Mercury Emissions Capture Efficiency with Activated Carbon Injection at a Russian Coal-Fired Thermal Power Plant

Scientific Report

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Disclaimer

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Acronyms and Abbreviations

AAS	Atomic absorption spectroscopy
AC	Activated carbon
ACAP	Arctic Council Action Plan
ACI	Activated carbon injection
CCR	Coal combustion residuals
CFR	United States Code of Federal Regulations
CO	Carbon monoxide
CTRL	Chronic toxicity reference level
DAF	Dilution attenuation factor
DOE	United States Department of Energy
EPRI	Electric Power Research Institute
ESP	Electrostatic precipitator
h	Hour
Hg	Mercury
Hg(0)	Elemental mercury
Hg(II)	Oxidized (+2) mercury
IOC	Zelinsky Institute of Organic Chemistry
ISTC	International Science and Technology Center (Moscow)
kg	Kilogram
kg/h	Kilogram per hour
kg/t	Kilogram per metric ton
kV	Kilovolt
L/S	Liquid-to-solid ratio
lb	Pound
lb/MMacf	Pounds (of AC) per million actual cubic feet of flue gas
LEAF	Leaching Environmental Assessment Framework
LSP	Liquid-solid partitioning
m	Meter
m/s	Meter per second
m^3	Cubic meter
m^3/h	Cubic meter per hour
mA	Milliampere
MCL	Maximum Contaminant Level
MCLG	Maximum Contaminant Level Goal
mg	Milligram
mg/m^3	Milligrams per cubic meter

mg/t	Milligrams per metric ton
mL/g	Milliliter per gram
MMacf	Million actual cubic feet
MSW	Municipal Solid Waste
MW	Megawatt
MW_e	Megawatt electric
ng	Nanogram
Nm^3/h	Normal cubic meter per hour
NO_X	Nitrogen oxides
NRMRL	USEPA National Risk Management Research Laboratory
NSDWR	National Secondary Drinking Water Regulations
OH	Ontario-Hydro Method
OSWER	USEPA Office of Solid Waste and Emergency Response
PAC	Powdered activated carbon
PM	Particulate matter
ppm	Parts per million
PTFE	Polytetrafluoroethylene
RfD	Reference dose
rpm	Revolutions per minute
RSD	Risk specific dose
SCR	Selective catalytic reduction
SO_2	Sulfur dioxide
TC	Total carbon
TCLP	Toxicity Characteristic Leaching Procedure
TPP	Thermal Power Plant
TT	Treatment technique
U.S.	United States
UNEP	United Nations Environment Programme
USEPA	United States Environmental Protection Agency
VI	Voltage-current
VTI	All-Russia Thermal Engineering Institute
ZIOC	Zelinsky Institute of Organic Chemistry
$\mu g/m^3$	Micrograms per cubic meter

Table of Contents

List of Tables

List of Figures

1 Executive Summary

Coal combustion is a major source of anthropogenic mercury (Hg) emissions. The United Nations Environment Programme (UNEP) estimates that coal combustion contributes about 24% of global anthropogenic Hg emissions. On an individual country basis, the largest emitters from this category are China, India, and the United States (U.S.). Meanwhile, the Arctic Council has found that within the eight Arctic countries, the Russian Federation (Russia) and the U.S. contribute the bulk of Hg emissions from power plants.

The purpose of this report is to present and interpret the results of a collaborative Hg test program designed to demonstrate the effectiveness of injection of activated carbon (AC) sorbents in controlling Hg emissions at a Russian coal-fired power plant. AC sorbents have been extensively studied for control of Hg in the U.S. and they are currently used at many U.S. coal-fired power plants. However, until now, Hg emission control using AC, has never been, to our knowledge, demonstrated in Russia.

The project testing was conducted by the All-Russia Thermal Engineering Institute (VTI) and the Zelinsky Institute of Organic Chemistry (ZIOC). The overall project was led by the U.S. Environmental Protection Agency (USEPA) in collaboration with UNEP and the Swedish Environmental Research Institute (IVL), with funding administered by the International Science and Technology Center (ISTC). The UNEP funds were sent directly to VTI.

Sorbents are best used with particulate emission collectors, such as fabric filters (FF) or electrostatic precipitators (ESPs). Gas phase Hg is adsorbed onto the surface of the powdered AC sorbent and subsequently removed by the downstream particulate control device. The captured AC and fly ash mixture may be either stored in a landfill or, in some countries, sold for beneficial re-use such as cement or concrete production. As shown in the results, adding a halogen such as bromine to the AC sorbent can enhance its effectiveness in controlling Hg.

Because Hg is an element that can neither be created nor destroyed, only transferred among media, the project also studied the leaching potential of selected constituents [Hg, arsenic (As), and selenium (Se)] as contained in three particulate fly ash residuals (baseline residual, i.e., no sorbent used; residual with use of standard AC sorbent; and, residual with use of brominated AC sorbent).

Testing was conducted at the 8 megawatt (MW), 940,000 cubic meter/hour (m^3/h) pilot plant facility at the 300 MW Cherepetskaya Thermal Power Plant (TPP) located approximately three hours by car south of Moscow. Coal from the Kuznetsk region (located east of Novosibirsk), the most abundantly used coal in Russia, was used for the tests.

Findings and Conclusions – Hg Emission Reduction

1. The emission tests indicated that under baseline conditions, i.e., without the use of sorbents, approximately 40% of the Hg was captured in the ESP on the fly ash.
2. The emission tests indicated that the use of standard AC with an injection rate of 160 milligrams per cubic meter (mg/m^3) [10 pounds of AC per million actual cubic feet of flue gas (lb/MMacf)] resulted in a range of capture efficiencies of total Hg removal in the ESP of 67% to about 80%.

3. The emission tests indicated that the use of brominated AC with an injection rate of up to 110 mg/m^3 (7 lb/MMacf) resulted in Hg emission capture efficiencies of up to 90%.

4. The range of 67% to 80% Hg removal from the flue gas achieved with the use of standard AC necessitated an AC injection rate of 160 mg/m^3 to achieve the higher removal. An injection rate of only about 32 mg/m^3 with the use of brominated AC was needed to achieve a comparable level of Hg removal.

5. These emission control efficiency results using Kuznetsk coal are similar to those obtained in earlier U.S. studies using U.S. coals.

Findings and Conclusions – Leaching Studies

Note: *Leach testing focused on the Leaching Environmental Assessment Framework (LEAF) test methods. These methods have been shown in previous studies to provide more data and better insight than the USEPA's Toxicity Characteristic Leaching Procedure (TCLP) in processes that influence the rate and extent of leaching of mercury (Hg), arsenic (As), and selenium (Se) from fly ash and AC mixes.*

1. The LEAF tests on concentrations of Hg, As, or Se, in each of the three fly ash residuals (baseline; standard AC; and, brominated AC) did not equal or exceed contaminant limits in USEPA's TCLP.

2. The LEAF tests indicated that Hg is strongly bound to the surface of the particles (either fly ash particles or sorbent particles). For all three types of fly ash residuals, none were found to exceed USEPA's Hg standard for drinking water quality.

3. The LEAF tests indicated that leachate concentrations of As from both baseline fly ash and sorbent-containing fly ashes did exceed USEPA's As drinking water standard by an order of magnitude. The LEAF results are considered a source term for use in fate and transport modeling to determine environmental release. In fate and transport modeling, attenuation and dilution are considered along with the specifics of how the ash is managed. Therefore, further modeling of As would be needed to determine potential impact on human health.

4. The LEAF tests indicated that the Se concentrations in leachate for baseline fly ash were below the USEPA's Se drinking water standard, except at the very highest pH values, where it was exceeded slightly. For standard AC fly ash wastes the drinking water standard was exceeded slightly at the lower and higher end pH values. For brominated AC, the concentrations were slightly above the drinking water standard at only at the highest pH values.

5. In such cases where, following fate and transport modeling, the predicted concentrations exceed the MCL, consideration should be given, where appropriate, to treatment/storage measures such as a lined landfill, that could address any concern owing to leaching potential.

6. This study indicated that pH is a key factor affecting As and Se leaching from fly ash. These results using the LEAF method are similar to those obtained in previous U.S. studies for Hg, As, and Se constituents from the three types of fly ash wastes that were tested.

2 Introduction

This project testing has been conducted by the All-Russia Thermal Engineering Institute (VTI) and the Zelinsky Institute of Organic Chemistry (ZIOC) under the project leadership of the United States Environmental Protection Agency (USEPA) in collaboration with the United Nations Environment Programme (UNEP) and the Swedish Environmental Institute (IVL). Funding has been administered through the International Science and Technology Center (ISTC) in Moscow.

Coal combustion is recognized by the UNEP and the Arctic Council as a major anthropogenic source of global Hg emissions. According to UNEP's most recent estimate (2013),[1] Hg emissions from coal combustion are estimated to comprise about 24% of the global anthropogenic Hg emission inventory. According to the Arctic Council Action Plan (ACAP) to Eliminate Pollution of the Arctic Hg Project, the United States (U.S.) and Russia (within the eight Arctic countries) contribute the bulk of such emissions[2]. In the U.S., concurrent with development of the USEPA's regulation to limit emissions of Hg and other toxic air pollutants from U.S. coal-fired power plants [3], the USEPA, U.S. Department of Energy (DOE) and others (e.g., Electric Power Research Institute (EPRI), private sector) have successfully demonstrated several approaches for controlling power plant Hg emissions[4-10].

One such approach involves the treatment of flue gas by injecting powdered sorbent materials. The most widely studied and used of the sorbent materials is powdered AC – a process referred to as AC injection (ACI). The ACI technology can use conventional powdered AC or brominated AC (i.e., AC that has been pre-treated to add bromine species to the carbon surface prior to injection). The AC captures gaseous Hg through physical and/or chemical reactions on the surface of the carbon. Conventional AC can be used effectively with coals and process configurations that naturally produce oxidized forms of Hg [i.e., coals that have high levels of halogens, such as chlorine; and plants that are equipped with selective catalytic reduction (SCR) reactors for control of oxides of nitrogen (NO_X)]. Brominated AC is used for facilities that are firing coals with lower levels of native halogen (i.e., low chlorine coals) that produce mostly elemental Hg vapor. The surface bromine species facilitate capture by oxidizing the elemental Hg so that it can more easily bind to the particle surface. The Hg-containing carbon is then captured by the plant's particulate control device, e.g., an electrostatic precipitator (ESP), and the resultant captured particulate material is then disposed of appropriately, e.g., in a landfill, or is sold for beneficial re-use, e.g., cement production.

Since the ACI technology had not been previously evaluated at a Russian power plant using Russian coals, the goal of this project was to demonstrate the effectiveness of the AC sorbents in controlling Hg emissions from a slip stream at a coal-fired power plant in Russia. The power plant fired the most abundantly used coal in Russia – Kuznetsk coal (from the Kuznetsk region). Kuznetsk coal is a low sulfur, high ash content coal with a heating value near that of lignite. The goal was to evaluate the use of ACI technology at a pilot plant that uses an ESP for particulate matter (PM) control and to compare the results with those from earlier studies in the U.S. (using U.S. coals). Because Hg cannot be destroyed but only be transferred among media, the project also studied the leaching potential of selected constituents [Hg, arsenic (As), and selenium (Se)] as contained in three particulate fly ash wastes (baseline waste, i.e., no sorbent used; standard AC waste; and, brominated AC waste). Testing was conducted at the 8 megawatt (MW) (40,000 m^3/h) pilot plant facility located at the 300 MW Cherepetskaya Thermal Power Plant (TPP) located three hours south of Moscow, Russia.

3 Description of the Cherepetskaya pilot plant

The pilot plant at Cherepetskaya TPP has been used for investigations of the performance of ESPs with various designs of discharge and collecting electrodes as well as investigations of power supply and rapping system modes. As illustrated below, the pilot plant has been installed at the bypass of ESP #4 of Unit #5. It consists of two parallel experimental ESPs, and each ESP contains two electric fields. Experimental ESP1 (experimental ESP) has been used for tests conducted during this study. A slip-stream of flue gas is pulled from the plant's flue gas duct located before the air heater or from the inlet gas duct of ESP #4. The slip stream is then treated through the experimental scale ESPs and treated flue gas is then returned into the outlet gas duct of ESP #4. Captured fly ash is deposited in the unit ESP hoppers using a hydro ash removal system. The Cherepetskaya pilot plant is located adjacent to boiler #4 of the plant. Photos of the pilot plant are shown as Figures 1 and 2.

Figure 1. Power supply of one section of experimental ESP.

Figure 2. Top of experimental ESP.

Figure 3 shows the configuration of the pilot plant facility. The experimental ESP is connected to the inlet and outlet gas ducts of ESP #4. The boiler flue gas reaches the experimental ESP when the gate valve is opened with the fan operating. The carbon injection lance and distribution system is located upstream of the experimental ESP. Hg was sampled at sampling points 1 and 2.

