Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.16573

Mineral surface roughness resulting from dissolution as a proxy for the reaction conditions: a potential biosignature?

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Determination of biosignatures based on the microtopography of naturally weathered mineral samples has often turned out to be equivocal. For instance, Fisk et al. showed that etching features and microchannels similar in 'size, shape, and distribution' to those found in naturally weathered basaltic glasses and attributed to the activity of microorganisms, can be reproduced through fully abiotic dissolution experiments [1]. However, solely relying on qualitative comparisons of the microtopography can hide some of its potential in differentiating between those processes. Here, we propose a statistical characterization (using power spectral density and semi-variogram) of the steady-state surface roughness resulting from dissolution as a tool to quantitatively differentiate between biotically and abiotically weathered mineral surfaces. First, we performed flow-through abiotic dissolution experiments at room temperature and atmospheric pCO2 with mechanically-polished calcite crystals at different degrees of fluid undersaturation ($0 \leq \Omega \leq 0.8$) in alkaline conditions (pH = 7.9). We showed that steady-state surface roughness could be used as a proxy for the reaction conditions [2]. The Ω-roughness relationship derived from experimental observations was explored through kinetic Monte Carlo (kMC) modeling of mineral dissolution, attempting to identify the atomic scale factors that might play a role in determining the trend detected. We then performed the corresponding biotic experiments by covering the calcite surface subject to dissolution with a biofilm of Chroococcidiopsis thermalis PCC 7203 cells. Our results suggest that, at $\Omega \leq 0.3$, the presence of a biofilm on the dissolving calcite substrate produces surface features that can be captured by our statistical characterization. Finally, the extent to which a statistical characterization of the steady-state surface roughness resulting from dissolution can be used to back-estimate the reaction conditions, while quantitatively differentiating between abiotic and biotic processes, will be discussed.

[1] Fisk, M. R.; Crovisier, J.-L.; Honnorez, J., Experimental abiotic alteration of igneous and manufactured glasses. *Comptes Rendus Geoscience* **2013,** *345* (4), 176-184, https://doi.org/10.1016/j.crte.2013.02.001.

[2] Stigliano, L.; Ackerer, P.; Benzerara, K.; Daval, D., Linking calcite surface roughness resulting from dissolution to the saturation state of the bulk solution. In *Goldschmidt*, Honolulu, **2022,** https://doi.org/10.46427/gold2022.9474.