



## Co-effects of sulfur dioxide load and oxidation air on mercury re-emission in forced-oxidation limestone flue gas desulfurization wet scrubber

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### HIGHLIGHTS

- ▶ We investigated FGD Hg<sup>0</sup> re-emission under full-scale operation conditions.
- ▶ Continuous Hg emission monitoring data and plant operation information were analyzed.
- ▶ Both oxidation air flow rate and sulfur load in FGD slurry showed significant effects.
- ▶ High Hg<sup>0</sup> re-emission occurred when operating under unfavorable sulfur to air ratio.
- ▶ Adjusting oxidation air operation might be an economic approach in controlling Hg<sup>0</sup> re-emission.

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### ABSTRACT

Re-emission of Hg<sup>0</sup> refers to the reduction of oxidized mercury (Hg<sup>2+</sup>) to the elemental mercury (Hg<sup>0</sup>) in a wet flue gas desulfurization process (WFGD), resulting Hg<sup>0</sup> re-emits to the flue gas and increasing mercury emission for a coal combustion power plant. The correlations between re-emission of Hg<sup>0</sup> and operation parameters of WFGD process, i.e., sulfur dioxide removal, oxidation air flow, and pH of FGD slurry, were investigated with mercury field data collected from two coal combustion power plants (Plant C and Plant O). At Units 31/32 of Plant C, Coupling mercury results and the plant information (PI) data suggested that an increase of the oxidation air flow in the WFGD effectively inhibited the re-emission of Hg<sup>0</sup>, and consequently increased the flue gas mercury removal efficiency from 63% to over 92%. But at Plant O, the increase of the oxidation air flow can sometimes increase Hg<sup>0</sup> re-emission. It was further found that the re-emission of Hg<sup>0</sup> at Plant O was strongly correlated not only to the flow rate of the oxidation air in the forced-oxidation operation conditions, but also to the amount of sulfur dioxide removed from the flue gas by the FGD scrubber (i.e., the sulfur dioxide load in the scrubber). Possible mechanisms were proposed.

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### 1. Introduction

Mercury is well-known as one of the most toxic metals that can be a threat to the health of people and wildlife at low concentrations. Approximately 50 tons of mercury is emitted annually by the utility industry in America and more than tripled in other developing countries (i.e. China) as a result of coal use [1,2]. The coal combustion energy generation utility has been identified as

the largest anthropogenic mercury pollution source [1,2]. On March 15, 2005, the Clean Air Mercury Rule (CAMR) was announced by the U.S. Environmental Protection Agency (EPA), which requires the power industry to implement the maximum achievable control technology to reduce mercury emission 20% by 2010 and 50% by 2018. The U.S. Court of Appeals for the District of Columbia Circuit later issued a unanimous decision vacating the CAMR. On December 16, 2011, the EPA Administrator signed the final MATS and Utility NSPS rulemakings, and these were published in the Federal Register on February 16, 2012. On November 16, 2012, the Environmental Protection Agency (EPA) proposed to update emission limits for new power plants under the Mercury and Air Toxics Standards (MATS) [3]. EPA will issue a final reconsideration in March of 2013. Thus, efforts have continued to limit the emission of mercury from the coal combustion process by

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exploring the fate of Hg in the flue gas after leaving the boiler, which is aimed at finding more effective and economical control approaches.

During the combustion process, mercury in coal is volatilized and predominantly exists in the flue gas in the elemental form (i.e.,  $\text{Hg}^0$ ) after leaving the boiler. As the temperature decreases after the flue gas passes an air heater unit and/or selective catalytic reduction process,  $\text{Hg}^0$  begins to be converted to gaseous mercuric ion ( $\text{Hg}^{2+}$ ) and/or particulate-bound mercury (i.e.,  $\text{Hg}^{\text{p}}$ ). The chemical species of gaseous mercury is an important factor for controlling mercury emission from a coal combustion process.  $\text{Hg}^0$  is volatile and insoluble in water, which makes it difficult to be removed from flue gas without advanced control devices (e.g., activated carbon injection). Different from  $\text{Hg}^0$ ,  $\text{Hg}^{\text{p}}$  can be effectively captured by baghouses [4,5] or electrostatic precipitators [4,6]. Gaseous  $\text{Hg}^{2+}$  is highly soluble and can be effectively removed in the wet scrubbers of the flue gas desulfurization (FGD) process, which, therefore, has been considered as a cost-effective multi-pollutant control approach for coal-fired power generation utilities. However, results obtained from many field studies carried out by the Institute for Combustion Science and Environmental Technology (ICSET) [5,10,12] and reported by other researchers [7] suggested that  $\text{Hg}^{2+}$  captured by wet FGD process can be chemically reduced within the scrubber and re-emitted back to the stack gas as  $\text{Hg}^0$ . As a result, the overall removal efficiency of Hg is hidden. Reducing the occurrence of  $\text{Hg}^0$  re-emission can be a promising Hg emission control approach for the coal combustion process.