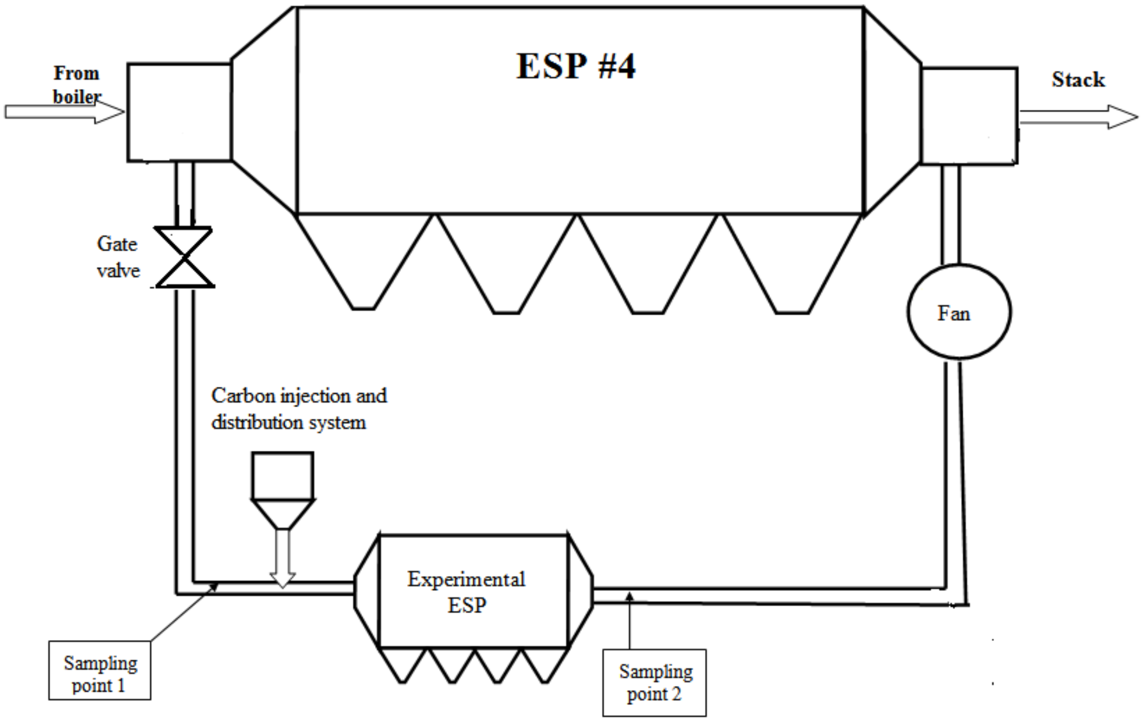


Figure 3. Configuration of the pilot plant for study of Hg emissions control with AC injection.

3.1 Characteristics of the experimental ESP

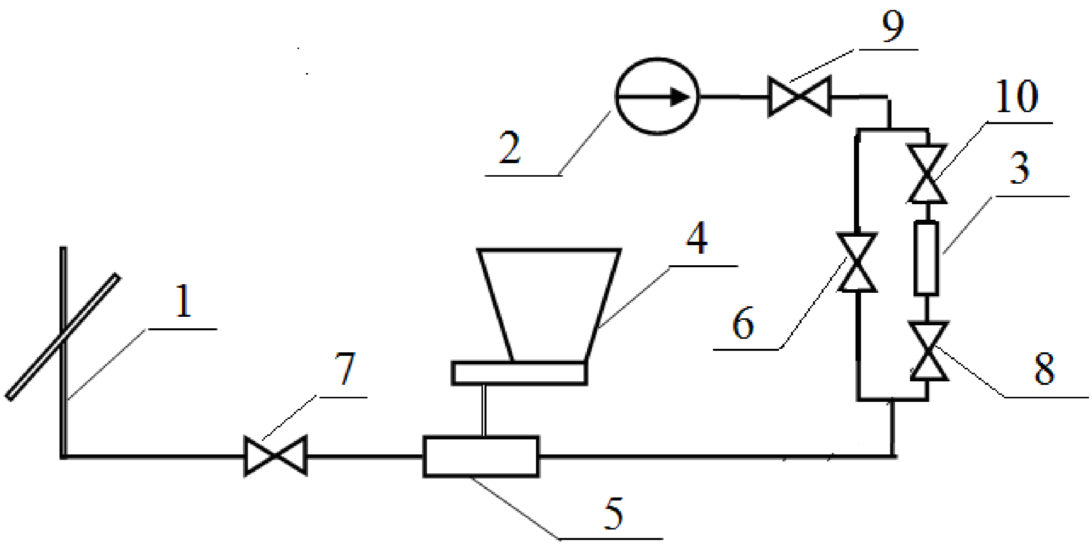
The experimental ESP consists of two independent housings, each having two sections. Experimental ESP’s technical parameters are given in Table 1.

3.2 Description of the injection and distribution system

The schematic of the injection and distribution system is shown in Figure 4. AC is loaded into the hopper of feeder (4). The feeder auger takes AC from the hopper and supplies it to the ejector (5). The AC injection rate is adjusted by varying the auger rotation speed. Parameter F03 of feeder power supply determines the screw rotation speed and, therefore, the ACI rate. Air from compressor (2) is mixed with AC as it enters the ejector. Valve (8) controls the air flow rate measured by rotameter (3). The mixture of carbon and air passes from the ejector to the distribution lattice (1), located in the experimental ESP inlet gas duct. A photo of the injection and distribution system is shown in Figure 5. A photo of the air-AC pipe lance inserted into the gas duct is shown in Figure 6.

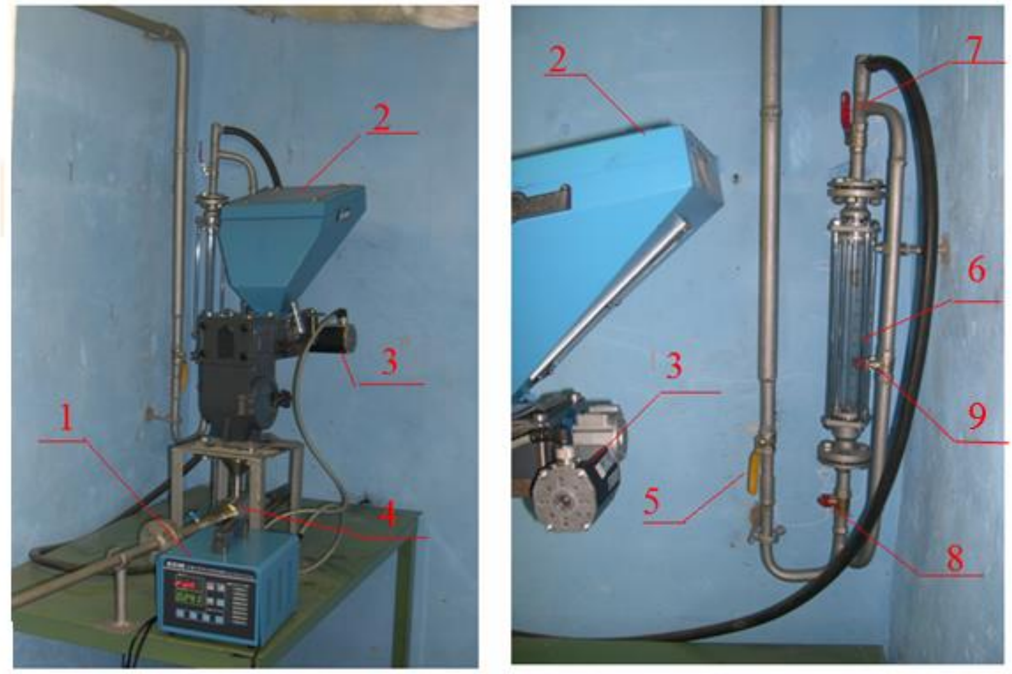
Table 1. Experimental ESP technical parameters.

Parameter	Value	Unit of measurement
ESP Design		
Number of housings	2	
Number of sections in one housing	2	
Number of gas passes per section	2	
Total gas flow rate	5,000 – 40,000	m^3/h
Total cross-sectional active area	2.8	m^2
Cross-sectional active area of one housing	1.4	m^2
Type of discharge electrodes	tape-needle	
Type of collection electrodes	S-type	
Number of discharge electrodes per section	28	
Number of collection electrodes per housing	6	
Total number of discharge electrodes	112	
Height of discharge electrode	1.7	m
Section lengths	2.7	m
Inter-electrode spacing	275	mm
Rapping System		
Type	hammer	
Ash Removal System		
Type	hydraulic	
Power Supply		
Number per section	1	
Number per ESP	4	
Control system	automatic	



1- Distribution lattice
2- Compressor
3- Rotameter PM-04-10GUS
4- Feeder SCM75-12
5- Ejector
6,7,8,9,10- Valves

Figure 4. Schematic of the injection and distribution system.



1- Feeder power supply
2- Feeder hopper
3- Feeder screw
4- Ejector
5,7,8,9- Valves
6- Rotameter

Figure 5. Photo of the injection and distribution system.

Figure 6. ACI lance inserted into the gas duct.

4 Measuring devices

Measuring devices used during tests are listed in Table 2 below.

Table 2. Measuring devices.

Device	**Measurement**	**Comment**
Flue gas flow meter KIMO MP 200	Flue gas flow rate before and after ESP	Permanent check of flue gas flow rate during tests
Gas analyzer QUINTOX Model KM 9106	Flue gas composition, flue gas temperature before and after ESP	Check of flue gas composition and temperature at the beginning and completion of any test
Ash analyzer GRAVIMAT Model SHC 501	Inlet and outlet ash load	Determination of ESP performance with and without sorbent injection
Rotameter PM-04-10GUS	Air flow rate in carbon injection and distribution system	Permanent check of air flow
Aspirator PU-4E	Flow rate of flue gas, sampled during tests	Determination of sampled gas volume
Temperature meter TESTO 925	Flue gas temperature	Permanent check of flue gas temperature during tests
HR-200	Weighing of dust collectors for GRAVIMAT	Measurement of ash weight in dust collectors for determination of ESP performance

5 Methods

5.1 Determination of necessary AC consumption

From a review of articles and reports, standard and brominated AC injection rates for Hg removal were determined to range from 0 – 160 mg/m^3 (0 – 10 per million actual cubic feet of flue gas [lb/MMacf]) and 0 – 80 mg/m^3 (0–5 lb/MMacf), respectively. The flue gas flow rate for an experimental ESP can vary from 2,500 to 20,000 m^3/h. A flue gas flow rate of about 7,000 m^3/h was used for all tests, and the flue gas velocity inside the ESP was 1.4 m/s for this gas flow rate. Such velocity is typical for most of the older Russian ESPs.

For a flue gas flow rate of about 7,000 m^3/h, AC consumption varied from 0.2 to 1.20 kilograms per hour (kg/h).

5.2 Test of distribution lattice

It was important to determine the range of air flow rate changes. Within this range, AC consumption had to depend upon the auger rotation speed and not depend upon the air flow. Therefore, all AC supplied from the feeder auger into the ejector was mixed with air and sent to the distribution lattice.

In addition, air flow had to be minimal because a higher air flow could change the flue gas composition and temperature. Before the test, the distribution lattice was removed from the duct to check uniformity of the AC flow through the distribution lattice holes, as shown in Figure 7.

The acceptable range of air flow was determined as 10 normal cubic meter per hour (Nm^3/h) to 30 Nm^3/h. This air flow did not influence the flue gas flow and the AC consumption did not depend on air flow. The AC distribution through the distribution lattice holes was uniform. An air flow rate of 20 Nm^3/h was used for all tests and was verified using a rotameter.

Figure 7. Test of distribution lattice.

5.3 Measurement of flue gas composition, temperature and flow rate

Monitoring devices used for measurement of flue gas parameters are shown in Figure 8. Flue gas composition was measured by a QUINTOX gas analyzer at the beginning and the end of every sampling test (see Figure 8a). Temperature was measured by a TESTO Model 925 temperature meter (see Figure 8b) and by a QUINTOX gas analyzer additionally during gas composition measurements. Flue gas composition varied slightly during tests and did not depend upon the boiler operation mode. The flue gas flow rate was maintained at 7,000 m^3/h during all tests and was monitored by a KIMO Model MP 200 flow meter (see Figure 8c).

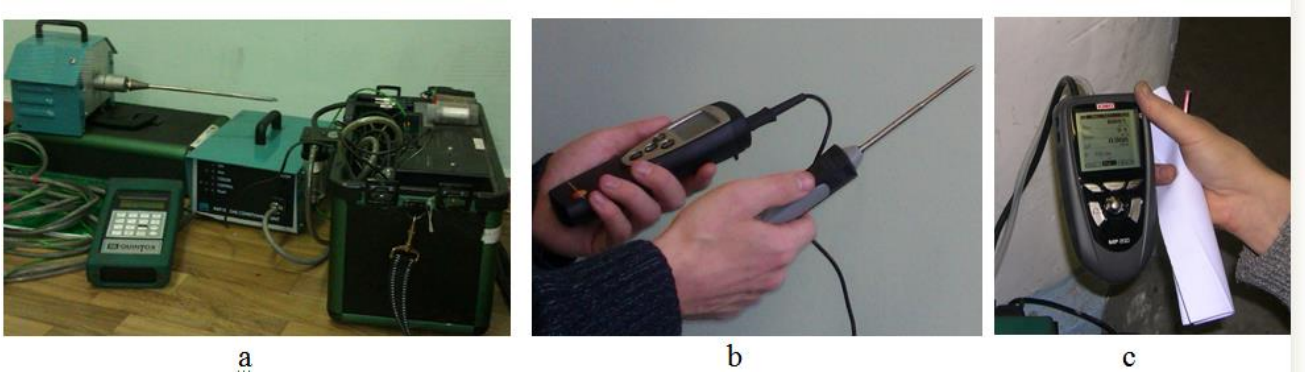


Figure 8. Monitoring devices: (a) Gas analyzer QUINTOX KM 9106, (b) Temperature meter TESTO 925, (c) Flow meter KIMO MP 200.

5.4 Characterization of experimental ESP performance

The operating parameters for experimental ESP were not changed during the tests. Experimental ESP power supplies were operated in an automatic mode. Operating voltages and currents for the first and second sections were 35 kilovolts (kV), 1.5 milliampere (mA) and 37 kV, 2.1 mA, respectively. Voltage-current (VI) curves for the experimental ESP are given in Figure 9.

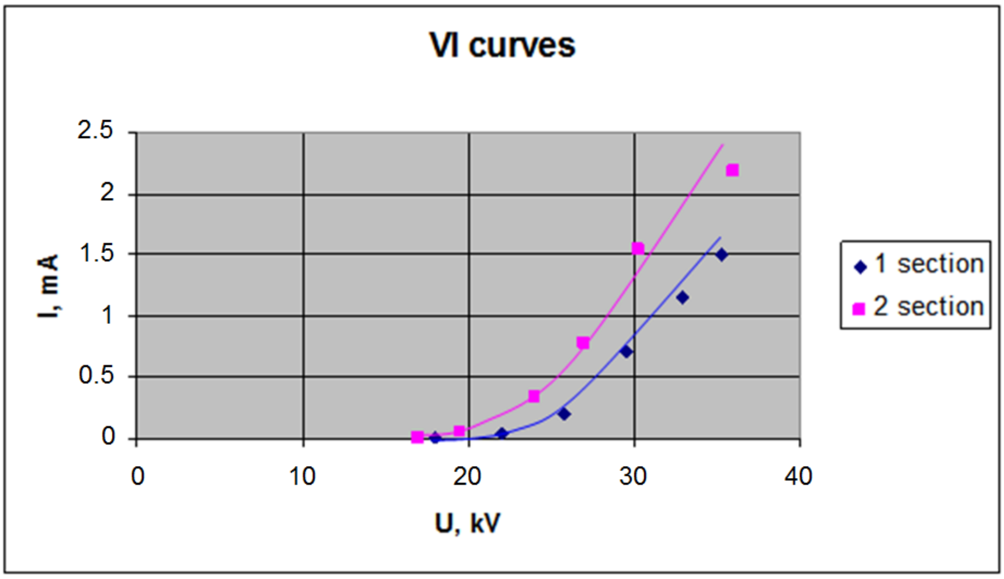


Figure 9. VI curves of experimental ESP.

