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Biotite oxidation, fracturing and subsurface particle transport under a granitoid watershed

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The chemical weathering rate of silicates at the Río Icacos watershed in the Luquillo Critical Zone Observatory (Puerto Rico) is among the highest reported for granitoid watersheds. The high weathering flux at Río Icacos has previously been attributed to fast infiltration of oxygenated meteoric water through the interconnected fractures, which is hypothesized to be initiated by biotite oxidation. However, the direct evidence of biotite oxidation and fracturing is still lacking. In this study, we used a multi-scale approach to investigate the spatial relationship between fractures and oxidation signature of biotite.

In Río Icacos, spheroidal weathering results in cm-sized, onion-like rindlets that wrap around increasingly spheroidal corestones. Each rindlet is separated from other rindlets by macro-fractures. Combining the measurements by micro Raman spectroscopy and electron microprobe on rindelts, we found an oxidation signature only on biotite grains (sub-mm in diameter) on the edge of the rindlets (near fractures). Transmission electron microscopic imaging, electron and X-ray diffraction and simulation of diffraction patterns through crystal models further illustrated that in situ transformation of biotite is initiated by oxidation of Fe(II) in the octahedral site and release of K in the interlayer to form vermiculite. Microscopic imaging shows extensive exfoliation parallel to the [001] basal plane of oxidized biotite. We found that the transformation of biotite to vermiculite, a volume expansion reaction, not only induces the formation of (micro)fractures but also weakens the rock around the fractures until particles (sub-mm to mm) detach and are mobilized by subsurface flow through fractures. The subsurface particle transport is evidenced by similar textures and compositions of particles collected from a seep to the exterior of rindlets, where weathered biotite grains are located. As such, the fracture surfaces serve as hot spots for weathering and subsurface erosion in Río Icacos. Our results highlighted the importance of biotite oxidation in exposing fresh particle surfaces to weathering fluids, which might be a key process to sustain the high rate of chemical weathering in Río Icacos.