

Pilot-Scale Study of the Effect of Selective Catalytic Reduction Catalyst on Mercury Speciation in Illinois and Powder River Basin Coal Combustion Flue Gases

Chun W. Lee and Ravi K. Srivastava

U.S. Environmental Protection Agency, Office of Research and Development, National Risk Management Research Laboratory, Research Triangle Park, NC

S. Behrooz Ghorishi

Babcock & Wilcox, Alliance, OH

Jarek Karwowski

ARCADIS Geraghty & Miller, Inc., Research Triangle Park, NC

Thomas W. Hastings

Cormetech, Inc., Durham, NC

Joseph C. Hirschi

Illinois Clean Coal Institute, Carterville, IL

ABSTRACT

A study was conducted to investigate the effect of selective catalytic reduction (SCR) catalyst on mercury (Hg) speciation in bituminous and subbituminous coal combustion flue gases. Three different Illinois Basin bituminous coals (from high to low sulfur [S] and chlorine [Cl]) and one Powder River Basin (PRB) subbituminous coal with very low S and very low Cl were tested in a pilot-scale combustor equipped with an SCR reactor for controlling nitrogen oxides (NO_x) emissions. The SCR catalyst induced high oxidation of elemental Hg (Hg⁰), decreasing the percentage of Hg⁰ at the outlet of the SCR to values <12% for the three Illinois coal tests. The PRB coal test indicated a low oxidation of Hg⁰ by the SCR catalyst, with the percentage of Hg⁰ decreasing from ~96% at the inlet

of the reactor to ~80% at the outlet. The low Cl content of the PRB coal and corresponding low level of available flue gas Cl species were believed to be responsible for low SCR Hg oxidation for this coal type. The test results indicated a strong effect of coal type on the extent of Hg oxidation.

INTRODUCTION

Coal combustion is the largest source of anthropogenic mercury (Hg) emissions in the United States, and this has caused environmental health concerns.¹ The U.S. Environmental Protection Agency (EPA) recently issued the Clean Air Mercury Rule for reducing Hg emissions from coal-fired power plants.² Speciation, the split among chemical forms of Hg species, has a great influence on the control and environmental fate of Hg emissions from coal combustion.³ Elemental Hg (Hg⁰) is released from coal during the high-temperature combustion process. However, a significant fraction of the Hg can be oxidized in the low-temperature, postcombustion environment of a coal-fired boiler. Relative to Hg⁰, oxidized Hg (Hg²⁺) is more effectively captured in conventional pollution control systems, such as flue gas desulfurization (FGD) systems. It is well known that FGD control of the emissions of Hg²⁺, the water-soluble mercuric chloride (HgCl₂) in particular, is much easier than controlling the insoluble Hg⁰.^{3–6}

Coal-fired power plants are also a significant source of anthropogenic nitrogen oxide (NO_x) emissions that are associated with several environmental concerns, such as acid rain, fine particulate matter (PM_{2.5}), and ozone (O₃) formation.⁷ Selective catalytic reduction (SCR) technology, which has commercially reduced NO_x emissions by >90% at utility power plants, is increasingly used to meet

IMPLICATIONS

Controlling the emissions of water-soluble oxidized Hg species from coal combustion is much easier than controlling the insoluble Hg⁰. Limited field data indicate that the SCR technology used for NO_x control also promotes Hg⁰ oxidation in bituminous coal-fired boilers. The effect of coal type on Hg oxidation via SCR catalysts is evaluated to assess the applicability of SCR systems as an effective Hg removal strategy. The test results presented here suggest the viability of reducing Hg emissions using SCR systems in Illinois bituminous coal-fired boilers that are equipped with flue gas desulfurization (FGD) systems. SCR applications on PRB subbituminous coal-fired boilers equipped with FGD systems may not achieve significantly increased Hg removal without augmentation of the amount of flue gas chlorine species.

