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The use of stable lead isotopes for the identification of the sources and transport processes leading to the release of radioactive contaminants downstream of former uranium mine sites

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Radioactive enriched materials generated by uranium mining activities potentially release radionuclides and associated heavy metals into the environment, leading to high uranium accumulation in soils and sediments around the former uranium mine. However, high uranium content comparable to that from mining activities, due to natural weathering and erosion of local bedrock was also reported (up to 6000 $mg.kg^{-1}$). Thus, highlighting the potential sources and mechanisms driving radioactive elements dissemination in the vicinity of former U mines is a key element for the decision-making process, effective management, and remediation of contaminated sediments and soils. This research aims to identify the sources of radioactive contaminants and their transport mechanism using stable Pb isotopes and radioactive disequilibrium of the ^{238}U-decay series.

The study area is a wetland located downstream of the former Rophin U mine (Zone atelier territoires uranifères, France). Soils and sediments were sampled in the wetland and radiological and chemical analyses were performed (platforms PATERSON and LUTECE, IRSN). The uranium content reaches to 16000 $mg.kg^{-1}$ in the surface organic layer (34 $mg.kg^{-1}$ in local bedrock). The radiogenic $^{206}Pb/^{207}Pb$ ratio (~1.73) and the alignments of samples in the three Pb isotopes (^{206}Pb, ^{207}Pb, ^{208}Pb) plot point out the impact of mining activities. Uranium originates probably from three sources related to the mining history of the site: i) pitchblende UO_2 due to mechanical and chemical processing from Bois Noirs ore ii) parsonsite $Pb_2(UO_2)(PO_4)_2$, the local dominant U mineralization and iii) U from the local geochemical background. The high content of common Pb in parsonsite hides its radiogenic Pb signature limiting thus the use of the Pb isotopes as a relevant fingerprint for this case. Additionally, observations of Pb-bearing phases associated with phosphate by SEM strengthen our hypothesis. The variation of activity ratio $^{230}Th/^{238}U$ measured by gamma spectrometry also indicates the mobility of uranium in the profile, probably due to the fluctuation of the water table in the wetland. Finally, a geostatistical approach was explored to highlight lateral radioelement mobility at the wetland scale based on the data of in situ and mobile gamma measurements. Preliminary findings will be shown during the presentation.