ADVANCED EMISSIONS CONTROL DEVELOPMENT PROGRAM

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Abstract

In cooperation with the U.S. Department of Energy, Federal Energy Technology Center and the Ohio Coal Development Office within the Ohio Department of Development, the Babcock & Wilcox Company, McDermott Technology, Inc., has evaluated the formation and control of trace element emissions from the combustion of Ohio bituminous coals. In response to industry concern over potential regulation of mercury emissions from utility boilers, testing in Phases II and III of the Advanced Emissions Control Development Program was focused on measurement of the quantity and species distribution of mercury downstream of the boiler and emissions control equipment. Testing included evaluation of the potential for controlling mercury emissions in coal-fired utility power plants using conventional particulate control and flue gas desulfurization (FGD) equipment. This paper presents the results of mercury emissions testing on pilot-scale facilities at the Alliance Research Center. The emissions control techniques discussed in this paper include wet limestone SO_2 scrubbing, sorbent injection systems, a baghouse, and an electrostatic precipitator (ESP).

Introduction

Under the Clean Air Act Amendments of 1990, the United States Environmental Protection Agency (US EPA) was mandated to evaluate emissions of hazardous air pollutants (HAPs) from fossil fuel-fired electric generating units and to provide a summary report to Congress on mercury emissions sources, controls, and health impacts. Figure 1 identifies the trace elements of primary concern. Field measurements sponsored by the United States Department of Energy (US DOE) and the Electric Power Research Institute (EPRI) have characterized HAP emissions from a variety of coals, boiler types, and emissions control equipment configurations. The results have indicated that existing particulate emissions control equipment - electrostatic precipitators (ESPs) and baghouses - provide high efficiency removal of most of the trace elements generated by coal combustion. However, for mercury, the data revealed that a wide range of removal efficiencies exist for commercial particulate and SO_2 emissions control equipment. The Babcock & Wilcox Company, McDermott Technology, Inc. is conducting testing to evaluate causes of the observed performance variations and optimize the use of conventional systems to provide near-term solutions for enhanced control of mercury emissions from coal-fired boilers.

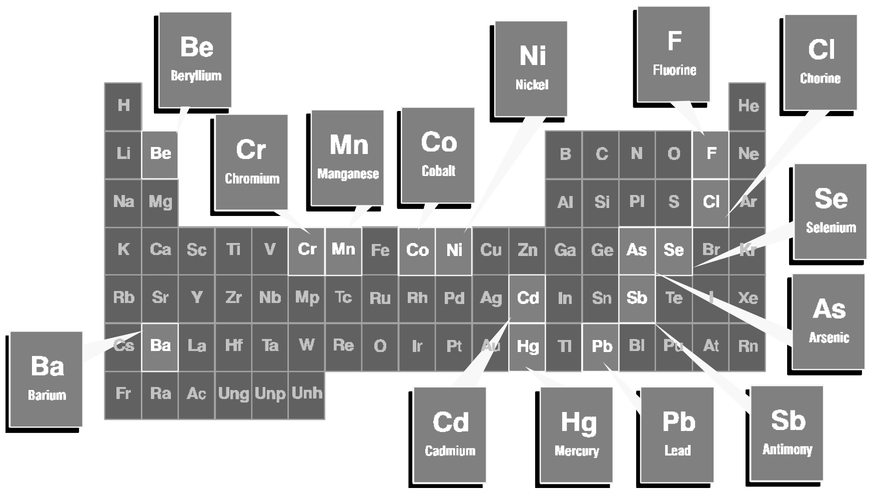

Figure 1. Trace Elements of Primary Concern.

The US EPA, state environmental agencies, and regional associations continue to evaluate the need for regulation of mercury emissions from coal-fired boilers to reduce human exposure to this persistent, bio-accumulative trace element. Mercury is emitted from coal-fired boilers in very low concentrations. Based on field sampling at utility sites, uncontrolled mercury emissions from coal combustion are generally in the range of 5 to 30 µg/dscm, already well below the regulated emissions limit of 80 :g/dscm for municipal solid waste (MSW) boilers. Annual mercury emissions from a coal-fired unit not equipped with SO_2 emission controls are on the order of one-third to one pound of mercury per MW of generating capacity[1]. However, as a group, coal-fired boilers represent a major unregulated source of mercury emissions to the environment. The US EPA and EPRI estimate that coal-fired utility boilers emit 50 to 55 tons of mercury per year in the U.S[2].

The variation associated with the reported mercury emissions control efficiency of commercial emissions control systems and the potential for mercury regulations, suggest that additional research is required. It is necessary to better define causes for the observed performance variations and maximize mercury emissions control performance of conventional flue gas emissions control systems. In cooperation with the US DOE and the Ohio Coal Development Office (OCDO) within the Ohio Department of Development, Babcock & Wilcox (B&W) is evaluating mercury emissions control performance of commercial FGD systems as well as advanced systems under development by B&W. The Advanced Emissions Control Development Program (AECDP) is directed toward demonstration of practical, cost-effective strategies for reducing HAP emissions from coal-fired boilers using conventional particulate and SO_2 control equipment.

