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Validation of a database of mean uranium, thorium and potassium concentrations in rock samples of Portuguese geological units, generated of literature data

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# **Validation of a database of mean uranium, thorium and potassium concentrations in rock samples of Portuguese geological units, generated of literature data**

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## **Highlights**

- A database of U, Th and K concentrations in rocks of Portugal established;
- Mean U, Th and K concentrations in rock per geological unit calculated;
- Validation by comparison of calculated to measured mean terrestrial dose rate;
- Investigation of variance components traceable to the geochemical analysis;
- Use of scattered local geochemical data to establish large scale maps enabled.

1        **Abstract**

2        The European Atlas of Natural Radiation has been under development for the past  
3        15 years. Among the lacunae of the Atlas are maps of U, Th and K concentrations in  
4        rocks due to lack of European-wide geochemical surveys of bedrock units. The objective  
5        of this paper is to investigate the usability of scattered geochemical data of rock samples  
6        for large-scale mapping of U, Th and K concentrations in geological units. For this  
7        purpose, geochemical data were compiled from grey literature sources to produce a  
8        geochemical database (LIT database) that includes 2817 results of U, Th and K  
9        concentrations measured in rock samples.

10       Given the methodical heterogeneity within LIT database, the influence of the  
11       geochemical analysis techniques was assessed through a three-way analysis of  
12       variance (ANOVA) using geological units, geochemical analysis techniques and loss on  
13       ignition (LOI) as categorical variables. The percentage of variation explained by  
14       geological factors was 53 % for U, 38 % for Th, 50% for K and 51 % for the terrestrial  
15       gamma dose rate (TGDR), while the percentage of variation explained by the  
16       geochemical analysis techniques and LOI was generally lower than 5 %. The geological  
17       factors were thereby the main source of variability in the data, followed by the error  
18       component which can be assumed to represent true spatial variability of geochemical  
19       concentrations, hence, variability within the geological units. The reliability of LIT  
20       database between the different geochemical analysis techniques was investigated  
21       through pairwise comparison of the least square (LS) means computed through the  
22       three-way ANOVA for each geochemical analysis technique. The results show an  
23       agreement of the LS means between the different geochemical analysis techniques,  
24       which indicates that LIT database can be considered consistent within itself, thus,  
25       reliable.

26       In order to validate the usability of literature data, the TGDR calculated from literature  
27       data ( $TGDR_{calc}$ ) was compared to the measured TGDR ( $TGDR_{obs}$ ) displayed in the

28 Radiometric Map of Portugal (RMP) using geological units as reference units. The  
29 correlation between  $TGDR_{calc}$  and  $TGDR_{obs}$  was highly significant ( $p < 0.001$ ) and the  
30 results of a paired sample t-test and Wilcoxon median tests indicate that the differences  
31 between the arithmetic means of  $TGDR_{calc}$  and  $TGDR_{obs}$  were not statistically significant  
32 ( $p = 0.126$  and  $p = 0.14$ , respectively). Distributions of  $TGDR_{calc}$  and  $TGDR_{obs}$  are  
33 seemingly equal according to the Kolmogorov-Smirnov and Anderson-Darling tests. The  
34 pooled standard deviation (SD) of the RMP dataset was set as criterion to assess the  
35 discrepancy between the arithmetic means of the RMP and LIT databases. About 14 out  
36 of 26 geological units are located within one pooled SD around  $TGDR_{obs}$  and only 1 out  
37 of 26 units is located outside 2 pooled SDs. Systematic discrepancies were observed for  
38 sedimentary rocks possibly due to the lack of representativeness of the data.  
39 Nonetheless, the compatibility of the RMP and LIT databases can be considered  
40 acceptable, which implies that the estimation of the contents of terrestrial radionuclides  
41 using literature data for large-scale mapping of U, Th and K contents in geological units  
42 is reasonable.

43 **Keywords:** Data compilation; Geochemical analysis techniques; Terrestrial  
44 radionuclides; Terrestrial gamma dose rate; Geological units.

## 45 **1. Introduction**

### 46 **1.1 Motivation**

47 A long-term European project, under way for about 15 years, is the European Atlas  
48 of Natural Radiation, of which web and printed versions exist (Cinelli et al., 2019; EC,  
49 2019). The Atlas is however incomplete, as certain compartments and environmental  
50 media are missing or fragmentary due to lack of data. Among these lacunae are maps  
51 of concentrations of naturally occurring radionuclides, namely isotopes of U, Th and K,  
52 in rock. While Europe-wide geochemical surveys of *agricultural and pasture soil* including  
53 these radionuclides exist (FOREGS and GEMAS, maps and references in the Atlas),  
54 literature data of U, Th and K in *rock* are scattered and methodically highly

55 heterogeneous. Therefore, creating Europe-wide maps from such data appeared  
56 questionable. The alternative, on the other hand, namely creating a methodically  
57 homogeneous geochemical database of European rocks from scratch, would pose a  
58 tremendous task which is beyond realistic feasibility. Therefore, it has been concluded  
59 by the board of experts of the Atlas (to which the authors of this paper belong) to explore  
60 the feasibility of maps of U, Th and K concentrations in rocks, derived from literature  
61 data. This requires, as very first step, to investigate whether literature data can be used  
62 to this end, observing standards of accuracy and precision, given methodical  
63 heterogeneity. The objective of this paper is thus, investigating the usability of scattered  
64 geochemical literature data for large-scale (in perspective, European scale) mapping. As  
65 test region for this study we use the territory of Portugal and as geochemical data  
66 sources, local studies on rock geochemistry performed in Portugal.

## 67 **1.2 Geochemical surveys**

68 Local surveys of geochemistry are profusely available in scientific literature. Large  
69 scale surveys are, however, scarce because the generation of a truly representative  
70 sample of the media is difficult with limited resources, and sampling of the entire  
71 population of landscape units is impractical and costly.

72 UNESCO initiated the first attempt to create a geochemical map of the world in 1988  
73 through the International Geochemical Mapping Project (IGCP 259) (Xuejing and  
74 Hangxin, 2001). The main goal of the project was to estimate the concentration of several  
75 elements at a global scale with particular focus on ore pathfinders and radioelements  
76 (Darnley 1995, Darnley et al. 1995, Xuejing and Hangxin, 2001). The project intended to  
77 overcome the lack of numerical equivalence between compiled datasets that are subject  
78 to different sampling and analytical techniques as well as different levels of quality control  
79 (Darnley et al. 1995). However, the compilation and normalization of existing datasets  
80 required that data at multiple spatial scales be interlinked (Darnley 1995, Darnley et al.  
81 1995).

82 A compilation of geochemical data hardly fulfills the assumptions of statistical  
83 inference (ex. the sampling must be random, unbiased and designed to adequately  
84 represent the population, Crumbling, 2001; Ramsey and Hewitt, 2005) as the data have  
85 been acquired for different purposes, at different scales and with different methods.  
86 However, every geochemical survey must rely on a suitable knowledge of the geology  
87 of the surveyed area (e.g. Rollinson, 1993). Thus, geochemical surveys can be  
88 presumed to be representative of the geological units sampled.

89 The estimation of the contents of terrestrial radionuclides, radon, gamma dose rate  
90 and air-absorbed dose rate using geochemical data has been attempted at both a local  
91 (e.g. Ielsch et al. 2010; Ye and Gan, 2013) and regional scale (e.g. Appleton et al. 2008;  
92 Beamish 2014; Ielsch et al. 2017; Marsac et al. 2016). However, the authors often resort  
93 to geochemical data available in national databases that have been acquired with  
94 consistent analytical techniques and quality control procedures.

### 95 **1.3 Heterogeneity of literature data**

96 Heterogeneity of literature data occurs on several levels. (1) The data come from  
97 regional or local studies of certain rock formations or types with certain geochemical  
98 objectives, such as mineral exploration, hydrological investigations etc. While  
99 representativeness for the study area can be assumed, it is unclear to which extent this  
100 applies to an entire lithological class.

101 (2) Sampling and sample geo-referencing methods are heterogeneous. For example,  
102 many literature sources do not quote locations of individual samples, but only regions of  
103 origin. Sometimes, no individual but only aggregated results are given, e.g. arithmetic  
104 mean concentrations and standard deviations. This circumstance inhibits the use of  
105 geostatistical tools.

106 (3) Finally, geochemical analytic methodology is heterogeneous. Comparability of  
107 results between different studies is therefore not given a priori. Both radiological and  
108 geochemical data can be used to evaluate U, Th and K concentrations and distributions



109 (IAEA, 2010). Radiological data refers to radionuclide concentrations determined  
110 through alpha- or gamma-spectrometry while geochemical data may be acquired  
111 through various geochemical analysis techniques (hereafter referred to as analytical  
112 techniques), such as X-Ray Fluorescence (XRF), Induced Coupled Plasma  
113 Spectrometry (ICP) or Instrumental Neutron Activation Analysis (INAA), among others.

114 It should be stressed again that heterogeneity between studies does not mean to  
115 invalidate the individual studies, but it is an obstacle to using them as input for maps  
116 beyond the scale of the individual ones. Investigating whether it is still possible, is the  
117 subject of this paper.

#### 118 **1.4 Variability of true and measured geochemical concentrations**

119 The idea underlying the analysis performed in this study is that there are essentially  
120 three components of variability:

121 (A) Variability between geological units, leading to variance between geochemical  
122 concentrations of samples (or studies, representing aggregated samples) located in  
123 different geological units, as this factor is assumed to control geochemical concentration;  
124 this component represents *true spatial variability* of the investigated variable  
125 (geochemical concentration).

126 (B) Variance due to different analytical techniques, as it is known that different  
127 techniques applied to the same sample, i.e. same true concentration, can lead to  
128 systematically different results (e.g. Rollinson, 1993). This component refers to the  
129 *observation process* of the variable, not to its true variability over a geological unit.

