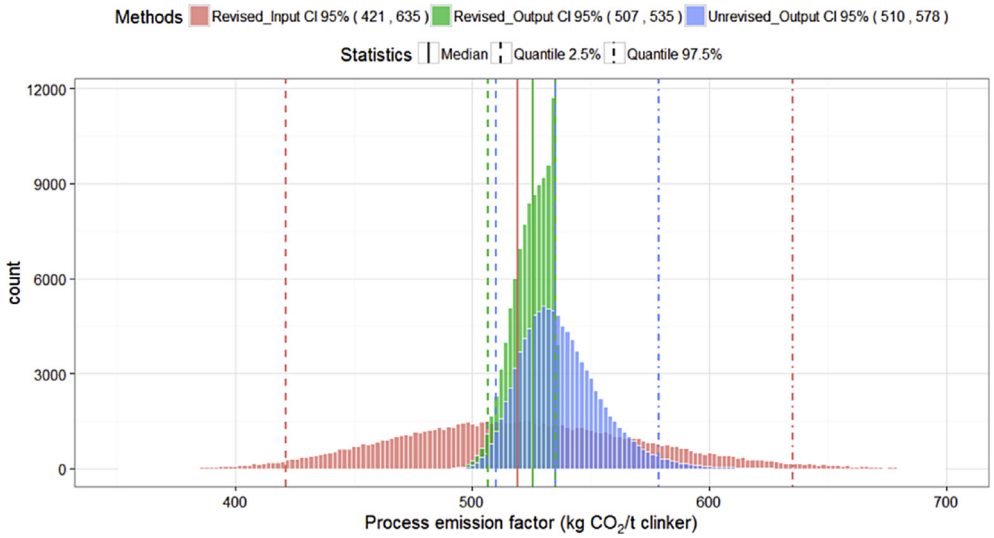

Fig. 2. Comparison between simulation results of revised input method, revised output method and unrevised output method.

and [-40.4%, 51.5%]. The one additional input parameter in the NCV method results in a wider range of uncertainties than the CC method. The unsubstantial difference of medians reflects that the NCV method can deduce relatively accurate estimate of fuel emission factor when carbon content data is not available. Results of the NCV method are acceptable; however, to produce a more precise estimation of synthesized emission factor, the results of the CC method are applied in the following sections. This is because the uncertainties of the CC method are relatively smaller.

4.3. Direct emission factor

The sum of process emission factor and fuel emission factor refers to the direct emission factor of cement production. Corrections for direct emission are presented in Table 6. The median of CKD correction fraction for input method is −0.507% with a 95% CIs of [-1.00%, +0.0108%] while the median of CKD correction fraction for output method is +2.87% with a 95% CIs of [-0.0613%, +5.67%]. IPCC provides a default CKD correction factor of +2%/-2% (IPCC, 2006), whilst the CKD correction calculation proposed by CSI is rather complicated for cement plants (CSI, 2011). The CKD largely originates from the upper kiln tail and lower kiln head. CKD released at the point of the upper kiln tail is partly calcinated. On the other hand, CKD released from the lower kiln head is fully calcinated and the composition of that is similar to clinker. Technically, a large portion of the cement kiln dust is reciprocated into the clinker-manufacturing loop through a dust catcher and bypass system (Maslehuddin et al., 2008). The degree to which the CKD is recycled depends on its composition (trace metal and contaminants) and the regional restriction for alkali content (Huntzinger and Eatmon, 2009).

Correction for incomplete decomposition, organic carbon and inorganic carbon are summarized in Table 6 as well. With the correction of CKD, incomplete decomposition, organic carbon and inorganic carbon, the distributions of direct emission factor are presented in Fig. 4. After corrections for CKD, incomplete decomposition, organic carbon and inorganic carbon, the median of direct emission factor is 919 kg CO_2/t clinker.

