FINAL TECHNICAL REPORT
November 1, 2000, through January 31, 2002

Project Title: **ROLE OF COAL CHLORINE AND FLY ASH ON MERCURY SPECIES IN COAL COMBUSTION ASH**

ICCI Project Number: 00-1/2.2C-2
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ABSTRACT

CONSOL R&D evaluated mercury concentrations around air heaters at two power plants in Illinois to evaluate the impact of coal chlorine on mercury speciation. The data suggest that, while coal chlorine is an important contributor, it is not the only factor important to mercury speciation. The air heater played a major role in oxidizing mercury. At both locations, the air heater inlet Hg speciation was mostly elemental Hg, while at the air heater outlet oxidized Hg predominated. Particulate Hg was not substantially present at the air heater inlet at either plant; however, at the air heater outlet, substantial particulate Hg was measured at Site 1 but not Site 2. The fraction of particulate mercury was substantially affected by the air heater outlet temperature at Site 1; the air heater outlet temperature had a negligible effect on Hg speciation at Site 2. A measurable amount of Hg was observed at the air heater air side outlet at both plants, which means that a small portion of the mercury entering the air heater can be recycled back to the boiler.

EXECUTIVE SUMMARY

The sampling program was performed during the week of October 29, 2001 at Site 1 and the week of November 5, 2001 at Site 2. Testing at both plants consisted of quadruplicate flue gas Hg measurements across the air heater. Feed coal samples and fly ash samples were obtained.

The flue gas Hg speciation data indicate that the elemental (Hg^0) and oxidized (Hg^{++}) fractions of the flue gas Hg at the air heater inlet are substantially different from the fractions at the air heater outlet. The air heater inlet Hg speciation estimated from the Ontario Hydro sampling method showed between 60% and 90% elemental Hg. Outlet speciation data showed between 10% and 25% elemental Hg. Particulate Hg was not substantially present at the air heater inlet at either plant; however, at the air heater outlet, substantial (nearly 50%) particulate Hg was measured at Site 1 but not Site 2 (less than 10% particulate Hg).

Air heater outlet mercury speciation was determined at two points in the outlet duct – the maximum temperature point and the minimum temperature point. At both sites, the difference was about 30 °F. At the Site 1 air heater outlet, the temperature had a substantial effect on mercury speciation. At the minimum temperature point (265 °F) about 55% of the mercury was particulate mercury; at the maximum temperature point (295 °F) the particulate mercury was about 30% of the total mercury. At site 2, the temperature (285-315 °F) had very little effect on mercury speciation.

A measurable amount of Hg was observed at the air heater air side outlet at both plants. This air is the combustion air going to the boiler, which means that a small portion (about 5%) of the mercury entering the air heater is recycled back to the boiler. This also provides additional indication that the air heater is interacting with the mercury in the flue gas. The results of testing at these sites demonstrated that the air heater had a substantial effect on Hg speciation. This information adds to the understanding of the behavior of mercury in plants burning Illinois basin coals.

OBJECTIVES

The objective for the proposed work is to develop fundamental information regarding the impact of coal chlorine on mercury speciation in coal combustion systems. This information is needed to develop cost-effective mercury controls beneficial to the Illinois coal market. Specific Technical Objectives of the program are:

- Determine the flue gas mercury speciation at two Illinois-based utilities at the inlet of the air heater, the combustion air exit of the air heater, and two flue gas temperatures at the outlet of the air heater.
- Collect an analyze coal, ESP ash, and if available, air heater deposit samples representative of the flue gas sampling period.
- Correlate the observed mercury speciation with analytical and operating data to identify factors that influence mercury speciation, specifically the role of coal chlorine.
- Identify the role of the air heater in mercury speciation and removal.

CONSOL R&D initiated this study to evaluate the role of coal chlorine in mercury speciation. When originally designed, one station was firing a coal containing 0.35% chlorine while the other was firing a coal containing 0.1% chlorine. Prior to testing, however, the first station switched fuel supplies, and the coal fired during the test period contained 0.17% chlorine. The change in coal chlorine content was not discovered until the coal sample was analyzed.

INTRODUCTION AND BACKGROUND

The Environmental Protection Agency (EPA), the U.S. Department of Energy (DOE), the Electric Power Research Institute (EPRI), and their subcontractors have extensively studied the partitioning of Hazardous Air Pollutants (HAPs) at coal-fired utility plants. The majority of HAPs were captured in existing particulate collection devices (ESPs, fabric filters). The exceptions are the elements or compounds denoted as Class III (Hg, Se, HF, HCl), which remain in the vapor state at the ESP or fabric filter exit. Potential HAP regulations could impact the Illinois coal industry.

