

Uptake of Dissolved Mercury by Coal Combustion Fly Ashes
Karl Schroeder, Mike Schoffstall, Ann Kim

Concern about mercury (Hg) emissions from power plants has been increasing since the Clean Air Act Amendments of 1990. Recent studies and reports by the EPA make it clear that they view emissions of Hg from coal-fired utilities as a serious environmental problem. Future regulation of the atmospheric emissions of Hg will shift the environmental burden from the flue gas to the solids formed as by-products of the combustion and flue-gas clean up processes, *i.e.* coal utilization by-products (CUB). The beneficial uses of CUB may then be in jeopardy. This is particularly true for those uses that may allow for transport of the mercury into surface or ground water. For example, the use of CUB as a grouting material, as backfill to prevent mine subsidence and/or acid mine drainage, and as a soil amendment, put the material in direct contact with either the ground or water. For this reason, it is important to understand the chemistry at the CUB-water interface, to be able to predict the environmental fate of the CUB-bound Hg, and to be able to anticipate the effect of additional Hg loads in the CUB material.

The stability of the CUB-bound Hg in a given application depends upon both the nature of the CUB and the nature of the environment in which it is placed. Studies have shown that Hg can be quite mobile in the environment although the sequestration of mercury by solid particles has been considered an important process controlling its transport in aqueous systems. Releases of Hg from both deeply buried areas, such as landfills, and surface soils have been found. Only a limited amount of data is available for the case of fly ash-bound Hg(II).

This investigation studies the Hg(II) chemistry associated with the CUB-water interactions. Hg(II) adsorption isotherms were measured under various conditions. Solutions providing an upper limit of about 20 mg Hg(II) per kg of ash were used to give concentrations relevant to US coals. The range included not only currently found Hg loadings but also the maximum attainable if all of the Hg in the coal were to be captured in the CUB. A number of samples at progressively longer times were taken to determine if equilibration between the aqueous and solid phases had been reached. Hg_(TOT) remaining in solution was determined by cold vapor atomic absorption spectrometry. The effect of pH was studied by pre-equilibration of the CUB at the desired pH prior to introduction of a Hg(II) solution of the same pH.

The results indicated that complete equilibrium was not attained, even after one month, although the changes were small. Prior leaching of the fly ash to remove materials soluble at a pH of 2 did not eliminate the problem. Because the trend was toward increasing Hg adsorption at longer times, the measured values underestimate the Hg retention capacity of the fly ashes. The solution pH did not appear to affect the extent of adsorption in the range of 2 < pH < 3. In this region, the mercury speciation would be expected to be predominately Hg²⁺. The data was analyzed using both the Freundlich and Langmuir adsorption isotherm equations. Neither gave exceptionally good fits although the Langmuir was better than the Freundlich.