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Mercury Mass Balances: A Case Study of Two North Dakota Power Plants

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ABSTRACT

The Energy & Environmental Research Center (EERC) conducted a mercury-sampling program to provide data on the quantity and forms of Hg emitted and on the Hg removal efficiency of the existing air pollution control devices at two North Dakota power plants—Milton R. Young Station and Coal Creek Station. Minnkota Power Cooperative, Great River Energy, the North Dakota Industrial Commission, and EPRI funded the project. The primary objective was to obtain accurate measurements of Hg released from each plant, as verified by a material balance. A secondary objective was to evaluate the ability of a mercury continuous emission monitor (CEM) to measure total Hg at the stack.

At both plants, speciated Hg measurements were made at the inlets and outlets of both the electrostatic precipitators (ESPs) and the flue gas desulfurization (FGD) systems. A Semtech Hg 2000 (Semtech Metallurgy AB) mercury CEM was used to measure the total Hg emissions at the stack in real time. Using these measurements and plant data, the measured Hg concentrations in the coal, FGD slurries, and ESP ash, a Hg mass flow rate was calculated at each sampling location. Excellent Hg mass balances were obtained (±15%). It was also found that the Hg was mostly in the elemental phase (~90%), and the small amount of oxidized Hg that was generated was removed by the FGD systems.

Insignificant amounts of particulate-bound Hg were measured at both plants. However, 10–20% of the elemental Hg measured prior to the ESP was converted to oxidized Hg across the ESP. The data show that, at these facilities, almost all of the Hg generated is being emitted into the atmosphere as elemental Hg. Local or regional deposition of the Hg emitted from these plants is not a concern. However, the Hg does become part of the global Hg burden in the atmosphere. Also, the evidence appears to indicate that elemental Hg is more difficult to remove from flue gas than oxidized Hg is.

IMPLICATIONS

As part of the *Study of Hazardous Air Pollutant Emissions from Electric Utility Steam Generating Units* and the *Mercury Study Report to Congress*,[1] the U.S. Environmental Protection Agency (EPA) concluded that Hg emitted from coal-fired utilities is a potential threat to human health. However, EPA also concluded that additional research/information is necessary before a determination can be made regarding regulation of Hg emissions from power plants. Toward that effort, this paper provides data on the quantity and forms of Hg emitted by two North Dakota power plants and the Hg removal efficiencies across their air pollution control devices.

INTRODUCTION

The 1990 Clean Air Act Amendments required the U.S. Environmental Protection Agency (EPA) to determine whether the Hg emitted from fossil-fuel-fired electric utility power plants poses an unacceptable public health risk. Congress required EPA to produce two mercury reports. These two reports, *Study of Hazardous Air Pollutant Emissions from Electric Utility Steam Generating Units* and the *Mercury Study Report to Congress*,[1] were issued late in 1998. The reports concluded that burning coal was the primary source of anthropogenic Hg emissions in the United States, accounting for 46% of the total of 143,500 kg/yr. The reports indicated that EPA views Hg as a potential threat to human health. They also concluded that there were no cost-effective Hg control technologies available for coal-fired

electric power stations. It was clearly stated that additional research is needed to develop new, more cost-effective Hg control strategies and a better understanding of the forms of Hg emitted from coal-fired power plants.

There is a general consensus that assessing the risk posed by power plant Hg emissions is very complicated.[2–7] Mercury is emitted in such small amounts that accurately measuring emission rates has been extremely difficult. In addition, researchers discovered that Hg is emitted in various physical and chemical forms, each possessing distinct properties that affect sampling and analysis methods.[8–10] It is only in the last two years that techniques have been developed to accurately measure speciated Hg in flue gas.[9] Accurate measurements of Hg emissions from fossil-fuel-fired power stations into the environment are required to meet a variety of needs, including

- estimating the anthropogenic flux of Hg to the environment on a local, regional, and global scale;
- identifying atmospheric transport and transformation processes;
- evaluating the removal efficiency of control technologies;
- determining partitioning among the various effluents of fossil fuel combustion systems; and
- ensuring compliance of sources with emission regulations, should they be promulgated.

