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The spatial-temporal pattern of sintered flue gas emissions in iron and steel enterprises of China

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ABSTRACT

This study provides a comprehensive survey of emissions from the sintering process in China to give an indication of the actual emission situation. Survey data were taken from China's continuous emission monitoring system (CEMS) from 209 sintering plants from 2015 to 2016. The standard-reaching rate of national emissions and the oxygen content ratio of the sintering process were considered as two indicators to evaluate the emissions level. Moreover, the CEMS-based emission factor (EF) estimation method was applied to improve the accuracy of EF estimation compared with existing methods. The emission concentration of total suspended particles (PM_{TPS}), SO_2 and NO_x decreased, and the standard-reaching rate of these three pollutants increased from 92%, 94% and 98%, respectively, to almost 100% during the research period. The average emission concentration in eastern China was lower than that in western China. More samples exceeded the oxygen regulation of <16% in 2016 than in 2015. This indicates that most plants should control air leakage to decrease the oxygen content. The national monthly average EF of PM_{TPS} and SO_2 decreased from 0.013 to 0.009 kg/ton-sintering and from 0.036 to 0.026 kg/ton, respectively, during the research period. The EF of NO_x remained around 0.068 kg/ton. These results are much lower than the results of former studies.

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1. Introduction

The iron and steel industry (ISI) is one of the most energy-intensive industries in China, representing 15% of China's total energy consumption (NBS, 2016). As one of the major iron and steel production processes, the sintering process improves the quality of iron ore (Cheng et al., 2016; Fernández-González et al., 2017), and provides raw material for the Basic Oxygen Furnace (BOF), which produces about 90% of crude steel in China (CSDRI, 2015; Hasanbeigi et al., 2014). Nevertheless, as an energy-intensive process, sintering emits a range of pollutants including SO_2, NO_x, total suspended particles (PM_{TPS}) and dioxins. In particular, about 70% of the SO_2 emissions and 50% of the total pollutants in an integrated steelworks are emitted from this process (Chun et al., 2017; Zhou et al., 2016). Therefore, reducing emissions from the sintering process will have a great impact on the cleaner production of the ISI.

The Chinese government has enacted a strict emission standard to reduce the emissions from the sintering process. For example, the Emission Standard of Air Pollutants for Sintering and Pelletizing of Iron and Steel issued by the Ministry of Environmental Protection of China in 2012 (GB28662-2012) dictated that the emission concentration of PM_{TPS} discharged from sintering facilities should be

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no more than 50 mg/m^3 from 2015. Moreover, the limitation of SO_2 emission concentration decreased from 600 in the former version to 200 mg/m^3, and the limitation of NO_x emission concentration was decreased to 300 mg/m^3. The standard is even stricter in environmentally sensitive regions, such as the Beijing-Tianjin-Hebei region and its surrounding area (M.E.E., 2012). For example, the emission standard in Hebei, which produces 24% of national crude steel, stipulates that concentrations of PM_{TPS}, SO_2 and NO_x emitted from sintering process should be no more than 50, 80 and 300 mg/m^3, respectively (Department of Environmental Protection of Hebei, 2015). Shandong province produces 9% of national crude steel. In this region, the sintering process should emit less than 30, 100 and 300 mg/m^3 of PM, SO_2 and NO_x, respectively (Departmeng of Environmental Protection of Shandong, 2013).

Studies have applied a variety of methods to reduce the greenhouse gas and air pollutant emissions from the perspective of the whole industry (Dong et al., 2013; Hasanbeigi et al., 2016; Li et al., 2018; Peng et al., 2018; Sun et al., 2017). However, few researchers have further explored the sintering process (Aries et al., 2006; Kasai et al., 2001). Model simulation is the most common research method as it can detail the reaction conditions in the iron ore sintering bed based on technology updating or policy implementations, and thus extrapolate the future emission reduction potential, energy intensity improvement or production process optimization (Hasanbeigi et al., 2014; Yang et al., 2004). Emission inventory research is an effective way to account for the historical carbon emissions from each production process in ISI according to the energy consumption, production technology and product output (Gao et al., 2019; Townsend-Small et al., 2015; Wang et al., 2016). Technology updating (Mayer et al., 2019), control and optimization of reaction temperature and other parameters in the sintering process (Scarlett et al., 2004) and introducing renewable energy as substitutes for fossil fuels (Gan et al., 2012; Ooi et al., 2008; Zandi et al., 2010) are the main means of reducing CO_2 and air pollutant emissions in the sintering process.

Besides the theoretical research mentioned above, the real emissions from the sintering process can be confirmed by field investigations. For example, Liu et al. (2012) investigated nine sintering plants in China and found that the polychlorinated naphthalene concentrations emitted from those plants ranged from 3 to 983 ng/m^3 under different operation scales. Nakano et al. (2009) detected dioxin emissions from the sintering process using pot tests. Similarly, Kasama et al. (2006) detected the release position of dioxin during the sintering production, taking the Oita No. 1 sintering plant as an example. The formation mechanism of dioxins was then estimated and corresponding dioxin emission reduction measures were proposed.

However, those existing studies mainly focused on a specific region or enterprise with limited sample size and detection time. Therefore, these studies are not sufficient to verify the theoretical accounting results, reflect the emission level in different regions and different production capacities, and consider the clean production level of the whole ISI in China. Moreover, these field observation studies neglected quantitative analysis of the air pollutant concentrations and the spatial characterization of the emissions. As a result, there is currently insufficient information about the priority, geographic guidance and time schedule of each iron and steel enterprise in terms of emission reduction. With increasingly stringent emission standards, the ISI in China is facing tremendous pressure to reduce emissions. It is therefore urgent to identify the current concentration and clean production levels for the sintering process within the ISI.

To fill in these gaps, this research selected 209 national-controlled sintering plants, including about one thousand sintering facilities in China, as the research objects. The PM_{TPS} and gaseous pollutant emissions from those facilities were stably detected by a continuous emission monitoring system (CEMS). Detection data in 2015 and 2016 were analyzed as these two years represent the implementation period of the 2012 version of the emission regulations, following a three-year grace period. This study aimed to (1) investigate the real-time emission of the sintering process with a large sample size and calculate the average emission concentrations of PM_{TPS}, SO_2 and NO_x at hourly, daily and monthly scales to reflect the emission variation trend of the sintering process; (2) compare the emission results with the national emission standard to analyze the standard-reaching rate at the national and provincial scale to understand the implementation of the policy; (3) detect the oxygen content to assess the emission dilution; and (4) calculate the emission factor (EF) of China's sintering process.

2. Data and methodology

China's CEMS is a direct and real-time measurement of emission concentrations for a variety of air pollutants and some auxiliary parameters such as flow rate, oxygen content, humidity and pressure (Fig. A-1). The continuous monitoring data were collected in terms of hourly averages at high spatial and temporal resolutions. This study extracted the emission data of PM_{TPS}, SO_2 and NO_x from China's sintering process in the CEMS system. A total of 1001 sintering blowdown stacks in 209 state-controlled[1] iron and steel plants from 28 provinces and municipalities in China are covered by the CEMS database (Fig. 1). The time span was from January 1, 2015 to December 31, 2016. Other auxiliary data, such as provincial GDP, were obtained from the China Statistical Yearbook 2016 and 2017 (NBS, 2016).

