

Post-doctoral position :

Geochemical behavior of radium in wetland impacted by former uranium mining activities : mineralogical characterizations and leaching experiments.

OFFICE/LABORATORY :

PSE-ENV/SEDRE/LELI under the responsibility of Arnaud MANGERET (arnaud.mangeret@irsn.fr) - Availability required from January 2022

Post-doctoral research topic :

In France, uranium ore extraction activities concerned 250 sites, from 1948 to 2001 (IRSN 2017), in which 75 % are located in the Loire-Bretagne watershed. Uranium and its daughter isotopes can be transported downstream of these sites, even several decades after rehabilitation operations. Several studies demonstrated the presence of uranium mining impacted deposits in wetlands soils located downstream of U extraction sites. Recent research efforts showed elevated activities of natural radionuclides, such as radium, which were significantly higher than the geochemical background and also the potential migration of these radionuclides to non-contaminated soils [2], [3]).

These wetlands are characterized by fluctuations of the water table level, leading to changes of redox conditions. In these natural systems, the geochemical behavior of uranium is largely known to be governed by these conditions ([4], [5]) while the fate of radium is not directly sensitive to redox conditions but rather related to the stability of its chemical species [6].

Radium migration is mainly controlled by sorption and coprecipitation processes onto mineral phases in oxidized surface media. Some of these phases, such as iron and manganese oxyhydroxides, are highly sensitive to reducing conditions, which are encountered in water-saturated media. It is thus important to determine radium-bearing solid species in these contaminated wetlands and if the radium-enriched mine deposits contamination may spread to overlying and underlying soil layers.

For addressing these scientific issues, laboratory scale studies will be performed. This approach usually requires small amounts of material, both in terms of solid and water samples. For this reason, the analysis of radium represents an important analytical challenge. The strategy that we want to utilize is to directly quantify the radium concentration by ICP-MS techniques after a chemical separation step. These analytical problems are also exacerbated for the determination of radium chemical species and its distribution in solid samples. Direct techniques usually used for the determination of solid speciation (e. g. for uranium) required Ra atom-rich samples, which are generally inconsistent with radioprotection provisions. However, current analytical advances, including mass spectrometry techniques, such as nano-SIMS, can lead to acquire new knowledges regarding radium distribution in mining-impacted solid samples.

The purpose of the post-doctoral work is to identify geochemical mechanisms controlling the radium fate from mining deposits under reducing conditions. For addressing this purpose, the study will be dedicated firstly to the determination of radium speciation in mining deposits. This characterization step will be carried out by combining i/ selective chemical extractions of mineral phases suspected to contain radium with ii/ some attempts to locate radium at the micrometric scale, according to nano-SIMS. This work will be undertaken with French experts of nano-SIMS analyses ; the post-doctoral student will actively contribute to this analytical development. For the second step, the student will perform laboratory experiments for mimicking the influence of water table fluctuations on radium migration in the wetland. On this basis, the student will monitor the radium contents in the liquid fraction by High-Resolution ICP-MS and the solid fraction by gamma spectrometry, these two techniques being available at the IRSN/LELI laboratory. Combined with the analyses of other major and trace chemical elements, these informations will be helpful to make assumptions regarding the mechanisms involved in the migration of radium towards uncontaminated wetland soils..

The results of this work will lead to a better assessment of management strategies for uranium mining impacted materials. This post-doctoral position is part of the european project Radonorm (<https://www.radonorm.eu/>) dedicated to bring an evaluation of public, workers and biota exposure to radon and NORM's (Naturally Occuring

Radioactive Materials). Acquired data will contribute to meet the purposes of the task 2.7 of the project where the effect of radionuclide speciation in NORM's on their mobility in the environment will be assessed.

References

- [1] IRSN (2021) MIMAUSA database, Memory and Impacts of uranium mines : synthesis and records, July 2021, <https://mimausa.bdd.irsn.fr/>.
- [2] Martin, A., Hassan-Loni, Y., Fichtner, A., Péron, O., David, K., Chardon, P., Larrue, S., Gourgiotis, A., Sachs, S., Arnold, T., Grambow, B., Stumpf, T., Montavon, G. (2020) An integrated approach combining soil profile, records and tree ring analysis to identify the origin of environmental contamination in a former uranium mine (Rophin). *Science of the Total Environment*. 747, 141295.
- [3] Mangeret, A., Blanchart, P., Alcalde, G., Amet, X., Cazala, C., Gallerand, M.-O. (2018) An evidence of chemically and physically mediated migration of ²³⁸U and its daughter isotopes in the vicinity of a former uranium mine. *Journal of Environmental Radioactivity*. 195, 67-71.
- [4] Schöner, A., Noubactep, C., Büchel, G., Sauter, M. (2009) Geochemistry of natural wetlands in former uranium milling sites (eastern Germany) and implications for uranium retention. *Chemie der Erde*. 52, 91-107.
- [5] Stetten, L., Blanchart, P., Mangeret, A., Lefebvre, P., Le Pape, P., Brest, J., Merrot, P., Julien, A., Proux, O., Webb, S. M., Bargar, J. R., Cazala, C., Morin, G. (2019) Redox fluctuations and organic complexation govern uranium distribution from U(IV)-phosphate minerals in a mining-polluted wetland soil, Brittany, France. *Environmental Science and Technology*. 52, 13099-13109.
- [6] Mangeret, A., Reyss, J.-L., Seder-Colomina, M., Stetten, L., Morin, G., Thouvenot, A., Souhaut, M., van Beek, P. (2020) Early diagenesis of radium 226 and radium 228 in lacustrine sediments influenced by former mining sites. *Journal of Environmental Radioactivity*. 222, 106324.

SKILL REQUIREMENTS :

Geochemistry, characterization of solid phases, analytical chemistry, ICP-MS

REQUIRED FORMATION :

Ph. D. in geochemistry