Acta Montanistica Slovaca
ISSN 1335-1788
actamont.tuke.sk

Natural radioactivity in select metamorphic and sedimentary rocks of the Opava Mountains (Poland): A comparison between laboratory and in situ measurements of ^{232}Th, ^{238}U, and ^{40}K activities

Agnieszka MOSKA[1], Dariusz MALCZEWSKI[2], Jerzy ŻABA[2] and Maria DZIUROWICZ[2]*

Authors' affiliations and addresses:
[1] Oil and Gas Institute - National Research Institute, Lubicz 25A. 31-503 Krakow, Poland
e-mail: moskaa@inig.pl

[2] Institute of Earth Sciences, University of Silesia, Będzińska 60, 41-200 Sosnowiec, Poland
e-mail: dariusz.malczewski@us.edu.pl

[2] Institute of Earth Sciences, University of Silesia, Będzińska 60, 41-200 Sosnowiec, Poland
e-mail: jerzy.zaba@us.edu.pl

[2] Institute of Earth Sciences, University of Silesia, Będzińska 60, 41-200 Sosnowiec, Poland
e-mail: maria.dziurowicz@us.edu.pl

***Correspondence:**
Dariusz Malczewski, Institute of Earth Sciences, University of Silesia, Będzińska 60, 41-200 Sosnowiec, Poland
tel.: +48 604991794
e-mail: dariusz.malczewski@us.edu.pl

Funding information:
Research program of the Institute of Earth Sciences, University of Silesia, Poland, ZB-14-2020

How to cite this article:
Moska, A., Malczewski, D., Żaba, J. and Dziurowicz, M. (2023). Natural radioactivity in select metamorphic and sedimentary rocks of the Opava Mountains (Poland): A comparison between laboratory and in situ measurements of ^{232}Th, ^{238}U, and ^{40}K activities. *Acta Montanistica Slovaca*, Volume 28 (1), 250-262

DOI:
https://doi.org/10.46544/AMS.v28i1.20

Abstract
This paper reports the natural radioactivities of eight characteristic rocks from the Opava Mountains region (Eastern Sudetes, Poland) as measured in the laboratory and under in situ field conditions. Activity concentrations for ^{232}Th, ^{238}U, and ^{40}K were obtained using an HPGe gamma-ray spectrometry system. Activities for ^{232}Th ranged from 2 to 39 Bq kg^{-1} as measured in the laboratory and from 5 to 38 Bq kg^{-1} measured under in situ field conditions. For ^{238}U, the activity concentrations ranged from 4 to 37 Bq kg^{-1} in the laboratory and from 7 to 42 Bq kg^{-1} under in situ field conditions. For ^{40}K, activity concentrations ranged from 12 to 820 Bq kg^{-1} in the laboratory and from 60 to 826 Bq kg^{-1} under in situ field conditions. Measured activity concentrations were compared with average activity concentrations of radionuclides in similar types of rocks from different global localities. The in situ field measurement and laboratory measurements did not show significant differences indicating that both methods provide reliable ^{232}Th, ^{238}U, and ^{40}K results for different lithologies.

Keywords
Natural radioactivity, Opava Mountains, laboratory measurements, in situ measurements, gamma-ray spectrometry

© 2023 by the authors. Submitted for possible open access publication under the terms and conditions of the Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).

Introduction

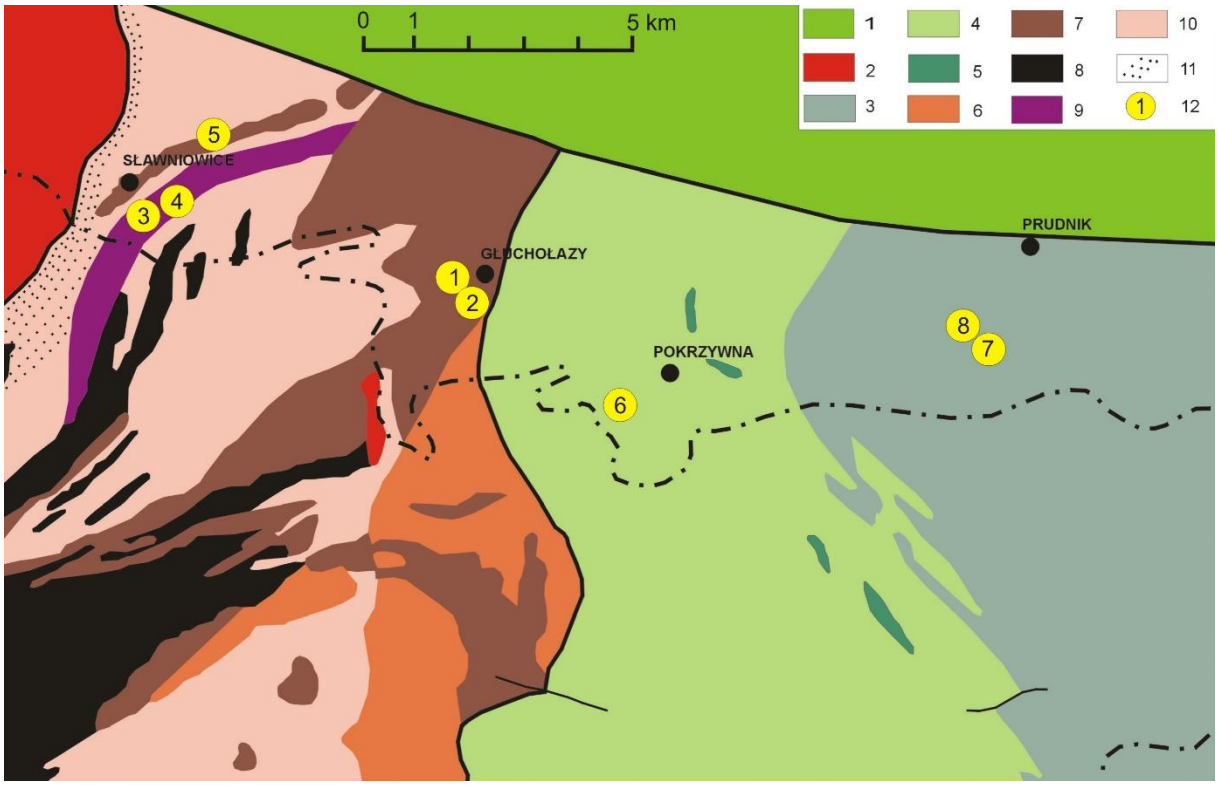
Due to implications for health and mineral exploration, many studies have addressed the subject of the radioactivity generated by naturally occurring radionuclides in rocks. Levels of natural radiation are a function of lithology, composition, and specifically, the ^{40}K, ^{232}Th and ^{238}U content of rocks. The rocks analyzed by this study here come from the Opava Mountains, which form the easternmost features of the Eastern Sudetes (SW Poland). These mountains strike approximately E-W along the Polish border with the Czech Republic and are well known for their lithological and mineral diversity. This region specifically hosts a number of mining heritage sites associated with gold exploration. Only a small area of the mountain range occurs in Poland with the larger proportion residing in the Czech Republic.

