JV TASK 28 – PILOT-SCALE EVALUATION OF THE IMPACT OF SELECTIVE CATALYTIC REDUCTION FOR NO_X ON MERCURY SPECIATION

Final Report

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ABSTRACT

Full-scale tests in Europe and bench-scale tests in the United States have indicated that the catalyst, normally vanadium/titanium metal oxide, used in the selective catalytic reduction (SCR) of NO_x, may promote the formation of Hg^{2+} and/or particulate-bound mercury (Hg_p). To investigate the impact of SCR on mercury speciation, pilot-scale screening tests were conducted at the Energy & Environmental Research Center. The primary research goal was to determine whether the catalyst or the injection of ammonia in a representative SCR system promotes the conversion of Hg^0 to Hg^{2+} and/or Hg_p and, if so, which coal types and parameters (e.g., rank and chemical composition) affect the degree of conversion.

Four different coals, three eastern bituminous coals and a Powder River Basin (PRB) subbituminous coal, were tested. Three tests were conducted for each coal: 1) baseline, 2) NH_3 injection, and 3) SCR of NO_x. Speciated mercury, ammonia slip, SO_3, and chloride measurements were made to determine the effect the SCR reactor had on mercury speciation.

It appears that the impact of SCR of NO_x on mercury speciation is coal-dependent. Although there were several confounding factors such as temperature and ammonia concentrations in the flue gas, two of the eastern bituminous coals showed substantial increases in Hg_p at the inlet to the ESP after passing through an SCR reactor. The PRB coal showed little if any change due to the presence of the SCR. Apparently, the effects of the SCR reactor are related to the chloride, sulfur and, possibly, the calcium content of the coal. It is clear that additional work needs to be done at the full-scale level.

EXECUTIVE SUMMARY

Full-scale tests in Europe[1] and bench-scale tests in the United States[2] have indicated that the catalyst, normally vanadium/titanium metal oxide, used in the selective catalytic reduction (SCR) of NO_x may promote the formation of oxidized mercury (Hg^{2+}) and/or particulate-bound mercury (Hg_p). To investigate the impact of SCR on mercury speciation, pilot-scale screening tests were conducted at the Energy & Environmental Research Center (EERC). The primary research goal was to determine whether the catalyst in a representative SCR system promotes the conversion of elemental mercury (Hg^0) to Hg^{2+} and/or Hg_p and, if so, which coal parameters (e.g., chemical composition) affect the degree of conversion.

Three bituminous coals and a Powder River Basin (PRB) subbituminous coal were fired in a 580-MJ/hr (550,000-Btu/hr) pilot-scale combustion system equipped with an ammonia (NH_3) injection system, SCR reactor, and electrostatic precipitator (ESP). Table ES-1 provides information about the coals. The selection criteria for the four coals investigated were the significant differences in their sulfur (S) and chloride (Cl) contents. Sulfur and Cl in coal are believed to affect mercury speciation and removal by air pollution control devices. Table ES-2 provides the analyses of each of the test coals. Mercury concentrations (as-received basis) of the two bituminous coals (Paradise and Blacksville) and the PRB coal (Cordero Rojo) were very similar at about 0.1 ppm, whereas the third bituminous coal (Band Mill) contained only 0.02 ppm mercury.

Mercury speciation of the combustion flue gases was determined under three different test conditions: 1) baseline, 2) NH_3 injection (bypassing the SCR), and 3) SCR of NO_x. The test conditions are shown in Table ES-3. The first test provided baseline mercury emission and speciation data for each test coal. For the second test condition, NH_3 was injected into the duct upstream of the SCR bypass loop at a temperature of about 340°C. The third test was conducted under SCR conditions. For the SCR tests, the NH_3 was injected immediately upstream of the SCR reactor at a targeted NH_3-to-NO_x stoichiometric ratio of 1. The Ontario Hydro mercury speciation method was used to collect speciated mercury samples. Ammonia, SO_3, and Cl measurements were conducted at the ESP inlet-sampling location for each test. ESP hopper ashes were collected and analyzed to investigate the effects of NH_3 injection and the SCR catalyst on fly ash composition and Hg_p formation.

For the Paradise and Blacksville SCR tests, the NH_3 and NO_x measurements showed that the NH_3-to-NO_x stoichiometric ratio was maintained at approximately 1, thus maximizing NO_x conversion while minimizing NH_3 slip ($\leq$5 ppmv). However, for the Cordero Rojo and Band

[1] Gutberlet, H.; Schlüten, A.; Lienta, A. SCR Impacts on Mercury Emissions on Coal-Fired Boilers. Presented at EPRI SCR Workshop, Memphis, TN, April 2000.
[2] Galbreath, K.C.; Zygarlicke, C.J.; Olson, E.S.; Pavlish, J.H.; Toman, D.L. Evaluating Mercury Transformation Mechanisms in a Laboratory-Scale Combustion System. *The Science of the Total Environment* **2000**, 261 (1–3), 149–155.

Table ES-1
Coal Information

Organization	State	Mine	Coal	Location	Rank[a]	Production, t/yr
KenAmerican Resources, Inc.	KY	Paradise	Western Kentucky No. 9	Illinois Basin	hvBb[b]	1,943,910[c]
Kennecott Energy Co.	WY	Cordero Rojo Complex	Wyodak–Anderson	Powder River Basin	subC[d]	37,011,000[c]
Arch Coal Sales, Inc.	VA and KY	Band Mill (Pardee Complex)	Taggart	Appalachian Basin	hvAb[e]	1,700,000[f]
Consolidation Coal Co.	PA	Blacksville No. 2	Pittsburgh	Appalachian Basin	hvAb	3,898,360[c]

[a]Determined according to American Society for Testing and Materials (ASTM) Designation D388-88.
[b]High-volatile B bituminous.
[c]1998 mine production statistic from *Keystone Coal Industry Manual*, Intertec Publishing Corporation, Chicago, Illinois, 2000, 793 p.
[d]Subbituminous C.
[e]High-volatile A bituminous.
[f]1999 mine production statistic from Arch Coal, Inc., http://www.archcoal.com/ab/ab03a.html (accessed 10/1/2000).

Table ES-2
Coal Hg and Cl Concentrations

Element	Laboratory	Paradise	Cordero Rojo	Band Mill	Blacksville
Hg, ppm	EERC	0.111 ± 0.002	0.085 ± 0.012	0.022 ± 0.001	0.094 ± 0.006
S, %	EERC	3.10 ± 0.07	0.52 ± 0.04	0.75 ± 0.05	2.00 ± 0.04
Cl, ppm	EERC	350 ± 44	<50	58 ± 12	758
Cl, ppm	HawkMtn[a]	454 ± 19	8.7 ± 2.6	59 ± 3	NA[b]

[a] Analysis done using oxidative hydrolysis microcoulometry, which is a more sensitive method than that done by the EERC.
[b] Not analyzed.

Table ES-3
Test Matrix

Test	Coal Mine	Injection*	SCR Unit
607	Paradise	None	Bypassed
608	Paradise	25 ppmv NH_3	Bypassed
609	Paradise	~750 ppmv NH_3	Flow through
610	Cordero Rojo	None	Bypassed
611	Cordero Rojo	10 ppmv NH_3	Bypassed
612	Cordero Rojo	~750 ppmv NH_3	Flow through
613	Band Mill	None	Bypassed
614	Band Mill	10 ppmv NH_3	Bypassed
615	Band Mill	~750 ppmv NH_3	Flow through
616	Blacksville	None	Bypassed
617	Blacksville	10 ppmv NH	Bypassed
618	Blacksville	~900 ppmv NH_3	Flow through

*The NH_3 injection for Test 608 was 25 ppm instead of 10 ppm. Also, the NH_3/NO_x was to be 1.0 for the SCR test, but instead was ~1.2 for Tests 612 and 615.

Mill SCR tests, NH_3/NO_x was closer to 1.2 because of a calibration error in the NH_3 injection system. As a result of this error, NH_3 slip averaged 143 and 189 ppmv during the Cordero Rojo and Band Mill SCR of NO_x tests, respectively. For all tests, the SCR reactor reduced NO_x concentrations by >97%.

Mercury speciation results for each of the four test coals are summarized in Figures ES-1 through ES-4 and in Table ES-4. Note: In order to make direct comparisons at each sampling location, the mercury results in Table ES-4 are presented as a percentage change in mercury species relative to the baseline condition (ΔHg^0, ΔHg^{2+}, and ΔHg_p). A negative value shows a decrease in a mercury species with respect to the baseline condition and a positive value indicates an increase. These figures and the table show that NH_3 injection and, possibly, the SCR catalyst promote the conversion of Hg^{2+} to Hg_p in the Paradise and Band Mill coal combustion flue gases, but not in the Cordero Rojo flue gas. Based on the results of these pilot-scale tests, it is more difficult to determine if either the NH_3 injection or SCR conditions impacted mercury speciation when Blacksville coal is fired. For the Paradise coal test, when the SCR reactor was used, there appeared to be conversion of Hg^0 to Hg^{2+} between the SCR inlet- and outlet-sampling locations.

Linear regression analyses were completed relating mercury speciation results to the chemical composition of the four test coals. It should be noted that the results of these regression analyses represent data that may not be totally representative of full-scale SCR performance owing to the smaller size and design of the pilot-scale combustor facility, limited number and range of coal types, and the level of ammonia slip.

For the regression analysis, the changes in mercury species through the SCR and ESP were correlated with the coal factors (Hg, S, Cl, and Ca concentrations) and operating factors (NH_3 injection and SCR). The most significant variables affecting the total mercury emitted are NH_3 injection, SCR, and Cl in coal. Changes in Hg^0 and Hg^{2+} were inversely related, as expected, but both experienced positive and negative changes. Apparently, the SCR resulted in oxidation for coals higher in Hg and Cl, but reduction for coals low in Hg and Cl and higher in Ca. The percentage of coal mercury emitted as Hg^0 at the exit of the ESP is positively correlated with the Ca, negatively with S and Cl, and negatively and secondarily with SCR; the correlation for these four variables combined has an R^2 of 0.84. It would appear that the chemistry of mercury on an SCR catalyst is quite complex.

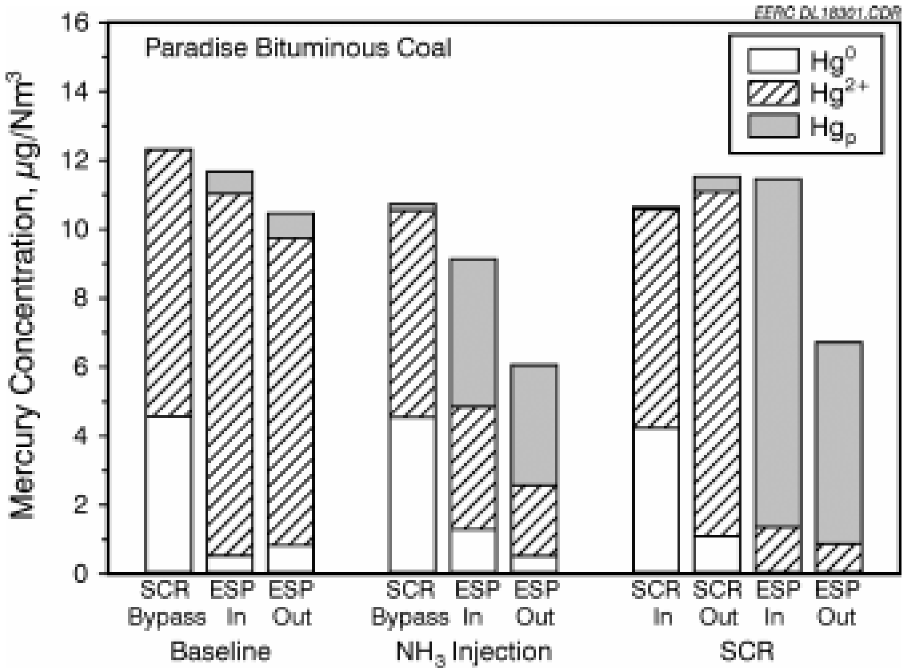

Figure ES-1
Average Mercury Speciation Results for the Paradise Test Series for Each of the Three Conditions

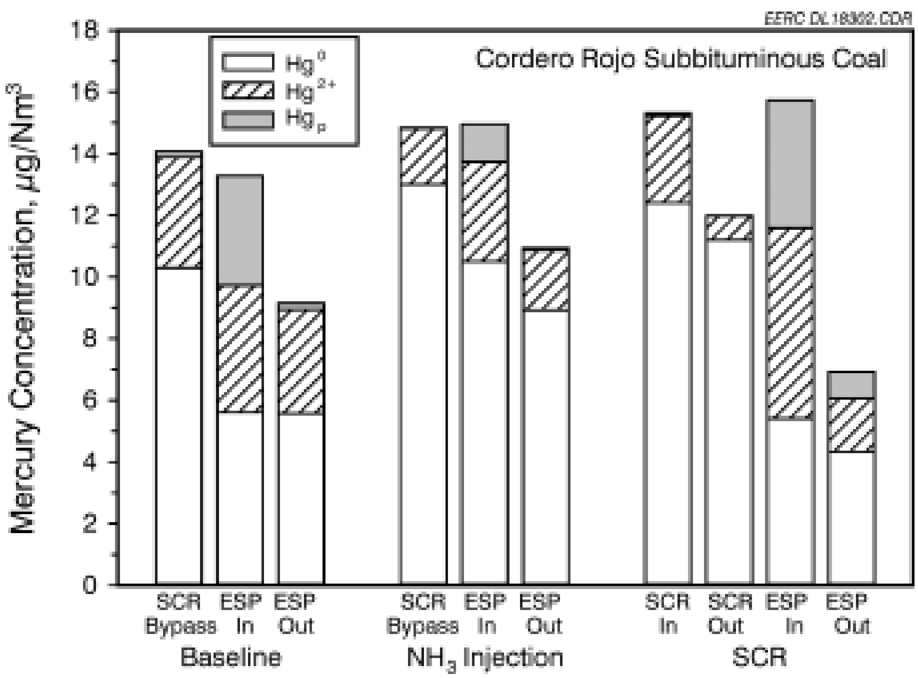

Figure ES-2
Average Mercury Speciation Results for the Cordero Rojo Test Series for Each of the Three Conditions

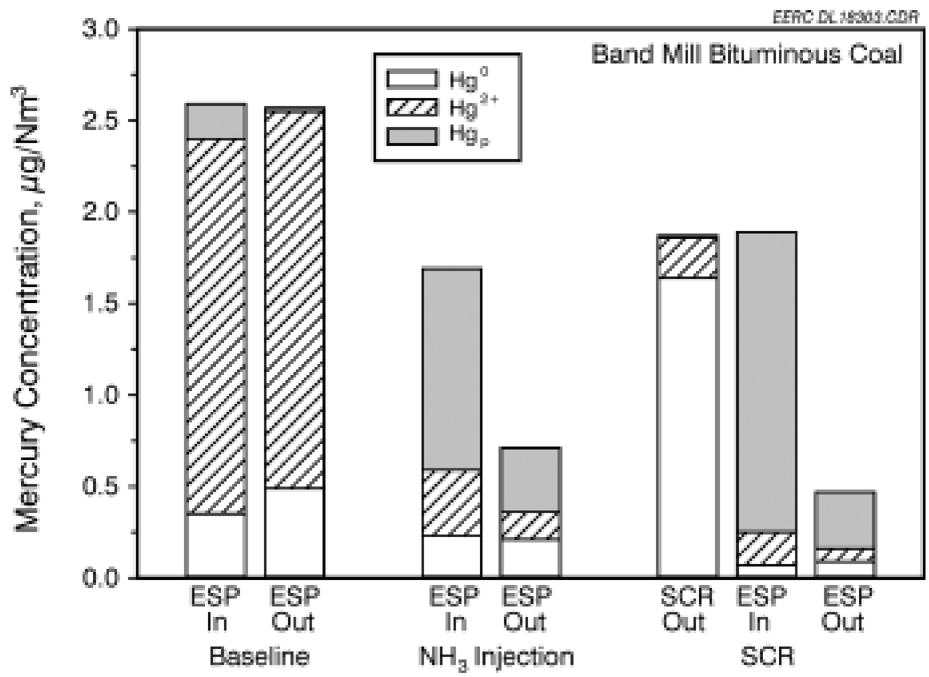

Figure ES-3
Average Mercury Speciation Results for the Band Mill Test Series for Each of the Three Conditions

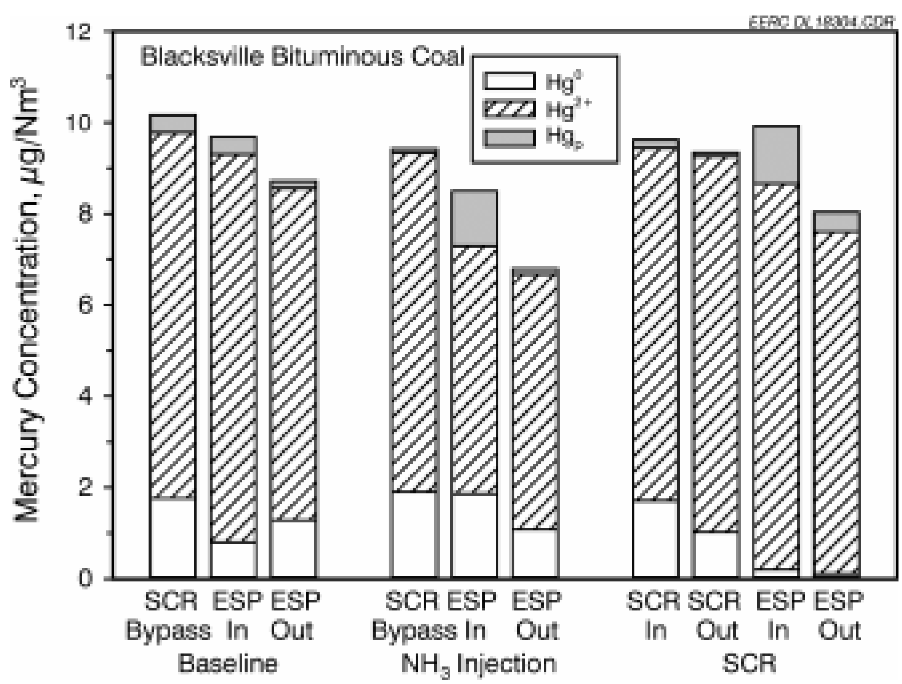

Figure ES-4
Average Mercury Speciation Results for the Blacksville Test Series for Each of the Three Conditions

Table ES-4
Percent Change in Mercury Species Proportions Relative to the Baseline Tests

Coal:	Paradise[a]			Cordero Rojo			Band Mill			Blacksville		
Sampling Location:	SCR Out	ESP In	ESP Out	SCR Out	ESP In	ESP Out	SCR Out	ESP In	ESP Out	SCR Out	ESP In	ESP Out
NH_3 Injection Only												
ΔHg^0	NA[b]	9.6	0.6	NA	27.9	20.6	NA	0.1	10.5	NA	13.6	1.2
ΔHg^{2+}	NA	−51.0	−51.3	NA	−9.3	−19.0	NA	−57.8	−59.0	NA	−23.9	−1.3
ΔHg_p	NA	41.4	50.7	NA	−18.6	−1.7	NA	57.8	48.5	NA	10.3	0.1
SCR Tests[c]												
ΔHg^0	−30.3	−4.3	−7.5	12.3	−7.9	1.6	ND[d]	−9.8	0.1	-6.9	−6.2	−13.3
ΔHg^{2+}	27.2	−78.3	−72.7	−11.9	8.3	−11.7	ND	−69.6	−65.3	7.9	−2.5	9.7
ΔHg_p	3.2	82.6	80.2	−0.5	−0.4	10.1	ND	79.4	65.2	−1.0	8.8	3.6

[a] For the Paradise NH_3 injection test, 25 ppm was added, compared to 10 ppm for the remainder of the tests.
[b] Not applicable.
[c] For the Cordero Rojo and Band Mill tests, the NH_3/NO_x was ~1.2, compared to ~1.0 for Paradise and Blacksville tests.
[d] Not determined because of a lack in repeatability among the three SCR bypass/inlet mercury speciation measurements.

Based on the results obtained from this screening evaluation, the following conclusions can be made:

- For some coals, NH_3 appeared to increase the Hg_p concentration, thereby increasing Hg removal in the downstream ESP. Because NH_3 slips were higher than expected in a full-scale SCR or selective noncatalytic reduction (SNCR) application and NH_3 concentration may directly impact Hg speciation and removal, these results may or may not be consistent with full-scale applications.
- The impact of SCR on mercury speciation and mercury capture appears to be very coal-dependent and quite complex.
- Based on a regression analysis, the chlorine, sulfur, and calcium appear to correlate with mercury speciation across the SCR.
- Relatively high concentrations of alkaline-earth metals (i.e., CaO and MgO) in the Cordero Rojo and Blacksville fly ashes may have limited the suspected interactions involving SO_3, NH_3, and Cl that promote Hg_p formation.
- NH_3 injection and/or the SCR catalyst promoted the conversion of Hg^0 to Hg^{2+} across the SCR for the Paradise coal, but not for any of the others.
- NH_3 injection, with and without the SCR reactions, converted Hg^{2+} to Hg_p when the Paradise and Band Mill coals were fired, but not for the Cordero Rojo PRB.
- The increased mercury removals as measured by the flue gas measurements were confirmed with mercury analyses of the corresponding fly ash. The Hg_p concentrations in Paradise and Band Mill ESP hopper ashes increased by 230%–460% relative to the baseline fly ashes as a result of NH_3 injection and SCR tests.
- Because of the high levels of Hg^{2+} in the baseline tests, it is not possible to determine whether there was an increase in Hg_p or oxidation of Hg^0 for the Blacksville coal.

The applicability of the conclusions from this pilot-scale investigation should be evaluated by performing similar flue gas and fly ash measurements at utility-scale boilers equipped with SNCR and SCR units. As part of this proposed utility-scale investigation, flue gas and fly ash samples should be collected when the SNCR and SCR units are off-line and on-line. Size-fractionated fly ash samples should also be collected and analyzed to investigate further the apparent role of particle size and composition on Hg_p formation. Additional coals also need to be tested because the impact of SNCR and SCR of NO_x on mercury speciation apparently depends on the coal’s chemical composition.

LIST OF ABBREVIATIONS

Abbreviation	Abbreviated Phrase
ASTM	American Society for Testing and Materials
CVAAS	Cold-vapor atomic absorption spectroscopy
EERC	Energy & Environmental Research Center
EPA	U.S. Environmental Protection Agency
ESP	electrostatic precipitator
Hg^0	elemental mercury
Hg^{2+}	mercuric compounds
Hg_p	particle-bound mercury
LOI	loss on ignition
N	Number of observations
$NH_xCl_ySO_z$	sulfated ammonia and chloride compounds
NO_x	nitrogen oxides
OH	Ontario Hydro mercury speciation method
P	Probability
ppm	parts per million
ppmv	parts per million by volume
PTC	particulate test combustor
PTFE	polytetrafluoroethylene
R^2	Correlation coefficient
SCR	selective catalytic reduction
SNCR	selective noncatalytic reduction
XRF	x-ray fluorescence spectrometry

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1 INTRODUCTION

Coal combustion from electric utilities is a large source of anthropogenic mercury emissions in the United States, according to the U.S. Environmental Protection Agency (EPA) [1], accounting for 52 of the 158 tons/yr of total point-source mercury emissions. More recent data indicate this number may be closer to 45 tons/yr [2]. Clearly, EPA views mercury from coal-fired utilities as a potential public health concern [3]. Many research and development organizations are striving to develop effective and economical mercury emission control technologies for coal-fired utility boilers. The development of mercury control technologies is being spurred by environmental and human health concerns and the resulting potential for mercury emission regulations [1–5].

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). Total mercury concentrations in coal combustion flue gas generally range from 3 to 10 µg/m^3; however, Hg^0, Hg^{2+}, and Hg_p concentrations are much more variable, depending on coal composition and combustion conditions [6].

During combustion, Hg^0 is liberated from coal. However, depending on the coal type, a significant fraction of the Hg^0 can be oxidized to Hg^{2+} 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 generally more effectively captured in conventional pollution control systems, such as wet scrubbers, fabric filters, and electrostatic precipitators (ESPs) [3, 7–10]. 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 increased acidification, 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 installation of low-NO_x burners. These burners have the capability of reducing NO_x emissions by 40%–60%. However, with possible establishment of $PM_{2.5}$, regional haze, and 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 to 90 U.S. utilities are planning to install SCR units [11].

1.1 Potential Impacts of NO_x SCR on Mercury Speciation

SCR units achieve lower NO_x emissions by reducing NO_x to N_2 and H_2O. Ammonia (NH_3) is the most common reductant used for the SCR of NO_x. The SCR process is generally performed on metal oxide catalysts such as titanium dioxide (TiO_2)-supported vanadium oxide catalysts (V_2O_5). These units are operated at about 343°–371°C (650°–700°F). Laboratory-scale testing indicates that metal oxides, including V_2O_5 and TiO_2, promote the conversion of Hg^0 to Hg^{2+} and/or Hg_p in relatively simple flue gas mixtures [12]. In addition, mercury speciation measurements at European coal-fired boilers equipped with SCR reactors have indicated that SCR catalysts promote the formation of Hg^{2+} [13, 14]. Therefore, it has been speculated that the installation of an SCR reactor to reduce NO_x emissions may improve the mercury control efficiency of existing air pollution control devices by promoting Hg^{2+} and/or Hg_p formation. Possible mechanisms that could result in the SCR of NO_x impacting mercury speciation include:

- Changing the flue gas chemistry. The significant reduction in flue gas NO_x and slight increase in NH_3 concentrations associated with SCR may affect mercury speciation. It is well known that NO_x, particularly NO_2, has a substantial effect on mercury speciation [20]. The gas-phase effects of NH_3 on mercury are unknown. However, tests are being conducted by the University of Cincinnati to help determine possible reactions.
- Catalyzing the formation of SO_3 and, potentially, Cl_2, which then may react with mercury [15 – 19].
- Changing the fly ash chemical composition. It is possible that the SCR process may change the surface chemistry of the fly ash particles such that their ability to adsorb or convert mercury species is changed.
- Catalytically oxidizing the mercury. As was reported by the German studies, there is some evidence that vanadium-based catalysts can promote the formation of Hg^{2+}[14]. However, the extent to which this can occur and at what temperatures are unknown.
- Increasing wall deposition. SCR systems may result in the deposition of ammonium bisulfate and ammonium sulfate in the air preheater as well as the duct walls. It is unknown whether increased deposition could impact mercury emissions or mercury speciation.

1.2 Project Objectives

The effects of NH_3 injection and the SCR process for NO_x reduction on mercury speciation were evaluated at the Energy & Environmental Research Center (EERC) using a pilot-scale combustion system equipped with an ESP. The primary research goal was to determine whether NH_3 injection and/or the catalyst in a representative SCR system promote the conversion of Hg^0 to Hg^{2+} and/or Hg_p and, if so, the coal types and parameters (e.g., rank and chemical composition) that affect the degree of conversion. Although this project was a screening evaluation and not a complete parametric study, potential mechanisms by which Hg^0 potentially transforms to Hg^{2+} and/or Hg_p were investigated.

