



Emission Characteristics of Hazardous Atmospheric Pollutants from Ultra-low Emission Coal-fired Industrial Boilers in China

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ABSTRACT

This study comprehensively investigated the emission characteristics of primary air pollutants (PM, SO₂ and NO_x) and trace elements (As, Cd, Cr, Hg and Pb) from twelve coal-fired industrial boilers, nine of which were ultra-low emission (ULE) and three of which were non-ULE, based on field measurements. The concentrations, release ratios and relative enrichment factors (REFs) of the trace elements in both the coal and the bottom ash were obtained. Furthermore, the influence of atmospheric pollutant control devices (APCDs) on the emission concentrations and emission factors (EFs) of these pollutants and elements, as well as on their removal efficiencies, was analyzed. The average release ratios for Hg, Cr, Pb and As from the coal were 96.28%, 59.95%, 65.34% and 84.85%, respectively, whereas the average overall removal efficiencies for PM, SO₂, NO_x, Hg, Cr, Pb and As with the APCD configurations of the ULE coal-fired industrial boilers were 99.5%, 95.9%, 81.0%, 95.6%, 95.6%, 99.3% and 96.0%, respectively. Using selective non-catalytic reduction (SNCR), and a hybrid of selective non-catalytic reduction and selective catalytic reduction (SNCR-SCR) in the ULE boilers, the EFs of the NO_x were $5.5 \times 10^{-1} \text{ kg t}^{-1}$ and $4.9 \times 10^{-1} \text{ kg t}^{-1}$, respectively. Overall, the removal efficiencies for NO_x and Hg were 2.1 and 2.8 times higher, respectively, with the ULE than the non-ULE coal-fired industrial boilers, which was mainly attributable to the higher denitrification efficiency, higher dedusting efficiency and higher liquid/gas (L/G) desulfurization ratio of the ULE boilers.

Keywords: Ultra-low emission; Coal-fired industrial boilers; Emission characteristics; Trace elements; Relative enrichment factors.

INTRODUCTION

Coal-fired industrial boilers are the general power equipment in China and the important consumers of coal, consuming about 700 million tons of coal, which accounted for 13.5% of the total coal consumption in China in 2015 (NBS, 2016). The emission of PM, SO₂ and NO_x from coal-fired industrial boilers were 1.6 million tons, 7.2 million tons and 2.7 million tons in 2011, respectively, which is responsible for releasing large quantities of not only primary air pollutants but also trace elements into the atmosphere (MEP, 2011, 2013; Zhao *et al.*, 2010; Tian *et al.*, 2015, 2018). Therein, trace elements in coal were volatilized into metallic vapor or sub-micrometer particles at high temperature in the furnace room and emitted

to the environment by adsorption onto the fly ash or along with flue gas (Lin *et al.*, 2005).

China still has numerous coal-fired industrial boilers, and the combustion efficiency and atmospheric pollutant control devices (APCDs) are still lagging behind. Environmental statistics showed that only 19% and 1.4% of coal-fired industrial boilers were equipped with denitrification devices and flue gas desulfurization (FGD) devices by the end of 2015 in China (NEMC, 2016). At present, coal-fired industrial boilers in China are dominated by small-capacity boilers. Grate-fired (GF) boilers, circulating fluidized-bed (CFB) boilers and pulverized coal (PC) boilers are the major boiler types. By 2015, GF boilers (chain grate-fired boilers, stoker-fired boilers and other grate-fired boilers) and CFB boilers accounted for 69% and 21% of the capacity in coal-fired industrial boilers, respectively (NEMC, 2016), where CFB boilers had been rapidly developed since the 1960s in China as a clean boiler type for SO₂ and NO_x emission control (Duan *et al.*, 2012).

With the enhancement of the environmental management requirement, coal-fired power plants in China have already

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completed the ultra-low emission (ULE) retrofit. Some coal-fired industrial boilers have implemented the ULE retrofit by adopting the same APCDs applied in coal-fired power plants to execute the ultra-low emission standards, such as hybrid of selective non-catalytic reduction and selective catalytic reduction (SNCR-SCR) for denitrification, more efficient wet flue gas desulfurization (WFGD) for desulfurization, and wet electrostatic precipitator (WESP) after WFGD systems for removal of PM and secondary PM produced by wet desulfurization (Zhu *et al.*, 2014; Yao *et al.*, 2019). In August 2015, Shandong Province required the in-use coal-fired industrial boilers to be carried out ULE retrofit to meet the ULE limits of 10 mg m^{-3} , 50 mg m^{-3} , and 200 mg m^{-3} for PM, SO_2 and NO_x , respectively, under the O_2 content of 9% (SEPD, 2015). In November 2018, the Chinese government required that the emission concentration of atmospheric pollutants from newly built coal-fired boilers in the key air pollution control region must meet the ULE limits (MEE, 2018). These stringent environmental management requirements will have significant impacts on the emission characteristics of atmospheric pollutants from coal-fired industrial boilers by altering the boiler types and levels of pollutant control devices.