more stringent regulations for O_3 , $PM_{2.5}$, and regional haze. Approximately 100 gigawatts of coal-fired capacity in the United States will use SCR for NO_x control in 2005.⁸ The operation of SCR units affects the speciation of Hg in the coal combustion flue gases. Limited field data obtained from European coal-fired boilers equipped with SCR systems suggest that SCR catalysts promote the formation of Hg^{2+} species.⁹ Results of pilot-scale screening tests on the impact of SCR on Hg speciation also indicate that SCR systems could promote the formation of Hg^{2+} species.¹⁰ The screening test results show that the impact of SCR on Hg speciation appears to be dependent on coal type. For three bituminous coals and one subbituminous Power River Basin (PRB) coal tested in that study, only the high-sulfur (S) bituminous coal showed a significant increase in Hg^{2+} in the outlet of the SCR. The PRB coal with low-S and also low-chlorine (Cl) content showed that the SCR had very little impact on Hg speciation. The results for the other two bituminous coals were between the two extreme cases. The recent full-scale field tests conducted in the United States at utility plants showed that there was an increase in Hg oxidation across the SCR catalysts for plants firing bituminous coals with S contents ranging from 1% to 3.9%.¹¹ Two plants firing subbituminous PRB coals, which contain significantly lower Cl and S content and higher calcium (Ca) content compared with those of the bituminous coals, showed very little change in Hg speciation across the SCR reactors.¹¹

The increase in oxidation of Hg^0 to Hg^{2+} across the SCR reactor could result from the changes in flue gas chemistry caused by the SCR catalysts. The SCR catalysts, which contain oxides of vanadium, titanium, and tungsten or molybdenum, have the potential to catalyze the formation of sulfur trioxide (SO_3) from sulfur dioxide (SO_2) and gas-phase chlorine (Cl_2) from hydrogen chloride (HCl) in coal combustion flue gases. Both SO_3 ¹² and Cl_2 ¹³ have been suggested to react with Hg^0 to cause its oxidation. However, not much is known about either the detailed mechanism or the effect of the flue gas species and chemical reactions on the extent of Hg^0 oxidation across SCR reactors.

This paper reports a study conducted by EPA, Cormetech, Inc., and Illinois Clean Coal Institute to investigate the effect of an SCR catalyst on Hg speciation in coal combustion flue gases. The study consists of pilot-scale experiments to evaluate the viability of SCR technology for Hg speciation modification in Illinois and PRB coal combustion flue gases. No study of the effect of SCR on Hg speciation for boilers firing Illinois coals had been reported at the time of this study, and very limited field data for PRB-fired boilers were available.¹¹ The effect of coal type on Hg oxidation via SCR catalysts is evaluated to assess the applicability of SCR systems as a component of effective Hg removal strategies for Illinois and PRB coals, which are widely used in U.S. coal-fired power plants. Because SCR and FGD are increasingly being used at power plants to control NO_x and SO_2 emissions, respectively, an understanding of Hg^0 reaction behavior over SCR catalysts for these coals has the potential to optimize the Hg control levels achievable with such a combination of pollution control devices.

Initial bench-scale experiments were conducted in EPA laboratories to investigate Hg^0 oxidation in the presence of simulated coal combustion flue gases and under SCR reaction conditions.¹⁴ Flue gas mixtures with different concentrations of HCl and SO_2 for simulating the combustion of Illinois and PRB coals and with different Cl and S contents were tested in these experiments. It was observed that HCl is the critical flue gas component that causes conversion of Hg^0 to Hg^{2+} under SCR reaction conditions, and SO_2 seems to have very little impact on Hg^0 oxidation. A subsequent pilot-scale research study to further evaluate the effect of the SCR catalyst on Hg speciation in the combustion of coals with different Cl and S contents is reported in this paper. A pilot-scale SCR system, designed and manufactured by Cormetech, Inc., was installed in a pilot-scale combustor located at the EPA Research Triangle Park facility to evaluate Hg^0 oxidation capabilities during combustion of three different Illinois coals (from high to low S and Cl) and one PRB coal (very low S and Cl). The objective of the research was to get a better understanding of the effect of coal type on the extent of Hg^0 oxidation over SCR catalysts and the resultant impact on Hg speciation.

