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Verifying and quantifying carbon fixation in minerals from serpentine-rich mine tailings using the Rietveld method with X-ray powder diffraction data

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ABSTRACT

Most carbon on Earth is bound within minerals, and increasing the uptake of atmospheric carbon dioxide into minerals may reduce the greenhouse gas content of the atmosphere. We document carbon disposal through the mineralization of mine tailings at Clinton Creek, Yukon Territory, and Cassiar, British Columbia. We confirm crystallographic binding of carbon in these tailings and quantify carbon dioxide uptake using quantitative phase analysis with the Rietveld method for X-ray powder diffraction data. Planar disorder in the structures of the kaolinite-serpentine group minerals makes Rietveld refinements of X-ray powder diffraction data for serpentinites problematic. Using structureless pattern fitting and with the addition of a known quantity of a well-crystallized material, the problem of structural disorder is overcome by considering the serpentine minerals as amorphous phases. We test the accuracy and precision of this refinement method using synthetic serpentine-rich mine tailings of known composition. Estimates of the abundance of hydrated magnesium carbonates in these tailings have a precision of approximately 5% relative for mineral species present in amounts greater than 10 wt%. Precise estimates of carbonate mineral content and crystallographically bound atmospheric CO_2 are made for samples of serpentine-rich tailings from Clinton Creek and Cassiar. Results for mine tailings are also compared to mineralogically similar samples from a carbonate playa at Atlin, British Columbia. The potential for decomposition of metastable hydrated magnesium carbonate phases to geologically stable magnesite may represent long-term stability of the products of mineral sequestration in mine tailings.

Keywords: Carbon disposal, mineral sequestration, quantitative phase analysis, Rietveld method, Pawley method, X-ray powder diffraction, serpentinite, mine tailings, carbonation, order-disorder