This study reports results of both in situ field and laboratory measurements of natural radioactivity from eight rock types found in the Opava Mountains. Rocks analyzed include quartzites, shales, and carbonates. The objective of this study was to determine any variation between in situ field and laboratory measurements of ^{40}K, ^{232}Th, and ^{238}U activities in different rock types. Previous studies have addressed radioactivity measurements of granites and gneisses found in the Opava Mountains region (Dżaluk et al., 2014, 2018). These studies provide rigorous evidence for optimal and/or efficient approaches to measuring radioactivity for different lithologies and different localities (Dziurowicz et al., 2017).

Geological settings and locations of rock samples

The Opava Mountains belong to the western part of the Upper Silesia Block, which together with the Brno Block, forms the Brunovistulicum structure (Żaba et al., 2005). These structures and their lithologies formed from Variscan deformation, overthrusts, and metamorphism during collisions between the Bohemian Massif and the Brunovistulicum (Schulmann and Gayer, 2000). The Opava Mountains host rocks of different ages and lithology associated with five structural elements that strike roughly N-S (Żaba et al., 2005). To the west, the East Sudetic nappe includes the Velké Vbrno and Keperník nappes resting on parautochthonous gneisses of the Desná unit and overlain by the Devonian allochthonous volcano-sedimentary Vrbno group (Mazur et al., 2007). The Upper Devonian/Lower Carboniferous Andělská-Hora Formation is the oldest sequence of a Variscan flysch formation. It consists of dark phyllites, metagreywackes, and subordinate metaconglomerates (Kozdrój, 2003). Metamorphism does not appear to have affected sandstones, mudstones, and conglomerates (Żaba et al., 2005) of the Lower Carboniferous Horní-Benešov Formation. To the west, the Horní-Benešov contacts the Andělská-Hora Formation, and to the east, it contacts the Moravice Formation. The Žulova Massif consists of Upper Carboniferous Variscan granitoid. It occurs as an approximately 200 m thick body within an apophysis and records acidic volcanism in the area. The Žulova Massif otherwise consists of Proterozoic Desna Series gneisses.

The lithologies analyzed by in situ field and laboratory measurements (the latter using sample collection) occurred at several localities. Devonian quartzite (Fig. 1, point 1) and quartzitic shale (Fig. 1, point 2) of the Vbrno group are exposed in the Głuchołazy area. Additional locations occurred in the Sławniowice quarry, which hosts calcitic (Fig. 1, point 3) and dolomitic marbles (Fig. 1, point 4). Localities in Burgrabice and Jarnołtówek include outcrops of quartzite (Fig. 1, point 5) belonging to the Vbrno group and of phyllite (Fig. 1, point 6) belonging to the Andělská-Hora Formation. Two sites in the Dębowiec quarry hosted mudstone (Fig. 1, point 7) and greywacke (Fig. 1, point 8) belonging to the Horní-Benešov Formation.



1: Sandstone and marble (Upper Cretaceous), Žulova Massif (Upper Devonian/Lower Carboniferous, Upper Carboniferous). 2: Granitoid and associated contact phenomena, Horn-Benešov Formation (Lower Carboniferous). 3: Sandstone, mudshale, and conglomerate; Andělská-Hora Formation (Upper Devonian/Lower Carboniferous). 4: Phyllite, metasandstone, crystalline limestone. 5: Greenstone, Vbrna Series (Lower Devonian). 6: Phyllite, graphitic shale, crystalline limestone, and metabasite (Middle Devonian). 7: Quartzite, quartzitic shale, and mica schist; Jesenik Massif. 8: Amphibolite (Lower and Middle Devonian) Desna Series (Proterozoic). 9: Marble 10-gneiss and biotite schist. 11: Contact zone of the Žulova granitoid intrusion.

Fig. 1. *Geological map of the Opava Mountains after Żaba et al. (2005) showing locations of in situ field measurements and sample collection sites. 1: quartzite, Głuchołazy; 2: quartzitic shale, Głuchołazy; 3: calcitic marble, Sławniowice quarry; 4: dolomitic marble, Sławniowice quarry; 5: metamorphic quartzite, Burgrabice; 6: phyllite, Jarnołtówek; 7: mudstone, Dębowiec quarry; 8: greywacke, Dębowiec quarry.*

Materials and methods

Activity concentrations of naturally occurring radionuclides were measured using a GX3020 gamma-ray spectrometry workstation. This system uses a high-purity germanium (HPGe) detector with 32% relative efficiency. The energy resolutions of the detector were 0.8 keV at 122 keV and 1.7 keV at 1330 keV.

During in situ measurements, the detector was mounted about 1 m above the ground and 0.2–0.5 m from the outcrop (Fig. 2). These geometries capture a majority of the gamma rays emitted from a 2–5 m radius area (Helfer and Miller, 1988). The calculated average minimum detectable activity (MDA) was 2 Bq kg^{-1} for ^{40}K and 1 Bq kg^{-1} for ^{214}Pb, ^{214}Bi, and ^{228}Ac. A single measurement lasted 2 h, and in situ, object counting software (ISOCS) was used in efficiency calibration. The consistency of activities calculated for gamma-ray transitions for a given multiline radionuclide (e.g., ^{208}Tl, ^{214}Bi, ^{214}Pb, ^{228}Ac) was checked using line activity consistency evaluator (LACE) analysis. For all measurements obtained by in situ field and laboratory techniques described here, activity ratios for the multiline radionuclides approached unity (Canberra Industries, 2004a).

A – location 1 (Głuchołazy, Devonian quartzite); B – location 2 (Głuchołazy, quartzitic shale); C – location 3 (Sławniowice, calcitic marble); D – location 4 (Sławniowice, dolomitic marble); E – location 5 (Burgrabice, metamorphic quartzite); F – location 6 (Jarnołtówek, phyllite); G – location 7 (Dębowiec, mudstone); H – location 8 (Dębowiec, greywacke)

Fig. 2. *Photos of the site locations and detector configuration for the in situ measurements.*

For laboratory measurements, rock samples were crushed and then analyzed in 450 ml Marinelli be akers. As with field measurements, each sample was analyzed for 48 hours. The obtained spectra were interpreted using the Genie 2000 v. 3.4 software package and LabSOCS (Laboratory Sourceless Calibration Software). Spectrometer energy was calibrated using ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, and ^{203}Hg radionuclides homogeneously dispersed in a silicone resin (certificate source type Marinelli Beaker Standard Source; MBSS, supplied by the Czech Metrological Institute). Activities were calculated from the following gamma-ray transitions (energy in keV): ^{40}K = 1460.8; ^{208}Tl = 583.1, 860.5, and 2614.5; ^{212}Pb = 238.6 and 300.1; ^{214}Pb = 242, 295.2, and 351.9; ^{214}Bi = 609.3, 1120.3, and 1764.5; ^{228}Ac = 338.32, 911.6, 964.6, and 969.1.

Both the LabSOCS and ISOCS procedures require the use of Geometry Composer in Genie 2000. For both laboratory and in situ measurements, Geometry Composer required parameterizations of the Marinelli beaker size, outcrop dimensions, rock densities, and chemical compositions. Geometry Composer further required parameters of the distance between rock outcrops and the detector end-cup for in situ measurements. The LabSOCS/ISOCS software generates the full-energy efficiency for a given source-detector geometry. The model and measurements

differed by less than 5% for point-like sources and less than 10%–15% for extended sources (Canberra Industries, 2004b).

Results and discussion

Table 1 lists gamma-ray activities for the ^{40}K, ^{232}Th, and ^{238}U series. **Fig. 3** shows in situ gamma-ray spectra from locations 4 and 7.