As a result of the 1990 Clean Air Act Amendments (CAAA), the U.S. EPA revised the human daily mercury intake standard (rfd). Subsequently, the environmental community sued the EPA and the Federal District Court issued an order requiring EPA to propose mercury control regulations by 2003 and to promulgate the regulations by 2004. EPA issued a Mercury Information Collection Request that required utilities to determine the mercury concentration in all coal purchased for one year. In addition, a number of individual boilers were identified that were required to measure the mercury concentration at the inlet and exit of existing control equipment (e.g. ESP's, FGD's, fabric filters, etc). These results have been published and discussed at a number of meetings.

One of the parameters that seem to influence the controllability of flue gas mercury is speciation. The oxidized form is more readily collected in ESP's and wet or dry FGD's. Potential mercury regulations threaten the Illinois coal industry because additional control costs would make switching to Power River Basin coal or natural gas more likely. If a method can be identified to increase the oxidized fraction of flue gas mercury, then the Illinois coal industry would be less affected by mercury regulations.

Some Illinois coals contain high chlorine contents (greater than 0.2%). Coal chlorine is a factor that influences the amount of oxidized mercury that reports to the control equipment; the higher the coal chlorine, the greater the oxidized mercury fractions. A second factor that influences mercury removal is flue gas temperature. The lower the flue gas temperature, the more mercury is removed in the ESP with the fly ash.

The results of this program will provide the Illinois coal industry with valuable information to evaluate no or low cost options to comply with the 2003 proposed regulations.

EXPERIMENTAL PROCEDURES

Two Illinois utility stations agreed to participate with CONSOL on the sampling test program. These stations, identified as sites 1 and 2, were evaluated in 2001. Site visits were conducted by the CONSOL sampling team in early 2001. During the site visits, program requirements were discussed with plant operating personnel, sampling locations were identified, and arrangements were made with plant personnel for assistance during the sampling.

Four tests were conducted at each station. Each test period included simultaneous flue gas sampling at the air heater gas side inlet, air heater gas side outlet, and air heater air side outlet. Gas sampling was conducted at two locations in the air heater gas side exit duct: at a point near the highest measured temperature and at a point near the lowest measured temperature. A diagram showing the sampling locations is presented in Figure 1. These samples were obtained by using the Ontario Hydro sampling train. The Ontario Hydro sampling train is capable of speciating Hg into three fractions; 1) Hg adsorbed on particulate matter, 2) oxidized Hg, and 3) elemental Hg. In addition to flue gas samples, coal and ESP ash samples were obtained during the test period. A diagram showing the Ontario Hydro method is shown in Figure 2.

The Ontario Hydro method is the generally accepted method for speciating mercury in flue gas. This method was developed for use and validated at 250 to 350 degrees F. However, the temperature at the air heater inlet was 600-700 degrees F. Whether the Ontario Hydro method properly speciates mercury at the high temperatures at the air heater inlet is not certain. For the purpose of this report, it will be assumed that the method is speciating properly, and the conclusions drawn will be based on this assumption.

Utility Operating Conditions

The operating staffs at both utilities were asked to maintain steady-state boiler operating conditions for each test period. Site 1 is a 250 MWe PC-fired boiler. Site 2 is a 366 MWe PC-fired boiler. The following tables show the operating parameters maintained at both sites during the sampling programs.

BOILER OPERATING PARAMETERS AT SITE 1

Plant Parameter	Test 1	Test 2	Test 3	Test 4	Average
Megawatts	251	241	232	238	240
Coal Feed, lb/hr (dry)	191300	198100	176700	208800	193725
$\%O_2$ @ Economizer	4.32	3.64	3.73	3.53	3.81

BOILER OPERATING PARAMETERS AT SITE 2

Plant Parameter	Test 1	Test 2	Test 3	Test 4	Average
Megawatts	360	360	360	360	360
Coal Feed, lb/hr (dry)	322800	318500	321600	318200	320275
$\%O_2$ @ Economizer	3.32	3.04	3.08	2.96	3.10

