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Analysis on differences of carbon dioxide emission from cement production and their major determinants

Tianming Gao [a,b], Lei Shen [a], Ming Shen [a,b,*], Fengnan Chen [a,b], Litao Liu [a], Li Gao [c,d]

[a] Institute of Geographic Sciences and Nature Resources Research (IGSNRR), CAS, 11A Datun Road, Chaoyang District, Beijing 100101, China
[b] University of Chinese Academy of Sciences, Beijing 100049, China
[c] School of Economics and Management, China University of Geosciences, Wuhan 430074, China
[d] Institute of Policy and Management, Chinese Academy of Sciences, Beijing 100190, China

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ABSTRACT

Based on 15 production lines we surveyed in China, the widely accepted input and output methods were applied to compare the process emissions with CSI (Cement Sustainability Initiative), and IPCC (Intergovernmental Panel on Climate Change) default values. We found that the output method would magnify CO_2 emissions from carbonate breakdown during clinker production. A reasonable method is to calculate carbonate content in raw meal using the CaO and MgO content in carbonate-containing material and their material ratio. Another finding is that the raw meals consumption recommended by CSI and CMBA (China Building Materials Academy) would enlarge and underestimate the calcining emissions, respectively. We applied the TC (total carbon) and LHV (lower heating value) methods for fuel emissions calculation and found that all of the samples' fuel emissions by the LHV method were higher than those by the TC method. Indirect emissions from different cement producing stages were also estimated by using regional electricity emission factor. In raw meal preparation and cement grinding stage, there were no differences in main production technologies, but in clinker production stage a remarkable difference appears. Replacing carbonate-containing materials with non-carbonate materials and changing clinker ratio are the main ways to reduce CO_2 content in raw meal and process emissions. Lowering fossil fuel intensity, using clean energy and alternative fuel were strongly recommended for reducing cement energy emissions.

1. Introduction

The cement industry is one of the most significant sources of greenhouse gases (GHG) in particular with carbon dioxide (CO_2) emissions. This sector accounts for about 1.8 Gt of CO_2 emissions in 2006 (Barker et al., 2009), approximately 7% of the total anthropogenic CO_2 emissions worldwide (Deja et al., 2010). This percentage is rapidly increasing since cement production is expected to increase faster. The cement industry will still play a more significant role in the future development though it is a major source of pollution. Recently, one of key goals of the global environmental agenda is to reduce emissions so as to protect world climate pattern. Ammenberg et al. (2014) discussed the extent of industrial symbiosis options can lead to reduced CO_2 emissions. After then Feiz et al. (2014) presented a systematic approach for assessing measures to reduce CO_2 emissions in cement industry. Control of thermo-chemical process emissions is significantly important to meet CO_2 emissions limitations (Mikulčić et al., 2014). Using demolished inorganic building materials and waste concrete powder as cement substitute materials is a recycling method for recycled cement (Oh et al., 2014). There are various approaches to improve energy efficiency. However, reduce limestone in raw meal and change its chemistry are becoming increasingly important to lower process emissions.

In general, carbon emissions from cement industry depend on cement production and emission factors. There are two widely accepted methods for estimating CO_2 emissions from cement production process. One is raw materials based (input) method; the other is defined as clinker based (output) method. The input method calculates calcinations CO_2 emissions based on volume and

* Corresponding author. Institute of Geographic Sciences and Nature Resources Research (IGSNRR), CAS, 11A Datun Road, Chaoyang District, Beijing 100101, China. Tel.: +86 10 64888073 13; fax: +86 10 64889005.
E-mail address: gaoming0920@aliyun.com (M. Shen).

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carbonate content of raw materials consumed in cement production (Cement Sustainability Initiative-CSI, 2005; CSI, 2011), while the output method is based on volume and composition of clinker produced (CSI, 2005, 2011). In theory, the raw material and clinker based methods are equivalent. In practice, however, the input method requires more extensive data than the clinker based method, thus not widely applied and only used in few countries such as the United States and Japan (CSI, 2005). Some rough estimation methods are also used in the absence of relevant data or for convenience. According to the clinker-based methodology, the CSI (2011) estimated the default emission factor as 547 kg CO_2 for per ton clinker production. However, in the absence of specific data, a default emission factor of 520 kg CO_2/t clinker is recommended by Intergovernmental Panel on Climate Change (IPCC, 1996, 2006).

China is the biggest cement producer and CO_2 emitter in the industry. The cement industry, accounting for 15% of total GHG emissions in China in 2009, is one of the main sectors to implement low carbon development (Wang et al., 2013). A life cycle inventory study on China cement industry is conducted by Li et al. (2014) to evaluate the environmental damages, including GHG, primary pollution and the hazardous air pollutants. Hu et al. (2014) selected two most common production processes in China to investigate material and energy use as well as pollutant emissions. Chen et al.'s (2014) study addressed pollutants generated by the cement industry in China, the impacts of these pollutants, and the potential for environmental improvement. Different estimations on cement production emissions of China vary significantly owing to different default emission factors. The Carbon Dioxide Information Analysis Center-CDICA (2013), for example, recommended 499 kg CO_2/t as emission factor to estimate China's cement production emissions. Wang (2008) adopted an emission factor of 425 kg CO_2/t to roughly estimate the process emissions from China's cement production in 2005 and 2007. Shen et al. (2014), using onsite surveys and sampling, developed a factory-level measurement for different types of clinker and cement production and classified the overall emission factors of cement production into three types (process emissions, combustion emissions and electricity emissions factors). Other scholar's estimations are listed in Table 1. Many scholars (Ke et al., 2013, 2012; Lei et al., 2011; Wang et al., 2013) estimated China's cement emissions with the default value. These studies' major advantage is increasing the energy efficiency in China's cement industry, yet they also have some weaknesses, which is they did not consider the historical improvement of clinker quality and the use of alternative resources in raw meal preparation. Nowadays most cement plants in China widely adopted new methods and technologies including non-carbonate sources, New Suspension preheater (NSP) kiln, waste heat recovery (WHR) power generation technologies and low clinker-to-cement ratio. As a result it is highly believed that the Chinese cement emission factors should be reviewed and corrected.

Above studies made significant contribution to CO_2 emissions inventory for China's cement industry, yet we strongly argue some existing limitations. First, the emissions factor value is not a fixed value which depends on the technology, equipments, fuels used and other factors. Due to the widely used steel slag and fly ash in raw meal preparation, the component of CaO and MgO from non-carbonate should be subtracted in clinker based method. Second, fuel emission factors, including electricity emission factors used in their studies, were not suitable for China's case. Third, previous studies just considered CO_2 emissions accounting, but did not identify their key driving forces. Considering these shortcomings mentioned above, we aim to provide some Chinese empirical studies and analysis. In this study, 11 NSP kilns from 8 cement plants and 4 shaft kilns from 3 cement plants were surveyed in Guangdong province of China. According to these 15 samples, we compared the estimation results for cement production process using different CO_2 calculating methods by IPCC, CSI, and other organizations. This study aims to evaluate the different estimation methodologies, understand their diversity in various estimation methodologies, identify the driving factors of CO_2 emissions from cement production, and provide policy implications for decision makers for appropriate mitigation policies towards China's "2020 strategic reduction target[1]".

