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Source identification of uranium-containing materials at mine legacy sites in Portugal

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ABSTRACT

Whilst prior nuclear forensic studies have focused on identifying signatures to distinguish between different uranium deposit types, this paper focuses on providing a scientific basis for source identification of materials from different uranium mine sites within a single region, which can then be potentially used within nuclear forensics. A number of different tools, including gamma spectrometry, alpha spectrometry, mineralogy and major and minor elemental analysis, have been utilised to determine the provenance of uranium mineral samples collected at eight mine sites, located within three different uranium provinces, in Portugal. A radiation survey was initially conducted by foot and/or unmanned aerial vehicle at each site to assist sample collection. The results from each mine site were then compared to determine if individual mine sites could be distinguished based on characteristic elemental and isotopic signatures. Gamma and alpha spectrometry were used to differentiate between samples from different sites and also give an indication of past milling and mining activities. Ore samples from the different mine sites were found to be very similar in terms of gangue and uranium mineralogy. However, rarer minerals or specific impurity elements, such as calcium and copper, did permit some separation of the sites examined. In addition, classification rates using linear discriminant analysis were comparable to those in the literature.

1. Introduction

Nuclear forensics has been a growing scientific field since the early 1990s. Between 1995 and the end of 2015, the International Atomic Energy Agency (IAEA) logged a total of 2889 incidents of nuclear materials outside of regulatory control on the Incident and Trafficking Database (ITDB) (IAEA, 2015). As a result, the IAEA has been working with the Nuclear Forensics International Technical Working Group (ITWG) to characterise such materials and determine their provenance, with a number of institutions analysing materials of known origin to produce representative sample databases (Kristo and Tumey, 2013). The aim of such work is to ensure that any obtained illicit materials can be analysed and the results directly compared to those within the databases to deduce a potential match and, hence, provenance. Since the system inception, and after the analysis of a considerable range of samples from around the world, a number of studies have successfully deduced the provenance of nuclear materials based on characteristic signatures, such as elemental impurities, rare earth element (REE) patterns and Pb, Sr, S, Nd, O and U isotopic ratios (Brennecka et al., 2010; Han et al., 2013; Keegan et al., 2008, 2012; Krajkó et al., 2014; Mayer et al., 2005, 2007; Švedkauskaitė-LeGore et al., 2008; Varga et al., 2009, 2010a, 2010b; Wallenius et al., 2006). However, the focus of those studies has been typically on distinguishing samples from geochemically-different types of uranium deposit, such as those controlled by low temperature redox sensitive processes (sandstone), high temperature redox sensitive processes (unconformity-related) and non-redox processes (placer) (Badaut et al., 2009; Varga et al., 2010a,b).

Unlike these earlier works, the objective of this study has been to provide a scientific basis for source identification of materials from uranium mine sites within three different mineralisation districts, which together form part of the same former mining region. The area sampled was the Beiras region of central Portugal, where hydrothermal mineral veins within the Hercynian Granites (which intruded ~290 million years ago) were economically mined for uranium throughout the last century (Carvalho et al., 2007, 2014; Pereira et al., 2014). The Serra de Estrela granite mountain range dominates the region, with the

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Table 1
Details of mine sites (1–8) sampled in the Beiras region (Portugal), including the number of samples collected at each site and the total number of EDS point analyses for all samples.

Mine Site	Location (Lat., Long.)	Mine Name	Locality	Province	No. of Samples	No. of EDS Analyses
1	40.5177, −7.2925	Forte Velho	City of Guarda	Guarda	5	100
2	40.8334, −7.4204	Reboleiro	Village of Reboleiro	Reboleiro	4	79
3	40.8698, −7.3849	Fonte Velha	Sebadelhe da Serra	Reboleiro	2	15
4	40.8282, −7.4248	Lenteiros	Village of Reboleiro	Reboleiro	4	74
5	40.8944, −7.2550	Murtórios	Marialva	Reboleiro	2	10
6	40.8197, −7.5571	Maria Dónis	Aguiar da Beira	Reboleiro	5	79
7	40.5204, −7.6398	Ribeira do Boco	Gouveia	Urgeiriça	1	14
8	40.5520, −7.5557	Castelejo	Gouveia	Urgeiriça	4	69

extensive mineral veins existing within the granite body to the north of the mountain range in a horse-shoe shape. There are four different uranium provinces where the concentration of these veins is significant (Cameron, 1982a). The three provinces sampled as part of this study were: the Guarda province to east; the Reboleiro province to the north; and the Urgeiriça province to the west (Cameron, 1982b). Samples were collected from spoil heaps at eight different disused mine sites across these three provinces, and the locations of each sample are included within the subsequent methods section.

