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MEMORANDUM

Subject: Control of Mercury Emissions from Coal-fired Utility Boilers

From: Frank T. Princiotta /s/
Director

To: John Seitz, Director
Office of Air Quality Planning and Standards (MD-10)

Attached is a technical memorandum that summarizes current knowledge on the control of mercury emissions from coal-fired utility boilers. This memorandum contains a summary of APPCD studies that were designed to inform EPA's determination on the control of toxic air pollution emissions from electric utility power plants. The memorandum summarizes information on: coals and technologies used at coal-fired power plants, mercury capture methods, potential retrofit technologies, the costs of mercury retrofits, the evaluation of ICR data, the behavior of mercury in coal combustion by-products, and recommendations.

A companion APPCD technology report that provides more detailed information has been peer reviewed by OAR, OAQPS and ORD staff members and is being prepared for publication by December 15, 2000. The attached memo is, in essence, an executive summary of that report.

APPCD's mercury control technology team has established an excellent working partnership with your staff during our mercury control technology studies. We hope to build on this relationship by helping to resolve future air emissions concerns with your Office.

Attachment

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Technical Memorandum

Control of Mercury Emissions from Coal-fired Electric Utility Boilers

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Summary

The National Risk Management Research Laboratory (NRMRL), Air Pollution Prevention and Control Division (APPCD) is conducting research to develop and evaluate mercury and multi-pollutant emission control technologies for stationary combustion sources. A primary objective of this research is to support the Office of Air Quality Planning and Standards (OAQPS) in the development of control technologies for coal-fired utility boilers.

This memorandum summarizes studies that were conducted to support EPA's decision on the need to regulate toxic air pollution emissions from electric utility boilers. These studies included: performance and cost estimates of retrofit technologies for the control of mercury emissions from existing plants; an evaluation of the information collection request (ICR) data to determine the performance of existing controls in capturing mercury; and a review of existing information on the stability of mercury contained in the residues and residue by-products for coal-fired power plants.

Coals and Technologies Used at Coal-fired Power Plants

In 1999, utility boilers in the U.S. burned 786 million tons of coal; of which about 52% was bituminous and 37% was subbituminous. Other fuels included lignite, anthracite, reclaimed waste coal, mixtures of coal and petroleum coke (pet-coke), and mixtures of coal and tire derived fuel (TDF). Pulverized coal-fired (PC) boilers represent approximately 86% of the total number and 90% of total utility boiler capacity. Based on capacity, other types of boilers include cyclone-fired boilers (7.6%), fluidized-bed combustors (1.3%), and stoker-fired boilers (1.0%).

The 1999 ICR responses indicate that a variety of emission control technologies are employed to meet requirements for sulfur dioxide (SO₂), nitrogen oxides (NO_x) and particulate matter (PM). Most utilities control NO_x by combustion modification techniques and SO₂ by the use of compliance coal. For post-combustion controls, 77.4% of units by number have PM

control only, 18.6% have both PM and SO₂ controls, 2.5% have PM and NO_x controls, and 1.3% have three post combustion control devices.

For PM control, electrostatic precipitators (ESPs) are found on 84% of units and fabric filters (FFs) on 14%. Post combustion SO₂ controls are less common. Wet flue gas desulfurization (FGD) systems are used on 15.1% of the units and spray dryer absorbers (SDA) are used on 4.6% of units surveyed. While the application of post-combustion NO_x controls are becoming more prevalent, only 3.8% of units used either selective non-catalytic reduction (SNCR) or selective catalytic reduction (SCR) systems in 1999.

Mercury Capture Methods

The capture of mercury by flue gas cleaning devices is dependent on the chemical and physical forms of mercury. There are three basic forms of mercury in the flue gas from coal combustion: elemental mercury (Hg⁰), ionic mercury [Hg(II)], and particulate-bound mercury [Hg(p)]. Both Hg⁰ and Hg(II) are in a vapor phase at flue gas cleaning temperatures. Hg⁰ is insoluble in water and cannot be captured in wet scrubbers. The predominate Hg(II) compounds in coal flue gas are weakly to strongly soluble and can be generally captured in wet FGD scrubbers. Both Hg⁰ and Hg(II) can be adsorbed onto porous solids such as fly ash, PAC, or calcium-based acid gas sorbents for subsequent collection in a PM control device. Hg(II) is easier to adsorb than Hg⁰. Hg(p) is attached to solids that can be readily captured in ESPs and FFs.

"Speciation" is a term used to denote the amounts of Hg⁰, Hg(II), and Hg(p) in flue gas. The Ontario-hydro (OH) modified method is the only method that is currently recognized for the measurement of mercury speciation in flue gas from the combustion of coal.