Approach

The work is made possible by the state-of-the-art Clean Environment Development Facility (CEDF). Figure 2 provides an isometric view of the CEDF. The 100 million Btu/hr CEDF integrates combustion and post-combustion testing capabilities to facilitate the development of the next generation of power generation equipment. The furnace has been carefully designed to yield combustion zone temperatures, flow patterns, and residence times representative of commercial boilers. Boiler convection pass and air heater simulators maintain representative conditions through the entire boiler system. Back-end systems include both a baghouse and an electrostatic precipitator for particulate control, sorbent injection systems, and wet and dry scrubbers for SO_2 control.

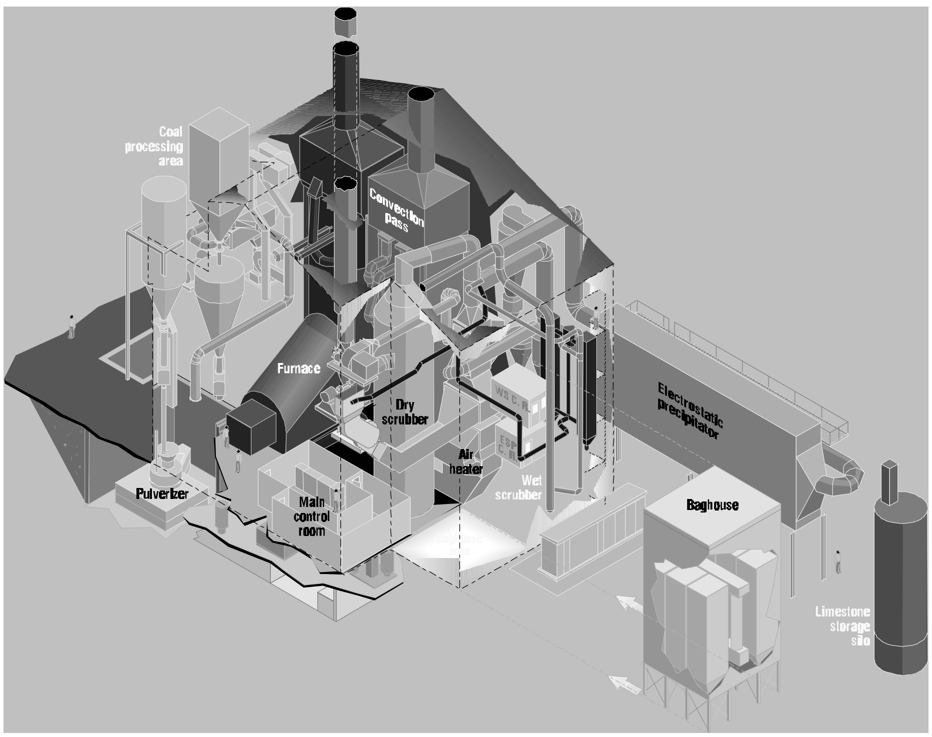

Figure 2. Isometric View of the Clean Environment Development Facility.

Phase I of the AECDP project focused on characterization work to verify that representative air toxic results were being obtained from the CEDF. Comparisons were made between uncontrolled trace metal emissions from the CEDF furnace and predicted levels for those emissions based on correlations developed by the EPA on the basis of utility boiler data. Excellent agreement was obtained, indicating that CEDF results can be reliably used to predict the air toxics emissions performance of full-scale systems. Phase I results laid the groundwork for the development activities in Phases II and III. This paper provides a summary of the Phase II results and some general information from Phase III, which is in progress.

Mercury Speciation

The form or species of mercury present in the flue gas impacts the performance of emissions control equipment. Mercury is generally present either as elemental mercury, Hg^0, or as oxidized compounds such as $HgCl_2$ and HgO. Industry experience to date suggests that Hg^0 and $HgCl_2$ are the dominant species in the flue gas from coal-fired boilers. The oxidized form of mercury is much more soluble in the aqueous solution present in Flue Gas Desulfurization (FGD) systems than elemental mercury and is, therefore, more likely to be removed from the flue gas. Elemental mercury tends to remain in the vapor state at the operating temperature of conventional emissions control equipment. A relatively higher proportion of oxidized mercury present as $HgCl_2$ would be expected to result in higher removal efficiency in a FGD system.

Mercury measurements for the AECDP project were made using the EPA Method 29 and Ontario Hydro methods. EPA Method 29 is a validated method for measuring total mercury emissions and is used as a benchmark for comparison of alternative speciation measurement methods. Much of the early mercury emissions testing cited in literature was performed using EPA Method 29. However, the method has been shown to report a significant fraction of the elemental mercury as oxidized mercury[3]. The Ontario Hydro Method is a modification of EPA Method 29 in which an alternative reagent is used in the initial impingers to prevent the oxidation of elemental mercury. The US DOE and EPRI are sponsoring efforts to evaluate various measurement techniques for quantifying the relative amount of elemental and non-elemental or oxidized forms of mercury in the flue gas[3]. This work has identified the Ontario Hydro Method as the preferred technique. The Ontario Hydro method was therefore used to characterize the relative distribution of mercury species in CEDF testing, with EPA Method 29 used as a check on the total mercury. The most recent modification of the Ontario Hydro Method was incorporated in Phases II and III of the AECDP testing, and involves the addition of a $KMnO_4/H_2SO_4$ solution to the KCl impingers immediately following the post-sampling leak check of the impinger train. This stabilizing agent prevents the loss of mercury from these impingers during the recovery procedure and improves the total mercury recovery[3].