Several studies have conducted on emission level and removal efficiency of hazardous air pollutants from ULE and non-ULE coal-fired power plants (Zhang *et al.*, 2008; Zhao *et al.*, 2017; Zheng *et al.*, 2017). Wu *et al.* (2018, 2020) established an integrated emission factors (EFs) database of size-fractionated particulate matter (PM) for typical ULE technical routes installed in coal-fired power plant. Zheng *et al.* (2017) investigated partitioning of hazardous trace elements from ULE coal-fired power plants. Zhang *et al.* (2019) investigated the removal efficiency of SO_3 in ULE coal-fired power plants. Compared with coal-fired power plant, there are few studies on coal-fired industrial boiler. Previous studies are mainly focus on emission characteristics of primary air pollutants from non-ULE coal-fired industrial boiler. The Electrical Low-Pressure Impactor (ELPI) was applied to investigate the emission characteristics of size-fractionated PM of industrial boilers (Cornette *et al.*, 2020). Zhao *et al.* (2014) studied the $\text{PM}_{2.5}$ emission characteristics of a CFB boiler equipped with fabric filters (FFs). Li *et al.* (2018, 2019) obtained NO_x , PM, SO_2 , and VOCs EFs of coal-fired boilers from more than 91 enterprises related to coal washing, iron-steel production, lime and gypsum making, coking, and cement industries. Ruan *et al.* (2019) studied the PM emission characteristics from two ULE coal-fired industrial boilers in Xi'an, China, and the PM removal efficiency of the FFs for a chain-grate boiler and a CFB boiler were obtained. Yue *et al.* (2018) evaluated the environmental impacts of coal-fired industrial boilers in Beijing, based on a comprehensive emission inventory established in this study. Comprehensive researches on the removal efficiency and EFs of primary air pollutants and trace elements from coal-fired industrial boilers, especially for ULE boilers in China, are still quite limited.

With the stricter requirement for pollutants emission from coal-fired industrial boilers, the boiler types and APCDs of coal-fired industrial boilers in China have been changed

significantly. However, due to the limited field measurement data of primary hazardous atmospheric pollutants from coal-fired industrial boilers in the context of current control technology, the true emission characteristics of primary air pollutants and trace elements are unclear.

In this study, we presented comprehensive investigations of the emission characteristics of primary air pollutants (PM, SO_2 and NO_x) and trace elements (As, Cd, Cr, Hg and Pb) by the field measurements of nine ULE coal-fired boilers. The data obtained from this study is valuable for systematically understanding the emission characteristics of primary air pollutants and trace elements from coal-fired industrial boilers with ULE.

METHODOLOGY

Unit Description and Test Conditions

Nine typical coal-fired industrial boilers after ULE retrofit were selected for conducting field tests considering the boiler capacity, boiler types and APCDs. The basic information of the nine ULE coal-fired industrial boilers is shown in Table 1 as the #1–9 units. To compare the emission characteristics of primary air pollutants and trace elements of ULE and non-ULE coal-fired industrial boilers, three coal-fired industrial boilers without ULE retrofit were chosen in this study. The basic information of the three coal-fired industrial boilers without ULE retrofit is shown in Table 1 as the #10, #11 and #12 units.

GF boilers and CFB boilers are two kinds of industrial boilers most widely used in China. Therefore, the nine ULE coal-fired industrial boilers selected in this study include five GF boiler units and four CFB boiler units. The #5 unit was equipped with SNCR, while the #6 and #9 units were equipped with hybrid SNCR-SCR, which is usually equipped in coal-fired industrial boilers to meet the NO_x emission limit. Like the #1–4 units, the combination of low- NO_x burner technology (LNB) and SNCR is usually applied in CFB boilers, because of the calcium poisoning of de- NO_x catalysts on SCR and the space limit after furnace (Shang *et al.*, 2013; Wang *et al.*, 2017; Zheng *et al.*, 2017). Meanwhile, in order to adapt to the low-temperature flue gas emitted from coal-fired industrial boilers, the oxidation denitrification (OD) technology using NaClO_2 or O_3 as the oxidant and sorbent to remove NO_x , has been used in recent years (Chien and Chu, 2000), such as the #7 and #8 units in this study. In terms of dust removal technologies, the #2, #3 and #4 units used electrostatic fabric filters (ESP-FFs) + WESP and the other seven boilers used fabric filters (FFs) + WESP. FFs and electrostatic precipitator (ESP) are two of the most effective and widely used post-combustion dust removal technologies. It was found that WESP had a good removal efficiency (7.72–94.41%) for ultrafine particles (Ruan *et al.*, 2019), and it is usually installed as a final APCD to ensure that the emission of air pollutants meet the ULE standards. WFGD (limestone-gypsum, sodium alkali, magnesia) is also commonly used in coal-fired industrial boilers. During the tests, the boilers and APCDs were all operated under normal conditions.

Sampling Methods

Sampling and testing were strictly in accordance with the relevant standards and technical criterion for the stationary sources (MEP, 1996, 2017). The APCDs' configuration and sampling points are presented in Table 1. The management of coal-fired industrial boilers is generally not as good as coal-fired power plants. The sampling port diameter and platform space of coal-fired industrial boilers cannot always meet the test requirements, so the real field condition is not always available for sampling at the inlet and outlet of all the APCDs. In this study, there were no sampling points for the denitrification equipment.

The sampling points are shown in Fig. 1. PM was sampled by the PM sampler (3012H-D; Qingdao LaoYing Environmental Science and Technology, Co., Ltd.) equipped with the low-concentration membrane sampling tube. Gaseous pollutants (NO_x and SO₂), O₂ content, flue gas velocity and the temperature were tested by the gas analyzer (testo 350; Testo). Trace elements (Hg, As, Pb, Cd and Cr) in the flue gas were sampled by the sampler (C-5000 series; ESC) following EPA Method 29 and analyzed by atomic fluorescence spectrophotometry (AFS) and inductively coupled plasma mass spectrometry (ICP-MS). The coal and bottom ash were collected as solid samples for all plants and the contents of trace elements (Hg, As, Pb, Cd and Cr) in the coal and bottom ash were also analyzed by AFS and ICP-MS. The detection limits of Hg, As, Pb, Cd and Cr were 0.002 mg kg⁻¹, 0.5 mg kg⁻¹, 2.1 mg kg⁻¹, 0.6 mg kg⁻¹ and 1.0 mg kg⁻¹, respectively. Pollutant concentrations were converted to the values under the O₂ content of 9%, as required by relevant criterion (MEP, 2014).

