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Advanced Emissions Control Development Program: Mercury Control

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Introduction

McDermott Technology, Inc. (a subsidiary of Babcock & Wilcox) is conducting the Advanced Emissions Control Development Project (AECDP) which is aimed at the development of practical, cost-effective strategies for reducing the emissions of hazardous air pollutants (HAPs) from coal-fired electric utility plants. The need for such controls may arise as the U. S. Environmental Protection Agency (EPA) proceeds with implementation of requirements set forth in the Clean Air Act Amendments (CAAA's) of 1990. Promulgation of air toxics emissions regulations for electric utility plants could dramatically impact utilities burning coal, their industrial and residential customers, and the coal industry. AECDP project work will supply the information needed by utilities to respond to potential HAPs regulations in a timely, cost-effective, environmentally-sound manner which supports the continued use of the Nation's abundant reserves of coal, such as those in the State of Ohio. The development work is being carried out using the 10 MW_c Clean Environment Development Facility wherein air toxics emissions control strategies can be developed under controlled conditions. The specific objectives of the project are to 1) measure and understand production and partitioning of air toxics species for a variety of coals, 2) optimize the air toxics removal performance of conventional flue gas cleanup systems, 3) develop advanced air toxics emissions control concepts, 4) develop and validate air toxics emissions measurement and monitoring techniques, and 5) establish a comprehensive, self-consistent air toxics data library. This project is supported by the Department of Energy, the Ohio Coal Development Office within the Ohio Department of Development and Babcock & Wilcox.

A comprehensive assessment of HAP emissions from coal-fired electric utility boilers sponsored by the Department of Energy and the Electric Power Research Institute concluded that with the exception of selenium and mercury, the majority of trace elements are well controlled due to their association with the particulate phase of flue gas. Reflecting the current focus of the USEPA and state environmental agencies on mercury as a potential candidate for regulation, the project specifically targets the measurement and control of mercury species. This paper discusses the results of testing on the quantity and species distribution of mercury while firing Ohio high-sulfur coal to assess the mercury emissions control potential of conventional SO₂ and particulate control systems. Results from recent AECDP tests are presented and two alternative mercury speciation methods are compared. The AECDP results clearly show that higher total mercury control efficiency can be achieved with a wet FGD scrubber than recently reported in the interim final USEPA report on HAP emissions from fossil-fired electric utility steam generating units.

Approach

The project objectives will be achieved through testing in McDermott Technology’s state-of-the art Clean Environment Development Facility (CEDF). The project has extended the capabilities of the 100 million Btu/hr CEDF to facilitate air toxics emissions control development work on “backend” flue gas cleanup equipment. Specifically, an electrostatic precipitator (ESP), a fabric filter (baghouse), and a wet scrubber for SO₂ control were added — all designed to yield air toxics emissions data under controlled conditions, and with proven predictability to commercial systems. A schematic of the CEDF and the project test equipment added is shown in Figure 1.

The CEDF design is based on extensive numerical modeling to replicate the thermal environment and flow characteristics of a “hot” utility boiler. Convection pass metal temperatures are maintained by way of a novel double-walled tube design and cooling with boiling water. Sufficient heat-transfer surface is available to cool the flue gas from the furnace exit temperature of around 2300 °F to the convection bank exit temperature of approximately 700 °F while burning high volatile, bituminous coal at full load. After the convection bank, the flue gas enters an air heater. Careful control of the flue gas cooling rate provides a gas time-temperature profile that is similar to commercial units to generate similar levels and forms of trace substances. The backend equipment including an ESP, baghouse, wet scrubber, and combined dry scrubber/baghouse configuration can be operated simultaneously for air toxics evaluation.