Furnace Mercury Emissions

Furnace mercury emissions were measured for six commercial eastern bituminous coals. While the total mercury emissions from the furnace are primarily dependent upon the mercury content of the coal, compositions of fly ash and flue gas can affect whether the mercury is associated with the vapor or solids and the fraction of the mercury that is present in the oxidized form. Partitioning of the total mercury emissions between the vapor phase and the particulate phase measured in pilot tests at the Alliance Research Center is summarized in Table 1. The flue gas was sampled using the Ontario Hydro Method downstream of the combustion air pre-heater before the particulate collection equipment. Mercury emissions data for the Ohio 5&6 coal blend was obtained while firing the $5x10^6$ Btu/hr Small Boiler Simulator (SBS). The remaining data in Table 1 was obtained while firing the coals in the $100x10^6$ Btu/hr CEDF. The measured loss-on-ignition (LOI) at 800^oC for the fly ash sampled isokinetically from the flue gas stream for each coal is noted.

Table 1 - Furnace Mercury Emissions and Partitioning

Coal	Flue Gas Temperature (EF)	Total Mercury (:g/dscm)	Vapor Phase (%)	Filter Catch (%)	Fly Ash LOI (Loss-On-Ignition) (%)
Ohio 5&6	333	17.6	88.8	11.2	2.5
Mahoning 7	328	22.3	74.4	25.6	5.7
Ohio 6A	342	20.1	93.8	6.2	5.0
Meigs Creek	358	11.2	95.2	4.8	1.8
Ohio 5,6,&7	342	23.5	72.1	27.9	3.4
Clarion 4A	359	23.0	84.4	15.6	4.0

On average, approximately 15% of the total mercury was present on the particulate collected in the sampling train for the bituminous coals fired. For the narrow range of relatively low LOI values in these tests, the distribution of mercury between the vapor phase and the particulate did not appear to be strongly correlated with fly ash LOI.

Coal Cleaning Impacts

The total mercury emissions shown in Table 1 represent those from the combustion of commercially cleaned, high-sulfur eastern bituminous coals. For three of these coals (Ohio 5&6, Ohio 6A, and Meigs Creek), chemical analyses were performed on the raw and cleaned coals to evaluate the effects of commercial cleaning on mercury emissions. The total emissions in Table 1 match well with predicted emissions based on the coal chemistry and the assumption that all of the mercury exits the furnace. Comparison of the emissions to those predicted for firing the raw Ohio bituminous coals indicated that the average reduction in mercury emissions resulting from the cleaning processes was 49 percent, with a range from 45 to 56 percent. This range can be expected to vary with different types of coals having more or less mercury associated with the inorganic fraction.

Vapor Phase Mercury Speciation

The distributions of vapor phase mercury species in the flue gas at the air pre-heater outlet were measured for the Ohio 5&6, Ohio 6A, and Meigs Creek coals presented in Table 1 at the indicated flue gas temperatures. The speciation measurements using the Ontario Hydro Method are summarized in Figure 3. For these bituminous coals, the vapor phase mercury is primarily present as oxidized species following the boiler, upstream of the particulate emissions control equipment.

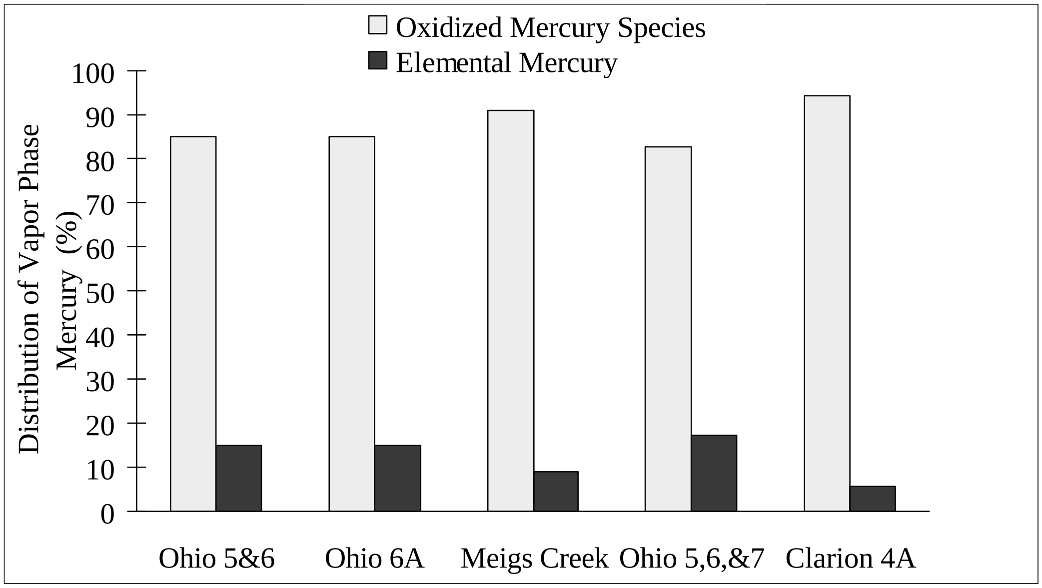

Figure 3. Mercury Speciation Downstream of the Air-Preheater.

