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Research article

Migration and emission characteristics of Hg in coal-fired power plant of China with ultra low emission air pollution control devices



Yi Zhang ^{a,b}, Jianping Yang ^a, Xuehai Yu ^{a,c}, Ping Sun ^{a,c}, Yongchun Zhao ^{a,*}, Junying Zhang ^a, Gang Chen ^a, Hong Yao ^a, Chuguang Zheng ^a

^a State Key Laboratory of Coal Combustion, School of Energy and Power Engineering, Huazhong University of Science and Technology, Wuhan 430074, China

^b Sanhe Power Plant Co., Ltd., Hebei, Lanfang 065201, China

^c Shenhua Guohua (Beijing) Electric Power Research Institute Co., Ltd., Beijing 100025, China

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ABSTRACT

The emission and migration characteristics of Hg from an ultra low emission (ULE) coal-fired power plant in China was investigated. During the tests, the flue gas was sampled simultaneously at the inlet and outlet of selective catalytic reduction (SCR) system, low temperature economizer (LTE), electrostatic precipitators (ESP), wet flue gas desulfurization (WFGD), and wet electrostatic precipitators (WESP) by EPA 30B method. The feed coal, lime, limestone slurry, process water, fly ash, bottom ash, gypsum, FGD effluent, and WESP effluent were also sampled. The results showed that Hg concentration in flue gas at the outlet of boilers and stacks was in the range of 4.46–5.17 μ g/m³ and 0.51–1.22 μ g/m³, respectively. The overall gaseous Hg removal efficiencies of SCR + LTE + ESP + WFGD + WESP equipped in the ULE power plant were about 88.5%–89.6%. Mass distribution of Hg in the whole system showed that about 70% of Hg present in solid and liquid combustion products and 30% of Hg emitted into atmosphere. After ULE reformation, the atmospheric emission factor of the power plant is in the range of 0.39–0.81 g/TJ, which is much lower that of power plants before ULE reformation (2.18–2.34 g/TJ). Thus, the ULE reformation for coal-fired plants is beneficial for the reduction of Hg emission to atmosphere.

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1. Introduction

The pollution of mercury (Hg) has attracted worldwide attention in recent years because of its high toxicity, bioaccumulation and worldwide migration [1]. Coal–fired power plants are the main anthropogenic emissions of Hg [1]. The Mercury and Air Toxics Standards (MATS) issued by the US Environmental Protection Agency (EPA) comes into effect for existing electric utility steam generating units (EGUs) on April 16, 2015 [2]. The MATS restricted emission limits of Hg in existing units: 0.013 lb/GWh for non-lignite units and 0.040 lb/GWh for lignite units [3]. China, as the largest coal-consuming country in the world, contributes about 25%–40% of global anthropogenic Hg emissions to the atmosphere every year [4]. The Chinese government issued the latest Emission Standard of Air Pollutants for Thermal Power Plants (GB13223-2011) and the emission limits of Hg is 30 μ g/m³. In April 2013, Chinese government approved the "Minamata Convention", which will further restrict the emission limits.

Hg in the coal combustion flue gas occurs in three forms: elemental mercury (Hg^0), oxidized mercury (Hg^{2+}), and particle-bound mercury

(Hg^P). Hg^p associated with fly ash particles could be captured effectively by the dust control devices like electrostatic precipitator (ESP) and fabric filter (FF). Hg²⁺ could be removed by the wet flue gas desulfurization (WFGD) system because of its water-solubility. However, Hg⁰ is difficult to be removed by existing air pollution control devices (APCDs) because of its insolubility in water and volatility. In recent years, extensive technologies for the reduction of Hg emission have been developed, which mainly focused on the following fields: (i) specific Hg removal technologies, (ii) co-beneficial control by existing APCDs. The specific Hg removal technologies like catalytic oxidation [5–11] and sorbent injection [12–21] could control Hg emission effectively. However, specific Hg control devices need to be installed in power plants, which involves high facility and operation cost. Thus, the efficient and low-cost Hg removal technology is needed for power plants to meet the more stringent emission limits.

Nowadays, conventional APCDs like SCR, ESP/FF, WFGD have been installed widely in power plants with an intention to reduce the emission of NO_x , particulates, and SO_x . Besides the targeted pollutants, these APCDs also significantly affect the emission and speciation of Hg. Thus, the cobenefit Hg removal by the existing APCDs is a promissing technology for Hg reduction. The US Information Collection Request (ICR) investigated the cobenefit Hg removal performance of PM, NO_x ,



^{*} Corresponding author. *E-mail address:* yczhao@hust.edu.cn (Y. Zhao).