Although the reliability of CEMS data has been improved by strict specification, the CEMS dataset still contains null observations and extreme values. Therefore, preprocessing of the CEMS database is necessary according to the related official regulations, such as *The Data Availability Verification Rules for Automatic Monitoring of Enterprises Pollution* (Department of Environmental Monitoring, 2010) and *Technical Regulations for Comparing Monitoring results of Automatically Monitoring Devices for Pollution Sources* (China National Environmental Monitoring Centre, 2010; Tang et al., 2019). Abnormal values were removed, including zeros during operation periods, and impossible values beyond the measurement ranges of CEMS monitors. The remaining data set was used to calculate the standard-reaching rate and pollutant emissions (the number of valid data points is provided in Table A-1). Modification to account for missing data was based on *Specifications for Continuous Emissions Monitoring in the Flue Gas Emitted from Stationary Sources* (HJ/75−2007).

After data preprocessing, the average emission concentration from each blowdown stack at hourly, daily and monthly scales using the valid measurements was generated by using Eq. (1).

$$AEC_i^{b,T} = \frac{\sum SEC_i^{b,T}}{TSN_i^T} \quad (1)$$

where AEC is the average emission concentration of each blowdown stack by different counting period T; b is the identification

[1] State-controlled enterprises are the key state monitoring enterprises selected according to the "notice on the dynamic update of the list of key national monitoring enterprises" required by "the comprehensive work plan for energy conservation and emission reduction during the 12th Five-Year Plan", "opinions of the state council on strengthening key environmental protection work" and the revised environmental protection law.

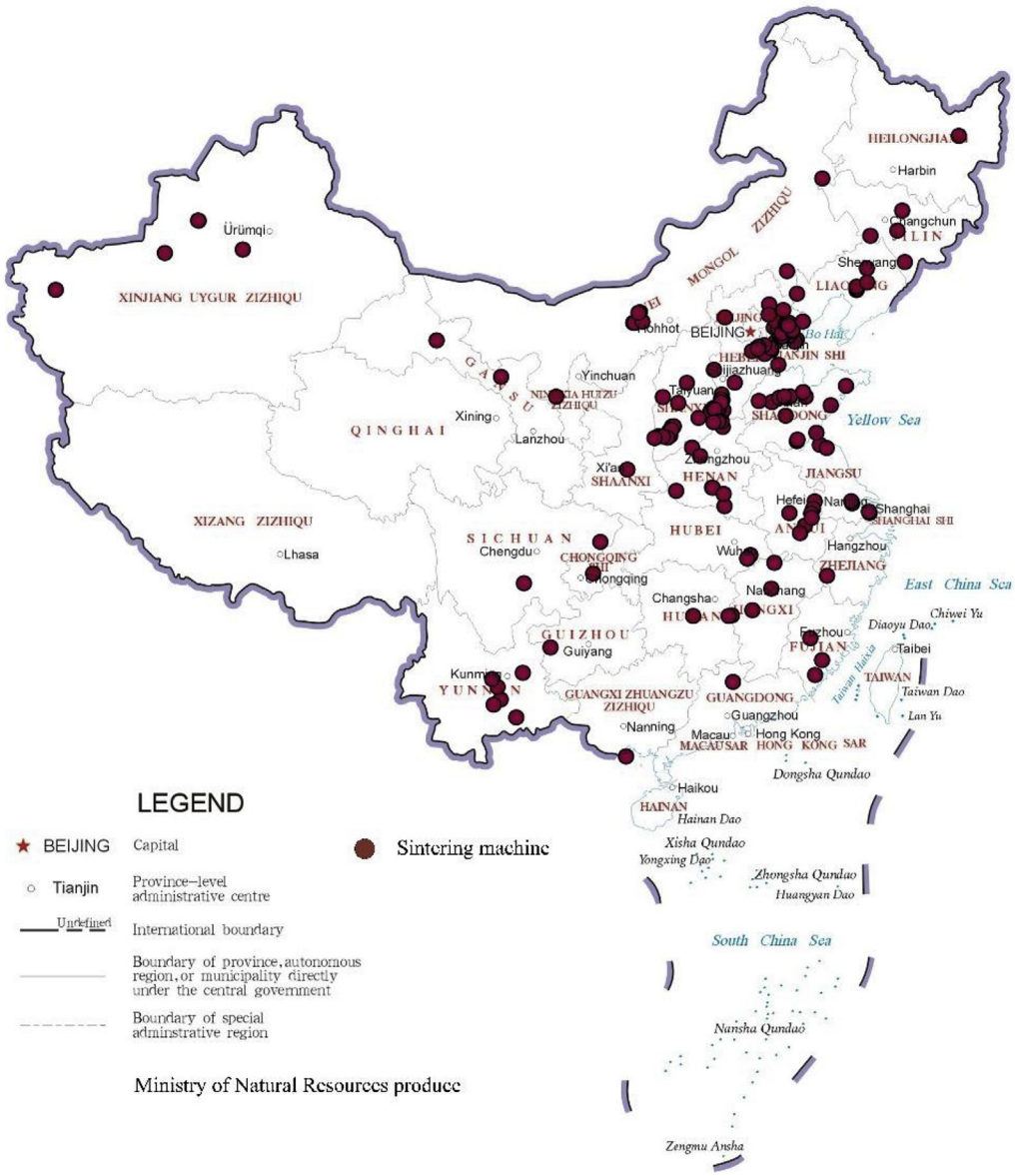


Fig. 1. Chinese sintering plant stacks included in the CEMS database in 2015 and 2016.

number of each blowdown stack; T is the counting period including hourly, daily and monthly scales; and i is the type of emissions; $\sum SEC_i^{b,T}$ is the sum of emission concentrations of i emitted from each blowdown stack detected during the calculation period T; TSN_i^T is the total sample size of each blowdown stack when the emission is i under counting period T.

Emissions from sintering plants can be assessed by comparing the emission concentration results from Eq. (1) with the national emission standards. The standard-reaching rate was proposed as an indicator and defined as the proportion of blowdown stack with emission concentrations lower than the national standards for the total number of gas flues (Eq. (2)). The more blowdown stacks emit less than national standard, the higher the standard reach rate. Consistent with the statistic above, the standard-reaching rate was also calculated on an hourly, daily and monthly basis. It is worth mentioning that to evaluate the standard-reaching rate uniformly, only the national emission standard was adopted and the regional standard was not considered.

$$SRR_i^{j,T} = \frac{NSR_i^{j,T}}{TSN_i^{j,T}} \tag{2}$$

where SRR is the standard-reaching rate of emission i in research scope j, such as national or provincial; NSR is the number of gas flue with emission concentrations lower than national standards in research scope j; and $TSN_i^{j,T}$ represents the total number of blowdown stacks in research scope j. For the standard-reaching rate of PM emissions (SRR_{PM}), NSR_{PM} includes the blowdown stacks with AEC_{PM} of $\leq$50 mg/m^3. Similarly, for SRR_{SO2} and SRR_{NOx}, the blowdown stack was counted if its AEC_{SO2} or AEC_{NOx} was no more than 200 and 300 mg/m^3, respectively.