130 (C) Variance due to lack of spatial representativeness: assume two studies using  
131 same analytical technique, performed in different parts of the same geological unit. Due  
132 to true spatial variability within geological units, the results will still be different, in general,  
133 in spite of variance sources (1) and (2) excluded. This effect can only be avoided  
134 (asymptotically with sample size) if the sample covers the domain (geological unit)  
135 representatively. (Representativeness means that the statistical distribution of the  
136 sample is asymptotically the same as the true distribution of the investigated variable,

137 here geochemical concentration.) This component refers to availability of data within a  
138 geological unit and their aggregation within and can be understood as sampling effect  
139 induced by true variability, component (1). Getting hold of this component is particularly  
140 difficult.

### 141 **1.5 Validation of usability of literature data**

142 The tool to validate the usability of geochemical literature data is comparison of  
143 terrestrial gamma dose rate (TGDR) calculated from geochemical concentrations with  
144 measured ones. Coincidence within tolerable deviation is a *necessary condition* of the  
145 validity of regional estimates from scattered literature data. The strategy is therefore, 1)  
146 estimate mean U, Th, K concentrations of geological units from literature data; 2)  
147 calculate the mean TGDR per unit and 3) compare these calculated with measured mean  
148 TGDR of the same units. This is possible for Portugal as test region, as a fine-grained  
149 TGDR map of Portugal is available. If it is proven that creating maps of rock geochemistry  
150 from scattered literature data is possible in Portugal, it can plausibly be assumed that  
151 this is possible also in other regions.

### 152 **1.6 Organization of this paper**

153 In the methods section (2), we first establish the geological classification scheme  
154 which leads to the set of geological units in which geochemical literature data are  
155 aggregated and mean dose rates are calculated. This is followed by a presentation of  
156 the compiled geochemical database (section 2.2) and the TGDR map of Portugal (2.3).  
157 In section 2.4, the conversion of geochemical concentration into TGDR is reviewed and  
158 in 2.5, we explain statistical techniques.

159 The results section (3) is firstly devoted to analyzing the sources of variance A and B  
160 (geology and analytical techniques) in section 3.1. Then an attempt is made to remove  
161 source B, to assess the effect of lack of representativeness at least qualitatively (section  
162 3.2). The validation analysis through terrestrial dose rate is shown in section 3.3.

## 163 **2. Materials and Methods**

## 164 **2.1. Geological classification scheme**

165 OneGeology-Europe (OGE) data were chosen as the geological map to be used as  
166 reference because they are available at the European level (at [http://www.onegeology-](http://www.onegeology-europe.org/)  
167 [europe.org/](http://www.onegeology-europe.org/)), providing access to geological maps at the scale of 1:1,000,000 (Baker and  
168 Jackson, 2010). The OGE map for Portugal was developed through collaboration with  
169 the Portuguese National Laboratory of Energy and Geology (LNEG), and comprises  
170 three distinct layers, namely lithology, age and geological structures. The code  
171 classification scheme of the OGE map is equivalent to the code classification scheme of  
172 the Portuguese Geological Map (PGM) at the scale of 1:1,000,000 published in 2010 by  
173 LNEG, and available at <http://geoportal.lneg.pt/geoportal/mapas/index.html>. The OGE  
174 classification comprises 76 geological units in mainland Portugal and 11 units in the  
175 Madeira and Azores archipelagos. A summary description of the geological units studied  
176 in the present work is presented in Table 1 according to Baker and Jackson (2010). A  
177 simplified geological map is presented in Figure 1a.

## 178 **2.2. The Radiometric Map of Portugal**

179 The Radiometric Map of Portugal (RMP, Figure 1b, Batista et al. 2013) displays the  
180 terrestrial gamma dose rate in mainland Portugal and is further denoted as  $TGDR_{obs}$ . A  
181 raster layer is available freely at <http://geoportal.lneg.pt/geoportal/mapas/index.html>. The  
182 data were acquired with hand-carried, vehicle-borne and air-borne equipment from 1955  
183 onwards (Batista et al. 2012). The equipment used were calibrated either using sources  
184 of terrestrial radionuclides with known concentrations of U, Th and K or by measuring  
185 the same sites with different equipment (Batista et al. 2012; Grasty et al. 1993). The first  
186 radiometric maps of Portugal were produced based on 733,201 measurement points by  
187 linear interpolation with the minimum curvature method. The estimation grid consists of  
188 250 m × 250 m cells. A minimum of 10 measurement points was defined necessary for  
189 estimation on each grid point, to assure reasonable statistical reliability. This required a  
190 search radius of 10 km around every grid point to make sure that the required minimum

191 number of observations could be achieved (Torres et al. 1997; Saraiva et al. 1998). The  
192 updated RMP was produced based on 841,440 measurement points following the  
193 acquisition of new data to fill the gaps in the previously developed maps (Batista et al.  
194 2012; Batista et al. 2013).

195 The cell size of the RMP constitutes an important limitation for geological units that  
196 have a large perimeter to area ratio, such as vein-type rocks (perimeter to area ratio  
197 generally higher than 5). The width of the f1 and f2 (basic vein-type rocks) and qz (quartz  
198 veins) units is generally lower than 500 m. Given the RMP cell size of 250 m and the  
199 search radius of 10 km used to reach a minimum of 10 measurement points for  
200 interpolation, the  $TGDR_{obs}$  for these units is highly influenced by the  $TGDR_{obs}$  of  
201 neighboring geological units. Therefore, the  $TGDR_{obs}$  of vein-type rocks displayed in the  
202 RMP is unreliable. For this reason, vein-type geological units were excluded from the  
203 analysis. This is unfortunate because these units often contain concentrations of  
204 radioactive elements strongly different from their surroundings and should therefore be  
205 given particular attention for radioprotection reasons. The methodical alternative would  
206 have been a different interpolation scheme for such units, but the original raw TGDR  
207 data which would be necessary for this, are not available to us.

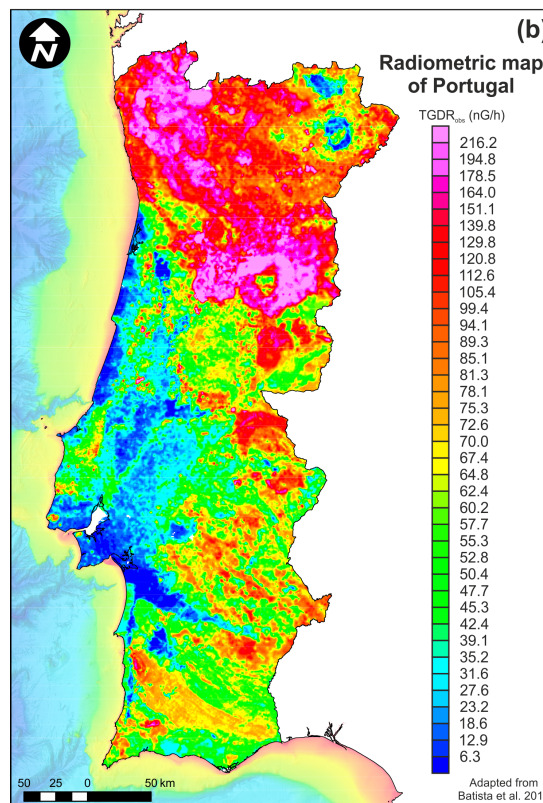
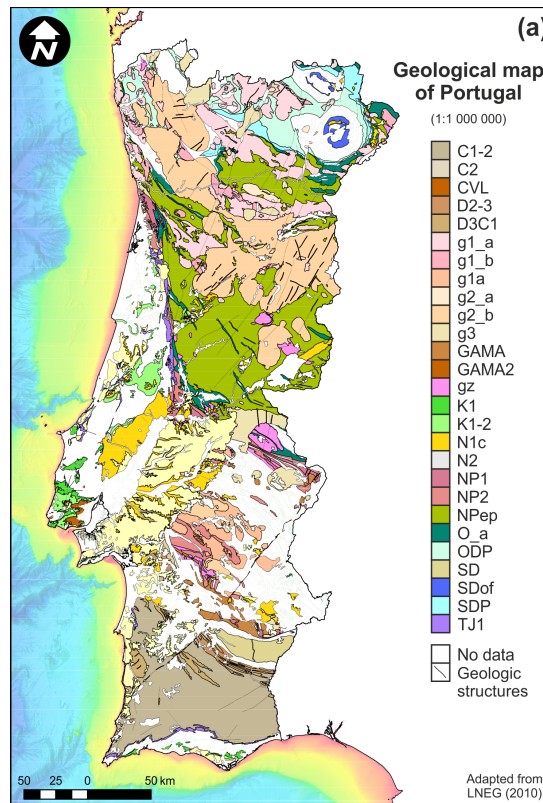
208 The lack of access to the original raw TGDR data inhibits the assessment of the  
209 influence of the presence of unconsolidated overburden and soil units in the measured  
210 TGDR. However, the RMP was primarily developed for the purpose of mineral  
211 exploration (Batista et al. 2012; Grasty et al. 1993, Saraiva et al. 1998; Tauchid and  
212 Grasty, 2002; Torres and Grasty, 1993). Sampling was focused on the main geological  
213 units, as surveys were carried preferentially over crystalline areas, followed by the  
214 Mesozoic and Cenozoic sedimentary basins (Torres and Grasty, 1993). Data from car,  
215 foot and air-borne surveys were calibrated through measurement of selected areas of  
216 exposed bedrock of the main geological units (Torres and Grasty, 1993). The RMP thus  
217 reflects the geological character of Portugal (Batista et al. 2012; Grasty et al. 1993,

218 Saraiva et al. 1998; Tauchid and Grasty, 2002; Torres and Grasty, 1993). Thus, the RMP  
219 data can be presumed to be representative of the geological units.

220 **Table 1.** Summary description of the geological units studied according to OGE data  
 221 (Baker and Johnson, 2010).

