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Natural radionuclides in rocks of King George Island (West Antarctica) as tracers of chemical processes

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Abstract: During the austral summer of 2002/03, 253 gamma radiation spectrometric measurements of uranium, thorium, and potassium (further as U, Th, and K, respectively) content on outcrops of magmatic-sedimentary succession of the Admiralty Bay (King George Island) were performed. Low content and stable U/Th/K ratios were found in most cases in unaltered igneous rocks. Three examples of anomalous Th/K and U/K ratios were found in the Ezcurra Fault Zone, a quartz-pyrite lode of the Keller Peninsula, and a Paleogene paleosol at Cytadela Hill. In the first case, an enrichment in K was found to occur due to hydrothermal alterations in the fault zone. The high Th content in the quartz lode is evidence of volcanogenic leaching of K and, to some extent, U from the granodioritic rock. A significant mass loss during the leaching resulted in the enrichment of the inert Th content in the residuum. Modification of Th/K ratios due to chemical weathering was found in the Paleogene paleosol, where K leaching occurred. The K/Th ratio is a valuable tool for mapping hydrothermal alteration phenomena and locating Paleogene chemical weathering zones.

Keywords: Antarctic, South Shetlands, uranium, thorium, potassium, gamma spectrometry.

Introduction

The geology of King George Island has been investigated since 1820 (Adie 1957); however, systematic geological field studies were not conducted until almost a hundred years later (Ferguson 1922). Since then, a profound understanding of petrology and geological evolution has accumulated, see the review by Bastias *et al.* (2023) and references therein. Moreover, detailed morphological and geological maps were published (Birkenmajer 1980c, 1982, 1997, 2003). Despite this, little is known about the geochemistry of the radionuclides in the area. Scarce data on the Th and K content in rocks have been published, based on radiometric and petrologic analyses by Birkenmajer *et al.* (1991), who reported K enrichment in two samples from the Ezcurra fault zone.

The first paper concerning local radioactivity on King George Island was published by Godoy *et al.* (1998), which primarily referred to lichen and moss samples. Still, it also contained 11 analyses of ^{226}Ra, ^{228}Ra, ^{40}K, and ^{137}Cs in soil and sediment samples, as well as four from other islands of South Shetlands. Subsequently, Evangelista and Pereira (2002) made 22 measurements of U and Th content but did not give a precise geological description of the sampling sites. According to these studies, the changes in the Th/U ratio in rocks result from the area's chemical weathering, which, if proven, might have strong paleoclimatic implications. An excellent example of a similar analysis was presented by Zeller and Dreschhoff (1980) for the pre-Beacon (Cambrian-Devonian) chemical weathering surface in the Transantarctic Mountains.

During the austral summer of 2002/03, 253 gamma radiation spectrometric measurements on outcrops of magmatic-sedimentary succession of the Admiralty Bay (King George Island) were performed by the present authors. Data on radon and thoron daughters activity from these measurements have been used in a paper about radon activity in soil gas, social rooms, and in the greenhouse bungalow of the Henryk Arctowski Station (Solecki 2004).

In addition to U and Th, K content was also measured, which formed the basis for the correlation analysis of all three radionuclides presented in this article. They are often correlated in unaltered igneous rocks but show different geochemical behaviour in weathering and alteration zones (Wedepohl 1969). Th is commonly regarded as the most inert element and is believed to remain in weathering residuum. U behaves likewise only if contained in resistant minerals like zircon. However, most of it is adsorbed on mineral grains, can be easily leached, and forms numerous secondary uranyl minerals in the zone of chemical weathering. K is mobilized in chemical weathering zones due to incongruent dissolution during clay mineral formation from its most common minerals, *i.e.*, feldspar and biotite.

Study area

King George Island (Fig. 1) is a part of the Shetland microplate separated by a back-arc extension in Bransfield Strait from the Antarctic Peninsula. It originated at the subducting Pacific margin of Gondwana (Bastias *et al.* 2022; 2024) and contains Cretaceous-early Miocene island arc volcanites, mainly andesites and basalts, and plutonites, including gabbros, diorites, and monzonites (Birkenmajer 2001). According to Nawrocki *et al.* (2021), the stratified volcanogenic rocks forming a part of the Martel Inlet Group, *i.e.*, Keller Peninsula, Visca Anchorage, and Domeyko Glacier Formation, were emplaced there near the early/late Palaeocene boundary (*ca.* 62.11 ± 0.66 Ma ago), in the Early Eocene (*ca.* 56.3–51.9 Ma), and near the Early/Middle Eocene boundary (*ca.* 49.9–47.9 Ma), respectively. Volcanogenic formations of Keller Peninsula are intruded by the copper-bearing mineral veins of the Wegger Peak Group in several places (Barton 1965; Paulo and Rubinowski 1987; Nawrocki *et al.* 2021).