The ash content in the gas ducts before and after the experimental ESP was measured by the GRAVIMAT Model SHC-501 (see Figure 10).

Figure 10. Measurements of experimental ESP ash load: (a) inlet, (b) outlet.

5.5 Measurement of Hg content in flue gas

The concentration of Hg in the flue gas was measured using Ontario-Hydro method (OH) sampling trains (see Figure 11) and using sorbent tubes (Method 30B).

Figure 11. OH sampling equipment.

6 Test results

Tests were conducted with the boiler load at 280 ± 10 megawatt electrical (MW_e), firing Kuznetsk coal. Three sets of test were conducted, each consisting of two phases:

1. Baseline tests

2. Tests with injection of standard AC

In every test set, baseline tests were conducted followed by the tests with standard AC injection. Initially, only two sets of tests were planned. However, the results of the first two baseline tests did not agree. The apparent reason for this disagreement was the varying Hg content in the coal. The Cherepetskaya TPP buys Kuznetsk coal from various brokers. Because of different supplies, the coal composition and the coal Hg content varied.

At the end of December 2012, the Cherepetskaya TPP bought a large, new batch of coal which was used in the third set of tests.

6.1 First set of tests

6.1.1 Baseline tests

Baseline tests were carried out for four days. The test conditions, coal composition and the coal Hg content were approximately the same for all tests.

Chemical coal composition was as follows:

- $W^p = 8.7\%$; $A^p = 17.4\%$; $V = 11.9\%$; $S^p = 0.46\%$

where W^p is water content, A^p is ash content, V^p is volatile content, and S^p is sulfur content of the coal by weight.

Fly ash concentrations measured by the GRAVIMAT Model SHC 501 ash analyzer were as follows:

- ESP inlet – 15.2-18.9 g/Nm^3
- ESP outlet – 1.4-1.8 g/Nm^3
- PM capture of the experimental ESP was about 91%
- Unburned carbon in the fly ash was 5.1%

Flue gas composition before ESP was measured by gas analyzer Quintox Model KM 9106 and was as follows:

- Oxygen (O_2) – 7.7-8.9% (air leakage in ESP was about 1.0%)
- Carbon monoxide (CO) – 4.0-6.0 parts per million (ppm)
- Nitrogen oxides (NO_X) - 500-570 ppm
- Sulfur dioxide (SO_2) - 240-260 ppm
- Flue gas temperature upstream of ESP was 115÷122 °C
- Average Hg content in the coal (four samples) was 84.7 milligrams per metric ton (mg/t); a sample was collected each test day

- Average chlorine content in the coal (four samples) was 2 kg per metric ton (kg/t) (0.02%); a sample was collected each test day
- Average Hg content in the ash (eight samples) was 192.5 mg/t; two samples were collected each test day

Measurement results of the flue gas Hg content are shown in Table 3.

Table 3. Measurement results of the flue gas Hg content for the first baseline tests.

No.	Hg content in flue gas before the ESP, µg/m³ at 6% O_2	Hg content in flue gas after the ESP, µg/m³ at 6% O_2	Sampling method	Hg capture on fly ash, %	Comment
1	3.370*	2.050*	30B	39.2	sorbent trap
2	2.930*	1.930*	30B	34.1	sorbent trap
3	4.270*	2.430*	30B	43.1	sorbent trap
4	3.990	2.040	30B	48.9	speciated trap
5	4.380	-	OH	-	sampling train
6	3.650	1.970	30B	46.0	sorbent trap
7	4.610	2.710	30B	41.2	sorbent trap
8	6.740	3.700	30B	45.1	sorbent trap

* average value for two samples taken at the same time.

Table 3 shows that the average efficiency of Hg removal in the experimental ESP was about 42.5%. One OH train was ruined because reagents in the bulbs were mixed during tests (the sampling pump vacuum was too high). The pump vacuum was corrected in the next set of OH tests. Measurement results of the flue gas Hg contents are shown in Table 4. All samples were taken under the same conditions.

Table 4. Measurement results of the flue gas Hg contents for the first baseline tests.

Total Hg, µg/m³ at 6% O_2	Hg(0), µg/m³ at 6% O_2	Hg(II), µg/m³ at 6% O_2	Location of sampling point	Sampling method
3.990	1.440	2.550	Before ESP	speciated trap
4.380	1.840	2.540	Before ESP	OH
2.040	0.920	1.120	After ESP	speciated trap

Hg	mercury
Hg(0)	elemental Hg
Hg(II)	oxidized Hg

The results of the form of Hg in flue gas (Hg speciation) as determined by OH and 30B methods were satisfactorily matched.

6.1.2 Tests with standard AC injection

The first tests with injection of standard AC were carried out seven days following the baseline tests. The test conditions for the standard ACI tests were the same as for the first baseline tests.

Hg content at the ESP inlet/outlet was measured each time during ACI tests. The average Hg content in the flue gas before ESP was 4.19 micrograms per cubic meter (µg/m^3). Dependence of Hg removal in the experimental ESP vs. standard sorbent injection rate is shown in Table 5.

Table 5. Hg removal in experimental ESP vs. standard sorbent injection rate.

Equivalent injection rate, mg/m^3	0	38.4	48.0	113.6	163.2
Total Hg content in flue gas after ESP, µg/m^3 at 6% O_2	2.12	1.18	1.01	0.80	0.68
Hg removal, %	42	68	72	78	81

6.2 Second set of tests

6.2.1 Baseline tests

Baseline tests were carried out over four days. The analysis of the Kuznetsky coal was as follows:

- W^p = 8.1%; A^p =26.3%; V = 18.4%; S^p = 0.36%
- Average chlorine content in the coal (3 samples) was 4 kg/t (0.04%)
- Experimental ESP performance was about 90%

Flue gas composition before ESP was measured by gas analyzer Quintox and was as follows:

- O_2 – 8.8-9.3% (air leakage in ESP was about 1.0%)
- CO – 5.0-9.0 ppm
- NO_X – 500-570 ppm
- SO_2 – 250 ppm
- Flue gas temperature before ESP was 110÷115°C

Over the test period, Hg content in the coal varied, as shown below:

- Day 1: 302 mg/t (1 sample)
- Day 2: 91 mg/t (1 sample)
- Day 3: 240 mg/t (1 sample)
- Day 4: 39, 91, and 61 mg/t (3 samples)
- Average Hg content in the ash was 380.7 mg/t (8 samples)

The results of Hg content determination in flue gas for the second baseline tests are shown in Table 6. The results of flue gas Hg measurements for the second set of baseline tests are shown in Table 7. All samples were taken at the same time.

Table 6. The results of Hg content determination in flue gas for the second set of baseline tests.

No.	Hg content in flue gas before ESP, µg/m^3 at 6% O_2	Hg content in flue gas after ESP, µg/m^3 at 6% O_2	Sampling method	Hg capture on fly ash, %	Comment
1	10.8[a]	6.2[a]	30B	42.6	sorbent trap
2	31.4	20.4	30B	35.0	speciated trap
3	26.2	17.1	OH	34.7	train
4	21.2[a]	12.7[a]	30B	40.1	sorbent trap
5	2.97[a,b]	2.01[a,b]	30B	32.3	sorbent trap

a. Average value between two samples taken at the same time.

b. To control the accuracy of the measurements, one of the tubes was 30B standard vapor-spiked sorbent trap (with 50 nanograms [ng] Hg).

Table 7. The results of Hg form determination in flue gas for the second baseline tests.

Total Hg, µg/m^3 for 6% O_2	Hg(0), µg/m^3 for 6% O_2	Hg(II), µg/m^3 for 6% O_2	The location of sampling point	Sampling method
31.4	24.4	7.0	Before ESP	speciated trap
26.2	21.3	4.8	Before ESP	OH
20.4	12.7	6.8	After ESP	speciated trap
17.1	12.6	4.5	After ESP	OH

The results confirm good agreement of Method 30B and OH. However, the Hg contents in the coal and in flue gas varied, as was shown above. Thus, the tests were repeated (third set of tests).

6.3 Third set of tests

6.3.1 Baseline tests

Baseline tests were conducted during 3 days. The test conditions, coal composition, and Hg content in the coal were approximately the same for baseline tests and tests with standard ACI.

Chemical coal composition for these tests is given below:

- W^p = 10.0%; A^p =22.4%; V = 19.2%; S^p = 0.34%
- Average chlorine content in the coal (4 samples) was 7.5 kg/t (0.075%)

The results of GRAVIMAT fly ash concentration measurements are given below:

- ESP inlet load – 16.5-20.3 g/Nm^3
- ESP outlet load – 1.2-2.0 g/Nm^3
- Unburned carbon in fly ash – 4.7%
- Average performance of experimental ESP was about 91%.

Flue gas composition before ESP measured by the Quintox gas analyzer was as follows:

- O_2 – 8.0-9.6% (air leakage in ESP was about 1.0%)
- CO – 4.0-7.0 ppm
- NO_X - 520-540 ppm
- SO_2 - 210-270 ppm
- Flue gas temperature before ESP was 110÷115°C.
- Average Hg content in the coal (6 samples) was 64.2 mg/t
- Average Hg content in the ash (8 samples) was 167.5 mg/t

The results of Hg in flue gas measurement for the third set of baseline tests are shown in Table 8.

Table 8. The results of Hg content determination in flue gas for the third set of baseline tests.

N	Hg content in flue gas before ESP, µg/m^3 at 6% O_2	Hg content in flue gas after ESP, µg/m^3 at 6% O_2	Sampling method	Hg Capture on Fly ash, %	Comment
1	4.220[a]	2.570[a]	30B	39.1	sorbent trap
2	4.270[a,b]	2.610[a,b]	30B	38.9	sorbent trap
3	3.830[a]	2.280[a]	30B	40.5	sorbent trap

a. Average value for two samples taken at the same time.

b. One of the tubes was a spiked sorbent trap (with 50 ng Hg).

6.3.2 *Tests with standard AC injection*

Tests with standard ACI were carried out for seven days following the baseline tests. Average Hg content in the flue gas before the ESP was 4.22 µg/m^3 and average Hg content in flue gas after ESP was 2.57 µg/m^3. Hg removal in ESP without sorbent injection was about 39%. Dependence of Hg removal in experimental ESP vs. standard sorbent injection rate is shown in Table 9.

Table 9. Hg removal in experimental ESP as a function of standard sorbent injection rate

Equivalent injection rate, mg/m^3	0	35.2	48.0	110.4	156.8
Hg content in flue gas after ESP at 6% O_2, µg/m^3	2.57	1.38	1.14	1.01	0.88
Efficiency of ESP Hg removal, %	39	67	73	76	79

Total Hg removal in the experimental ESP as a function of standard AC injection rate is shown in Figure 12.

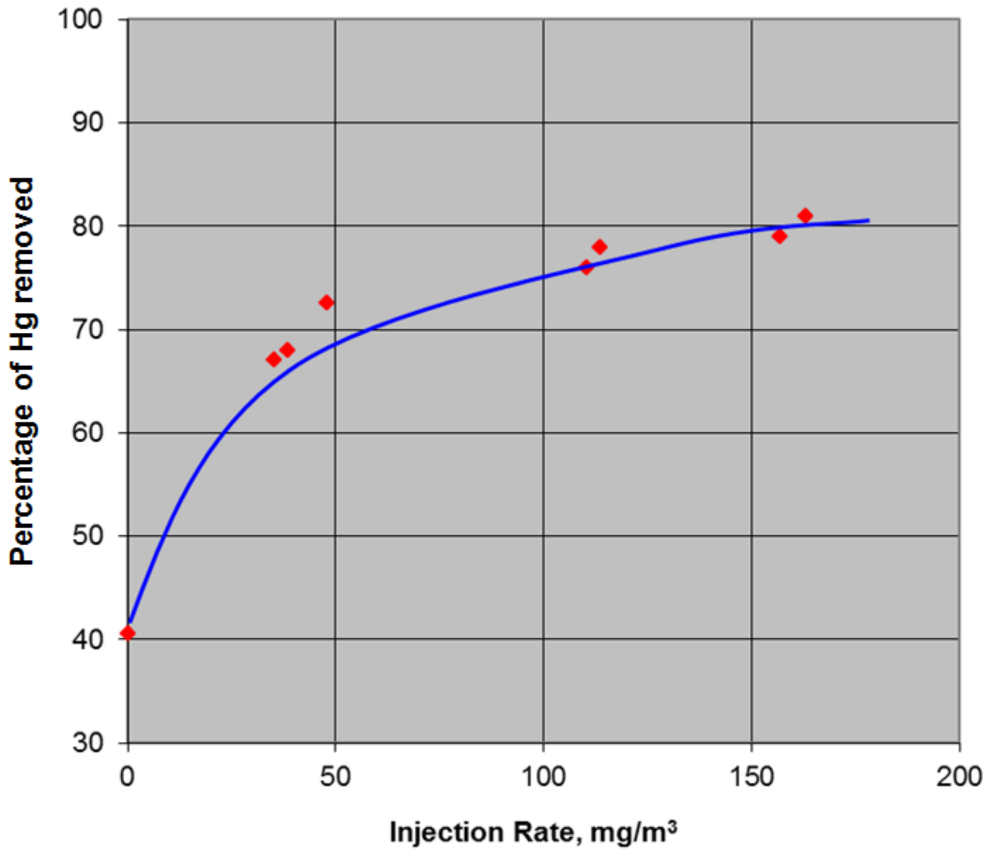


Figure 12. Parametric test results of Hg removal by standard AC.