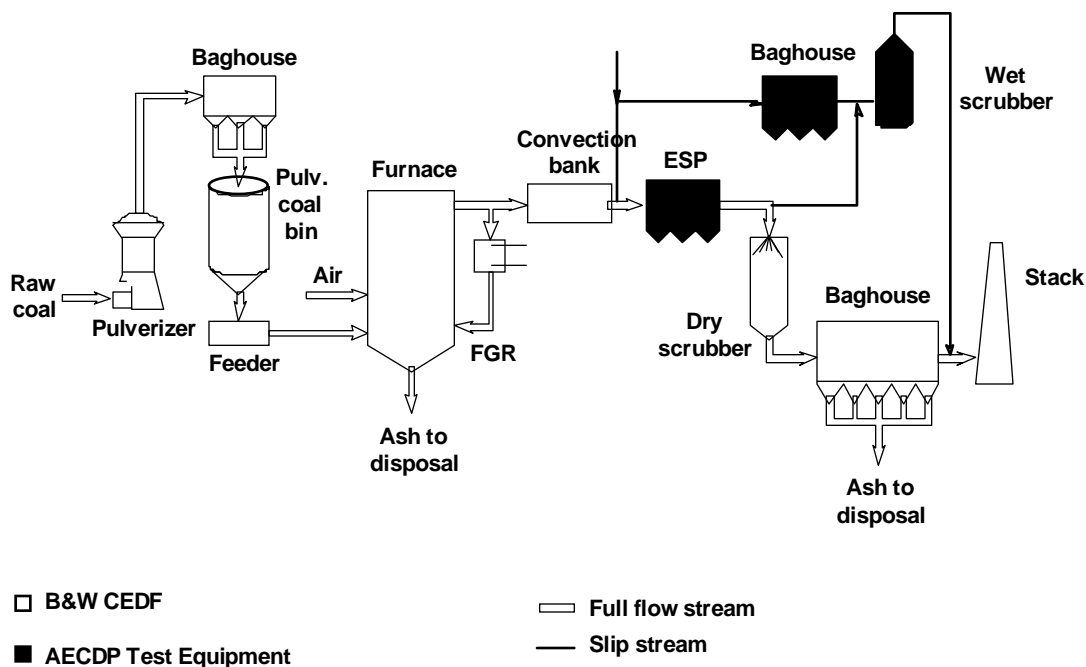


Figure 1 – Clean Environment Development Facility

Project Phases

The project is divided into three phases. Phase I (Facility Modification and Benchmarking) consisted of installation, shakedown, validation, and benchmarking of the test equipment added (ESP, slipstream baghouse, and wet SO₂ scrubber) to the CEDF. Baseline air toxics emissions and capture efficiency were established for each of the major flue gas cleanup devices: ESP, baghouse, and wet SO₂ scrubber. All tests were conducted with a high sulfur Ohio steam coal. The work in this phase culminated in the development of a data library for use by project participants.

Phase II (Optimization of Conventional Systems) testing involves the development of air toxics control strategies based on conventional particulate and SO₂ control equipment. Pilot testing, engineering and evaluation will be done to optimize the performance of these devices for the capture of air toxic species. Phase II testing will also provide data on the impacts of coal properties on air toxics emissions for several steam coals. The air toxics measurement techniques and monitoring instrumentation will also be investigated in this phase. Phase II work started in early 1996.

Phase III (Advanced Concepts and Comparison Coals) testing will be directed at the development of new air toxics emissions control strategies and devices, to further reduce the emissions of selected toxics. Testing will also be conducted to extend the air toxics data library to include a broader range of coal types. Finally, the development work on advanced air toxics emissions measurement and monitoring techniques begun in Phase II will continue in Phase III.

Facility Operation

The first test series of Phase II was completed in June, 1996. The test focus was the characterization of trace metal and particulate emissions from the particulate control devices as a function of ESP and slipstream baghouse operating conditions. The resulting information was used to guide subsequent evaluation of commercial control concepts for utilities that are equipped with wet scrubbers.

The focus of the second test series was the characterization of mercury emissions control across the wet scrubber as a function of key operating conditions designed to cover a range of commercial wet scrubber practise. The test matrix included the evaluation of the impacts of forced versus natural oxidation, open spray versus tray tower design, the liquid-to-gas ratio and the pH of the scrubbing solution. The wet scrubber was also evaluated for the control of other species (HCl, SO₂ and particulates). These tests were completed in February, 1997.

During the first test series, the CEDF was operated at constant, full load conditions (100 million Btu/hr, 10 MW_e equivalent). Key CEDF operating parameters (coal feed rate, load) had standard deviations of approximately 1.5%. A blend of Ohio 5 and 6 (Lower and Middle Kittanning) was fired with a low NO_x burner at a rate of approximately 4 tons per hour.

During the second test series, an alternate combustion facility operated at constant rate of 4 million Btu/hr was employed for the operation of the air pollution control devices. The same Ohio 5 and 6 coal blend was fired with a low NO_x burner at a rate of approximately 455 lbs/hour. The average boiler flue gas composition as measured at the air heater outlet during the two test series is summarized in Table 1. Low carbon carryover in the hopper ash was measured by loss on ignition (LOI) .

Table 1 Average Flue Gas Composition

Flue Gas Component	Test I	Test II
NO _x , ppm	< 200	< 350
SO ₂ , ppm	2,370	2151
CO ₂ , %	15.4	14.27
O ₂ , %	3.28	4.1
LOI, %	2.3	3.0
Particulate, lb/10 ⁶ Btu	3.82	1.52

Multiple pulverized coal samples were collected throughout each test day to generate a daily composite coal sample. Coal samples were isokinetically collected downstream of the pulverizer with an aspirator and cyclone collector in accordance with ASTM D197-87 “Standard Test Method for Sampling and Fineness Test of Pulverized Coal” and ASME Performance Test Code PTC 4.2 “Pulverized Coal Sampling”. The average as-fired mercury and chlorine concentrations in the Ohio 5 & 6 coal blend fired in Phase I and Phase II are compared in Table 2. The percent relative standard deviation of the mercury and chlorine content of the Ohio coal is 20% or less, reflecting a fairly uniform product from the single coal supplier over time. As NIST mercury certified coals are not currently available, the accuracy of the mercury analysis procedure was determined by the analysis of certified European coal standards. Coal mercury recovery ranged between 97 - 103% for the European coal standards during Phase II.

