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Study on the Hg emission and migration characteristics in coal-fired power plant of China with an ammonia desulfurization process

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ABSTRACT

Migration and emission characteristics of mercury in coal-fired power plant with an ammonia-based wet flue gas desulfurization (Am-based WFGD) process were studied based on the Ontario Hydro Method (OHM). The temperature-programmed desorption (TPD) method was used to identify the mercury species enrichment mechanisms in fly ash and the by-products of desulfurization. The SCR has a 95.41% mercury oxidation efficiency attributed to the high chlorine content in feed coal. SCR + ESP + WFGD combination has 94.15% gaseous mercury removal efficiency. Mercury distribution analysis showed that most of incoming mercury was deposited in solid (76.46%), and only 4.94% gaseous mercury was emitted to atmosphere. The TPD results indicate that $HgCl_2$ and HgO are the dominant mercury species in fly ash while mercury in WFGD products exists in $HgCl_2$ form. The re-emission of mercury during ammonia desulfurization did not occur due to the presence of forced oxidation process, while mercury release from desulfurization product was monitored suggesting the mercury re-emission from the ammonium sulfate separate and drying system could not be ignored. The mercury emission factor of the coal-fired power plant is calculated as 0.319 g/10^{12} J, less than the mean value of Chinese power plant. With the equipment of Am-based WFGD, the mercury content in ammonium sulfate is 1.126 mg/kg, therefore the environmental influence from ammonium should be taken into account.

1. Introduction

Mercury and its compounds are considered as the dangerous heavy metal to both humans and the ecosystem because of the properties of high toxicity, bioaccumulation, and ability to transmit for long time and long distance [1]. Mercury pollution has attracted global attention since Hg emission had continuously increased with a certain annual rate [2,3]. Fossil fuel production and combustion, nonferrous metal smelting, gold smelting, Hg mining, iron and steel production, and cement production are identified as important anthropogenic sources of airborne Hg in the previous studies [4–6]. Among those, coal combustion is considered to be the largest anthropogenic mercury emission source, accounting for 23% global anthropogenic mercury emission to the atmosphere [7]. China is the largest coal consumer around the world because of its unique energy consumption structure [8]. The Hg content of most Chinese coals is between 0.1 and 0.3 mg/kg. The average Hg content of Chinese coals is 0.19 mg/kg [9], which means a huge amount of anthropogenic mercury is emitted in the process of coal utilization.

Gaseous mercury in coal-fired flue gas mainly exist in three forms: elemental (Hg^0), oxidized (Hg^{2+}) and particulate-bound mercury (Hg^p) [10]. Hg^0 with high volatility, chemical inertness and low water solubility [11], is recognized as the most abundant but hard to removal form in the atmosphere with residence time of 0.5–2 years [6]. The Hg^0 emission control by conventional pollution control systems is difficult due to its physical and chemical inertia. However, the reactivity of gaseous Hg^p and Hg^{2+} are relatively higher than Hg^0 that can be easily captured by dust collector and wet scrubber in power plants [12]. The proportions of Hg^0, Hg^{2+} and Hg^p in the flue gas released from a pulverized-coal (PC) boiler, averaging about 56%, 34% and 10%, respectively [13]. Activated carbon injection (ACI) method is certified as an effective mercury removal technology. However, it is difficult to reach large-scale industrial application due to high cost. Therefore, making the most use of existing air pollution control devices (APCDs) to control mercury in coal-fired flue gas is known as reliable alternative method in China. According to the on-site Hg sampling results in coal-fired power plants and industrial boilers, existing APCDs possess a synergistic removal effect on Hg. The total mercury removal efficiency of electrostatic precipitator (ESP) during on-site measurement has shown an average mercury removal efficiency of 29% with a large range of 1–74% [13]. The average removal rate with the combination of ESP and wet flue gas desulfurization (WFGD) was 74% [14]. While power plant

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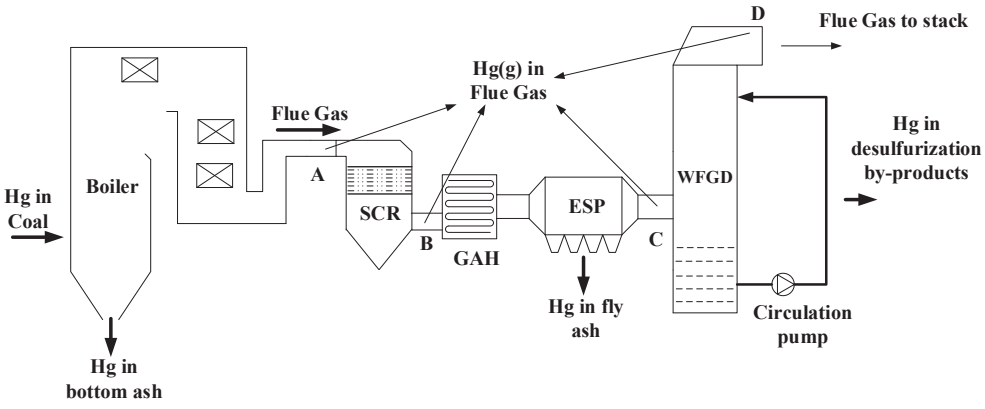