Flue gas cleaning technologies that are used on combustion sources use three basic methods to capture mercury: the capture of Hg(p) in PM control devices; the adsorption of Hg⁰ and Hg(II) onto entrained sorbents for subsequent capture in PM control devices; and the solvation of Hg(II) in wet scrubbers. Factors that affect the speciation and capture of mercury in coal-fired combustion systems include the type and properties of coal, the combustion conditions, the type of flue gas cleaning technologies employed, and the temperatures at which the flue gas cleaning systems operate.

Potential Retrofit Technologies

A practical approach to controlling mercury emissions at existing utility plants is to minimize capital costs by adapting or retrofitting the existing equipment to capture mercury. Potential retrofit options for control of mercury were investigated for units that currently use the following post combustion emission control methods: (1) ESPs or FFs for control of PM; (2) dry FGD scrubbers for control of PM and SO₂; and (3) wet FGD scrubbers for the control of PM and SO₂.

ESP and FF Systems

ESPs and FFs are either cold-side or hot-side devices. Hot-side devices are installed upstream of the air pre-heater while cold-side devices are installed downstream. Flue gas temperatures in hot-side devices typically range from 350 to 450 °C while cold-side devices typically operate at temperatures ranging from 140 to 160 °C. Based on current information, it appears that little mercury can be captured in hot-side ESPs or FFs.

Least cost retrofit options for the control of mercury emissions from units with ESP or FF are believed to include:

1. Injection of a sorbent upstream of the ESP or FF. Cooling of the stack gas or modifications to the ducting may be needed to keep sorbent requirements at acceptable levels.
2. Injection of a sorbent between the ESP and a pulsejet FF retrofitted downstream of the ESP. This approach will increase capitol costs but reduce sorbent costs.
3. Installation of a semi-dry circulating fluidized-bed absorber (CFA) upstream of an existing ESP used in conjunction with sorbent injection. The CFA re-circulates both fly ash and sorbent to create an entrained bed with a high number of reaction sites. This leads to higher sorbent utilization and enhanced fly ash capture of mercury and other pollutants.

Units equipped with a FF require less sorbent than units equipped with an ESP. ESP systems depend on in-flight adsorption of mercury by entrained fly ash or sorbent particles. FFs obtain in-flight capture and capture as the flue gas passes through the FF.

In general, the successful application of cost effective sorbent injection technologies for ESP and FF units will depend on: (1) the development of lower cost and/or higher performing sorbents; and (2) appropriate modifications to the operating conditions or equipment being currently used to control emission of PM, NO_x, and SO₂.

Dry FGD Systems

SDA systems that use calcium based sorbents are the most common dry FGD systems used in the utility industry. An aqueous slurry containing the sorbent is sprayed into an absorber vessel where the flue gas reacts with the drying slurry droplets. The resulting, particle-laden, dry flue gas then flows to an ESP or a FF where fly ash and SO₂ reaction products are collected.

CFAs are “vertical duct absorbers” that allow simultaneous gas cooling, sorbent injection and recycle, and gas absorption by flash drying of wet lime reagents. It is believed that CFAs can potentially control mercury emissions at costs lower than those associated with use of spray dryers.

Dry FGD systems are already equipped to control emissions of SO₂ and PM. The modification of these units by the use of appropriate sorbents for the capture of mercury and other air toxics is considered to be the easiest retrofit problem to solve.

Wet FGD Systems

Wet FGD systems are typically installed downstream of an ESP or FF. Wet limestone FGD scrubbers are the most commonly used scrubbers on coal-fired utility boilers. These FGD units generally capture more than 90% of the Hg(II) in the flue gas entering the scrubber. Consequently, existing wet FGD scrubbers may lower mercury emissions by about 20% to more than 80%, depending on the speciation of mercury in the inlet flue gas.

Improvements in wet scrubber performance in capturing mercury depend primarily on the oxidation of Hg⁰ to Hg(II). This may be accomplished by (1) the injection of appropriate oxidizing agents or (2) the installation of fixed oxidizing catalysts upstream of the scrubber to promote oxidization of Hg⁰ to soluble species.

An alternative strategy for controlling Hg emissions from wet FGD scrubbing systems is to inject sorbents upstream of the PM control device. In wet FGD systems equipped with ESPs, performance gains are limited by the in-flight oxidization of Hg⁰, and the in-flight capture of Hg(II) and Hg⁰. In systems equipped with FFs increased oxidization and capture of mercury can be achieved as the flue gas flows through the FF. Increased oxidization of Hg⁰ in the FF will result in increased mercury removal in the downstream scrubber.