Coal Analyses

The literature suggests that coal composition can influence Hg speciation in the flue gas. Site 1 was firing a 50/50 blend of Illinois and Kentucky coals. Site 2 was firing 100% Illinois coal. Comprehensive coal analyses were completed on daily test coal samples from each plant. The coal analyses from samples collected at the test sites are summarized as follows:

ANALYSES OF COAL FEED SAMPLES AT SITE 1

(Units are % dry basis unless noted)

	Test 1	Test 2 & 3	Test 4	Avg
Volatile Matter	36.19	35.97	36.11	36.09
Ash	8.27	8.68	10.37	9.11
Carbon	74.46	74.08	71.57	73.37
Hydrogen	4.61	4.57	4.43	4.54
Nitrogen	1.44	1.44	1.44	1.44
Oxygen	9.60	9.60	10.62	9.94
Total Sulfur	1.51	1.52	1.47	1.5
Chlorine	0.114	0.109	0.097	0.107
Mercury, ppm	0.09	0.10	0.10	0.10
Heating Value, Btu/lb	13,400	13,354	12,735	13,163

The Hg concentration in the coal showed little variability and averaged 0.10 ppm (µg/g) on a dry basis. Assuming that all the Hg in the coal volatilizes during combustion, the

mercury concentrations would result in a nominal Hg flue gas concentration of ~10µg/m^3. A number of studies have linked the chlorine concentration of the coal with flue gas Hg speciation. The chlorine content of this coal would result in a theoretical flue gas HCl concentration of ~74 ppmv.

ANALYSES OF COAL FEED SAMPLES AT SITE 2
(Units are % dry basis unless noted)

	Test 1	Test 2 & 3	Test 4	Avg
Volatile Matter	41.79	41.54	41.93	41.75
Ash	9.73	9.62	9.62	9.66
Carbon	69.80	69.75	69.71	69.75
Hydrogen	4.95	4.93	4.96	4.95
Nitrogen	1.39	1.42	1.45	1.42
Oxygen	9.72	9.8	9.72	9.75
Total Sulfur	4.24	4.31	4.37	4.31
Chlorine	0.175	0.165	0.172	0.171
Mercury, ppm	0.06	0.06	0.06	0.06
Heating Value, Btu/lb	12,831	12,854	12,827	12,837

The Hg concentration in the coal showed no variability and averaged 0.06 ppm (µg/g) on a whole coal basis. Assuming that all the Hg in the coal volatilizes during combustion, the flue gas mercury concentration would be ~7 µg/m^3. The chlorine content of this coal would result in a theoretical flue gas HCl concentration of ~125 ppmv.

Flue Gas Sampling Data
The key flue gas sampling data for Site 1 is shown in the following tables for each location.

SUMMARY OF FLUE GAS MEASUREMENTS FOR TEST SITE 1
Air Heater Exit, Air Side Measurement

	Test 1	Test 2	Test 3	Test 4
Sample Time, min	120	120	120	120
Bar. Pres., inches of Hg	29.56	29.36	29.25	29.15
Static Pres., inches of H_2O	3.8	3.8	3.8	2.69
% O_2	20.9	20.9	20.9	20.9
% H_2O	1.4	0.0	0.6	1.3
Flue Gas Temp, °F	559	550	550	552
Gas Sample Volume, dry std cubic feet	49.52	56.67	56.59	56.78

Air Heater Inlet, Gas Side Measurement

	Test 1	Test 2	Test 3	Test 4
Sample Time, min	120	120	120	120
Bar. Pres., inches of Hg	29.56	29.36	29.25	29.15
Static Pres., inches of H_2O	-3.70	-3.72	-3.72	-4.82
% O_2	4.4	3.5	3.6	3.0
% H_2O	8.9	8.2	9.0	10.1
Flue Gas Temp, °F	662	643	620	647
Gas Sample Volume, dry std cubic feet	56.88	52.26	46.43	49.20
Particulate Rates: gr/dry standard cubic feet	2.276	2.579	2.974	2.992
% Isokinetic	103	98	97	100

Air Heater Outlet, Gas Side Measurement, Maximum Temperature

	Test 1	Test 2	Test 3	Test 4
Sample Time, min	139	122	121	121
Bar. Pres., inches of Hg	29.56	29.36	29.25	29.15
Static Pres., inches of H_2O	-8.96	-7.15	-7.15	-9.26
% O_2	3.8	5.4	3.5	4.3
% H_2O	8.7	6.3	8.4	9.3
Flue Gas Temp, °F	299	294	290	295
Gas Sample Volume, dry std cubic feet	45.47	38.11	38.62	41.44
Particulate Rates: gr/dry standard cubic feet	2.987	1.167	3.911	3.517
% Isokinetic	103	109	106	104

