



## Evolution of four-decade atmospheric mercury release from a coal-fired power plant in North China

Zhonggen Li<sup>a,\*</sup>, Xufeng Chen<sup>b</sup>, Wenli Liu<sup>b</sup>, Taishan Li<sup>b</sup>, Ji Chen<sup>a</sup>, Che-Jen Lin<sup>c,d,a</sup>, Guangyi Sun<sup>a,e</sup>, Xinbin Feng<sup>a,\*\*</sup>

<sup>a</sup> State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, 550081, China

<sup>b</sup> Tangshan Environmental Monitoring Center, Tangshan, 063000, China

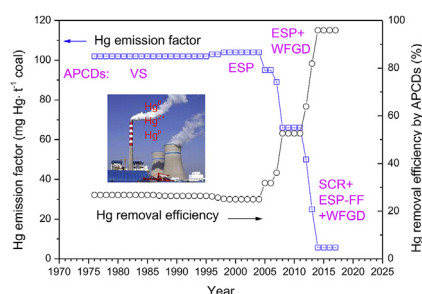
<sup>c</sup> Center for Advances in Water and Air Quality, Lamar University, Beaumont, TX, USA

<sup>d</sup> Department of Civil and Environmental Engineering, Lamar University, Beaumont, TX, USA

<sup>e</sup> University of Chinese Academy of Sciences, Beijing, 100049, China



### GRAPHICAL ABSTRACT



### ARTICLE INFO

#### Keywords:

Mercury  
Coal-fired power plants  
APCDs  
Co-benefit removal  
Emission history

### ABSTRACT

Coal-fired power plants (CFPPs) are an important anthropogenic emission source of atmospheric mercury (Hg). Understanding the co-benefits of conventional air pollution control devices (APCDs) on Hg reduction is crucial for reducing Hg emission from this source. In this study, atmospheric Hg emission from a pulverized coal (PC) utility boiler in North China was investigated along the emission control stage of selective catalytic reduction (SCR), electrostatic precipitator (ESP) and fabric filter (FF) and lime stone-gypsum wet flue gas desulfurization (WFGD). Historical atmospheric Hg emission from this CFPP with total 1550 MW capacity since its operation in 1976 was estimated based on the implementation of different configurations of APCDs. The results showed that a combination of SCR + ESP-FF + WFGD leads to a co-beneficial Hg control of 96%, with the in-stack Hg concentration ( $0.87 \mu\text{g m}^{-3}$ ) much lower than the Chinese national standard for CFPPs ( $30 \mu\text{g m}^{-3}$ ), and the stack Hg speciation ( $\text{Hg}^0:\text{Hg}^{2+}:\text{Hg}^{\text{p}}$ ) was in 86%:13%:1%. Mass balance analysis indicates that Hg from the input of feed coal and limestone was mainly ended up in fly ash (89%), followed by gypsum (5%), stack gas (5%) and bottom ash (1%), respectively. During the past 42 years of operation (1976–2017), the total air Hg emission from this CFPP summed to 17.62 tonnes, in which  $\text{Hg}^0$  make up 14.02 tonnes (79.5%),  $\text{Hg}^{2+}$  make up 3.15 tonnes (17.9%) and  $\text{Hg}^{\text{p}}$  0.45 tonne (2.6%). Although the speciation of emitted Hg into the atmosphere changed with respect to the configuration of APCDs,  $\text{Hg}^0$  was the most dominated emission species (64–98%) for all time. Nearly 90% of total Hg emissions occurred in the first three decades when venturi scrubber (VS) and ESP were the primary APCDs. The installation of WFGD and SCR substantially decreased the Hg emission factors by 17

\* Corresponding author. No. 99 Lincheng Road West, Guanshanhu District, Guiyang, 550081, PR China.

\*\* Corresponding author. No. 99 Lincheng Road West, Guanshanhu District, Guiyang, 550081, PR China.

E-mail addresses: [lizhonggen@mail.gyig.ac.cn](mailto:lizhonggen@mail.gyig.ac.cn) (Z. Li), [Fengxinbin@mail.gyig.ac.cn](mailto:Fengxinbin@mail.gyig.ac.cn) (X. Feng).

<https://doi.org/10.1016/j.atmosenv.2019.06.045>

Received 19 November 2018; Received in revised form 18 June 2019; Accepted 22 June 2019

Available online 24 June 2019

1352-2310/© 2019 Elsevier Ltd. All rights reserved.

times compared to the peak values during 1998–2004. This study demonstrates the combination of SCR + ESP-FF + WFGD as a best available technology (BAT) for Hg control in CFPPs in China.

## 1. Introduction

Mercury (Hg) is a toxic and persistent pollutant that cycles globally (Driscoll et al., 2013). Hg pollution is mainly spread through atmospheric transport due to the long atmospheric lifetime and continuous emission-deposition cycle of gaseous elemental mercury ( $\text{Hg}^0$ ) (Schroeder and Munthe, 1998; Lin and Pehkonen, 1999; Gustin et al., 2008). Both natural and anthropogenic sources emit Hg into the atmosphere (Pirrone et al., 2010). Human activities has dramatically disturbed the global Hg pool since the industrial revolution (Mason et al., 1994; Streets et al., 2017) and coal combustion has become a predominant anthropogenic emission source of Hg globally since the second half of twenty century (Streets et al., 2011). Hg exists at a trace level (ca. 0.10–0.20 mg kg<sup>-1</sup>) in coal (Yudovich and Ketris, 2005; Zhang et al., 2012) and the extensive coal use has resulted in large Hg emissions. Globally, coal combustion has released 38 Gg (10<sup>9</sup> g) of Hg to air, land and water since pre-1850 to 2010, and more than 97% of the atmospheric release occurred after 1850 (Streets et al., 2018). The high temperature (850–1500 °C) during coal combustion liberates Hg in coal into flue gases in the forms of  $\text{Hg}^0$ , gaseous oxide mercury ( $\text{Hg}^{2+}$ ) and particulate bound mercury ( $\text{Hg}^p$ ) (Galbreath and Zygarlicke, 2000).  $\text{Hg}^{2+}$  and  $\text{Hg}^p$  are easily deposited nearby the emission sources and impacted the local environment, while  $\text{Hg}^0$  could be a global concern (Schroeder and Munthe, 1998). Recently, the legally-binding Minamata Convention on Mercury enters into force in 2017 to curb global Hg emissions from anthropogenic sources and intentional Hg uses, where coal-fired power plants (CFPPs) are listed as the top priority for Hg control among other emission sources including smelting and roasting processes, waste incineration facilities and cement clinker production facilities.

As the largest coal consumer in the world, China annually consumed about a half of the world's coal production (Dai and Finkelman, 2018) and emitted 202–250 tonnes of Hg into atmosphere each year from this source during 1999–2010 (Streets et al., 2005; Zhang et al., 2015), of this emission quantity, 57–100 tonnes was emitted from CFPPs each year during 2000–2015 (Zhang et al., 2015; Liu et al., 2018). The air pollution control devices (APCDs) for CFPPs in China has improved dramatically in the past two decades. Devices including the high efficient dust collector (such as electrostatic precipitator (ESP) and fabric filter (FF)), wet flue gas desulfurization (WFGD), flue gas denitrification (especially selective catalytic reduction (SCR)) have been gradually installed since the late 1990s (Wu et al., 2016). The combination of different APCDs provide substantial co-benefits for Hg removal from the flue gas in addition to the removal of particulate matter (PM), SO<sub>2</sub> and NO<sub>x</sub> (Wang et al., 2010a). There have been reports documenting the enhanced Hg removal in CFPPs in China provided by ESP or ESP + WFGD (Guo et al., 2007; Chen et al., 2007; Wang et al., 2010b; Wang et al., 2011a, b; Tang and Pan, 2013). Much fewer studies reflect the recent advances of flue gas denitrification (especially SCR) (Zhang et al., 2017; Zhao et al., 2017a; Zhou et al., 2013). More importantly, their associated co-benefits on Hg emission reduction and speciation changes along with the flue gas desulfurization and particulate removal are not well understood. Such information is needed to appropriately update the Hg emission inventories for CFPPs in China.