EXPERIMENTAL WORK

Pilot-Scale Coal Combustion and SCR System

A pilot-scale 34.9-kW (150,000 Btu/hr), refractory-lined, down-fired cylindrical furnace fired with coal was used in this study. Natural gas was fired during the startup and shutdown of the unit. The furnace, which is shown in Figure 1 and referred to in this paper as the innovative furnace reactor (IFR), has an inner diameter of 15.2 cm and overall length of ~4 m. The IFR was used to simulate and generate a coal combustion environment and flue gas quenching conditions similar to those upstream of SCR units in coal-fired utility boilers. Viewing ports and injection or probe ports were provided along the length of the furnace for testing flexibility.

The IFR system was retrofitted with a downward-flow pilot-scale SCR reactor containing two full-length catalyst elements. Figure 1 illustrates the dimensions of this two-stage SCR system. The first stage of the SCR catalyst was equipped with a flow straightener, a soot blower, and a honeycomb catalyst. The second stage had a soot blower and a honeycomb catalyst. The two honeycomb catalysts were a commercial SCR catalyst manufactured by Cormetech with a vanadia/titania/tungsten oxide formulation having a length of 1250 mm each consisting of 64 flow channels in an array of 8×8 cells. The two-stage SCR reactor was operated at a space velocity of 2943 hr^{-1} , which is within the normal range for commercial SCR units operated in the field. Sampling ports along the horizontal ducts at the inlet and the outlet of the vertical SCR reactor allow gas and particle monitoring.

Emission Measurement and Hg Sampling Methods

The IFR was equipped with a continuous emission monitoring system to measure flue gas composition (O_2 , CO_2 , CO, NO_x , and SO_2) at the inlet and outlet of the SCR reactor. The IFR was also equipped with 12 thermocouples within and downstream of the combustor. A fabric filter

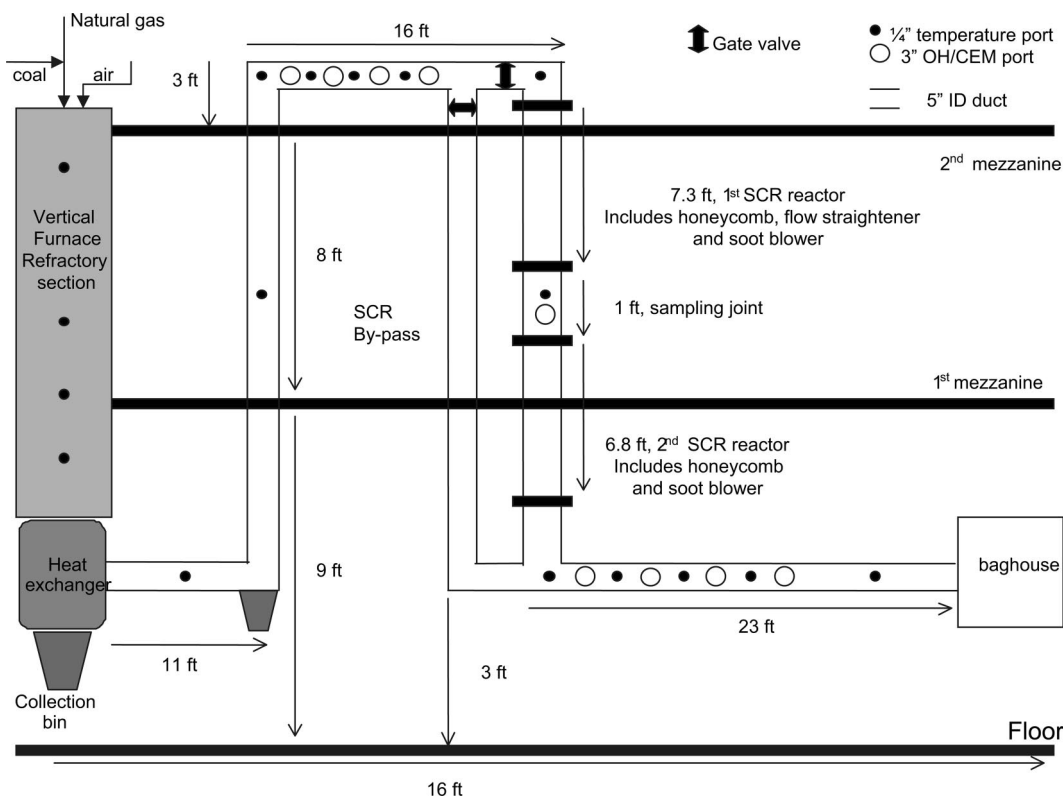


Figure 1. Schematic of the pilot-scale IFR equipped with SCR used for the Hg speciation tests.

