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Radiometric characterization of the different environmental components of a high background radiation area: a comprehensive review

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Abstract— Airborne radiometric surveys showed that Aja heights, of granitic composition, represent a radiometric anomaly area and should be of interest for detail study in a purpose of radiation protection. Ground surveys and radiometric analysis of rocks, surface soil, building materials and groundwater samples have been conducted. Radon-222 has been measured in groundwater and in the atmosphere (indoor and outdoor) of the inhabited area. Root uptake of natural uranium by vegetation grown in farms lie in the foot of the granitic massif has been investigated.

Ground surveys showed an average effective dose rate, due to terrestrial γ-radiation, of 1.49 mSv/y. This level is about 3.3 times greater than the world average external effective dose rate (0.46 mSv/y) that estimated in normal background areas. The average concentrations (249 and 383 Bq/kg, respectively) of ^{238}U and ^{232}Th in the collected rock samples were higher than their worldwide average (about 61 Bq/kg for both radionuclides) in granites. The average concentrations of ^{238}U and ^{232}Th (156 and 187 Bq/kg, respectively) in the top-soil samples were greater than the normal soil-background of 40-50 Bq/kg for both radionuclides. Samples of building materials, collected from utilized quarries dispersed randomly in the area, indicated that the highest activity was found in the fragmented granites and the lowest activity was found in the black rock materials. The average activity concentrations of ^{238}U, ^{234}U, ^{226}Ra and ^{228}Ra in the groundwater were 0.40, 0.77, 0.29 and 0.46 Bq/L, respectively. These values exceeded the national guideline values set out for their concentration in drinking water. The average ^{222}Rn concentrations in the groundwater and in the indoor and outdoor atmospheric air were 30.3 kBq/m^3, 54.6 Bq/m^3 and 10.5 Bq/m^3, respectively. Root uptake of uranium by vegetation grown in the region showed demonstrable differences in uranium concentration between plant parts and types, which generally, followed the sequence: roots > leaves > stems or branches > fruits. Plant-soil transfer factors, based on the edible parts of the plant, for uranium isotopes were calculated. Some conclusions and recommendations are presented.

Keywords—environmental radioactivity, uranium, radium, radon, radiation exposure, environmental pollution.

I. Introduction

Several studies proved that igneous rocks of granitic composition contain higher levels of uranium and thorium, compared to rocks of other composition [e.g. 1-3]. The Arabian shield represents a basement of igneous and metamorphic rocks and lies in the western part of Saudi Arabia. It occupies about one-third of the kingdom area and contains many randomly dispersed cities and villages. Systematic airborne radiometric exploration of the Shield indicated that Aja granitic massif of Hail province is a radiometric anomaly area and should be of interest for detail study. A national research project (grant No.: 8-ENV-128-3, KACST, SA) was dedicated to conduct the recommended study. The study provided detail radiological data which were essential in providing smart recommendations to relevant authorities, decision makers and inhabitants to react with the raised situation, in a purpose of radiation protection. This article presents a comprehensive review summarizing the whole work.

II. Nature of the study area

A. Topography and hydrology

The study area includes Aja massif and the surrounding inhabited zones [4]. It is bounded by lat. 27o 00’ and 28o 05’, and long. 41o 00’ and 42o 15’ E, and occupies an area of approximately 10500 km2, in the northern part of the middle region of Saudi Arabia (Fig. 1). It represents almost the major part of Hail quadrangle (Sheet 27E; international index NG-37-4) that bounded by lat. 27o 00’ and 28o 00’ N., and long. 40o 30’ and 42o 00’E. Hail city lies at the foot of the Aja massif, in the central part of the study area, at an elevation of about 980 m above sea level. It is one of the largest cities in north-central Saudi Arabia.

Annual rainfall occurs mostly between November and March, resulting, on occasion, in temporary lakes that evaporate to form small sabkhahs. Drainage in the area is controlled by the rugged Aja range, and wadis, intermittently filled by runoff, radiate towards the west, northwest, east and northeast (major portion). The use of groundwater for farming is rapidly lowering the water table, as little if any aquifer recharge is taking place in the extremely arid climatic conditions. Natural vegetation is restricted to perennial shrubs and seasonal grasses. Agricultural activity is limited, although significant areas of cultivation have developed, since the mid-1970s, in the northeastern corner of the study area (Fig. 1), where the groundwater is comparatively more available.