Mercury emissions from anthropogenic sources occur in three main forms: solid particle-associated mercury, gaseous divalent mercury (Hg^{2+}), and gaseous elemental mercury (Hg^0). Until recently, estimates of the relative proportions of these species in fossil-fuel-fired power plant emissions and in the ambient air were scarce because of the lack of reliable sampling and analysis methods for the different Hg species.

In an effort to more accurately determine the amount and type of Hg being emitted into the atmosphere by coal-fired power plants, in late 1998 EPA issued an Information Collection Request (ICR).[10] Two primary activities required by the ICR impacted the coal-fired electric utility industry. First, all coal-fired plants were required to submit coal analyses that include Hg to EPA on a quarterly basis. Second, about 80 power stations were required to measure the speciated Hg concentration in the flue gas. These plants were selected on the basis of plant configuration and coal type.

In an effort to provide data on the fate of Hg from North Dakota power plants, a consortium was established with the Energy & Environmental Research Center (EERC) and the following groups:

- Minnkota Power Cooperative, Inc., the operator and part owner of the Milton R. Young (MRY) Power Station located near Center, ND;
- Great River Energy, the owner and operator of the Coal Creek (CC) Station located near Underwood, ND;
- North Dakota Industrial Commission;
- U.S. Department of Energy; and
- EPRI.

OBJECTIVES

The overall goal of the project was to determine the concentration of speciated Hg emitted from two North Dakota power plants and then relate these emissions to the concentration of Hg in sediments as a function of geological time in one of the lakes downwind from the plants. A final project report was issued in January 1999.[12] This paper focuses on the concentrations of Hg across the air pollution control devices and the forms of Hg emitted from the two North Dakota power plants. Specific objectives relative to the two stations were to

- determine the speciated Hg emissions from the coal-fired electric generating stations;
- evaluate the ability of a continuous emission monitor (CEM) to measure total Hg at the stack;
- determine Hg removal efficiencies across the air pollution control devices for each of the Hg species; and
- calculate Hg material balances for each plant and air pollution control device.

UNITS TESTED

As shown in Table 1, the two North Dakota power plants are similar in that they both burn a North Dakota lignite and they have similar air pollution control devices. Electrostatic precipitators (ESPs) are used for particulate removal, and wet flue gas desulfurization (FGD) systems scrub SO_2 from the gas. Differences between the two plants are that the MRY Unit 2 is a wet-bottom cyclone boiler compared with CC Unit 2, which is a pulverized coal-fired system with low-NO_x burners. The stations use different alkaline additives for scrubbing. MRY uses a mixture of fly ash and lime, while CC operates exclusively with lime. At both plants, the wet scrubbers bypass a substantial amount of flue gas. Because North Dakota lignites are relatively low in sulfur, only a portion of the flue gas must be scrubbed to meet SO_2 emission standards. As will be discussed later, the FGD bypass complicates Hg mass balance calculations.

APPROACH

Mercury speciation flue gas sampling was completed at four different locations. These are as follows:

- at the inlet to the ESP;
- at the outlet of the ESP (which is the same as the inlet to the FGD);

Table 1. Plant descriptions.

Station	Boiler Type	Generating Capacity	Coal	Particulate Control	Sulfur Control	NO_x Control
MRY, Unit 2	Wet-bottom cyclone	438 MW	BNI ND lignite	ESP	Wet scrubber	None
CC, Unit 2	Pulverized coal-fired	550 MW	Falkirk ND lignite	ESP	Wet scrubber	Low-NO_x burners with overfire air

- at the outlet of the FGD; and
- at the stack.