The EFs of the sintering process were estimated based on the actual emission concentration data from the CEMS system using Eq. (3). As CEMS monitors were installed in the sintering stacks, detected emission concentrations were measured after the effect of pollution control technology. Therefore, the CEMS-based EF

estimation method is a direct measurement and the result can reflect the actual emission level of the sintering process in China. Thus, this method improves the accuracy of EF estimation greatly because the average and invariable EFs are estimated by many indirect parameters and are affected by the associated assumptions in previous studies, such as the net heating value and the removal efficiency of control technology (Zhao et al., 2008). Moreover, national EFs and those for different time series can also be provided using this method on the basis of real-time CEMS data (Eq. (3)). For more details, please refer to Tang et al. (2019).

$$EF_{i,\,t} = Con_{i,t} VCos \tag{3}$$

where *EF* represents the emission factor of the sintering process, expressed as pollutant emissions per unit of sintering product, g/kg-sintering; *Con* is the emission concentration from the sintering blowdown stack detected by the CEMS system (g/m^3); and *V* is the theoretical flue gas rate that is the flue gas volume per unit of fuel consumption (m^3/kg). Here, the energy source used for sintering is assumed to be 30% of coal and 70% of coke (Gui et al., 2013). Theoretical flue gas rates of coal and coke refer to the study of Tang et al. (2019) and *The Engineering Toolbox (www.EngineeringToolBox.com)*. *Cos* is the fuel consumption per unit of sintering product (kg fuel/ton), based on data from the China Steel Yearbook 2017. Finally, i and t represent the emission types and statistical time series, respectively.

It should be noted that as there is no information about raw material quality and production technologies, the differences in quality of raw materials and in technologies in each plant were not quantitatively analyzed. Moreover, provincial controlled enterprises were also not taken into consideration.

3. Results

3.1. Standard-reaching rate analysis

3.1.1. Hourly analysis

Fig. 2 summarizes the hourly standard-reaching rate at the national scale. In 2015 and 2016, NO_x reached 99% of the standard-reaching rate, the highest standard-reaching rate among the three analyzed pollutants. The standard-reaching rate of PM_{TPS} and SO_2 increased from 95% in 2015 to around 99% in 2016 and became more stable. Although the standard-reaching rate of PM and SO_2 fluctuated slightly throughout the day, there was no significant difference between each hour.

Table 1 shows the provincial scale analysis. Some regions, such as Tianjin, Guangxi, Shanghai and Sichuan, reached 100% of the standard-reaching rate of PM emissions in 2015 and 2016. Only 25% of emission data achieved the national standard in Guizhou in 2015. As most of the blowdown stacks in Guizhou were closed (no valid monitoring data) in 2016, the standard-reaching rate was much better in 2016. Results in Liaoning, Hebei, Inner Mongolia, Jiangxi, Guangdong and Shandong also improved greatly during the research period. However, the standard-reaching rate in Shanxi decreased slightly.

The standard-reaching rate of SO_2 was 100% in Tianjin, Heilongjiang, Shanghai, Jiangsu, Zhejiang, Anhui, Fujian, Shandong, Henan, Hubei, Guangxi and Ningxia in 2015 (Fig. A-2). In 2016, only Liaoning and Yunnan were around 90% of the standard-reaching rate for SO_2, whereas other provinces all achieved the national SO_2 emission standard (100%).

The standard-reaching rate of NO_x emissions in most provinces reached 100% in 2015 (Fig. A-3). However, Gansu only had 67%–75% of emission data meeting the national regulations. Xinjiang only had an 80% standard-reaching rate in 2016, whereas other provinces fully satisfied the national NO_x emission limitation.

3.1.2. Daily analysis

The daily standard-reaching rate of NO_x emissions from the sintering process increased from 97% in early 2015 to almost 100% in 2016 (Fig. 3). In early 2015, 93% of discharge concentrations from sintering blowdown stacks achieved the national standard of SO_2 emissions and this ratio increased to almost 100% by the end of 2016. The PM_{TPS} emissions from the sintering process had the lowest standard-reaching rate compared with other pollutants, at between 92% and 97% in the first half of 2015 and increased gradually to about 99% by the end of 2016.

From the provincial perspective, Tianjin, Heilongjiang, Zhejiang, Guangxi and Ningxia achieved a 100% standard-reaching rate of PM_{TPS} emissions for daily data in 2015. The PM_{TPS} emissions in Shanghai, Anhui and Fujian exceeded the national emission standard on several days. The standard-reaching rate of PM_{TPS} in Jiangsu and Henan province was lowest in early August and mid-July, respectively. Similarly, Tianjin, Heilongjiang, Zhejiang, Fujian, Guangxi, Shaanxi, Ningxia achieved a 100% standard-reaching rate of SO_2 emissions in 2015. The SO_2 emissions in Shanghai and Anhui did not meet the national standard for only a few days. Standard-

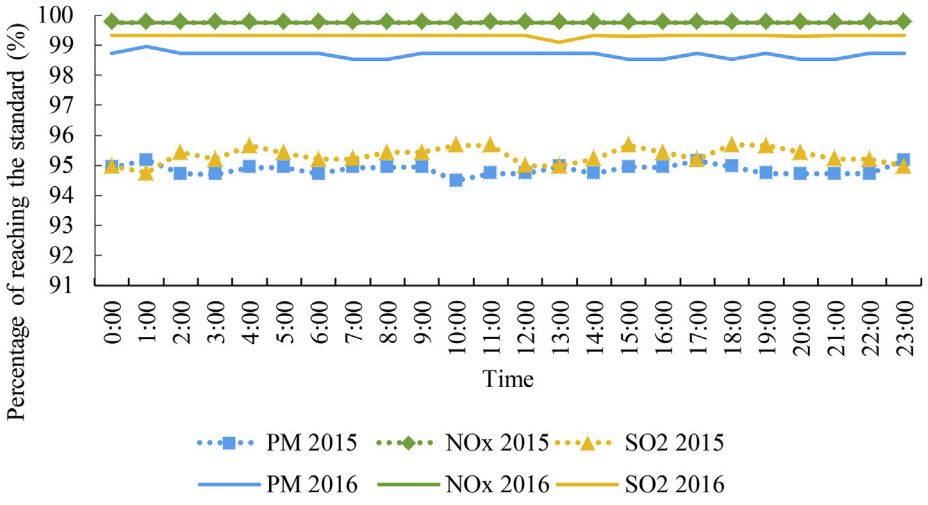


Fig. 2. Hourly average analysis of standard-reaching rates for PM, NO_x and SO_2 emissions.

Table 1
Standard-reaching rate of hourly average PM emissions in each province in 2015 and 2016.

