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Subtask 1.17 - Subcritical Water Extraction of Mercury From Soils and Sediments

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SUBTASK 1.17 - SUBCRITICAL WATER EXTRACTION OF MERCURY FROM SOILS AND SEDIMENTS

1.0 INTRODUCTION

The U.S. Environmental Protection Agency (EPA) "National Sediment Quality Survey" lists the top pollutants responsible for toxicity in watersheds as 1) PCBs (polychlorinated biphenyls), 2) mercury, and 3) other organics such as PAHs (polycyclic aromatic hydrocarbons) and pesticides. In addition, these same pollutants are major contributors to chemical pollution on U.S. Department of Energy (DOE) and other contaminated sites (e.g., industrial sites and harbors). An ideal remediation method would allow cost-effective removal of both organic and mercury contamination using a single process.

The Energy & Environmental Research Center (EERC) has demonstrated that controlling the temperature (and to a lesser extent, the pressure) of water can dramatically change its ability to extract organics and inorganics from matrices ranging from soils and sediments to waste sludges and coal. The dielectric constant of water can be changed from ca. 80 (a very polar solvent) to <5 (similar to a nonpolar organic solvent) by controlling the temperature (from ca. ambient to ca. 400°C) and pressure (from ca. 5 to 350 bar). The EERC has shown that hazardous organic pollutants such as pesticides, PAHs, and PCBs can be completely removed from soils, sludges, and sediments at temperatures (250°C) and pressures (<50 atm) that are much milder than typically used for supercritical water processes (temperature >374°C, pressure >221 atm). In addition, the process has been demonstrated to be particularly effective for samples containing very high levels of contaminants (e.g., parts per thousand).

The EERC has also demonstrated that mercury can be extracted using supercritical water at much harsher conditions (400°C, and >300 atm). However, the removal of mercury from contaminated solids at the lower temperature and pressure conditions (e.g., 250°C, 50 atm) has not been investigated. If successful, this project will provide the basis for using hot/liquid water to extract both organic contaminants and mercury from contaminated solids in a single-step process.

2.0 OBJECTIVE

- To determine the ability of hot/liquid water to extract mercury from contaminated soils and/or sediments using the same conditions of temperature and pressure that have previously been successful for removing organic pollutants including PAHs, PCBs, and pesticides.

3.0 SPECIFIC DELIVERABLES

- The ability (and conditions) of hot/liquid water to remove mercury from three or more highly contaminated soils and/or sediments will be reported.

4.0 ACCOMPLISHMENTS

4.1 Sample Selection

Three samples were selected for initial extractions. All samples are "real-world"; e.g., they were collected from contaminated environmental sites and used without significant preparation. One of the samples was obtained from the National Institute of Standards and Testing (NIST), so that certified values of the mercury concentrations were available. The three samples were contaminated (total mercury values) at 50, 213, and 3380 ppm.

4.2 Initial Extractions

Initial extractions have been performed using a subcritical water extraction apparatus developed for this project. Problems were encountered with plugging of the 1/16-in-OD outlet lines for the most highly contaminated samples. Since the extraction cell outlet contained a 0.5-μm frit, the plugging appeared to be deposition of extracted material as the extractant water was cooled. Successful extractions of the most highly contaminated sample were performed by using 2-g samples in a 3.4-mL extraction cell and placing an empty 0.8-mL cell after the sample cell before the water was cooled.

Since the goal of the extractions was to determine the removal efficiency of total mercury using subcritical water extractions, the concentration of mercury was determined in the original soils and in the soil residues after extraction. The results of initial extractions are summarized in Table 1. As shown, the conditions typically used for removal of organic pollutants from soils show substantial removal of mercury from the test soils in 15 to 30 minutes at conditions previously used to remove organic pollutants such as PAHs, PCBs, and pestcides (e.g., 250°C). Increasing the temperature to 300°C also increases the rate of extraction (Table 1). It should be noted that the results in Table 1 are based on single determinations. Future work will determine the reproducibility of the removal efficiencies at 250° and 300°C.

TABLE 1

Mercury Removal Efficiencies Using Subcritical Water

	Soil A	Soil B	Soil C (NIST)
Initial Concentration, ppm	213	3380	50 (43)*
Concentration after 250°C, 15 min, ppm	61	2190	6.2
Hg Removed after 250°C, 15 min, %	71	35	87
Concentration after 250°C, 30 min, ppm	10.5	1860	-
Hg Removal after 250°C, 30 min, %	95	45	-
Concentration after 300°C, 30 min, ppm	-	540	3.4
Hg Removal after 300°C, 30 min, %	-	84	93

*The concentration certified by NIST was 50 ppm; 43 ppm was determined by the EERC.