Air Heater Outlet, Gas Side Measurement, Minimum Temperature

	Test 1	Test 2	Test 3	Test 4
Sample Time, min	141	121	123	106
Bar. Pres., inches of Hg	29.56	29.36	29.25	29.15
Static Pres., inches of H_2O	-8.96	-7.15	-7.15	-9.26
% O_2	5.3	4.9	4.6	4.9
% H_2O	8.2	7.5	7.3	9.0
Flue Gas Temp, °F	265	263	261	265
Gas Sample Volume, dry std cubic feet	51.99	41.61	46.05	47.45
Particulate Rates: gr/dry standard cubic feet	3.411	3.310	3.519	3.581
% Isokinetic	104	101	99	99

These data show excellent reproducibility for each test and location. The data obtained from these samples are used to calculate the Hg concentrations in the flue gas.

The key flue gas sampling data for Site 2 are summarized in the following tables.

SUMMARY OF FLUE GAS MEASUREMENTS FOR TEST SITE 2

Air Heater Exit, Air Side Measurement

	Test 1	**Test 2**	**Test 3**	**Test 4**
Sample Time, min	120	120	120	120
Bar. Pres., inches of Hg	29.40	29.33	29.26	29.27
Static Pres., inches of H_2O	8.64	7.18	7.18	6.10
% O_2	20.9	20.9	20.9	20.9
% H_2O	0.9	1.2	1.2	1.6
Flue Gas Temp, °F	611	606	606	592
Gas Sample Volume, dry std cubic feet	57.00	55.23	52.20	51.91

Air Heater Inlet, Gas Side Measurement

	Test 1	**Test 2**	**Test 3**	**Test 4**
Sample Time, min	121	110	101	116
Bar. Pres., inches of Hg	29.40	29.33	29.26	29.27
Static Pres., inches of H_2O	-6.69	-5.83	-5.83	-5.60
% O_2	4.2	3.9	4.0	3.6
% H_2O	9.3	9.1	11.6	9.8
Flue Gas Temp, °F	677	672	663	674
Gas Sample Volume, dry std cubic feet	63.14	57.18	50.28	58.84
Particulate Rates: gr/dry standard cubic feet	2.050	2.358	2.746	2.501
% Isokinetic	96	97	95	97

Air Heater Outlet, Gas Side Measurement, Maximum Temperature

	Test 1	**Test 2**	**Test 3**	**Test 4**
Sample Time, min	91	114	122	120
Bar. Pres., inches of Hg	29.40	29.33	29.26	29.27
Static Pres., inches of H_2O	-9.13	-8.72	-8.72	-8.72
% O_2	5.0	2.4	2.6	2.6
% H_2O	9.6	8.9	10.7	10.2
Flue Gas Temp, °F	306	316	319	319
Gas Sample Volume, dry std cubic feet	48.36	43.87	41.83	43.89
Particulate Rates: gr/dry standard cubic feet	2.809	1.014	3.610	3.320
% Isokinetic	96	104	104	103

Air Heater Outlet, Gas Side Measurement, Minimum Temperature

	Test 1	Test 2	Test 3	Test 4
Sample Time, min	116	81	102	114
Bar. Pres., inches of Hg	29.40	29.33	29.26	29.27
Static Pres., inches of H_2O	-9.13	-8.72	-8.72	-8.72
% O_2	4.3	3.6	4.4	4.2
% H_2O	9.0	11.0	10.2	9.9
Flue Gas Temp, °F	279	280	287	286
Gas Sample Volume, dry std cubic feet	59.03	38.03	51.35	57.04
Particulate Rates: gr/dry standard cubic feet	3.004	3.622	3.156	2.979
% Isokinetic	98	100	101	97

As with the first plant, these data show consistent duct measurements for the three test days and serve as the basis for the Hg concentration calculations.

