

EVALUATION OF MERCURY SPECIATION AT POWER PLANTS USING SCR AND SNCR NO_x CONTROL TECHNOLOGIES

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ABSTRACT

Since the U.S. Environmental Protection Agency (EPA) regulatory determination in 2000 that it intended to regulate emissions from coal-fired power plants, many utilities have stepped up proactive steps to investigate methods to control and reduce emissions. However, a lack of sound data still exists which documents the effect of selective catalytic reduction (SCR), selective noncatalytic reduction (SNCR), and ammonia (NH₃) on the speciation and removal of mercury at power plants. Although both SCR and SNCR systems are effective at nitrogen oxide reduction, each system may impact mercury speciation differently. In addition, some utilities have utilized NH₃ and/or sulfur trioxide (SO₃) to improve electrostatic precipitator performance by changing the resistivity characteristics of the ash.

This project investigates the impact that SCR, SNCR, and flue gas-conditioning systems have on total mercury emissions and on the speciation of mercury. If SCR and/or SNCR systems enhance mercury conversion/capture, then they could be thought of as multipollutant technologies. Data from this project can be used for environmental planning purposes as well as to assist in regulatory decisions. Previous Energy & Environmental Research Center pilot-scale tests investigated the role that coal type plays in mercury speciation with the addition of NH₃ and the use of SCR. The results indicated that SCR and NH₃ may enhance mercury capture, although it appeared that the impact was highly coal-specific. However, there were significant concerns as to the applicability of the pilot-scale results to full-scale power plants. To validate pilot-scale results, sampling must be conducted at the full scale.

Seven power plants were chosen for full-scale sampling to investigate the role that SCR and NH_3 had in mercury speciation. For a 10–12-day period, sampling was conducted both prior to and after the SCR unit or NH_3 injection using both the wet-chemistry Ontario Hydro method and near-real-time continuous mercury monitors. Mercury variability, speciation, and concentration were evaluated. Additional sampling involved the use of EPA Method 26A to test for chlorides, a selective condensation method to measure SO_3 , and EPA Method 27 for NH_3 slip. Fly ash and coal samples were also collected to obtain the mercury balance across the control devices.

Although laboratory analyses and data interpretation are still ongoing, preliminary results indicate that SCRs can assist in converting elemental mercury to oxidized mercury. However, the effect appears to be coal-specific and possibly catalyst-specific. NH_3 injection, whether directly as a gas or indirectly as urea, did not appear to have an effect on mercury speciation and removal.

INTRODUCTION

Coal combustion by electric utilities is a large source of anthropogenic mercury emissions in the United States, according to the U.S. Environmental Protection Agency (EPA).¹ Recent data indicate that the total mercury emissions from coal-fired power plants in the United States are about 45 tons/yr.² Clearly, EPA views mercury from coal-fired utilities as a potential public health concern.³

Mercury emissions from coal-fired boilers can be empirically classified, based on the capabilities of currently available analytical methods, into three main forms: elemental mercury (Hg^0), oxidized mercury (Hg^{2+}), and particle-bound mercury (Hg_p). The concentration of Hg^0 , Hg^{2+} , and Hg_p primarily depends on coal composition and combustion conditions.⁴

During combustion, Hg^0 is liberated from coal. However, depending on the coal type, a significant fraction of the mercury can be oxidized, as well as become associated with the fly ash particles in the postcombustion environment of a coal-fired boiler. Relative to Hg^0 , Hg^{2+} and Hg_p are more effectively captured in conventional pollution control systems, such as flue gas desulfurization (FGD) systems, fabric filters, and electrostatic precipitators (ESPs).^{5–7} The identification of a process for converting Hg^0 to Hg^{2+} and/or Hg_p forms could potentially improve the mercury removal efficiencies of existing pollution control systems.

In addition to mercury, coal-burning power plants are a significant anthropogenic source of nitrogen oxide (NO_x) emissions to the atmosphere. NO_x emissions are an environmental concern primarily because they are associated with acid precipitation, as well as fine-particle and ozone formation. Depending on the size and type of boiler, the 1990 Clean Air Act Amendments require specific reductions in NO_x emissions from coal-fired electric utilities. The most common NO_x reduction strategy is the use of low- NO_x burners. These burners have the capability of reducing NO_x emissions by 40%–60%. However, with possible establishment of particulate matter with an aerodynamic diameter less than $2.5\text{ }\mu\text{m}$, regional haze, and more strict ozone regulations, there is increased incentive to reduce NO_x emissions to a level below what can be achieved using low- NO_x burners. Selective catalytic reduction

(SCR) technology, which can reduce NO_x emissions by >90%, is therefore becoming more attractive, particularly because catalyst costs continue to decrease and the knowledge base for using SCR reactors is expanding. Within the next 5 years, 80–90 U.S. utilities are planning to install SCR units.⁸

