Evaluation of the Effect of SCR NOx Control Technology on Mercury Speciation

Thomas J. Feeley, III
U.S. Department of Energy
National Energy Technology Laboratory
626 Cochrans Mill Road
Pittsburgh, PA 15236
Phone: 412-386-6134
Fax: 412-386-4822
E-mail: thomas.feeley@netl.doe.gov

Lynn A. Brickett
U.S. Department of Energy
National Energy Technology Laboratory
626 Cochrans Mill Road
Pittsburgh, PA 15236
Phone: 412-386-6574
Fax: 412-386-5917
E-mail: lynn.brickett@netl.doe.gov

James T. Murphy
Science Applications International Corporation
626 Cochrans Mill Road
Pittsburgh, PA 15236
Phone: 412-386-4115
Fax: 412-386-4516
E-mail: james.murphy@netl.doe.gov

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Abstract

The U.S. Environmental Protection Agency (EPA) performed an Information Collection Request (ICR) in 1999 to gather additional information on the control and emission of mercury from coal-fired power plants. The ICR data indicates that a significant, but highly variable, amount of mercury removal can occur across a power plant's conventional air pollution control (APC) equipment used for the capture of particulate matter, nitrogen oxide (NOx), and sulfur dioxide (SO_2) emissions. Such equipment includes electrostatic precipitators (ESP), fabric filters (FF), and flue gas desulfurization (FGD) systems.

One of the more important operating variables that influence the degree of co-benefit mercury capture in these devices is the chemical speciation of the mercury in the flue gas. The mercury in coal-fired power plant combustion flue gas exists as elemental, oxidized, or particulate-bound species. Although elemental mercury is not readily captured, oxidized and particulate mercury can be effectively removed by conventional APC equipment.

Recent testing has indicated that the selective catalytic reduction (SCR) devices used for the control of NOx emissions may further enhance mercury capture in existing pollution control equipment by oxidizing elemental mercury across the SCR catalyst. However, the degree of SCR mercury oxidation is quite variable and appears to be both coal- and catalyst-specific. Also uncertain at this time is whether the oxidation capacity degrades after extended operation of the SCR catalyst.

Since the number of coal-fired power plants equipped with SCR controls is expected to increase significantly in response to current and future NOx regulations, the potential for SCR systems to enhance the removal of mercury could play an important role in a plant's strategy to comply with future restrictions on mercury emissions. This paper presents the results of recent testing sponsored by the Department of Energy's National Energy Technology Laboratory (DOE/NETL), and other organizations, to study the impact of SCR systems on mercury capture and to identify the important design and operational parameters that affect capture performance.

Background

Mercury emissions from U.S. coal-fired power plants are likely to be regulated within the next several years either as a hazardous pollutant under §112 of the Clean Air Act (CAAA Title III – Hazardous Air Pollutants, 42 U.S.C.A. §7412) or as a result of new federal multipollutant control legislation. In December 2000, EPA issued a regulatory determination recommending that power plant mercury emissions be reduced. The Agency is currently developing a §112 Maximum Achievable Control Technology (MACT) standard that could call for mercury reductions from power plants in excess of 90%.[1] EPA plans to issue a proposed MACT rule by December 2003 and a final rule by December 2004. Based on this schedule, implementation of mercury controls could be required as early as December 2007.

Meanwhile, multipollutant control legislation has been under discussion in both the Administration and the Congress. The May 17, 2001, National Energy Policy Report recommended that the President direct the EPA to work with Congress to propose multi-pollutant control legislation "that would establish a flexible, market-based program to significantly reduce and cap emissions of SO_2, NOx, and mercury from electric power generators." On February 14, 2002, the Bush Administration announced its Clear Skies Initiative

(CSI). CSI would require a more modest phased-in reduction in mercury compared to MACT. Using 1999 mercury emission levels as a baseline, CSI would require a 45% reduction beginning in 2010 and a 70% reduction beginning in 2018. The Clear Skies Act was introduced in both the House and Senate of the 2nd Session of the 107th Congress. Although the Clear Skies Act of 2002 did not become law, it is expected to be re-introduced in the 108th Congress in 2003.