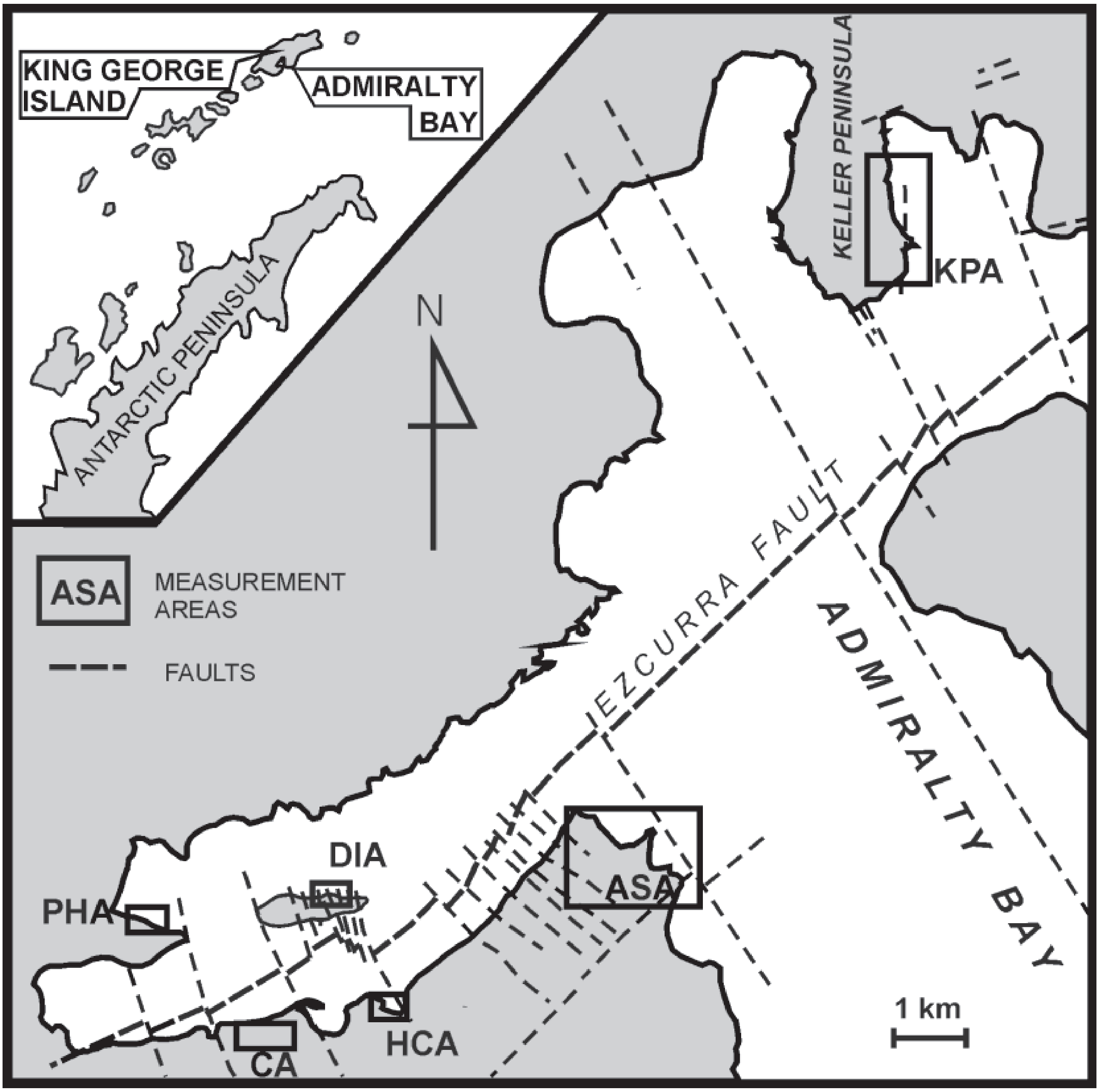

Fig. 1. Localization of the area under investigation.

The late subduction stage, connected with the opening of the Bransfield Rift and the development of the back-arc Bransfield Basin, separated the South Shetland Islands from the Antarctic Peninsula (Burton-Johnson *et al.* 2023). Basalts and their volcaniclastics are associated with this stage. Numerous quartz veins, veinlets, and red hematite alterations indicate the significant role of superimposed hydrothermal activity. Two main tectonic blocks separated by the Ezcurra Fault (Fig. 1) exist in the area under investigation: the northern, uplifted Barton Horst and the southern, downthrown Warszawa Block (Birkenmajer 2001).