Analytical Methods

Trace Elements

The mass balance method is calculated by normalizing the concentration of each trace element for the entire process. It is widely used to investigate the EFs and removal efficiency for trace elements (Zhang et al., 2016; Zheng et al., 2017; Wu et al., 2018). In this study, based on the inlet and outlet concentration of trace elements for APCDs, the EFs and removal efficiency of trace elements were calculated by Eqs. (1) and (2):

$$EF_{te} = C_{te_fluegas} \times \frac{(21 - \varphi(O_2))}{1000 \times (21 - \varphi'(O_2))} \times V \quad (1)$$

$$\eta_{te} = 1 - \left(\frac{EF_{te}}{R_{EC} \times C_{te_coal}} \right) \quad (2)$$

where EF_{te} is the emission factor for trace elements, g t⁻¹; $C_{te_fluegas}$ is the concentration of trace elements in the flue gas, μg m⁻³; C_{te_coal} is the concentration of trace elements in the feed coal, mg kg⁻¹; V is the volume of the flue gas per unit of fuel consumption, m³ kg⁻¹; R_{EC} is the release ratio for trace elements, %; $\varphi(O_2)$ is the reference oxygen content where 9% was applied according to GB13271-2014; and $\varphi'(O_2)$ is the oxygen content measured by the gas analyzer.

Table 1. Basic information of the 12 boilers in the field tests.

Unit#	Boiler Type	Capacity	Denitrification	Dedusting	Desulfurization	Sampling positions			EPA Method 29
						Before Dedusting	Before Desulfurization	Outlet	
1	CFB	75 t h ⁻¹	LNB + SNCR	FFs + WESP	limestone-gypsum	✓	✓	✓	✓
2	CFB	75 t h ⁻¹	LNB + SNCR	ESP-FFs + WESP	limestone-gypsum	✓*	✓*	✓	✓
3	CFB	75 t h ⁻¹	LNB + SNCR	ESP-FFs + WESP	natrimum-alkali	✓	✓	✓	✓
4	CFB	75 t h ⁻¹	LNB + SNCR	FFs + WESP	limestone-gypsum	✓	✓	✓	✓
5	GF	35 t h ⁻¹	SNCR	FFs + WESP	natrimum-alkali	✓	✓	✓	✓
6	GF	35 t h ⁻¹	SNCR-SCR	ESP-FFs + WESP	limestone-gypsum	✓	✓	✓	✓
7	GF	83 t h ⁻¹	OD	FFs + WESP	limestone-gypsum	✓	✓	✓	✓
8	GF	41 t h ⁻¹	OD	FFs + WESP	natrimum-alkali	✓	✓	✓	✓
9	GF	15 t h ⁻¹	SNCR-SCR	FFs + WESP	natrimum-alkali	✓	✓	✓	✓
10	CFB	35 t h ⁻¹	SNCR	ESP-FFs	natrimum-alkali	✓	✓	✓	✓
11	GF	10 t h ⁻¹	SNCR	FFs	natrimum-alkali	✓	✓	✓	✓
12	CFB	40 t h ⁻¹	LNB + SNCR	FFs	natrimum-alkali	✓	✓	✓	✓

* #2 and #3 share the same chimney.

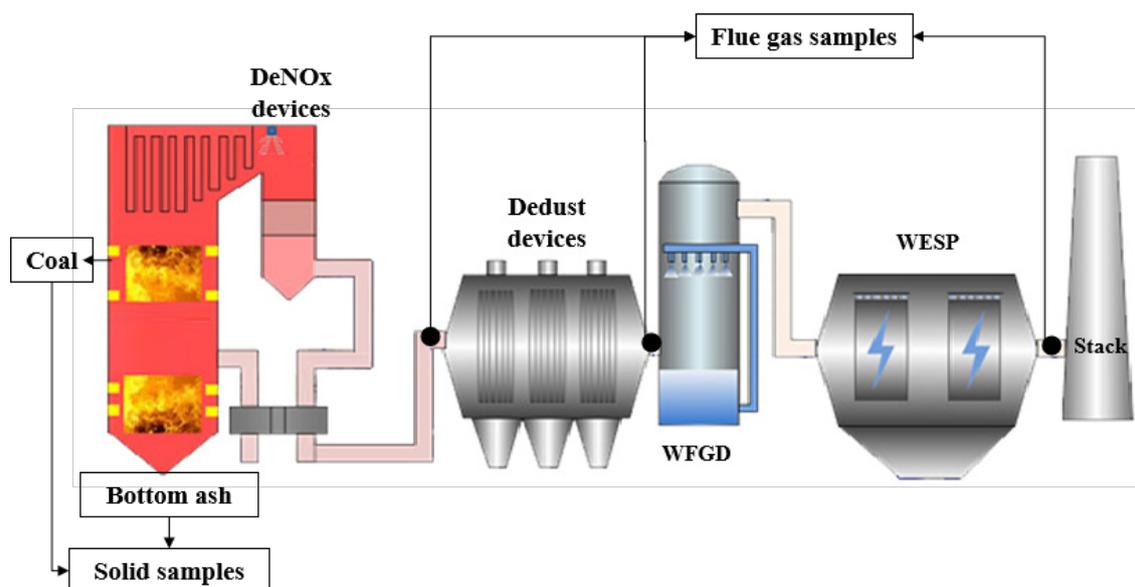


Fig. 1. APCDs' configurations and sampling points.

Because of the volatility of different trace elements at high temperature in the furnace, the release characteristics of trace elements are different. The term “release ratio” represents the proportion of trace elements in the feed coal released to the flue gas and fly ash, which can be calculated by Eq. (3):

$$R_{EC} = 1 - \frac{C_{bottom\ ash} \times M_{bottom\ ash}}{C_{coal}} \quad (3)$$

where R_{EC} represents the release ratio of trace elements from the coal to the flue gas and fly ash, %; $C_{bottom\ ash}$ represents the concentration of trace elements in the bottom ash, mg kg^{-1} ; C_{coal} represents the concentration of trace elements in the feed coal, mg kg^{-1} ; and $M_{bottom\ ash}$ represents the mass of bottom ash produced by coal combustion, kg kg^{-1} , which can be estimated by the ash content in the feed coal by Eq. (4):

$$M_{bottom\ ash} = k_{boiler\ type} \times A_{coal} \quad (4)$$

where A_{coal} represents the ash content in the coal, %, and $k_{boiler\ type}$ represents the production factor of the bottom ash from different boiler type. For CFB boilers, $k_{boiler\ type} = 5.25$, and for GF boilers, $k_{boiler\ type} = 9.24$ (CNPSS, 2011).