In support of its regulatory efforts, EPA issued a mercury data information collection request (ICR) in November 1998 that required electric utilities to sample and analyze their coal shipments for mercury and chlorine content throughout 1999. In addition, approximately 80 plants were required to conduct mercury emissions testing to determine the effectiveness of existing APC equipment on the reduction of mercury emissions. Based on the results of the 1999 ICR plant testing, a significant, but highly variable, amount of co-benefit mercury capture was shown to occur across a power plant's existing ESPs, FFs, and FGD systems. The average mercury capture of existing APC equipment varied from 0 to 98%, and the level of control appeared to be dependent on coal properties, combustion conditions, and type of control equipment in use at the plant. For example, plants burning bituminous coal equipped with a cold-side ESP averaged 36% mercury capture, while plants burning subbituminous coal equipped with a cold-side ESP averaged only 3% mercury capture.[2]

One possible cause for this large variation in mercury capture is that the mercury entrained in coal-fired power plant combustion flue gas can exist as elemental, oxidized, or particulate-bound species, and the degree of mercury speciation can vary significantly from plant-to-plant depending on coal properties and combustion conditions. EPA's analysis of the 1999 ICR data indicates that power plants that burn bituminous coal typically have higher levels of oxidized mercury than plants that burn lignite or subbituminous coal possibly due to the higher chlorine content of bituminous coal. ICR data showed that plants burning bituminous coal equipped with a cold-side ESP averaged only 7% elemental mercury at the inlet to the ESP, while plants burning subbituminous coal equipped with a cold-side ESP averaged 70% elemental mercury.[2]

While oxidized and particulate mercury can be effectively captured in a plant's ESP, FF, or FGD system, elemental mercury is not readily captured. The oxidized mercury is more likely to be adsorbed onto flyash particles and collected along with the ash in either an ESP or FF. Also, since oxidized mercury is water- soluble, it is absorbed in the scrubbing slurry of plants equipped with wet FGD systems compared to elemental mercury, which is not water-soluble. Therefore, methods to further increase the oxidation of elemental mercury to enhance its capture in existing APC equipment could be a cost-effective compliance strategy, particularly for coal-fired power plants equipped with wet FGD systems.

The potential mercury oxidation that occurs across the catalyst used in SCR NOx reduction systems represents one control method currently under investigation by DOE/NETL and others to enhance the removal of elemental mercury. At the time the 1999 ICR data was being collected, very few SCR systems were in operation and, therefore, only one plant with an SCR device was included in the mercury emissions testing phase of the ICR. As a result, the 1999 ICR data did not provide much insight into the possible role of SCR systems on the oxidation of elemental mercury. However, pilot-scale testing conducted by the University of North Dakota's Energy and Environmental Research Center (UNDEERC), cosponsored by DOE/NETL, EPA, and EPRI, indicated that the catalyst and/or ammonia reagent associated with SCR and selective non-catalytic reduction (SNCR) NOx control technologies might enhance overall mercury capture.[3] As a result of the pilot-scale testing, it was concluded that full-scale would be

necessary to determine the effectiveness of SCR at promoting the oxidation of elemental mercury in full-scale commercial power plants.

The oxidation of mercury across SCR systems could be a cost-effective approach to mercury control for many coal-fired power plants in the U.S. In addition to the multipollutant control legislation discussed previously, there are several other regulatory drivers leading to more stringent control of NOx emissions from coal-fired power plants. First, the EPA's so-called NOx SIP Call regulations will take effect in 2003 and 2004 across the Eastern U.S. The NOx SIP call imposes a cap on NOx emissions within a region covering 19 states east of the Mississippi River[a] plus the District of Columbia. The NOx emissions cap for the region is the equivalent of a 0.15 lb/MBtu NOx emission rate covering the five-month ozone season from May 1 through September 30. The SIP Call is being implemented through a regional cap-and-trade compliance program similar to the SO_2 acid rain program. The Energy Information Administration (EIA) has estimated that the equivalent of over 90,000 MW of SCRs would need to be installed to comply with the NOx SIP Call.[4]