Glaciations on King George Island (Fig. 2) began in the Oligocene, as the Polonez glaciation *ca.* 26.7 Ma, and lasted until the Miocene, represented by the Melville and Legru glaciations 22–21 Ma and ≤ 10 Ma, respectively, with short interglacial periods, see Fig. 2 based on Smellie *et al* (2021b). Nowadays, > 90% of the island is covered with glaciers, so rock material is accessible only locally in recent and raised pebble-cobble beaches, recent and subrecent moraines, and rocky slopes, often covered with shattered material derived from the surrounding cliffs. According to Birkenmajer (1982), the most critical weathering agent is the action of frequent successive freezing and melting. Soil cover is practically non-existent.

Methods

Measurements were performed using an Exploranium GR-320 gamma radiation spectrometer with a standard NaI (Tl) GPX-21A detector of 0.35 L volume. Impulses supplied by the detector units were classified using channels 70–204 of the spectrometer's 256 channels, which cover the energy window 850–2810 keV. Three bands (Regions of Interest, ROI) corresponding to the energy windows of the radionuclide peaks ^{40}K, ^{214}Bi, and ^{208}Tl have been set up (Table 1).

The problem of stabilizing the energy windows of channels was solved by continuously measuring cesium 662 keV photons from an internal source in the band RO/1, covering channels 51–60 (600–730 keV). The gain parameter responsible for fitting channels to energy windows was continuously updated using the least-squares fit of a Gaussian cesium peak shape every time the 5000 level of Cs counts was exceeded. This ensured that system gain was always correct and selected channels corresponded to the desired energy windows.

The instrument was set to ASSAY evaluation, which uses calibration coefficients computed during calibration on the calibration pads traceable to the Geological Survey of Canada to display data at the end of the sample period as:

TOTAL COUNT in ppm eU.
K in %,
U in ppm,
Th in ppm.

The last two concentrations were calculated assuming the existence of equilibrium in the ^{238}U and ^{232}Th decay series, which is a common practice in this kind of measurement marked by eU and eTh notation in case of U and Th content (IAEA 2003). The detector was deployed in the field for each measurement using a geometry as close as possible to 2Π. Depending on the local radionuclide content, sampling time ranged from 15 to 30 minutes to reach statistically significant counts in all ROIs.

Results

Gamma radiation spectrometric measurements have been performed in six areas: Arctowski Station area (ASA), Cytadela area (CA), Dufayel Island area (DIA), Hervé Cove area (HCA), Keller Peninsula area (KPA), and Pond Hill area (PHA), located on outcrops of various rocks along the shore of the Admiralty Bay indicated in Fig. 1. Table 2 and

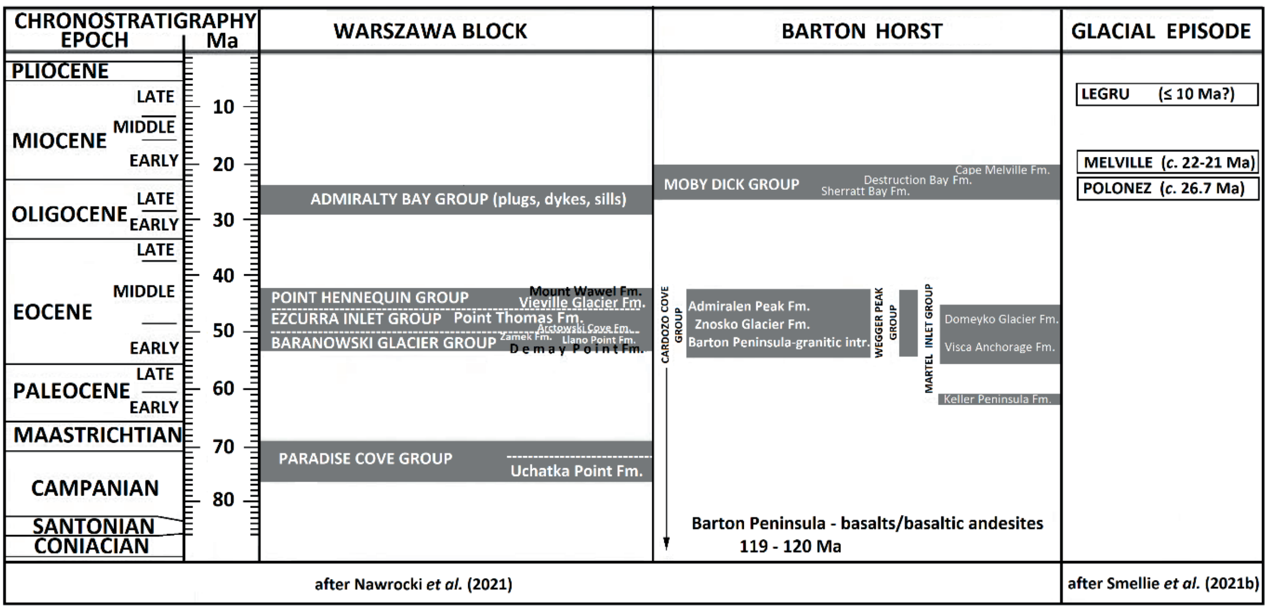

Fig. 2. Stratigraphic position of analyzed rock series and glacial episodes after Nawrocki *et al.* (2021) and Smellie *et al.* (2021b), simplified.