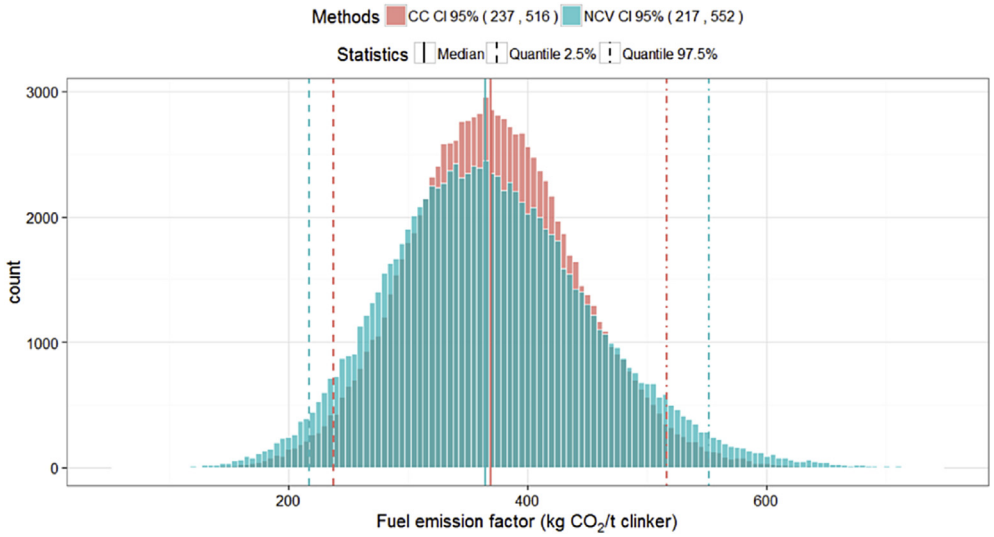

Fig. 3. Comparison between simulation results of the CC method and NCV method.

Table 6
Simulation results of corrections for process and fuel emission factor.

Corrections	Unit	Process/fuel	Median	95% CI
CKD	%	Process (input)	−0.507	[-1.00, +0.0108]
	%	Process (output)	+2.87	[-0.0613, +5.67]
Incomplete decomposition	%	Process	99.7	[99.4, 99.9]
Organic carbon	kg CO_2/t clinker	Fuel	7.72	[0.828, 23.8]
Inorganic carbon	kg CO_2/t clinker	Process	1.94	[0.504, 3.69]

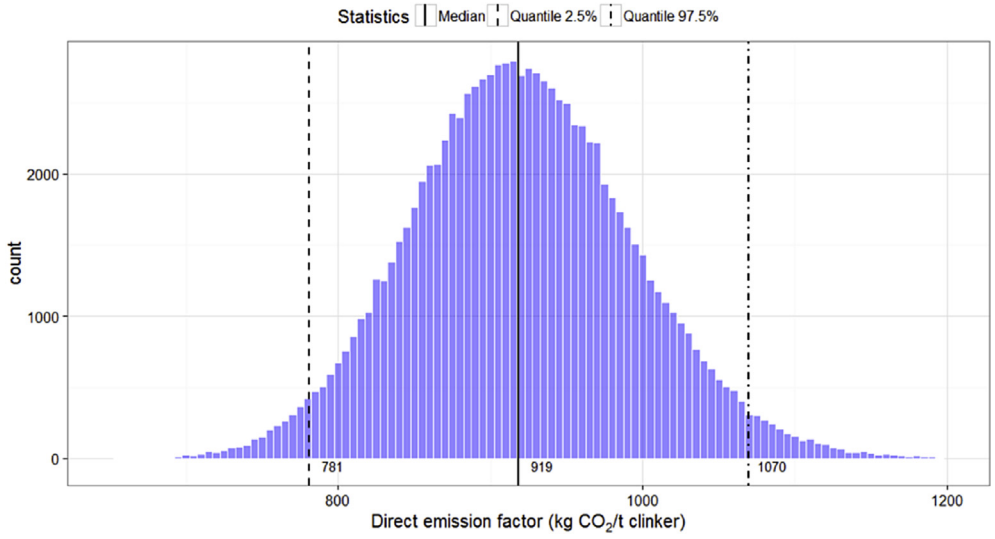

Fig. 4. Simulation result of direct emission factor for cement production.

4.4. Electricity emission factor and synthesized emission factor

Fig. 5 depicts the distributions of electricity emission factor and synthesized emission factor for cement product which is the final product of cement production. The median of electricity emission factor is 74.9 kg CO_2/t cement with a relative uncertainty (95% CIs) of [26.0, 128]. The synthesized emission factor of cement is derived based on direct emission factor, electricity emission and clinker/cement ratio. The median cement emission factor is 761 kg CO_2/t cement with relative uncertainties of [-34.8%, +31.6%].