Additional NOx reductions, and subsequent SCR installations, could also result within the next ten years in response to the EPA's 1997 revised ozone and fine particulate National Ambient Air Quality Standards and the Best Available Retrofit Technology requirements of the 1999 regional haze regulation. Future enactment of the Administration's CSI or similar multi-pollutant control legislation would also likely increase the utilization of SCR controls to meet the additional NOx reduction requirements. Therefore, the possible increased use of SCR as a NOx control technology offers the added potential to simultaneously improve the capture of mercury. A more complete understanding of this potential co-benefit will be critical as the electric-utility sector begins to consider how it will comply with future mercury regulations. This paper addresses DOE/NETL's research and development (R&D) activity that is attempting to provide this information.

Mercury Control Technology R&D Program

DOE/NETL is carrying out a comprehensive, integrated R&D program under its Innovations for Existing Plants (IEP) Program. It focuses on advanced, low-cost environmental control technology that can assist the existing fleet of coal-based power plants in meeting current and future environmental requirements. The program also provides high-quality scientific information on present and emerging environmental issues for use in regulatory and policy decisionmaking, and directly supports CSI and the May 2001 National Energy Policy recommendations concerning the environmental performance of coal-based power systems.

The IEP program includes bench-scale through field-scale R&D related to the control of mercury, NOx, particulate matter, and acid gas emissions from power plants, as well as research in the area of ambient air quality, atmospheric chemistry, and solid by-products. Furthermore, the program recognizes the importance of emerging water-related issues and their relationship to reliable and efficient power plant operations. Partnership and collaboration with industry, Federal and state agencies, research organizations, academia, and non-governmental organizations are key to the success of the IEP program.

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[a] The SIP Call area consists of Alabama, Connecticut, Delaware, District of Columbia, Georgia, Illinois, Indiana, Kentucky, Maryland, Massachusetts, Michigan, Missouri, New Jersey, New York, North Carolina, Ohio, Pennsylvania, Rhode Island, South Carolina, Tennessee, Virginia, West Virginia, and Wisconsin.

The mercury control technology portion of the IEP program includes a short-term goal to develop mercury control technologies to achieve 50 to 70% mercury capture at no more than 75% of the current estimated costs for powdered activated carbon injection.[b] These technologies should be ready for commercial demonstration by 2005. The IEP program also includes a long-term goal to develop advanced mercury control technologies to achieve 90% or greater capture at one-half to three-quarters the cost of existing technology and would be ready for commercial demonstration by 2010.

The current IEP mercury control technology R&D portfolio includes a mix of in-house and extramural laboratory-scale through larger-scale projects. A number of novel concepts are being developed at the bench- and pilot-scale to address the long-term goal of >90% mercury capture. In addition, the performance of two promising mercury control technologies – sorbent injection and wet FGD enhancement – has recently been evaluated in the field at six operating power plants. A second phase of field-testing is planned to begin in early 2004 to evaluate emerging mercury control technologies on a broader suite of coal types and power plant configurations over a longer period of time.

Evaluating the Effect of Post-Combustion NOx Controls on Mercury Oxidation

An important part of the IEP mercury research has been the continued investigation of the effect of SCR and SNCR on the oxidation and subsequent capture of mercury. In partnership with EPRI and EPA, DOE/NETL has cosponsored bench-scale and full-scale studies with the University of North Dakota Energy and Environmental Research Center (UNDEERC). The purpose of this research is to assess the effects that SCR, SNCR, and flue-gas-conditioning systems have on the speciation of mercury and total mercury removal. The following is a summary of the results-to-date from this research.