In addition, in order to investigate the partitioning behavior of trace elements from the feed coal into the bottom ash, the term “relative enrichment factors” was introduced to describe the behavior of trace elements remaining in the bottom ash (Meij, 1994), as shown in Eq. (5). Generally, REFs presented the partitioning of trace elements after coal combustion and a high REF means a strong enrichment capacity for the trace element in the ash (Bhattacharyya et al., 2009; Zheng et al., 2017):

$$REFs = \frac{C_{ash}}{C_{coal}} \times \frac{A_{coal}}{100} \quad (5)$$

where C_{ash} represents the concentration of trace elements in the bottom ash, mg kg^{-1} ; C_{coal} represents the concentration of trace elements in the feed coal, mg kg^{-1} ; and A_{coal} represents the ash content in the feed coal, %.

Primary Pollutants

The EFs of PM, SO_2 and NO_x from coal-fired industrial boilers can be calculated by Eq. (6) (Zhao et al., 2010; CNPSS, 2011):

$$EF_p = C_p \times \frac{(21 - \varphi(\text{O}_2))}{1000 \times (21 - \varphi'(\text{O}_2))} \times V \quad (6)$$

where EF_p is the emission factor of pollutant p , kg t^{-1} ; C_p is the emission concentration of pollutant p in the flue gas, mg Nm^{-3} ; and V is the volume of the flue gas per unit of fuel consumption, $\text{m}^3 \text{kg}^{-1}$.

The flue gas volume is mainly affected by the lower heating value of coals, and bituminous is the main coal type of coal-fired industrial boilers in China. V in Eq. (6) and Eq. (4) can be calculated by Eq. (7) (Zhao et al., 2010):

$$V = 1.04 \times \frac{Q_L}{4187} + 0.77 + 1.0161 \times (\alpha - 1) \times \left(0.251 \times \frac{Q_L}{1000} + 0.278 \right) \quad (7)$$

where Q_L is the lower heating value, kJ kg^{-1} , and α is the excess air coefficient where 1.75 was applied according to GB13271-2014. Meanwhile, the removal efficiency for APCDs is calculated by Eq. (8):

$$\eta = 1 - (C_{outlet}/C_{inlet}) \quad (8)$$

where C_{outlet} and C_{inlet} are the pollutant concentrations in the

flue gas from the outlet and inlet of each APCD, respectively, mg Nm^{-3} .

RESULTS

Enrichment Characteristics of Trace Elements in the Feed Coal and Bottom Ash

The REFs of trace elements in the bottom ash are shown in Tables S1 and S2. Because the concentrations of Cd in the feed coal and bottom ash were below the detection limit, Cd was not discussed in this study.

Fig. 2 shows the distribution of trace elements content in the feed coal and bottom ash. The average content of Cr and Pb were 6.18 mg kg^{-1} and 7.43 mg kg^{-1} in the feed coal, respectively, which were 71.8–74.6% lower than those in bottom ash. The average content of As in feed coal was 6.92 mg kg^{-1} , which was 1.5 times higher than that in bottom ash. While the average content of Hg in feed coal was 0.048 mg kg^{-1} in feed coal, higher than 0.007 mg kg^{-1} in bottom ash. This can be explained by the REFs as follows.

Meij and Winkel (2007) classified the trace elements into three classes based on their behavior during the combustion in the boiler with their REFs. In this study, the REFs of Cr, Pb and As were less than 0.7, classified as Class II. Cr, Pb and As were volatile in the boiler, but they were completely condensed in ESP on the ash particles. Cr, Pb and As originally existed in the vapor phase and had no chance to be condensed on the bottom ash. The REFs of Hg was very small ($\ll 1$) in the bottom ash, and Hg was classified as Class III, which manifested that Hg tended to remain in the flue gas, resulting in the emission of high concentrations of Hg into the atmosphere.

EFs and Removal Efficiency of Trace Elements

The boiling points of Hg, Cd, Cr, Pb and As are 356°C , 767°C , 2672°C , 1749°C and 613°C , respectively. Researchers have found that the combustion temperature in the furnace and the composition of the flue gas have the effect on the

proportion of trace elements in the bottom ash (Hiraoka *et al.*, 1980; Gerstle and Albrinck, 1982; Chang *et al.*, 2000; Zheng *et al.*, 2017). Since trace elements in some feed coal and bottom ash samples were under the detected limits, the release ratio of available data has been calculated, as shown in Table S3.

Due to the optimum desulfurization temperature (generally $850\text{--}900^\circ\text{C}$) and strong turbulent motion in CFB boilers, the emission characteristics of trace elements for CFB and GF boilers are different (Xu *et al.*, 2004). The average release ratio of Hg, Cr, Pb and As in CFB boilers were 97.87%, 57.94%, 52.41% and 86.64%, respectively, while the average release ratio of Hg, Cr, Pb and As in GF boilers were 95.14%, 62.96%, 86.90% and 81.27%, respectively, as shown in Fig. 3. The average release ratio of Hg and As in CFB boilers were higher than that in GF boilers, and the range of release ratio for Cr and Pb in CFB boilers was larger than that in GF boilers.