Table 2 Mercury and Chlorine Content of Ohio 5/6 Coal Blend, ppm

Analyte	Phase I	Phase II, Test I	Phase II, Test II
Mercury	0.24 ± 0.05	0.25 ± 0.02	0.17 ± 0.04
Chlorine	1,154 ± 30	1,018 ± 288	1,327 ± 40

Sampling and Analytical Procedures

Mercury and particulate emissions were the primary targets of the first test series. In addition, other trace metals (arsenic, cadmium, chromium, and selenium) were selectively analyzed to provide ESP and baghouse performance data. Sampling time was generally 4 hours to ensure quantification (at levels at least 10 times the detection limit) of trace metals that are well controlled across particulate devices. The 4-hour sample time was twice as long as necessary to quantify the mercury emissions. Sampling time was reduced to two hours during the second test series.

Sampling was conducted according to EPA Method 29 and Ontario Hydro procedures. EPA Method 29 was selected as the current EPA-approved grab sampling method for total mercury and other metals. The Ontario Hydro method, which has been evaluated by several organizations to provide an improved measure of the elemental and oxidized mercury species relative to EPA Method 29, was most frequently employed. The Ontario Hydro method was selected based on initial reports that Ontario Hydro total mercury emissions were comparable to Method 29 and the ease of sample recovery relative to Method 29. To leverage operating time, an independent sampling contractor (ATS, Inc.) performed near simultaneous measurements along side McDermott Technology personnel.

In Method 29, oxidized mercury is collected in the initial impingers (nitric acid/peroxide) and the remaining elemental mercury is collected in the final impingers (potassium permanganate solution). It is suspected that the nitric acid/peroxide may oxidize the elemental mercury resulting in an over-reporting of the oxidized mercury fraction. In the Ontario Hydro (OH) method, the initial impinger solutions of Method 29 are modified. Potassium chloride (KCl) is substituted for the nitric acid/peroxide solutions to capture the oxidized component. This modification is believed to minimize the potential for oxidation of elemental mercury in the peroxide impinger solutions and provide a better measure of the mercury species distribution in the flue gas.

The evaluation of alternative sampling methods to quantify mercury species in coal-fired flue gas is the subject of several on-going research projects funded primarily by the DOE and EPRI. Inherent limitations of Method 29 for quantifying mercury species have been recognized and are driving the development of alternative sampling techniques. EPA Method 29, however, is still a benchmark for comparison of total mercury emissions for alternative methods.

Observations on the mercury sampling methods include:^[1]

During the OH recovery procedure, a substantial portion of the mercury can be lost. To minimize mercury loss primarily from the KCl impingers, the use of a preservative (permanganate, dichromate, or peroxide) is recommended.

The amount of mercury captured on the sampling filter, and therefore the amount reported as particulate-phase mercury, is influenced by filter temperature.

Oxidation of elemental mercury across high sulfur, bituminous coal flyash is suspected.

Characterization of wet scrubber mercury behavior with Method 29 often results in higher elemental mercury emissions measured at the scrubber outlet relative to the inlet. This has been attributed to the higher SO₂ levels at the scrubber inlet that result in an over-reporting of oxidized mercury.

Many of these phenomena were observed at a scale between 0.1 - 4 MW_e and frequently with simulated flue gas combined with mercury spiking. As discussed, similar phenomena at the 10 MW_e scale has been independently observed with unaltered coal-fired flue gas.

Methods Discussion

Total mercury emissions from the boiler and at the baghouse outlet as measured by EPA Method 29 and the OH method during the first test series are compared in Figure 2. Mercury emissions are reported in $\mu\text{g}/\text{dscm}$ normalized to 3% O_2 . The lower mercury levels measured by the OH method compared to Method 29 coincide with observations made by other researchers when dichromate is used as a preservative.

