

Constraining the role of low-temperature Mg-clays on carbon dioxide sequestration in natural and engineered alkaline environments

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Although Mg-rich clays are common in sediments, rocks and mineral wastes produced by mining, little is known about how they might contribute to carbon dioxide (CO₂) sequestration. The observed correlation of Mg-phyllsilicates (Mg-clays) and Mg-carbonates in microbialites and sediments from alkaline lakes could offer answers to the “silica problem”, which is the observation that co-precipitation of Mg-clays with Ca- and Mg-carbonate minerals decreases the efficiency of CO₂ sequestration during mineral carbonation^[1]. For instance, lakes on the finely ground basaltic glacial sediment of the Cariboo Plateau, British Columbia, Canada, precipitate Mg-carbonates and Mg-clays^[2,3]. The precipitation of these phases heavily influences the aqueous geochemistry, as the combination of evaporation and mineral precipitation consumes cations and alkalinity^[4].

We have conducted a series of field and experimental studies to seek answers to the “silica problem”. These involve examining the mineralogy and aqueous geochemistry of sediments and microbial mats found in alkaline lakes of the Cariboo Plateau. Results show that Mg-clay precipitation is highly favored within microbial mats over Mg-carbonates in alkaline lakes, stressing the role of microorganisms in Mg-clay precipitation. Additionally, we performed a series of batch experiments testing the hypothesis that authigenic 2:1 Mg-clays, while less reactive to CO₂ than other Mg-rich minerals (*e.g.*, brucite and serpentines), can transform into carbonates under high alkalinity conditions (100–1187 mEq/L). Although we observed precipitation of a Mg-carbonate, dypingite [Mg₅(CO₃)₄(OH)₂·~5H₂O], on the surface of synthetic stevensite [(Na_{0.082}Ca_{0.013}·nH₂O)Mg_{2.92}Si₄O₁₀(OH)₂] in laboratory experiments, our field data suggest that Mg partitioning between authigenic clays and carbonates in alkaline lakes is strongly influenced by Si_(aq) concentration, alkalinity, and pH. Lastly, we discuss the implications of Mg-clay dissolution and precipitation for carbon sequestration projects. These findings provide important insights and observations that can contribute towards geochemical models of environments where we observe carbonation of clay minerals (*e.g.*, alkaline lakes) which can serve as natural analogues for industrial carbon sequestration projects.

^[1]Oelkers, E.H., et al. (2019). *Geochimica et Cosmochimica Acta*, 245, 542-555.

^[2]Renaut, R.W. (1990). *Saline Lakes*. May 1988 (pp. 67-81).

^[3]Raudsepp, M.J., et al. (2024). *Chemical Geology*, 648, 121951.

^[4]Eugster, H.P. & Hardie, L.A. (1978). *Lakes: chemistry, geology, physics* (pp. 237-293).