and a scrubber were installed downstream of the SCR reactor for treating the flue gas before it was released into the atmosphere. The Hg speciation method¹⁵ developed by Ontario Hydro (OH) was used for measuring Hg^0 , Hg^{2+} , and particulate-bound Hg (Hg_p) in the pilot-scale tests. Isokinetic OH trains were used for making Hg speciation measurements at the inlet and the outlet of the SCR reactor simultaneously during the tests. A heated out-of-stack filter similar to the EPA Method 5-type filter was used for collecting fly ash at the outlet of the sampling probe. The sampling probe and the filter were heated to 120 °C. The 5-in. inner-diameter sampling duct was too small for applying the in-stack filter similar to the EPA Method 17-type filter for fly ash collection. The advantage of the in-stack filter is that it is heated to the same flue gas temperature where the sample is taken to minimize the potential for adsorption of Hg by the fly ash collected in the filter. It should be noted that the OH method was developed for stack measurements, and it has not been validated for use at the SCR operating temperatures (300–400 °C). However, it is the only method available for sampling Hg emissions from coal combustion sources, and reasonably good Hg speciation measurement results were obtained recently from coal-fired SCR units in the field by using the OH method.¹¹

RESULTS AND DISCUSSION

Three different Illinois coals designated as Turriss, Crown II, and Galatia Mines, as well as one PRB coal designated as Black Thunder were tested in the pilot-scale combustor. Table 1 summarizes the characteristics of the coals tested. Turriss coal contains medium levels of S (3.1%) and Cl

(0.17%). Crown II is a high-S (3.5%) and low-Cl (0.13%) content coal. Galatia contains high Cl (0.29%) and low S (1.1%). Black Thunder is a PRB coal with very low S (0.3%) and Cl (<0.01% detection limit) content. The Hg content of the Illinois bituminous coals is within the 0.07–0.09 ppmw range, and the PRB coal has low-Hg content (0.05 ppmw). The Illinois and PRB coals were tested under combustion conditions listed in Table 2. The IFR was operated at firing rates of ~140,000 Btu/hr for the Illinois coal tests. The PRB coal was tested at a slightly higher firing rate (156,000 Btu/hr) than those for the Illinois coal tests. Very good combustion conditions were achieved for all of the tests as indicated by the low CO emissions (30–40 ppm) measured in the tests. There was no in-leakage between the inlet and the outlet of the SCR. The temperature data in Table 2 show that the SCR tests were performed within the typical field SCR temperature range (300–400 °C). The axial temperature gradient across the SCR reactor was ~50 °C, and temperatures at any axial location varied <26 °C for the entire set of test conditions. Uncontrolled levels of NO_x emissions were varied from relatively high levels for the Turriss test (850 ppm) to relatively low levels for the PRB test (525 ppm). NH_3 was injected at a nominal NH_3/NO_x molar ratio of 0.9. Consistently high NO_x reduction (86–90%) across the SCR was observed for all of the tests. The emissions of SO_2 from the Illinois coals varied significantly, from the high-S Turriss coal (2921 ppm) to the low-S Galatia coal (929 ppm), and emissions from the very low S PRB coal were much lower (222 ppm). The measured SO_2 emissions are all in very good agreement with values from combustion calculations using the coal composition data.

Table 1. Characteristics of three Illinois bituminous coals and one PRB subbituminous coal used in the study.