7 Tests with injection of brominated AC

7.1 Measurement of flue gas composition, temperature, and flow rate

Flue gas composition was measured by a QUINTOX gas analyzer at the beginning and the end of every sampling test. Temperature was measured by temperature meter TESTO Model 925 continuously and by a QUINTOX gas analyzer during gas composition measurements. Flue gas composition changed slightly during tests and did not depend upon the boiler operation mode. The flue gas flow rate through the experimental ESP was monitored by flow meter KIMO MP 200 and was kept at 7,000 m^3/h during all tests.

7.2 Baseline tests

Baseline tests were carried out for four days. The test conditions, coal composition, and the coal Hg content were approximately the same for all tests.

The chemical coal composition test results were as follows:

- Wp = 9.2-10.0%; Ap =20.0-23.1%; V = 14.5-17.9%; Sp = 0.4%.
- Average chlorine content in the coal was 0.02%.
- Unburned carbon in fly ash – 4.5%.
- Fly ash concentration in the flue gas was measured by a GRAVIMAT and fly ash results were as follows:

  - ESP inlet – 16.5 - 18.0 g/Nm^3.
  - ESP outlet – 1.3 - 1.6 g/Nm^3.

- Average experimental ESP performance was about 91.6%.

Flue gas composition before ESP was measured by the Quintox gas analyzer and was as follows:

- O_2 – 7.8-9.4% (air leakage in ESP was about 1.0%).
- CO – < 5 ppm.
- NO_X –540-580 ppm.
- SO_2 – 200-290 ppm.

- Flue gas temperature before ESP was 105-115 °C.
- Flue gas flow rate was about 7,000 m^3/h for all tests and the flue gas velocity inside the ESP was 1.4 m/s in this case.
- Average Hg content in the coal (4 samples) was 84.7 mg/t; one sample was taken each test day.
- Average Hg content in the fly ash was 192.5 mg/t
- Average Hg content in flue gas before the ESP was 4.61 µg/m^3

The flue gas Hg content was measured using 30B Method and OH.

7.3 Tests results with brominated AC injection (first set of tests)

The first set of tests with brominated AC injection was carried out for seven days after completion of baseline tests. The test conditions were the same as for the baseline tests. The results of the first test set with injection of brominated AC are shown in Table 10.

Table 10. The results of the first set of tests with injection of brominated AC.

Equivalent injection rate, lb/MMacf	0	2.6	3.5	7.0	7.0
Hg content in flue gas after ESP, µg/m^3 at 6% O_2	2.72	0.55	0.69	0.52	0.44*
Total Hg removal, %	41.0	88.1	85.0	88.7	90.4

* Speciated trap

7.4 Tests results with brominated AC injection (second set of tests)

Coal composition for the second set of tests did not vary significantly. Average Hg content in the coal (six samples) was 64.2 mg/t. Average chlorine content in the coal was 0.15% and average Hg content in the ash (8 samples) was 167.5 mg/t. The results of the second set of tests with injection of brominated AC are given in Table 11.

Table 11. The results of the third set of tests with injection of brominated AC.

Equivalent injection rate, lb/MMacf	0	2.5	3.6	6.9	6.8	6.8
Hg content in flue gas before ESP, µg/m^3 for 6% O_2	4.64	4.84	4.82	4.51	4.45[a]	4.75[b]
Hg content in flue gas after ESP, µg/m^3 for 6% O_2	2.75	0.98	0.73	0.65	0.53[a]	0.89[b]
Total Hg removal, %	40.7	79.7	84.8	85.6	88.1	81.3

a. Speciated trap

b. OH was used

The results of flue gas Hg measurements are shown in Table 12. All samples were taken at the same time. The results confirm good agreement of Method 30B and OH.

Table 12. The results of Hg form determination in flue gas.

Total, µg/m^3 at 6% O_2	**Hg(0), µg/m^3 at 6% O_2**	**Hg(II), µg/m^3 at 6% O_2**	**The location of sampling point**	**Sampling method**
0.44	0.25	0.19	after ESP	speciated trap
4.45	2.03	2.42	before ESP	speciated trap
4.75	2.21	2.54	before ESP	OH
0.53	0.29	0.24	after ESP	speciated trap
0.89	0.51	0.38	after ESP	OH

Total Hg removal in the experimental ESP as a function of standard and brominated AC injection rate is shown in Figure 13.

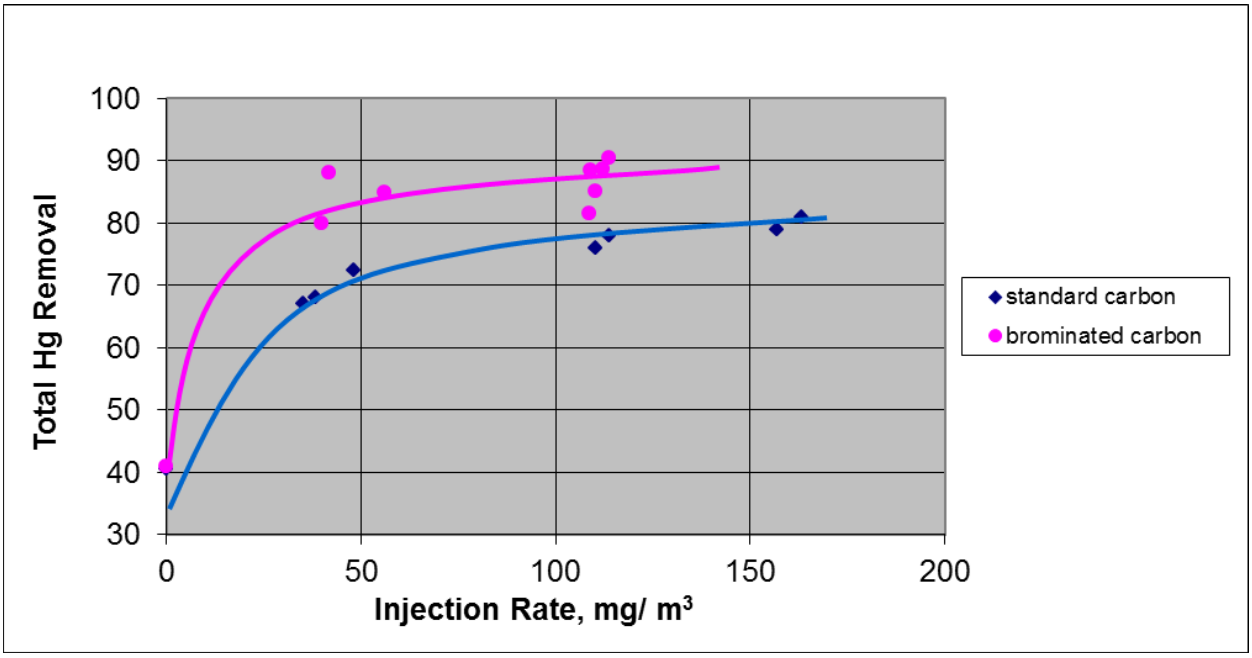


Figure 13. Total Hg removal in the experimental ESP for standard and brominated ACI.

Figure 13 shows that the maximum efficiency of Hg removal by brominated AC with an injection rate of 110 mg/m^3 was about 90%. The efficiency of Hg removal with injection of brominated AC was 10% higher than with standard AC. Maximum efficiency of total Hg removal was about 90% for a brominated AC injection rate of 110 mg/m^3 and about 80% for the standard AC injection rate of 160 mg/m^3.

7.5 Findings and conclusions – Hg emission reduction

1. The emission tests indicated that under baseline conditions, i.e., without the use of sorbents, between 39% to 42% of the Hg was captured on the fly ash.

2. The emission tests indicated that the use of standard AC injection with an injection rate of 160 mg/m^3 (10 lb/MMacf) resulted in a range of capture efficiencies of total Hg removal in the ESP of 67% to about 80%.

3. The emission tests indicated that the use of brominated AC with an injection rate of up to 110 mg/m^3 (7 lb/MMacf) resulted in Hg emission capture efficiencies of up to 90%.

4. The range of 67% to 80% Hg removal from the flue gas achieved with standard AC necessitated an AC injection rate of 160 mg/m^3 to achieve the higher removal. An injection rate of only about 32 mg/m^3 for brominated AC was needed to achieve a comparable level of Hg removal.

5. These emission control efficiency results using Kuznetsk coal are similar to those obtained in earlier U.S. studies using U.S. coals.

8 Leaching tests

Leaching tests were conducted using a variety of methods on samples of the particulate matter wastes collected: (1) under baseline conditions, i.e., when no sorbent was used; (2) when standard AC was used; and (3) when brominated AC was used. Constituents examined were Hg, As and Se.

8.1 Method 1311 Toxicity Characteristic Leaching Procedure (TCLP)

The leaching test procedure follows USEPA Method 1311 (Toxicity Characteristic Leaching Procedure [TCLP], SW-846) [11]. While recognizing that fly ash has many constituents, based on the budget and a review of the available literature of constituents examined in previous studies, three metals were selected for this study. To examine the leaching potential of Se and As, USEPA Method 6020 was also applied.

The samples were exposed to an extraction solution with pH=4.95 on a rotary tumbler at 30 revolutions per minute (rpm) for 18 hours. After the extraction, final pH of the leachate was measured. The leachate was subjected to chemical analyses to determine the concentration of Hg, Se, and As. The Hg content was measured immediately after the leaching tests by Atomic Absorption Spectroscopy (AAS) method using the Lumex Hg analyzer RA-915+, attachment RP-91C. Each test used 12 ESP-captured fly ash samples that were:

- Six baseline test samples (without AC injection) - samples 1-6
- Three samples with standard AC injection - samples 7-9
- Three samples with brominated AC injection – samples 10-12

Each sample was tested four times, Tables 13 through 15 show the average values for the four measurements for each sample. The leaching test results of fly ash without sorbent injection are shown in Table 13.

Table 13. Leaching test results for the fly ash without sorbent injection.

	Sample 1	**Sample 2**	**Sample 3**	**Sample 4**	**Sample 5**	**Sample 6**	**Regulatory Limit TCLP, µg/L**
Hg, µg/L	<0.1	0.8	9.0	<0.1	0.1	0.4	200
As, µg /L	--	--	--	77.0	86.9	89.3	5000
Se, µg /L	--	--	--	12.8	14.9	16.2	1000
Leachate pH	5.57	6.33	8.50	6.15	5.83	6.77	--
Hg content in captured ash, mg/t	167.1	380.1	520.3	157.2	171.2	190.5	--

Hg leaching by TCLP tests showed Hg stability, even for very high Hg content in fly ash. Content of all investigated elements in the leachate does not exceed USEPA’s regulatory limit, as shown in Tables 13-15. The TCLP is USEPA’s mandated test method for obtaining extracts of wastes when determining whether waste and its constituents can be designated as “hazardous,” and, thus, requiring the waste to be treated/stored/disposed in hazardous waste facilities, e.g., a hazardous waste landfill. The TCLP includes regulatory levels for assessing individual contaminant concentrations (see ANNEX II for details).

The leaching test results of ash with standard AC injection are shown in Table 14.

Table 14. Leaching test results for the ash with standard AC injection.

	Sample 7	**Sample 8**	**Sample 9**	**Regulatory Limit TCLP, µg/L**
Hg, µg /L	<0.1	0.1	0.1	200
As, µg /L	78.8	78.6	88.6	5000
Se, µg /L	20.5	22.2	28.5	1000
Leachate pH	6.25	6.83	9.18	--
Hg content in captured ash, mg/t	163.2	175.7	194.7	--

Leaching tests of fly ash from injection of standard AC as well as without AC injection showed no Hg leaching. The As content in leachate remains approximately equal with and without the sorbent injection. The Se content in leachate is approximately twice as high with the standard ACI. Apparently, the injected sorbent effectively captures Se and some of the captured Se is leachable.

The leaching test results with brominated ACI are shown in Table 15.

Table 15. Leaching test results for the ash with brominated ACI.

	Sample 10	**Sample 11**	**Sample 12**	**Regulatory Limit TCLP, µg/L**
Hg, µg /L	0.3	<0.1	0.4	200
As, µg /L	30.3	29.8	31.2	5000
Se, µg /L	17.7	30.6	21.1	1000
Leachate pH	6.15	5.83	6.77	--
Hg content in captured ash, mg/t	160.1	190.7	197.3	--

As and Se concentrations in leachate after completion of leaching tests for extraction solution with pH=4.95 are shown in Figure 14.

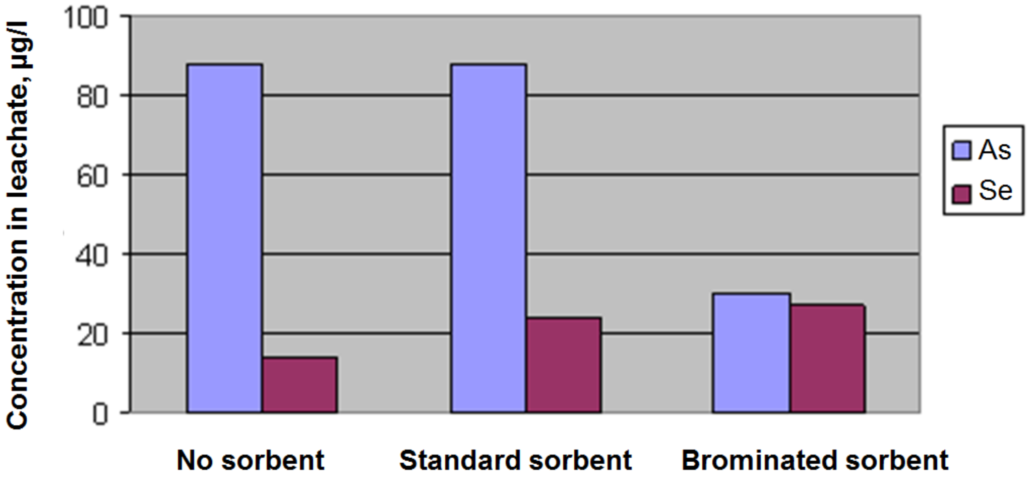


Figure 14. As and Se concentrations in leachate for extraction solution with pH=4.95.

