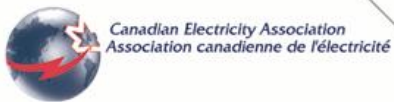


MERCURY INFORMATION **CLEARINGHOUSE**



Quarterly 1 - Sorbent Injection Technologies for Mercury Control



MERCURY INFORMATION CLEARINGHOUSE

QUARTERLY 1 – SORBENT INJECTION TECHNOLOGIES FOR MERCURY CONTROL

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QUARTERLY 1 – MERCURY MEASUREMENT

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MERCURY INFORMATION CLEARINGHOUSE

QUARTERLY 1 – SORBENT INJECTION TECHNOLOGIES FOR MERCURY CONTROL

EXECUTIVE SUMMARY

Mercury is an immediate concern for the Canadian and U.S. electric power industries because of pending regulation of mercury emissions. Canada has established a consultative process to develop Canadawide Standards (CWS) for mercury emissions from coal-fired electricity generation. Although the process has not been completed, an estimate of the reduction in mercury emissions within Canada is likely to be in the range of 60%–90% and aligned with U.S. standards.

Options for mercury control in existing coal-fired power plants consist of the enhancement of existing air pollution control systems and the addition of new multipollutant control options. Enhancement of existing air pollution control devices includes sorbent injection with and without sorbent enhancement agents upstream of existing particulate control systems and mercury oxidation upstream of wet and dry scrubbers. This report focuses on the injection of sorbents, specifically activated carbon.

The use of sorbent and/or activated carbon injection as a means of removing mercury from coal combustion flue gas is widely accepted as the most developed and commercially viable method. Application of sorbent injection in coal-fired utility boilers is very challenging because of lower concentrations of mercury and the range of mercury forms in coal combustion flue gases. The level of mercury in flue gases is typically 10 micrograms/normal cubic meter, and the mercury is in three forms: elemental, oxidized, and particulate. The distribution of mercury in the various forms is dependent upon coal composition. Typically, lower-rank subbituminous and lignitic coals with low chlorine levels produce flue gases where the mercury is dominated by the elemental form. Higher-rank bituminous coals that contain over 200 ppm chlorine produce flue gases where the mercury is in the oxidized or particulate form. The oxidized and particulate forms of mercury are more easily controlled by existing air pollution control devices than the elemental form. Flue gases that contain high levels of elemental mercury require the addition of a sorbent enhancement agent in order to increase the capture efficiency of mercury.

Activated carbon sorbent injection to achieve a required level of removal must be maintained at a minimum amount in order to avoid adverse impacts on air pollution control devices or ash disposal and utilization. The ability of the activated carbon sorbent to control mercury depends upon flue gas composition and temperature as well as the sorbent particle size, reactivity, and capacity. Research and demonstration projects for mercury control technologies are currently ongoing at laboratory, pilot, and field scales. Activated carbons show promise; however, their effectiveness depends on coal type and power plant configuration.

MERCURY INFORMATION CLEARINGHOUSE

QUARTERLY 1 – SORBENT INJECTION TECHNOLOGIES FOR MERCURY CONTROL

INTRODUCTION

In response to the need identified by the Canadian Electricity Association (CEA), the Energy & Environmental Research Center (EERC) was contracted to create and maintain an information clearinghouse on global research and development activities related to mercury emissions from coal-fired electric utilities. With the support of CEA, Center for Air Toxic Metals[®] (CATM[®]) Affiliates, and the U.S. Department of Energy (DOE), the EERC is developing comprehensive quarterly information updates to provide a detailed assessment of developments in mercury monitoring, control, policy, and related research advances.

Recent developments in the area of mercury regulations from coal-fired power plants in both Canada in the form of Canadawide Standards (CWS) and the United States in the U.S. Environmental Protection Agency's (EPA's) Proposed Utility Mercury Reduction Rule illustrate the need for effective mercury control strategies for coal-fired electric utilities. Sorbent injection upstream of a particulate control device is the retrofit technology that has demonstrated the widest application for mercury control in plants not equipped with flue gas desulfurization (FGD) scrubbers and is discussed in detail in this quarterly report.

In order to adequately address topics and provide the detail necessary for the various stakeholders to make informed decisions, selected topics will be discussed in detail in each quarterly report. Issues related to mercury from coal-fired utilities include the general areas of measurement, control, policy, and transformations. Specific topics that will be addressed in subsequent quarterly reports include, but are not limited to, the following:

- Mercury Policy
 - Upcoming events and news releases
 - Regulation, policy, compliance strategies, and health developments
- Mercury Measurement
 - Continuous mercury monitors
 - Advanced mercury-sampling systems
 - Wet-chemistry mercury measurement techniques
 - Baseline mercury levels and emissions
- Mercury Control
 - Sorbent technologies and control in unscrubbed systems
 - Advanced and developmental mercury control technologies
 - Summary of large-scale test activities and associated economics
 - Mercury oxidation and control for scrubbed systems
 - Multipollutant control strategies
 - Impact of mercury control on combustion by-products/fate of captured mercury

- Summary of mercury-related economics for commercial systems
Mercury Chemistry and Transformations
- Mercury chemistry fundamentals, modeling, prediction, and speciation
- Mercury fate and transport – Impacts on health

One objective of this quarterly report is to provide timely information on developments in the field of mercury. In order to address timely issues as well as provide necessary detail on selected topics, additional subject headings will be provided to summarize recent developments not related to the quarterly topic. In this manner, updated information can be provided on topics previously covered or in advance of topics not yet discussed.

The primary subject areas for this first quarterly report include an introduction to policy regarding regulations for mercury control as well as a detailed discussion on mercury control using sorbent injection. In the second quarterly report to be submitted in April 2004, mercury measurement will be covered in detail as well as CWS developments and a review of EPA's Proposed Utility Mercury Reduction Rule.

MERCURY POLICY – BRIEF OVERVIEW OF CURRENT REGULATIONS

Mercury is an immediate concern for the Canadian and U.S. electric power industries because of pending regulation of mercury emissions. Canada has established a consultative process to develop CWS for mercury emissions from coal-fired electricity generation. A process is well under way to evaluate and discuss, in conjunction with a multistakeholder advisory group, options for achieving cost-effective reductions in mercury emissions. Recently, the Canadian Council of Ministers of the Environment (CCME) agreed that consultation should continue with regard to setting CWS in 2005 (1). CCME is committed to developing mercury standards by 2005. The most common discussion points for this standard are to achieve significant (>50%) emission reductions by 2010, with ongoing review to address the emerging science in the United States and elsewhere on mercury control. Standards within Canada are likely to be in the range of 60%–90% and to align with U.S. standards.