In the present study, a representative and large scale pulverized coal (PC) fired power plant in North China was systematically studied. A special emphasis is placed on the behaviors of Hg in each APCDs such as SCR, FF-ESP and WFGD, hence the mass balance of Hg and the ultimate Hg emission factors can be obtained. Based on the APCD implemented in different periods and associated mercury emission factors obtained

by the literature, historical Hg emissions from this CFPP since its operation in 1970s was estimated. This provides insights into Hg emissions from this CFPP over a relatively long period. The information obtained from this study serves as an important case study for evaluating Hg emission and speciation changes over time for existing CFPPs in China.

## 2. Methods

### 2.1. Studied coal-fired power plant

The studied CFPP locates in Tangshang city, Hebei province, North China, 170 km east of Beijing. It has eight utility boilers that put into operation during 1976–1987, totally with an installation capacity of 1550 MW. The information of each utility boiler and the associated APCDs was showed in Table S1. All boilers are pulverized coal fired boilers, Unit #1 and #2 (each 125 MW) were built in 1976 and 1977, respectively; Unit #3 and #4 (each 250 MW) were built in 1978; Unit #5 and #6 (each 200 MW) were built in 1983 and 1984; and Unit #7 and #8 (each 200 MW) were built in 1986 and 1987, respectively. The only APCD was Venturi Scrubber (VS) before 1995 except for Unit #7 and #8 that equipped with cold side ESP since their operation. From 1996 to 1998, all VS was upgraded to cold side ESP; and from 2005 to 2008, the limestone-gypsum WFGD was implemented. In 2012 and 2013, the cold side ESP was retrofitted into the ESP-FF. During the second half of 2012 to Mid-2014, the SCR system was installed.

The studied CFPP once was the largest CFPP in China during the 1970s–1980s, not only its single utility boiler capacity but also the total installed capacity, and the CFPP served as an important electricity source to Beijing and North China (Zheng et al., 2015). While, with the policy of “developing large units and suppressing small ones”, the Unit #1 and #2 were shut down at the end of 2008, and this CFPP was scheduled to construct two 350 MW coal-fired units and two 770 MW natural gas-fired units to replace the existing six coal-fired units during 2018–2020. The APCDs used in this CFPP for the past four decades represent the contemporary, or the most advanced techniques among Chinese CFPPs, and this CFPPs abide by the air pollution control regulations strictly at different periods. Hence, the results obtained in this research would be representative for Hg emission from this source in China.

### 2.2. Sampling and analysis

In this research, the on-site testing was conducted in Unit #3 (250 MW) in August 2013, and the actual boiler's load remained at a consistent level of 190 MW during this study of a four day's period. All boilers in this CFPP before 2018 are PC boilers as aforementioned and feed by the same type of coal, which produced from the local Kai-Luan coal mine, a very big coal mine in Tangshan city that operated since 1878 and was the cradle of the China's coal mining industry (Li et al., 2017), and the APCDs installed among different units were identical for the same time period as indicated in Table S1, hence, the Hg removal efficiencies by a specific APCD or a combination of different APCDs, as well as the final Hg emission factors would be consistent among different units, this has been verified by previously studies that the type of coal and the configuration of APCDs was the fundamental influencing factors for Hg emissions from a same type of boiler, namely, PC or circulating fluidized bed (CFB) utility boiler (Yokoyama et al., 2000; Tang and Pan, 2013; Zhang et al., 2016; Pudasainee et al., 2017), while the boiler load (68–100%) has little effect on the Hg distribution's

among different coal combustion products for a same PC boiler (Zhao et al., 2017a). So, the results obtained in Unit #3 would reflect the overall average Hg emission levels of this CFPP.

In Unit #3, both the flue gas, solid and liquid samples were simultaneously collected (Fig. 1). The flue gas were collected at four sites, namely SCRinlet, ESP-FFinlet, ESP-FFoutlet and WFGDoutlet, using the Ontario Hydro Method (OHM) according to ASTM Method 6784-02 (ASTM, 2008) (Fig. S1) with APEX XC-572 (Apex Instruments, USA). OHM separates three operationally defined Hg species, i.e. Hg<sup>0</sup>, Hg<sup>2+</sup> and Hg<sup>p</sup>. Hg<sup>p</sup> is first collected on the glass fiber filter, and an additional glass cyclone is applied in front of the glass fiber filter for high particulate loads (mainly before the dust collector). Three impingers containing KCl solution (1 mol L<sup>-1</sup>) are utilized to capture Hg<sup>2+</sup>. One impinger with H<sub>2</sub>O<sub>2</sub> + HNO<sub>3</sub> (10% v/v + 5% v/v) and three impingers with H<sub>2</sub>SO<sub>4</sub> + KMnO<sub>4</sub> solution (10% v/v + 4% m/v) jointly collect Hg<sup>0</sup> through oxidation and absorption processes. At the end, an impinger with silica gel is used to remove the moisture in flue gas. Both the probe and the filters were heated to at least 120 °C to avoid the water condensation and Hg adsorption on the tubing walls. Each flue gas sample lasted for 1–1.5 h with a flue gas volume of about 1.0–1.5 m<sup>3</sup>. After sampling, Hg measurements were made in the laboratory where Hg in each impinger was recovered by SnCl<sub>2</sub> reduction and then quantified by Cold Vapor Atomic Absorption Spectrophotometry (CVAAS, F732S, Shanghai Huaguang Instrument Corp), which has a detection limit of 0.05 µg L<sup>-1</sup>. Duplicate or triplicate measurements were conducted for each sample to gain a mean value.

Solid materials (including feed coal, bottom ash, ESP-FF fly ash, limestone, gypsum) and liquid sample such as WFGD waste water were collected in synchronism with the flue gas sampling, taking consideration of the different rhythms of material in the coal power plant sections. Each sample (~1 kg or ~1 L) represent a composite sample at each time. The coal samples were collected on the conveyor belt to the coal mill and limestone powder were taken from the limestone mill 30 min before each flue gas sampling. Bottom ash, fly ash, gypsum and WFGD waste water were collected immediately after each flue gas sampling. Fly ash was a mixture of ESP ash and FF ash since they were mixed together to the fly ash bucket for transportation. The solid samples were dried at 40 °C, then homogenized and grounded to < 150 µm. US EPA Method 7473 was adopted to determine the Hg concentration in solid samples (U.S. EPA, 2007), which heat solid samples at 800° and measure the released Hg<sup>0</sup> by cold-vapor atomic absorption spectrometry (CVAAS) (Lumex RA915+, Russia) with a detection limit of 0.1 µg kg<sup>-1</sup>. Each solid sample was determined at least three times to obtain the mean. Liquid samples were determined

by cold-vapor atomic fluorescence spectrometry (CVAFS) (Tekran 2500, Canada) following US EPA method 1631 (U.S. EPA, 2002).

Proximate analysis of coal was performed using the Chinese national standard method (GB/T 212–2008). Ultimate analysis of coal was accomplished by an elemental analyzer (vario MACRO cube, Elementar, Germany). For total sulfur in coal, it was detected by the Eschka method according to GB/T 214–2007. The calorific value of coal was determined by GB/T 213–2008.

### 2.3. Quality assurance and quality control

All sampling lines (quartz glass and Teflon tubing), impingers and bottles used in the flue gas sampling were soaked in a 20% nitric acid liquid overnight in the laboratory and washed with deionized water before sampling. The systematic blank was determined by sampling the ambient air, which was found to be 0.05 µg m<sup>-3</sup>. Hg concentrations in reagents used in the experiments were below the detection limit, except H<sub>2</sub>SO<sub>4</sub> (at ~1.2 ng Hg·mL<sup>-1</sup> concentrated), which compromised about 80% of the systematic blank. Certified reference materials (CRMs) for coal, fly ash and soil (NIST 1632d for coal, NIST 1633c for fly ash, GBW 08401 for fly ash, GBW07405 for soil) were utilized to guarantee the analytical quality for Hg. The recovery of Hg of different CRMs was found to be in the range of 98%–105%. For proximate analysis and ultimate analysis of coal, certified reference materials (GSB06-2105-2007 for anthracite coal and GSB06-2113-2007 for bituminous coal) were detected, the measured values were close to the certified contents and the variation coefficients were lower than 5%.

