Dissolved Oxygen stable isotopes in groundwater: a new tracer of biogeochemical reactivity for the underground Critical Zone

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The persistence of dissolved oxygen (DO) in deep groundwater sustains microbial life and biogeochemical reactivity, with potential impacts on large scale nutrient cycling. In aquifers, dissolved oxygen is often heterogeneous and intermittent, but the driving factors of the spatial and temporal DO distributions remain poorly identified. This study aims to identify the reactive processes controlling DO dynamics in a fractured-bedrock catchment in Brittany, characterized by strong surface-groundwater connectivity, rapid hydrologic response and deep microbial hot-spots.

Oxygen stable isotopes of DO and of the water molecule, major gases, CFC concentrations and major cations and anions were analyzed on rainwater and on groundwater sampled from 10 150m-deep piezometers located in recharge and discharge zones of the catchment. We show that DO concentrations decrease with increasing water residence time while the $\delta^{18}O-DO$ signatures show two different trends depending on DO concentrations. DO concentrations ranging from 10 - 1.5 mg/L are associated with an unexpected depletion of $\delta^{18}O-DO$ suggesting that the DO pool enriches in ^{16}O as it is consumed. Conversely, DO concentrations below 1.5 mg/L (the so-called microaerobic range) are associated with an increase of the $\delta^{18}O-DO$ consistent with an enrichment fractionation of 0.9950 – 0.9980. Our results reveal a shift in the reactive processes controlling DO dynamics and $\delta^{18}O-DO$ at catchment scale.

Water chemical data as well as laboratory reactivity batch experiments indicate that, at high DO concentrations, the abiotic oxidation of dissolved Fe(II) is the main reactive process consuming DO but is not responsible for the observed $\delta^{18}O-DO$ depletion. Isotopic equilibrium exchanges of ^{18}O between DO and water, which $\delta^{18}O-H_2O$ compositions range from -5 to -5.2 ‰, could however explain the light DO isotopic signatures observed despite short (below 50 years) residence times. On the other hand, the observed enrichment in $\delta^{18}O-DO$ at low DO concentrations reflects a clear oxygen consumption by the activity of microaerobic iron-oxidizing bacteria (FeOB).

The present study shows a spatial heterogeneity of biogeochemical reactive processes controlled by oxygen concentrations. DO stable isotopes proved useful to unravel biotic and abiotic processes at stake in the deep critical zone with possible applications for the assessment of primary production of deep biomass.

Keywords: isotopic fractionation, dissolved oxygen isotopes, biogeochemical reactivity, Critical Zone, groundwater.