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Uranium decay daughters from isolated mines: Accumulation and sources

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ABSTRACT

This study combines in situ gamma spectrometry performed at different scales, in order to accurately locate the contamination pools, to identify the concerned radionuclides and to determine the distribution of the contaminants from soil to bearing phase scale. The potential mobility of several radionuclides is also evaluated using sequential extraction. Using this procedure, an accumulation area located downstream of a former French uranium mine and concentrating a significant fraction of radioactivity is highlighted. We report disequilibria in the U-decay chains, which are likely related to the processes implemented on the mining area. Coupling of mineralogical analyzes with sequential extraction allow us to highlight the presence of barium sulfate, which may be the carrier of the Ra-226 activities found in the residual phase ($Ba(Ra)SO_4$). In contrast, uranium is essentially in the reducible fraction and potentially trapped in clay-iron coatings located on the surface of minerals.

1. Introduction

An abundant literature is available concerning the dispersion in the environment of natural radionuclides associated with uranium mining and milling activities (Neame et al. 1982; Waite et al. 1988; Mosinets, 1991; Bunzl et al. 1994; Jurado Vargas et al. 1997, McConnell et al. 1998; Fernandes et al. 1998; Fernandes and Franklin, 2001; Fernandes et al. 2006; Lozano, 2000, 2002; Vera Tomé et al. 2002; Carvalho et al. 2007, 2014; Frostick et al. 2008, 2011; Kipp et al. 2009; Momčilović et al. 2013; Blanco Rodríguez et al. 2014; Kayzar et al. 2014). However, only few published studies offer maps of radionuclide distribution (Dragović et al. 2014; Erenturk et al. 2014), in particular for uranium mining impacted environments (Vandenhove et al. 2006; Bollhöfer et al. 2006, 2014). Moreover, the considered areas usually vary from 10 to 100 km^2 and do not allow us to determine local accumulation areas. Clearly, there is a lack of published and accessible natural radionuclide distribution maps in the small-scale environment surrounding uranium mining sites. Especially maps that can help to consider the extent and the level of the contamination, in particular for the rehabilitation of former minor mining sites either abandoned or simply isolated.

The occurrence of soils and sediments enriched in uranium and decay products – often located downstream from former mines –

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may induce a local increase of the dose rate and of the external exposure to local inhabitants. The remobilization of accumulated radionuclides cannot be discarded, a process that poses a risk for biota. Thus there is a need for a precise radiological characterization of uranium and decay product accumulations. A technique widely used to understand the distribution of trace elements in the different solid phases is sequential selective chemical extraction. With that in mind, the revised standardized 3-step procedure of Rauret et al. (1999), developed within the framework of the Standards, Measurements and Testing Program (SMTP), offers many benefits, since it is easy to use, rapid, and frequently applied in the case of environmental contamination due to uranium mining and/or processing (Dhoum and Evans, 1998; Martin et al. 1998; Howe et al. 2002; Meca et al. 2012; Smodiš et al. 2012; Štrok and Smodiš, 2013).

Consequently, we propose a methodology combining gamma spectrometer at different scales in order to better constrain the multi-scale distribution and the potential mobility of radionuclides, around uranium mines. First, an initial overview of the studied environment using a coupling of mobile and fixed in situ gamma spectrometers was conducted in order to determine in real time the nature and the location of the radionuclide accumulation areas. Then, activity data were interpolated using spatial modeling software to obtain activity distribution maps. We used these maps to build the best sampling strategy. These in situ activities were then validated by collected discrete field samples and analyzing them in the laboratory. On a smaller scale, the geochemical behavior of the natural radionuclides is also investigated using geochemical and radioactive measurements coupled with sequential extraction and mineralogical analyses.

2. Methods

2.1. Study site

2.1.1. Climate regime

The Bertholène area is characterized by a warm temperate climate with mean annual rainfall of 732 mm recorded from 1981 to 2010 by the Millau weather station (data from Météo-France 2015). The average annual temperature is 10.5 °C. Winds directions are mainly W and S–W, even if N-direction is mainly encountered in winter.