2 EXPERIMENTAL

2.1 Coals Tested

Three bituminous coals from the Paradise, Band Mill, and Blacksville No. 2 coal mines and a subbituminous Power River Basin (PRB) coal from the Cordero Rojo mine were selected for this investigation, based on anticipated differences in their sulfur (S) and chloride (Cl) concentrations. Sulfur and Cl concentrations were used as coal selection criteria because it is believed that these parameters affect mercury speciation and removal [2, 18]. Information on the four coals selected for this investigation is presented in Table 2-1.

2.2 Facilities

2.2.1 Pilot-Scale Combustion System

The pilot-scale tests were conducted using the EERC particulate test combustor (PTC), with an ESP as the downstream particulate control device. This combustor has been extensively used by the EERC for a variety of work over the years, including tests to evaluate a catalytic fabric filter for NO_x reduction, projects to evaluate mercury measurement methods and control technologies, and projects for removing fine particulate matter. The following is a short description of the pilot-scale facilities.

The PTC is a 580-MJ/hr (550,000-Btu/hr) pulverized coal-fired unit designed to generate fly ash and flue gas chemistry representative of that produced in a full-scale utility boiler. Coal is introduced to the primary air stream via a screw feeder and ejector. An electric air preheater is used for precise control of the combustion air temperature. The PTC instrumentation permits system temperatures, pressures, flow rates, flue gas constituent concentrations, and ESP operating data to be monitored continuously and recorded on a data logger.

The PTC (shown in Figure 2-1) is designed to operate in conjunction with an ESP. The ESP, shown in Figure 2-2, is a single-wire, tubular ESP, with a specific collection area of 125 ft^2 /1000 acfm (0.41 m^2/m^3) at 149°C (300°F) and a plate spacing of 27.9 cm (11 in.). Since the flue gas flow rate for the PTC is 3.67 scmm (130 scfm), the gas velocity through the ESP is 1.5 m/min (5 ft/min). The ESP has an electrically isolated plate that is grounded through an ammeter, allowing continual monitoring of the actual plate current to ensure consistent operation of the ESP from test to test.

Table 2-1
Coal Information

Organization	State	Mine	Coal	Location	Rank[a]	Production, t/yr	Average S, wt%
KenAmerican Resources, Inc.	KY	Paradise	Western Kentucky No. 9	Illinois Basin	hvBb[b]	1,943,910[c]	5.5
Kennecott Energy Co.	WY	Cordero Rojo Complex	Wyodak–Anderson	Powder River Basin	subC[d]	37,011,000[c]	0.32
Arch Coal Sales, Inc.	VA and KY	Band Mill (Pardee Complex)	Taggart	Appalachian Basin	hvAb[e]	1,700,000[f]	0.77
Consolidation Coal Co.	PA	Blacksville No. 2	Pittsburgh	Appalachian Basin	hvAb	3,898,360[c]	1.97

[a]Determined according to American Society for Testing and Materials (ASTM) Designation D388-88.
[b]High-volatile B bituminous.
[c]1998 mine production statistic from *Keystone Coal Industry Manual*, Intertec Publishing Corporation, Chicago, Illinois, 2000, 793 p.
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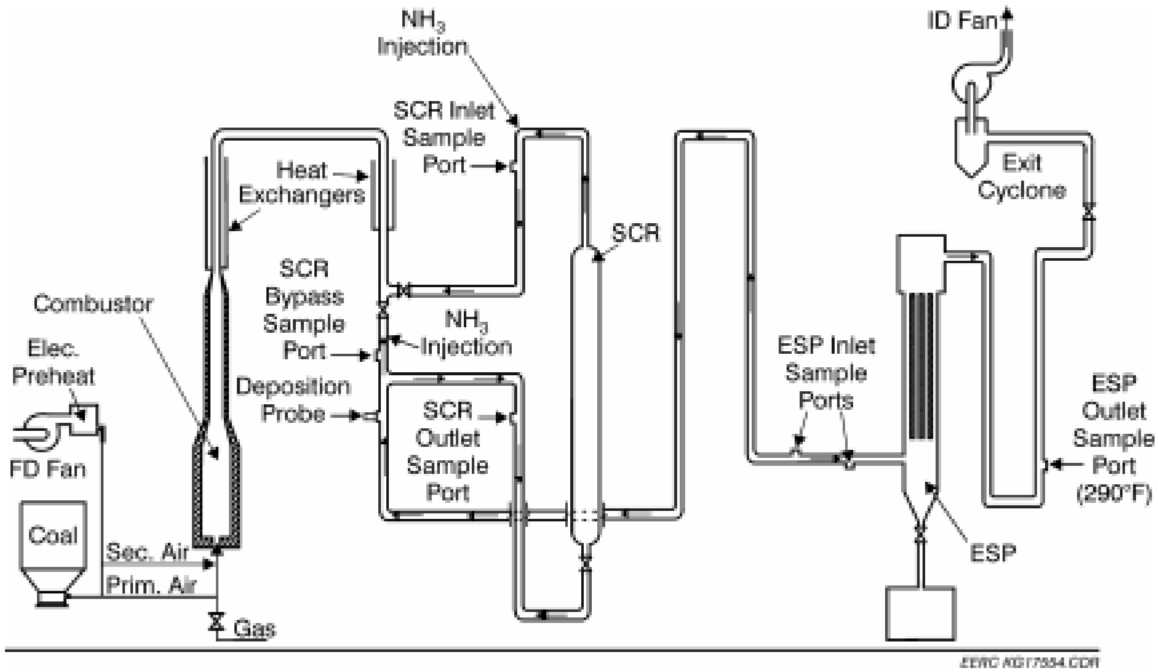

Figure 2-1
Schematic of the EERC Pilot-Scale Combustor

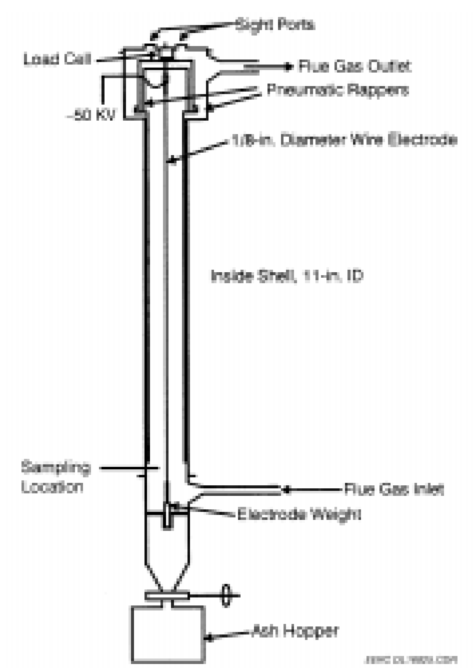

Figure 2-2
Schematic of the EERC ESP

2.2.2 SCR Reactor

An SCR reactor, manufactured and supplied by Cormetech, Inc., was installed on the PTC. The SCR reactor is 5.5 m (18 ft) tall and 0.02 m (0.67 ft) in diameter and contains 0.088 m^3 (3.1 ft^3) of catalyst surface area, providing a space velocity of 2500 hr^{-1}. This is approximately the same space velocity of the SCR installed by Cormetech at TVA's Paradise Station. The SCR designed for the PTC was built in three sections. Each section held one module of a honeycomb-type vanadium/titanium catalyst. Enough space was provided between each module so that the modules could be backflushed. Photographs of the SCR and the catalyst are shown in Figures 2-3 and 2-4. At the request of Cormetech, prior to being used for the tests, the SCR catalyst was conditioned for several days with flue gas and sulfur dioxide (SO_2). The flue gas was generated by firing natural gas in the PTC during which SO_2 was added directly to the combustor to achieve about 1000 ppm in the flue gas.

The SCR reactor was designed to operate at 343°–371°C (650°–700°F) with minimal temperature loss. This was accomplished using electric heaters and insulation. Following the SCR reactor, a cooling loop was added to bring the ESP inlet temperature down to about 350°F (173°C).

2.2.3 Ammonium Injection System

The NH_3 used for all the tests was obtained from a tank of anhydrous NH_3. As shown in Figure 2-1, the NH_3 was injected in the SCR bypass and SCR inlet locations. These locations were selected to ensure that good mixing of the NH_3 with the flue gas occurred prior to its

Figure 2-3
Photograph of the SCR Reactor

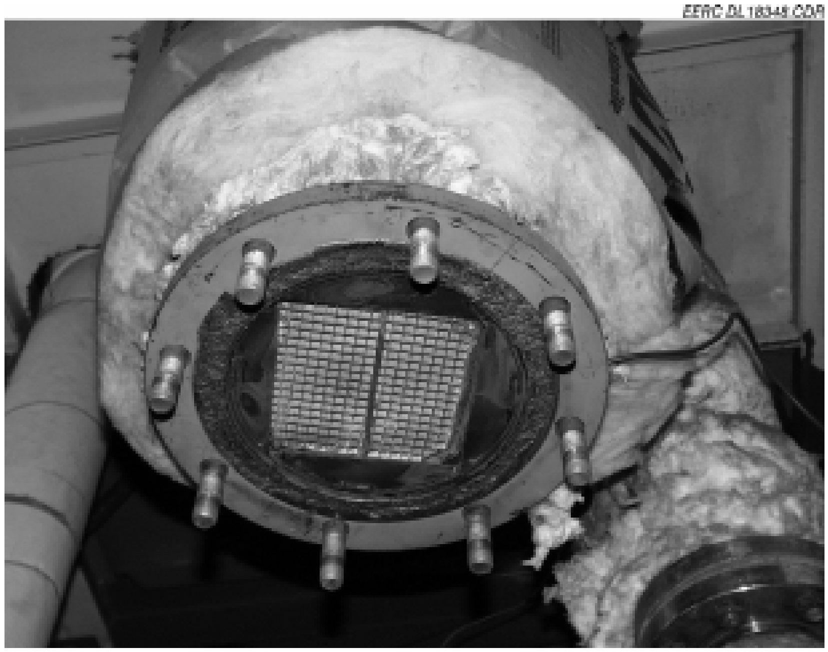

Figure 2-4
Photograph of the SCR Catalyst Following Test

entering the SCR reactor; the NH_3 injection nozzle was designed to aid good mixing. To accurately meter the NH_3 into the combustion system, flowmeters were used. The flowmeters were calibrated prior to the first test firing the Pardise coal; unfortunately, during the tests firing the Cordero Rojo and the Band Mill coals, the calibration of the flowmeter was in error, and about 20% more NH_3 was added than was thought. After another calibration check was conducted, a correction was made prior to testing the Blacksville coal. It is unknown why the flowmeters went out of calibration, as the results from the first test appeared to indicate that they were within specifications. It is possible that the anhydrous NH_3 tank pressure or temperature changed. Gaseous NH_3 and particle-associated NH_3 concentrations were measured at the ESP inlet during every combustion test.

2.3 Test Matrix

As described in Table 2-2, four test series (Tests 607–609, 610–612, 613–615, and 616–618) were conducted firing four different coals in the PTC. The first test condition (Tests 607, 610, 613, and 616) was to provide baseline mercury emission and speciation data. The second test condition (Tests 608, 611, 614, and 617) was to determine if adding a small amount of NH_3 (10 or 25 ppm) to the flue gas impacted mercury speciation. The third condition with each coal (Tests 609, 612, 615, and 618) was the SCR tests. NH_3 was added just upstream of the SCR reactor at a targeted NH_3-to-NO_x stoichiometric ratio of 1. Again, this is the stoichiometric ratio used at TVA's Paradise Station. By comparing the baseline results to the results obtained by injecting NH_3 to those obtained during SCR tests, the impact of NH_3 injection and/or SCR catalyst on mercury speciation was determined.

Table 2-2
Combustion Test Matrix

Test	Coal	Duration, hr	NH_3 Injection, ppmv	SCR Unit	Test Condition
607	Paradise	34.2	None	Bypassed	Baseline
608	Paradise	30.5	25*	Bypassed	NH_3 injection
609	Paradise	38.1	~750	Flow through	SCR
610	Cordero Rojo	38.3	None	Bypassed	Baseline
611	Cordero Rojo	27.4	10	Bypassed	NH_3 injection
612	Cordero Rojo	33.9	~750	Flow through	SCR
613	Band Mill	37.1	None	Bypassed	Baseline
614	Band Mill	28.4	10	Bypassed	NH_3 injection
615	Band Mill	37.6	~750	Flow through	SCR
616	Blacksville	32.6	None	Bypassed	Baseline
617	Blacksville	31.7	10	Bypassed	NH_3 injection
618	Blacksville	37.7	~900	Flow through	SCR

* Reduced to 10 ppm for subsequent tests.

Unless otherwise noted, for the purposes of this report, the following conventions will be used to describe the test conditions:

- Test Condition 1 – Baseline
- Test Condition 2 – NH_3 injection
- Test Condition 3 – SCR

2.4 Sampling and Analytical Methods

2.4.1 Coal and Ash Sampling and Analysis

Coal and ESP hopper ash were sampled and analyzed using the methods indicated in Table 2-3. In addition, a deposition probe, maintained at a surface temperature of ≈ 400°F (204°C), was inserted downstream of the SCR reactor during each test to simulate an air preheater tube. The deposits that collected on this tube, however, could only be analyzed for a very limited number of analytes because of the small amount of deposition that occurred during each test period (~30 hr).

Table 2-3
Coal and Fly Ash Sampling and Analytical Methods

Sample Type	Sampling Method(s)	Analyte(s)	Analytical Method(s)
Coal	Grab composite sampling (ASTM D2234)	Hg	CVAAS[a] (EPA 245.1 and SW-846 Method 7470)
		Na, Mg, Al, Si, P, S, K, Ca, Sr, Mn, Ti, Ba, and Fe	XRF[b] (ASTM D4326)
		Cl	ASTM D4208 and oxidative hydrolysis microcoulometry (EPA SWA-846[c])
		S, C, H, N, O, moisture, ash, heating value, fixed C, and volatile matter	Ultimate (ASTM D3176) and Proximate (ASTM D3172 and D5142)
Fly Ash	Grab composite sampling (EPA Method S007) Cascade 5-stage impactor Sampling filter from the OH mercury speciation sampling method	Hg	CVAAS (EPA SW-846 Method 7470)
		NH_3 (P)	Selective ion electrode (Standard Method 4500-B NH_3)
		Na, Mg, Al, Si, P, S, K, Ca, Ti, Mn, and Fe	XRF (ASTM D4326)
		C	Leeman Labs Model CE440 elemental analyzer
		LOI[d]	ASTM C114
		Cl	Ion chromatography

[a]Cold-vapor atomic absorption spectroscopy.
[b]X-ray fluorescence spectrometry.
[c]Performed by HawkMtn Labs, Inc.
[d]Loss on ignition (LOI).

In addition to collecting and analyzing ESP hopper ash samples, ash samples were also collected at the ESP inlet during Test 609 using a heated 5-stage cascade multicyclone sampler developed by Smith and Wilson [21]. The purpose for doing this sampling procedure was to determine the particle-size distribution of the fly ash and to determine if the mercury is preferentially concentrated on the finer particle fraction. The cascade cyclones were coated with polytetrafluoroethylene (PTFE) to minimize metal contamination. The 50% cutoff diameters (d_{50}) for the cyclone stages were 7.2, 4.0, 2.2, 1.5, and 0.6 µm, based on the actual flue gas conditions (temperature, pressure, gas composition, etc.). The filter catch d_{50} was assumed to be

≈0.4 µm. Multicyclone measurement results were extrapolated using a cubic spline fit procedure described by McCain et al. [22] to evaluate the particle-size distribution of fly ash.

The third method used to collect fly ash samples was the sample filter used as part of the Ontario Hydro (OH) mercury speciation sampling method. These samples were collected isokinetically according to the method (discussed in the next section). The ash collected on the sample filters was then analyzed as shown in Table 2-3.

2.4.2 Flue Gas Sampling and Analysis

As part of each test series (Table 2-2), flue gas samples were collected and analyzed for mercury, NH_3, SO_3, and Cl. The number of samples collected at each location and the average temperature at the sampling location are summarized in Table 2-4. The actual measured temperatures at each sampling location are shown in Appendix A. The temperatures were generally consistent during

Table 2-4
Flue Gas Sampling Methods, Frequencies, Locations, and Temperatures at Each Test Condition

Test Condition	Analyte	Sampling Method	Samples Taken	Sampling Location[a]	Av. Temp., °F
Baseline and NH_3 Injection	Hg	OH method	1	SCR bypass	615
Baseline and NH_3 Injection	Hg	OH method	3	ESP inlet	350
Baseline and NH_3 Injection	Hg	OH method	3	ESP outlet	295
Baseline and NH_3 Injection	Cl	EPA Method 26A	2	ESP inlet	350
Baseline and NH_3 Injection	NH_3	EPA Method 27	2	ESP inlet	350
Baseline and NH_3 Injection	SO_3	Selective condensation[b]	2	ESP inlet	350
SCR	Hg	OH method	1	SCR inlet	650
SCR	Hg	OH method	3	SCR outlet	615
SCR	Hg	OH method	3	ESP inlet	350
SCR	Hg	OH method	3	ESP outlet	295
SCR	Cl	EPA Method 26A	2	ESP inlet	350
SCR	NH_3	EPA Method 27	2	ESP inlet	350
SCR	SO_3	Selective condensation[b]	2	ESP inlet	350

[a]The sampling locations are shown in Figure 2-1.
[b]Described by DeVito and Smith [23].

all the coal combustion tests. In addition to the flue gas samples collected (Table 2-4) the flue gas was also analyzed for SO_2, NO_x, CO, CO_2, and O_2 concentrations at the furnace outlet. SO_2, NO_x, and O_2 concentrations were also measured at the ESP inlet. These gas concentration measurements were recorded every 5 minutes by a data acquisition system.

2.4.2.1 Ontario Hydro (OH) Method

Mercury speciation analyses for each test were conducted using the OH mercury speciation method. The mercury emission and speciation measurement capabilities of the OH method have been validated through dynamic spike tests and method intercomparisons [24]. Accordingly, this method is being considered by the ASTM Subcommittee D22.03.01 on Sampling and Analysis of Atmospheres as a standard test method for measuring mercury species from coal-fired stationary sources. A detailed description of the OH method is available on an EPA Web site at http://www.epa.gov/ttn/emc under preliminary methods.

It should be noted that the three OH samples were taken as pairs at the ESP inlet and outlet sampling locations. Each of the triplicated pairs was sequential. By comparing the average mercury results, the effect of the SCR reactor and NH_3 injection on mercury speciation was determined and the ESP mercury removal efficiencies calculated.

The samples were analyzed using CVAAS, as stated in the OH method. The mercury mass balances were calculated from the average coal mercury concentrations, coal feed rates, average flue gas flows, total mercury concentrations measured at each sampling location, dust loadings, ESP fly ash collection efficiencies, and ESP hopper ash mercury concentrations. Mercury mass balance closures of 75%–125% are considered acceptable based on experience with the OH method [24, 25]. Sample calculations and mass balance data for mercury are shown in Appendices B and C, respectively.

2.4.2.2 Mercury On-Line Analyzers

During Tests 616–618 (Blacksville coal), two continuous mercury analyzers, a PS Analytical and a Semtech Hg 2010, were used to measure Hg^0 and total mercury concentrations at the ESP outlet sampling location. The PS Analytical instrument is based on atomic fluorescence principles, whereas the Semtech Hg 2000 is a portable Zeeman-modulated CVAAS. These instruments are discussed in detail in a paper recently presented at the Air Quality II Conference [26]. The fly ash-sampling components of an EPA Method 29 sampling train, a glass nozzle and probe, and quartz-fiber filter maintained at the flue gas temperature, were used to obtain particle-free gas samples for analysis. After particle filtration, a proprietary flue gas-conditioning system was used to remove acid gases and reduce any Hg^{2+} present to Hg^0 for subsequently measuring total mercury.

2.4.2.3 Ammonia Analysis and Measurements

Once the NH_3 samples were collected using EPA Method 27, the solutions were analyzed using a selective ion electrode. NH_3 sample calculations and mass balance data for the Paradise, Cordero

Rojo, Band Mill, and Blacksville combustion flue gases are also presented in Appendices B and C. The flue gas NH_3 measurement results for the baseline tests firing the Paradise and Blacksville coals were greater than corresponding results when NH_3 injected. This suggests that the baseline NH_3 concentrations for these two coals are biased high. The suspected positive bias in the baseline Paradise and Blacksville NH_3 measurements is manifested in low NH_3 mass balance closures for the NH_3 injection tests. The reason for the high NH_3 measurements in the two baseline tests is unknown. It is possible that there was some contamination on the filter or solution or there was something that was interfering with the measurement.

The targeted NH_3-to-NO_x stoichiometric ratio for all of the SCR tests was 1. As discussed previously, because of a flowmeter calibration error, the ratio for the Cordero Rojo and Band Mill tests was about 1.2. The flowmeter calibration was corrected prior to the Blacksville test. Table 2-5 presents the NH_3-to-NO_x stoichiometric ratio for each test series.

Table 2-5
Comparison of Triplicate-Measured NH_3/NO_x During the SCR Tests*

Sample	Paradise	Cordero Rojo	Band Mill	Blacksville
1	0.89	1.31	1.37	0.97
2	0.97	1.15	1.22	0.96
3	1.10	1.13	1.16	0.98
Average	0.99 ± 0.11	1.20 ± 0.10	1.25 ± 0.11	0.97 ± 0.01

* These results are based on the average NO_x concentration measured at the combustor outlet and the NH_3 mass balance (NO_x reduction plus NH_3 slip).

3 RESULTS

3.1 Coal Compositions

The proximate–ultimate analysis results for the four test coals are presented in Table 3-1. Composite coal samples were taken directly at the outlet of the pulverized coal feeder. As can be seen, the Paradise and Blacksville bituminous coals have a much higher sulfur concentration than the Cordero Rojo subbituminous and Band Mill bituminous coals. As expected, the moisture content was much higher and the heating value lower for the PRB subbituminous coal compared to the three bituminous coals.

The concentrations of the major and minor elements in the coal ash as measured by XRF are presented in Table 3-2. Again, as expected, the Cordero Rojo PRB subbituminous coal had much higher concentrations of the alkaline elements than the bituminous coals, particularly calcium. The bituminous coals, however, had much higher concentrations of silica and alumina in the coal ash. Comparing the three bituminous coals, there are also clear differences. In addition to high S and Cl concentrations, the Paradise and Blacksville coals had a higher Fe_2O_3 concentration than the Band Mill coal. The Blacksville coal had a higher concentration of CaO and MgO in the coal ash than the other two bituminous coals. As will be discussed in Chapters 3 and 4 of this report, these differences may be important in explaining some of the results observed in this study.

As shown in Table 3-3, there is a difference in the mercury and Cl concentrations among the four coals. The mercury contents of the Paradise, Cordero Rojo, and Blacksville coals are very similar at ≈0.1 ppm. The Band Mill coal, however, is distinguished by a very low mercury content. Chloride concentration was much higher in the Paradise and Blacksville coals relative to the other two coals. Chloride in the Cordero Rojo coal was not detected by the EERC using ASTM Method D4208; however, HawkMtn Labs, Inc., was able to quantify Cl using oxidative hydrolysis microcoulometry (EPA SWA-846). As anticipated, the S:Cl ratios of the four coals range widely.

3.2 Pilot-Scale Coal Combustion Tests

3.2.1 Paradise Bituminous Coal (Illinois Basin)

3.2.1.1 Flue Gas Compositions

Average Paradise coal combustion flue gas compositions are presented in Table 3-4. SO_3 concentrations decreased for the NH_3 injection and SCR tests compared to the baseline. This

Table 3-1
Coal Ultimate–Proximate Analysis Results, as-received wt%

Analysis Parameters	Paradise (Illinois Basin)	Cordero Rojo (PRB)	Band Mill (Appalachian Basin)	Blacksville (Appalachian Basin)
Proximate Analysis				
Moisture	5.40 ± 0.27	23.1 ± 2.7	1.43 ± 0.10	2.00 ± 0.00
Volatile Matter	39.2 ± 0.6	36.7 ± 1.4	35.5 ± 0.7	36.6 ± 0.3
Fixed Carbon	45.8 ± 0.8	34.7 ± 1.0	55.8 ± 0.5	52.8 ± 0.2
Ash	9.56 ± 0.15	5.56 ± 0.23	7.22 ± 1.10	8.61 ± 0.15
Heating Value, Btu/lb	12,040 ± 190	8,610 ± 700	13,650 ± 550	13,050 ± 110
Ultimate Analysis				
Carbon	67.0 ± 0.2	50.6 ± 2.1	77.8 ± 1.5	72.7 ± 1.1
Hydrogen[a]	4.64 ± 0.03	3.60 ± 0.11	4.95 ± 0.06	4.71 ± 0.11
Nitrogen	2.22 ± 0.05	1.35 ± 0.13	2.32 ± 0.05	2.31 ± 0.07
Sulfur	3.10 ± 0.07	0.52 ± 0.04	0.75 ± 0.05	2.00 ± 0.04
Number of Analyses	4	3	3	3
Ash	9.56 ± 0.15	5.56 ± 0.23	7.22 ± 1.10	8.61 ± 0.15
Oxygen (by difference)[a]	8.11 ± 0.31	15.3 ± 0.6	5.56 ± 0.95	7.71 ± 1.15
Total Moisture	5.40 ± 0.27	23.1 ± 2.7	1.43 ± 0.10	2.00 ± 0.0
Number of Analyses	4	3	3	3

[a]Hydrogen and oxygen do not include H and O in sample moisture.

observation is believed to result from the formation of bisulfate and/or sulfates because of reaction with NH_3. Chloride concentrations were very consistent, ranging from 25–29 ppmv. NO_x concentrations increased because of the gradual temperature increase that occurred in the combustion system with increased operating time as refractory in the combustor reached thermal stability. On average, the SCR reactor reduced the Paradise flue gas NO_x concentration by 98%. As indicated in Figure 3-1, most of the NH_3 slip is associated with ash particles, although probably in a sulfated form.