Table 2 presents the sampling matrix that was used for the project. All flue gas Hg speciation measurements were made using the Ontario Hydro method, which has been validated at the pilot scale and in field-testing by the EERC.[8] The Ontario Hydro method was selected as the method of choice for the Hg speciation sampling to be done under the ICR. Mercury EPA Method 17 (in-stack filtration) was used at all sampling locations except at the stacks, where the temperature was well below 120 °C and EPA Method 5 (out-of-stack filtration) was used.[13]

At both power plants, a Semtech Hg 2000 (Semtech Metallurgy AB) mercury CEM was used to continuously measure the Hg emissions at the stacks. The Semtech Hg 2000 is a portable Zeeman-modulated cold-vapor atomic adsorption (CVAA) instrument that can monitor Hg^0 continuously. To measure total Hg, all the Hg in the flue gas is converted to Hg^0 in a separate unit prior to being introduced into the Semtech instrument.[14] The operating range of the analyzer is 0.3 mg/Nm^3–20 mg/Nm^3 Hg^0. The Semtech Hg 2000 has been certified by TUEV Rheinland for determining compliance with the German legal limit of 50 mg/Nm^3 for total Hg from waste incinerators.

Table 2. Mercury speciation test program matrix.

Process Stream	Target Mercury Species Hg^0 (elemental)	Hg^{2+} (oxidized)	Total Hg
Flue Gas Sample Streams			
Air preheater inlet	X	X	X
ESP inlet	X	X	X
ESP outlet/FGD inlet	X	X	X
FGD outlet	X	X	X
Stack	X	X	X
Solid Samples			
Coal mills			X
ESP hoppers			X
Liquid Samples			
FGD slurry		X	
Lime/Fly ash slurry			X
Ash sluice water			X
FGD pond return water			X

At both plants, the flue gas flow is divided into two streams immediately after it exits the boiler. The flue gas-sampling activities were completed on only one of the gas streams. It was assumed that there was a good mixing of the flue gas and that the concentration of Hg in each of the gas streams was the same.

Two series of tests were conducted at the MRY Station. During the first test series, the FGD system was operated using only fly ash (Condition A); the second series of tests was conducted with the FGD system using only lime as the scrubbing medium (Condition B). As stated earlier, for the CC Station, lime was the only scrubbing medium used.

Methods for Determining Mercury Material Balances

The total Hg balance across each of the plants was calculated by comparing the rate of Hg entering the plant to the rate of Hg leaving each plant. To express the data on the same basis, all flue gas flow rates were calculated on a moisture-free 3% oxygen basis and are expressed in units of g/hr. The coal feed rate was also calculated on a dry basis.

The Hg input to the system as a result of burning coal was calculated on the basis of the plant coal feed rate and Hg concentration in the coal. The Hg concentrations in the other sources of Hg to the plant, lime, and makeup water were at or near the detection limit of the measurement and were, therefore, insignificant in determining the Hg balance. The Hg flow rate calculations for the various sample locations were based on the measured Hg flue gas concentrations shown in Table 3.

Knowing the FGD slurry volumetric flow rates and the Hg concentration in the slurry allowed the rate of Hg removed from the flue gas stream by the FGD system to be calculated. However, the rate of Hg leaving with the scrubber blowdown was difficult to accurately calculate. The blowdown is controlled by the suspended solids. The actual flow rate was not directly measured, because it was not continuous and occurred several times a day. On the basis of discussions with plant personnel and the measured level of suspended solids in the slurry, the average volume of FGD liquor expelled was estimated and a count was made of the number of times during the day in which this occurred. It is believed that this resulted in a reasonably accurate approximation of the volumetric flow rate of the scrubber blowdown.

Table 3. Flue gas Hg concentrations. All units are $\mu g/Nm^3$.