In December 2000, EPA decided that regulation of mercury from coal-fired electric utility steam-generating units is appropriate and necessary under Section 112 of the Clean Air Act (2). EPA determined that mercury emissions from power plants pose significant hazards to public health and must be reduced. The EPA *Mercury Study Report to Congress* (1997) (3) and the *Utility Hazardous Air Pollutant Report to Congress* (1998) (4) both identified coal-fired boilers as the largest single category of atmospheric mercury emissions in the United States, accounting for about one-third of the total anthropogenic emissions. On December 15, 2003, EPA published the proposed Utility Mercury Reduction Rule in order to solicit comments on two approaches for mercury emission control. The proposed EPA rule is being reviewed extensively by many organizations and is summarized here; however, a more comprehensive evaluation of the proposed rule will be provided in the next quarterly report. Under one approach, coal-fired power plants in the United States would be required to install currently available control devices defined as maximum achievable control technologies (MACT) under Section 112 of the Clean Air Act. This approach requires utilities to comply by the end of 2007 and is expected to result in mercury emission reduction of 14 tons (29%). The second approach, proposed under Section 111 of the Clear Air Act would create a market-based “cap-and-trade” program. This alternative would apply to both new and existing sources and takes advantage of co-pollutant mercury control associated with SO₂ and NO_x reductions required by the Interstate Air Quality Rule that was also proposed by EPA in December 2003. Under this approach, a mandatory declining cap would be set for total mercury emissions from all U.S. coal-fired power plants. Emissions trading would be allowed, and a mercury cap of 34 tpy would be enforced in 2010 and a lower cap of 15 tpy would be enforced in 2018, resulting in a total mercury emission reduction of nearly 70%. Implementation of this alternative would require EPA to revise its December 2000 finding that it is appropriate and necessary to regulate utility hazardous air emissions under the MACT standard of the Clear Air Act.

In light of these proposed regulations, it is important to note that there are no commercially available mercury removal technologies that can be universally applied to coal-fired boilers because of the array of coals and plant configurations present in North American coal-fired electric generating facilities. It is critical, therefore, to maintain an accurate and timely review of the issues associated with both control technologies and measurement in the context of changing environmental policies. Further development of sampling and measurement methods are necessary to improve accuracy and reduce the costs currently associated with mercury monitoring and future

compliance. This is especially important with respect to the CWS effort and as U.S. and Canadian stakeholders develop commercially viable control methods.

QUARTERLY 1 FOCUS: SORBENT INJECTION TECHNOLOGIES FOR MERCURY CONTROL

The use of sorbent and/or activated carbon injection as a means of removing mercury from coal combustion flue gas is widely accepted as the most developed and commercially viable alternative. Many other technologies are being investigated, but the demonstrated performance of activated carbon in the municipal waste combustor industry has generated much interest for application in the coal-fired utility industry. However, application of sorbent injection in coal-fired utility boilers is far more challenging, owing to the lower concentrations of mercury that must be treated, the lower equilibrium capacities and mass-transfer rates associated with these lower concentrations, the wide range of concentrations of acid gases and chlorine species present, and the shorter gas residence time upstream of the particulate control device.

Sorbent Injection Background and Fundamentals

A detailed review of sorbent injection technologies has been conducted by the EERC and was published by Elsevier in *Fuel Processing Technology* in 2003 (5). This report provides a summary of the key findings in that review and a comprehensive look at the issues related to mercury control using sorbent injection. This report covers the following general topics: 1) mercury control requirements to be met, 2) laboratory studies on variables affecting sorbent capacity, 3) pilot-scale tests on capture performance for different coals, 4) research on sorbent properties to identify improved sorbents, and 5) full-scale demonstration results. The review provided goes beyond the presentation of descriptive information and attempts to explain cause-and-effect relationships; thus the interpretation may need to be revised as the understanding of mercury chemistry advances.

Requirement

Sorbent injection can be used as an effective control technology, provided a required level of mercury removal is achieved with a minimum amount of activated carbon while avoiding adverse impacts on air pollution control devices or on ash disposal and utilization. At the present time, activated carbon at typically 50 cents per pound represents a cost factor of about 0.4 mils/kWh at an injection ratio of C/Hg of 10,000:1 for coal containing 0.1 ppm mercury. Injection at this level results in approximately 1%–2% in fly ash loading in a coal combustion system. This carbon content in the ash adversely impacts the use of fly ash for cement replacement in concrete. Therefore, low carbon injection rates or sorbents other than carbon are desired to minimize the impact on ash marketability and reduce sorbent costs.

Equilibrium Sorption Capacities Determined in the Laboratory

Numerous bench-scale, fixed-bed tests have been conducted (6–11) to evaluate the mercury adsorption capacity of sorbents, including a range of carbons such as iodine- and sulfur-impregnated carbons and inorganic compounds. The capacities have been measured in nitrogen, air, or simulated flue gas of varying compositions. The testing conducted in simulated flue gas is the most relevant to actual conditions to which a sorbent is exposed. Some of the key factors that influence sorption capacity include temperature, oxidation state of the mercury, and composition of flue gas.

Lignite-derived DARCO™ FGD activated carbon has been studied as a mercury sorbent for coal-fired systems more extensively than any other sorbent. FGD activated carbon has a bulk density of 0.51 g/cm³ and a surface area of 600 m²/g. Baseline tests conducted at Radian (6, 10) and the EERC (7, 11) were performed in a simulated flue gas atmosphere typically containing 6% O₂, 12% CO₂, 8% H₂O, 1600 ppm SO₂, 50 ppm HCl, and 20–80 µg/Nm³ Hg⁰ or HgCl₂. The variables investigated included mercury concentration; concentration of the acid gases SO₂, HCl, and NO_x (NO and NO₂); water vapor concentration; and mercury oxidation. Tables 1 and 2 present details on carbon properties and test conditions (6). Important findings for DARCO™ FGD activated carbon are presented in Figures 1A–II and in the discussion that follows:

- Increasing temperature (Figure 1A [10]) results in decreased equilibrium adsorption capacity.
- Equilibrium adsorption capacity using baseline simulated flue gas composition and a bed temperature of 135°C was found to increase linearly with increasing inlet levels of either Hg⁰ or HgCl₂, as shown in Figure 1B (6). This is consistent with a physical adsorption mechanism. It is also believed that chemisorption of an oxidized species to a basic site on the carbon is the binding site on the carbon.