Leaching tests for fly ash after injection of brominated AC showed that no Hg leached from the sample. The As concentration in the brominated AC leachate is significantly lower (two to three times) than concentration in both the standard AC leachate and in the "no sorbent" leachate. The Se concentration is equal to that in the standard AC leachate, and is approximately two times higher than the "no sorbent" leachate concentration.

8.2 Additional tests with Method 1311

Leaching tests with the same ash samples have been carried out for an extraction solution with pH = 2.88 to evaluate the effect of pH on leaching. The leaching test results for the extraction solution with pH=2.88 are shown in Table 16. Leaching tests were conducted for Hg content in captured ash ranging from 157.2 to 197.3 mg/t.

Table 16. Leaching test results for fly ash and its mixtures with standard and brominated AC.

Sample type	**Captured fly ash**			**Captured ash with standard AC**			**Captured ash with brominated AC**		
Sample number	4*	5*	6*	7*	8*	9*	10*	11*	12*
Hg, µg /L	<0.1	0.1	0.3	<0.1	0.1	0.1	0.3	<0.1	0.4
As, µg /L	56.2	61.2	55.5	22.4	22.6	20.1	34.5	35.4	37.2
Se, µg /L	5.5	5.3	7.3	0.6	0.5	1.7	11.1	14.7	12.3
Leachate pH	4.40	4.13	5.2	5.01	4.80	5.17	3.94	4.31	4.05
Hg content in captured ash, mg/t	157.2	171.2	190.5	163.2	175.7	194.7	160.1	190.7	197.3

* Sample numbers the same as in Tables 13-15

Concentrations of As and Se in leachate after completion of the leaching tests for extraction solution with pH=2.88 are shown on Figure 15.

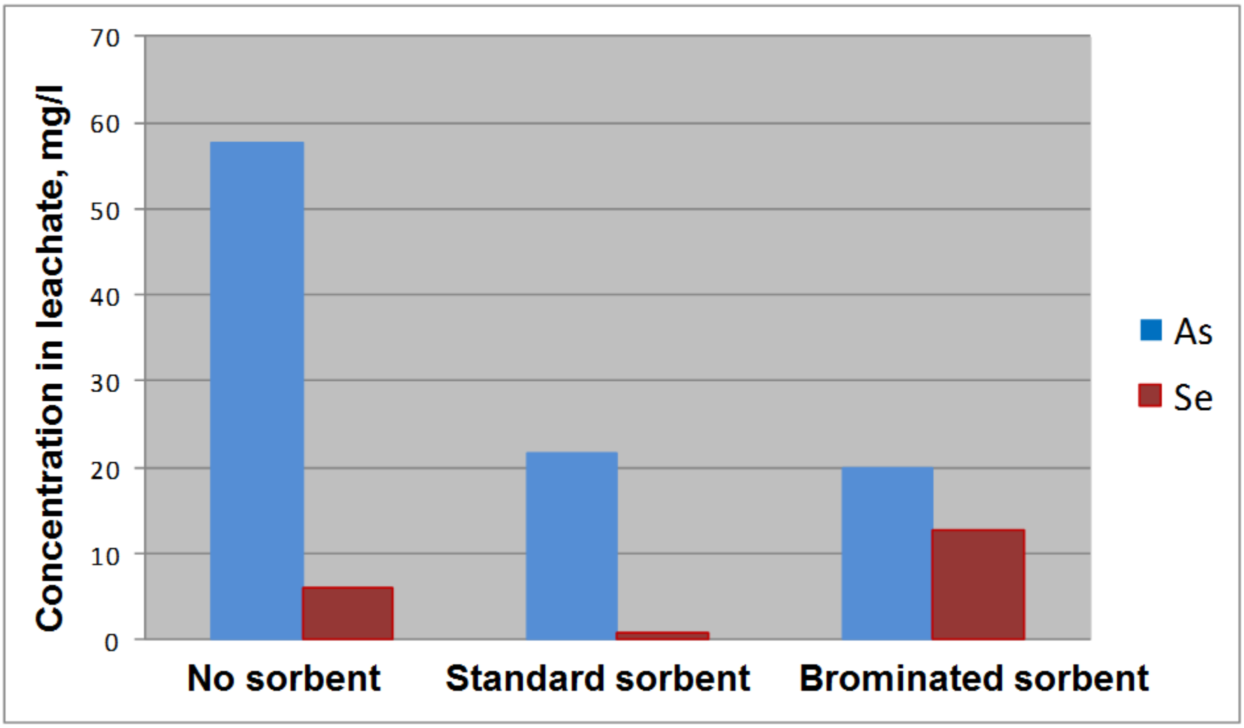


Figure 15. As and Se concentrations in leachate for extraction solution with pH=2.88.

8.3 Discussion of Method 1311 results

The leaching tests carried out with Method 1311 showed that the levels of Hg, As and Se in leachate did not exceed USEPA’s TCLP regulatory limit. The Hg content in all tests at pH=4.95 was negligible (or below the instrument detection limit). When using standard ACI, the As content in leachate remained at the same level, and the Se content approximately doubled. In the case of brominated ACI, Se content is equal to the level when standard AC is used, and the As content is 2.5 to 3 times lower than with the standard AC. Tests at pH = 2.88 showed a remarkable reduction in the leachate of As and Se, especially for the standard AC. Thus, with brominated ACI, the amount of Se that leached out was more than double that of standard AC. The As concentration in leachate does not change with standard sorbent but decreases 2-3 times with brominated AC.

8.4 USEPA Methods 1313 through 1316

For more than 10 years, the USEPA has been working to develop leach-testing alternatives for use in cases where environmental conditions anticipated by the current regulatory test (TCLP) are unlikely to occur. As the regulatory test, TCLP has been used very widely, even when it is not required by regulation and it is implausible for the Municipal Solid Waste (MSW) landfill conditions anticipated by TCLP to occur. This work is in response to concerns raised by the Science Advisory Board in 1991 and 1999 about over-broad use of TCLP. This work also responds to legal challenges to use of TCLP in cases where waste disposal conditions differ significantly from TCLP conditions because of chemical and/or physical factors.

The Leaching Environmental Assessment Framework (LEAF) methods evaluate the impact of three parameters that strongly affect leaching from both granular and monolithic materials, and whose values vary in the environment:

- Leachate pH – the leachate concentrations of many constituents, including several toxic metals of environmental concern (e.g., lead, As, chromium), vary significantly at different pH values.
- Liquid-to-Solid Ratio (L/S) – the volume of water contacting materials (L/S) can affect initial leaching concentrations and the amount of the constituent leached over time.
- Physical Form of the Waste – the physical characteristics (e.g., "particle" size or monolithic form, exposed surface area, pore structure) affects the rate of constituent leaching from a waste either by percolation or diffusion.

By varying the values of these parameters in laboratory leaching tests, a better understanding of how environmental conditions affect leaching is gained. Using test results in conjunction with the anticipated conditions of waste disposal or reuse, more accurate estimates of the leaching potential of a material as used or disposed can be determined than can be determined than by use of many other leach test approaches. LEAF provides a science-based approach for determining leaching behavior that allows consistent, risk-informed evaluation of disposal and beneficial use applications, thus ensuring more effective and efficient environmental protection while allowing natural resources to be conserved through safe use of secondary materials. This is consistent with the USEPA's goal of sustainability.

The LEAF testing framework comprises four laboratory test methods:

- Method 1313 determines equilibrium leaching concentrations as a function of pH
- Method 1314 determines leaching concentrations as a function of L/S in a percolation column
- Method 1315 determines the rates of leaching from monolithic and compacted granular materials as a function of time
- Method 1316 determines equilibrium leaching concentrations as a function of L/S as batch extractions

All four of these methods (Methods 1313, 1314, 1315, and 1316) have been validated through inter-laboratory comparisons and are posted as "new methods" on USEPA/Office of Solid Waste and Emergency Response's (OSWER) analytical methods guidance (SW-846) in the New Methods section of "Test Methods for Evaluating Solid Waste, Physical/Chemical Methods" (SW-846) of October 2012. [11-13]

The USEPA is working to develop guidance on how LEAF results can be used in decision making. The web link for the four LEAF methods is:

- http://epa.gov/wastes/hazard/testmethods/sw846/new_meth.htm

Additional information and reports on LEAF and its applications can be found at:

- http://www.vanderbilt.edu/leaching/

For this study, USEPA LEAF Methods 1313 and 1316 were used to evaluate the leaching behavior of Hg, As, and Se as a function of pH and L/S. Microwave assisted acid digestion was used to determine the total chemical composition of the fly ash samples. The total elemental contents (As, Se) in fly ash and raw coal samples were analyzed by AAS methods (Lumex AAS MGA-915). Hg contents were measured using a Lumex Hg analyzer RA-915+ with attachment RP-91C for direct Hg analysis. Fly ash samples were homogenized with a ball mill to reduce the particle size to less than 0.3 µm. Following the method given in literature,[14-15] the fly ash was decomposed in the microwave oven Ethos ONE (Milestone) for analysis of As and Se.

The procedure for analyzing fly ash microwave-oven decomposition follows. First, about 250 mg of the sample and 10 ml of 65% nitric acid were placed in a polytetrafluoroethylene (PTFE) container inside the microwave oven. Then six bottles containing six samples were tightly capped and placed on the microwave oven carousel. The microwave heating program consisted of a three-stage power and time setting. The system operated at a power level of 50% for 8 minutes at full power for 4 minutes and then 80% for 7 minutes. Third, after completion of the heating cycle, the rack was removed from the oven and bottles were cooled in a water bath for about 15 minutes. Fourth, the beaker contents were filtered through a 0.5 µm membrane filter. Finally, the solution was put into a 100-ml calibrated flask, diluted to volume, and analyzed.

In this study leaching characterization was conducted by the USEPA test Methods 1313 and 1316 [14-15]. The two methods were intended to characterize the liquid-solid partitioning (LSP) at conditions approaching equilibrium as a function of either final extract pH or L/S.

Method 1313: Liquid-Solid Partitioning as a Function of Extract pH for Constituents in Solid Materials using a Parallel Batch Extraction Procedure [12]. This equilibrium-based leaching test is designed to provide the LSP curve of constituents as a function of eluate pH, and consists of nine parallel batch extractions of a particle size reduced solid material in dilute acid or base in order to achieve final extract pH values at specific target values. Experimental conditions: temperature = 21-22 ºC, leaching time = 24 hours. Agitation: end-over-end rotation at 30 rpm.

Method 1316: Liquid-Solid Partitioning as a Function of Liquid-Solid Ratio for Constituents in Solid Materials Using a Parallel Batch Extraction Procedure [16]. This equilibrium-based test method is designed to provide LSP of inorganic constituents at the natural pH of the solid material (i.e., no acid or base addition) as a function of the ratio of the amount of liquid per solid mass unit.

Hg, As and Se contents were measured immediately after the leaching tests by AAS method with Lumex Hg analyzer RA-915+, attachment RP-91C for Hg and Lumex AAS MGA-915 for As and Se.

Each test used 9 ESP captured ash samples which were labeled as follows:

- Three baseline test samples, without ACI (samples 1-3)
- Three samples with standard ACI (samples 4-6)
- Three samples with brominated ACI (samples 7-9)

Each sample was tested four times. Tables AI.1-AI.12 (ANNEX I) show average values for the four measurements. In all tables and figures the Hg concentration is given in nanograms/liter (ng/L), As and Se concentration is given in micrograms/liter (µg/L).

Contents of Hg, As and Se in the tested fly ash samples are given in Table 17, which also shows the analysis results for three samples of raw coal.

Table 17. Contents of Hg, As, Se in fly ash and coal samples.

Sample	Description	Hg ng/g	As µg/g	Se µg/g
1	Fly ash, no AC	152.5	41.76	6.07
2	Fly ash, no AC	165.2	40.09	6.94

3	Fly ash, no AC	174.7	40.35	6.75
4	Fly ash, standard AC	213.1	102.35	8.65
5	Fly ash, standard AC	205.4	112.73	7.93
6	Fly ash, standard AC	190.7	105.47	8.12
7	Fly ash, brominated AC	198.8	77.62	10.26
8	Fly ash, brominated AC	210.5	81.15	8.70
9	Fly ash, brominated AC	232.7	88.53	9.46
C-1	Raw coal	65.1	22.5	2.7
C-2	Raw coal	84.3	10.2	4.1
C-3	Raw coal	91.4	6.3	3.4

Method 1313 leaching test results are shown in Figures 16 through 18 (raw data is given in Tables AI.1-AI.9 in ANNEX I). Inherent sample pH values are circled in Figures 16-18. Maximum Contaminant Levels (MCLs) noted in Figures 16-18 are standards that are set by the USEPA for drinking water quality (see also ANNEX II).

