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Mapping soil radioactivity in the Fernando de Noronha archipelago, Brazil

Fernando C. A. Ribeiro[1] · Dejanira da C. Lauria[1] · Mônica A. P. do Rio[1] · Fernanda G. da Cunha[2] · Wanderson de Oliveira Sousa[1] · Enjôlras de Albuquerque Medeiros Lima[3] · Melissa Franzen[3]

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Abstract A radioactivity survey was conducted on some of the most alkaline volcanic islands in the world. Seventy soil samples were analysed using gamma spectrometry. ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs (Bq kg^{-1}) activity concentration varied from 4.6 to 550, 10 to 298, 13 to 1280 and <0.3 to 2.0, respectively. Although the concentration ranges for ^{232}Th (^{228}Ra) and ^{226}Ra exceeded the worldwide range, ^{40}K levels remained low. The lowest values of natural radionuclides were observed over the Quixaba Formation, and the highest values were observed over the Remédios Formation. The median effective dose from external radiation exposure was 0.45 mSv a^{-1}.

Keywords Environmental radioactivity · ^{137}Cs · Soil · Radium isotopes · Mapping · Volcanic archipelago

Introduction

Studies of the distribution of radioactivity in soils have been performed worldwide to obtain radiological maps (of radioactivity levels), which provide useful information for mineral exploration and geological and geophysical investigations and can be used to assess the exposure of humans to radiation, to perform epidemiological studies and to build a "reference" database that can be useful for evaluating changes in environmental radioactivity due to nuclear, industrial or other human activities [1–14].

Natural environmental radioactivity levels are related to the presence of the primordial radioactive isotopes ^{238}U and ^{232}Th, their decay products, and ^{40}K. These radionuclides are present at different concentrations in all components of the environments. However, their levels in the soil depend on the geological and geographical conditions and vary in different parts of the world [15]. Although soil radionuclide contents are directly related to the radionuclide contents in the parent rock, the processes of soil formation may promote changes in radionuclide contents due to their addition, loss, transformation and translocation, which depend on additional factors such as topography, climate, organisms and time. Natural radioactive isotopes are responsible for approximately 86 % of human exposure to radiation [15].

Besides natural radioactive isotopes, which have been present since the beginning of the earth, atomic weapon tests and the Chernobyl and Fukushima accidents introduced man-made radionuclides to the atmosphere; these radionuclides were spread throughout the environment and have been incorporated in all components of the environment [16, 17]. Because few atomic tests were carried out in the southern hemisphere, the highest concentrations of ^{137}Cs in the soil were mainly found at middle and high latitudes in the northern hemisphere and in central Australia [17–20]. According to the estimated global distribution of ^{137}Cs in 1996 that originated from nuclear testing, the fallout in the equatorial region was minimal [21]. Therefore, many studies have been conducted to establish the ^{137}Cs baseline in soils in Europe, America, China, and Australia [16–20]. However, limited amounts of data from

✉ Fernando C. A. Ribeiro
fribeiro@ird.gov.br

[1] Instituto de Radioproteção e Dosimetria (IRD), Comissão Nacional de Energia Nuclear (CNEN), Av. Salvador Allende, s/n°, Rio de Janeiro, RJ 22780-160, Brazil

[2] Serviço Geológico do Brasil (CPRM), Av. Pasteur, 404, Rio de Janeiro 22292–040, Brazil

[3] Serviço Geológico do Brasil (CPRM), Av. Sul, 2291, Recife, PE 50770-011, Brazil

systematic surveys of this radionuclide in the equatorial region are available in the literature [22].

Some surveys have described the abiotic characteristics of the Fernando de Noronha archipelago (FNA), raising important data for the preservation of the ecosystem [23–28]. However, no studies concerning the occurrence of natural or man-made radioactive isotopes on these islands have been performed. Therefore, this study contributes to the knowledge of the island environments and establishes a "baseline" of the natural radioactive isotope and ^{137}Cs contents in the soils of this low-latitude volcanic archipelago in the southern hemisphere. Overall studies of these small oceanic islands can be used to gather relevant information for improving our understanding of the nature of the oceanic crust and the processes related to its spreading.

Materials and methods

Study site

The Fernando de Noronha archipelago is located in the Southern Equatorial Atlantic around 400 km from the northeast coast of Brazil (at 3°51′S and 32°25′W). The archipelago consists of one main island encircled by 20 islands and islets [24]. The total area of the archipelago is approximately 20 km^2. The main island, which the archipelago is named after, comprises 16.4 km^2 and is the only inhabited island.

The climate is tropical (Awi type according to the Köppen classification system), marked by the oceanic domain, with annual average temperature of approximately 25 °C, and the annual rainfall is approximately 1400 mm. There are two distinct seasons, a wet season from March to July and a dry season with a water deficit from August to January [25].

The archipelago has approximately 2700 inhabitants, and 100 % of the population resides in the urban area. Tourism is the main economic activity (in 2014 were about 90,000 visitors), followed by the service sector [26]. The inhabitants practice subsistence farming and artisanal fisheries.

The archipelago is the top of an extinct volcano, with a wide variety of rocks of igneous origin, ranging from ultrabasic to intermediate. Almeida [23] reports that an area of only 1.5 km^2 of the archipelago contains 14 different types of volcanic rocks. Geologically, the 21 islands and islets that form the Archipelago represent the top of a mountain chain that developed along an east–west fracture zone of the ocean floor and was built up by volcanic and subvolcanic alkaline and subsaturated rocks. These rocks are products of two distinct volcanic episodes. The first episode, which is represented by the Remédios Formation of the Late Miocene, is composed of pyroclastic deposits at the base and cut by intrusions in the form of necks, plugs, domes and dikes of subsaturated alkaline rocks. The composition ranges from intrusive basic-ultrabasic (lamprophyres, tephrites, basanites and alkali basalts) to intermediate (trachytes and phonolites) rocks. The second episode, represented by the Quixaba Formation of the Late Pliocene to the Early Pleistocene, is composed by a stack of melanocratic ankaratrite lava flows, subordinate pyroclastic deposits and some nefelinite dikes. A small amount of basanite flow with a controversial age is represented by the São José Formation [26].

The relief of the main island is made up of beaches, dunes, plains, slopes, low plateaus, hills, shore cliffs and a high plateau, which is where the small town of Vila dos Remédios lies and where most of the inhabitants of the island live.

The volcanism represented by the exposed rocks in the archipelago is characterized by its strongly sodium-alkaline nature, subsaturation of silica (there is no quartz in the rocks, although it is the most abundant mineral of the Earth's crust) and a high degree of differentiation. The silica contents of the rocks vary between 34.4 (melilite ankaratrite) and 60.8 % (alkali-trachyte). Among the oceanic volcanic islands of the world, the magmatic province of Fernando de Noronha is one of the most alkaline [24]. The lighter fractions of the magmatic differentiates were produced earlier, originating from the Remédios Formation, which corresponds to the oldest rocks of the archipelago that are dated between 8 and 12 Ma. After a period of time that may have lasted 5 to 6 Ma, ultrabasic volcanism of the Quixaba Formation was initiated, with an age of between 1.7 and 3 Ma [27].

The strongly melanocratic rocks are the youngest rocks of the Remédios Formation and contain little or no feldspar. Special emphasis is given to the xenoliths and ejectolites of the Remédios Formation because they may represent very deep source rocks of the volcanic edifice.