Impact of Particulate Control Systems on Mercury Control and Speciation

The particulate control equipment upstream of the FGD system can potentially remove the majority of the mercury associated with the particulate phase. Data provided below represent results from sampling using the Ontario Hydro Method. Emissions measurements following the ESP show that mercury speciation across the ESP appears to be unaffected by the particulate control system. However, measurements downstream of the baghouse treating a flue gas slipstream indicate an apparent difference in mercury speciation, with decreases in elemental mercury measured across the system.

Electrostatic Precipitator

Mercury emissions control by the AECDP ESP has been measured for three high-sulfur bituminous coals. The ESP operating conditions and particulate emissions control performance for each coal are summarized in Table 2.

Table 2 - ESP Operations Summary

	Meigs Creek	Ohio 6A	Ohio 5&6
SCA (ft^2/kacfm)	281	285	310
Inlet Temperature (EF)	379	359	319
Average Temperature (EF)	368	349	304
Inlet Loading (gr/dscf)	4.60	2.25	2.32
Outlet Loading (gr/dscf)	0.022	0.021	0.017
Particulate Removal Efficiency (%)	99.52	99.07	99.27
Particulate Emissions (lb/10^6 Btu)	0.032	0.029	0.029

Figure 4 summarizes the total mercury measured at the inlet and outlet of the ESP for each coal, the partitioning of mercury between the particulate and vapor phases, and the distribution of mercury species in the vapor phase. The range bars in Figure 4 represent the range of the triplicate or quadruplicate individual measurements from which the average total mercury emissions value is obtained.

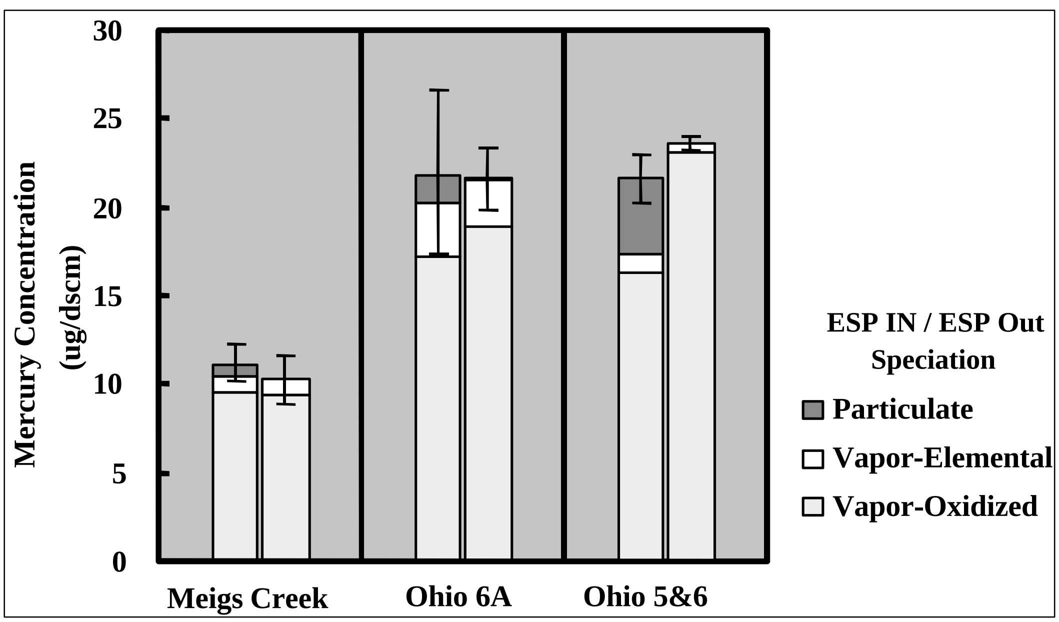

Figure 4. ESP Mercury Measurements.

For these three test coals, no significant reduction in total mercury emissions was measured across the ESP. Even though the potential exists to remove nearly all of the particulate phase mercury from the flue gas, the ESP impact was minimized because most of the mercury was in the vapor phase. Figure 4 also indicates that the ESP had no significant impact on elemental mercury in the flue gas.

Baghouse

The impact of a pulse-jet baghouse on mercury emissions was evaluated for the same three Ohio bituminous coals. The average operating conditions for the baghouse are summarized in Table 3. The same GoreTex™ membrane filter bags were installed in the baghouse for each of the test periods.

Table 3 - AECDP Baghouse Operating Summary

	Meigs Creek	Ohio 6A	Ohio 5&6
Air-to-Cloth Ratio (ft/min)	4.0	4.0	4.2
Pressure Drop (inches water)	5.6	5.2	5.3
Inlet Temperature (EF)	332	328	335
Average Temperature (EF)	313	310	316
Inlet Loading (gr/dscf)	4.60	2.25	0.94
Outlet Loading (gr/dscf)	0.009	0.002	0.002
Particulate Removal Efficiency (%)	99.80	99.91	99.79
Particulate Emissions (lb/10^6 Btu)	0.013	0.015	0.003

The impact of the baghouse on total mercury emissions and vapor phase mercury speciation is presented in Figure 5. As with the ESP, the average total mercury emissions reduction across the baghouse was minimal for each coal. For each coal, the concentration of elemental mercury in the vapor phase was reduced across the baghouse, while the oxidized mercury in the vapor phase increased. It should be noted that for these coals the elemental mercury concentrations were low, and the decreases in elemental mercury across a baghouse should be confirmed with coals that provide higher levels of elemental mercury at the furnace exit.