APCDs have co-benefits to trace elements, as most of the trace elements were enriched in the fly ash (Zhang *et al.*, 2017; Zheng *et al.*, 2017). To obtain the EFs and removal efficiency of trace elements, the concentrations of trace elements in the feed coal, the bottom ash and the flue gas were analyzed. Fig. 4 shows the distribution of trace elements' concentrations for Hg, Cr, Pb and As in the flue gas at outlet for six ULE coal-fired industrial boilers (#1–6), which were $0.02\text{--}0.42 \mu\text{g m}^{-3}$, $5.92\text{--}20.01 \mu\text{g m}^{-3}$, $0.91\text{--}6.61 \mu\text{g m}^{-3}$ and $0.63\text{--}11.39 \mu\text{g m}^{-3}$, respectively. Besides, the Cd concentration in #8 was $0.02 \mu\text{g m}^{-3}$.

Table 2 shows the EFs and removal efficiency of Hg, Cr, Pb and As. The average EFs of Hg, Cr, Pb and As for ULE coal-fired industrial boilers were 0.0007 g t^{-1} , 0.135 g t^{-1} , 0.038 g t^{-1} and 0.055 g t^{-1} , respectively, while the average EFs of Hg, Cr, Pb and As for non-ULE coal-fired industrial boilers were 0.006 g t^{-1} , 0.09 g t^{-1} , 0.045 g t^{-1} and 0.015 g t^{-1} , respectively. EF_{Hg} of the ULE coal-fired industrial boilers was approximately 83% lower than that of the non-ULE coal-fired industrial boilers. The overall removal efficiency of Hg, Cr, Pb and As for ULE boilers were 90.8–99.2%,

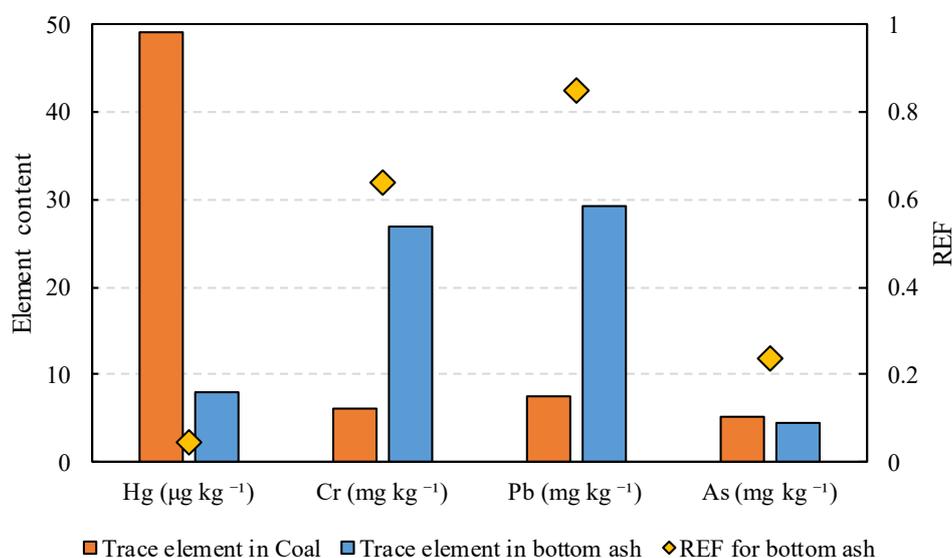


Fig. 2. Average trace element content in the feed coal and bottom ash.

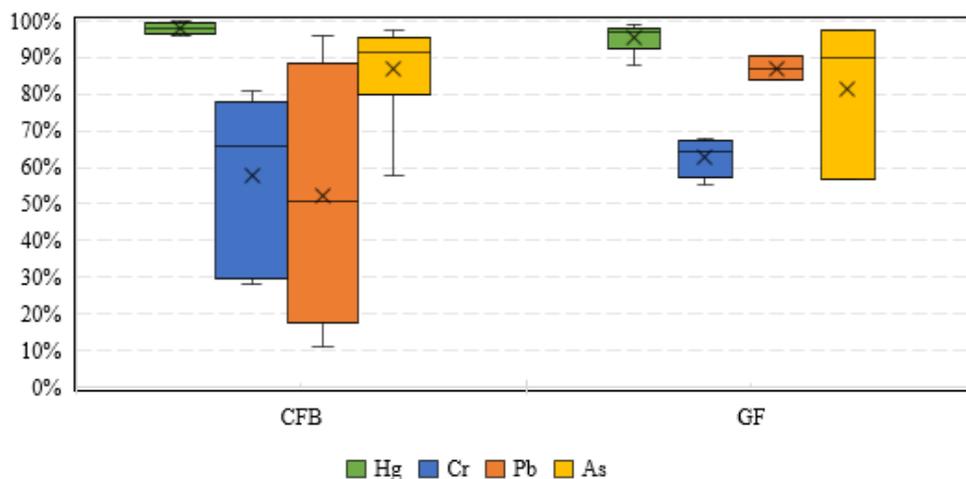


Fig. 3. Heavy metal release ratio for GF and CFB boilers.

