



## Effects of existing energy saving and air pollution control devices on mercury removal in coal-fired power plants



Zi-Jian Zhou<sup>a</sup>, Xiao-Wei Liu<sup>a,\*</sup>, Bo Zhao<sup>a</sup>, Zhen-Guo Chen<sup>a</sup>, Hai-Zhong Shao<sup>a</sup>, Le-Le Wang<sup>b</sup>, Ming-Hou Xu<sup>a,\*\*</sup>

<sup>a</sup> State Key Laboratory of Coal Combustion, Huazhong University of Science and Technology, Wuhan 430074, China

<sup>b</sup> Xi'an Thermal Power Research Institute Co., Ltd. Suzhou Branch, Suzhou 215011, China

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### ABSTRACT

The impacts of a low temperature economizer (LTE) on mercury removal across an electrostatic precipitator and influence of load variation on mercury conversion over selective catalytic reduction (SCR) catalysts were determined at two coal-fired boilers. When the LTE was on, the total and elemental mercury removal efficiency increased by 42.87% and 18.85%, respectively, due to the improvement of adsorption and oxidation capacity of the fly ash at lower temperature. Mercury speciation at the inlet and outlet of the SCR system were analyzed, and the impacts of load variation and catalyst aging on Hg<sup>0</sup> conversion were discussed. The variable loads resulted in simultaneous changes of the gas hourly space velocity, the ambient temperature, and the oxygen content. The results showed the load ratio was significant for Hg<sup>0</sup> conversion by the SCR catalysts and load reduction benefitted Hg<sup>0</sup> conversion. When the load ratios were 100%, 75% and 60%, the Hg<sup>0</sup> conversion were 61.78%, 65.71% and 72.12%, respectively. Moreover, Hg<sup>0</sup> conversion was more significantly affected by the catalyst aging than NO<sub>x</sub> reduction. Among the three factors, the most important one is the flue gas temperature based on the grey relational analysis.

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### 1. Introduction

The mercury (Hg) emitted during coal combustion has become an increasing concern due to its high toxicity, bioaccumulation and worldwide migration. Coal-fired power plants are the main sources responsible for the anthropogenic mercury emissions into the atmosphere. The anthropogenic mercury emission is a serious concern in both developing and developed countries [1]. In China, coal contributes about 70% of the total primary energy consumption [2], and a large amount of Hg is released during coal combustion [3]. In 2010, about 576 tons of Hg were emitted into the atmosphere, accounting for about 35% of the global anthropogenic mercury emissions and about 97 tons of mercury were emitted from coal-fired power plants [4]. In the United States, Hg has been listed as a hazardous pollutant under Title III of the Clean Air Act Amendments (CAAA) since 1990 [5]. However, mercury emissions from power plants have not been controlled efficiently until recent years. In 2011, the Environmental Protection Agency (EPA) has established the first ever national standards to reduce mercury emission from coal-fired power plants [6].

Hg in the flue gas occurs in three primary forms [7]: elemental mercury (Hg<sup>0</sup>), oxidized mercury (Hg<sup>2+</sup>) and particle-bond mercury (Hg<sup>p</sup>). Different species of mercury have different physical and chemical properties [8]. Hg<sup>p</sup> is associated with the fly ash particles and is in the solid form. Hg<sup>p</sup> can be removed easily by the electrostatic precipitator (ESP) used for dust removal. Hg<sup>2+</sup> is water-soluble and can be captured by the wet flue gas desulfurization (WFGD) scrubbers designed for SO<sub>2</sub> removal. However, Hg<sup>0</sup> is difficult to capture because it is extremely volatile and water-insoluble and remains in the atmosphere for a long time [8]. Fortunately, in recent years, it is observed that the mercury emissions have decreased in power plants equipped with SCR systems [9], due to the promotion of Hg<sup>0</sup> conversion to Hg<sup>2+</sup> by the commercial SCR catalyst (V<sub>2</sub>O<sub>5</sub>-WO<sub>3</sub>(MoO<sub>3</sub>)/TiO<sub>2</sub>) [10]. As stated above, the existing APCDs, such as the ESP and SCR system, in the coal-fired power plants can realize the benefits of Hg capture [11]. Therefore, the performances of mercury removal across the ESP and the SCR are discussed as follows.

