

# Canada Wide Standards Mercury Measurement Methodologies for Coal-fired Power Plants

## Paper # 15

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EPRI-EPA-DOE-AW&MA Power Plant Air Pollutant Control “MEGA” Symposium, August 28-31, 2006, Baltimore, Maryland, USA

## ABSTRACT

In support of the proposed Canada Wide Standards (CWSs) for Mercury Emissions from Coal-fired Electric Power generation Plants, the Canadian Council for Ministers of the Environment (CCME) commissioned a study of mercury monitoring options for Canada’s coal-fired power plants. Broad study questions were developed to explore monitoring options. Questions posed were: what to measure, how to monitor, over what averaging timeframe, and how to minimize variability? A review was conducted of the spatial and temporal environmental health impacts and experience with measurement methodologies. CCME and the Canadian Electricity Association (CEA) coordinated a two year program of weekly coal and ash data collection (mass balance) from 20 Canadian power plants. Analyses using these coal and ash data are presented. Additionally, a smaller set of stack CEMS and Ontario Hydro Method stack test data are presented. One of CCME’s goals is to develop predictive models for mercury emissions, and regression analyses were employed to develop statistical models of emission rates (kg/TWh) as a function of coal mercury and chlorine content using Canadian and U.S. data. The most useful model was a statistically significant regression equation for plants with a cold-side ESP, no post-combustion NOx controls, and no post-combustion SOx controls. The study recommended monitoring of total and oxidized mercury, although oxidized emissions would be for information only because the draft CWSs only specify total mercury for determining compliance. To benefit from anticipated performance and cost improvements in CEMS technology, mercury monitoring by mass balance is recommended for Canadian plants until one or two years after U.S. plants are required to have CEMS. Monitoring by CEMS or sorbent trap is recommended for the long run. A 12-month rolling average is recommended with exceedances corrected within 36 months. It is emphasized that this study was commissioned to perform analyses and make recommendations. However these results and recommendations were not intended to be automatically adopted into CCME’s monitoring protocol.

## INTRODUCTION

### STUDY OBJECTIVES

This paper is based on a study performed under contract (project # 369-2006) to the Canadian Council for Ministers of the Environment (CCME) in support of development of a Canada-wide standards (CWSs) mercury monitoring protocol for coal-fired power plants[1]. The study focused principally on answering the following four questions:

- What parameters need to be measured to assess CWSs achievement?
- What are the appropriate measurement methodologies to determine CWSs achievement?
- What is an appropriate averaging time?
- How should variability in measurements be addressed for determining achievement of CWSs?

The technical assessment was designed to address CCME terms of reference for this project, and was comprised of four main components:

1. Review experience with measurement methodologies.
2. Perform statistical analyses of Canadian and U.S. data.
3. Assess and compare the variability and uncertainties of different mercury monitoring methods for determining CWSs achievement.
4. Make recommendations.

CCME works collaboratively to establish national standards, strategies, and objectives such as the proposed *Canada-wide Standards for Mercury Emissions from Coal-Fired Electric Generating Plants* [2-4]. CCME has approved-in-principle CWSs for mercury emitted from coal-fired power plants aimed to reduce emissions approximately 60-90%. The draft CWSs specify annual caps on mass emissions of mercury at the provincial level for existing facilities. For new facilities, the proposed CWSs are that plants achieve either a percent mercury removal or stack emission rate (kg/TWh). As part of the CWSs development, CCME is tasked to develop a monitoring protocol for measurement of mercury from coal-fired power plants. In support of this goal, a uniform data collection program (UDCP) was initiated through a cooperative effort of CCME and the Canadian Electricity Association (CEA) which consisted of a laboratory round robin exercise, and a program to collect and analyze coal and ash samples (mass balance) over approximately a 2-year time period[5, 6]. This study is intended to support the development of CCME's monitoring protocol scheduled for completion by the end of 2006.

### Review of Mercury in the Environment and Human Health Impacts

The purpose here is to briefly summarize the emissions, ecological processes, and impacts of mercury. We focus only on issues that have important implications for measurement (e.g. what to measure; averaging time) of emissions from coal-fired power plants in Canada.

Mercury in the environment comes from biogenic and anthropogenic processes, and results in human health and ecological impacts. The current consensus is that most impacts result

(indirectly) from mercury emitted into the air. Air emissions come from various processes that then deposit in soils and bodies of water.

Mercury emitted from coal-fired power plants is commonly classified into three forms: elemental - Hg<sup>0</sup>, oxidized mercury - Hg<sup>II</sup>, and particulate bound mercury - HgP. In 1999, it was estimated that U.S. coal-fired power plants emitted 41 tonnes of mercury from coal containing 68 tonnes (i.e. ~ 40% native removal efficiency)[7]. Of this 41 tonnes, 59% was Hg<sup>0</sup>, 39% Hg<sup>II</sup>, and 2% HgP.

The chemical form emitted is one important factor that affects the spatial scale of mercury deposition[8-11]. Hg<sup>II</sup> and HgP are more easily scrubbed out of the atmosphere and thus tend to deposit more locally, while Hg<sup>0</sup> oxidizes in the atmosphere to form Hg<sup>II</sup> and tends to result in longer range deposition. The distance from the source to deposition location is also a function of meteorology, which is an important factor in the conversion of Hg<sup>0</sup> to Hg<sup>II</sup> in the atmosphere. While currently there are concerns that Hg<sup>II</sup> could deposit more locally under certain meteorological conditions, the balance of evidence indicates that local deposition occurs less often and substantial deposition occurs over long distances[7, 12, 13]. For example, remote Canadian lakes that are 1,400 km away from industrial activity show elevated levels of mercury[14]. The worldwide distribution of emission sources and exposure via consumption of non-local fish are additional factors that confirm mercury as a global problem[15, 16].

In water, a small portion (~1%) of Hg<sup>II</sup> transforms to methyl mercury, an organic compound. Methyl mercury is the only known mercury compound that bio-accumulates, and is considered to have the largest human health impact via exposure by eating fish[7, 14, 17-19]. Methyl mercury is a mutagen (changes genetic information), teratogen (causes abnormal cell masses during fetal growth), and carcinogen (promotes cancer)[12, 14, 18]. In the U.S., mercury impacts on children's health alone are estimated to result in lost productivity of US\$8.7 billion annually, of which US\$1.3 billion is attributable to mercury emissions from American coal-fired power plants[20]. For comparison, EPA estimates the incremental annual cost of its clean air mercury rule will range from US\$0.16 billion in 2010 to US\$1.04 billion in 2020[12].

The temporal and spatial scales of mercury in the environment are important to highlight because they have implications for regulations and emissions monitoring. An indicator of the temporal scale of mercury in the environment is its residence time in environmental reservoirs[7, 12, 14, 19]

**Table 1.** Typical residence times for mercury in the environment

<b>Environmental Reservoir</b>	<b>Approximate Residence Time (half life)</b>
Atmosphere	0.02 to 1 year (1 week to 1 year)
Soils	1,000 years
Freshwater	0.3 to 5 years (3 to 60 months)
Ocean water	2,000 to 3,000 years
Ocean sediments	1,000,000 to 300,000,000 years

Another important indicator of time scale is provided by the results of EPA's ecological models which predict that new steady state concentrations of mercury in sediments (in response to changes in anthropogenic emissions) can be reached in 5 to 30 years for most freshwater bodies (but as little as 1 year for some) [12].