Table 1. Properties of DARCO™ FGD Powdered-Activated Carbon (5)

Carbon Property	Lab Data	Norit America's Datasheet
<i>General Properties</i>		
Bulk Density, g/mL	–	0.51
Surface Area, m ² /g	–	600
Molasses Decolorizing Efficiency	–	90
Iodine Number	–	600
<i>Particle Size</i>		
% Passing 325 mesh	94	95 minimum
Avg. Size from SEM Analysis, µm	15	–
Avg. Size from Microtrac Analysis, µm	14	–
<i>Pore-Size Distribution, mg/g</i>		
Micro, <20 Å	–	0.18
Meso, 20–50 Å	–	0.25
Macro, 50–150,000 Å	–	1.06
<i>Chemical Composition, wt%</i>		
Oxygen	28	–
Carbon	22	–
Silicon	14	–
Calcium	13	–
Iron	7.4	–
Aluminum	7.1	–
Sulfur	3.7	1.8
Magnesium	2.9	–

Table 2. Bench-Scale Test Sorption Capacity Experiments for DARCO™ FGD PAC (5)

Parameter	Baseline Value	Range Tested
Gas Rate (1/min at 75 °F)	1	–
<i>Gas Composition</i>		
HgCl ₂ , µg/N m ³	40–80	10–130
Hg ⁰ , µg/N m ³	40–80	–
SO ₂ , ppm	1600	0–3000
HCl, ppm	50	1–100
O ₂ , %	6	–
CO ₂ , %	12	–
H ₂ O, %	7	–
Adsorption Temperature, °F	275	225–400

- Sorbent particle size determines the minimum sorbent requirement needed to effect mass transfer from the bulk gas to sorbent particles (12, 13). Based on the mass transfer calculations, 90% removal in 2 seconds at a C/Hg ratio of 10,000 for a flue gas stream that contains 10 µg Hg/Nm³ in flue gas requires a mean particle size of about 4 µm for a typical size distribution. The capacity of the sorbent would be 100 µg Hg/g C at an exiting mercury concentration of 1 µg Hg/Nm³. In-flight mercury capture using DARCO™ FGD may be limited by mass transfer.
- Water vapor in flue gas increases the equilibrium sorption capture for mercuric chloride and Hg⁰ (6).
- HCl increases the equilibrium adsorption capacity of the DARCO™ FGD for Hg⁰ as shown in Figure 1D (6).
- SO₂ in the absence of NO_x, as shown in Figure 1E (6), reduces the equilibrium adsorption capacity dramatically for Hg⁰ and mercuric chloride. The effect of a combination of SO₂ and NO₂ reduces the capture of Hg⁰ even more severely (Figure 1F), with significant reductions in sorption capacity noted at concentrations as low as 100 ppm SO₂ and 2.5 ppm NO₂ (8, 11, 14).
- NO_x (10% NO₂ and 90% NO) has an impact on Hg⁰ capacity in the presence of SO₂ and HCl, as shown in Figure 1G (6). The equilibrium sorption capacity of Hg⁰ is minimal in the absence of both NO_x and HCl, and it increases as NO_x alone increases. In the presence of HCl, the capacity for Hg⁰ drops as NO_x increases. The combined effect of NO₂ and SO₂ is discussed in detail elsewhere (8, 11, 14). The capacity effects suggest that HCl and NO_x/NO₂ can promote the oxidation and capture of Hg⁰ and no chemisorption capture appears to occur in the absence of mercury oxidation.

- The equilibrium sorption capacity for Hg^0 increases with increasing levels of oxidation occurring across the carbon test bed, as determined by changing the concentrations of HCl and SO_2 , as shown in Figure 1H (6). This indicates that mercury oxidation is an essential step in capturing mercury on sorbents, as shown in Figure 1I.

Mercury Capture on Activated Carbon in Pilot-Scale Tests

DARCO™ FGD activated carbon was tested in pilot-scale facilities to capture mercury through injection upstream of a fabric filter (FF). Figure 2 shows trends in mercury removal across FF systems burning Powder River Basin (PRB) subbituminous coal and low-sulfur eastern bituminous coal as a function of injection rate. These coals contained similar levels of mercury but quite different concentrations of chlorine, sulfur, iron, and calcium. Overall, the results for the PRB coal indicate 1) significant mercury capture on fly ash without sorbent injection in both electrostatic precipitator (ESP) and FF systems, 2) lower removals in the ESP as compared to the FF, and 3) moderate increases in removal associated with either increased carbon injection or lower temperatures.

Very different trends were observed when DARCO™ FGD activated carbon was injected into pilot FF systems burning two eastern bituminous coals: low-sulfur Evergreen coal tested at the DOE National Energy Technology Laboratory (NETL) (15) and Blacksville coal tested at the EERC (7). Overall results for the bituminous coals indicate 1) for the Evergreen coal, increasing the injection rate from a C/Hg ratio of 2500–10,000 resulted in an increase in removal from 30% to 85% and extrapolation to $\text{C}/\text{Hg} = 0$, which suggests that very little removal occurred on fly ash without carbon injection (Figure 2); 2) for Blacksville bituminous coal, increasing the injection rate over a range of C/Hg ratios between 3500 and 13,000 resulted in little if any increase in removal at any given temperature (7), and removals increased from 10% to 95% at a constant C/Hg ratio of 3800 when the flue gas was cooled from 175° to 100°C, as shown in Figure 3, and 3) temperature on mercury removal is less pronounced for the Evergreen bituminous coal and is still less for the Belle Ayr PRB coal (Figure 3).

