

Laboratory study of air–water–coal combustion product (fly ash and FGD solid) mercury exchange

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Abstract

Recent laboratory research has indicated that coal fly ash derived from subbituminous and bituminous type coals is a sink for atmospheric mercury (Hg), however lignite-based ash was found to emit Hg to the air. Solids collected from systems with components that enhance Hg removal (i.e. activated carbon injection (ACI), flue gas desulfurization (FGD), and selective catalytic reduction (SCR) or selective non-catalytic reduction (SNCR)) may have higher Hg concentrations and therefore a higher potential for Hg release. For this study we investigated the potential for Hg release to the air and water from coal combustion products (CCPs) collected from coal-fired units with FGD equipment, SCR and SNCR equipment, and sorbent injection for Hg removal. In the laboratory study, most dry samples acted as sinks for atmospheric Hg in the dark at 25 °C. When exposed to light or increased temperature (45 °C), deposition of Hg to the fly ash substrates in most cases continued but decreased. Wet FGD samples emitted Hg. However, they became a sink for atmospheric Hg or exhibited low Hg emission rates when dried. Mercury flux in the dark at 25 °C was correlated with fly ash carbon content (LOI). Most liquid extracts derived using the synthetic precipitation leaching procedure (SPLP EPA method 1312) had very low Hg concentrations (<13 ng/l).

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1. Introduction

Mercury (Hg) occurs naturally in coal and is vaporized during combustion [1], making coal-fired power plants a source of atmospheric mercury [2]. In United States an estimated 75 tons of Hg are contained in the fuel burned annually at coal-fired power plants; about 60% is released to the atmosphere, and the remaining 40% is retained in coal combustion products (CCPs) [3].

CCPs are the solid residues remaining after coal combustion for the production of electricity. High-volume CCPs include fly ash, bottom ash, boiler slag, and flue gas desulfurization (FGD) material. Coal ash is composed largely of oxides of silicon, aluminum, iron, and calcium, with smaller quantities of magnesium, potassium, titanium, phosphorus, sulfur, and alkali compounds. Flue gas desulfurization solids are composed primarily of alkaline–sulfur compounds, depending on the sorbent used for scrubbing. The characteristics of CCPs depend greatly upon the type of coal (e.g. anthracite, bituminous, subbituminous, and lignite) from which they are produced, combustion conditions, and air emission control equipment [4].

Control of mercury emissions from coal-fired power plants is currently achieved via existing pollution controls used to remove particulate matter (PM) (i.e. electrostatic

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precipitator (ESP), fabric filter (FF), compact hybrid particulate collector (COHPAC)), sulfur dioxide (SO₂) (wet and dry FGD systems), and nitrogen oxides (NO_x) (low NO_x burners (LNBs), selective catalytic reduction (SCR), and selective non-catalytic reduction (SNCR)) [5]. A leading candidate in technology currently being investigated for enhanced mercury control is activated carbon injection (ACI) [3]. The efficiency of Hg capture in air pollution control devices depends on Hg speciation in flue gas. Factors such as the type of coal, combustion conditions, SCR/SNCR, and SO₂ controls used at each power plant determine the speciation and therefore capture efficiency of Hg [6]. Wet FGD systems use an alkaline scrubber, most commonly limestone, and produce either sulfitic FGD material (sludge or unoxidized FGD material) or FGD gypsum (from forced-oxidation systems). FGD systems can capture soluble compounds of Hg²⁺ while PM control device can capture Hg_p [6]. Recent research suggests that catalysts used in SCR units could oxidize Hg⁰ to Hg²⁺ in the flue gas therefore enhancing the capture of Hg in a downstream wet FGD [6,7]. Unburned carbon is thought to play a significant role in mercury capture on fly ash [8]. Loss on ignition (LOI) is commonly used as an indicator of unburned carbon content in fly ash [9].

Approximately 122 million tons per year of CCPs were generated in U.S. in 2004 [10]. On March 15, 2005, EPA issued the Clean Air Mercury Rule to establish mercury emission limits for coal-fired utility boilers, potentially increasing the Hg concentration of fly ash and FGD materials [1,6,11]. This could restrict combustion products use [12]. Because the number of coal-fired plants with FGD systems is anticipated to grow over the next several years, the annual production of FGD materials is expected to

grow as well [10]. An additional concern is the possibility for captured mercury to be re-emitted from the fly ash and/or FGD material once it is in a landfill or pond [11]. According to a previous study, mercury in coal fly ash is unlikely to enter the environment (air or ground water) through volatilization or leaching [12]. In fact, the data showed coal fly ash could act as a sink for atmospheric Hg [12].

This project, which is an extension of a previous study [12], examined the potential for Hg release from 21 CCP samples using laboratory techniques. Investigation of Hg exchange between air and CCPs was conducted in a controlled experimental setting simulating in situ conditions using a single-pass gas exchange system. Total, recoverable, and leachable Hg concentrations in the CCP samples were determined. Samples included fly ash, FGD materials collected using wet scrubbers, and fly ash with activated carbon collected during Hg removal demonstration projects. In situ Hg emissions associated with fly ash, vegetated topsoil covered fly ash, and FGD solid residues have been studied and are reported in two papers [12,13].

2. Experimental

2.1. CCP sample information

CCP samples were collected from 10 power plants using lignite, bituminous, and subbituminous coal, and a variety of pollution controls (Table 1). All the samples were shipped to the University of Nevada, Reno in clean, sealed amber glass bottles. The bottles were stored in a cool dark cabinet under same conditions as described in the previous study to allow for comparability [12].

Table 1
Summary of relevant data from power plants where substrate samples were collected

Plant	Unit	Capacity (MW)	Primary coal type	Primary coal source	Particulate control ^a	Boiler type	SO ₂ control ^b	NO _x control SCR/SNCR ^c	NO _x control boiler ^d	Other ^e
B1	A	1300	Bituminous	WV, OH	ESPc	Wall fired	FGDw	SCR	LNB	
B8	A	640	Bituminous	WV	ESPc	Wall fired	FGDw (mag-L)/ Inhib	SCR	LNB and OFA	
B9	A	272	Bituminous	AL	ESPh and FF	Wall fired			LNB	COHPAC
B10	A	175	Bituminous ^f	CO	ESPc	Wall fired				
B11	A	165	Bituminous	S. App	ESPc	Tangential fired		SNCR		
B11	B	165	Bituminous	S. App	ESPc	Tangential fired				
B12	A	566	Bituminous	PA, IL	ESPc	Tangential fired	FGDw/LSFO	SCR		
L3	A	140	Lignite	ND	ESPc	Wall fired			LNB	
S4	A	350	Subbituminous	WY	ESPh	Tangential			LNB	
S5	A	233	Subbituminous	NM	FF	Tangential	FGDw		OFA	

^a ESPc: cold-side electrostatic precipitator; ESPh: hot-side electrostatic precipitator; FF: fabric filter.

^b FGDw: wet FGD; FGDw/LSFO: wet FGD with limestone forced-oxidation FGDw (mag-L)/Inhib: wet FGD using magnesium–lime sorbent with inhibited oxidation.

^c SCR: selective catalytic reduction; SNCR: selective non-catalytic reduction.

^d LNB: low nitrogen burner; OFA: overfire air.

^e COHPAC: compact hybrid particulate collector.

^f Blended fuel: 70% bituminous/30% subbituminous.

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