## 3. Results

### 3.1. Coal properties

The property of all feed coal samples (N = 7) collected during the campaign is highly consistent (Table 1). This is anticipated since the coal was produced in the same coal mine. The feed coal represents a typical bituminous coal, with average volatile matter of 25.4 ± 1.9%. Feed coal had relatively high ash yield (30.6 ± 4.6%), which was about 2 times of the national average (16.85%, Li and Zhai, 1994). Sulfur in feed coal was relatively low, with average of 0.52 ± 0.21%, which belongs to low sulfur coal in China (0.51–1.00%, GB/T 15224.2–2010) and was about a half of the national average 1.15% (Li and Zhai, 1994). Hg in the coal averaged at 139 ± 27 µg kg<sup>-1</sup>, which was slightly lower than the national average of 170 µg kg<sup>-1</sup> (Zhang et al., 2012).

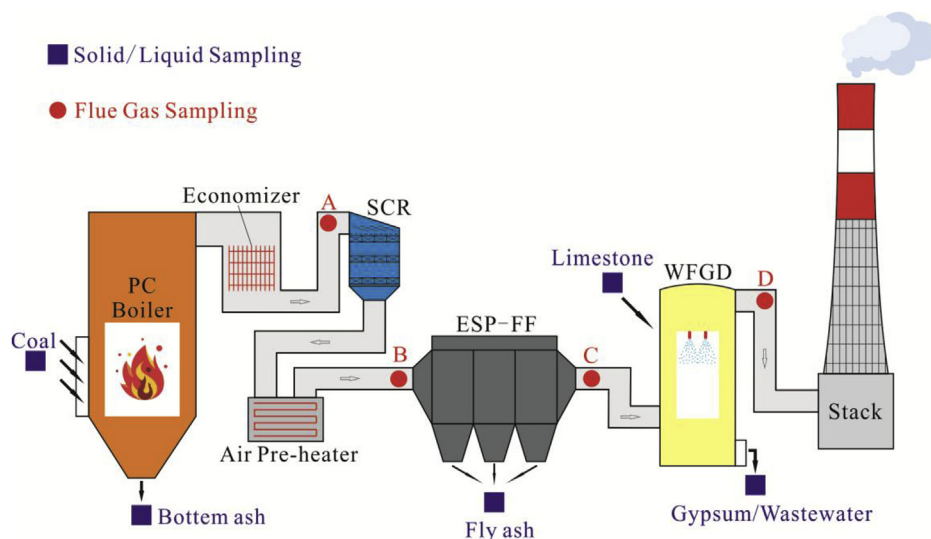


Fig. 1. Sampling locations of the tested utility boiler.

**Table 1**  
Proximate and ultimate analysis of feed coal.

No.	Proximate analysis					Ultimate analysis					
	Mad (%)	Vad (%)	Aad (%)	FCad (%)	Qad (MJ/kg)	Cad (%)	Had (%)	Nad (%)	Oad (%)	Sad (%)	Hg ( $\mu\text{g kg}^{-1}$ )
#1	3.8	25.6	28.96	41.7	20.65	53.2	3.56	0.89	9.17	0.40	86
#2	5.0	26.6	28.77	39.6	20.22	51.9	3.54	0.87	9.03	0.90	175
#3	2.3	25.0	33.92	38.8	19.34	50.0	3.34	0.88	9.33	0.25	129
#4	4.0	27.7	30.83	37.5	20.10	51.6	3.46	0.86	8.55	0.68	144
#5	2.7	22.9	37.07	37.3	18.12	46.5	3.25	0.87	9.03	0.50	141
#6	3.8	23.1	32.43	40.6	18.97	49.1	3.36	0.87	9.95	0.44	145
#7	6.1	26.9	22.45	44.6	19.48	50.4	3.42	0.87	9.30	0.45	152
Mean $\pm$ SD	4.0 $\pm$ 1.3	25.4 $\pm$ 1.9	30.63 $\pm$ 4.63	40.0 $\pm$ 2.6	19.55 $\pm$ 0.85	50.4 $\pm$ 2.2	3.42 $\pm$ 0.11	0.87 $\pm$ 0.01	9.20 $\pm$ 0.42	0.52 $\pm$ 0.21	139 $\pm$ 27

Note: M, moisture; V, volatile matter; A, ash yield; FC, fixed carbon; Q, calorific value; C, carbon; H, hydrogen; N, nitrogen; O, oxygen; S, sulfur; Hg, mercury; subscript “ad”, air-dried basis.

It should be noted that there has a slight variation of each investigated parameter abovementioned during a four days' sampling period, this situation might be true for the whole Kai-Luan coal mine, that covering an area of 550 km<sup>2</sup> in Tangshan city and have a coal resource of 4.1 billion tonnes in 1990s (Zhuang et al., 1999). The background values of trace elements in coal were dominated by sediment source regions (Dai et al., 2012), and coal in Kai-Luan coal mine was bituminous which formed in large inland depression basin during late Carboniferous and early Permian (C<sub>2</sub>-P<sub>1</sub>) (Zhuang et al., 1999). Due to the similar coal-forming and setting environment and without external interruptions such as hydrothermal-fluid intrusion and volcanic-ash incorporation, the trace element in this coal mine was anticipated to be confined in a narrow range. And this was confirmed by a previously report showing the average Hg content in Kai-Luan coal mine was 163  $\mu\text{g kg}^{-1}$  (Ren et al., 2006), much close to the average (139  $\pm$  27  $\mu\text{g kg}^{-1}$ ) of this research.

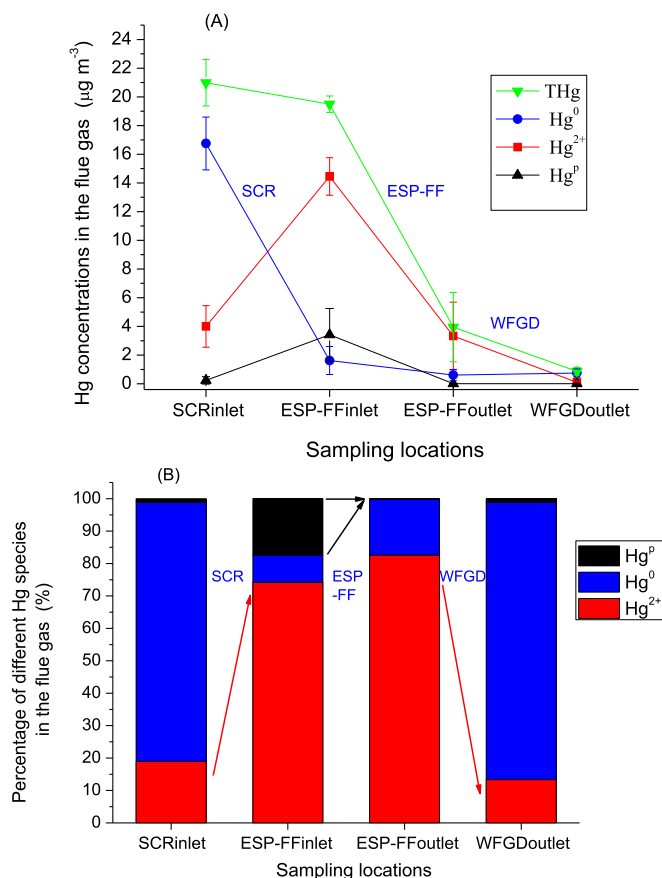
### 3.2. Hg in flue gas across different APCDs

Hg concentration and mass percentage of Hg species in the flue gas at four sampling locations from the upper stream (boiler outlet) to downstream stack were illustrated in Fig. 2 and Table S2.