Group	OGE unit	Perimeter/Area ratio	Lower age	Upper age	Predominant Lithology	Subordinate lithology
Sedimentary rocks	N2	0.9	Pliocene	Pliocene	Sandstone	Conglomerate, Siltstone
	N1c	1.2	Miocene	Miocene	Sandstone	Conglomerate, Siltstone
	K1-2	1.4	Cretaceous	Cretaceous	Limestone	Sandstone, Impure carbonate sedimentary rock, Dolomite
	K1	1.8	Early/Lower Cretaceous	Early/Lower Cretaceous	Sandstone	Limestone, Impure carbonate sedimentary rock, Dolomite
	TJ1	2.2	Late/Upper Triassic	Middle Jurassic	Sandstone	Claystone, Evaporite, Limestone
	C2	3.6	Pennsylvanian	Pennsylvanian	Conglomerate	Sandstone, Claystone, Coal
Metamorphic rocks	C1-2	0.3	Carboniferous	Carboniferous	Phyllite	-
	D2-3	1.9	Middle Devonian	Late/Upper Devonian	Phyllite	Quartzite
	SDof	1.2	Silurian	Devonian	Amphibolite	Schist, Fine grained igneous rock
	SD	0.4	Silurian	Early/Lower Devonian	Phyllite	-
	SDP	0.7	Ordovician	Devonian	Phyllite	-
	ODP	0.8	Ordovician	Devonian	Quartzite	-
	O_a	1.6	Ordovician	Ordovician	Quartzite	Phyllite
	gz	1.1	Furongian	Middle Ordovician	Orthogneiss	Granite, Diorite
	Npep	0.6	Ediacarian	Cambrian	Phyllite	-
	NP2	1.4	Neoproterozoic	Neoproterozoic	Phyllite	-
NP1	1.6	Neoproterozoic	Neoproterozoic	Schist	Gneiss, Migmatite	
Igneous rocks	GAMA2	3.8	Late/Upper Cretaceous	Late/Upper Cretaceous	Diorite	Gabbro
	CVL	2.2	Late/Upper Cretaceous	Late/Upper Cretaceous	Basalt	Pyroclastic material, Gabbro,
	g1_b	1.1	Pennsylvanian	Cisuralian	Granite	-
	g2_b	0.3	Pennsylvanian	Cisuralian	Granite	-
	g3	0.7	Pennsylvanian	Cisuralian	Granite	-
	g1_a	0.7	Mississippian	Pennsylvanian	Granite	-
	g2_a	0.7	Mississippian	Pennsylvanian	Granite	-
	g1a	0.8	Late/Upper Devonian	Pennsylvanian	Tonalite	Granodiorite,
	GAMA	1.3	Late/Upper Devonian	Pennsylvanian	Gabbro	Anorthositic rock, Diorite
D3C1	1.4	Late/Upper Devonian	Pennsylvanian	Fine grained igneous rock	Phyllite	

\*The perimeter to area ratio was computed as the ratio between the sum of the perimeter to the sum of the area of all polygons of the geological unit.



222

223 **Figure 1. a)** Geological units studied. Adapted from the Geological Map of Portugal at  
 224 the scale of 1:1,000,000 (LNEG, 2010). **b)** Radiometric Map of Portugal (Batista et al.  
 225 2013). Bathymetry was retrieved from [http://portal.emodnet-bathymetry.eu/mean-depth-](http://portal.emodnet-bathymetry.eu/mean-depth-full-coverage)  
 226 [full-coverage](http://portal.emodnet-bathymetry.eu/mean-depth-full-coverage) (last accessed in February 2017).

### 227        **2.3. The geochemical database**

228        A compilation of geochemical surveys available in mainland Portugal was carried out  
229        in order to produce a new geochemical database, called LIT to denote that it consists of  
230        literature data. The LIT database includes 2817 observations of U, Th and/or K  
231        concentrations measured in rock samples presumed to be representative of the  
232        geological units studied. The data included in the LIT database were retrieved from the  
233        references listed in table A1 (Supplementary material). All samples included in the LIT  
234        database were collected in areas of bedrock exposed on the earth surface excluding  
235        data retrieved from Cerejo (2013), Pinto (2014), Oliveira (2015) and Dinis et al. (2011)  
236        whom also studied rock samples retrieved from boreholes and/or underground galleries.  
237        Due to the inherent modifications caused by in-situ chemical and physical weathering,  
238        erosion and transport, among others, soil samples and unconsolidated materials are not  
239        considered representative of geological units, hence, were excluded from the analysis.  
240        Data for the Azores and Madeira archipelagos were not included in the database as the  
241        RMP only covers mainland Portugal (Batista et al. 2013).

242        Three types of samples were considered as observations: (1) individual analyses of  
243        rock samples; (2) analyses of composite samples (mixtures of several individual samples  
244        taken within an area, analyzed as one sample; the purpose is to reduce variability) and  
245        (3) averages of individual samples' analyses. Statistically, sample averages were treated  
246        as individual samples; (3) can be thought of as mathematical mixing in analogy to physical  
247        mixing in (2). The accurate location of the sampling sites of the observations are mostly  
248        unknown, inhibiting the use of geostatistical tools. This is because in earlier surveys, geo-  
249        referencing was not considered necessary. Due to this limitation, the regionalization of  
250        the available data was carried out using geological information.

251        Each observation of the LIT database was classified with a code of the geological  
252        classification scheme described in section 2.1. according to the location of the  
253        observations or the study area (when provided by the authors), or, in alternative, through



254 the lithologic, petrographic or paleontological features of the samples. A summary of the  
255 data available by geological unit (Table A1) and by analytical techniques (Table A2) are  
256 given as supplementary material.

257 For each observation of U, Th and/or K, the element concentration and analytical  
258 techniques were registered as provided by the authors. No corrections to U, Th and/or K  
259 concentrations were applied for loss on ignition (LOI) or the volatile content. A binary  
260 categorical variable for the computation of the effects of LOI in an Analysis of Variance  
261 (ANOVA) was defined. Each observation was classified as “TRUE” if the authors  
262 estimated LOI (and/or the volatile content) but did not correct the data for LOI and  
263 “FALSE” if the authors estimated and corrected the concentration of terrestrial  
264 radionuclides using LOI data.

265 Values reported as below the lower limit of detection (LLD) as well as zeroes were  
266 set to 0.65LLD or to an arbitrary small value in case the LLD was not provided by the  
267 authors. The overall percentage of values below the LLD was 6% for U, 4% for Th and  
268 less than 1% for K.

#### 269 **2.4 Calculation of terrestrial gamma dose rate TGDR**

270 U, Th and K concentration were converted into activity concentration using  
271 appropriate conversion factors (Table 2). The TGDR, further denoted as  $TGDR_{calc}$ , was  
272 computed from U, Th and K activity concentration (in Bq/kg) according to the following  
273 equation:

$$274 \quad TGDR_{calc} = 0.0417 \times {}^{40}K + 0.462 \times {}^{238}U + 0.604 \times {}^{232}Th \quad (1)$$

275 The dose conversion factors (in Bq/kg) of 0.0417, 0.462 and 0.604 were from  
276 UNSCEAR (2010). All observations were considered valid, regardless of the analytical  
277 technique used in the analysis, and representative of the geological unit sampled by the  
278 authors. The database of U, Th and K contents in activity concentration is presented as  
279 Supplementary material (Table B1).

280

**Table 2.** Conversion factors.

<i>From</i>	<i>To</i>	<i>Conversion</i>
U (mg/kg)	<sup>238</sup> U (Bq/kg)	<sup>238</sup> U (Bq/kg) = U (mg/kg) x 12.35
U (mg/kg)	<sup>226</sup> Ra (Bq/kg)	<sup>226</sup> Ra (Bq/kg) = U (mg/kg) x 12.35
Th (mg/kg)	<sup>208</sup> Tl (Bq/kg)	<sup>208</sup> Tl (Bq/kg) = Th (mg/kg) x 4.063
K (%)	<sup>40</sup> K (Bq/kg)	<sup>40</sup> K (Bq/kg) = K (%) x 312.5

281

282 **2.5. Data analysis**283 **a) Influence of analytical techniques**

284 The influence of the analytical techniques in the LIT database was assessed through  
285 a three-way Analysis of Variance (ANOVA) using the OGE classification scheme, the  
286 analytical techniques and LOI as categorical variables. The ANOVA design was  
287 unbalanced due to the unequal number of observations resulting from the level  
288 combinations of each factor (OGE, LOI and analytical techniques). Given an unbalanced  
289 design, the differences between the geochemical analysis techniques were assessed by  
290 pairwise comparisons of the Least square (LS) means calculated with the three-way  
291 ANOVA (the LS mean is an estimate of the population mean, gained from grouped data  
292 by adjusting for the influence of grouping, thereby adjusted for the imbalance of the  
293 ANOVA design). If the results of the pairwise comparison of the LS means show an  
294 agreement of the LS means between the different geochemical analysis techniques, the  
295 compiled database can be considered consistent within itself, thus, reliable. Data  
296 showing strong deviations from normality were transformed through the Box-Cox  
297 transformation (Zar, 2010).

298 **b) Accuracy of the LIT database**

299 One objective of the study is to evaluate whether the LIT database is accurate in the  
300 sense that mean geochemical concentrations per geological unit are representative for  
301 the units. Representativeness means that the frequency distribution of concentrations in  
302 samples is equal the true, but unknown one; in particular, the sample mean must be  
303 equal to the true mean. Deviation is called bias or inaccuracy. Since the true distributions

304 are not known, accuracy of samples cannot be validated by comparing sample and true  
305 distributions. However, a necessary – though not sufficient – condition for accuracy is  
306 that for each geological unit mean  $TGDR_{calc}$  is equal (up to permitted tolerance) to mean  
307  $TGDR_{obs}$  in the same unit, taken from the RMP.

