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Are Chlorine stable isotopes a tracer of anthropogenic inputs in the critical zone?

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Chloride ion (Cl^-) is a conservative solute widely used as a hydrological tracer in watershed studies. Its low background levels and substantial presence in anthropogenic sources make it an indicator of human impact on water bodies. Prior works[1] on the Cl stable isotopic composition ($\delta^{37}Cl$) of chlorides have helped elucidate the geodynamic cycle of Cl, ion transport in porous media, magmatic and volcanic processes, etc. The homogeneity of oceanic $\delta^{37}Cl$ (at 0 ‰)[2] provides a global reference standard, while the salts and organochlorine compounds used in industrial and agricultural applications display a wide $\delta^{37}Cl$ range (-6 to +6 ‰). Due to this variability, $\delta^{37}Cl$ has the potential to serve as a valuable tracer for anthropogenic contamination in freshwater systems, particularly when compared to ocean-derived atmospheric inputs.

However, the natural variations in $\delta^{37}Cl$ of chlorides from freshwater systems remain largely unexplored. Our research aims to fill this gap by studying the $\delta^{37}Cl$ composition of water samples from different compartments of the critical zone (CZ). We analyzed water samples from French CZ observatories[3] across a range of geographic and climatic contexts. Preliminary results suggest the presence of a coast-inland gradient, with the coast-proximal Naizin catchment ($\delta^{37}Cl$ mean: 0.06‰, SD 0.15, n=29) clustering close to that of seawater and indicating the dominance of oceanic input through rain. Contrastingly, results from the inland, agricultural-dominated Orgeval catchment show a notable depletion relative to seawater ($\delta^{37}Cl$ mean: -0.22‰, SD 0.34, n=56). This observation is concurrent with lower observed [Na]/[Cl] at Orgeval, indicating the chlorine-enriching alteration of oceanic inputs possibly through fertilizer inputs and urban emissions.

This study marks one of the first efforts to establish the Cl-isotopic composition in rivers and in defining the riverine input to the global budget. Ongoing work will refine our understanding by analyzing potential end-members, studying additional catchments and shedding light on fractionation mechanisms.

[1] Eggenkamp (2014), The geochemistry of stable chlorine and bromine isotopes.

[2] Godon et. al. (2004), Chemical Geology 207, 1–12

[3] Gaillardet et. al. (2018), Vadose Zone Journal 17, 1-24