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DISTRIBUTION OF NATURALLY OCCURRING RADIONUCLIDES (K, TH AND U) IN WEATHERED ROCKS OF VARIOUS LITHOLOGICAL TYPES FROM THE URANIUM BEARING REGION OF FORNOS DE ALGODRES, PORTUGAL

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ABSTRACT

The distribution of K, Th and U in various types of weathered rocks (granite, schist, dolerite and aplite), belonging to the Beiras uraniferous province in the Fornos de Algodres area of Northern Portugal, was studied. Since the concentration of naturally occurring radionuclides in rocks depends much on the geological context, levels of ionizing radiation and radon emission may vary as a function of geological origins and processes. Our study aims to contribute to the knowledge of the background concentrations of these elements from rocks in a region that is generally enriched in uranium, and consequently with higher potential risk. These are used to estimate dose rates to which people might be exposed at the land surface, as well as dose rates within each context, relevant to luminescence dating, and identify sources of enhanced radon emission.

The concentrations of natural radionuclides were obtained using two methods for comparison, the field gamma spectrometry (FGS) and instrumental neutron activation analysis (INAA), as their combination may help detect spatial variations and disequilibrium in the U series. The mineralogical study of rock types was performed by X-ray diffraction of powder samples.

INAA and FGS results were commonly similar. The K, Th and U contents in the studied area are variable, but usually are higher than average upper continental crust values. A granitic weathering profile was encountered in which significant leaching of K had occurred, producing a ca. 30% difference in dose rate. The greatest U contents were observed in a weathered aplite dyke intersecting the granite; radon escape of up to 590 Bq kg^{-1} was inferred, producing a 300% difference between dose rates based on INAA or FGS data. However, the enrichment in U is not restricted to aplite; its heterogeneous distribution may cause localized enrichment, which was detected in the other lithologies: notably, soils on moderately radioactive bedrock were enriched in Th and U by colluvial movement.

KEYWORDS: FGS, INAA, Natural radionuclides, Uraniferous province

INTRODUCTION

In general, the concentrations of natural radionuclides (K, Th and U) in rock forming minerals and soils are not a concern to the environment (Elles and Lee, 2002); however there are areas with relatively high natural concentrations of these elements due to their geological context. In such places, ionizing radiation and especially radon emission from decay of U may reach hazardous radiological levels and represent a potential risk to the environment or human health (Singh et al., 2009).

As the population is exposed mainly to terrestrial radiation of natural origin (UNSCEAR, 1988) it is important to study the background values of natural radionuclides to assess the gamma radiation dose for the population in order to know the health and ecological risks and to have a baseline for future changes in the environmental radioactivity due to human activities (Singh et al., 2009).

The present study contributes to the compilation of mineralogical, geochemical and in situ gamma-ray data as a basis to evaluate the radon levels emanated from rocks, by studying the concentration and distribution of natural radionuclides in various lithologies from the Fornos de Algodres area (Northern Central Portugal).

The studied area is located in the Beira uraniferous province, known for its U-Ra metallogenic deposits, which were exploited since the beginning of the 20th century, until recently (Carvalho et al., 2007), and so the interest of studying the background values of radionuclides in this area.

Some information on the geochemistry and mineralogy of clay-rich materials from the Fornos de Algodres region may be encountered elsewhere (Dias et al., 2000; Trindade et al., 2010; Trindade et al., 2011a).

MATERIALS

The Fornos de Algodres region, located in the Northern Iberian Hercynian massif, is mainly composed of granitic rocks that are correlated with the third deformation phase (D3) of the Variscan orogeny (Matte, 1986); most of them are late-post-kinematic plutons (315-270 Ma) and a minority are deformed syn-D3 granitoids (340-320 Ma) (Ferreira et al., 1987; Pinto et al., 1987) (Fig. 1).

The whole batholith was emplaced into metasedimentary rocks of Cambrian to Lower Ordovician age, from which only small bodies can be seen in the area.

Subvertical shearing and faulting related with late-stage activity of Variscan deformation phases originated the installation of dolerite and aplite-pegmatite veins (Azevedo and Nolan, 1998) that appear frequently intersecting the granitic rocks.