Total Hg (Hg<sup>T</sup>) in flue gas at the SCRinlet was 20.99  $\pm$  1.62  $\mu\text{g m}^{-3}$ , it dropped to 0.87  $\pm$  0.26  $\mu\text{g m}^{-3}$  at the WFGDoutlet or stack, the discharged concentration was much lower than the Chinese national standard for coal-fired power plant (GB13223-2011), 30  $\mu\text{g m}^{-3}$ . The observed concentration was much lower than pulverized coal utility boilers that only equipped with ESP (9.2–32.1  $\mu\text{g m}^{-3}$ , Tang et al., 2007; Guo et al., 2007; Chen et al., 2007) and ESP + WFGD (1.6–14.5  $\mu\text{g m}^{-3}$ , Wang et al., 2010b; Wang et al., 2011a,b; Tang and Pan, 2013), while similar to CFPPs that installed SCR + ESP + WFGD (range: 0.1–2.8  $\mu\text{g m}^{-3}$  and > 50% cases were less than 1.0  $\mu\text{g m}^{-3}$ , Tang and Pan, 2013; Zhang et al., 2017; Tang et al., 2016; Zhao et al., 2017a; Zhou et al., 2013; Pudassanee et al., 2012; Yokoyama et al., 2000).

Both the Hg species and concentrations in flue gas changed remarkably along the sampling point of APCDs train (Fig. 2 and Table S2), reflecting the effectiveness of each APCD on Hg transforming and removals. For SCR, although total Hg concentration was similar at its inlet and outlet (here refers to ESP-FFinlet), Hg species changed from Hg<sup>0</sup> dominated (79.9% of total Hg) at the SCRinlet to Hg<sup>2+</sup> dominated (74.3% of total Hg) at the ESP-FFinlet. The high Hg<sup>0</sup> fraction in SCRinlet might ascribe to the low chlorine content (mean 110 mg kg<sup>-1</sup>, range 70–150 mg kg<sup>-1</sup>, Ren et al., 2006) in the Kailuan coal mine. The oxidation of Hg<sup>0</sup> inside the SCR was facilitated by the catalyst of SCR layer, such as V<sub>2</sub>O<sub>5</sub>-WO<sub>3</sub>-CeO<sub>2</sub>, and flue gas components such as halogens (Zhang et al., 2016; Yang et al., 2017; Chen et al., 2007). Subsequently, the formed Hg<sup>2+</sup> in flue gas was adsorbed on fly ash to form particulate Hg and decrease the fraction of Hg<sup>0</sup> in total Hg from 79.9% to 8.3%, while increases Hg<sup>P</sup> from 1.1% to 17.4%. The overall Hg<sup>0</sup>

oxidation efficiency was 90.3%, higher than the typical reported oxidation efficiency (mean = 74%) of SCR (Zhang et al., 2016). For ESP-FF, it decreased total Hg from 19.49  $\mu\text{g m}^{-3}$  to 3.95  $\mu\text{g m}^{-3}$  (~80% reduction) and efficiently removed Hg<sup>P</sup> (99.8%), meanwhile, it removed Hg<sup>0</sup> and Hg<sup>2+</sup> at a lesser level of 62–77% (Table 2). This suggests that Hg<sup>0</sup> was continuously convert into Hg<sup>2+</sup> and then adsorbed onto fly ash inside ESP-FF, since the fly ash cake formed on the fabric filter can serve as an active Hg sorption layer, and the adsorption process is influenced by a range of factors (Gao et al., 2015). The result of this study was similar to that of Senior et al. (2000), which reported 73% of gaseous Hg was captured in FF. After ESP-FF, the fraction of Hg<sup>2+</sup> increased to 86.2%. WFGD mainly absorbed the soluble Hg<sup>2+</sup> and have little effect on Hg<sup>0</sup> and Hg<sup>P</sup> in the flue gas (Table S2 and Table 2). Gaseous oxide mercury (Hg<sup>2+</sup>) was efficiently (96.8%) scrubbed by the limestone slurry and the share of Hg<sup>2+</sup> declined to



**Fig. 2.** Concentration (A) and mass percentage (B) of different Hg species in flue gas at different sampling locations of the tested utility boiler.

**Table 2**  
Hg removal efficiency (%) across different air pollutant control devices (APCDs) in tested utility boiler.

APCDs	Hg <sup>2+</sup>	Hg <sup>0</sup>	Hg <sup>p</sup>	Hg <sup>T</sup>
SCR	−261.5	90.3	−1337.8	7.1
ESP-FF	76.9	62.2	99.8	79.7
WFGD	96.8	−23.0	−43.5	78.0
The entire APCDs (SCR + ESP-FF + WFGD)	97.3	95.5	96.3	95.9

13.4% in WFGD outlet. In contrast, Hg<sup>0</sup> increased from 0.61 to 0.75  $\mu\text{g m}^{-3}$  and Hg<sup>p</sup> increased 6  $\text{ng m}^{-3}$  to 9  $\text{ng m}^{-3}$  across the WFGD, respectively, indicating slight re-emission of Hg<sup>0</sup> and the gypsum rain formed inside the WFGD system (Wu et al., 2010). After WFGD, Hg<sup>0</sup> became the predominant species (86%), followed by Hg<sup>2+</sup> (13%) and Hg<sup>p</sup> (1%).

The removal efficiency of Hg<sup>T</sup> across the whole flue gas treating system (SCR + ESP-FF + WFGD) was 95.9%, and the removals for different Hg species (Hg<sup>0</sup>, Hg<sup>2+</sup> and Hg<sup>p</sup>) ranged from 95.5% to 97.3% (Table 2). Collectively, the high Hg removal occurred in this CFPP shows a synergistic effect of SCR, ESP-FF and WFGD, with SCR convert 90.3% of Hg<sup>0</sup> into Hg<sup>2+</sup>, and ESP-FF removed 99.8% of Hg<sup>p</sup> downstream, and WFGD absorbed 96.8% of Hg<sup>2+</sup>. Such removal efficiency (95.9%) was high compared to 58.8–73.3% that obtained from a 350 MW CFPPs with the same APCDs by Zhao et al. (2017a), but comparable to other CFPPs in China, South Korea and Netherland that achieved over 90% of Hg removals with similar APCDs (Tang et al., 2016; Zhang et al., 2017; Pudasainee et al., 2012; Meij and Winkel, 2006).

### 3.3. Hg mass balance and emission factors

Based on Hg concentration in different input and output solid/li-liquid/flue gas samples, and their associated mass flow (Table S3), Hg mass balance in the utility boiler #3 system was calculated and shown in Fig. 3 and Table S3. The average Hg content in feed coal, bottom ash, fly ash, limestone, gypsum were 139  $\pm$  27, 58  $\pm$  28, 465  $\pm$  89, 2  $\pm$  1, 1012  $\pm$  142  $\mu\text{g kg}^{-1}$  (air dried basis, N = 7), respectively, and for the WFGD wastewater was 8.26  $\pm$  0.49  $\mu\text{g L}^{-1}$  (N = 4). And the material input rate was 2158 and 61  $\text{t d}^{-1}$  for feed coal and limestone, respectively; the material output rate was 60, 538 and 93  $\text{t d}^{-1}$  for bottom ash, fly ash and FGD gypsum, respectively, and 61  $\text{t d}^{-1}$  for the desulfurization waste water and 15.56 million  $\text{m}^3 \text{d}^{-1}$  for the stack flue gas, respectively. Combined this, Hg input into Unit #3 was 284.7  $\text{g d}^{-1}$

while the output was 282.5  $\text{g d}^{-1}$ , the ratio of Hg output/input was 99.2%, indicating near complete recovery. The main input of Hg into the system was feed coal (99.96%), whilst limestone accounts a small share of 0.04%. The main Hg output was through fly ash (88.6%), followed by WFGD gypsum (5.2%), bottom ash (1.2%) and WFGD wastewater (0.2%). Only a small fraction (4.8%) of Hg output is released into the atmosphere through the stack flue gas, comparable to a Korea CFPP (3.7%) that equipped with SCR + ESP + WFGD (Pudasainee et al., 2012).