and SO₂ control devices from typical coal-fired power plants [22]. The effects of coal rank, combustion conditions and APCDs configuration on the degree of cobenefit Hg removal were studied. Meij et al. carried out extensive field tests at Dutch coal-fired power plants over a period of 25 years [23]. The results showed that the following percentages of Hg abatement could be achieved with the presence of APCDs: (i) cold side ESP 50%, (ii) cold side ESP + wet FGD 75%, (iii) cold side $ESP + wet FGD + DENO_x$ (SCR) up to 90%. Pudasainee et al. [24,25] investigated the Hg emissions concentration at inlet of APCDs and stack. On average, Hg emission concentrations are in the range of 16.3- $2.7 \,\mu\text{g/m}^{-3}$, $2.4-1.1 \,\mu\text{g/m}^{-3}$, $3.1-0.7 \,\mu\text{g/m}^{-3}$ from power plants burned anthracite with the equipment of CS-ESP, power plants burned bituminous with CS-ESP and WFGD, and power plants burned bituminous with SCR + CS-ESP + WFGD, respectively. The studies of Ochoa-González et al. [26] and Cordoba et al. [27] showed that the cobenefit Hg removal efficiency by WFGD is highly influenced by the Hg concentration entering the system, which is determined by the amount of Hg fed to the boiler and the cobenefit removal efficiency of the particle control device. The working conditions in the system especially the liquid/ gas (L/G) ratio are important for the removal of Hg²⁺ because of the mass transfer and gas residence time in the scrubber. Moreover, fly ashes and gypsum are the main wastes produced in coal-fired power plants, which may be disposed of in landfills or deposit outdoor. Thus, the leach of Hg from fly ashes and gypsum might also cause potential risk. Rallo et al. [28] studied the leaching of mercury in fly ashes and gypsum from a Dutch co-combustion power station equipped with SCR. The results showed that the fly ash and gypsum could be acceptable at landfills as non-hazardous waste. Thus, the cobenefit Hg removal by existing APCDs is a promising technology for the reduction of Hg emission in power plants.

Recently, the Chinese government has issued the "Reformation and Upgrading Action Plan for Coal Energy Conservation and Emission Reduction", where the emission limit of PM, NO_{x_0} and SO_2 in some provinces like Jiangsu and Zhejiang was 5 mg/m³, 35 mg/m³, and 50 mg/m³, respectively. To meet the ULE standard, the SCR, ESP, WFGD system have been transformed in several power plants. Particularly, low temperature economizer (LTE) was installed at the inlet of ESP to collect the heat generated from flue gas, and wet electrostatic precipitator (WESP) was applied to capture ultrafine particles and aerosols. To the author's knowledge, little literatures were reported about the emission characteristics of Hg in ULE power plants.

In the present study, the field tests of Hg emission were studied in three utility boilers equipped with ULE APCDs. The Hg concentration and speciation distribution was simultaneously measured at the inlet and outlet of SCR, LTE, ESP, WFGD, WESP, and stack emission. The feed coal, lime, limestone slurry, WESP process water, fly ash, bottom ash, gypsum, FGD effluent, WESP effluent were collected during the sampling period. The migration and emission of Hg across each APCDs as well as the co-benefit Hg removal efficiency of APCDs were studied. The Hg mass balances in the whole system, Hg partition in combustion by-products and atmospheric Hg emission factor were discussed.

2. Experimental section

2.1. Introduction of utility boiler

The emission and speciation of Hg were studied in three ULE utility boilers. The tested boilers are all pulverized coal boilers with electricity generation capacity of 330 MW (boiler #1), 330 MW (boiler #2), and 300 MW (boiler #4), respsctively. The APCDs installed in boiler #1 and boiler #4 include SCR for NO_x control, ESP for particulate removal, WFGD for SO₂ removal, and WESP for ultrafine particles or aerosols removal. The APCDs installed in boiler #2 include SCR, ESP, and WFGD, whereas WESP is not installed. In addition to the APCDs, LTEs are all installed at the inlet of each ESP. Fig. 1 shows the schematic configuration of the APCDs, the LTE, and the sampling points. During the tests, the load of each boiler is 100% output. The properties of the feed coal during the sampling process are shown in Table 1.

2.2. Sampling procedures and analytical method

The flue gas was sampled simultaneously at the inlet and outlet of each APCDs by EPA 30B method. Traps (Ohio Lumex 30B Carbon Traps) filled with KCl and activated carbon were employed to capture the gaseous Hg^{2+} and Hg^{0} , respectively. In each test, two parallel traps were used to ensure the reliability. The traps were maintained at 120 °C to avoid moisture and Hg condensation on the lines. A gas flow rate of 500 mL·min⁻¹ was maintained for 60 min for each trap. Coal, lime, limestone slurry, WESP process water, fly ash, bottom ash, gyp-sum, FGD effluent, WESP effluent were also collected during the sampling period. Hg content in traps and other samples were analyzed by RA–915M mercury analyzer with PYRO–915 + pyrolysis equipment (Ohio Lumex Company).

 SO_3 in flue gas was determined by EPA method 8. Briefly, the flue gas was extracted isokinetically from the gas duct. Four impingers were equipped in the sampling system, where the first one was empty in case of suck-back; the second and third one was placed 100 mL of 80% isopropanol; the fourth one was placed desiccant to remove the water before entering into aspirator pump. The second and third impingers were placed into ice. In each test, 500 L flue gas sample was extracted. After sampling, the reagent in the second and third impingers were collected and the SO_4^2 ⁻ was analyzed by ion chromatography (IC). The SO_3 concentration in flue gas was calculated by the SO_4^2 ⁻ content of in reagent. The flue gas temperatures at each sampling points were measured by platinum-rhodium thermocouple.



Fig. 1. Schematic configuration of the APCDs and the sampling points.

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