308 As a first step, the arithmetic mean of the  $TGDR_{obs}$  for each geological unit studied  
309 of the OGE classification scheme (Table 1 and Figure 1) was estimated. The zonal  
310 statistics tool in ArcGis 10.5.1 (ESRI, Redland CA, USA, 2017) was used for this  
311 purpose. Given that the RMP data corresponds to data modelled by linear interpolation,  
312 due to the absence of outliers, the arithmetic mean is a robust measure of central  
313 tendency. Since the TGDR per pixel of the RMP are given as class values, i.e. belonging  
314 to a class “i”,  $TGDR \in (x_i, x_{i+1})$ , the AM cannot be simply calculated from pixel values.  
315 This would be the straight-forward way, but the means per pixel are not available. An  
316 approximation is the weighted mean of class means,  $(x_{i-1}+x_i)/2$ , with weight the number  
317 of pixels  $N(i-1, i)$  in this class divided by the total number of pixels,  $N$ .  $x_0$  is set equal zero,  
318 which is the physically possible lower bound of a geochemical concentration. The  
319 question is how to deal with the highest class, labeled “ $>x_n$ ”, as no class mean can be  
320 calculated, since its upper bound is not known. This can be done approximately by  
321 assuming a tentative analytical model of the upper tail of the TGDR distribution and  
322 calculate its contribution to the AM analytically. The maximum  $TGDR_{obs}$  was estimated  
323 by polynomial regression of the TGDR values displayed in the legend of the RMP (Batista  
324 et al. 2013). Polynomials of degree 6 and 7 were applied to maximize the multiple R-  
325 squared and minimize the residual standard error. A maximum  $TGDR_{obs}$  of 236.911  
326 nGy/h and 238.358 nGy/h were predicted from the 6-degree and 7-degree polynomial  
327 regression model fittings, respectively. The mean value of 237.6 nGy/h was set as the  
328 maximum  $TGDR_{obs}$  in order to estimate the arithmetic means of  $TGDR_{obs}$  for each  
329 geological unit. We tried two additional tail models, but the results are very similar and  
330 these models are not further discussed here.

331 As second step, the comparison of the arithmetic means between the LIT database  
332 and the RMP for each geological unit was carried out using the Student's paired sample  
333 t-test, the Wilcoxon median test and Pearson's correlation coefficient. The distributions  
334 of  $TGDR_{calc}$  and  $TGDR_{obs}$  were compared with the Anderson-Darling and Kolmogorov-  
335 Smirnov tests. Statistical analyses were performed with Statistica 7.0 (StatSoft Inc.,  
336 USA) and R (Lenth, 2016; R Core Team, 2017).

### 337 **3. Results and discussion**

#### 338 **3.1. Analysis of the influence of the analytical techniques on LIT database**

##### 339 **a) Effect size**

340 The results of a three-way ANOVA performed on Box-Cox transformed data using  
341 the OGE classification scheme, LOI and the analytical techniques as categorical  
342 variables are shown in Table 3. The differences between the mean values of the  
343 analytical techniques and geological factors were significant at a 0.001 level of  
344 significance for U, Th, K and  $TGDR_{calc}$ . The effects of LOI were not significant at a 0.01  
345 level of significance for U, Th, K and  $TGDR_{calc}$ .

346 The percentage of variation explained by the analytical techniques and LOI given by  
347 the partial eta-squared was close to or lower than 2 %, being lower than the variance  
348 explained by geological factors. According to Cohen's (1977) rule of thumb, the size of  
349 the effects of the analytical techniques and particularly LOI can be considered small (<  
350 2 %) while the size of the effects of the geological factors was large (> 14 %). The  
351 variance explained by the LOI effects was generally lower than the variance explained  
352 by the analytical techniques (Table 3). The size of the effects of the analytical techniques  
353 was higher for the  $TGDR_{calc}$  compared to the individual radioelements which is likely due  
354 to the combination of analytical techniques that results in a higher number of categories,  
355 hence, in a higher variability of geochemical analysis techniques used (Table 3).

356 The percentage of variation explained by geological factors was on average 53 % for  
357 U, 38 % for Th, 50% for K and 51 % for the  $TGDR_{calc}$ . The variance explained by

358 geological factors was much higher than the variance explained by the analytical  
 359 techniques and LOI (Table 3). The geological factors are thereby the main source of  
 360 variability in the data, followed by the error component. The latter can be assumed to  
 361 mainly represent true spatial variability of geochemical concentrations – and in  
 362 consequence the calculated dose rate  $TGDR_{calc}$  – within geological units.

363 **Table 3.** Summary table of a three-way ANOVA performed on U, Th, K and  $TGDR_{calc}$   
 364 Box-Cox transformed data using the OGE classification, LOI and the analytical  
 365 techniques as categorical variables.

		SS	df	MS	F	p	Partial eta-squared (%)
U	OGE	2009.7	25	80.4	95.7	<0.001	52.7
	LOI	5.4	1	5.4	6.5	0.011	0.3
	Analytical techniques	33.0	2	16.5	19.7	<0.001	1.8
	Error	1801.1	2144	0.8			
Th	OGE	3457.9	26	133.0	58.70	<0.001	37.6
	LOI	0.9	1	0.9	0.38	0.536	<0.1
	Analytical techniques	37.9	2	18.9	8.35	<0.001	0.7
	Error	5741.2	2534	2.3			
K	OGE	1909.4	26	73.4	106.8	<0.001	50.4
	LOI	0.7	1	0.7	1.0	0.306	<0.1
	Analytical techniques	21.9	5	4.4	6.4	<0.001	1.2
	Error	1880.5	2735	0.7			
$TGDR_{calc}$	OGE	37380	25	1495	83.1	<0.001	50.7
	LOI	40	1	40	2.2	0.137	0.1
	Analytical techniques <sup>a</sup>	1882	9 <sup>a</sup>	209	11.6	<0.001	4.9
	Error	36311	2018	18			

SS – Sum of squares; df – degrees of freedom; MS – Mean square; F – F-test; p – p-value.

<sup>a</sup>The effects of the group where U and Th were determined by NAA and K by AAS are not estimable for the  $TGDR_{calc}$ , thus were excluded from the analysis.

### 366 **b) Component effects**

367 The differences between the LS means of the analytical techniques were investigated  
 368 in Figure 2. The results of the pairwise comparison of the LS means are presented as  
 369 supplementary material for U, Th and K (Table A3) and  $TGDR_{calc}$  (Table A4). The ICP-  
 370 MS LS mean was lower than the NAA and XRF means for both U and Th (Figure 2). For  
 371 U, the LS means for NAA and XRF were 35 % higher, on average, than the ICP-MS  
 372 mean. For Th, the XRF LS mean was 8 % higher while the NAA LS mean was 20 %

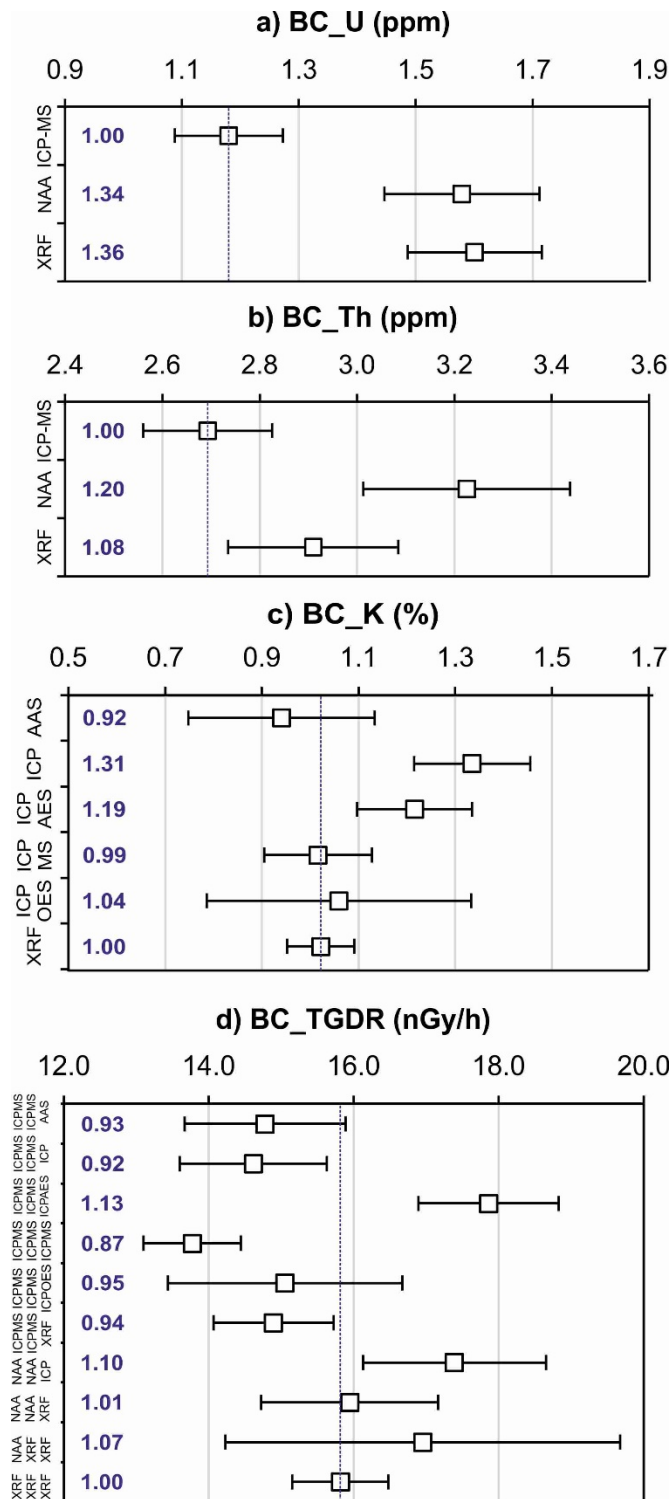
373 higher than the ICP-MS mean. The pairwise comparison of the LS means showed a  
374 systematic statistically significant differences between the ICP-MS data compared to  
375 other datasets (NAA and XRF) for U (Table A3). For Th, a statistically significant  
376 difference was observed between NAA and ICP-MS datasets, however, the differences  
377 between XRF and NAA, and XRF and ICP-MS were not significant (Table A3).