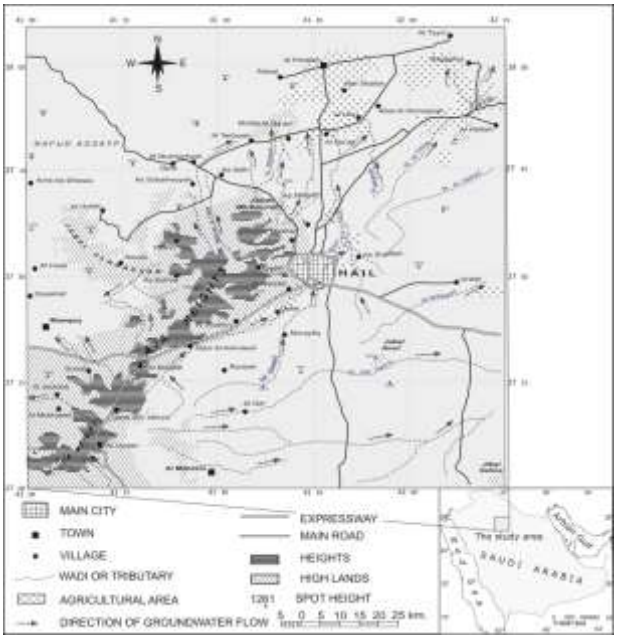

Fig. 1. Topographical map of the study area showing the groundwater flow directions.

B. Geology

Quaternary surficial deposits overlie most of the Phanerozoic bedrocks and parts of the Proterozoic basement of the study area [4]. The deposits predominantly consist of eolian sand, and small occurrences of gravel, alluvium, and sabkhah. The region is underlain by late Proterozoic volcano-sedimentary and intrusive rocks, and a Cambrian to early Silurian succession of essentially sedimentary rocks. The Proterozoic rocks crop out in the south western part of the study area, and predominantly consist of relatively young granitic intrusions. They include monzogranite of the Rughayghith suite and more evolved alkali-feldspar granites of the Abanat suite. The suite occurs as large batholiths in the center of the Hail quadrangle, where they form the topographically conspicuous Aja massif (Figs. 1 and 2).

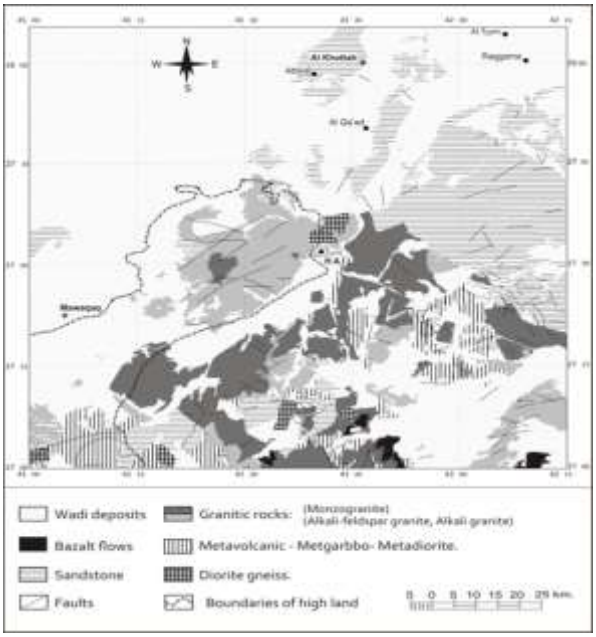

Fig. 2. Geological map of the study area

III. Ground surveys

Ground radiometric surveys were conducted on different sites scattered in the study area [4] to measure the outdoor γ-ray dose rate. It was measured 1 meter above the ground and the obtained data were arranged in relation to lithology. The average dose rate was 1.18, 1.85 and 2.57 mSv/y on the soil of the inhibited and cultivated zones, Wadi deposits of Aja heights and outcropping rocks, respectively. These values are higher than the global average value (0.46 mSv/y) of normal background areas. The highest exposure rate was found on the later rocks with some extreme values in the rugged zones. The pattern of variation in exposure rate with lithology was consistent with the relative average abundance of uranium and thorium in common lithologic units [5]. The data indicated that the source of radiation in the region is the granitic rocks.