2.1.2. Description of the site and of the U-ore

The Bertholène mine is located in the Palanges forest, at 1.2 km from the Bertholène village, within the Palanges orthogneissic massif. This mine is one of the nine French uranium mines with tailings disposal and effluent treatment plant and selected due to the availability of previous environmental data and the low tonnage of uranium extracted (<1000 tons) (http://mimausabdd.irsn.fr/#). Uranium ore and associated weathering were described by Schmitt et al. (1987) and Levêque (1990), but the ore is mainly constituted by vanadates in the oxidized area and coffinite in the reduced area. The Balaures catchment covers 2.6 km^2 and Balaures stream flows in a north/south direction to the confluence with the Aveyron River located 2 km from the mine, but the part flowing through the mine was re-directed and canalized. The flow rate recorded at the mine exit is 0.04 $m^3\ s^{-1}$.

2.1.3. Description of the mine operation

The exploitation of the Bertholène uranium ore was performed between 1981 and 1995 as underground (1981–1992) and open pit (1983–1995) mines, leading to the production of 744 tons of uranium and 470 000 tons of tailings. The site was equipped with an in-situ uranium ore pre-processing unit, as described by Humbert (1986). Briefly ores were sorted depend on their grade and their origin (underground or open-pit), and crushed to reach a 0–8 mm grain size. Ground ore was then placed as piles in process tanks and successively leached with various reactants, as sulfuric acid and sodium chlorate, in order to extract uranium which then immobilized on ion-exchange resins. The resins were driven to a local processing plant. At the mine closure, between 1995 and 2001, muds from settlement of effluent waters were collected and transported to a local plant for processing. Nowadays, the muds are stocked in tanks located on the waste rock pile of the site. Since 1985, effluent waters from mines, tailings dams and barren materials were collected in tailing ponds and neutralized with lime, flocculating agents and occasionally sodium hydroxide, in order to precipitate uranium. The supernatant water was transferred in another tank for clarification and densification, then pH verification before being released in the Balaures stream. At the mine closure, during the mud recovery phase, barium chloride ($BaCl_2$) was added to the treatment in order to immobilize radium. Presently, same treatments, without $BaCl_2$ are performed on effluent waters from the open pit and underground mines, the waste rock pile and the tailing storage area, before being released back in the stream. A monitoring of the releases of U-238 and Ra-226 in the downstream part of the Balaures has been implemented by AREVA NC, since the closure of the mine. Over the 2000–2009 periods, the activity concentrations of the downstream waters ranged from 1.97 to 5.57 Bq l^{-1} for U-238 and 0.05–0.10 Bq l^{-1} for Ra-226 in the soluble fraction. The activity concentration of the insoluble fraction is less than 0.02 Bq l^{-1} for Ra-226 (AREVA NC, Cadoret, 2010). To our knowledge, the determination of the activity ratios of the different kinds of solid wastes produced at the Bertholène mine was never performed but the Ra-226/U-238 activity ratio of the drainages from the Bertholène wastes ranged from 0.007 to 0.08, depending on the source, during the year 2013 (AREVA, 2013 annual report).

2.2. Mapping: kilometer to meter scale

2.2.1. Mobile in situ gamma spectrometry

A first view of the environment was done using the self-supporting tool for mapping activity level of contaminated sites (MARCASSIN) of the French *Institut de radioprotection et de sûreté nucléaire* (IRSN) (Lemercier, 2007; Panza and Guarrarian, 2009, Panza, 2012; Al-Masri and Doubal, 2013) (Fig. 1), i.e. a quad equipped with a high-purity germanium (HPGe) P-type semiconductor spectrometer of coaxial construction and of relative efficiency equal to 22%. A ZnS treated plastic scintillator detector of 3 × 3 inch was also associated with a photomultiplier. The scanned energies ranged from 23 keV to 7 MeV allowing the measurement of equivalent dose rates H*(10). The operating speed was 10 km h^{-1} and every 2 s, nuclear measurements were stored and associated with the GPS coordinates. The calibration was performed from equations of ICRU 53 (Bethesda, 1994) and allowed calculation of the transfer coefficient between the count rate and the soil activity. Calculations of the Minimum Detectable Activity (MDA) were performed from GTN 5 (Millies-Lacroix, 1994) and given in Table 1.

Using the appropriate radiation energy, it was possible to select in the spectrum only gamma rays from particular radionuclides (Table 1). The combination of the measurements of soil emission rate (or activity) and their distributions allowed creating accumulation radioactivity maps.

Subsequently, data were reprocessed in order to improve the estimation of the count rate by summing all spectra measured in a circle of 5 m around the given measurement point. The number of combined spectra was a function of the measurement density and ranged from 5 (uncontaminated and low-contaminated areas) to