The next sections will examine how CO_2 emissions are originated from cement industry and its amounts through various calculation methods, justify with onsite surveyed samples in Guangdong province which is a developed coastal area in China. Some main factors affecting cement emissions are also presented. The last section is our conclusions.

Table 1
China's cement CO_2 emission factor estimated by scholars (kg CO_2/t).

	Energy consumption	Calcining process	Total
Zhu, 2000	367–393	365	732–758
Worrell et al., 2001	467	415	883
WBCSD, 2002	–	–	900–950
NDRC, 2004	–	374	–
Cui and Liu, 2008	168(259)	395	563(654)
Boden et al., 2009	–	496–507	–
Lei et al., 2011	248–336	395	643–731
Wang, 2009	226	425	651

Note: the numbers in parentheses are the clinker emission factors.

2. CO_2 emissions from cement industry and its calculating methods

During the cement producing process, CO_2 is emitted from three different sources. Combustion of fossil fuel and calcination of calcium carbonate in processing stage, are defined as direct emissions. An indirect amount of CO_2 comes from electricity for raw materials transportation and electricity generating consumed by electrical motors and facilities. During the cement manufacturing process, almost 90% of CO_2 is direct emissions, and 10% is from the convey of raw material and some other production processes (Mikulčić et al., 2013).

2.1. Direct emissions

2.1.1. Process emissions

Calcium oxide (CaO) and magnesium oxide (MgO) are the typical clinker composition, which account for 64–68% of the clinker weight. Limestone is a major raw material used in the production of cement. The typical limestone used in cement production has 75–90% $CaCO_3$ in raw meal. Most CO_2 is produced in converting calcium carbonate ($CaCO_3$) and magnesium carbonate ($MgCO_3$) into CaO and MgO. When temperature is above 900 °C, the calcinations take place:

$$CaCO_3 \rightarrow CaO + CO_2\uparrow$$
$$MgCO_3 \rightarrow MgO + CO_2\uparrow$$

At this stage, the CO_2 leaves the materials, the raw meal losing over one third of its original weight. When temperature reaches 1300–1450 °C, the reaction of clinkerisation takes place, parts of the material become liquid, forming nodules known as clinker.

[1] China promised to reduce carbon emission intensity per unit of gross domestic product (GDP) by 40–45%, using 2005 as the benchmark year, and to increase the percentage of non-fossil fuels in the primary energy consumption to approximately 15% by 2020.

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2.1.2. Process emission calculating method

Cement production is a complex crafts due to various materials and energy flows. So the CO_2 emissions inventory for cement industry is also complicated. During the last two decades, many organizations and scholars (China Building Materials Academy-CMBA, 2011; CSI, 2005, 2011; IPCC, 1996, 2006; Ke et al., 2013; Netherlands Environmental Assessment Agency-PBL, 2008; Wang et al., 2013) have made many efforts on calculating CO_2 emissions inventory in cement industry. These methods differ only in the range of operating boundaries, specific calculation methods for each emission process, and the specific CO_2 emission coefficient. These are valuable sources of calculating cement production CO_2 emissions. However, to determine whether these methods are scientific and applicable in China, we need to conduct empirical research and comparative studies first.

2.1.2.1. IPCC method. The IPCC provides three basic methodologies for estimating emissions from cement production, which focus on process emissions from carbonate minerals during the process of clinker production. And it is widely accepted and cited default emission factor is 510 kg CO_2/t clinker, without including MgO emissions.

In tier 1 method, emissions are calculated by estimating clinker production inferred from cement production data, correcting for imports and exports of clinker. The clinker based output method has been adopted by IPCC as tier 2 method for national GHG inventory calculations. It requires collection of clinker production data and information of the CaO content of clinker and fraction of CaO from carbonate. If a plant is deriving a significant fraction of CaO from a non-carbonate source (such as steel slag or fly ash), this component of CaO should be subtracted first in emissions estimation (IPCC, 2006). Tier 3 emission calculation is based on actual CO_2 contents of carbonates, which requires full accounting of carbonates (species and sources). This method need ensure that all carbonate inputs (i.e., types, amounts, all sources) to the kiln are fully investigated, and repeat full investigation whenever there is any significant change in materials or processes. However, a large number of raw material inputs and the need of continuously chemical composition monitor make this approach impractical in many cement plants (CSI, 2011).

2.1.2.2. CSI clinker based method. CSI calculation method was based on the IPCC guidelines and has been applied by over 100 countries in their major cement plants. For the process emissions, CSI method presents two calculation methods: a carbonate mineral based method and a composition of clinker based method. The CSI clinker based method is basically the same with IPCC method, but considered the influence of MgO in clinker, bypass dust and cement kiln dust (CKD) in clinker production, and the influence of no-fuel carbon from raw material.

A small amount of MgO (1–2%) in clinker is desirable, because it acts as a flux (Van Oss and Padovani, 2002). Part of MgO may come from carbonate source. In IPCC tires 1 and 2 default emission factors might be underestimate as they do not include the CO_2 emissions from the calcinations of $MgCO_3$ (Ke et al., 2013). CSI (2005) suggested that calculating CO_2 emissions from calcinations process is based on the CaO and MgO content in the clinker. Corresponding to IPCC default value, a default of 525 kg CO_2/t clinker should be used (CSI, 2005).

Emissions from fuel are emitted from two different sources: kiln fuels and non-kiln fuels. These fuels defined by CSI as conventional fuels and alternative fuels, mixed fuels and biomass fuels (CSI, 2011).CSI (2005) gave the emission factor of alternative fuels. CKD, TOC and indirect emissions from electricity consumption were also calculated and reported by CSI.

2.1.2.3. CBMA method. CBMA method was jointly developed by China Building Materials Academy (CBMA), China Business Council for Sustainable Development (CBCSD), Institute of Geographic Sciences and Natural Resources Research, CAS (IGSNRR,CAS), and China Clean Development Mechanism Fund (CCDMF), and approved by the China National Standardization Committee in 2013. This method took fully considerations of the emissions influence factors mentioned above into its calculation. It provides a basic CO_2 calculation method for enterprises, especially the producers with alternative raw materials and alternative fuel, and a conversion factor (1.04) for coal ash added in clinker. However, this approach is based on the composition of CaO and MgO in clinker, instead of the carbonate content in raw material. This will expand emission factor, especially when there are many producers using steel slag and fly ash as raw material, in which CaO and MgO exist in the form of non-carbonate.