Considering the time and spatial scales of emissions and impacts, and without yet considering monitoring methods or costs, there are three key implications for monitoring and regulation that are supported by the current scientific knowledge:

1. Because the chemical species of mercury is critical in determining its fate and transport in the environment and ultimately human health impacts, monitoring of speciated mercury is important, not just total mercury. Hg<sup>0</sup> and Hg<sup>II</sup> dominate emissions from coal-fired power plants, hence these two fractions are the most important.
2. Monitoring and regulation should focus on long term emissions such as rolling averages of 1 year or more. Hourly, daily, and even monthly emissions are of much less importance from a regulatory or environmental management perspective – e.g., an exceptionally low or high daily value implies little incremental benefit or impact on human or ecological health. This is in contrast to other air pollutants such as SO<sub>2</sub> which can trigger reactions in asthmatics over a 15 minute time period, or PM<sub>2.5</sub> which is shown to cause increased mortality when daily concentrations increase.
3. Because the spatial and temporal scales of mercury impacts are very large, emissions from sources other than power plants and from other countries are important to Canada. Hence Canada's efforts to reduce mercury releases from other domestic sources, and to negotiate internationally for reductions elsewhere, are well founded.

A final remark on this brief review of environmental and health impacts is that it is based on *current* science as described in the literature. Mercury is a highly active area of research and some new findings in coming months or years could alter recommended monitoring strategies for coal fired power plants.

## REVIEW OF MONITORING METHODOLOGIES

An important part of the study scope was to overview and compare methods for directly or indirectly monitoring stack emissions of mercury, with the focus being issues related to measurement variability and accuracy. Sources of information included study team members' experience, literature from U.S. and Canadian non-peer reviewed sources (e.g. EPRI, EPA, Environment Canada), and peer-reviewed literature (searched Elsevier Science Direct database and Google Scholar with a variety of key words). In this study we reviewed the following methods for directly or indirectly monitoring stack emissions of mercury:

- Predictive models, such as the statistical models developed later in this paper[21, 22].
- Mass balance: measuring mercury input through the fuel and output in residues (e.g. bottom ash, fly ash, scrubber liquor), then determining stack emissions of total mercury by difference[23-26].
- Wet chemistry measurements at the stack by EPA Method 101A, Method 29, and Ontario Hydro Method[27, 28]

- Stack measurements using Sorbent traps[29-32]
- Stack measurements using CEMS (continuous and semi-continuous)[33-35]

A summary of study findings is shown in Table 2 (see the original study for more details of the monitoring methodology review). Capital costs<sup>a</sup> are annualized at an 8% discount rate over 20 years, which is equivalent to an annual cost of about 10% of the capital cost (e.g. \$250k capital adds \$25k to the annual cost). These costs are very much approximations and contain a mix of “hard” costs to the plant to purchase equipment and services, and in-house costs (e.g. to collect coal and ash samples). It is noted, however, that based on a study of mercury control costs for Canadian power plants, even the highest cost for monitoring appears to be only a small fraction of control costs; control costs were generally estimated at over \$1 million per year per generating unit (1999 Canadian dollars)[36].

**Table 2: Summary and comparison of mercury monitoring methods.**

Option	Accuracy:		Precision	Speciation	Approximate annual cost
	at low levels <sup>b</sup> ( $< 0.5 \mu\text{g}/\text{m}^3$ )	representative of long term average			
Predictive models	very low	low	low	HgT	<\$25k
Mass balance (coal & ash)	low	med	med	HgT	\$25k (assume monthly sampling @ 1 week composite sample per month)
EPA Method 101A	med-high	low	med-high	HgT	\$20k
EPA Method 29	med-high	low	med-high	HgT	\$20k
Ontario Hydro Method stack tests	med-high	low	high	Hg0, HgII, HgP	\$25k (one triplicate set/year)
Sorbent trap	high	med-high (dependent upon how conducted)	high	Hg0, HgII (can include HgP, or only Hg0)	\$50k (assume \$100k capital + lab costs + annual OH certification)
CEMS (continuous and semi- continuous)	med-high	high	med-high	Hg0, HgII (by difference)	\$75k (assume \$250k capital + annual OH certification)

A few assessments of variability of mercury emissions have been found in the literature and discussed herein. Chu et al (2003)[35] assessed variability in mercury emissions with one month studies on 16 different coal-fired boilers firing: lignite-Powder River Basin blend, bituminous, Powder River Basin alone, and western bituminous. They measured and analyzed hourly and

<sup>a</sup> Costs in this study are in 2005 Canadian dollars unless otherwise specified.

<sup>b</sup> Descriptions of accuracy at low levels for stack test methods have been modified from the original study.

daily averages with CEMS validated by Ontario Hydro Method tests. For a typical unit, about ¼ of the hourly measurements over a one month period were  $<1.5 \mu\text{g}/\text{m}^3$ , and ¼ were  $>3.8 \mu\text{g}/\text{m}^3$ . This is a strong indicator of temporal variation over one hour timeframes, and it was reported that such variation was typical for all of the 16 units tested. Furthermore, for some plants the daily averages varied by a factor of five over the one month of testing. This study illustrates the large amount of short term variability in emissions over hours and days (even though the study is titled “longer-term”). Three additional published EPRI reports[37-39] that would likely be helpful in assessing variation in mercury emissions and removal efficiencies were not reviewed due to limited study funds. Analyses of emission and mercury removal variability obtained from EPA’s web site highlight the variability of removal efficiency as a function of coal quality and emission control systems[21, 22].