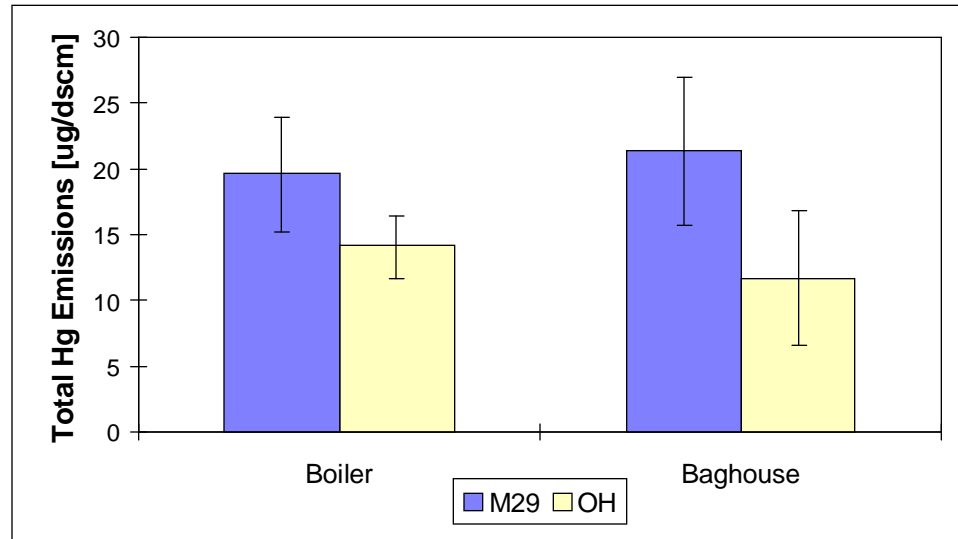


Figure 2 Method 29 and Ontario Hydro Comparison - Dichromate Preservation

Ontario Hydro Preservation

The lower mercury emissions measured by the OH method were primarily due to the consistently lower oxidized mercury concentrations (compared to Method 29) suggesting a loss of mercury from the OH KCl impingers. In the absence of an oxidizing species such as peroxide, oxidized mercury could be reduced to more volatile elemental mercury. A number of preservatives (permanganate, dichromate, or peroxide) have been recommended for use in the OH KCl solutions to retard mercury loss during recovery.^[1] For the first test series, dichromate was used as a preservative. The results of several in-house test programs suggested a correlation of the time between the completion of sampling and the addition of preservative to the amount of mercury loss from the OH impinger solutions which have been since confirmed by other researchers including the Energy & Environmental Research Center (EERC), CONSOL and Carnot.^[2] Immediate preservation after leak checking and rapid analytical recovery will reduce mercury loss. In second test series, permanganate was selected as the preservative on the basis of the resulting visual color change and stronger oxidizing properties than dichromate. As illustrated in Figure 3, the use of permanganate preservative during the second test series resulted in improved agreement between the two methods.

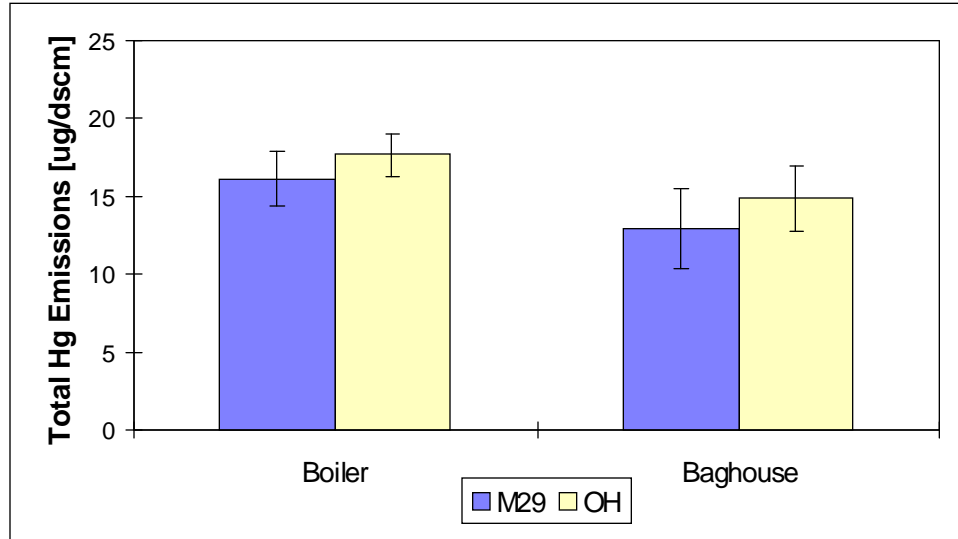


Figure 3 Method 29 and Ontario Hydro Comparison - Permanganate Preservation

Filter Temperature Impacts

The impact of the filter temperature on the amount of mercury measured on the particulate at a constant flue gas temperature of 300 °F is shown in Figure 4. Even though the filter temperatures were maintained within the protocols of EPA Method 29 (248 ± 14 F), the fraction of mercury measured on the particulate increased with incremental reductions in filter temperature for the Ohio 5 & 6 coal flyash. To overcome impacts of filter temperature on mercury speciation results, the filter should be maintained at the flue gas temperature. Similar observations for other coal fly ashes have been reported by other investigators including EERC and Radian. In the case of measurements downstream of a wet scrubber, the filter temperature should probably be maintained at 250 °F as prescribed by EPA Method 29.

