

Air Quality II Conference

Introduction

The Air Quality II Conference (AQII) on mercury (Hg) and fine particulates was held September 19–21, 2000, in McLean, Virginia. The reduction of Hg emissions from coal-fired utility boilers is an immediate concern since the U.S. Environmental Protection Agency's (EPA) December 2000 determination that control is required under Section 112 of the Clean Air Act. Based on presentations and discussions at AQII, it appears that there are new technologies under development, but to date none has been shown to provide widely effective control at acceptable cost. Only within the last few years has a fundamental understanding of the chemical transformations and capture mechanisms of Hg begun to emerge, which should guide the development of more effective control methods. The following is a brief overview and status of the Hg issues discussed at AQII.

Mercury Regulations and Cost

EPA's 2000 determination requires a proposed regulation by December 2003, a final rule by December 2004, and implementation by the end of 2007. Future regulatory determination will be influenced by the amounts and forms of Hg emitted from coal-fired boilers and the capabilities of control technologies (1, 2a). Two major reports on Hg prepared by EPA for the U.S. Congress, *the Mercury Study Report to Congress* (1) and *the Utility Hazardous Air Pollutant Report to Congress* (3), identified coal-fired utility boilers as the largest single category of Hg emissions in the United States, accounting for about one-third of the total anthropogenic emissions. Further assessment based on EPA's 1999 Information Collection Request (ICR) indicates that approximately 40% of the 75 tons of Hg contained in the coal burned in the United States is captured in ash and scrubber residues and 60% is emitted to the atmosphere (2b, 2c). The percentage emitted by different plants varies from about 10% to 90%. Since U.S. emissions account for only 3% of the worldwide total (3), effective control will require international agreements such as the Great Lakes Binational Toxics Strategy signed by the United States and Canada in April 1997.

Several regulatory approaches have been suggested. A regulatory approach that allows emissions trading under a cap on total Hg emissions would minimize cost by exempting plants with low emissions from

further control and targeting reduction on plants with relatively high emissions and low unit control costs. The drawback of this approach is that certain sources would likely not be controlled.

An alternative to cap and trade would be a percentage reduction requirement. Combinations of available methods can provide up to 90% control for some plants but not for others. Costs for 90% control using activated carbon injection are estimated by EPA at \$5000–\$28,000 per pound of Hg removed and by the U.S. Department of Energy (DOE) at \$25,000–\$70,000 per pound of Hg removed (4), which corresponds to an increase in power-generating costs of 0.2–6 mil/kWh.

Policy discussions at AQII indicated growing support for a multipollutant approach to achieve greater improvement in environmental quality, reduce compliance cost, and provide a more consistent regulatory framework for compliance decisions. Industry would benefit if consistent rules could be maintained for up to 10 years (2d). Environmental advocates look at multipollutant control as a way to achieve greater overall improvement in the quality of the environment (2e, 2f). Strategic trade-offs could be part of a multipollutant compliance strategy, e.g., regulations that favor the installation of additional scrubbers to control both SO₂ and Hg emissions.

Health Effects

A Congressionally mandated study of the toxicological effects of Hg issued by the National Research Council (NRC) in August 2000 reaffirmed EPA's Hg exposure reference dose (RfD) of 0.1 µg/kg/day as the level necessary to protect public health (5). The ingested methylmercury from fish consumption, which is estimated at 1–6 µg/kg Hg/day, is converted to mercuric Hg, which is slowly eliminated from the kidneys but remains fixed in the brain indefinitely (2g). Epidemiological studies conducted in the Faroe Islands and New Zealand have shown an association between chronic, low-dose prenatal exposure to methylmercury and neurodevelopmental effects on attention, motor function, language, visual-spatial, and verbal abilities, which were, however, not seen in a third study conducted in the Seychelles Islands. The Faroe Islands study was used in the NCR study because of its quantifiable findings for a large study

population and extensive peer review. The RfD of 0.1 µg/kg/day represents the 5% statistical lower bound for the most sensitive response, which was the Boston Naming Test, and an uncertainty factor of 10 to account for biological variability and database insufficiencies.

