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Field analyses of ^{238}U and ^{226}Ra in two uranium mill tailings piles from Niger using portable HPGe detector

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ABSTRACT

The radioactivities of ^{238}U and ^{226}Ra in mill tailings from the U mines of COMINAK and SOMAÏR in Niger were measured and quantified using a portable High-Purity Germanium (HPGe) detector. The ^{238}U and ^{226}Ra activities were measured under field conditions on drilling cores with 600s measurements and without any sample preparation. Field results were compared with those obtained by Inductive Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) and emanometry techniques. This comparison indicates that gamma-ray absorption by such geological samples does not cause significant deviations. This work shows the feasibility of using portable HPGe detector in the field as a preliminary method to observe variations of radionuclides concentration with the aim of identifying samples of interest. The HPGe is particularly useful for samples with strong secular disequilibrium such as mill tailings.

1. Introduction

Uranium (U) mining and milling are the first steps of the nuclear fuel cycle and result in the accumulation of various Technology Enhanced Naturally Occuring Radioactive Materials (TENORM). Mill tailings are the solid mineral powder generated after U beneficiation from the ore. Most of U deposits have low ore grades, resulting in the accumulation of large amounts of tailings all over the world (Abdelouas, 2006). The worldwide volume of U mill tailings was estimated to be more than 900×10^6 m^3, stored on more than 180 piles in more than 30 countries (IAEA, 2004). After ore processing, a small proportion of U, as well as all other radioactive members of the ^{238}U decay series (such as ^{226}Ra), remain in the tailings. Proper storage and long-term stabilization of U mill tailings is required to limit the release of radionuclides (Martin et al., 2003; Carvalho et al., 2005), the contamination by heavy metals inherited from the ore (Essilfie-Dughan et al., 2012; Shaw et al., 2011) as well as acid mine drainage in the case of acidic ore processing.

In the laboratory, several techniques can be used to measure U and ^{226}Ra concentration in Naturally Occuring Radioactive Materials (NORM), TENORM and other environmental samples. Uranium measurements and quantification are often performed by destructive methods, such as Inductive Coupled Plasma Mass Spectrometry (ICP-MS) and Inductive Coupled Plasma Atomic Emission Spectroscopy (ICP-AES). Radiation produced along with U decay can also be used for U quantification. Alpha spectrometry is often used to quantify U isotopic composition (Boulyga et al., 2001; Sill, 1977). High Resolution Gamma Spectrometers and absolute calibration tools for High-Purity Germanium (HPGe) detectors are also used for non-destructive quantification of gamma-emitting radionuclides. High-purity Ge detectors have been routinely used to quantify radionuclides of the ^{238}U series in geological samples (e.g. Tzortzis et al., 2003a,b; Huy and Luyen, 2004; Oyedele et al., 2010) and contaminated soils (e.g. Papachristodoulou et al., 2003; Karangelos et al., 2004; Ocone et al., 2004). Since ^{226}Ra concentration in natural samples is usually below the limits of detection of ICP techniques, it is often quantified with scintillation or radiation

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counting techniques (e.g. emanometry after ^{222}Rn emanation, alpha and gamma spectrometers and liquid scintillation).

In the field, qualitative assay of radionuclides of the ^{238}U decay series can be easily obtained with radiation counters such as Geiger–Müller tubes or pulse counting scintillators. However these instruments can lead to over/under estimations since they do not distinguish the different gamma-emitting radionuclides. This is especially the case if ^{238}U is not in secular equilibrium with its radio-emitting daughters (Bourdon et al., 2003). In the case of mill tailings, ^{238}U is in strong secular disequilibrium with its daughters (except with ^{234}Th and ^{234M}Pa that reach equilibrium about six months after U extraction) because U is chemically extracted from the ore independently from its progeny. However, recent availability of portable electrically cooled HPGe detectors has allowed on-site quantitative measurements (e.g. Pierson et al., 2013; Keyser and Hagenauer, 2008). Because of their excellent spectral resolution, HPGe detectors are particularly suited for determining the activities of individual radionuclides in mill tailings, thus not requiring the samples to be in secular equilibrium.

In this study, a portable HPGe detector was used on two ~30 m drilling cores in order to determine the vertical U and ^{226}Ra distribution in the tailings piles of the COMINAK (COmpagnie MINière d'AKouta) and SOMAÏR (SOciété des Mines de l'AÏR) U mines in Niger. Quantitative measurements of ^{238}U and ^{226}Ra using HPGe were performed in France at the AREVA laboratory of Bessines-sur-Gartempe. In order to be close to the field conditions, measurements were performed without any sample preparation and in a room with significant background gamma radiation. In order to validate the HPGe quantification method, the results obtained by HPGe were then compared with those obtained in the laboratory by ICP-AES for U and by emanometry for ^{226}Ra.