Table 3-2
Coal Ash – Major and Minor Element Oxide Compositions, wt%

Element	Paradise	Cordero Rojo	Band Mill	Blacksville
SiO_2	46.2 ± 0.6	24.3 ± 0.5	54.6 ± 0.6	42.1 ± 0.3
Al_2O_3	20.9 ± 0.4	15.2 ± 0.8	27.6 ± 3.0	22.2 ± 0.8
Fe_2O_3	23.6 ± 0.2	6.66 ± 0.71	7.12 ± 0.13	17.3 ± 0.8
MnO	NA[a]	0.036 ± 0.003	0.054 ± 0.007	0.029 ± 0.002
TiO_2	1.00 ± 0.00	1.43 ± 0.11	1.45 ± 0.06	0.92 ± 0.03
BaO	NA	0.46 ± 0.04	0.08 ± 0.03	0.073 ± 0.016
P_2O_5	0.10 ± 0.00	0.91 ± 0.05	0.17 ± 0.01	0.49 ± 0.01
CaO	1.73 ± 0.28	23.5 ± 1.6	0.82 ± 0.04	5.53 ± 0.22
MgO	1.50 ± 0.00	4.46 ± 0.42	1.67 ± 0.12	1.58 ± 0.05
Na_2O	0.40 ± 0.00	1.26 ± 0.05	0.61 ± 0.01	0.89 ± 0.03
K_2O	2.20 ± 0.00	0.32 ± 0.04	2.66 ± 0.28	1.59 ± 0.03
SO_3	2.28 ± 0.44	23.1 ± 5.0	0.67 ± 0.40	7.04 ± 0.12
Total	99.8 ± 0.1	101.6 ± 0.7	97.4 ± 2.3	99.8 ± 0.1
No. of Samples	4	3	3	3

Table 3-3
Coal Hg and Cl Concentrations (as-received ppm) and S–Cl Ratios (as-received basis)

Element	Laboratory	Paradise	Cordero Rojo	Band Mill	Blacksville
Hg	EERC	0.111 ± 0.002[a]	0.085 ± 0.012[b]	0.022 ± 0.001[b]	0.094 ± 0.006[b]
Cl	EERC	350 ± 44[a]	<50	58 ± 12[b]	758[c]
Cl	HawkMtn	454 ± 19[b]	8.7 ± 2.6[b]	59 ± 3[b]	NA[d]
S/Cl	EERC	77	598	128	26

[a]Calculated from four analyses.
[b]Calculated from three analyses.
[c]Average of duplicate analyses.
[d]Not analyzed.

Table 3-4
Average Paradise Bituminous Coal (Illinois Basin) Combustion Flue Gas Compositions

	Baseline		NH_3 Injection		SCR	
	Average	Std. Dev.[a]	Average	Std. Dev.	Average	Std. Dev.
SO_2, ppmv[b]	2480	130	2480	160	2500	170
SO_2, ppmv[c]	2400	130	2370	130	2400	210
SO_3, ppmv[c]	8.2	NA[d]	0.7	NA	0.5	NA
CO, ppmv[b]	21.0	9.5	24.1	17.0	29.3	19.6
CO_2, mol%[b]	14.1	0.8	14.1	1.0	14.5	1.2
O_2, mol%[b]	4.27	0.79	4.36	1.01	4.37	0.95
O_2, mol%[c]	5.34	1.33	5.51	1.25	4.47	1.36
NO_x, ppmv[b]	632	82	648	95	760	120
NO_x, ppmv[c]	586	76	605	89	16.5	18.9
NH_3, ppmv[c]	6.69	NA	2.50	0.17	1.14	NA
Cl, ppmv[c]	26.3	NA	29.0	NA	25.2	NA

[a] Population or sample standard deviation.
[b] Measured at combustor outlet.
[c] Measured at ESP inlet (gaseous NH_3 only).
[d] Not applicable because only one or two measurements were made.

3.2.1.2 Mercury Speciation

Average mercury speciation results and mass balances for the Paradise coal combustion flue gases produced during the baseline, NH_3 injection, and SCR tests are presented in Tables 3-5–3-7 and shown graphically in Figures 3-2 through 3-4, respectively. Mass balances were acceptable, except during NH_3 injection tests when they were low (≤75%). This appears to be attributable to the very low dust loading collected at the ESP inlet. About 60% of the Hg^0 liberated from Paradise coal during combustion was oxidized at ≥340°C (≥650°F) at the SCR bypass and SCR inlet locations. An additional 30% of the Hg^0 oxidized through the cooling loop as the temperature decreased to≥175°C (≥350°F) as measured at the ESP inlet. Comparing the mercury species concentrations in Figures 3-2 and 3-3, it appears that the injection of 25 ppmv NH_3 promoted the formation of Hg_p upstream of the ESP inlet. A concurrent decrease in Hg^{2+} and increase in Hg_p suggests that NH_3 promoted the capture of Hg^{2+} by the fly ash. For the SCR test, a comparison of the SCR inlet and outlet mercury species concentrations in Figure 3-4 indicates that the SCR of NO_x promoted Hg^{2+} formation.

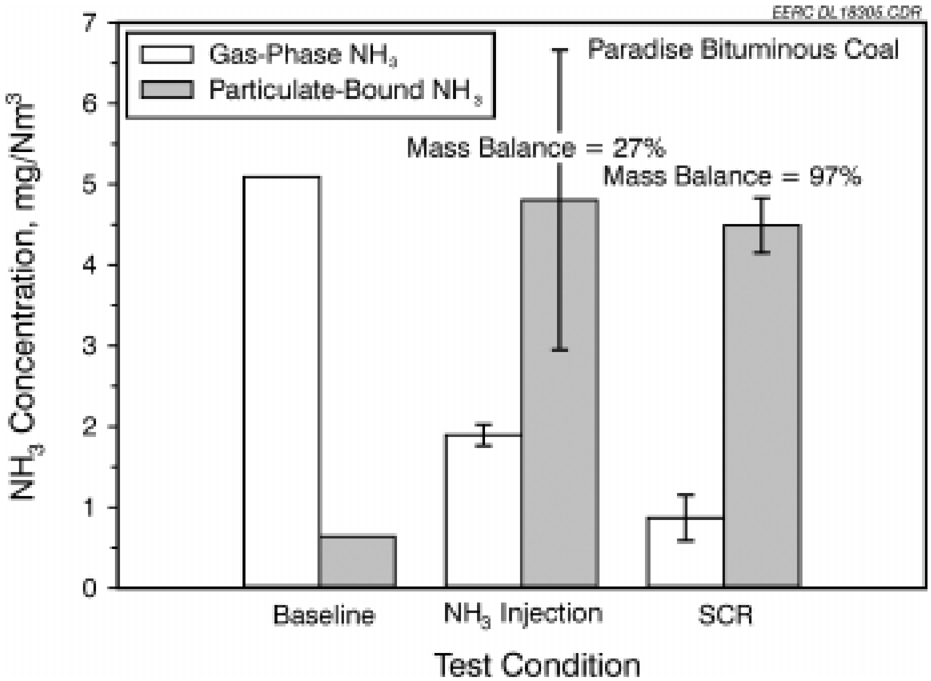

Figure 3-1
Ammonia Concentrations and Mass Balances for the Paradise Tests

Table 3-5
Mercury Speciation Results for the Paradise Baseline (Test 607) Test, on a dry, 3% O_2 basis

Sample Location	Hg^0, µg/Nm^3	Hg^{2+}, µg/Nm^3	Hg_p, µg/Nm^3	Total Hg, µg/Nm^3	Mass Balance, %
SCR Bypass	4.57	7.74	0.01	12.3	90
ESP Inlet	0.44	11.2	0.65	12.3	90
ESP Inlet	0.76	11.0	0.47	12.2	89
ESP Inlet	0.38	9.34	0.82	10.5	77
Average	0.53	10.5	0.65	11.7	85
Std. Dev.	0.20	1.0	0.18	1.0	7
ESP Outlet	0.87	8.71	0.37	9.95	85
ESP Outlet	1.28	9.80	0.03	11.1	94
ESP Outlet	0.35	8.23	1.77	10.4	88
Average	0.83	8.91	0.72	10.5	89
Std. Dev.	0.47	0.80	0.92	0.6	4

Table 3-6
Mercury Speciation Results for the Paradise NH_3 Injection (Test 608) Test, on a dry, 3% O_2 basis

Sample Location	Hg^0, µg/Nm^3	Hg^{2+}, µg/Nm^3	Hg_p, µg/Nm^3	Total Hg, µg/Nm^3	Mass Balance, %
SCR Bypass	4.54	6.00	0.19	10.7	75
ESP Inlet	1.40	2.75	3.69	7.84	55
ESP Inlet	0.92	4.41	3.82	9.15	64
ESP Inlet	1.54	3.52	5.37	10.4	73
Average	1.29	3.56	4.29	9.14	64
Std. Dev.	0.33	0.83	0.93	1.30	9
ESP Outlet	0.62	1.98	2.56	5.16	45
ESP Outlet	0.31	1.65	4.43	6.39	54
ESP Outlet	0.63	2.51	3.47	6.61	55
Average	0.52	2.05	3.49	6.05	51
Std. Dev.	0.18	0.43	0.94	0.78	5

The SCR of NO_x also enhanced Hg_p formation between the SCR outlet and the ESP inlet. The increase in Hg_p concentration was much more pronounced when the SCR reactor was utilized compared to the NH_3 injection test.

3.2.1.3 Chemical Composition of Fly Ashes and Deposits

In Figure 3-5, the mercury concentrations of the collected fly ash samples are compared. As can be seen in Figure 3-5, the simulated air preheater deposits are depleted in mercury relative to the other fly ash samples. Also, it appears that ash samples collected during the NH_3 injection and SCR tests are more concentrated in mercury relative to corresponding samples collected from the baseline test. Mercury is most concentrated in the ESP outlet ash samples, indicating that mercury is associated primarily with fine ash particles that escape the ESP. As would be expected, the mercury concentration is greatest on the OH filter. The OH filter was a much more efficient collector than the EERC ESP. In addition, it is possible that additional deposition occurs because of the better ash-to-gas contact.

The chemical compositions of ESP hopper ashes generated during the Paradise coal tests are shown in Table 3-8. Fly ashes produced during the NH_3 injection and SCR tests have higher CaO, SO_3, NH_3, and Cl concentrations relative to the baseline. These increases, with the exception of CaO, may be a result of NH_3 and SO_3 and NH_3 and Cl reactions. LOI and carbon analysis results for the Paradise ESP hopper ashes are compared in Figure 3-6. LOI concentrations are greater than the corresponding carbon concentrations for a given ash,

Table 3-7
Mercury Speciation Results for Paradise SCR (Test 609) Test, on a dry, 3% O_2 basis

Sample Location	Hg^0, µg/Nm^3	Hg^{2+}, µg/Nm^3	Hg_p, µg/Nm^3	Total Hg, µg/Nm^3	Mass Balance, %
SCR Bypass (Test 607)	4.57	7.74	0.01	12.3	90
SCR Bypass (Test 608)	4.54	6.00	0.19	10.7	75
SCR Inlet (Test 609)	3.58	5.29	0.01	8.88	68
Average	4.23	6.34	0.07	10.6	77
Std. Dev.	0.56	1.26	0.10	1.7	12
SCR Outlet	0.88	8.30	1.21	10.4	79
SCR Outlet	1.30	8.14	0.10	9.54	73
SCR Outlet	1.08	13.6	0.01	14.7	112
Average	1.09	10.0	0.44	11.5	88
Std. Dev.	0.21	3.1	0.67	2.8	21
ESP Inlet	<0.03	0.03	12.1	12.1	92
ESP Inlet	0.06	3.95	7.19	11.2	85
ESP Inlet	0.03	<0.03	11.0	11.0	84
Average	0.03	1.33	10.1	11.4	87
Std. Dev.	NA[a]	NA	2.6	0.60	5
ESP Outlet	<0.03	<0.03	8.30	8.30	98
ESP Outlet	0.06	2.51	5.72	8.29	98
ESP Outlet	0.03	<0.02	3.52	3.55	62
Average	0.03	0.84	5.85	6.71	86
Std. Dev.	NA	NA	2.39	2.74	21

[a] Not applicable because less than three analyte concentrations were greater than the lower limit of quantitation.

indicating that additional volatile components (bisulfates/sulfates, carbonates, etc.) were released from ashes during the LOI analysis. It can be seen in Figure 3-6 that the LOI increased for the two conditions with NH_3 injection. Based on the carbon concentration, the LOI for the SCR test is due to an increase in other volatile components and may be attributable to the greater S, N, and Cl contents (Table 3-4). Carbon is more concentrated in fly ashes produced during the NH_3 injection and SCR tests.

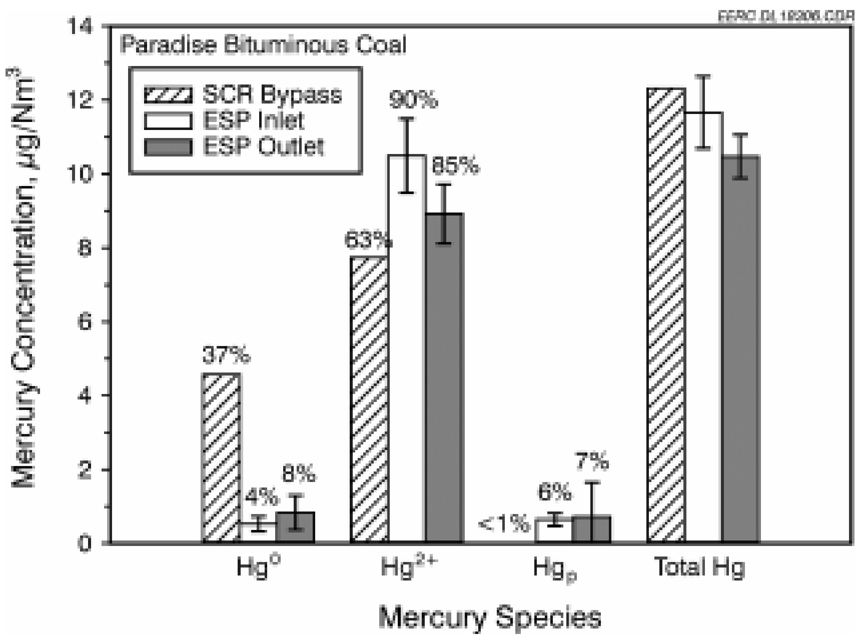

Figure 3-2
Mercury Speciation Results for the Paradise Baseline Test (The relative proportions of each mercury species are shown by the percentages above the bars.)

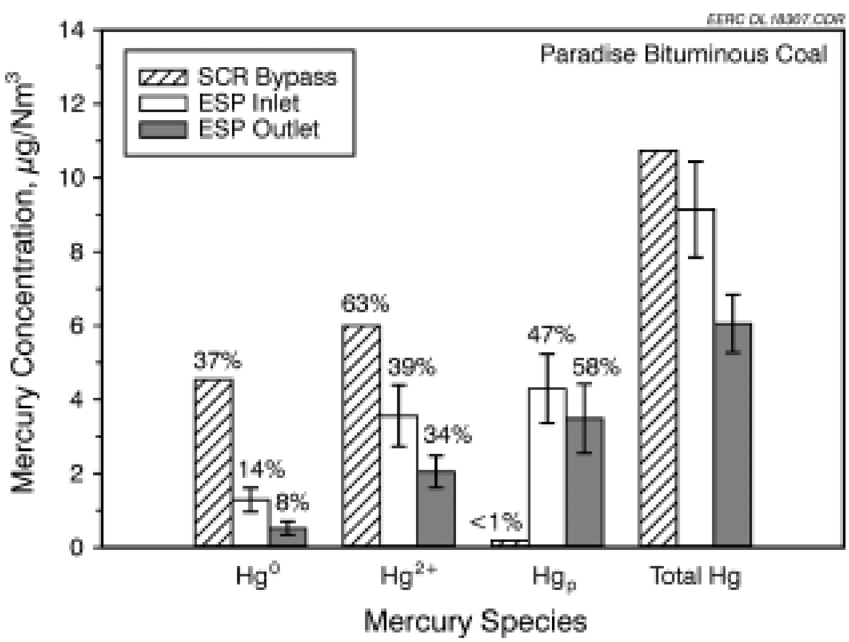

Figure 3-3
Mercury Speciation Results for the Paradise NH_3 Injection Test (The relative proportions of each mercury species are shown by the percentages above the bars.)

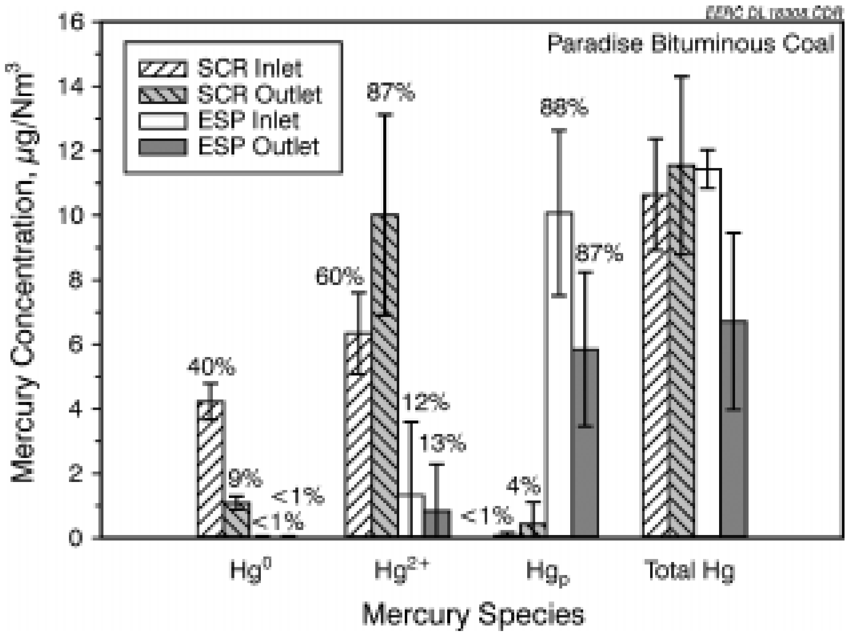

Figure 3-4
Mercury Speciation Results for the Paradise SCR Test (The relative proportions of each mercury species are shown by the percentages above the bars.)

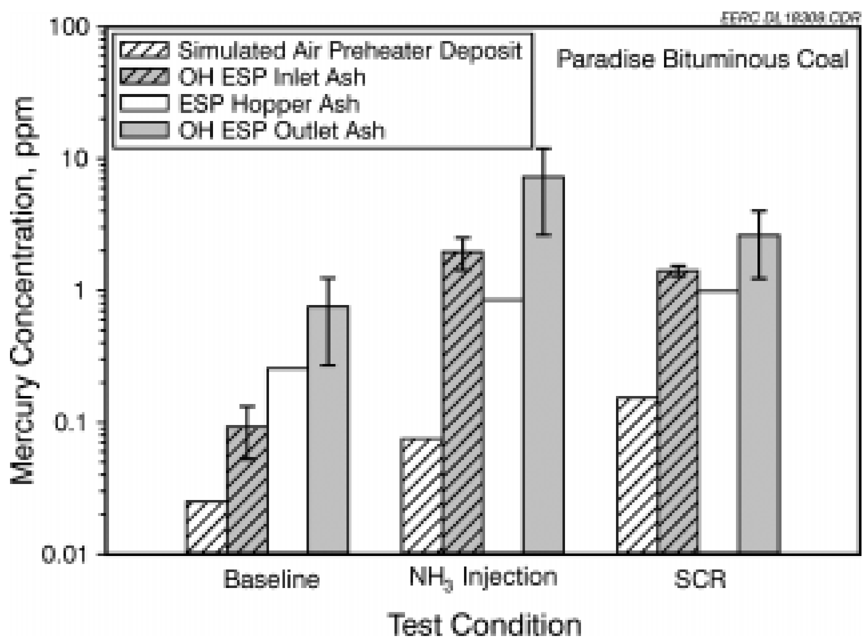

Figure 3-5
Mercury Concentration in the Paradise Ash Samples

Table 3-8
Paradise ESP Hopper Ash – Major, Minor, and Trace Element Compositions

Element, wt%	Baseline Test	NH_3 Injection Test	SCR Test
SiO_2	50.9	50.4	50.9
Al_2O_3	21.9	21.8	21.8
Fe_2O_3	24.5	24.8	24.2
TiO_2	1.11	1.05	1.12
P_2O_5	0.12	0.12	0.11
CaO	1.53	1.91	2.20
MgO	1.36	1.35	1.36
BaO	0.049	0.054	0.042
Na_2O	0.36	0.36	0.37
K_2O	2.45	2.41	2.46
SO_3	0.80	0.95	1.15
NH_3	0.004	0.155	0.366
Cl, ppm	19	25	256
Total, wt%	105.1	105.4	106.1

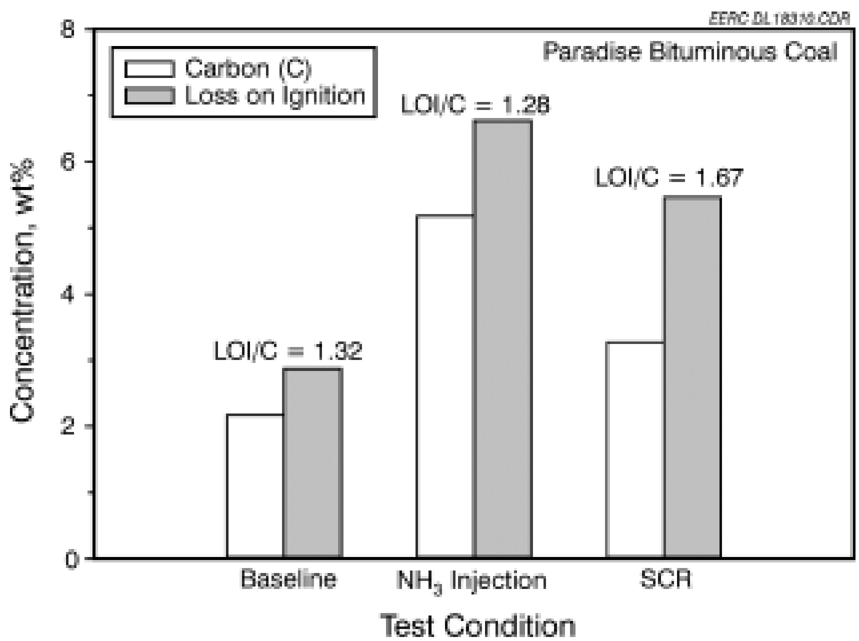

Figure 3-6
Loss on Ignition and Carbon in the Paradise ESP Hopper Ashes

The chemical compositions of the simulated air preheater deposits produced during each of the tests firing the Paradise coal were compared. As was the case for the ESP hopper ashes, these deposits are much more sulfated for the two conditions when NH_3 is injected relative to the baseline deposit. The deposits were not analyzed for Hg, Cl, or N because not enough sample was available.

3.2.1.4 ESP Fly Ash and Total Mercury Collection Efficiencies

The removal efficiencies of the ESP for particulate matter, total mercury, and Hg_p for the Paradise coal tests are shown in Table 3-9. Fly ash removal efficiencies were variable and generally low during the tests. The average ESP removal for the three test conditions was only 87%, 70.8%, and 65.3%, respectively. Modifications to the ESP, however, greatly improved fly ash removal efficiencies during subsequent tests with the other three coals. The ESP was relatively ineffective in removing mercury during the baseline combustion test. Total mercury removal efficiencies were significantly greater during the NH_3 injection and SCR tests relative to the baseline condition. The improvement in ESP mercury removal is attributable to the increase in Hg_p when NH_3 is injected (Figures 3-3 and 3-4). This was the case, even though the ESP efficiency decreased for the three tests.

Table 3-9
Hg_p Removal Efficiency of the ESP for the Paradise Coal

Run No.	Hg_p	% Hg_p Removal	% Total Hg Removal	% Fly Ash Removal
607	0.65	0.0	10.3	87.0
608	4.29	18.6	33.8	70.8
609	10.08	42.0	58.7	63.3

3.2.2 Cordero Rojo Subbituminous Coal (PRB)

3.2.2.1 Flue Gas Compositions

Average Cordero Rojo flue gas compositions are presented in Table 3-10. Chloride and SO_3 concentrations in the flue gases were below detection because the Cordero Rojo coal contains such low S and Cl concentrations (Tables 3-2 and 3-3) and high alkali in relation to the S content. The SCR reactor reduced NO_x concentrations in the Cordero Rojo flue gas by 99%. NH_3 concentrations during the SCR tests were relatively high because of a calibration error in the NH_3 injection system as discussed in Section 2.4.2.3. NH_3 concentrations in the Cordero Rojo baseline, NH_3 injection, and SCR flue gases are compared in Figure 3-7. In contrast to the Paradise tests, gaseous NH_3:particle-associated NH_3 ratios are much greater than 1, indicating that NH_3–ash sorption does not occur to any great extent.

Table 3-10
Average Cordero Rojo Subbituminous Coal (PRB) Combustion Flue Gas Compositions

	Baseline Test		NH_3 Injection Test		SCR Test	
	Average	**Std. Dev.**[a]	**Average**	**Std. Dev.**	**Average**	**Std. Dev.**
SO_2, ppmv[b]	396	61	377	26	349	39
SO_3, ppmv[c]	<0.03	NA[d]	<0.03	NA	<0.03	NA
CO, ppmv[b]	26.6	13.2	21.8	9.2	28.2	15.6
CO_2, mol%[b]	14.3	1.1	14.4	0.9	14.6	0.9
O_2, mol%[b]	4.45	0.84	4.56	0.60	4.43	0.73
O_2, mol%[c]	4.53	0.99	4.48	0.70	4.81	0.90
NO_x, ppmv[b]	860	78	848	61	750	55
NO_x, ppmv[c]	852	67	844	56	6.31	2.93
NH_3, ppmv[c]	0.82	NA	13.5	0.6	143	64
Cl, ppmv[c]	<2	NA	<2	NA	<2	NA

[a] Population or sample standard deviation.
[b] Measured at combustor outlet.
[c] Measured at ESP inlet (gaseous NH_3 only).
[d] Not applicable because the analyte concentration was less than the lower limit of quantitation or only one or two measurements were performed.

3.2.2.2 Mercury Speciation

Average mercury speciation and mass balance results for the Cordero Rojo tests are presented in Tables 3-11–3-13 and graphically in Figures 3-8 through 3-10, respectively. Mercury mass balances were acceptable. Mercury speciation measurements at the SCR bypass and inlet locations indicate that about 25% of the Hg^0 released during combustion of the Cordero Rojo coal is oxidized upstream of the SCR at ≥320°C (610°F). For the Cordero Rojo baseline test (Figure 3-8), about 25% of the Hg^0 becomes associated with the particulate matter between the SCR bypass and ESP inlet sampling locations. Mercury speciation results in Figure 3-9 indicate that the injection of 10 ppmv NH_3 did not promote the formation of Hg^{2+} or Hg_p downstream from the SCR bypass. The mercury speciation results in Figure 3-10 indicate that Hg^0 was not effectively oxidized in the SCR reactor. The relative proportions of Hg^{2+} and Hg_p species downstream from the SCR at the ESP inlet and outlet show an increase in Hg^0 and a decrease in Hg^{2+} compared to the baseline Cordero Rojo flue gas, indicating that the SCR was ineffective in enhancing mercury oxidation and Hg_p formation. This was true, even though the NH_3 slip was very high, >140 ppm.