4.5. Comparison with other sources

Emission factors of diverse data sources are presented in Fig. 6. The green bars are the medians of simulation results and the error bars represent the upper and lower bound of 95% CI. In the second report of 'The People's Republic of China: National Communication on Climate Change', the process CO_2 emission of cement in 2005 was 411.67 Mt while the clinker production was 674 Mt (NDRC, 2013b). Thus, 611 kg CO2/t clinker is simply assumed to be process emission factor in this authoritative report. IPCC provides a default value of 527 kg CO2/t clinker with default fraction of CaO (65%) and MgO (1.5%). CSI has released a database (Getting the Numbers Right, GNR) for CO_2 and energy performance of worldwide cement industry based on emissions data from individual cement production line (CSI, 2013). The GNR database covers 21% of global cement production. By 2013, 5 Chinese cement companies[3] (including 80 cement production lines) have participated into CSI and delivered data to this database.

For process emission factor, the simulation result is lower than other four data sources. The process emission factor from Chinese National Development and Reform Commission' report obviously deviates far from the real situation. The IPCC default value and CSI estimate are slightly higher. It is indicated that previous researchers have overestimated Chinese cement process emission by using the unrevised output method (Shen et al., 2014). Industrial by-products can be used as substitutes for traditional natural raw materials (Song et al., 2016), e.g., slag (Monshi and Asgarani, 1999) and fly ash (Gäbel and Tillman, 2005). Since alternative raw materials would affect the process CO_2 emissions (Cai et al., 2015; Oh et al., 2014), the non-carbonate CaO and MgO should be deducted.

For fuel emission factor, the underestimation of GNR database is mainly caused by the one-tail distribution of Chinese samples in GNR database. The Chinese members of CSI are 5 large cement enterprises which are more willing to adopt fuel conservation technologies. Thus, the emission factor of Chinese cement production from GNR database may not be representative (Li et al., 2013). The samples in the present study have included the medium-scale and small-scale cement production lines. For electricity emission factor in CSI, the cogeneration is not subtracted from the electricity consumption that leads to overestimation emission factor.

The process emission factor, fuel emission factor, electricity emission factor and clinker/cement ratio of GNR database are 536 kg CO_2/t clinker, 314 kg CO_2/t clinker, 93 kg CO_2/t cement and 72%. Thus, the synthesized emission factor of cement product within GNR database is 705 kg CO_2/t cement which is 7.36% lower than the median of simulation result.

[3] The 5 Chinese members of CSI are China Resources Cement, China National Building Material (CNBM), China National Materials Group (Sinoma), Tianrui Group and West China Cement.