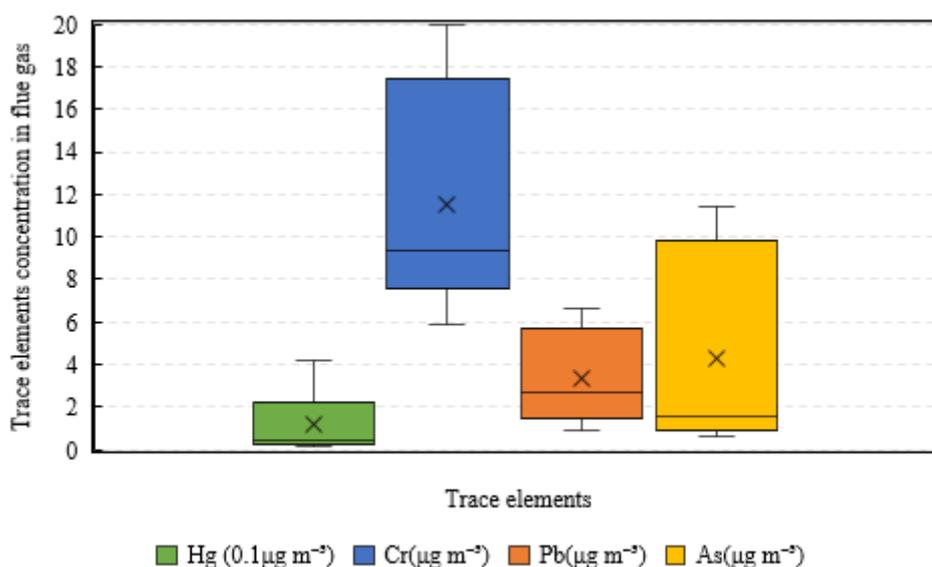


Fig. 4. Trace elements' concentration in the flue gas at the outlet of ULE coal-fired industrial boilers.

96.7–99.0%, 96.8–99.9% and 93.7–99.9%, respectively, and those for non-ULE coal-fired industrial boilers were 20.7–64.3%, 96.4–99.2%, 93.6–99.5% and 99.5–99.8%, respectively. The average overall Hg removal efficiency for the ULE coal-fired industrial boilers was 2.8 times higher than the non-ULE coal-fired industrial boilers. Hg is divided into three chemical forms, including gaseous elemental mercury (Hg^0), particle-bound mercury (Hg_p) and reactive gaseous mercury (Hg^{II}), accounting for average 56%, 10% and 34% released from coal combustion sources, respectively (Zhang *et al.*, 2015). With the decreasing temperature of flue gas, hazardous trace elements tend to be adsorbed on particulate matter and subsequently captured by dust removal devices (Zhao *et al.*, 2017; Zheng *et al.*, 2017). Meanwhile, WFGD is the crucial step in the co-benefit mercury control technologies by removing Hg^{II} with average 45% overall mercury removal efficiency (Zhang *et al.*, 2017). ULE coal-fired industrial boilers were equipped with WESP and high-efficiency WFGD, with high dust removal efficiency and

high liquid/gas (L/G) ratio of desulfurization, which enhanced the removal efficiency of Hg (Zheng *et al.*, 2017).

EFs and Removal Efficiency of Primary Air Pollutants NO_x

The requirements of NO_x emission for ULE coal-fired industrial boilers are different in different regions. Under the O_2 content of 9%, the emission limit of NO_x for ULE coal-fired boilers is 200 mg m^{-3} in general and 100 mg m^{-3} in key region in Shandong Province, while in Beijing-Tianjin-Hebei region the requirement is 50 mg m^{-3} under the O_2 content of 6%. Because of the regional differences of the NO_x emission limit for ULE coal-fired industrial boilers, some denitrification facilities with low efficiency are still used.

The denitrification technologies applied in coal-fired industrial boilers are mainly from the coal-fired power plants due to the similar combustion features, namely SNCR and SCR. Compare with coal-fired power plant, coal-fired industrial boilers were relatively small in capacity. And due

Table 2. Overall EFs and removal efficiency of trace elements.

Boiler Type	Pollutant Control Devices	Unit	Emission Factor (g t ⁻¹)				Removal Efficiency (%)			
			Hg	Cr	Pb	As	Hg	Cr	Pb	As
CFB	LNB + SNCR + FFs + limestone-gypsum FGD + WESP	#1, #4	0.0005	0.08	0.02	0.04	97.9	99.0	99.7	99.4
CFB	LNB + SNCR + ESP-FFs + limestone-gypsum FGD + WESP	#2, #3	0.0004	0.11	0.01	0.02	99.2	97.0	99.9	93.7
GF	SNCR + FFs + sodium-alkali FGD + WESP	#5	0.0003	0.13	0.09	0.15	90.8	97.5	96.8	99.8
GF	SNCR-SCR + limestone-gypsum FGD + WESP	#6	0.0017	0.22	0.03	0.01	94.0	96.7	99.6	99.9
CFB	SNCR + ESP-FFs + sodium-alkali FGD	#10	0.0013	0.06	0.02	0.01	64.3	99.2	99.5	99.8
GF	SNCR + FFs + sodium-alkali FGD	#11	0.0099	0.12	0.07	0.02	20.7	96.4	93.6	99.5

to the limit of initial investment costs, installation space, flue gas temperature and furnace structure, SNCR were widely used in denitrification for coal-fired industrial boilers (Wang, 2018). Meanwhile, to adapt to the low-temperature flue gas emitted from the industrial boilers, oxidation denitrification (OD) with NaClO₂ or O₃ as the oxidant and sorbent to remove NO_x has been used in recent years (Chien *et al.*, 2000; Hu *et al.*, 2018).

To meet the ultra-low emission limit for NO_x, hybrid SNCR-SCR is generally applied in GF boilers to improve the denitrification efficiency, which can pre-reduce the NO_x emission by SNCR and lighten the burden of SCR (Ruan *et al.*, 2019). Besides, the combination of low-NO_x burner technology (LNB) and SNCR is usually applied in CFB boilers, because of the calcium poisoning of de-NO_x catalysts on SCR and the space limit after furnace (Shang *et al.*, 2013; Wang *et al.*, 2017; Zheng *et al.*, 2017; Ruan *et al.*, 2019).