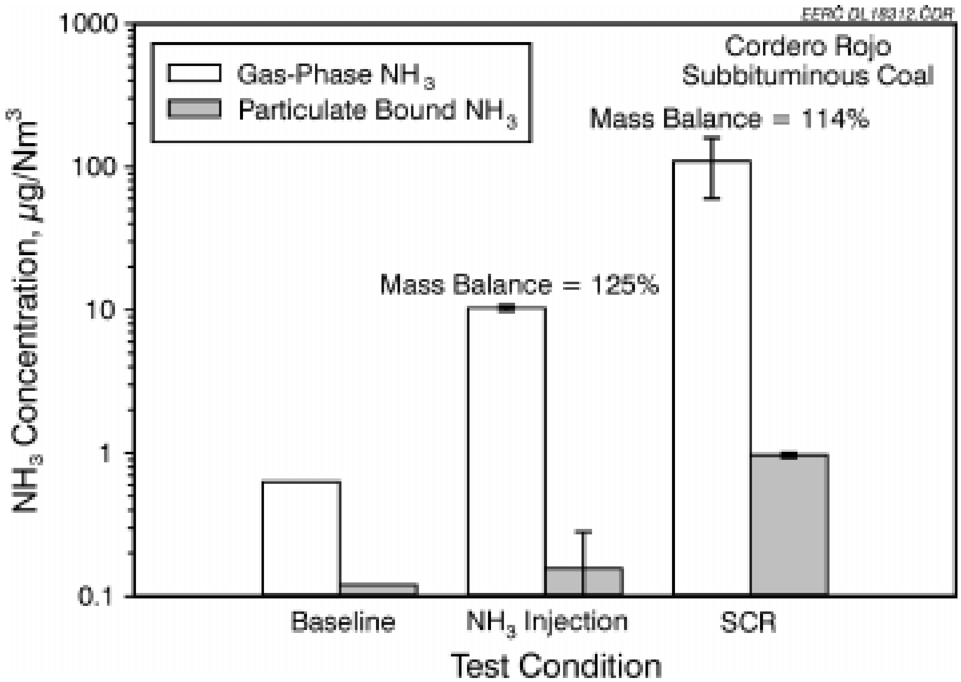

Figure 3-7
Ammonia Concentrations and Mass Balances for the Cordero Rojo Tests

Table 3-11
Mercury Speciation Results for the Cordero Rojo Baseline (Test 610) Test, on a dry, 3% O_2 basis

Sample Location	Hg^0, μg/Nm^3	Hg^{2+}, μg/Nm^3	Hg_p, μg/Nm^3	Total Hg, μg/Nm^3	Mass Balance, %
SCR Bypass	10.3	3.59	0.18	14.1	96
ESP Inlet	3.84	4.23	5.84	13.9	95
ESP Inlet	6.16	2.79	3.87	12.8	87
ESP Inlet	6.84	5.33	0.95	13.1	90
Average	5.61	4.12	3.55	13.3	91
Std. Dev.	1.57	1.27	2.46	0.56	4
ESP Outlet	4.68	2.99	0.29	7.96	67
ESP Outlet	5.28	2.70	0.28	8.26	69
ESP Outlet	6.75	4.39	0.07	11.2	89
Average	5.57	3.36	0.21	9.14	75
Std. Dev.	1.07	0.90	0.12	1.80	12

Table 3-12
Mercury Speciation Results for the Cordero Rojo NH_3 Injection (Test 611) Test, on a dry, 3% O_2 basis

Sample Location	Hg^0, µg/Nm^3	Hg^{2+}, µg/Nm^3	Hg_p, µg/Nm^3	Total Hg, µg/Nm^3	Mass Balance, %
SCR Bypass	13.0	1.82	0.02	14.8	96
ESP Inlet	6.94	3.70	2.69	13.3	87
ESP Inlet	11.7	3.34	0.72	15.8	102
ESP Inlet	12.9	2.68	0.24	15.8	102
Average	10.5	3.24	1.22	15.0	97
Std. Dev.	3.1	0.52	1.30	1.4	9
ESP Outlet	7.33	2.07	0.15	9.55	68
ESP Outlet	9.05	1.93	0.03	11.0	78
ESP Outlet	10.4	1.84	0.04	12.3	86
Average	8.92	1.95	0.07	10.9	77
Std. Dev.	1.53	0.12	0.07	1.4	9

3.2.2.3 Chemical Compositions of Fly Ashes and Deposits

In Figure 3-11, the Hg_p concentrations of the ash samples collected during the Cordero Rojo test are compared. The mercury concentration in the simulated air preheater deposits was $\leq$0.02 ppm. Only the NH_3 injection test produced enough deposit to be analyzed. As was the case for the Paradise tests, there did not appear to be any deposition of mercury, due to the OH filter, as the ESP hopper ash on Hg_p concentration was similar to the Hg_p concentration in ash collected on the ESP inlet OH filter. The Hg_p is more concentrated in the ESP outlet filters for all test conditions, indicating that mercury is preferentially associated with the finer-particle fraction.

The chemical compositions of Cordero Rojo ESP hopper ashes are compared in Table 3-14. In contrast to the Paradise ESP hopper ashes (Table 3-8), neither NH_3 injection nor the SCR catalyst greatly affected the chemical composition of Cordero Rojo fly ash. The NH_3 concentration in the ash is very low for all tests, even though the NH_3 slip was high. This may be related to the very low SO_3 concentration in the flue gas. Combustion efficiency firing Cordero Rojo coal was very high as indicated by low LOI values of <0.01, 0.05, and 0.09 wt% in Figure 3-12 for each of the three test conditions. The carbon content of the ash was <0.01% in all cases. The chemical compositions of Cordero Rojo simulated air preheater deposits could not be determined because insufficient amounts of ash were deposited.

Table 3-13
Mercury Speciation Results for Cordero Rojo SCR (Test 612) Test, on a dry, 3% O_2 basis

Sample Location	Hg^0, μg/Nm^3	Hg^{2+}, μg/Nm^3	Hg_p, μg/Nm^3	Total Hg, μg/Nm^3	Mass Balance, %
SCR Bypass (Test 610)	10.3	3.59	0.18	14.1	96
SCR Bypass (Test 611)	13.0	1.82	0.02	14.8	96
SCR Inlet (Test 612)	14.0	3.04	0.05	17.1	128
Average	12.4	2.82	0.08	15.3	107
Std. Dev.	1.9	0.91	0.09	1.6	19
SCR Outlet	11.5	0.76	0.01	12.2	92
SCR Outlet[a]	1.79[b]	1.43[b]	0.01[b]	3.23[b]	24
SCR Outlet	10.9	0.80	0.01	11.7	88
Average	11.2	0.78	0.01	12.0	90
Std. Dev.	NA[b]	NA	NA	NA	NA
ESP Inlet	2.32	2.69	10.4	15.4	116
ESP Inlet	6.70	5.04	1.42	13.2	99
ESP Inlet	7.22	10.8	0.56	18.6	140
Average	5.41	6.18	4.14	15.7	118
Std. Dev.	2.69	4.18	5.47	2.7	20
ESP Outlet	2.47	0.90	2.41	5.78	54
ESP Outlet	4.75	1.65	0.15	6.55	60
ESP Outlet	5.76	2.65	0.03	8.44	74
Average	4.33	1.73	0.86	6.92	63
Std. Dev.	1.69	0.88	1.34	1.37	10

[a]Value was not considered in calculating an average or in constructing the bar plot associated with these results.
[b]Not applicable because less than three analyte concentrations were greater than the lower limit of quantitation.

3.2.2.4 ESP Fly Ash and Total Mercury Collection Efficiencies

As shown in Table 3-15, the ESP effectively (>99% removal efficiency) captured the Cordero Rojo fly ash. The ESP mercury removal efficiencies were similar for baseline and NH_3 injection test conditions and just slightly better for the SCR test condition. A comparison of ESP inlet and outlet mercury speciation results in Figures 3-8 through 3-10 also suggests that the ESP was more effective in removing Hg^{2+} when the SCR was used. It should be noted that the first sample taken at the ESP inlet location for both NH_3 injection and SCR tests had much higher Hg_p concentrations than the last two. If these two samples were ignored, there would be a reduction in Hg_p as a result of NH_3 injection. Table 3-15 presents the Hg_p removal efficiency for each test as a function of the ESP.

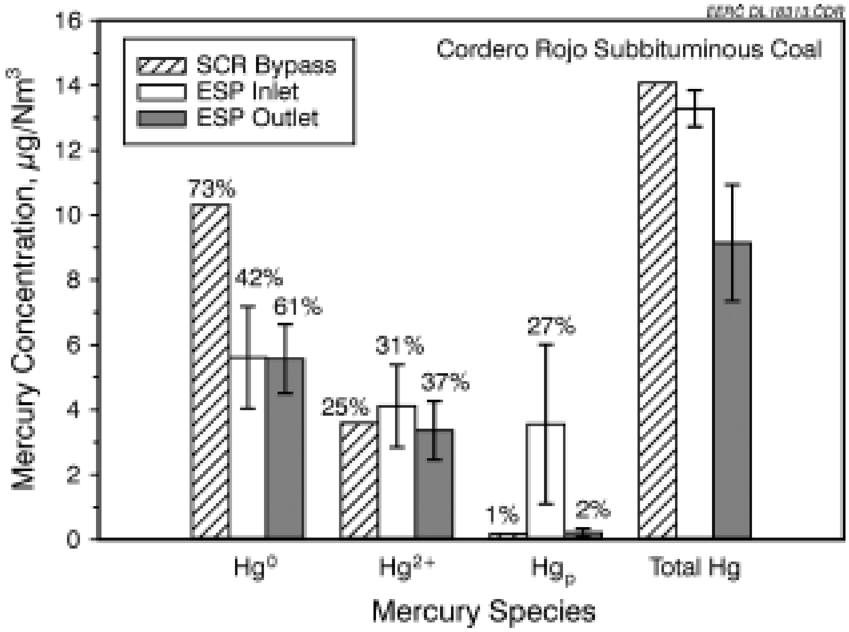

Figure 3-8
Mercury Speciation Results for the Cordero Rojo Baseline Test (The relative proportions of each mercury species are shown by the percentages above the bars.)

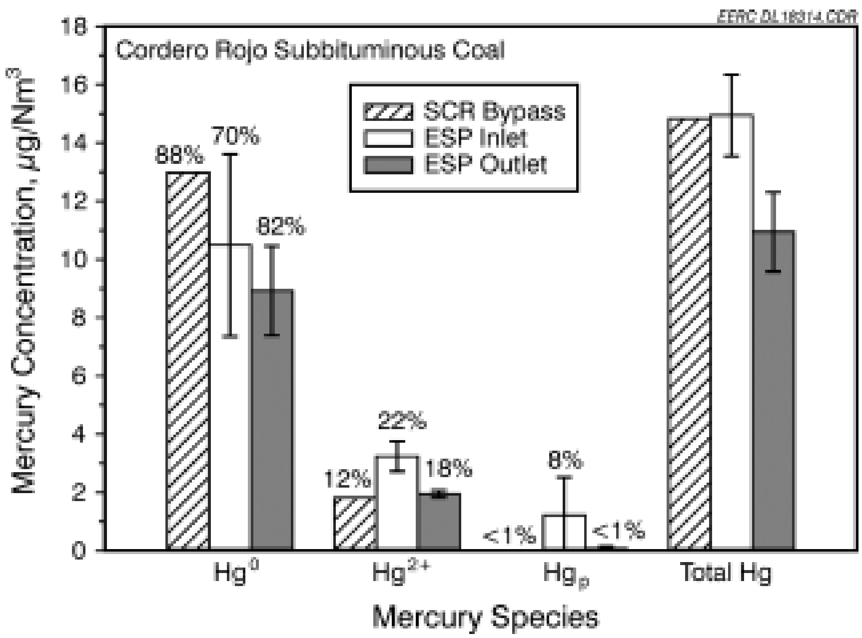

Figure 3-9
Mercury Speciation Results for the Cordero Rojo NH_3 Injection Test (The relative proportions of each mercury species are shown by the percentages above the bars.)

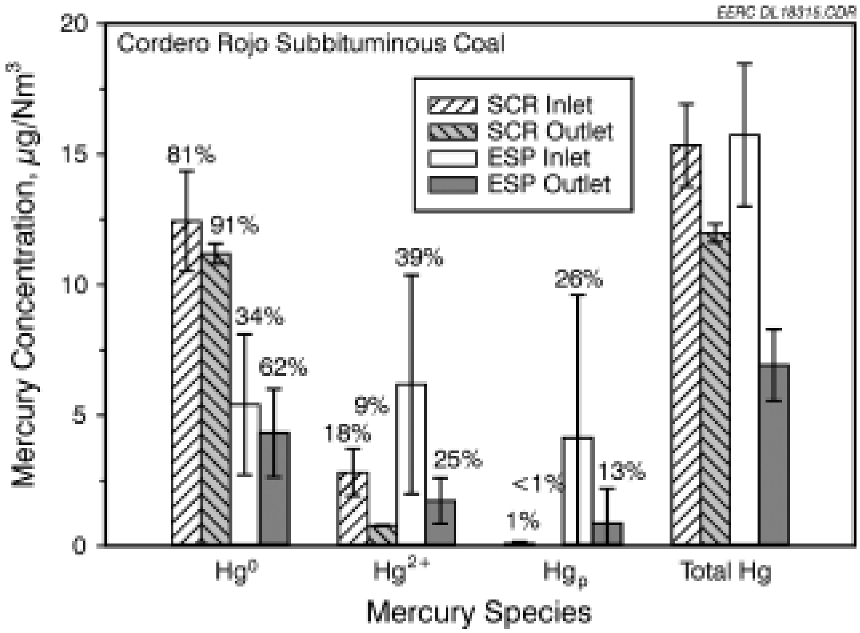

Figure 3-10
Mercury Speciation Results for the Cordero Rojo SCR Test (The relative proportions of each mercury species are shown by the percentages above the bars.)

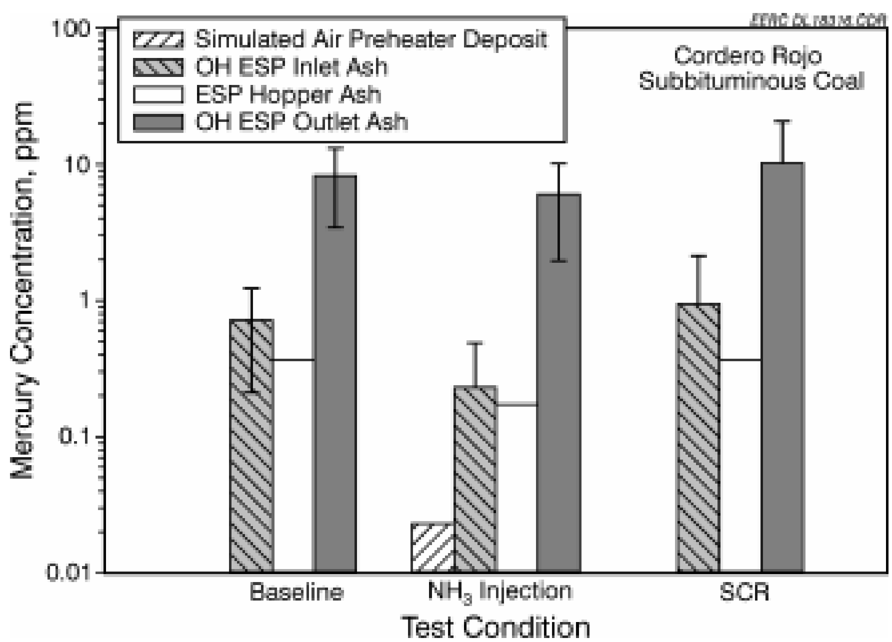

Figure 3-11
Mercury Concentration in Cordero Rojo Ash Samples (The relative proportions of each mercury species are shown by the percentages above the bars.)

Table 3-14
Cordero Rojo ESP Hopper Ash – Major, Minor, and Trace Element Compositions

Element, wt%	Baseline Test	NH_3 Injection Test	SCR Test
SiO_2	32.8	34.3	32.4
Al_2O_3	17.3	17.6	18.4
Fe_2O_3	7.66	7.40	7.01
MnO	0.044	0.042	0.043
TiO_2	1.67	1.71	1.65
BaO	0.77	0.69	0.80
P_2O_5	1.06	1.03	1.16
CaO	28.2	28.2	28.5
MgO	4.45	4.56	4.57
Na_2O	2.41	2.38	2.73
K_2O	0.37	0.37	0.38
SO_3	3.60	3.55	3.91
NH_3	0.002	0.003	0.016
Cl, ppm	<20	<20	<20
Total, wt%	100.3	101.8	101.6

Figure 3-12
Loss on Ignition in the Cordero Rojo ESP Hopper Ashes

Table 3-15
Hg_p Removal Efficiency of the ESP for the Cordero Rojo Coal

Run No.	Hg_p	% Hg_p Removal	% Total Hg Removal	% Fly Ash Removal
610	3.55	94.1	21.2	99.5
611	1.22	94.3	26.8	99.7
612	4.14	79.2	41.5	99.7

3.2.3 Band Mill Bituminous (Appalachian Basin) Coal

3.2.3.1 Flue Gas Compositions

The average Band Mill coal combustion flue gas compositions are presented in Table 3-16. The Band Mill flue gas was characterized by relatively low concentrations of SO_2, SO_3, and Cl and low total Hg. SO_2 and Cl concentrations are higher than those obtained firing the Cordero Rojo subbituminous coal but much lower than those obtained burning either the Paradise or Blacksville coal. The SCR reactor reduced NO_x concentrations in the Band Mill flue gas on average by 99%. As was the case for the Cordero Rojo tests, NH_3 concentrations were greater than expected because the NH_3 injection system was improperly calibrated. NH_3 analysis results for the Band Mill tests are presented in Figure 3-13. NH_3 mass balances were acceptable. The ratio of gas-phase NH_3 to particle-associated NH_3 was about 1 for the NH_3 injection test and more than an order of magnitude higher for the SCR test as a result of excess NH_3 slip. It should be noted that the particle-bound NH_3 concentration did not change.

3.2.3.2 Mercury Speciation

Average mercury speciation and mass balance analysis results for the Band Mill coal tests are provided in Tables 3-17–3-19 and presented graphically in Figures 3-14–3-16. Total mercury concentrations were very low (<3 $\mu g/Nm^3$) for these tests; therefore, mercury speciation results were generally more variable compared to those obtained for firing the other three coals. For example, results obtained at the SCR bypass and SCR inlet sampling locations are not used for constructing Figures 3-14–3-16 because of a lack in agreement among the triplicate mercury speciation measurements. These results, however, are shown in Table 3-19. These samples were taken during three different days during the week of testing. Although this was the case for all of the coals tested, the Band Mill appeared to have the greatest variation. Total mercury mass balances were acceptable at the SCR outlet and ESP inlet locations; however, mass balances were somewhat low (≈60%) at the ESP outlet for the NH_3 injection and SCR tests. The baseline Band Mill flue gas (Figure 3-14) consists of about 80% Hg^{2+}, 15% Hg^0, and 5% Hg_p. Mercury speciation results in Figure 3-15 indicate that the injection of 10 ppmv NH_3 converted Hg^{2+} to Hg_p, thus greatly improving the total mercury removal efficiency of the ESP. For the SCR tests (Figure 3-16), the concentration of Hg_p is even higher relative to only injecting NH_3.

Table 3-16
Average Band Mill Bituminous Coal (Appalachian Basin) Combustion Flue Gas Compositions

	Baseline Test		NH_3 Injection Test		SCR Test	
	Average	Std. Dev.[a]	Average	Std. Dev.	Average	Std. Dev.
SO_2, ppmv[b]	451	41	429	28	439	33
SO_2, ppmv[c]	436	43	413	26	381	48
SO_3, ppmv[c]	<0.03	NA[d]	<0.03	NA	<0.03	NA
CO, ppmv[b]	14.8	6.4	16.3	5.4	17.7	6.6
CO_2, mol%[b]	13.9	1.1	14.2	1.0	14.2	0.8
O_2, mol%[b]	4.54	1.19	4.51	0.86	4.49	0.65
O_2, mol%[c]	5.20	1.30	5.15	1.08	4.99	0.91
NO_x, ppmv[b]	694	67	725	47	749	43
NO_x, ppmv[c]	689	70	722	50	8.8	7.0
NH_3, ppmv[c]	0.43	NA	4.67	0.49	189	66
Cl, ppmv[c]	3.7	NA	2.4	NA	3.4	NA

[a] Population or sample standard deviation.
[b] Measured at combustor outlet.
[c] Measured at ESP inlet.
[d] Not applicable because the analyte concentration was less than the lower limit of quantitation or only one or two measurements were performed.

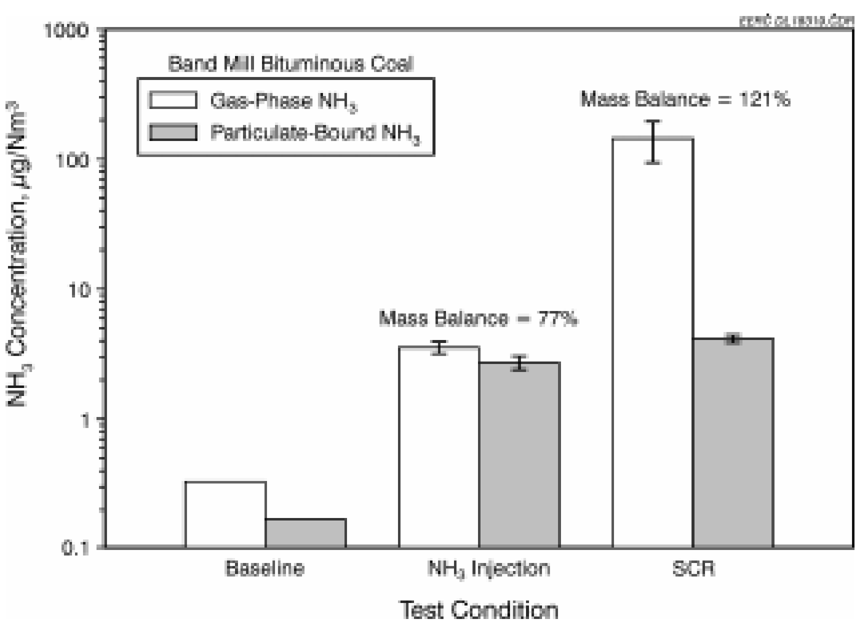

Figure 3-13
Ammonia Concentrations and Mass Balances for the Band Mill Tests

Table 3-17
Mercury Speciation Results for the Band Mill Baseline (Test 613) Test, on a dry, 3% O_2 basis

Sample Location	Hg^0, µg/Nm^3	Hg^{2+}, µg/Nm^3	Hg_p, µg/Nm^3	Total Hg, µg/Nm^3	Mass Balance, %
SCR Bypass	1.46	3.36	<0.01	4.82	192
ESP Inlet	0.28	2.32	0.23	2.83	113
ESP Inlet	0.36	2.11	0.22	2.69	107
ESP Inlet	0.42	1.72	0.13	2.27	91
Average	0.35	2.05	0.19	2.60	104
Std. Dev.	0.07	0.30	0.06	0.29	12
ESP Outlet	0.93[a]	5.65[a]	0.02	6.60[a]	277[a]
ESP Outlet	0.51	2.28	0.02	2.81	125
ESP Outlet	0.47	1.84	0.02	2.33	106
Average	0.49	2.06	0.02	2.57	116
Std. Dev.	NA[b]	NA	0.00	NA	NA

Table 3-18
Mercury Speciation Results for the Band Mill NH_3 Injection (Test 614) Test, on a dry, 3% O_2 basis

Sample Location	Hg^0, µg/Nm^3	Hg^{2+}, µg/Nm^3	Hg_p, µg/Nm^3	Total Hg, µg/Nm^3	Mass Balance, %
SCR Bypass	0.25	0.44	0.04	0.73	30
ESP Inlet	0.32	0.31	1.49	2.12	88
ESP Inlet	0.02[a]	0.54[a]	0.57[a]	1.13[a]	47[a]
ESP Inlet	0.35	0.22	1.23	1.80	75
Average	0.23	0.36	1.10	1.68	81
Std. Dev.	0.18	0.17	0.47	0.51	NA
ESP Outlet	0.22	0.19	0.21	0.62	57
ESP Outlet	0.25	0.21	0.43	0.89	68
ESP Outlet	0.15	0.05	0.42	0.62	57
Average	0.21	0.15	0.35	0.71	61
Std. Dev.	0.05	0.09	0.12	0.26	6

[a]Value was not considered in calculating an average or in constructing the bar plot associated with these results.

Table 3-19
Mercury Speciation Results for the Band Mill SCR (Test 615) Test, on a dry, 3% O_2 basis

Sample Location	Hg^0, µg/Nm^3	Hg^{2+}, µg/Nm^3	Hg_p, µg/Nm^3	Total Hg, µg/Nm^3	Mass Balance, %
SCR Bypass (Test 613)	1.46	3.36	<0.01	4.82	292
SCR Bypass (Test 614)	0.25	0.44	0.04	0.73	30
SCR Inlet (Test 615)	0.75	2.17	0.03	2.95	124
Average	0.82	1.99	0.03	2.83	116
Std. Dev.	0.61	1.47	NA[a]	2.05	81
SCR Outlet	1.72	0.24	<0.01	1.96	82
SCR Outlet	1.53	0.17	0.02	1.72	72
SCR Outlet	1.67	0.25	<0.01	1.92	81
Average	1.64	0.22	0.01	1.87	79
Std. Dev.	0.10	0.04	NA	0.13	5
ESP Inlet	0.07	0.34	1.33	1.74	73
ESP Inlet	0.07	0.07	1.95	2.09	88
ESP Inlet	0.08	0.14	1.65	1.87	79
Average	0.07	0.18	1.64	1.90	80
Std. Dev.	0.01	0.14	0.31	0.18	7
ESP Outlet	0.17	0.15	0.18	0.50	60
ESP Outlet	0.06	0.02	0.35	0.43	57
ESP Outlet	0.04	0.04	0.40	0.48	59
Average	0.09	0.07	0.31	0.47	59
Std. Dev.	0.07	0.07	0.12	0.04	2

[a] Not applicable.
[b] Not applicable because <3 analyte concentrations were greater than the lower limit of quantitation.

Mercury speciation results at the SCR outlet in Figure 3-16 suggest that only about 10% of the Hg^0 released firing Band Mill coal is oxidized at temperatures, ≥345°C (≥655°F). The use of the SCR to reduce NO_x appeared to promote the formation of Hg_p between the SCR and the ESP inlet sampling locations compared to the baseline condition. However, the NH_3 slip was much higher (189 ppm) than would be expected in a full-scale system.

Figure 3-14
Mercury Speciation Results for the Band Mill Baseline Test (The relative proportions of each mercury species are shown by the percentages above the bars.)

Figure 3-15
Mercury Speciation Results for the Band Mill NH_3 Injection Test (The relative proportions of each mercury species are shown by the percentages above the bars.)