Potential Impacts of SCR on Mercury Speciation

SCR units achieve lower NO_x emissions by catalytically reducing NO_x to N₂ and H₂O. Ammonia (NH₃) is the reductant used for the SCR of NO_x. The SCR process is generally performed on metal oxide catalysts such as titanium dioxide (TiO₂)-supported vanadium pentoxide catalysts (V₂O₅). These units are operated at about 340°–400°C (640°–750°F). Laboratory-scale testing indicates that metal oxides, including V₂O₅ and TiO₂, promote the conversion of Hg⁰ to Hg²⁺ and/or Hg_p in relatively simple flue gas mixtures.⁹ In addition, mercury speciation measurements at European coal-fired boilers equipped with SCR reactors have indicated that SCR catalysts promote the formation of Hg²⁺.^{10,11} Therefore, it has been hypothesized that the use of an SCR to reduce NO_x emissions may improve the mercury control efficiency of existing air pollution control devices by promoting Hg²⁺ and/or Hg_p formation. Possible mechanisms that could result in the SCR of NO_x impacting mercury speciation include:

- Catalytically oxidizing the mercury.
- Changing the flue gas chemistry.
- Changing the fly ash chemical composition through changes in flue gas chemistry.
- Increase residence time.

EERC Pilot-Scale Tests

In an attempt to evaluate the effects of SCR on mercury speciation, pilot-scale tests were conducted at the Energy & Environmental Research Center (EERC).¹² Tests were conducted firing four different coals: three bituminous coals and one Powder River Basin (PRB) subbituminous coal. The general conclusion reached based on these tests was that SCR has the potential to impact mercury speciation, but the effects are coal-dependent. Because of the inherent concerns related to small pilot-scale tests (surface area-to-volume ratios, different flue gas chemistries, and time and temperature profiles), it was decided that it was necessary to conduct sampling at full-scale power plants. Therefore, EPRI, the U.S. Department of Energy's National Energy Technology Laboratory, and EPA funded a project with the EERC to conduct mercury sampling at six different power plants with different types of air pollution control devices. These included four plants using SCR, one using selective noncatalytic reduction (SNCR), and one using NH₃ and sulfur trioxide (SO₃) for flue gas conditioning.

DESCRIPTION OF THE POWER PLANTS AND COAL

The units tested ranged from 160 to 1300 MW in size. The coals burned at the plants included two PRB coals, five eastern bituminous coals, and a blended PRB–eastern bituminous coal. The plant configurations of air pollution control devices included SCR reactors, an SNCR injection system, ESPs, a wet scrubber, and a Venturi scrubber.

Information about each of the plants is provided in Table 1. The coal analysis for each unit tested is shown in Table 2.

EXPERIMENTAL APPROACH

The overall objective of the project was to determine the effects of SCR operations on mercury speciation and, ultimately, removal efficiencies for a variety of coals. To achieve this objective for each unit/coal, a sampling plan was developed for various operating conditions so that the effects of the catalyst and the NH_3 injection could be determined separately. The mercury measurements were conducted using the manual Ontario Hydro (OH) method as well as continuous mercury monitors (CMMs). The approach was set up to obtain samples at the exit of the boiler (prior to NH_3 injection), at the stack, and in between each of the air pollution control devices. The testing done at each power plant is described below. At the four sites with SCRs (Sites S1–S4), all the plants were tested as follows:

- SCR operated under normal conditions
- SCR operated with the NH_3 turned off
- SCR bypassed

The one exception was Site S2, where tests could not be conducted with and without NH_3 , so tests were only conducted under normal SCR conditions and with the SCR bypassed. At Site A1, which employs NH_3 and SO_3 conditioning to improve ESP performance, tests were conducted for two different coals with and without NH_3 injection (SO_3 conditioning only). At Site A2, which had an SNCR system (urea injection), tests were conducted firing two different eastern bituminous coals. For both coals, the sampling was conducted with and without urea injection.

MERCURY RESULTS AND DISCUSSION

Site S1

The mercury results for Site S1 are summarized in Figure 1. At the SCR inlet, the mercury is present primarily in the gas phase and as Hg^0 . At Site S1, the fly ash generated resulted in over 60% of the mercury being Hg_p at the ESP inlet for all three tests conditions. This is a relatively high level of Hg_p for a PRB coal and may be caused by the large amount of unburned carbon (15%) in the fly ash.

The mercury oxidation at the SCR inlet and outlet indicates that normal SCR operation increased the percentage of Hg^{2+} from 6% to 26%, yielding a 20 percentage point increase. However, comparing the mercury speciation results at the stack, both with and without SCR, there is little “net” improvement in the percentage of Hg^{2+} at Site S1. At the stack, 20% of the mercury is oxidized with SCR compared to 28% without SCR. Similarly, the Hg^0 is comparable. This small difference of 8 percentage points is probably not significant compared with measurement variability.

Table 1. Information about the power plants tested

Plant	Category	Coal	Boiler Type	Boiler Size, MW	Low-NO _x Burners	Catalyst Vendor and Type	Catalyst Age	SCR Space Velocity, hr ⁻¹	Particulate Control	Sulfur Control
S1	SCR	PRB subbitum.	Cyclone	650	No	Cornetech honeycomb	~8000 hr	1800	ESP	None
S2 ^a	SCR	OH bitum.	Wall-fired	1300	Yes	Siemens/Westinghouse plate	~2500 hr	2125	ESP	Wet FGD
S3	SCR	PA bitum. ^b	Tangential-fired	750	Yes, with overfire air	KWH honeycomb	~3600 hr	3930	ESP	None
S4	SCR	KY bitum.	Cyclone	650	No	Cornetech honeycomb	~3600 hr	2275	Venturi scrubber	Venturi scrubber
A1 Unit A	NH ₃ /SO ₃ gas conditioning	PRB-bitum. blend	Opposed-fired	500	Yes	NA ^c	NA	NA	ESP	None
A1 Unit B	NH ₃ /SO ₃ gas conditioning	PRB subbitum.	Opposed-fired	500	Yes	NA	NA	NA	ESP	None
A2	SNCR	OH bitum. ^b	Tangential-fired	160	No	NA	NA	NA	ESP	None

^a Two identical units sampled.

