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Concentrations of the Terrestrial Radionuclides and Their Radiological Risk at Wadi Nugrus, South Eastern Desert, Egypt

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Abstract: Fourteen samples were collected from a minor tributary at Wadi Nugrus, SED, Egypt. The average values of the activity concentrations of the terrestrial radionulclides U-238and Th-232 in the studied samples were 325 and 277 (Bq/kg) much higher than the worldwideaverages. The average value of K-40, 820 (Bq/kg), is comparable to the worldwide average concentration of this radionuclide. Four samples have safe values for the hazard indices Ra_{eq}, H_{ex} and H_{in}. This represented a contradiction with values of the absorbed dose rate D which hasa minimum value of 110 (nGy/h) exceeding the worldwide average of 60 (nGy/h). At the announced safe unity for both H_{ex} and H_{in}, the total annual effective dose appears to have morethan the value of 6 (mSv/y) which far exceeds the worldwide average effective dose rate of 1.5(mSv/y). The effective dose approach established by UNSCEAR is reliable since the effective dose reacts directly with the dose limits recommended by the radiation protection regulations.

Keyword: Terrestrial radionuclides; Building materials; Hazard indices; Absorbed dose rate; Effective dose rate; Excess lifetime risk.

1. Introduction

Wadi Nugrus lies in the southern part of the Eastern Desert (SED) of Egypt. It is located 90 km.southwest of Marsa Alam and lies between latitudes 24^0 40\ and 24^0 50\ North and longitudes 34^0 32\ and 34^0 40\ East, Fig 1. The area could be reached from the Red Sea coast through WadiEl-Gemal area.

The stream sediments along Wadi Nugrus are formed due to the weathering processes of the different surrounding rocks. These rocks are almost of granitic origin. Granitic rocks have relatively high concentration of radionuclides, the members of the natural radioactive series ofU-238, Th-232 and U-235. The presence of these naturally occurring radionuclides along withthe non-series radionulclide ^{40}K and their daughter products in building materials is a source ofindoor radioactive pollution. Wadi Nugrus is subjected to several geological activities to explore precious minerals such as gold, corundum, zircon and pyrite, etc... This explains the establishing of many important development projects at this area. The dweller of houses and office buildings, where they spend about 80% of their time, are exposed to radiation emitted bythese radionuclides.

Continuous exposure to even low level radiation may adversely affect human health. Therefore, it is important to monitor the concentration of radionuclides in building materials and to assessthe radiation exposure to the people. Most of the developed and developing countries in the world are carrying out nation-wide surveys to assess the amount of radioactivity in order to establish possible radiological hazards and to take safety measures if necessary [1-5].

Previously, the stream sediments at nine locations along Wadi Nugrus, SED were evaluated as building materials [6]. This study extends the evaluation of the stream sediments as building materials at a different tributary at Wadi Nugrus.

Fig. 1. Study area at the South Eastern Desert, Egypt [7].

2. Samples and Experimental Methods

2.1. Sample collection and preparation

Fourteen sedimentary samples were collected from one of the minor tributaries of Wadi Nugrusto the east of Wadi Abu-Rusheid. Locations of the samples are shown in Fig. 2, while their GPSpositions (in meters) are represented in Table 1.

Each sample was thoroughly stirred, and homogenized as a representative sample for its location. Friable sediment samples were collected from holes having about 70 cm diameter andabout 90cm depth. These samples were extracted from each hole, at depths between 20 and 90cm from the surface level of the Wadi after stripping off the surficial windblown sand layer, i.e.20cm. The sample weight is about 50 kg and the grains with pebbles (>2 mm) size were first excluded by sieving through a 2mm size aperture sieve.