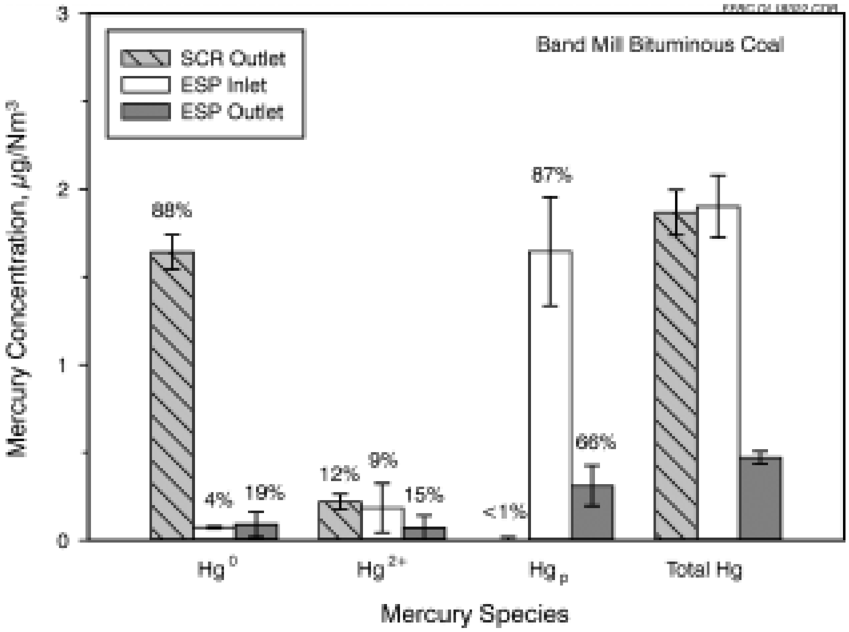

Figure 3-16
Mercury Speciation Results for the Band Mill SCR Test (The relative proportions of each mercury species are shown by the percentages above the bars.)

3.2.3.3 Fly Ash Chemical Compositions

The Hg_p concentrations in Band Mill ash samples are compared in Figure 3-17. Interestingly, for the baseline condition, the mercury concentration in the simulated air preheater deposit is essentially the same as the ESP hopper ash and filter ash samples. However, the deposits collected from the simulated air preheater tube during the NH_3 injection and SCR tests have a very low mercury concentration, $\leq$0.02 ppm. The Hg_p concentration in the ESP hopper ash and filter ash collected during the two test conditions with NH_3 injection was greater than those collected at the baseline condition. Similar to the Paradise and Cordero Rojo fly ashes, mercury is most concentrated in the OH filter ash samples taken at the ESP outlet.

The chemical compositions of Band Mill ESP hopper ash samples are compared in Table 3-20. Fly ashes produced during the NH_3 injection and SCR tests contain greater CaO, SO_3, and NH_3 concentrations relative to the baseline Band Mill fly ash. Similar enrichments were observed in the Paradise fly ashes. It would be expected that the SO_3 would increase due to SO_2 oxidation by the SCR. Also, an increase in NH_3 concentration in the ash is expected because of the addition of NH_3 to the system. However, the increase in CaO is more than likely not real. The increase is small and may be just the variability in the coal. Also, there does not appear to be a subsequent decrease in the percentage of any other ash component. The total mass balance simply increases. Carbon and LOI analysis results for the Band Mill fly ashes are presented in Figure 3-18. LOI/carbon is much greater for the fly ashes produced for the two tests with NH_3 injection, indicating that these ashes contain greater concentrations of volatile noncarbon components relative to the baseline fly ash.

Table 3-20
Band Mill ESP Hopper Ash – Major, Minor, and Trace Element Compositions

Element, wt%	Baseline Test	NH_3 Injection Test	SCR Test
SiO_2	49.3	47.9	49.0
Al_2O_3	24.7	25.8	26.3
Fe_2O_3	6.51	6.94	7.03
MnO	0.056	0.058	0.058
TiO_2	1.28	1.43	1.47
BaO	0.12	0.14	0.14
P_2O_5	0.14	0.18	0.19
CaO	0.78	0.95	1.24
SrO	0.11	0.14	0.15
MgO	1.68	1.62	1.63
Na_2O	0.53	0.58	0.59
K_2O	2.72	2.49	2.48
SO_3	0.37	1.03	1.12
NH_3	0.002	0.150	0.380
Cl, ppm	<20	<20	<20
Total, wt%	88.3	89.4	91.8

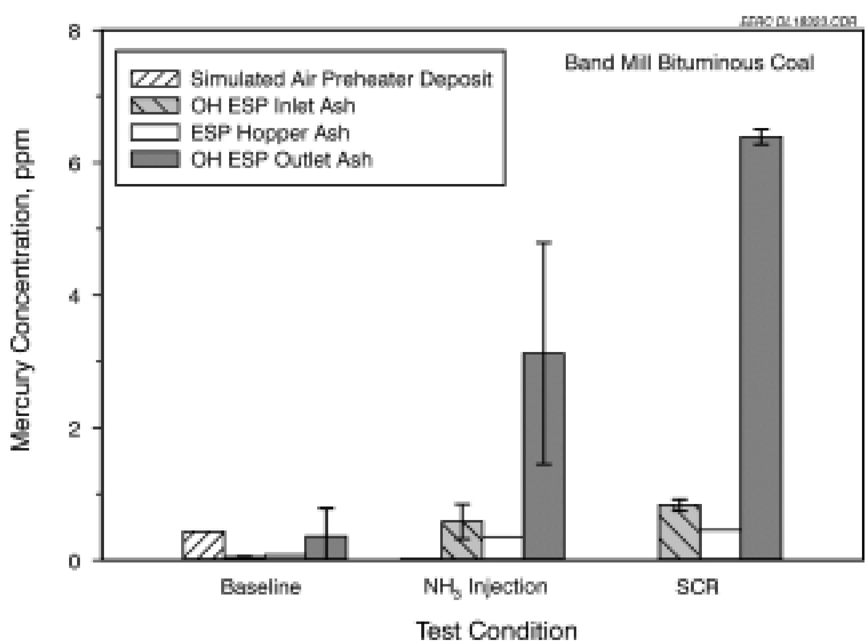

Figure 3-17
Mercury Concentration in the Band Mill Ash Samples

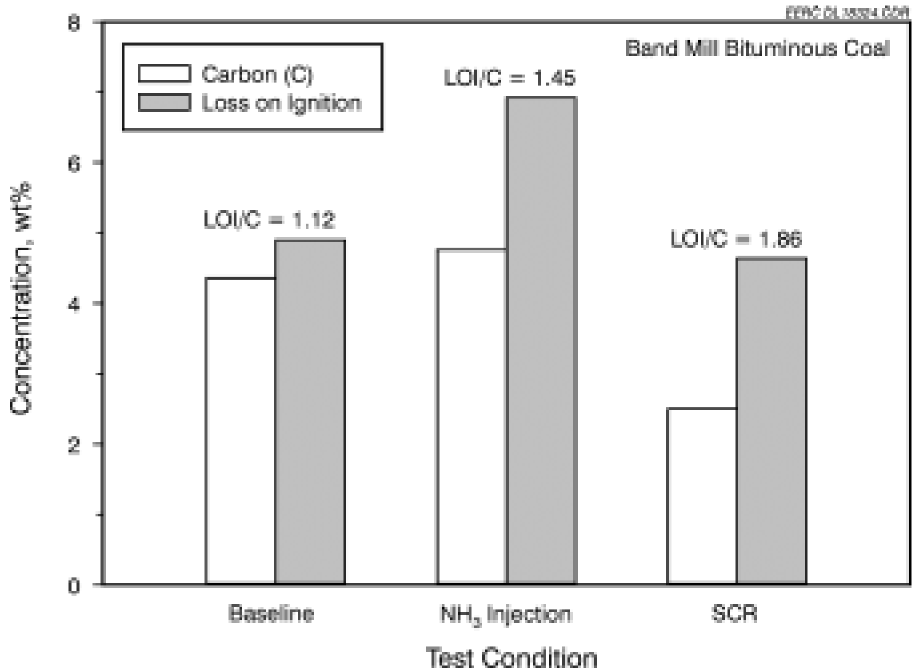

Figure 3-18
Loss on Ignition and Carbon in Band Mill ESP Hopper Ashes

The Band Mill deposits from the simulated air preheater tubes were not chemically analyzed in detail because of a lack of ash deposition on the simulated air preheater tube. Enough sample was obtained, however, for NH_3 analyses. Deposits produced during baseline, NH_3 injection, and SCR conditions contained 0.068, 0.692, and 1.02 wt% NH_3, respectively. NH_3 was much more concentrated in these deposits relative to the fly ashes, possibly as a result of ammonium bisulfate or sulfate reactions.

3.2.3.4 ESP Fly Ash and Total Mercury Collection Efficiencies

The mercury removal by the ESP is compared to the fly ash removal efficiency in Table 3-21. As can be seen from the table, the fly ash removal efficencies for the ESP were >92% for the Band Mill coal combustion tests. For the baseline test, the ESP was ineffective at removing mercury. However, the NH_3 injection and SCR tests showed greatly improved mercury removal efficiencies. As was discussed earlier, this was primarily due to an increase in Hg_p concentrations. ESP mercury removal efficiencies for the NH_3 injection and SCR tests were very similar, suggesting that the presence of NH_3 rather than the SCR catalyst was more of a factor in promoting Hg_p formation.

Table 3-21
Hg_p Removal Efficiency of the ESP for the Band Mill Coal

Run No.	Hg_p, µg/Nm^3	% Hg_p Removal	% Total Hg Removal	% Fly Ash Removal
613	0.19	89.5	1.2	94.0
614	1.10	68.2	57.7	92.6
615	1.64	81.1	76.1	97.6

3.2.4 Blacksville Bituminous Coal (Appalachian Basin)

3.2.4.1 Flue Gas Composition

As shown in Table 3-22, the Blacksville coal combustion flue gas contained the highest NO_x and Cl concentrations of the four coals tested. SO_2 concentrations are also high in comparison to the Cordero Rojo and Band Mill flue gases, but lower than Paradise flue gas. The SCR test resulted in an average NO_x reduction of 97%. As was the case for the Paradise coal test, there was a

Table 3-22
Average Blacksville Bituminous Coal (Appalachian Basin) Combustion Flue Gas Compositions

	Baseline Test		NH_3 Injection Test		SCR Test	
	Average	Std. Dev.[a]	Average	Std. Dev.	Average	Std. Dev.
SO_2, ppmv[b]	1458	86	1449	74	1459	67
SO_2, ppmv[c]	1392	76	1358	74	1395	50
SO_3, ppmv[c]	2.4	NA[d]	<0.3	NA	1.0	NA
CO, ppmv[b]	19.7	12.2	27.1	10.8	20.6	9.1
CO_2, mol%[b]	14.1	1.1	14.3	0.8	13.8	0.8
O_2, mol%[b]	4.36	0.63	4.38	0.68	4.46	0.56
O_2, mol%[c]	5.31	0.60	5.55	0.85	4.81	0.83
NO_x, ppmv[b]	861	82	913	97	924	40
NO_x, ppmv[c]	764	76	819	96	29	19
NH_3, ppmv[c]	2.18	NA	0.64	0.18	0.40	0.18
Cl, pmv[c]	38.6	NA	25.9	NA	38.2	NA

[a] Population or sample standard deviation.
[b] Measured at combustor outlet.
[c] Measured at ESP inlet (gaseous NH_3 only).
[d] Not applicable because the analyte concentration was less than the lower limit of quantitation or only one or two measurements were performed.

measurable amount of NH_3 in the baseline flue gas; in fact, the baseline NH_3 concentration was higher than the NH_3 slip measured in the tests when NH_3 was added to the system. This results in poor mass balances for the NH_3 injection tests. As was suggested previously, it is possible that the baseline NH_3 concentration is biased high. Average NH_3 measurement results for the Blacksville flue gases are shown in Figure 3-19. It is clear that most of the unreacted NH_3 in Blacksville flue gas was adsorbed on ash particles, most likely as bisulfates or sulfates.

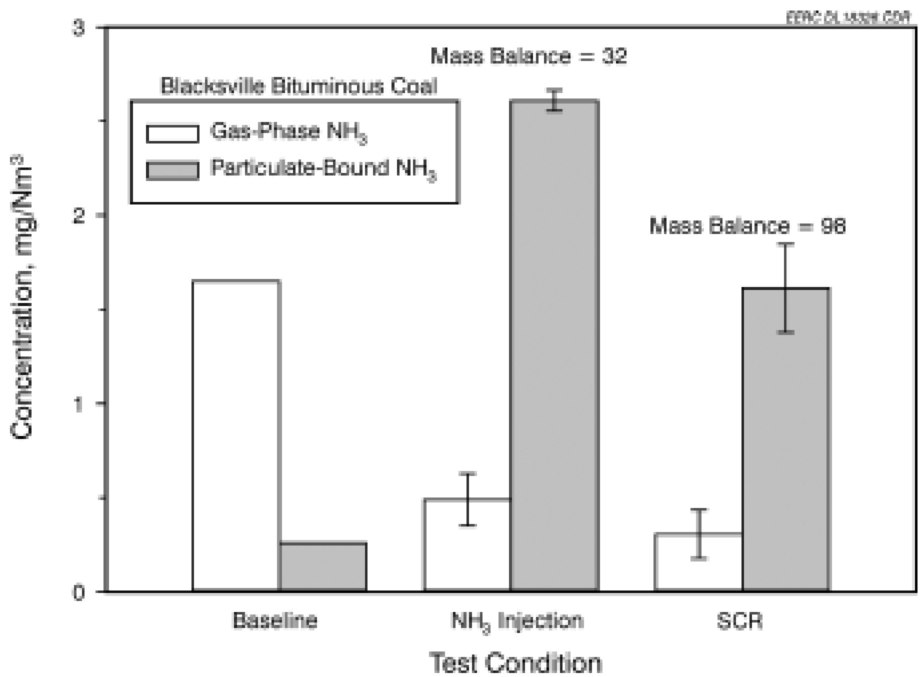

Figure 3-19
Ammonia Concentrations and Mass Balances for the Blacksville Tests

3.2.4.2 Mercury Speciation

Mercury speciation results for the Blacksville coal tests are compared in Tables 3-23–3-25 and shown graphically in Figures 3-20–3-22. Average mass balances for the Blacksville flue gases are acceptable. As shown in Figure 3-20, mercury speciation of the baseline tests was characterized by very high Hg^{2+} concentrations, accounting for ≥85% of the total mercury. Measurements at the SCR bypass location indicate that most of the conversion of Hg^0 to Hg^{2+} occurs at ≥335°C (≥635°F). The ESP was relatively ineffective in removing Hg^{2+} from the baseline Blacksville flue gas. Mercury speciation results in Figure 3-21 suggest that the injection of 10 ppmv NH_3 into Blacksville flue gas promoted the conversion of Hg^{2+} to Hg_p to a small extent. Mercury speciation at the ESP outlet, however, did not change significantly as a result of NH_3 injection. A comparison of Figures 3-20 and 3-22 suggests that passing the flue gas through the SCR reactor did little to increase the Hg_p concentration.

Table 3-23
Mercury Speciation Results for the Blacksville Baseline (Test 616) Test, on a dry, 3% O_2 basis

Sample Location	Hg^0, μg/Nm^3	Hg^{2+}, μg/Nm^3	Hg_p, μg/Nm^3	Total Hg, μg/Nm^3	Mass Balance, %
SCR Bypass	1.76	8.02	0.38	10.2	94
ESP Inlet	1.31	8.55	0.21	10.1	93
ESP Inlet	0.50	8.78	0.28	9.56	88
ESP Inlet	0.60	8.16	0.66	9.42	87
Average	0.80	8.50	0.38	9.68	90
Std. Dev.	0.44	0.31	0.24	0.34	3
ESP Outlet	1.75	7.44	0.22	9.41	96
ESP Outlet	1.18	7.43	0.03	8.64	89
ESP Outlet	0.91	6.97	0.20	8.08	84
Average	1.28	7.28	0.15	8.71	90
Std. Dev.	0.43	0.27	0.10	0.67	6

For the Blacksville tests, two continuous mercury analyzers were used at the outlet of the ESP and compared to the OH method. The results are shown in Figures 3-23–3-25. Total mercury concentrations measured with the PS Analytical instrument compared very well to the corresponding OH measurements. Conversely, those measured with the Semtech analyzer were generally biased low relative to the OH total mercury measurements. The Hg^0 measurements with the PS Analytical instrument were lower than those measured using the OH method and also measured by the Semtech. In general, the mercury analyzers gave results that were similar to those obtained with the OH method.

3.2.4.3 Fly Ash Chemical Compositions

The Hg_p concentrations in Blacksville fly ash samples are compared in Figure 3-26. Blacksville simulated air preheater deposits are lower in Hg_p relative to fly ashes sampled from the ESP hopper and OH filter samples at the ESP inlet and outlet. Hg_p concentrations in the ESP hopper ashes and on the ash collected on the OH filter at the ESP inlet location are very similar. This indicates that the OH filter was not collecting additional mercury relative to the ESP. However, the Hg_p concentration is higher in the ash on the OH filter used at the ESP outlet. Hg_p is slightly more concentrated in fly ashes produced during the NH_3 injection and SCR tests relative to the baseline test, based on the ESP inlet filter and hopper ash data.

Table 3-24
Mercury Speciation Results for the Blacksville NH_3 Injection (Test 617) Test, on a dry, 3% O_2 basis

Sample Location	Hg^0, µg/Nm^3	Hg^{2+}, µg/Nm^3	Hg_p, µg/Nm^3	Total Hg, µg/Nm^3	Mass Balance, %
SCR Bypass	3.42	4.69	0.90	9.01	86
SCR Bypass	1.90	7.43	0.09	9.42	90
ESP Inlet	2.54	5.48	0.89	8.91	85
ESP Inlet	1.69	4.55	2.00	8.24	79
ESP Inlet	1.34	6.25	0.74	8.33	80
Average	1.86	5.43	1.21	8.49	81
Std. Dev.	0.62	0.85	0.69	0.36	3
ESP Outlet	1.38	5.69	0.07	7.14	79
ESP Outlet	0.83	5.00	0.25	6.08	69
ESP Outlet	1.03	6.09	0.03	7.15	79
Average	1.08	5.59	0.12	6.79	76
Std. Dev.	0.28	0.55	0.12	0.61	6

The chemical compositions of Blacksville fly ashes sampled from the ESP hopper during each of the three different test conditions are presented in Table 3-26. Relative to the baseline condition, SO_3 is slightly more concentrated in the fly ashes produced for the two test conditions with NH_3 injection. NH_3 and Cl concentrations in the fly ash produced during the SCR test are greater than those produced in either the baseline or NH_3 injection test. The chemical compositions of Blacksville ash deposits could not be determined because insufficient amounts of ash were deposited on the simulated air preheater tubes.

LOI and carbon analysis results for the Blacksville ESP hopper ash samples are compared in Figure 3-27. Fly ashes produced during NH_3 injection and SCR conditions had a greater LOI/carbon partly because of their greater SO_3 and NH_3 concentrations (Table 3-22).

Table 3-25
Mercury Speciation Results for Blacksville SCR (Test 618) Test, on a dry, 3% O_2 basis

Sample Location	Hg^0, µg/Nm^3	Hg^{2+}, µg/Nm^3	Hg_p, µg/Nm^3	Total Hg, µg/Nm^3	Mass Balance, %
SCR Bypass (Test 616)	1.76	8.02	0.38	10.2	94
SCR Bypass (Test 617)	3.42[a]	4.69[a]	0.90[a]	9.01[a]	86[a]
SCR Bypass (Test 617)	1.90	7.43	0.09	9.42	90
SCR Inlet (Test 618)	1.84	7.61	0.24	9.69	89
SCR Inlet (Test 618)	1.33	7.86	0.02	9.21	84
Average	1.71	7.73	0.18	9.62	89
Std. Dev.	0.22	0.23	0.14	0.36	4
SCR Outlet	1.12	8.12	0.02	9.26	85
SCR Outlet	0.91	8.42	0.06	9.39	86
SCR Outlet	1.03	8.19	0.17	9.39	86
Average	1.02	8.24	0.08	9.35	86
Std. Dev.	0.11	0.16	0.08	0.08	1
ESP Inlet	0.17	8.18	1.01	9.36	86
ESP Inlet	0.24	8.63	2.11	11.0	101
ESP Inlet	0.19	8.56	0.67	9.42	91
Average	0.20	8.46	1.26	9.92	92
Std. Dev.	0.04	0.24	0.75	0.92	8
ESP Outlet	0.09	7.12	0.35	7.56	80
ESP Outlet	0.24	6.96	0.55	7.75	81
ESP Outlet	<0.01	8.39	0.39	10.2	84
Average	0.11	7.49	0.43	8.51	82
Std. Dev.	0.12	0.78	0.11	1.49	2

[a]Value was not considered in calculating an average or in constructing the bar plot associated with these results.

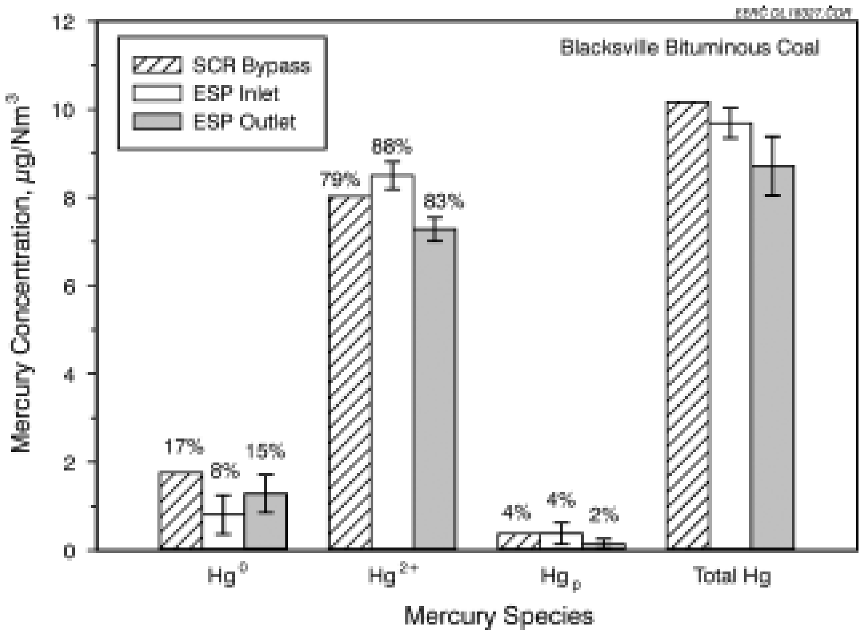

Figure 3-20
Mercury Speciation Results for the Blacksville Baseline Test (The relative proportions of each mercury species are shown by the percentages above the bars.)

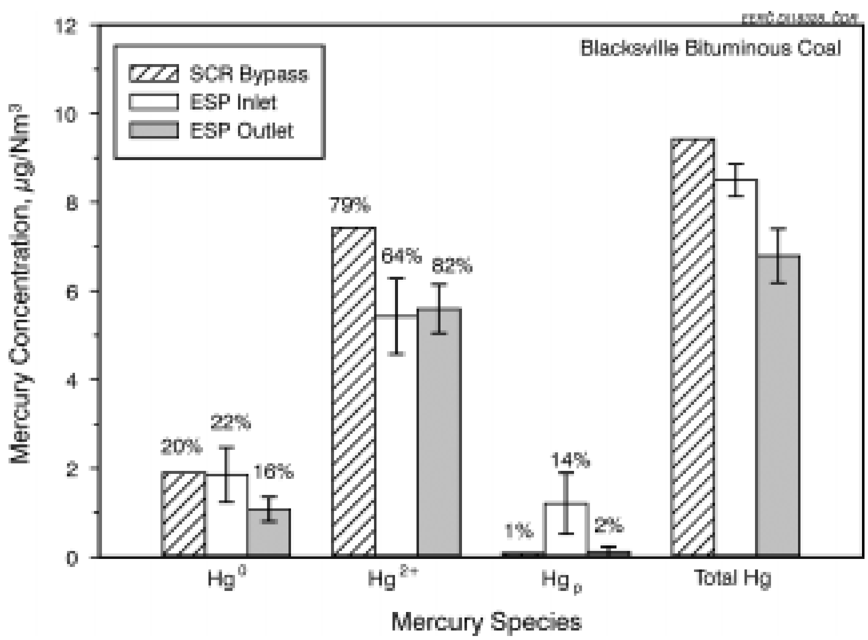

Figure 3-21
Mercury Speciation Results for the Blacksville NH_3 Injection Test (The relative proportions of each mercury species are shown by the percentages above the bars.)

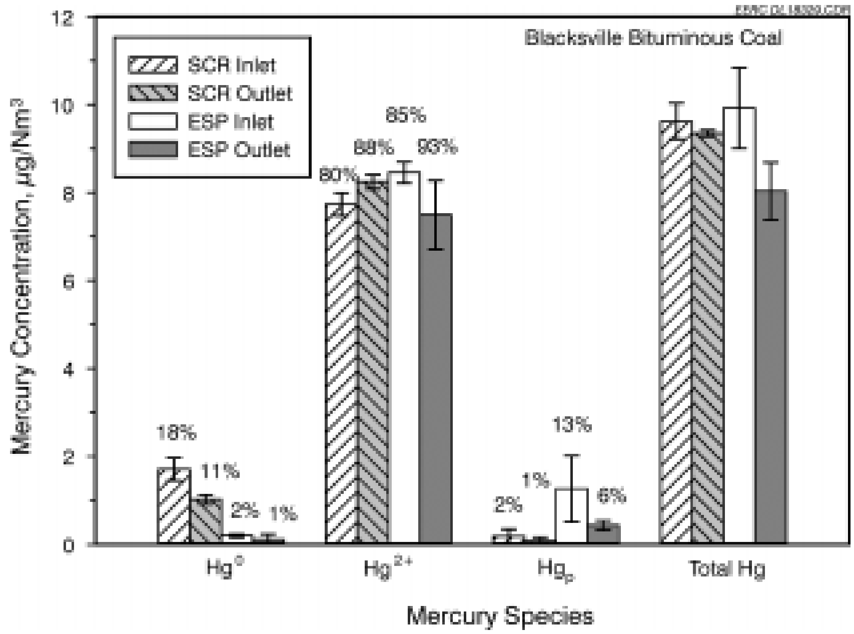

Figure 3-22
Mercury Speciation Results for the Blacksville SCR Test (The relative proportions of each mercury species are shown by the percentages above the bars.)