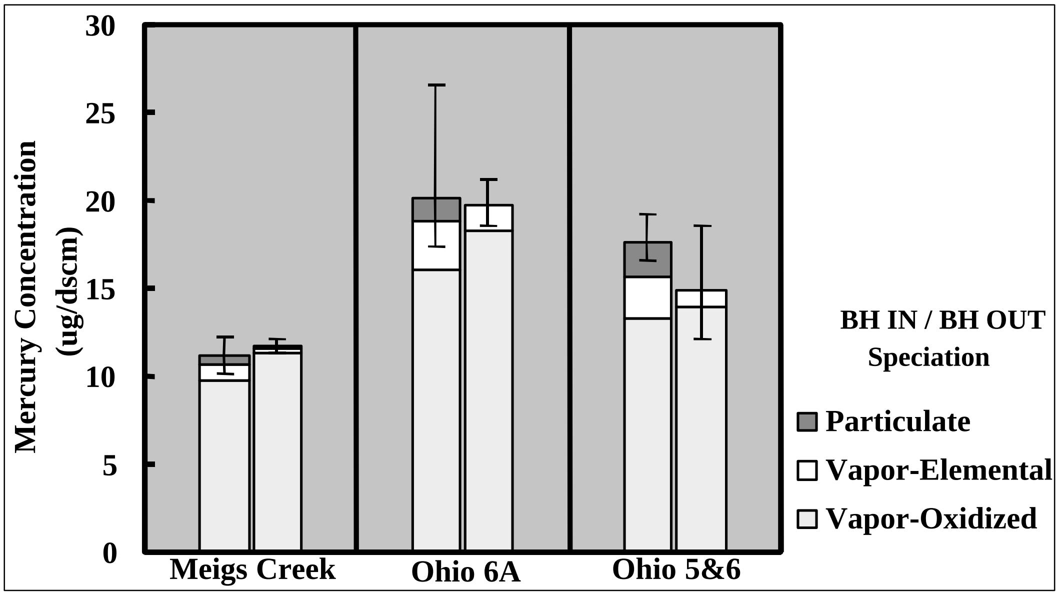

Figure 5. Baghouse Mercury Measurements.

Wet FGD Mercury Emissions Control

Wet FGD Pilot System

A wide range of mercury emissions control performance for wet scrubbers in bituminous coal applications (0 to 96%) appears in the literature with a number of factors contributing to this variability[2,4,5,6,7]. Significant differences in the mercury content of U.S. coals result in a wide range of mercury concentrations in the flue gas from the boiler. The form or species of mercury (elemental mercury or an oxidized compound such as $HgCl_2$) in the flue gas is thought to affect FGD system mercury removal efficiency. Mercury speciation in the flue gas is believed to be influenced by the type of coal fired, with sub-bituminous coals generating a higher relative proportion of elemental mercury than bituminous coals. EPRI pilot data indicates that at a flue gas temperature of 300EF, 68% of the total vapor phase mercury was present as elemental mercury for the sub-bituminous coal, compared to 6% as elemental mercury for the specific bituminous coal evaluated[8]. The coal chlorine content and ash characteristics may also influence partitioning between the solid and vapor phases and the mercury species in the vapor phase. The scrubber spray tower configuration, liquid-to-gas ratio (L/G), and slurry chemistry may also impact the reported mercury emissions control.

Wet FGD systems are currently installed on about 25% of the coal-fired utility generating capacity in the U.S., representing about 15% of the number of coal-fired units. FGD systems provide a cost-effective, near-term mercury emissions control option with a proven history of commercial operation. For boilers already equipped with FGD systems, the incremental cost of *any* vapor phase mercury removal achieved is minimal. The extent of the publicly available information base concerning the impact of basic wet scrubber design and operating conditions on mercury emissions control for bituminous coal applications needs to be expanded to provide a representative sampling of commercial FGD systems.

The AECDP wet scrubber was designed to allow simulation of commercial FGD systems. The wet scrubber system includes the absorber tower, a slurry recirculation tank, a reagent feed system, and a mist eliminator wash system. The 50-ft high by 2-ft diameter absorber tower is constructed of Plexiglas to permit visual observation of the slurry sprays. Pre-pulverized limestone is mixed with make-up water in the reagent feed tank. The solid content of the recirculating slurry is maintained at 12 to 15%. To achieve the desired L/G, any combination of four levels of single-spray nozzles may be used. The wet scrubber is equipped with a removable gas flow distribution plate to simulate both tray tower and open spray tower scrubber designs. An air sparger ring in the bottom of the recirculation tank is used for forced oxidation operation. Spent slurry from the scrubber is dewatered using a hydroclone circuit. The hydroclone underflow is discharged to settling tanks where the solids settle out and water is decanted to the clarified recycle water tank for re-use in the scrubber. A variable speed ID fan located downstream of the scrubber is used to control the gas flow rate through the scrubber. Typical scrubber operating conditions are summarized in Table 4. The wet scrubber was run at a higher oxidation air stoichiometry than a commercial unit to maintain the desired level of near complete oxidation because of the limited available height in the recirculation tank.