^b Two different bituminous coals were used.

^c Not applicable.

Table 2. Analysis of coals fired during field tests^a

	S1	S2	S3	S4	A1, Coal 1	A1, Coal 2	A2, Coal 1	A2, Coal 2
Moisture Content, %	27.5	7.6	7.0	10.5	17.3	27.3	6.2	7.3
Ash, %	3.73	11.67	14.04	9.08	7.03	4.77	6.99	8.17
Sulfur, %	0.19	3.90	1.67	2.88	0.61	0.36	2.62	2.64
Heating Value, kJ/kg	20,866	25,827	26,592	26,404	24,818	20,576	29,137	27,679
Mercury, µg/g dry	0.102	0.168	0.400	0.131	0.118	0.115	0.087	0.143
Chlorides, µg/g dry	<60	573–1910	721–1420	350–1280	632–958	110–200	1210–1360	743–1410

^a As received, unless otherwise noted.

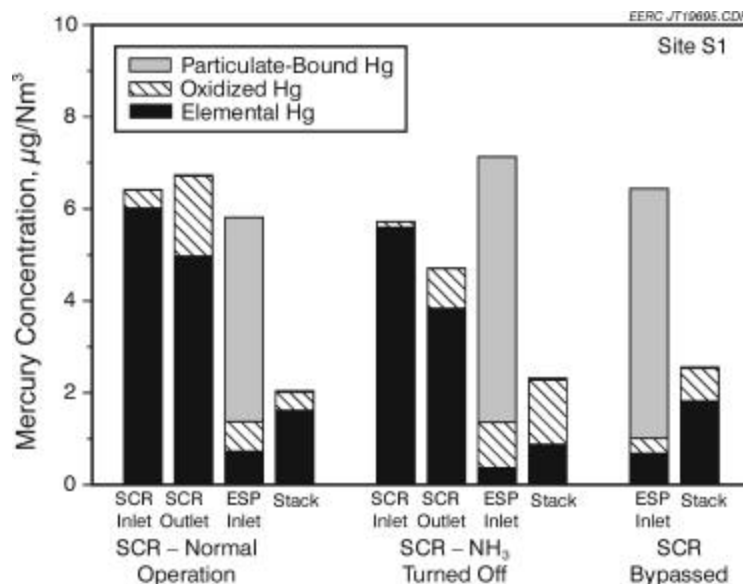


Figure 1. Mercury results showing the effect of SCR on mercury for Site S1.

With the NH₃ turned off to the SCR reactor (but with the flue gas still passing through the SCR reactor), there was no significant effect of SCR on Hg.²⁺

Mercury removal across the ESP was estimated by comparing the ESP inlet total mercury to that obtained at the stack. As shown below, there was a small increase in mercury when the SCR was operating normally. However, the data set is small for PRB coals, and it is not known if this is real or simply data variability.

- Mercury removal across the ESP was 78% when SCR was operating normally.
- Mercury removal across the ESP was 67% when SCR was operating without NH₃.
- Mercury removal across the ESP was 60% when SCR was bypassed.

Site S2

The results for Site S2 are summarized in Figure 2. As can be seen, there was a significant effect of SCR on both Hg²⁺ and its subsequent removal in the wet scrubber. There is an increase in Hg²⁺ with the SCR off-line, but shortly after the SCR is back on-line the concentration of Hg²⁺ decreases again. Comparing the mercury oxidation at the SCR unit inlet and outlet indicates that normal SCR increased the percentage of Hg²⁺ from 48% to 91%, yielding an increase of 43 percentage points across the SCR reactor.

Comparing the mercury speciation results at the ESP inlet with and without SCR shows that 97% of the mercury is Hg²⁺ with SCR, compared to 74% without SCR, which is a net increase of 23 percentage points. This increase in Hg²⁺ led to increased mercury capture in the downstream wet scrubber. Mercury removal across the ESP/FGD is defined by comparing the total ESP inlet mercury concentration to total mercury at the stack. Note that the ESP outlet mercury concentration is greater than the measured inlet concentration using the OH method. It is unknown why this was the case; however, this uncertainty does not

