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# Bimodal fly ash size distributions and their influence on gas-particle mass transfer during electrostatic precipitation

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#### Abstract

Electrostatic precipitators (ESPs) have previously been demonstrated to achieve substantial (up to 60–70%) removal efficiency of mercury from coal-fired power plants (CFPPs). However, a high degree of scatter exists in the pilot- and full-scale data, suggesting an incomplete understanding of the mechanism by which mercury is adsorbed within an ESP, particularly by native fly ash. The present analysis explores the influence of bimodal particle size distributions (PSDs) on the gas-particle mass transfer underlying mercury adsorption by fly ash within an ESP. The analysis is motivated by the recent discovery by other investigators of bimodal fly ash PSDs resulting from coal combustion. Results of the present analysis show that, relative to similar monomodal PSDs, bimodal PSDs exhibit greatly increased gas-particle mass transfer potential during electrostatic precipitation. For bimodal PSDs, gas-particle mass transfer potential increases with increasing particle mass in the second mode and decreasing geometric mean diameter and geometric standard deviation of the second mode. A supplemental analysis compares the mercury removal potential of native fly ash, injected fly ash, and injected powdered activated carbon (PAC) during their collection within an ESP, using representative mercury adsorption capacities and particle mass loadings for each. Results showed only marginal differences in mercury removal efficiency between the three sorbents.

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#### 1. Introduction

Mercury emissions control is a priority for coal-fired power plants (CFPPs) in the U.S. as a result of the Clean Air Mercury Rule (CAMR) promulgated by the U.S. EPA in 2005. Mercury emissions control for CFPPs is complicated by a number of challenges generally not faced by other mercury-emitting combustion processes such as municipal and medical waste incinerators. The unique technical challenges include the ultratrace mercury concentrations as well as complex mercury partitioning in both species and phase for which chemical kinetic mechanisms are still under development (e.g., [1–4]). Reviews by Pavlish et al. [5] and Brown et al. [6] summarize the technical challenges faced in developing mercury emissions control for CFPPs. In addition to the technical challenges, practical challenges include the integration of mercury emissions control

processes into existing CFPP exhaust trains, fly ash handling, and the sensitivity of mercury partitioning kinetics to the combustor and exhaust train configurations. This sensitivity is exacerbated by facility repairs, upgrades, and modernization projects that can take place as facilities age. As more than two-thirds of the coal-fired electric generating capacity in the U.S. is more than 25 years old [7], the substantial nonuniformity among CFPP facility configurations is believed to contribute to the nonuniformity in mercury emissions characteristics (e.g., oxidized mercury fraction) that are important to the design of effective control technologies.

Adsorption is the most mature mercury emissions control approach under consideration for CFPPs. Fixed bed adsorption is commonly used by medical and municipal waste incinerators (collectively, MWIs) to remove trace pollutants, including mercury, from their exhausts. Capitalizing on this demonstrated success, fixed bed adsorption has also been tested on a number of CFPPs by injecting a powdered sorbent into the flue gas upstream of a fabric filter (FF). However, fabric filters are currently installed on only a small fraction (less than 10%) of CFPPs in the U.S. [6],

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meaning that implementing fixed bed adsorption would require the vast majority of CFPPs to be retrofitted with FFs. Fabric filters often have large footprints that make them difficult to site at an existing CFPP. Fabric filters also impose a large pressure drop of 1250 to 2500 Pa (5 to 10 in H<sub>2</sub>O) in the exhaust stream. Consequently, a fabric filter retrofit may involve not only the capital costs of the FF, but also capital and O&M costs associated with compensating for the higher pressure drop, such as adding induced draft (ID) fans downstream of and reinforcing ductwork upstream of the FF. An often-cited advantage of fixed bed adsorption is that the retrofitted fabric filter collects the injected sorbent separately from the native fly ash. This maintains the low (less than 1%) carbon content of the fly ash, which concrete manufacturers require as a condition of its use. However, on average only about one-third of coal combustion fly ash undergoes beneficiation, suggesting that separate sorbent and fly ash collection offered by FF retrofits is not a priority for the majority of CFPPs, although ash beneficiation can exceed 90% for some facilities.