Property	Unit	Black Thunder (PRB)	Turris (Illinois)	Crown II (Illinois)	Galatia (Illinois)
Moisture	wt %	14.0	17.0	16.1	11.3
Ash	wt %	5.9	9.3	7.3	6.3
Volatile	wt %	37.3	33.9	37.1	34.2
Fixed C	wt %	42.8	39.9	39.6	48.2
HHV	Btu/lb	9,903	10,531	10,877	12,179
C ^a	wt %	59.7	59.0	60.5	68.3
H ^a	wt %	3.8	4.3	4.7	4.5
N ^a	wt %	0.82	1.19	1.07	1.5
S ^a	wt %	0.3	3.1	3.5	1.1
O ^a	wt %	15.4	6.0	5.7	6.9
Cl ^a	wt %	<0.01	0.17	0.13	0.29
Hg ^a	ppmw	0.05	0.07	0.07	0.09

^aUltimate analysis on as-received basis.

Hg Speciation Results for Illinois Coal Tests

The Hg speciation measurement results obtained from the OH method for the three Illinois coal tests are shown in Figure 2. In general, the results indicate that Hg⁰ was the predominant Hg species present at the inlet of the SCR. The outlet Hg species was dominated by Hg²⁺ for the three Illinois coal tests. The results show that significant Hg⁰ oxidation was occurring across the SCR catalyst for these tests. For example, concentration of Hg⁰ was decreased from 4.6 to 0.5 μg/dry standard cubic meter (dscm; a corresponding decrease from 84% to 12% of total Hg, respectively), and Hg²⁺ concentration was increased

from 0.8 to 3.5 μg/dscm (an increase from 15% to 85%, respectively) across the SCR catalyst for the Crown II test. Such high levels of Hg⁰ oxidation are consistent with the high levels of Hg⁰ oxidation to Hg²⁺ observed in the previous bench-scale tests for simulating the combustion of three different bituminous coals with Cl and S contents similar to those of the Illinois coals tested in the current pilot-scale tests.¹⁴ The pilot results are also consistent with the high levels of Hg⁰ oxidation to Hg²⁺ observed over several commercial SCR systems firing bituminous coals.¹¹

The Hg²⁺ concentration (2.1 μg/dscm) measured at the inlet of the SCR for the Galatia test was higher than

Table 2. Combustion conditions for the three Illinois bituminous coals and the PRB subbituminous coal tests.

Parameter	Unit	Turris	Galatia	Crown II	PRB
Coal feed rate	lb/hr	14.0	11.5	13.4	15.7
IFR firing rate	Btu/hr	148,000	140,000	145,000	156,000
Total air flow	scfm	28.6	28.2	28.4	28.9
Excess air	%	11	18	11	5
Top SCR temperature	°C	373 (±8)	354 (±1)	363 (±4)	380 (±2)
Middle SCR temperature	°C	365 (±5)	342 (±1)	352 (±3)	364 (±1)
Bottom SCR temperature	°C	323 (±4)	304 (±2)	315 (±2)	325 (±1)
Measured CO	ppm (dry)	38 (±6)	30 (±7)	40 (±4)	0 (±1)
Uncontrolled NO _x	ppm (dry)	960	850	650	525
NO _x reduction across SCR	%	90	86	90	90
Measured SO ₂	ppm (dry)	2921 (±49)	929 (±63)	2739 (±26)	222 (±2)
Calculated SO ₂	ppm (dry)	3060	920	3010	270
Measured O ₂	% (dry)	2.7	4.0	4.4	4.1
Calculated O ₂	% (dry)	1.8	3.0	1.9	0.1
In-leakage based on O ₂	%	5.0	5.0	13.0	19.0
Measured CO ₂	% (dry)	15.6	14.8	14.3	15.6
Calculated CO ₂	% (dry)	16.0	15.4	15.7	18.0
In-leakage based on CO ₂	%	3	3	9	13
Measured HCl	ppm (wet)	NM	246±(23)	NM	NM
Calculated HCl	ppm (wet)	140	210	96	7.9
Calculated Hg	μg/dscm	9.2	9.7	8.2	6.0
PM conc., SCR inlet ^a	mg/dscm	5860	3070 ± 560	3950	2420 ± 500
PM conc., SCR outlet ^a	mg/dscm	3310	1740 ± 190	2560	1610 ± 160

Notes: NM = not measured; conc. = concentration; scfm = standard cubic foot per minute. Values with (±) represent average and range of continuous emission monitor and temperature measurement values except for PM where it indicates standard uncertainty for calculated PM values from duplicate measurements. ^aMeasured using the filter weight of the isokinetic OH method at the inlet and outlet of the SCR, ± values represent duplicate analyses performed for Galatia and PRB coal tests.