378 The analytical technique used to determine U was identical to the analytical technique  
379 used to determine Th (ICP-MS, NAA or XRF in 2084 out of the 2173 observations  
380 available for U (96% of the available data). As the ICP-MS LS means were generally  
381 lower for both elements, a significant bias of this analytical technique is suggested. The  
382 bias could be due to (1) a preference of the authors for analyzing rock samples with lower  
383 U and Th concentrations with the ICP-MS technique; (2) lower detection limits achieved  
384 by ICP-MS compared to other analytical techniques (e.g. Gill, 1997) or (3) by the  
385 technique limitations such as an inefficient digestion of typically resistant U- and Th-  
386 bearing minerals (e.g. zircon) or selective coprecipitation of these elements with fluorides  
387 when hydrofluoric acid is used in the digestion process (Preusser and Kasper, 2001).

388 For K, the pairwise comparison of the LS means indicated a general agreement  
389 between datasets apart from the observed statistically significant differences of the LS  
390 means where K was determined by the ICP technique (Table A3). These results are  
391 congruent with the observed LS mean of the ICP analysis technique which was 30 %  
392 higher than the reference mean chosen (XRF, see Figure 2). Given that only 8 out of  
393 2768 observations present a K concentration below the LLD, it is unlikely that the  
394 observed difference could be caused by LLD values. Given that the ICP results were  
395 retrieved from older literature sources (ex. Mitjavila et al. 1997; Ribeiro 1998; Table A1)  
396 and that the ICP LS mean was higher than the reference mean chosen, the observed  
397 difference could be due to lack of reporting of values below the LLD by the authors. Apart  
398 from ICP and ICP-AES, deviations of the LS means from the reference mean chosen  
399 were generally lower than 10 %.

400 The results of the pairwise comparison of the LS means for  $TGDR_{calc}$  showed a  
401 general agreement of the LS means between different geochemical analysis techniques  
402 (Table A4). Statistically significant differences between the LS means were only  
403 observed in 6 out of the 45 pairwise comparisons performed (Table A4). The compiled  
404 database can thus be considered consistent within itself, hence reliable. Apart from the  
405 dataset where K was determined by ICP-AES and ICP-MS, deviations of the LS means  
406 from the reference mean chosen are equal to or lower than 10 %. The LS means where  
407 U and Th data were acquired with ICP-MS are systematically lower than the LS means  
408 where U and Th data were acquired with NAA and XRF, which is consistent with the  
409 results observed for U and Th individually and can thus be caused by error propagation.

410 The LS means for the OGE units and LOI are presented as supplementary material  
411 (Figures A1 and A2). The LS means display large differences between the various OGE  
412 units for U, Th, K and  $TGDR_{calc}$  (Figure A1), which are in accordance with the high  
413 percentage of variation explained by the OGE classification system (Table 3). With  
414 respect to LOI, higher LS means were expected in data corrected for the LOI and/or  
415 volatile content (corresponding to effect named "FALSE"), as are observed for K and the  
416  $TGDR_{calc}$  (Figure A2). However, lower LS means in LOI corrected data compared to  
417 uncorrected data are observed for U and Th (Figure A2 in Supplementary material).  
418 These results support the lack of a systematic influence of LOI. Given that the effects of  
419 LOI are not significant for U, Th and K (Table 3), it can be concluded that the correction  
420 of U, Th and K data for LOI or lack thereof does not affect significantly the results.



421

422 **Figure 2.** Plots of the least squares (LS) means of the analytical techniques effect for U,  
 423 Th, K and TGDR<sub>calc</sub> Box-Cox transformed data. The bars indicate the 95% confidence  
 424 interval of the LS mean. A LS mean was chosen as reference for comparison according  
 425 to higher sample size and number of geological units sampled (see Tables A1 and A2,  
 426 supplementary material). The proportion of the LS means were calculated assuming the  
 427 ICP-MS mean as reference for U and Th; the XRF mean for K and the dataset where U,  
 428 Th and K were determined by XRF for the TGDR<sub>calc</sub>.



429 **3.2. Variance component originating from spatial variability**

430 In the previous analysis, variance has been attributed entirely to geology (component  
431 A, see section 1.4) and analytical techniques (B), while component C was ignored, i.e.  
432 possible lack of representativeness of data for a geological unit. A possible procedure  
433 for resolving this would be performing an even higher stage ANOVA including individual  
434 data sources (each consisting of a number of data) as additional classification variable,  
435 or as simplification, simple ANOVA for data sources within individual geological units and  
436 for same analytical technique. However, data are not sufficient for this.

437 The sources of the error terms in Table 3 are measurement uncertainty and probably  
438 dominantly, spatial variability within units. Its contribution to the SS (sum of squares) is  
439 in the same order of magnitude as the one due to OGE. Given the data situation, it is  
440 however difficult to say to which degree possibly unrepresentative samples from the  
441 variable population contribute to bias of the mean. The question deserves further  
442 investigation in the future.

443 **3.3. Comparison of the LIT and RMP datasets**

444 The arithmetic means of the  $TGDR_{calc}$  from U, Th and K compiled data were  
445 compared to the  $TGDR_{obs}$  for each geological unit in Figure 3 and Table 4. The  
446 correlation between  $TGDR_{calc}$  and  $TGDR_{obs}$  was highly significant ( $p < 0.001$ ). The results  
447 of a paired sample t-test and Wilcoxon median tests indicate that the differences between  
448 the arithmetic means of  $TGDR_{calc}$  and  $TGDR_{obs}$  were not statistically significant ( $p = 0.126$   
449 and  $p = 0.14$ , respectively). Distributions of  $TGDR_{calc}$  and  $TGDR_{obs}$  were about equal  
450 according to Kolmogorov-Smirnov and Anderson-Darling tests. If deviation from  $TGDR_{obs}$   
451 by pooled standard deviation (SD) is taken as criterion, 14 out of 26 OGE units are  
452 located within one pooled SD around  $TGDR_{obs}$  (dashed lines in Figure 3), only 1 out of  
453 26 is located outside 2 pooled SDs.

454 (The pooled SD is defined as

455 
$$s_{pooled} = \sqrt{\frac{(n_1 - 1)s_1^2 + (n_2 - 1)s_2^2 + \dots + (n_k - 1)s_k^2}{n_1 + n_2 + \dots + n_k - k}} \quad (2)$$

456 with  $s_i$  the individual SDs and  $n_i$  the number of data in OGE unit (i). Its value is 29  
 457 nGy/h.)

458 A systematic overestimation of the  $TGDR_{calc}$  compared to  $TGDR_{obs}$  was observed for  
 459 the sedimentary rocks (N2, N1c, K1-2, K1, TJ1 and the C2 units) coupled with absolute  
 460 differences of the  $TGDR_{calc}$  generally higher than 29 nGy/h (the pooled SD of the RMP  
 461 dataset; Table 4, Figure 3). This effect may be caused, by (1) the lack of spatial  
 462 representativeness of the data (as sampling of stratigraphic sequences is commonly  
 463 carried out along vertical profiles instead of spatial grids) or (2) the lack of data from  
 464 carbonate rocks. Carbonate rocks are a common subordinate lithology of the  
 465 abovementioned geological units (see Table 1), presenting average radionuclides  
 466 concentrations generally lower than siliciclastic rocks (e.g. Sêco et al. 2016).

467 **Table 4.** Comparison of the arithmetic means ( $\pm$  standard deviation) of the TGDR  
 468 between LIT ( $TGDR_{calc}$ ) and the RMP datasets ( $TGDR_{obs}$ ).

Group	OGE unit	$TGDR_{calc}$ [1]	$TGDR_{obs}$ [2]	Difference [2] - [1]
Sedimentary rocks	N2	78 $\pm$ 26	32 $\pm$ 19	-46
	N1c	70 $\pm$ 23	36 $\pm$ 22	-34
	K1-2	106 $\pm$ 21	42 $\pm$ 28	-63
	K1	73 $\pm$ 21 <sup>b</sup>	46 $\pm$ 21	-27
	TJ1	85 $\pm$ 29	44 $\pm$ 20	-41
	C2	94 $\pm$ 15	59 $\pm$ 34	-35
Metamorphic rocks	C1-2	78 $\pm$ 27 <sup>b</sup>	65 $\pm$ 14	-13
	SDof	71 $\pm$ 18	19 $\pm$ 13	-52
	SD	39 $\pm$ 41	90 $\pm$ 22	51
	SDP	107 $\pm$ 37 <sup>b</sup>	91 $\pm$ 21	-16
	ODP	81 $\pm$ 58 <sup>b</sup>	104 $\pm$ 34	23
	O_a	69 $\pm$ 38 <sup>b</sup>	88 $\pm$ 27	18
	gz	77 $\pm$ 25 <sup>b</sup>	87 $\pm$ 28	10
	Npep	82 $\pm$ 23 <sup>b</sup>	80 $\pm$ 29	-2
	NP2	46 $\pm$ 30 <sup>b</sup>	60 $\pm$ 21	14
NP1	31 $\pm$ 31 <sup>b</sup>	56 $\pm$ 29	25	
Igneous rocks	gama2	68 $\pm$ 35 <sup>b</sup>	63 $\pm$ 16	-4
	CVL	52 $\pm$ 23	14 $\pm$ 10	-38
	g1_b	194 $\pm$ 101 <sup>b</sup>	157 $\pm$ 39	-37
	g2_b	137 $\pm$ 59 <sup>b</sup>	172 $\pm$ 40	35

	g3	157 ± 52 <sup>b</sup>	142 ± 46	-15
	g1_a	157 ± 88 <sup>b</sup>	136 ± 32	-21
	g2_a	150 ± 49 <sup>b</sup>	121 ± 24	-29
	g1a	61 ± 34 <sup>b</sup>	62 ± 23	1
	gama	9 ± 10	51 ± 19	43
	D3C1	50 ± 40 <sup>b</sup>	75 ± 26	25
<sup>a</sup> TGDR <sub>obs</sub> was calculated assuming an arbitrary maximum TGDR <sub>obs</sub> of 237.6 nGy/h.				
<sup>b</sup> The TGDR <sub>calc</sub> is located within one pooled SD of the TGDR <sub>obs</sub> .				