2.1.3. Fuel emissions and calculating method

It is widely recognized that there is an additional source of CO_2 emissions from fuel burning during calcining process. Fuel is combusted to produce heat for the process of clinker production. The CO_2 emissions form fuel combustion during the cement production process is influenced by the type of producing process, operating way, fuel used and the ratio of carbon content.

The approach is to calculate CO_2 from conventional fuels based on fuel consumption, lower heating values (LHV) and the correspondent CO_2 emission factors. Coal is the traditional fuel used in China's cement industry. Other types of fuels used include diesel, coke, coal gangue, fuel oil, as well as municipal wastes, while they shared only about 2.4% of the final energy consumption in 2009 (Ke et al., 2012). Missions from biomass alternative fuels are considered carbon-neutral. As a result, alternative fuel (AF) could reduce considerably fuel-related CO_2 emissions in cement industry.

2.2. Indirect emissions and calculating method

Date of indirect emissions is useful to assess overall carbon footprint of an industry. Four categories of indirect emission were listed by CSI (2005). But, in our surveyed samples, external production of electricity consumed by cement producer is the only sources. The CO_2 emissions from electricity consumed by cement production are estimated by using the annual national or regional average emission factors and power bought from external grid. Due to different technology and energy mix in different years and regions, emission factors of regional power grids may vary significantly (Lindner et al., 2013). In China, there are 7 power grids with different emission factors.

3. Case studies: comparing cement emissions based on onsite samples in Guangdong Province of China

Guangdong Province is the fourth largest cement producer in China. In 2010, 102 Mt of cement were produced, which accounted for 5.43% of China's cement production. There are 230 cement plants producing cement in complete manufacturing cycle, 60 cement grinding plants, of which include 51 NSP kilns with a clinker production capacity of 58.4 Mt, accounting for 61.5% of the total production capacity, and 430 other process lines with the production capacity of 37.1 Mt. In this study, 4 shaft kilns (abbreviated as S) and 11 NSP kilns (abbreviated as N) production lines were surveyed. These 8 NSP kiln plants are Taini yingde (N-1), Yingde hailuo (N-2), Guanying (N-3), Zhongcai hengda (N-4), Tapai longmen (N-5), Guanzhou yuebao (N-6), Guanzhou zhujiang (N-7), Meizhou longteng (N-8), and the other 3 shaft kiln plants are

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Heyuan hexing (S-1), Meizhou ganyuan (S-2), and Meixian hanjiang (S-3).

Generally, a cement production process can be divided into three main stages after mining and quarrying. This study involves crushing and grinding of raw materials, materials calcining in a rotary kiln, clinker cooling, mixing of the clinker with gypsum; and milling, storing and bagging of the finished cement (Fig. 1).

Detailed data collection forms were developed and used to collect information of onsite cement and clinker production, raw material and electricity consumption and energy use from the surveyed cement plants. The number of production lines at these plants, their clinker and cement production capacity and actual clinker and cement production quantity in 2012, energy used for per unit clinker and cement production, the alternatives of raw material and their composition were also collected. The systems datum are documented accordingly of continuous 12 months in 2012. Readings are taken repeatedly to minimize errors; and averaged values are employed in this paper. The composition of the raw materials, coal, and produced clinkers are similar to those of home and abroad Portland cement plants. The fuel intensity represents the energy efficiency and the level of China's cement industry. In addition, limestone, clay, shale, fly ash, coal, raw meal, clinker and cement outputs were also collected by a stainless steel spatula. These dry samples were collected from various points within the stockpiles and stored in zip locked plastic bags for analysis. All the collections and measurements are carried out during normal plant operations.

3.1. Process emissions based on input and output methods

At the calcining process, there are three sources of process CO_2 emissions: CO_2 from calcium carbonate breakdown, CO_2 from non-recycled CKD leaving the kiln system, and CO_2 from organic carbon during pyro-processing of the raw meal (Wang et al., 2013). CO_2 from carbonates can basically be calculated by raw material way issued by IPCC tier 3 which based on the volume and carbonate content of the raw meal consumed. But IPCC tier 3 is impractical in many cement plants. In general, the test carbonate content of raw material is a complex process. However, in many cement plants the homogenized mass flow of raw meal is routinely monitored including its chemical analysis for the purpose of process and product quality control. The testing of CaO and MgO content of raw material is a regular process. In typical cement plant, the carbonate rocks, such as limestone and dolomite, are the dominantly used raw materials. From the analysis of Ma et al. (2005), Zuo (1981), and Fan et al. (2001), we know that there could be a carbonate component in these raw materials: clay, shale, sandstone, and other supplementary materials. Steel slag, magnesium slag and fly ash were widely used as raw materials in many Chinese cement plants. In these materials, the CaO and MgO exist not in the form of carbonate but non-carbonate. Therefore, we assume that CaO and MgO are all from carbonate minerals in carbonate-containing material.

Based on this assumption and raw material ratio, we could use the CaO and MgO content in carbonate-containing material and their material ratio to calculate CO_2 content in raw meal (Eq. (1)).

$$Ra_{CO_2} = \sum\left(Rca_i \times r_i \times \frac{44}{56} + Rmg_i \times r_i \times \frac{44}{40}\right) \quad \left(i = 1,2,3\ldots\right) \quad (1)$$

where Ra_{CO_2} represents CO_2 content in raw meal, %; Rca_i, Rmg_i represents calcium carbonate and magnesium carbonate content in raw material i, %; r_i represents the proportions of raw material i in raw meal, %; 44/56, 44/40 represent CO_2 content in calcium carbonate and magnesium carbonate; $i = 1, 2, 3\ldots$ represents carbonate material species. Then we analyzed the collected raw meal samples from these production lines, and their CO_2 content are showed in Fig. 2. Comparing the content value of calculated results to the testing samples (Fig. 2), we found that the calculated CO_2 content is very close to the actual value of raw meals, the calculated and testing average values are 34.78% and 34.80% for NSP kilns and 31.33%, and 31.55% for shaft kilns. As a result, we could use CaO and MgO content in carbonate-containing material and their material ratio to calculate the carbonate content in raw meal. The CO_2 content from the raw meal method is lower than the clinker based method, and can better reflect the actual carbon dioxide emissions in clinker/cement production.