Parameters (e.g., pH [8–10], total sulfite and/or bisulfite concentrations [6,9],  $\text{Cl}^-$  contents [6–8], temperatures [7,8,11], and presence of transition metals [12]) controlling the re-emission of  $\text{Hg}^0$  have, therefore, been investigated and mechanisms were proposed by researchers [7,8] using bench-scale experiments under simulated wet scrubber conditions. It has been demonstrated that  $\text{Hg}^0$  re-emission can be effectively inhibited in a FGD slurry with higher  $\text{Cl}^-$  concentrations and/or lower pH [8–10]. Formation of an unstable mercuric-sulfite complex, e.g.,  $\text{HgSO}_3$ , is speculated to be involved in the reduction of  $\text{Hg}^{2+}$  in wet scrubbers [7,8], which is supported by the observations reported from the atmospheric cycle of Hg [13]. However, there have been disagreements in the literature data on the effects of slurry oxidation and total S(IV) concentrations (i.e., sulfite and bisulfite). Niksa and Fujiwara [14] suggested that the re-emission of  $\text{Hg}^0$  only occurred in the much lower oxygen level in the inlet of a WFGD system. In the study carried out by Chang and Ghorishi [7], they showed forced oxidation can enhance  $\text{Hg}^{2+}$  reduction and  $\text{Hg}^0$  emission as the concentration of sulfite and bisulfate in the scrubber slurry decreased. In addition, Wu et al. [10] showed that the reduction of the oxidized mercury was enhanced as the total aqueous S(IV) (sulfite and/or bisulfite) increased, which was also observed by other researchers [9]. However, results obtained from Chang and Ghorishi [7], and Blythe et al. [8] suggested otherwise.

With respect to decreasing mercury emission in a wet FGD process, it is beneficial to understand how the observations discovered in the laboratory-scale studies regarding  $\text{Hg}^0$  re-emission reflect in a full-scale operation. Considering the fact that a large quantity of  $\text{SO}_2$  is continuously removed from the flue gas and forced oxidation

has become more favorable in a full-scale operation, the effects of slurry oxidation and concentration of S(IV) in FGD slurry in a wet-scrubber should be concurrently considered when investigating their effects on  $\text{Hg}^0$  re-emission. In this study, the correlations between re-emission of  $\text{Hg}^0$  and sulfur dioxide removal, as well as the operation of oxidation air flow rate, were investigated under full-scale operation conditions with continuous mercury emission monitoring data and corresponding plant information (PI). The objective of this study is to demonstrate how the inhibition of  $\text{Hg}^0$  re-emission from a wet FGD scrubber might be achieved by modifying FGD operation without chemical additives. This study was carried out at four coal combustion power generation utilities, i.e., Units 31 and 32 of power plant C (Plant C) located in central Illinois and Units 1 and 2 of Plant O in western Kentucky.

## 2. Experimental

The configurations of each test unit can be seen in Table 1. The flue gases generated from Plant C Units 31 and 32 flowed into a common duct before entering the FGD unit. In the case of Plant O, the flue gases from Units 1 and 2 were combined before the electrostatic precipitator unit. Using duct dampers, the combined flue gas was either divided into two streams before the FGD plant, which had two wet scrubbers (i.e., Modules A and B), or directed to one of the scrubbers.

Two continuous mercury emission monitoring systems (CMMs) provided by Thermo Fisher Scientific and PS analytical were set up at the inlet of the FGD process and stack, respectively, for the testing carried out at Plant C. At Plant O, only the Thermo CMM system was set up at the stack. After setup, CMM systems were calibrated with three known concentrations of oxidized and elemental mercury vapors, which were produced by a HovaCal (EcoChem Analytics) and an 80i mercury calibrator (Thermo Fisher Scientific), respectively. Throughout the testing period, the systems were calibrated everyday with a known concentration of elemental Hg calibration vapor. Ontario Hydro (OH) and sorbent trap methods, as per the ASTM D6784 and USEPA Method 30B, respectively, were also conducted periodically at the inlet of FGD process and stack for comparison purposes. Plant information (PI) data provide by the utilities, e.g., unit load, coal flow,  $\text{SO}_2$  concentration at the inlet and outlet of the FGD process, lime feed rate, absorber blowdown rate, chloride blowdown, recycle pump status, force oxidation flow rate, recycle slurry density, absorber pH, slurry height in each absorber, total air flow rate, and/or flue gas oxygen concentration before FGD process, was collected.

During the testing carried out at Plant C, coal, bottom ash, ESP ash, FGD reagent slurry, FGD gypsum, and filtrates were also collected everyday for mercury analysis. Procedures described in ASTM methods ASTM D6357 and D6722, and Electric Power Research Institute (EPRI) Method H1 (EPRI, 2007) were followed.

## 3. Results and discussion

Two series of flue gas Hg emission measurements, Test C1 and Test C2, were carried out in different years at Plant C Units 31

**Table 1**  
Coal and boiler parameters of the tested units.

Plant	Boiler	Fuel types	Capacity (MWe)	Boiler types	Air pollution control device configurations		
					$\text{NO}_x$	Particulates	$\text{SO}_2$
C	Unit 31	Bituminous	90	Cyclone	SCR with $\text{NH}_3$ injection	Cold-side ESP	Limestone wet scrubber
	Unit 32	Bituminous	90	Cyclone	SCR with $\text{NH}_3$ injection	Cold-side ESP	
O	Unit 1	Bituminous	151	Cyclone	SCR with $\text{NH}_3$ injection	Cold-side ESP	Limestone wet scrubber
	Unit 2	Bituminous	280	Wall fired	SCR with $\text{NH}_3$ injection		

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