For this work we collected samples from: (i) a coarse- to medium-grained porphiroid two-mica monzonite granite, (ii) two aplite veins intersecting the granite, (iii) a complex of dolerite veins in the granite, and (iv) an outcrop of the schist-greywacke complex of Cambrian age (Fig. 1).

Twenty-nine samples of clay-rich rocks and soils of the various lithologies were sampled for analysis:

(i) four samples from a vertical profile of about 3 meters height (weathered granite, from base to top: GMX0, GMX1, GMX2; and soil: GMX3), and two more samples from the outcrop in the other side of the road (samples GMX2L and GMX1R),

(ii) five samples from an aplite dyke intruded in the granite of the previous outcrop (weathered aplite, from base to top: VGMX1, VGMX2, VGMX3; and soil: VGMX4, VGMX5), and 5 samples from another aplite dyke sited more at North (weathered aplite, from base to top: FQC0, FQC1, FQC2; and soil: S2FQC, S1FQC),

(iii) four samples from a dolerite dyke (weathered dolerite: F5SP; and soil, from base to top: S1F5, S2F5, S3F5), three samples from another dolerite dyke (weathered dolerite, from base to top: F1SP1, F1SP2; and soil: SF1) and two more samples of weathered dolerite from different dykes (F2SP and F3SP), and

(iv) four samples from a vertical profile of about 8 m height (weathered schist, from base to top: XMA1, XMA2, XMA3; and soil: SXMA).

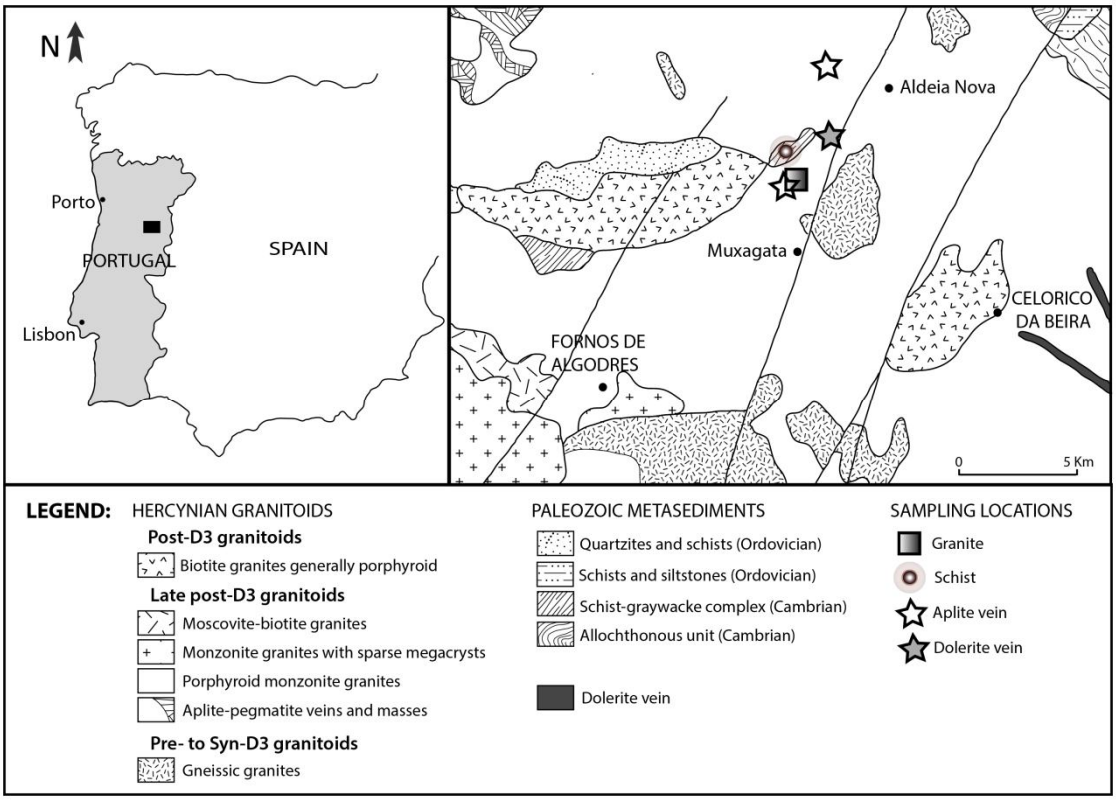

Figure 1: Geological sketch map of the Fornos de Algodres area (Northern Central Portugal). Drawing based on the digital Carta Geológica de Portugal, 1:500 000, from the geoPortal LNEG (http://geoportal.lneg.pt/geoportal/mapas/index.html).