The calcinations CO_2 emission factor is based on determining the amount of raw material consumed for per ton clinker production and the carbonate source content in raw meal. Raw meal clinker mass ratio is determined by the types of raw materials and composition, fuels ash and the quality of clinker. Since the temperature in clinker kiln is about 1300–1450 °C, in theory carbonate decomposition reaction is complete. But actually, the carbonate material is incompletely calcined in the outdate kilns, due to the uneven heating, insufficient amount of air into the kiln and uneven air distribution, etc. In dry process, the proportion of CO in exhaust gas emitted by the kiln was negligible (Qiu et al., 2012). This means that the oxidation reaction was complete in this craft. But in other process, especially the shaft kiln, the CO proportion varies at 1.3%–1.6% according to the different calcination technologies (National Building Materials Test Center-NMBTC and NMBTC CMBA, 2007). In

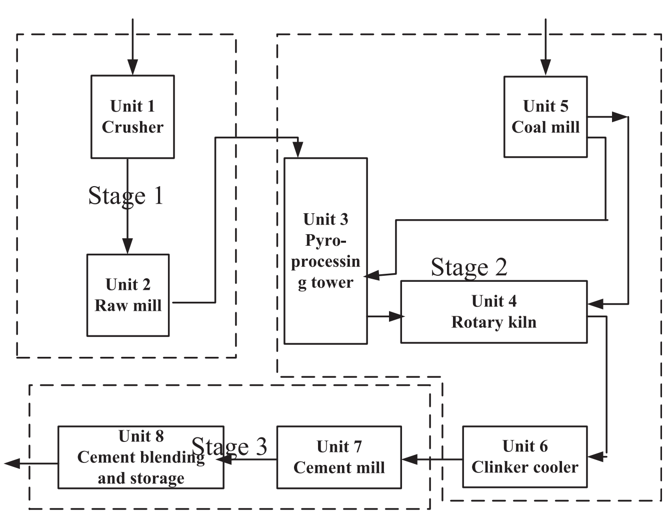

Fig. 1. The scope and system boundary of cement production.

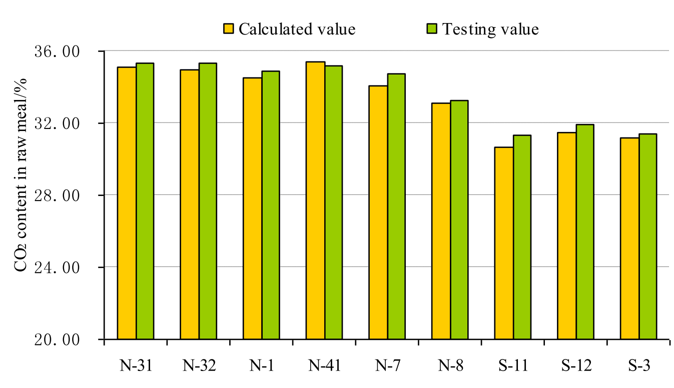

Fig. 2. Compare the calculated and testing CO_2 content in collected real meals.

the absence of data, a default value of 1.4% shall be used for incomplete combustion in outdate kilns.

Based on the CO_2 content in the raw meals from the 15 onsite cement plants, and their raw meal consumed in each clinker production lines, we calculated their process CO_2 emissions from clinker production. As Fig. 3 shows, the production line S-11 gives the lowest estimate for process CO_2 emissions (498.17 kg/t clinker) from clinker production, while the clinker production line N-31 gives the highest estimate (537.31 kg/t clinker). Given the large discrepancies between the two production lines, the proportion of limestone in raw meal is very different. We note that based on the raw materials method the average process CO_2 emissions (522.66 kg/t clinker) from all NSP kilns is very close to CSI default value (525 kg/t clinker). The relative uncertainty of the estimation of the process CO_2 emission from the surveyed NSP cement plant is 4.31–6.46%. But for the shaft kiln, their process emissions (507.46 kg/t clinker) are lower than the default values. And the relative uncertainty of the estimation for shaft cement plant is 2.76–4.14%. To compare, the calcination emissions estimation based on clinker method are also listed in Fig. 3. It clearly presents that almost all of the process emissions from production lines by raw materials are lower than those by the clinker method. Their difference is that the CaO and MgO from clinker is not all from carbonate materials. Therefore, we believe that the clinker method will exaggerate CO_2 emissions from $CaCO_3$ and $MgCO_3$ breakdown during clinker production. According to the 15 sample production lines, the process emissions based on raw material is about 13 kg and 11 kg lower than clinker method for NSP and shaft kilns, respectively (Fig. 3).

To produce certain quality clinker, the materials putting into the raw meals are defined within a certain extent. The four variables, limestone, clay, shale and iron ore are interacted in raw meal production. These four parameters are assumed as independent with each other for the local sensitivity analysis. Limestone content in raw meal representing the variable was taken into account during the sensitivity calculations. Because we have no more data to describe the distribution of the process emissions, computing sensitivities is based on a design of experiment, and we assume a 2% variation based on the basic data of 83.90% for the parameter limestone content in raw meal. Table 2 shows that if the limestone content in raw meal decreased from 83.91% to 82.22%, the clinker quality will present a decline (K was decreased from 0.89 to 0.85), and the process emission will be reduced by 12.36 kg for per ton clinker production. While, if the limestone content in raw meal increased by 2% (up to 85.17%), the process emission factor will lift up to 531.9 kg. The data show linear regression plots created between limestone content in raw meal and process emission factors

Table 2
Sensitivity of limestone content in raw meals responses to the process emissions.

Limestone content (%)	Chang rate (%)	Limestone saturation coefficient (K)	Raw meal intensity (kg/t-clinker)	CO_2 content in raw meal (%)	Process emissions (kg CO_2/t-clinker)
85.585	2	0.919	1500	0.355	531.9
85.167	1.50	0.914	1501	0.354	530.7
84.752	1	0.909	1499	0.353	528.6
84.324	0.50	0.899	1498	0.351	526.3
83.905	0	0.889	1494	0.35	522.5
83.484	−0.50	0.869	1488	0.348	518.2
83.061	−1	0.864	1486	0.347	515.7
82.639	−1.50	0.859	1484	0.346	513.1
82.216	−2	0.849	1481	0.345	510.2

($y = 6.380x - 51.20$). The R_2 values for this linear regression line shows that the limestone content in raw meal account for approximately 98.1% of the variance in process emissions. This suggests that the process emissions have the highest impact on the limestone content in raw meal if all the parameters are varied within a certain data range.

3.2. Fuel emissions based on LHV and TC method

The LHV and default emission factors for coal, fuel oil and natural gas were first from IPCC (1996) and corrected by the 2006 IPCC guidelines. CSI (2011) recommended companies to use plant- or country-specific emission factors if reliable data are available. This emission factor of fuels shall be based on the total carbon (TC) content. Direct calculation of emissions based on fuel consumption and fuel carbon content is acceptable given that material variations in the composition of the fuel, and especially its water content, are adequately accounted for (CSI, 2011).