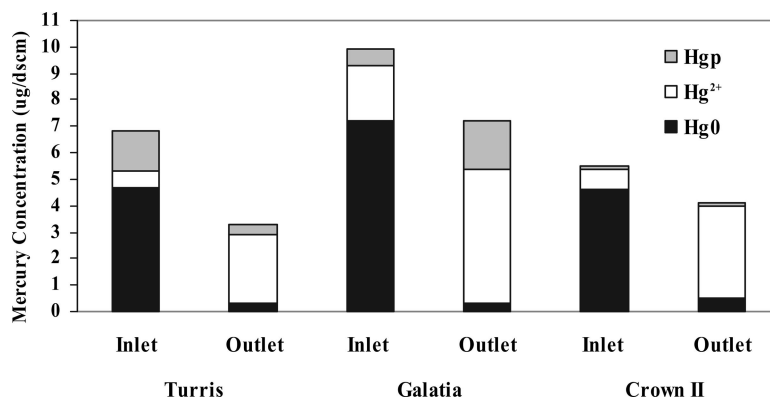


Figure 2. Summary of Hg speciation results measured at the inlet and outlet of the SCR reactor for the three Illinois coal tests.

those of the other two tests (0.6 and 0.8 $\mu\text{g}/\text{dscm}$ for Turriss and Crown II, respectively), suggesting that a low level of Hg^0 oxidation may be occurring before the SCR reactor for this bituminous coal, which has relatively high Cl content (0.29%) compared with those with the other two bituminous coals (0.17% and 0.13%). Coal Cl content has been shown to correlate positively with Hg^0 oxidation in coal combustion flue gas.³

The Turriss coal test showed relatively high Hg_p in the inlet of the SCR (1.5 $\mu\text{g}/\text{dscm}$, 23% of total Hg). The isokinetic OH filter weights were used to determine the particulate matter (PM) concentration values in the flue gas that are reported in Table 2. High PM concentration (5863 mg/dscm) was measured at the inlet of the SCR during the Turriss test, which is consistent with the high ash content (9.3%) of the Turriss coal. The high fly ash concentration in flue gas of this test may create the bias of higher Hg_p measured by the OH method. More fly ash captured by the OH sampling train filter may adsorb additional Hg from the gas stream drawn through the sampling train and may partly explain higher Hg_p measured by the OH method. The out-of-stack filter used in the tests was kept at a temperature (120 °C) significantly lower than the SCR temperature, which may have led to artificially high Hg_p values for the Turriss and Galatia tests. Table 3 shows a summary of the input coal Hg recovered at the SCR inlet and outlet locations and the mass balance closure across the SCR reactor (SCR outlet total Hg relative to SCR inlet total Hg). For the Illinois bituminous coals, calculated Hg recovery values relative to coal input ranged from 67% to 102% at the SCR inlet and 36% to 74% at the

SCR outlet. Across the SCR, Hg mass balance closure for the Illinois bituminous coals tested was 48% to 75% (73–75% excluding the Turriss test). Low Hg mass balance closure across the SCR was also observed in other pilot-scale¹⁰ and field studies,¹¹ which may be because of adsorption of Hg species by the catalyst or by unburned carbon in the fly ash collected on the OH filter. Significant reduction in fly ash concentration across the SCR was also found for the Turriss test (see Table 2).