The Costs of Mercury Retrofits

Annualized costs of mercury controls using powdered activated carbon (PAC) injection have been estimated based on recent pilot-scale evaluations with commercially available adsorbents (see Table 1). These control costs range from 0.31 to 3.78 mills/kWh, with the highest costs associated with plants having hot-side electrostatic precipitators (HS-ESPs). For plants representing 89% of current capacity and using controls other than HS-ESPs, the costs range from 0.31 to 1.92 mills/kWh. Assuming a 40% reduction in sorbent costs by use of a composite lime-PAC sorbent for mercury removal, cost projections range from 0.18 to 2.27 mills/kWh with higher costs again being associated with plants using HS-ESPs.

In comparison the estimated annual costs of mercury controls, as a function of plant size, lie mostly between the costs for low NO_x burners (LNBS) and SCR systems. The costs of mercury control will dramatically diminish if retrofit hardware and sorbents are employed for control of other pollutants such as NO_x, SO₂, or fine PM.

The performance and cost estimates of PAC injection-based mercury control technologies presented in this memorandum are based on relatively few data points from pilot-scale tests and are considered to be preliminary. However, based on pilot-scale tests and the results of ICR data

evaluations, we are confident that the better sorbents and technologies now being developed will reduce the costs of mercury controls beyond current estimates.

Within the next 2-3 years, the evaluation of retrofit technologies at plants where co-control is being practiced will lead to a more thorough characterization of the performance and costs of mercury control. Future cost studies will focus on the development of performance and cost information needed to: refine cost estimates for sorbent injection based controls, develop cost estimates for wet scrubbing systems that employ methods for oxidizing Hg⁰, and determine the costs of various multi-pollutant control options.

The issue of mercury in residues will also be examined to address concerns related to the release of captured mercury species into the environment. These evaluations will be conducted in conjunction with the development and evaluation of air pollution emission control technologies.

Table 1. Estimates of Current and Projected Annualized Operating Costs for Mercury Emission Control Technologies.

Coal		Existing Controls ^b	Retrofit Control ^c	Current Cost (mils/kWh)	Projected Cost (mils/kWh)
Type ^a	S%				
Bit	3	CS-ESP+FGD	PAC	0.727 – 1.197	0.436 – 0.718
Bit	3	FF+FGD	PAC	0.305 – 0.502	0.183 – 0.301
Bit	3	HS-ESP+FGD	PAC+PFF	1.501 – NA ^d	0.901 – NA ^d
Bit	0.6	CS-ESP	SC+PAC	1.017 – 1.793	0.610 – 1.076
Bit	0.6	FF	SC+PAC	0.427 – 0.753	0.256 – 0.452
Bit	0.6	HESP	SC+ PAC+PFF	1.817 – 3.783	1.090 – 2.270
Subbit	0.5	CS-ESP	SC+PAC	1.150 – 1.915	0.69 – 1.149
Subbit	0.5	FF	SC+PAC	0.423 – 1.120	0.254 – 0.672
Subbit	0.5	HESP	SC+PAC+PFF	1.419 – 2.723	0.851 – 1.634

- a. Bit = bituminous coal; Subbit = subbituminous coal
- b. CS-ESP = cold-side electrostatic precipitator; HS-ESP = hot-side electrostatic precipitator; FF= fabric filter; FGD = flue gas desulfurization
- c. PAC = powdered activated carbon; SC = spray cooling; PFF= polishing fabric filter
- a. NA = not available

The Evaluation of ICR Data

The ICR data collection effort was conducted in three phases. In Phase I, information was collected on the fuels, boiler types, and air pollution control devices used at all coal-fired utility boilers in the U.S. In Phase II, coal data was collected and analyzed by the utility industry for 1140 coal fired units. Each sample was analyzed for Hg content, chlorine (Cl) content, sulfur content, moisture content, ash content, and calorific value. In Phase III, mercury measurements were made using the modified OH Method for total and speciated mercury. Additional, Phase III coal samples were collected and analyzed in conjunction with the OH Method sampling tests.

The methods used to evaluate the ICR data were based on two interrelated objectives. The first, which was primarily an OAQPS objective, was to estimate the speciation, amount and geographical distribution of national mercury emissions from coal-fired power plants in 1999. The second, an ORD objective, was to characterize the effects of coal properties, combustion conditions, and flue gas cleaning methods on the speciation and capture of mercury.

National Emission Estimates

The method selected by OAQPS to estimate national Hg emissions used: (1) the mean Hg content of coal burned in the unit during 1999; (2) the amount of coal burned in the unit during 1999; and (3) coal-boiler-control device emission factors developed from ICR emission tests.