Coal is the traditional fuel used in China's cement industry. And the share of alternative fuel (AF) is negligible. Just as showed in the collected cement plants, all coals of cement plants are used as clinker production. We compared the fuel emissions in clinker production based on LHV and TC methods. As shown in Fig. 4, all of the fuel emissions based on LHV method are higher than those on TC method. Their gaps fall into the ranges of 0.59–74.81 kg for per ton clinker production. In production line N-51, the emissions from both methods are much approximate, while for production lines N-41 there is a difference about 74.81 kg in them. TC content of coal is an important source of heat. LHV of coal is determined by ash, volatile and moisture in dry basis. Therefore, LHV value is closely related to TC content and depends on other factors. In clinker

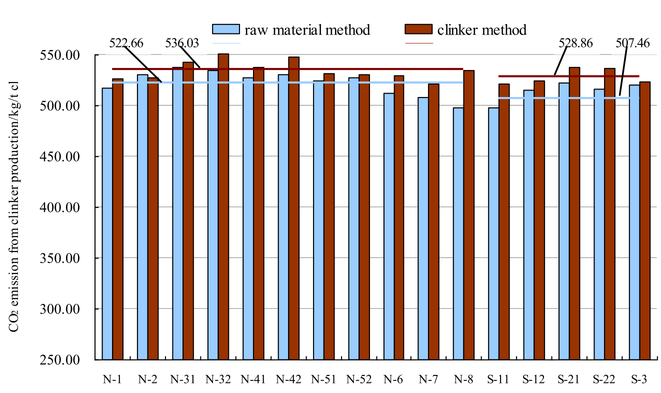

Fig. 3. Process emissions form clinker production based on raw materials and clinker method.

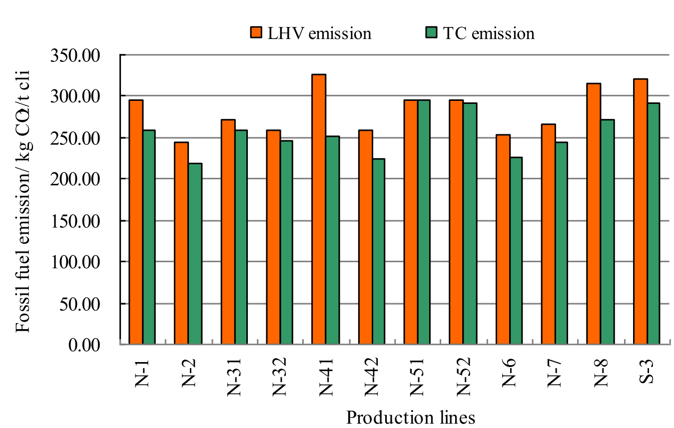

Fig. 4. Fossil fuel emissions in clinker production based on LHV and TC method.

production, the CO_2 emission factors of fuels should always be determined by the TC content. Thus, keeping the fuel consumption constant, and increasing the TC content in fuel, the fuel emission from the clinker production will increased at the same rate, and vice versa. According to TC method, the production line N-2 gives the lowest estimate (218.29 kg/t clinker) for fuel CO_2 emissions, while N-51 gives the highest estimate (295.36 kg/t clinker). The relative uncertain range of the estimation for the fuel CO_2 emission is larger (17.56%–26.35%) than the process emissions (4.31%–6.46%), due to the difference of production technologies and operated way.

3.3. Indirect emissions based on Guangdong electricity grid emission factors

Different countries usually adopt the same method in the calculation of CO_2 emissions from purchased electricity, which is based on power consumption and electricity carbon emission factors excluding electricity lost in transport and distribution (United States Environmental Protection Agency-EPA, 2008). Fossil fuel based electricity production directly emits a large amount of CO_2. In China, fossil fuel fired thermal power contributes more than 81% of the total electricity production and coal is the main fossil fuel for thermal power (National Bureau of Statistics-NBS, 2012a). Fired thermal power also dominates the electricity production in Guangdong province of China, but its proportion (79.35%) is lower than national average (NBS, 2012b). Therefore, the electricity emission factor (0.9344) (National Development and Reform Commission-NDRC, 2012) is lower than national emission factor. And the indirect emissions show an exact linear relationship with electricity consumption.

The electricity used for cement production is mainly purchased from the regional grid, but some Chinese cement plants use waste heat recovery (WHR) power generation technologies to self-generate electricity. This technology is widely used for the NSP kiln in China. In the onsite 11 NSP kilns lines, 9 of them are the WHR power generation system which recovers the energy in waste heat and does not consume additional fossil fuels thus reduces the total energy consumption and CO_2 emissions. Therefore the electricity consumed from the WHR power generation should be excluded while evaluating indirect emissions.

According to the electricity emission factor in Guangdong, we estimated the electricity emissions of the collected samples at different cement production stages. According to Fig. 5, we found that the electricity consumption range at raw meal preparation stage is from 23.14 to 38.39 KWh per ton clinker for both of the main production technology. Their average indirect emission factor at this stage is about 26.33 kg CO_2/t clinker. At clinker calcining stage, however, the amount of electricity consumption has significant discrepancies. For NSP kilns the average electricity consumption is about 34.51 KWh, and its indirect emission factor is 32.25 kg. For shaft kilns, the electricity consumption is 17.84 KWh and indirect emissions factor is 16.66 kg, which are almost half of NSP kilns' emissions. This discrepancy is resulted from the clinker movement in NSP kilns by power engine while shaft kilns depend on gravity. Cement grinding is the most electricity intensity stage, which used up 36.17 KWh, about 33.79 kg CO_2 emissions for grinding per ton cement. Other emissions from electricity are about 2.9 kg CO_2/t clinker in the surveyed samples. For the WHR generation system in NSP kilns, there were about 28.45 KWh electricity produced for per ton clinker production, meaning that 26.58 kg CO_2 was saved. The electricity emissions range from the shaft production lines is from 58.17 kg/t cement to 63.76 kg/t cement. The relative uncertainty of the estimation of the indirect CO_2 emission by the onsite shaft cement plant is 5.98–8.98%. Whatever the application of the WHR power generation technologies in the NSP cement plant, the uncertainty of indirect emissions falls into the range of 18.62%–27.93%.

4. Main factors affecting cement emissions

4.1. Factors affecting the process emissions

4.1.1. Raw meal consumed

The process emissions with input method are determined by raw meal consumed and its carbonate content. Due to the absence of better data, a default of 1.55 t raw meal/t clinker should be used (CSI, 2011). And in the CBMA method, a default value 1.04 was used as conversion factors of coal ash added in clinker. Plant-specific raw meal to clinker ratios should exclude the ash content of the fuels used, to avoid double counting (CSI, 2011). According to the principle of mass balance, we get the calculation method for raw meal clinker mass ratio. It is expressed in Eq. (2):

$$r_a = \frac{1 - C_{cl} \times A_c}{1 - R_l} \quad (2)$$

Here, r_a represents raw meal clinker mass ratio; C_{cl} represents coal consumption (dry) for per ton clinker production, t; A_c represents ash content of coal, %; R_l represents ignition loss of raw meal, %. According to the equation and the composition of fuel and raw meal, we calculated the plant specific raw meal clinker mass ratio of the surveyed samples (Fig. 6). Fig. 6 provides summary information on raw meal consumed for production of pre ton clinker in the 15