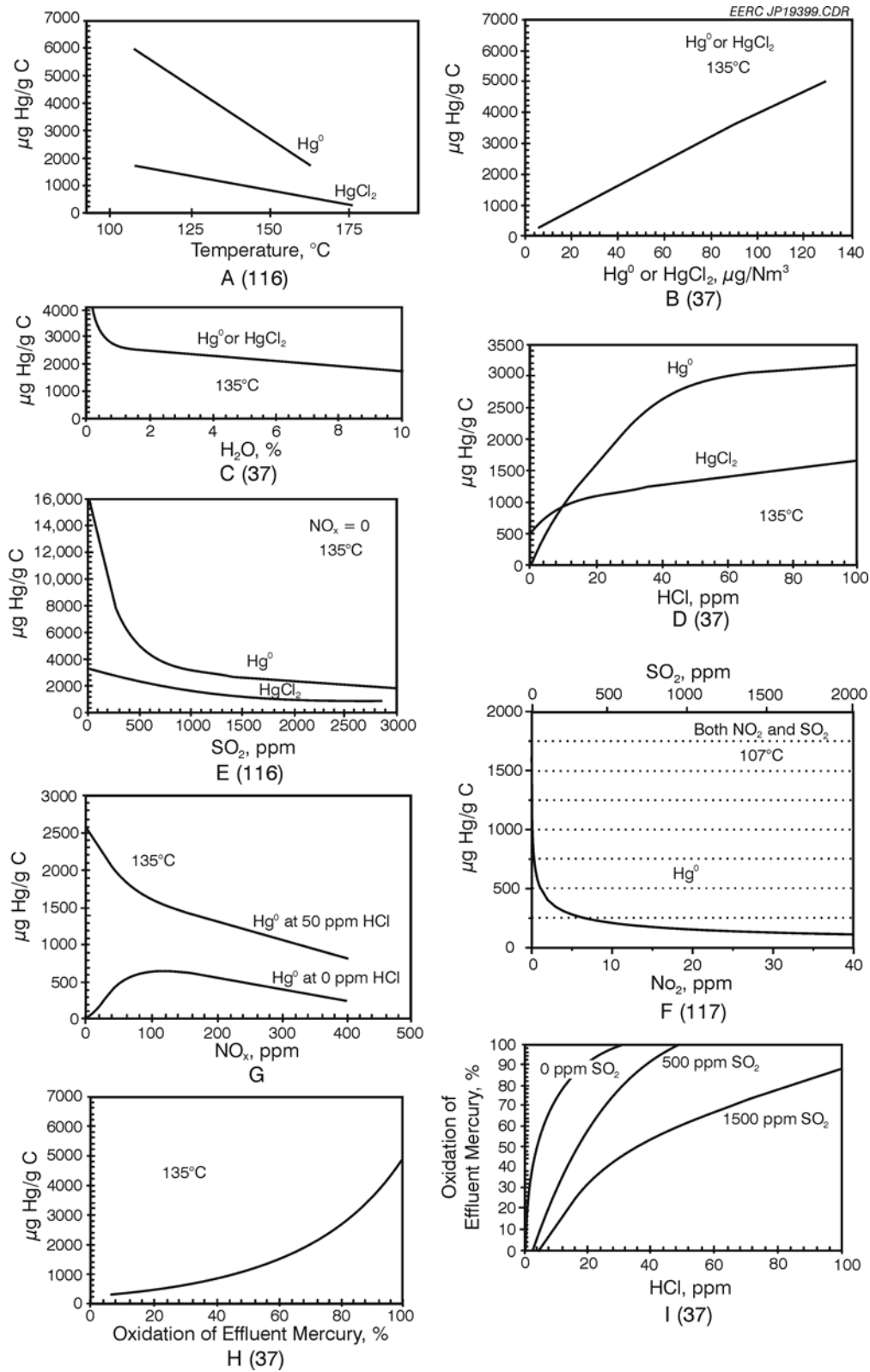


Figure 1. Effects of temperature, gas concentration, and oxidation on the Hg⁰ or HgCl₂ sorption capacities of DARCO™ FGD activated carbon in µg Hg/g C (5).

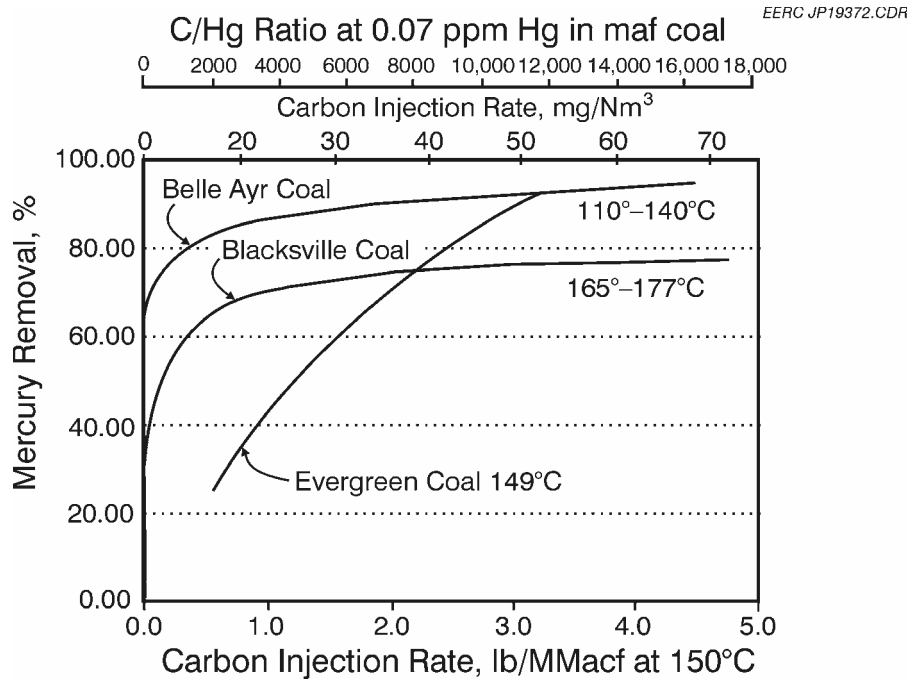


Figure 2. Effect of carbon injection rate on mercury removal in pilot system FF burning Belle Ayr, Wyoming, subbituminous coal and Evergreen bituminous coal (5).