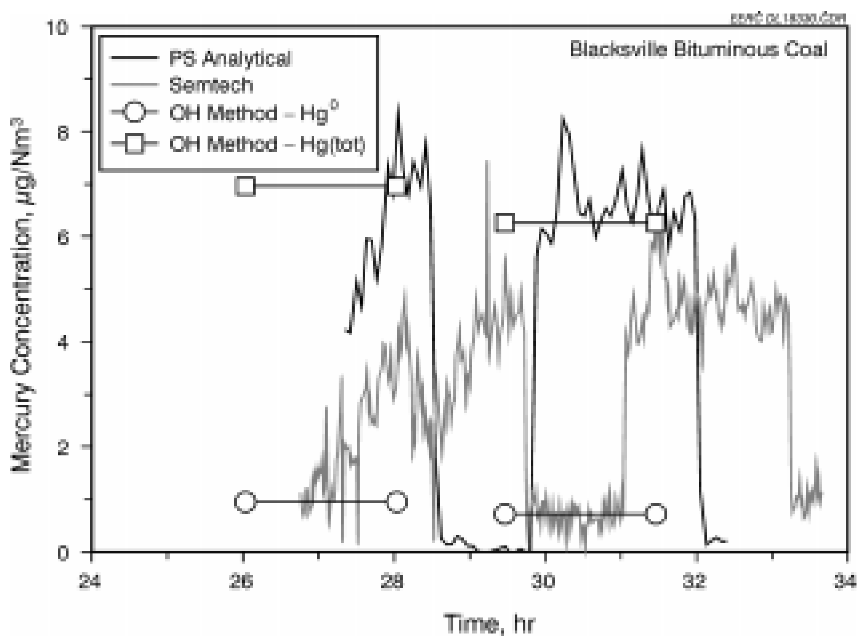

Figure 3-23
Comparison of Two Continuous Mercury Analyzers to the OH Method for the Blacksville Baseline Test

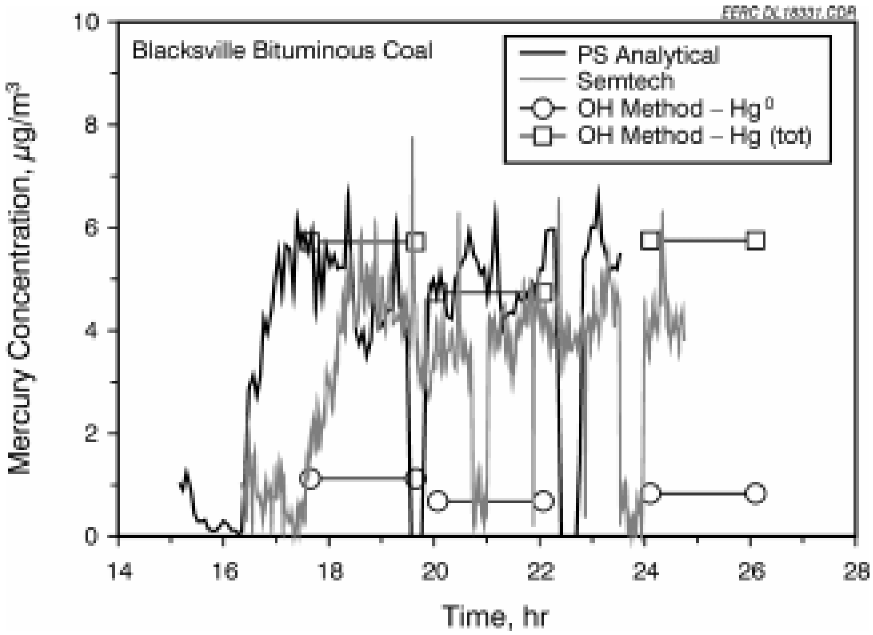

Figure 3-24
Comparison of Two Continuous Mercury Analyzers to the OH Method for the Blacksville NH_3 Injection Test

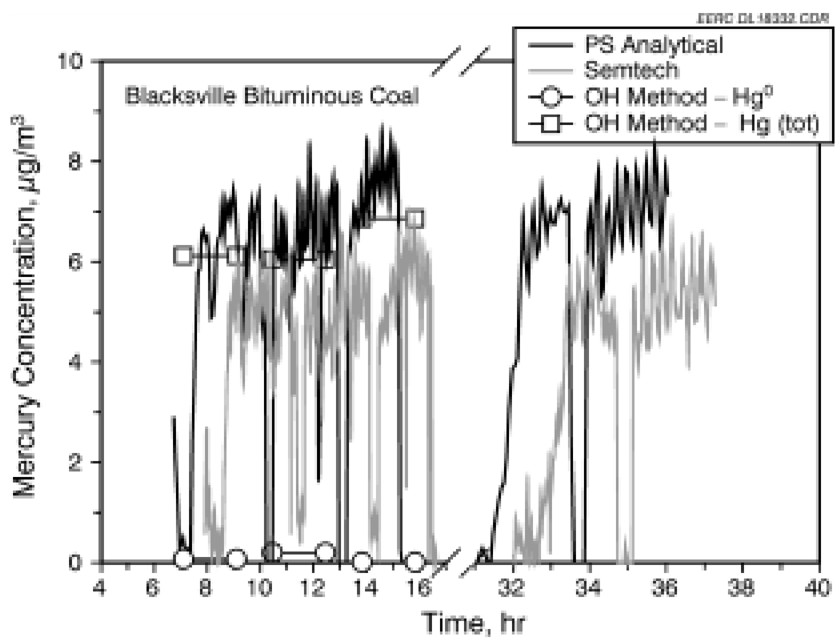

Figure 3-25
Comparison of Two Continuous Mercury Analyzers to the OH Method for the Blacksville SCR Test

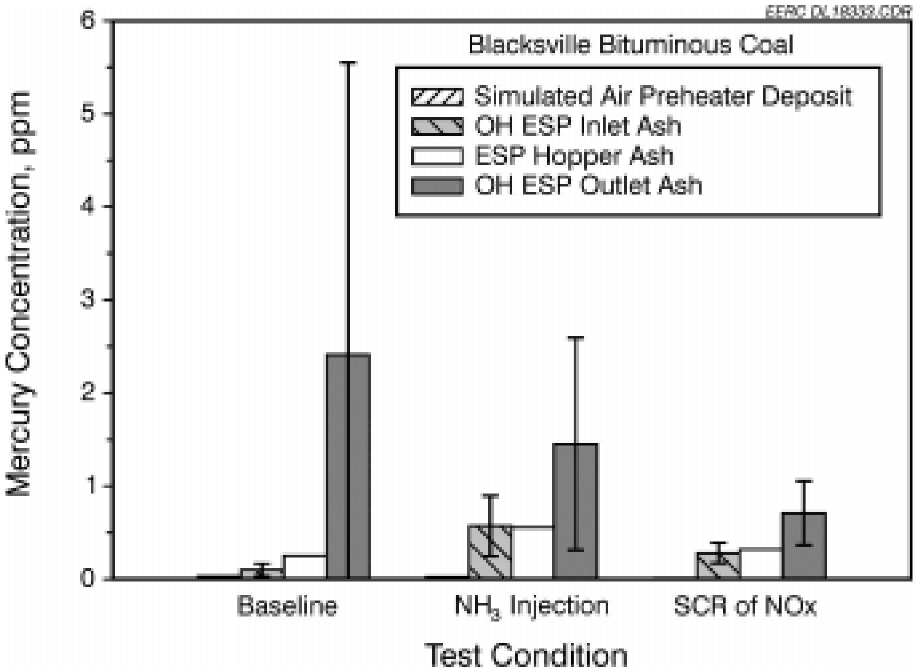

Figure 3-26
Mercury Concentration in the Blacksville Ash Samples

Table 3-26
Blacksville ESP Hopper Ash – Major, Minor, and Trace Element Compositions

Element, wt%	Baseline Test	NH_3 Injection Test	SCR Test
SiO_2	45.1	44.6	44.8
Al_2O_3	23.6	23.2	23.2
Fe_2O_3	17.2	16.7	16.8
MnO	0.029	0.031	0.032
TiO_2	1.08	1.08	1.11
BaO	0.11	0.13	0.12
P_2O_5	0.49	0.54	0.54
CaO	5.38	5.84	5.66
MgO	1.45	1.50	1.52
Na_2O	0.97	0.97	1.02
K_2O	1.80	1.78	1.86
SO_3	2.58	3.31	3.06
NH_3	0.0309	0.00435	0.11
Cl, ppm	71	69	90
Total, wt%	99.8	99.7	99.8

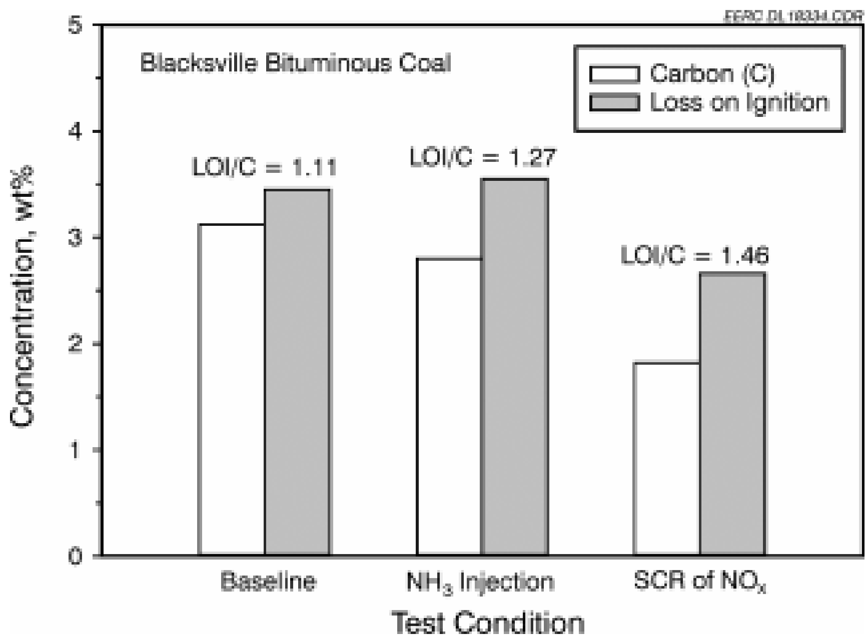

Figure 3-27
Loss on Ignition and Carbon in the Blacksville ESP Hopper Ash

3.2.4.4 ESP Fly Ash and Total Mercury Collection Efficiencies

Table 3-27 presents a comparison of the ESP removal efficiencies for total mercury and fly ash at each of three test conditions conducted firing the Blacksville coal. As can be seen in Table 3-27, there was degradation in the operation of the ESP for the third test as the particulate collection efficiency was much lower. There did not appear to be much, if any, change in the total mercury removal by the ESP for any of the three test conditions.

Table 3-27
Hg_p Removal Efficiency of the ESP for the Blacksville Coal

Run No.	Hg_p, µg/Nm^3	% Hg_p Removal	% Total Hg Removal	% Fly Ash Removal
616	0.38	60.5	10.1	96.9
617	1.21	90.1	20.1	96.4
618	1.26	65.9	22.9	82.6

4 DISCUSSION

4.1 Mercury Speciation

A primary goal of this investigation was to do a screening evaluation at the pilot-scale level to determine the effects on mercury speciation of NH_3 injection and an SCR reactor for the reduction of NO_x. In order to make direct comparisons at each sampling location, as a function of coal type and test condition, the mercury results are presented in Table 4-1 as a percentage change in mercury species relative to the baseline condition ($\triangle Hg^0$, $\triangle Hg^{2+}$, and $\triangle Hg_p$). For example, Hg^{2+} at the ESP inlet for the baseline Paradise test was 90% of the total mercury (Figure 3-2), whereas in the NH_3 injection test, the Hg^{2+} made up only 39% of the total mercury at the ESP inlet (Figure 3-3). Therefore, injecting NH_3 resulted in a 51% decrease in Hg^{2+} (i.e., $\triangle Hg^{2+}$ = –51%) compared to the baseline condition when the Paradise coal was fired. $\triangle Hg^0$, $\triangle Hg^{2+}$, and $\triangle Hg_p$ for each of the tests are presented in Table 4-1. Values of <15% in Table 4-1 are considered insignificant based on variability in the mercury speciation results.

4.1.1 The Effects of NH_3 Injection Without the SCR Catalyst on Mercury Speciation

As shown in Table 4-1, there was an increase in Hg_p and a subsequent decrease in Hg^{2+} as a result of NH_3 injection for the Paradise and Band Mill tests. However, with the Cordero Rojo coal, there appeared to be an increase in Hg^0 as a result of NH_3 injection. However, it is more than likely a result of data variability. For the Blacksville coal test, NH_3 injection showed little, if any, significant change.

4.1.2 The Effects of SCR of NO_x on Mercury Speciation

$\triangle Hg^0$, $\triangle Hg^{2+}$, and $\triangle Hg_p$ in Table 4-1 show that SCR did not significantly affect the mercury speciation of Cordero Rojo and Blacksville flue gases. However, for the Paradise SCR test, a significant and quantifiable amount of Hg^0 was converted to Hg^{2+} between the SCR inlet and SCR outlet sampling locations. There was also increased conversion of Hg^{2+} to Hg_p for the Paradise and Band Mill fuels for the SCR test compared to the NH_3 injection test.

4.2 Fly Ash Chemical Compositions

As was discussed in the previous sections, Paradise, Cordero Rojo, Band Mill, and Blacksville fly ashes produced during the three different test conditions and captured in the ESP were chemically analyzed to evaluate the effects of NH_3 injection and SCR of NO_x on ash composition including the concentration of Hg_p. The results are shown in Table 4-2. As can be

Table 4-1
Percent Change in Mercury Species Proportions Relative to the Baseline Tests

Coal:	**Paradise**[a]			**Cordero Rojo**			**Band Mill**			**Blacksville**		
Sampling Location:	**SCR Out**	**ESP In**	**ESP Out**	**SCR Out**	**ESP In**	**ESP Out**	**SCR Out**	**ESP In**	**ESP Out**	**SCR Out**	**ESP In**	**ESP Out**
NH_3 Injection Only												
ΔHg^0	NA[b]	9.6	0.6	NA	27.9	20.6	NA	0.1	10.5	NA	13.6	1.2
ΔHg^{2+}	NA	−51.0	−51.3	NA	−9.3	−19.0	NA	−57.8	−59.0	NA	−23.9	−1.3
ΔHg_p	NA	41.4	50.7	NA	−18.6	−1.7	NA	57.8	48.5	NA	10.3	0.1
SCR Tests[c]												
ΔHg^0	−30.3	−4.3	−7.5	12.3	−7.9	1.6	ND[d]	−9.8	0.1	-6.9	−6.2	−13.3
ΔHg^{2+}	27.2	−78.3	−72.7	−11.9	8.3	−11.7	ND	−69.6	−65.3	7.9	−2.5	9.7
ΔHg_p	3.2	82.6	80.2	−0.5	−0.4	10.1	ND	79.4	65.2	−1.0	8.8	3.6

[a] For the Paradise NH_3 injection test, 25 ppm was added, compared to 10 ppm for the remainder of the tests.
[b] Not applicable.
[c] For the Cordero Rojo and Band Mill tests, the NH_3/NO_x was ~1.2, compared to ~1.0 for Paradise and Blacksville tests.
[d] Not determined because of a lack in repeatability among the three SCR bypass/inlet mercury speciation measurements.

Table 4-2
Percent Change in ESP Hopper Ash Hg_p Concentrations Relative to Baseline Ashes

	NH_3 Injection Test	SCR Test
Paradise	232	286
Cordero Rojo	−53	−0.3
Band Mill	330	456
Blacksville	120	27

seen, there is a substantial increase in Hg_p for two of the bituminous coals. The unburned carbon particles in fly ash are potential mercury sorbents [27–30]. The carbon and LOI contents of fly ashes measured in the ESP hopper ashes, however, do not strongly correlate to Hg_p concentrations as shown in Figures 4-1 and 4-2. The R^2 values for carbon and LOI as a function of Hg_p are 0.211 and 0.456, respectively. The lack of a strong correlation in Figures 4-1 and 4-2 suggests that fly ash components other than carbon are more important in promoting the formation of Hg_p. These may include the NH_3 concentration and the reactivity of the carbon.

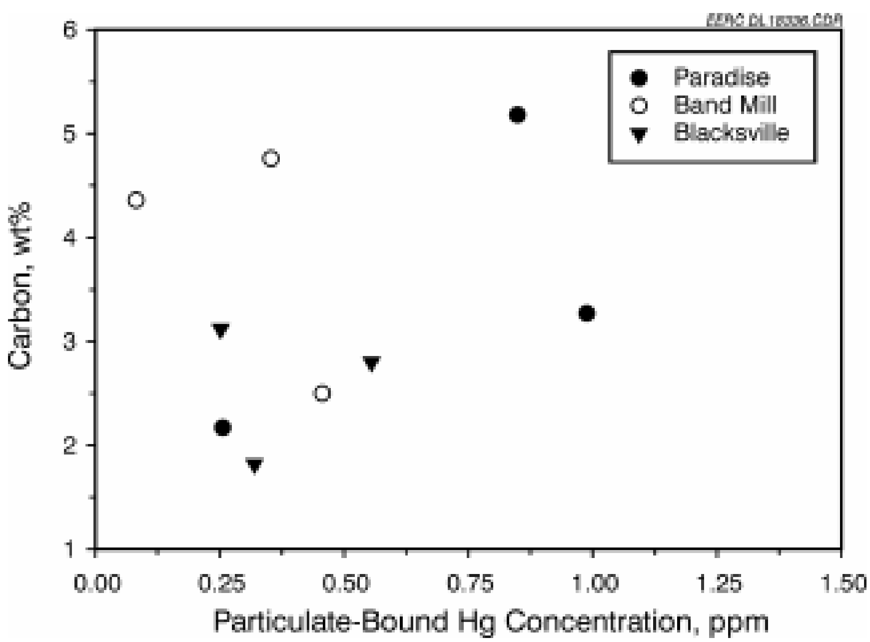

Figure 4-1
Mercury Concentration in the ESP Hopper Ashes as a Function of Carbon Content

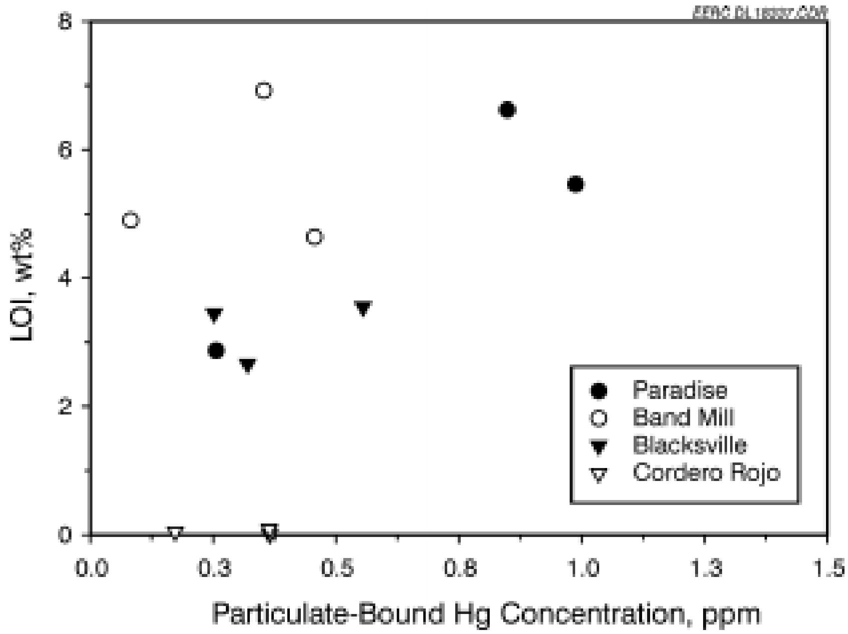

Figure 4-2 Mercury Concentration in the ESP Hopper Ashes as a Function of Loss on Ignition

Figures 4-3–4-6 show an increase in the NH_3 and Hg_p concentrations in Paradise and Band Mill ESP hopper ashes as a result of adding NH_3 (both the NH_3 injection only and SCR test conditions), but not in the Cordero Rojo tests. Chloride concentrations in Paradise fly ashes (Figure 4-3) also increased. As shown in Table 4-2, the Hg_p concentrations in the Paradise and Band Mill ESP hopper ashes increased significantly (230%–460%). The concentration of Hg_p in the Blacksville ash increased as a result of injecting NH_3 into flue gas, but not when the flue gas was passed through the SCR. In all of the tests, the NH_3 concentration in the flue gas was somewhat different. This may have had an impact on the results. However, it is clear that increasing the Hg_p concentration is advantageous. The collection efficiency of the ESP for Hg_p is directly proportional to the ESP collection efficiency, as shown by Figure 4-7.

In addition to sulfur, the positive correlations between NH_3 and Hg_p concentrations in Figure 4-8 imply that the formation of N-rich particles or sorption of N species on the Paradise and Band Mill fly ashes promoted Hg_p formation. A similar relationship between chlorides and Hg_p concentrations in Figure 4-8 for Paradise ashes suggests that Cl was involved in enhancing Hg_p formation. However, Cl does not positively correlate to Hg_p in the Blacksville ashes. When Hg_p concentration is compared as a function of Cl concentration (Figure 4-9), there is a positive correlation of $R^2 = 0.331$.

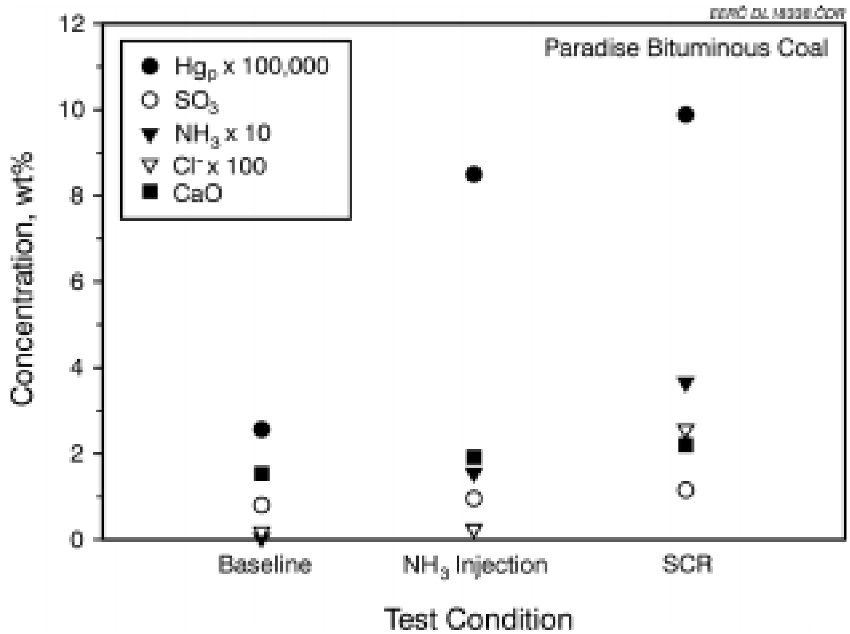

Figure 4-3
The Concentration of Hg_p, CaO, SO_3, NH_3, and Cl in the ESP Hopper Ash for the Paradise Tests

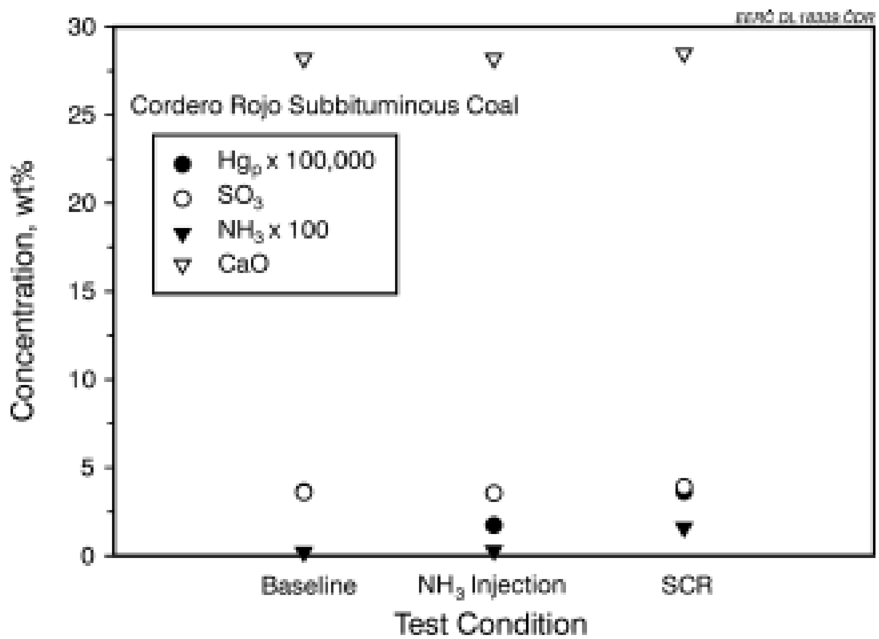

Figure 4-4
The Concentration of Hg_p, CaO, SO_3, and NH_3 in the ESP Hopper Ash for the Cordero Rojo Tests

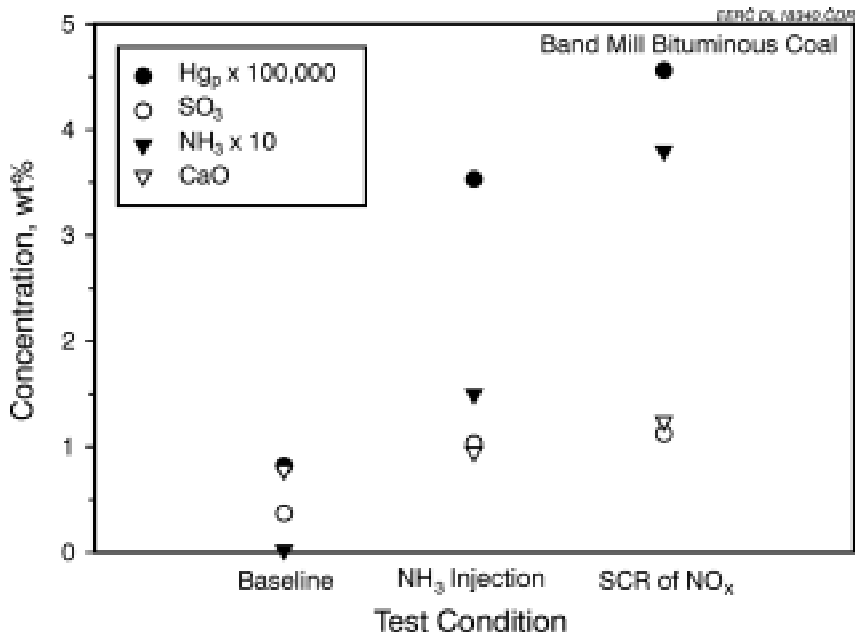

Figure 4-5
The Concentration of Hg_p, CaO, SO_3, and NH_3 in the ESP Hopper Ash for the Band Mill Tests