	2015								2016							
	0:00	3:00	6:00	9:00	12:00	15:00	18:00	21:00	0:00	3:00	6:00	9:00	12:00	15:00	18:00	21:00
Tianjin	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Hebei	99.5%	99.5%	99.5%	99.5%	99.5%	99.5%	99.5%	99.5%	99.5%	99.5%	99.5%	100.0%	100.0%	100.0%	99.5%	99.5%
Shanxi	87.5%	91.7%	87.5%	91.7%	91.7%	91.7%	87.5%	88.0%	90.6%	90.6%	90.6%	90.6%	90.6%	90.6%	90.6%	90.6%
Inner Mongol	92.9%	92.9%	92.9%	92.9%	92.9%	92.9%	92.9%	92.9%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Liaoning	77.3%	77.3%	77.3%	77.3%	77.3%	77.3%	77.3%	77.3%	90.9%	90.9%	90.9%	90.9%	91.3%	91.3%	90.9%	91.3%
Jiling	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Heilongjiang	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Shanghai	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Jiangsu	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Zhejiang	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Anhui	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Fujian	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Jiangxi	85.7%	85.7%	85.7%	85.7%	87.5%	85.7%	85.7%	85.7%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Shandong	82.1%	82.1%	82.1%	82.8%	82.8%	89.7%	85.7%	85.7%	100.0%	100.0%	100.0%	100.0%	100.0%	97.1%	100.0%	100.0%
Henan	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Hubei	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Hunan	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Guangdong	85.7%	85.7%	85.7%	85.7%	71.4%	71.4%	85.7%	85.7%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Guangxi	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Chongqing																
Sichuan	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Guizhou	25.0%	25.0%	25.0%	25.0%	25.0%	0.0%	25.0%	25.0%	100.0%	100.0%	100.0%	75.0%	75.0%	75.0%	75.0%	75.0%
Yunnan	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	92.3%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Shaanxi	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Gansu	75.0%	75.0%	75.0%	75.0%	66.7%	66.7%	75.0%	75.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Qinghai																
Ningxia	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%
Xinjiang	100.0%	80.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%	100.0%

Note: a darker green shade represents a low standard-reaching rate.

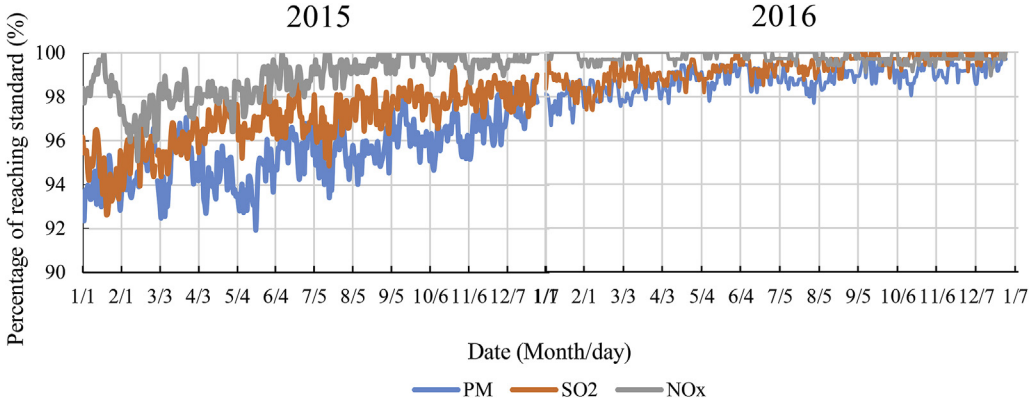


Fig. 3. Daily standard-reaching rate of PM, NO_x and SO_2 emissions.

reaching rates in Shandong, Henan and Hubei were lower in spring and summer than in other seasons. The NO_x emissions in most provinces met the national standard, except for Inner Mongolia, Liaoning and Shanghai.

3.1.3. Monthly analysis

Similar to the daily analysis, the monthly standard-reaching rate of the sintering process increased rapidly from 2015 to 2016 (Fig. 4). The PM_{TPS} emissions had a larger improvement than SO_2 and NO_x emissions, as the standard-reaching rate increased from 93% in January 2015 to almost 100% in December 2016. The standard-reaching rate of SO_2 emissions also increased by 6% during these two years. It even reached 100% in October and November of 2016 but declined slightly in December 2016. The NO_x emissions still had the highest standard-reaching rate among these three pollutants; although its percentage reduced to 97% in February 2015, it increased quickly and reached 100% as early as October 2015.

Considering the average monthly PM_{TPS} emission standard-reaching rate, Tianjin, Shanghai, Anhui, Fujian, Hubei, Heilongjiang, Zhejiang, Guangxi and Ningxia fully met the national standard in 2015 (Table 2). The PM_{TPS} emissions in Jilin, Jiangsu, Henan, Hunan, Sichuan, Shanxi and Xinjiang exceeded the national

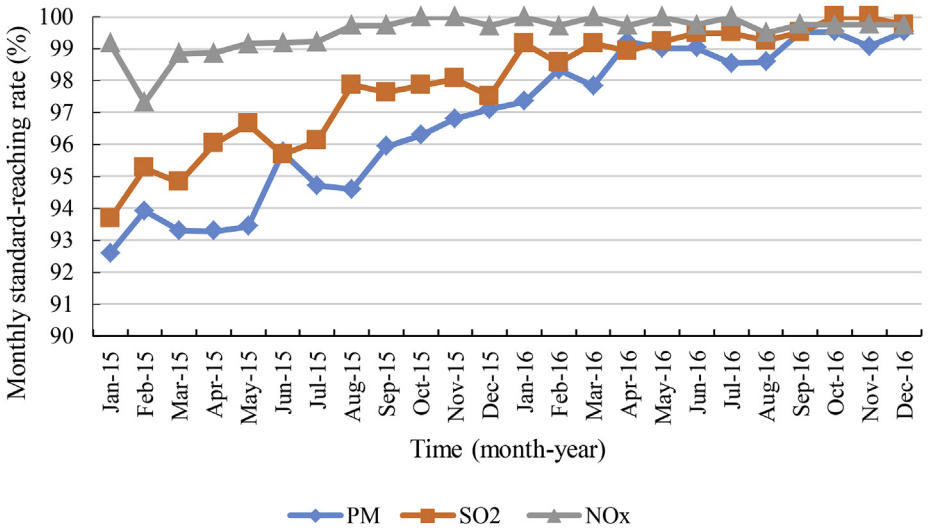


Fig. 4. Monthly standard-reaching rate of PM, NO_x and SO_2 emitted from the sintering process.

Table 2
Monthly analysis of the standard-reaching rate of PM in each province for 2015–2016.

	2015						2016					
	Jan	Mar	May	Jul	Sept	Nov	Jan	Mar	May	Jul	Sept	Nov
Tianjin	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%
Hebei	99%	98%	99%	97%	99%	100%	99%	100%	99%	100%	100%	100%
Shanxi	81%	93%	82%	94%	89%	94%	90%	90%	96%	96%	96%	93%
Inner Mongo	80%	64%	82%	93%	92%	92%	92%	100%	100%	100%	100%	100%
Liaoning	90%	90%	90%	83%	85%	89%	83%	89%	89%	90%	100%	91%
Jilin	75%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%
Heilongjiang	100%	100%	100%	100%	0%	0%	0%	0%	0%	0%	100%	100%
Shanghai	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%
Jiangsu	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%
Zhejiang	100%	100%	100%	100%	0%	100%	100%	100%	100%	100%	100%	100%
Anhui	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%
Fujian	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%
Jiangxi	71%	71%	86%	88%	75%	86%	100%	100%	100%	100%	100%	100%
Shandong	79%	84%	80%	84%	88%	91%	88%	96%	100%	100%	100%	100%
Henan	100%	100%	100%	97%	100%	100%	100%	100%	100%	100%	100%	100%
Hubei	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%
Hunan	100%	100%	100%	67%	100%	100%	100%	100%	100%	100%	100%	100%
Guangdong	60%	43%	40%	80%	100%	100%	100%	100%	100%	100%	100%	100%
Guangxi	100%	100%	100%	100%	100%	100%	100%	0%	100%	100%	100%	100%
Chongqing												
Sichuan	100%	100%	100%	100%	0%	0%	0%	0%	0%	100%	100%	100%
Guizhou	25%	0%	25%	100%	75%	25%	100%	50%	100%	50%	100%	100%
Yunnan	75%	83%	89%	90%	100%	100%	100%	100%	100%	100%	100%	100%
Shaanxi	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%
Gansu	100%	67%	50%	100%	100%	100%	100%	100%	100%	100%	100%	100%
Qinghai												
Ningxia	100%	100%	100%	100%	100%	100%	0%	100%	100%	100%	100%	100%
Xinjiang	0%	0%	67%	100%	100%	100%	100%	100%	100%	50%	80%	100%