RESULTS AND DISCUSSION

Analysis of Hg Oxidation Across Air Heater Systems

The flue gas Hg speciation data indicate that the elemental (Hg^0) and oxidized (Hg^{++}) fractions of the flue gas Hg at the air heater inlet are substantially different from the fractions at the air heater outlet. The air heater inlet Hg speciation estimated from the Ontario Hydro sampling method showed between 60% and 90% elemental Hg. Outlet speciation data showed between 10% and 25% elemental Hg. Particulate Hg was not substantially present at the air heater inlet at either plant; however, at the air heater outlet, substantial (nearly 50%) particulate Hg was measured at Site 1 but not Site 2 (less than 10% particulate Hg).

At the Site 1 air heater outlet, the temperature had a substantial effect on mercury speciation. At the minimum temperature point (265 °F) about 55% of the mercury was particulate mercury; at the maximum temperature point (295 °F) the particulate mercury was about 30% of the total mercury. At site 2, the temperature (285-315 °F) had very little effect on mercury speciation.

Total Hg and speciated Hg at the air heater inlet, air heater outlet (maximum temperature), and air heater outlet (minimum temperature) for Site 1 are summarized as follows:

FLUE GAS Hg SPECIATION AT TEST SITE 1

	Air Heater Inlet	Air Heater Outlet (Max Temp)	Air Heater Outlet (Min Temp)
Particulate, µg/m^3			
Test 1	0.05	0.82	5.15
Test 2	0.06	1.36	7.20
Test 3	1.70	7.07	7.41
Test 4	0.07	6.19	7.05
Average	0.47	3.86	6.70
Oxidized, µg/m^3			
Test 1	2.46	10.56	7.06
Test 2	5.54	4.94	0.66
Test 3	6.74	4.02	1.43
Test 4	3.42	2.33	0.00
Average	4.54	5.46	2.29
Elemental, µg/m^3			
Test 1	8.44	1.10	0.33
Test 2	7.72	0.98	2.56
Test 3	5.62	1.01	0.08
Test 4	8.52	0.43	0.16
Average	7.57	0.88	0.78
Total, µg/m^3			
Test 1	10.95	12.47	12.55
Test 2	13.32	7.29	10.41
Test 3	14.06	12.09	8.92
Test 4	12.00	8.95	7.20
Average	12.58	10.20	9.77

The speciation breakdown at the air heater inlet was 4% particulate, 36% oxidized, and 60% elemental. The air heater outlet (maximum temperature) speciation breakdown was 38% particulate, 54% oxidized, and 9% elemental. The air heater outlet (minimum temperature) speciation breakdown was 69% particulate, 23% oxidized, and 8% elemental.

Total Hg and speciated Hg at the air heater inlet, air heater outlet (maximum temperature), and air heater outlet (minimum temperature) for Site 2 are summarized as follows:

FLUE GAS Hg SPECIATION AT TEST SITE 2

	Air Heater Inlet	Air Heater Outlet (Max T)	Air Heater Outlet (Min T)
Particulate, μg/m^3			
Test 1	0.03	0.44	0.20
Test 2	0.16	0.63	0.34
Test 3	0.03	0.03	0.09
Test 4	0.08	0.01	0.34
Average	0.07	0.28	0.24
Oxidized, μg/m^3			
Test 1	1.80	5.17	4.68
Test 2	0.18	1.57	3.61
Test 3	0.53	4.32	4.14
Test 4	0.62	4.27	3.63
Average	0.78	3.83	4.01
Elemental, μg/m^3			
Test 1	6.03	1.72	1.79
Test 2	8.82	1.63	1.42
Test 3	7.09	0.68	1.29
Test 4	6.77	1.50	2.07
Average	7.18	1.38	1.64
Total, μg/m^3			
Test 1	7.86	7.33	6.67
Test 2	9.16	3.83	5.37
Test 3	7.64	5.03	5.52
Test 4	7.46	5.78	6.04
Average	8.03	5.49	5.90

The speciation breakdown at the air heater inlet was 1% particulate, 10% oxidized, and 89% elemental. The air heater outlet (maximum temperature) speciation breakdown was 5% particulate, 70% oxidized, and 25% elemental. The air heater outlet (minimum temperature) speciation breakdown was 4% particulate, 68%, and 28% elemental.

Mercury Recycle from the Gas Side to the Air Side of the Air Heater

At both sites, measurable concentrations of mercury were found in the air exiting the air heater. Presumably, mercury in the flue gas is absorbing on the air heater surface (or ash deposits on the surface), and then re-evolving from the air heater to be released into the combustion air. This also provides additional indication that the air heater is interacting with the mercury in the flue gas. The total mercury concentration measured in the air stream for each test site is shown as follows.