Three AECDP pilot test programs have been performed to characterize the mercury emissions control performance of wet scrubbers over a range of operating conditions for several coals. Following a brief series of tests to demonstrate that variation of scrubber operating conditions can impact mercury removal efficiency, an extensive program to characterize the impact of key scrubber design and operating parameters on mercury emissions control was completed. These tests covered the range of operating conditions reported in the field measurements summary used by the EPA as a basis for the *Mercury Study Report to Congress*. The impact of inlet vapor phase mercury speciation on scrubber mercury emissions control performance was evaluated in the third test series of Phase II.

Table 4 - Wet Scrubber Pilot Operating Parameters for Mercury Emissions Control Testing

Operating Parameter	Range of Operation
Inlet Flue Gas Flow (acfm)	2000 to 3000
Slurry pH	5 to 6
L/G (gal/1000 acf)	35 to 130
Slurry Spray Flux (gpm/ft^2)	20 to 70
Oxidation Air Stoichiometry (mol O_2/mol SO_2 absorbed)	0 to 8

Impact of Absorber Configuration

Most of the existing U.S. wet FGD capacity may be classified as open-spray tower or tray tower designs. Packed towers and venturi scrubbers represent smaller segments of the market. B&W markets the tray tower absorber design for controlling utility SO_2 emissions and has approximately 27,000 MW of wet FGD systems installed or under contract.

The AECDP scrubber was operated as both a tray tower and an open spray tower downstream of the baghouse while firing the Ohio 5&6 coal blend. Scrubber operations covered a wide range of slurry spray flux rates with a common tower velocity representative of conventional commercial scrubber operation. Operation with the gas flow distribution tray installed enhanced both SO_2 and mercury emissions control over a wide L/G range as illustrated in Figures 6 and 7. The error bars shown represent the range of the triplicate measurements for each set of operating conditions. For all of the tests presented in Figures 6 and 7, the oxidation air stoichiometry was greater than 5 mol O_2/mol of SO_2 absorbed to maintain near complete oxidation and the absorber slurry pH was maintained at 5.4 to 5.5. Additional tests to compare the tray tower and open spray tower configurations at nominal scrubber operating pHs of 5.0 and 5.9 showed comparable relative performance to that presented in Figures 6 and 7.

The tray tower configuration provided more consistent SO_2 and mercury emissions control than the open spray tower over the two-week test period. The tray significantly improved mercury emissions control at the lower L/G operating condition. For the four points shown in Figure 7, the average tray tower mercury emissions were 19 to 46 percent lower than the average measured for the open spray tower on a :g/dscm basis.

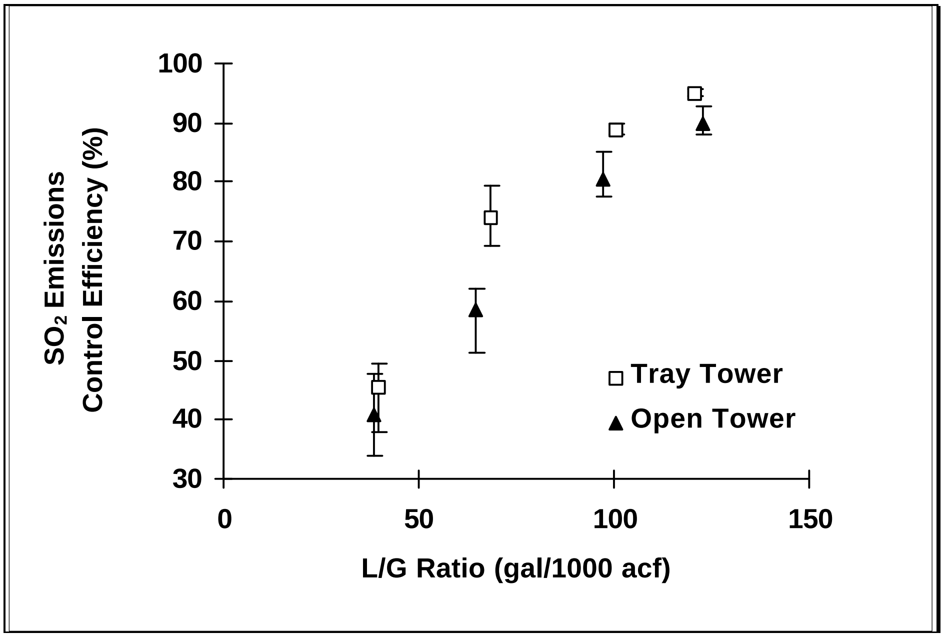

Figure 6. Impact of Absorber Tower Configuration on SO_2 Emissions Control.