Table 1. Energy window characteristics of the GR-320 gamma-ray spectrometer.

Band	Radionuclide peak	Channels	Energy window (keV)	Sensitivity
RO/2	^{40}K 1460 keV	109–122	1370–1570	0.661 cps/%
RO/3	^{214}Bi 1760 keV	129–142	1660–2810	0.067 cps/ppm
RO/4	^{214}Tl 2620 keV	179–204	2410–2810	0.025 cps/ppm

Table 2. Results obtained for selected rock types.

Lithology-Stratigraphy-Area	K (%)	eU (ppm)	eTh (ppm)	eTh/eU	eTh/K 10^{-4}	eU/K 10^{-4}
Basalt, PTF, ASA	0.7±0.1	0.3±0.1	1.2±0.3	4.4	1.8	0.4
Agglomerate, PTF, HCA	1.2±0.1	1.1±0.1	4.5±0.1	4.0	3.6	0.9
Beach, Recent, ASA	1.3±0.1	1.5±0.1	4.9±0.3	3.3	3.7	1.1
Andesite, various	1.7±0.1	1.7±0.1	6.2±0.4	3.7	3.6	1.0
Ezcurra Fault zone, DIA	4.1±0.4	1.6±0.1	5.7±0.5	3.6	1.4	0.4
Lode, WPG, KPA	0.2±0.2	2.8±0.1	12.7±0.4	4.5	63.2	14.2
Granodiorite drop-stone, WPG?, ASA	3.4	4.7	18.4	3.9	5.4	1.4
Paleosol, PTF, CA	0.7	1.5	4.9	3.3	7.0	2.14
Rusty andesite, CCG, PHA	0.9	1.4	5.6	4.0	6.2	1.56
PTF- Point Thomas Formation, CCG - Cardozo Cove Group, WPG - Wegger Peak Group, KPF - Keller Peninsula Formation (Martel Inlet Group), VAF - Visca Anchorage Formation (Martel Inlet Group)						
ASA - Arctowski Station area, CA - Cytadela area, DIA - Dufayel Island area, HCA - Hervé Cove area, KPA - Keller Peninsula area, PHA - Pond Hill area						

Fig. 2 present the stratigraphic position of the analyzed rocks based on results of Nawrocki *et al.* (2010, 2011, 2021) and Smellie *et al.* (2021a, 2021b). The statistical parameters of the obtained results are presented in the upper part of Table 3, and their frequency distribution is presented in Fig. 3.

The most unimodal pattern can be observed in the case of K distribution. Its content varies in the range of 0–5.8 %. The mean value is 1.11%, higher than the modal value of 0.6–0.7%, as shown in Fig. 3. The asymmetry of the distribution is primarily due to a few measurements with increased K content (4–5%). U content varies between 0 and 4.7 ppm. The mean value is 1.18 ppm, but Fig. 3 shows three distinct groups with modal values: 0.2–0.3, 1.1–1.2, and 2.7–2.8. Th content varies between 0.6 and 18.4, with a mean value of 4.59. In this case, Fig. 3 shows three distinct groups with modal values 1.0–1.5, 4.5–5.0, and 11.5–12.0.

Results obtained for some characteristic rock types and localities are shown in Table 2. The mean values are joined with the standard error. Single measurements are presented without error data. The lowest content of all three radionuclides (Table 2) has been measured in the basalts of the Point Thomas Formation (PTF) in the Arctowski Station area. Significantly greater values have been obtained for andesites of various localities and stratigraphic positions. Basalts and andesites data form two main groups in Fig. 3 of lowermost and slightly higher eU and eTh contents, respectively.

Discussion

The obtained results are generally lower than those of Evangelista and Pereira (2002), but fit much better to the published data of Godoy *et al.* (1998) and Wedepohl (1969), see Table 3. Similarly, is the range of results published for rocks of island arc areas (Larsen and Gottfried 1960; Gottfried *et al.* 1963; Heier and Rogers 1963; Heier and Carter 1964; Heier *et al.* 1964; Rogers and Donelly 1966; Tatsumoto *et al.* 1965).

Intermediate values between basalts and andesites data have been measured for PTF agglomerate in the

Table 3. Statistical parameters of K, eU, and eTh concentrations.

