



Full Length Article

Emission characteristic and transformation mechanism of hazardous trace elements in a coal-fired power plant



Shilin Zhao^a, Yufeng Duan^{a,*}, Yaning Li^a, Meng Liu^a, Jianhong Lu^a, Yanjun Ding^b, Xiaobing Gu^c, Jun Tao^c, Mingsheng Du^c

^a Key Laboratory of Energy Thermal Conversion and Control of Ministry of Education, School of Energy and Environment, Southeast University, Nanjing 210096, China

^b State Key Laboratory of Power Systems, Dept. of Thermal Engineering, Tsinghua University, Beijing 100084, China

^c Datang Environment Industry Group Co., Ltd, Beijing 100097, China

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ABSTRACT

Coal-fired power plant is considered to be main anthropogenic emitter for emission of hazardous trace elements (HTEs), which can cause great damage on environment and human health. Emission characteristic of nine HTEs, namely Hg, Zn, Sb, Pb, Cd, As, Cr, Mn and Ba was carried out on a 320 MW coal-fired power plant equipped with SCR, ESP and WFGD. US EPA method 29 was used for flue gas HTEs sampling at inlet or outlet of each APCD simultaneously. Solid or liquid samples like feed coal, ESP ash, bottom ash, fresh slurry, waste water, gypsum were collected at the same time. Scanning electron microscope, energy dispersive spectrometer and X-ray powder diffraction spectrometry were adopted to characterize the physical and chemical structure characteristic of some solid samples. Results show that mass balance rate of the HTEs for each device and the system is in acceptable range. Mass amount of HTEs distributed in bottom ash and ESP ash accounts for large proportion, while little of them escapes into atmosphere ($< 0.05\%$). The mass amount of Mn and Ba in bottom ash is higher than that of other HTEs. Particulate HTEs are the main form in flue gas before ESP. ESP + WFGD has high removal efficiency for the HTEs in flue gas (99.96–99.99%), in which ESP makes a great contribution. During coal combustion, Mn and Ba enrich in bottom and fly ash equally while As, Cd, Sb, Zn, Pb and Cr will be prone to enrich in fly ash. Concentration of the HTEs in flue gas emitted to atmosphere is relatively low (3.5×10^{-3} – $4.30 \mu\text{g}/\text{m}^3$) with emission factor of 0.001–1.473 g/10¹² J. Finally, transformation mechanism of the studied HTEs during coal combustion is discussed.

1. Introduction

Hazardous trace elements (HTEs, including Hg, Zn, Sb, Pb, Cd, As, Cr, Mn, Ba, etc.) emitted from coal combustion have done great harm on the environment and human health, which is due to their high toxicity [1–3]. In China, more than 30 serious poisoning cases associated with HTEs pollution have happened successively since 2009, which include pollution incidents of Cd, As, Pb in Hunan province, Shandong province, and Guangdong province, respectively [4–6]. The US Clean Air Act Amendments have listed Sb, As, Cr, Pb, Cd, Hg, Ni, Se, Be, Mn and Co as key toxic air pollutants [7], and some trace elements (like As, Pb, Cd, Hg, and Ni) have been given the main environmental concern by the European Union [8]. In the Ambient Air Quality Standard of China (GB 3095-2012), emission of Pb to the atmosphere has been restricted [9]. Coal-fired power plant has been recognized as the main anthropogenic emitter of the HTEs [10,11].

Lots of work about the emission and distribution of HTEs in coal-

fired power plants have been conducted. Goodarzi et al. [12] used NAA, ICPES, ICP-MS, GFAA, CVAA and XANES spectroscopy to determine content and speciation of As, Cr, Ni and/or Hg in feed coal, ash and stack-emitted materials in a Canadian power plant burning bituminous coal. Swanson et al. [13] collected the feed coal, bottom ash, economizer fly ash and fly ash from two coal-fired power plants in US to discover the partitioning of As, Cr, Hg, Pb, and Se in coal combustion products. Klika et al. [14] studied the effects of boiler output on trace elements partitioning during coal combustion in two fluidised-bed power stations in Czech Republic. Bhangare et al. [15] sampled coal and ash from five different thermal power plants and detected concentration of trace elements in them, which aimed to understand their distribution, enrichment, and partitioning behavior. Tang et al. [10,16] conducted experimental analysis of the simultaneously collected feed coal, bottom ash, fly ash and FGD products to study the distribution and fate of trace elements in coal-fired power plants in Anhui province, China. However, the field data about the direct measurement of HTEs in

* Corresponding author.

E-mail address: yfduan@seu.edu.cn (Y. Duan).