The Quixaba Formation is a sequence of black flows of alternating ankaratrite lava and pyroclasts containing components of the same lava [28]. This formation rests on two step-like plateaus of the main island. The eastern one may reach an exposed thickness of 180 m above sea level. This formation also occurs on some islands, particularly on the Rata Island and the small São José, Cuscuz and de Fora Islands.

Another formation called the Calcarenite of Caracas consists of light cream coloured sediment, also called Modern Sediment, and was built by fluctuations of sea level that covered the island platforms with deposits of sand, gravel, reef coralline algae and marine sands. During the Pleistocene, when the sea level was approximately six

metres below the current sea level, extensive sandy beaches were built to the south and southeast of the archipelago. These sands were moved by wind and formed dunes that likely reached a thickness of approximately 20 m. With the increase in sea level that followed, some of these sands were submerged and currently form the Calcarenite of Caracas Formation. These sediments are light cream in colour and are almost entirely composed of calcium carbonate grains of biogenic origin (quartz is virtually nonexistent in the islands). On Rato Island, phosphate deposits have been partially replaced by calcium phosphate that originated from seabird guano. In addition, the soils on Rata Island are rich in phosphates due to the interactions between the weathering of volcanic rocks and guano [28].

Sample collection, preparation and measurement

This survey is part of a cooperative agreement between the Geological Survey of Brazil (CPRM) and the Instituto de Radioproteção e Dosimetria (IRD/CNEN). The CPRM collects soil samples throughout all Brazil territories as part of its Low Density Geochemistry Project, and some of the collected samples have been sent to the IRD for radionuclide analysis.

Soil samples were collected at a depth of approximately 20 cm from residual soils that had not been displaced or used for agriculture by using a manual stainless steel auger. The sampling grid was approximately 500 × 500 m. Seventy samples were collected to achieve a sample density of 3.7 samples.km^{-2}. A Garmin Global Positioning System Receiver (GPS 12 XL) was used to record the latitude and longitude of each sample location. Figure 1 shows a map of the area and the locations of the sampling points.

Afterwards, 1–3 kg of the samples were stored in polythene bags and sent to the IRD. Later, approximately 1 kg subsamples of the soil samples were oven dried at 40 °C in the laboratory in a stove for 48 h or until a constant weight was achieved. Next, the samples were ground and passed through a nylon 2 mm sieve before further analysis. For gamma spectrometry analysis, sediment subsamples of approximately 300 g were transferred to polyethylene tubes, sealed and stored for at least 30 days to reach radioactive equilibrium among ^{226}Ra and its decay products [29].

Then, the samples were analysed for 60,000 s using two high purity germanium detectors (HPGe) and associated electronic devices (amplifier, HV source and MCA Analyser) from Canberra, Inc. (USA) with a relative efficiency of 20 %. Energy and efficiency calibrations were performed using standard sources supplied by the National Laboratory of Metrology of Ionizing Radiation (LNMRI) of the IRD. The activity of ^{228}Ra (^{232}Th) was derived from the 911.6 keV peak of ^{228}Ac. The peaks at 351.9 keV (^{214}Pb) and 609.3 keV (^{214}Bi) were used to determine the activity of ^{226}Ra. ^{40}K activities were calculated from its photo peak of 1460.8 keV, and ^{137}Cs activities were calculated from its photo peak of 661 keV [30]. The activities concentrations and respective uncertainties were determined according to the statistical uncertainties of the peak areas provided by the Genie2000 software. The value of minimum detectable activity (MDA) was 12.1 Bq kg^{-1} for ^{40}K, 2.3 Bq kg^{-1} for ^{226}Ra, 2.8 Bq kg^{-1} for ^{232}Th (^{228}Ra) and 0.3 Bq kg^{-1} for ^{137}Cs when a counting time of 60,000 s was used. The certificated reference material MAPEP-09-MaS21 and EML Analysis QAP59 were used for quality assurance.

All statistical data processing in this study was carried out using the commercial statistics software package *SPSS 19.0 for Windows* (IBM, USA) and ProUCL 5.0 [31, 32]. Maps were designed using the QGIS 2.8.2 software (Open Source Geospatial Foundation Project), which is free to download [33].

Results and discussion

Radionuclide concentrations

As usually found in environmental surveys, the dispersion of the radionuclide concentrations resulted in a skewed data distribution in this study (Fig. 2). The goodness-of-fit (GOF) tests for normal, lognormal and gamma distributions showed that the concentrations of ^{232}Th (^{228}Ra) and ^{226}Ra do not fit the three distributions, and the ^{40}K data distribution could be fit as a lognormal distribution (0.05 of significance level) [31]. Thus the statistical analysis should be performed using nonparametric methods [32]. For comparison purposes, the central tendencies of the data, including the average (arithmetic mean) and median, are reported in Table 1 that also shows the observed range of values of the radionuclide concentrations and the coefficients of variation obtained from the datasets. The uncertainties of the measurement values were around 4 % for ^{226}Ra, 3 % to ^{228}Ra and 9 % for ^{40}K.

The most abundant of these three nuclides is potassium, which is found in alkali feldspars and micas. A significant correlation was found between ^{226}Ra and ^{228}Ra, with a correlation coefficient of 0.542, and between ^{226}Ra and ^{40}K, with a correlation coefficient of 0.433 (0.05 significant). No correlation was found between ^{228}Ra and ^{40}K. Figure 3 shows the spatial concentration of ^{40}K obtained from the interpolation of the obtained results.

For most of the samples (90 %), the ^{226}Ra concentrations are lower than the ^{228}Ra concentrations. The ratio of radium isotopes ^{228}Ra/^{226}Ra varied between 0.16 and 4.37,