METHODS

The total samples, except fragments with dimensions above 2 mm that were separated by dry sieving, were crushed into a fine powder using an agate mortar and then prepared for mineralogical and chemical analysis.

The mineralogical composition of the various rock types studied was obtained by X-ray diffraction (XRD), using a Philips X'Pert Pro diffractometer, with a PW 3050/6x goniometer, CuKα radiation, and fixed divergence slit, operating at 45 kV and 40 mA. Scans were run from 4º to 60º 2θ, using a step size of 0.02º 2θ and a scan step time of 2 s. The identification of crystalline phases by XRD was carried out using the International Centre for Diffraction Data Powder Diffraction Files (ICDD PDF).

The chemical data were obtained by instrumental neutron activation analysis (INAA), by irradiating 200-300 mg of powdered samples and standards (GSD-9 and GSS-1, from the Institute of Geophysical and Geochemical Prospecting. Reference concentration values from Govidaraju, 1994) in the core grid of the Portuguese Research Reactor (IST/ITN, Sacavém) for 7 hours (long irradiation) at a thermal flux of 4.4 x 10^{12} n cm^{-2} s^{-1}; $\varphi_{epi}/\varphi_{th}$=1.4%; $\varphi_{th}/\varphi_{fast}$=12.1, which allowed to obtain the concentration of the naturally occurring elements K, Th and U. Two γ-ray spectrometers were used, a 150 cm^3 coaxial Ge detector and a low energy photon detector (LEPD), both connected through Canberra 2020 amplifiers to Accuspec B (Canberra) multichannel analyzers. The relative precision and accuracy of the method are, in general, to within 5%, and occasionally within 10%. Details concerning the measurement and processing of the gamma spectra can be found elsewhere (Gouveia and Prudêncio, 2000; Dias and Prudêncio, 2007).

For selected residual clay and soil samples the apparent concentrations of parent element of K, Th and U were also measured by field gamma spectrometry (FGS). FGS measurements were conducted in situ at the points of sampling, using a 3"x3" NaI probe with a HPI Rainbow MCA. Stripped counts in the windows 1380-

1530 keV, 1690-1840 keV, and 2550-2760 keV (designed to obtain signals dominated by ^{40}K, ^{214}Bi and ^{208}Tl respectively), were calibrated relative to previous measurements in the Oxford and the Gif-sur-Yvette blocks (Richter et al., 2003), to obtain apparent parent element concentrations, assuming equilibrium in the ^{232}Th and ^{238}U series.

This combination of INAA and FGS measurements enables inferences related to dosimetric homogeneity, and parent element concentrations as compared with post radon emissions can help detect disequilibrium in the U series, including radon emission.

RESULTS AND DISCUSSION

Mineralogical composition

The mineralogical composition of the various rock types can be summarized as follows:

(i) the granite samples are mainly composed of quartz, plagioclase, K-feldspar, mica and clay minerals; the proportion of clay minerals increases to the top of the profile;

(ii) the aplite samples have similar composition to that of the granite, but generally with higher K-feldspar/plagioclase ratio. Soils over the aplite dikes are much richer in quartz;

(iii) the residual clays of dolerite have augite and Ca-plagioclase as the original mafic minerals; it also contains quartz, K-feldspar, magnetite, ankerite and clay minerals, the last consisting mainly of smectite, serpentine (clinochrysotile), talc and illite (Trindade et al., 2011b).

The soils above dolerite veins have compositional features similar to that of the granite, with quartz, K-feldspar, plagioclase, mica and clay minerals; and finally

(iv) the weathered schists consist mainly of quartz, plagioclase, K-feldspar, mica, clay minerals and may include traces of amphibole, magnetite, and hematite; the amount of plagioclase and mica decreases to top of the profile. The soil sample in top of schist profile is richer in quartz, K-feldspar and mica.