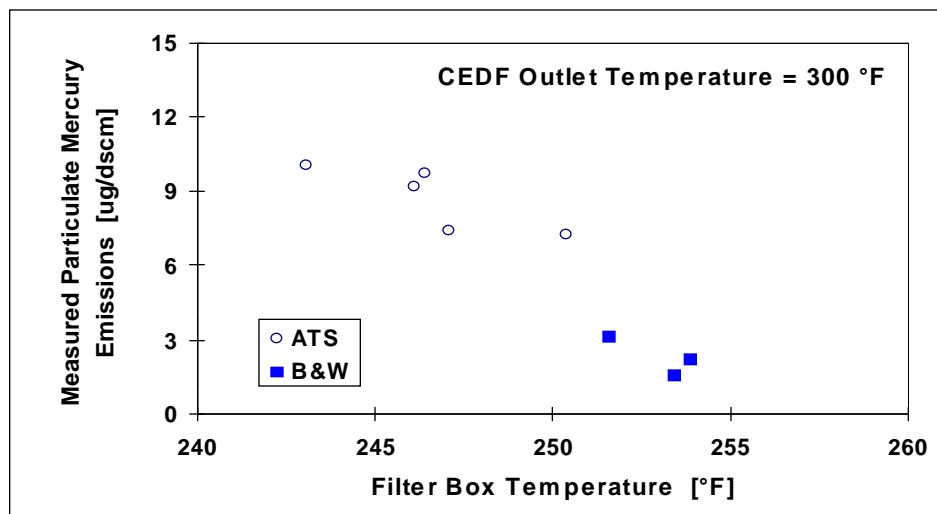


Figure 4 – Impact of Filter Box Temperature on Mercury Particulate Emissions

Mercury Emissions Control

Pulse-Jet Baghouse

Two fabrics, Ryton and GORE-TEX®, were evaluated for combined particulate and mercury emissions control in the first test series. Particulate emissions lower than the current New Source Performance Standard of 0.03 lb/million Btu were readily achieved with both the Ryton and GORE-TEX® fabrics. The Ryton fabric reduced particulate emissions to less than 0.01 lb/million Btu over a range of air-to-cloth ratios (3 - 4.3 ft/min) and baghouse operating temperatures (240 - 300 °F). The GORE-TEX® fabric provided significant improvement in particulate control. Particulate penetration through the GORE-TEX® fabric was less than 0.005 lb/million Btu over a similar range of air-to-cloth ratio and baghouse operating temperature.

The majority of the mercury emissions exiting the baghouse and entering the wet scrubber were measured as the oxidized form. However, due to the possible loss of oxidized mercury in the OH KCl impinger solutions and presumed inability of Method 29 to adequately speciate at high SO₂ concentrations, the discussions of mercury behavior observed during the first test series will be generally limited to elemental mercury as measured by the OH method and total mercury as measured by Method 29.

The elemental mercury emissions entering the slipstream baghouse averaged 2.8 ± 1.0 µg/dscm as measured by both sampling teams in the flue gas temperature range of 300 - 350 °F. Table 3 compares the average elemental mercury emissions exiting the baghouse based on OH measurements for both fabric types. The number within the parentheses signifies the number of replicate measurements. The results from both sampling teams suggest that baghouse fabric type does not impact elemental mercury emissions. Elemental mercury levels downstream of both fabrics were consistently lower than measured at the baghouse inlet, suggesting a transformation or removal of elemental mercury. Radian has also reported elemental mercury conversion across Ryton fabric.^[3] Since the elemental mercury emissions were comparable between fabrics (for each sampling team) the transformation or removal is likely due to the high-sulfur bituminous coal flyash. Although suspect due to loss of oxidized mercury from the OH KCl solutions, higher levels of oxidized mercury were generally measured at the baghouse exit than at the inlet, supporting the possible oxidation of elemental mercury across the slipstream baghouse coal flyash filter cake.

Table 3 Baghouse Mercury Emissions - Ontario Hydro

Elemental Mercury Emissions		
Fabric	ATS Measurements [µg/dscm]	B&W Measurements [µg/dscm]
Ryton	0.27 ± 0.19 (6)	1.61 ± 0.51 (5)
GORE-TEX®	0.36 ± 0.18 (4)	1.26 ± 0.57 (3)

Similar results based on OH sampling observed in the second test series while firing the same Ohio 5 & 6 coal blend are presented in Figure 5. On the basis of replicate preserved OH sample trains, 11% of the total mercury was present on the particulate entering the baghouse. On average, 76% of the vapor-phase mercury was measured as the ionic form. Total mercury removal across the pulse-jet baghouse averaged 16% indicating a small amount of mercury adsorption onto the baghouse filtercake. The primary mercury control mechanism for a conventionally operated baghouse appears to be the conversion of elemental mercury by the flyash filtercake. The combination of a baghouse and wet scrubber has a high potential for total mercury emissions reduction due to the conversion of elemental mercury to a more soluble form. Other benefits would include compliance with phase II SO₂ requirements and potential fine particulate regulations.