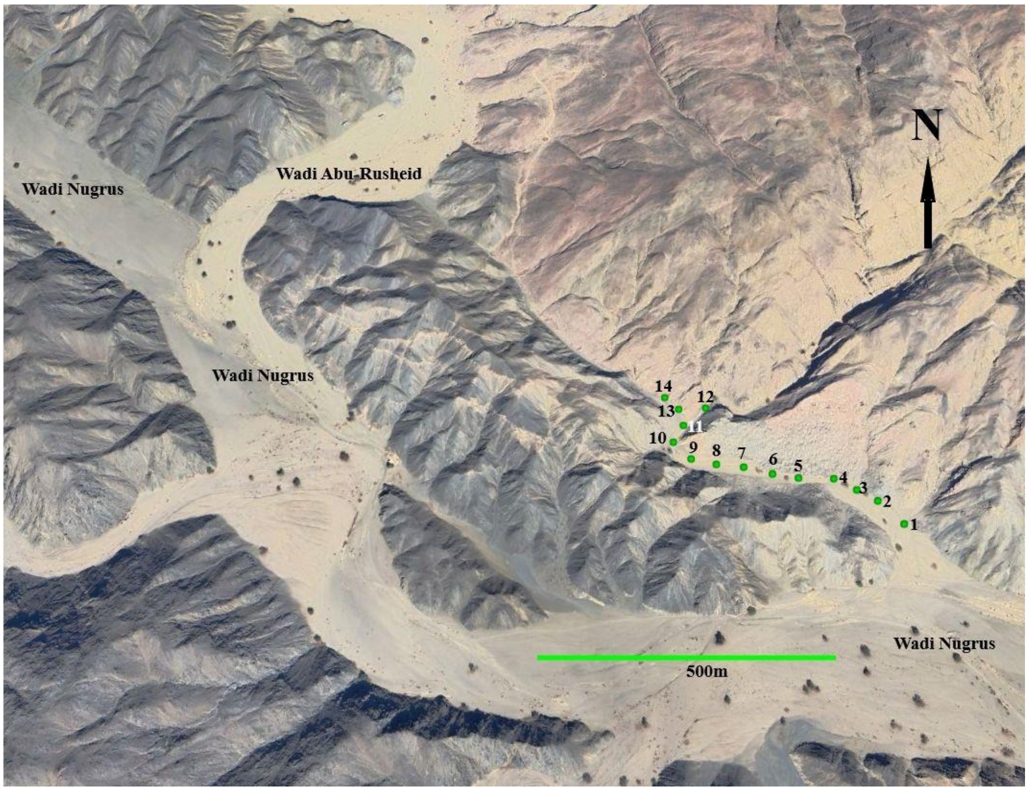

Fig. 2. Locations of the fourteen samples collected from Wadi Nugrus, SED [7].

2.2. Radiometric measurements

About 300-350g from each sample was packed in a plastic container, sealed well and stored forabout 30 days before analysis. This allowed the in-growth of uranium and thorium decay products prevent the escape of radiogenic gases ^{222}Rn and ^{220}Rn and allowed secular equilibrium between ^{238}U, ^{232}Th and their decay products. After attainment of secular equilibrium, each of the prepared samples was measured in the laboratory for their U, Th and K contents using a high resolution multichannel analyzer of γ-ray spectrometer (HPGe detector). Each sample was counted for 1000s. The radiometric measurement for the studied radionuclides was carried out at exact lines (energies). Uranium activity concentrations (Bq/kg)were determined at the lines of Pa-234, Ra-226, Pb-214 and Bi-214 at energies 100.1, 186.1, 295.1, 352.1, 609.3, 934.1, 1120.3 and 1764.5 (keV). Thorium was measured using the lines ofAc-228 and Tl-208 at 911 and 584.45 (keV), respectively. Concentrations of potassium were determined at its characteristic line, 1460 (keV).

3. Results and Discussions

Other

3.1. Activity concentrations in the studied stream sediments

Activity concentrations of U-238, Th-232, and K-40 were measured in the chosen stream sediments at Wadi Nugrus as in Table 1. The activity concentrations of U-238 range between 85.11 and 622.8 with an average of 325.1 (Bq/kg) while Th-232 activity concentration varies between 56.9 and 600.1 (Bq/kg) with 277.7 (Bq/kg) as an average. K-40 concentration ranges between 674.1 and 996.2 (Bq/kg) with an average of 820.1 (Bq/kg). The worldwide average concentrations of ^{238}U, ^{232}Th and ^{40}K are 33, 45 and 412 (Bq/kg), respectively, [8,9]. This indicates that the studied stream sediments at Wadi Nugrus have much higher values of the activity concentrations of the radionuclides; U-238 and Th-232 compared to the reported average values [9]. The average value of K-40 is comparable to the worldwide average [9].