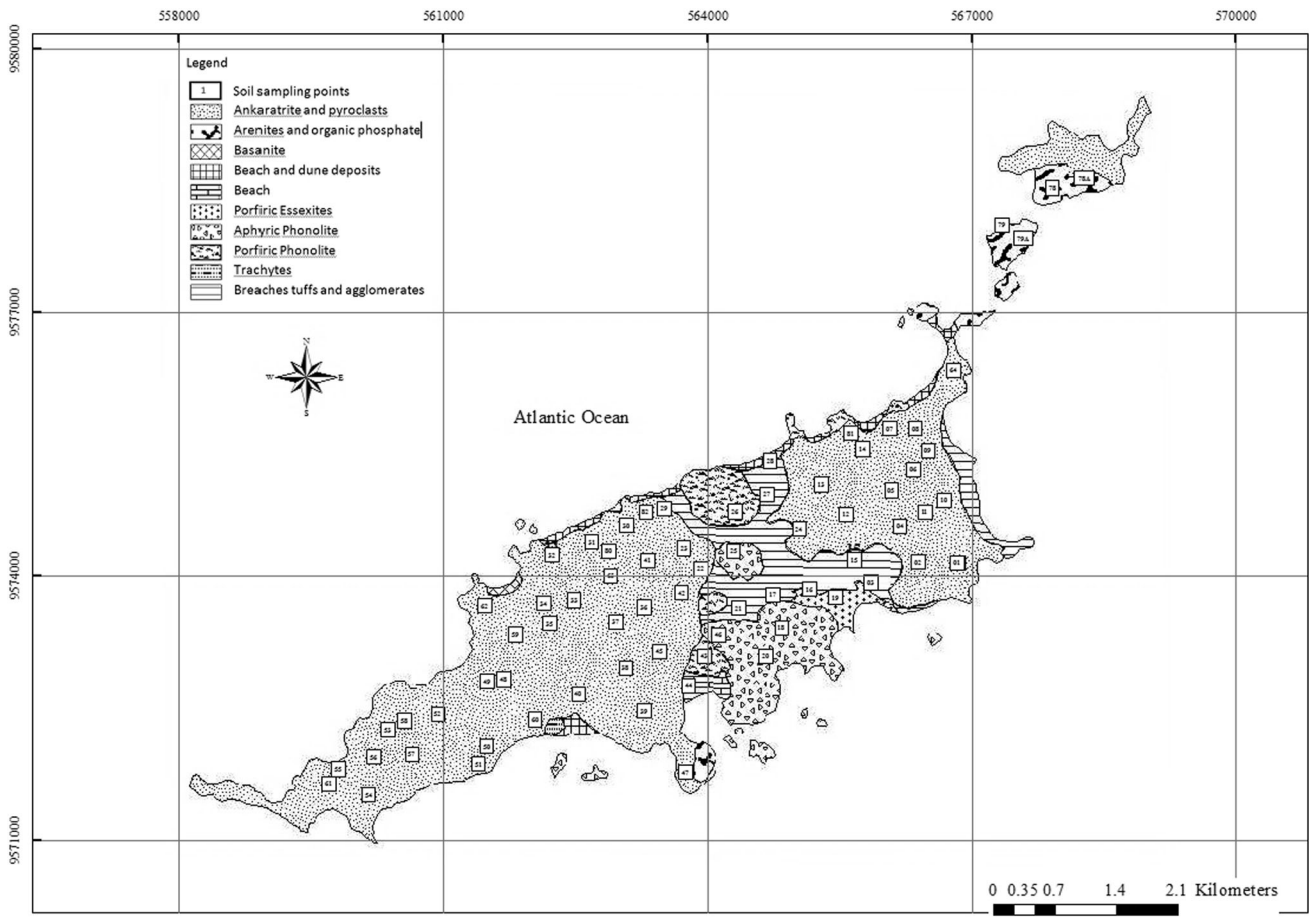

Fig. 1 Geologic map (adapted from Almeida [33]) and locations of the sampling points

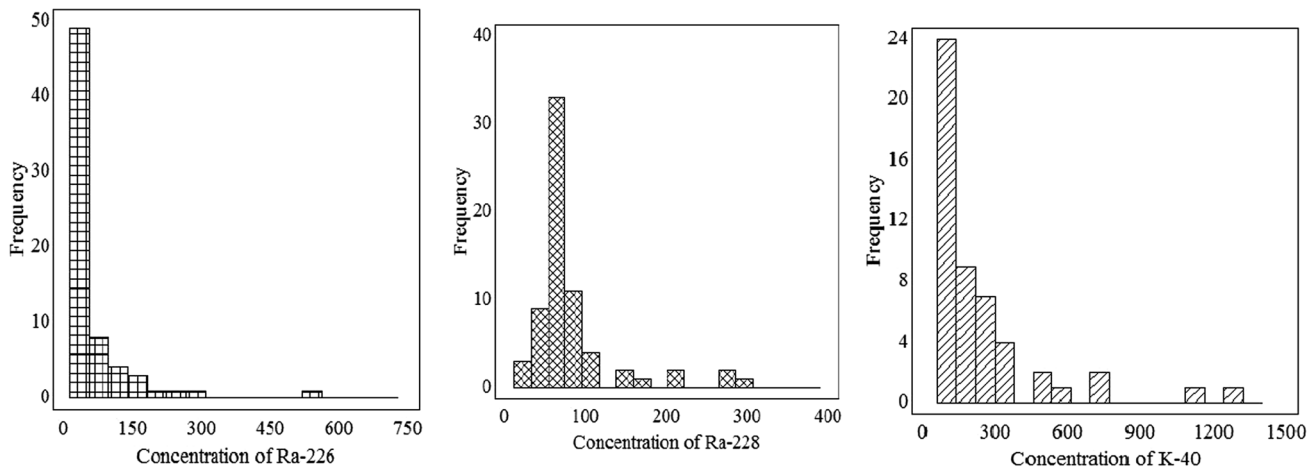

Fig. 2 Distribution of natural radionuclides in the FNA soil samples

with average and median values of 1.85 and 1.75, respectively. The high value of the radium isotope ratio resulted from the high stability of thorium during weathering processes. Although thorium remains stable in all minerals, uranium is only stable in zircon and monazite and is very soluble in other minerals containing carbonates, sulfates and phosphates [34]. Thus, during the weathering process, uranium tends to weather out (as the soluble form of uranyl) as metamorphism begins while thorium remains in the rock, resulting in a higher thorium concentration in the igneous rocks than in the sedimentary rocks. However, this trend only changed for samples collected from the

Table 1 Descriptive statistical analysis of the natural radionuclide concentrations (in Bq kg^{-1}) in the soils from Fernando de Noronha

	^{226}Ra	$^{232}Th(^{228}Ra)$	^{40}K
Minimum	4.7	10.5	12.7
Maximum	550	298	1280
Average	62	82	179
Median	43	70	108
CV (%)*	125	68	129

* CV coefficient of variation

sandstones and phosphates of Rata Island, where the concentration of ^{226}Ra activity was higher than that of ^{228}Ra. This outcome can be attributed to the greater affinity of ^{238}U than ^{232}Th to the biological phosphate (guano) present on this island [35].

The highest concentrations of ^{226}Ra and ^{228}Ra were found in some small areas, as shown in Figs. 4 and 5.

For comparison, the reported average radionuclide concentrations in rocks from different countries are shown in Table 2.

The average concentrations of ^{232}Th (^{228}Ra) and ^{226}Ra in this survey are higher than the values reported world wide, while the average concentration of ^{40}K is lower than average. Regarding natural radioactivity, igneous rocks usually exhibit higher contents of Th and U than sedimentary rocks, except for some shale and phosphate rocks [16]. Thus the relatively high concentrations of ^{226}Ra and Th^{232} (Ra^{228}) in the soil of the FNA are related to the igneous origins of the archipelago. On the other hand, the low ^{40}K concentration of the soil in the FNA is related to the low feldspar content of most of the rocks on the island.