IV. Uranium and thorium in the granitic rocks

Some rock samples were collected randomly from different outcropping rock-sites of Aja Massif and analyzed by α-spectrometry for uranium and thorium. The samples were mainly monzogranite (the predominant lithology in the region). Some of the collected samples were fresh samples while others were of different degrees of weathering.

The results indicated that ^{238}U activity concentration in the measured samples ranged from 141 to 532 Bq/kg, with an average value of 294 Bq/kg, whereas ^{232}Th activity concentration ranged from 96 to 669 Bq/kg, with an average value of 383 Bq/kg. These average values of uranium and thorium in the analyzed rock samples indicated that these granitic rocks are remarkably enriched with uranium and thorium isotopes in levels roughly estimated by 12 orders of magnitude greater than their normal level in the Earth's crust, and about 5 orders of magnitude greater than their global average in the granitic rocks. The average concentration of uranium and thorium in the Earth's crust are 1.8 and 7.2 ppm, respectively [6], and in the igneous rocks of granitic composition are 5 and 15 ppm, respectively [7], where, I ppm of uranium and of thorium corresponds to 12.36 and 4.04 Bq/kg, respectively.

The theoretical expected $^{232}Th/^{238}U$ mass ratio for the primary granitic rocks is about 4 (corresponds to activity ratio about unity) [6]. This ratio is indicative for the relative depletion or enrichment of the radioisotopes. However, ^{238}U concentration was plotted versus ^{232}Th concentration (Fig. 3). The ^{232}Th concentration is positively correlates with ^{238}U ($R^2 = 0.62$). This observation may be related to the primary U-Th fractionation in the granitic magma (melt), where there is no secondary redistribution of both radionuclides was occurred due to secondary geological processes. The $^{232}Th/^{238}U$ activity ratio is about 1.15 (Fig 3). The distribution of the plot indicated that ^{238}U was more depleted in the collected samples relative to ^{232}Th.

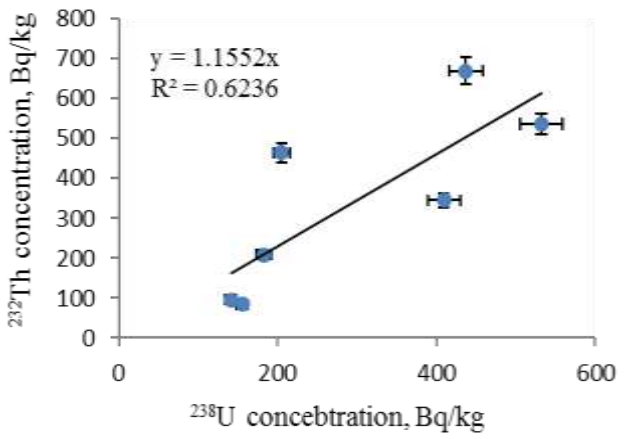

Fig. 3. Correlation between ^{238}U and ^{232}Th in the analyzed rock samples.

In most cases, ^{238}U and ^{234}U isotopes were not exist in quite secular equilibrium. The daughter/parent radionuclide activity ratios were not equal unity in levels exceeding the statistical error. The average value was about 0.79 (Fig. 4), indicating that the rock has been disturbed, probably due to water-rock interaction. Groundwater may transport the more soluble radionuclide into or away from the rock. The isotopic ratios indicated losses without gains of the more mobile ^{234}U. This is not the case with the less mobile thorium isotopes (^{232}Th and ^{228}Th), where the $^{228}Th/^{232}Th$ activity ratio was about unity.