Hg emission factors (MEF) were estimated by three methods (Table S3), namely, based on the coal consumption (MEF #1, 6.3  $\text{mg Hg t}^{-1}$  coal), or the electricity generated (MEF #2, 2.96  $\mu\text{g Hg (kW-h)}^{-1}$ ) or the coal heat value (MEF #3, 0.32  $\text{g Hg-TJ}^{-1}$ ). Compared with MEFs obtained from sixteen Chinese PC utility boilers equipped with ESP + WFGD (11–131  $\text{mg Hg t}^{-1}$  coal or 14.09–56.08  $\mu\text{g Hg (kW-h)}^{-1}$ , Wang et al., 2010b; 2011a; 2011b), the MEF of this study was relatively low, and comparable to a CFPP in Japan (4.43  $\mu\text{g Hg (kW-h)}^{-1}$ , Ito et al., 2006) and a CFPP in China (0.92–1.56  $\text{g Hg-TJ}^{-1}$ , Zhao et al., 2017a,b) that equipped with SCR + ESP + WFGD. The high mercury removals of PC boilers equipped with SCR should be ascribe to the oxidation of Hg<sup>0</sup> and the subsequent retention/absorption of Hg<sup>2+</sup> in the downstream ESP/WFGD that already installed, the additional Hg removals in CFPPs by SCR were achieved in the range of 82–85% in a south Korea CFPP (Pudasainee et al., 2012) and a Chinese CFPP (Tang and Pan, 2013).

### 3.4. Cumulative Hg emission from the studied CFPP

For a specific utility boiler, atmospheric Hg emission is mainly constrained by the APCDs if the coal property remains the same (Chen et al., 2018; Pudasainee et al., 2012; Tang and Pan, 2013). For the studied CFPP that use the local bituminous coal, Hg atmospheric release is largely dependent on the gradual APCD implementation in the past four decades, as shown in Table S1.

Hg removal with various APCDs configurations was obtained for the average value through literature reviews with similar APCDs and Hg speciation profiles in the flue gas, as well as data from this study and our data obtained from 12 PC CFPPs in Guizhou province, China (Table 3). In summary, the total Hg removals by VS, cold ESP, cold side ESP + WFGD, cold side ESP-FF + WFGD, SCR + cold side ESP-FF + WFGD were 26%, 24%, 52%, 72% and 96%, respectively, similar to those found in Pavlish et al. (2003) and Wu et al. (2016). Based on this study, 99% of Hg in feed coal was lost into flue gas during the combustion, with 1% remained in the bottom ash, and the Hg

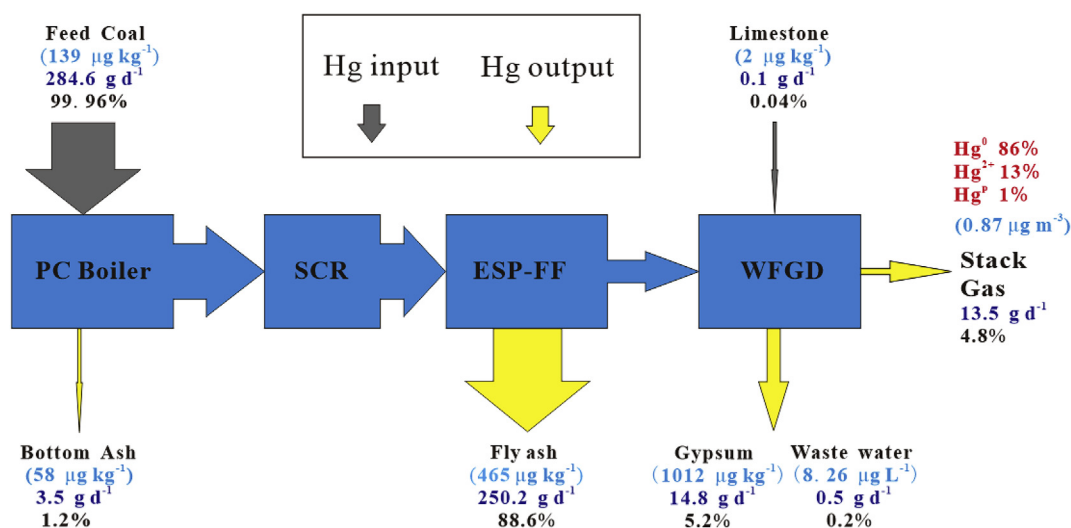


Fig. 3. Diagram of Hg input/output in the tested utility boiler.

**Table 3**  
Hg removal efficiencies of different APCDs for the studied CFPP based on this study and literature reviews.

APCDs	Ratio of Hg in flue gas out of the boiler (%)	Hg species in the inlet of APCDs (%)			Hg removals (%)				Reference
		Hg <sup>P</sup>	Hg <sup>2+</sup>	Hg <sup>0</sup>	Hg <sup>P</sup>	Hg <sup>2+</sup>	Hg <sup>0</sup>	Hg <sup>T</sup>	
VS	99	10	30	60	60.3	88.9	−11.0	26.1	Tang et al. (2007); Xue, 2013
Cold side ESP	99	10	30	60	98.2	9.8	19.2	24.3	Chen et al. (2007); Wang et al. (2010b)
Cold side ESP + WFGD	99	10	30	60	99.8	88.7	25.9	52.2	Wang et al. (2010b); 12 PC CFPPs in Guizhou, unpublished data
Cold side ESP-FF + WFGD	99	10	30	60	99.7	99.2	53.7	72.0	Based on this study
SCR + Cold side ESP-FF + WFGD	99	1.11	18.98	79.91	96.3	97.3	95.5	95.9	This study

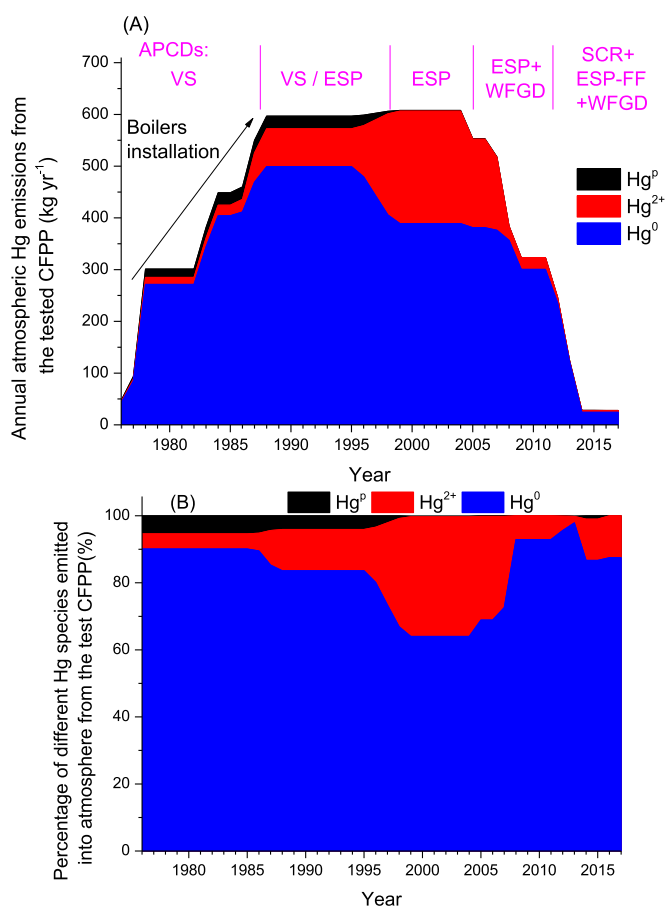
speciation in the flue gas of SCR inlet were 1.11%, 18.98% and 79.61% for Hg<sup>P</sup>, Hg<sup>2+</sup> and Hg<sup>0</sup>, respectively. While, for APCD configurations without SCR, Hg speciation in the flue gas before entering the APCDs was taken as 10%, 30% and 60% for Hg<sup>P</sup>, Hg<sup>2+</sup> and Hg<sup>0</sup>, respectively, taking into account the additional reactions in the flue gas between the SCR and the VS/ESP due to the temperature drop and the additional reaction time (Tao et al., 2010). The result for atmospheric Hg emission from this CFPP during 1976–2017 was shown in Fig. 4. There were three distinct stages of atmospheric Hg emission in the past 42 years (Fig. 4A). Phase I (1976–1987), the annual atmospheric Hg emission increased from 47 kg yr<sup>-1</sup> to 550 kg yr<sup>-1</sup>, mainly due to the installation of eight boilers in this period. Phase II (1988–2004), the annual atmospheric Hg emission remained at a high level (597–608 kg yr<sup>-1</sup>), although VS changed to ESP in this period, the emission of total Hg did not change significantly. The annual emission peaked during 1998–2004 with a rate of 608 kg yr<sup>-1</sup>. Phase III (2005–2017), Hg emission reduced dramatically during 2005–2008 and 2012–2014. The reduction during these two periods corresponded to the installation of WFGD, ESP-FF and SCR. During 2005–2008, atmospheric Hg emission reduced from 553 to 384 kg yr<sup>-1</sup>, and during 2012–2014, the emission was further reduced from 247 to 28 kg yr<sup>-1</sup>. After 2014, Hg emissions from this CFPP has remained at low level of 28 kg yr<sup>-1</sup>, only 1/22 of its peak release.