Considering the fractionation processes that occur as a result of radioactive decay and the distinct chemical characteristics of the decay products, radioactive equilibrium among the radionuclides of uranium and thorium decay chains cannot be expected to exist in environmental samples. Due to its greater environmental mobility in comparison with ^{238}U and ^{230}Th, ^{226}Ra may be present at slightly different concentrations than ^{238}U in soil samples [2, 16]. Although similar considerations can be made for ^{232}Th and ^{228}Ra concentrations, the ^{228}Ra half-life (5.75 years) may allow a quite similar concentration values for these radionuclides in soils. Despite of the above considerations, an approach of ^{238}U and ^{232}Th concentrations can be estimated considering the equilibrium in the chains. The concentrations of ^{238}U (^{226}Ra) and ^{232}Th (^{228}Ra) can be converted to the elemental concentrations (μg g^{-1}) of the radionuclides by using ^{238}U and considering that

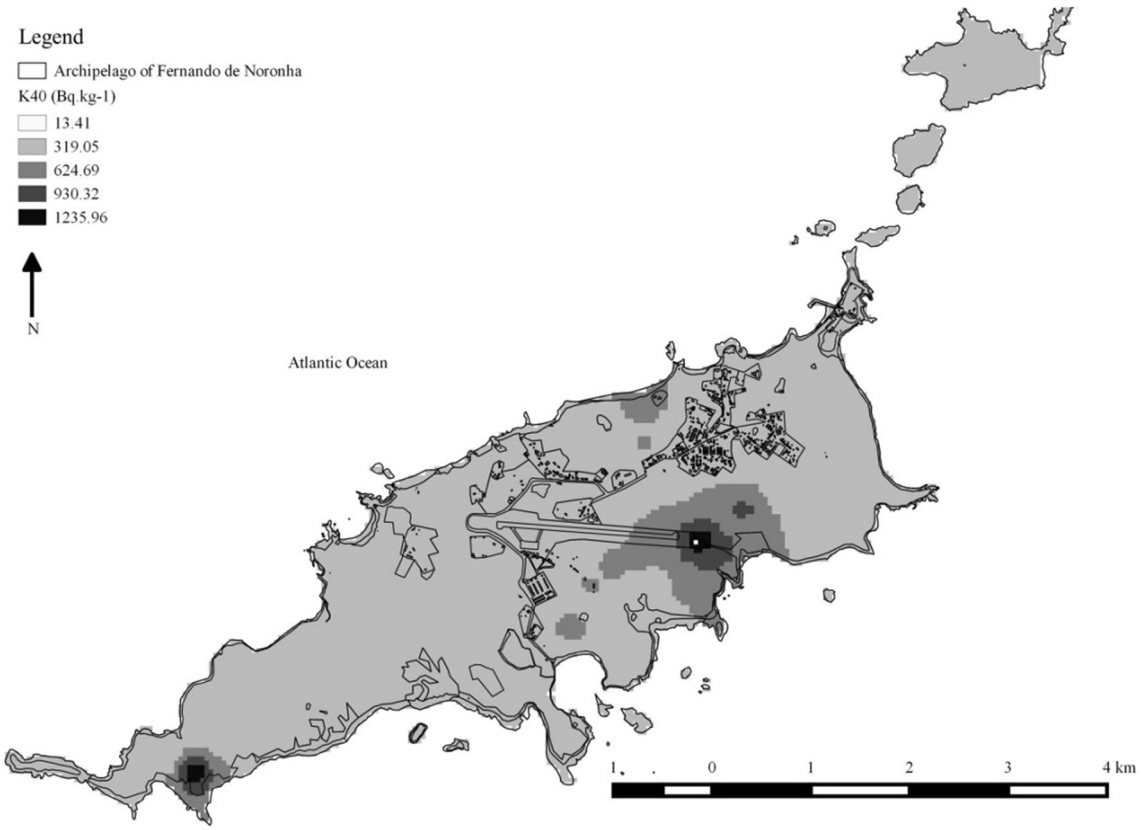

Fig. 3 Distribution of ^{40}K in the soils of the Fernando de Noronha Archipelago

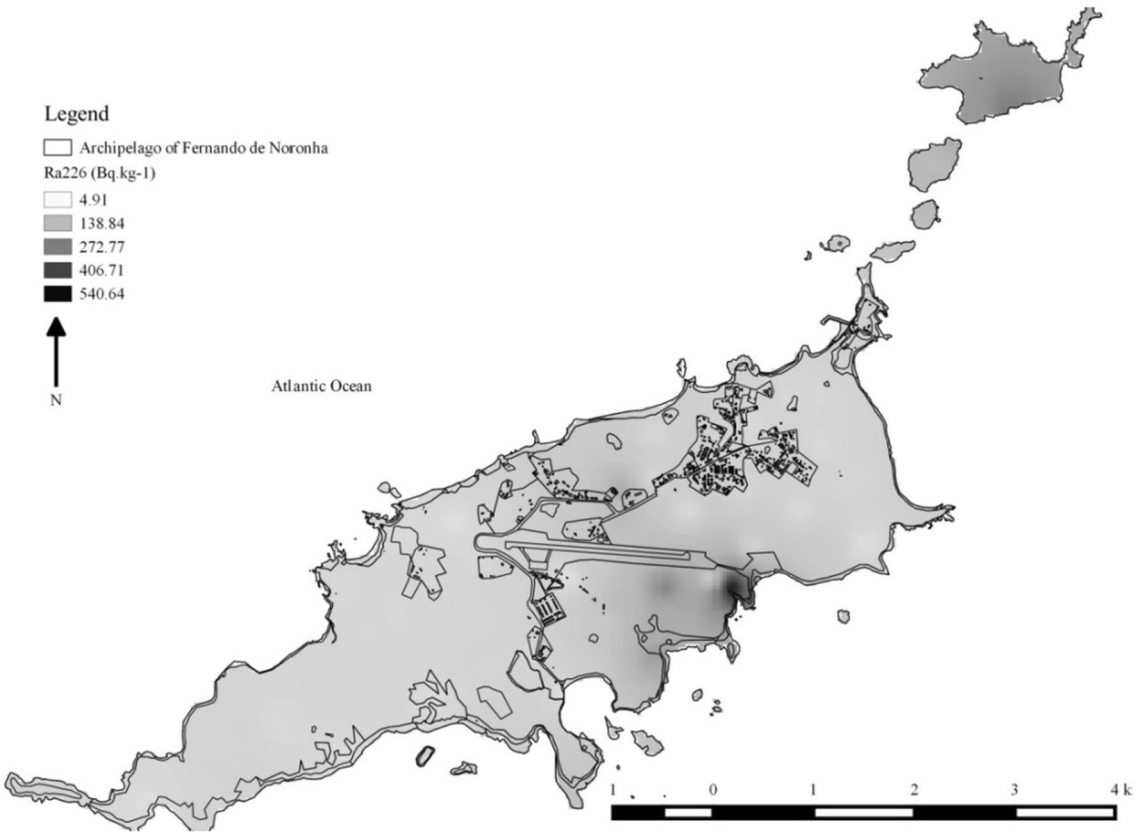

Fig. 4 Spatial distribution of ^{226}Ra in the soils of the FNA

1 μg g^{-1} of ^{238}U is equivalent to 12.35 Bq of ^{238}U kg^{-1}. For ^{232}Th, 1 μg g^{-1} of ^{232}Th was considered to correspond to 4.06 Bq of ^{232}Th kg^{-1}. The percentage of K (%) was obtained by dividing the ^{40}K concentration by 2.76, and the concentration of K was obtained by dividing the ^{40}K concentration by 2.76 10^{-2}.