Table 1. GPS positions (m) of fourteen samples collected from Wadi Nugrus and theactivity concentrations (Bq/kg) of the radionuclides; U-238, Th-232 and K-40.

No.	(m) E	(m) N	U-238 (Bq/kg)	Th-232 (Bq/kg)	K-40 (Bq/kg)
1	679691.00	2723971.00	85.11±3.47	70.05±1.66	683.7±10.43
2	679651.00	2724015.00	97.61±11.8	68.44±1.7	674.1±10.4
3	679616.00	2724036.00	158.00±5.3	190±2.7	832.9±12.0
4	679577.00	2724058.00	161.2±9.4	156.2±2.54	855.6±12.06
5	679512.00	2724058.00	94.91±2.7	64.33±1.73	910.3±9.53
6	679465.00	2724065.00	85.56±8.2	56.90±1.84	911.2±12.86
7	679413.00	2724078.00	584.1±17.29	528.1±4.51	850.4±12.62
8	679363.00	2724083.00	461.5±18.01	339.1±4.68	911.8±16.52
9	679317.00	2724093.00	330.2±12.86	259.8±2.52	877.5±9.51
10	679286.00	2724127.00	552.7±12.46	600.1±4.66	776.6±11.73
11	679307.00	2724162.00	514.3±12.72	504.9±4.33	748.9±11.80
12	679351.00	2724198.00	219.8±9.38	114.6±2.24	996.2±13.44
13	679300.00	2724196.00	622.8±16.18	472.9±3.93	731.3±12.35
14	679275.00	2724221.00	583.5±16.85	462.5±4.44	721.4±12.16

3.2. Hazard indices

Historically, several indices were proposed to evaluate the radiation hazard received by the members of public as a result of using the building materials containing the terrestrial;radionuclides; U-238, Th-2342 and K-40. These indices were named Ra_{eq} H_{ex} to evaluate external hazards from gamma rays while the third H_{in} to evaluate the internal hazards due to theinhalation of radon gas and its short-life decay products [1,10].

3.2.1. Radium Equivalent Activity (Ra_{eq})

The radium equivalent activity for the samples was calculated. The exposure to radiation can be defined in terms of the radium equivalent activity (Ra_{eq}), [10]. Ra_{eq} can be expressed by thefollowing equation:

$$Ra_{eq} = U\text{-}238 + 10/(7\times Th\text{-}232) + 10/(130\times K\text{-}40)\ [Bq/kg] \quad (1)$$

where: U-238, Th-232 and K-40 are the specific activities (Bq/kg) of the radionuclides U, Th and K, respectively.

For a safe use, the maximum value of Ra_{eq} in stream sediments from Wadi Nugrus must be lessthan 370 (Bq/kg) [10]. The range and the mean value of Ra_{eq} of the studied samples are presented in Table 2. The values of Ra_{eq} for these samples are higher than the criterion limit of370 (Bq/kg) except at four locations; 1, 2, 5 and 6, Fig. 2. Apart from these four samples, it is unsafe to use the stream sediments from the chosen tributary at Wadi Nugrus as building materials.

3.2.2. External and internal hazard indices (H_{ex} and H_{in})

Another index is suggested to assure the safety of the external gamma-rays for the samples under investigation, the external hazard index (H_{ex}). H_{ex} is obtained by dividing Equation (1) by 370 (Bq/kg) according to the following equation [10]:

$$H_{ex} = U\text{-}238/370 + Th\text{-}232/259 + K\text{-}40/4810 \quad (2)$$

The internal exposure to Rn-222 and its radioactive progeny is controlled by the internal hazardindex (H_{in}), which is given by [10]:

$$H_{in} = U\text{-}238/185 + Th\text{-}232/259 + K\text{-}40/4810 \quad (3)$$

The values of both indices must be less than unity for the radiation hazards to be acceptable. Table 2 represents the values of the external hazard index H_{ex} and the internal hazard index H_{in}as calculated for the chosen fourteen samples from Wadi Nugrus. Like radium equivalent Ra_{eq},the values of both external and internal hazard indices H_{ex} and H_{in} for the studied samples are higher than unity except at the four locations; 1, 2, 5 and 6. External and internal hazard indicesfor the remaining samples are higher than unity indicating that they cannot be used as buildingor decorative materials at the dwellings.