Table 1. *Activity concentrations measured from different rock types under in situ field and laboratory conditions. Uncertainties are quoted as 1σ.*

		Activity (Bq kg^{-1})							
		Sample no.							
	Nuclide	**1 Quartzite**		**2 Quartzitic shale**		**3 Calcitic marble**		**4 Dolomitic marble**	
		in situ	laboratory	*in situ*	laboratory	*in situ*	laboratory	*in situ*	laboratory
Non series	^{40}K	345 ± 7.5	310 ± 7.0	517 ± 12	409 ± 9	60 ± 1.8	12.4 ± 1.3	121 ± 3.0	122 ± 3.0
^{232}Th series	^{*208}Tl	9.3 ± 0.2	8.5 ± 0.4	12.1 ± 0.3	11.2 ± 0.2	1.8 ± 0.1	0.75 ± 0.06	3.7 ± 0.1	1.7 ± 0.3
	^{212}Pb	23.4 ± 1.0	26.2 ± 0.9	34.1 ± 1.1	34.4 ± 1.2	5.2 ± 0.2	2.2 ± 0.2	8.4 ± 0.4	5.2 ± 0.2
	^{228}Ac	23.8 ± 0.8	24.7 ± 0.8	29.7 ± 0.7	33.2 ± 1.0	5.0 ± 0.3	2.4 ± 0.2	9.7 ± 0.3	5.0 ± 0.4
^{238}U series	^{214}Pb	21.9 ± 0.9	18.2 ± 0.6	22.0 ± 1.0	21.5 ± 0.5	21.7 ± 0.6	16.8 ± 0.4	7.2 ± 0.3	13.2 ± 0.3
	^{214}Bi	22.3 ± 0.5	17.0 ± 0.3	21.5 ± 0.4	19.0 ± 0.7	21.7 ± 0.3	16.0 ± 0.2	7.2 ± 0.2	11.9 ± 0.2
	$^{\#226}$Ra	22.1 ± 1.0	17.6 ± 0.7	21.8 ± 1.1	20.3 ± 0.9	21.7 ± 0.7	16.4 ± 0.4	7.2 ± 0.4	12.6 ± 0.4

		Activity (Bq kg^{-1})							
		Sample no.							
	Nuclide	**5 Quartzite**		**6 Phyllite**		**7 Mudstone**		**8 Greywacke**	
		in situ	laboratory	*in situ*	laboratory	*in situ*	laboratory	*in situ*	laboratory
Non series	^{40}K	826 ± 19	820 ± 18	367 ± 9	380 ± 8	647 ± 15	611 ± 13	586 ± 11	560 ± 12
^{232}Th series	^{208}Tl	6.2 ± 0.2	2.4 ± 0.3	11.8 ± 0.3	10.5 ± 0.4	15.7 ± 0.4	13.7 ± 0.5	13.5 ± 0.3	12.3 ± 0.5
	^{212}Pb	16.5 ± 0.7	6.9 ± 0.3	33.9 ± 1.1	30.8 ± 1.1	42.3 ± 1.4	40.9 ± 1.4	35.5 ± 1.2	36.1 ± 1.3
	^{228}Ac	17.0 ± 0.5	6.8 ± 0.2	30.8 ± 0.6	30.3 ± 0.9	37.8 ± 0.9	38.9 ± 1.2	33.4 ± 0.8	35.9 ± 1.1
^{238}U series	^{214}Pb	6.3 ± 1.2	4.2 ± 0.2	25.0 ± 1.1	20.8 ± 0.7	41.3 ± 1.8	37.1 ± 1.1	41.3 ± 1.0	36.2 ± 1.1
	^{214}Bi	7.5 ± 0.3	3.9 ± 0.2	24.9 ± 0.4	20.8 ± 0.3	42.8 ± 0.7	37.7 ± 0.4	41.1 ± 0.6	36.7 ± 0.4
	^{226}Ra	6.9 ± 1.2	4.1 ± 0.3	25.0 ± 1.2	20.8 ± 0.8	42.1 ± 1.9	36.5 ± 1.2	41.2 ± 1.2	36.5 ± 1.2

**Branching ratio 36%; $^{\#}$Based on the ^{214}Pb and ^{214}Bi activities.*

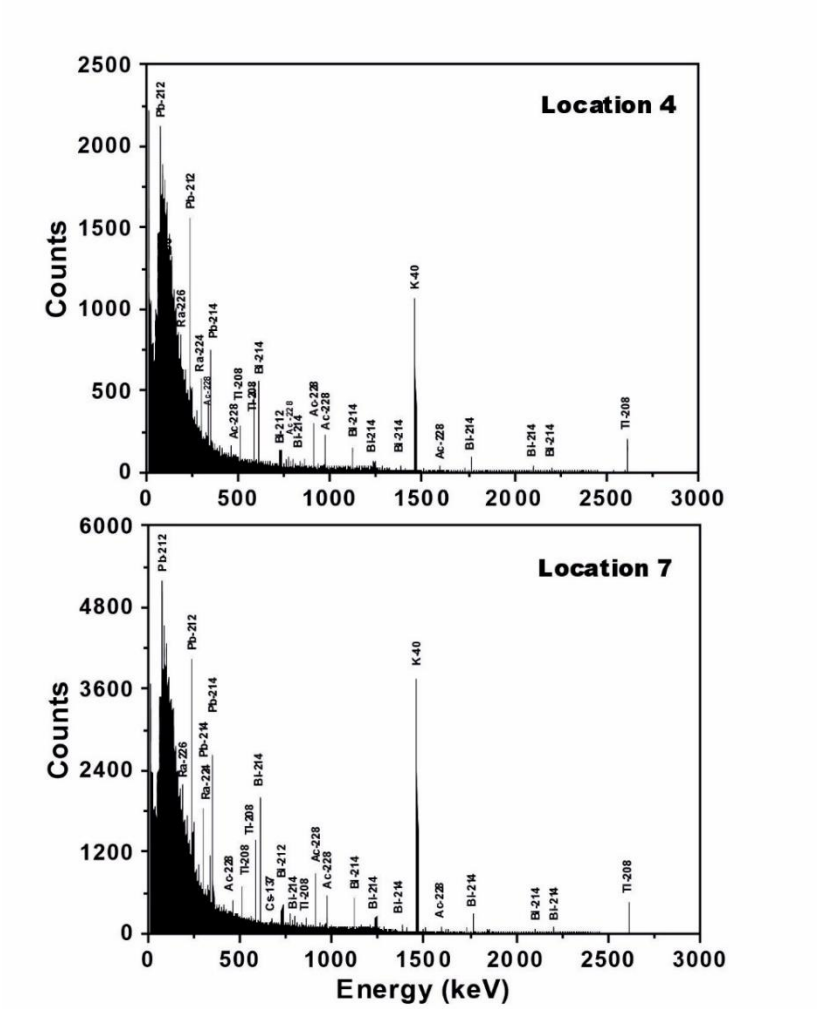


Fig. 3. *In situ gamma-ray spectra for locations 4 (dolomitic marble) and 7 (mudstone). The characteristic γ-ray emitters are marked above the corresponding peaks.*

^{40}K

As is shown in Table 1, the Burgrabice quartzite exhibited the highest ^{40}K activities observed (826 Bq kg^{-1}) under both in situ and laboratory conditions (826 Bq kg^{-1} and 820 Bq kg^{-1}, respectively). The calcitic marble from the Sławniowice quarry exhibited the lowest ^{40}K activities observed under both in situ and laboratory conditions (60 Bq kg^{-1} and 12.4 Bq kg^{-1}, respectively).