## DATA ANALYSIS

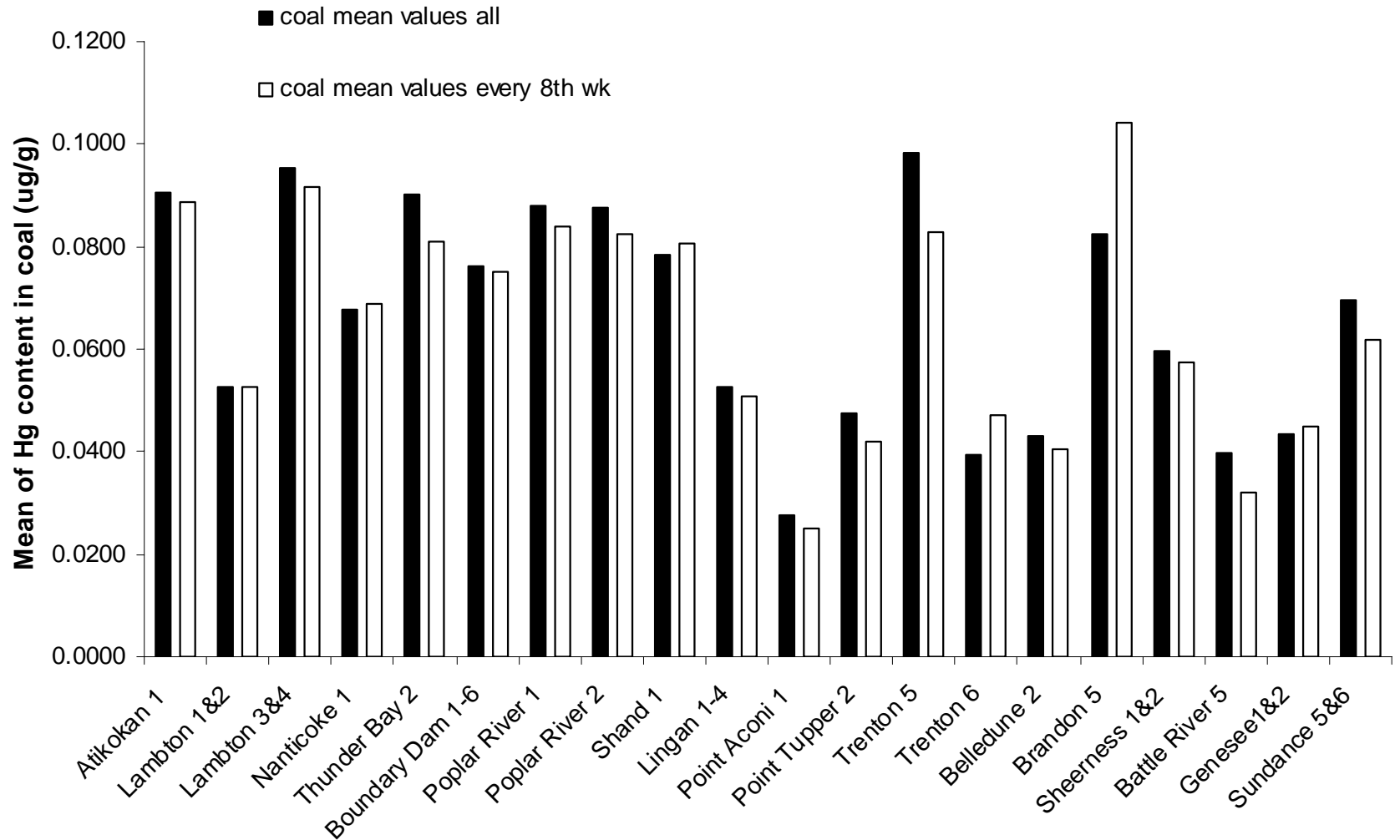
Coal and ash data collected per the CCME protocol are available from the CEA web site ([www.ceamercuryprogram.ca](http://www.ceamercuryprogram.ca)). Weekly sampling was conducted for up to seven quarters for each unit from 2002 to 2005 (although most sampling occurred in 2003-04). The calculated descriptive statistics included means, number of samples (n), coefficient of variation (COV), and range for all data that was provided. Values below the detection limit were assigned ½ the value of the detection limit. A few outliers were removed based upon a value that exceeded other values by an order of magnitude, without conducting any rigorous statistical tests for outliers. Complete tables of descriptive statistics for all data are available in the original project report[1].

To assess the effect of different sampling frequencies on variability in measured coal and ash mercury content, descriptive statistics were recalculated based on hypothetical sampling frequencies of every 4 weeks and every 8 weeks (i.e. ignoring data for 3 out of 4 weeks, or 7 out of 8 weeks). The results are plotted in Figures 1 and 2 for coal means and COV comparing all measurements with those from every 8<sup>th</sup> week (see the full study report for every 4<sup>th</sup> week results). A somewhat surprising insight from these calculations is that, in general, the mean and COV values differ little for the less frequent sampling. However, it is critical to note that these data are based on weekly composite samples collected at a minimum of 3 different days of a given week (as per UDCP protocol). The hourly and daily variability in coal mercury content and stack emissions that has been found in various test programs must be assumed to be occurring in these plants as well[35, 40]. A possible reason for the apparent low variability in coal mercury data when comparing weekly results with bimonthly (i.e. every 8<sup>th</sup> week) results, is that the hourly and daily temporal variations simply are not capable of being captured in weekly composite fuel samples.

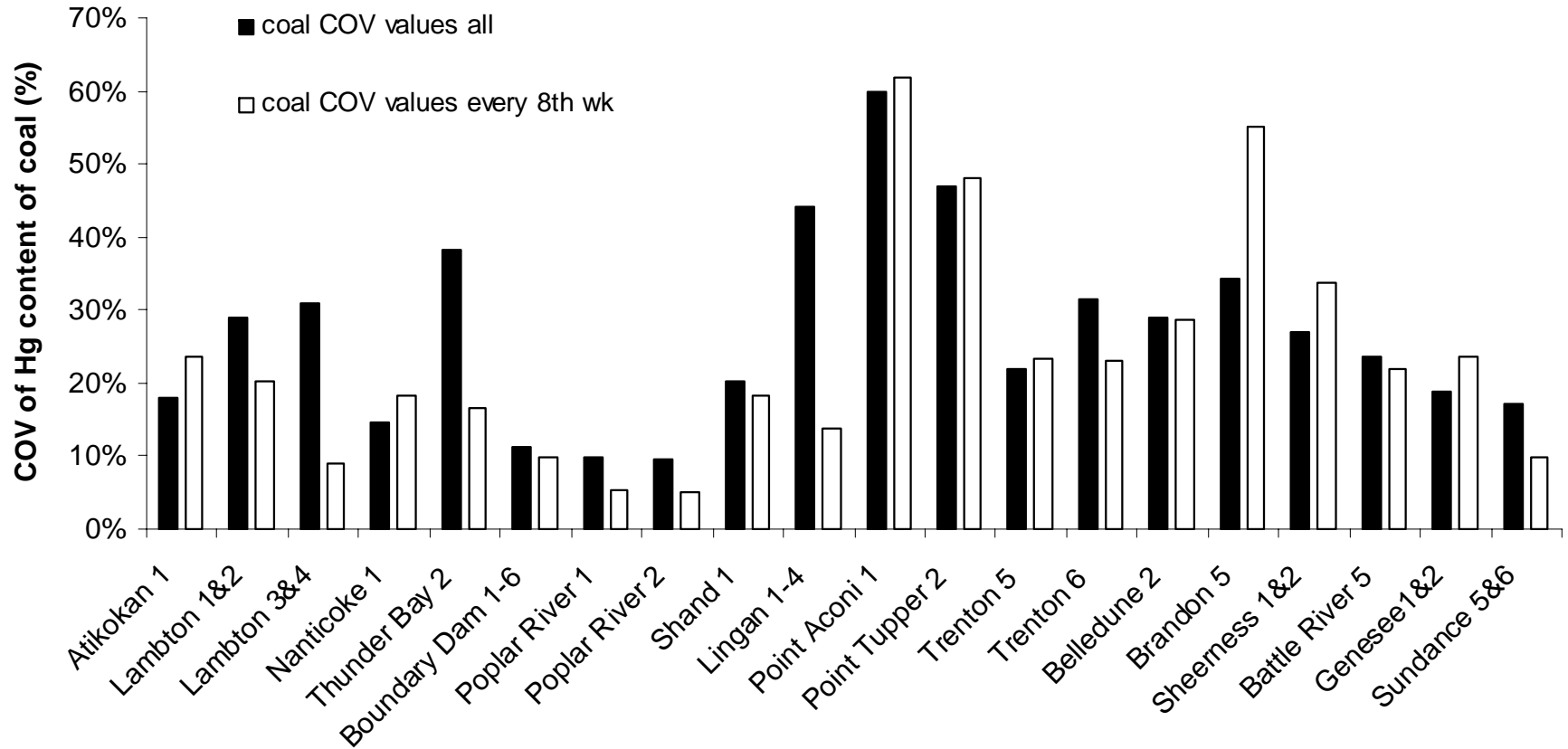
Coal and ash data were used to estimate mercury emissions (kg/TWh) by mass balance. These calculations are summarized in Table 3.