PRB Coal Test Results

The Black Thunder PRB coal was tested in duplicate runs. The combustion conditions were identical for both tests and are listed in Table 2 under the PRB column. Figure 3 summarizes the results of the OH sampling for the first and second PRB coal combustion tests. A reasonable Hg recovery was obtained across the SCR catalyst during the second PRB test (5.1 $\mu\text{g}/\text{dscm}$ at the inlet vs. 4.6 $\mu\text{g}/\text{dscm}$ at the outlet), which was better than those obtained during the first test (7.2 $\mu\text{g}/\text{dscm}$ at the inlet vs. 5.1 $\mu\text{g}/\text{dscm}$ at the outlet). At the inlet, almost all of the Hg was Hg^0 with no appreciable Hg^{2+} or Hg_p . The comparison of data measured between inlet and outlet of the catalyst shows only a small drop in Hg^0 , from 97% to 88%, for the first test and from 96% to 76% for the second test. This is equivalent to only 9% and 20% oxidation of Hg^0 , respectively. Available field data indicate similarly low Hg^0 oxidation results for two PRB coal-fired SCR systems.¹¹ As shown in Table 3, total Hg recovery as a percentage of coal input Hg was 77–120% at the SCR locations, whereas Hg

Table 3. Summary of input coal Hg recovery at SCR reactor sampling locations and Hg mass balance closure across SCR reactor.

Mercury Measurement	Unit	SCR Sampling Location	Test				
			Turriss	Galatia	Crown II	PRB	PRB ^a
Total concentration	$\mu\text{g}/\text{dscm}$	Inlet	6.9	9.9	5.5	7.2	5.1
Hg recovery, SCR inlet	%	Coal Input	75	102	67	120	85
Total concentration	$\mu\text{g}/\text{dscm}$	Outlet	3.3	7.2	4.1	5.1	4.6
Hg recovery, SCR outlet	%	Coal Input	9.2	9.7	8.2	6.0	6.0
Hg mass balance closure across SCR	Outlet as % of inlet		48	73	75	71	90

^aPRB repeat test.

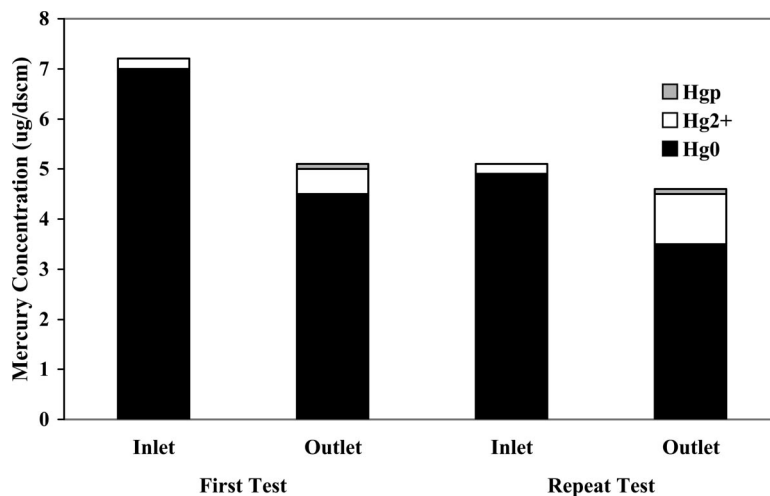


Figure 3. Summary of Hg speciation results measured at the inlet and outlet of the SCR reactor for the PRB coal tests.

mass balance closure across the SCR was 71–90% for the PRB tests.

Discussion of Results

Subbituminous coals generally have significantly lower Cl and higher alkaline content than those of bituminous coals. It is possible that the alkaline species may tie up Cl and may reduce the level of available Cl in the PRB coal combustion flue gas, although this may be implausible at typical SCR temperatures. The low oxidation of Hg⁰ across the SCR catalyst observed for the PRB coal test supports the hypothesis that HCl, which serves as the Cl source, is critical in Hg⁰ oxidation across the SCR catalyst. Figure 4 correlates the degree of conversion of Hg⁰ to Hg²⁺ (shown as a percentage of SCR inlet Hg⁰ converted to Hg²⁺) with coal Cl content for the bituminous and PRB coal tests performed in this study. Figure 4 shows a trend of increased Hg⁰ conversion with increased coal Cl content. It appears that coal Cl content is a strong indicator for SCR Hg oxidation activity. It is well known that most of the Cl in coal is converted into HCl during the coal combustion process. The importance of HCl in Hg⁰ oxidation across the SCR catalyst had been demonstrated by the authors in a bench-scale SCR Hg oxidation simulation study reported previously.¹⁴ It has been suggested that V₂O₅ is a

reactive catalyst for the absorption of HCl to produce vanadium oxychloride complexes, for example, V₂O₃(OH)₂Cl₂ and VO₂Cl.¹⁶ Further reaction of such complexes with Hg⁰ might produce oxidized Hg species, such as HgCl₂.