Pilot-Scale Testing

Beginning in 1998, pilot-scale testing was conducted using UNDEERC's 550,000 Btu/hr pulverized coal combustor as retrofitted with an ammonia injection system and an SCR reactor loaded with titanium dioxide (TiO_2) and vanadium oxide (V_2O_5) catalyst.[3] Three bituminous coals and a Powder River Basin (PRB) subbituminous coal were tested. The impact of the catalyst on mercury speciation was determined under three conditions: 1) baseline (SCR bypass), 2) ammonia injection with the SCR bypassed, and 3) normal SCR operation. The test results indicated that both ammonia injection and SCR catalyst promoted the conversion of oxidized mercury to particulate mercury for two of the bituminous coals, but not the PRB coal. The results were inconclusive for the third bituminous coal.

2001 Field Testing

Based on the conclusion that the pilot-scale tests may not have been truly representative of full-scale SCR systems, DOE/NETL, EPRI, and EPA sponsored UNDEERC to investigate the effects of SCR on mercury speciation at four commercial power plants. The four plants were identified as Sites S1, S2, S3, and S4.[5] (Note: This project also included mercury speciation testing at two other power plants using SNCR, NH_3, and SO_3 flue gas conditioning.) Plant Site S1 fired a PRB

[b] Baseline cost estimates for PAC technology are in the range of $50,000 to $70,000 per pound mercury removal.

subbituminous coal and the other three plants fired eastern bituminous coals. Each of the tested SCRs is located downstream of the plant's economizer and upstream of the air preheater in a high-dust environment. Table 1 contains additional design information for each of the plants.

Plant Design Data - 2001 SCR Field Testing

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	Cormetech honeycomb	~8000 hr	1800	ESP	None
S2[a]	SCR	OH bitum.	Wall-fired	1300	Yes	Siemens/ Westinghouse plate	~2500 hr	2125	ESP	Wet scrubber
S3	SCR	PA bitum.[b]	Tangential-fired	750	Yes, with overfire air	KWH honeycomb	~3600 hr	3930	ESP	None
S4	SCR	KY bitum. coal	Cyclone	650	No	Cormetech honeycomb	~3600 hr	2275	Lime venturi scrubber	Lime venturi scrubber

[a] Two identical units sampled.
[b] Two different bituminous coals were used.
[c] Not applicable.
Reference: EPRI Report 1005400, December 2002

Table 1 - Plant Design Data

Similar to the bench-scale studies, the objective of the field-testing was to determine the effect of SCR operation on mercury speciation and capture efficiency in the downstream APC equipment. Mercury speciation testing was conducted at each site at the inlet and outlet of the SCR, the ESP inlet, and the stack. The location of the test sampling points for a typical plant is shown on Figure 1.