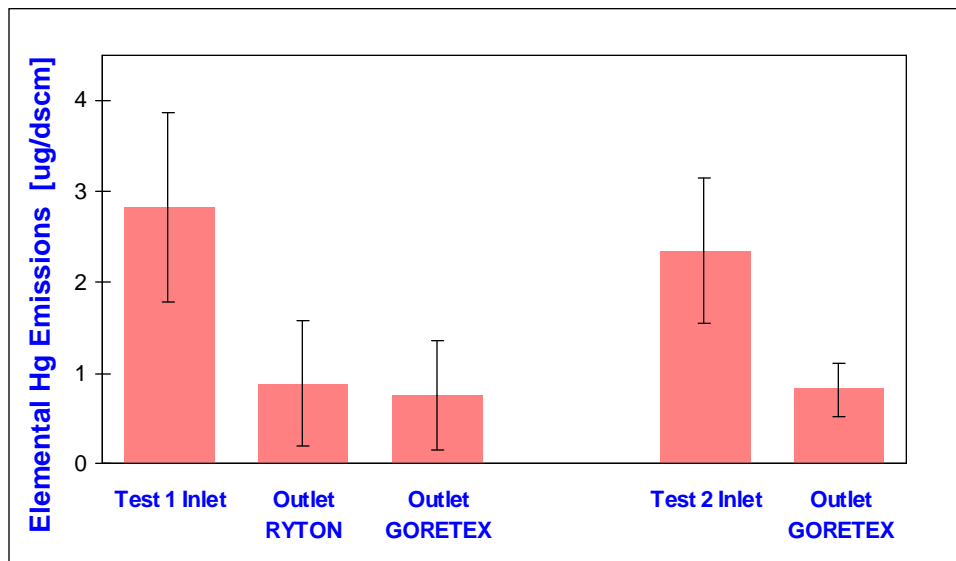


Figure 5 Elemental Mercury Transformation across Pulse-Jet Baghouse

Electrostatic Precipitator

The CEDF test facility provides the opportunity for direct comparison of ESP and baghouse performance. Elemental mercury emissions entering the ESP (and baghouse) were measured at 2.8 ± 1.0 $\mu\text{g/dscm}$ throughout the first test period as previously discussed. The average elemental mercury emissions (based on OH sampling) from the ESP as a function of ESP outlet temperature are presented in Table 4. Unlike the elemental mercury behavior across the pulse-jet baghouse, elemental mercury passed unaffected through the ESP over operating temperatures of 250 - 300 °F. The ESP temperature was controlled with a flue gas cooler or by flue gas humidification. Total ESP vapor-phase mercury removal as measured by Method 29 was negligible at an ESP temperature of 330 °F.

Table 4 ESP Mercury Emissions - Ontario Hydro

Elemental Mercury Emissions			
Location	ESP Inlet	ESP Outlet @ 250 °F	ESP Outlet @ 300 °F
[µg/dscm]	2.8 ± 1.0 (4)	3.4 ± 1.2 (4)	3.2 ± 1.1 (3)

Wet Scrubber

The second test series focused on total mercury emissions control using a limestone wet FGD system. Pilot and commercial scale tests have indicated that wet FGD systems have the potential for high mercury emissions control efficiency. FGD systems are currently installed on approximately 20% of the U.S. coal-fired generating capacity and, therefore, represent a mercury emission control option with a proven history of commercial operation.

The wide range of mercury emissions control performance reported in the literature (10-90%) for wet scrubbers is summarized in Table 5. The EPA interim final report on hazardous air pollutant (HAP) emissions from fossil-fired electric utility steam generating units presented a median mercury emissions control efficiency of only 17% for wet scrubbers and a range of 0 to 59% based on sampling at five commercial plants. The characterized wet scrubber population does not reflect the mercury emissions control potential of the existing commercial units in the eastern U.S.^[3] Bituminous coal is fired at only two of these plants. Four of these scrubbers have an open spray tower design and the fifth is a one-of-a-kind U.S. installation of the Chiyoda jet bubbling reactor (JBR) system. Three of the five units are designed for 60% SO₂ removal or less. All of the units were designed to operate at an L/G of less than 70 with one unit designed for an L/G of 22. In current commercial practice, a design L/G of 90 to 100 is typical for a limestone forced oxidation FGD system designed for 90 to 95% SO₂ removal efficiency.

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 sound, scientific basis for the EPA to assess the need for regulation of mercury emissions from coal-fired utilities. The results of the second AECDDP test series are intended to contribute to this data base.

The pilot wet scrubber was operated in several modes to simulate existing commercial scrubber installations. The scrubber configurations and number of operating conditions evaluated were limited to four primary target design and operating conditions. These conditions were selected to represent the design and operating characteristics of a majority of the existing U.S. population of commercial scrubber installations. Mercury emissions from the slipstream wet scrubber were measured using the Ontario Hydro method (with permanganate preservation).

The bulk of existing commercial wet scrubbers in the U.S. are either open spray towers or have a perforated tray installed to distribute the gas flow over the cross section of the scrubber. The tray also provides a region of relatively lower slurry pH which may impact the absorption of mercury

Table 5 Mercury Control Summary - Flue Gas Desulfurization

FGD System (Coal Type)	Data Source	Reported Total Mercury Emissions Reduction (%)	
		Range	Average
Wet FGD (Bituminous)	AECDP Pilot	20 - 97	
	EPRI Pilot ^[5,6]	85 - 96	
	Radian ^[6]		50
	SRI ^[7]		45
	KEMA ^[8]	8 - 72	52
	USEPA	0 - 59	17
	USDOE ^[9]		44
	Consol ^[10]	50 - 77	62
	EPRI ^[9]		84
Wet FGD (Sub-bituminous)	EPRI ^[9]	10 - 69	
Dry FGD (Bituminous)	AECDP Pilot	60 - 65	

from the flue gas or the retention of absorbed mercury in the slurry. Mercury control performance was evaluated over a range of pH and L/G ratio for tray and open tower operation. As illustrated in Figure 6, the inclusion of the tray enhanced total mercury control. This is not an unexpected finding, as operation with the tray also provides for incremental improvements in SO₂ removal. The major contribution towards the improvement in total mercury removal was the reduction of soluble oxidized mercury emissions (Figure 7) when the tray was installed. Elemental mercury emissions were generally unaffected by tray configuration.