ESP is one of the most common particulate matter control devices. In recent years, mercury capture via an ESP has attracted increasing attention. The flue gas temperature, flue gas compositions, and characteristics of fly ash all influence the performance of mercury removal across the ESP [12]. Previous studies [13,14] have shown that Hg removal efficiency across an ESP is higher in plants equipped with cold-side ESPs than that with hot-side ESPs and this result indicates that lower temperature benefits the mercury removal via an ESP. The acid gases in the flue gas, such as the HCl and SO<sub>2</sub>, always have great impacts on the distribution of the mercury [13]. In a field test, Lu et al. [15] found

\* Correspondence to: X.-W. Liu, 1037 Luoyu Road, Wuhan 430074, China. Tel.: +86 27 87544779 8207; fax: +86 27 87545526.

\*\* Correspondence to: M.-H. Xu, 1037 Luoyu Road, Wuhan 430074, China. Tel.: +86 27 87544779 8309; fax: +86 27 87545526.

E-mail addresses: [xwliu@hust.edu.cn](mailto:xwliu@hust.edu.cn) (X.-W. Liu), [mhXu@hust.edu.cn](mailto:mhXu@hust.edu.cn) (M.-H. Xu).

that the concentration of the  $\text{Hg}^0$  in flue gas decreased and the  $\text{Hg}^{2+}$  concentration increased after the ESP because the  $\text{Hg}^0$  may be oxidized by the acid gases with the help of the fly ash. The ICR data show that the average mercury removal efficiency by the ESP is ~27%, but the variation is 7–92% [16]. The different efficiencies may be caused by the unburned carbon (UBC) content in the fly ash, and the efficiency increases as the UBC content increases [14]. The UBC acts as the adsorbent for gaseous mercury capture. Hence, taking the advantage of the UBC in the fly ash seems a potential and promising way to remove the gaseous mercury efficiently across an ESP. According to literature [13–17], the ESP mainly removes the  $\text{Hg}^0$  and the performance of gaseous Hg removal via the ESP is limited. However, some new technologies to improve the dust removal efficiency or boiler thermal efficiency may also affect the mercury removal performance through an ESP. For example, to improve the thermal efficiency of the boilers, a low temperature economizer (LTE), an energy saving device, is introduced into the low pressure heating system to make use of the heat of flue gas, which can heat the return water from the steam turbine. The dust removal efficiency can be also improved. The LTE is always placed at the inlet of the ESP. As an energy saving device, the LTE itself cannot reduce any pollutants directly. When the LTE is running, the temperature of flue gas flowing across the ESP will be reduced to some extent. Thus, the characteristics of the particles and properties of flue gas may change significantly, such as the fly ash resistivity, the viscosity and velocity of flue gas. However, there is no available studies on the influences of the LTE on the Hg removal across the ESP. When the LTE is on, the effect it will exert on the performance of the mercury removal by an ESP is still unknown. Furthermore, even if mercury removal across the ESP can be promoted by the LTE, the underlying causes also need to be further investigated. It is also unknown if the existence of an LTE has an effect on the speciation of the mercury in the flue gas at the ESP. All these issues need to be further studied.

The performance of  $\text{Hg}^0$  conversion across the SCR catalysts in coal-fired power plants also receives much attention since the oxidized mercury can be captured by the WFGD easily. Chlorine compounds, like HCl, serve as oxidizers for mercury conversion [17] in the oxidation process. In addition to the chlorine compounds, the flue gas temperature, the gas hourly space velocity (GHSV) and the flue gas compositions [18] can affect  $\text{Hg}^0$  conversion across the SCR catalysts. Unfortunately, these factors tend to fluctuate frequently when the boilers are run in a routine mode. Senior et al. [19] investigated the effect of temperature on Hg conversion and found that at typical operating temperatures (315 °C–345 °C),  $\text{Hg}^0$  conversion was as high as 60–80%. The mercury conversion was high within a relatively wide temperature window (300 °C–400 °C), and an optimal temperature exists for the highest efficiency. When the temperature is lower or higher than the optimal temperature, the efficiency will decrease [20–22]. The GHSV always has an inverse relationship with the conversion in field tests [17,23]. In a study by Machalek et al. [24], the GHSV increased from 3000  $\text{h}^{-1}$  to 7800  $\text{h}^{-1}$ , while the conversion decreased from 40% to 5%, illustrating the strong dependence of efficiency on the GHSV. Although the performance of  $\text{Hg}^0$  oxidation by the SCR catalysts has been reported, most studies have only discussed a single factor on the mercury conversion. With the boiler run in normal mode, load variation occurs frequently, resulting in simultaneous changes of many variables, including GHSV, flue gas temperature and the  $\text{O}_2$  content. In the process, the variations of some factors benefit the mercury conversion while the other ones inhibit conversion of mercury. However, the dominant factors in the conversion process are still unknown. Hence, the impacts of simultaneous variables of the GHSV, the temperature, the  $\text{O}_2$  content on the mercury conversion should be analyzed.

