TITLE: CONTROL OF MERCURY EMISSIONS FROM COAL-FIRED BOILERS

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Objective:

The "Development of Advanced Environmental Control Technology" project at Argonne is designed to investigate new concepts leading to advanced control technologies for fossil-energy systems. The objective of this new task on air toxics control is to develop new or improved, cost-effective control technology for the abatement of emissions of hazardous air pollutants (HAPs) from fossil-fuel combustion plants and to evaluate the possible effects of any captured species on waste disposal. The HAPs to be investigated initially in this task include mercury and arsenic compounds.

Accomplishments and Conclusions:

Information on mercury concentrations in U.S. coals, emission rates and emissions control for mercury in flue gas from coal combustion plants, and methods for the sampling and analysis of mercury in flue gas has been collected from reports, papers, conferences, and discussions with other researchers. A thorough evaluation of this information was then performed, and a topical report summarizing the information was prepared.

As shown in Table 1, anthracite and bituminous coals have the highest mean-mercury concentrations (0.23 ppm and 0.21 ppm, respectively), and subbituminous coals have the lowest mean-mercury concentration (0.1 ppm). However, subbituminous coals have the greatest range of reported mercury concentrations. The concentration of mercury in U.S. coals also varies by the geographic region in which the coal is mined. As shown in Table 2, coals from the Appalachian and Gulf states have the highest mean-mercury concentration, whereas coals from the Alaska region have the lowest mean-mercury concentration. However, coals from the Alaska region also have the greatest range of mercury concentrations.

With respect to mercury emissions from coal-combustion sources, it was found that the utility and industrial sectors are the most-characterized combustion sources, while few data are available for the commercial/institutional and residential sectors. Information on measured mercury emissions from coal-fired utility and industrial boilers is summarized in Tables 2-4 for bituminous, subbituminous, and lignite coals, respectively. It should be pointed out that, because of the difficulties associated with the sampling and analysis of mercury compounds in flue gas, the mass balances often do not close well for field tests, and significant differences in measured mercury emissions between tests exist. Furthermore, a major deficiency in prior field tests involving mercury measurements is that essentially no determinations were made of the types of mercury compounds present in the flue gas. Since some mercury compounds are more toxic than others, the lack of information on the concentrations of individual mercury compounds in the flue gas makes a credible risk assessment impossible.

DISCLAIMER

The data in Tables 3-4 show considerable variation in mercury emissions from large coal-fired boilers. No significant differences in mercury emissions exist between different boiler types or different end-use sectors. However, the available test data indicate that for coal-combustion systems equipped with flue-gas-cleaning devices, significant reductions in mercury emissions could be realized. Also, preliminary data on vapor-phase mercury emissions from a small atmospheric-pressure fluidized-bed combustion unit were found to be significantly lower, when compared to a small stoker burning the same coal. In contrast, preliminary results from tests with a large utility boiler modified for low-NO_x operation showed greater mercury emissions than the baseline mercury emission levels associated with normal, unmodified operation.

Of the four types of devices widely used for particulate collection--cyclone, wet scrubber, electrostatic precipitator (ESP) and fabric filter (FF)--most of the available data on trace-element emissions have been obtained from boilers equipped with cold ESPs, and limited information on the removal of trace elements by cyclones and hot ESPs indicates that the removal efficiencies for mercury are negligible. With cold ESPs (see Table 5), the calculated mercury removal efficiencies range from 22 to 91% for utility boilers and from 40 to 91% for industrial stokers equipped with a mechanical collector and an ESP in series. The high-end number is suspected to result from deficiencies in the sampling and analytical techniques employed during the field tests, and the effectiveness of the cold ESP for mercury control probably would be in the range of 20-50%. Only two paired data points have been found for coal-fired utility boilers equipped with high-efficiency wet scrubbers, which show a removal efficiency of about 70%; the mass balance of the data, however, is poor. No information has been obtained from the open literature on the mercury emissions from coal-fired boilers equipped with fabric filters for particulate control; however, data obtained by the Electric Power Research Institute (EPRI) indicate high mercury-removal efficiencies ranging from 85 to 90% with fabric filters. These numbers need further verification.

Of the two types of systems for flue-gas desulfurization, wet scrubbing and spray-dryer (SD) scrubbing, very limited information on the mercury removal efficiencies for the latter system on coal-fired boilers has been found in the open literature. However, available results on the removal efficiencies of mercury from municipal- and hazardous-waste incinerators indicate good mercury removal efficiencies in the range of 75-99% with SD/FF systems and in the range of 35-45% with SD/ESP systems, so long as the flue-gas temperature at the spray-drying chamber outlet is kept below 150 °C, but essentially no mercury removal when the flue gas temperature at the SD outlet in higher than 200 °C. These numbers cannot be directly translated to coal-fired boilers since the mercury levels in flue gas from coal-fired boilers are about one to two orders of magnitude lower than those from the incinerators, but they do suggest that a significant amount of mercury removal could probably be attained with an SD/FF combination because the flue-gas temperature at the SD outlet is normally maintained at below 75°C for coal-combustion systems. With wet FGD systems, the removal efficiencies for mercury are found to range from 30% to as high as 95%. The variations in the features for different types of wet-scrubber designs may contribute to the wide range of mercury removal efficiencies, but very limited information on the designs and operating conditions of the wet scrubbers tested has been reported. Again, further verification of the high mercury-removal efficiency is warranted.