Table 2. Hazard indices; Ra_{eq} (Bq/kg), H_{ex} and H_{in} calculated for fourteen samples collected from Wadi Nugrus, SED, Egypt.

No.	Ra_{eq} (Bq/kg)	H_{in}	H_{ex}
1	235.4	0.86	0.64
2	233.9	0.86	0.63
3	508.2	1.84	1.37
4	447.7	1.64	1.21
5	246.3	0.89	0.67
6	236.1	0.87	0.64
7	1401.5	5.36	3.79
8	1026.5	4.0	2.8
9	769.1	2.97	2.08
10	1406.1	5.12	3.78
11	1242.3	4.61	3.36
12	443.7	1.75	1.2
13	1462.2	5.92	3.95
14	1192.3	4.5	3.2
Ave.	775.1	2.94	2.09
min	233.9	0.86	0.63

max	1462.2	5.92	3.95
Permissible level	370	1	1

3.3. Absorbed dose rate in air D

The absorbed gamma dose rates in air at 1m above the ground surface for the uniform distribution of radionuclides; U, Th and K were calculated as follows, [8]:

$$D\,[\mathrm{nGy/h}] = 0.462\times\mathrm{U\text{-}238} + 0.604\times\mathrm{Th\text{-}232} + 0.0417\times\mathrm{K\text{-}40}, \quad (4)$$

where: U-238, Th-232 and K-40 are the average specific activities (Bq/kg) of U, Th and K, respectively.

In the above conversions, it is assumed that all the decay products of U-238 and Th-232 are inradioactive equilibrium with their precursors. Table (3) represents the estimated values of the absorbed dose rate D (nGy/h) at 1m above the stream sediments at the chosen tributary at WadiNugrus, SED. The D values at the fourteen locations range from 110.1 to 650.2 (nGy/h) with an average of 352.1 (nGy/h). According to the UNSCEAR (2000) [8], the corresponding worldwide average values range from 18 to 93 (nGy/h) and a typical variability range for measured absorbed dose rates in air outdoors is from 10 to 200 (nGy/h). The population weighted values of the absorbed dose rate in air outdoors from terrestrial gamma radiation estimated by the spectroscopic analysis of soil samples give an average of 60 (nGy/h). The D values for all the studied samples are much higher than the worldwide average value for soils (60 nGy/h) alarming the use of the stream sediments from the chosen tributary at Wadi Nugrusas building materials.

3.4. External effective dose rate E_{ext}

The outdoor external effective dose rate E_{ext}^{out} (mSv/y) is calculated from the absorbed dose rate D (nGy/h) by applying the dose conversion factor of 0.7 (Sv/Gy) and the outdoor occupancy factor of 0.2 [8] as clarified by the equation:

$$E_{\mathrm{ext}}^{\mathrm{out}}\,[\mathrm{mSv/y}] = D\,[\mathrm{nGy/h}]\times 0.7\,[\mathrm{Sv/Gy}]\times 8760\,[\mathrm{h/y}]\times 0.2\times 10^{-6}. \quad (5)$$

Values of the annual outdoor external effective dose E_{ext}^{out} (mSv/y) for the fourteen studied samples are listed in Table 3. The values obtained varied between 0.135 and 0.797 obtaining an average of 0.432 (mSv/y). These values are much higher than the worldwide average of theoutdoor external effective dose rate of 0.07 (mSv/y) as reported [8].