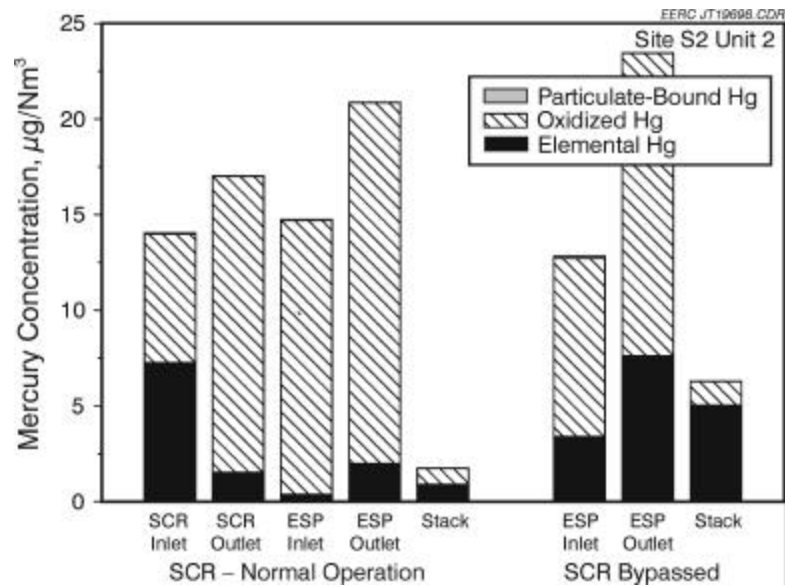


Figure 2. Mercury results showing the effect of SCR for Site S2, Unit 2.

affect the conclusion. Overall, mercury removal increased from 51% to 88% with the SCR reactor in service. In Figure 3, the total mercury results using a CMM are presented. The CMM clearly shows the increase in mercury emissions at the stack when SCR was bypassed.

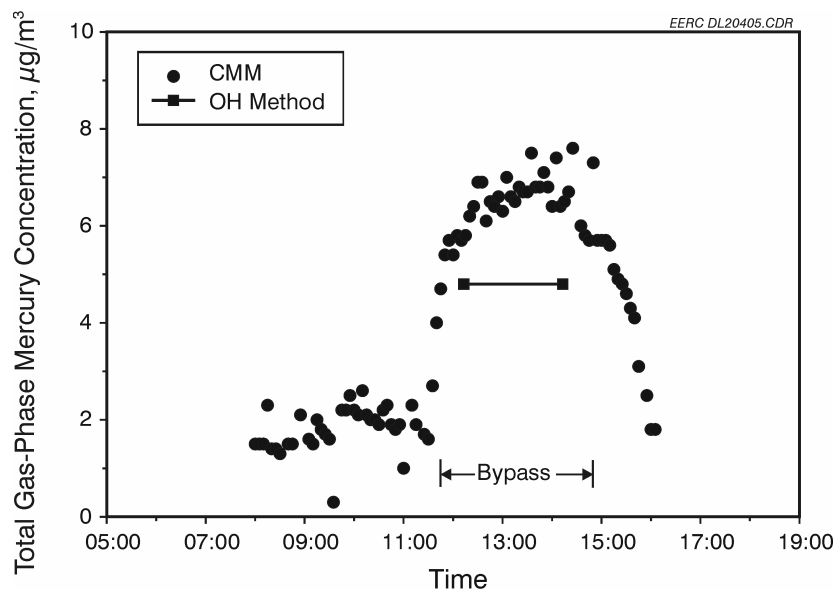


Figure 3. Mercury monitor results showing the effect of bypassing the SCR reactor on total mercury at Site S2.

Site S2, which has a wet scrubber, represents an ideal scenario in that there is clearly oxidation of Hg^0 across the SCR reactor. The result is about a 90% reduction in mercury emissions at the stack, compared to the concentration at the ESP inlet.

Site S3

The mercury results for Site S3 are shown in Figures 4 and 5. From Figure 4, it can be seen that SCR had little effect on overall mercury oxidation. With normal SCR operation, the Hg^{2+} increased from 55% to 65% across the SCR reactor. Comparing the measured mercury speciation at the ESP inlet location both with and without SCR, the Hg^{2+} and Hg_p were similar. These results were somewhat surprising, considering the relatively high chlorine and sulfur content of the coal. One possible contributing factor to the low mercury oxidation across the SCR reactor is that the space velocity of the SCR reactor was at 3930 hr^{-1} , which is nearly double the space velocity for Sites S1, S2, and S4. Space velocity is inversely proportional to residence time, and recent studies have shown that residence time may impact mercury oxidation.¹³ In addition, the inlet mercury concentration was more than double the concentration of the other SCR sites tested. Comparing the total ESP inlet mercury concentration to those obtained at the stack for each of the test conditions gives the following ESP mercury removal results:

- Mercury removal across the ESP was 13% when SCR was operating normally.
- Mercury removal across the ESP was 10% when SCR was operating without NH_3 .
- Mercury removal across the ESP was 16% when SCR was bypassed.

Under these different operating conditions, the results are essentially the same; therefore, based on these measurements, the SCR reactor as currently configured had an insignificant impact on mercury emissions.

A comparison of the OH results and the CMMs is shown in Figure 5. The CMM results support the results obtained from the OH sampling. The error bars for the curve data are based on the standard deviation obtained from the total gas-phase mercury concentrations.