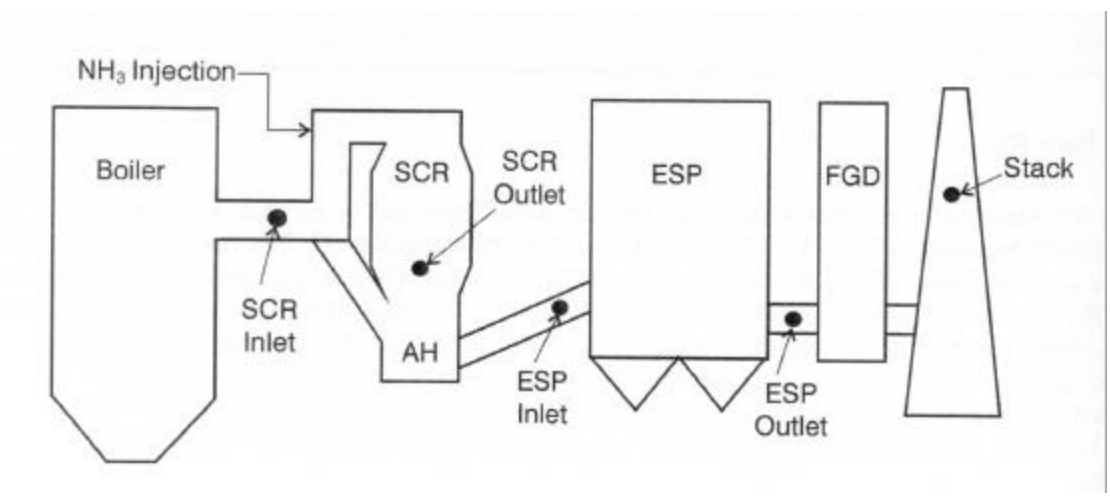

Figure 1 - Sampling Locations per EPRI Report 1005400 - Dated December 2002

In addition, testing was conducted during various operating conditions, including normal SCR operation, SCR operation without ammonia feed, and with the SCR bypassed. The mercury speciation testing included the use of both the manual Ontario Hydro (OH) method and semi-continuous emission monitors (SCEMs). In general, the OH samples were taken as paired samples (i.e., simultaneous SCR inlet and outlet) and were done in duplicate. At each test location, the OH sampling was based on a single-point extraction rather than a full-duct traverse.

The OH sampling was conducted using the EPA Method 17 configuration with the sample filters heated to approximate the local flue-gas temperature. The following is a summary of the OH test results for each of the plants:

Site S1 -- Site S1 burns PRB coal in a cyclone boiler and the average mercury concentration for the coal samples taken during the testing was 0.087 µg/g on a dry basis and the chloride content was less than 60 ppm. The mercury in the flue gas is present primarily as elemental mercury at the SCR inlet (>90%). Surprisingly, a majority of the mercury (84%) is already present as particulate-bound mercury at the ESP inlet, even without the SCR in operation, which is atypical for PRB plants. This unusually high level of particulate-bound mercury for a PRB coal may be attributed to adsorption of elemental mercury onto the flyash, which contains a relatively high level of unburned carbon (approximately 15% Loss on Ignition).

Operation of the SCR resulted in a modest increase in oxidized mercury from 8% to 18% across the SCR.[c] At the inlet to the ESP, the combination of particulate-bound and oxidized mercury increased from about 89% to 95% as a result of SCR operation. Consequently, mercury capture across the ESP increased from 60% to 78% as a result of SCR operation. On the other hand, there was no significant change in mercury oxidation across the SCR when the ammonia was turned off. A graphical summary of the Ontario Hydro test results for Site S1 is shown in Figure 2.

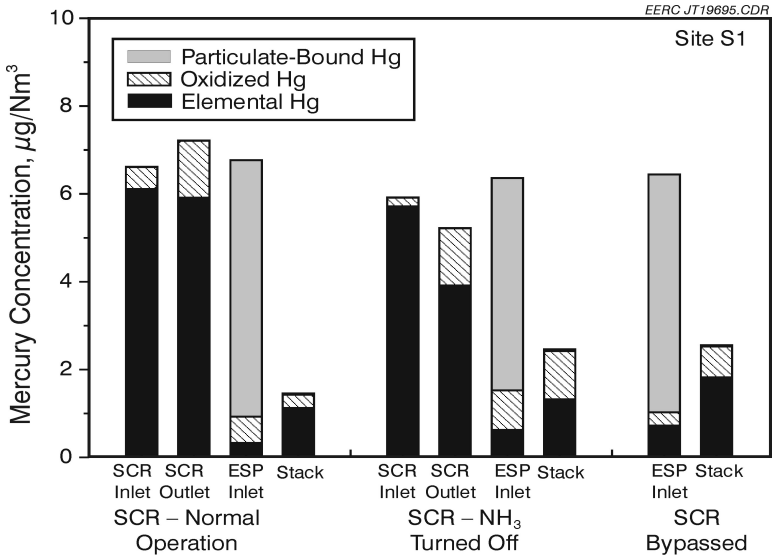

Figure 2 - Site S1 OH Test Results per EPRI Report 1005400 --Dated December 2002

Site S2 -- Site S2 burns a high-sulfur eastern bituminous coal in a wall-fired boiler with an average mercury concentration of the coal samples taken during the testing of 0.168 µg/g (dry basis). The daily coal sample chloride concentrations varied considerably from 573 ppm to 1910 ppm. Operation of the SCR resulted in a significant increase in oxidized mercury from 48% to 91% (of total mercury) across the SCR. The oxidized mercury increased from 73% (SCR bypassed) to 97% (SCR operating) at the ESP inlet and from 68% (SCR bypassed) to 90% (SCR

[c] Note that pilot-scale slipstream testing conducted at Site S1 with a fresh catalyst resulted in an 80% increase in mercury oxidation. However, the catalyst deactivated after about 1700 hours of operation, which suggests that catalyst age may be an important factor in mercury oxidation performance.

operating) at the ESP outlet. Overall mercury capture across the ESP/FGD combination increased from 51% to 88% as a result of SCR operation, and the oxidized mercury capture across the wet FGD system was 94%. A graphical summary of the Ontario Hydro test results for Site S2 is shown in Figure 3.