Emission and Transport of Mercury

Mercury is a multimedia pollutant that is emitted, deposited, and reemitted on a local and global scale in both terrestrial and marine environments (2h). Elemental Hg remains in the atmosphere for up to a year and can be transported transcontinentally, whereas oxidized gaseous and particulate forms are deposited near their source (2i, 2j). Approximately 47% of the Hg deposited in the northeastern United States comes from within that region, 30% from other U.S. sources, and 23% from global sources (2k).

Source-receptor models show that Hg deposition is concentrated in populated areas close to anthropogenic sources (2i). Asian emissions, which represent about half of the world's total, account for only 10%–20% of deposition of Hg on the West Coast of the United States. Nonanthropogenic sources contribute only 4%–7% to deposition in the eastern United States and 10%–12% in the West. Hence, most Hg deposition appears to be generated from anthropogenic sources close to the point of deposition.

Information on the amount of Hg in U.S. coals and Hg emissions from coal-fired utility plants was obtained from the EPA three-part ICR. Part I identified the size and configuration of all coal-fired utility boilers and associated pollution control devices. Part II obtained data quarterly on the origin, quantity, and analysis of coal shipments delivered to generating units with a capacity greater than 25 MW_e, including a minimum of three analyses per month for Hg and Cl contents, together with any other available analyses on ash, sulfur, heating value, etc. Part III required emission tests on 84 generating units selected at random from 36 categories representing different plant configurations and coal ranks to measure total and speciated Hg concentrations in the flue gas before and after the final air pollution control device upstream of the stack. Over 40,000 fuel samples were analyzed by many different laboratories using a range of sampling and

analysis methods. A round-robin evaluation of the commonly used methods showed that about one-third of the analyses fell below reliable lower quantitative limits, which were 0.060 ppm for Hg and 220 ppm for Cl (2c). Error analysis indicated that this imprecision had little effect on the calculated values of total Hg in coal or the total annual Hg emission, but did influence the accuracy of estimates for some individual plants.

The table shown provides a summary of ICR coal data by point of origin for six regions and corresponding coal ranks. The total Hg tonnage of 75.1 tons agrees with the more rigorous estimates performed by EPRI and EPA (2b, 2c). Appalachian bituminous and western subbituminous coals, which together account for 75% of U.S. coal production and 81% of the Hg entering coal-fired plants, have very different analyses affecting Hg emissions. Appalachian coals have high Hg, Cl, and S contents and low Ca content. Subbituminous coals typically have low concentrations of Hg, Cl, and S and a high Ca content.

Recent estimates of Hg emissions based on ICR data (2c, 2l) compare favorably with EPA's 1996 estimates used in *Utility Hazardous Air Pollutant Report to Congress* (3). Units with cold electrostatic precipitation (ESP), either alone or combined with wet flue gas desulfurization (FGD), represent 74% of the Hg entering U.S. plants, and their removal estimates have a predominant effect on the calculation of total Hg emissions. EPRI's estimates, based on separate calculation for each control category, predict a total annual Hg emission of 49 tons before correcting for Cl content. The 1996 EPA removals by category would lower the total to 45 tons, and Laumb's estimates (2l), based on multiple regression analysis, would lower the total emission to 40 tons. The EPRI value of 49 tons compares most favorably with EPA's earlier estimate of 51.3 tons for 1994 (3).

Wide variations in removals were observed within each of the ICR control categories, indicating that effects due to fuel properties and operating variables need to be considered to arrive at more exact predictions. Differences by coal rank are most immediately evident. For example, removal across a cold ESP averaged 35% for bituminous coal compared to 10% for low-rank coal (LRC) (lignite and bituminous), and removal across a cold ESP followed by wet FGD averaged 65% for bituminous compared to 35% for LRC. These differences reflect an effect from Cl content, which can introduce a bias in the estimate of total Hg emissions. An analysis of the distribution of ICR coal