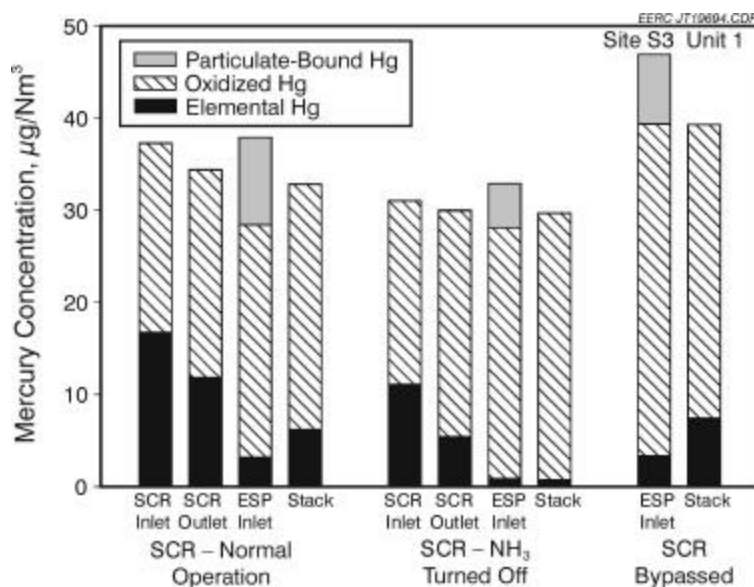


Figure 4. Mercury results showing the effect of SCR on mercury for Site S3, Unit 1.

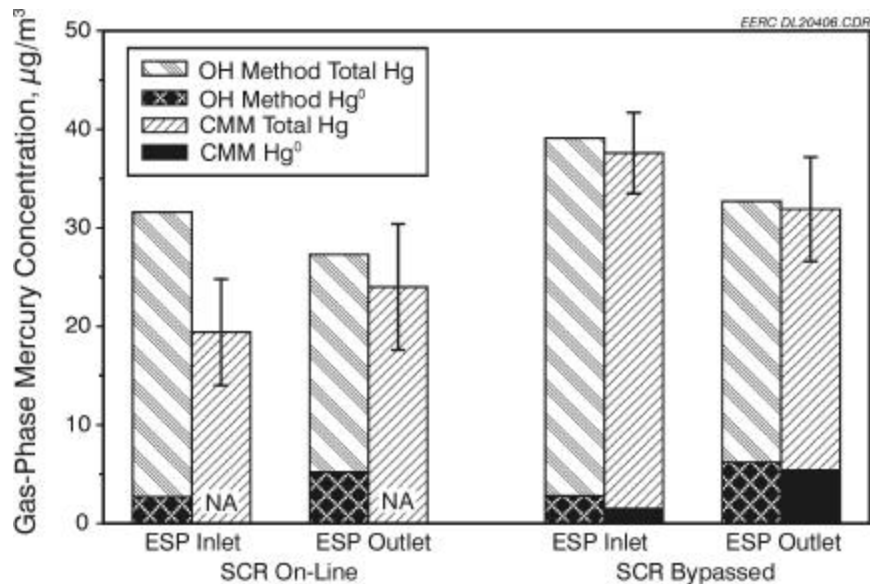


Figure 5. Comparison of a mercury monitor with the OH mercury-sampling method at Site S3 (NA shows Hg⁰ was not measured).

Site S4

The mercury results for Site S4 are shown in Figures 6 and 7. At Site S4, there was significant mercury oxidation and removal by the FGD as a result of SCR operation. Comparing the mercury speciation results (Figure 6) at the air heater outlet (Venturi scrubber inlet) with and without SCR shows that 87% of the mercury is oxidized with SCR in service compared to 56% without SCR in service, yielding a net increase of 31 percentage points. There was also significant oxidation across the SCR reactor when the NH₃ was turned off, with the overall oxidation increasing to 93% (air heater outlet). The mercury oxidation increased from 80% at the SCR inlet to 90% at the SCR outlet.

This increased mercury oxidation also led to increased mercury removal in the Venturi scrubber. Comparing the total air heater outlet mercury concentrations to those obtained at the stack gives the following Venturi scrubber removal results:

- Mercury removal was 90% when SCR was operating normally.
- Mercury removal was 85% when SCR was operating without NH₃.
- Mercury removal was 46% when SCR was bypassed.

In Figure 7, the total mercury results using a CMM are presented. The results clearly show the increase in mercury emissions at the stack when the SCR reactor was bypassed. As was the case at Site S2, this facility represents an ideal scenario in that there is clearly oxidation of Hg⁰ across the SCR reactor and the plant has an FGD system that removes a high percentage of Hg²⁺.

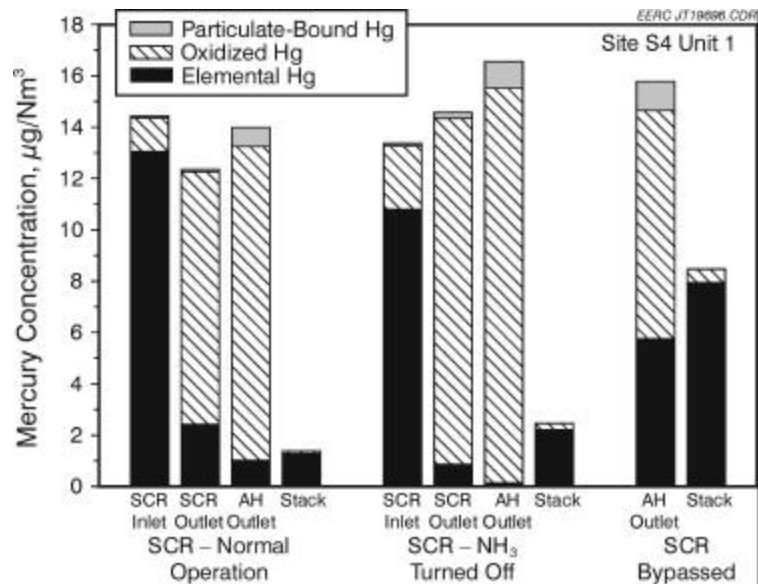


Figure 6. Mercury results showing the effect of SCR on mercury for Site S4, Unit 1.

