Associations of Mercury in Indiana Coals and Their Fly Ash; Insights from a Sequential Extraction Technique

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Coal from two of Indiana's economically important Pennsylvanian coal beds, the high-sulfur Springfield Coal Member of the Petersburg Formation and the low-sulfur Danville Coal Member of the Dugger Formation, along with their respective fly ashes were studied with regard to mercury geochemistry. Both coals are of high-volatile bituminous rank; mercury (Hg) content is 0.13 mg/kg in the Springfield and 0.03 mg/kg in the Danville. Study of the combustion products of these coals show relatively high Hg concentrations in the low temperature fly ash, which is especially pronounced in the Danville coal. To better understand the fate of Hg during the combustion process, a sequential extraction procedure, adapted from published sources was used to evaluate Hg affinity to four geochemically distinct fractions. In step I, ammonium acetate (CH_3COONH_4, or "AmmAcet") was used to decompose some of the least resistant organic matter and some of the carbonates. In step II, the residue of step I was further leached with HCl, removing remaining carbonates, Fe oxides, sulfates, monosulfides, and certain chelated organic compounds. In step III, HF dissolved the silicate minerals, and finally, in step IV, HNO_3 was added to decompose disulfides, especially pyrite, any remaining phosphates, and refractory organic matter. In this study, solid residues of all four phases were analyzed for Hg concentration using a Leco Advanced Mercury Combustion analyzer.

The results show that, in both coals, the majority of the Hg is associated with disulfides, particularly pyrite, and refractory organics, whereas negligible amounts of Hg appear to be associated with silicates and carbonates. The main difference between the two coals was that in the Springfield coal (high S and high Hg), the refractory organic matter had much higher concentrations of Hg than these in the Danville coal (0.01 ppm versus 0.001 ppm in the Danville). In the fly ash from the Danville coal, there is a major difference in Hg associations between baghouse fly ash (low-temperature) and rear pass hopper fly ash (high-temperature); residue after step III has the highest Hg content in baghouse fly ash, whereas in rear pass hopper ash, step IV residue (refractive) contains the most Hg. It is interpreted that in the rear pass hopper fly ash, Hg does not condense on fly ash particles because of high temperature, and the only Hg that is present is a very resistant one associated with the unburned carbon. Indeed, rear pass hopper fly ash has very low Hg content (<0.02 ppm). In contrast, in baghouse fly ash, condensation and adsorption of Hg onto fly ash particles took place (resulting in an elevation of Hg concentrations to 0.2 ppm). We suggest that this adsorbed Hg is resistant and is extracted only by HNO_3. Fly ash from the Springfield coal shows similar Hg associations to those in baghouse fly ash from the Danville coal, with the adsorbed at low temperature Hg being the dominant.

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