Note: a darker green shade represents a low standard-reaching rate.

standard for a few months. Guizhou presented a low standard-reaching rate of PM_{TPS} in winter and spring. In 2016, most of the provinces fully achieved the standard-reaching rate, except Shanxi, Liaoning, Hebei, Inner Mongolia, Shandong, Guizhou and Xinjiang.

The standard-reaching rate of SO_2 in 2015 was better than that of PM_{TPS} emissions. Besides the provinces mentioned above, Jiangsu, Shandong and Shaanxi also achieved a 100% standard-reaching rate of SO_2 emissions. Moreover, the standard-reaching rate in most of the remaining provinces was higher than 75%, except for a few months in Guangdong, Hunan, Gansu and Xinjiang. In 2016, only Liaoning, three months in Guizhou, spring in Yunnan and April in Hebei did not achieve the 100% standard-reaching rate

(Fig. A-4).

For the NO_x emissions, besides spring in Gansu, other provinces had a high standard-reaching rate in 2015. In 2016, only autumn and winter in Xinjiang, June in Guizhou, May in Jilin and April in Hebei did not achieve the 100% standard-reaching rate of NO_x emissions (Fig. A-5).

3.2. Emission concentration of air pollutants from the sintering process

3.2.1. Average hourly emission concentration

The average hourly emission concentration of air pollutants from the sintering process in each province was calculated and is shown in Fig. 5. In 2015, the PM_{TPS} concentration in Guizhou was higher than that of the other provinces, reaching 58–62 mg/m^3 in one day. Conversely, Shanghai, Tianjin and Heilongjiang presented the lowest PM_{TPS} concentrations of 13–17 mg/m^3 in one day. In 2016, the PM_{TPS} concentration in Guizhou decreased to 40 mg/m^3, but remained the province with the highest PM_{TPS} emissions. Tianjin had the second lowest PM_{TPS} emissions, at only about 13 mg/m^3.

For SO_2, all areas reached the national standard except for Xinjiang, which had concentrations of 11 mg/m^3 higher than the national standard. Sintering equipment in Shanghai and Tianjin emitted the lowest SO_2 concentrations with average values of 23 and 33 mg/m^3, respectively. However, the emissions in Guizhou and Sichuan province were much higher than other provinces, at 155 and 146 mg/m^3, respectively. In 2016 the SO_2 emissions in Xinjiang decreased to 46 mg/m^3. Instead, Guizhou became the biggest SO_2 emitter, and the average emissions within one day were 127.60 mg/m^3 (data not presented).

Shanxi had the highest NO_x concentration in 2015, followed by Shanghai and Guangdong. Xinjiang emitted the least NO_x. In 2016, Guangdong and Shanghai became the biggest NO_x emitters, and Heilongjiang emitted the least NO_x during the production process (data not presented).

3.2.2. Average monthly emission concentration

Monthly average emissions of PM_{TPS} in each province are shown in Fig. 6. Guizhou was the largest emission source of PM_{TPS}, and the average concentration was 57 mg/m^3. However, there were no obvious temporal characteristics. In 2016, the average concentration of PM_{TPS} in Guizhou decreased to 39.0 mg/m^3.

For the SO_2 emissions in 2015, Xinjiang had the highest SO_2 concentration, especially in winter and spring (Fig. A-6). Shanghai had the lowest SO_2 emissions and the average concentration was only 23 mg/m^3. This was followed by Tianjin with an average SO_2 concentration of 33 mg/m^3. The SO_2 concentration generally declined in 2016. Shanghai still had the lowest SO_2 emissions, and the average concentration was reduced to 18 mg/m^3. Guizhou emitted the highest SO_2 concentration of 224 mg/m^3.

The highest NO_x emissions in 2015 were found in Gansu, with a value of 442 mg/m^3 in May; however, the average value was only 168 mg/m^3 for other months. Xinjiang had the lowest NO_x emissions. In 2016, the NO_x concentration in Shaanxi decreased, whereas it increased quickly in Xinjiang in the second half of the year (Fig. A-7).

3.3. Oxygen content analysis

The oxygen content reflects the degree to which emissions are diluted by air. To avoid air dilution and reveal the actual emission level, the revised Emission Standard of Air Pollutants for Sintering and Pelletizing of Iron and Steel (GB28662-2012) specified that the baseline of oxygen content from sintering flue gas was 16%. If the oxygen content from one blowdown stack is higher than 16%, it means that the air pollutants emission is overly diluted, and the actual emission concentration is higher than the detected result. Thus, the detected concentration of air pollutants should be converted to the concentration when the oxygen content is 16% to show the actual emission level. Then, the converted result was used in determining whether the emissions reach the standard (M.E.E., 2017). Although this revised standard was implemented on October 1, 2017, we discuss it in terms of the emission database in 2015 and 2016 to evaluate the existing emission improvement and understand what efforts are required to meet the new standard.

In 2015, 20% to almost 100% of detected data in each province did not meet the oxygen content regulation, which indicates that dilution of emissions by air leakage in the sintering process is a common phenomenon (Fig. 7). Correspondingly, the actual emissions from those samples should be higher than the current results and the standard-reaching rate would correspondingly become lower.

In 2016, air leakage became more serious in some provinces such as Xinjiang, Gansu, Yunnan, Heilongjiang, Sichuan, Hunan, Hubei, Shanxi and Anhui because the ratio of high oxygen content was higher than that in 2015. In particular, the number of samples with a 18%–19% oxygen content rate greatly increased from 2015 to 2016 in Heilongjiang, Zhejiang, Sichuan and Guizhou province. This result also implies that the high air leakage is one of the reasons for