The elemental concentrations ranged from 0.38 to 44.5 μg g^{-1}, with an average concentration of 3.00 and a median concentration of 5.02 μg g^{-1} for ^{238}U, from 2.59 to 73.3 μg g^{-1}, with an average concentration of 20.2 μg g^{-1} and a median concentration of 16.5 μg g^{-1} for ^{232}Th, and from 0.05 to 4.64 %, with an average concentration of 0.65 % and a median concentration of 0.32 % for K. The worldwide median values reported by UNSCEAR (2000) are 35, 30 and 400 Bq kg^{-1} for ^{238}U, ^{232}Th and ^{40}K respectively, which correspond to 2.8 mg kg^{-1}, 7.4 mg kg^{-1} and 1.4 %. Thus, the U and Th contents in the FNA soil are higher, while the K content is smaller than the median value for these radionuclides worldwide. The mean value of the Th/U ratio (5.6 ± 2.6) is higher than the theoretically expected Th/U ratio of 2.8 for the Earth's crust. The average K/U ratio found for the FNA (1.7 10^3, with a median value of 9.7 10^2) is lower than the estimated terrestrial and ocean basalt values of approximately 1.0 10^4 and lower than the lower value of the range for the Earth (from 0.5 10^4 to 1.4 10^4) [36]. The low K/U ratio observed for FNA is consistent with the depletion of K observed in the archipelago. This value is similar to the determined value of 2.8 10^3 for the superficial soil of Cyprus [37] The K/Th values range from 1.9 10^1 to 3.2 10^3, with an average of 3.9 10^2 and a median of 8.5 10^2.

From the seventy investigated samples, ^{137}Cs was detected in only sixteen samples (23 % of the analysed samples). The statistical code ProUCL 5.0 can be used to perform statistical analysis of a data population with non-detected values. Because the number of non-detected data points is not lower than ten [31], ^{137}Cs descriptive analysis was performed. The ^{137}Cs was calculated using the Kaplan–Meier method, which resulted in a mean of 0.62 Bq kg^{-1} and a range of <0.3–2.4 Bq kg^{-1}. The 50 % percentile and 99 % percentile are 1.1 and 2.4 Bq kg^{-1}, respectively. Considering only the sixteen detected values, the values range from 0.4 to 2.4 Bq kg^{-1}, with a mean value of 1.04 and a median value of 0.97 Bq kg^{-1}. These data are comparable with the data obtained by LaBrecque and Cordoves [22] for topsoil in Venezuela, where only 28 % of the 90 analysed samples had ^{137}Cs values higher than 2 Bq kg^{-1}. The values of the detected samples varied between less than 2 and 33.4 Bq kg^{-1}. The low ^{137}Cs content in the FNA soil confirms the predictions reported

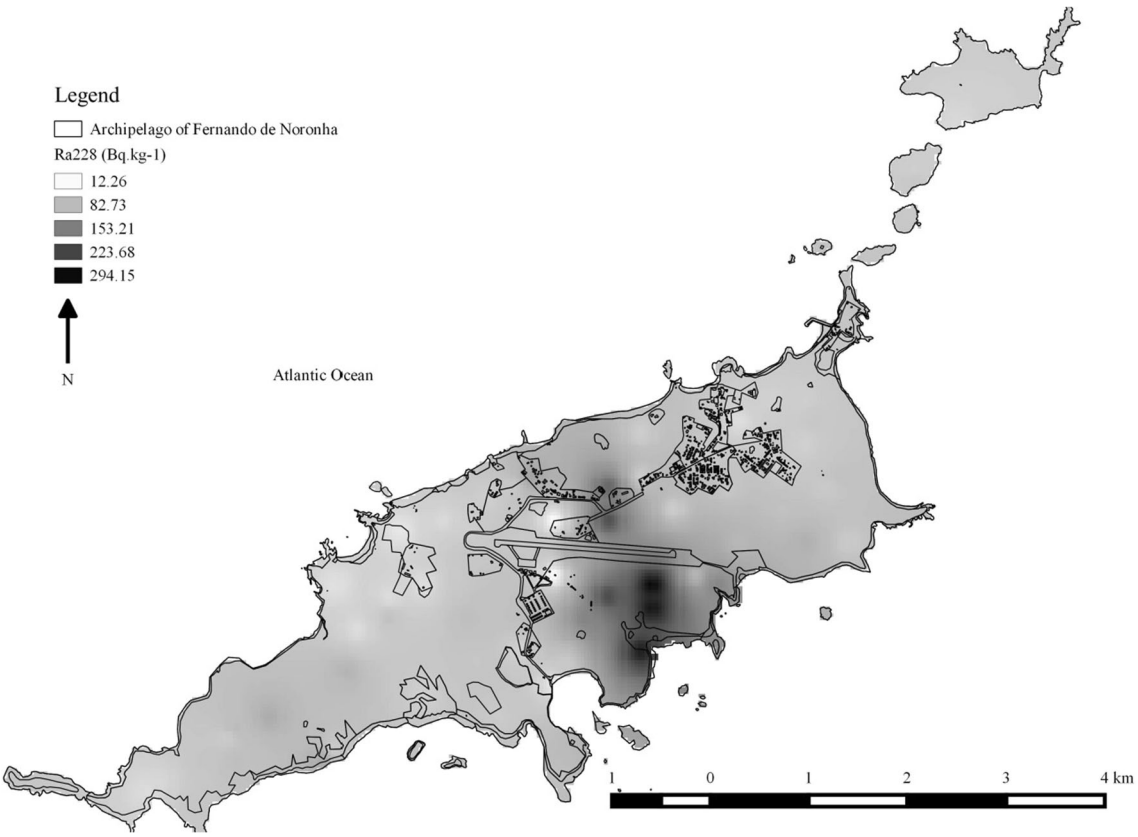

Fig. 5 Spatial distribution of ^{228}Ra in the FNA

Table 2 Worldwide averages of soil radionuclide concentrations

Average concentration (Bq kg^{-1})				Location	References
^{226}Ra	^{232}Th (^{228}Ra)	^{40}K	^{137}Cs		
62	82	179	<0.3–2.4	Fernando de Noronha	This survey
30	35	400	–	Worldwide	[16]
29	33	448	2–25	Turkey	[2]
23	31	300	2–5.4	Texas, USA	[36]
6.5	61.7	379.8	<1–2.8	Tamil Nadu, India	[15]
19.2	24.2	304	–	Uzhgorod City, Ukraine	[38]
43	21	283	0.9–148	Norway	[40]
29.2	47.8	708	–	Rio Grande do Norte, Brazil	[41]

by UNSCEAR [21] regarding the low level of fallout radionuclides that reached the equatorial region.

Relationships with the main geological formations

The maximum and minimum values and the average and median concentrations of radionuclides obtained in this study for the main formations of FNA, Quixaba (Q), Remédios (R) and Modern Sediments (MS) are summarized in Table 3.

Most of samples were collected from the Quixaba Formation (52 samples, corresponding to 74 % of all the samples), 14 samples were collected from the Remédio Formation, only 4 samples were collected from the Modern Sediments Formation and no samples were collected from the San José Formation.

ANOVA using Fisher's test showed that the mean of the data from the Quixaba Formation (Q) for ^{226}Ra is lower than the mean of the data from the Remédios and Modern Sediments Formations, which had similar means (with 95 % of significant). The ^{228}Ra and ^{40}K concentrations are both higher for the Remédios Formation than for the Quixaba and Modern Sediments Formations, which are similar. The box-and-whisker graphs in Fig. 6 show the

Table 3 Average, median and maximum and minimum ^{226}Ra, ^{228}Ra and ^{40}K concentrations in Bq kg^{-1} according to geological formation: Quixaba (Q), Remédios (R) and Modern Sediments (MS)

Formation	Average	Median	CV (%)	Minimum	Maximum
^{226}Ra					
Q	40.3	34	88	4.7	268
R	130	90	104	29	550
MS	108	108	72	12	201
^{228}Ra					
Q	64.5	65	31	10.5	109
R	155.2	144	55	61	298
MS	51	53	32	29	68
^{40}K					
Q	115	67	134	12.6	1100
R	425	354	77	39	1280
MS	155	167	66	35	248

distributions of the concentration data obtained from the different formations.