	MRY–A				MRY–B[a]		CC			
Hg Species	ESP Inlet	ESP Outlet	FGD Outlet	Stack	FGD Outlet	Stack	ESP Inlet	ESP Outlet	FGD Outlet	Stack
Total Hg	9.97	10.04	8.64	10.64	7.81	9.22	12.16	11.27	8.84	10.88
Std. Dev.	0.75	0.59	0.32	0.01	0.67	0.97	1.16	0.51	0.63	1.22
PB-Hg[b]	0.49						<0.01			
Std. Dev.	0.11									
Hg^0	8.91	8.81	8.32	9.56	7.68	8.24	11.16	8.27	8.69	8.94
Std. Dev.	0.50	0.45	0.22	0.01	0.67	0.76	1.37	0.42	0.69	1.21
Hg^{2+}	0.56	1.23	0.31	1.08	0.12	0.99	1.00	3.00	0.15	1.94
Std. Dev.	0.31	0.57	0.11	0.01	0.04	0.23	0.53	0.46	0.10	0.20

[a]ESP inlet and ESP outlet Hg concentrations are the same at MRY Condition A; [b]Particulate-bound Hg.

Milton R. Young Station. At most coal-fired power plants, the fly ash collected in the ESP is either sold as a combustion by-product or landfilled. In either case, the Hg captured by the fly ash must be considered in the mass balance equation. However, during normal operation (Condition A) at the MRY Station, a substantial amount of fly ash generated is recycled back to the FGD unit. The exact percentage of fly ash used for scrubbing purposes by the FGD system is not measured by the plant and varies over any given time. However, a high percentage of the fly ash is typically recycled, and the amount of Hg measured in the ash was very low. Therefore, the Hg in the fly ash was not included in the overall Hg balance. This does impart a small low bias to the overall Hg mass balance.

Because the only sampling ports available to measure the Hg concentrations were located prior to the bypassed flue gas being reintroduced to the system, to complete the Hg mass balance across the FGD system it was necessary to know the percentage of the flue gas bypassing the FGD. The amount of flue gas bypassing the FGD is a function of the need for flue gas reheat, coal quality, and compliance with permitted SO_2 emission rates. The actual flue gas bypass flow rate is not directly measured, so the bypass flow rate was calculated using an energy balance. It should be noted, however, that the energy balance was very sensitive to the gas temperature and flue gas moisture content.

On the basis of the energy balance calculation, the flue gas bypass for MRY was 60% for Condition A and 90% for Condition B. The high level of bypass at Condition B was due to the higher reactivity of lime compared with that of fly ash. Very little scrubbing was necessary to meet SO_2 emission standards.

Coal Creek Station. The fly ash collected by the ESP is either sold or landfilled, depending on the season. To obtain the rate of Hg leaving with the ESP ash, the amount of ash removed was calculated on the basis of ESP inlet and outlet dust-loading data. Then, on the basis of the known Hg concentration in the ash, the Hg leaving the system via the ESP was calculated.

As was the case at MRY, to calculate the Hg mass balance around the FGD system, it was necessary to do an energy balance. On the basis of the energy balance calculation, the bypass was 55% for CC. At CC, there was an independent measure of FGD flow bypass, and although the plant personnel considered it unreliable, the measured FGD bypass was the same as the calculated value.

RESULTS AND DISCUSSION

Mercury in Coal

Analysis of the two North Dakota lignites showed that the Hg concentration in the lignites was very similar for the two plants. The CC coal averaged 98 ppb (on a dry basis) for six coal samples, with a variation of 7.4 ppb, compared with 104 ppb for 13 coal samples with a variation of 27 ppb at MRY. Although the variation was greater at MRY than at CC, 26 and 8%, respectively, this is still good reproducibility for coal Hg concentrations. For the material balance calculations, the average coal Hg concentration for each plant was used.

Total Plant Mass Balances

Table 4 presents all the Hg mass flow rates entering and leaving the plants. From these data, the Hg mass balances across the entire plant and across each of the air pollution control devices were calculated. For both the MRY and the CC stations, the only source of Hg to the plant is the coal being burned. The Hg in the lime and makeup water were measured and were either at or near the detection level of the instrument and, therefore, insignificant. On the basis of ambient Hg concentrations, the Hg in the combustion air was also insignificant compared with the Hg in the coal.