Regional Summary of ICR Data on Hg in Utility Coal

Region/Rank	Hg, ppm	Hg, lb/TBtu	Cl, ppm	Utility Coal Produced, MMt	Hg in Utility Coal, t
Appalachian Bituminous	0.126	9.5	948	342	42.1
Interior Bituminous	0.086	6.6	1348	67	5.4
Western Bituminous	0.049	3.9	215	75	3.5
Western Subbituminous	0.068	5.7	124	336	18.4
Fort Union Lignite	0.088	8.3	139	23	1.3
Gulf Coast Lignite	0.119	12.5	221	57	4.5
Totals				900	75.2

Cl analysis performed by EPRI showed that Cl contents were lower in Part III emission tests than in the more broadly representative Part II coal sampling (2c). To correct this bias, regression correlations expressing Hg removal and speciation as a function of Cl content were developed for ten control categories. These regression correlations were subsequently used to recalculate Hg emissions as a function of coal Hg and Cl contents, which predicted that a more accurate estimate of the total annual Hg emission from all plants would be approximately 45 tons.

Measurement of Mercury Emissions

Methods for analyzing Hg emissions from coal-fired boilers have advanced rapidly in the past three years. The Ontario Hydro (OH) wet-chemical method for measuring total and speciated Hg, which was selected by EPA for making flue gas measurements under the ICR, has been submitted to the American Society for Testing and Materials (ASTM) for adoption as a standard reference method. A modified solid OH method that substitutes solid reagent cartridges for liquid impingers is under development and appears to provide good agreement with the wet-chemical method on Hg⁰ and Hg²⁺, but does not measure Hg_p (2m).

Continuous emission monitors (CEMs) for Hg will be needed at coal-fired power plants to provide real-time measurements for control and compliance monitoring. Robust instruments suitable for routine use in power plants are expected to be on the market within two to five years, in time for the implementation of Hg controls in 2007. CEM measurements on coal-derived flue gases are difficult, because of the low concentrations of Hg and the presence of fine particulate and acid gases, which interfere with measurements. The available instruments measure either Hg⁰ or total Hg and must be equipped with a pretreatment system to measure Hg²⁺ by difference. They do not measure Hg_p, and there is concern that fly ash particles collected on pretreatment filters may be

adsorbing or oxidizing vapor-phase Hg. Eight leading CEMs under development are variously based on cold-vapor atomic absorption, atomic fluorescence (AF), Zeeman-modulated atomic absorption spectroscopy, and differential optical atomic spectroscopy (2n). CEMs are being evaluated under EPA's Environmental Technology Verification program (2o), but results have not yet been published.

Mercury Transformations During Combustion

Mercury transformations were reviewed to identify fundamental mechanisms that may explain Hg⁰/Hg²⁺ variability in boiler systems (2p). Mercury in coal commonly occurs in association with pyrite (FeS₂) and cinnabar (HgS) and may also be organically bound to coal macerals. All forms of Hg decompose in the combustion flame to form Hg⁰. Equilibrium predicts almost complete conversion to oxidized forms of volatile Hg²⁺X and Hg_p upon cooling to 400°C, but measurements of 35%–95% oxidation for different coals indicate that conversion is kinetically controlled. High levels of Hg oxidation correlated with high Cl in coal and indicate that chlorination is a predominant oxidation mechanism. Formation of HgCl₂ is believed to involve the homogeneous reaction of Hg⁰ with atomic chlorine. Other transformation reactions have been suggested to involve acid gases such as HCl, NO₂, and SO₂ and catalytic sites on fly ash and unburned carbon.

Studies on the oxidation and capture of Hg show that individual mineral and gas constituents produce different results depending on the coal being burned. Adding HCl to flue gas from a Powder River Basin (PRB) subbituminous coal did not promote oxidation, nor did cofiring with a high-Cl biomass (2p). Addition of calcium acetate to PRB coal increased the emission of Hg⁰ by further inhibiting the oxidizing effect of coal Cl. NO₂ promoted the oxidation of Hg⁰ in tests on high-Cl Blacksville bituminous, but not for PRB coal (2n). Increased Hg capture on the finer fractions of PRB fly ash was proportional to the reciprocal of particle diameter, suggesting that adsorption occurred by a slow surface reaction. Al₂O₃ and TiO₂ promoted oxidation in N₂-O₂ and Ar-O₂ atmospheres, but not in coal-derived flue gas. Hematite (alpha-Fe₂O₃) has been reported to promote Hg oxidation in simulated flue gases (6a, 7, 6b–c), but addition of hematite and NO₂ did not promote oxidation of Hg⁰ for either PRB coal or North Dakota lignite (6d). A broad range of experiments

suggests that the fundamental mechanisms of Hg oxidation are substantially different for western LRCs and eastern bituminous coals, owing to differences in Hg, Cl, S, Ca, and Fe contents.