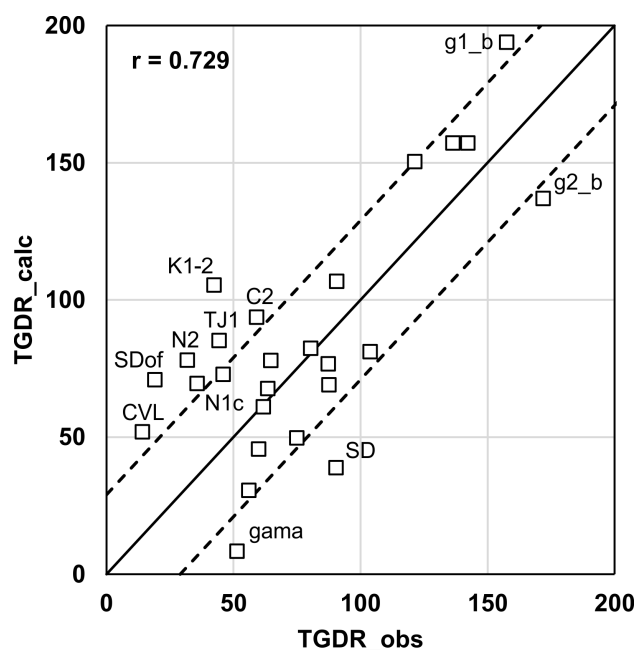
469 The arithmetic means of the TGDR<sub>calc</sub> of granitic rocks are also generally  
470 overestimated in LIT database (g1\_b, g3, g1\_a and g2\_a). In this case, the limitations of  
471 the RMP data must be taken into account, namely the fact that the maximum TGDR<sub>obs</sub>  
472 in mainland Portugal is unknown. A value of 237.6 nGy/h was set as the maximum  
473 TGDR<sub>obs</sub> in order to compute the mean TGDR<sub>obs</sub> by geological unit. This issue only  
474 affects the geological units which display a TGDR<sub>obs</sub> higher than 216.2 nGy/h, which are  
475 listed in Table 5 and include the abovementioned granitic rocks (g1\_b, g2\_b, g3, and  
476 g1\_a). These units present a high percentage of their area (> 3 %) characterized by a  
477 TGDR<sub>obs</sub> above 216.2 nGy/h. The underestimation of TGDR<sub>obs</sub> of these units may be  
478 caused by the underestimation of the maximum TGDR<sub>obs</sub> chosen (of 237.6 nGy/h),  
479 biasing the estimation mean TGDR<sub>obs</sub> by geological unit.

480 **Table 5.** Percentage of the area characterized by a TGDR<sub>obs</sub> higher than 216.2 nGy/h.

	OGE units	% of Area
Sedimentary rocks	N2	0.01
	N1c	0.05
	K1_2	0.68
	ODP	1.20
Metamorphic rocks	O_a	0.16
	gz	0.06
	NPep	0.47
	g1_b	5.36
Igneous rocks	g2_b	15.15
	g3	9.54
	g1_a	3.04
	g2_a	0.37

481 From a health perspective, the overestimation of the TGDR<sub>calc</sub> is not as concerning  
482 as its underestimation, which was observed systematically in the SD, g2\_b and the gama  
483 units. In the case of the g2\_b unit, the difference observed between the TGDR<sub>calc</sub> for

484 Model 1 is, however allocated within one SD of the  $TGDR_{obs}$ . The underestimation of the  
 485  $TGDR_{calc}$  of the g2\_b may be due to the underrepresentation of quantitative data from  
 486 metallogenic deposits of uranium and/or radium that are common in the g2\_b unit (ex.  
 487 Marques et al. 2010; Neiva et al. 1987; Trindade et al. 2013). Because the g2\_b dataset  
 488 corresponds to one of the largest datasets available (Table A1, Supplementary material),  
 489 this enforces the hypothesis that the major source of error within the compiled database  
 490 was the insufficient representativeness of the data, which leads to a biased estimate of  
 491 the mean, rather than the amount of data or the analytical techniques. Apart from the SD  
 492 and SDof units, the absolute deviations between the arithmetic means of  $TGDR_{calc}$  and  
 493  $TGDR_{obs}$  were generally lower than 29 nGy/h for the metamorphic rocks (Table 4). For  
 494 the SD unit, the only dataset available was retrieved from Dahn et al. (2014) and refers  
 495 to mafic rocks outcropping in the SD unit. Thus, the underestimation of the  $TGDR_{calc}$   
 496 reflects the lack of data from other types of rocks within the unit, particularly of the  
 497 predominant lithology of the unit namely phyllites (Table 1). The underestimation of the  
 498  $TGDR_{calc}$  of the gama unit may be due to the lack of data from differentiated rocks, such  
 499 as anorthositic rocks and diorites (the subordinate lithologies of the gama unit, Table 1)  
 500 that may present higher contents of terrestrial radionuclides than gabbros as U, Th and  
 501 K become enriched with magmatic differentiation (Heier and Rogers, 1963).



502

503 **Figure 3.** Scatterplot of the arithmetic means of the TGDR (in nGy/h) of each geological  
504 unit. The dashed lines indicate deviations of 29 nGy/h (the pooled SD) from the identity  
505 line.

506 The pooled SD of the RMP dataset was set as criterion to appraise the discrepancy  
507 between the arithmetic means of the RMP and LIT databases. Given the statistical  
508 parameters of the differences between  $TGDR_{calc}$  and  $TGDR_{obs}$  and their distributions, the  
509 compatibility of the RMP and LIT databases can be considered acceptable, but not  
510 perfect. For some geological units, the exact reason for discrepancy remains to  
511 investigated, in order to improve further attempts of the kind.

512 As the  $TGDR_{calc}$  was calculated from U, Th and K data compiled from scientific  
513 literature, the compatibility of the TGDR between the LIT and the RMP datasets (namely,  
514  $TGDR_{calc}$  and  $TGDR_{obs}$ ) implies that the estimation of the contents of the terrestrial  
515 radionuclides in bedrock from compiled datasets is also reasonable. Putting it more  
516 precisely, since the approximate equality of  $TGDR_{calc}$  and  $TGDR_{obs}$  is only a necessary  
517 condition for the hypothesis to be true that the LIT database represents U, Th and K  
518 concentration in rocks reasonably well, the result of this investigation does not reject the  
519 hypothesis.

520 Vein-type rocks were excluded from the comparison exercise of LIT and RMP  
521 databases due to their large perimeter to area ratio. Given the compatibility of the RMP  
522 and LIT datasets, the  $TGDR_{calc}$  of vein-type rocks was estimated resorting to data  
523 compiled from local geochemical surveys. The mean  $TGDR_{calc}$  for the f1 and f2 (basic  
524 veins) and qz (quartz veins) units are presented in Table 6. The  $TGDR_{calc}$  of basic vein-  
525 type rocks (f1 and f2) is similar to other mafic rocks units (ex. CVL, gama, and SDof units,  
526 Tables 1 and 4) as expected. Quartz veins present a similar  $TGDR_{calc}$  to sedimentary  
527 and metamorphic rocks, being lower than the average  $TGDR_{calc}$  of granitic rocks (Tables  
528 1 and 4).

529 **Table 6.** Estimation of the TGDR of vein-type units using LIT data.

	OGE units	Number of samples	TGDR <sub>calc</sub> (nGy/h)		References
			Mean	Standard deviation	
Vein-type rocks	f1	26	14	5	Cebriá et al. (2003); Martins (1999); Martins et al. (2009); Youbi et al. (2003);
	f2	17	34	14	Trindade et al. (2013); Alves (2010);
	qz	16	83	20	Lima (2000)

#### 530 4. Conclusions

531 Two databases were compared with the aim of validating the compilation of literature  
532 data to estimate the mean concentrations of terrestrial radionuclides in bedrock using  
533 geological units as reference units. The results show that the compatibility of the  
534 databases was essentially reasonable. The differences between the arithmetic means  
535 were generally lower than the maximum allowable discrepancy set and the differences  
536 between the two databases were not statistically significant. A systematic overestimation  
537 of the contents of radionuclides calculated from the compiled data and observed in  
538 sedimentary rocks probably reflects the lack of representativeness of the data compiled  
539 for these units, while the differences observed for some granitic rocks may reflect the  
540 limitations of the TGDR<sub>obs</sub>. Despite the significance of the effects of the analytical  
541 techniques, the variance explained by them, ranging from less than 1% to 5%, is  
542 sufficiently low, thereby lacking a significant impact on the comparability of the results  
543 acquired with different analytical techniques. The major sources of variability in the data  
544 are geological factors as demonstrated by the high percentage of variation explained by  
545 this factor in the ANOVA computed, as well as the true spatial variability of geochemical  
546 concentrations within geological units.

547 Geochemical characterization of geological units based on scattered literature data,  
548 as attempted in this study, allows the development of maps of terrestrial radionuclides  
549 contents in bedrock without large-scale sampling efforts, since a large amount of data is  
550 already available. Where extensive TGDR surveys are available, validation may be  
551 repeated e.g. along the lines presented here.

552 While the estimates computed for some units may be unreliable, they also set the  
553 groundwork needed to support further research. Certainly, a key problem is  
554 representativeness of the data, on which depends the reliability of the estimates of the  
555 mean contents of U, Th and K per geological units. It can be improved when more data  
556 with better coverage of geological units are available. Nonetheless, we think that  
557 generation of maps of U, Th and K concentrations in rock based on geological units is  
558 feasible.

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1029 **Supplementary material**

1030 **Table A1.** Summary of the data available in LIT database for the OGE units sampled by  
 1031 geological unit. 2173 observations were compiled for U, 2564 for thorium and 2768 for K  
 1032 acquired with 7 different analytical techniques.