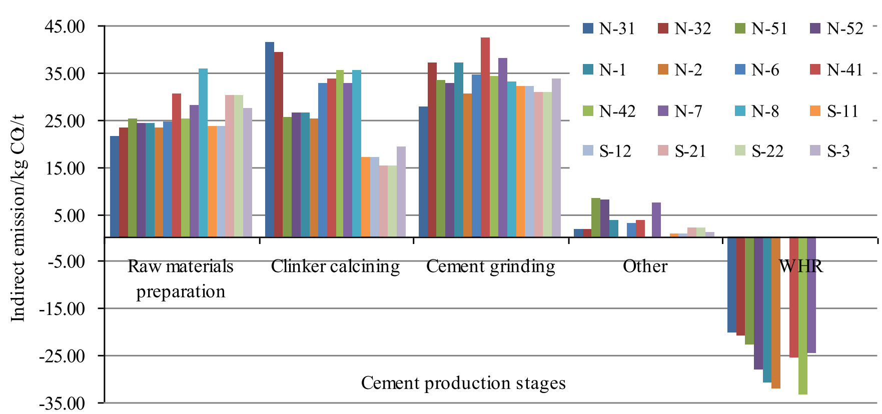

Fig. 5. Indirect emissions from cement production stages in the collected samples.

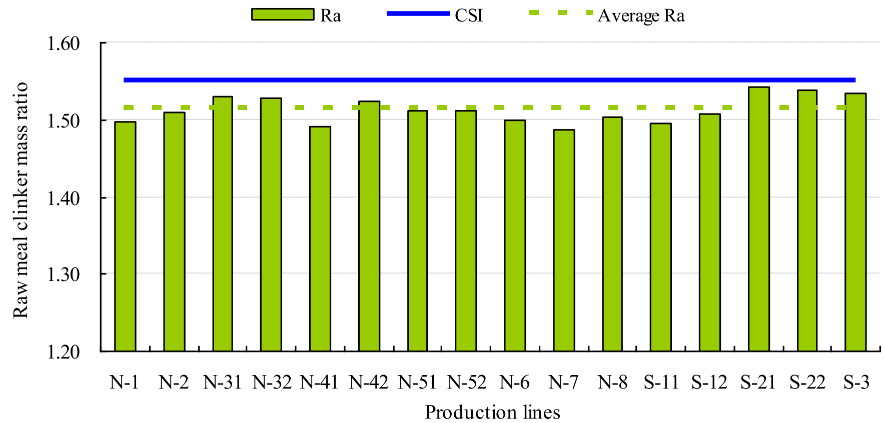

Fig. 6. Raw meal clinker mass ratio in surveyed production lines.

surveyed plants in Guangdong province. Production line S-21 consumed the highest amount of raw meal (1.542 t) in the production of per ton clinker, whereas production line N-7 had the lowest raw meal intensity (1.486 t). The all plant specific raw meal clinker mass ratio in surveyed production lines was lower than the CSI default value (Fig. 6). Their average raw meal clinker mass ratio is 1.5132 (Fig. 6).

According to the coal and raw meal consumed, we calculated the amount of coal ash mixed in the clinker in the surveyed NSP kilns samples. The proportion of coal ash added into clinker is listed in the Table 3. We note that coal ash added in clinker is determined by coal consumption and its ash content. Production line N-8 is the biggest one, which mixed about 40.75 kg coal ash into clinker, whereas only 22.20 kg coal ash was added in clinker in production line N-31. The average coal ash added in clinker in the collected NSP kilns is about 30.34 kg, accounting for 3.04% of per ton clinker. This proportion is less than the CMBA method's default value (1.04) 4%. This means that less raw meal was consumed for per ton clinker based on the CMBA method, and less process emissions were emitted.

4.1.2. Carbonate content in raw meal

Another important reason to affect calcining CO_2 emissions is the carbonate source content in raw meal, which depends on the carbonate content in the carbonate-containing materials and their share in raw meal. Siliceous and ferrous materials share 10–20% of raw meal, and carbonate content in these materials is negligible. Thus, replace siliceous and ferrous materials with coal ash, sulfuric acid residue, copper slag or lead slag to reduce process emissions is not significant. Their reduction potential is from 0.49 kg to 6.92 kg CO_2/t clinker, accounted for 0.01%–1.27% of the process emissions because of the different proportion of siliceous and ferrous materials in raw meal in the surveyed plants. So, using alternative materials of limestone is the main way to decrease CO_2 emissions from raw materials decomposition. Bauxite, calcium carbide residue (CCR), steel slag, magnesium slag are the main non-carbonate materials to replace limestone. A future challenge of the cement industry is to use more alternative raw materials originating as byproducts from other industries or directly from other waste streams.

Table 3
Quantity and proportion of coal ash added in to pre ton clinker.

Production lines	Coal consumption/kg	Ash content/%	Coal ash added/kg	Share of clinker/%
B-1	153.71	24.24	37.27	3.73
B-2	123.02	21.50	26.45	2.64
B-31	135.70	16.36	22.20	2.22
B-32	132.45	17.73	23.48	2.35
B-41	151.24	24.17	36.56	3.66
B-42	134.40	24.17	32.48	3.25
B-51	138.98	22.60	31.41	3.14
B-52	137.53	22.78	31.33	3.13
B-6	124.86	21.75	27.16	2.72
B-7	130.31	19.73	25.71	2.57
B-8	137.43	29.65	40.75	4.07
Average	–	–	30.44	3.04

4.1.2.1. The reduction potential of replacing limestone by CCR. The properties of CCR cements were approved when the raw materials replaced by CCR (Krammart and Tangtermsirikul, 2004; Rattanashotinunt et al., 2013). In China there are more than 20 of the carbide slag cement production lines in production or under construction, the annual clinker production capacity is about 11.5 Mt. Based on a typical carbide slag producers, we design 5 ingredients programs to evaluate their process emissions reduction capability (Table 4). From Table 4, we know that with the increased incorporation of CCR, the reduction capability of process emissions from 110.95 kg increase to 401.83 kg per ton clinker by the incorporation of CCR from 15% to 60%. This shows that CCR cement has a significant process emissions reduction capacity. Thus, we should fully utilize the 20 Mt CCR as a by-product of the acetylene gas production.

4.1.2.2. Reduction potential by sulphoaluminate cement production. The manufacture of Ordinary Portland Cement (OPC) produces large amounts of CO_2 mainly due to the high calcium carbonate content of raw meal. While calcium sulphoaluminate cement production demands less limestone in the raw feed, a lower burning zone temperature, and ease of cement grinding due to higher clinker porosity (Martín-Sedeño et al., 2010). So, sulphoaluminate cement represents a low CO_2 alternative to OPC, mainly because they generate lower CO_2 emissions in the clinkering process and grinding (Pelletier-Chaignat et al., 2012). Calcium sulphoaluminate cements have been used in China for about 40 years, but their development and use in other countries is not widespread currently.