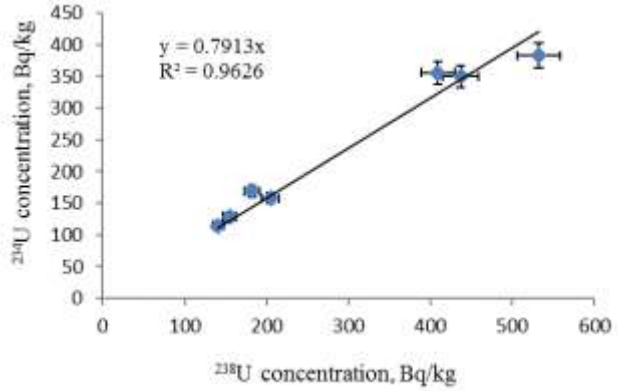

Fig. 4. Correlation between ^{238}U and ^{234}U in the analyzed rock samples.

V. Natural radioactivity in some building materials

Samples of building materials from local quarries were analyzed for ^{226}Ra, ^{232}Th and ^{40}K by γ-spectrometry [8]. The common used materials were fragmented weathered granites, granite gravels mixed with clays, red-yellow to yellow sands and crushed mafic metavolcanic rocks (known as black rocks). The highest activity concentrations were found in the fragmented granite materials. Average values of 194, 912 and 1320 Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K, respectively, were obtained. The lowest activity concentrations were found in the crushed black rocks; average values of 24, 82 and 255 Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K, respectively, were obtained. The values of the other materials lie in between. The activity concentration of both ^{226}Ra and ^{232}Th follows the same common pattern of variation of natural radioactivity with lithology and in agreement with the present ground survey data.

VI. Natural radioactivity in the groundwater

A. Gross α and gross β activities

Gross α and gross β activity measurements were conducted for the groundwater of the scattered wells in the inhabited and cultivated zones [9]. The gross activity varied markedly from site to site. The average gross α and gross β activity concentrations were 2.15 Bq/L and 2.60 Bq/L, respectively. These values are about 3.9 and 1.9 times greater than the national limit values set out for gross α and gross β activities, respectively, in the drinking water. Most samples (about 81%) exceeded the national regulation limit values.

B. Uranium and radium in groundwater

Uranium and radium isotopes were analyzed in the groundwater of the sampled wells [9]. Uranium concentration exceeded the national limit value of 30 µg/L (corresponds to about 0.37 Bq/L of ^{238}U) in about 38% of the samples. Uranium-238 concentration positively correlates with ^{234}U (R^2 = 0.97) and the $^{234}U/^{238}U$ activity ratio was greater than unity in all cases and ranging from 1.5 to 2.8 with an average value of about 1.9. This ratio may vary widely from 0.5 to 10 with, in most cases, a ratio >1 [10].

The average ^{226}Ra concentration was 0.29 Bq/L, which exceeded the national regulation limit value (0.11 Bq/L) set out for ^{226}Ra in drinking water in significant number of samples (about 49%) and was below the detection limit in few samples of shallow waters.

The average ^{228}Ra concentration was 0.46 Bq/L. The national regulation limit set out as a maximum contaminant level for combined radium ($^{226}Ra+^{228}Ra$) is 0.185 Bq/L. This value was also exceeded in significant number of samples. Poor correlation (R^2 = 0.29) was observed between ^{226}Ra and ^{228}Ra levels and was explained to be related to different $^{238}U/^{232}Th$ ratios in the minerals of the aquifer rock-structures [9]. The anomalous radioactive source in the groundwater of the study area was different in the different aquifer structures and may differ in the same structures [9], reflecting the complexity of the aquifer-water system.

In an attempt to find out some link between some chemical parameters and the radionuclide content, higher uranium concentrations were found in waters of relatively lower pH values (within the pH range 6.25 to < 8 of in the region), and in higher HCO_3^- content, where the HCO_3^-

anion is stable in this pH range. The positive correlation (R^2 = 0.61) between uranium and HCO_3^- concentrations was clear, where the role of CO_3^- and HCO_3^- in uranium leaching is known [11]. Oppositely, the higher radium concentration was found in waters of low SO_4^{2-} and low HCO_3^- content (< 300 µg/L), reflecting their partly limiting effect on radium dissolution [12]. Some samples from shallow water wells have low radium content although they have low SO_4^{2-} and low HCO_3^- concentrations because the water table is underlain by nonporous rocky-layer keeping the rock-water interaction to minimum. This explanation was supported by the presence of low uranium and TDS concentrations in these samples.