As stated above, the SCR and ESP have positive effects on the mercury removal in a coal-fired power plant. However, the mercury removal by the APCDs is a complex process that is affected by many factors. In the present work, we will discuss some new aspects in the process of mercury removal across the SCR and ESP, respectively. Firstly, in order to

clarify the effects of LTE on the mercury capture across an ESP, a series of tests will be performed in a coal-fired boiler (boiler #1) equipped with LTEs. The mercury concentrations and speciation before and after the ESP with the LTE on or off will be measured. Secondly, the influences of load variation on the  $\text{Hg}^0$  conversion across the SCR catalysts were studied, since the load variation resulted in the simultaneous changes of the GHSV, the flue gas temperature and the  $\text{O}_2$  content. The mercury concentrations and speciation are measured when the load ratio are 60%, 75% and 100% in another coal-fired boiler (boiler #2). The coal and fly ashes were collected and analyzed in the laboratory. The flue gas compositions before and after the ESP and SCR were also measured.

Furthermore, a quantitative analysis was done to determine the significance level of the aforementioned factors on the performance of mercury removal across the SCR catalysts. In the present work, the results will have significance for the mercury removal in coal-fired power plants. Besides, a new way was provided to realize the energy-saving and mercury emission control simultaneously. In addition, the study will also improve understanding of the simultaneous removal of mercury and  $\text{NO}_x$  across the SCR catalysts. The results can also provide support for making pollutant emissions regulations.

## 2. Material and methods

### 2.1. Introduction of the boilers and sampling sites

Both of the two boilers (boiler #1 and boiler #2) are 660 MW ultra-supercritical boilers. The two steam turbines have a main steam pressure of 27.4 MPa with an outlet steam temperature of 605 °C. The two boilers are single furnace, ultra-supercritical [1]boilers. The boiler maximum continuous ratings (BMCR) are 2060 t/h and 2098 t/h for boiler #1 and boiler #2, respectively. Both of the excess air factors measured at the outlet of the two furnaces are 1.18. Boiler #1, has a width of 22162.4 mm, depth of 15456.8 mm and the elevation of the main girder bottom is 84,900 mm. For boiler #2, the width of the boiler is 22162.4 mm, the depth is 16,640 mm and the elevation of the main girder bottom is 85,100 mm. The flue gas temperature tested at the furnace outlet of boiler #1 and boiler #2 were 1039 °C and 1027 °C, respectively. The coal feed rates, flow rates, excess air factors, and flue gas temperatures of different sites are listed in Table 1.

Both of the two boilers are equipped with common APCDs, including an SCR system, a cold-side ESP and a WFGD. In addition to the APCDs, boiler #1 has an energy saving device, a low-temperature economizer (LTE), at the inlet of the ESP. Fig. 1 shows the location of the APCDs, the sampling sites and the location of the LTE.

SCR catalysts with a honeycomb structure were used in the two boilers. The SCR catalysts were based on  $\text{V}_2\text{O}_5$  and  $\text{WO}_3$  and dispersed on the  $\text{TiO}_2$  carrier and produced by a Chinese catalyst factory with the same chemical compositions. For the fresh catalysts, the  $\text{V}_2\text{O}_5$  content is generally low (2.15 wt%), and  $\text{WO}_3$  is employed in larger amounts (7.14 wt%). The catalysts in boiler #1 have been in service for approximately 35,000 h, while those in boiler #2 have been in service for less than 3000 h. Three layers of catalysts were installed, and the third layer considered a spare.

The location of the LTE is shown in Fig. 1 at the inlet of the ESP. When the LTE is on, the standard coal consumption rate decreases by approximately 2.0 g/kW · h. Moreover, the LTE also helps raise dust removal efficiency of ESP.

At boiler #1, the sampling sites were at the inlets and outlets of the ESP and SCR system, respectively. The inlet of the ESP is just before the LTE. At boiler #2, the sampling sites were located before and after the SCR system. The operating conditions are listed in Table 2. During the experiments, boiler #1 maintained nearly a full load, and the LTE was only operated as needed. The load of boiler #2 slightly varied, but fluctuated less than 5% during all the tests.

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