The NO_x emission concentration for different denitrification technologies obtained in this study are shown in Fig. 5. The average NO_x emission concentrations for LNB + SNCR, SNCR, SNCR-SCR and OD were 37 mg m⁻³, 158 mg m⁻³, 45 mg m⁻³ and 111 mg m⁻³, respectively. The average NO_x emission concentration of LNB + SNCR and SNCR-SCR were significantly lower than that of SNCR and OD. The EFs of NO_x for different denitrification technologies are shown in Table 3. The NO_x EFs of LNB + SNCR, SNCR, SNCR-SCR and OD were 0.44 kg t⁻¹, 1.92 kg t⁻¹, 0.49 kg t⁻¹ and 1.36 kg t⁻¹, respectively. The efficient working temperature, division and layout of the spray gun, and reducing agent concentrations were the main operating parameters affecting removal efficiency of SNCR (Wang *et al.*, 2017). The pH, solution temperature and degree of oxidation of NO have significant influence on the denitrification of OD (Xiao *et al.*, 2011). With ULE retrofit, the operating parameters of SNCR were optimized, and LNB or SCR were coupled to removal NO_x. Therefore, due to unstable coal quality, frequently changed operating loads, unoptimized operating parameters and individual NO_x removal process, the NO_x EFs of SNCR and OD were higher than LNB + SNCR and SNCR-SCR. Since there were no sampling ports at the inlet of the denitrification devices, the NO_x removal efficiency cannot be obtained by the field tests. In this study, NO_x removal efficiency was estimated by the unabated EFs for different types of boilers in Table S4 (CNPSS, 2011). The average NO_x removal efficiency of LNB + SNCR, SNCR, SNCR-SCR and OD were 82%, 30%, 83% and 54% respectively, which are consistent with existing researches (Xiao *et al.*, 2011; Wang *et al.*, 2017; Wang, 2018). The average NO_x removal efficiency of LNB + SNCR and SNCR-SCR was 2.7 times higher than η_{NO_x} of SNCR and 1.5 times higher than η_{NO_x} of OD. Therefore, to meet the stricter requirement for NO_x emission, such as 50 mg m⁻³ under O₂ content of 6% in Beijing-Tianjin-Hebei region, it is recommended to apply LNB + SNCR for CFB boilers and SNCR-SCR for GF boilers.

SO₂

WFGD (including limestone-gypsum and sodium alkali) is commonly used in coal-fired industrial boilers to reduce

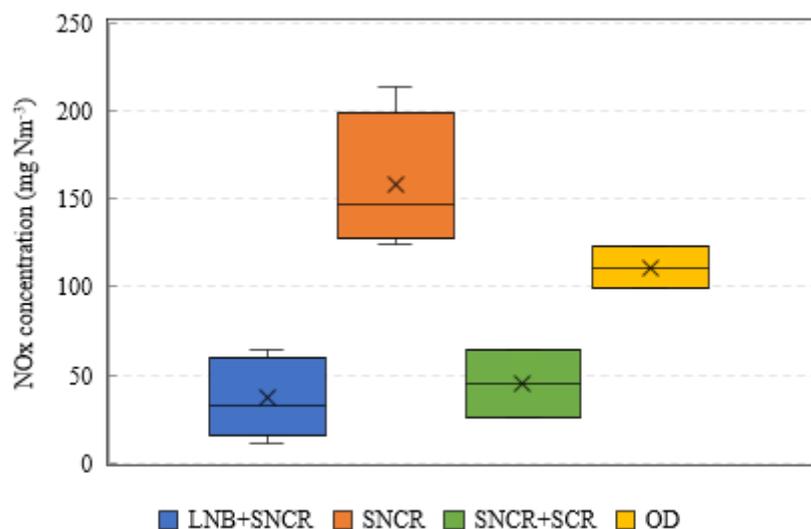


Fig. 5. NO_x emission concentration for different NO_x control devices.

Table 3. NO_x emission factor of different NO_x control devices (kg t⁻¹).

	LNB + SNCR	SNCR	SNCR-SCR	OD
AM	0.44	1.92	0.49	1.36
SD	0.28	0.52	0.20	0.11
NM	5	4	2	2
MX	0.77	2.76	0.69	1.46
MI	0.08	1.4	0.29	1.25

Note: AM—arithmetic mean; ST—standard deviation; NM—number of samples; MX—maximal; MI—minimum.

SO₂. Liquid gas ratio, pH value, imported sulfur dioxide concentration and the oxidation of sulfite were main factors for desulfurization efficiency in WFGD system (Chen and Xu, 2005; Zhong *et al.*, 2008). To meet the ultra-low emission limit for SO₂, the WFGD can be upgraded by improving the liquid/gas ratio, increasing the spray layer and adding trays in the absorption tower (Lu, 2014; Zhao *et al.*, 2015).

In this study, the inlet and outlet concentration of SO₂ and the removal efficiency of WFGD for five ULE coal-fired industrial boilers were obtained. As shown in Table 4, the average SO₂ removal efficiency for ULE coal-fired industrial boilers was 96%, which is significantly higher than it for non-ULE coal-fired industrial boilers reported by CNPSS, with the value of 60–80% (2011). Meanwhile, due to calcium carbonate precipitation by absorbing carbon dioxide from flue gas, sodium alkali WFGD required stricter operating parameters than limestone-gypsum (Mo *et al.*, 2007). In Table 4, the desulfurization efficiency of limestone-gypsum WFGD and sodium alkali WFGD were 97% and 94% respectively, which was probably caused by operating conditions, and more field test were needed to identify the variance of desulfurization efficiency.

Table 5 shows the EFs of SO₂ for WFGDs based on the field measurements in this study. The average SO₂ EFs for the limestone-gypsum WFGD and the sodium alkali WFGD of ULE coal-fired industrial boilers and the sodium alkali WFGD of non-ULE coal-fired industrial boilers were 0.22 kg t⁻¹, 0.27 kg t⁻¹ and 0.40 kg t⁻¹, respectively. In terms of the sodium alkali WFGD, the SO₂ EFs of ULE coal-fired

industrial boilers reduced by 33% compared with non-ULE coal-fired industrial boilers. In order to improve desulfurization efficiency, dual-pH value control technology and composite desulfurization tower technology were used in ULE retrofit, leading to higher mass-transfer efficiency and resistance time for sorbent (Guo *et al.*, 2019). Therefore, the desulfurization efficiency of ULE coal-fired industrial boilers improved compared to the three coal-fired industrial boilers with sodium alkali WFGD but without the ULE retrofit.