The absence of correlation between the alkaline earths (Ba and ^{226}Ra or ^{228}Ra isotopes) ruled out the probability of chemical dissolution mechanism for radium leaching, however, direct recoil mechanism may has the major effect [13, 14]. The wells of different water tables did not indicate correlation between well-depth and uranium or radium concentration in the groundwater. Generally the higher uranium content in groundwater was found in the granitic aquifers where the waters have relatively lower pH and higher HCO_3^- content, whereas the higher radium content was found in the waters of the sandstone aquifers of relatively lower SO_4^{2-} and HCO_3^- content. However, the complexity of the aquifer-water system resulted in lack of very clear correlations between the different parameters, where may be more than one factor is influencing the dissolution or precipitation processes of the radionuclides.

VII. Radon-222 distribution

A. *Radon-222 in groundwater*

Radon-222 was measured in the groundwater of the scattered wells in the study area [15] using RAD7 instrument. Radon levels varied widely (ranging from 2.5 to 95 kBq/m^3), with average value of about 30.5 kBq/m^3. In most wells (75% of the samples) radon values exceeded the limit value set out for radon in drinking water (11.1 kBq/m^3). The radon concentration range in the groundwater is in good agreement with the worldwide range, but the average concentration is about 3 times greater than the worldwide average value. The worldwide average radon concentration in groundwater is 10 kBq/m^3, with values ranging from 1-100 kBq/m^3 [16]. However, the obtained results indicated that the groundwater in the area is a radon prone water. The lowest average radon concentrations were found in the sandstone aquifers, whereas the highest average concentration, as well as the highest individual values, were found in the granitic aquifers. The quaternary surfacial deposits, which overlie most of the region, showed intermediate average radon concentrations.

B. *Radon-222 in the atmosphere (indoor and outdoor)*

Radon-222 was measured in the atmosphere of some sites, distributed between Hail city and other two towns (Al-Qa'ad and Al-Khottah) (Fig. 1), using RAD7 instrument. The activity concentration of indoor radon varied widely, ranging from 12.0 to 125.6 Bq/m^3, with an average value of about 54.6 Bq/m^3. This wide range was explained by the extreme variable migration of radon to the Earth's surface in the different lithologies due to the various aspects of local geology as fractures, porosity and other openings. Other factors have a significant effect on radon emanation as weather conditions, home ventilation, type of dwelling construction materials and seasons of the year [17-19]. In addition, some natural parameters as local atmospheric pressure gradients, soil moisture, groundwater movement, can affect the radon release. No much significant indoor ^{222}Rn levels were observed and were found to be below the U.S. EPA recommended action level of 148 Bq/m^3 [20], but 4 samples exceeded the encourage action starting at 74 Bq/m^3. The average concentration of indoor radon in the high radiation area of Pocos de Caldas, Brazil, was 61 Bq/m^3 with a wide range of 12-920 Bq/m^3 [21], which is comparable to the present data, but very low compared to indoor Rn level (100-5000 Bq/m^3) in Stockholm province, Sweden [22]. Indoor radon levels range from 10-100 Bq/m^3 are reported in some European countries [23] which is in agreement with the present findings. The average indoor radon concentration is almost 1.4 times greater than the global average concentration of 40 Bq/m^3 [24].

The outdoor Rn concentrations were much lower than that of the outdoor radon. It was varied within a narrow range (from 6.2 to 13.3 Bq/m^3), with an average value of about 10.3 Bq/m^3. Outdoor radon usually diluted by winds keeping lower concentration with little variation. The average outdoor ^{222}Rn concentration well agree with the global average of 10 Bq/m^3 [16]. The average indoor value is higher than that of the outdoor by a factor of about 5. This factor was 1.5 in lower level data [25] and was about 5 in similar level data [21, 26].

