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Colloid-controlled arsenic transport from carbonate-rich mine tailings to surface waters

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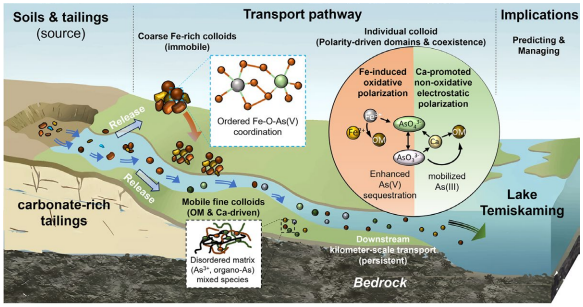

Particulate pollutants play a central role in contaminant transport across environmental compartments, yet their transformation and mobility under neutral, carbonate-buffered conditions are not well understood. In mine-impacted systems, arsenic (As) is often thought to be immobilized by Fe (oxyhydr)oxides at circumneutral pH. However, persistent As contamination in surface waters downstream of reclaimed or naturally neutralized tailings suggests that particulate-mediated pathways may dominate long-distance transport.

This study explores how colloids and mineral–organic matter associations (MOAs) influence As transport from carbonate-rich mine tailings through soils, creeks, and lakes in the historic Cobalt mining district (Ontario, Canada). We integrate size-resolved filtration (coarse: 450 nm–5 µm; fine: 20–450 nm; truly dissolved: <20 nm), water chemistry, zeta potential measurements, transmission electron microscopy (TEM), electron energy-loss spectroscopy (EELS), and As K-edge X-ray absorption spectroscopy (XAS) to link watershed-scale As fluxes with processes occurring at the particle and molecular levels.

Across transport, arsenic is predominantly associated with particulate and colloidal fractions rather than the truly dissolved phase. Near tailings, Fe-rich colloids host As(V) in structurally ordered scorodite-like and ferrihydrite-associated forms and preferentially accumulate in coarse fractions, limiting transport. With increasing distance, fine colloids enriched in organic matter and Ca dominate As transport. Correlation analyses show that these colloids remain electrostatically stabilized despite neutral pH, decoupling surface charge from classical pH-dependent mineral behavior.

TEM and XAS reveal that organic-rich colloids host mixed As(III)/As(V) species in structurally disordered environments, while EELS demonstrates nanoscale polarity changes in organic matter induced by Fe and Ca. Fe promotes oxidative polarization and As(V) immobilization, whereas Ca enhances non-oxidative polarization that stabilizes As(III) within organic matrices. These nanoscale transformations allow mobile and immobile As species to coexist within individual particles.

Our findings show that fine colloids act as persistent particulate pollutants, linking tailings, soils, creeks, and downstream lakes such as Lake Temiskaming (Figure 1). The study underscores how the transformation of particulate matter from Fe-rich aggregates into Ca- and organic-associated colloids affects contaminant mobility across environmental compartments. These findings have direct implications for aquatic ecosystems, drinking water safety, and long-term human health risks in areas affected by legacy mining.