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An evidence of chemically and physically mediated migration of ^{238}U and its daughter isotopes in the vicinity of a former uranium mine

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ABSTRACT

The present study reports the evidence of a radioactive contamination in a wetland located downstream from a former French U mine in Brittany. This situation is demonstrated according to the measurements of gamma dose rates and activity ratios of ^{238}U and ^{232}Th-decay series nuclides, which give the justification regarding the accumulation of significant amounts of ^{238}U, ^{230}Th and ^{226}Ra in this wetland.

The dose rate map highlights an increase of radiation level along the former mine water pathway compared to the background value, with a maximum value of 1500 nSv.h^{-1} reached in the wetland. Activities of ^{238}U, ^{230}Th and ^{226}Ra and ^{232}Th/^{238}U ratios measured in surface wetland soils are significantly higher than the geochemical background. ^{230}Th/^{238}U ratios less than unity suggest a preferential accumulation of U in the wetland, compared to its daughter isotopes. Moreover, the loss of ^{226}Ra compared to ^{230}Th raises its higher mobility compared to its parent isotope. In far-field sediments, ^{226}Ra/^{238}U ratio of 1.76 implies a different geochemical behavior of U, which could be explained by the occurrence of mobile U species. The results suggest that contamination of wetland soils and far-field sediments could result from discharges of underground mine waters.

1. Introduction

In France, roughly 240 sites were operated for uranium extraction activities, which generated the production of 76,000 tons of uranium metal from 1948 to 2001 (MIMAUSA, 2018 version).[1] These activities led to the production of a large quantity of mining waste, including approximately 52 Million tons (Mt) of mill tailings and 200 Mt of waste rock.

In the vicinity of former uranium mining sites, natural systems such as soils (Štrok and Smodiš, 2010), sediments (Morin et al., 2016; Stetten et al., 2018) and especially wetlands (Cuvier et al., 2015; Schöner et al., 2009; Wang et al., 2013) are known to sequester significant levels of uranium. As a result, uranium concentration in wetlands is generally significantly higher than the geochemical background. In some cases, natural and constructed wetlands are used for water purification including U removal from anthropogenic sources (Li et al., 2014, 2015).

Several studies refer to wetlands contaminated by uranium processing facilities releases and uranium mines (Cuvier et al., 2015; Kaplan et al., 2017). Most of these studies focus specifically on uranium accumulation processes. Conversely, only a few of them deal with the behavior of its daughter isotopes. Indeed, accumulation of ^{238}U-series nuclides in soils and sediments may result from deposition of ore particles and/or precipitation and sorption of dissolved radionuclides.

The present study focuses on a wetland located downstream from the former uranium mine of Ty-Gallen in Brittany (France) and impacted by former discharges of underground uranium mine waters 30 years ago. The purpose of this work is to identify the surface areas still impacted by deposition of radionuclide-enriched materials and to explore mechanisms of radionuclide migration through the analysis of the different disequilibrium states in the ^{238}U-series decay chain.

2. Materials and methods

2.1. Geology and history of the Ty-Gallen uranium mine

The underground mine of Ty-Gallen (Brittany, France) is located in the granitic Massif of Pontivy containing fine to medium-sized leucogranites interspersed by a large number of aplitic and pegmatitic veins (AREVA, 2014). The mineralization occurs as lenses which have grown up along a fracture network characterized by a N 40°W orientation and

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[1] MIMAUSA: Mémoire et Impact des Mines d'urAniUm; Synthèse et Archives. 2018 version. IRSN.

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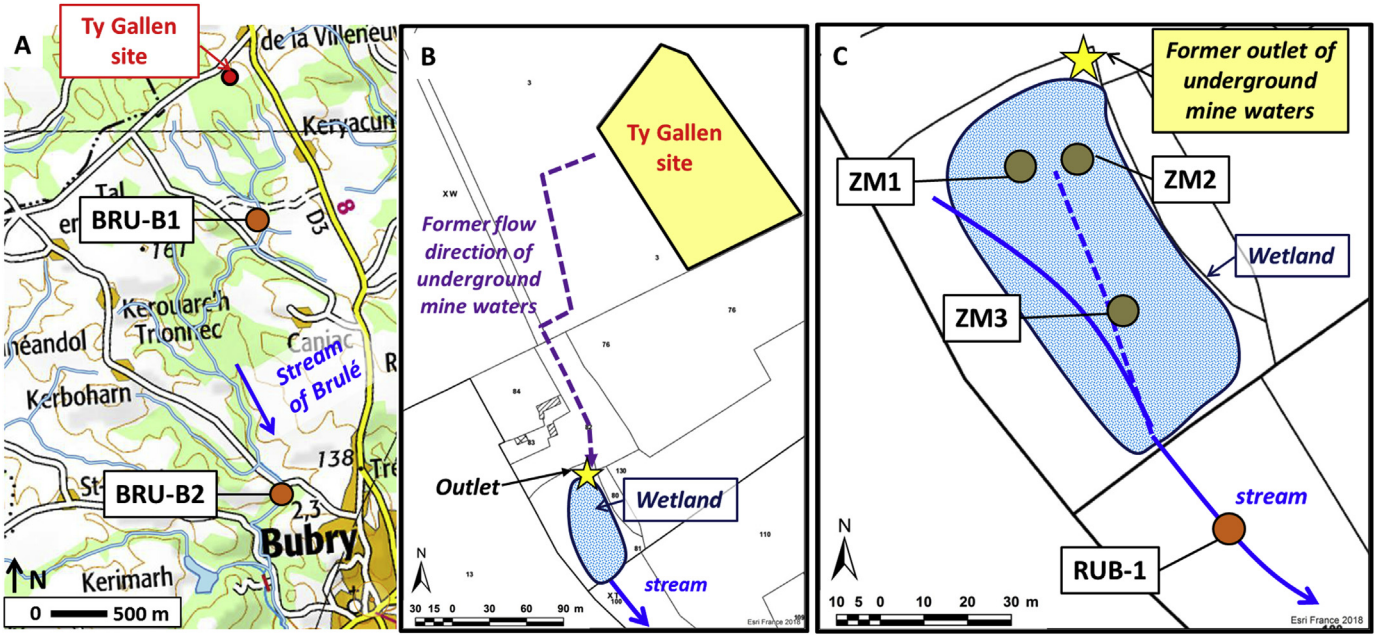