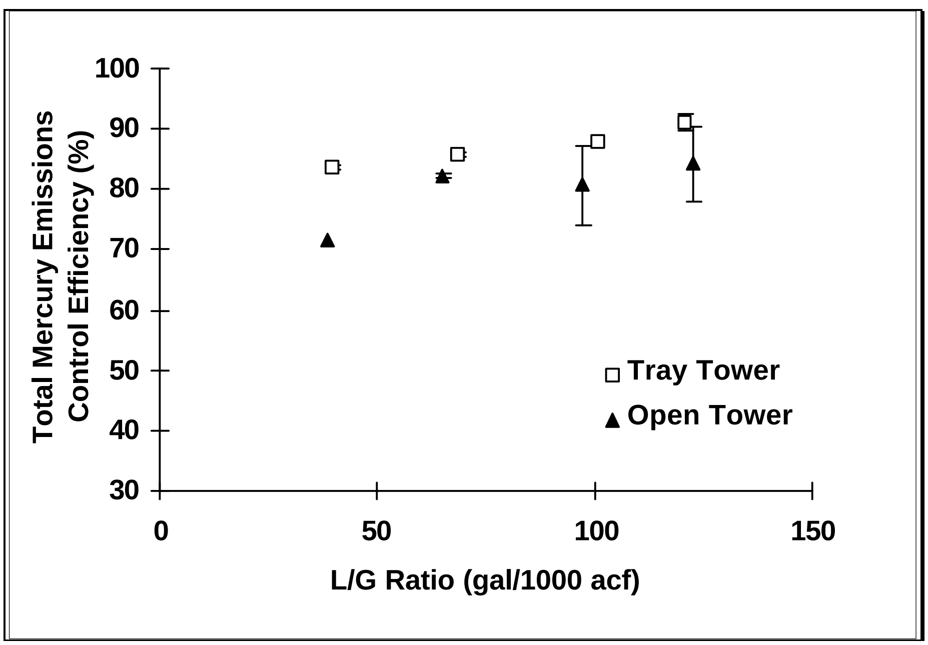

Figure 7. Impact of Absorber Tower Configuration on Mercury Emissions Control.

Impact of Scrubber Operation

The influence of slurry pH and L/G on mercury emissions control was evaluated for both scrubber configurations while firing the Ohio 5&6 coal blend. Figure 8 presents the impact of LSFO scrubber operation on total (vapor phase and particulate phase) mercury emissions for the tray tower. The superficial flue gas velocity was maintained at a steady value, and the slurry spray flux was varied to obtain a range of L/G operating conditions. Total mercury concentration downstream of the baghouse at the scrubber inlet averaged 14.8 :g/dscm. The distribution of mercury species at the scrubber inlet was 94% oxidized species and 6% elemental mercury as measured using the Ontario Hydro Method.

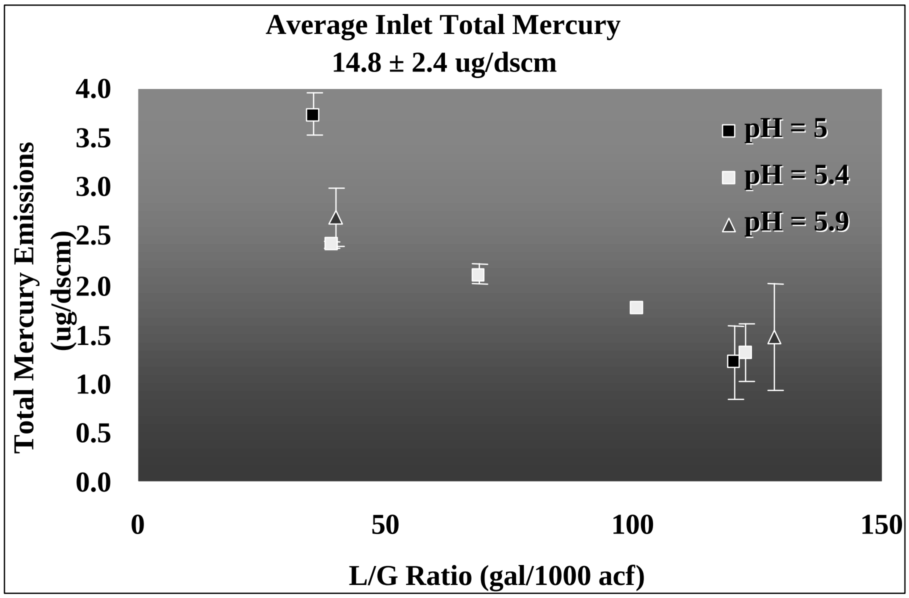

Figure 8. Impact of Scrubber Operating Conditions on Total Mercury Emissions - Tray Tower Following Baghouse for Ohio 5&6 Coal Blend.

Impact of Upstream Particulate Control

Operation of the AECDP wet scrubber at a common set of conditions for three bituminous coals provided data on the impacts of the upstream particulate control equipment and inlet mercury concentration on the mercury removal performance of the scrubber. The scrubber was configured as a tray tower and operated as a limestone-forced oxidation system. The pH of the recirculating slurry was maintained at 5.4. A common L/G of 125 to 130 gal/1000acf was used for each test series, with a nominal slurry spray flux of 67 gpm/ft^2 and an oxidation air stoichiometry of 6 moles of O_2/mole of SO_2 absorbed. Figure 9 provides the total mercury removal and mercury speciation at the scrubber inlet and outlet for two process configurations. Only the baghouse was used for the Ohio 5&6 coal blend test data shown.

