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Enhancing environmental radionuclides tracking: High-resolution isotopic analysis using NanoSIMS

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Human activities such as nuclear operations, and uranium (U), coal, and phosphate mining have induced the release of radionuclides into the environment, raising questions about their persistence and effects on populations and ecosystems due to ionizing radiation exposure. Understanding the sources, dispersion mechanisms, and spatiotemporal evolution of these contaminants is crucial for developing strategies to manage contaminated sites and associated waste.

Traditional bulk sample analyses often conceal critical information about contamination sources because of isotopic mixing and/or concentration dilution caused by the matrix elements of the geochemical background. They also fail to precisely identify the nature and past evolution of radionuclide-bearing phases. To overcome these challenges, microscale Secondary Ion Mass Spectrometry (SIMS and NanoSIMS) proves to be a suitable technique. This study focuses on the development of an analytical method for SIMS to trace contamination sources in Rophin's wetland (France) impacted by U-mining and milling activities, and NanoSIMS to understand radionuclide mobility at the grain scale. Stable lead and uranium decay products (^{238}U, ^{230}Th, ^{226}Ra) were calibrated with a natural autunite ($Ca(UO_2)_2(PO_4)_2$ • 10-12 H_2O) mineral from Margnac (France) and synthesized barite ($BaSO_4$) standards containing 2.89 and 9.65 $mg.kg^{-1}$ ^{226}Ra. Additionally, this study introduces a novel approach for NanoSIMS data processing using spatial analysis tools from geostatistics. This enhances the interpretation of isotopic ratio distributions at the grain scale, offering insights into the mechanisms of radionuclide incorporation, stability, and mobility in complex environmental settings.

Bulk soil analyses indicated an initial U-ore signature dominated by parsonsite ($Pb_2(UO_2)(PO_4)_2$) ($^{206}Pb/^{208}Pb \approx 0.48$), with slight isotopic variations in certain soil layers ($^{206}Pb/^{208}Pb$ up to 0.6), suggesting secondary contamination sources. SIMS analysis highlighted U-bearing phases ($^{206}Pb/^{208}Pb \approx \sim 9$) inherited from pitchblende (UO_2), a U-ore from Bois-Noir Limouzat that was also processed at the Rophin site. NanoSIMS provided new insights into the heterogeneous distribution of ^{238}U, ^{226}Ra, and ^{230}Th within individual grains, distinguishing between newly formed and inherited U-bearing phases. These results bring information on processes governing radionuclides mobility (alteration, adsorption, precipitation). Finally, this work provides insights into the historical functioning of this wetland which can be considered in models predicting radionuclide mobility for future projections.