Hg Measured in the Combustion Air Exiting the Air Heater

	Site No. 1	Site No. 2
Test 1	2.06	0.46
Test 2	0.89	0.33
Test 3	0.15	0.39
Test 4	0.76	0.16
Average	0.85	0.34

ESP Ash Analyses

The ESP ash sample analyses from samples collected at the test sites are summarized as follows.

ANALYSES OF ESP ASH SAMPLES AT SITE 1

	Test 1	Test 2 & 3	Test 4	Avg
Carbon, dry wt%	10.3	5.09	19.37	11.59
Mercury, ppm	0.25	0.14	0.58	0.32

The carbon and Hg concentrations in the ESP ash showed large variability from day to day.

ANALYSES OF ESP ASH SAMPLES AT SITE 2

	Test 1	Test 2 & 3	Test 4	Avg
Carbon, dry wt%	5.36	3.62	3.37	4.12
Mercury, ppm	0.07	0.08	0.06	0.07

The carbon and Hg concentrations in the ESP ash at Site 2 showed less day to day variability than observed at Site 1.

CONCLUSIONS AND RECOMMENDATIONS

In 2001, CONSOL measured the Hg speciation around the air heater at two Illinois coal-fired utilities. The ICCI test program demonstrated that substantial mercury oxidation occurs across the air heater at plants burning Illinois basin coals.

At both locations, the air heater inlet Hg speciation was mostly elemental Hg, while at the air heater outlet oxidized Hg predominated.

Particulate Hg was not substantially present at the air heater inlet at either plant; however, at the air heater outlet, substantial particulate Hg was measured at Site 1 but not Site 2. Site 2 had an average fly ash carbon concentration of 4.1%. Site 1 had an average fly ash

carbon concentration of 11.6%. The difference in fly ash carbon content may impact the particulate mercury concentration – the fraction removed.

The fraction of particulate mercury was substantially affected by the air heater outlet temperature at Site 1; the air heater outlet temperature had a negligible effect on Hg speciation at Site 2. At Site 1, the fly ash captured approximately 32% of the coal mercury. At Site 2 the fly ash captured approximately 11% of the coal mercury. This, again, can be attributed to the change in fly ash carbon content, mentioned above.

A measurable amount of Hg was observed at the air heater air side outlet at both plants, which means that a small portion of the mercury entering the air heater can be recycled back to the boiler.

DISCLAIMER STATEMENT

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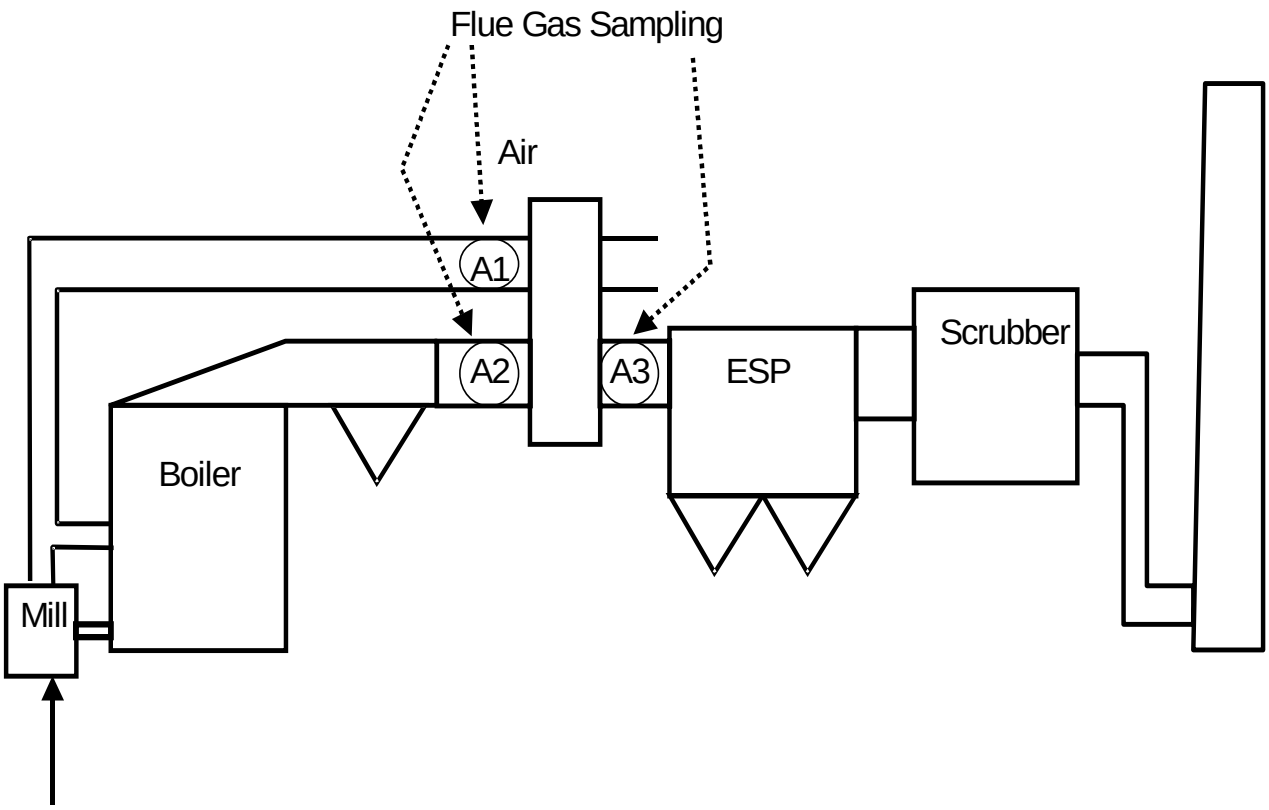