Thus, except for the similar data of Remédios and Modern Sediments Formations for ^{226}Ra, the data set from the Remédios Formation, which is the oldest formation in the archipelago, had the highest concentrations of natural radionuclides.

The greater affinity of ^{238}U than ^{232}Th for biological phosphate (guano) in the Modern Sediment Formation was underlined by the nonparametric Wilcoxon–Mann–Whitney (WMW) test, which showed a significant difference (95 % significance) among the data of the ^{228}Ra/^{226}Ra ratios of the Modern Sediment Formation (data ranging from 0.24 to 2.38, average of 0.95 and median of 0.58) and the ^{228}Ra/^{226}Ra ratios from Quixaba (data ranging from 0.16 to 4.36, average of 1.99 and median of 1.79) and Remédios (data varying between 0.26 and 2.57, average and median values of 1.60 and 1.71, respectively). No significant difference was observed between the radium isotope ratios of the Quixaba and Remédios Formations. Considering the elemental concentrations, the WMW test indicated a significant difference between the U/Th ratios of the sediment from the Modern Sediment Formation and the sediments from the Quixaba and Remédios Formations, which are similar. However, the WMW test showed no significant differences among the K/U ratios and among the K/Th ratios for the three geological formations.

Absorbed gamma dose rate and corresponding annual effective dose

The gamma dose rate can be estimated from the concentrations of soil ^{226}Ra, ^{232}Th (^{228}Ra), and ^{40}K (Eq. 1) [21]. Using this approach, the infinite source geometry, homogeneous distribution of radionuclides and radioactive equilibrium in the ^{238}U and ^{232}Th decay series should be considered. For this estimation, the concentrations of natural radionuclides are multiplied by the corresponding values of the kerma rate conversion factors (K) in nGy h^{-1} per Bq kg^{-1} (Eq. 1).

$$D_{nat}\left(\eta G.h^{-1}\right) = 0.0417 \times C_{K40} + 0.463 \times C_{Ra226} + 0.604 \times C_{Ra228} \quad (1)$$

where D is the rate of the dose in the air (nGy h^{-1}), the Ks values were obtained by Saito and Jacob [39] and correspond to 0.463 for the ^{238}U series, 0.604 for the ^{232}Th series and 0.0417 for ^{40}K, respectively, and C is the radionuclide concentration in Bq kg^{-1}.

The estimated gamma rate dose varied between 27 and 369 nGy h^{-1}, with mean and median values of 86 and 72

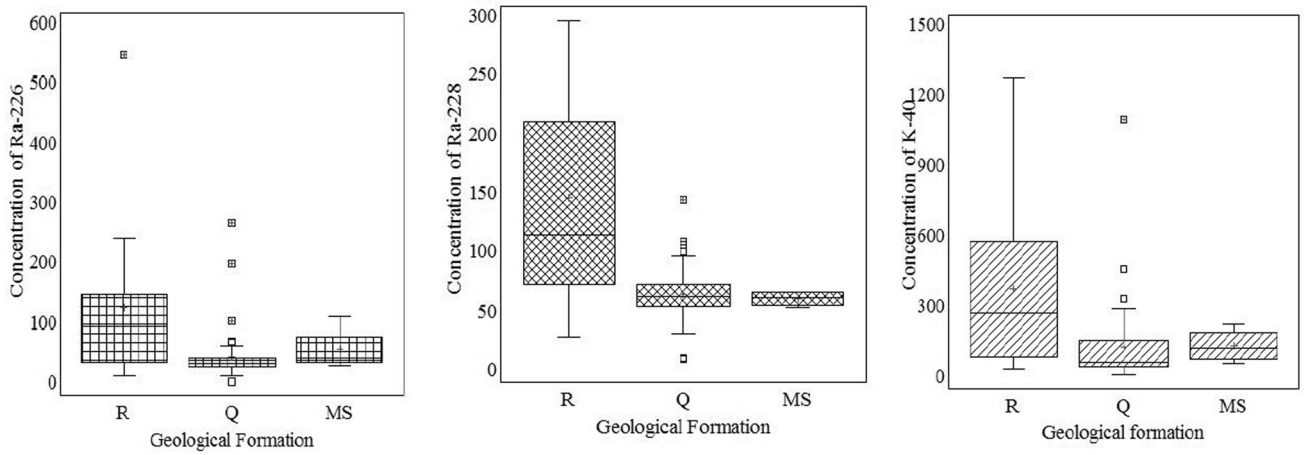

Fig. 6 Distributions of ^{226}Ra, ^{228}Ra and ^{40}K data in the main geological formations of the FNA (*R* Remédios formation, *Q* Quixaba formation and MS Modern sediments formation)