PM

FFs, due to their high dust removal efficiency and regardless of coal types, have been widely used in coal-fired industrial boilers. Although WFGD can not only effectively reduce SO₂ but also remove coarse particles, the removal efficiency of the PM with size 0.2–2 μm was relatively low, and even can increase the mass concentration of PM with size 0.094–0.946 μm (Liu *et al.*, 2016; Ruan *et al.*, 2019). Meanwhile, the particle size distributions of GF and CFB were different (Zhao *et al.*, 2017). The mass size distributions (MSDs) of the PM_{2.5} emitted from GF presented bimodal distribution of the peak existed on 0.12–0.20 μm and > 1 μm (Li *et al.*, 2009; Wang *et al.*, 2009). Due to different combustion method and combustion temperature, the MSDs of the PM₁₀ emitted from CFB showed no peak (Zhao *et al.*, 2014). To meet the PM ultra-low emission limit and remove the PM produced by WFGD, WESP is installed downstream of the WFGD to capture the ultrafine particulates and other pollutants (Zheng *et al.*, 2017).

Table 4. SO₂ removal efficiency of different desulfurization technologies in ULE coal-fired industrial boilers.

WFGD type	C _{inlet} (mg Nm ⁻³)	C _{outlet} (mg Nm ⁻³)	η _{SO₂} (%)
Limestone-gypsum	226	10	95.57
	778	14	98.24
	944	16	98.31
natrium-alkali	244	14	94.26
	815	44	94.60

Table 5. SO₂ EFs of different desulfurization technologies in ULE and non-ULE coal-fired industrial boilers (kg t⁻¹).

	Limestone-gypsum WFGD (ULE)	natrium-alkali WFGD (ULE)	natrium-alkali WFGD (non-ULE)
AM	0.22	0.27	0.40
ST	0.15	0.18	0.05
NM	5	4	3
MX	0.52	0.57	0.45
MI	0.10	0.14	0.33

Note: AM—arithmetic mean; ST—standard deviation; NM—number of samples; MX—maximal; MI—minimum.

Table 6. PM removal efficiency at different process of APCDs.

Unit	Dedusting (mg Nm ⁻³)				Desulfurization + WESP (mg Nm ⁻³)				APCD η
	Type	C _{inlet}	C _{out}	η	Type	C _{inlet}	C _{out}	η	
#1	FFs	26,732	–	–	limestone/lime-gypsum FGD + WESP	–	8	–	99.97%
#2	EFs	11,952	36	99.7%	natrium alkali FGD + WESP	36	8	77.7%	99.93%
#4	FFs	11,711	43	99.6%	limestone/lime-gypsum FGD + WESP	43	5	87.3%	99.95%
#5	FFs	942	–	–	natrium alkali FGD + WESP	–	8	–	99.15%

PM removal efficiency of four ULE coal-fired industrial boilers was measured in this study, as shown in Table 6. The #1, #2 and #4 units were CFB boilers with installed capacities of 75 t h⁻¹, and #5 unit was GF boiler with installed capacity of 20 t h⁻¹. The mass concentration of PM emitted from CFB were higher than GF, which was mainly produced by fragmentation mechanism and incomplete combustion respectively (Ruan *et al.*, 2019). The average removal efficiency for four ULE coal-fired industrial boilers were 99.75%. And the average removal efficiency for CFB boilers were 99.95% higher than GF boiler (#5 unit) notably (99.15%). It probably due to the different MSDs and mass concentration for PM emitted from boiler and unstable operating condition for small-capacity boilers (Zhao *et al.*, 2017; Ruan *et al.*, 2019). Meanwhile, the outlet of WFGD had no measuring point; we obtained the average hybrid PM removal efficiency of 82.52% for WFGD and WESP in #2 and #3. Ruan *et al.* (2019) found the PM removal efficiency of CFB and GF with ULE retrofit were in the range of 98.12–99.56% and 90.0–93.6% respectively, which were consistent with this study. Fig. S1 shows the PM removal efficiency and EFs for each ULE coal-fired industrial boilers in this study, and the average PM concentration and EFs were 7.2 mg m⁻³ and 0.08 kg t⁻¹, respectively.

CONCLUSION

This study evaluated nine ULE coal-fired industrial boilers, specifically, the emission and enrichment characteristics of primary air pollutants (PM, SO₂ and NO_x) and trace elements (As, Cd, Cr, Hg and Pb), and the removal efficiency of the

APCDs, based on field measurements.

The emission intensities of the ULE boilers for primary pollutants was significantly lower than those of non-ULE coal-fired industrial boilers. The APCDs of the ULE boilers exhibited average overall removal efficiencies of 99.5%, 95.9% and 81.0% for PM, SO₂ and NO_x, respectively, and the dedusting performance of these boilers was also enhanced by a WFGD + WESP process, which displayed a dedusting efficiency of 85%. Compared with non-ULE boilers, the ULE boilers showed a 37% higher desulfurization efficiency due to a higher mass-transfer efficiency, longer resistance by the sorbent and optimized operating parameters and a 96% higher denitrification efficiency due to the coupling of multiple denitrification techniques. Additionally, NO_x EFs of 0.44 kg t⁻¹ and 0.49 kg t⁻¹ were obtained when LNB + SNCR and SNCR-SCR were utilized in the ULE boilers, respectively.

The APCDs used to retrofit the ULE boilers demonstrated higher co-benefit removal efficiencies than those in the non-ULE boilers for the trace elements, with average overall removal efficiencies of 95.6%, 95.6%, 99.3% and 96.0% for Hg, Cr, Pb and As, respectively. In particular, the removal efficiency for mercury increased by 180% due to the speciation of this element and the greater efficiency of the APCDs.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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