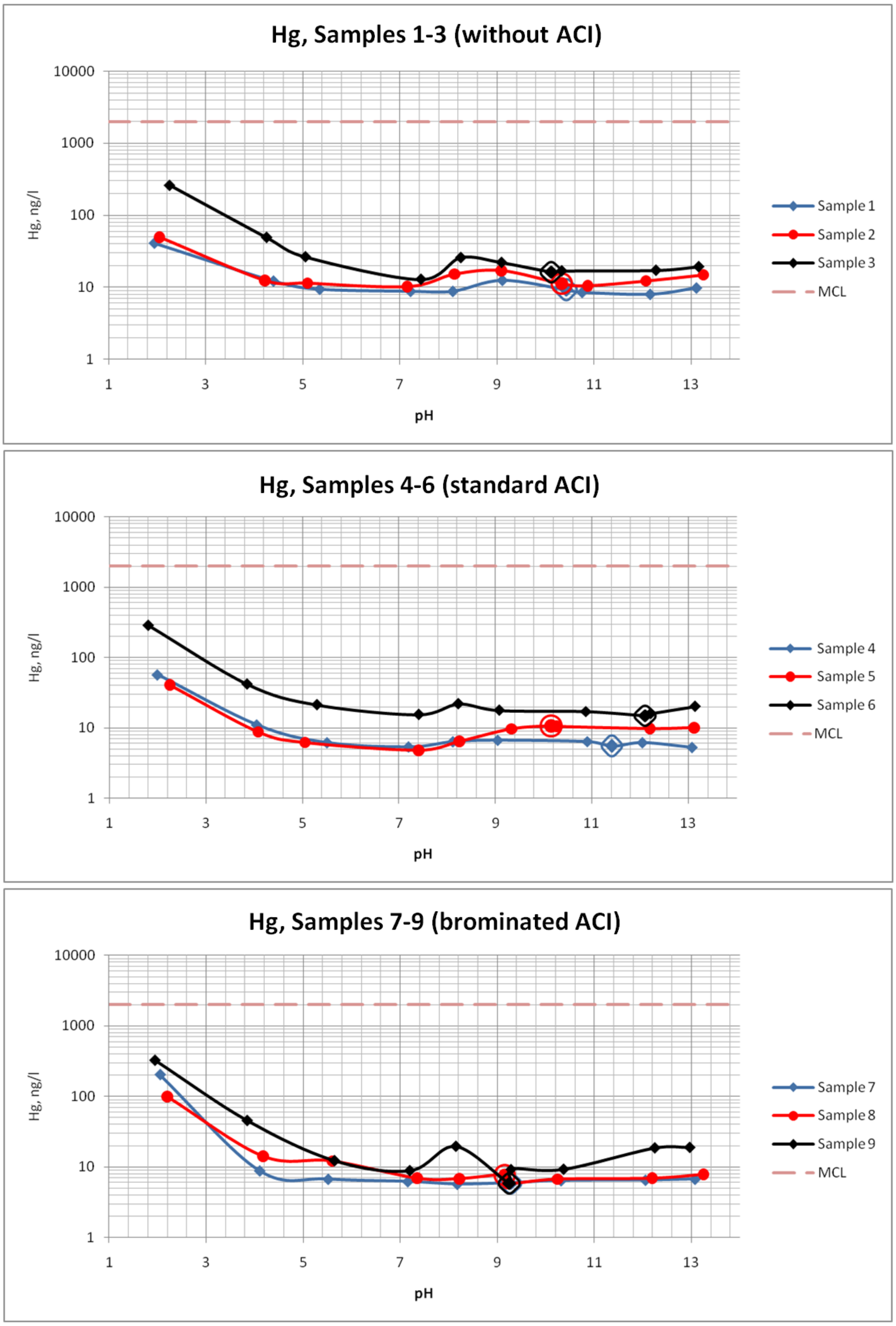


Figure 16. pH-dependent Hg leaching results.

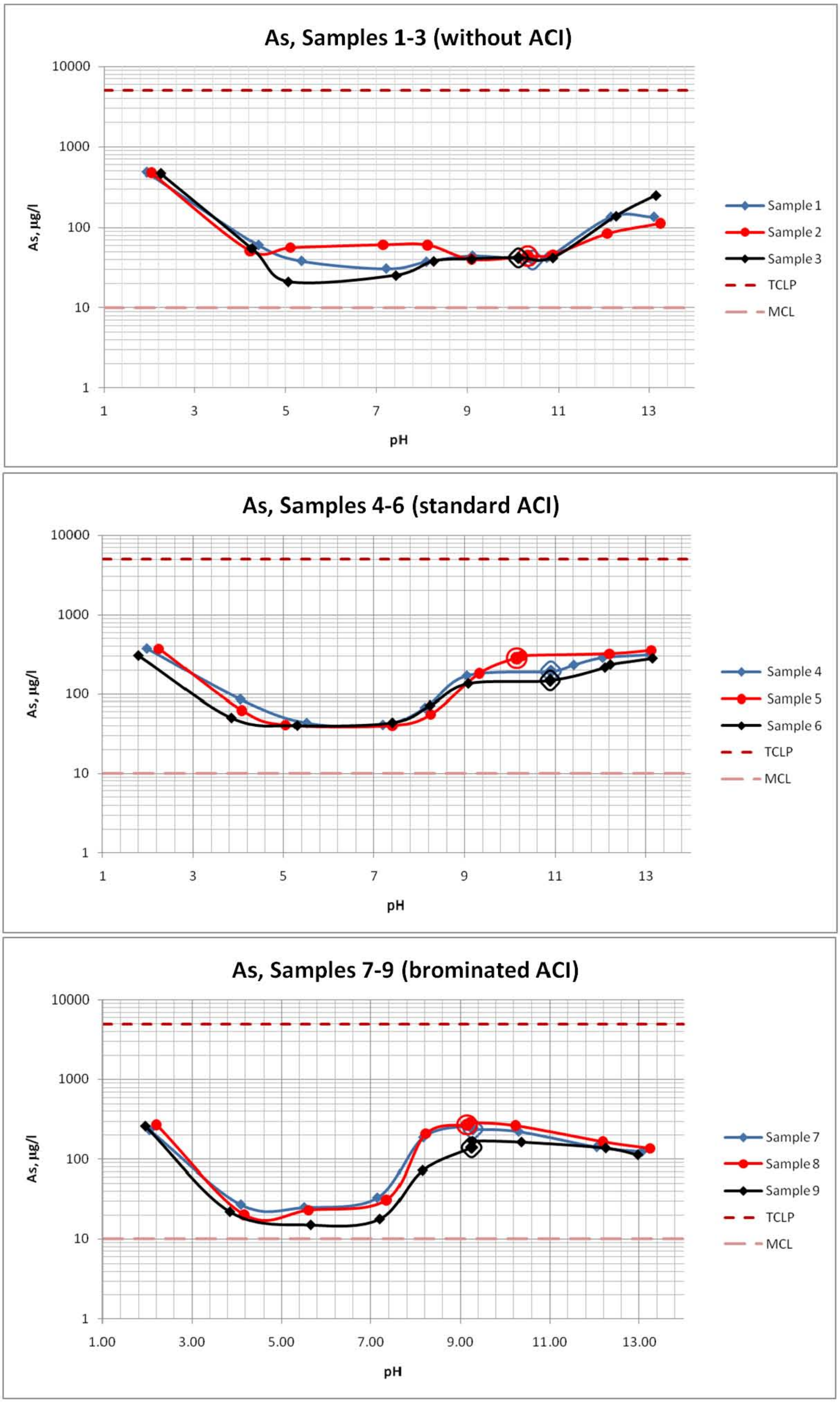


Figure 17. pH-dependent As leaching results.

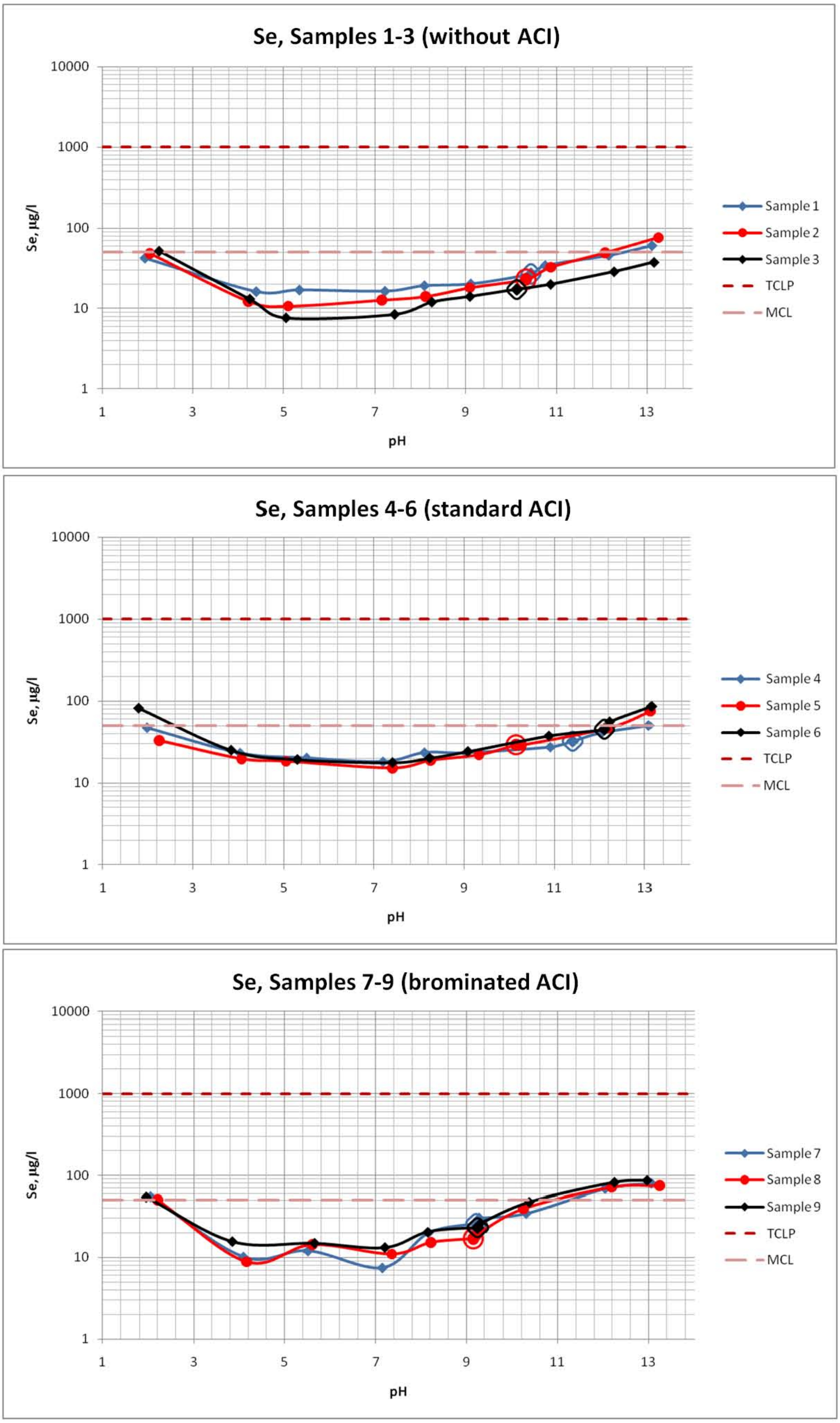
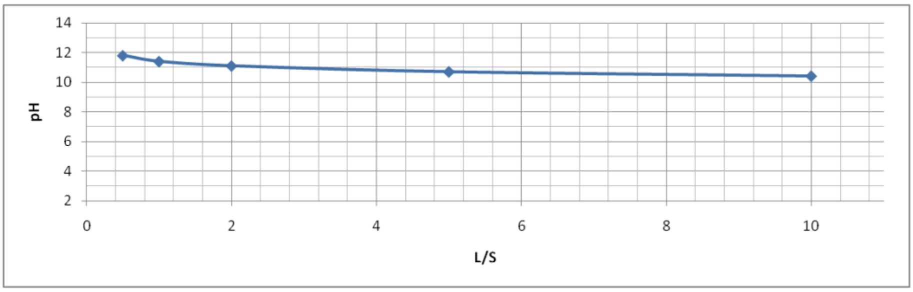
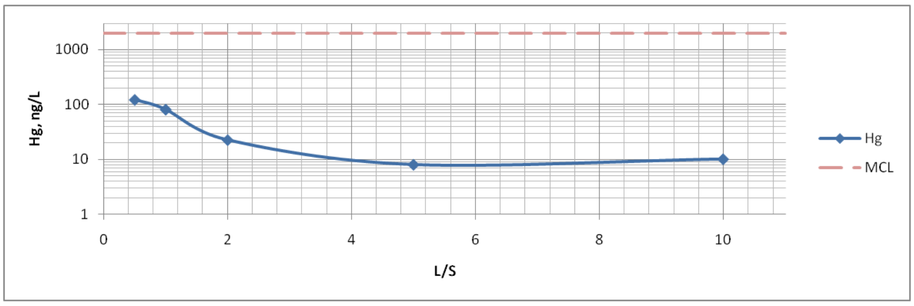


Figure 18. pH-dependent Se leaching results.

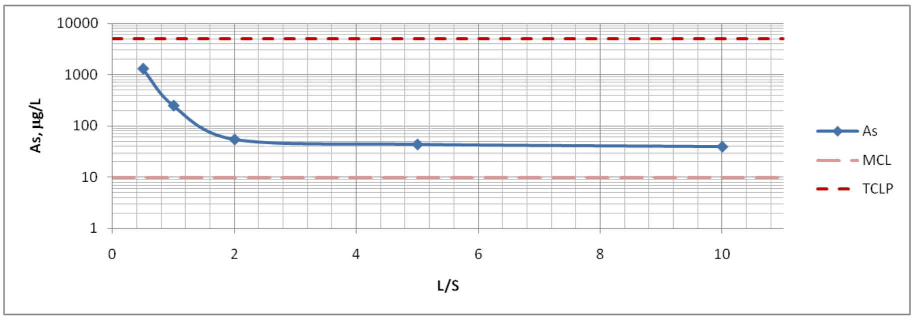
Method 1316 leaching test results are shown in Figures 19 through 21 (raw data is given in Tables AI.10-AI.12 in ANNEX I). The following nomenclature is used in Figures 19-21: MCL=Maximum Contaminant Level in drinking water, TCLP=Regulatory limit TCLP, L/S=Liquid-solid ratio.