During the past four decades (1976–2017), this CFPP cumulatively emitted 17.6 ± 3.4 tonnes (range:10.9–22.2 tonnes) of Hg into the atmosphere (Figs. 4B and 5, Table S4) based on the error propagation method that incorporate the coal Hg content and Hg emission factor in different stages. This figure accounted for 1.3‰ of the national total anthropogenic emissions (about 13 × 10<sup>3</sup> t) or 2.1% of Hebei provincial emissions (831 t) in the almost same period (1978–2014, Wu et al., 2016), noting that there have more than 600 thousands of Hg point sources in China (Chen et al., 2015). The average speciation profile of Hg<sup>0</sup>:Hg<sup>2+</sup>:Hg<sup>P</sup> emitted from this CFPP was at 79.5%:17.9%:2.6% in the past four decades. The share of Hg<sup>P</sup> to total Hg in stack flue gas was largest (5.4%) when VS was the dominant APCDs before 1985, it became negligible (< 1%) after ESP or ESP-FF was installed. Hg<sup>2+</sup>% in total Hg peaked in 1999–2007 (35.7%) when ESP was the dominate control measure (Fig. 4B). For Hg<sup>0</sup>, it was the leading Hg species emitted into atmosphere from this CFPP for all times, its share in total Hg ranged from 64% to 98%, with lowest for ESP (1999–2004) and highest for ESP-FF + WFGD (2013). For total Hg, it was mainly exhausted before 2008 when the WFGD were completed installed, and 89.6% of the total emissions was occurred in the first three decades, only 10.4% occurred during the last ten years (2008–2017).

Mercury emission factor (MEF) and Hg removal efficiency (MRE), both highly depended on the APCDs (Pudasainee et al., 2012; Tang and Pan, 2013), had undergone remarkable changes during the past four decades (Fig. 6). Before 2005, the MEF were relative stable and kept in a high level of 102–104 mg Hg t<sup>-1</sup> coal; during 2005–2008, MEF dramatically reduced to about a half level (66 mg Hg t<sup>-1</sup> coal), and from 2012 to 2014, the MEF was further reduced to a very low level (5.7 mg Hg t<sup>-1</sup> coal). The substantial Hg emission reduction was occurred after 2005. Since 2014, the MEF was only 5.9%, or 1/17, of the levels before

2005.

During the 42 years operation, the total Hg input by coal was 28.5 ± 5.6 tonnes (range:17.6–35.9 tonnes), with 17.6 ± 3.4 tonnes emitted into atmosphere, hence the total emission ratio was 61.8%, and the average MRE was 38.2% (Table S4), the obviously emission reduction was achieved in the recent 10 years. The atmospheric mercury emission rate before 2005 (73.7%) of this CFPP was similar to different utility and industrial boilers (64.0%–78.2%) in China during 1978–1995 (Wang et al., 2000). This CFPP located closely to Beijing and implemented the air pollution control policies completely and promptly after the emission control policy strengthened for each time, hence could be a typical case of Hg emissions from Chinese CFPPs in the past four decades. At the same time, the results obtained from this study could provide important information for assessing the environmental impact of Hg release from this CFPP, such as the atmospheric Hg levels, Hg wet/dry depositions, and Hg accumulation in the surrounding soils



**Fig. 4.** Dynamics of atmospheric Hg emissions from the tested CFPP during 1976–2017: (A) Amount of different Hg species (kg yr<sup>-1</sup>); (B) Percentage of different Hg species (%)

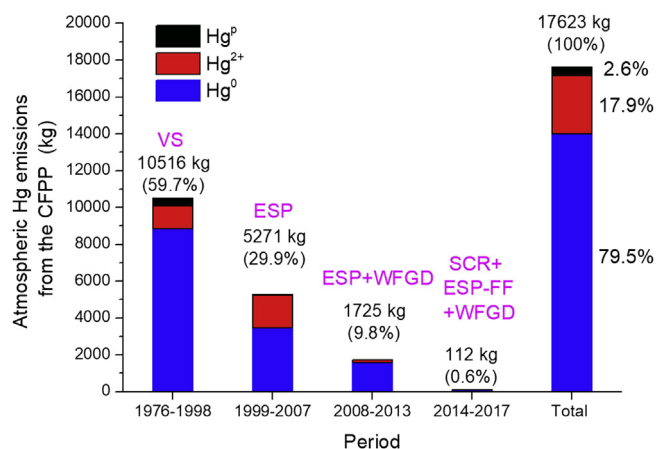


Fig. 5. Atmospheric Hg emissions from the tested CFPP in different periods with varying configuration of APCDs.

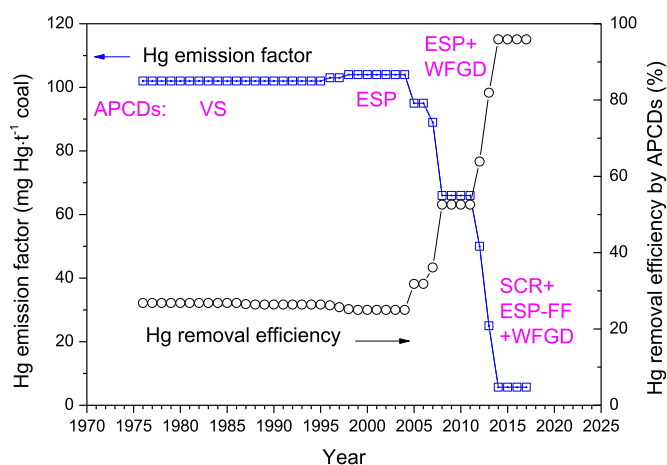


Fig. 6. Dynamics of Hg emission factor versus Hg removals by different APCDs in the tested CFPP during 1976–2017.

(Li et al., in prep.).

It should be noted that, although there have not been specific Hg removal techniques (such as active carbon injection) used in CFPPs, the co-beneficial Hg removal by APCDs is high (> 95%) with ESP/ESP-FF, WFGD and SCR. This combination of APCDs has the potential to be a best available technique (BAT) for Hg control in Chinese CFPPs. Although CFPPs has been listed as the No.1 priority for Hg control in Minamata Convention, CFPPs in China have achieved a much higher Hg reduction in the past decade. However, the re-mobilization of Hg from the APCDs byproducts (such as fly ash and gypsum) must be carefully monitored to avoid the second Hg release into the environment.