Table 4. Mercury mass flow rates at each plant. (All Hg mass flow rates are in units of g/hr.)

Plant	Coal	Lime/Fly Ash	Makeup Water	ESP Inlet	ESP Outlet	FGD[a] Inlet	FGD[a] Outlet	Stack	ESP Ash	Blowdown Water
MRY–Condition A	20.76	2.12	18.06	18.06	18.96	7.57	6.27	17.65	0.93	1.83
MRY–Condition B	20.76	<0.03	18.03	18.03	17.19	1.72	1.39	16.86	0.90	0.71
CC	24.20	<0.03	22.91	22.91	22.36	10.05	7.93	21.49	<0.03	2.12

[a]Values were calculated based on the FGD bypass of 60 and 90% for MRY Conditions A and B, respectively, and 55% for CC.

The primary sources of Hg leaving the plant were the stack emissions. Some Hg also left the plant through the scrubber blowdown and with the ESP fly ash. Two other materials that are generally disposed of are the coal "pyrites" and the bottom ash. For both of these materials, the Hg mass flow rate was insignificant. In the pyrites, the Hg concentration was relatively high, but the quantity was very low. As would be expected, the very high temperatures in the boiler resulted in the Hg concentration in the bottom ash being below detection limits. Table 5 presents the overall Hg balance for each plant—93.8% for MRY Condition A, 89.1% for MRY Condition B, and 96.1% for CC.

Mercury Mass Balances across Air Pollution Control Devices

The Hg balances across the ESP and FGD systems are shown in Table 6. The Hg balances across each of these systems were very good (±15%). A summary of all the calculated Hg material balances is shown in Table 7.

Both of the lignites used at these two power plants generate mercury that has a very high percentage of Hg^0 compared with Hg^{2+}. At the inlet to the ESP, 89% of the total Hg was identified as Hg^0 at MRY and 92% at CC. It should also be noted that MRY had ~5% particulate-bound Hg compared with almost zero at CC. As a consequence of the high percentage of Hg^0 in the flue gas, almost all of the Hg generated from burning the coal exited the stack. However, because Hg^0 is not water-soluble, it is not deposited either locally or regionally and can remain in the atmosphere for up to a year.[15,16]

The Hg speciation results across each of the air pollution control devices for the two plants are shown in Figures 1 and 2. Figure 1 shows that some Hg^0 is oxidized to Hg^{2+}, and there is little Hg removed across the ESP. Evidence has been presented in the literature showing that some conversion of Hg species can occur across an ESP.[13] The effect appears to be more pronounced for CC. It is unknown whether this is caused by differences in ESP operation or fly ash chemistry. As expected, Figure 2 shows

Table 5. Overall plant Hg material balance.

	MRY Condition A, g/hr	MRY Condition B, g/hr	CC, g/hr
Inlet Hg			
Hg from coal	20.76	20.76	24.05
Hg from makeup water	<0.03	<0.03	<0.03
Hg from lime	NA[a]	<0.03	<0.03
Outlet Hg			
Hg exiting with stack	17.65	16.86	21.49
Hg in ESP hopper ash	NA[a]	0.90	<0.03
Hg in scrubber blowdown	1.83	0.74	2.10
Outlet/Inlet Balance	93.8%	89.1%	96.1%

[a]Not applicable—no lime was used, and the fly ash was recycled back through the FGD system.

Table 6. Mercury mass flow rates across air pollution control devices.

Hg Source	MRY Condition A, g/hr	MRY Condition B, g/hr	CC, g/hr
Hg at the ESP inlet	18.06	18.03	22.91
Hg at the outlet of the ESP	18.96	17.19	22.36
Hg in the ash	0.93	0.90	<0.03
Outlet/Inlet Balance	110.1%	100.3%	97.6%
Hg at the inlet of FGD	7.57	1.72	10.05
Hg in fresh FGD slurry	0.93	0.90	<0.03
Hg at the outlet of FGD	6.27	1.39	7.93
Hg in scrubber blowdown	1.83	0.71	2.12
Outlet/Inlet Balance	83.4%	111.6%	100.0%

Table 7. Summary of Hg mass balances.