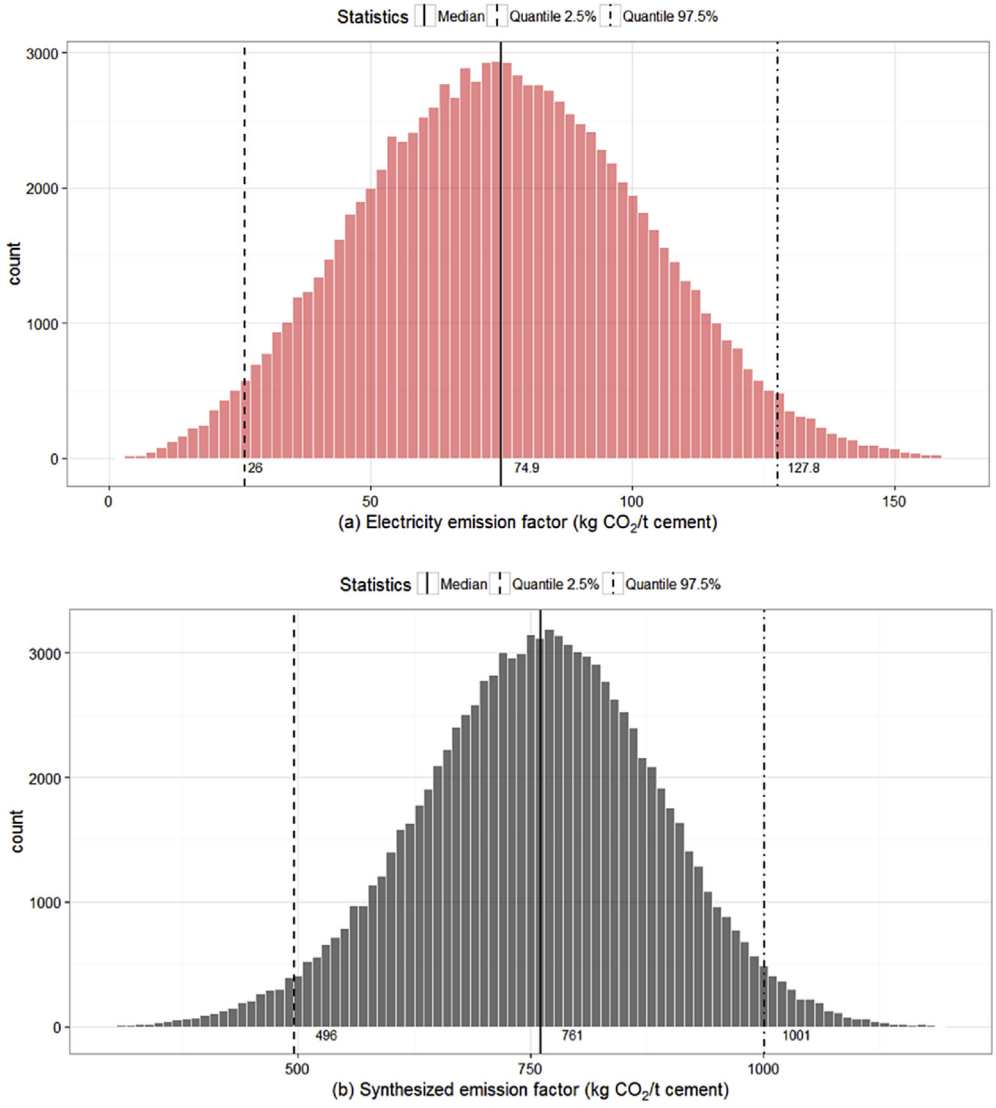

Fig. 5. Simulation results for electricity emission factor and synthesized emission factor.

4.6. Discussion

The data used in the two revised calculation methods for process emission factor are easily available from the cement plants in China. To control the quality of clinker and cement product, the laboratory in cement plant conventionally collects and assays the chemical content of raw materials, fuels, raw material mix and clinker several times a day. These two revised methods will not add burden on cement plants. Instead, the overestimation of industrial CO_2 emission from non-carbonate raw materials is avoided. The corollary is jointly proved by the results of the two revised methods.

The use of default emission factors would cause substantial uncertainty as long as the fuel qualities vary considerably across from the default value (Konstantinaviciute and Bobinaite, 2015). Previous studies of Chinese cement industry have estimated the coal-related CO_2 emission by various default emission factors (Table 7). These estimations will result in uncertainty in CO_2 emission accounting of cement production. The primary fuels used in Chinese cement production are thermal coals, mixed with a little proportion of low grade coal. It is noted that the coal quality in China has a significant discrepancy with that of the IPCC default value (Guan et al., 2012). Recently, a re-evaluation of Chinese coal is carried out based on 4845 coal samples in which the net carbon content per calorific value of Chinese coal is 26.32 or 26.59 t C/TJ (i.e., 96.51 or 97.50 t CO_2/TJ) within a 3% uncertainty (Liu et al., 2015b). In the present study, the median (simulated) of net carbon content per energy is 26.05 t C/TJ (95.52 t CO_2/TJ) and the 95% CI of net carbon content per calorific value is [24.07, 28.19] which are close to Liu et al.'s result. The slight difference can be attributed to that quality of coal burned in cement production is not exactly identical with the entire sectors. Future studies of Chinese cement CO_2 emissions should adopt country-specific fuel emission factor since cement industry is a major source of CO_2 emissions for China.