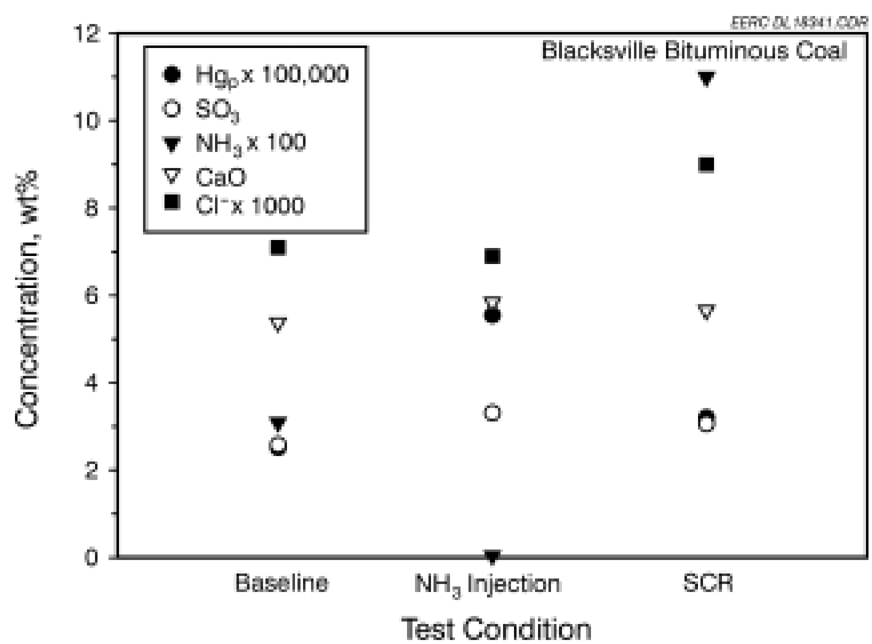

Figure 4-6
The Concentration of Hg_p, CaO, SO_3, NH_3, and Cl in the ESP Hopper Ash for the Blacksville Tests

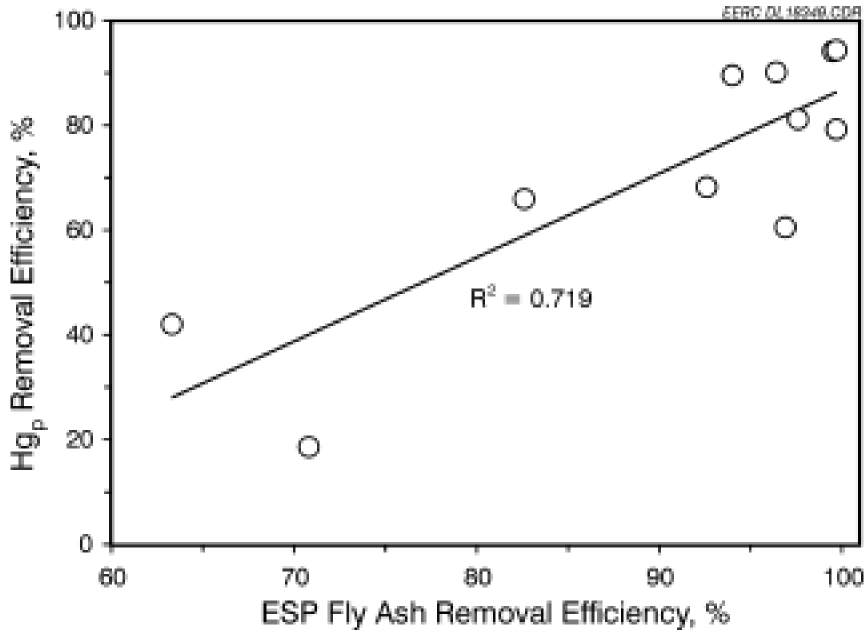

Figure 4-7
Hg_p Removal Efficiency as a Function of ESP Collection Efficiency

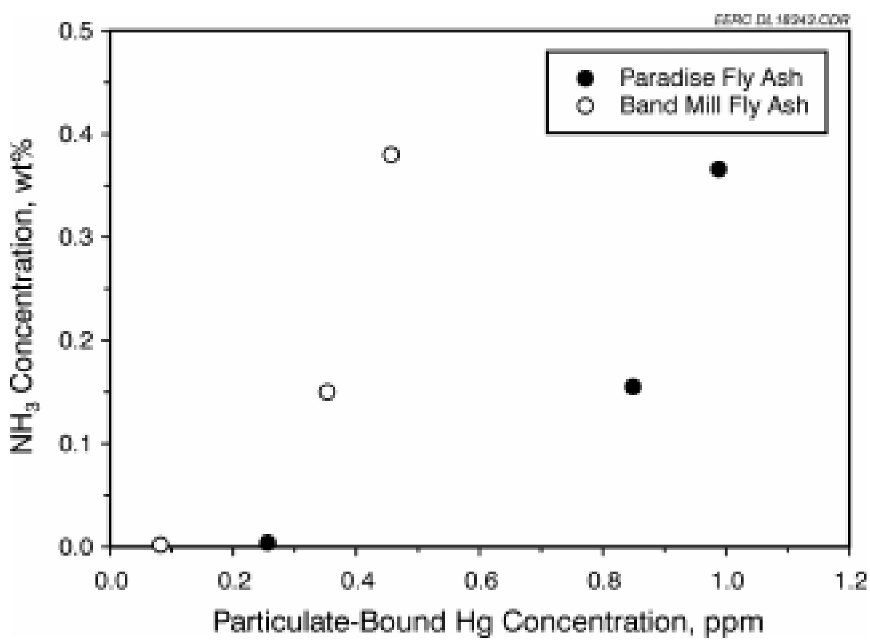

Figure 4-8
Mercury Concentration in the Paradise and Band Mill ESP Hopper Ashes as a Function of NH_3 Concentration

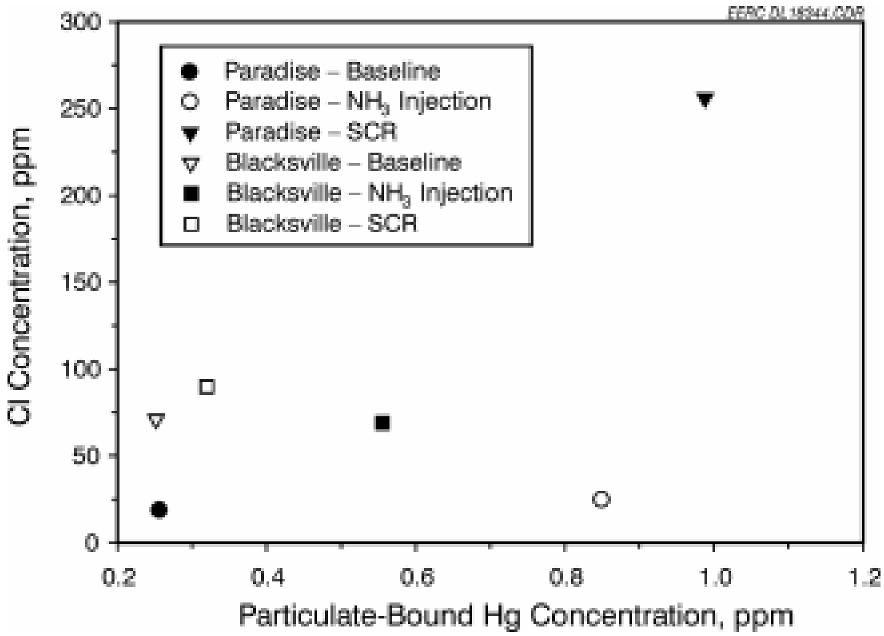

Figure 4-9
Mercury Concentration in the Paradise and Blacksville ESP Hopper Ashes as a Function of Cl Concentration

4.3 Linear Regression Analyses of the Results

Linear regression analyses were performed on mercury speciation results in reference to the chemical composition of the four test coals (Hg, S, Cl, and Ca concentrations in parts per million on a moisture-free basis). Selected regression correlations are shown in Appendix D. Appendix D tabulates three statistical parameters which are referred to in the discussion below: 1) a value of R^2 for a specified number of data points (N), 2) the positive or negative "x coefficient" which indicates proportional dependence on the independent regression variable, and 3) the probability (P) that the x coefficient is significantly different from zero. The only independent variables presented are those with the highest statistical significance for a given set of data points. Some of the variables discussed, particularly where N is small, still have what would be considered to be a low statistical significance as indicated by their P values, where a P value of 0.95 is often used to identify a statistically significant variable. For most of the regression data sets, N has a value of 12 (4 coals $\times$ 3 test conditions). However, the regression data sets for the Hg^0 and Hg^{2+} species leaving the furnace and entering the SCR are a special case and have an N of only 10 because the three measured values of these species for Band Mill coal were averaged to eliminate a wide divergence at essentially the same furnace conditions. N has a value of 4 for those data sets that represent SCR operation. Because of low values of N and scatter in the data, the regression equations presented in Appendix D should not be interpreted as reliable predictors of speciation but only as general indications of trends. Also, it should be noted that several factors concerned with the overall data set limit the validity of the regressions for full-scale plants. These factors include possible differences in mercury speciation as a function of the much smaller size of the pilot-scale combustor, the limited number and range of coal types, and differences in ammonia slip concentrations. These will be discussed in the recommendation section of this report.

The following discussion of regression equations relates to the order of the four tables in Appendix D, which respectively deal with 1) mercury speciation leaving the furnace and entering the SCR, 2) changes in mercury speciation across the SCR, 3) changes in speciation across the cooling loop, and 4) the percentages of coal mercury emitted as different species at the exit of the ESP. The discussion will identify the positive or negative effects of the coal and operating variables that are most significantly correlated with speciation at these four points.

The speciation of mercury leaving the EERC pilot-scale combustor and entering the SCR was principally correlated with the concentrations of Cl, Ca, and S in the coal (Table D-1). The percentages of coal mercury occurring as elemental mercury increased along with Ca and were reduced by Cl, with both elements indicated to be statistically significant at the 99% confidence level (P = 0.99) and the value of R^2 being 0.95 for N = 10. The percentages occurring as Hg^{2+} decreased with increasing Ca and S and increased along with Cl, meaning that the correlations for Hg^{2+} are the inverse of those for Hg^0. This correlation was also statistically significant as indicated by P values of 0.99 for both Ca and S and a lower value of 0.88 for Cl; the value of R^2 was 0.94 at N = 10. The percentages occurring as Hg_p, which were less well correlated at an R^2 of 0.58 for N = 10, increased along with Ca and Cl and were reduced by higher levels of coal Hg and SCR operation. Chlorine was the most significant variable with a P value of 0.96, followed by Hg at 0.88, SCR at 0.86, and Ca at 0.64.

Changes in mercury speciation across the SCR are represented by the regression equations in Table D-2. All of these correlations are based on the four tests performed with the SCR in operation (N = 4), and all changes are expressed as percent of mercury in coal. The change in total mercury across the SCR was negative for three of the four coals, possibly indicating adsorption. The upward trend from a negative to a positive change increased along with higher levels of coal Hg and higher values of Cl/Ca, with both variables being statistically significant at P values of 0.98 and 0.97 respectively. These high values of P for a two-variable regression with N = 4 (1 degree of freedom) indicate that the data points fall very close to the regression line, which was confirmed by graphing the data. The value of R^2 was 0.9997. Both positive and negative changes were observed in both elemental and oxidized mercury, suggesting that either oxidation or reduction could have occurred for different coals. The change in Hg^0 was negatively correlated with both Hg and Cl in coal at P levels of 0.99 and 0.86, respectively. The R^2 for the regression on changes in Hg^0 had a value of 0.9997. The change in Hg^{2+} was positively correlated with Hg and Cl/Ca at P levels of 0.98 and 0.95, with an R^2 of 0.9992.

Changes in mercury species are greater across the cooling loop than across the SCR, but the changes that occur with cooling are not nearly as well correlated with coal and operating factors as are changes across the SCR (Table D-3). The changes in elemental mercury Hg^0 increase from negative to positive along with higher levels of Cl and Hg in coal at P levels of 0.79 and 0.66 respectively; R^2 has a value of 0.37 at N = 12. The change in Hg^{2+} is positively correlated with Hg and Cl/Ca, and negatively correlated with S in coal, at P levels of 0.84, 0.56, and 0.66, respectively; R^2 is 0.32. The changes in particulate mercury increase with either NH_3 injection or SCR operation and are reduced by higher levels of Cl and Hg in coal; P values are 0.81, 0.65, 0.72, and 0.67, respectively, and R^2 is 0.58.

Regression equations for the mercury species emitted at the exit of the ESP are given in Table D-4. All of these regressions are for N=12, and the emissions are expressed as percent of Hg in coal. The percentages of particulate mercury emitted were influenced by the efficiency of the ESP, but values for Hg^0 and Hg^{2+} are believed to represent the chemistry of the system. The total mercury emitted increased along with Cl in coal and was reduced by NH_3 injection, at P levels of 0.68 and 0.92, respectively, and an R^2 of 0.41. The emission of elemental mercury was quite well correlated and was increased by Ca and reduced by S and Cl in coal and by SCR operation. The P levels are 0.99, 0.75, 0.62, and 0.94, respectively, and R^2 is 0.84. The emission of oxidized mercury was increased by Cl and reduced by Ca and NH_3 injection, which are the opposite of the coal effects shown for elemental mercury. P levels are 0.56, 0.86, and 0.97, respectively, and R^2 is 0.60. The emission of particulate mercury was increased by S in coal, NH_3 injection, and SCR operation and reduced by Cl and Ca in coal. P levels are 0.96, 0.73, 0.66, 0.94, and 0.66, indicating that coal S and Cl contents are the most significant variables.

Some of the trends described above are in agreement with the understanding of mercury combustion chemistry emerging from other research. The statistically indicated effect of coal Cl to increase Hg^{2+} and reduce Hg^0 is in this category. The opposite effects indicated for coal Ca and S, tending to increase Hg^0 and reduce Hg^{2+}, support trends that have been observed in other research, but the mechanisms responsible for these effects are less well understood. It is in the area of the statistically significant trends in mercury speciation through the SCR that the present tests and regression results may be providing new insights. The statistical correlations for changes in the SCR suggest a combination of adsorption and chemical oxidation or reduction depending on the levels of Hg, Cl, and Ca in the coal. These trends very much need to be confirmed by additional laboratory research and field testing before they are used generally to describe the effects of SCR on mercury. The positive correlation of an increase in particulate Hg indicated to occur with NH_3 injection, both through the cooling loop and at the EST exit, is another trend indication that may warrant follow-up. For now, as was stated previously, the correlation equations in Appendix D should not be treated as reliable predictors of speciation but only as trend indicators.

4.4 Potential Hg_p Formation Mechanisms

During combustion, sulfur is liberated from coal and subsequently oxidized to SO_2. A fraction of the SO_2, generally 1%–3%, is oxidized to SO_3 [30]. The oxidation of SO_2 to SO_3 is catalyzed by transition metal oxides, such as the V_2O_5-based catalyst in the SCR reactor, but is neutralized by alkali and alkaline-earth metals in ash [31]. SO_3 can react downstream from the SCR with excess NH_3 (i.e., NH_3 that did not react with NO_x) to create aerosols (particles 0.1–0.2 µm in diameter) that subsequently condense, predominantly as ammonium sulfate or ammonium bisulfate. These small, sticky particles can cause major clogging problems in an air preheater and on SCR catalyst surfaces. More importantly, with regard to this research, these small particles provide significant surface area, possibly for heterogeneous reactions involving mercury. Research indicates that heterogeneous reactions and physisorption are important mechanisms for transforming Hg^0 to Hg^{2+} and Hg_p [16–18].

Alkaline-earth metals such as calcium will react with SO_2 and SO_3, especially on surfaces, to form calcium sulfite ($CaSO_3$) and calcium sulfate ($CaSO_4$). The $CaSO_3$ and $CaSO_4$ particles generated during combustion are probably very fine and widely distributed, thus increasing the potential for reactions to occur on air heater tube and catalyst surfaces. In addition to SO_2 and SO_3, CaO will react with Cl [32]. The Cordero Rojo and Blacksville fly ashes possess greater CaO concentrations relative to Paradise and Band Mill fly ashes.

It is expected that sulfated compounds ($NH_xCl_ySO_z$) are likely to be small, sticky aerosols/particulate matter in the 0.1–0.2-µm size. Assuming these types of compounds form, they would also significantly increase surface area for mercury heterogeneous condensation reactions. Additionally, because these compounds are chlorinated, they may increase mercury oxidation. If the assumptions stated above are accurate, this may offer an explanation as to why increased mercury and Cl are measured in the Paradise SCR test fly ash but not in the other two Paradise coal tests. This may also explain why the Blacksville ash for the SCR test had less mercury than the NH_3 injection tests. That is, during the Blacksville SCR, there was little unreacted NH_3 to react with SO_3 or possibly Cl.

4.5 Particle–Gas Partitioning of Mercury

The solid–gas partitioning of mercury at the ESP inlet and outlet sampling locations was measured using a part of the OH method. It has been previously seen that particle–gas filtration through a fixed bed of ash particles, especially fine ash particles, can promote Hg_p formation [33]. In order to investigate whether this occurred during the Paradise test, a heated 5-stage cascade multicyclone sampler was used to collect aerodynamically sized fly ash samples at the ESP inlet. Simultaneously, ash samples were collected at the same location using a standard filter. The measured particle-size distribution using the multicyclone is presented in Figure 4-10. Submicron fly ash particles that most likely penetrated the ESP account for only about 1.5% of the total fly ash mass. However, the total number and surface area of these small particles are high relative to their mass concentration. Mercury concentrations for the size-fractionated Paradise fly ash samples produced during the SCR test are presented in Figure 4-11. Although mercury is most concentrated in the smallest particle-size fraction (i.e., filter catch) and least concentrated in the largest particle-size fraction, on a mass basis, most of the mercury appears to be associated with the larger particles. Mercury concentrations for the bulk fly ashes sampled on the OH sample filter and multicyclones were 1.29 and 1.28 ppm, respectively. These values are essentially identical, within sampling and analytical errors, indicating that the particle–gas filtration through a fixed bed of Paradise fly ash did not promote Hg_p formation any more than did cyclonic filtration.

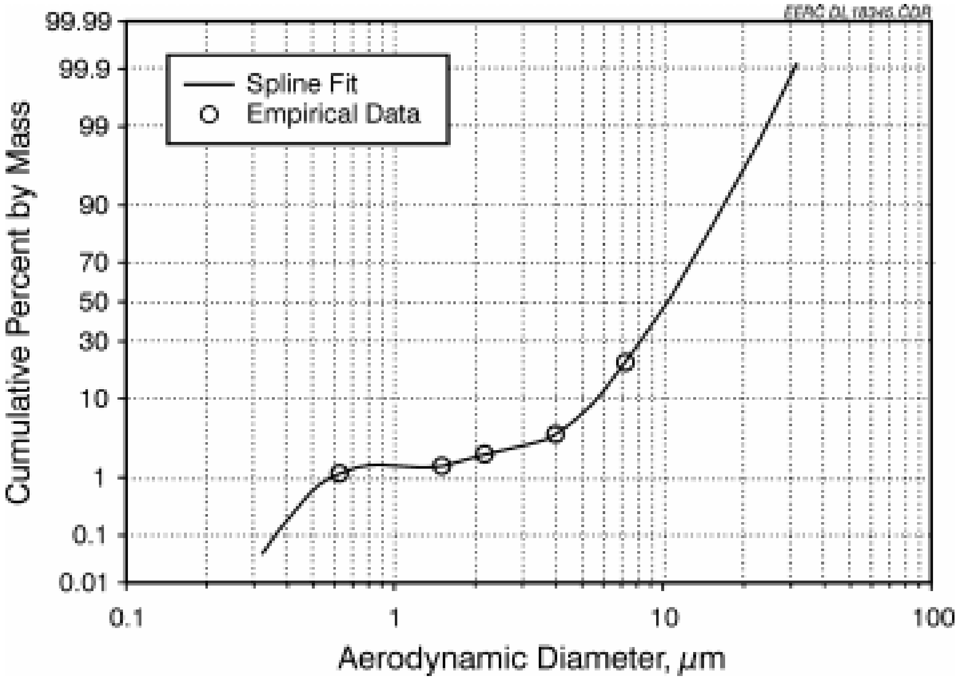

Figure 4-10
Particle-Size Distribution at the ESP Inlet for the Paradise SCR Test

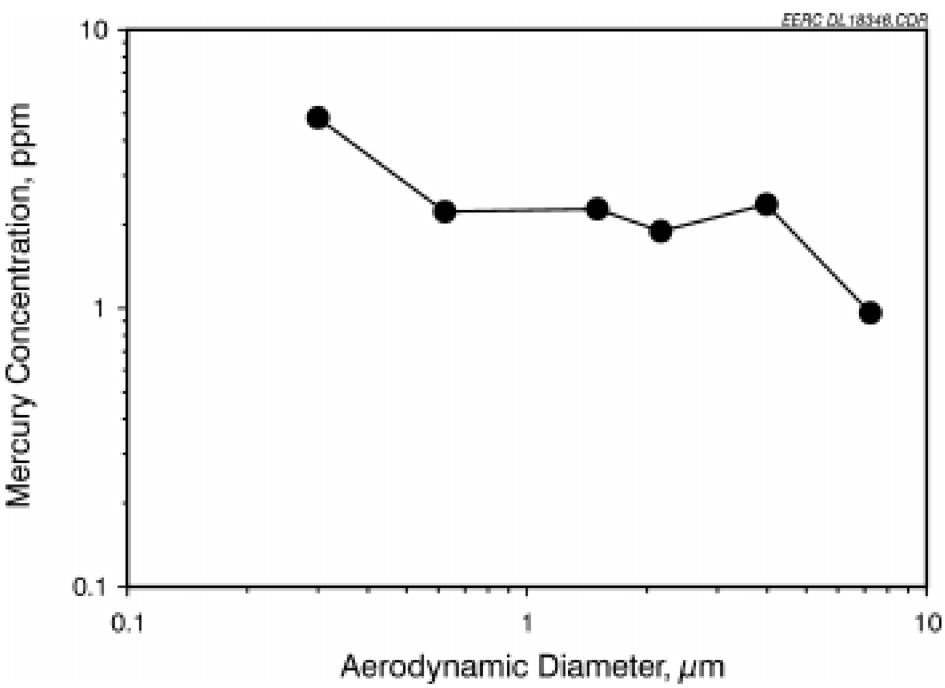

Figure 4-11
Mercury Concentration as a Function of ESP Inlet Particle Size for the Paradise SCR Test

5 CONCLUSIONS

Based on the results obtained from this screening evaluation, the following conclusions can be made:

- For some coals, NH_3 appeared to increase the Hg_p concentration; thereby increasing Hg removal in the downstream ESP. Because NH_3 slips were higher than expected in a full-scale SCR or SNCR application and NH_3 concentration may directly impact Hg speciation and removal, these results may or may not be consistent with full-scale applications.
- The impact of SCR on mercury speciation and mercury capture appears to be very coal dependent and quite complex.
- Based on a regression analysis, the chlorine, sulfur, and calcium appear to correlate with mercury speciation across the SCR.
- Relatively high concentrations of alkaline-earth metals (i.e., CaO and MgO) in the Cordero Rojo and Blacksville fly ashes may have limited the suspected interactions involving SO_3, NH_3, and Cl that promote Hg_p formation.
- NH_3 injection and/or the SCR catalyst promoted the conversion of Hg^0 to Hg^{2+} across the SCR for the Paradise coal, but not for any of the others.
- NH_3 injection, with and without the SCR reactions, converted Hg^{2+} to Hg_p when the Paradise and Band Mill coals were fired, but not for the Cordero Rojo PRB.
- The increased mercury removals as measured by the flue gas measurements were confirmed with mercury analyses of the corresponding fly ash. The Hg_p concentrations in Paradise and Band Mill ESP hopper ashes increased by 230%–460% relative to the baseline fly ashes as a result of NH_3 injection and SCR tests.
- Because of the high levels of Hg^{2+} in the baseline tests, it is not possible to determine whether there was an increase in Hg_p or oxidation of Hg^0 for the Blacksville coal.

The applicability of the conclusions from this pilot-scale investigation should be evaluated by performing similar flue gas and fly ash measurements at utility-scale boilers equipped with selective noncatalytic reduction (SNCR) and SCR units. As part of this proposed utility-scale investigation, flue gas and fly ash samples should be collected when the SNCR and SCR units are off-line and on-line. Size-fractionated fly ash samples should also be collected and analyzed to investigate further the apparent role of particle size and composition on Hg_p formation. Additional coals also need to be tested because the impact of SNCR and SCR of NO_x on mercury speciation apparently depends on the coal's chemical composition.

6 RECOMMENDATIONS

These pilot-scale tests were completed only as a screening evaluation of the impact of SCR on mercury speciation. In a number of ways, these tests did not mimic a full-scale combustor, SCR, and ESP. For example, it is possible that the mercury flue gas chemistry may be different firing coal in the PTC compared to a full-scale pulverized coal combustor. It appears that PTC generates more Hg^{2+} than predicted using the EPRI correlations developed from the ICR results [2]. These correlations are based on the sulfur and chloride content of the coal. Table 6-1 compares the EPRI correlation with the EERC speciation baseline results at the ESP inlet sampling location for the four test coals. The difference may be due to shorter residence time along with a different time/temperature profile for the PTC compared to a full-scale boiler.

Table 6-1
Comparison of EERC Mercury Speciation Results to Those Predicted by EPRI's Correlations

Fuel	Hg^0 Concentration based on EPRI Correlations, %	Hg^0 Concentration based on EERC Measurement, %
Paradise Bituminous Coal	46	7
Cordero Rojo Subbituminous Coal	71	61
Band Mill Bituminous Coal	69	16
Blacksville Bituminous Coal	36	13

The EERC's ESP was never intended to represent a full-scale system. Therefore, for a number of the tests, the particulate collection efficiency was low and varied considerably. The collection efficiency for the three bituminous coal tests averaged about 90% compared to >99% for a well-operating full-scale ESP. Also, because of a calibration error, several of the tests had much higher NH_3 concentrations in the flue gas than would have been desired. This may have complicated the interpretation of the data and resulted in more variability.

Therefore, the applicability of the conclusions from these pilot-scale test must be evaluated by performing similar flue gas and fly ash measurements at utility-scale boilers equipped with SNCR and SCR units.

A final recommendation is that bench-scale studies be completed to provide a better understanding of mercury, NH_3, SO_3, and fly ash chemistry as it relates to SCRs. Currently, some of this work is being done by the University of Cincinnati, but these pilot-scale results may warrant an expansion of that program or the initiation of a new bench-scale project.

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A FLUE GAS TEMPERATURE

Table A-1
Paradise Bituminous Coal Average Flue Gas Temperatures, °F

	Baseline		NH_3 Injection		SCR	
	Average	**Std. Dev.**	**Average**	**Std. Dev.**	**Average**	**Std. Dev.**
SCR Bypass	610	68	603	38	—*	—
SCR Inlet	—	—	—	—	651	48
SCR	—	—	—	—	642	22
SCR Outlet	—	—	—	—	627	39
ESP Inlet	349	13	372	17	332	16
ESP Outlet	294	12	309	14	283	15
Deposition Tube	399	14	398	12	399	15

*Not applicable.

Table A-2
Cordero Rojo Coal Average Flue Gas Temperatures, °F

	Baseline		NH_3 Injection		SCR	
	Average	**Std. Dev.**	**Average**	**Std. Dev.**	**Average**	**Std. Dev.**
SCR Bypass	607	42	614	21	—*	—
SCR Inlet	—	—	—	—	661	—
SCR	—	—	—	—	655	—
SCR Outlet	—	—	—	—	643	12
ESP Inlet	339	7	341	7	345	7
ESP Outlet	290	8	291	7	294	7
Deposition Tube	401	6	402	8	403	5

*Not applicable.