Both in situ and laboratory measurements gave relatively low ^{40}K activities for the dolomitic marble from the Sławniowice quarry (121 and 122 Bq kg^{-1}, respectively). Both marbles gave an average ^{40}K activity of 79 Bq kg^{-1}, which exceeds an average activity estimated from carbonates samples of 70 Bq kg^{-1} (Fig. 4) reported in Eisenbud and Gesell (1997). The ^{40}K activities for the marbles analyzed here fell below values reported for marbles from Turkey (1260 Bq kg^{-1}, Baykara et al., 2007), Algeria (310 Bq kg^{-1}, Amrani and Tahat, 2001), India (242 Bq kg^{-1}, Ramasamy et al., 2005), and the Sinai area of Egypt (302 Bq kg^{-1}, Fares, 2016). ^{40}K activities for dolomitic marble reported here resemble values reported for similar lithologies in Tunisia (117 Bq kg^{-1}, Manai et al., 2012), Egypt (143 Bq kg^{-1}, Fares et al., 2011), Nigeria (90 Bq kg^{-1}, Isinkaye et al., 2015), and France (78 Bq kg^{-1}, Malczewski and Żaba., 2012). Similar values were obtained through in situ measurements of calcitic marbles quarried in Jordan (79 Bq kg^{-1}, Khatiebeg et al., 1997) and Pakistan (57 Bq kg^{-1}, Iqubal et al., 2000; 58 Bq kg^{-1}, Tufail et al., 2000). Marbles from Sicily and India gave significantly lower ^{40}K activities (16 Bq kg^{-1}, Rizzo et al., 2001 and 10 Bq kg^{-1}, Sahoo et al., 2007, respectively). These values resembled laboratory measurements from calcitic marbles reported here.

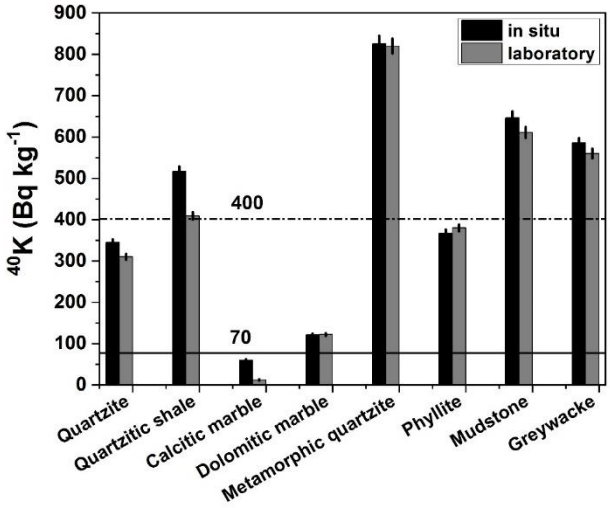


Fig. 4. *Comparison between in situ (black bars) and laboratory (gray bars) measurements of ^{40}K. Thick solid line: average ^{40}K in typical carbonate rocks. Dash dot line: average ^{40}K in typical quartzites.*

Among quartzites and quartzitic shales, the quartzite from Głuchołazy gave the lowest ^{40}K activity measured under either in situ or laboratory conditions (345 Bq kg^{-1} and 310 Bq kg^{-1}, respectively). The metamorphic quartzite from Burgrabice gave the maximum activity recorded overall (826 Bq kg^{-1} and 820 Bq kg^{-1}) for either type of measurement. Eisenbud and Gesell (1997) found that quartzites gave an average of ^{40}K activities of 400 Bq kg^{-1} (Fig. 4) showing that the metamorphic quartzite exceeded the average values noted above by a factor of two. The mudstone and greywacke from the Dębowiec quarry also exhibited higher activity values. Quartzites and shales from the Opava Mountains gave an average of ^{40}K activities of 532 Bq kg^{-1}. Sandstones can vary considerably in their ^{40}K activities. The greywacke analyzed here gave ^{40}K activity values less than half those measured from sandstones in Turkey (1158 Bq kg^{-1}, Baykara et al., 2011) but about five times greater than those measured from similar rocks in Egypt (108 Bq kg^{-1}, El Aassy et al., 2011). Metamorphic quartzite from the Opava Mountains also showed higher ^{40}K activity concentrations than those measured from quartzites in India (697 Bq kg^{-1}, Sengput et al., 2005), France (572 Bq kg^{-1}, Malczewski and Żaba, 2012), and Nigeria (257 Bq kg^{-1}, Joshua et al., 2009). The values reported here also greatly exceed those recorded from metamorphic bodies containing quartzites in the Peduase and Weija areas of Ghana (108 and 9 Bq kg^{-1}, respectively, Yeboah et al., 2001). A phyllite from Jarnołtówek gave lower ^{40}K activities than those measured from rocks collected from the Alps-Apennines transition zone (764 Bq kg^{-1}, Chiozzi et al., 2001) and the Aburi area of Ghana (727 Bq kg^{-1}, Yeboah et al., 2001). Rock samples from Nepal showed a higher average ^{40}K activity concentrations as well (477 Bq kg^{-1}, Wallova et al., 2010). The ^{40}K activity value recorded for mudstones from the Dębowiec quarry resembles that reported for similar lithologies from Nigeria (470 Bq kg^{-1}, Joshua et al., 2009). However, similar lithologies from the Alps-Apennines transition zone gave higher values (790 Bq kg^{-1}, Chiozzi et al., 2001). A quartzitic shale from the Alps-Apennines transition zone also gave higher values (543 Bq kg^{-1}, Chiozzi et al., 2001) relative to those measured for rocks from Głuchołazy.

^{232}Th series (^{208}Tl, ^{212}Pb, ^{228}Ac)

Data listed in Table 1 indicate radioactive equilibrium among ^{232}Th series nuclides for the rocks analyzed here. Since ^{228}Ac is the second radionuclide in the thorium series, the activity concentration of ^{232}Th can be estimated from the ^{228}Ac activity (Helfer and Miller, 1988). A mudstone from the Dębowiec quarry gave the highest ^{232}Th activities observed: 37.8 Bq kg^{-1} measured under in situ conditions and 38.9 Bq kg^{-1} measured under laboratory conditions. A calcitic marble from the Sławniowice quarry gave the lowest activity values measured: 5 Bq kg^{-1} under in situ conditions and 2.4 Bq kg^{-1} under laboratory conditions.