The indoor to outdoor ratios range from 0.6 to 2.3, with a population-weighted value of 1.4. Thus indoor exposures are, in general, 40% greater than outdoor exposures. Besides, the occupancy factor for the public indoors is 0.8. So, the indoor external effective dose rate is estimated as follows [8]:

$$E_{\mathrm{ext}}^{\mathrm{ind}}\,[\mathrm{mSv/y}] = 1.4\times D\,[\mathrm{nGy/h}]\times 0.7\,[\mathrm{Sv/Gy}]\times 8760\,[\mathrm{h/y}]\times 0.8\times 10^{-6}. \quad (6)$$

Table 3 lists the calculated values of E_{ext}^{ind}. The annual indoor effective dose E_{ext}^{ind} results fromthe external exposure to γ-rays emitted from the radionuclides in the stream sediments from thedifferent locations at Wadi Nugrus studied tributary when used as building materials range between 0.76 (mSv) and 4.47 (mSv) with an average of 2.42

(mSv), All values of E_{ext}^{ind} are much higher than the worldwide average of 0.41 (mSv/y) [8].

Finally, values of the external effective dose rate E_{ext} which is the summation of the outdoor and indoor external effective dose rates E_{ext}^{out} and E_{ext}^{ind} calculated as follows:

$$E_{ext}\ [mSv/y] = E_{ext}^{out}\ [mSv/y] + E_{ext}^{ind}\ [mSv/y], \quad (7)$$

are tabulated in Table 3. Like the absorbed dose rate D, the external effective dose rate E_{ext} hasmuch higher values due to all samples collected from the chosen locations at Wadi Nugrus compared to the worldwide average of 0.48 (mSv/y).

3.5. Excess lifetime cancer risk ELCR

Based upon calculated values of the annual external effective dose E_{ext}, excess lifetime cancerrisk (ELCR) was calculated using the following equation, [5];

$$ELCR = E_{ext}\ [mSv/y] \times 10^{-3} \times LE[y] \times RF[Sv^{-1}], \quad (8)$$

where E_{ext} is the annual external effective dose, LE life expectancy (70 years) and RF is the fatal risk factor per Sievert, which is 0.05 (Sv^{-1}) [11]. Table 3 represents the values of the dimensionless factor ELCR due to exposure to the studied samples. All the values of ELCR forthe fourteen samples collected from the chosen tributary at Wadi Nugrus, SED are much higherthan the worldwide average value of ELCR, 1.45×10^{-3} [5].

Table (3): The absorbed dose rate D (nG/h), outdoor and indoor external effective dose rates, E_{ext}^{out} and E_{ext}^{ind} (mSv/y) and their sum E_{ext} (mSv/y) along with the excess life cancer risk ELCR calculated for fourteen samples collected from Wadi Nugrus, SED, Egypt.

No.	D (nGy/h)	E_{ext}^{out} (mSv/y)	E_{ext}^{ind} (mSv/y)	E_{ext} (mSv/y)	ELCR×10^{-3}
1	110.1	0.14	0.76	0.90	3.12
2	114.5	0.14	0.79	0.93	3.25
3	222.5	0.27	1.53	1.80	6.30
4	204.5	0.25	1.41	1.66	5.79
5	120.7	0.15	0.83	0.98	3.42
6	111.9	0.14	0.77	0.91	3.17
7	624.3	0.77	4.29	5.06	17.69
8	456.0	0.56	3.13	3.69	12.92
9	346.1	0.42	2.38	2.80	9.80
10	650.2	0.8	4.47	5.27	18.42
11	573.8	0.7	3.94	4.64	16.26
12	212.3	0.26	1.46	1.72	6.01
13	603.8	0.74	4.15	4.89	17.11
14	579.0	0.71	3.98	4.69	16.40
Ave.	352.1	0.43	2.42	2.85	9.98
min	110.1	0.14	0.76	0.90	3.12
max	650.2	0.8	4.47	5.27	18.42
Worldwide average	60	0.07	0.41	0.48	1.45×10^{-3}

