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Chemical controls on carbonate sediment production and phosphate concentration in alkaline lakes

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As some of the most biologically productive environments on the modern Earth, alkaline lakes may have figured prominently in the evolution of the global carbon cycle and in the chemical origins of life. Significant $CaCO_3$ supersaturation is a ubiquitous trait of alkaline lakes, controlling the geochemical evolution of lake waters and the mineralogy and sedimentary expression of their deposits. By definition, $CaCO_3$ supersaturation requires compounds that interfere or inhibit precipitation, yet the processes controlling $CaCO_3$ saturation in many alkaline lakes are poorly understood. In this study we focus on the role of aqueous phosphate in influencing the dissolved carbonate system. We investigate a compilation of alkaline lakes' water chemistry data from 14 countries using a Pitzer ion interaction activity coefficient model to explore carbonate geochemistry and the potential role of phosphate in maintaining $CaCO_3$ supersaturation. Our observations are compared with new experimental fluid chemistry results from Na-Ca-Mg-CO_3-Cl-PO_4 solutions mimicking the extreme end of alkaline waters under evaporitic conditions. Our findings support previous work pointing out that phosphate can reach high concentrations under high alkalinity [1,2]. However, mass-balance calculations show that initial Ca^{2+} removal from solution was >90% in all experiments, producing Mg/Ca ratios close to 20 during approximately 2/3 of evaporation, even after their introduction in the ratio 1:1. Under these circumstances, Mg^{2+} acts as a stronger inhibitor, maintaining >25% more dissolved Ca^{2+} compared to those solutions with phosphate alone. This independently shows how Mg- and Ca-poor solutions can achieve remarkably high Mg/Ca ratios, leaving other ions, such as dissolved silica, as principal controls on soluble Mg^{2+} concentrations [3]. Together, these data confirm that interactions between phosphate and Mg^{2+} primarily control the dissolved carbonate chemistry of many alkaline lakes and influence kinetic processes governing carbonate sedimentation. Furthermore, they provide additional evidence that high soluble phosphate concentrations may have been attained in alkaline settings on early Earth, thereby not representing a major hindrance for prebiotic chemistry.

[1] Toner & Catling (2020) PNAS 117, 883–888. [2] Ingalls et al. (2020) GRL 47 doi:10.1029/2020GL088804. [3] Tutolo & Tosca (2018) GCA 225, 80–101.