In the IPCC guideline, correction for cement kiln dust (CKD) has already been incorporated into the Tier 2 method (IPCC, 2006). Further, CSI has taken all of influential factors into consideration to ensure the completeness of the inventory (Ke et al., 2013). However, these corrections depend on extra test and computation that would be a burden for cement plants. For the principle of good practice, several simplified calculation methods developed by the present study might be appropriate in the current situation of Chinese

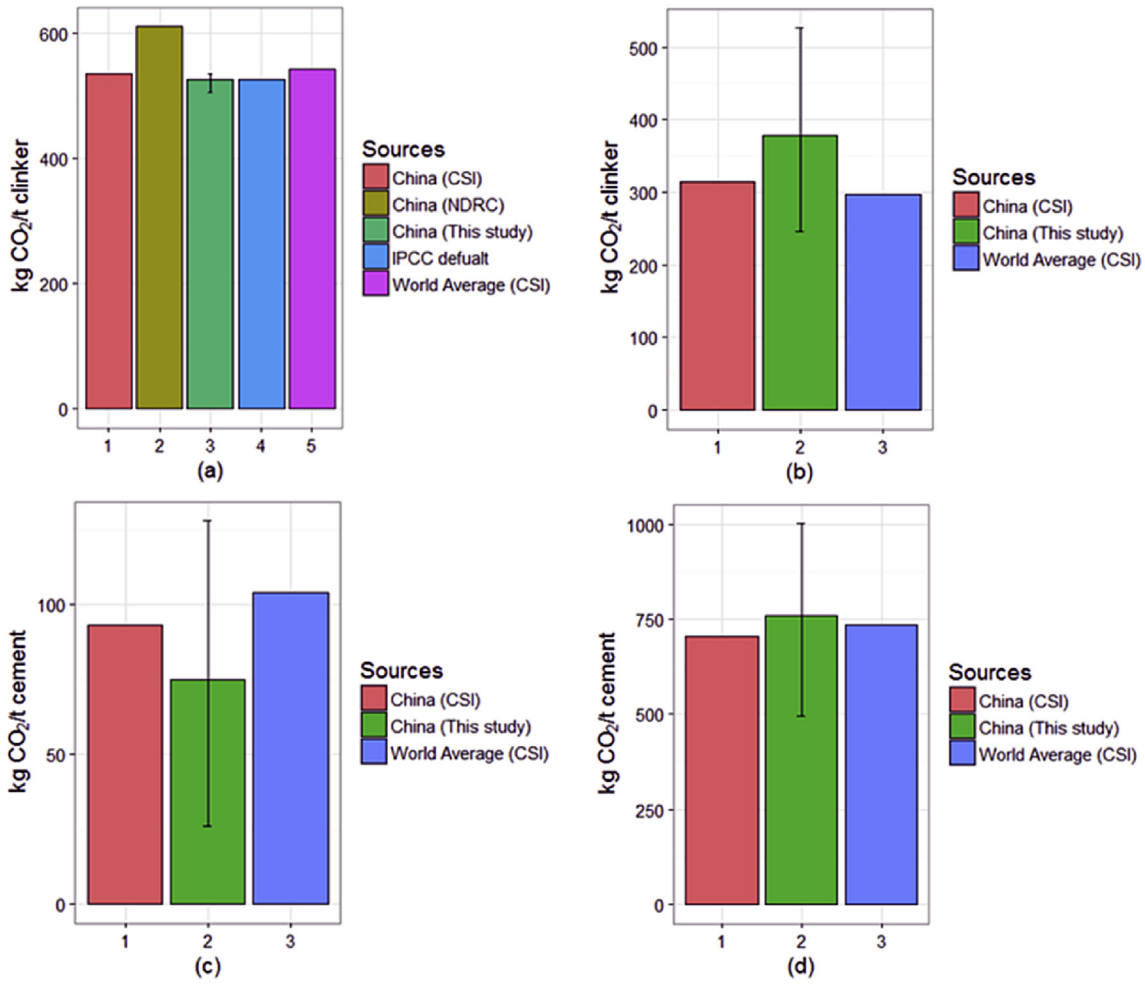

Fig. 6. Comparisons of (a) process emission factor, (b) fuel emission factor, (c) electricity emission factor and (d) synthesized emission factor of cement production in China between different sources and simulation results.