Chemical composition of a typical sulphoaluminate cement producer is listed in Table 5, which shows that the ratio of limestone and bauxite is 48:52 in raw meal. The process emission in this cement plant is 271.52 kg for per ton clinker, which is 250 kg less than OPC. Sulphoaluminate cement manufacture in a modern cement plant can give CO_2 emissions reductions of up to 35% per mass of cement produced, relative to OPC (Martín-Sedeño et al., 2010).

Table 4
Different CCR cement ingredients programs and their reduction capability.

Program	CCR/%	Limestone/%	Sandstone/%	Clay/%	Iron ore/%	Process emissions/kg CO_2/t clinker	Reduction capability/%
A_1	0.00	86.21	3.9	8.79	1.1	527.89	0
A_2	15.00	70.41	3.86	9.42	1.31	416.94	21.02
A_3	30.00	54.82	6.29	7.19	1.7	314.24	40.47
A_4	45.00	39.30	8.69	4.91	2.1	218.37	58.63
A_5	60.00	23.40	8.70	5.55	2.35	126.06	76.12

4.1.3. *Adjusting clinker ratio value*

The quality of clinker is determined by limestone saturation coefficient (KN), silicic ratio (N) and aluminum oxide ratio (P), whose value is based on the content of SiO_2, Al_2O_3, CaO, and Fe_2O_3 in clinker. KN can vary within a certain range (0.85–0.95). Moderately lowering KN, less raw meal is consumed, less resources is demanded and less carbon emissions is emitted. If the KN decreased from 0.95 to 0.88, the content of CaO in clinker will decreased from 68% to 64%. This means that CO_2 emissions from per ton clinker produced will be reduced by about 30 kg (Liu and Li, 2010).

4.2. *Factors affecting the fuel emissions*

4.2.1. *TC content in fossil fuel and carbon oxidation factor*

TC content of fuel primary based on carbon content, but which also contain varying amounts of hydrogen, oxygen, nitrogen, sulfur and other elements. Tu et al. (2003) studied the correlation between fixed carbon and carbon content of anthracite coal, which value varied from 0.993 to 0.999. TC in all kiln fuels should be treated as fully oxidized, due to very high combustion temperatures and long residence time in kilns and no, or minimal, residual carbon found in clinker (CSI, 2011).

The default carbon content and carbon oxidation factor for fuels was estimated by IPCC (2006). Many fossil fuels, such as fuel oil, natural gases are lower carbon content and CO_2 emissions per MJ of energy than coal. Natural gas and fuel oil with around 38.5% and 17.2% less CO_2 emissions per GJ compared with the coal (Pardo et al., 2011). So, using other fossil fuel rather than coal is a way to reduce fuel CO_2 emissions in cement industry.

4.2.2. *Fuel intensity*

The main factors determining energy consumption in the cement industry are the structural shifts between kiln types, the kiln efficiency improvement, and the sophistication of kiln technologically. The fuel intensities of shaft kilns span a wide range, from 110 to more than 220 kgce/t clinker, depending on the levels of mechanization and operation skills.

NSP kilns differ from shaft kilns is because the existence of pre-heater or pre-calciner unit. Pre-heater towers consist of a series of vertical cyclone chambers which allow part of the heat of the exhausting gases of the kiln to be recovered. The energy consumption of kilns with suspension pre-heaters is much smaller than previous kilns. Pre-calciner kilns have an extra combustion chamber installed between the pre-heater and the kiln. This pre-calciner chamber consumes 60% of the fuel used in the kiln, and 80–90% of the calcination takes place there. This reduces energy consumption by 8–11% (Ali et al., 2011). Table 6 shows specific thermal energy consumption for different types of clinker manufacturing process. Energy management and process control system, kiln combustion system improvement, waste heat recovery were mainly used for improving kiln efficiency, and they saved 24.6% energy comparing 2009 to 1990 in China's cement industry (Xu et al., 2012).

Table 5
Chemical composition of a typical sulphoaluminate cement producer.

	Loss	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Ratio of raw material
Limestone	42.13	1.63	0.64	0.35	53.99	0.3	48
Bauxite	14	7.79	69.85	2.19	1.25	0.4	52
Raw meal	27.50	4.83	36.63	1.31	26.57	0.35	
Coal ash	–	34.22	22.63	9.36	19.48	2.06	
Clinker	–	7.35	51.07	2.17	35.37	0.72	

4.2.3. *AF used for reduction fuel emissions*

Due to the increased environmental awareness, cement plant operators are starting to use AF. The use of AF for cement clinker production is crucially important to the reduction of fuel emissions in cement manufacturing. AF has been used broadly. Mokrzycki and Uliasz-Bocheńczyk (2003) classified AF into three basic groups: gas (e.g., landfill gas, pyrolytic gas, and biogas), liquid (e.g., used oils and solvents), and solid (tires, wood waste, plastics, meat and bone animal meal (MBM), municipal solid waste (MSW), sewage sludge (SS) and textiles). MSW, MBM and SS are the most widely used AF in the EU cement industry (Aranda Usón et al., 2013). Using these AF as substitute fuel helps in saving conventional fuels and was accepted as renewable fuel energy sources. As biomass fuels, they help the cement plant reduce its CO_2 conventional fuel emissions.

4.2.3.1. Municipal solid waste (MSW). MSW has become a common AF in cement industry. There are notable different ash, chlorine, sulfur, and water contents among MSW according to different sources. Their different physical and chemical properties can cause difficulties in the kiln combustion process if MSW is unsorted. Rovira et al. (2010) performed a study to analyze the effects of increasing the substitution ratio of MSW in a conventional fuel. By investigating the effect of MSW combustion in cement calcining system, the performance of clinker and cement, and the environment impact, the suitable rate of heat replaced by MSW disposing in NSP kilns is up to 30% (Wang, 2006). It means that a reduction of 30% emissions from fossil fuels has been achieved.

4.2.3.2. MBM replace of coal. Gulyurtlu et al. (2005) performed several tests with different MBM/coal ratios to determine the influence of various MBM parameters on the process of co-combustion with coal. The reduction of CO_2 emissions was evident when the ratio of MBM increased. Gulyurtlu et al. (2005) illustrated that the CO_2 emissions from MBM was only about two-thirds from coal if the coal was fully replaced by MBM. Cascarosa et al. (2011) showed that increasing MBM content raised the production of H_2 but tended to decrease CO_2 and CH_4. Another important aspect we need consider is the NO_x emissions increased. The test of using leather as an alternative fuel was carried out in China. And the result showed that 1 ton leather can replace 0.77 ton coal (Zhang, 2008). While in the production practice, the 15 kg coal was replaced by 30 kg leather, and the emissions reduction from the mixed fuels was 39 kg than the parent coal, accounting for 13.73% of the fuel emissions (Zhou et al., 2009).