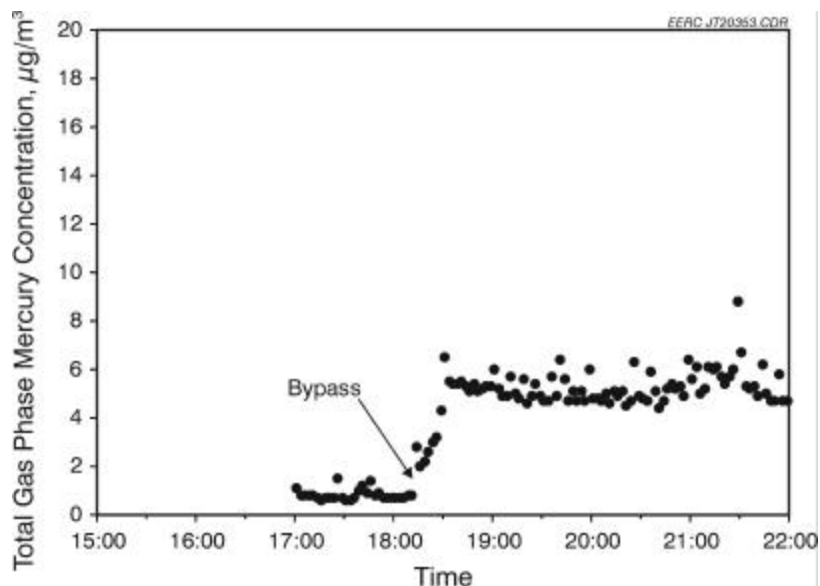


Figure 7. Mercury monitor results showing the effect of bypassing SCR (beginning at 18:20) on total mercury at Site S4.

Site A1

Measurements were conducted at two sister units (A and B) at Site A1. The results for Unit A (50:50 PRB-bituminous blend) are shown in Figure 8. Both with and without NH_3 addition, there is a high percentage of the mercury in the particulate phase. The addition of NH_3 increased the particulate mercury by 29 percentage points (79% compared to 50%). This increase in particulate mercury led to increased mercury removal in the ESP. The resulting

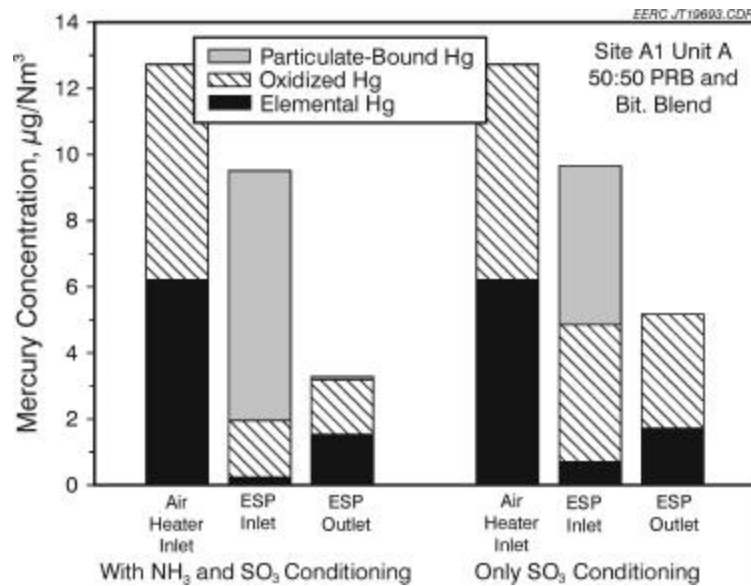


Figure 8. Mercury results showing the effect of adding NH₃ to the flue gas at Site A1, Unit A.

mercury removal efficiencies of the ESP are 66% with NH₃ addition as compared to 46% without NH₃ addition.

For Unit B (100% PRB coal), the mercury results are shown in Figure 9. The mercury is predominantly Hg⁰ both with and without NH₃ injection, 77% and 69%, respectively. The ESP removal efficiency for the two test conditions compared to those obtained using the blended coal was only 21% with NH₃ injection compared to 10% without NH₃ injection.