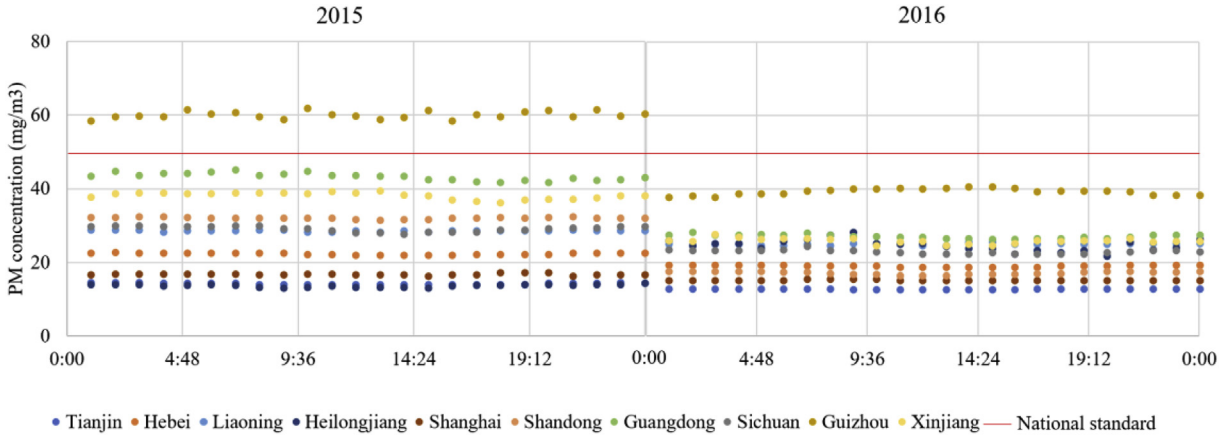


Fig. 5. Hourly analysis of PM_{TPS} concentrations from sintering equipment in a selection of representative provinces in 2015 and 2016. The red line indicates the national PM_{TPS} emission standard. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

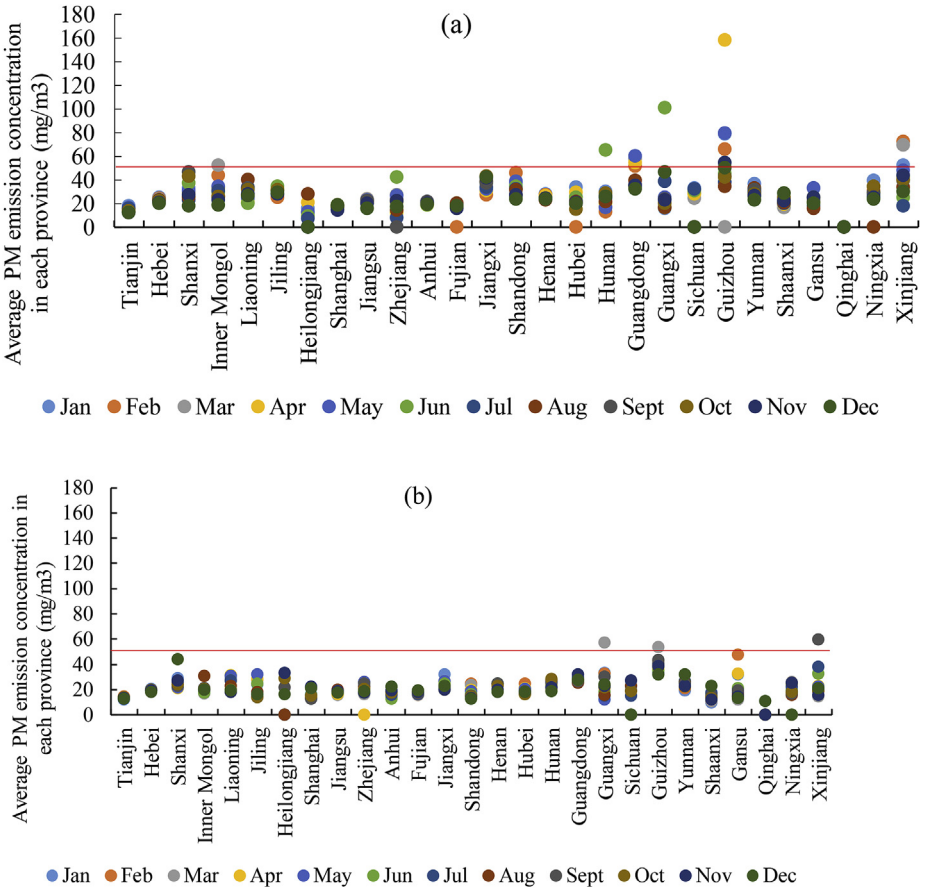


Fig. 6. Monthly analysis of PM emission concentrations from sintering equipment in each province in 2015 (a) and 2016 (b). The red line indicates the national PM emission standard. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

the high standard-reaching rate in these provinces in 2016, in addition to technological progress. Thus, decreasing the oxygen content ratio and improving emission quality in these provinces may have become more difficult once the new regulation came into effect in 2017. In contrast, Hubei, Jilin, Shanghai, Shandong, Guangxi and Guangdong reduced the air leakage, and more samples met the national oxygen content ratio standard (Fig. 7).

3.4. Emission factors of the sintering process

Fig. 8 shows the monthly average PM_{TPS} EF of the sintering process at the national and provincial level during the research period. The national average EF decreased from 0.013 to 0.009 kg/ton-sintering during the research period. The monthly average EF in each province ranged from 0.001 to 0.075 kg/ton. Most of the province values were concentrated between 0.009 and 0.017 kg/ton. Chongqing and Guizhou had the highest EF among 28 provinces in 2015. Conversely, most of the blowdown stacks were closed (no valid monitoring data) in 2016, and the remaining finite samples had lower EFs than the level in 2015. Some other provinces in western China, such as Inner Mongolia and Xinjiang, also had relatively higher EF of PM_{TPS} than other regions. The national average EF of SO_2 decreased from 0.036 to 0.026 kg/ton during the research period. In contrast, the EF of NO_x remained steady during the research period. More information about the provincial EFs of SO_2 and NO_x is shown in Figs. A-8 and A-9.

The oxygen content could be a source of uncertainty as a higher oxygen content may underestimate the emissions, as well as the EF. However, in some provinces, such as Guangxi, Guangdong, Shanghai and Jilin, the oxygen content decreased, and more samples met the national regulation in 2016 compared with 2015 levels. Whereas, the EFs decreased in these provinces during the research period.

3.5. Comparison with other studies

Table 3 summarizes the annual average EFs of air pollutants from China's ISI in different studies. The estimation results of PM_{TPS} EF and SO_2 EF in this work were much lower than other studies, and NO_x EFs were similar among the different studies. Different estimation methods may result in these differences. As mentioned in the methodology section, the CEMS-based EF estimation method provides direct and actual measurements. In contrast, other studies depend mainly on indirect parameters, such as energy consumption, and the remove effect of pollutant control facilities. Different study years may also explain the different results as the pollutant control facilities for the ISI developed rapidly in the last two decades in China.