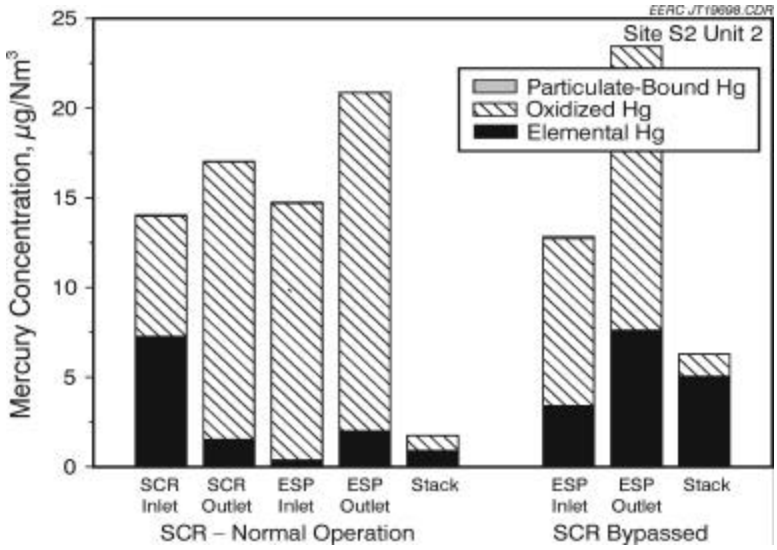

Figure 3 - Site S2 OH Test Results per EPRI Report 1005400 -- Dated December 2002

Site S3 -- The majority of the testing at Site S3 was conducted on Unit No. 1. Site S3 burns a low-to-medium sulfur blended eastern bituminous coal in a tangential-fired boiler. The average mercury concentration of the coal samples taken from Unit No. 1 during testing was 0.400 µg/g (dry basis), and the daily coal sample chloride concentrations varied considerably ranging from 721 ppm to 1420 ppm. Operation of the SCR resulted in only a slight increase in oxidized mercury from 55% to 65% across the Unit No. 1 SCR.[d]

Despite the slight increase in oxidized mercury at the SCR outlet, the non-elemental mercury at the inlet to the ESP was essentially unchanged from the 93% portion without SCR operation. Consequently, mercury capture across the ESP, primarily the particulate-bound species, was only 16% without the SCR and 13% with the SCR in operation. One possible explanation for the relatively low oxidation rate of the SCR is the relatively high space velocity (low-gas-residence time) of the SCR design, which, at 3930 hr^{-1}, is nearly double the space velocity compared to Sites S1, S2, and S4. In addition, the total inlet mercury concentration was more than twice the levels seen at the other test sites. Interestingly, during operation of the SCR without ammonia feed, the oxidized mercury increased from 64% to 82% of total mercury concentration compared with normal SCR operation with ammonia. A graphical summary of the Ontario Hydro test results for Site S3 Unit No. 1 is shown in Figure 4.

[d] Note that additional testing conducted on Unit No. 2 indicated a higher increase in oxidized mercury across the SCR from 35% to 61%, even though the Unit No.2 SCR had been in operation longer than Unit No. 1. However, the coal sample mercury concentration from Unit No. 2 was only 0.169 µg/g compared to 0.400 µg/g for Unit No. 1.