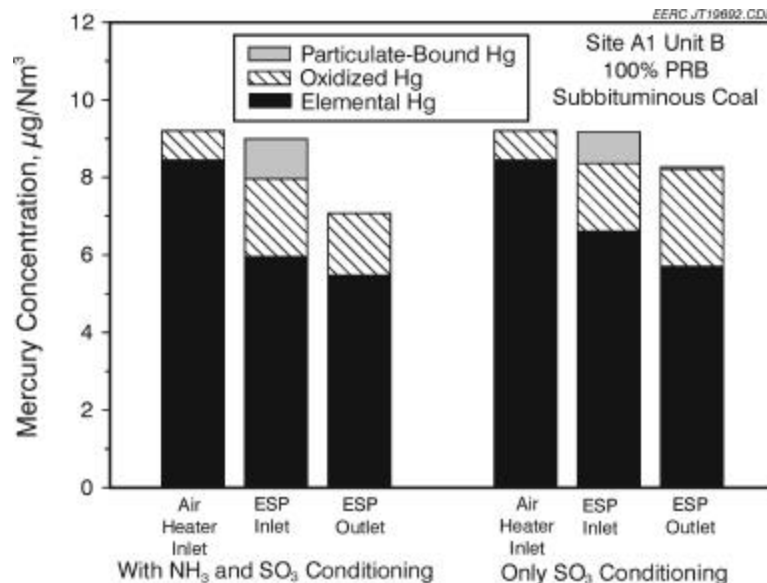


Figure 9. Mercury results showing the effect of adding NH₃ to the flue gas at Site A1, Unit B.

Site A2

The mercury results for Site A2 are shown in Figures 10 and 11. As shown in Figure 10, the addition of urea just upstream of the boiler appeared to result in less mercury oxidation at the ESP inlet—62% as compared to 81% without urea. However, both with and without urea injection, there was little, if any, mercury removal across the ESP. For the test using the second coal, shown in Figure 11, there was little, if any, effect of urea injection on mercury speciation or removal. The CMM data generated for this test, as shown in Figure 12, support the results of the OH mercury sampling.

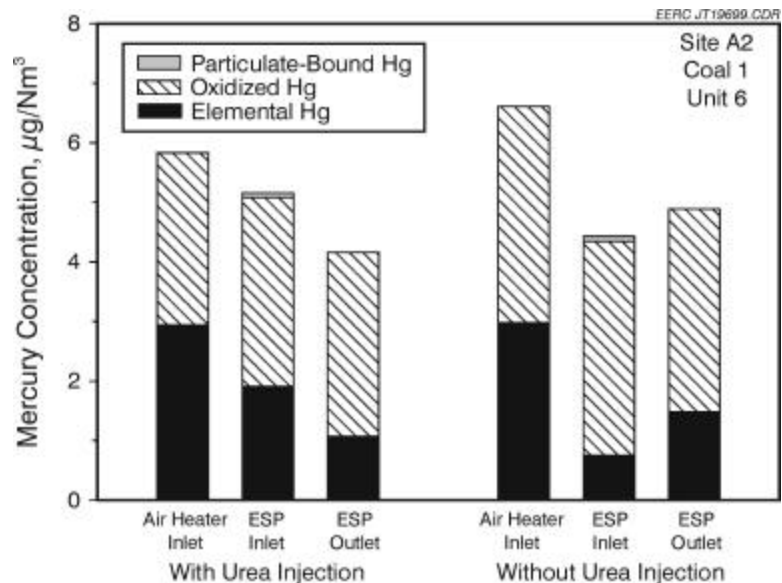


Figure 10. Coal 1 mercury results showing the effect of adding urea to the flue gas at Site A2.

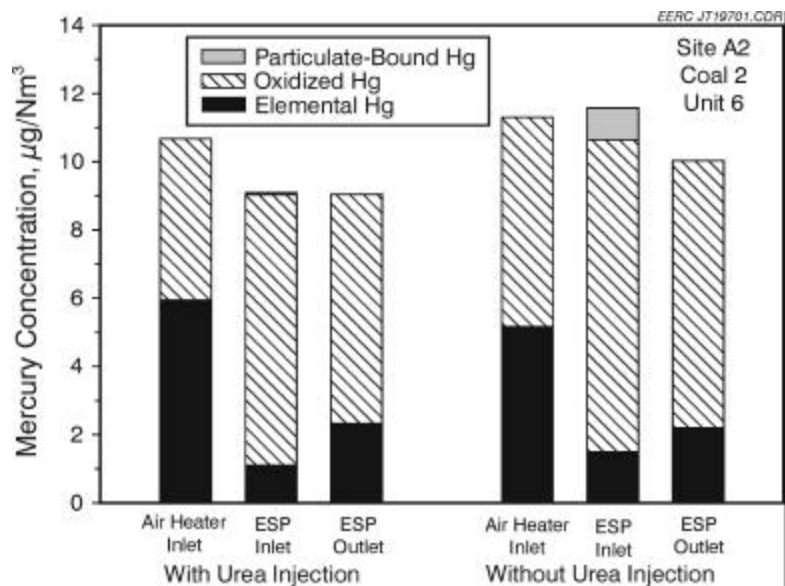


Figure 11. Coal 2 mercury results showing the effect of adding urea to the flue gas at Site A2.