Plant	Total Plant %	ESP %	FGD %
MRY–Condition A	93.8	110.1	83.4
MRY–Condition B	89.1	100.3	111.6
CC	96.1	97.6	100.0

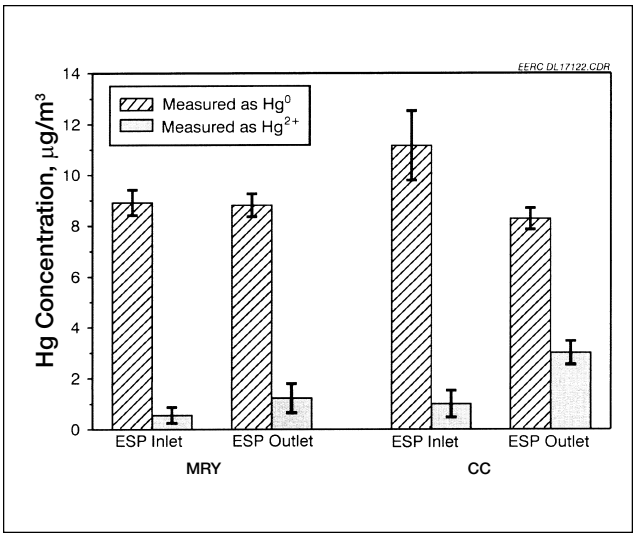

Figure 1. The change in vapor-phase speciated Hg across the ESPs.

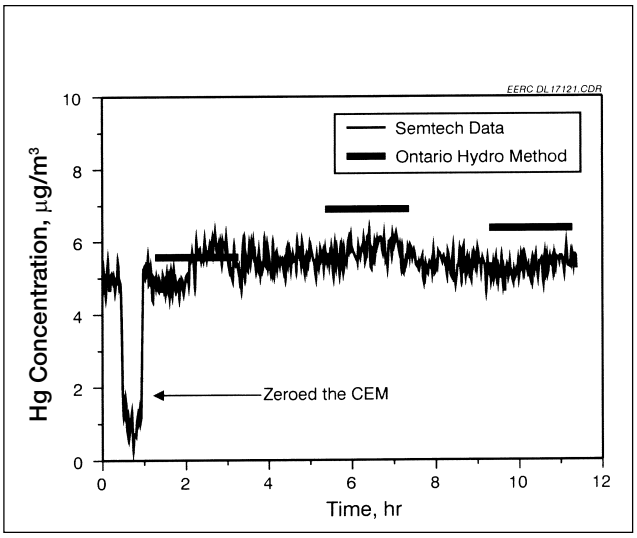

Figure 3. Comparison of the results from a mercury CEM to the Ontario Hydro method results at MRY.

that the FGD systems remove a high percentage of the Hg^{2+} and little, if any, Hg^0.

Comparison of the Mercury CEM Stack Emission Data to the Ontario Hydro Method

At both the MRY and the CC plants, a Semtech Hg 2000 mercury CEM was used to continuously monitor Hg stack emissions. Figures 3 and 4 show that the mercury CEM results compared very favorably with the Ontario Hydro method at both plants. These results demonstrate that mercury CEMs can be used to accurately measure Hg at coal-fired power plants. However, their use is still relatively labor-intensive. To measure the Hg concentration in flue gas from coal-fired systems, it is necessary that a flue gas pretreatment/conversion system be used in conjunction with the mercury CEM.[14] Research is ongoing at the EERC to optimize the flue gas pretreatment/conversion systems.

CONCLUSIONS

- The average mercury-in-coal concentration was about 0.100 ppm (on a dry basis) for the two North Dakota lignites tested.
- At both power stations, the Hg was ~90% Hg^0 at the inlet to the ESPs.
- At both stations, 10–20% of the Hg^0 measured at the inlet to the ESPs was converted to Hg^{2+} across the ESPs.
- The particulate-bound Hg concentration was low at both plants, and at CC it was at or below CVAA detection limits.

