Radium mineral associations within abandoned mine drainages and treatment systems with perspective to the future of rare earth element extraction

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Discharges from abandoned coal mines and unearthed coal waste products [i.e., acid mine drainage (AMD)] are persistent sources of contamination representing a major water quality issue in the U.S. state of Pennsylvania. Unearthed coal waste products and treatment solids have been the focus of a nationwide agenda toward establishing a domestic U.S. supply chain of critical minerals, specifically rare earth elements (REE), for clean energy development. Rare earth elements have been documented to sorb and/or co-precipitate with abundant Fe, Mn and Al oxyhydroxides and possibly other compounds within AMD treatment systems. Ideally, processes may be identified and used to concentrate and separate REE from associated contaminants in AMD for economic recovery. Radium (Ra), a known carcinogen with the longest radionuclide half-life of 1600 vears, has also been documented to sorb to Fe and Mn oxyhydroxide solids. To this end, it is useful to establish a baseline assessment of the partitioning of coal-derived radium within sediments that naturally exist in coal-bearing rock (e.g. clays), precipitate from AMD discharges as a function of atmospheric exposure (e.g. Fe, Mn, and Al oxyhydroxides), or precipitate due to AMD treatment or general water chemistry (e.g. limestone beds, caustic soda addition, high sulfate concentrations). To our knowledge, a baseline assessment of Ra in AMD or associated solids has never been conducted. To test the hypothesis that Ra may be accumulated with AMD solids, we collected water and sediment samples from 4 abandoned and 12 treated (both active and passive systems) AMD discharges in Pennsylvania, USA, within both bituminous and anthracite coal regions. Our data suggest Ra concentrations were relatively low in corresponding AMD water and solid samples (< 0.5 Bq/L and upwards of 850 Bq/kg, respectively). Sequential leaching of solids, largely composed of amorphous Fe, Al and Mn oxyhydroxide solids, indicate the low Ra concentrations are largely retained in recalcitrant leach residues. REE concentrations in the leached residues are also largely retained in the solids throughout the leaching process. These results provide an understanding of overall Ra activities and sediment associations in AMD solids and are discussed with perspective toward guiding targeted REE extraction activities.