The dolomitic marble samples gave ^{232}Th activity concentrations of 9.7 Bq kg^{-1} under in situ conditions and 5 Bq kg^{-1} under laboratory conditions. All marbles analyzed from the Opava Mountains gave average ^{232}Th activity concentrations of 5.5 Bq kg^{-1}. These values resemble values measured from other carbonate rocks (8 Bq kg^{-1}, Eisenbud and Gesell, 1997) (Fig. 5). Other studies have observed higher ^{232}Th activities in marbles from Pakistan (32 Bq kg^{-1}, Iqubal et al., 2000; 26 Bq kg^{-1}, Tufail et al., 2000), the Sinai area of Egypt (49 Bq kg^{-1}, Fares, 2016), and Turkey (20 Bq kg^{-1}, Baykara et al., 2007). The values reported here are also lower than those obtained for similar rocks in Algeria (18 Bq kg^{-1}, Amrani and Tahat, 2001) and India (12 Bq kg^{-1}, Ramasamy et al., 2005). The dolomitic marble from the Sławniowice quarry gave similar ^{232}Th activities to those reported for rocks from Jordan (10 Bq kg^{-1}, Khatiebeh et al., 1997), Tunisia (8 Bq kg^{-1}, Manai et al., 2012), Nigeria (8 Bq kg^{-1}, Isinkaye et

al., 2015), and Egypt (6 Bq kg^{-1}, Fares et al., 2011). Both calcitic and dolomitic marbles found in the Sławniowice quarry gave higher ^{232}Th activity values than those measured in similar rocks from India (3 Bq kg^{-1}; Sahoo et al. 2007) and marbles from Spain (3 Bq kg^{-1}, Walley El-Dine et al., 2001) and France (2 Bq kg^{-1}, Malczewski and Żaba., 2012). The values reported here exceed those measured in similar rocks from Greece (1 Bq kg^{-1}, Walley El-Dine et al., 2001) and Sicily (1 Bq kg^{-1}, Rizzo et al., 2001).

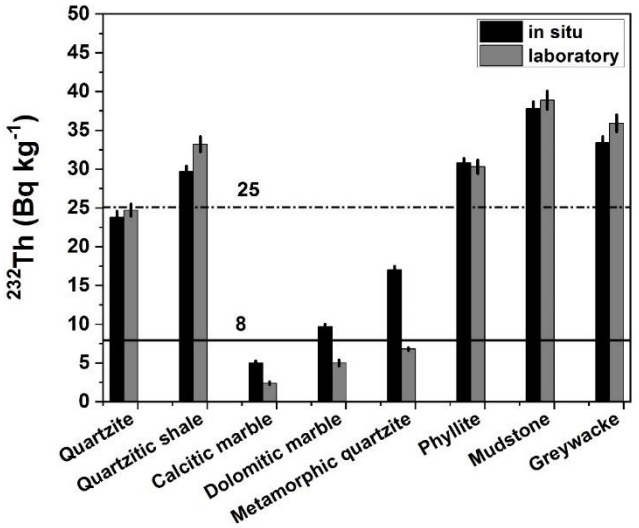


Fig. 5. *Comparison between in situ (black bars) and laboratory (gray bars) measurements of ^{232}Th. Thick solid line: average ^{232}Th in typical carbonate rocks. Dash dot line: average ^{232}Th in typical quartzites.*

As noted above, a mudstone from the Dębowiec quarry gave the highest ^{232}Th activity concentrations observed, while quartzites and quartzitic shales from Burgrabice gave the minimum ^{232}Th activities observed (17 Bq kg^{-1} under in situ field conditions and 6.8 Bq kg^{-1} under laboratory conditions). The ^{232}Th activity values reported here gave an arithmetic mean of 28.5 Bq kg^{-1}, which strongly resembles that reported for typical quartzites (25 Bq kg^{-1}; Fig. 5) by Eisenbud and Gesell (1997). Excluding the quartzites from Burgrabice and Głuchołazy, almost all rocks analyzed here gave higher activity concentrations than the 25 Bq kg^{-1} mean noted above. The ^{232}Th activity values reported here fell significantly below values reported for quartzites from Nigeria (75 Bq kg^{-1}; Joshua et al., 2009) and slightly below values reported for rocks from India (29 Bq kg^{-1}; Sengput et al., 2005). Similar rocks from France (Malczewski and Żaba, 2012) and the Peduase and Weija areas of Ghana (Yeboah et al., 2001), however, gave lower respective ^{232}Th activity concentrations (10, 6, and 1 Bq kg^{-1}, respectively, for the Ghanaian rocks). The phyllite from Jarnołtówek gave ^{232}Th activity concentrations that fell below the average activity reported for a phyllite from the Aburi area of Ghana (112 Bq kg^{-1}, Yeboah et al., 2001) by a factor of four. The Jarnołtówek phyllite ^{232}Th activity values were also lower than those reported for similar rocks from Nepal (92 Bq kg^{-1}, Wallova et al., 2010) and the Alps-Apennines transition zone (43 Bq kg^{-1}, Chiozzi et al., 2001). Quartzitic shales from the Alps-Apennines transition zone gave lower activity concentrations (43 Bq kg^{-1}) than those reported here for analogous rock types from the Opava Mountains region, indicating that these relationships vary with lithology. Mudstones also show considerable variation. Chiozzi et al. (2001) reported ^{232}Th activity values of 51 Bq kg^{-1} for mudstones. Mudstones from Nigeria gave ^{232}Th activity values that exceed those measured from Opava Mountain mudstones by a factor of two (80 Bq kg^{-1}, Joshua et al., 2009). Greywackes from the Dębowiec quarry gave ^{232}Th activity values that exceed those reported in the literature for similar rock types. A sandstone from Nigeria which gave ^{232}Th activity values of up to 88 Bq kg^{-1} (Joshua et al., 2009) represents an exception to this trend.

^{238}U series (^{214}Pb, ^{214}Bi, ^{226}Ra)

The activity concentrations of ^{238}U were estimated assuming radioactive equilibrium in the ^{238}U → ^{226}Ra → ^{222}Rn → ^{214}Pb → ^{214}Bi decay chain. The ^{238}U activity concentration was estimated from the ^{226}Ra activity concentration, which itself was calculated as the arithmetic mean of ^{214}Pb and ^{214}Bi activity values. Results in Table 1 show that mudstone from the Dębowiec quarry gave the maximum ^{238}U activity concentrations observed (42.1 Bq kg^{-1} under in situ conditions and 36.5 Bq kg^{-1} under laboratory conditions). These values resemble values obtained for the greywacke from the same quarry. In situ measurements gave slightly different values. The metamorphic quartzite gave the lowest observed ^{238}U activity values of 6.9 Bq kg^{-1} under in situ conditions and 4.1 Bq kg^{-1} under laboratory conditions.

Among carbonate rocks from the Opava Mountains, in situ measurements of a calcitic marble gave the highest ^{238}U activity values of 21.7 Bq kg^{-1}. In situ measurements of a dolomitic marble gave the lowest observed ^{238}U activity values of 7.2 Bq kg^{-1}. Eisenbud and Gesell (1997) reported an average of ^{238}U activity values for carbonate rocks of 25 Bq kg^{-1} (**Fig. 6)**. This exceeds ^{238}U activity values measured from marbles analyzed here (14.5 Bq kg^{-1}).