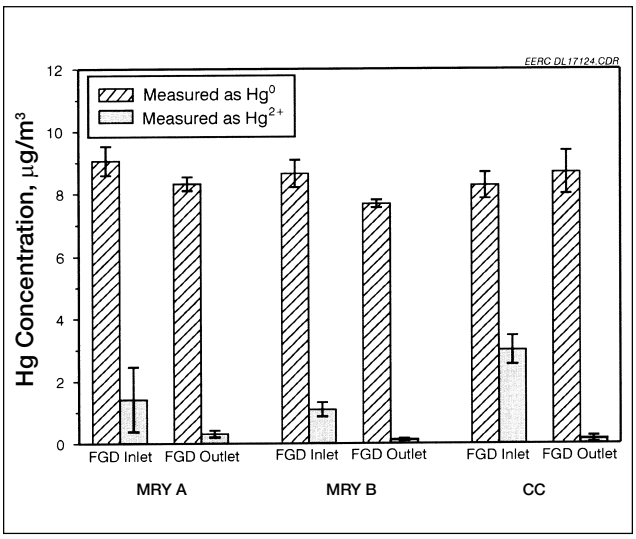

Figure 2. The change in vapor-phase speciated Hg across the FGD systems.

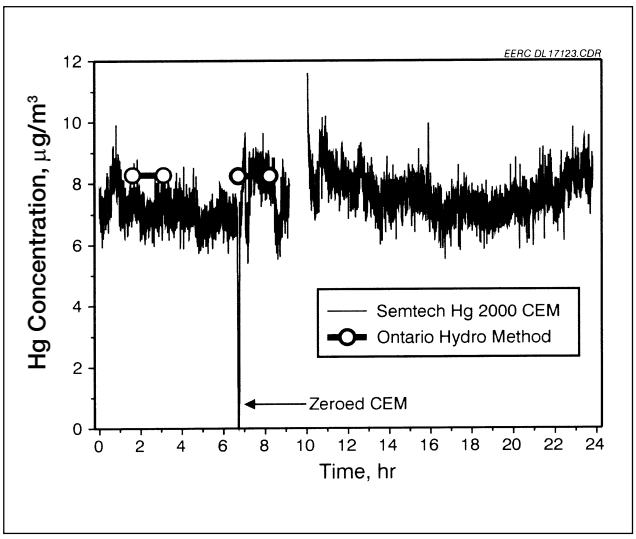

Figure 4. Comparison of results from a mercury CEM to the Ontario Hydro method results at CC.

- Although the Hg^{2+} concentration in the flue gas was low at both plants, the FGD systems removed almost all of it. Conversely, less than 5% of the Hg^0 was removed by the FGD systems.
- The Semtech Hg 2000 CEM (used at the stack) gave Hg results that were highly comparable to those obtained using the Ontario Hydro Hg speciation sampling method.
- Good total Hg mass balances were obtained at both plants: 93.8% for MRY Condition A, 89.1% for MRY Condition B, and 96.1% for CC.
- The Hg mass balances obtained across the air pollution control devices at both plants were excellent. Across the ESPs, the results were 110.0% for MRY Condition A, 100.3% for MRY Condition B, and 96.7% for CC. The Hg mass balances obtained across the FGD systems were 83.4% for MRY Condition A, 111.6% for MRY Condition B, and 100.0% for CC.
- Mercury captured by the scrubber sludge was associated with the solids, with little of the Hg found in the scrubber sludge water.
- Mercury levels in the ash sluice water and ash pond water were very low, at or near the detection limits of the CVAA instrument.

These tests show that good Hg mass balances can be obtained across coal-fired electric utilities and air pollution control devices when great care is taken and sound quality assurance/quality control procedures are followed.

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