Statistical parameters of the obtained results	K (%)	eU (ppm)	eTh (ppm)
Mean	1.11	1.18	4.59
Median	0.90	1.20	4.50
Standard deviation	0.75	0.78	2.95
Standard error	0.05	0.05	0.19
Kurtosis	1.83	1.20	2.99
Skewness	2.80	0.84	1.37
Minimum	0.00	0.00	0.60
Maximum	5.80	4.70	18.40
Number of measurements	253	253	253
Mean values calculated for published data:			
other South Shetland islands (Godoy *et al.* 1998)	0.2	0.3	1.3
King George Island soil (Godoy *et al.* 1998)	1.3	1.5	5.3
King George marine sediments (Godoy *et al.* 1998)	1.7	1.1	3.7
Admiralty Bay soil (Evangelista and Pereira 2002)	-	3.34	14.12
Earth - basalts and andesites (Wedepohl 1969)	-	0.5	1.6

Hervé Cove area. It is quite reasonable, taking into account the fact that it is a mixture of basalt and andesitic pebbles, cobbles, and boulders up to 2 m in diameter deposited as lahar sediment (Birkenmajer 2001). Similar is the explanation of values obtained for the Recent cobble beach in the Arctowski Station area (Table 2).

Changes in the K, U, and Th contents of the analyzed rocks are positively correlated. Pearson's r correlation coefficients calculated for all combinations of analyzed radionuclides are presented in Table 4. The best correlation exists between U and Th. The eU-eTh correlation coefficient for all analyzed rocks of King George Island is much higher than the same coefficient, see the last row in Table 4, calculated using published data for basalts and andesites from other parts of the world (Larsen and Gottfried 1960; Gottfried *et al.* 1963; Heier and Rogers 1963; Heier and Carter 1964; Heier *et al.* 1964; Rogers and Donelly 1966; Tatsumoto *et al.* 1965; Wedepohl 1969).

The high correlation between U and Th indicates that the eTh/eU ratio is stable. The data presented in Table 2 show that the eTh/eU ratio reaches its extreme values of 3.3 and 4.5 for paleosol and lode, respectively. Considering the error range in the case of basalts and agglomerate, most obtained results are within the range of average values of 2.55–3.92 of normal mid-ocean ridge basalt and ocean island basalts published by Plant *et al.* (1999). The only, evidently outstanding value is 4.5 in the case of the lode from the Keller Peninsula (Table 2). The conclusion of Evangelista and Pereira (2002) that a relatively large spread of the normalized correlation between U and Th suggests that the region of investigation may have been subjected to "geochemical weathering processes that are probably more intensive than the reported results of Dreschhoff for Terra de Victoria" seems to be no longer valid. Fruitful studies of Th/U/K ratios, performed by Zeller and Dreschhoff (1980, 1986, 1987, 1990) in the Transantarctic Mountains, proved that the authors were able to investigate the chemical weathering of the pre-Beacon surface of Cambrian-Devonian age. The application of their ideas and techniques in areas of much younger geology, such as King George Island, requires careful geological examination, as periods of intensive chemical weathering were relatively unique during the Cainozoic of Antarctica.

The correlation of U and Th with K is very weak. This fact is illustrated in Fig. 4, which shows the interrelation between K, eU, and eTh. The well-defined central linear trend among all three radionuclides (marked with a black line) is disturbed by two groups of measurements, including Ezcurra Fault zone and Keller Peninsula lode (Fig. 4). Removing these anomalous measurements results in a significant increase of eU-K and eTh-K Pearson's coefficient, shown by values in brackets in Table 4, without change of Th-U correlation.

The first group of anomalous measurements is connected with increased K content combined with moderate eTh and eU results from the Ezcurra Fault zone in Dufayel Island area (Fig. 1 and Table 2) and their shifting right from the linear trend of Fig. 4. Anomalous results have been obtained within the secondary faults of the Ezcurra Fault zone developed in andesites of the Cardozo Cove Group. Two samples from this location described by Birkenmajer *et al.* (1991) are "extensively altered; only relicts