Table A-3
Average Band Mill Coal Combustion Flue Gas Temperatures, °F

	Baseline		NH_3 Injection		SCR	
	Average	**Std. Dev.**	**Average**	**Std. Dev.**	**Average**	**Std. Dev.**
SCR Bypass	637	67	624	34	—*	—
SCR Inlet	—	—	—	—	671	18
SCR	—	—	—	—	664	11
SCR Outlet	—	—	—	—	655	16
ESP Inlet	341	4	360	26	346	16
ESP Outlet	291	3	300	11	291	13
Deposition Tube	391	57	398	22	399	9

*Not applicable.

Table A-4
Average Blacksville Coal Combustion Flue Gas Temperatures, °F

	Baseline		NH_3 Injection		SCR	
	Average	**Std. Dev.**	**Average**	**Std. Dev.**	**Average**	**Std. Dev.**
SCR Bypass	636	19	625	25	—*	—
SCR Inlet	—	—	—	—	655	6
SCR	—	—	—	—	653	11
SCR Outlet	—	—	—	—	642	19
ESP Inlet	344	9	351	10	350	7
ESP Outlet	293	12	303	9	301	7
Deposition Tube	399	16	397	22	398	20

*Not applicable.

B SAMPLE CALCULATIONS

Example Calculations of NH_3 Flue Gas Flow Rates and Mass Balances

Constants

Molar volume of an ideal gas at 0°C and 1 atm pressure = 22.41 L
Formula weight of NH_3 = 17.031 g/mol
Combustor flue gas flow rate ≈ 175 Nm^3/hr = 175,000 L/hr

$$700\ ppm \times \frac{17.031\ g/mol \times 175{,}000\ L/hr}{22.41\ L/mol \times 10^6} = 93.1\ g/hr$$

Conversion of 5 ppmv NH_3(g) to a flue gas flow rate, g/hr

Conversion of 5000 µg/Nm^3 NH_3(p) to a flue gas flow rate, g/hr

$$5000\ \mu g/Nm^3 \times 175\ Nm^3/hr \times \frac{1g}{1{,}000{,}0000} = 0.88\ g/hr$$

Conversion of NO_x removal to an equivalent NH_3 removal rate, g/hr

SCR inlet NO_x = 700 ppmv
ESP inlet NO_x = 20 ppmv
NO_x removal = 680 ppmv

$$680\ ppm \times \frac{17.031\ g/mol \times 175{,}000\ L/hr}{22.41\ L/mol \times 10^6} = 90.4\ g/hr$$

Example of Mercury Mass Balance Calculations for the SCR of NO_x Paradise Coal Test (609)

Mercury mass balances were calculated from the average coal Hg concentrations, coal feed rates, average flue gas flows, total Hg concentrations measured at each sampling location, electrostatic precipitator (ESP) dust loadings and fly ash collection efficiencies, and ESP hopper ash Hg concentrations. An example of these data is presented in Table B-1 for the selective catalytic reduction (SCR) of NO_x Paradise Coal Test (609).

Table B-1
Average Measurement Data for the SCR of NO_x Paradise Coal Test (609)

Parameter, unit	Value
Coal Feed Rate, g/hr	22,093
Coal Hg, as-received, µg/g	0.111
Flue Gas Flow Rate, dNm^3 @3% O_2/hr	186.51
ESP Inlet Dust Load, g/hr	1355.7
ESP Ash Collection Efficiency, %	63.3
ESP Hopper Ash Hg, µg/g	0.988
Total Hg at the SCR Bypass/Inlet, µg/dNm^3 @3% O_2	10.6
Total Hg at the SCR outlet, µg/dNm^3 @3% O_2	11.5
Total Hg at the ESP Inlet, µg/dNm^3 @3% O_2	11.4
Total Hg at the ESP Outlet, µg/dNm^3 @3% O_2	6.71

Hg mass balances for each sampling location are calculated relative to the average coal Hg feed rate during the combustion test where:

$$\text{coal Hg feed rate} = \text{coal feed rate} \times \text{coal Hg}$$

Therefore, the coal Hg feed rate was 2452 µg/hr (22,093 g/hr × 0.111 µg/g) during the SCR of NO_x Paradise coal test.

The removal of Hg from the flue gas by the ESP is accounted for using the following equation:

$$\text{ESP Hg removal rate} = \text{ESP inlet dust load} \times \text{ESP hopper ash Hg} \times (\text{ESP ash collection efficiency} \div 100)$$

According to the values in Table B-1, the ESP removed Hg from the flue gas at a rate of 847.8 µg/hr (1355.7 g/hr × 0.988 µg/g × [63.3 ÷ 100]).

Total Hg flow rates at the SCR bypass/inlet, SCR outlet, ESP inlet, and ESP outlet are calculated by simply multiplying the total Hg concentrations in Table B-1 by the flue gas flow rate, for example:

$$\text{total Hg flow rate at the SCR bypass/inlet} = 10.6\ \mu g/dNm^3\ @3\%\ O_2 \times 186.51\ dNm^3\ @3\%\ O_2/hr = 1977\ \mu g/hr$$

Total Hg flow rates for the four sampling locations are presented in Table B-2.

Percent Hg mass balances for the SCR inlet/bypass, SCR outlet, and ESP inlet are calculated by dividing the Hg flow rates in Table B-2 by the average coal Hg feed rate and multiplying by 100. For example, Hg mass balance at the ESP inlet was:

$$2126\ \mu g/hr \div 2452\ \mu g/hr \times 100 = 86.7\%.$$

A Hg mass balance for the ESP outlet is calculated by combining the ESP outlet Hg flow rate and ESP removal rate then dividing by the average coal Hg feed rate and multiplying by 100. For example, Hg mass balance at the ESP outlet was:

$$(1251\ \mu g/hr + 847.8\ \mu g/hr) \div 2452\ \mu g/hr \times 100 = 85.6\%$$

Mass balance results for the four sampling locations are presented in Table B-3.

Table B-2
Calculated Hg Feed, Flow, and Removal Rates for the SCR of NO_x Paradise Coal Test (609)

Parameter	µg/hr
Coal Hg Feed Rate	2452
ESP Hg Removal Rate	847.8
Total Hg Flow Rate at the SCR Bypass/Inlet	1977
Total Hg Flow Rate at the SCR Outlet	2145
Total Hg Flow Rate at the ESP Inlet	2126
Total Hg Flow Rate at the ESP Outlet	1251

Table B-3
Hg Mass Balance Results for the SCR of NO_x Paradise Coal Test (609)

Location	Hg Mass Balance, %
SCR Bypass/Inlet	80.6
SCR Outlet	87.5
ESP Inlet	86.7
ESP Outlet	85.6

C MERCURY AND NH_3 MASS BALANCE DATA

Mercury Mass Balance Data

Table C-1
Paradise Bituminous Coal – Tests 607 through 609 Mercury Mass Balance Data

Coal and Mercury Feed Rates			
Total Coal Burned, lb	5006		
Coal Feed Total Time, hr	102.8		
Coal Feed Rate, lb/hr	48.71		
Coal Feed Rate, g/hr	22,093		

Test No.	**607**	**608**	**609**
Coal Hg, as-received µg/g	0.11	0.114	0.111
Coal Hg Feed Rate, µg/hr	2430	2519	2452
Average Flue Gas Flow Rate, scfm	130.9	130	130
Average Flue Gas O_2, %	5.34	5.51	4.47
Average Flue Gas H_2O, %	8.02	7.87	8.06
Flue Gas Flow, dNm^3 @3%O_2/hr	177.99	175.13	186.51
Hooper Ash Mercury, µg/g	0.256	0.849	0.988
Avg. ESP Inlet Dust Load, grains/scf	2.6334	0.7625	2.6822
Avg. ESP Dust, g/hr	1340.2	385.4	1355.7
Avg. ESP Collection Efficiency, %	87	70.8	63.3
Hopper Ash Mercury, µg/hr	298.5	231.7	847.8

Table C-2
Paradise Bituminous Coal – Tests 607 through 609 Mercury Mass Balance Results

Run No.			**607**					**608**					**609**		
Total Mercury Flow Rate, µg/hr															
Sample	**1**	**2**	**3**	**Avg.**	**Std. Dev.**	**1**	**2**	**3**	**Avg.**	**Std. Dev.**	**1**	**2**	**3**	**Avg.**	**Std. Dev.**
SCR In	2193	–	–	–	–	1879	–	–	–	–	1656	–	–	1909*	270*
SCR Out	–	–	–	–	–	–	–	–	–	–	1938	1179	2740	1952	781
ESP In	2179	2173	1876	2076	173	1373	1602	1827	1601	227	2259	2089	2050	2133	111
ESP Out	1771	1977	1842	1863	105	904	1119	1158	1060	137	1548	1546	662	1252	511
Mercury Mass Balance, %															
SCR In	90.2	–	–	–	–	74.6	–	–	–	–	67.5	–	–	77.5*	11.6**
SCR Out	–	–	–	–	–	–	–	–	–	–	79.0	72.6	111.7	87.8	21.0
ESP In	89.6	89.4	77.2	85.4	7.1	54.5	63.6	72.5	63.6	9.0	92.1	85.2	83.6	87.0	4.5
ESP Out	85.2	93.7	88.1	89.0	4.3	45.1	53.6	55.2	51.3	5.4	97.7	97.6	61.6	85.6	20.8

Table C-3
Cordero Rojo Subbituminous Coal – Tests 610 through 612 Mercury Mass Balance Data

Coal and Mercury Feed Rates	
Total Coal Burned, lb	6812
Coal Feed Total Time, hr	99.68
Coal Feed Rate, lb/hr	68.34
Coal Feed Rate, g/hr	30,998

Test No.	**610**	**612**	**613**
Coal Hg, as-received µg/g	0.087	0.091	0.077
Coal Hg Feed Rate, µg/hr	2697	2821	2387
Average Flue Gas Flow Rate, scfm	133.1	131.1	132.1
Average Flue Gas O_2, %	4.53	4.48	4.81
Average Flue Gas H_2O, %	11.06	10.42	11.30
Flue Gas Flow, dNm^3 @3%O_2/hr	184.05	183.15	179.08
Hooper Ash Mercury, µg/g	0.365	0.173	0.364
Avg. ESP Inlet Dust Load, grains/scf	1.8002	1.9956	1.4126
Avg. ESP Dust, g/hr	932	1017	726
Avg. ESP Collection Eff., %	99.47	99.73	99.72
Hopper Ash Mercury, µg/hr	338.2	175.5	263.3

Table C-4
Cordero Rojo Subbituminous Coal – Tests 610 through 612 Mercury Mass Balance Results

Run No.			**610**					**611**					**612**		
Total Mercury Flow Rate, µg/hr															
Sample	**1**	**2**	**3**	**Avg.**	**Std. Dev.**	**1**	**2**	**3**	**Avg.**	**Std. Dev.**	**1**	**2**	**3**	**Avg.**	**Std. Dev.**
SCR In	2591	–	–	–	–	2714	–	–	–	–	3066	–	–	2790[a]	247[a]
SCR Out	–	–	–	–	–	–	–	–	–	–	–	2190	578	2101	2146[b]63[b]
ESP In	2560	2360	2415	2445	103	2441	2879	2890	2737	256	2767	2357	3331	2818	489
ESP Out	1465	1520	2063	1683	330	1749	2016	2247	2004	249	1035	1173	1511	1240	245
Mercury Mass Balance, %															
SCR In	96.1	–	–	–	–	96.2	–	–	–	–	128.4	–	–	106.9[a]	18.6[a]
SCR Out	–	–	–	–	–	–	–	–	–	–	91.8	24.2	88.0	89.0[b]	2.7[b]
ESP In	94.9	87.5	89.5	90.7	3.8	86.5	102.1	102.5	97.0	9.1	115.9	98.7	139.6	118.1	20.5
ESP Out	66.9	68.9	89.0	74.9	12.3	68.2	77.7	85.9	77.3	8.8	54.4	60.2	74.4	63.0	10.3

[a]Average and standard deviation of the three runs (610–612).
[b]Sample Number 2 was not used to calculate average and standard deviation.

Table C-5
Band Mill Bituminous Coal – Tests 613 through 615 Mercury Mass Balance Data

Coal and Mercury Feed Rates	
Total Coal Burned, lb	4501
Coal Feed Total Time, hr	103.28
Coal Feed Rate, lb/hr	43.58
Coal Feed Rate, g/hr	19,768

Test No.	**613**	**614**	**615**
Coal Hg, as-received µg/g	0.023	0.022	0.022
Coal Hg Feed Rate, µg/hr	455	435	435
Average Flue Gas Flow Rate, scfm	131.2	129.9	130.6
Average Flue Gas O_2, %	5.20	5.15	4.99
Average Flue Gas H_2O, %	7.36	7.12	7.34
Flue Gas Flow, dNm^3 @3%O_2/hr	18.28	180.5	182.89
Hooper Ash Mercury, µg/g	0.082	0.353	0.456
Avg. ESP Inlet Dust Load, grains/scf	1.5559	0.8229	0.7559
Avg. ESP Dust, g/hr	794	416	384
Avg. ESP Collection Eff., %	94.0	92.6	97.6
Hopper Ash Mercury, µg/hr	61.2	135.9	170.8

Table C-6
Band Mill Bituminous Coal – Tests 613 through 615 Mercury Mass Balance Results

Run No.	613					614					615				
Total Mercury Flow Rate, µg/hr															
Sample	**1**	**2**	**3**	**Avg.**	**Std. Dev.**	**1**	**2**	**3**	**Avg.**	**Std. Dev.**	**1**	**2**	**3**	**Avg.**	**Std. Dev.**
SCR In	874	–	–	–	–	132	–	–	–	–	540	–	–	515*	372*
SCR Out	–	–	–	–	–	–	–	–	–	–	358	315	351	341	24
ESP In	513	488	412	471	53	383	204	325	304	128	318	382	342	347	32
ESP Out	1196	509	422	709	424	112	161	112	128	28	91	79	88	86	7
Mercury Mass Balance, %															
SCR In	192.2	–	–	–	–	30.3	–	–	–	–	124.1	–	–	115.5*	81.3*
SCR Out	–	–	–	–	–	–	–	–	–	–	82.4	72.3	80.7	78.5	5.4
ESP In	112.8	107.3	90.5	103.5	11.6	88.0	46.9	74.7	69.9	21.0	73.2	87.9	78.6	79.9	7.4
ESP Out	276.6	125.5	106.4	115.9	93.3	57.0	68.2	57.0	60.7	6.5	60.3	57.4	59.5	59.0	1.5

[a]Average and standard deviation of the three runs (613–615).

Table C-7
Blacksville Bituminous Coal – Tests 616 through 618 Mercury Mass Balance Data

Coal and Mercury Feed Rates	
Total Coal Burned, lb	4688.0
Coal Feed Total Time, hr	103.5
Coal Feed Rate, lb/hr	45.31
Coal Feed Rate, g/hr	20,551

Test No.	**616**	**617**	**618**
Coal Hg, as-received, µg/g	0.0950	0.0899	0.0973
Coal Hg Feed Rate, µg/hr	1952	1848	2000
Average Flue Gas Flow Rate, scfm	131.5	130.5	129.9
Average Flue Gas O_2, %	5.31	5.55	4.81
Average Flue Gas H_2O, %	7.24	7.32	7.76
Flue Gas Flow, dNm^3 @3%O_2/hr	180.67	176.40	183.12
Hooper Ash Mercury, µg/g	0.252	0.555	0.320
Avg. ESP Inlet Dust Load, grains/scf	1.4053	0.7500	1.5684
Avg. ESP Dust, g/hr	718.5	380.5	792.1
Avg. ESP Collection Eff., %	96.9	96.4	82.6
Hopper Ash Mercury, µg/hr	175.4	203.6	209.4

Table C-8
Blacksville Bituminous Coal – Tests 616 through 618 Mercury Mass Balance Results

Run No.			**616**					**617**					**618**		
Total Mercury Flow Rate, μg/hr															
Sample	**1**	**2**	**3**	**Avg.**	**Std. Dev.**	**1**	**2**	**3**	**Avg.**	**Std. Dev.**	**1**	**2**	**3**	**Avg.**	**Std. Dev.**
SCR In	1836	–	–	–	–	1589	1662	–	–	–	1774	1687	–	1710[*]	97[*]
SCR Out	–	–	–	–	–	–	–	–	–	–	1696	1720	1720	1712	14
ESP In	1819	1727	1702	1749	62	1572	1454	1469	1498	64	1714	2011	1817	1847	151
ESP Out	1700	1561	1460	1574	121	1259	1073	1261	1198	109	1384	1419	1470	1425	43
Mercury Mass Balance, %															
SCR In	94.0	–	–	–	–	86.0	89.9	–	–	–	88.7	84.3	–	88.6	4.1
SCR Out	–	–	–	–	–	–	–	–	–	–	84.8	86.0	86.0	85.6	0.7
ESP In	93.2	88.5	87.2	89.6	3.2	85.1	78.7	79.5	81.1	3.5	85.7	100.6	90.8	92.4	7.5
ESP Out	96.1	88.9	83.8	89.6	6.2	79.2	69.1	79.3	75.8	5.9	79.7	81.4	84.0	81.7	2.2

[a]Average and standard deviation of the three runs (616–618).

NH_3 Mass Balance Data

Table C-9
Paradise NH_3(g,p) Analysis and Mass Balance Results

Test	NH_3(g), ppmv	NH_3(g), mg/Nm^3	NH_3(p), mg/Nm^3	Mass Balance, %
Baseline	6.69	5.09	0.64	NA[a]
25 ppm – NH_3 Injection	2.44	1.85	2.66	18
25 ppm – NH_3 Injection	2.36	1.79	5.93	31
25 ppm – NH_3 Injection	2.69	2.04	5.82	32
Average	2.50	1.89	4.80	27
Std. Dev.[b]	0.17	0.13	1.86	8
SCR of NO_x	1.41	1.07	4.25	83[c]
SCR of NO_x	0.88	0.67	4.73	103[c]
SCR of NO_x	86.4	65.7	29.4	106[c]
Average	29.6	22.5	12.8	97
Std. Dev.	49.2	37.4	14.4	12

[a] Not applicable.
[b] Sample standard deviation calculated from the average of three analyses.
[c] Calculated assuming that the ratio of converted moles of NO_x and NH_3 is 1.

Table C-10
Cordero Rojo NH_3(g,p) Analysis and Mass Balance Results

Test	NH_3(g), ppmv	NH_3(g), mg/Nm^3	NH_3(p), mg/Nm^3	Mass Balance, %
Baseline	0.82	0.63	0.12	NA[a]
25 ppm – NH_3 Injection	13.7	10.4	0.07	126
25 ppm – NH_3 Injection	12.9	9.80	0.10	119
25 ppm – NH_3 Injection	14.0	10.7	0.30	131
Average	13.5	10.3	0.16	125
Std. Dev.[b]	0.6	0.5	0.13	6
SCR of NO_x	216	165	0.95	120[c]
SCR of NO_x	119	90.4	0.94	116[c]
SCR of NO_x	94.7	72.0	1.01	105[c]
Average	143	109	0.97	114
Std. Dev.	64	49	0.04	8

[a] Not applicable.
[b] Sample standard deviation calculated from the average of three analyses.
[c] Calculated assuming that the ratio of converted moles of NO_x and NH_3 is 1.

Table C-11
Band Mill NH_3(g, p) Analysis and Mass Balance Results

Test	NH_3(g), ppmv	NH_3(g), mg/Nm^3	NH_3(p), mg/Nm^3	Mass Balance, %
Baseline	0.43	0.33	0.17	NA[a]
25 ppm – NH_3 Injection	4.61	3.51	2.35	72
25 ppm – NH_3 Injection	4.21	3.20	2.97	76
25 ppm – NH_3 Injection	5.19	3.95	2.82	84
Average	4.67	3.55	2.71	77
Std. Dev.[b]	0.49	0.38	0.32	6
SCR of NO_x	262	199	4.47	129[c]
SCR of NO_x	173	132	4.03	118[c]
SCR of NO_x	132	101	3.96	114[c]
Average	189	144	4.15	121
Std. Dev.	66	50	0.28	8

[a] Not applicable.
[b] Sample standard deviation calculated from the average of three analyses.
[c] Calculated assuming that the ratio of converted moles of NO_x and NH_3 is 1.

Table C-12
Blacksville NH_3(g, p) Analysis and Mass Balance Results

Test	NH_3(g), ppmv	NH_3(g), mg/Nm^3	NH_3(p), mg/Nm^3	Mass Balance, %
Baseline	2.18	1.65	0.26	NA[a]
25 ppm – NH_3 Injection	0.85	0.65	2.66	35
25 ppm – NH_3 Injection	0.53	0.40	2.55	31
25 ppm – NH_3 Injection	0.55	0.42	2.62	32
Average	0.64	0.49	2.61	32
Std. Dev.[b]	0.18	0.14	0.06	2
SCR of NO_x	0.60	0.45	1.52	98[c]
SCR of NO_x	0.26	0.20	1.44	96[c]
SCR of NO_x	0.35	0.27	1.88	100[c]
Average	0.40	0.31	1.61	98
Std. Dev.	0.18	0.13	0.23	2

[a] Not applicable.
[b] Sample standard deviation calculated from the average of three analyses.
[c] Calculated assuming that the ratio of converted moles of NO_x and NH_3 is 1.

D LINEAR REGRESSION ANALYSIS FOR THE PILOT-SCALE SCREENING TESTS EVALUATING THE IMPACT OF SCR ON MERCURY SPECIATION

Linear Regression

Appendix D presents linear regression equations for mercury speciation in reference to the chemical composition of the four test coals (Hg, S, Cl, and Ca concentrations in parts per million on a moisture-free basis) and operating conditions (NH_3 and SCR are assigned a value of 1 when these operations were used and a value of 0 when they were not). These equations respectively deal with 1) mercury speciation leaving the furnace and entering the SCR, 2) changes in mercury speciation across the SCR, 3) changes in speciation across the cooling loop, and 4) the percentages of coal mercury emitted as different species at the exit of the ESP. All speciation predictions are expressed as percent of coal mercury. The statistical parameters included in these equations include 1) a value of R^2 for a specified number of data points (N), 2) the positive or negative "x coefficient" which indicates proportional dependence on the independent regression variable, and 3) the probability (P) that the x coefficient is significantly different from zero. The only independent variables presented are those with the highest statistical significance for a given set of data points. Some variables, particularly where N is small, still have what would be considered to be a low statistical significance as indicated by their P values, where a P value of 0.95 is often used to identify a statistically significant variable. For most of the regression data sets, N has a value of 12 (4 coals $\times$ 3 test conditions). The regression data sets for the Hg^0 and Hg^{2+} species leaving the furnace and entering the SCR are a special case and have an N of only 10 because the three measured values of these species for Band Mill coal were averaged to eliminate a wide divergence at essentially the same furnace condition. N has a value of 4 for those data sets that represent SCR operation. Because of low values of N and scatter in the data, these regression equations should not be interpreted as reliable predictors of speciation but only as general indications of trends. Also, it should be noted that several factors concerned with the overall data set limit the validity of the regressions for full-scale plants. These factors include possible differences in mercury speciation as a function of the much smaller size of the pilot-scale combustor, the limited number and range of coal types, and differences in ammonia slip concentrations. These are discussed in the recommendation section of this report.

D-1 Regression Equations for Mercury Speciation Leaving the Furnace and Entering the SCR

Elemental Mercury

N = 10, R^2 = 0.95

$\%X^0$ (percent of Hg in coal) = 39.0 + 0.00211 Ca – 0.0391 Cl
P = 0.99 P = 0.99

Oxidized Mercury

N= 10, R^2 = 0.94

% X^{2+} (percent of Hg in coal) = 109 – 0.00471 Ca − 0.00205 S + 0.0190 Cl
P = 0.99 P = 0.99 P = 0.88

Particulate Mercury

N = 10, R^2 = 0.58

% X_p (percent of Hg in coal) = 1.8 + 0.0000701 Ca + 0.00361 Cl – 21.67 Hg – 1.12 SCR
P = 0.64 P = 0.96 P = 0.88 P = 0.86

D-2 Regression Equations for Changes in Mercury Species Across the SCR

Total Mercury

N = 4, R^2 = 0.9993

Change in Hg_{total} (percent of Hg in coal) = –78.5 + 484 Hg + 174 Cl/Ca
P = 0.98 P = 0.97

Elemental Mercury

N = 4, R^2 = 0.9997

Change in Hg^0 (percent of Hg in coal) = 54.7 – 594 Hg – 0.00648 Cl
P = 0.99 P = 0.86

Oxidized Mercury

N = 4, R^2 = 0.9992

Change in Hg^{2+} (percent of Hg in coal) = – 131 + 1063 Hg + 178 Cl/Ca
P = 0.98 P = 0.95

D-3 Regression Equations for Changes in Mercury Species Across the Cooling Loop, Including When the SCR Is in Operation

Elemental Mercury

N = 12, R^2 = 0.37

Change in Hg^0 (percent of Hg in coal) = –46.2 + 0.0340 Cl + 186 Hg – 0.000210 S
P = 0.79 P = 0.66 P = 0.2*

Oxidized Mercury

N = 12, R^2 = 0.32

Change in Hg^{2+} (percent of Hg in coal) = –32.6 + 623 Hg + 219 Cl/Ca – 0.00326 S
P = 0.84 P = 0.56 P = 0.66

Particulate Mercury

N = 12, R^2 = 0.58

Change in Hg_p (percent of Hg in coal) = 31.5 + 21.2 NH_3 + 14.7 SCR – 0.0253 Cl – 178 Hg
P = 0.81 P = 0.65 P = 0.72 P = 0.67

*S was included because of a value of P = 0.78 for S in a regression on it alone.

D-4 Regression Equations for Mercury Species Emitted at the Exit of the ESP

Total Mercury

N = 12, R^2 = 0.47

Total Hg Emitted (percent of Hg in coal) = 68.6 + 0.0195 Cl – 25.7 NH_3 – 2.65 SCR
P = 0.68 P = 0.92 P = 0.2*

Elemental Mercury

N = 12, R^2 = 0.84

Hg^0 Emitted (percent of Hg in coal) = 21.0 + 0.00143 Ca – 11.6 SCR – 0.000391 S – 0.00994 Cl
P = 0.99 P = 0.94 P = 0.75 P = 0.62

Oxidized Mercury

N = 12, R^2 = 0.60

Hg^{2+} Emitted (percent of Hg in coal) = 51.0 + 0.0395 Cl – 38.0 NH_3 – 0.000771 Ca
P = 0.56 P = 0.97 P = 0.86

Particulate Mercury

N = 12, R^2 = 0.74

Hg_p Emitted (percent of Hg in coal) =

–1.8 + 0.000889 S + 7.7 NH_3 + 6.55 SCR – 0.0262 Cl – 0.000467 Ca
P = 0.96 p = 0.73 P = 0.66 P = 0.94 P = 0.66

* SCR was included because of a value of P = 0.74 in a regression on it alone.