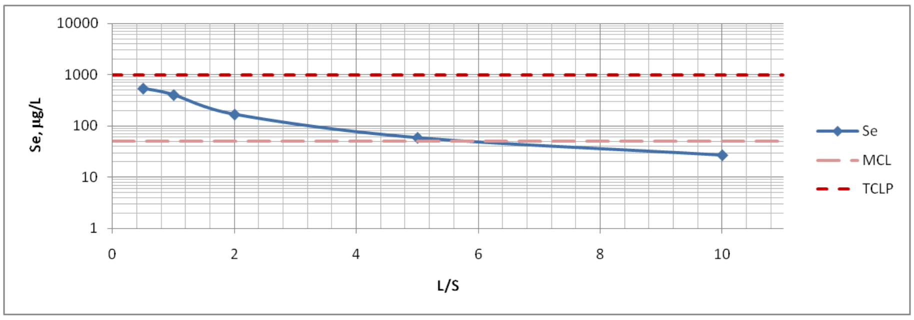
pH as a Function of LS Ratio, sample 1



Hg Release as a Function of LS Ratio, sample 1

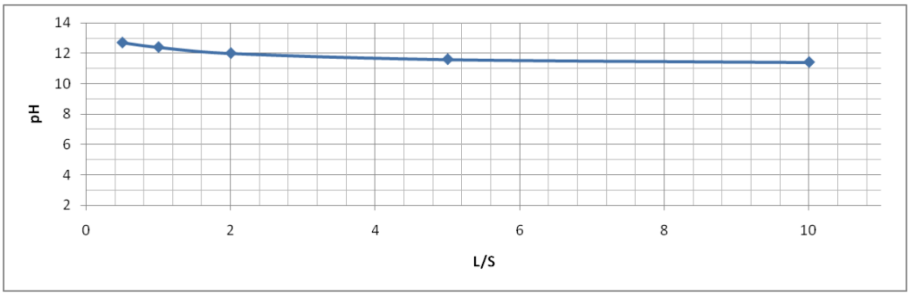


As Release as a Function of LS Ratio, sample 1

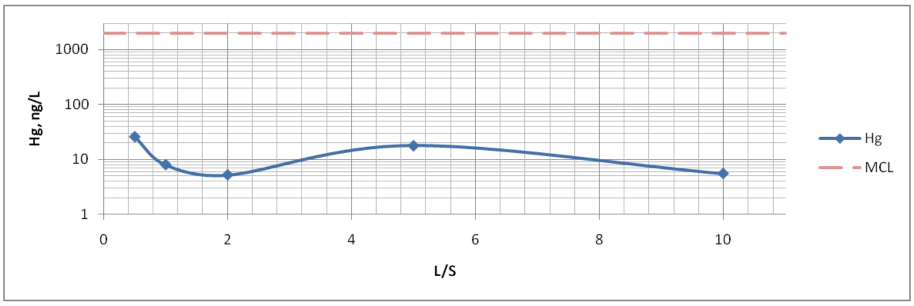


Se Release as a Function of LS Ratio, sample 1

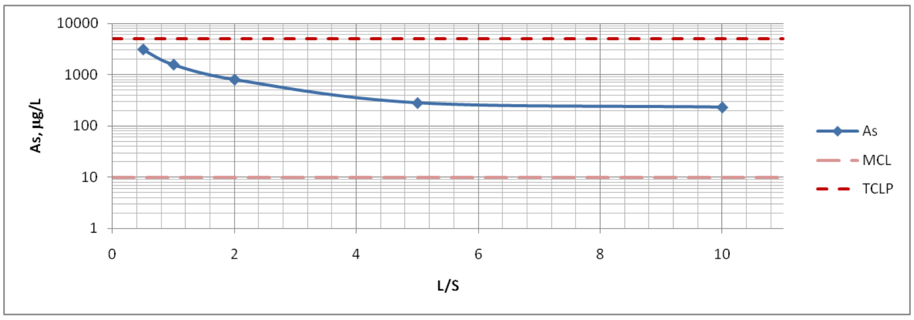
Figure 19. Fly ash leaching test results (Method 1316), sample 1 (without ACI).



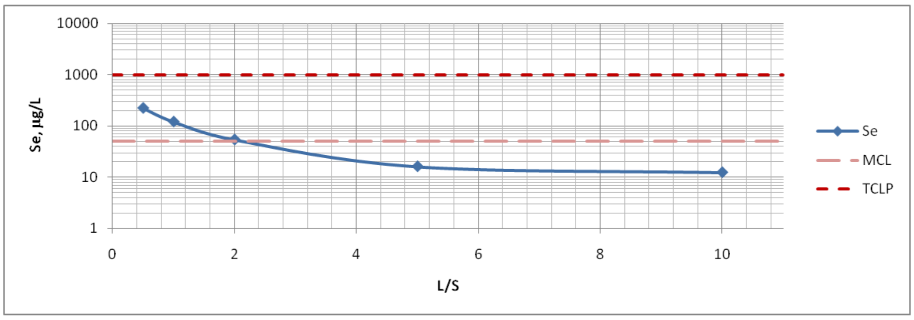
pH as a Function of LS Ratio, sample 4



Hg Release as a Function of LS Ratio, sample 4

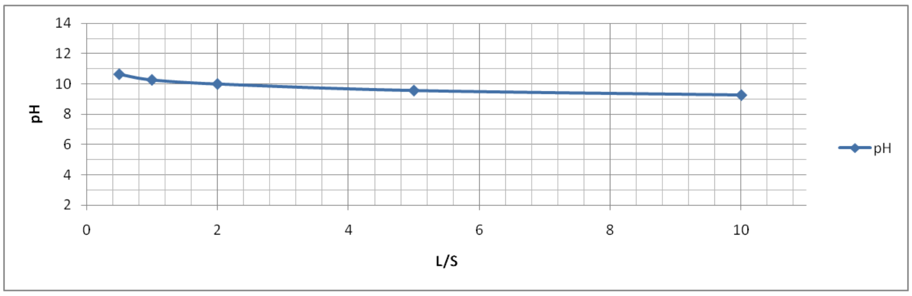


As Release as a Function of LS Ratio, sample 4

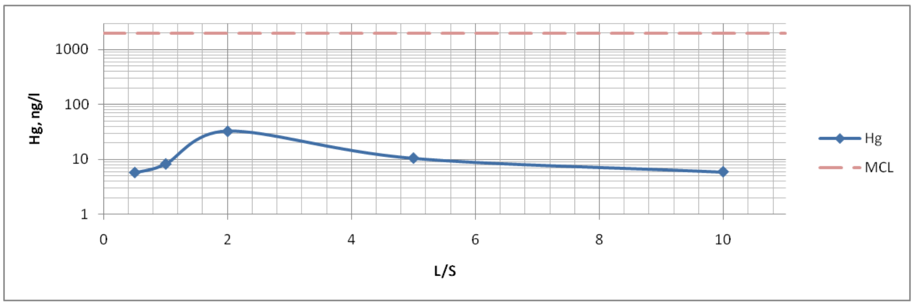


Se Release as a Function of LS Ratio, sample 4

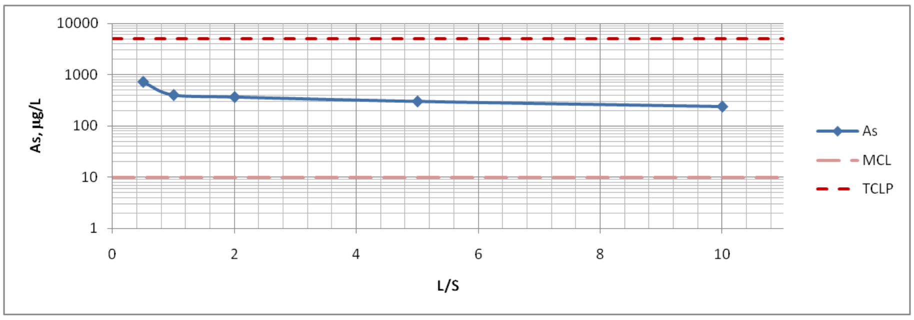
Figure 20. Fly ash leaching test results (Method 1316), sample 4 (standard ACI).



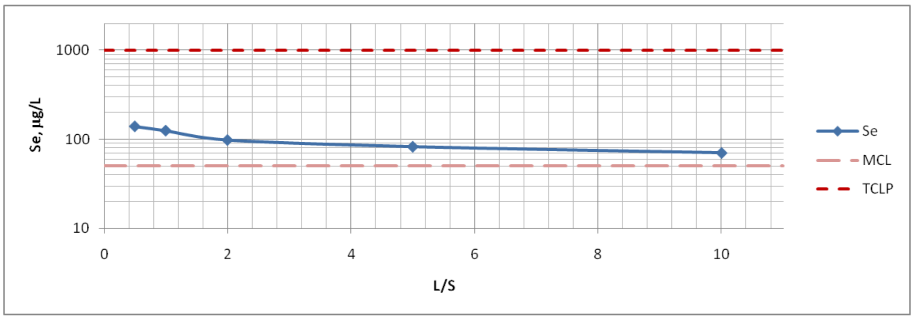
pH as a Function of LS Ratio, sample 7



Hg Release as a Function of LS Ratio, sample 7



As Release as a Function of LS Ratio, sample 7



Se Release as a Function of LS Ratio, sample 7

Figure 21. Fly ash leaching test results (Method 1316), sample 7 (brominated ACI).

9 Leaching Discussion

Leaching tests showed Hg stability in fly ash, both with and without ACI. In all leaching tests the Hg concentrations were below USEPA's total carbon (TC) RLs based on use of the TCLP (see and drinking water MCLs shown in Table 18 (see also ANNEX II).

Table 18. Limits of toxic elements contents in water.

	Regulatory limit TCLP, µg/L	Drinking water maximum contaminant levels (MCL), µg/L
Hg	200	2
As	5,000	10
Se	1,000	50

In contrast with the Hg, the As and Se leaching behavior is more complicated. Comparison of sample contents values (Table 17) and the leachate data indicates that the total content and the leachate concentrations are not correlated. Thus, the total As and Se element contents cannot provide the only basis for prediction of the leaching behavior. Through USEPA research one of the conclusions[16] indicated that total content is not a good predictor of environmental release. Historically total content has been used to predict environmental release. The use of LEAF data in fate and transport models allows for a more accurate prediction of environmental release of contaminants found in coal ash.

In this study, the variable responses of the eluate concentrations from leaching as functions of pH (Figures 16 through 18) and L/S (Figures 19 through 21) show that multiple LSP phenomena determine the observed behavior. The results show that the use of total content, single-point leaching tests (e.g., TCLP Method 1311) and similar approaches are not sufficient for prediction of leaching behavior over the wide range of potential use and disposal scenarios with various chemical and hydraulic conditions. The leaching behavior may be predicted with better accuracy by use of the LEAF test methods.

Analysis of results shows that in the leaching test the eluate As was higher than the drinking water MCL, and Se concentrations for some pHs were higher than the MCL. As stated earlier, the LEAF data are considered inputs to fate and transport modeling that take into account attenuation and dilution to determine potential concern to human health and the environment. If values, such as for Se analysis, are above the MCL, then fate and transport modeling is needed to determine if there is a potential concern to human health and the environment, taking into account local conditions such as topography, hydrology, precipitation, and environmental conditions encountered during coal ash management.

9.1 Findings and conclusions – leaching of particulate fly ash wastes

> *Note*: *Leach testing focused on the Leaching Environmental Assessment Framework (LEAF) test methods. These methods have been shown in previous studies to provide more data and better insight than the USEPA's Toxicity Characteristic Leaching Procedure (TCLP) in processes that influence the rate and extent of leaching of mercury (Hg), arsenic (As), and selenium (Se) from fly ash and AC mixes.*

1. In no cases did the LEAF tests on concentrations of Hg, As, or Se, in each of the three fly ash wastes (baseline waste, i.e., no sorbent used; standard AC waste; and, brominated AC waste) equal or exceed contaminant limits in USEPA's TCLP.

2. The LEAF tests indicated that Hg is strongly adsorbed onto the fly ash. For all three types of fly ash wastes, no exceedances were found of USEPA's standard for drinking water quality. This is expressed as the MCL of 2 µg/L.

3. The LEAF tests indicated that leachate concentrations of As from both baseline fly ash and AC-containing fly ashes did exceed the drinking water MRL of 10 µg/L by an order of magnitude.

4. The LEAF tests indicated that the Se concentrations in leachate for baseline fly ash were below the drinking water standard, except at the very highest pH values, where it was exceeded slightly. For standard AC fly ash wastes the MCL was exceeded slightly at the lower and higher end pH values. For brominated AC, the concentrations were slightly above the MCL only at the highest pH values.

5. This study indicated that pH is a key factor affecting As and Se leaching from fly ash. We note that these results using the LEAF method are similar to those obtained in previous U.S. studies for Hg, As and Se constituents from the three types of fly ash wastes that were tested.

6. Although these tests indicate that neither baseline fly ash nor fly ashes with the two sorbents are determined to be "hazardous wastes" using the TCLP and as defined by the TCLP RLs, in some cases following fate and transport modeling, the predicted concentrations exceed the MCL. In such cases consideration should be given, where appropriate, to treatment/storage measures, e.g., a lined municipal landfill, that could address any concern owing to leaching potential.

10 References

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ANNEX I: Fly Ash Leaching Test Results

Table AI.1. Fly ash leaching test results (EPA Method 1313), sample 1 (without ACI).

pH	1.95	4.40	5.35	7.22	8.09	9.11	10.43*	10.75	12.15	13.10
Hg, ng/L	41.0	12.2	9.4	8.8	8.8	12.5	9.2	8.5	8.0	9.8
As, µg/L	482.5	60.2	38.2	30.6	37.2	44.3	39.7	41.8	135.2	135.3
Se, µg /L	42.2	16.3	17.1	16.5	19.3	20.3	26.9	34.2	45.7	60.2

*Natural pH at LS 10 mL/g-dry (no acid/base addition).

Table AI.2. Fly ash leaching test results (EPA Method 1313), sample 2 (without ACI).

pH	2.05	4.22	5.10	7.15	8.12	9.09	10.33*	10.87	12.07	13.25
Hg, ng/L	49.2	12.4	11.4	10.2	15.2	17.0	11.2	10.5	12.2	14.8
As, µg /L	477.2	51.5	56.3	61.2	60.7	40.1	44.0	46.1	84.2	113.1
Se, µg /L	48.1	12.3	10.7	12.7	14.1	18.1	23.1	32.5	49.2	76.4

*Natural pH at LS 10 mL/g-dry (no acid/base addition).

Table AI.3. Fly ash leaching test results (EPA Method 1313), sample 3 (without ACI).

pH	2.25	4.25	5.05	7.43	8.25	9.09	10.12*	10.33	12.27	13.15
Hg, ng/L	262.1	49.2	26.4	12.8	25.7	22.0	16.3	16.8	17.1	19.2
As, µg /L	470.1	55.2	21.1	25.3	38.1	40.5	42.2	42.0	139.12	252.7
Se, µg /L	52.1	13.2	7.7	8.5	12.1	14.2	17.4	20.1	28.9	37.8

*Natural pH at LS 10 mL/g-dry (no acid/base addition).