Figure 1. Schematic of Sampling Locations for Test Sites 1 and 2.

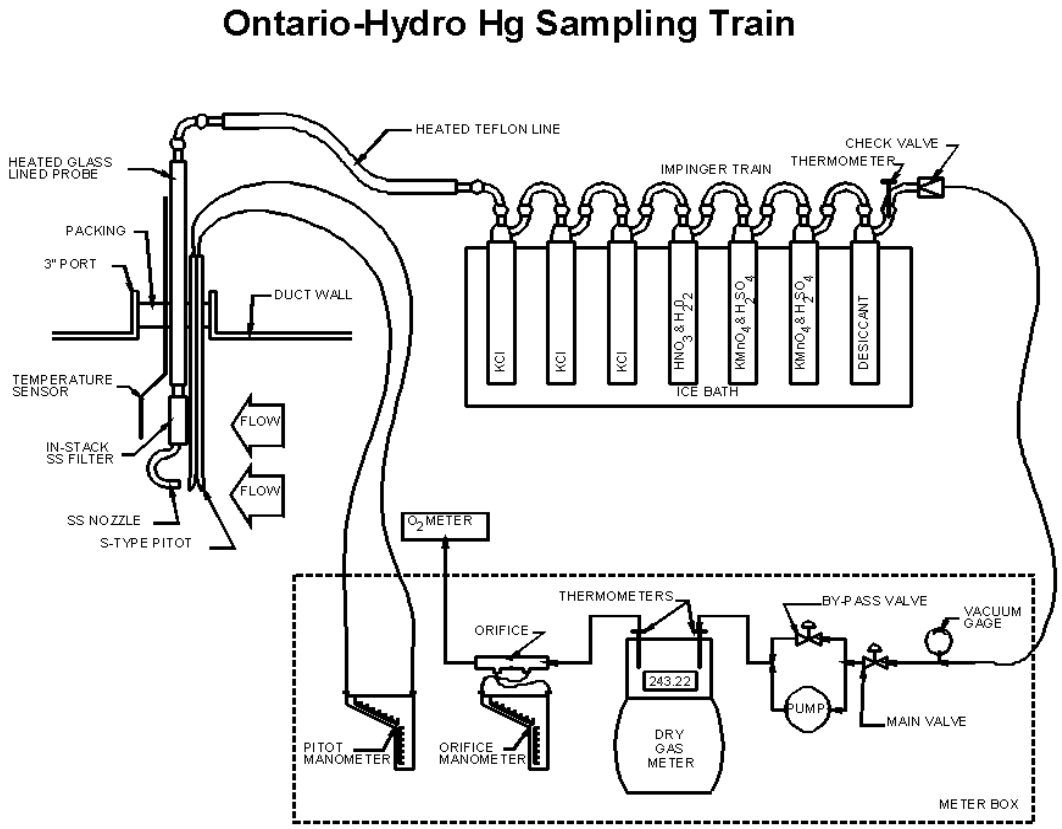

Figure 2. Schematic of Ontario Hydro Method for Mercury Speciation.