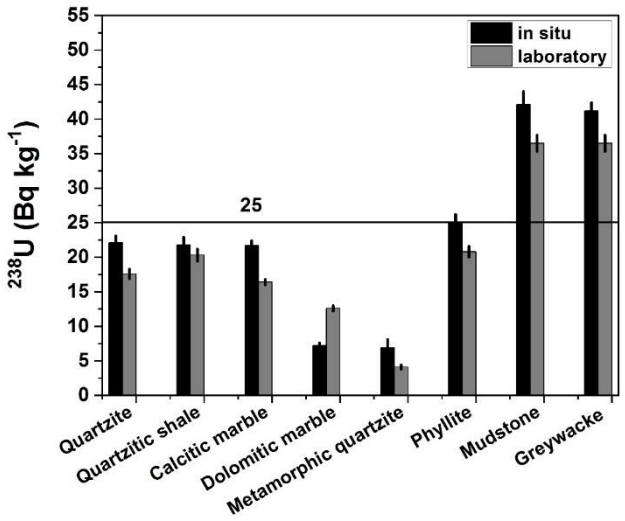


Fig. 6. *Comparison between in situ (black bars) and laboratory (gray bars) measurements of ^{238}U. Thick solid line: average ^{238}U in typical carbonate and quartzite rocks.*

Generally, the ^{238}U activities reported here resembled values reported for similar rocks from different localities. Khatibeth et al. (1997) reported ^{238}U activity concentrations of 116 Bq kg^{-1} for marbles from Jordan. Rocks from Egypt gave ^{238}U activity values three times as high (57 Bq kg^{-1}, Fares et al., 2011) as in situ measurements of calcitic marbles from the Sławniowice quarry. Rocks from the Sinai area of Egypt (54 Bq kg^{-1}, Fares, 2016), Tunisia (33 Bq kg^{-1}, Manai e. al., 2012), Turkey (30 Bq kg^{-1}, Baykara et al., 2007), and Pakistan (27 Bq kg^{-1}, Iqubal et al., 2000; 33 Bq kg^{-1}, Tufail et al., 2000) also gave higher ^{238}U activity values. Malczewski and Żaba (2012) obtained similar results for rocks from France (26 Bq kg^{-1}), and Amrani and Tahtat (2001) obtained similar results for rocks from Algeria (23 Bq kg^{-1}). Rocks from India (8 Bq kg^{-1}, Ramasamy et al., 2005; 12 Bq kg^{-1}, Sahoo et al., 2007) and marbles from Nigeria (11 Bq kg^{-1}, Isinkaye et al., 2015) gave lower ^{238}U activity values than the calcitic marble from the Opava Mountains. These former values, however, resembled those obtained for the dolomitic marble analyzed here. The ^{238}U activity values for marbles reported here exceed those reported for marbles from Sicily (4 Bq kg^{-1}, Rizzo et al., 2001), Spain, India, Greece (6 Bq kg^{-1}), and Italy (8 Bq kg^{-1}, Walley El-Dine et al., 2001).

The mudstone from the Dębowiec quarry exhibited the maximum ^{238}U activity concentration observed among quartzites and shales. Quartzite and quartzitic shales from Głuchołazy and the phyllite from Jarnołtówek gave average values that were lower by a factor of two. The quartzite from Burgrabice gave the lowest ^{238}U activity values observed: 6.9 Bq kg^{-1} measured under in situ conditions and 4.1 Bq kg^{-1} measured under laboratory conditions. Quartzitic rocks gave an average ^{238}U value of 24.6 Bq kg^{-1} which resembled that reported for typical quartzites (25 Bq kg^{-1}; Fig. 6) by Eisenbud and Gesell (1997). The ^{238}U activity concentrations for Opava Mountains quartzites fell below values reported for quartzites from Nigeria (44 Bq kg^{-1}, Joshua et al., 2009) but exceeded those reported for rocks from India (19 Bq kg^{-1}, Sengput et al., 2005), France (9 Bq kg^{-1}, Malczewski and Żaba, 2012), and from the Peduase and Weija areas of Ghana (12 and 1 Bq kg^{-1}, respectively, Yeboah et al., 2001). Phyllites from Nepal (42 Bq kg^{-1}, Wallova et al., 2010) and the Alps-Apennines transition zone (36 Bq kg^{-1}, Chiozzi et al., 2001) gave higher ^{238}U activity concentrations than the phyllite from Jarnołtówek. A phyllite from the Aburi area of Ghana gave a ^{238}U activity value of 15 Bq kg^{-1} (Yeboah et al., 2001), lower than that reported here. The ^{238}U activity values from the Głuchołazy quartzitic shale exceeded those measured for rocks from the Alps-Apennines transition zone (15 Bq kg^{-1}, Chiozzi et al., 2001). Mudstones from the Alps-Apennines transition zone gave ^{238}U activity values similar to those measured from Debowięc quarry rocks. The average ^{238}U activity in sandstones from Egypt (103 Bq kg^{-1}, El Aassy et al., 2011) exceeded values measured from the Dębowiec quarry greywacke by a factor of two. A sandstone from Nigeria gave values similar to those reported here (38 Bq kg^{-1}, Joshua et al., 2009). Only sandstones from Turkey have given lower ^{238}U activity values (25 Bq kg^{-1}; Baykara et al., 2011) than those measured from Opava Mountains samples.

Comparison between *in situ* and laboratory measurements

As seen in Figs. 4-6, in situ field and laboratory measurement techniques gave similar activity values for different lithologies and isotopic systems. The in situ field measurements gave average ^{232}Th and ^{238}U activity values that equated within error to those obtained by laboratory methods. In situ and laboratory measurements gave ^{40}K activity values that varied by 108 Bq kg^{-1} only for the quartzitic shale (Fig. 4). For other rock types, in situ and laboratory measurement techniques gave average values that differed in absolute terms ($|^{40}K_{in\ situ} - {}^{40}K_{lab}|$) by 23 Bq kg^{-1}. This value resembles individual uncertainties listed in Table 1. Fig. 5 shows that ^{232}Th activities show the smallest differences between the in situ and laboratory measurement techniques. The average absolute difference in $|^{232}Th_{in\ situ} - {}^{232}Th_{lab}|$ was 3.3 Bq kg^{-1}. Among ^{238}U activity values, the mudstone exhibited the largest difference (5.6 Bq kg^{-1}) between measurements made under laboratory and in situ field conditions (Fig. 6). The average absolute differences in $|^{238}U_{in\ situ} - {}^{238}U_{lab}|$ was 4.3 Bq kg^{-1} for all locations and samples.

Estimates of K (%), Th (ppm), and U (ppm)

Table 2 lists the average K (%), Th (ppm), and U (ppm) concentrations as calculated based on activities listed in Table 1.

Table. 2. *Concentrations of K (%), Th (ppm), and U (ppm).*

	1 Quartzite	2 Quartzitic shale	3 Calcitic marble	4 Dolomitic marble	5 Metamorphic quartzite
K (%)	1.08 ± 0.02	1.53 ± 0.04	0.12± 0.007	0.4 ± 0.1	2.71 ± 0.06
Th (ppm)	5.96 ± 0.2	7.73 ± 0.21	0.91 ± 0.06	1.81 ± 0.09	2.93 ± 0.09
U (ppm)	1.61 ± 0.07	1.70 ± 0.08	1.55 ± 0.05	0.8 ± 0.03	0.45 ± 0.06

	6 Phyllite	7 Mudstone	8 Greywacke
K (%)	1.23 ± 0.03	2.08 ± 0.04	1.89 ± 0.04
Th (ppm)	7.51 ± 0.19	9.43 ± 0.26	8.52 ± 0.24
U (ppm)	1.85 ± 0.08	3.22 ± 0.13	3.14 ± 0.09

Potassium concentrations ranged from 0.12 wt.% (calcitic marble) to 2.71 wt.% (metamorphic quartzite). Similar to K concentrations, the lowest Th concentration (0.91 ppm) occurred in the calcitic marble, while the highest Th concentration (9.43 ppm) occurred in the mudstone. Uranium concentrations varied from 0.45 ppm in metamorphic quartzite to 3.22 ppm in mudstone.