2. Background

2.1. Site description

The mines of COMINAK and SOMAÏR are part of the Tim Mersoï basin, located near Akokan and Arlit in Niger, about 200 km north of Agadez and west of the Aïr Mountains (Fig. 1). The underground mine of COMINAK opened in 1978. The deposit occurs in lower Carboniferous *Guezouman* sandstones, with an ore grade of about 4300 ppm U and a tonnage of 44 000 tons U. The open-pit mine of SOMAÏR opened in 1971. The deposit occurs in higher Carboniferous *Tarat* sandstones with an ore grade of about 2000–3000 ppmU and a tonnage of ~50 000 tons U. In both deposits, U-bearing phases mainly consist of micro-phases of uraninite containing variable amount of Si (Forbes, 1989; Pagel et al., 2005; Cavellec, 2006).

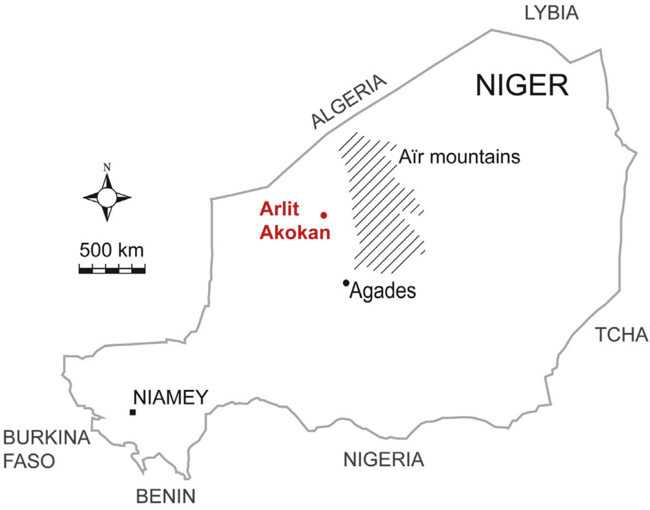

Fig. 1. Map of Niger. The mines of COMINAK and SOMAÏR are located near the towns of Akokan and Arlit, respectively.

2.2. Milling process

In both mines, the ore is crushed and oxidized in order to liberate micro-phases of U associated with the gangue and to solubilize U. The leaching operation is performed simultaneously with sulfuric acid and minor nitric acid. After flocculation, the solid fraction (i.e. the mill tailings) is washed and the U-rich pulp is filtered. The process yield is about 90–96%, thus some residual U remains in the tailings after this step. The dissolved U is then stripped of the pregnant liquor using liquid solvents. U is finally precipitated as MgU_2O_7 (s) at COMINAK and as $Na_2U_2O_7$(s) at SOMAÏR (types of yellow cake) and stored in drums. Further details on the process are available in the technical report IAEA (1993).

2.3. Tailings management

In both mines, fresh tailings are carried by a conveyor belt from the mill to the top of the pile. Fresh tailings are watered with mine water to increase their flowability down the pile slope. At present, the tailings piles are about 30 m high and 500 × 1100 m wide (more than 16 million tons) at COMINAK and about 40–45 m high and 700 × 1000 m wide (more than 17 million tons) at SOMAÏR. Both tailings piles lay on a clay layer and a hardened crust is formed at the surface of the pile within the first meter, after evaporation of the water contained in fresh tailings.

3. Materials and methods

3.1. Field sample collection

Two ~30 m cores were drilled in the tailings pile of COMINAK and SOMAÏR, from the surface to the underlying clay layer. The cores were drilled with a triple tube MAZIER® sampler in order to collect undisturbed samples. The drilling cores were retrieved by 1.5 m subsections (or less, depending on the drilling recovery rate) and stored in transparent PVC tubing (1 mm thickness and 88 mm inner diameter) with taped end caps.

3.2. ^{238}U and ^{226}Ra analyses by HPGe

Measurements were performed with a portable Canberra Falcon 5000® HRGS using an electrically cooled (104 K) broad energy HPGe detector crystal with a relative efficiency of the order of 20%. The unit also integrates a signal pre-amplifier, an analog-to-digital converter, a multi-channel analyzer and a high voltage power supply. The instrument was operated with an external power supply, but it can also be operated in the field in a stand-alone mode for up to 3 h using a dual hot swappable Li-ion battery system. The instrument was previously energy calibrated with known radioactive sources at AREVA-Canberra in Montigny (France). The HPGe detector was placed directly on the drilling core subsections, without any sample preparation (Fig. 2). In order to limit the influence of the background radiation, a lead shielding device was designed to isolate both the head of the detector and the core sample. The lead shield assembly was designed to allow for a 100 × 100 mm cavity for core sample fitting. The shield dimensions were 170 × 200 × 350 mm and the bricks were 50 mm thick (over 100 kg weight). The head of the detector was positioned against the lead shielding walls, at ~2 cm from the core samples. The detector