4. Discussion

4.1. Standard-reaching rate and regional differences

The ISI in China has experienced rapid development in the past two decades and produces almost half of the world's total crude steel (Worldsteel, 2017). With the improvement of environmental

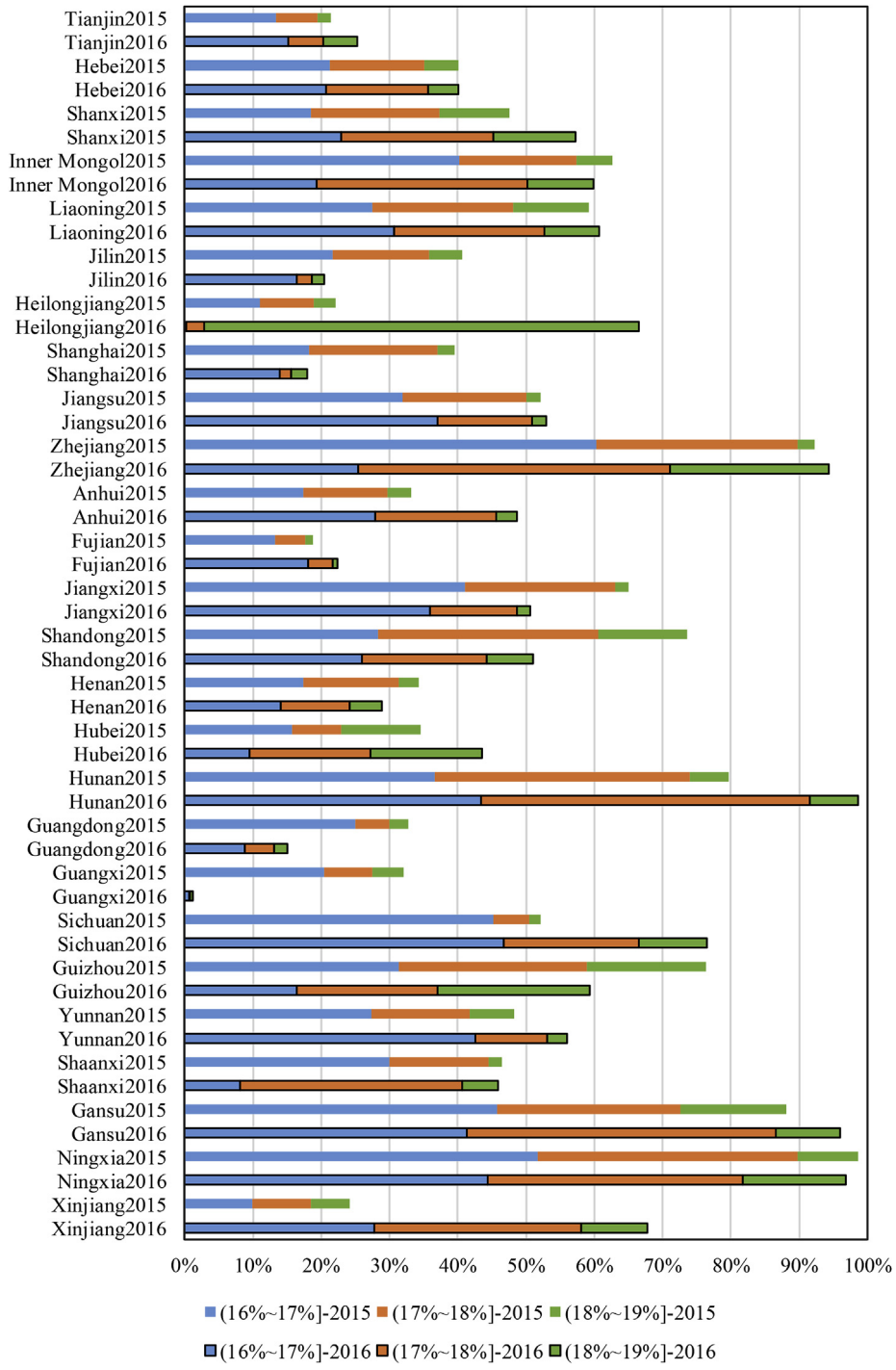


Fig. 7. Percentage of the CEMS database with a >16% oxygen content rate in 2015 and 2016.

awareness, the Chinese government has released a series of policies to reduce the pollutant emissions of sintering plants (Table A-2). The sintering process is an essential technique by which to realize clean production in the ISI and the regulation of the sintering process is more stringent than that of other procedures in the latest ultra-low standards. In this research, we analyzed the effect of the national standard by collecting the CEMS data from sintering equipment and then calculated the EF for the sintering process. More than one thousand sites of sintering equipment distributed across 28 provinces in China were included.

The Emission Standard of Air Pollutants for Sintering and Pelletizing of Iron and Steel (GB 28662–2012) issued in 2012 is used as the reference policy and required that all sintering plants followed the rule since 2015 after a three-year grace period. Even so, some enterprises still failed to meet this standard. Fortunately, the results indicate a rapid improvement of the standard-reaching rate from 93% in 2015 to 98% in 2016. As the production process is continuous, the standard-reaching rate and emission concentration show no obvious temporal features.

The daily or monthly average emission concentrations

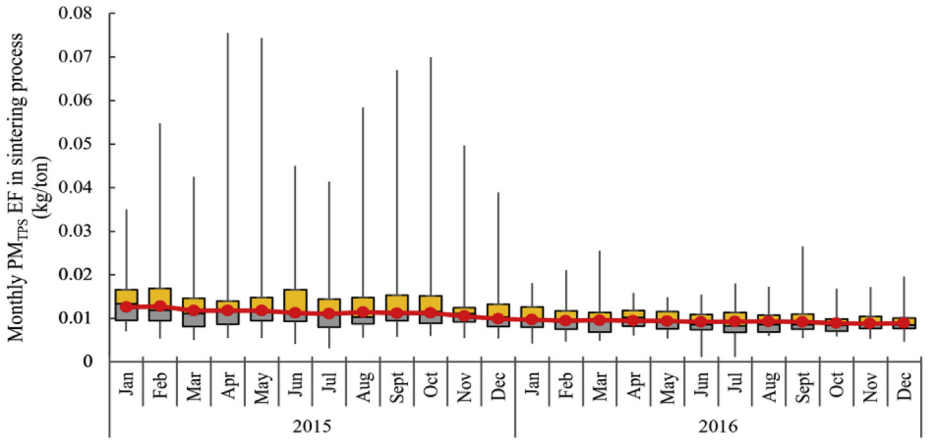


Fig. 8. Monthly EF of PM_{TPS} in the sintering process. Note: Boxplot shows the distribution of EF in each province. The red line is the national monthly average EF. . (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 3
Emission factors (EFs) of air pollutants from the sintering process in China in different sources.

	EF of PM_{TPS} (kg/ton sinter)	EF of SO_2 (kg/ton sinter)	EF of NO_x (kg/ton sinter)	Year
This work	0.011	0.033	0.068	2015
This work	0.009	0.028	0.068	2016
Zhao et al. (2017)	0.121	–	–	–
Guo et al. (2017)	0.14	0.62	0.62	2015
Wang et al. (2016)	1.0	3.2	0.6	2011
Wu et al. (2015)	0.402–1.590	2.400–2.800	0.522–0.612	2012

decreased during the research period and the standard-reaching rate showed a fluctuating but generally increasing trend. Technology improvement is an important factor. The China Environmental Statistics Bulletin (http://www.mee.gov.cn/) reported the installation of terminal treatment equipment by investigating 1000–1100 sintering facilities. It showed that number of dust-cleaning apparatus was 16162 in 2014, which increased to 16337 in 2015. As there is no economical and effective end treatment technology for denitration of flue gas in the sintering process, the reduction in the emission concentration of NO_x is mainly achieved by improving the quality of raw materials and controlling the reaction conditions (Zhou et al., 2016). Therefore, the number of denitration facilities was only 39 devices in 2014, which increased to 63 in 2015. The sintering process contributes about 70% of the SO_2 emissions of the ISI (Chun et al., 2017). Therefore, desulfurization is an important task in the sintering process. The proportion of sintering machines with desulfurization facilities was only 29.5% in 2012 and increased to 62.5% in 2015 (M.E.E., 2015).