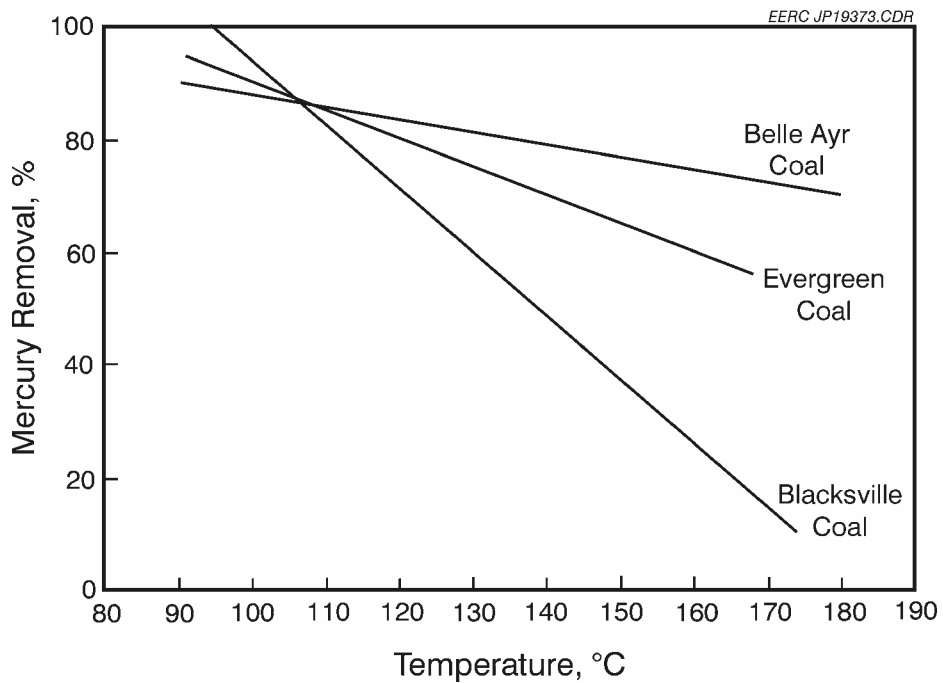


Figure 3. Effect of temperature on mercury removal showing sensitivity to coal type: Evergreen, Blacksville, and Belle Ayr coals (5).

An analysis of mass-transfer control for the three test coals based on selected experimental data points relating mercury capture at 150°C to carbon injection ratios (C/Hg) between 3760 and 6400 is summarized in Table 3. Results of mass-transfer calculations presented in Table 3 were used to predict removal levels for the three test coals at a C/Hg ratio between 0 and 10,000 and also the C/Hg ratio estimated to be required to achieve 90% removal. The removals calculated for the Belle Ayr and Evergreen test coals agree with the observed trend presented in Figure 2. It is predicted that C/Hg ratios of 12,872 and 15,822, respectively, would be required to achieve 90% removal.

Observed Hg removals were independent of injection rate above a C/Hg ratio of 3760 for Blacksville coal. This indicates that other factors besides mass transfer need to be considered. The dramatic increase in removal from 10% to 95% observed for Blacksville coal when the flue gas was cooled from 175° to 100°C at a constant C/Hg injection ratio of 3800 suggests an increase in the effective sorption capacity at the lower temperature. Therefore, it is proposed that the effective sorption capacity of Blacksville coal available in the 5-second residence time was increased dramatically because of the combined effects of decreasing temperature and heterogeneous catalytic oxidation promoted by maghemite.

Effects of Sorbent Properties

Carbon sorbents of different types have been tested to identify methods to improve performance and reduce cost. Four types of activated carbon, including lignite-derived, bituminous coal-derived, iodine-impregnated, and sulfur-impregnated carbons, are listed in Table 4. The properties of the sorbents and Hg⁰ sorption capacities are presented.

In general, the sorption capacities for activated carbons derived from lignite and bituminous coal are similar. Table 4 shows that the carbons differ significantly with respect to surface area, pore diameter, porosity, and particle size. Overall, there is no conclusive evidence at this time that the precursor coal is a critical factor in selecting an activated carbon for sorbent injection.

Carbons that had been treated with different chemical agents to increase their mercury capture performance are compared in Table 4. Iodine-impregnated activated carbon exhibited capacities between 507 and 8530 µg Hg/g C, depending on Hg⁰ concentration and flue gas temperature. These capacities are comparable to those shown for untreated coal-based carbons at similar conditions. The iodine-impregnated carbon performed better than either lignite- or bituminous coal-derived activated carbons in pilot-scale injection tests (7). Sulfur-impregnated carbons have higher sorption capacities than their untreated precursors (16–21).

Catalytic carbons derived from bituminous coal-derived activated carbons (e.g., Calgon F 400) by impregnating with nitrogen compounds and recarbonizing were shown to have mercury sorption capacities in air similar to those of lignite-derived FGD carbon (22, 23). Treatment with calcium chloride has been shown to produce a significant increase in capacity (20), and Ca(OH)₂-based sorbents (alone and admixed with fly ash) have been reported to provide superior capture of mercuric chloride but not Hg⁰ (24).

Impregnated carbons may offer a practical advantage over the coal-derived activated carbons but need more testing. Currently, the differences observed in equilibrium capacity do not

Table 3. Comparison of Activated Carbon Injection for Pilot Test Using Norit FGD Carbon (5)

	Belle Ayr Wyoming PRB Subbituminous	Blacksville Pittsburgh No. 8 Bituminous	Evergreen Low-Sulfur Bituminous
<i>Coal Analysis, mf</i>			
Hg, ppm	0.07	0.069	0.09
Cl, ppm	50	650	1247
S, ppm	5000	24,000	10,000
Fe, ppm	2600	14,000	NA
Ca, ppm	14,000	3100	NA
<i>Flue Gas Conditions</i>			
Hg Equivalent to Coal Hg, $\mu\text{g}/\text{N m}^3$	7.54	6.4	8.57
Characteristic Mercury Oxidation, % Hg(II) ^a	36 [55]	75	81
HCl Equivalent to Coal Cl, ppm	3	37	71
NO _x , ppm	750	550	600
SO ₂ , ppm	260	1670	770
<i>Selected Experimental Results on Mercury Capture at 150 °C</i>			
C/Hg Ratio Tested	4000	3760	6400
Carbon Concentration, $\text{g}/\text{N m}^3$	0.03	0.024	0.055
Total Mercury Removal at Designated C/Hg, %	74	37	62
Removal on Fly Ash w/o Carbon Injection, %	60	6 [55]	6 (est)
<i>Analysis of Mass Transfer Control Assuming Fast Reaction 35 on Carbon and Slow Reaction on Ash^b</i>			
Total Removal at Designated C/Hg, %	74	37	62
Removal on Fly Ash with Carbon Injection, %	39	4	2.4
Adjusted Removal on Carbon, %	35	33	60
Carbon Particle Size Required, μm	15.1	14	14
Hg on Injected Carbon, $\mu\text{g Hg}/\text{g C}$	88	88	93
Nominal Capacity at 150 °C	250	500	500
Percent of Capacity Utilized	35	18	19
<i>Removal Versus Injection Rate, %</i>			
C/Hg			
0	60	6	6
2000	68	24	29
4000	74	39	47
10,000	86	68	77
12,872	90		
15,822			90
21,038		90	

^a Reported oxidation levels ahead of a FF baghouse, not necessarily at the same temperature.

^b This assumption causes the injected carbon to be initially exposed to a gaseous Hg concentration corresponding to the mercury content of the coal, which is consistent with the reported conclusion that capture on fly ash occurs via a slow surface reaction.