Ontario Hydro Method tests and mass balance data and calculations for selected Canadian power plants were also provided by CCME. All data provided are summarized in Table 4, except two test reports which were not included based on reported QA problems.

**Figure 1: Comparison of mean values of mercury in coal ( $\mu\text{g/g}$ ) as calculated by two methods: the solid bars use all values (i.e. collected every week up to 7 quarters); the white bars use only the values from every 8<sup>th</sup> week to simulate the effect of bimonthly sampling (i.e. one week every two months).**



**Figure 2: Comparison of coefficient of variation (COV) values for mercury in coal ( $\mu\text{g/g}$ ) as calculated by two methods: the solid bars use all values (i.e. collected every week up to 7 quarters); the white bars use only the values from every 8<sup>th</sup> week to simulate the effect of bimonthly sampling (i.e. one week every two months).**





**Table 3: Summary of Canadian power plant mercury emissions as calculated by mass balance using UCDP coal and ash data.**

		Atikokan 1	Lambton 1&2	Lambton 3&4	Nanticoke 1	Thunder Bay 2	Sheerness 1&2	Battle River 5	Genesee 1&2	Sundance 5&6	Brandon 5
		Mean	Mean	Mean	Mean	Mean	Mean	Mean	Mean	Mean	Mean
Province		ON	ON	ON	ON	ON	AB	AB	AB	AB	MB
Coal rank - primary		Lig	Bit/Sub	Bit/Sub	Bit/Sub	Lig	Sub	Sub	Sub	Sub	Sub
Coal rank - secondary		none	Pet Coke	Pet Coke	none	Sub	none	none	none	none	Lig
Boiler Type		Pulv	Pulv	Pulv	Pulv	Pulv	Pulv	Pulv	Pulv	Pulv	Pulv
Particulate control device		cold ESP	cold ESP	cold ESP	cold ESP	hot ESP	cold ESP	cold ESP	cold ESP	cold ESP	cold ESP
SO2 control device		none	none	FGD wet	none	none	none	none	none	none	none
NOx control device		LNB	LNB	SCR	LNB	none	none	none	none	none	none
<b>Mercury Balance</b>											
Mercury in (fluid bed limestone)	kg	-	-	-	-	-	-	-	-	-	-
Mercury In (Coal)	kg	68.653	147.067	249.236	605.759	70.264	93.943	37.289	93.943	162.078	18.957
Mercury Out (Fly Ash)	kg	1.735	68.814	55.890	388.688	0.119	27.376	9.197	27.376	40.442	2.256
Mercury Out (Bottom Ash)	kg	0.074	2.096	6.768	2.044	0.664	0.465	0.474	0.465	2.348	0.123
Mercury Emissions (Stack)	kg	66.845	76.157	186.579	215.026	69.481	66.103	27.618	66.103	119.289	16.578
Mercury Capture	%	2.634	48.216	25.140	64.503	1.114	29.635	25.934	29.635	26.400	12.548
<b>Annualized Rates</b>											
Annualized Generation	GWh	949	4495	5413	19844	1462	6376	2848	6376	5351	559
Annualized Mercury Input	kg/yr	39.23	98.04	166.16	524.00	56.21	125.26	49.72	125.26	216.10	18.96
Annualized Mercury Emission	kg/yr	38.20	50.77	124.39	172.02	55.59	88.14	36.82	88.14	159.05	16.58
Calculated Mercury Emission Rate	kg/TWh	40.27	11.29	22.98	8.67	38.03	13.82	12.93	13.82	29.73	29.66

		Belledune 2	Lingan 1 to 4	Point Aconi 1	Point Tupper 2	Trenton 5	Trenton 6	Boundary Dam 1 to 6	Polar River 1	Polar River 2	Shand 1
		Mean	Mean	Mean	Mean	Mean	Mean	Mean	Mean	Mean	Mean
Province		NB	NS	NS	NS	NS	NS	SK	SK	SK	SK
Coal rank - primary		Sub/PetCoke	Bit/Sub	Pet Coke	Bit/Sub	Bit/Sub	Bit/Sub	Lig	Lig	Lig	Lig
Coal rank - secondary		none	Pet Coke	Bit/Sub	Pet Coke	Pet Coke	Pet Coke	none	none	none	none
Boiler Type		Pulv	Pulv	Fluid Bed	Pulv	Pulv	Pulv	Pulv	Pulv	Pulv	Pulv
Particulate control device		hot ESP	cold ESP	Fab Filter	cold ESP	cold ESP	cold ESP	cold ESP	cold ESP	cold ESP	cold ESP
SO2 control device		FGD wet	none	limestone	none	none	none	none	none	none	sorb inj
NOx control device		LNB	OFA	none	OFA	none	none	OFA	OFA	LNB	LNB
<b>Mercury Balance</b>											
Mercury in (fluid bed limestone)	kg	-	-	5.308	-	-	-	-	-	-	-
Mercury In (Coal)	kg	89.976	116.111	17.131	32.000	53.355	28.383	505.842	225.575	247.502	140.183
Mercury Out (Fly Ash)	kg	8.355	1.085	18.288	0.166	3.975	3.364	42.544	8.901	27.712	8.367
Mercury Out (Bottom Ash)	kg	0.437	0.040	0.404	0.054	0.008	0.036	4.605	0.160	0.120	0.119
Mercury Emissions (Stack)	kg	81.184	114.986	3.747	31.780	49.372	24.983	458.694	216.514	219.670	131.696
Mercury Capture	%	9.772	0.969	83.300	0.688	7.465	11.979	9.321	4.017	11.245	6.054
<b>Annualized Rates</b>											
Annualized Generation	GWh	3958	4685	1343	1177	944	1130	6115	2141	2308	2055
Annualized Mercury Input	kg/yr	59.98	77.41	11.42	25.60	35.57	18.92	55.15	70.25	71.48	54.58
Annualized Mercury Emission	kg/yr	54.12	76.66	2.50	25.42	28.21	14.28	305.80	144.34	146.45	105.36
Calculated Mercury Emission Rate	kg/TWh	13.67	16.36	1.86	15.42	29.88	12.63	50.01	67.43	63.44	51.28