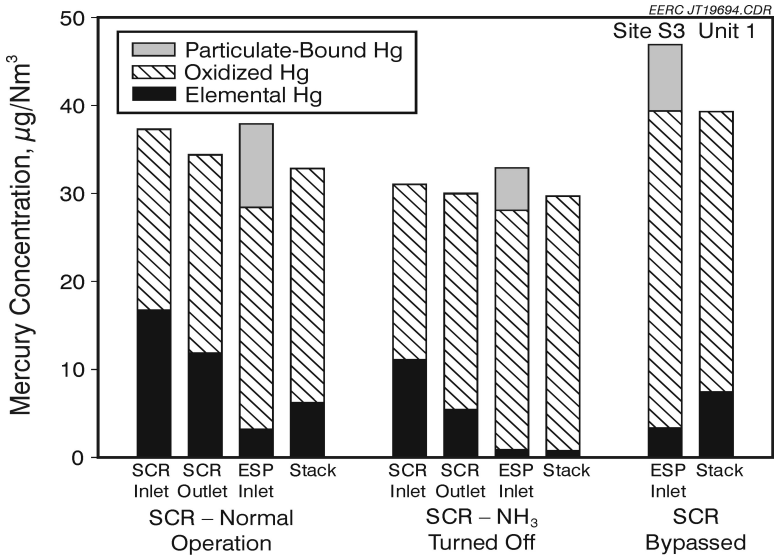

Figure 4 - Site S3 OH Test Results per EPRI Report 1005400 -- Dated December 2002

Site S4 -- Site S4 burns a blend of medium-sulfur eastern bituminous coals in a cyclone boiler with an average mercury concentration of the coal samples taken during testing of 0.131 µg/g (dry basis). The daily coal sample chloride concentrations varied considerably, ranging between 357 ppm to 1160 ppm. Operation of the SCR resulted in an increase in oxidized mercury from 9% to 80% of total mercury concentration, and the oxidized mercury at the inlet to the lime venturi scrubber increased from 56% to 87%. Mercury capture across the lime venturi scrubber system increased from 46% to 90% as a result of SCR operation. Unlike site S3, there was no significant change in mercury oxidation across the SCR when the ammonia was turned off. A graphical summary of the Ontario Hydro test results for Site S4 is shown in Figure 5.

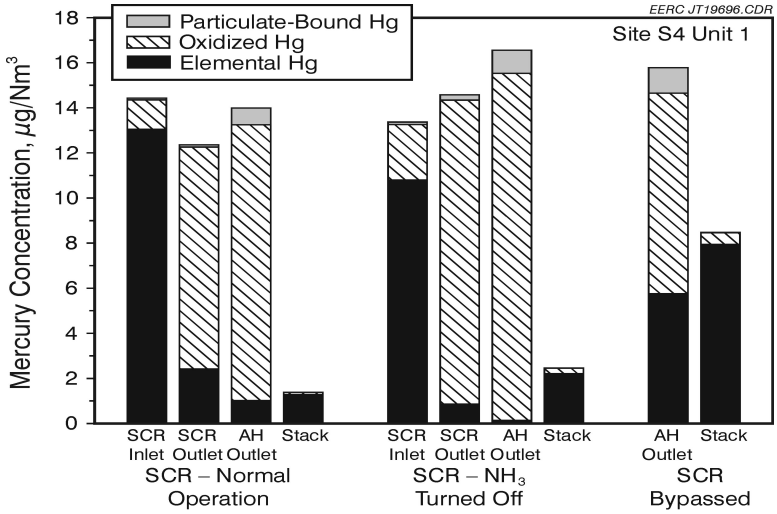

Figure 5 - Site S4 OH Test Results per EPRI Report 1005400 -- Dated December 2002

In summary, the following major observations can be made from the results of the UNDEERC 2001 field-testing program:

- SCR systems can promote the oxidation of elemental mercury. However, the oxidation effect was quite variable and appears to be coal-specific and possibly catalyst-specific. In particular, the catalyst type, space velocity, and catalyst age may all be important variables. Enhanced mercury oxidation was measured across the SCR for two of the four plants (S2 and S4), resulting in over 90% non-elemental mercury at the inlet to the APC equipment. Of the two plants that did not show significant oxidation across the SCR, one burned Powder River Basin coal (S1) and the other had an SCR with a relatively high space velocity (S3).