Table 4. Selected Properties of Activated Carbons (5)

Sorbent Type	Sorbent Identification	Ash, wt%	Surface Area, m ² /g	Pore Volume, cm ³ /g	Avg. Pore Diameter, nm	Porosity, %	Mass Mean Particle Size, μm	Impregnation Treatment			Carrier Gas	Elemental Mercury Capacity		
								Reagent	Amt., %	Temp., °C		Hg ⁰ Conc., μg/N m ³	Temp., °C	Capacity, μg/g
<i>Lignite Activated Carbon</i>														
Norit Americas	DARCO™ FGD	32.1	546	0.611	3.8	57.9	15				Simulated flue gas w/o NO _x	45	135	2590
Norit Americas	DARCO™ FGD		500–700				9					54	135	3627
												52	163	2460
<i>Bituminous Coal-Derived Carbon</i>														
Norit Americas	PC-100		900		1.8		<30				Simulated flue gas w/o NO _x	330	100	1780 ^a
Prepared by the Illinois State Geological Survey from Illinois Bituminous Coal	AC-36	18.9	680									54	135	2091
	AC-C	18.8	688	0.382		46.2	8					76	135	2188
<i>Iodine-Impregnated Carbon</i>														
Barnebey & Sutcliffe			750				3.5	Iodine			Simulated flue gas w/o NO _x	60	107	8530
												60	135	1929
												60	163	892
												20	135	1697
												20	163	507
<i>Sulfur-Impregnated Carbons</i>														
Calgon, Bituminous Coal-Derived Activated Carbon Treated with Sulfur	BPL		1007				210	None	0.6					
	BPL-S-4/1-250		168					Sulfur	37.4	250	Nitrogen	55	140	550 ^b
	BPL-S-4/1-400		632					Sulfur	10.5	400		55	140	1450 ^b
	BPL-S-4/1-600		835					Sulfur	10.1	600		55	140	2200 ^b
	BPLH-0.25		570					H ₂ S	12.7	150		55	140	200 ^b
			27				3–8	Sulfur	5.5		Simulated flue gas w/o NO _x	50		1500 ^b
								Sulfur	33	400		50		13,831
								Sulfur	33	400		50		3260

^a The value of 1780 μg/g was for 24-h exposure at 100°C to gas containing 330 μg/N m³ Hg⁰ (24). Hsi et al. (18) reported a capacity of 230 μg/g at 4 h, 140°C, and 40 μg/N m³.

^b These capacity values were estimated by graphical integration with extrapolation to breakthrough and may be quite approximate.

appear to be important since most of the capacity values are adequate at C/Hg ratios well below 10,000. The minimum C/Hg ratio of 5000 to 10,000 is anticipated to be required based on diffusion control. The final criteria is sorbent cost. Impregnated sorbents require additional processing, resulting in higher costs. Finally, the limits on carbon in fly ash and the loss of revenue due to the inability to sell fly ash also need to be considered in the economics of mercury control.

Large-Scale Demonstrations of Sorbent Control Technologies

An overview of full-scale test results shows that the effectiveness of sorbent injection is highly variable for various particulate control devices and coal rank (25). Results from Phase I field tests performed by ADA-ES under a cooperative agreement with NETL for a unit burning bituminous coals show very high removals, greater than 90%, at low injection rates of 3 lb/MMacf for a COHPAC™ FF installed in series with a hot-side ESP (EPRI's TOXECON™ control technology)¹. Much lower removals were observed at similar injection rates for a cold-side ESP where capture trended linearly toward 90% only at high injection rates of 20 lb/MMacf.(25–28). Retrofit ESP–FF technology can be applied for multipollutant control to improve particulate collection efficiency and remove mercury separately without affecting the marketability of a majority of the fly ash. Hot-side ESPs alone, operating at 450°–750°F, do not effectively capture mercury. Mercury capture from a PRB coal-burning unit equipped with a cold-side ESP reached 50%–60% at low injection rates of 5 lb/MMacf but leveled off between 60%–65% at higher rates up to 30 lb/MMacf. These data suggest that high injection rates alone do not ensure high capture in ESPs for low-rank coals having both low chlorine content and low unburned carbon in ash. Mercury capture in this PRB test was not improved, with decreased temperature achieved by spray cooling the flue gas from 300° to 250°F. However, raising the flue gas temperature from 300° to 350°F in the ESP case burning bituminous coal reduced mercury capture from 90% to only 45% at 20 lb/MMacf, indicating that cooling may be required where flue gas temperatures exceed 340°F (26).

Full-scale testing of five different activated carbons chosen on the basis of laboratory adsorption capacity demonstrated similar trends in removal versus injection rate when tested at 300°F. These results indicated that all had sufficient capacity and reactivity to capture mercury from bituminous flue gas in an ESP. Comparison of in-duct capture with total removal across the ESP showed that the majority of the capture and almost all of the capture at high injection rates of 15–20 lb/MMacf occurred in less than a half-second of residence time in the duct (29). In bench-scale tests, equilibrium absorption capacities have been evaluated for several sorbents and compared to that of Norit FGD carbon (30). Oil-based activated carbons tested under PRB simulated flue gas conditions demonstrated an elemental mercury absorption capacity ranging from 1168 to 2267 µg/g, which was similar to Norit FGD carbon at 2070 µg/g under the same conditions. A number of activated carbons derived from vehicle tire rubber char have also been tested and have been shown to oxidize 95%–99% of the inlet elemental mercury and possess absorption capacities ranging from 444 to 2310 µg/g when tested with PRB simulated flue gas. In lignite flue gas in which chloride concentrations are very low, many activated carbons have not performed well. However, in tests of iodated carbons in a lignite simulated flue gas, mercury absorption capacities ranged from 1430 to 1930 µg/g, compared to only 1140 µg/g for the Norit

¹ Note: injecting a sorbent downstream of an ESP and upstream of an FF is a technology configuration that has been used by Alstom under the designation Filsorption in Europe to control mercury in waste-to-energy plants and is patented by EPRI in the United States as TOXECON™ for controlling mercury in power plants.

FGD carbon. In these tests, absorption capacity appeared to increase with iodine content as well as surface area of the carbon. Initial testing of carbon nanotube and fixed-structure carbon materials have demonstrated promise; however, adsorption capacity values are still much lower than more commercially available activated carbon.