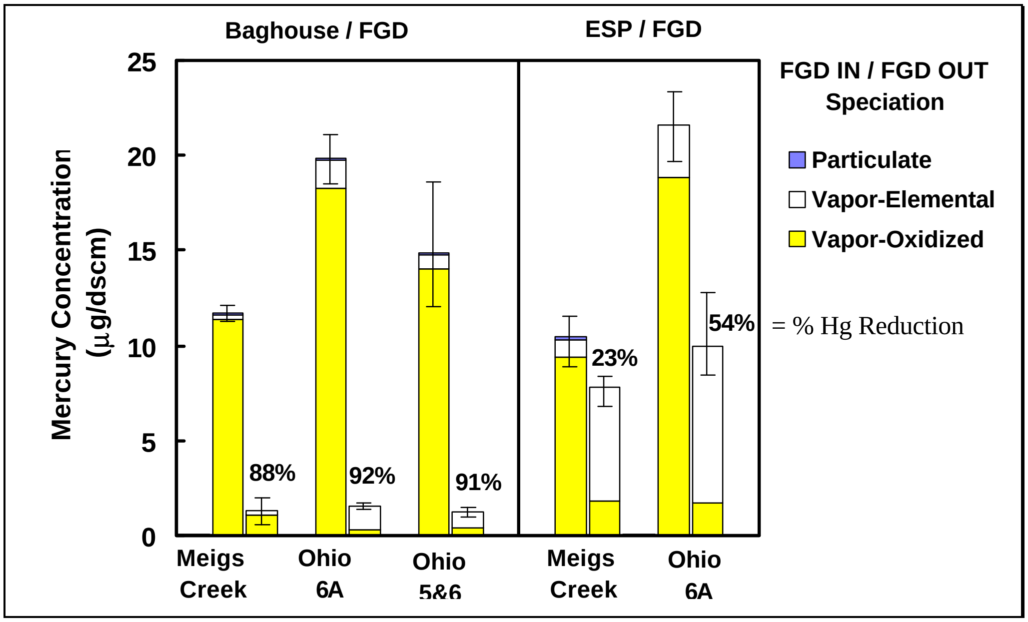

Figure 9. Wet Scrubber System Mercury Removal and Speciation Summary.

For the baghouse/FGD configuration, total mercury emissions were reduced by approximately 90% for all three coals despite the wide range of inlet mercury concentrations (11.7 to 19.8 :g/dscm). This is consistent with the expected high efficiency removal of oxidized mercury which accounted for 92 to 98% of the vapor phase mercury for each coal upstream of the scrubber with the baghouse in service. As shown in Figure 9, the level of elemental mercury in the flue gas upstream and downstream of the scrubber was consistent for each coal for the baghouse/scrubber configuration.

The scrubber mercury removal efficiency appeared to be significantly lower with the scrubber operating downstream of the ESP rather than the baghouse. Even though the inlet speciation was only moderately different with a slightly higher fraction elemental mercury downstream of the ESP relative to the baghouse, total mercury control across the scrubber was markedly lower for the ESP configuration, with a significant increase in the amount of elemental mercury in the outlet flue gas. The reasons for this are unclear at present and are the subject of further investigation in Phase III.

The apparent increase in the concentration of elemental mercury across the scrubber suggests that oxidized mercury absorbed by the scrubbing reagent may be reduced and off-gassed as elemental mercury. This explanation assumes that the speciation measurements at the scrubber inlet and outlet adequately reflect the actual speciation at these two locations. The measurements have been repeated to confirm these results, and as mentioned previously the Ontario Hydro method has been shown to perform well for measurements of total mercury and mercury speciation.

The baghouse/FGD system data in Figure 9 indicates that variation of the inlet mercury concentration over a range of 12 to 20 :g/dscm had little impact on mercury emissions from the scrubber. For all three coals, emissions from the scrubber remained steady at approximately 1.4 :g/dscm.

FGD Mercury Emissions Control Summary

Wet scrubber mercury removal efficiencies measured over a wide variety of operating conditions for several bituminous coals in the AECDP pilot tests are consistent with that reported for commercial installations and other pilot operations[9]. The wet scrubber FGD system research completed to date has demonstrated that many factors impact the overall system mercury emissions control efficiency. The particulate emissions control upstream of the FGD system as well as the absorber tower design and operating conditions can have a significant influence on mercury emissions for a given coal. Although the distribution of mercury species at the scrubber inlet is a key variable influencing mercury control, it may not be the dominant factor in predicting overall mercury emissions control efficiency.

Commercial and pilot data indicate that high-efficiency mercury emissions control can be achieved with a wet FGD system. The pilot data also indicate that FGD system design and operation impact mercury removal performance. Based on these results, a tray retrofit of an existing open tower scrubber may be a cost-effective means of enhancing both SO_2 and mercury removal efficiency. Application of an average mercury emissions modification factor to predict mercury emissions based on measurements of mercury in the coal does not differentiate the measured influences of scrubber design and operation on emissions control. The Babcock & Wilcox Company, McDermott Technology, Inc. continues to evaluate various aspects of wet scrubber design, operation, and scrubber chemistry to develop techniques for enhancing mercury removal in FGD systems.

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Advanced Emissions Control Development Program

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