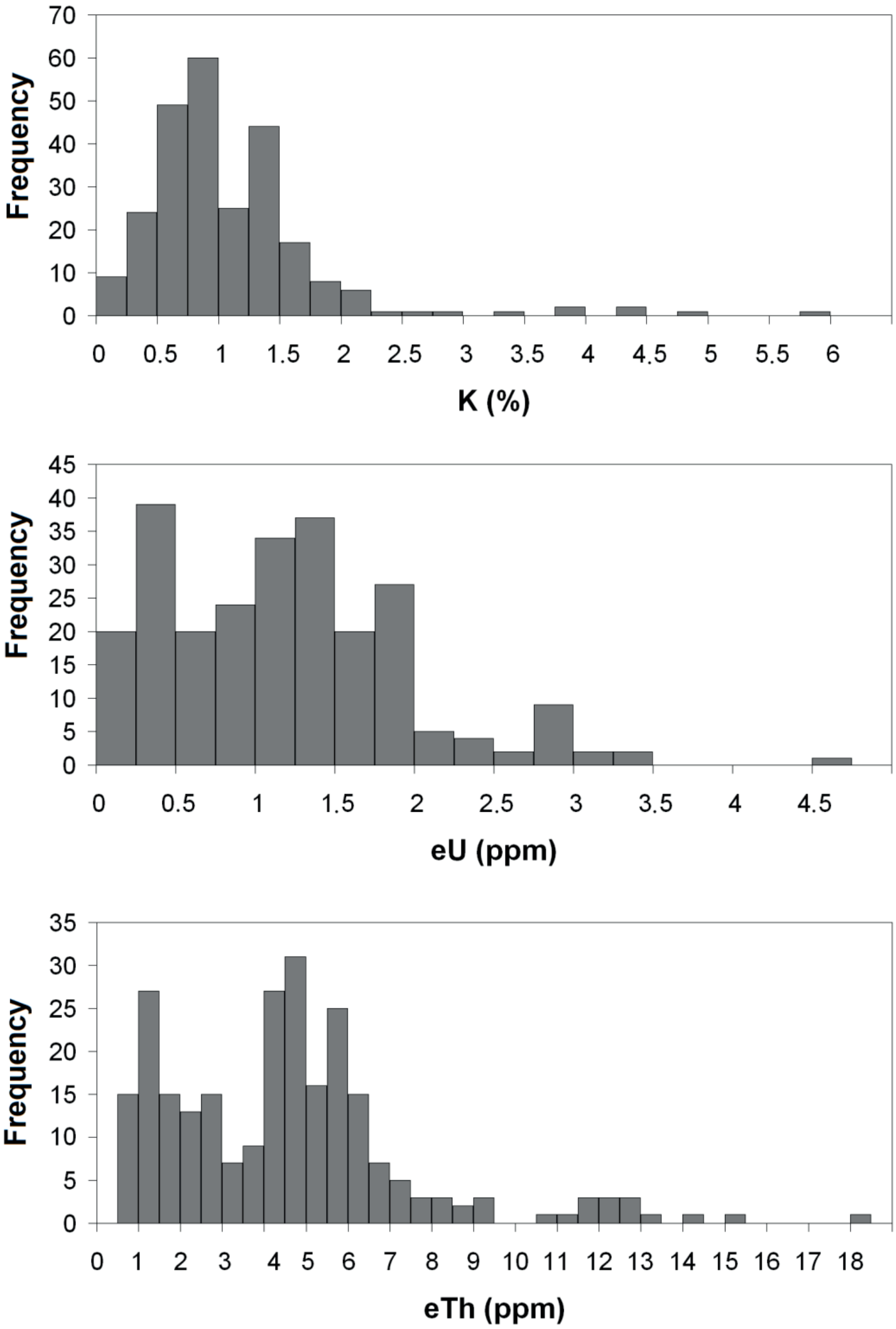

Fig. 3. Frequency distribution of K, eU, and eTh content in the studied rocks.

Table 4. Correlation coefficients K, eU, eTh for the analyzed rocks and published global data on basalts and andesites (in parentheses, the values obtained after excluding the anomalous measurements from the Keller Peninsula lode and Ezcurra Fault zone).

	Pearson's ***r***
Th-U	0.89
U-K	0.29 (0.70)
Th-K	0.21 (0.73)
Th-U world	0.84.

of plagioclases completely transformed to saussurite, and ghosts of chloritized mafic phases can be observed in a strongly altered groundmass containing small calcite veinlets". Their K content, determined by X-ray fluorescence on pure powder rock pellets using the method of Franzini *et al.* (1972), was 3.8 and 4.2, which is in excellent agreement with the results of gamma radiation spectrometry. The alteration described by Birkenmajer *et al.* (1991) is typical of low-temperature, high-pressure fault zone transformations. Geochemical changes in this case are limited to K enrichment, and the eTh/eU ratio remains similar to other localities.