The presence of soils with granitic composition and clearly enriched in quartz and K-feldspar, even the ones sited above dolerite veins, suggests that they are not residual as a consequence of in situ weathering of parent rocks, but contain significant portions of detritic components derived from alteration of surrounding granitic rocks, which were then transported and accumulated elsewhere over different types of rocks.

K, Th and U concentrations in the various rock types

The concentrations of K, Th and U obtained by INAA and the apparent concentration of parent elements obtained for some samples by FGS for comparison are listed in Table 1.

Total infinite matrix (4π steradians) α, β, γ dose rates to etched 100 μm quartz grains as commonly used for luminescence dating (further details of calculations in Burbidge et al., 2014) ranged from 2 to 21 mGy a^{-1} based on parent K, Th and U concentrations from INAA, and from 2 to 9 mGy a^{-1} based on ^{40}K, ^{208}Tl and ^{214}Bi activities from FGS.

2π gamma dose rates at the surfaces of the soil and the exposed sections, estimated from 4π FGS measurements, ranged from 0.4 to 1.8 mGy a^{-1} (ca. 0.05 to 0.2 μSv hr^{-1}). The most radioactive locations would therefore slightly exceed ICRP minimum reference levels if exposure were permanent (ICRP, 2007).

INAA and FGS results are commonly similar, despite spatial variability evident in the INAA results. In the absence of other effects, slightly lower values from the field measurements are observed, being related to in situ water.

Occasional large differences in values for uranium are indicative of U uptake and / or radon loss. This is especially relevant for the aplite sample FQC1, where the U value obtained through field measurements is about five times lower than that measured in lab by INAA.

Given the high U levels in the other residual clays (samples FQC0 and FQC2) of the same profile, this suggests strong radon emission rather than localized enrichment, but may include a component of both. If the difference were entirely explained by radon loss it would imply the escape of ca. 590 Bq kg^{-1} from material in this vein.

The INAA and FGS results for FQC1 give dose rates for luminescence dating of 18 and 5.7 mGy a^{-1}, calculated as above assuming equili-

brium. Thus, a 300% difference in the age estimate could be obtained, depending on the assumptions used in relation to U disequilibrium and spatial homogeneity when combining the INAA and FGS results. Improved constraint of these factors based on additional FGS measurements and high resolution gamma and/or alpha spectrometry would be required for accurate dating analyses.

Low Th/U ratios were observed in other sampled sections, but ratios for INAA were similar to those for FGS, where measured. This is consistent with little radon emission and ancient enrichment, i.e. with pre-radon isotopes near equilibrium. However, the U contribution to the environmental dose rate for luminescence dating at these sites was often 50% to 80%. Dose rate evaluations for dating are therefore highly sensitive to disequilibrium, and the importance of element- and isotope- specific attenuation factors is increased.

The distribution of naturally occurring radionuclides (concentrations obtained by INAA) among the profiles studied in the various rock types is presented in Fig. 2.

Table 1: Concentrations of natural radionuclides obtained by INAA and FGS (element concentration for INAA and apparent concentration of parent element for FGS).