#### 4. Conclusions

The configuration of APCDs with SCR + ESP-FF + WFGD in a 250 MW pulverized coal-fired utility boiler achieves high (96%) Hg removal, with SCR oxidizing 92% of Hg<sup>0</sup> and WFGD scrubbing 97% Hg<sup>2+</sup> downstream and ESP-FF retaining nearly all Hg<sup>p</sup>. Hg concentration in stack gas was 0.87 μg m<sup>-3</sup> with a speciation profile of Hg<sup>0</sup>:Hg<sup>2+</sup>:Hg<sup>p</sup> at 86%:13%:1%. The detected total Hg concentration is substantially below than the Chinese National Standard for coal-fired power plant (GB13223-2011), 30 μg m<sup>-3</sup>. This CFPP cumulatively emitted 17.6 tonnes of Hg into the atmosphere during the past four decades (1976–2017), with a majority of the Hg release (89%) taking place before the WFGD was installed in 2008. Although the quantity and speciation of emitted Hg changed significantly with gradual APCDs

implementation, the predominant emission species remains consistently as Hg<sup>0</sup> (64–98%). Significant Hg<sup>2+</sup> and Hg<sup>p</sup> emission only occurred when ESP and VS were the primary APCDs. The installation of WFGD, ESP-FF and SCR dramatically lowered the Hg emission factor by 17 times compared to the peak emission. In the future, more attention should be directed to the stability of Hg in fly ash and gypsum products and the associated Hg re-emissions.

#### Conflicts of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper “Evolution of four-decade atmospheric mercury release from a coal-fired power plant in North China”.

#### Acknowledgements

The authors greatly appreciate the two anonymous reviewers for their constructive suggestions. This work is financially supported by the Science & Technology Research and Development Plan of Tangshan (No. 12130215A), the National Key Basic Research Program of China (No. 2013CB430001) and the National Natural Science Foundation of China (U1612442, 41428301). And we also appreciate the hard work during the field works for Xu Yang and Zhuo Lu, and the help of the staff of the studied CFPP.

#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.atmosenv.2019.06.045>.

#### References

- ASTM Standard (Method 6784-02) Test Method for Elemental, 2008. Oxidized, Particle-Bound, and Total Mercury in Flue Gas Generated from Coal-Fired Stationary Sources (Ontario-Hydro). ASTM International, West Conshohocken, P.A.
- Chen, L., Duan, Y., Zhuo, Y., Yang, L., Zhang, L., Yang, X., Yao, Q., Jiang, Y., Xu, X., 2007. Mercury transformation across particulate control devices in six power plants of China: the co-effect of chlorine and ash composition. *Fuel* 86, 603–610.
- Chen, L., Li, S., Du, X., Sun, Y., 2015. The dilemma and solutions for implementation of Minamata Convention. *Chinese Ecol. Civ. (Zhongguoshengtaiwennming)* 74–79 (in Chinese).
- Chen, Z., Wang, R., Sun, R., Wang, H., Hu, H., Huang, H., 2018. Distribution and enrichment of mercury in utility boiler of Huainan coal-fired power plant. *Environ. Chem.* 37 (2), 193–199 (In Chinese with English Abstract).
- Dai, S., Ren, D., Chou, C.L., Finkelman, R.B., Seredin, V.V., Zhou, Y., 2012. Geochemistry of trace elements in Chinese coals: a review of abundances, genetic types, impacts on human health, and industrial utilization. *Int. J. Coal Geol.* 94, 3–21.
- Dai, S., Finkelman, R.B., 2018. Coal geology in China: an overview. *Int. Geol. Rev.* 60 (5–6), 531–534.
- Driscoll, C.T., Mason, R.P., Chan, H.M., Jacob, D.J., Pirrone, N., 2013. Mercury as a global pollutant: sources, pathways, and effects. *Environ. Sci. Technol.* 47, 4967–4983.
- Galbreath, K.C., Zygarlicke, C.J., 2000. Mercury transformations in coal combustion flue gas. *Fuel Process. Technol.* 65–66, 289–310.
- Gao, Z., Lv, S., Chen, S., Yang, P., Zheng, S., Yin, L., Chen, C., 2015. ESP effects on particulate mercury speciation and thermal stability in a coal-fired utility boiler. *J. North China Electr. Power Univ. (Soc. Sci.)* 42 (1), 63–68 (in Chinese with English abstract).
- Guo, X., Zheng, C., Xu, M., 2007. Characterization of mercury emissions from a coal-fired power plant. *Energy Fuel.* 21, 898–902.
- Gustin, M.S., Lindberg, S.E., Weisberg, P.J., 2008. An update on the natural sources and sinks of atmospheric mercury. *Appl. Geochem.* 23 (3), 482–493.
- Ito, S., Yokoyama, T., Asakura, K., 2006. Emissions of mercury and other trace elements from coal-fired power plants in Japan. *Sci. Total Environ.* 368 (1), 397–402.
- Li, W., Zhai, J., 1994. Both ash and sulfur content and calorific value in Chinese steam coals. *Coal convers. (Meitanzhuanhua)* 17 (1), 12–25 (In Chinese with English Abstract).
- Li, D., Lan, G.Z., Kraeger, P., Wei, M., 2017. Tangshan-China's one time industrial pioneer striving for ecological excellence. *Cities* 65, 1–9.
- Li, Z., Chen, X., Liu, W., Li, T., Wang, M., Qiu, G., Yan, H., Sun, G., Lu, Z., Yang, X., Chen, J., 2019. Linkage between Mercury Enrichment in Atmosphere/upland Agricultural Soil and Hg Emissions from a Nearby Large Scale Coal-Fired Power Plant in North China. *Atmosphere*. (in prep.).
- Lin, C.J., Pehkonen, S.O., 1999. The chemistry of atmospheric mercury: a review. *Atmos. Environ.* 33 (13), 2067–2079.