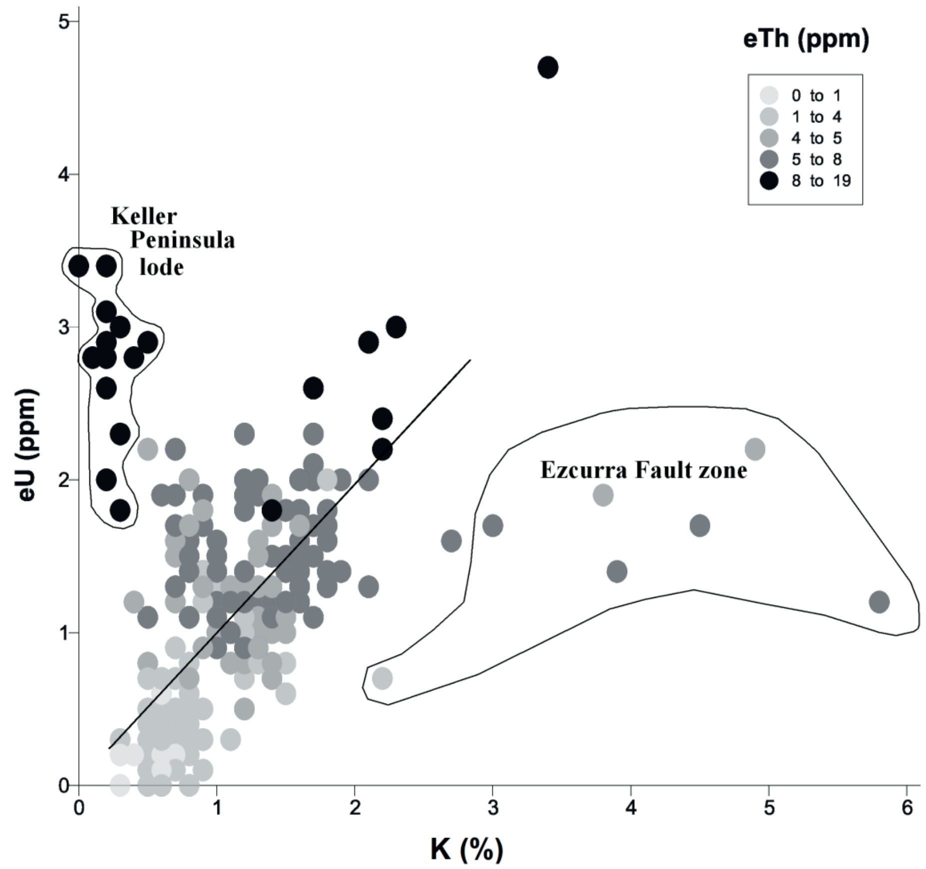

Fig. 4. Interrelation among eU, K, and eTh contents of rocks in analyzed sites.

The second group of anomalous results, which is shifted upward from the linear trend of Fig. 4, has been obtained in the area of the Keller Peninsula (Fig. 5 and Table 2). Anomalous results of low K and high Th have been obtained on the outcrops of one of the numerous lodes with pyrite mineralization (Table 2), discovered by Ferguson (1922), who explained their origin as a result of the replacement of andesites. Hawkes (1961) interpreted them as a result of metasomatic alteration of Jurassic lavas by Andean intrusions. Barton (1961, 1965) described their connection with fault zones, while Mueller (1963) and Littlefair (1978) pointed out the role of fumarolic-solfatara alteration. Birkenmajer (1979, 1980a, 1980b, 1980c, 1982) described them as a result of Andean pluton intrusion (Wegger Peak Group-WPG) into the Martel Inlet Group volcanites, locally reworked by younger fault zones.

A detailed petrologic study of these veins has been published by Paulo and Rubinowski (1987), who described them as bleached pyrite-bearing zones. The results of their standard chemical analyses, where K content does not exceed 0.17%, fit well with the gamma-ray spectrometric results of the present authors. Geochemical change, in this case, is connected with significant eTh enrichment, significant K depletion, and a slight increase in eU content (Table 2). Since these changes are in inversed relation with the geochemical mobility of these elements, the most reasonable explanation is that, leaching of primary material which resulted in K depletion (Table 2) was connected with significant mass loss and relative, inert Th enrichment in the siliceous residuum.

The only two cases of increased eTh/K ratio (Table 2), which may be interpreted as a result of chemical weathering, have been found within rusty zones in andesites of the Cardozo Cove Group on the slopes of Pond Hill and paleosol of the Point Thomas Formation on the slopes of Cytadela Hill. Exogenous chemical weathering of pyrite-bearing zones, caused by acidification resulting from the oxidative decomposition of iron sulfide, led to a color change to green, rusty, and creamy white (Birkenmajer 1982). Chemical weathering must have been especially active in the Eocene when fossil plant horizons and lignite seams developed. Not only paleobotanic studies (Cortemiglia *et al.* 1981; Stuchlik 1981; Jagmin 1987; Birkenmajer and Zastawniak 1989a, 1989b) but also mineralogical evidence (Birkenmajer and Łydka 1990) supports the existence of a warm climate at the Eocene-Oligocene boundary. Taking into account the stratigraphic position of the Point Thomas Formation (Fig. 2), the chemical weathering might have started in the warm and humid Eocene climate preceding the Polonez Glaciation (Birkenmajer 2001) and continued in temperate climate periods before Melville and Legru glaciations (Barton 1964; Zastawniak 1981; Zastawniak *et al.* 1985; Birkenmajer and Zastawniak 1989a; Smellie *et al.* 2021b).