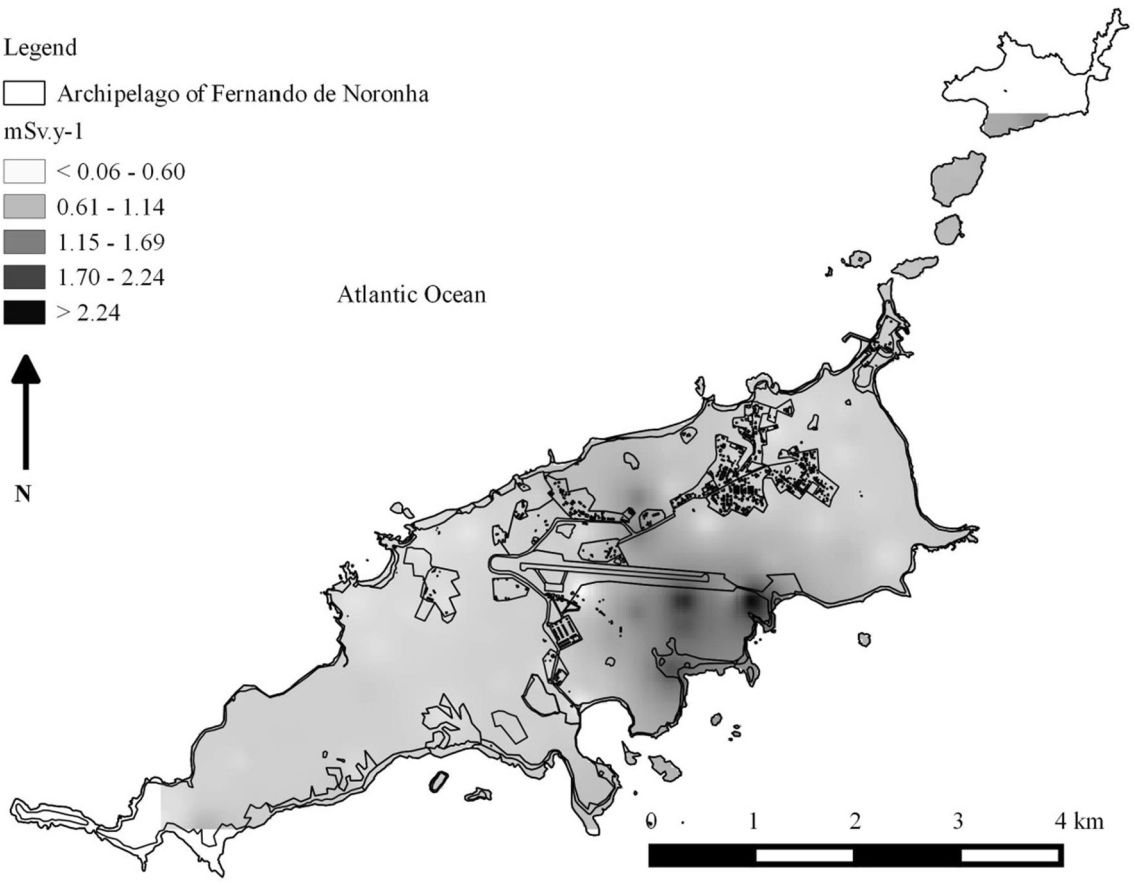

Fig. 7 Dose resulting from the natural radionuclides in the soils from the FNA

nGy h^{-1}, respectively. The range of values estimated for FNA surpass the range of variability of the measured absorbed dose rates in the air cited by UNSCEAR [16], which range from 10 to 200 nGy h^{-1}, and the median values are slightly higher than the worldwide median of 51 nGy h^{-1}.

The annual effective dose due to outdoor external exposure E_m (mSv a^{-1}) from the soil can be calculated using the following equation:

$$E_m = D_{nat} \times O_f \times 0.7\,Sv\,Gy^{-1} \times 10^{-6} \quad (2)$$

where O_f is the fraction of the year for which a hypothetical member of the public is exposed to the outdoors. The suggested O_f value is 20 % (0.2), which means that each individual spends an average of 1752 h outdoors each year. The factor 0.7 Sv Gy^{-1} is the conversion factor from the absorbed dose in the air to the effective dose received by adults at a height of 1 m above the ground surface [15].

The calculated median value of the annual effective external dose outdoors was 0.08 mSv a^{-1}, with an average value of 0.10 mSv a^{-1} and a range of 0.03–0.45 mSv a^{-1}.

If no flooring cover would be considered, and considering that individuals spend 80 % of their time indoors, the external dose due to soil exposure indoors varies between 0.17 and 2.26 mSv a^{-1}. Thus, the average and the median total contributions of natural radionuclides in the soil to the total external effective dose are 0.52 and 0.31 mSv a^{-1}, respectively, which are similar to the worldwide values of 0.48 and 0.45 mSv a^{-1} established be UNSCEAR [15]. However, one location with Aphyric phonolites in the south and middle area of the main island would present a total annual external dose of between 1.1 and 2.3 mSv, which surpasses the worldwide average external dose of 0.3–0.6 mSv (Fig. 7).

Conclusions

This research generated a database on naturally occurring radionuclides and ^{137}Cs levels in the soils of the Fernando de Noronha archipelago, whose soils were analysed using gamma spectrometry. The median concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the soil samples from FNA were 43, 70 and 108 Bq kg^{-1}, respectively. The contents of ^{226}Ra and ^{232}Th in the soil samples were higher than the world median concentrations of 30 for ^{226}Ra and 35 for ^{232}Th

Bq kg^{-1}. However, due its rock characteristics, the levels of ^{40}K in the soils from the FNA are four times lower than the worldwide ^{40}K estimate of 400 Bq kg^{-1}. The distribution of radionuclides in the soils of the archipelago is very heterogeneous, which could be attributed to geological characteristic of the archipelago. The soils from the Remédios Formation, which is the oldest formation of the archipelago, have the highest concentrations of natural radionuclides. No significant differences were observed between the U/Th ratios of the Quixaba and Remédios Formations. However, this trend changed for samples collected from the soils overlying sandstones and phosphates on Rata Island (Modern Sediment), where the concentrations of ^{226}Ra were higher than the concentrations of ^{228}Ra due to the greater affinity of ^{238}U than ^{232}Th for sorption to the biological phosphate (guano) present on this island. In this study, the average concentrations of 3.00 μg g^{-1} for ^{238}U, 20.2 μg g^{-1} for ^{232}Th and 0.65 % for K were derived. The measured elemental ratios exhibit a wide range of values. Specifically, the Th/U ratios varied between 0.5 and 13, with an average value of 5.6, which was higher than the expected value when considering the average value for the Earth's crust of approximately 2.8. This result potentially indicates that the fractionation of radioactive isotopes during weathering occurred. The K/Th ratio ranged from 1.9 10^1 to 3.2 10^3, with an average value of 3.9 10^2, and K/U ratio varied between 6.9 10^1 and 1.2 10^4, with an average value of 1.7 10^3, which is similar to the ratio observed in the soils of Cyprus Island. The K/U and K/Th ratios were similar for the three geological formations. Although ^{137}Cs was detected in only sixteen samples, the range of values from less than 0.3 to 2.4 Bq kg^{-1} can be used as a reference for assessing the contamination of FNA soils. The low soil ^{137}Cs content confirms the prediction of low radionuclide fallout in equatorial regions. A gamma dose rate of 72 nGy h^{-1} was estimated, which slightly exceeded the world median rate of 51 nGy h^{-1}. The main contributor for the dose rate is the Th decay series, which accounts for 58 %, followed by the U-series and ^{40}K, which account for 33 and 9 %, respectively. The level of external exposure to naturally occurring radionuclides in soils indicates that the population in most areas of the FNA are exposed to normal radiation levels. However, a small area containing Aphyric phonolites has higher levels of external exposure to radiation than the worldwide range.