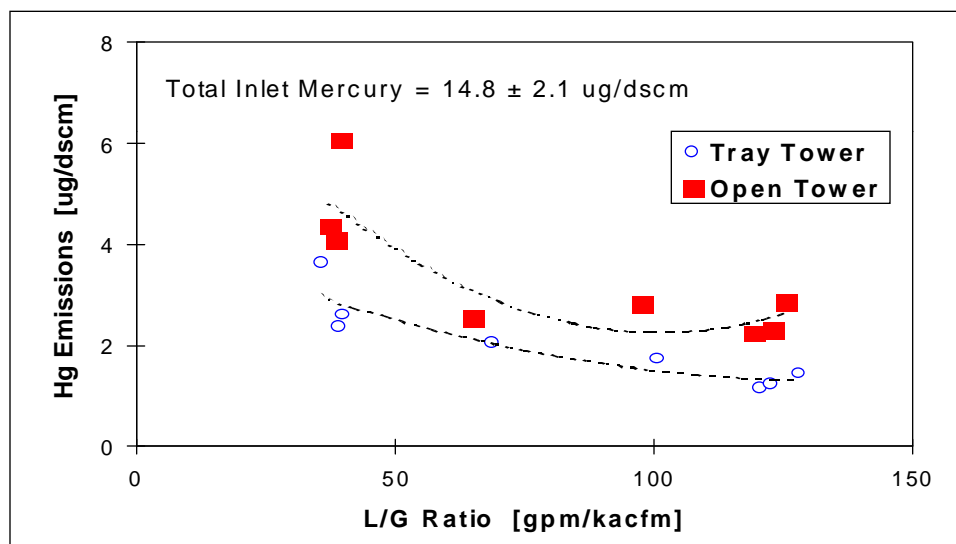


Figure 6 Impact of Tray on Total Mercury Emissions - Wet Scrubber

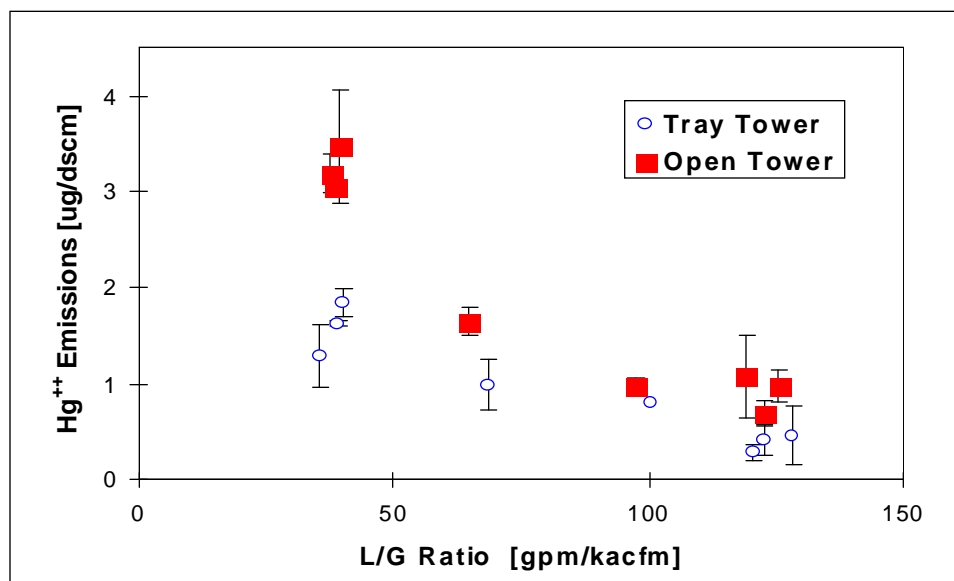


Figure 7 Impact of Tray on Oxidized Mercury Emissions - Wet Scrubber

Figure 7 also illustrates the reduction in oxidized mercury emissions as a function of L/G ratio. For operation with the tray, the average oxidized mercury removal increased from 80 to 91% as the L/G ratio was increased from approximately 40 to 120 gpm/kacfm. During the open tower tests, the average oxidized mercury removal increased from 67 to 83 % with increased L/G ratio.

Total Mercury Control Summary

In summary, the total mercury emissions measured from the various particulate and SO₂ control devices during the first test series are illustrated in Figure 8. Emissions from the particulate control devices, primarily in the oxidized form, were within the measured range of mercury emissions from the boiler. On the basis of Ontario Hydro sampling, total mercury removals greater than 60% were obtained across the wet scrubber system conservatively operated downstream of the pulse-jet baghouse. Mercury emissions from the wet scrubber were mainly in the elemental form due to the consistently high levels of oxidized mercury removal (greater than 94%).