Lignites like subbituminous coals typically have low chlorine content, high calcium content, and very low levels of unburned carbon in fly ash. Comparing carbon injection results for lignite from EERC field tests with the ADA-NETL tests discussed above, mercury removals for lignite using a cold-side ESP followed by a FF approached 90% at an injection rate of 7 lb/MMacf, approximately twice the injection rate needed for bituminous coal (31). Field results for lignite using a cold-side ESP alone showed the same level of removal at high injection rates as for PRB coal, except that capture for lignite peaked at 40%–50% instead of 50%–60%. Results of recent pilot-scale tests performed to optimize capture for lignite using various particulate control devices were reported for northern Great Plains test lignites that typically generate 85% elemental mercury at the inlet to particulate control devices (31). Mercury removals for cold-side ESP and ESP-FF configurations (EPRI's TOXECONTM and the EERC's *Advanced Hybrid*TM filter) were in general agreement with results of earlier field tests, with some difference observed for different lignites. Increasing temperature from 300° to 400°F required only a modest increase in injection rates (e.g., 14%) to achieve the same removals observed at 300°F in an ESP or ESP-FF configuration, respectively. However, a 70% increase in injection rate was required for a FF alone, possibly because of increased frequency of pulse cleaning at the higher temperature. Reducing the particle size of the activated carbon from a mean diameter of 20 to 5 microns increased mercury removals for a Poplar River lignite test from a range of 30%–40% to a range of 30%–75% in the interval of 8–18 lb/MMacf. A similar size reduction had no significant effect for another lignite, possibly because of particle agglomeration or coal-specific differences in the oxidants present. The addition of HCl to flue gas at two different temperatures (1700° and 300°F) to improve downstream sorbent effectiveness did not significantly improve mercury removal. However, the addition of NaCl to the lignite feed substantially improved mercury capture to levels approaching or exceeding 90% for three configurations tested: ESP-FF, *Advanced Hybrid*TM, and ESP alone.

The EERC's *Advanced Hybrid*TM particulate filter combines the best features of ESPs and FFs in a unique configuration that provides multipollutant control capability for ultrahigh collection of fine particulate and additional advantages as an excellent gas-solid contactor for mercury control (32). The *Advanced Hybrid*TM technology has demonstrated over 90% mercury control at a low carbon (DARCOTM FGD) injection rate of 1.5 lb/MMacf in tests on a 2.5-MW test unit operated on a slipstream from a cyclone-fired boiler burning PRB coal along with 4% supplemental fuel that included tire-derived fuel (TDF). Without supplemental TDF fuel, mercury removals at 1.5 lb/MMacf were 54%–74% in field tests and 50%–60% in smaller pilot-scale tests, which are levels comparable to the 60% removals observed for lignite in EERC field tests with the ESP-FF at 1–3 lb/MMacf (31). The effect of firing TDF based on a comparison of field- and pilot-scale data was to increase the level of unburned carbon from 0.2% to the range of 0.5%–1.9% and the level of HCl in the flue gas from 1–2 ppm to about 5 ppm (compared to 50–100 ppm for most bituminous coals), resulting in a large decrease in the inlet percentage of elemental mercury from 67% to 6%. Without TDF, injection of 10 ppm HCl upstream of the *Advanced Hybrid*TM filter had no appreciable effect on mercury capture. When operating without high-voltage power, the *Advanced Hybrid*TM filter distributes flue gas and sorbent evenly among all of the filter bags to make optimum use of sorbent capacity. With power on, most of the

sorbent is collected on the charged plates. Under this condition, mass transfer between the gas and the sorbent particles in-flight may be relatively more important than the sorbent capacity in limiting mercury removal. However, nearly identical removals of 93.8% and 94.8% were measured with the power on and off, indicating that gas–solid contact and residence time were adequate to achieve very high removals at the low carbon injection rate of 1.5 lb/MMacf in either case. Increasing the air to cloth ratio (A/C) from 5 to 10 ft/min at a constant injection rate of 1.5 lb/MMacf reduced mercury removals only marginally from the range of 94%–97% to 91%–92%, whereas increasing A/C from 6 to 14 ft/min at a constant sorbent rate, effectively reducing the volumetric injection rate from 2.51 to 1.07 lb/MMacf, lowered mercury removal from 94% to 77%. Carbon injection in the *Advanced Hybrid*TM filter had no adverse effect on particulate filtration, bag-cleaning interval, or pressure drop.

The activated carbon used in most pilot- and full-scale ACI tests has been DARCOTM FGD made by Norit Americas Inc., which has become the benchmark. In general, only slight differences in performance have been noted between these or other untreated commercial carbons. The criteria for selecting an activated carbon or other sorbent are minimum cost, including transportation, handling, feeding, and disposal, and ability to maintain marketability of fly ash. An extensive evaluation funded by NETL with technical support from EPRI was performed to identify cost-effective alternatives to commercially available activated carbons, starting with a broad screening and narrowing the selection by performing injection tests on boiler slipstreams (33). Overall results showed that there are alternatives that could be commercially developed to provide higher performance at lower cost but that not all sorbents will perform universally (e.g., for all coals). The initial screening was based on laboratory sorbent capacities at a mercury concentration of 50 µg/Nm³, testing 47 sorbents in simulated low-sulfur eastern bituminous flue gas and 27 in simulated PRB flue gas. Seventeen low-cost candidates were chosen for subsequent injection testing on a slipstream from a pulverized-fuel-fired boiler burning 85% bituminous coal and 15% petroleum coke, and five were chosen for similar testing on a cyclone-fired boiler burning PRB coal. DARCOTM FGD was tested as a reference carbon. Small-scale injection tests were performed to simulate two configurations: 1) in-flight capture in 2–4 seconds representative of the first section of an ESP and 2) capture for sorbent injection downstream of an ESP and ahead of a FF. The 10-acfm test unit used in this evaluation has been shown to provide results comparable to those measured in the field tests on the TOXECONTM technology (28).

In the injection tests performed on carbons selected for bituminous coal application, five of the 17 and the FGD reference carbon all demonstrated greater than 90% capture in the ESP–FF configuration. Parametric tests in this configuration identified two, a corn char and a treated activated carbon, that achieved higher removals at lower injection rates than FGD carbon over the range of 1–2 lb/MMacf with removals 5%–10% higher at 350° than at 315°F. In-flight tests showed that a soot-derived carbon having a significantly smaller particle size (d_{50}) of 6.2 µm provided higher removals than corn char carbon with a particle size of 15 µm. In-flight mercury capture generally increased with residence time and injection rate, but maximum removals were all less than 70% in the range of 2.5–15 lb/MMacf.