Large-scale fracturing of this granite as a result of extensive tectonism allowed infiltration of low temperature hydrothermal fluids, capable of leaching, to ascend and subsequently dissolve minerals at the edges of these fractures. These low-temperature fluids then became saturated in various elements as the edges of the granite body became sericitised. Over time, these transported elements were re-precipitated alongside the predominant gangue mineral quartz, within the widespread network of fractures emanating from the central region (Cameron, 1982b).

The aim of the study is to provide a scientific basis for source identification of materials from uranium mining sites within the same region that can potentially be used within nuclear forensics. This study has particularly focussed on differences in mineralogy and elemental impurities between samples from different sites. A number of samples were additionally analysed by gamma and alpha spectrometry, after a cumulative gamma spectrum was taken by an unmanned aerial vehicle (UAV) to deduce the sampling locations.

The ability to attribute any seized material to an exact source mine rather than a broad geographical area may help resolve issues, such as illegal dumping of radioactive waste, origin of materials abusively taken for use in landfills or construction and illicit traffic of uranium ore.

2. Materials and methods

2.1. Airborne radiation mapping system

The UAV, radiation mapping system and associated post-analysis software used in this work were developed in-house at the University of Bristol (UK). Previous applications of this technique include the characterisation of land contaminated from uranium mining in Cornwall (UK) and fallout from the disaster at Fukushima Daiichi Nuclear Power Plant in 2011 (Martin et al., 2016, 2015). Details regarding the UAV may also be found in these publications.

During mapping flights, a consistent ground equivalent speed of between 1.5 m s^{-1} and 2.0 m s^{-1} was employed, at an altitude of between 5 m and 10 m above the ground, depending on topography and obstructions. This speed at which the survey was undertaken, through the calibration discussed subsequently, was selected to ensure the calibrated collection of dose-rate for the system's sampling rate (2 Hz). A grid-spacing of 2 m was used to ensure adequate ground coverage. This setup produced an on-ground resolution of approximately 1 m radius. Control of the system was performed autonomously using the UAV's autopilot platform (ArduPilot Mega APM 2.8) via a series of predetermined waypoints, with both take-off and landing performed manually using remote controls.

A map of radiation intensity was constructed, consisting of a number of scaled circles overlain onto a satellite base-map. At locations where multiple data-points overlapped producing constructive interference, an average of the two measurements was used for plotting. A conversion between the counts per second (cps) values obtained from the detector and a dose-rate (μSv.hr^{-1}) from a number of various isotopic sources was conducted, as described in (Martin et al., 2017). Further inter-calibrations with the aerially-measured results were performed using a calibrated handheld dosimeter from RADEX™ (Moscow, Russia) carried on the ground during the work.

To provide additional ground coverage where it was not possible to obtain aerial data due to extreme topography, vegetation or overhead obstructions, small regions were surveyed on foot, with the detector swept by hand in uniform patterns. These areas were limited to reduce the overall impact that their results may have on those from the airborne survey. Where these measurements were conducted, the detection system was placed in the operator backpack at a height corresponding to 1 m above the ground (as per the normalisation conducted for altitude by the rangefinder on the UAV).

2.2. Sample collection

At each site, following the airborne radiation survey, ore samples were collected from areas exhibiting elevated activity relative to the surrounding area. By subsequently manually scanning the metre-scale area of elevated activity using a hand-held Geiger counter, rock fragments consisting of uranium ore were identified amongst the spoil and collected. At each sampling location, a photograph was taken and the GPS location recorded. Samples were then double-bagged and labelled. The location of each mine site is outlined in Table 1 as well as depicted on a map of the area in Fig. 1. The number of samples collected at each site and total number of EDS point analyses is also listed in Table 1. Although a large amount of time was spent identifying areas of elevated activity at some sites, only a small number of samples could be collected. In addition, some samples had limited uranium mineralisation; hence, only a small number of EDS point analyses were taken.

2.3. Gamma spectrometry

For the gamma spectrometry measurements, aliquots of large milling and mine tailing samples, were sieved to remove stones (> 2 mm diameter). The sieved samples were collected in 100 ml thick polyethylene bags and securely closed. Samples were stored for a period of > 35 days, to allow for the formation of radium–radon daughters radioactive equilibrium, before the samples were measured by gamma spectrometry. Gamma spectrometry was performed using BeGe large volume detectors (Canberra, Meriden, CT, USA) and gamma spectra analysis and computations were made using Genie2000™ software (Canberra, Meriden, CT, USA). Quality control was ensured by regular