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Germanium redistribution during weathering of Zn mine wastes: Implications for environmental mobility and recovery of a critical mineral

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Legacy mine wastes are abundant globally and can contain toxic and/or valuable elements, including critical minerals. Often little is known about the biogeochemical behavior of critical minerals in the waste environment. Investigating the mineral hosts and speciation of these elements in mine waste, and how these change during weathering, is fundamental to improving understanding, and can shed light on the potential for solid and aqueous transport, and strategies for isolation, remediation, and/or recovery. The Tar Creek Superfund Site (TCSS) in northeast Oklahoma, a location producing Zn, Pb, and Ge until the 1970s, is a natural laboratory for understanding the behavior of Ge and other critical minerals in mine wastes. At the TCSS we studied Ge in bulk and size-fractionated surficial mine waste samples using bulk and microanalytical techniques. In the sampled waste, locally known as chat, Ge has been redistributed from sphalerite (ZnS), its original host mineral, to hemimorphite ($Zn_4Si_2O_7(OH)_2 \cdot H_2O$), a weathering product that forms rinds of fine needle-like particles on both quartz and sphalerite grains. Despite hemimorphite and sphalerite being commonly <2 wt.% in bulk chat, hemimorphite hosts 64% on average of Ge in the chat sampled. Sphalerite hosts only 10% on average of the Ge. Quartz, an abundant mineral (~90 wt.% in bulk chat) with low Ge concentration (~2 ppm compared to averages of 320 ppm and 240 ppm in hemimorphite and sphalerite, respectively), hosts the remaining Ge (26% on average). Geochemical modeling shows that hemimorphite provides a thermodynamically stable secondary repository for Ge in chat. However, hemimorphite is fine-grained and readily soluble in simulated gastric and lung fluid, potentially releasing Ge if ingested or inhaled. While Ge has been linked to adverse health impacts via ingestion at high concentrations associated with dietary supplements, acute toxicity from ingestion of chat is unlikely at this site. However, little is known about low-level, chronic toxicity of Ge, or effects from its inhalation. This study shows how weathering can directly influence the distribution and mineral hosts of Ge in mine wastes, which in turn can control the environmental stability, potential bioavailability, and metallurgical recovery strategies for Ge.