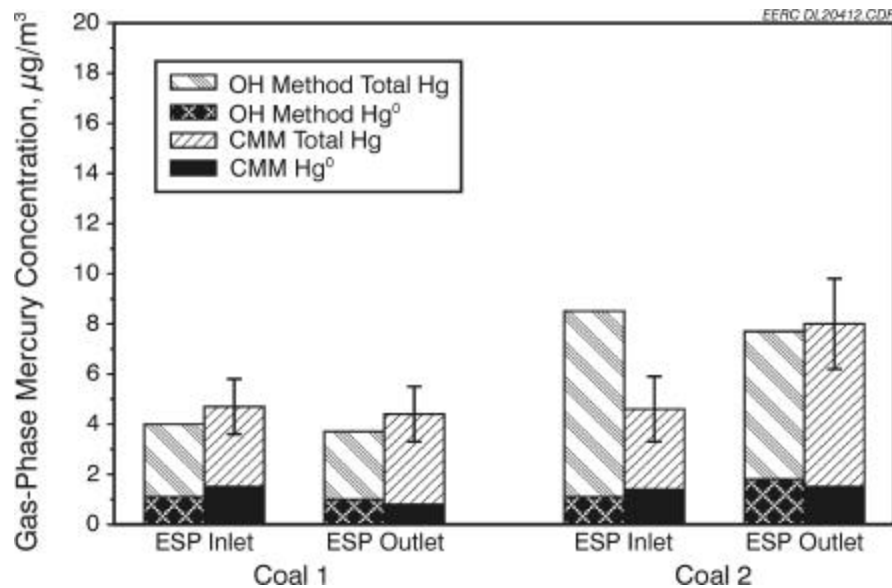


Figure 12. Comparison of mercury monitor results to the OH method for Site A2.

The data from Table 3 show that there is a significant amount of nonelemental mercury even at the economizer outlet (SCR/AH inlet) for some coals. This appears to correlate with the amount of chloride in the coal. For Sites S1, S4, and A1–2, where the coal contained less than 700 µg/g chlorides (Cl), the nonelemental mercury averaged 8% at the SCR inlet (or air heater inlet). For the remaining five sites, where the coals contained greater than 700 µg/g Cl, the nonelemental mercury averaged 50% at the SCR inlet/air heater inlet.

Table 3. Percentage of nonelemental mercury for all units/coals^a.

	S1	S2	S3	S4	A1-1	A1-2	A2-1	A2-2
SCR Inlet/AH Inlet, %	6	48	55	10	51	8	50	44
SCR Outlet, %	26	91	66	80				
APCD Inlet, %	87	97	92	93				
	S1	S2	S3	S4				
APCD Inlet w/o SCR, %	89	74	93	64				

^a Sum of Hg²⁺ and Hg_p, all units in %.

The data from Table 3 also show that with SCR all four sites showed greater than 87% nonelemental mercury at the air pollution control device (APCD) inlet, with an average of 92%. When the SCR reactor was bypassed, only two of the four sites (S1 and S3) showed greater than 87% nonelemental mercury at the APCD inlet.

For two of the plants (S2 and S4), significant oxidation of mercury was shown across the SCR reactor. It is interesting to note that these are the plants that show less than 85% nonelemental mercury at the APCD inlet when the SCR was bypassed.

Hg_p was only a factor at two of the units tested (Sites S1 and A1) and only appeared after the air heater (temperature drop). Additionally, these two sites generated high loss-on-ignition ashes (>10%).

The potential contributing factors that can affect mercury oxidation across the SCR reactor include reaction with the catalyst, increased residence time, change in flue gas chemistry (including, but not limited to, the reduction in NO_x concentration), and reaction with NH₃. It appears that the addition of NH₃ does not have a significant effect on mercury oxidation across the SCR reactor. This also means that the change in flue gas chemistry related to the reduction of NO_x did not significantly affect mercury oxidation. This leaves reaction with the catalyst, an increase in residence time, and other changes in flue gas chemistry as the contributing factors to the increase in oxidation seen across some of the SCR reactors. Unfortunately, the data generated from this project do not allow the determination of these variables separately.

CONCLUSIONS

The observations from these data are as follows:

- It appears that SCR units can assist in converting Hg⁰ to Hg²⁺. However, the effect appears to be coal-specific and, possibly, catalyst-specific. Significant oxidation of mercury across the SCR reactor was shown for two of the four sites tested, with the nonelemental mercury being greater than 90% at the APCD inlet.
- The results of this study, as expected, show a high percentage of Hg²⁺ is removed by the FGD system and a high percentage of the Hg_p is removed by the ESP.
- For the two SCR sites with only minimal increase in mercury oxidation across the SCR reactor, the results showed greater than 85% nonelemental mercury at the APCD inlet. Site S1 burned a PRB coal in a cyclone boiler and produced a high-unburned-carbon fly ash, which may not be representative. The SCR unit at Site S3 operates at a space velocity of 3930 hr⁻¹, which may explain the lower oxidation.
- NH₃ injection, whether directly as a gas or indirectly as urea, did not appear to have a significant effect on mercury speciation.

It should be noted that the data set is small, so the reader should exercise caution in extrapolating the results until further data can be obtained to develop a more robust database to verify these observations.

FUTURE TEST PLANS

Based on a review of these test results, there are numerous data gaps and uncertainties. Currently, several other power plants are being tested to fill in some of the data gaps. Tests are planned to evaluate the effect of SCR on mercury speciation for a power plant firing a low-sulfur compliance coal as well as a plant using a PRB coal in a pulverized coal-fired

boiler. To evaluate the effect of catalyst age, it is planned to retest the two “high-performing” SCR sites (S2 and S4) after the SCR unit has had an additional ozone season of service. Finally, it is important to note that all of the measurements provided in this report represent only short-term measurements lasting hours to several days. Additional measurements are being planned to characterize mercury emissions for up to a month by placing more reliance on CMMs.

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