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Chemical weathering processes in the 0-10 m of the critical zone developed on the Tégulines clay formation (Aube, France)

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Abstract

Interactions between clayrock, atmosphere and hydrosphere induce mineralogical, chemical, pore water composition, and also petrophysical changes in the clay formation [1]. The Lower Cretaceous Tégulines clay formation is an aquitard outcropping near Brienne-le-Chateau (north-eastern France) that Andra has investigated in the context of repository for low level radioactive waste. During the first drilling campaign, a multi-disciplinary approach highlighted the development of a critical zone (CZ) on Tégulines clay to depths of 20-25 m, with a highly reactive zone at depths of 10-12 m [2]. In this work, the main mineral and organic matter reactivities are presented as well as associated chemical weathering processes concerning the first 0-10 meters in which root network develops.

The first meters of sediments overlying Tégulines Clay (1-8 m) consist of soils developed on Quaternary clay silty loams, then carbonate-free and Fe-hydroxide-rich colluvium. At the loam-colluvium/Tégulines clay interface, claystone show often slip surfaces along hillslopes. They have rather yellowish-greenish color, and are swelling clay-rich, quartz-poor, and carbonate-poor. Gypsum and iron hydroxide entirely replaced pyrite. Claystone is marked by a high Cation Exchange Capacity (CEC) up to 37 meq/100 g (reduced Tégulines clay: 16-19 meq/100 g), and dominated by Ca cation (~80 %) in interlayer.

The first meters of Tégulines clay corresponds to the most reactive greenish clay. Clay fraction consists of a mixing between kaolinite (~35 %), illite-smectite mixed layers (~40 %) and illite-micas including glauconite (20-25 %). Clayrock shows CEC of 18-21 meq/100 g slightly higher than reduced clay, and Ca remains the dominant cation on the exchanger. The top of this zone is characterized by the highest dissolved inorganic carbon concentrations (DIC up to ~10 mmol/L compared to reduced clay: 1.5 ± 0.5 mmol/L) and sulfates (up to 30 mmol/L compared to reduced clay: 2 mmol/L) in pore water, associated with a peak of [O_2] and [N_2]. The presence of O_2 drives oxidative weathering of pyrite, organic matter and other Fe^{2+} bearing minerals including glauconite. The oxidation of pyrite releases protons, which leads to the calcite dissolution. Both oxidation of pyrite and organic matter contribute to the DIC increase. Based on the $\delta^{13}C$ of CO_2, organic matter degradation is the major source of CO_2 (~50-60 %) at the top of the reactive zone, then decreases to ~20-30 % at the bottom. Iron-hydroxides and gypsum are secondary minerals formed in the entire zone, whereas secondary carbonate concretions only precipitated at the top. The isotopic dating of concretions with $\delta^{13}C$, $\delta^{18}O$, $^{87}Sr/^{86}Sr$ and ^{14}C indicate that they precipitated from recent surface-derived fluids (<50 000 years).
Calcite dissolution, pyrite oxidation, organic matter and glauconite oxidation reactions, and secondary minerals precipitation represent “reaction fronts” associated with chemical weathering that can be used to infer the depth at which dissolved oxygen infiltrate the

subsurface profile and to provide information on the main flow paths and acids (CO_2, organic acids…) at the area scale.

[1] Brantley et al., (2018) Vadose Zone J. 17, Article ID 180092.
[2] Lerouge et al. (2018) Geofluids, doi.org/10.1155/2018/1606753.