In the injection tests for PRB coal, three of five test carbons and the FGD carbon demonstrated similar performance of 80%–86% removal in the ESP–FF configuration at an injection rate of 1.5 lb/MMacf, including carbons derived from corn, soot, and German brown coal. Parametric tests resulted in similar removals for FGD carbon and the corn- and brown coal-

derived test carbons trending upward from 65% to 95% over the range of 0.5–5 lb/MMacf, with no appreciable difference in removals between 300° and 350°F. In time-of-flight tests at 300°F, all removals were less than 50% for both soot- and corn-derived carbons in the range of 2.5–5 lb/MMacf, with the smaller particle size of the soot-derived carbon again providing somewhat higher removals. A commercial iodine-impregnated carbon demonstrated much higher removals in this configuration and greater improvement between 2- and 4-second residence time, suggesting that the small amount of HCl present in the PRB flue gas was not adequate to provide in situ conditioning of the untreated sorbents at the high injection rates required in an ESP.

Bench- and pilot-scale testing of a non-carbon-based sorbent have shown promising results for mercury capture at high temperatures, typical for a combined physisorption and chemisorption process. The sorbent is currently marketed as a specialty mineral additive for concrete applications, positively affecting compressive strength and pozzolanicity. Fixed-bed testing suggested mercury capture could reach 75% with increasing temperature up to 1100°F. However, at temperatures below 600°F, no mercury sorption was observed. Pilot-scale tests were conducted at Southern Research Institute's 1.75-MW Combustion Research Facility burning Choctaw bituminous coal. Mercury capture was measured at near 50% at the baghouse inlet, approximately 4 seconds downstream of the sorbent injection point, at a rate of 22lb/MMacf. For this test, the temperature of the injection point was 1600°F, and at the baghouse inlet, 320°F. Mercury measurements at the baghouse outlet illustrated a general downward trend in mercury concentration over 8 hours of intermittent sorbent injection, ending in a maximum removal of near 90% at the conclusion of testing (34).

The effect of sorbent injection on mercury capture can be substantially impacted by the dispersion of sorbent within the flue gas (34). In an effort to quantify the mercury capture that occurs as a result of sorbent injection relative to residence time and turbulent mixing, bench-scale tests have been conducted using Norit FGD carbon, a nitrogen carrier gas, and a batch-mixing chamber. By monitoring mercury concentration at the various stages of injection, mixing, and residence time, the relative effects of each stage on mercury capture was determined. On average, 80% of the capture occurred immediately after sorbent injection. Additional residence times of 5 and 30 seconds under turbulent mixing conditions, made up less than 5% of the 30% mercury capture measured. Results from this research indicate that the way sorbent is injected into full-scale ductwork can impact mercury capture. Additional in-duct residence time may not be sufficient to ensure maximum efficiency, especially when applying sorbent injection technologies to ESP configurations where in-flight mass transfer is of primary importance.

Lessons Learned in Conducting the EPA Information Collection Request

In 1998, EPA issued an Information Collection Request (ICR) for calendar year 1999 to obtain mercury and chlorine analyses on coal and mercury speciation and emission data for 84 U.S. generating units representing different plant configurations burning various coals. Based on these data, it was estimated that, on average, 60% of the 75 tons of mercury in coal burned in utility boilers during 1999 was emitted into the atmosphere. Statistical correlations of percentage emissions, varying from under 10% to over 90%, were obtained to identify the most significant differences based on coal type and plant design. However, correlation and interpretation of the ICR data were limited by what are now recognized as deficiencies in data collection. Following is a summary of issues identified that can be addressed in future data-gathering efforts:

- Mercury speciation and capture in air pollution control devices downstream of the boiler are now known to be influenced by other elements in the coal and ash besides chlorine, which was analyzed as part of the ICR. At a minimum, future studies should include analyses for sulfur in coal, unburned carbon in ash, and the elements Ca, Mg, Na, K, and Fe. In addition, samples of coal and fly ash should be retained under suitable laboratory storage conditions to facilitate future analysis, as needed.
- Several different methods were used to analyze mercury and chlorine concentration in the coal during the ICR. The current methods of choice for mercury analysis are acid leaching (American Society for Testing and Materials [ASTM] D 6414) microwave digestion and direct combustion (ASTM 6722). The methods used for analyzing coal for chlorine were quite imprecise below 100 ppm. Improvements in analysis, including the use of ASTM D 6721-03, have decreased the lower limit of quantification to approximately 10 ppm. The Ontario Hydro wet chemical method used for measuring total and speciated mercury in the ICR tests on power plant flue gas is still the method of choice for research quality studies at plant sites. However, rigorous quality assurance and control, which appeared to be lacking in some ICR results, are required to ensure reliable results.

UPCOMING EVENTS

Winter International Activated Carbon Conference
March 1–2, 2004, Mexico City, Mexico
<http://members.aol.com/hnpacs/conferences/IACC11techSum.htm>

227th American Chemical Society National Meeting – Spring 2004
March 28–April 1, 2004, Anaheim, California

Electric Power 2004
March 30–April 1, 2004, Baltimore, Maryland
Mercury Session March 29, 2004, 1:30–4:30 p.m.
http://www.electricpowerexpo.com/conference_program.asp

Symposium on Air Quality Measurement Methods and Technology
April 19–22, 2004, Research Triangle Park, North Carolina
<http://www.awma.org/events/confs/Measurements/default1.asp>

Principles, Applications, and Opportunities with Activated Carbons
Professional Analytical and Consulting Services – Dr. Nowicki will provide a
2-day short course for \$950
June 17–18, 2004, Toronto, Ontario, Canada
<http://members.aol.com/hnpacs/news/MercurySorbentOpp.htm>

International Conference on Mercury as a Global Pollutant
June 7–July 2, 2004, Ljubljana, Slovenia
http://congress.cd-cc.si/icmgrp04/?menu_item=welcome&menu_level=2

Air & Waste Management Association 97th Annual Conference and Exhibition
June 22–25, 2004, Indianapolis, Indiana
<http://www.awma.org/ACE2004/>

228th ACS National Meeting – Fall 2004
August 22–26, 2004, Philadelphia, Pennsylvania

EPRI–EPA–DOE–A&WMA Power Plant Air Pollutant Control “Mega” Symposium August 30–
September 2, 2004, Washington, D.C.
Sign-up deadline April 15, and final presentations due July 23, 2004
<http://www.awma.org/events/mega.pdf>

14th International Activated Carbon Conference
October 7–8, 2004, Pittsburgh, Pennsylvania
<http://members.aol.com/hnpacs/conferences/IACC11techSum.htm>

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