Air pollutant emissions in western China (e.g., Sichuan, Yunnan, Shanxi and Xinjiang) were higher than those in other regions, and southeast provinces emitted more NO_x than other regions. There are two main factors that may lead to this result. The first is the higher EF and lower raw material quality in western China. For example, the SO_2 EF of sintering in western China is between 2 and 3.4 kg/ton, whereas it is between 1.9 and 2.6 kg/t in other regions (Gao et al., 2019). Coal quality is another important factor. The ash content in coal and coke can reduce the calorific value and affect production efficiency. The coke ash content in developed countries is around 10%, whereas it is 13% in China. In southwest China, the coke ash content can reach as high as 18% (China Metallurgical Technology Network, 2011; Ouyang and Lin, 2015), leading to higher energy consumption and pollutant emissions. Eastern China has convenient water and land transportation and therefore high quality raw materials from both domestic and international sources are available in Shanghai (Association, 2018; China Iron and Steel Association, 2015). Secondly, the strength of policy implementation could lead to the variation in air pollutant concentrations. East China is facing serious environmental problems and therefore policy enforcement in this region is stronger than that of other regions (MIIT, 2012). However, eastern China, especially Shanghai, has a geographical advantage in terms of economic strength and policy support for updating the technologies (Diao et al., 2016; Mok and Nam, 1999) (Yao et al., 2015).

4.2. Oxygen content analysis and EFs

The oxygen content ratio of sintering chimneys can be decreased by controlling the air leakage. Correspondingly, new sintering facilities should stabilize equipment operation and strengthen equipment management to reduce air leakage. The existing facilities should be aware of the gap between actual emission concentration and the national standard. As for the high oxygen content in current samples, the pollutants concentration will increase after reducing the oxygen content caused by air leakage. However, after reducing the air leakage during production process, the gas removal efficiency might be improved by reducing the workload of the waste gas purification apparatus. Thus, the final concentration of discharged air pollutants could be further decreased.

The main source of anthropogenic carbon emissions and air pollutant emissions is fossil fuel combustion. The Intergovernmental Panel on Climate Change (IPCC) provided an emission calculation guideline to unify the accounting method of carbon emissions from energy consumption (IPCC, 2006). It provides the default values of different kinds of fossil fuels to support the calculation. However, by considering the regional differences in resources, the EFs for Chinese coal are on average 40% lower than the default values recommended by the IPCC. Moreover, emissions

calculated by different organizations based on this "energy consumption approach" differ by 0.3 Gton (Liu et al., 2015). The CEMS-based emission calculation method promoted in this study can eliminate the uncertainty of emission estimation as it bypasses different energy characteristics, alongside eliminating uncertainty from mixing energy combustion and the operating efficiency of environmentally friendly equipment. This approach is therefore an effective way to account for actual carbon emissions and verify the results calculated by the "energy consumption approach".

4.3. Policy suggestions

Through the analyses above, several concerns and suggestions can be proposed to promote clean production in the ISI in China. Because of the increasing oxygen content and relatively lower standard-reaching rate in some provinces in western China, it is of vital importance to undertake sustainable development in this region. Technology is always the foundation to reduce emissions and achieve clean production. Therefore, advanced technologies should be installed on a wider scale in all sintering facilities to cope with the stricter national emissions standard. For example, the install ratio of small sintering process was only 40% by 2010. This technology could reduce energy consumption by 10%–15% and improve production efficiency by 15%–20%. Some other technologies with a lower installation rate, such as sintering flue gas desulfurization technology, are also effective to improve the emission quality (MIIT, 2012).

In addition, waste energy reuse is an efficient way to decrease energy consumption and air pollutant emissions, which will be the future development direction for cleaner production. Recycled waste energy is a good alternative to fossil fuels. For example, the recovered heat can be reused in the production process or used as an energy source for raw material preheating. It also can be provided to other industries, such as the food processing industry, or to households to generate profit (Kim et al., 2018). Waste heat can also be used for power generation to reduce power consumption during the production process.

Finally, upgrading the production structure is another way to conserve energy and decrease pollutant emissions. Development of the electric arc furnace (EAF) to produce crude steel can effectively reduce air pollutant emissions from the sintering process because of the cleaner production process and lower energy intensity of EAF compared with BOF. As the raw material of EAF, limited ferrous scrap resources represent one of the factors restricting the development of EAF. Fortunately, some studies have shown that ferrous scrap will increase quickly after 2016 because of the huge steel stock in China after a rapid infrastructure construction. Correspondingly, the EAF usage is predicted to increase to more than 40% of total by 2050 (Li et al., 2017, 2019; Sun et al., 2018a, 2018b).

5. Conclusion

This study analyzed the emission of air pollutants from sintering equipment and the proportion of plants reaching the national emission standard of the sintering process in China in 2015 and 2016 using CEMS database. The CEMS is a real-time data source with good quality and good consistency. Sampling sites in this database have a good broad coverage of China. By using this database, the effect of policy implementation and spatial distribution of gas emissions from the sintering process was analyzed. Results show that the air pollutant concentrations decreased gradually from 2015 to 2016, indicating effective policy execution. Eastern China had good performance in decreasing the emissions in the sintering section. In contrast, the standard-reaching rate in western China was lower than eastern China. The oxygen content increased in the tested samples, indicating increased air leakage. It is therefore necessary to promptly improve policy of the oxygen content to show the actual pollutant emission levels. The CEMS-based EF calculation method was promoted in this research, and it was found that the energy consumption-based EF calculation method overestimated the EF of the sintering process. Based on the results of this research, policy suggestions were proposed including promoting advanced technologies, implementing stricter policies and comprehensively investigating the impact on enterprises before implementing a new standard.

By analyzing the real-time emission monitoring data in the sintering process, it is possible to identify the reaction of enterprises to policy, so that the appropriate measures for pollutants control from iron and steel enterprises in China can be proposed for different regions and different plants. However, this research did not consider other harmful emissions in the sintering process, such as dioxin. Moreover, the analysis of oxygen content ratio in this research focused only on the distribution number of detection data in each interval value, which also causes the uncertainty in EF calculations. In the next step, a greater range of different types of pollutants will be studied. Oxygen content ratio data will be matched to the time series emission information, and the emission concentration will be converted to the actual level by removing the effects of oxygen dilution. Estimations of the actual emission amount will also be provided in the subsequent study to allow the analysis and policy suggestions to become be more targeted.

CRediT authorship contribution statement

Xin Bo: Project administration, Funding acquisition, Data curation, Writing - original draft, Writing - review & editing. **Zhaoling Li:** Methodology, Supervision, Writing - review & editing, Writing - original draft. **Jiabao Qu:** Data curation. **Bofeng Cai:** Project administration, Supervision. **Beihai Zhou:** Methodology, Writing - review & editing. **Lu Sun:** Methodology, Writing - review & editing. **Weigeng Cui:** Project administration. **Xiaohong Zhao:** Funding acquisition. **Jun Tian:** Conceptualization. **Hui Kan:** Conceptualization.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2020.121667.

There is no conflict of interest exits in the submission of this manuscript, and manuscript is approved by all authors for publication. I would like to declare on behalf of my co-authors that the work described was original research that has not been published previously, and not under consideration for publication elsewhere, in whole or in part. All the authors listed have approved the manuscript that is enclosed.

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