**Table 4: Summary of Ontario Hydro Method mercury stack emission rates and mass balance calculations.**

		Atikokan 1	Lambton 4	Nanticoke 5	Nanticoke 7	Nanticoke 7	Nanticoke 7	Nanticoke 7	Nanticoke 7	Nanticoke 3	Thunder Bay 2&3
Province		ON	ON	ON	ON	ON	ON	ON	ON	ON	ON
Unit rating	MW	230	505 net	564	564	564	564	564	564	564	163
Coal rank - primary		Lig	Bit/Sub	Bit/Sub	Sub/Bit	Sub/Bit	Sub/Bit	Sub/Bit	Sub/Bit	Bit/Sub	Lig/Sub
Coal rank - secondary		none	Pet Coke	none	none	none	none	none	none	none	Sub
Boiler Type		Pulv	Pulv	Pulv	Pulv	Pulv	Pulv	Pulv	Pulv	Pulv	Pulv
Particulate control device		cold ESP	cold ESP	cold ESP	cold ESP	cold ESP	cold ESP	cold ESP	cold ESP	cold ESP	hot ESP
SO2 control device		none	FGD wet	none	none	none	none	none	none	none	none
NOx control device		LNB	SCR	LNB	SCR	SCR	SCR	SCR	SCR	LNB	none
<b>Test conditions</b>											
Test date(s)	month-year	Sep-98	Sep-03	Apr-??	Mar-04	May-04	Apr-05	Apr-05	Apr-05	May-05	??-98
Electricity generation	gross MW	219	521	536	510	510	511	511	506	508	310
Capacity factor (assume when needed)	%	95%		95%							95%
<b>Coal</b>											
Moisture	%	32.47	5.97	21.2	22.1	22.9	24.4	23.9	23.9	25.1	30.350
Chlorine	ug/g dry	16.333	859.0	442.000	413	486	224	244	244	1248	12.833
Mercury	ug/g dry	0.1016	0.104	0.056	0.069	0.073	0.067	0.066	0.066	0.062	0.096
Chlorine (calc)	ug/g wet	11.0	808	348	322	374	169	186	186	934	8.9
Mercury (calc)	ug/g wet	0.0686	0.0978	0.0442	0.0538	0.0560	0.0503	0.0504	0.0504	0.0463	0.0667
<b>Hg Stack Emissions</b>											
Particulate	mg/s	0.011	<0.002	0.537	0.166	0.008	0.075	0.190	0.190	0.133	0.003
Oxidized	mg/s	0.190	0.068	0.733	1.084	1.164	1.110	0.890	0.890	1.000	0.089
Elemental	mg/s	3.160	0.137	0.226	0.078	0.202	0.120	0.095	0.095	0.108	2.250
Total	mg/s	1.662	0.205	1.496	1.329	1.374	1.305	1.175	1.175	1.240	2.342
Total	kg/TWh	27.4	1.41	10.0	9.2	9.6	9.2	8.4	8.4	8.8	27.2
<b>Hg Mass Balance</b>											
out (ash + residue + stack) / in (coal)	%	131%	125%	113%	103%	153%	112%	113%	113%	106%	133%

		Brandon 5	Belledune 2	Grand Lake	Lingan 1&2	Point Aconi 1	Point Tupper 2	Trenton 5	Trenton 6	Bound Dam 1&2	Bound Dam 5
Province		MB	NB	NB	NS	NS	NS	NS	NS	SK	SK
Unit rating	MW	105	480	57	150 each	165	150	150	160	62 net each	139 net
Coal rank - primary		Sub	Sub/PetCoke	indigenous	Bit/Sub	Pet Coke	Bit/Sub	Bit/Sub	Bit/Sub	Lig	Lig
Coal rank - secondary		Lig	none	none	Pet Coke	Bit/Sub	Pet Coke	Pet Coke	Pet Coke	none	none
Boiler Type		Pulv	Pulv	Pulv	Pulv	Fluid Bed	Pulv	Pulv	Pulv	Pulv	Pulv
Particulate control device		cold ESP	hot ESP	cold ESP	cold ESP	Fab Filter	cold ESP	cold ESP	cold ESP	cold ESP	cold ESP
SO2 control device		none	FGD wet	none	none	limestone	none	none	none	none	none
NOx control device		none	LNB	none	OFA	none	OFA	none	none	OFA	OFA
<b>Test conditions</b>											
Test date(s)	month-year	Apr-05	Sep-04	Oct-03	Jun-02	Nov-01	Nov-01	Jul-03	Jun-03	Aug-04	Aug-04
Electricity generation	gross MW	102	488	57	318	161	159	147	167	108	144
Capacity factor (assume when needed)	%										
<b>Coal</b>											
Moisture	%	13.99	9.43	4.70	11.260	9.51	9.06	6.82	6.27	33.8	34.0
Chlorine	ug/g dry	32.0	215	289	136	156	317	1040	1267	17.0	14.0
Mercury	ug/g dry	0.050	0.033	0.623	0.079	0.032	0.056	0.093	0.051	0.110	0.124
Chlorine (calc)	ug/g wet	27.5	194.7	275.4	120.7	141.2	288.3	969.1	1187.6	11.3	9.2
Mercury (calc)	ug/g wet	0.0430	0.0299	0.5937	0.0701	0.0290	0.0509	0.0867	0.0478	0.0728	0.0818
<b>Hg Stack Emissions</b>											
Particulate	mg/s	0.00064	0.019	0.0083	0.006	0.003	0.003	0.003	0.003	0.00	0.00
Oxidized	mg/s	0.0703	0.094	3.508	1.408	0.042	0.528	0.736	0.317	1.88	2.06
Elemental	mg/s	0.5533	0.478	1.008	0.967	0.019	0.425	0.461	0.275	9.57	10.08
Total	mg/s	0.6244	0.5917	4.5250	2.381	0.064	0.956	1.200	0.594	11.45	12.14
Total	kg/TWh	22.0	4.4	285.8	26.9	1.4	21.6	29.4	12.8	55.1	58.4
<b>Hg Mass Balance</b>											
out (ash + residue + stack) / in (coal)	%	125%	115%	116%	108%	107%	109%	96%	92%	108%	96%

## STATISTICAL ANALYSIS OF ICR AND CCME DATA

### The Usefulness and Limitations of Predictive Statistical Models

Ideally, the movement of mercury through the power generation process could be modeled completely from principles of physics and chemistry. While advances have been made, the development of widely validated models has yet to be achieved[26, 41]. Hence there is potentially a role for statistical analysis to reveal some quantitative relationships. It is to be emphasized that statistical models are only possible when there is sufficient understanding of the underlying physical processes so that hypotheses can be made about which variables are potentially important.

In our view, there are three specific uses for the statistical models developed here:

1. To predict changes in mercury emission rates for a specific plant when changes are planned that affect coal properties (e.g. mercury or chlorine content of purchased coal) or plant equipment (e.g. new ESP).
2. To estimate plant emissions when direct measurements have not been made.
3. To assess quantitatively the mercury emissions performance (e.g. the removal rate) of a particular power plant as compared to other plants of similar designs.

Outside of these specific uses, we view statistical models as having limited usefulness.

### Regression Model Results<sup>c</sup>

Linear ordinary least square (OLS) statistical equations (i.e. “models”) were developed of the following form:

$$(\text{Hg kg/TWh emission rate}) = a + b1 * \ln(\text{coal Hg content}) + b2 * \ln(\text{coal Cl content}) + \text{error term}$$

In this analysis, the “explained” (dependent) variable is:

HgT emission rate: kg/TWh electrical output

Two “explanatory” (independent variables) were modelled<sup>d</sup>:

Coal mercury content: ppm,wet (ppmw)

Coal chlorine content: ppm,wet (ppmw)

Wet concentrations of mercury and chlorine were chosen because the ICR data were all presented in these units (i.e. not dry basis). When using the UDCP Canadian data, concentrations were converted from dry to wet basis.