					INAA				FGS			
Rock type		Sample	Lab Reference	Depth (cm)	K (%)	Th (ppm)	U (ppm)	Th/U	K (%)	Th (ppm)	U (ppm)	Th/U
Granite	soil	GMX3	A10/126	15	1.39	15.8	3.07	5.1				
	res. clay	GMX2	A10/125	90	2.91	19	9.83	1.9				
	res. clay	GMX1	A10/124	200	4.17	18.8	28	0.7				
	res. clay	GMX0	A10/123	300	3.78	10	5.15	1.9				
	res. clay	GMX2L	A10/092	350	4	14.5	10.2	1.4				
	res. clay	GMX1R	A10/093	380	4.06	14	17.7	0.8	3.5±0.04	12.2±1.1	13.9±1.3	0.9
Aplite	soil	VGMX5	A10/091	15	5.35	11.6	8.31	1.4				
	res. clay	VGMX4	A10/090	40	3.95	12.1	9.09	1.3	2.8±0.03	9.0±0.8	10.0±0.9	0.9
	res. clay	VGMX3	A10/089	130	4.45	9.86	14.4	0.7				
	res. clay	VGMX2	A10/088	240	4.13	13.2	14.4	0.9				
	res. clay	VGMX1	A10/087	280	3.91	12.2	10.8	1.1	3.7±0.04	13.4±1.2	18.5±1.7	0.7
	soil	S1FQC	A10/121	15	4.97	27.1	4.46	6.1				
	soil	S2FQC	A10/120	30	7.31	30.1	4.49	6.7				
	res. clay	FQC2	A10/119	150	2.65	8.11	45	0.2				
	res. clay	FQC1	A10/115	320	3.03	7.91	57.8	0.1	2.1±0.03	7.1±0.7	12.1±1.1	0.6
	res. clay	FQC0	A10/122	330	6.53	7.51	55	0.1				
Dolerite	soil	SF1	A10/100	20	4.83	25.7	8.62	3.0	2.2±0.03	12.4±1.1	3.0±0.3	4.1
	res. clay	F1SP2	A10/098	80	1.43	4.4	2.15	2.0	1.3±0.02	5.8±0.6	1.7±0.2	3.4
	res. clay	F1SP1	A10/094	180	1.56	3.48	1.2	2.9	1.3±0.02	4.6±0.4	1.8±0.2	2.6
	soil	S3F5	A10/110	20	5.17	23.1	8.03	2.9				
	soil	S2F5	A10/109	50	1.86	9.57	14.4	0.7				
	soil	S1F5	A10/108	80	2.16	11.9	15.2	0.8				
	res. clay	F5SP	A10/107	120	1.69	3.97	33.2	0.1				
	res. clay	F2SP	A10/101	100	1.78	4.37	3.08	1.4				
	res. clay	F3SP	A10/103	140	1.5	3.8	11.1	0.3				
Schist	soil	SXMA	A10/114	20	3.14	36.9	6.64	5.6				
	res. clay	XMA3	A10/113	200	1.09	11.1	13.9	0.8				
	res. clay	XMA2	A10/112	350	2.8	9.98	4.15	2.4	1.9±0.03	8.6±0.8	3.2±0.3	2.7
	res. clay	XMA1	A10/111	500	4.06	10.6	3.39	3.1	2.4±0.03	8.4±0.8	3.3±0.3	2.5