The results of these estimates indicated that:

- Coal and related fuels burned in coal-fired utility boilers in 1999 contained 75 tons of mercury,
- The APCDs currently installed on existing units captured 43% of the mercury in these as-burned fuels, and
- Forty-three (43) tons of mercury was emitted to the atmosphere in 1999 from coal-fired utility power plants.

Coal, Boiler, and Control Technology Effects

The air pollution control technologies now used on coal-fired utility boilers exhibit levels of mercury control that range from 0 to 99%, as shown in Table 2. The best levels of control are generally obtained by emission control systems that use FFs. The amount of mercury captured by a given control technology is better for bituminous coal than for either subbituminous coal or lignite. The lower levels of mercury capture in plants firing sub-bituminous coal and lignite is attributed to low fly ash carbon content and the higher relative amounts of Hg^o in the flue gas from combustion of these fuels. For example the average capture of mercury based on OH inlet measurements in PC fired plants equipped a cold-side ESP is 48% for bituminous coal, 16% for subbituminous coal and near zero for lignite.

Plants that only employ post combustion PM controls display mercury emission reductions ranging from 0% to 93%. The highest levels of control were observed for units with FFs. Decreasing levels of control were shown for units with ESPs, PM scrubbers, and mechanical collectors.

Units equipped with dry scrubbers (SDA/ESP or SDA/FF systems) exhibited average mercury captures ranging from 98% for units burning bituminous coals to 3% for units burning subbituminous coal. The predominance of Hg⁰ in stack gases units fired with subbituminous coal and lignite results from low levels of Hg⁰ oxidization.

The capture of mercury in units equipped with wet FGD scrubbers is primarily dependent on the relative amount of Hg(II) in the inlet flue gas. Average mercury captures in wet FGD scrubbers ranged from 33% for one PC fired ESP + FGD unit burning subbituminous coal to 96% in a PC fired FF + FGD unit burning bituminous coal. The high mercury capture in the FF + FGD unit is attributed to increased oxidization and capture of mercury in the FF.

Table 2. Mean Mercury Emission Reductions for PC Fired Boilers^a, %

Add-on Controls	Type of Coal		
	Bitum.	Subbitum.	Lignite
PM Only			
CS-ESP	46	16	0
HS-ESP	12	13	NT
CS-FF	83	72	NT
PM Scrubber	14	0	33
Dry FGD Scrubbers			
SDA + ESP	NT	38	NT
SDA + FF	98	25	17
SCR + SDA + FF	98	NT	NT
Wet FGD Scrubbers			
CS-ESP + Wet FGD	81	35	44
HS-ESP + Wet FGD	55	33	NT
CS-FF + Wet FGD	96	NT	NT

^a Based on OH train data. NT= not tested. Mean reduction from 3-test averages for each unit.

Mercury capture in PC fired units equipped with spray dry scrubbers and wet scrubbers appear to provide similar levels of control on a percentage reduction basis. However, this observation is based on a small number of short-term tests at a limited number of facilities.

Additional testing will be required to characterize the effects of fuel, combustion conditions, and APCD conditions on the speciation and capture of mercury.

Co-Controls

Preliminary evaluation of ICR data indicates that technologies currently in place for control of criteria pollutants achieve reductions in mercury emissions that range from 0% to >90%. Current levels of mercury control can be increased by application of mercury retrofit technologies or methods designed to increase capture of more than one pollutant. This approach can utilize the synergism's that accrue through the application of multi-pollutant control technologies.

Bench-and-pilot scale tests have shown that Hg capture in PM control devices generally increase as the carbon content of fly ash increases. Increased use of combustion modification techniques that increase ash carbon content will generally increase the amount and capture of Hg(p).

Tests on bituminous coal-fired units equipped with SNCR + CS-ESP and SCR + SDA/FF systems exhibited average mercury emission reductions of 91% and 97% respectively. By comparison, average mercury reductions in similar units without post-combustion NO_x controls were 48% and 98%, respectively. Recent tests on a pilot-scale combustion system equipped with an ESP have also indicated that SNCR and SCR systems may enhance mercury capture in PM control devices. However, the level of capture appears to be highly dependent on the type of coal burned.

It is believed that the use of combustion modification techniques and post combustion NO_x control technologies that are applied on NO_x SIP units will also increase the capture of mercury in these units.

The retrofit of utility units to control emission of SO₂ and fine PM will also provide co-benefits in the control of mercury. This is apparent from the increased control of mercury on units equipped with FFs, dry FGD scrubbers, and wet FGD scrubbers. Mercury or multi-pollutant sorbents will add only marginal costs to units retrofitted with FFs or SD/FF. The use of multi-pollutant or mercury sorbents would be more costly, but, the added costs of mercury control would be modest. Technologies developed for use on existing wet FGD units could also be used for new scrubber retrofits intended to control SO₂ and the precursors to secondary fine PM.