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<sup>c</sup> The terminology used in this section is standard and can be defined in just about any statistics book.

<sup>d</sup> It should be noted that these two parameters (coal mercury and chlorine content) clearly have the most impact on mercury emissions for a given plant configuration, but there are other variables that have less, but still significant, impact on emission such as sulphur content, moisture, heating value, and process temperatures. It was outside the scope of this study to perform an exhaustive analysis.

The OLS analysis tool in Microsoft Excel was used to calculate the constants  $a$ ,  $b1$ , and  $b2$ . All models were performed with log transformed independent variables (i.e. just by taking the natural log of the value), and some were additionally performed with the dependent variable log transformed. Acceptance criteria were defined as P-values (a measure of the statistical significance)  $<0.1$  for  $a$ ,  $b1$ , and  $b2$  and F-statistic  $<0.1$  (another indicator of a statistically valid model). Additionally, a visual review of the residual plots (i.e. model error terms) was performed to look for gross asymmetries.

Groupings (i.e. stratification) and independent variables for statistical analyses were chosen based important parameters from the literature, particularly what is known about the EPRI statistical analyses and other engineering literature[22, 26, 41].

All groupings chosen for our analyses are shown in Table 5, along with the modeling results – i.e. whether the model result was “valid” or “not valid” by applying the stated criteria. All “not valid” models were characterized by a low number of data points.

**Table 5: Regression model summary**

Unit type	PM control device	Post-combustion NOx control	SO <sub>2</sub> control equipment	Primary fuel	Model result
<i>pulverized coal</i>	<i>cold ESP</i>	<i>none</i>	<i>none</i>	<i>all</i>	<i>valid</i>
pulverized coal	cold ESP	none	none	bituminous	not valid
pulverized coal	cold ESP	none	none	subbituminous	not valid
pulverized coal	cold ESP	SCR	none	all	not modeled; insufficient data
<i>pulverized coal</i>	<i>cold ESP</i>	<i>none</i>	<i>wet scrubber</i>	<i>all</i>	<i>valid</i>
pulverized coal	hot ESP	none	wet scrubber	all	not valid
pulverized coal	hot ESP	none	none	all	not valid
pulverized coal	baghouse	none	Spray dryer	all	not valid
pulverized coal	baghouse	none	none	all	not valid
pulverized coal	all	all	all	lignite	not valid
pulverized coal	all	all	all	subbituminous	not valid
pulverized coal	all	all	all	bituminous	not valid
fluidized bed	all	all	all	all	not valid

Only two regression models were assessed to be valid (those shown in *bold italics* in Table 5) using the ICR data. These model results met the aforementioned criteria for P-values and F-statistics, and resulted in fairly high R<sup>2</sup> values as shown in Table 6. The R<sup>2</sup> value is a measure of how much explanatory power a model has – i.e the two valid models for predicting mercury emission rates describe 69% and 81% of the variation.

A third model with ICR data was developed using the *log transform* of the dependent variable as shown in Table 6. This model form is useful because the  $b1$  and  $b2$  regression coefficients are “elasticity” values<sup>e</sup> to predict changes in the dependent variable. Thus the model in Table 6 for

<sup>e</sup> “elasticity” values are a ratio of the % change in one value to the % change in another value.

units with cold ESP, no post-combustion NO<sub>x</sub> control, and no SO<sub>2</sub> control equipment predicts the following:

- a +1% change in coal chlorine content results in a -.23% change in Hg kg/TWh.
- a +1% change in coal mercury content results in a +1.31% change in Hg kg/TWh

Just one Canadian power station (Lambton) has a cold ESP with wet scrubber (i.e. the 2<sup>nd</sup> line in Table 6), but several stations have cold ESP with no post-combustion NO<sub>x</sub> controls and no SO<sub>2</sub> control equipment.

A second predictive model was developed for units with cold ESP, no post-combustion NO<sub>x</sub> controls, and no SO<sub>2</sub> control equipment, this time using pooled Canadian and ICR data (i.e. all available data combined into one data set). This should, ideally, improve the model with more observations and better statistical power. This turned out to be the case as the confidence intervals did reduce, the P-values were much lower, and the R<sup>2</sup> value increased from 69% to 74%. Hence the “pooled” statistical model listed in Table 6 was used to predict emission rates as shown in Table 7.

## Summary and Comparison of Emission Rates

A summary and comparison of mercury emission rates for Canadian power plants is shown in Table 8 as determined by three methods: UDCP mass balance, Ontario Hydro Method stack tests, and regression model prediction. It is of interest that there is general agreement between the three methods. However, it should be cautioned that these three methods inherently have important differences in predicting emission rates. The Ontario Hydro Method is considered the “gold standard,” but it is still just a snap shot that represents emissions over a narrow window of time. The mass balance is representative of emissions at the time samples are taken and within the limits of accuracy of that method. The regression models, by definition, are created from a combination of mass balance and Ontario Hydro Method data sets, so it is no surprise that they generally agree.

The correlation coefficient between the Ontario Hydro Method tests and UDCP mass balance estimates of stack emissions is 0.77. This is based on comparing the first two columns of Table 8. This R<sup>2</sup> indicates good agreement<sup>f</sup>, although this does not resolve the potential limitations of mass balance estimates (capture of short term variations, representative of long term emissions, lack of speciation).

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<sup>f</sup> There are not exact criteria for what comprises “good” or “poor” correlation. In this study, r<sup>2</sup> values of about 0.75 are considered “good”, while those below 0.5 are considered “poor.”

**Table 6: Statistically significant regression models using ICR data sets, and pooled ICR with Canadian data sets.**

data set	boiler type	PM control	post-comb NOx control	SO2 control equipment	coefficients			lower 90% confidence interval			upper 90% confidence interval			maximum P-value	F-statistic	R <sup>2</sup>
					a	b1	b2	a	b1	b2	a	b1	b2			
ICR only*	pulv	cold ESP	none	none	109.13	-3.91	24.03	70.27	-7.52	14.66	147.99	-0.29	33.41	0.0786	0.0031	0.69
ICR only*	pulv	cold ESP	none	wet scrub	159.36	-8.34	40.69	101.96	-15.72	18.98	216.76	-0.95	62.40	0.0721	0.0163	0.81
pooled*	pulv	cold ESP	none	none	124.84	-5.17	26.53	99.99	-7.01	18.72	149.70	-3.32	34.34	0.0001	0.000003	0.74
ICR only**	pulv	cold ESP	none	none	7.77	-0.23	1.31	5.73	-0.42	0.82	9.81	-0.04	1.80	0.0493	0.0023	0.70

\* model form:  $\text{HgT stack kg/TWh elec} = a + b1 * \ln(\text{ppmw Cl}) + b2 * \ln(\text{ppmw Hg})$

\*\* model form:  $\ln(\text{HgT stack kg/TWh elec}) = a + b1 * \ln(\text{ppmw Cl}) + b2 * \ln(\text{ppmw Hg})$