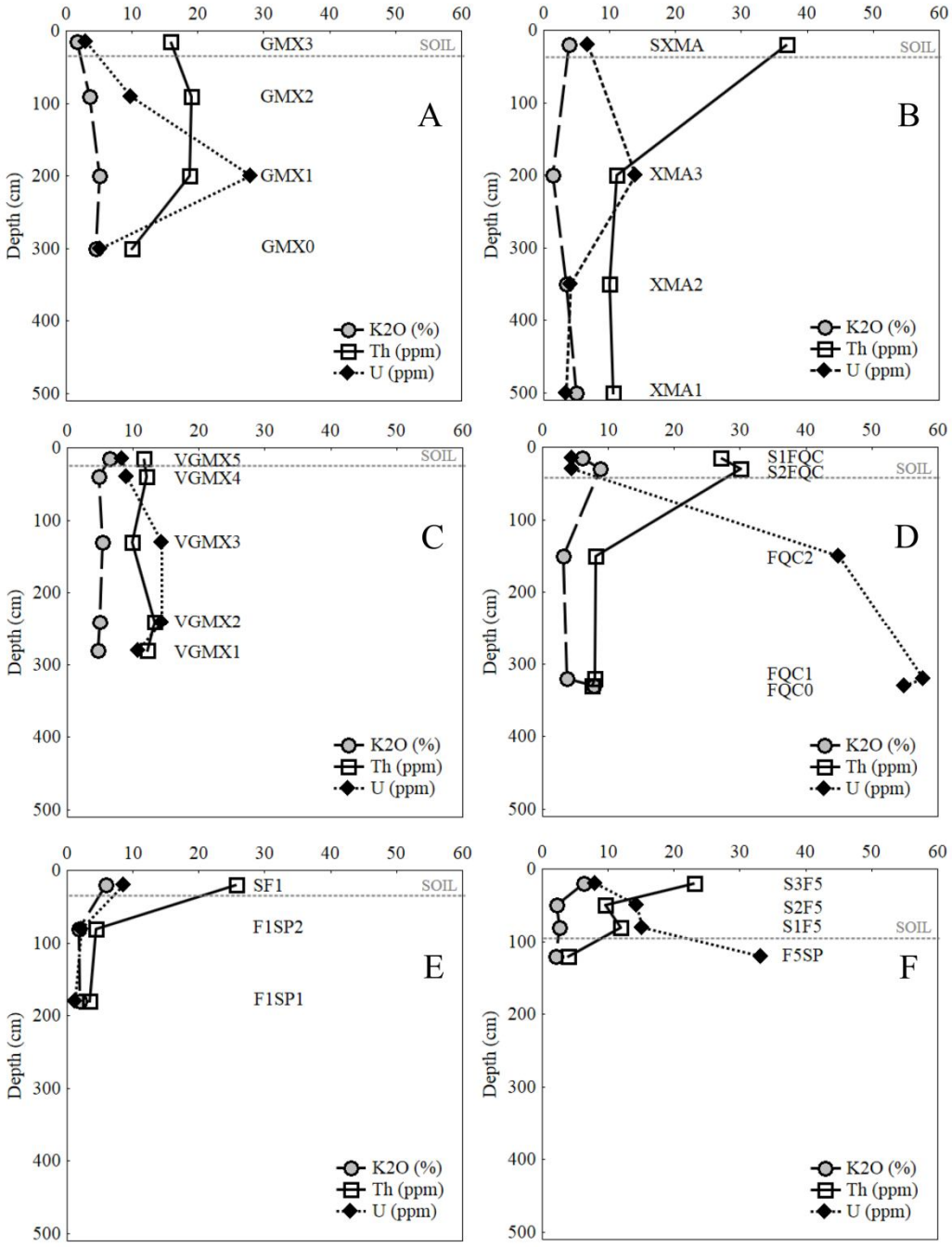

Figure 2: Distribution of the naturally occurring radionuclides (K, Th and U) in vertical profiles, including weathered rock and soil, in granite (A), schist (B), and aplite (C and D) and dolerite (E and F) veins. The average Upper Continental Crust (UCC) values are K_2O = 2.8 %; Th = 10.5 ppm and U = 2.7 ppm (Rudnick and Gao, 2003).

In general, the K contents in residual clays of felsic rocks are slightly higher than average upper continental crust (UCC) value (K_2O = 2.8 %, Rudnick and Gao, 2003), whereas in dolerite veins the K concentrations are slightly lower, as expected for mafic rocks. Some variations in the K concentration are observed across the profiles, the most relevant concern the clay-soil transition. The soil sample over the granite presents a K content ca. 1/3 that of the clay, but similar Th values, suggesting an in situ weathered granite profile where K was lost as a function of leaching during soil formation processes affecting the clay. Average K, Th and U values for clay samples GMX1, GMX0, GMX2L and GMX1R give a luminescence dose rate of 8.9 mGy a^{-1} (see previous description). Substitution of the K content of sample GMX3 reduces this by 29%, indicating

that changes in K content over time could have a significant effect on the luminescence dating of all but recent soil layers in such contexts, and should be evaluated in addition to U mobility (e.g. Zacharias et al., 2005). For all other rock types the soils are enriched in K, especially those above dolerite veins. This is consistent with mineralogical observations, indicating the soil is not residual from weathering of dolerite. The weathered granite exhibits commonly, with exception of one sample, values of Th that are considerably higher than UCC values and, inversely, the weathered dolerite has much lower Th contents, as is predictable due to the fact that Th is an incompatible element; in the remaining lithological types the Th contents are similar to that of UCC. The concentration of Th often increases abruptly (up to almost 3 times higher than UCC value) in the clay-soil transition, as occurs in the schist, in one of the aplite veins and in both dolerite veins.