References

1. Turhan Ş, Köse A, Varinlioğlu A, Şahin NK, Arıkan İ, Oğuz F, Yücel B, Özdemir T (2012) Distribution of terrestrial and anthropogenic radionuclides in Turkish surface soil samples. Geoderma 187–188:117–124
2. Laubenstein M, Plastino W, Povinec PP, Fabbri V, Aprili P, Balata M, Bella F, Cardarelli A, Deo M, Gallese B, Ioannucci L, Nisi S, Antonecchia D, Del Pinto C, Giarrusso G (2013) Radionuclide mapping of the Molise region (Central Italy) via gamma-ray spectrometry of soil samples: relationship with geological and pedological parameters. J Radioanal Nucl Chem 298:317–323
3. Akhtar N, Tufail M, Ashraf M (2005) Natural environmental radioactivity and estimation of radiation exposure from saline soils. Int J Environ Sci Technol 1:279–285
4. Garba NN, Ramli AT, Saleh MA, Sanusi MS, Gabdo HT (2015) Terrestrial gamma radiation dose rates and radiological mapping of Terengganu state, Malaysia. J Radioanal Nucl Chem 303:1785–1799
5. Matiullah A, Ahad A, Rehman S, Rehman S, Faheem M (2004) Measurement of radioactivity in the soil of Bahawalpur division, Pakistan. Radiat Prot Dosim 112:443–447
6. Ahmad N, Matiullah A, Khatibeh AJAH (1997) Indoor radon levels and natural radioactivity in Jordanian soils. Radiat Prot Dosim 71:231–233
7. Al-Jundi J (2002) Population doses from terrestrial gamma exposure in areas near to old phosphate mine, Rusaifa, Jordan. Radiat Meas 35:23–28
8. Fatima I, Zaidi J, Arif M, Daud M, Ahmad SA, Tahir SN (2008) Measurement of natural radioactivity and dose rate assessment of terrestrial gamma radiation in the soil of southern Punjab, Pakistan. Radiat Prot Dosim 128:206–212
9. Karahan G, Bayulken A (2000) Assessment of gamma dose rates around Istanbul (Turkey). J Environ Radioact 47:213–221
10. McAulay IR, Moran D (1988) Natural radioactivity in soil in the Republic of Ireland. Radiat Prot Dosim 24:47–49
11. Quindos LS, Fernandez PL, Soto J, Rodenas C, Gomez J (1994) Natural radioactivity in Spanish soils. Health Phys 66:194–200
12. Saleh IH, Hafez AF, Elanany NH, Motaweh HA, Naim MA (2007) Radiological study on soils, foodstuff and fertilizers in the Alexandria region, Egypt. Turk J Eng Environ Sci 31:9–17
13. Tahir SNA, Jamil K, Zaidi JH, Arif M, Ahmed N, Ahmad SA (2005) Measurements of activity concentrations of naturally occurring radionuclides in soil samples from Punjab province of Pakistan and assessment of radiological hazards. Radiat Prot Dosim 113:421–427
14. Pillai GS, Hameed PS, Khan SMM (2016) Natural radioactivity levels in the soils and human risk assessment in Tiruchirappalli district (Tamil Nadu, India). J Radioanal Nucl Chem 307:1265–1277
15. UNSCEAR (2000) Sources and effects of ionizing radiation, Report to the general assembly, with scientific annexes. United Nations Scientific Committee on effects of atomic radiation, vol 1. United Nations Publication, New York
16. Zhao Y, Yan D, Zhang Q, Zhan J, Hua H (2012) Spatial distributions of^{137}Cs in surface soil in Jing-Jin-Ji Region, North China. J Environ Radioact 113:1–7
17. Godoy JM, Schuch LA, Nordemann DJR, Reis VRG, Ramalho M, Recio JC, Brito RRA, Olech MA (1998)^{137}Cs, 226,^{228}-Ra,^{210}Pb and 40 K Concentrations in Antarctic soil, sediment and selected moss and lichen samples. J Environ Radioact 41:33–45
18. Ritchie JC, McHenry JR (1990) Application of radioactive fallout ^{137}Cs for measuring soil erosion and sediment accumulation rates and patterns: a review. J Environ Qual 19:215–233
19. Ritchie JC, Nearing MA, Rhoton FE (2009) Sediment budgets and source determinations using fallout Caesium-137 in a semi-arid rangeland watershed, Arizona, USA. J Environ Radioact 100:637–643

20. Walling DE, He Q (1999) Improved models for estimating soil erosion rates from Caesium-137 measurement. J Environ Qual 28:611–622
21. UNSCEAR (1969) Report. Annex A: Radioactive contamination of the environment by nuclear tests. United Nations Scientific Committee on effects of atomic radiation. United Nations Publication, New York
22. LaBreque JJ, Cordoves PR (2005) Caesium-137 spatial activity in surface soils near and surrounding the Gurí Reservoir (Venezuela). J Radioanal Nucl Chem 265:91–94
23. Almeida FFM (1955) Geologia e petrologia do arquipélago de Fernando de Noronha. Departamento Nacional de Produção Mineral-DNPM, Rio de Janeiro
24. Almeida FFM (2002) Arquipélago de Fernando de Noronha. Schobbenhaus C, Campos D A, Queiroz ET, Winge M, Berbert-Born M. (Eds.). Serviço Geológico do Brasil-CPRM, Brasília
25. Batistella AM (1993) Cartografia ecológica de Arquipélago de Fernando de Noronha. Master of Science Thesis. Universidade de São Paulo. São Paulo
26. IBGE. Instituto Brasileiro de Geografia e Estatística. http://www.ibge.gov.br/cidadesat/painel/painel.php?codmun=260545. Accessed 27 Jan 2016
27. Cordani UG (1970) Idade do vulcanismo no Oceano Atlântico Sul, vol 1. Instituto de Geociências e Astronomia, Boletim IGA, Brasília
28. Oliveira SMB, Pessenda LCR, Gouveia SEM, Favaro DIT (2009) Evidência geoquímica de solos formados pela interação de guanos com rochas vulcânicas, Ilha Rata, Fernando de Noronha (PE). Rev Inst Geoc 9:3–12
29. Canet A, Jacquemin R (1990) Methods for measuring radium isotopes: Gamma spectrometry In: Environmental behaviour of radium, Technical Report Series No. 310. IAEA, Vienna, pp 189–204
30. International Atomic Energy Agency (1989). Measurement of radionuclides in food and the environment. Technical Report Series 295, A Guidebook. IAEA
31. Singh A and Singh A (2013) Statistical software for environmental applications for data sets with and without non detect observations. ProUCL. U.S. Environmental Protection Agency. EPA/600/R-07/041
32. U.S. Environmental Protection Agency (EPA). 2006b. Data Quality Assessment: Statistical Methods for Practitioners, EPA QA/G-9S. EPA/240/B-06/003. Office of Environmental Information, Washington, DC. Available from http://www.epa.gov/quality/qs-docs/g9s-final.pdf
33. QGIS Development Team, 2009. QGIS Geographic information system. Open Source Geospatial Foundation. URL http://qgis.osgeo.org
34. Ivanovich M, Harmon RS (1992) Uranium-series disequilibrium: applications to earth, marine and environmental sciences, 2nd edn. Oxford Univ Press, New York
35. Almeida FFM (2000) The Fernando de Noronha archipelago. In: Schobbenhaus,C., Campos DA, Queiroz,ET, Winge,M, Berbert-Born,M (Edit.) Sítios Geológicos e Paleontológicos do Brasil. http://sigep.cprm.gov.br/sitio066/sitio066english.htm. Accessed 27 Jan 2000
36. Hannan M, Wahid K, Nguyen N (2015) Assessment of natural and artificial radionuclides in mission (Texas) surface soils. J Radioanal Nucl Chem 305:573–582
37. Tzortzis M, Tsertos H (2004) Determination of thorium, uranium and potassium elemental concentrations in surface soils in Cyprus. J Environ Radioact 77:325–338
38. Potoki I, Parlag O, Maslyuk V, Lengyel A, Torich Z (2015) Long-term monitoring of natural radionuclides in Uzhgorod city, Ukraine. J Radioanal Nucl Chem 306:249–255
39. Saito K, Jacob P (1995) Gamma-ray fields in the air due to sources in the ground. Radiat Prot Dosim 58:29–45
40 Dowdall M, Gerland S, Lind B (2003) Gamma-emitting natural and anthropogenic radionuclides in the terrestrial environment of Kongsfjord, Svalbard. Sci Total Environ 305(1–3):229–240
41 Malanca A, Gaidolfi L, Pessina V, Dallara G (1996) Distribution of 226Ra, 232Th, and 40K in soils of Rio Grande do Norte (Brazil). J Environ Radioact 30(1):55–67