Fig. 1. Mapping of the wetland, the former mining site of Ty-Gallen and the watershed of the Brûlé stream (modified from the database Cartage®, supplied by the French National Institute of Geography, IGN, France).

a 50° W dip. The most significant lense is 60 m long and a width ranging from 1.5 to 8 m as a function of depth. The extraction of the Ty-Gallen deposits led to the production of 49 tons of U from 1963 to 1981 (AREVA, 2014). During the former mining operations, underground mine waters were pumped via a technical borehole and directly discharged and channeled up to a wetland located in the hamlet of Guern en Tal. This wetland seasonally supplies the "Brulé" stream (Fig. 1).

2.2. Gamma dose rate characterization

In order to identify radionuclide-enriched materials, gamma dose rate measurements were performed at 50 cm above the soil surface in the vicinity of the mine. A Saphymo 6150 ADB probe was used with a Saphymo 6150 AD5 radiation meter. The radiation range of the probe was chosen between 5 and 100,000 nSv per hour, with a scanning energy from 23 keV to 7 MeV.

In addition the MARCASSIN system (self standing vehicle for performing mapping of radiological activities in contaminated sites) developed by IRSN was used to establish a gamma dose rate map. The system is similar to the previously described one but operates with a GPS. MARCASSIN was used from the mining place to the wetland outlet and especially the wetland.

2.3. Soil/sediment sampling procedures

The two uppermost centimeters of wetland soils were sampled on the base of gamma dose rate measurements in order to collect sample with high radiological content (ZM1 to ZM3, Fig. 1). In addition three stream sediment samples were collected (RUB_1, BRU_B1 and BRU_B2). The first one is located 25 m downstream from the wetland on "stream 2", the second and the third respectively at 0.5 and 3.5 km downstream of the wetland on the "ruisseau du Brulé" (Fig. 1). Samples were collected during Autumn 2015. Soils and sediments were sampled with plastic shovels and immediately placed in plastic bags. They were stored at 4 °C until the return to the IRSN laboratory.

2.4. Gamma spectrometry

In the laboratory, samples were dried in a heat chamber at 105 °C and sieved at 2 mm to remove gravels and twigs. The < 2 mm fraction was sealed in air-tight 500 ml plastic boxes and put away for three weeks to ensure ^{220}Rn and ^{222}Rn and their short half live daughters ingrowth to the equilibrium with their respective parents ^{228}Ra and ^{226}Ra. Radiometric analyses were performed on 38–56 g of samples for a 24 h gamma counting. A HP Ge detector (EGPC 20, Intertech) was used for gamma spectrometry measurements. The efficiencies and backgrounds were periodically controlled with certified IRSN standards such as the 131SL300PM lake sediment sample and the RGU-1 IAEA standard. The activity measurements were given with a two sigma statistical error.

^{238}U activity was determined on ^{234}Th at 63.3 and 92.4 keV. ^{226}Ra activities were determined on ^{214}Pb (295.2 and 351.9 keV) and ^{214}Bi (609.3 and 1764.5 keV). For ^{228}Ra activities, ^{228}Ac at 338.3, 911.2 and 968.9 keV, ^{212}Pb at 238.6 keV and ^{208}Tl at 583.2 keV were used. Finally, ^{230}Th was measured for uranium-enriched soils (ZM1, ZM2, ZM3 and RUB1 soil samples) at 67.7 keV. Background noise was estimated to be 1.826 counts per second from 40 to 3000 keV.

3. Results and discussion

3.1. Gamma dose rate mapping

Gamma dose rates ranged from the geological background level (~150 nSv.h^{-1}) to 1500 nSv.h^{-1}. (Fig. 2). The highest values are encountered along the ditch and within the wetlands. Intermediate dose rates (700–900 nSv.h^{-1}) were measured downstream from the wetland along the "stream 2" and "ruisseau du Brulé" banks.

The spatial distribution of gamma dose rate indicates that the radioactive contamination is located on the former mine water discharge pathway. It gives an evidence of a long term (more than 30 years) contamination of the environment. These data also demonstrate the accumulation of gamma-emitting radionuclides in the wetland located 2 km downstream from the site of Ty-Gallen.

Gamma dose rates measurements performed in the vicinity of the wetland soil points (ZM1, ZM2 and ZM3) range from 1 to 2.1 μSv.h^{-1}., which support the presence of the radioactive contamination of the wetland. In contrast, no data are available for the RUB-1 sample and the far-field sediments BRU-B1 and BRU-B2.