Fig. 1. Simultaneous sampling locations in the power plant.

with the combination of selective catalytic reduction (SCR), ESP, fabric filter (FF) and WFGD was reported to achieve an 87.6% of mercury removal efficiency [15]. The power plant which adopts ultra-low emission technology might further increase the mercury removal rate [16]. It shows that mercury can be co-beneficially controlled through reasonable arrangement of APCDs. The previous mercury field measurement conducted on an ultra-low emission coal-fired power plant confirmed that the total mercury removed across all APCDs in order of most to least was WFGD > ESP > WESP [17].

WFGD as an efficient desulfurization technology, has been applied worldwide [18], and is also the crucial step in the co-benefit mercury control technologies in the coal-fired power plants [13]. However, the traditional limestone wet flue gas desulfurization is facing the plight of Hg^0 re-emission [19,20]. Recently, owing to its lower investment, higher desulfurization efficiency, no secondary pollution, and useful byproducts comparing to others flue gas desulfurization (FGD) [21], ammonia-based desulfurization (Am-based WFGD) has aroused widespread attention among China [22], but the aerosol problems arising in the desulfurization process have been reported frequently, being the major challenge in the development of this technology [23]. Furthermore, little researches on subsequent pollution of desulfurization byproducts, mercury migration and emissions in the Am-based WFGD process were conducted. In this study, the field sampling test about mercury emission and migration were conducted on a coal-fired power plant equipped with SCR, ESP and Am-based WFGD in China. The Ontario-Hydro method (OHM) was applied for the mercury field sampling. OHM is considered to be the standard method for vapor phase mercury (Hg(g)) speciation measurement in coal-derived flue gas [24] and widely used to quantify the mercury concentration of utility power plants [1,11,25]. This paper studied the concentration of different forms of mercury in the gas phase and the solid discharge, the mercury mass balance was then calculated. The mercury distribution and speciation as well as the synergistic removal effect over APCDs in this coal-fired power plant were also analyzed. The temperature programmed desorption (TPD) technique was used to explore the enriched form of mercury in solid products. Moreover, the mercury emission factor (MEF) was calculated to assess the environmental pollution of mercury emitted from this plant. The results will provide assistance for mercury emission control and the corresponding environmental issue management and a good understanding on the Hg emission and migration characteristics in the coal-fired power plant with an ammonia desulfurization process.

2. Experimental

2.1. Site description and configuration

On-site tests were conducted on a pulverized coal power plant which located in China. In order to meet the pollutant emission standard of China [26], the power plant was installed with SCR which using traditional V_2O_5-WO_3/TiO_2 catalyst, ESP for particle removal, and ammonia-based wet desulfurization process for controlling sulfur dioxide emissions. A schematic diagram of the sampling locations is shown in Fig. 1. Flue gas sampling were carried out simultaneously at four sites: inlet and outlet of SCR, the front and rear of WFGD tower. In addition, the furnace coal, bottom ash, fly ash, and diverse concentrations of absorption solutions from the different locations of desulfurization tower (marked as AS1 and AS2 in this paper, and the concentration of ammonium sulfate in AS2 is higher than that in AS1) were also collected simultaneously during the sampling period. The desulfurization slurry was recycled through a circulation pump so as to improve desulfurization efficiency. The desulfurization by-product was continuously produced through the separation system. The air is continuously carried into the desulfurization tower through a fan so that

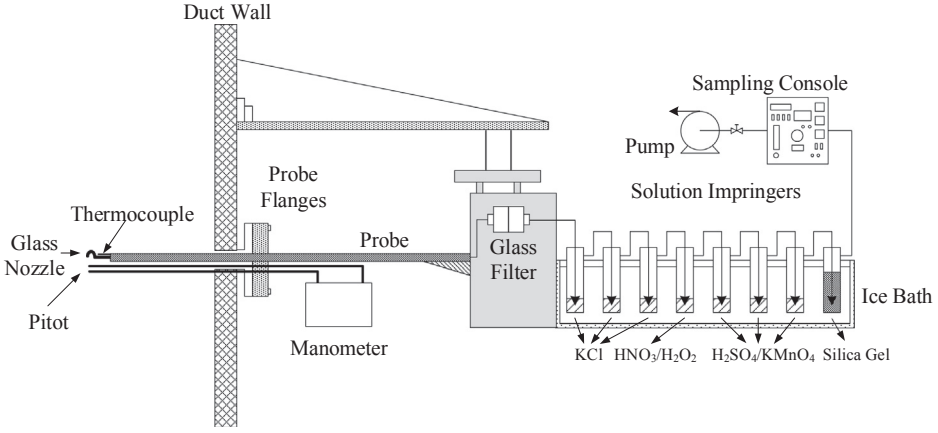

Fig. 2. Schematic of the OHM mercury sampling device.