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Table 6
Specific thermal energy consumption of a kiln process.

Kiln process	Thermal energy consumption (Gj/t clinker)	Coal consumption (kgce/t clinker)[a]
Wet rotary kiln	5.86–6.28	199.73–214.04
Dry long rotary kiln	4.60	156.78
Dry long rotary kiln with 1-stage cyclone preheater	4.18	142.47
Dry long rotary kiln with 2-stage cyclone preheater	3.77	128.49
Dry long rotary kiln with 3-stage cyclone preheater	3.55	121.00
Dry long rotary kiln with 4-stage cyclone preheater	3.14	107.02
Dry long rotary kiln with 4-stage cyclone preheater, calciner and high efficiency cooler	3.01	102.59
Dry long rotary kiln with 4-stage cyclone preheater, calciner and high efficiency cooler	<2.93	99.86

[a] Author's calculation.
Source: Ali et al. (2011)

4.2.3.3. Sewage sludge (SS). Normally, SS is placed in the main furnace of the cement kiln and burned as a fuel. At the same time the residual non-combustible components of the sludge are used as raw materials in cement production. In addition to moisture content, but also other important parameters, the release and combustion of volatile compounds and the combustion of the high ash content, may potentially affect the overall combustion process of SS. To control other parameters, a maximum SS feed rate of 5% of the clinker production capacity was suggested by Aranda Usón et al. (2013).

4.3. Factor affecting indirect emissions

4.3.1. Electricity emission factor

Electrical production of China has long been based on coal-fired electricity generation. Other power sources, such as hydropower, nuclear power, and wind power, are relatively clean. Thus, China's electricity emission factor is based on power coal intensity. Yu et al. (2014) utilized life-cycle assessment to assess the effect of carbon emissions and calculated the coefficient of carbon emissions in coal-to-energy chains. In this study, 4 processes (coal mining, selecting and washing, transport and electricity generation) were calculated to estimate the GHG emissions. Results show that the carbon emissions coefficient of coal to energy chain in China is 875 g/kWh. So, lowering the percentage of coal-fired electricity, enhancing coal mining energy conservation, increasing the proportion of railway transportation in coal transportation and improving energy-power conversion efficiency are the main ways to decrease electricity emission factor.

4.3.2. Electricity consumption

Electricity is used in the cement production process for raw materials extraction, blending, grinding, homogenization, clinker production, cement grinding and convey, packing and loading. Gu et al. (2012) reported that average electrical energy consumption was about 110 kWh/t cement in China's cement industry. Grinding remains the biggest source of energy consumption in cement production. Approximately 60–70% of the total electrical energy is used for the grinding of raw materials, coal and clinker. So, the use of new grinding equipment is the main way to reduce power consumption in cement production, and an important method to reduce CO_2 emissions. Vertical mill and roller mill have about 30% energy saving than ball mill (Ali et al., 2011). Adoption and utilization of waste heat recovery (WHR) power generation technology is another way to reduce China's cement energy intensity. In the year of 2011, the installed capacity of WHR power generation reached 4786 megawatts (MW) (Zuo and Yang, 2011). Ke et al. (2013), based on the clinker production, assumed that 36 kWh electricity was generated by the production progress of per ton clinker, which can typically provide 25–33% of a cement facility's electricity demand for cement production (Zeng, 2009). Other equipments, such as high-efficiency classifiers, high efficiency motors and frequency implementations are also used for decrease electricity consumption and reduce CO_2 emissions.

5. Conclusions

Cement production as one of main sources of CO_2 emissions has received worldwide concern. China is the biggest producer and CO_2 emitter in cement industry. Thus the cement industry is a critical sector in China to meet its national 40–45% carbon intensity reduction target. Conventional cement emission factor can be used to evaluate roughly national/regional cement emissions, but it ignores some changing facets like the alternative materials used in clinker production, low clinker to cement ratio, and other factors. After onsite empirical studies we argued that the estimations by the conventional emission factor might overestimate the actual state. The input method, TC method, and regional electricity emission factor based on 15 collected cement plants are applied to compare the discrepancies of three different emissions with traditional method. The input method based on the three elements, CaO and MgO content in carbonate-containing material, materials ratio in raw meal, and raw meal clinker mass ratio are some reasonable parameters to measure the process emission in clinker production by getting rid of the CaO and MgO content in non-carbonate materials. It helps provide more insights about alternative material use and clinker quality changes in cement production, and can be applied to many other countries in the world.

This article reports the CO_2 emission results of onsite cement plants in China. Accordingly, almost all of the process emissions by the input method are lower than those by the output method. About 13 kg and 11 kg process emissions based on input method are lower than output method for NSP kiln and shaft kilns, respectively. This comparative result shows that CaO and MgO in clinker are not in the form of carbonate materials, and the output method magnifies CO_2 emissions by the $CaCO_3$ and $MgCO_3$ decompose. Therefore, we strongly suggest to calculate clinker process emission with CO_2 content in raw meal and raw meal clinker mass ratio. The CO_2 content in raw meal is estimated by the CaO and MgO content in carbonate-containing material and their material ratio to calculate CO_2 content in raw meal; and the plant-specific raw meal clinker mass ratios based on the principle of mass balance is 1.51 t, lower than CSI default value (1.55 t). Compared with fuels emissions from collected plants, all fuel emissions based on LHV method is higher than that of TC method. Thus, CSI encourages companies to use TC content method in calculating plant-specific fuels emissions. Regional electricity emission factor are used to evaluate the indirect emissions from cement production; and the values indicate that there is no significant discrepancy of electricity emissions per ton clinker/cement in raw meal preparation and cement grinding stage, while this emissions are observably different in clinker production stage for NSP kilns and shaft kilns. According to the analysis, we conclude that replace carbonate-containing materials with non-carbonate materials is the main way to reduce calcining emissions, and the reduction potential of process emissions by adjusting clinker quality is limited (≤6%). Application of advanced kiln, efficiency equipments and technologies to decline fuel intensity, and adopting lower carbon content fuel are two main way to reduce the fuel emissions in cement production. Lower electricity emission factor and consumption by using more efficiency electrical facilities and recycling waste energy means lower indirect emissions.

The cement industry is a high pollutant emitting industry. From above comparison we found that the input method can reasonably and reliably represents actual process emission in present production situation, and can be widely used in other production lines. This method may be of interest for future studies to discuss the sensitivity of each input variables towards low process emissions. TC method is encouraged by international organization to calculate fuel emissions. Using more efficiency electrical facilities and recycling waste energy could decrease electricity consumption in cement production. The analysis of some major affecting factor for CO_2 emissions provides quantitative information to improve the production technology, use alternative resource and energy, and suggest policy-makers to set up more sustainable development strategy for cement industry.

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