Control Technology Overview

Since 1995, DOE, EPRI, electric utilities, and research organizations such as the EERC have collaborated in an accelerated effort to develop cost-effective Hg control options that involve either improving Hg capture in existing air pollution control devices, perfecting sorbent injection technology, or identifying entirely new and novel methods for Hg control (2q–2t). For some plants, existing technologies may prove to be effective; for others, new technologies will be required. Below is a brief description of how well existing technologies work in a broad sense and some of the new technologies that are under development.

Trends in Mercury Control Using Existing Technologies

Trends in Hg speciation and subsequent removal (control) are largely derived from Part III of the ICR data and are summarized below.

Boilers – On average, the percentage of coal Hg emitted at the furnace exit ahead of a particulate control device was close to 100% in the 18 ICR tests on wall-fired and tangentially fired boilers where these measurements were made (2j). EPA's previous estimates of emission modification factors (EMFs, or % emission) for pc-fired furnaces ranged from 41%–94% and were lower for units equipped with NO_x controls (3). The data suggest that no substantial amounts of Hg are retained in the furnace or in the bottom ash of a pc-fired boiler, but cyclone-fired boilers and fluidized-bed combustors (FBCs) do appear to retain Hg (2j). An average EMF of 66% was determined for four cyclone-fired units without NO_x control in ICR tests, compared to EPA values of 54% with NO_x control and 93% without. The average EMF for four FBCs in ICR tests was 79%.

Mercury speciation at the furnace exit was principally influenced by coal Cl content and temperature (2j), with the percentage of Hg leaving the furnace in elemental form dropping sharply from >85% to about 10% for coal Cl contents greater than 150–200 ppm. At the higher temperatures of a hot-side ESP, 250°–400°C, the percentage of Hg⁰ remained high even at high levels of Cl, and virtually no Hg_p was formed. At the lower temperatures of a cold-side ESP, 130°–170°C, Hg was largely converted to an oxidized form at high Cl levels and was typically 75% particulate.

Particulate Control Devices – Three-fourths of all coal-fired power plants have only particulate controls. Two-thirds of the Hg in coal used by the power industry goes to plants having either a cold- or hot-side ESP or a fabric filter. Average Hg removals for these three categories in the ICR were 27%, 4%, and 58%, and the respective percentages of Hg exiting in elemental form were 47%, 66%, and 23%. Higher levels of coal Cl content were significantly correlated with an increase in Hg capture and a decrease in percent Hg⁰ for all classes of particulate control (2c, 8). For cold-side ESPs only, higher levels of coal S were correlated with reduced Hg capture.

ICR data have been reviewed to identify changes in Hg speciation across the three principal particulate control devices (2j). Substantially all of the Hg_p entering either a cold-side ESP or a fabric filter was removed, whereas essentially no Hg_p was present at the inlet of a hot-side ESP, owing to its higher temperature. Changes in gaseous Hg²⁺ differed depending on coal Cl content. Below 200 ppm Cl, an increase in Hg²⁺ suggested that Hg⁰ was oxidized but not removed. At higher Cl levels, Hg²⁺ removals increased along with coal Cl for fabric filters but not for ESPs. Removal of total gaseous Hg across a fabric filter was close to zero below 200 ppm coal Cl, but increased to an average of 73% over the range of 200–1400 ppm Cl. Fabric filters were shown to remove significant percentages of both Hg⁰ and gaseous Hg²⁺, owing to the oxidation and capture that occurs because of excellent gas–solids contact across the dust cake on the filter. No significant change in total gaseous Hg was observed across a cold-side ESP.