The literature search and personal communications with other researchers also revealed that several processes have been investigated for the reduction of mercury emissions in gas streams. These processes include improved spray-dryer scrubbing with the addition of activated carbon powder, a low-temperature plasma process, dry adsorption processes with carbon or selenium filters, and polysulfide wet scrubbing. While the former three technologies have been evaluated on flue gas from combustion or incineration flue gases, the polysulfide wet-scrubbing process has only been tested on mercury-containing natural gas. Both the improved spray-dryer scrubbing and the dry-adsorption processes have demonstrated high mercury-removal efficiencies on incineration flue gases, but the improved processes have yet to be adequately evaluated on mercury levels representative of those encountered in coal-combustion flue gases. To ensure that cost-effective mercury-control technologies are available for coal-fired systems when needed, a systematic evaluation on a laboratory scale of the effectiveness of the above technologies and other additives for reducing low levels of mercury (less than 10 ppb) is recommended.

Plans:

In order to be able to screen a variety of chemical additives and a range of process variations in a rapid and economical manner, a laboratory-scale fixed-bed reactor facility is currently being constructed at Argonne, and a laboratory-scale spray-dryer/fabric-filter (SDFF) facility previously developed will be used. The SDFF facility includes a gas-handling system for preparing synthetic flue gas; a gas analysis system capable of measuring CO_2, O_2, SO_2, NO, and NO_x in flue gas; and a computer-controlled data acquisition system. In the experiments with the fixed-bed reactor facility, however, a simpler gas-preparation system will be used since the reaction of mercury with other major gaseous compounds in the flue gas is believed to be negligible. A mercury-containing permeation tube will be used to generate a known concentration of mercury vapor in the feed gas entering the reactor section. To provide an adequate analysis of the low concentration of mercury vapor in the gas streams entering into and exiting from the fixed-bed reactor, a Jerome gold-film mercury-vapor analyzer made by the Arizona Instrument Corp. will be used. The schematic flow diagram of the fixed-bed reactor facility is shown in Figure 1.

After the fixed-bed reactor facility is constructed, experiments will be conducted to investigate the potential for enhanced mercury reduction in the filter cake as a result of various chemical additives and process operating conditions. Before the investigations with chemical additives are started, however, experiments will be carried out on $Ca(OH)_2$ solely to develop the baseline results, against which the data from future experiments with additives and process variations will be compared. Chemical additives to be investigated will include two types of activated carbons, alkali sulfide, and other chemicals to be identified as the task progresses. Process operating conditions to be investigated include the process operating temperature, filter cake thickness, and other potential concepts to be identified later.

Once promising chemical additives and/or process modifications have been identified in the laboratory research with the fixed-bed reactor, additional experiments will be carried out by using the SDFF facility to further confirm the results. If warranted, demonstration tests will be conducted with Argonne's industrial-scale SDFF facility.

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Table 1. Concentrations of Mercury in U.S. Coals

Coal Type	Mercury Concentration (ppm) Mean	Range
Bituminous	0.21	0.01 - 3.3
Subbituminous	0.10	0.01 - 8.0
Anthracite	0.23	0.16 - 0.3
Lignite	0.15	0.03 - 1.0

Table 2. Concentrations of Mercury in U.S. Coals

Region	Mercury Concentration (ppm) Mean	Range
Appalachian	0.24	0.01 - 3.3
Interior	0.14	0.01 - 1.5
Illinois Basin	0.21	0.03 - 1.9
Gulf Province	0.24	0.03 - 1.0
Northern Plains	0.11	0.01 - 3.8
Rocky Mountains	0.09	0.01 - 8.0
Alaska	0.08	0.02 - 6.3

Table 3. Summary of Mercury Emissions from Bituminous Coal-Fired Boilers

Boiler Type	Emission Range ($lb/10^{12}$ Btu)
UTILITY	
Pulverized Dry Bottom	
Uncontrolled	3.9 - 308
With ESP	0.4 - 22
Pulverized Wet Bottom	
Uncontrolled	NA
With ESP	2.6 - 6.3
Cyclone	
Uncontrolled	10
With ESP	4.0 - 18
INDUSTRIAL	
Pulverized Dry Bottom	
Uncontrolled	NA
With ESP	4.2 - 4.4
Spreader Stoker	
Uncontrolled	0.8 - 12
With Multiclone	5.8 - 25
With ESP	1.0 - 4.2
Overfeed Stoker	
Uncontrolled	0.01 - 2.1
Dust Collector	0.4 - 1.2

