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Study on the mercury emission and transformation in an ultra-low emission coal-fired power plant

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HIGHLIGHTS

- The distribution, removal and emission characteristic of mercury in the ULE power plant were reported.
- Mercury transformation process was discussed systematically for each APCD by new method.
- Mercury in bottom and ESP ash had no effects on the soil.
- Attention should be given to the gypsum disposal, especially when using thermal treatment.

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ABSTRACT

Mercury emissions from a Chinese ultra-low emission (ULE) coal-fired power plant burning a low chlorine and sulfur bituminous coal was investigated by using the Ontario Hydro method (OHM). A temperature-programmed decomposition desorption (TPDD) instrument, scanning electron microscope (SEM), and X-ray power diffraction spectrometry (XRD) were used to identify the mercury species, apparent morphology, and chemical composition of the coal and combustion by-products. As measured by the OHM, the elemental mercury emitted from the stack is the largest proportion of the total mercury (about 38.95%), almost 13 times more than the oxidized mercury. Mercury in bottom ash was found to be only 0.07% of the incoming total mercury. The elemental mercury was found to oxidize across the SCR, with 45.47% converted to oxidized mercury. This increase in mercury oxidation is significant, as it was shown to be removed by the WFGD system. The total mercury removed across all APCDs in order of most to least was WFGD > ESP > WESP. The WESP was shown to further remove mercury, whose removal rate is about 13.63%, resulting in lower mercury stack emissions. For this plant arrangement, the mercury emission factor was calculated to be 1.56 $g/10^{12}$ J, less than the mean value of Chinese plants. The mercury content in the gypsum is higher than the limit (0.50 mg/kg) while bottom and ESP ash were shown to have no effect on the soil. Further investigation of thermal treatment on mercury in gypsum is needed before definitive conclusions on disposal can be drawn. Concentration of mercury measured in the WFGD and WESP waste water is higher than the limit (0.001 mg/L), and more importance should be given on the waste water from WFGD.

1. Introduction

Mercury is well known as the toxic pollutant, which can damage human health and the environment because of its long lifetime, volatility, bioaccumulation, high toxicity and long-distance transboundary transportation in the atmosphere [1–3]. It can be found in the environment worldwide, even in the area far away from any emission source. Mercury has been regarded as a global pollutant by the United National Environment Programme (UNEP) [4]. Coal combustion for electricity and heat generation is the dominant source for anthropogenic mercury emissions [5,6]. It is estimated that coal-fired power plants accounts for 26% global anthropogenic mercury emission to the atmosphere [7]. In China,

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Nomenclature

ULE	ultra-low emission
OHM	Ontario Hydro method
SEM	scanning electron microscope
XRD	X-ray power diffraction spectrometry
SCR	selective catalytic reduction
WFGD	wet flue gas desulfurization
APCDs	air pollution control devices
ESP	electrostatic precipitator
MEF	mercury emission factor
TPDD	temperature-programmed decomposition desorption
APCD-x	individual air pollution control device or the whole system, in which x = SCR, ESP, WFGD, WESP, or SCR + ESP + WFGD + WESP
Hg^y	gaseous mercury in the flue gas including Hg^0, Hg^{2+}, Hg^p, Hg^T ($Hg^T = Hg^0 + Hg^{2+} + Hg^p$)
$Hg^y_{APCD\text{-}x,in}$	concentration of the mercury Hg^y at the inlet of the APCD-x.
$Hg^y_{APCD\text{-}x,out}$	concentration of the mercury Hg^y at the outlet of the APCD-x.
WESP	wet electrostatic precipitator
Hg^0	elemental mercury
Hg^{2+}	oxidized mercury
Hg^p	particle-bound mercury
UBC	unburned carbon
XAFS	X-ray absorption fine structure
$Hg^0_{in,SCR}$	Hg^0 concentration at inlet of SCR
$Hg^0_{out,SCR}$	Hg^0 concentration at outlet of SCR
UNEP	United National Environment Programme

over 2000 coal-fired power plants result in the largest single atmospheric mercury emitter in the world.

