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Dissolved gas (CO₂, alkanes, O₂, N₂) in critical zone developed on claystone

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Weathering processes in clay environments are of major importance because they participate to regulate elemental cycling and mass transfer in the critical zone with major implications for carbon and nitrogen cycling.

In that aim, we measured 1) dissolved CO₂, alkanes, O₂ and N₂ concentrations in clay pore waters by rock degassing, 2) soil gas flux and concentrations, and 3) $\delta^{13}\text{C}$ of CO₂ and alkanes in two contrasted tectonic contexts.

The first context is the marine Jurassic black marls in the French Alps, characterized by deep burial, high erosion rates and dominant physical weathering processes. These marls are well-known for their occurrences of natural methane gas seeps. In this area, we carried out rock degassing on outcropping weathered claystone in complement of soil flux measurements to constrain the implication of weathering processes on the natural gas releases. These measurements are also tested as a new component of environmental baseline assessment in the field of unconventional hydrocarbons.

The second context is the marine Cretaceous Tégulines Clay of the north-eastern part of the Paris Basin, characterized by low burial, low erosion rates, and dominant chemical weathering processes. In this area, we carried out rock degassing on soil and weathered claystone accessible by deep boreholes, in order to define the depth of the critical zone and major reactions controlling the weathering profile.

Oxygen and nitrogen concentrations are the record of the atmospheric diffusion through the formations. Some values are higher than the gas solubility, which could be attributed to rock desaturation and air bubbles, and clay sorption (only for nitrogen).

Weathering processes induce a significant CO₂ increase and a large range of $\delta^{13}\text{C}_{\text{CO}_2}$, providing evidence of two major CO₂ sources: CO₂ internally controlled by carbonates and organic-derived CO₂ of internal and external origins. In Alpine black marls, field observations suggest a low depth affected by weathering, due to intense erosion. In Tégulines Clay, the CO₂ increase provides evidence of a ~ 20 m-thick critical zone. The lowest $\delta^{13}\text{C}_{\text{CO}_2}$ indicates that the highest reactive zone

(organic matter degradation, calcite dissolution and pyrite oxidation) is ~10 m deep, in agreement with the depth of the root network.

Nature and amounts of alkanes are contrasted in the two contexts. In deep burial environment, alkanes are abundant, in particular, in the "fontaines ardentes" gas seeps in the French Alps. Composition of hydrocarbon gas and $\delta^{13}\text{C}$ of methane strongly suggest a thermogenic origin. Outcropping black marls contain methane, suggesting oxidation of higher alkanes. That assumption is supported by $\delta^{13}\text{C}_{\text{CO}_2}$ of soil close to $\delta^{13}\text{C}$ of alkanes. In low burial environment, small amounts of methane are present that rapidly disappear with weathering. Some methane concentrations could be attributed to diffusion of external methane formed by degradation of organic matter under reducing conditions in soil.

Overall those results suggest that dissolved gas and their isotopic signature are good markers of weathering processes in the critical zone.

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