Table 4. Summary of Measured Mercury Emissions from Coal-Fired Boilers

Boiler Type	Emission Range ($lb/10^{12}$ Btu) SUBBIT.	LIGNITE
UTILITY		
Pulverized Dry		
Uncontrolled	NA	NA
With ESP	4.1	<0.23
With Multiclone	NA	4.4 - 6.5
Cyclone		
Uncontrolled	81	NA
With Scrubber	4.9	NA
With Cyclone	NA	22
With ESP	NA	0.46
INDUSTRIAL		
Spreader Stoker		
Uncontrolled	0.6 - 17	NA
With Multiclone	NA	5.6
With ESP	NA	0.53
With Cyclone/ESP	0.4 - 0.6	NA

Table 5. Summary of Removal Efficiencies of Mercury in Coal-Fired Boilers

Control Device	Removal Efficiency (%)
UTILITY BOILER	
Cold ESP	22 - 91 (20 - 90*)
Hot ESP	0
Fabric Filter	--- (85 - 90*)
Scrubber	70
INDUSTRIAL BOILER	
Cyclone/Multiclone	0 - 40
ESP	40 - 90
Fabric Filter	No Data Available
Scrubber	No Data Available

* From EPRI PISCES data base

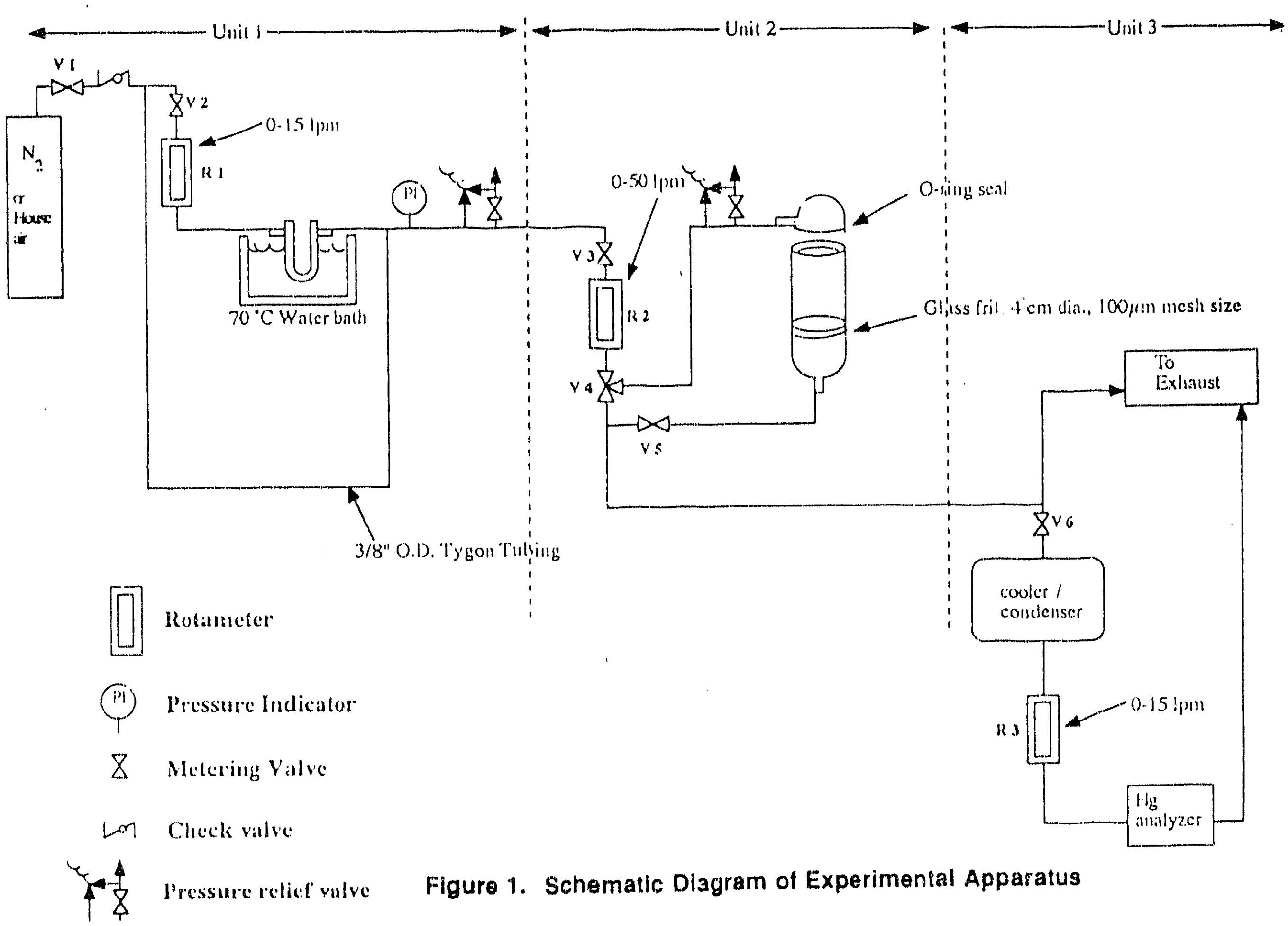

Figure 1. Schematic Diagram of Experimental Apparatus

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