Generally, the control of mercury emissions via multi-pollutant control technologies can provide a cost-effective method for collectively controlling the various pollutants of concern.

Behavior of Mercury in Coal Combustion By-products

The operation of power plants results in solid discharges including fly ash, bottom ash, boiler slag, and FGD residues. These residues already contain mercury, presumably as bound mercury that is relatively insoluble and non-leachable. In 1998, approximately 98 million Mg (108 million tons) of coal combustion residues (CCRs) were generated. Of this amount, about 70 million Mg (77 million tons) was landfilled, and about 28 million Mg (31 million tons) was used for the manufacture of by-products.

Increased control of mercury emissions from coal-fired power plants may change the amount and composition of CCRs. Such changes may increase the potential for release of mercury to the environment from either landfilling or uses of CCRs. Available data have been collected to help characterize this potential and to identify data gaps that may exist in order to understand the potential life-cycle environmental burdens associated with current and future management practices of CCRs.

Mercury volatilization or leaching is possible during any phase of CCR life cycle (production, usage, or disposal). Available CCR data report a range of values for mercury concentration based on varying mercury contents in the input coal, varying mercury capture rates for PM and SO₂ control devices, and differences in analytical and sampling protocols in use. Fly ash and FGD residues typically have higher mercury concentrations than bottom ash and boiler slag. The mercury concentrations in the APCD residues are typically in the sub-ppm range (i.e., <1.0 ppm).

Over 70% of the total mass of CCRs produced in 1998 went to land disposal (mostly landfilling, although some went to surface impoundments) and the remainder was managed in beneficial uses. Mercury volatilization of CCRs in landfills and/or surface impoundments is expected to be low due to the low temperatures involved and the existence of a relatively small surface area per unit volume of residue. Since the residues are typically alkaline and the mercury is adsorbed on alkaline particles, acid leaching potential is expected to be minimal. Limited available data indicate non-detectable mercury levels in the leachate. However, due to the large volumes of residues being land-disposed and the increase in mercury content, some limited work would help provide direct measurements to ensure that mercury would not be released into the environment.

There are several commercial uses of CCRs where available data on which to characterize the mercury emission potential are lacking. These include:

1. Cement production - mercury volatilization is a concern if fly ash is input to cement kilns in which temperatures reach 1500 °C (2700 °F);
2. Structural fill - mercury volatilization is not a concern, but leaching is possible, although the alkaline nature of fly ash reduces this concern;
3. Mining applications - there is concern that leaching may occur due to the acidic conditions occurring during mining applications;

4. Wallboard - the substitution of FGD residues for natural gypsum has been growing rapidly. Volatilization of mercury in the FGD by-products during manufacturing (during the drying process) is of concern;
5. Mineral filler - mercury volatilization during the production and application of asphalt using fly ash fillers is of concern since the production and application of asphalt occurs at temperatures above 175 °C (350°F).
6. Agriculture - fly ash, bottom ash, and FGD sludge are used as soil amendments, generally replacing lime. The potential for mercury leaching or uptake in plants needs to be evaluated in order to determine the potential fate of mercury. This has been a concern of environmental interest groups, and data are needed to be able to determine the ultimate fate of mercury.

Recommendations

Additional efforts are planned to characterize the behavior of mercury in coal combustion systems. These efforts must include further research on the speciation and capture of mercury, and the stability of mercury in CCRs and residue by-products. The control capabilities and costs of potential mercury retrofit technologies currently under pilot-scale development must be continued and appropriate control technologies must be evaluated on full-scale units. Additionally, an evaluation of the co-control of mercury with available PM, SO₂, and NO_x controls is needed.

Continued collaboration between OAR and ORD is important. A team approach is deemed necessary to ensure that appropriate regulatory and scientific guidance is available in the development of mercury and multi-pollutant control technologies. Mercury measurement and monitoring capabilities must be consistent with the regulatory approaches being considered, e.g. speciated vs. total mercury emissions. Field activities need to be coordinated to: (1) improve the emissions data base; (2) develop the technologies most appropriate for Agency goals (e.g. mercury-specific vs. multi-pollutant); and (3) refine cost data and models based on actual field experience.

Finally, ORD must continue to work closely with DOE, EPRI and the utility industry to develop mercury and multi-pollutant control technologies. Only a collaborative effort will provide all of the scientific knowledge, engineering skills and financial resources needed to develop effective control technologies and regulatory requirements.