- The two plants with wet FGD systems (S2 and S4) were able to capture 94% and 99% of the oxidized mercury.
- Operation of the SCRs at all four sites did not appear to increase the amount of particulate-bound mercury. However, the majority of the particulate-bound mercury was effectively captured in the APC equipment.
- The ammonia injection used for SCR systems did not appear to have a significant effect on mercury speciation, indicating that the SCR catalyst likely promotes the oxidation.

- Average daily coal sample chloride content varied significantly at all test sites and, therefore, could have independently affected the degree of mercury oxidation measured, thus distorting the observed effects of SCR operation.

- Caution is urged in drawing conclusions from this limited set of data. The results are based on short-term tests that might be misleading due to the potential for substantial variation in total and speciated mercury concentrations. In addition to possible differences resulting from the coal properties for each of the four plants tested, the plants also varied considerably in SCR design and operating variables including catalyst manufacturer, catalyst space velocity (flue gas residence time), and catalyst age (hours of operation). All of these parameters may dependently or independently affect the degree of mercury oxidation.

- Several uncertainties need to be taken in account when interpreting the OH test data. Although the OH method has become the industry-accepted method for conducting combustion flue-gas-mercury measurements, there are concerns with test data bias and precision. Using the OH method in the high-dust locations upstream of the particulate control device can result in biases due to mercury reactions with the flyash collected on the sampling filter. The flyash reactions can lead to either higher particulate-bound mercury or higher oxidized mercury measurements. In addition, the precision of OH mercury measurements is estimated in the range of $\pm$ 10% to 30%. Therefore, apparent differences or similarities between two measurements may be an artifact of this measurement error. Also, using single point OH samples may not be representative of the average flue gas mercury concentration due to stratification within the ductwork.

<u>2002 Field-Testing</u>

In 2002, UNDEERC continued the investigation of the effects of SCR on mercury speciation at four plants burning bituminous coal, including two of the plants that were evaluated during the 2001 field-testing program. The test results from the two previously tested plants should provide information on the long-term effectiveness of the SCR catalyst impact on mercury speciation. The two new plants tested in 2002 will provide information on the mercury speciation effect of different SCR catalyst design parameters relative to the plants tested in 2001. The results of the

2002 field-testing are under review and a preliminary test report should be available by summer 2003.

Future Plans

An investigation of the effects of SCR on mercury speciation at another plant burning PRB subbituminous coal will be performed by UNDEERC in 2003. Two new DOE/NETL projects will be initiated in 2003 that also focus on SCR mercury speciation. The first project involves field measurements at several bituminous coal-fired power plants equipped with both SCR and wet FGD systems. The second project involves field measurements at plants burning low-rank coals equipped with SCR.

Conclusions

The results from the 2001 field-testing program and prior pilot-scale tests indicate that while oxidation of mercury across SCR systems does occur, it is a complex process dependent on a number of key factors, including coal properties, SCR catalyst type, and age. Increased mercury oxidation was measured across the SCR for only two of the four plants tested. Of the two plants that did not show significant oxidation across the SCR, one burned Powder River Basin coal and the other had a relatively small SCR system that results in a high-space velocity. It also appears that ammonia injection, by itself, did not significantly affect mercury oxidation. Based on these results, it is not possible at this time to predict with a high degree of certainty the level of oxidation that might occur for any given coal type or SCR system.

Clearly, however, reductions in NOx emissions required under the NOx SIP Call and multi-pollutant control legislation, such as the President's Clear Skies Initiative, has the potential to significantly increase the use of SCR technology. And, as such, could lead to the co-removal of mercury. Having a more complete understanding of the potential impact of SCR on mercury oxidation will, therefore, be key to the electric-utility sector as it begins to consider how to comply with future mercury regulations. Further evaluation of SCR impacts on mercury speciation, which will be carried out by DOE/NETL and its industry partners, will help provide the knowledge base needed to design the most cost-effective pollution control compliance strategy.

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