**Table 7: Predicted stack emissions of total mercury using regression models from both ICR-only data and pooled data.**

		Atikokan 1	Nanticoke 5	Brandon 5	Grand Lake	Lingan 1&2	Point Tupper 2	Trenton 5	Trenton 6	Bound Dam 1&2	Bound Dam 5
Province		ON	ON	MB	NB	NS	NS	NS	NS	SK	SK
Unit rating	MW	230	564	105	57	150 each	150	150	160	62 net each	139 net
Coal rank - primary		Lig	Bit/Sub	Sub	indigenous	Bit/Sub	Bit/Sub	Bit/Sub	Bit/Sub	Lig	Lig
Coal rank - secondary		none	none	Lig		Pet Coke	Pet Coke	Pet Coke	Pet Coke	none	none
Boiler Type		Pulv	Pulv	Pulv	Pulv	Pulv	Pulv	Pulv	Pulv	Pulv	Pulv
Particulate control device		cold ESP	cold ESP	cold ESP	cold ESP	cold ESP	cold ESP	cold ESP	cold ESP	cold ESP	cold ESP
SO2 control device		none	none	none	none	none	none	none	none	none	none
NOx control device		LNB	LNB	none	none	OFA	OFA	none	none	OFA	OFA
<b><u>Predicted emissions from ICP model</u></b>											
predicted stack emission rate	kg/TWh	35.4	11.3	20.6	74.6	26.5	15.4	23.5	8.4	36.7	40.3
lower 90% confidence interval	kg/TWh	12.9	-19.5	-0.8	20.4	-4.8	-16.0	-17.3	-27.6	13.7	16.8
upper 90% confidence interval	kg/TWh	57.8	42.1	41.9	128.9	57.8	46.9	64.3	44.3	59.8	63.7
<b><u>Predicted emissions from pooled model</u></b>											
predicted stack emission rate	kg/TWh	41.4	11.8	24.2	82.0	29.6	16.6	24.4	7.6	42.8	46.9
lower 90% confidence interval	kg/TWh	33.0	0.5	17.8	50.8	16.6	4.5	6.0	-6.6	34.0	37.5
upper 90% confidence interval	kg/TWh	49.7	23.1	30.6	113.1	42.5	28.7	42.9	21.8	51.7	56.4

**Table 8: Summary and comparison of mercury stack emission rates (kg/TWh)**

	UDCP mass balance	Ontario Hydro stack test	Pooled regression model
Atikokan 1	40.3	27.4	41.4
Lambton 1&2	11.3	-	-
Lambton 3&4	23.0	1.4	-
Nanticoke 1	8.7	-	-
Nanticoke 3	-	8.8	-
Nanticoke 5	-	10.0	11.8
Nanticoke 7	-	9.1	-
Thunder Bay 2&3	38.0	27.2	-
Sheerness 1&2	13.8	-	-
Battle River 5	12.9	-	-
Genesee 1&2	13.8	-	-
Sundance 5&6	29.7	-	-
Brandon 5	29.7	22.0	24.2
Belledune 2	13.7	4.4	-
Lingan 1&2	-	26.9	29.6
Lingan 1 to 4	16.4	-	-
Point Aconi 1	1.9	1.4	-
Point Tupper 2	15.4	21.6	16.6
Trenton 5	29.9	29.4	24.4
Trenton 6	12.6	12.8	7.6
Boundary Dam 1 to 6	50.0	-	-
Bound Dam 1&2	50.0	55.1	42.8
Bound Dam 5	50.0	58.4	46.9
Polar River 1	67.4	-	-
Polar River 2	63.4	-	-
Shand 1	51.3	-	-

## STUDY RECOMMENDATIONS

Study recommendations were made based on how to best determine compliance with CWSs over the near term (present through 2010) and beyond. The recommendations are made with the aim to reduce variability (improve precision), and also to maximize accuracy. It should be noted that these recommendations are not made based on the U.S. requirements (e.g. the recommendation to eventually install CEMS or STM does not mean we recommend adoption of U.S. CEMS standards for certification and reporting of emissions). All recommendations made apply equally to existing plants and new plants.

**Recommendation: monitor Hg<sup>0</sup> and Hg<sup>II</sup> stack emissions.** Both species are essential considering human health impacts and ecological processes, and both are substantial components of emissions from coal-fired power plants. This recommendation is somewhat different than the emphasis in the U.S. which does not consistently require Hg<sup>II</sup> monitoring. Nonetheless, the processes that mercury is presently understood to follow in the environment that lead to health

impacts are dependent upon the chemical species, not just total mercury. Reporting of HgII would be for information only because the draft CWSs only specify HgT for determining compliance.

**Recommendation: monitor Hg0 and HgII by CEMS or STM, certified by Ontario Hydro Method stack tests, as the long term goal.** It is clarified that CEMS include those that monitor on a continuous or semi-continuous basis. Additionally, estimating HgII emissions by difference between total gaseous mercury and Hg0 is considered sufficient. The rationale for this recommendation is in part determined by elimination of the basic methods that were summarized in Table 2:

- Predictive models are characterized by large uncertainties for assessing achievement of CWSs (but are useful for other reasons described elsewhere in this paper). Large uncertainties should not be considered sufficient to protect public health and ecological integrity.
- Ontario Hydro Method tests alone provide the best “snap shot” of plant emissions, but cannot capture hourly and daily variability in emissions in a cost-effective manner. Nor can this method cost effectively capture representative emission rates on time scales of a year or more. Nonetheless, it is an invaluable method as the “gold standard” to ascertain speciated emission rates and to certify the accuracy of other measurement methods.
- Method 101A and Method 29 wet chemistry measurements have similar detection limits<sup>9</sup> than the Ontario Hydro Method, but do not provide speciation measurements. Method 101A does not provide significant cost savings relative to the Ontario Hydro Method, but Method 29 is an economical method for measuring total mercury emissions for plants that need to measure stack emissions of the other metals.
- Coal and ash mass balance is generally characterized by relatively large uncertainties, even though fairly consistent and accurate results appear to be achievable at some plants. This method also has the limitation that mercury species cannot be measured, nor can short term variations (hourly, daily) in emission rates. Although long term variations are most critical from an ecological and human health impact view, it cannot be ruled out that methods that do not measure short term variations might have cumulative errors in determining long term averages.

Hence, the remaining available options are CEMS or STM. An important consideration is that future concentrations of mercury in exhaust stacks should be lower as plants implement emission controls. Therefore low detection limit methods are to be emphasized (i.e. a method that works well during baseline characterization may be less effective once mercury controls are implemented).

**Recommendation: bi-monthly (i.e. one week every 2 months) sampling, analysis, and reporting of mercury content of coal (but not ash) following UDCP procedures.** This would involve collecting samples on 3 separate days for the week that sampling is performed. The analyses in this report (Figures 1 and 2) show that average results changed little when the average

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<sup>9</sup> The same laboratory instrumentation is used (CVAA), so any differences in detection limit will be a function of the sampling (e.g. duration) and the analytical preparations (e.g. aliquots used).



of samples collected every week are compared with the average of samples collected every 8<sup>th</sup> week (i.e., ignoring 7 out of every 8 weeks). Monitoring mercury content in coal is useful to explain and validate any changes in emissions over time. Increased sampling frequency should be considered for specific plants that experience large variability in results, or when critical changes are made (e.g. procurement of coal from new sources).

Collection and analysis of ash is not viewed as essential to determine CWSs compliance, as the primary method is recommended to be CEMS or STM. There may be other reasons to monitor mercury content in ash and other residues such as (a) concerns that focus on the fate of disposed ash and residue[42] which will have increasing mercury content as controls are installed, and (b) to understand and assess plant mercury control equipment performance.

