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Adsorption to Al-, Fe-, and Mn-oxyhydroxides dominates rare earth element (REE) and P mobility in a headwater stream in Vermont, USA

Sleepers River watershed W-9, a U.S. Geological Survey site in Vermont USA, is underlain by Waits River Formation carbonaceous schist with impure rusty-weathering limestone beds. Quartz, feldspar, two micas, garnet, calcite, and pyrite dominate. Trace apatite, monazite, and possibly calcite are sources of REEs. Till, containing abundant local bedrock and erratics of granite, ranges up to 4 m thick. We evaluated REE and P mobility for three discharge events. Events included eight samples, distributed from baseflow to peak(s) to nearly baseflow, and speciated for **Total** (unfiltered and acidified) and **Dissolved** (0.45 µm-filtered and acidified).

E1 (large Fall rainstorm; 0.014 to 3.696 mm/h): **Total** Al, Fe, and Mn increased from 0.21, 0.83, and 0.85 µmol/L, respectively, to 376(1790X), 161(194X), and 38(45X) µmol/L; **Total** Al/Fe≈2. **Total** Ca **(≈Dissolved**) was diluted, as Mg and K increased slightly (leaf senescence). **Total** Na (**≈Dissolved** Na) remained low and relatively constant. Concurrently, **Total** La, Ce, Pr, and Nd increased from 0.15, 0.17, 0.04, and 0.13 nmol/L, respectively, to maxima of 87(580X), 114(671X), 15(375X), and 53(408X) nmol/L, nearly concurrently. At or near peak discharge, **Dissolved** La, Ce, Pr, and Nd were <3% of **Total**. **Total** La, Ce, Pr, and Nd during events had negative hysteresis, suggesting that particulates of Al, Fe, and Mn were mobilized from the stream bed, while precipitation of fresh Al-, Fe-, and Mn-oxyhydroxides occurred during higher discharge of groundwater, higher pH (degassing of excess CO_2), and higher PO_2. **Total** export for E1 was Ce>La>Nd>Pr for the light REEs while four heavier REEs were typically Dy>Ho>Er>Tm, also with negative hysteresis.

E2 (snowmelt; 0.407 to 0.941 mm/h): Maximum **Total** Al was 10% of **Total** Al for E1 and constant, while maximum Al/Fe was ≈0.10. Base cations (Ca>Mg>K>Na) remained nearly constant with **Total≈Dissolved**. **Total** La, Ce, Pr, and Nd increased 33X, 27X, 25X, and 24X, respectively, while **Total** Mn increased 80X, greater than Al(≈0X) or Fe(12X) during the first peak.

E3 (small Fall rainstorm; 0.006 to 0.073 mm/h): As discharge increased from 0.006 to only 0.013 mm/h, mobilization of most REEs surged 3X-5X. Base cations remained nearly constant (Ca>Mg>K>Na), with **Total≈Dissolved**. During E3, **Total** Al/Fe≈0.67-1.59, consistent with base flow dominated by weathering pyrite. Maximum **Dissolved** P declined from 0.89 (E1) to 0.27 µmol/L (E3); **Total** P declined from 15 (E1) to <1 µmol/L (E3). REEs and P are dominantly exported adsorbed on particulate Al-, Fe-, and Mn-oxyhydroxides (partitioning unknown), with Al>Fe>Mn during E1.