

Changes to the mineralogical, geochemical, and isotopic compositions of mineral feedstocks during enhanced rock weathering: Implications for carbon verification

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Mg- and Ca-rich rocks are potential feedstocks for enhanced rock weathering (ERW) as their reaction with CO₂ can lead to the formation of secondary carbonates. Kimberlite residues from the Venetia diamond mine (South Africa) and powdered brucite- and wollastonite-rich rocks were tested as ERW feedstocks. Automated wet-dry cycling (4/day) experiments ran for 1 yr under laboratory conditions to simulate weathering of rock powders. Two primary reaction pathways were identified: A) dissolution and reprecipitation of carbonates (undesirable), and B) silicate/hydroxide dissolution leading to carbonate precipitation (desirable). Kimberlite residues that contained serpentine (19–33 wt.%) and calcite (4–5 wt.%) experienced variable and minor changes in total inorganic carbon (TIC). The δ¹³C and δ¹⁸O values of the carbonate minerals approached those expected for atmospheric-derived CO₂ but are best explained by the exchange of carbonate CO₂ with atmospheric CO₂ as opposed to net CO₂ sequestration (pathway A). In contrast, there was substantial carbonation of brucite (1.22 to 5.98 ‰C) and wollastonite (0.22 to 1.01 ‰C) as indicated by the increases in TIC (pathway B); yet, decreases in δ¹³C values resulted from kinetic isotope fractionation due to carbonation being CO₂-limited. Unweathered (blue ground) and weathered (yellow ground) kimberlites from the Voorspoed diamond mine (South Africa) were compared to kimberlite residues from experiments. Yellow ground contained twice the TIC content relative to blue ground, while quantitative scanning electron microscopy showed that most additional carbonate occurs as calcite veinlets. Depletion in ¹³C and enrichment in ¹⁸O of the yellow ground carbonates supports weathering by meteoric waters that are out of equilibrium with atmospheric CO₂. Furthermore, both Voorspoed kimberlites were essentially radiocarbon dead. However, during weathering experiments, substantial modern carbon was incorporated into kimberlite residues and brucite powder, regardless of the reaction pathway. Although carbon isotopes have been used to verify atmospheric CO₂ sequestration in mine wastes [1–2], these isotopes are affected by CO₂ exchange and kinetic fractionation during weathering that can be misleading and thus, should not be relied on as the sole method