Nomenclature	
HTEs	hazardous trace elements
APCD	air pollution control device
SCR	selective catalytic reduction
ESP	electrostatic precipitator
WFGD	wet flue gas desulfurization
SEM	scanning electron microscope
EDS	energy dispersive spectrometer
XRD	X-ray power diffraction spectrometry
UBC	unburn carbon content
$R_{furnace}$	mass balance rate of HTEs across the furnace, %
R_{SCR}	mass balance rate of HTEs across SCR, %
R_{ESP}	mass balance rate of HTEs across ESP, %
R_{WFGD}	mass balance rate of HTEs across WFGD, %
$R_{furnace, out}$	mass balance rate of HTEs from feed coal to the outlet of furnace, %
$R_{SCR, out}$	mass balance rate of HTEs from feed coal to the outlet of SCR, %
$R_{ESP, out}$	mass balance rate of HTEs from feed coal to the outlet of ESP, %
$R_{WFGD, out}$	mass balance rate of HTEs from feed coal to the outlet of WFGD, %
$R_{whole system}$	mass balance rate of HTEs across the whole system, %
$HTE_{furnace, out}$, $HTE_{SCR, out}$, $HTE_{ESP, out}$, $HTE_{WFGD, out}$	mass amount of HTEs in flue gas at the outlet of furnace, SCR, ESP and WFGD per unit time, respectively, g/h
$HTE_{SCR, in}$, $HTE_{ESP, in}$, $HTE_{WFGD, in}$	mass amount of HTEs in flue gas at the inlet of SCR, ESP and WFGD per unit time, respectively, g/h
$HTE_{ESP, removal}$, $HTE_{WFGD, removal}$	mass amount of HTEs removed by ESP and WFGD per unit time, respectively, g/h
$HTE_{fresh slurry}$	mass amount of HTEs in the fresh slurry getting in WFGD system per unit time, g/h
$HTE_{DeSO_2 wastewater}$	mass amount of HTEs in the waste water leaving WFGD system per unit time, g/h
CR_{ESP} , CR_{WFGD}	contribution rate of ESP, WFGD for the total flue gas HTEs removal, respectively
$C_{ESP, in}$, $C_{WFGD, in}$	concentration of total HTEs in the flue gas at the inlet of ESP, WFGD, respectively
$C_{ESP, out}$, $C_{WFGD, out}$	concentration of total HTEs in the flue gas at the outlet of ESP, WFGD, respectively
REI	relative enrichment index
$C_{HTEs, ash}$	concentration of HTEs in bottom ash or fly ash, mg/kg
$A_{coal, ad}$	content of ash in feed coal on air-dry basis, %
$C_{HTEs, coal}$	concentration of HTEs in feed coal, mg/kg
EF	emission factor, $g/10^{12} J$
HTE_{stack}	emission amount of flue gas HTEs to atmosphere, g/h
FC	feed coal amount per hour, t/h
$Q_{ar, net}$	low heat value of the feeding coal on as-received basis, MJ/kg

coal-fired flue gas is seldom reported. Although emission characteristic of trace elements has been performed in our previous work [17–19], the field test is far from enough based on diversity of coal, electricity generation capacity, air pollution control devices (APCDs) configuration, etc. In addition, transformation mechanism of HTEs during coal combustion needs further discovery, which is not clear at present.

According to the volatility and migration behavior, HTEs can be classified into three groups, which have been accepted by many researchers [20,21]. Group 1: Hardly volatile elements, which mainly exist in coarse residual particles, like Mn, Ba, etc. Group 2: Moderately volatile elements, which evaporate in furnace and be prone to condense with flue gas cooling, like Zn, Sb, Pb, Cd, As, etc. Group 3: Highly volatile elements, which escape from coal-fired power plant mainly in gaseous form, like Hg, Br, Cl, etc. Some HTEs like Cr, Cu, Co, etc. will be partitioned in different groups in some studies, which may be related to their occurrence in coal, flue gas component, coal type, mineral composition of fly ash and bottom ash [16].

In this study, U.S. EPA Method 29 [22] was used to sample HTEs involving the three groups in the flue gas at inlet or outlet of each APCD in a 320 MW coal-fired power plant simultaneously. Feed coal, fly ash from electrostatic precipitator (ESP), bottom ash, desulfurization fresh slurry and products were also collected at the same time. Scanning electron microscope (SEM), energy dispersive spectrometer (EDS) and X-ray power diffraction (XRD) spectrometry were adopted to characterize the physical and chemical structure characteristic of some solid samples. The main contents include: (1) Mass balance rate and distribution; (2) Flue gas HTEs partitioning and co-removal efficiency; (3) Enrichment characteristic of HTEs in fly ash and bottom ash; (4) Emission to the atmosphere; (5) Transformation mechanism during coal

combustion. The main object is to obtain basic emission characteristic and explore transformation mechanism of HTEs, which can provide a guide for their emission and control in coal-fired power plants.

2. Experimental section

2.1. Plant description and tested coal

The studied 320 MW coal-fired power plant is located in Zhangjiakou city, China. It is installed with selective catalytic reduction (SCR) for NO_x emission control, ESP to collect particulate matter and wet flue gas desulfurization (WFGD) for SO₂ capture.

During the field test, the bituminous coal was used, the proximate and element analysis of which can be found in Table 1. Concentration of Zn, Sb, Pb, Cd, As, Cr, Mn, and Ba in the coal is detected by the inductively coupled plasma-mass spectrometry (ICP-MS) after digestion. Mercury in coal is detected by DMA-80 Direct Mercury Analyzer (Italy Milestone Srl. Company). Concentration of the studied HTEs in coal is listed in Table 2. To better know the concentration level of HTEs, their average value of Chinese and world’s coal is also given in Table 2 [23,24]. It shows that content of Hg, Pb, Cr, Ba in the coal is higher than the average value of Chinese and world’s coal while the concentration of Zn, Sb, Cd is lower. Content of As is higher than the mean value in Chinese coal but lower than that in world’s coal. Concentration of Mn is less than the average value of Chinese coal.

2.2. Sampling and analysis

HTEs in flue gas, which exist in particulate and gaseous form, are

Table 1
Proximate and element analysis of the tested coal.

Proximate analysis (% , air dried basis)				$Q_{net, ar}$ MJ/kg	Element analysis(% , air dried basis)					
M	A	V	FC		C	H	O	N	S	Cl
0.98	28.50	25.42	45.10	19.88	55.52	3.56	9.73	0.82	0.89	0.010

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