The eTh/eU ratio in the case of paleosol (Table 2) is the lowest one, but still within the average range of Plant *et al.*

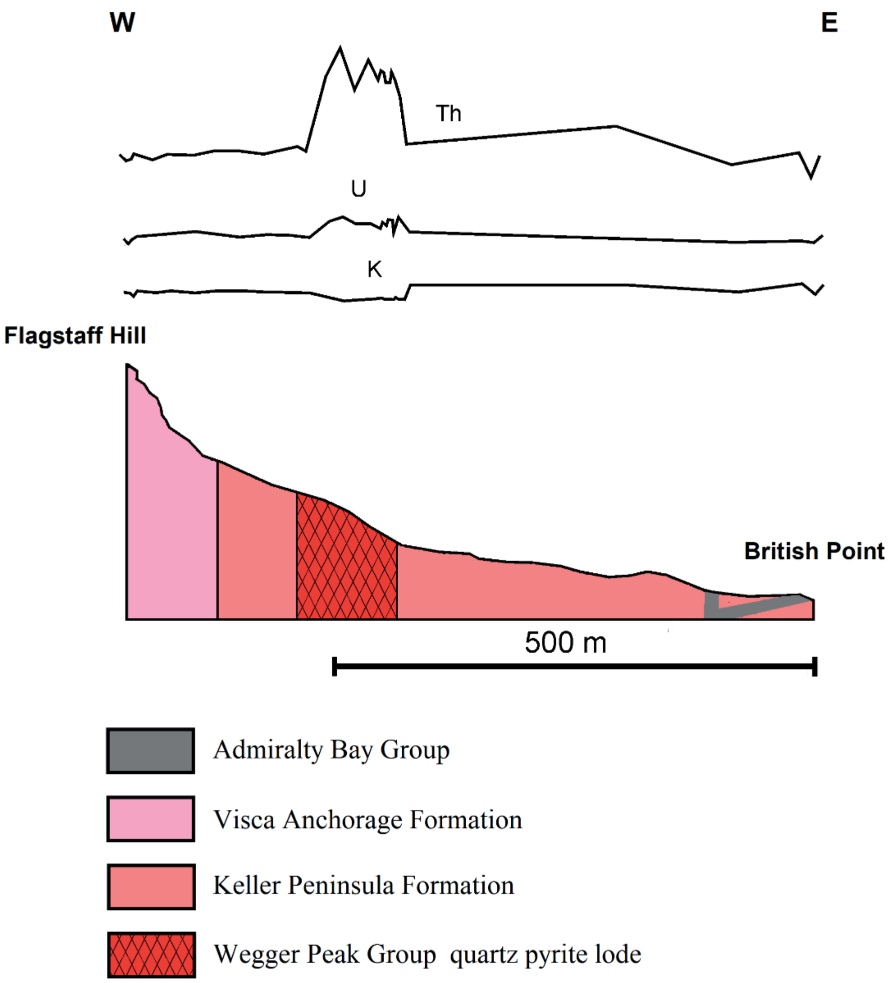

Fig. 5. Radiometric profile of the Keller Peninsula mineral vein. Simplified geology after Birkenmajer (1982) and Nawrocki *et al.* (2021).

(1999) for mid-ocean ridge basalt and ocean island basalts. Its change due to weathering seems to be much smaller than natural diversification among various rock types. This lowermost eTh/eU ratio value may be connected with the sorption of leached U by the organic matter of silicified wood. The high eTh/K ratio of paleosol (Table 2) is similar to the results of Hampson *et al.* (2005) from kaolinitic zones (paleosol formation) in a fluvial sequence of the Bookcliffs in Utah and is the result of chemical weathering. It is limited to a very thin and unique stratigraphic horizon that developed in a warmer climate.

Increased eTh/K ratio in Eocene paleosol and rusty andesite of the Cardozo Cove Group, may be interpreted as a result of chemical weathering during warm or temperate climates of Paleogene interglacials. Th/K ratio of rock seems to be a quite sensitive indicator of chemical weathering and leaching due to contrast of geochemical mobility of these two elements. In contrast, the eTh/eU ratio is too stable in the analyzed rocks to be a good indicator of chemical weathering. Its application in this study area requires careful geological investigation, as periods of intense chemical weathering were exceptional in the Cenozoic of Antarctica.

Conclusions

The K content and eTh/K ratio of the analyzed rocks change widely. The most significant changes in K content are limited to two localities with very special geological settings: the Ezcurra fault zone and the Keller Peninsula lode. Endogenic metasomatic processes connected with volcanic and tectonic activity can fully explain these changes. Their limited spatial extent excludes climatic factors and supports a connection with local geology.

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