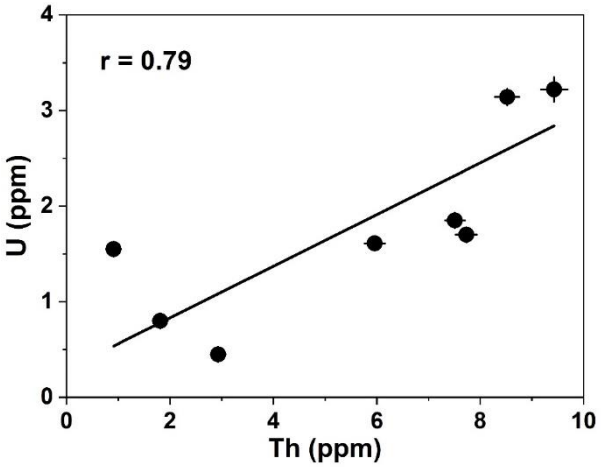


Fig. 7. *Correlation between Th (ppm) and U (ppm). The solid line represents the linear fit: U (ppm) = 0.24 x Th (ppm) + 0.47. The correlation coefficient r = 0.79.*

As shown in Fig. 7, a strong positive correlation occurs between U and Th concentrations (correlation coefficient r = 0.79). Moderate and weak correlations occur between K and Th (r = 0.51) and between K and U (r = 0.20). This likely reflects abundant potassium in the metamorphic quartzite (Fig. 8). Regional metamorphism (e.g., Polański and Smulikowski, 1969) caused potassium enrichment in micas observed in these rocks. Excluding the metamorphic quartzite, the K and Th data gave a correlation coefficient of 0.98, and the K and U data gave a correlation coefficient of 0.84 (Fig. 8).

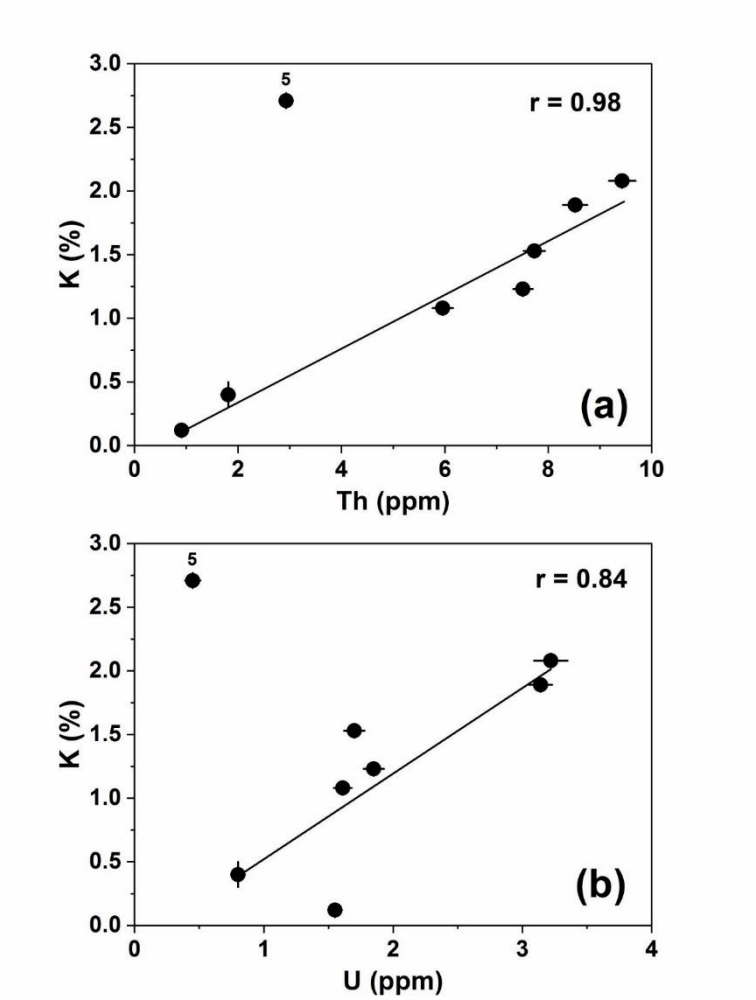


Fig. 8. *Correlation between* ***(a)*** *K (%) and Th (ppm) and* ***(b)*** *K (%) and U (ppm). The solid lines represent the linear fit excluding point 5: K (%) = 0.21 × Th (ppm) and K (%) = 0.69 × U (ppm). The correlation coefficients are r = 0.98 and 0.84, respectively.*

Conclusions

Laboratory and in situ measurements of gamma-ray radiation from rocks in the Opava Mountains gave consistent values for ^{40}K, ^{232}Th, and ^{238}U activities. A metamorphic quartzite gave the highest ^{40}K activity concentration observed. A mudstone gave the highest ^{232}Th and ^{238}U activities observed. Marbles gave average ^{40}K, ^{232}Th, and ^{238}U activity concentrations of 79, 5.5, and 14.5 Bq kg^{-1}, respectively. Quartzites and shales gave average ^{40}K, ^{232}Th, and ^{238}U activity concentrations of 532, 28.5, and 24.6 Bq kg^{-1}, respectively. The Th (ppm) and U (ppm) concentrations showed strong positive correlations. Excluding metamorphic quartzite, rocks also showed strong K (%) and Th (ppm) correlations.

References

Amrani, D. and Tahtat, M. (2001). Natural radioactivity in Algieria building materials. Applied Radiation and Isotopes, 54 (4), pp. 687-689, DOI: 10.1016/s0969-8043(00)00304-3.

Attendorn, H. G. and Bowen, R. N. C. (1997). Radioactive and stable isotope geology. Chapman & Hall, London.

Baykara, O., Doğru, M. and Karatece, S. (2007). Assesment of natural radioacticity and its significant hazards of different kinds of marbles in Elazig (Turkey). Natural and Applied Sciences, 2 (4), pp. 256-264.

Baykara, O., Karatepe, S. and Doğru, M. (2011). Assessments of natural radioactivity and radiological hazards in construction materials used in Elazig, Turkey. Radiation Measurements, 46 (1), pp. 153-158, DOI: https://doi.org/10.1016/j.radmeas.2010.08.010.

Canberra Industries (2004a). Genie 2000 3.0 operations manual, Meriden, USA.

Canberra Industries (2004b). ISOCS/LabSOCS detector characterization. Report ACK#0403379, Meriden, USA.

Chiozzi, P., Pasquale, V. and Verdoya, M. (2002). Naturally occurring radioactivity at the Alps–Apennines transition. Radiation measurements, 35 (2), pp. 147-154.

Dziurowicz, M., Malczewski, D. and Żaba, J. (2017). 222Rn and 220Rn concentrations in selected soils developed on the igneous rocks of the Kaczawa Mountains (Sudetes, Poland). Acta Montanistica Slovaca, 22 (3), pp. 238-245.

Dżaluk, A., Malczewski, D., Żaba, J. and Dziurowicz M. (2014). In situ measurements of natural radioactivity in selected igneous rocks of the Opava Mountain region. Contemporary Trends in Geosciences, 3 (1), pp. 5-13.

