Goldschmidt 2025 Abstract
https://doi.org/10.7185/gold2025.29314

Identification of natural and anthropogenic sources of geogenic radionuclides by using a multi-isotope tracing approach

ARNAUD MANGERET[1], JEAN-LOUIS REYSS[2], DR. JOSSELIN GORNY[3], TINGTING GENG[1], LUCIE STETTEN[4], GUILLAUME MORIN[5], MARIE-ODILE GALLERAND[1] AND ALKIVIADIS GOURGIOTIS[1]

[1]Autorité de sûreté nucléaire et de radioprotection (ASNR) - PSE/ENV/SPDR/LT2S, USDR
[2]Laboratoire des Sciences du Climat et de l'Environnement
[3]Autorité de sûreté nucléaire et de radioprotection (ASNR) – PSE-ENV/SPDR/LT2S, PSE-ENV/SIRSE/LER-NORD F-92260, Fontenay-aux-Roses, France.
[4]Institut de Minéralogie, de Physique des Matériaux et de Cosmochimie (IMPMC), Sorbonne Université, Muséum National d'Histoire Naturelle, CNRS UMR 7590, IRD
[5]Institut de Minéralogie, de Physique des Matériaux et de Cosmochimie (IMPMC), UMR 7590 Sorbonne Université-CNRS-MNHN-IRD

Environmental monitoring programs of nuclear legacy sites, formerly producing U-enriched materials, are mainly based on the interpretation of radionuclide concentrations in soils and sediments sampled downstream from these sites. However, the variations of these concentrations can be very low in upstream and downstream sampling locations. In addition, the different geological origins of soils and sediments can lead to different geochemical background values, which can be attributed wrongly to anthropogenic sources. It is therefore important to use geochemical tracers for accurately deciphering natural and anthropogenic radionuclide sources. This information is important for a better understanding of transport mechanisms of these substances in the critical zone and improving the evaluation of site management strategies.

The methodology proposed aims to provides tools for discriminating anthropogenic radioactive sources linked to former U mining activities, with regard to those originating from the natural weathering and erosion of rocks. This approach was performed according to a multi-isotope tracing approach, including the determination of (^{226}Ra/^{228}Ra) and (^{234}U/^{238}U) Activity Ratios (ARs) combined with the knowledge of the other ^{238}U decay series nuclide disequilibria and Pb stable isotope ratios.

Applied to a former U-mining-influenced watershed and lake, these isotopic tools reveal the existence of anthropogenic signatures in surface environments, several kilometers downstream from the mining source, several decades after the end of site rehabilitation operations. The integration of these results into the chronology of downstream lake sediment deposits showed that even if U contents remained twice the local geochemical background, these deposits sampled in a lake recorded that a part of U has been brought as dissolved species from the site into the sedimentary column. Moreover, the analyses of older lake sediment deposits indicated a clear signature of radiogenic Pb (i. e. Pb from U ores) associated to significant ^{230}Th activities, indicating the past transport of U decay series-enriched particles. Overall, this study emphasizes the high potential of multi-isotope tracing approaches for documenting the origin of geogenic radionuclides in the critical zone. These results yield further mechanistic explanations for the observed evolution of U concentration and speciation previously reported from mineralogical and spectroscopic analyses of these lake sediments.