Uranium shows a heterogeneous distribution, varying widely even among the residual clays of the same profile, as in the case of granite where the U contents changes more than 20 ppm. The U concentrations vary from values similar to that of UCC to values several times superior; the weathered aplite presents the higher values of U, reaching 58 ppm and being 17-21 times UCC. Lower but also considerable enrichments of U can be observed in samples of weathered granite, schist, and dolerite.

Uranium and thorium are highly incompatible elements; therefore they are more abundant in felsic rocks and sediments. U and Th can form their own phases in sedimentary rocks, uranite and thorite, but they are quite rare. Usually, in igneous and metamorphic rocks, U and Th are either dispersed as trace elements in major phases, or concentrated in accessory minerals such as zircon and monazite. Th is relatively immobile under most circumstances. In its reduced form, U^{4+} is insoluble and therefore relatively immobile, but in the U^{6+} form, which is stable under a wide range of conditions at the surface environments, U forms the soluble oxyanion complex, UO_4^{2-}; as a result, U can be relatively mobile.

The large change in U/Th ratio observed in most of the profiles clearly points to high mobility of the U under oxidizing conditions that normally prevail at the surface of the Earth. The distribution of U and sometimes K, indicates they were leached by the rain waters and transported within the vertical profiles and reconcentrated in certain weathered parts of the profile. However, the distribution of Th and K along the vertical profiles is greatly controlled by the distribution of resistates, such as zircon, and secondary stable minerals.

CONCLUSIONS

Beyond a slight enrichment in clay minerals towards the top, no significant mineralogical differences were found in the clay-rich sediments below the topsoil. The soil samples have higher amounts of quartz and feldspars, even the soil above dolerite veins, suggesting a distinct source area for detrital components of the soil.

The concentrations of radiogenic elements in weathered rocks of the studied area vary strongly, but frequently they are higher than average upper continental crust values (K = 2.8 %, Th = 10.7 ppm and U = 2.8 ppm, Rudnick and Gao, 2003): i) granite – K = 2.9-4.2 %, Th = 10-19, U = 5.2-17.7 ppm; ii) aplite – K = 2.7-6.5 %, Th = 7.5-13.2 ppm, U =9.1-57.8 ppm; iii) dolerite – K = 1.4-1.8 % K, Th = 3.5-4.4 ppm, U = 1.2-33.2 ppm; and iv) schist – K = 1.1-4.1 %, Th = 10-11.1 ppm Th, U = 3.4-13.9 ppm.

The combination of INAA and FGS is an efficient means of comparing the spatial distribution of both parent and daughter radioisotopes from NORM. The present results illustrate how this is important for the evaluation of gross spatial and temporal effects on dose rates for luminescence dating and radiation exposure. Radiation counting methods or the use of thermoluminescence dosimeters would not permit such evaluation, and large numbers of laboratory spectrometric analyses would be required to achieve the same coverage, though the present results would be enhanced by the addition of high resolution gamma or alpha spectrometric analyses in parallel with INAA.

In general, the clay-rich veins have higher U contents than the soils, and frequently are considerably enriched in U. INAA and FGS results were commonly similar, but lower post-radon U

values from FGS were considered to indicate spatial variability or radon loss and could cause systematic errors of up to 300% in luminescence dose rate.

The highest concentrations of U were found in one aplite vein, 20 times UCC, but possibly losing sufficient radon (590 Bq kg^{-1}) that any local habitation would have to be carefully planned to minimize exposure.

Surficial gamma dose rates from the clay in this location exceeded minimum reference levels despite the radon escape. However, the enrichment in U has a heterogeneous distribution, appearing in all the lithological types studied and not only in the aplite.

The soil samples of the various lithologies show higher concentrations of Th and K, except for granites (Fig. 2A), than the clay rich sediments below, which in association with the higher amount of sand fraction and mineralogical composition, suggest that most of the soils were not fully developed in situ; abundant detrital components came probably from a different weathered source area, most likely granites.

Thus, it is plausible to consider the effect of colluviation/slopewash in the development of the soils (not in situ). However, on the granite itself the soil sequence appeared in situ, and this exhibited significant temporal variability in dose rate as a function of the leaching of K.

Thus, although focusing on a region of uraniferous lithology, the present study serves to highlight the geochemical and geomorphological processes that lead to redistribution and alteration in each of the major radioelements K, Th and U.

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