Dżaluk, A., Malczewski, D., Żaba, J. and Dziurowicz M. (2018). Natural radioactivity in granites and gneisses of the Opava Mountains (Poland): a comparison between laboratory and in situ measurements. Journal of Radioanalytical and Nuclear Chemistry, 316, pp. 101-109.

Eisenbud, M. and Gesell, T. (1997). Environmental radioactivity from natural, industrial and military sources. Academic Press, San Diego, CA.

El Aassy, I. E., El Galy, M. M., Nada, A. A., El Feky, M. G. and Abd El Maksound, T. M. (2011). Effect of alteration processes on the distribution of radionuclides in uraniferous sedimentary rocks and their environmental impact, southwestern Sinai, Egypt. Journal of Radioanalytical and Nuclear Chemistry, 289 (1), pp 173-184. DOI: https://doi.org/10.1007/s10967-011-1059-1.

Fares, S., Yassene Ali, A. M., Ashour, A., Abu-Assy, M. K. and Abd El-Rahman, M. (2011). Natural radioactivity and the resulting radiation doses in some kinds of commercially marble collected from different quarries and factories in Egypt. Natural Science, 3 (10), pp. 895-905. DOI: 10.4236/ns.2011.310115.

Fares, S. (2016). Natural radioactivity emitted from granite and marble samples collected from Sinai Area Egypt and excess lifetime cancer risk. International Journal of Scientific & Engineering Research, 7 (12), pp. 432-444.

Helfer, I. K., and Miller, K. M. (1998). Calibration factors for Ge detectors used for field spectrometry. Health Physics, 55 (1), pp. 15-29. DOI: 10.1097/00004032-198807000-00002.

Isinkaye, O. M., Jibiri, N. N. and Olomide, A. A. (2015). Radiological health assessment of natural radioactivity in the vicinity of Obajana cement factory, North Central Nigeria. Journal of Medical Physics, 40 (1), pp. 52-59. DOI: 10.4103/0971-6203.152256.

Iqbal, M., Tufail, M. and Mirza, S. (2000). Measurement of natural radioactivity in marble found in Pakistan using a NaI(Tl) gamma-ray spectrometer. Journal of Environmental Radioactivity, 51 (2), pp. 255-265. DOI: 10.1016/S0265-931X(00)00077-1.

Joshua, E. O., Ademola, J. A., Akpanowo, M. A., Oyebanjo, O. A. and Olorode, D. O. (2009). Natural radionuclides and hazards of rock samples collected from Southeastern Nigeria. Radiation Measurements, 44 (4), pp. 401-404. DOI: https://doi.org/10.1016/j.radmeas.2009.04.002.

Khatibeh, A. J. A. H., Ahmad, N., Matiullah and Kenawy, M. A. (1997). Natural radioactivity in marble stones-Jordan. Radiation Measurements, 28 (1-6), pp. 345-348. DOI: 10.1016/S1350-4487(97)00097-8.

Kozdrój, W. (2003). Metabasites from the Polish of the Andelska Hora formation (Moravo-Silesian zone): Geochemistry, metamorphic history and geotectonic meaning. Journal of the Czech Geological Society, 48 (1-2), p. 82.

Malczewski, D. and Żaba, J. (2012). Natural radioactivity in rocks of the Modane–Aussois region (SE France). Journal of Radianalitycal and Nuclear Chemistry, 292 (1), pp. 123-130. DOI: 10.1007/s10967-011-1428-9.

Manai, K., Ferchichi, C., Oueslati, M. and Trablesi, A. (2012). Gamma radiation measurements in Tunisian marbles. World Journal of Nuclear Science and Technology, 2 (03), pp. 80-84. DOI: 10.4236/wjnst.2012.23013.

Mazur, S., Aleksandrowski, P., Turniak, K. and Awdankiewicz, M. (2007). Geology, tectonic evolution and Late Paleozoic magmatism of Sudetes – an overview. In: A. Kozłowski and J. Wiszniewska, ed., AM Monograph No. 1: Granitoids in Poland, KNMPAN-WG UW, Warszawa.

Polański, A., and Smulikowski, K. (1969). Geochemia. Wydawnictwa Geologiczne, Warszawa.

Ramasamy, V., Ponnusamy, V., Hemalatha, J., Meenakshisundaram, V. and Gajendrian, V. (2005). Evaluation of natural radioactivity and radiological hazards caused by different marbles of India. Indian Journal of Pure and Applied Physics, 43 (11), pp. 815-820.

Rizzo, S., Brai, M., Basile, S., Bellia, S. and Hauser, S. (2001). Gamma activity and geochemical features of building materials: Estimation of gamma dose rate and indoor radon levels in Sicily. Applied Radiation and Isotopes, 55 (2), pp. 259-265. DOI: 10.1016/S0969-8043(00)00384-5.

Sahoo, B. K., Dipen, Nathwani, Eappen, K. P., Ramachandran, T. V., Gaware, J. J. and Mayya Y. S. (2007). Estimation of radon emanation factor in Indian building materials. Radiation Measurements, 42 (8), pp. 1422-1425. DOI: 10.1016/j.radmeas.2007.04.002.

Schulmann, K. and Gayer, R. (2000). A model for a continental accretionary wedge developed by oblique collision: the NE Bohemian Massif. Journal of the Geological Society, 157 (2), pp. 401-416. DOI: 10.1144/jgs.157.2.401.

Sengupta, D., Ghoshb, A. and Mamtani, M. A. (2004). Radioactivity studies along fracture zones in areas around Galudih, East Singhbhum, Jharkhand, India. Applied Radiation and Isotopes, 63 (3), pp. 409-414. DOI: 10.1016/j.apradiso.2005.04.007.

Tufail, M., Iqubal, M. and Mirza, S. M. (2000). Radiation doses due to the natural radioactivity in Pakistan marble. Radioprotection, 35 (3), pp. 299-310. DOI: 10.1051/radiopro:2000110.

Walley El-Dine, N., E-Shershaby, A., Ahmed, F. and Abdel-Haleem, A. S. (2001). Measurements of radioactivity and radon exhalation rate in different kinds of marbles and granites. Applied Radiation and Isotopes, 55 (6), pp. 853-860. DOI: 10.1016/S0969-8043(01)00107-5.

Wallova, G., Acharya, K. K. and Wallner, G. (2010). Determination of naturally occurring radionuclides in selected rocks from Hetaunda Area, Central Nepal. Journal of Radioanalytical and Nuclear Chemistry, 283 (3), pp. 713-718. DOI: 10.1007/s10967-009-0401-3.

Yeboah, J., Boadu, M. and Darko, E. O. (2001). Natural radioactivity in soils and rocks within the Greater Accra Region of Ghana. Journal of Radioanalytical and Nuclear Chemistry, 249 (3), pp. 629-632. DOI: 10.1023/A:1013262702436.

Żaba, J., Ciesielczuk, J., Malik, K, and Strzyżewska-Konieczna, S. (2005). Structural and structural evolution of the Devonian and Carboniferous rocks in the Opava Mountains. In: J. Jureczek, Z. Buła and J. Żaba, ed., Geology and Environmental Protection of the Upper Silesia Region. LXXVI Scientific Conference of Polish Geological Society. Państwowy Instytut Geologiczny, Polskie Towarzystwo Geologiczne, Warszawa.