VIII. Uptake of uranium by vegetation

Soluble ions (including radionuclide ions) in the irrigation water and soil can be transferred into specific plant tissues as a function of their behavior during plant metabolism, potentially resulting in higher accumulation in a particular part of the plant. This accumulation may causing extra radiation dose to the consumer, if this part is edible. Carvalho, etal. [27] reported that the main source of radionuclides to the plant is their content in the irrigation water rather than the soil, however, the uptake of uranium isotopes (the predominant radionuclides in the irrigation water) by the plant and their translocation inside plant parts was investigated in a cultivated zone lies adjacent to the northeast of Hail City. The analyzed plants were the available plants of the season. They were green pepper, eggplant, lemon, fig, narenj, carrot, mint, alfalfa and lettuce. Uranium isotopes are also enhanced in the soil (156 and 130 Bq/kg, for ^{238}U and ^{234}U, respectively) [28] and may be partially available to the plant.

The uranium content was the highest in the roots (average values for ^{238}U and ^{234}U were 11.93 and 31.23 Bq/kg, respectively), whereas it was the lowest in the fruits (the average values were 0.05 and 0.13 Bq/kg, respectively). The

general distribution pattern was: roots > leaves > stems or branches > fruits [28]. The data are in agreement with previous findings for uranium distribution in plants [29-31].

A. Transfer of natural uranium to the edible parts of the plants

Root-uptake of uranium from soil to plant was evaluated by calculating the observed transfer factor (TF), which is the ratio between the radionuclide concentration in the edible part of the plant (Bq/kg fresh weight), and its concentration in the soil (Bq/kg dry weight).

The TF values ranged from $0.09x10^{-3}$ to $5.72x10^{-3}$ and from $0.23x10^{-3}$ to $20.06x10^{-3}$ for ^{238}U and ^{234}U, respectively. These values were higher in roots and leaves (ranged from $1.23x10^{-3}$ to $5.72x10^{-3}$ and from $5.69x10^{-3}$ to $18.34x10^{-3}$ for ^{238}U and ^{234}U, respectively), and were several orders of magnitude lower in the fruit samples (ranged from $0.09x10^{-3}$ to $0.72x10^{-3}$ and from $0.23x10^{-3}$ to $2.54x10^{-3}$ for ^{238}U and ^{234}U, respectively). These ^{238}U TF values are comparable with the range (10^{-3} - 10^{-1}) reported by other workers [32] for most of the TF values for ^{238}U root-uptake from soil to plant. Unclear correlation was observed between the TF values and the uranium concentration in the soil reflecting the complexity of uranium transfer process from soil to plant [33].

IX. Potential radiation hazards to the inhabitants

A. Hazards due to terrestrial γ-radiation

From the outdoor ground surveys, the average exposure rate values due to terrestrial γ-radiation were estimated by 0.88, 1.55, 2.27 mSv/y on the soil, wadi deposits and rocks, respectively [4], which was 2 to 5 times higher than the global normal average value (0.46 mSv/y) [16]. The average value in the residential zones of the study area was about 1.9 times higher than the global average.

The average annual effective dose rates for individuals living in the region were estimated by 1.13, 1.80, and 2.52 mSv/y on soil, wadi deposits and granitic rocks, respectively, with an average value of 1.74 mSv/y. The average fatal cancer risk, to an individual living in the region due to exposure to terrestrial γ-radiation from the soil was estimated by about $0.57 x10^{-4}$ for individuals living in the Hail city and the habited zones, and was about $0.90x10^{-4}$ for individuals living on the deposits of the wadis lie among the Aja heights. The highest average fatal cancer risk ($1.26x10^{-4}$) was for the rare individuals living on the heights. The average value of the fatal cancer risk in the whole region was estimated by $0.87x10^{-4}$. This means that a one cancer case per about 11,150 individuals, living permanently in the region, may occur. Although the area is considered as a high radiation area, the average cancer risk due to terrestrial γ-radiation is too small to cause alarm.