OGE unit	Analytical Technique <sup>a</sup>	N (U)	N (Th)	N (K)	References
C1-2	ICP-MS	74	74	0	Jorge et al. (2013); Trindade (2007)
	ICP-OES	0	0	74	
	NAA	7	7	0	
	XRF	0	0	7	
C2	ICP-MS	18	20	18	Dinis et al. (2011); Ribeiro et al. (2010)
	XRF	0	0	2	
CVL	ICP-MS	62	62	62	Miranda (2010); Miranda et al. (2009)
D2-3	XRF	0	30	32	Boulter et al. (2004)
D3C1	ICP	0	0	59	Boulter et al. (2004); Mitjavila et al. (1997); Rosa et al. (2004, 2006); Schützet al. (1987)
	NAA	59	129	0	
	XRF	28	27	135	
g1_a	AAS	0	0	17	Coelho et al. (2007); Costa et al. (2014); Ferreira (2013); Godinho et al. (1986, 1991); Gomes (1989); Gomes and Neiva (2002, 2005); Gomes et al. (2011, 2013); Neiva and Gomes, (1991); Neiva et al. (2008, 2009); Neves et al. (1999); Silva (2000, 2014a); Teixeira (2008); Teixeira et al. (2012)
	ICP	0	0	32	
	ICP-AES	0	0	7	
	ICP-MS	77	77	0	
	NAA	4	13	0	
	XRF	150	205	230	
g1_b	ICP-AES	0	0	15	Costa (2011); Costa et al. (2014); Gomes and Neiva (2005);
	ICP-MS	15	15	0	
	NAA	6	6	0	
	XRF	0	0	6	
g1a	ICP-AES	0	0	22	Antunes et al. (2010); Lima et al. (2013, 2014); Moita et al. (2009)
	ICP-MS	57	79	57	
g2_a	ICP-AES	0	0	12	Cerejo (2013); Neves et al. (2007); Silva and Neiva (1990)
	ICP-MS	12	12	0	
	NAA	10	10	0	
	XRF	8	8	18	
g2_b	AAS	0	0	43	Antunes (2006); Antunes et al. (2008, 2013); Carvalho et al. (2012); Costa (2006, 2011); Godinho et al. (1991); Lima et al. (2013, 2014); Marques et al. (2010); Martins et al. (2013); Neiva et al. (1987, 2009, 2011a, 2011b); Ramírez and Grundvig (2000); Silva (2014b); Silva et al. (2000); Trindade et al. (2013); van der Weijden and van der Weijden (1995)
	ICP-AES	0	0	134	
	ICP-MS	165	164	24	
	NAA	23	13	0	
	XRF	185	259	281	
g3	ICP-AES	0	0	64	Costa (2006, 2011); Gomes (2008); Jaques et
	ICP-MS	56	56	6	

OGE unit	Analytical Technique <sup>a</sup>	N (U)	N (Th)	N (K)	References
	NAA	0	3	0	al. (2016); Martins et al. (2009); Neiva (1993); Neves and Pereira, (2007); Ramirez and Menéndez (1999)
	XRF	14	14	32	
GAMA	ICP	0	0	60	Caldeira et al. (2007); Jesus et al. (2014, 2016); Pedro (2004); Quesada et al. (1994)
	ICP-MS	92	97	138	
	NAA	32	32	0	
	XRF	0	0	15	
GAMA2	ICP-MS	16	16	16	Grange et al. (2010); Miranda (2010)
gz	ICP-MS	8	8	4	Antunes (2006); Antunes et al. (2009); Cruz (2007); Godinho et al. (1986); Neves et al. (1998); Pereira et al. (2006, 2011); Sánchez-García et al. (2010, 2013)
	XRF	25	57	62	
K1	NAA	52	52	0	Trindade (2007); Trindade et al. (2010)
	XRF	0	0	52	
K1-2	ICP-MS	45	45	0	Dinis et al. (2016)
	XRF	0	0	45	
N1c	ICP	0	0	32	Arribas et al. (2014); Bustillo et al. (2012); Lisboa et al. (2015); Marques et al. (2010); Reis et al. (2012)
	ICP-AES	0	0	5	
	ICP-MS	48	48	0	
	NAA	16	16	0	
	XRF	0	0	16	
N2	ICP-MS	45	72	5	Dinis and Oliveira (2016); Dinis et al. (2011); Pereira et al. (2006); Trindade (2007)
	NAA	6	6	0	
	XRF	0	0	88	
NP1	ICP-AES	0	0	69	Gómez-Pugnaire et al. (2013); Henriques et al. (2017); Silva (2007)
	ICP-MS	90	90	0	
	XRF	0	0	21	
NP2	ICP	0	0	14	Dinis et al. (2011); Henriques et al. (2006, 2016); Lorda et al. (2013); Pereira et al. (2006, 2011)
	ICP-AES	0	0	5	
	ICP-MS	16	63	42	
	XRF	1	1	3	
Npep	AAS	0	0	16	Aires et al. (2011); Candeias et al. (2015); Cerejo (2013); Coelho et al. (2007); Dinis et al. (2011); Fuenlabrada et al. (2016); Godinho et al. (1991); Marques et al. (2010); Neves et al. (1999); Noronha et al. (2012); Oliveira (2015); Pinto (2014); Ribeiro (1998); Silva (2014a); Trindade et al. (2013); Ugidos et al. (1997a, 1997b, 2003a, 2003b, 2010); Valladares et al. (2000, 2002); Villaseca et al. (2014)
	ICP	0	0	35	
	ICP-AES	0	0	176	
	ICP-MS	310	341	86	
	ICP-OES	0	0	24	
	NAA	9	9	0	
	XRF	22	22	30	
O_a	ICP	0	0	1	Oliveira (2015); Pereira et al. (2011); Ribeiro et al. (2010); Silva (2014a)
	ICP-MS	2	2	1	
	XRF	15	15	15	
ODP	ICP	0	0	52	Dias (2011); Noronha et al. (2012); Ribeiro (1998)
	ICP-MS	41	41	0	
	NAA	46	46	0	
	XRF	6	6	47	
SD	XRF	33	33	33	Dahn et al. (2014)

OGE unit	Analytical Technique <sup>a</sup>	N (U)	N (Th)	N (K)	References
SDof	AAS	0	0	5	Fuenlabrada et al. (2010, 2012); Teixeira et al. (2010)
	ICP-MS	48	43	0	
	ICP-OES	0	0	43	
SDP	ICP	0	0	60	Noronha et al. (2012); Ribeiro (1998)
	NAA	54	55	0	
	XRF	3	3	3	
TJ1	NAA	62	62	0	Trindade (2007); Trindade et al. (2010)
	XRF	0	0	60	

N – number of observations.  
<sup>a</sup>ICP-MS – Inductively Coupled Plasma Mass Spectrometry; ICP-OES – Inductively Coupled Plasma Optical Emission Spectrometry; NAA – Neutron Activation Analysis; XRF – X-Ray Fluorescence; ICP - Inductively Coupled Plasma; ICP-AES – Inductively Coupled Plasma Atomic Emission Spectrometry; AAS – Atomic Absorption Spectrometry.

1033

1034 **Table A2.** Summary of the data available by analytical technique.

	Analytical technique			Number of samples (N)	% N	Number of OGEs sampled	%OGE
	U	Th	K				
U	NAA			386	17.8	14	51.9
	XRF			490	22.5	12	44.4
	ICP-MS			1297	59.7	21	77.8
Th		NAA		459	17.9	15	55.6
		XRF		680	26.5	13	48.1
		ICP-MS		1425	55.6	21	77.8
K			AAS	81	2.9	4	14.8
			ICP-OES	141	5.1	3	11.1
			ICP-MS	459	16.6	12	44.4
			ICP	345	12.5	9	33.3
			ICP-AES	509	18.4	10	37.0
			XRF	1233	44.5	23	85.2
TGDR <sub>calc</sub> <sup>a</sup>	ICP-MS	ICP-MS	ICP-MS	330	16.1	12	44.4
	ICP-MS	ICP-MS	ICP-OES	141	6.9	3	11.1
	ICP-MS	ICP-MS	ICP	143	7.0	5	18.5
	NAA	NAA	ICP	176	8.6	5	18.5
	NAA	NAA	XRF	160	7.8	8	29.6
	XRF	XRF	XRF	421	20.5	11	40.7
	ICP-MS	ICP-MS	XRF	217	10.6	10	37.0
	ICP-MS	ICP-MS	ICP-AES	380	18.5	8	29.6
	ICP-MS	ICP-MS	AAS	76	3.7	3	11.1
	NAA	XRF	XRF	10	0.5	1	3.7

<sup>a</sup>The analytical techniques listed for the TGDR<sub>calc</sub> refer to the compilation of the analytical techniques used to analyse U, Th and K.

1035

1036 **Table A3.** Pairwise comparison of the LS means of the analytical techniques for U, Th  
 1037 and K. P-values were adjusted with the false discovery rate (FDR) method (Lenth, 2016).

	<b>Analytical techniques under comparison</b>	<b>Difference between the LS means</b>	<b>SE</b>	<b>t</b>	<b>p</b>
U (df = 2144)	NAA x ICP-MS	-0.399	0.085	-4.691	<0.001(*)
	NAA x XRF	-0.022	0.091	-0.240	0.9688
	ICP-MS x XRF	-0.421	0.075	-5.589	<0.001(*)
Th (df = 2534)	NAA x ICP-MS	-0.532	0.132	-4.033	<0.001(*)
	NAA x XRF	0.316	0.145	2.171	0.076
	ICP-MS x XRF	-0.217	0.115	-1.890	0.142
K (df = 2735)	AAS x ICP	-0.394	0.116	-3.411	0.009(*)
	AAS x ICP-AES	-0.276	0.112	-2.469	0.134
	AAS x ICP-MS	-0.076	0.114	-0.667	0.986
	AAS x ICP-OES	-0.119	0.169	-0.708	0.981
	AAS x XRF	-0.081	0.099	-0.822	0.964
	ICP x ICP-AES	0.119	0.076	1.562	0.624
	ICP x ICP-MS	0.319	0.077	4.163	0.001(*)
	ICP x ICP-OES	0.275	0.155	1.770	0.485
	ICP x XRF	0.313	0.067	4.664	<0.001(*)
	ICP-AES x ICP-MS	0.200	0.075	2.674	0.081
ICP-AES x ICP-OES	0.156	0.148	1.055	0.899	
ICP-AES x XRF	0.194	0.066	2.923	0.041	
ICP-MS x ICP-OES	-0.044	0.154	-0.282	1.000	
ICP-MS x XRF	-0.006	0.071	-0.078	1.000	
ICP-OES x XRF	0.038	0.151	0.253	1.000	

SE – standard error; t, p – t-test and respective p-value. Significant values at  $\alpha = 0.01$  are indicated with (\*).