**Recommendation: monitor a rolling average up to a maximum of 3 years to assess CWSs achievement, with a condition that corrective action plans are implemented in the event that the CWSs annual caps are exceeded in any 12 month time period.** This recommendation is based on the *current* science that reveals the smallest time frame for mercury environmental and health processes is on the order of 3 years<sup>h</sup>. This recommendation is made with the intent to allow individual plants flexibility to achieve and maintain mercury emission limits in a cost-effective manner. It is emphasized that this study was not exhaustive, and that this recommendation is *not* made with the intent to change the proposed CWSs which are based on annual emission caps.

**Recommendation: plants with low annual emissions of mercury.** Another issue is that of specific plants with low estimated annual emissions. The EPA has annual allowance thresholds of 9 and 29 lbs (approximately 4kg and 13 kg) that exempt plants from CEMS requirements and specify alternate requirements such as semi-annual or annual Ontario Hydro Method stack tests. Similar exemptions are recommended for CCME.

**Recommendation: interim monitoring requirements.** The dilemma with the above set of recommendations is that presently neither CEMS nor STM are fully commercial, reliable, and proven to be robust over sufficient time frames. The expectation is that this will occur based on technological gains likely to be forced by the EPA regulations. As a result, *interim measures* are recommended as follows for all plants subject to CWSs requirements:

- Bi-monthly (i.e. at least one week out of every 2 months) sampling and analysis of *coal and ash* as per UDCP procedures. Increased sampling frequency should be considered for specific plants that experience large variability in results, or when critical changes are made (e.g. procurement of coal from new sources, major plant upgrades expected to affect mercury capture, etc.).
- Annual triplicate stack tests by Ontario Hydro Method of at least 2 hour duration for each test.

These interim monitoring requirements are recommended to remain until CEMS and STM methods become fully commercial. The definition of “fully commercial” is difficult to define, but it is intended here to mean that at least a few (say, 3 or more) manufacturers offer systems that are

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<sup>h</sup> Specifically, the EPA modeling that shows new steady state environmental levels of mercury in freshwater sediments can be achieved in 5 to 30 years, and as little as 1 year in some cases.

fully certified to EPA requirements and have demonstrated reliable operation for roughly one year or more. On the basis that U.S. coal-fired power plants are presently required to install CEMS (or STM as an allowable alternate) by the end of 2009, we recommend that CCME consider lagging the U.S. timeline by a year or two to take advantage of expected technical improvements and cost reductions. Therefore it is recommended to consider CEMS or STM be installed for reporting CWSs achievement by the end of 2010 or 2011. In the interim, the coal and ash mass balance methods as per UDCP procedures are recommended.

**Recommendations for further work** to address information gaps related to mercury measurements and monitoring include:

- Continued or increased Canadian participation in CEMS and STM demonstration programs.
- CEMS and STM detailed study to review and compare system accuracy (i.e. monitors, calibration systems, laboratory analyses, and data systems working together), costs, reliability, and maintainability. A focus on how methods will perform at decreasing concentrations must be emphasized since stack emissions are expected to decline as mercury control technologies are implemented. The expected stack concentrations of mercury can be estimated for all Canadian plants, as well as quantifying known interfering species (e.g. SO<sub>2</sub>). Thus a CEMS/STM study could focus on identifying specific systems expected to be most effective on Canadian power plants. Such a study would provide specific guidance on which systems to demonstrate on Canadian power plants, firing Canadian coals.
- Additional work on predictive models with more robust and detailed estimates of emission factors (e.g. testing additional explanatory variables and different statistical models), and removal rates (i.e. % removal in addition to kg/TWh rates). This would include purchasing EPRI reports to review and build upon this previous work.
- Continued or increased comprehensive testing of representative Canadian units, to include monitoring by selected CEMS or STM systems over periods of weeks to months. The goals of such testing could include: (a) assess emission reductions as well as CEMS/STM performance at low levels, (b) detailed comparisons of different methods (e.g. mass balance compared to CEMS over long time periods), or (c) trial tests of emission reduction techniques.

## **ACKNOWLEDGMENTS**

The authors would like to acknowledge contributions by CCME's mercury monitoring working group for their helpful critical review and feedback on the project report.

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## ACRONYMS & ABBREVIATIONS

AA – atomic absorption

ASTM – American Society for Testing and Materials

BACTEA – best available control technology economically achievable

CCME – Canadian Council of Ministers of the Environment

CEA – Canadian Electricity Association

CEMS – continuous emission monitoring system(s)

COV – coefficient of variation (mean divided by standard deviation)

CVAAS – cold vapor atomic absorption spectroscopy

CVAFS – Cold vapor atomic fluorescence spectroscopy

CWSs – Canada Wide Standards

EC – Environment Canada  
EPA – U.S. Environmental Protection Agency  
EPRI – Electric Power Research Institute  
ESP – electrostatic precipitator  
FAMS – flue gas absorbent mercury speciation  
FGD – flue gas desulphurization  
Hg0 – elemental mercury  
HgII – oxidized mercury  
HgP – particulate bound mercury  
HgT – total mercury  
ICR – U.S. mercury Information Collection Request program  
n – number of samples (observations)  
PM – particulate matter  
STM – sorbent trap method  
TWh – terawatt hours of electricity generation (gross) =  $10^{12}$  watt hours  
UDCP – uniform data collection program (under CCME)  
XRF – X-ray fluorescence

Mercury can harm pregnant women and put infants and children at risk of developmental problems. The calculations used at the time, however, accounted for how pollution-control equipment at coal plants would reduce emissions of particulate matter and other harmful substances that come out of smokestacks, in addition to mercury. Republican President Donald Trump's EPA said in 2018 it had reviewed the justification for the rule and believed it was inappropriate to have included the benefits of reducing emissions other than mercury. It proposed withdrawing the justification. Thursday's announcement... But the U.S. coal industry has railed against the mercury rule, and blamed it for putting hundreds of coal-fired power plants out of business in recent years. Coal-fired-derived flue gas mercury measurement mainly includes Ontario Hydro Method, semicontinuous emission monitor, and Appendix K. The Ontario Hydro Method (OHM) is the standard method of measuring and speciating mercury in flue gas, and its solutions were analyzed using a Leeman Labs Hydra AA. The semicontinuous emission monitor (Hg SCEM) uses a gold trap to collect the mercury from the flue gas before analysis with an atomic fluorescence detector. We also measured the mercury emissions by OHM, CMM, SCEM, and Appendix K in two plants. Jiang Wu, Yuying Du, Weiguo Pan (2009) Study on different measurement methods of mercury emission in the coal-fired power station. The final rule allows power plants to submit certain emissions reports as PDFs until July 1, 2018, while EPA works to put a single e-reporting system in place for MATS. The final rule also clarifies two mercury measurement quality assurance instructions. EPA continues to review comments on the other issues that were part of the MATS e-reporting streamlining proposal. Read the final rule. December 19, 2014 "EPA is proposing a technical corrections memo on the Mercury and Air Toxics Standards for power plants that would resolve conflicts between preamble and regulatory text and clarify some of the language in the regulatory text. The public will have 30 days to review and comment on the proposal after publication in the Federal Register.