Wet Flue Gas Desulfurization (FGD) – Wet FGD systems are installed on 151 U.S. power plants representing about 25% of coal-fired utility generating capacity and a similar percentage of the total Hg entering utility boilers. ICR tests on wet FGD included units where the scrubber followed either a cold-side ESP (11 sites), a hot-side ESP (six sites), or a fabric filter (two sites). Performance across the scrubber itself was consistent for nearly all sites, showing removal of about 90% of the gaseous Hg²⁺ entering. Elemental Hg was not removed and appeared to increase slightly in some tests, possibly due to chemical reduction of ionic Hg by sulfite in the scrubber solution followed by reemission as Hg⁰ (2j).

Mercury removals across scrubbers and their associated particulate control devices were again strongly influenced by the coal Cl content (2c, 8). On average,

removals across the FGD alone approximately doubled from 30%–60% between 50 and 1000 ppm coal Cl following a cold-side ESP and increased from about 20%–50% between 200 and 1000 ppm Cl following a hot-side ESP (8). Total removal averaged 88% for FGD following a fabric filter, where the fabric filter contributed 58% removal and 77% of the Hg entering the scrubber was in an oxidized form. Removal for FGD following a cold-side ESP averaged 49%, where the ESP contributed 27% removal, and 53% of the Hg entering the scrubber was in an oxidized form. Removal averaged only 26% for FGD following a hot-side ESP, where the hot-side ESP contributed only 4% removal, and 34% of the Hg entering the scrubber was in an oxidized form.

Spray Dryer Absorbers – Spray dryer absorbers, which use an alkaline slurry to absorb SO₂, also absorb about 90% of the gaseous Hg²⁺. The dry sorbent and fly ash particulates generated are captured in either a fabric filter or an ESP. Total Hg removal across a spray dryer and fabric filter ranged from 0 to 99% at ten ICR test sites (38% average), with removals being either very low at coal Cl contents below 100 ppm or very high above 900 ppm Cl (8). Total Hg removal across a spray dryer following an ESP averaged 18% at three ICR test sites, with no evident trend in relation to coal Cl content.

Fluidized-Bed Combustors (FBCs) with Fabric Filters – FBCs with fabric filtration represented the highest Hg removals in the ICR data, ranging from 66%–99% and averaging 86%, due in part to the relatively high carbon content of the FBC bed ash (8). The elemental fraction in the Hg emission is higher than for a fabric filter alone, averaging 56% versus 23%, due to removal of Cl by Ca in the bed.

Two integrated gasification combined cycle (IGCC) plants were indicated to have essentially zero Hg removal, with 96% of the Hg emitted in elemental form (8). Reducing conditions in the gasifiers kept Hg in its volatile elemental state, and acid gas-cleaning systems used to remove Cl and S had little effect on downstream speciation or capture. Combustion in the gas turbine in an atmosphere free of Cl and fly ash did not promote the oxidation of Hg.

Selective Catalytic Reduction (SCR) – Installation of SCR or selective noncatalytic reduction (SNCR) for reduction of NO_x between now and 2010 could potentially increase oxidation and improve Hg removal in U.S. coal-fired plants. ICR tests performed at two plants with SCRs and three plants with SNCRs did not provide conclusive answers because of masking

effects by other variables. European investigators have reported a decrease in the elemental fraction of Hg, from 40%–60% to 2%–12%, in full-scale plants (9). Recent pilot-scale tests at the EERC showed an increase in the fraction of Hg_p across the SCR when burning two high-Cl eastern bituminous coals, but essentially no effect on speciation when burning a low-Cl PRB subbituminous coal (2n). Coal-specific effects appear to be related to the chloride, S, and Ca content of the coal. Further tests at full-scale plants are planned to confirm pilot results.