B. Hazards due to radon

The annual effective dose due to radon inhalation was estimated and the probability of lung cancer mortality was calculated [15, 16]. The dose to individuals living in this region comes mainly from inhalation of the released radon from the soil and contaminated tap water. The average annual effective dose to individuals living permanently in the region was estimated by 1.53 mSv, and most of the dose was due to inhalation of indoor radon. The lifetime excess risk was estimated by $87.2x10^{-4}$. In other words, a one lung cancer case due to radon inhalation may occur along the lifetime per 115 individuals living permanently in Hail region.

C. Hazards due to utilization of local building materials

The radiation impact due to utilization of building materials originating from the study area was estimated based on the average activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, and calculating the average dose rate (absorbed and annual effective) [8]. The proposed radiation hazard indices that used as radiation measures for building material evaluation are also calculated [34, 35]. The utilized building materials were fragmented weathered granites, granite gravels mixed with different clay percentages, sands and crushed black rocks (mafic metavolcanic rocks). The fragmented granites and granite gravels with high clays percentage causing an average absorbed dose of about 12 times higher than that of the global normal average of 55 nGy/h [36], and causing an average annual external effective dose rate (0.83 mSv/y) about twice higher than that of the global normal average of 0.46 mSv/y [37, 38]. The absorbed dose and annual indoor effective dose reported by Dziri etal. (<230 nGy/h and <1.1 mSv/y, respectively) are lower [39].

A building material is classified as safe material if the proposed hazard indices limits are not exceeded. The radium equivalent activity index (Ra_{eq}) should be < 370 and the other external and internal hazard indices (H_{ex} and H_{in}) should be < unity [34]. The γ-radiation hazard index ($I_{\gamma r}$) should be ≤ 3 [35]. The average activity concentration of each lithologic group was used to calculate its hazard indices, to examine if the non-dimensional value of the hazard index does not exceeded. The complied material can be used without restriction, whereas the exceeded materials should be only used under specific circumstances or excluded from utilization as safe building materials. The results indicated that the proposed indices were exceeded for the fragmented granites and granite gravels that mixed with clays, whereas sands and crushed black rocks were compiled [8].

Conclusions

From the obtained data, the following conclusions could be drawn:

- The average exposure dose rate values due to terrestrial γ-radiation are 2-5 times higher than the global average value (0.46 mSv/y) of normal background areas.

- The granitic rocks are remarkably enriched with uranium and thorium isotopes in levels roughly estimated by 12 orders of magnitude greater than their normal level in the Earth's crust and about 5 orders of magnitude greater than their global average in the granitic rocks.
- The highest activity concentrations in the building materials were found in the fragmented granite materials, causing average absorbed dose of about 12 times higher than that of the global normal average. The lowest activity concentrations were found in the crushed black rocks; the values of other materials lie in between.
- The average gross α and gross β activity concentrations in groundwater were about 3.9 and 1.9 times greater than the national limit values set out for gross α and gross β activities, respectively, in the drinking water. Most samples (about 81%) exceeded the national regulation limit values
- The anomalous radioactive source in the groundwater was different in the different aquifer structures.
- In most wells (75%), radon concentration exceeded the limit value set out for radon in drinking water. It was the lowest in the sandstone aquifers and the highest in the granitic aquifers. Generally, it was uranium in wells of granitic aquifers and was radium in the wells of sandstone aquifers.
- The general distribution pattern of uranium in the plant tissues due to root uptake was: roots > leaves > stems or branches > fruits.
- No much significant indoor ^{222}Rn level was observed. It was found to be below the U.S. EPA recommended action level of 148 Bq/m^3. The dose to individuals due to radon comes mainly from inhalation of the indoor radon released from the soil.

Recommendations

- Remediation measures, based on the obtained information, should be justified and considered.
- Farming activities in this area should be restricted to growing fruitful trees and shrubs and avoiding growing leafy plants, including alfalfa (the animal feeding plant of interest).
- To reduce radon exposure, aeration of homes daily for few minutes should be of concern and considering mitigation programs for water aeration. Development of building codes requiring measures for radon prevention and radiation dose reduction in existing houses or houses under construction have to be planned.
- The groundwater should be subjected to an appropriate treatment to remove uranium and radium with salinity before use for drinking or other uses.

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