CONCLUSIONS

A pilot-scale combustion system was constructed to closely simulate the SCR emission control conditions in the field. The system was used for the combustion of three Illinois Basin coals and a PRB coal with different S and Cl levels to test their Hg speciation and oxidation behavior under SCR reaction conditions. High NO_x reduction levels (86–90%), similar to those observed in the field, were achieved for all of the coals tested in this study. It was found that the SCR catalyst has a strong ability to promote Hg⁰ oxidation, decreasing Hg⁰ to an insignificant level (<12%) at the outlet of the SCR for the three Illinois bituminous coals tested. The SCR catalyst showed a very low amount of Hg⁰ oxidation for the PRB coal tested. The effect of SCR on promoting Hg⁰ oxidation appears to be coal-type dependent. Such effect was not found for the PRB coal, which has Cl content ≥1 order of magnitude lower than those of the Illinois coals. These observations suggest that a Cl-containing species, such as HCl, is an

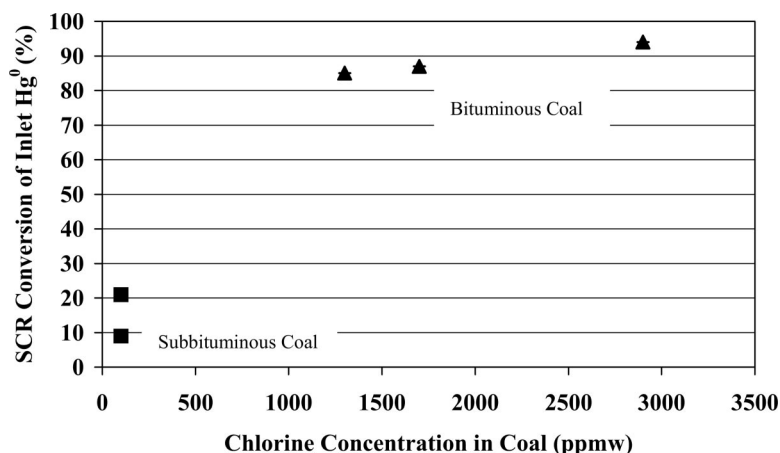


Figure 4. Effect of coal Cl content on conversion of Hg⁰ to Hg²⁺.

important source of Cl for Hg⁰ oxidation under SCR emission control conditions. The use of SCR systems in Illinois coal-fired boilers that are equipped with SCR systems seems to be a viable option for increasing the oxidized Hg content in flue gases, implying viability for reducing Hg emissions when used in conjunction with effective downstream equipment for capture of oxidized Hg, such as FGD systems. However, the low Hg⁰ oxidation observed for PRB coal combustion flue gases over SCR catalyst needs to be improved to achieve lower stack emissions of Hg when coupled with effective downstream Hg²⁺ capture equipment.

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About the Authors

Chun W. Lee is a senior research scientist and Ravis K. Srivastava is a senior research engineer with the U.S. Environmental Protection Agency, Office of Research and Development. S. Behrooz Ghorishi is a principle engineer with Babcock & Wilcox in Alliance, OH. Jarek Karwowski is a scientist with ARCADIS G&M, Inc. Thomas W. Hastings is a new business development manager with Cornetech, Inc. Joseph C. Hirschi is a project manager with the Illinois Clean Coal Institute. Address correspondence to: Chun W. Lee, Air Pollution Prevention and Control Division, E305-01, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711; phone: +1-919-541-7663; fax: +1-919-541-0554; e-mail: lee.chun-wai@epa.gov.