The total mercury emissions and corresponding mercury control measured for the baghouse and wet scrubber devices during the second test series are provided in Figure 9. The decrease in the uncontrolled mercury emissions from the boiler relative to the first test correlates to a reduction in the coal mercury content. The low level of total mercury removal measured across the baghouse (less than 15%) was associated with the mercury present on the particulate. The high percentage of oxidized mercury emissions from the baghouse resulted in mercury emission control across the wet scrubber greater than 80%. Data from the second test series indicate that wet scrubber configuration (tray) and operation (L/G ratio) impact mercury emissions. Operation with the tray tower resulted in lower SO₂ and mercury emissions at all conditions relative to the open spray tower. Overall total

mercury control was significantly higher than the median 17% total mercury removal as reported for wet scrubbers in the EPA interim final report on hazardous air pollutant (HAP) emissions from fossil-fired electric utility steam generating units. The results of the second test series suggest that the EPA interim final report understates the potential for mercury emissions control for commercial wet scrubbers treating flue gas generated from high-sulfur bituminous Ohio coal.

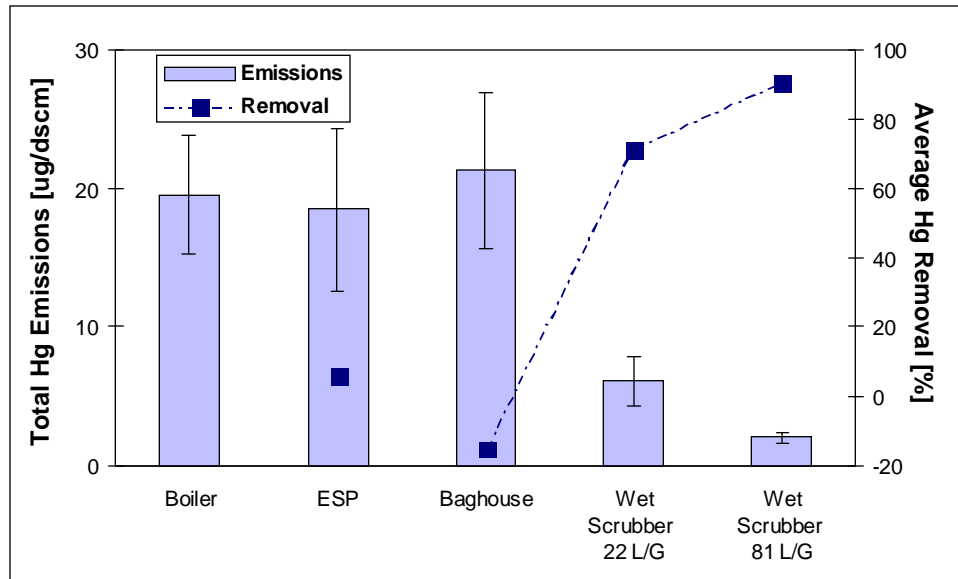


Figure 8 Mercury Emissions Control Summary for Ohio 5/6 Coal - Test I

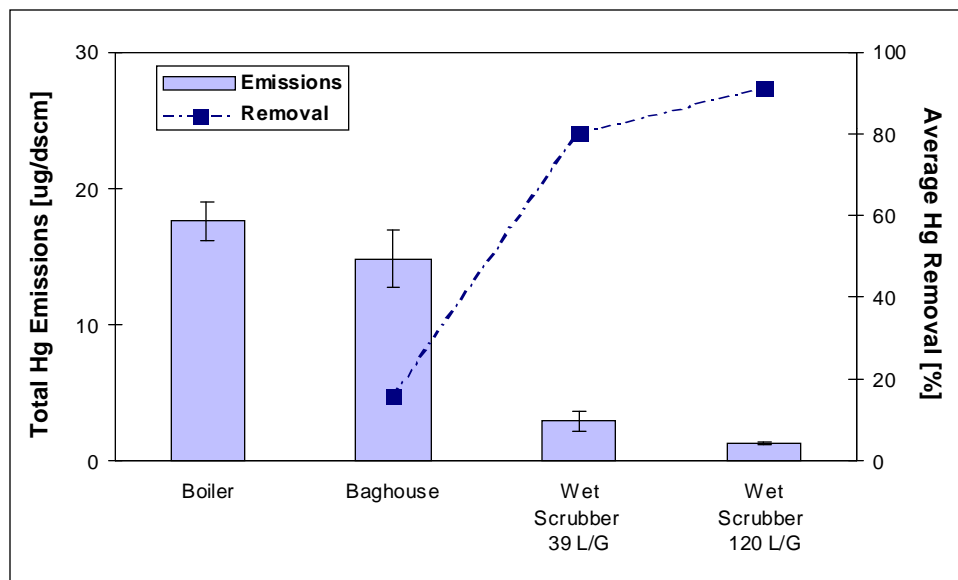


Figure 9 Mercury Emissions Control Summary for Ohio 5/6 Coal - Test II

Acknowledgments

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