- Liu, K., Wang, S., Wu, Q., Wang, L., Ma, Q., Zhang, L., Li, G., Tian, H., Duan, L., Hao, J., 2018. A highly resolved mercury emission inventory of Chinese coal-fired power plants. *Environ. Sci. Technol.* 52 (4), 2400–2408.
- Mason, R.P., Fitzgerald, W.F., Morel, F.M.M., 1994. The biogeochemical cycling of elemental mercury: anthropogenic influences. *Geochem. Cosmochim. Acta* 58, 191–198.
- Meij, R., te Winkel, H., 2006. Mercury emissions from coal-fired power stations: the current state of the art in The Netherlands. *Sci. Total Environ.* 368, 393–396.
- Pavlish, J.H., Sondreal, E.A., Mann, M.D., Olson, E.S., Galbreath, K.C., Laudal, D.L., Benson, S.A., 2003. Status review of mercury control options for coal-fired power plants. *Fuel Process. Technol.* 82, 89–165.
- Pirrone, N., Cinnirella, S., Feng, X., Finkelman, R.B., Friedli, H.R., Leaner, J., Mason, R., Mukherjee, A.B., Stracher, G.B., Streets, D.G., Telmer, K., 2010. Global mercury emissions to the atmosphere from anthropogenic and natural sources. *Atmos. Chem. Phys.* 10, 5951–5964.
- Pudasainee, D., Kim, J.H., Yoon, Y.S., Seo, Y.C., 2012. Oxidation, reemission and mass distribution of mercury in bituminous coal-fired power plants with SCR, CS-ESP and wet FGD. *Fuel* 93, 312–318.
- Pudasainee, D., Seo, Y.C., Sung, J.H., Jang, H.N., Gupta, R., 2017. Mercury co-beneficial capture in air pollution control devices of coal-fired power plants. *Int. J. Coal Geol.* 170, 48–53.
- Ren, D.Y., Zhao, F.H., Dai, S.F., Zhang, J.Y., Luo, K.L., 2006. Chapter 5 “Different trace elements in coal”. In: *Geochemistry of Trace Elements in Coal*. Science Publishing House, Beijing (In Chinese).
- Schroeder, W.H., Munthe, J., 1998. Atmospheric mercury—an overview. *Atmos. Environ.* 32, 809–822.
- Senior, C.L., Helble, J.J., Sarofim, A.F., 2000. Predicting the speciation of mercury emissions from coal-fired power plants. In: *Proceedings of the Conference on Air Quality II: Mercury, Trace Elements, and Particulate Matter*, McLean, VA, September 19–21, 2000.
- Streets, D.G., Hao, J., Wu, Y., Jiang, J., Chan, M., Tian, H., Feng, X., 2005. Anthropogenic mercury emissions in China. *Atmos. Environ.* 39, 7789–7806.
- Streets, D.G., Devane, M.K., Lu, Z., Bond, T.C., Sunderland, E.M., Jacob, D.J., 2011. All-time releases of mercury to the atmosphere from human activities. *Environ. Sci. Technol.* 45 (24), 10485–10491.
- Streets, D.G., Horowitz, H.M., Jacob, D.J., Lu, Z., Levin, L., ter Schure, A.F.H., Sunderland, E.M., 2017. Total mercury released to the environment by human activities. *Environ. Sci. Technol.* 51 (11), 5969–5977.
- Streets, D.G., Lu, Z., Levin, L., ter Schure, A.F.H., Sunderland, E.M., 2018. Historical releases of mercury to air, land, and water from coal combustion. *Sci. Total Environ.* 615, 131–140.
- Tang, N., Pan, S.W., 2013. Study on mercury emission and migration from large-scale pulverized coal fired boilers. *J. Fuel Chem. Technol.* 41 (4), 484–490.
- Tang, S., Feng, X., Qiu, J., Yin, G., Yang, Z., 2007. Mercury speciation and emissions from coal combustion in Guiyang, southwest China. *Environ. Res.* 105 (2), 175–182.
- Tang, S., Wang, L., Feng, X., Feng, Z., Li, R., Fan, H., Li, K., 2016. Actual mercury speciation and mercury discharges from coal-fired power plants in Inner Mongolia, Northern China. *Fuel* 180, 194–204.
- Tao, Y., Zhuo, Y., Zhang, L., Chen, C., Xu, X., 2010. Mercury transformation across various air pollution control devices in a 200 MW coal-fired boiler of China. *Asia Pac. J. Chem. Eng.* 5, 756–762.
- U.S. Environmental Protection Agency (U.S. EPA), 2002. US EPA Method 1631, Revision E: Mercury in Water by Oxidation, Purge and Trap, Cold Vapor Atomic Fluorescence Spectrometry. (Washington, DC).
- U.S. Environmental Protection Agency (U.S. EPA), 2007. US EPA Method 7473, Mercury in Solids and Solutions by Thermal Decomposition, Amalgamation, and Atomic Absorption Spectrophotometry. (Washington, DC).
- Wang, Q., Shen, W., Ma, Z., 2000. Estimation of mercury emission from coal combustion in China. *Environ. Sci. Technol.* 34, 2711–2713.
- Wang, S., Zhang, L., Wu, Y., Ancora, M.P., Zhao, Y., Hao, J., 2010a. Synergistic mercury removal by conventional pollutant control strategies for coal-fired power plants in China. *J. Air Waste Manag. Assoc.* 60, 722–730.
- Wang, S., Zhang, L., Li, G., Wu, Y., Hao, J., Pirrone, N., Sprovieri, F., Ancora, M.P., 2010b. Mercury emission and speciation of coal-fired power plants in China. *Atmos. Chem. Phys.* 10, 1183–1192.
- Wang, S., Wang, H.M., Zhu, F.H., Chen, H., Sun, X.L., Zuo, Y., Liu, G., 2011a. Mercury emission characteristics from coal-fired power plants based on actual measurement. *Environ. Sci.* 32 (1), 33–37 (In Chinese with English abstract).
- Wang, J., Wang, W., Xu, W., Wang, X., Zhao, S., 2011b. Mercury removals by existing pollutants control devices of four coal-fired power plants in China. *J. Environ. Sci.* 23 (11), 1839–1844.
- Wu, C., Cao, Y., Dong, Z., Pan, W., 2010. Impacting factors of elemental mercury re-emission across a lab-scale simulated scrubber. *Chin. J. Chem. Eng.* 18 (3), 523–528.
- Wu, Q., Wang, S., Li, G., Liang, S., Lin, C.-J., Wang, Y., Cai, S., Liu, K., Hao, J., 2016. Temporal trend and spatial distribution of speciated atmospheric mercury emissions in China during 1978–2014. *Environ. Sci. Technol.* 50 (24), 13428–13435.
- Xue, Z., 2013. Atmospheric Mercury Emission Inventory in China and the Controls. Study Report of State Environmental Protection Commonweal Trade Scientific Research in China. (200909024). Beijing.
- Yang, Y., Liu, J., Wang, Z., Liu, F., 2017. Heterogeneous reaction kinetics of mercury oxidation by HCl over Fe<sub>2</sub>O<sub>3</sub> surface. *Fuel* 159, 266–271.
- Yokoyama, T., Asakura, K., Matsuda, H., Ito, S., Noda, N., 2000. Mercury emissions from a coal-fired power plant in Japan. *Sci. Total Environ.* 259, 97–103.
- Yudovich, Ya E., Ketris, M.P., 2005. Mercury in coal: a review, Part 1. *Geochemistry. Int. J. Coal Geol.* 62, 107–134.
- Zhang, L., Wang, S., Meng, Y., Hao, J., 2012. Influence of mercury and chlorine content of coal on mercury emissions from coal-fired power plants in China. *Environ. Sci. Technol.* 46, 6385–6392.
- Zhang, L., Wang, S., Wang, L., Wu, Y., Duan, L., Wu, Q., Wang, F., Yang, M., Yang, H., Hao, J., Liu, X., 2015. Updated emission inventories for speciated atmospheric mercury from anthropogenic sources in China. *Environ. Sci. Technol.* 49 (5), 3185–3194.
- Zhang, L., Wang, S., Wu, Q., Wang, F., Lin, C., Zhang, L., Hui, M., Yang, M., Su, H., Hao, J., 2016. Mercury transformation and speciation in flue gases from anthropogenic emission sources: a critical review. *Atmos. Chem. Phys.* 16, 2417–2433.
- Zhang, Y., Yang, J., Yu, X., Sun, P., Zhao, Y., Zhang, J., Gang, C., Yao, H., Zheng, C., 2017. Migration and emission characteristics of Hg in coal-fired power plant of China with ultra low emission air pollution control devices. *Fuel Process. Technol.* 158, 272–280.
- Zhao, S., Duan, Y., Chen, L., Li, Y., Yao, T., Liu, S., Liu, M., Lu, J., 2017a. Study on emission of hazardous trace elements in a 350 MW coal-fired power plant. Part 1. Mercury. *Environ. Pollut.* 229, 863–870.
- Zhao, S., Duan, Y., Yao, T., Liu, M., Lu, J., Tan, H., Wang, X., Wu, L., 2017b. Study on the mercury emission and transformation in an ultra-low emission coal-fired power plant. *Fuel* 199, 653–661.
- Zheng, R., Liu, Q., Wu, W., 2015. The construction history of Douhe power plant—a result of Four-Three Plan. *Energy (Nengyuan)* 8 (1), 108–112 (in Chinese).
- Zhou, J., Luo, Z., Zhu, Y., Fang, M., 2013. Chapter 2: “Mercury Sampling and Measurement in Coal-Fired Power Plants” in “Mercury Emissions and its Control in Chinese Coal-Fired Power Plants”. Springer, pp. pp11–20.
- Zhuang, X.G., Yang, S.K., Zeng, R.S., Xu, W.D., 1999. Characteristics of trace elements in coals from several main coal districts in China. *Geol. Sci. Technol. Inf.* 18 (3), 63–66 (in Chinese with English Abstract).