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Remobilization of chromium by organic ligands in paddy soils

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Long-term pollution mitigation requires a better understanding of the biogeochemical processes that regulate the behavior and fate of trace metals, such as chromium (Cr), in subsurface environments. Cr has two oxidation states, hexavalent Cr(VI) and trivalent Cr(III), which are respectively soluble (toxic) and insoluble (less toxic). The redox cycles of Cr, iron (Fe), and manganese (Mn) are coupled during the early diagenetic transformations, whereas the Cr-Fe-Mn redox interplay is complicated by naturally occurring low molecular weight organic ligands, which include, e.g., organic acids and biogenic substances such as siderophores.

To better constrain the relative importance of Cr reduction and mobilization processes in subsurface environments, we conduct flow-through column experiments with soil samples collected from paddy fields that are representative of organic-rich reducing environments. Our column-flow experiments show increased levels of dissolved Cr(III) in the presence of citric acid and DFOB despite Cr reduction. Solid phase analyses suggest that the increased mobility of Cr is likely related to incongruent dissolution of Fe and Mn oxides. Cr isotopic composition ($\delta^{53}Cr$) is further validated as a tracer for Cr(III)-ligand complexation.

This study is complemented by pot experiments with the same paddy soil material under continuous and intermittent flooding conditions, from which we show that the addition of citric acid and DFOB can influence the Cr-Fe-Mn interplay in the soil-plant system over time and ultimately the accumulation of Cr in rice (*Oryza sativa*). These results highlight the role of organic molecules as complexing agent *vs.* electron donor and highlight ligand-promoted Cr remobilization as an overlooked but important process in a diversity of Earth's environments.