In general, mercury emitted from coal combustion exists in three primary forms in the flue gas, elemental mercury (Hg^0), oxidized mercury (Hg^{2+}) and particle-bound mercury (Hg^p) [8]. With high volatility and insolubility in water, Hg^0 can stay in the atmosphere for 0.5–2 years [8,9]. Compared to Hg^0, Hg^{2+} and Hg^p are more chemically reactive, less volatile, and water soluble, which make them be with much shorter lifetime (lasting for several days to a few weeks) and much fast deposition in both dry and wet processes [9–11]. Therefore, Hg^{2+} and Hg^p can be easily removed by the wet scrubber like wet flue gas desulfurization (WFGD) and particle control system like electrostatic precipitator (ESP) or fabric filter (FF) [12,13]. The characteristics of the Hg^0 make it difficult to be removed by the air pollution control device (APCD) in coal-fired power plants.

In recent years, lots of researchers around the world have conducted field tests on the synergistic effects of mercury removal across the APCD in the coal-fired power plants [14]. Bilirgen [15] studied the mercury capture at an American 810 MW coal-fired power plants with a low-NO_x system + WFGD, finding that combination of optimal boiler control settings for reduced Hg emission operation resulted in a 34.5% reduction in the total mercury at the stack. Zhang et al. [16] investigated the mercury emissions from six coal-fired power plants with ESP, ESP+FGD, FF, ESP in China, concluding that about 0.02%–1.2% of the mercury remained in the bottom ash while most of the mercury was emitted to the atmosphere for the pulverized coal boiler. Shah et al. [17] conducted the mercury speciation at five different coal-fired power plants with ESP or FF across Australia. Results showed that the total mercury concentration emitted from the plants was in the range of 1.9–5.6 μg/Nm^3 while Hg^p accounted for a low proportion of 0.3–3.7%. Pudasainee et al. [18] studied the mercury emission trend influenced by stringent air pollutants regulation for coal-fired power plants with ESP, ESP+WFGD, SCR (selective catalytic reduction) + ESP + WFGD in Korea. The mercury emission concentrations were 16.3–2.7 μg/Sm^3, 2.4–1.1 μg/Sm^3 and 3.1–0.7 μg/Sm^3, respectively. Yokoyama et al. [19] conducted the mercury emission at a 700 MW coal-fired power plant with a low-NO_x burner + SCR + ESP + WFGD in Japan. Results showed that the relative distribution of mercury in ESP, FGD and stack ranged from 8.3 to 55.2%, 13.3 to 69.2% and 12.2% to 44.4%, respectively. However, researches about the mercury transformation process across APCD and the effects of mercury in coal combustion by-products on the environment are seldom reported.

Recently, with much attention paid to the pollutant emitted from the coal-fired power plants, Chinese government has put forward the ultra-low emission (ULE) for thermal power units. It requires the emission limit values of dust, NO_x, and SO_2 are 5 mg/m^3, 35 mg/m^3 and 50 mg/m^3, respectively in some provinces including Jiangsu and Zhejiang province [20,21]. The mercury emission characteristics in the ULE coal-fired power plants are seldom investigated in the world [22]. The temperature-programmed decomposition desorption (TPDD) has been recognized as an effective method for identifying mercury species in solids, which is based on different decomposition temperature of various mercury forms [23–25]. In this study, the field test about mercury emission and transformation was conducted on an ULE demonstration coal-fired power plant equipped with SCR + ESP + WFGD + WESP (wet electrostatic precipitator) in China. The Ontario Hydro method (OHM) [26], the internationally recognized standard method, was used for the flue gas mercury sampling. TPDD was applied to the identification of mercury compounds in the coal and combustion by-products. The scanning electron microscope (SEM) and X-ray power diffraction spectrometry (XRD) were used to characterize the apparent morphology and chemical composition of the coal and combustion by-products. The main purpose includes the following: (1) mercury balance and its distribution in the ULE power plant, (2) mercury transformation process across the whole system, (3) mercury removal rate of the APCD and mercury emission factor, (4) contamination of mercury on the environment. The results can update the mercury emission data in the Chinese coal-fired power plants, which provides the theoretical basis and guidance for mercury control.

2. Experimental

2.1. Utility boiler

The ULE demonstration coal-fired power plant was a tangentially fired, pulverized coal boiler with electricity generation capacity of 660 MW. To achieve the ultra-low emissions, this power plant was installed with SCR capable of achieving NO_x emission conversion rate of about 80.86–82.87%, ESP used for PM removal, WFGD with SO_2 removal efficiency of 96%, and WESP for ultrafine particles or aerosols removal. The SCR catalyst used in this power plant was honeycomb with main component of V_2O_5-WO_3/TiO_2, which was arranged in high dust way. The WFGD uses the counter-current spray tower based on limestone-gypsum method, which is consisted of circulating slurry pump, oxidation zone, absorption