Development of New Mercury Control Technologies

In the last five years, there has been a focused effort to develop new Hg control technologies that are both effective at capturing Hg and economical to implement. Examples of technologies under development or testing include advanced fuel-cleaning processes, activated carbons, fly ash, zeolites, sulfur, ammonia, regenerable sorbents, in situ generated sorbents, oxidation catalysts, additives for improved capture in wet scrubbers or spray dryers, and corona discharge methods (2u). Recognizing that cost and ease of installation are primary considerations, many of the development efforts have focused on sorbent technologies utilizing either novel or commercially available sorbents or fly ash. Laboratory adsorption capacities at 150°C have been determined to be about 50 µg Hg/g for fly ashes, 200 µg Hg/g for activated carbons, and 800–1000 µg Hg/g for FGD solids. Modeling predicts Hg removals for sorbent injection upstream of an ESP between 0 and 60% for a residence time of 1 sec and up to 80% for 2 sec at injection rates of 0–4 lb/MMacf, corresponding to sorbent:Hg ratios of 0 to 110,000:1 for a coal containing 0.1 ppm Hg. Predicted Hg removals for injection upstream of a fabric filter approach 95% for a 60-min cleaning cycle. Field test results for sorbent injection ahead of a fabric filter show little change in removal between 127° and 163°C where sorbent capacities were adequate. A more thorough discussion on activated carbons and fly ash is provided below, given the attention and priority they have received.

Mercury Removal on Fly Ash – Mercury removals while western coals were burned without sorbent injection in four full-scale boilers operated by Public Service of Colorado ranged from a low of 28% for an ESP to 61%–99% for units equipped with reverse-gas baghouses (2v). Higher Hg removals were observed where there was a high level of unburned carbon in the fly ash (high loss on ignition, LOI). Pilot tests rein-

jecting power plant fly ashes ahead of a baghouse indicated removals of 13%–80%, and again removals increased along with LOI. Both carbon and Hg were enriched in the smaller size fractions of the fly ash, suggesting that size separation could be used to minimize reinjection rates, reduce cost, and avoid operating problems. One low-carbon fly ash showed a significant Hg sorption capacity even after heating to remove all carbon. Spray cooling to 110°C improved Hg capture. Fly ash reinjection was estimated to offer potential savings of 80% over using activated carbon. Carbon contents above 1% from either LOI or activated carbon injection may adversely affect the sale of fly ash.

Mercury Capture on Activated Carbon – Pilot tests run by ADA at Public Service of Colorado on a lignite-derived Norit Darco FGD activated carbon showed Hg removals ranging from 30%–90% for carbon injection rates of 0.5–4.5 lb/MMacf (equivalent to carbon-to-mercury ratios of ca. 1300–11,500:1 for coal containing 0.1 ppm Hg) (10). Removal increased along with injection rate and as gas temperatures were reduced from 177°–104°C and was limited to 70% or less for injection ahead of an ESP. Injection ahead of a baghouse provided 70%–90% removal at injection rates as low as 1 lb/MMacf, with little increase at higher rates, owing to the predominating influence of capture on PRB fly ash.

Laboratory tests have evaluated Hg adsorption capacities of activated carbons derived from lignite or bituminous coal and other sorbents including iodine- or sulfur-impregnated carbons and mineral compounds. The similar capacities reported for most activated carbons when tested in nitrogen limited their value because of the effect of flue gas constituents. Tests on combinations of SO₂, HCl, NO, and NO₂ in synthetic flue gas identified an interaction between SO₂ and NO₂ that severely impaired the capture of Hg⁰, whereas HCl, NO, and NO₂ individually or in combination enhanced Hg capture (2w, 11). Sorption capacity was found to be inversely proportional to the concentrations of both SO₂ and NO₂, with changes in capacity noted at concentrations as low as 100 ppm SO₂ and 2.5 ppm NO₂ (2x). With NO₂ present, oxidation and capture of Hg⁰ occur in the absence of O₂ and HCl, indicating that NO₂ can provide the electron sink for the surface oxidation of Hg. Removal of water from flue gas enhances Hg capture, but subsequent introduction of even a small amount of moisture causes the immediate release of Hg²⁺, suggesting that Hg is captured in a

nonvolatile form and is released as a volatile hydrate. Elemental Hg continues to be converted to a volatile oxidized form after a test carbon reaches its maximum capacity (breakthrough), and some of the Hg initially captured is slowly released over time if the carbon remains in flue gas containing SO₂ and NO₂.

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* For more complete reference information, please visit the EERC CATM web page at <http://www.undeerc.org/catm/cathome.html>.

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