Compared to fabric filters, electrostatic precipitators (ESPs), both hot- and cold-side, are commonplace, found on more than three-quarters of the coal-fired electric generating capacity in the U.S. [6]. Given the prevalence of ESPs, there is continuing interest in the potential to achieve significant mercury emissions control within an ESP. Pilot- and full-scale test results show that mercury capture occurs within ESPs, both with and without upstream sorbent injection [6,8–10]. However, there exists a high degree of scatter in most of the reported data that suggests an asyet incomplete understanding of the processes that contribute to mercury adsorption within ESPs. Recent mass transfer analyses have shown this to be true. For example, reductions of 60 to 70% in gas-phase mercury concentrations have been observed between the inlet and the outlet of ESPs (see, for example, Ref. [5]). It is a common assumption that these reductions are the result of Hg adsorption by the collected particulate matter ("dust cake") on the ESP plate electrodes. However, detailed mass transfer analyses [11] showed that for typically-sized ESPs (based on specific collection area, SCA) no more than 15 to 20% of the inlet gasphase mercury can be adsorbed by the dust cake. Subsequent analyses revealed that gas-particle mass transfer during particulate (either fly ash or injected sorbent) collection within an ESP presents a much greater potential for mercury capture [12], and that the characteristics of a monomodal particle size distribution (PSD) strongly influence gas-particle mass transfer as well as particulate collection efficiency [13].

Improved understanding of the mechanisms underlying mercury capture within ESPs enhances their potential to serve as highly flexible platforms for co-control of mercury and particulate matter. This potential involves not only Hg adsorption by injected powdered sorbents but also by native fly ash. The capacity of fly ash to adsorb mercury is thought to be related to its residual carbon content [6,14] known as LOI (loss-on-ignition). Senior and Johnson [15] estimate that fly ash derived from bituminous coals has on the order of one-tenth the equilibrium mercury adsorption capacity of powdered activated carbon (PAC), although they caution that this figure likely cannot be extrapolated to other ranks of coal. However, the results of

Maroto-Valer et al. [16] reveal additional complexities; in comparing fly ash before and after activation, they found that activation increased the surface area of the ash by a factor of 15, but decreased the equilibrium mercury adsorption capacity by almost a factor of eight. They speculated that the activation process drove off halogens that had been present on the precursor fly ash surface and had served to enhance mercury capture.

The objective of the present analysis is to elucidate the gasparticle mass transfer that supports adsorption of gas-phase mercury by fly ash within an ESP. Such analysis complements numerous prior studies of fly ash equilibrium adsorption capacity and oxidation potential with respect to gas-phase mercury. Of particular interest is the potential impact of bimodal particle size distributions (PSDs). Recent pilot-scale measurements [17] revealed the presence of a secondary mode in the measured PSDs of fly ash produced from coal combustion. Given the typically coarse characterization of fly ash (e.g., often only mass median diameter is reported), the extremely high total surface area available in the previously unnoticed secondary mode of the fly ash PSD may help explain the scatter in measured mercury removal efficiencies by fly ash within ESPs (see, for example, Ref. [5]). It must be emphasized that a comparison between the present analytical results and experimental data is not possible because no suitable experimental data exist. Pilot- and full-scale tests of mercury capture by fly ash within ESPs typically report only fly ash mass median diameter, which alone is insufficient for reconstructing even a monomodal PSD, much less the bimodal PSDs reported by Linak et al. [17] and considered here. Thus, the present analysis should be viewed not as a model for reproducing existing experimental results, but used instead to gain insight into fundamental mass transfer mechanisms and help explain and reduce the large degree of scatter in observed mercury removal efficiencies within ESPs.

Linak et al. [17] measured PSDs of fly ashes produced by two bituminous (Utah and Western Kentucky) coals and one subbituminous (Montana) coal. In addition to the expected coarse mode peaks in volume distribution  $(dV/d(\log d_p))$  for the three coals around 10-20 µm, they also measured a significant accumulation mode around 1-2 µm (referred to in Linak et al. [17] as the central mode) as well as a much smaller ultrafine mode around 0.08 µm (referred to in Linak et al. [17] as the accumulation mode). Although it is known that fine combustion aerosols can become enriched in a variety of elements, the data from Linak et al. show that largely similar values of LOI were measured for the <2.5  $\mu$ m fraction as for the >2.5  $\mu$ m fraction. LOI values measured by Linak et al. (1.7-15.7 wt.%) fall within the range of values  $(\sim 0-1)$ 30 wt.%) considered by Senior and Johnson [15] in their analysis of the correlation between LOI and mercury adsorption capacity of fly ash for pulverized coal combustors. Thus, given the evidence of a positive correlation between LOI and mercury adsorption capacity, the determination of non-negligible LOI within accumulation mode fly ash particles suggests that they may play a significant role, not previously considered, in the adsorption of mercury within an ESP. The present analysis seeks to quantify the impact of bimodal fly ash PSDs on the potential for mercury adsorption during particle collection within an ESP.

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