Table 7
Fuel CO_2 emission factors adopted in various studies.

F_f (kg CO_2/TJ)	Original emission factor	Data source
96,400	96.4 g CO_2/MJ	Kajaste and Hurme, 2016
92,800	92.8 kg CO_2/GJ	Xu et al., 2012
94,600	94.6 t CO_2/TJ[a]	Hasanbeigi et al., 2013
89,500–99,700	89.5–99.7 t CO_2/TJ	Ke et al., 2013
67,902–92,801	1.99–2.72 t CO_2/tce	Wen et al., 2015
92,128	2.7 t CO_2/tce[b]	Tan et al., 2015
95,700	26.1 kg C/GJ	Chen et al., 2015
83,826	2.4567 t CO_2/tce	Shen et al., 2015
_	2.6 t CO_2/t coal	Xi et al., 2013
_	1–3.04 t CO_2/t coal	Wang et al., 2013
94,686	25.8 10-6 kg C/kJ	Cui and Liu, 2008

Note:
[a] The original emission factor is 94.6 Mt/TJ of which the unit is not practical.
[b] The original emission factor is 2.7 t CO_2/kg ce of which the unit is not practical.

cement industry.

5. Conclusions

The 21st century has seen a huge growth in Chinese cement production. In parallel with the peaking output, the CO_2 emissions produced by this industry have been brought to the forefront of public and academic attention. A domestic and comprehensive CO_2 emission factor database of the cement industry is a fundamental tool for relocating reduction task and achieving reduction target, e.g., emission cap and trade permits. The motivation of this work is to compile a more reliable and comprehensive CO2 emission factor database, which is promoted by Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories (Penman et al., 2000). Moreover, this work has provided uncertainties of emission factors taking advantage of bootstrap simulation and Monte Carlo simulation.

On the basis of a database consisted of 197 cement production samples, the simulation results show that the previous authoritative estimates are likely to deviate from the actual situation of Chinese situation. For process emission factor, it is rather accessible to obtain the data of clinker output other than that of carbonate decomposed in China. The original input method for process emission factor should be converted into a clinker-based method compatible with the output method. As well, the original output method is revised to eliminate the CO_2 emission overestimated from the no-carbonate CaO and MgO content. Several critical corrections for the emission factors are also taken into account. The data required for these corrections have been a routine of quality control in Chinese cement plants without claiming for any extra data.

The estimation for energy-related CO_2 emissions of Chinese cement industry is diverse, even somewhat arbitrary, in previous

studies. Chinese coal quality, in fact, differs from the benchmarking in IPCC. This work has provided a domestic emission factor of coals actually burned in the cement production.

Although new CO_2 emission factor database for Chinese cement in this work is based on a comprehensive and factory-level samples. To develop a finer resolution emission factor database, the spatial sampling and interpolation should be applied to cement industry. Knowing the spatial variation of cement CO_2 emission factor could deduce the distribution for population of the cement CO_2 emission factor with more robust estimation.

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Appendix

Probability density function:

(1) Normal distribution:

$$f(x) = \frac{1}{\sqrt{2\pi} \times B} \times e^{-\frac{(x-A)^2}{2\times B^2}}$$

where A and B refer to mean and standard deviation, respectively;

(2) Uniform distribution:

$$f(x) = \frac{1}{B-A}, \quad A < x < B$$

where A and characteristic B refer to lower and upper boundary, respectively;

(3) Lognormal distribution:

$$f(x) = \frac{1}{x \times \sqrt{2\pi} \times B} \times e^{-\frac{(ln(x)-A)^2}{2\times B^2}}$$

where A and B refer to mean and standard deviation, respectively;

(4) Weibull distribution:

$$f(x;A,B) = \frac{B}{A} \times \left(\frac{x}{A}\right)^{B-1} \times e^{-\left(\frac{x}{A}\right)^B}, x \geq 0$$

where A and B refer to scale and shape, respectively;

(5) Gamma distribution:

$$f(x;A,B) = \frac{A^B \times x^{B-1} \times e^{-x\times A}}{\Gamma(B)}, \quad x \geq 0$$

$$\Gamma(B) = \int_0^\infty t^{B-1} \times e^{-t} dt$$

where A and B refer to rate and shape, respectively.

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