1038

1039 **Table A4.** Pairwise comparison of the LS means of the analytical techniques for TGDR<sub>calc</sub>  
 1040 (df = 2018). P-values were adjusted with the false discovery rate (FDR) method (Lenth,  
 1041 2016).

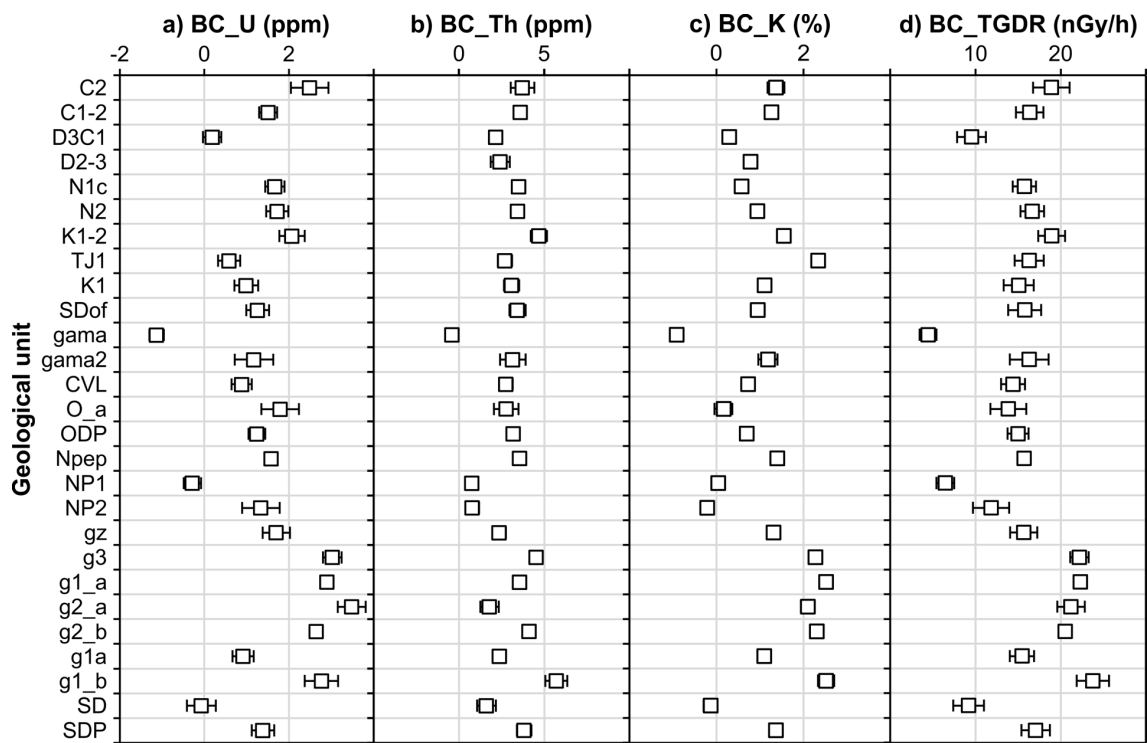
Analytical techniques <sup>a</sup> under comparison				Difference between the LS means			SE	t	p	
U	Th	K		U	Th	K				
ICP-MS	ICP-MS	AAS	x	ICP-MS	ICP-MS	ICP	0.162	0.780	0.208	1.000
				ICP-MS	ICP-MS	ICP-AES	-3.082	0.761	-4.052	0.002(*)
				ICP-MS	ICP-MS	ICP-MS	1.005	0.653	1.538	0.877
				ICP-MS	ICP-MS	ICP-OES	-0.275	1.058	-0.260	1.000
				ICP-MS	ICP-MS	XRF	-0.118	0.712	-0.166	1.000
				NAA	NAA	ICP	-2.611	0.900	-2.902	0.106
				NAA	NAA	XRF	-1.168	0.888	-1.316	0.950
				NAA	XRF	XRF	-2.175	1.434	-1.517	0.886
ICP-MS	ICP-MS	ICP	x	XRF	XRF	XRF	-1.038	0.544	-1.907	0.664
				ICP-MS	ICP-MS	ICP-AES	-3.245	0.516	-6.289	<0.001(*)
				ICP-MS	ICP-MS	ICP-MS	0.842	0.561	1.501	0.892
				ICP-MS	ICP-MS	ICP-OES	-0.437	0.901	-0.485	1.000
				ICP-MS	ICP-MS	XRF	-0.280	0.587	-0.478	1.000
				NAA	NAA	ICP	-2.774	0.786	-3.530	0.015
				NAA	NAA	XRF	-1.330	0.798	-1.668	0.814
				NAA	XRF	XRF	-2.338	1.498	-1.560	0.867
ICP-MS	ICP-MS	ICP-AES	x	XRF	XRF	XRF	-1.200	0.630	-1.906	0.665
				ICP-MS	ICP-MS	ICP-MS	4.087	0.527	7.749	<0.001(*)
				ICP-MS	ICP-MS	ICP-OES	2.807	0.828	3.392	0.025
				ICP-MS	ICP-MS	XRF	2.964	0.545	5.439	<0.001(*)
				NAA	NAA	ICP	0.471	0.778	0.606	1.000
				NAA	NAA	XRF	1.914	0.782	2.448	0.298
				NAA	XRF	XRF	0.907	1.482	0.612	1.000
				XRF	XRF	XRF	2.045	0.616	3.318	0.031
ICP-MS	ICP-MS	ICP-MS		ICP-MS	ICP-MS	ICP-OES	-1.280	0.895	-1.429	0.918
				ICP-MS	ICP-MS	XRF	-1.123	0.555	-2.024	0.582
				NAA	NAA	ICP	-3.616	0.760	-4.759	<0.001(*)
				NAA	NAA	XRF	-2.173	0.751	-2.894	0.108
				NAA	XRF	XRF	-3.180	1.429	-2.225	0.440
				XRF	XRF	XRF	-2.042	0.492	-4.155	0.001(*)
				ICP-MS	ICP-MS	XRF	0.157	0.932	0.168	1.000
				NAA	NAA	ICP	-2.336	1.074	-2.176	0.474
ICP-MS	ICP-MS	ICP-OES	x	NAA	NAA	XRF	-0.893	0.988	-0.903	0.996
				NAA	XRF	XRF	-1.900	1.661	-1.144	0.980
				XRF	XRF	XRF	-0.763	0.963	-0.792	0.999
				NAA	NAA	ICP	-2.493	0.695	-3.586	0.013
				NAA	NAA	XRF	-1.050	0.782	-1.343	0.944
				NAA	XRF	XRF	-2.057	1.456	-1.413	0.924
				XRF	XRF	XRF	-0.920	0.542	-1.696	0.798
				NAA	NAA	XRF	1.443	0.944	1.528	0.881
NAA	NAA	ICP	x	NAA	XRF	XRF	0.436	1.558	0.280	1.000
				XRF	XRF	XRF	1.574	0.769	2.047	0.566
				NAA	XRF	XRF	-1.007	1.552	-0.649	1.000
				XRF	XRF	XRF	0.130	0.764	0.170	1.000
				NAA	NAA	XRF	1.137	1.375	0.827	0.998

SE – standard error; t, p – t-test and respective p-value. Significant values at  $\alpha = 0.01$  are indicated with (\*). <sup>a</sup>The analytical techniques listed for the TGDR<sub>calc</sub> refer to the compilation of the analytical techniques used to analyse U, Th and K.

1042

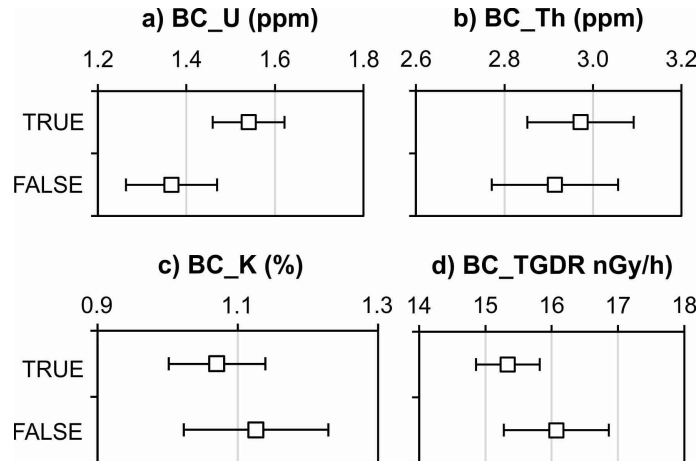
1043 **Table B1.** Compiled geochemical database.





1044

1045 **Figure A1.** Plots of the least squares (LS) means of the OGE effect for U, Th, K and  
 1046 TGDR<sub>calc</sub> Box-Cox (BC) transformed data by geological unit. The bars indicate the 95%  
 1047 confidence interval of the LS mean.



1048

1049 **Figure A2.** Plots of the least squares (LS) means of the LOI effect for U, Th, K and  
 1050 TGDR<sub>calc</sub> Box-Cox (BC) transformed data. The bars indicate the 95% confidence interval  
 1051 of the LS mean. Each observation of U, Th, K and TGDR<sub>calc</sub> is classified as “true” if the  
 1052 authors estimated but did not corrected the data for LOI (or the volatile content) and  
 1053 “false” otherwise.