Table AI.4. Fly ash leaching test results (EPA Method 1313), sample 4 (standard ACI).

pH	1.99	4.05	5.51	7.20	8.12	9.05	10.91	11.41*	12.05	13.08
Hg, ng/L	57.2	11.1	6.2	5.4	6.4	6.7	6.4	5.5	6.2	5.3
As, µg /L	377.2	86.7	43.5	41.2	67.6	171.2	193.4	233.4	288.7	315.7
Se, µg /L	47.3	23.1	20.1	18.1	23.4	23.3	27.4	32.4	41.1	50.3

*Natural pH at LS 10 mL/g-dry (no acid/base addition).

Table AI.5. Fly ash leaching test results (EPA Method 1313), sample 5 (standard ACI).

pH	2.25	4.08	5.05	7.41	8.25	9.33	10.15*	10.27	12.20	13.12
Hg, ng/L	41.1	8.8	6.2	4.8	6.4	9.7	10.7	10.5	9.8	10.1
As, µg /L	370.8	62.3	40.9	40.2	55.7	182.3	285.1	303.2	321.1	353.9
Se, µg /L	33.1	19.7	18.4	15.2	18.9	22.1	29.2	29.0	47.3	75.1

*Natural pH at LS 10 mL/g-dry (no acid/base addition).

Table AI.6. Fly ash leaching test results (EPA Method 1313), sample 6 (standard ACI).

pH	1.80	3.85	5.30	7.41	8.23	9.08	10.88	12.10*	12.22	13.14
Hg, ng/L	287.2	41.8	21.2	15.4	21.9	17.7	17.1	15.0	15.9	20.1
As, µg /L	305.1	50.1	40.1	43.5	71.7	135.1	147.3	215.1	235.3	281.5
Se, µg /L	82.1	25.1	19.3	17.7	20.1	24.2	37.7	45.4	55.9	86.2

*Natural pH at LS 10 mL/g-dry (no acid/base addition).

Table AI.7. Fly ash leaching test results (EPA Method 1313), sample 7 (brominated ACI).

pH	2.05	4.10	5.51	7.15	8.17	9.22	9.28*	10.31	12.05	13.07
Hg, ng/L	203.1	8.8	6.7	6.2	5.7	5.9	5.9	6.4	6.5	6.7
As, µg /L	234.5	27.2	25.1	33.2	190.0	263.1	241.1	224.7	143.2	125.5
Se, µg /L	55.1	10.2	12.1	7.5	20.2	26.3	30.1	34.7	69.1	80.1

*Natural pH at LS 10 mL/g-dry (no acid/base addition).

Table AI.8. Fly ash leaching test results (EPA Method 1313), sample 8 (brominated ACI).

pH	2.20	4.17	5.60	7.35	8.22	9.15*	9.21	10.24	12.19	13.25
Hg, ng/L	99.0	14.2	12.1	6.9	6.8	7.7	5.8	6.7	6.9	7.8
As, µg /L	271.7	20.1	23.1	31.1	210.1	271.9	288.2	265.3	166.1	137.2
Se, µg /L	50.1	8.8	14.3	11.0	15.3	17.1	18.7	39.3	71.5	74.4

*Natural pH at LS 10 mL/g-dry (no acid/base addition).

Table AI.9. Fly ash leaching test results (EPA Method 1313), sample 9 (brominated ACI).

pH	1.95	3.85	5.65	7.20	8.15	9.25*	9.28	10.37	12.25	12.97
Hg, ng/L	325.1	45.3	12.2	8.8	19.5	5.9	9.3	9.2	18.5	18.8
As, µg /L	259.1	22.1	15.1	17.9	72.5	140.9	167.1	163.2	137.5	113.1
Se, µg /L	54.1	15.5	14.8	13.1	20.5	23.3	25.5	47.1	81.7	86.2

*Natural pH at LS 10 mL/g-dry (no acid/base addition).

Table AI.10. Fly ash leaching test results (EPA Method 1316), sample 1 (without ACI).

L/S	0.5	1.0	2.0	5.0	10.0
pH	11.82	11.38	11.10	10.71	10.41
Hg, ng/L	122.2	81.3	22.7	8.1	10.2
As, µg /L	1,300	250.3	55	44	39.7
Se, µg /L	540.5	404.1	168.4	58.9	26.9

Table AI.11. Fly ash leaching test results (EPA Method 1316), sample 4 (standard ACI).

L/S	0.5	1.0	2.0	5.0	10.0
pH	12.73	12.42	12.01	11.59	11.41
Hg, ng/L	26.1	8.3	5.2	17.8	5.5
As, µg /L	3,077.1	1,566.6	803.4	283.3	233.4
Se, µg /L	225.1	121.7	54.3	16.2	12.4

Table AI.12. Fly ash leaching test results (EPA Method 1316), sample 7 (brominated ACI).

L/S	0.5	1.0	2.0	5.0	10.0
pH	10.65	10.28	10.01	9.58	9.28
Hg, ng/L	5.7	8.2	32.6	10.5	5.9
As, µg /L	717.1	401.1	367.2	303.1	241.1
Se, µg /L	140.1	125.4	98.2	82.5	70.1

ANNEX II: Descriptions of USEPA's TCLP and Maximum Contaminant Level Goal (MCLG)/Maximum Contaminant Level (MCL)

The following (excerpted verbatim from p. 181 of the USEPA's National Risk Management Research Laboratory's (NRMRL's) report, *Characterization of Coal Combustion Residues from Electric Utilities – Leaching and Characterization Data*; EPA-600/R-09/151, Dec. 2009) offers important context for its leaching results presented therein:

> "Provided below in a summary table for each coal combustion residual (CCR) category are the range of leach results over the pH range of 5.4 and 12.443, along with comparison to available regulatory or reference indicators including TC, MCL, and DWEL. In making such comparisons, it is critical to bear in mind that these test results represent an estimate of constituent release from the material as disposed or used on the land. They do not include any attempt to estimate the amount of constituent that may reach an aquifer or drinking water well. Leachate leaving a landfill is invariably diluted in ground water or constituent concentration attenuated by sorption and other chemical reactions in groundwater and sediment. Also, groundwater pH may be different from the pH at the site of contaminant release, and so the solubility and mobility of leached contaminants may change when they reach groundwater. None of these dilution or attenuation processes is incorporated into the leaching values presented, and so comparison with regulatory reference values, particularly drinking water values, must be done with caution."

- Toxicity Characteristic Leaching Procedure (TCLP):
  - A waste exhibits the "characteristic of toxicity" if the concentration of one or more of toxicity characteristic analytes in the TCLP's aqueous extract equals or exceeds the assigned regulatory action limits (see 40 Code of Federal Regulations [CFR] 261.24 and list of regulated constituents inserted below).
  - Regulatory Levels (RL) for each TCLP constituent was determined by multiplying Chronic Toxicity Reference Level (CTRL) by Dilution Attenuation Factor (DAF) i.e., RL = CTRL x DAF
  - The CTRL is a level below which health effects are not expected to occur, including drinking water standards MCL; or for carcinogens, the risk specific dose (RSD); or for noncarcinogens, the reference dose (RfD).
  - Using a subsurface fate and transport model (EPACML), the Agency also confirmed the adequacy of DAFs of 100 for all of the listed toxic constituents, which reflects an assumption that "drinking water wells were situated 500 feet down gradient from the [MSWLF] site," (45 FR 33111)
  - Regulatory limits represent a back-calculation from an acceptable CTRL in a receptor well, through the unsaturated and saturated zones, back to the source (i.e., the bottom of an unlined landfill) and are based on the assumptions that wastes would be co-disposed with actively decomposing MSW in a landfill, and that chemicals placed in a landfill leach at a uniform rate into the GW
  - TCLP is viewed by many as being inappropriate for assessing CCRs since it is a single-point pH leach test whose acetic acid leaching medium is designed to recreate the organic acid-leachate produced by the decomposition of putrescible wastes in a municipal solid waste landfill (MSWLF). Although co-disposal of CCRs with MSW is plausible, in the U.S., the vast majority of CCRs are not being managed through co-disposal with MSW (i.e., CCRs are often disposed of in monofills that are neutral or alkaline in nature).

- 40 CFR 261.24 Toxicity characteristics:
  - A solid waste (except manufactured gas plant waste) exhibits the characteristic of toxicity if, using the Toxicity Characteristic Leaching Procedure, test Method 1311 in ‘‘Test Methods for Evaluating Solid Waste, Physical/Chemical Methods,’’ USEPA Publication SW–846, as incorporated by reference in § 260.11 of this chapter, the extract from a representative sample of the waste contains any of the contaminants listed in table 1 at the concentration equal to or greater than the respective value given in that table. Where the waste contains less than 0.5% filterable solids, the waste itself, after filtering using the methodology outlined in Method 1311, is considered to be the extract for the purpose of this section.
  - A solid waste that exhibits the characteristic of toxicity has the USEPA Hazardous Waste Number specified in Table 1 which corresponds to the toxic contaminant causing it to be hazardous.

Table AII.1. Maximum concentration of contaminants for the toxicity characteristic (mg/L).

EPA HW No.[1]	Contaminant	CAS No.[2]	Regulatory Level (mg/L)
D004	Arsenic	7440–38–2	5.0
D005	Barium	7440–39–3	100.0
D018	Benzene	71–43–2	0.5
D006	Cadmium	7440–43–9	1.0
D019	Carbon tetrachloride	56–23–5	0.5
D020	Chlordane	57–74–9	0.03
D021	Chlorobenzene	108–90–7	100.0
D022	Chloroform	67–66–3	6.0
D007	Chromium	7440–47–3	5.0
D023	o-Cresol	95–48–7	[4]200.0
D024	m-Cresol	108–39–4	[4]200.0
D025	p-Cresol	106–44–5	[4]200.0
D026	Cresol		[4]200.0
D016	2,4-D	94–75–7	10.0
D027	1,4-Dichlorobenzene	106–46–7	7.5
D028	1,2-Dichloroethane	107–06–2	0.5
D029	1,1-Dichloroethylene	75–35–4	0.7
D030	2,4-Dinitrotoluene	121–14–2	[3]0.13
D012	Endrin	72–20–8	0.02
D031	Heptachlor (and its epoxide)	76–44–8	0.008
D032	Hexachlorobenzene	118–74–1	[3]0.13
D033	Hexachlorobutadiene	87–68–3	0.5
D034	Hexachloroethane	67–72–1	3.0
D008	Lead	7439–92–1	5.0
D013	Lindane	58–89–9	0.4
D009	Mercury	7439–97–6	0.2

EPA HW No.[1]	Contaminant	CAS No.[2]	Regulatory Level (mg/L)
D014	Methoxychlor	72–43–5	10.0
D035	Methyl ethyl ketone	78–93–3	200.0
D036	Nitrobenzene	98–95–3	2.0
D037	Pentrachlorophenol	87–86–5	100.0
D038	Pyridine	110–86–1	[3]5.0
D010	Selenium	7782–49–2	1.0
D011	Silver	7440–22–4	5.0
D039	Tetrachloroethylene	127–18–4	0.7
D015	Toxaphene	8001–35–2	0.5
D040	Trichloroethylene	79–01–6	0.5
D041	2,4,5-Trichlorophenol	95–95–4	400.0
D042	2,4,6-Trichlorophenol	88–06–2	2.0
D017	2,4,5-TP (Silvex)	93–72–1	1.0
D043	Vinyl chloride	75–01–4	0.2

[1] Hazardous waste number.

[2] Chemical abstracts service number.

[3] Quantitation limit is greater than the calculated regulatory level. The quantitation limit therefore becomes the regulatory level.

[4] If o-, m-, and p-Cresol concentrations cannot be differentiated, the total cresol (D026) concentration is used. The regulatory level of total cresol is 200 mg/L. [55 FR 11862, Mar. 29, 1990, as amended at 55 FR 22684, June 1, 1990; 55 FR 26987, June 29, 1990; 58 FR 46049, Aug. 31, 1993; 67 FR 11254, Mar. 13, 2002; 71 FR 40259, July 14, 2006]"

Maximum Contaminant Level Goal (MCLG) -- Means the maximum level of a contaminant in drinking water at which no known or anticipated adverse effect on the health of persons would occur, and which allows an adequate margin of safety. MCLGs are non-enforceable health goals (40 CFR 141.2). The MCLG is derived in a three-step process that includes the calculation of an RfD. The RfD is an estimate of the amount of a chemical that a person can be exposed to on a daily basis that is not anticipated to cause adverse systemic health effects over the person's lifetime. When determining an MCLG, EPA considers the risk to sensitive subpopulations (infants, children, the elderly, and those with compromised immune systems) of experiencing a variety of adverse health effects. (See 40 CFR §141.2 for codified definition.)

Maximum Contaminant Levels (MCLs) -- Are the enforceable threshold limits on the amount of each contaminant that is allowed in public water systems (i.e., serves as the maximum permissible level of a contaminant in water which can be delivered to any user of a public water system). The limit is usually expressed in milligrams or micrograms per liter of water with samples must be collected at each *entry point* to the *distribution system* To set a MCL for a contaminant, EPA first determines the MCLG and then set the limits as close to the MCLG as feasible after considering difficulties in measuring small quantities of a contaminant, whether there's a lack of available treatment technologies or whether the costs of treatment would outweigh the public health benefits of a lower MCL. In the last case, EPA is permitted to choose an MCL that balances the cost of treatment with the public health benefits. For some contaminants, USEPA establishes a Treatment Technique (TT) instead of an MCL. TTs are enforceable procedures that drinking water systems must follow in treating their water for a contaminant.

Some contaminants may cause aesthetic problems with drinking water, such as the presence of unpleasant tastes or odors, or cosmetic problems, such as tooth discoloration. Since these contaminants do not cause health problems, there are no legally enforceable limits on their presence in drinking water. However, EPA recommends maximum levels of these contaminants in drinking water. These recommendations are called National Secondary Drinking Water Regulations (NSDWRs), or secondary standards. (See 40 CFR §141.2 for codified definition.)