3.6. Hazard indices, absorbed dose rate and excess lifetime cancer risk

The decision about the safety of the stream sediments collected from locations; 1,2,5 and 6 as building materials was positive depending on the calculated values of the hazard indices; Ra_{eq},H_{ex} and H_{in}. This makes a contradiction with the decisions of unsafe materials based on the values of the absorbed dose rate D and all the dependent quantities; E_{ext}^{out}, E_{ext}^{ind}, E_{ext} and ELCR. This contradiction is recognized elsewhere [12,13]. Xinwei et. al. studied the building materials in Yan'an, China [12]. They abstracted that the values of Ra_{eq} for all samples are below 370 (Bq/kg), the values of H_{ex} are below unity, the average values of D, 101-177 (nGy/h),are higher than the worldwide average and the annual external effective dose E_{ext}, 0.5-0.87 (mSv/y). Amazingly, they found that these materials may be used safely as construction materials and do not pose significant radiation hazards to inhabitants [12]. Also, Wanyama et. al. investigated 30 samples from Rosterman gold mine, Kenya [13]. They reported that the values of Ra_{eq} and H_{ex} are within the permissible limits. Although the average value of D is 124(nGy/h) and the average value of E_{ext} is 0.7 (mSv/y), they concluded that mining of gold at Rosterman has no significant radiological health implication on the miners and the surroundingpopulation [13].

This contradiction is explained referring to the values and the average values of Ra_{eq}, H_{ex}, D and E_{ext} represented in Tables 2 and 3. The ratios Ra_{eq}/D and H_{ex}/D reveal that the safe value of Ra_{eq}, 370 (Bq/kg) and the safe unity of H_{ex} correspond to a value of 168 (nGy/h) for D whichis nearly triple the worldwide average value 60 (nGy/h). Also, the ratios Ra_{eq}/E_{ext} and H_{ex}/E_{ext} conclude that the safe values of both Ra_{eq} and H_{ex} correspond to 1.36 (mSv/y) as a value of theannual external effective doe E_{ext} which is almost triple the worldwide average, 0.48 (mSv/y). However the indices Ra_{eq} and accordingly H_{ex} and their safe values were suggested to evaluatethe building materials during the era of "permissible" dose in the seventies and eighties of the past century. The permissible dose was 5 (mSv/y) for the members of public from the sum of the external and internal exposures. This justified the condition that the studied building material is considered safe if the absorbed dose rate received by the members of public from the external gamma rays only is kept below 150 (mrad/y) or 1.5 (mGy/y) [1, 10]. This conditionwas fulfilled at a value of 370 (Bq/kg) for Ra_{eq} and a unity of H_{ex} as discussed above. The International Commission on Radiological Protection ICRP replaced the expression "permissible dose" by the expression "dose limit" and saved the limit for the public at 5 (mSv/y)from the total radiation exposures [14]. Later, ICRP reduced the dose limit for the members ofpublic to 1 (mSv/y) [11]. At the end of the century, the Scientific Committee on the Effects of Atomic Radiation UNSCEAR reported the worldwide average of the total annual effective dosereceived from the terrestrial radionuclides to have the value 1.5 (mS/y) [8].

Indeed, the evaluation of a building material should be assessed on the total annual effective dose received as a result of the exposure to this material. So far, only the external dose rate E_{ext}due to the external gamma rays from Nugrus samples is calculated. The internal effective doserate due to the inhalation of radon gas and its decay products and accordingly the total effectivedose rate due to the studied samples are not.

Refereeing to Equation (3), and neglecting the two terms of Th-232 and K-40, safety of a building material, i.e. H_{in} is less than unity, is verified if the activity concentration of

U-238 is185 (Bq/kg). At this concentration of U-238 in equilibrium with the successive decay product Ra-226, the parent source of Rn-222, the internal effective dose rate due to the exposure to thismaterial will exceed 5 (mSv/y) since only 40 (Bq/kg) of Ra-226 result in an internal effective dose rate of 1.095 (mSv/y), [8]. Accordingly, the values of the internal hazard index H_{in} for thesamples collected from locations 1, 2, 5 and 6 which were decided as safe values; these valueswill correspond to an internal effective dose rate of ~2.5 (mSv/y). Accordingly, the values of the total annual effective dose due to the use of the samples 1,2,5 and 6 will exceed 3 (mSv/y)which is double the worldwide average of 1.5 (mSv/y).

Remembering that the permissible unity of H_{ex} corresponds to an E_{ext} value of 1.36 (mSv/y) and the permissible unity of H_{in} corresponds to an annual internal effective dose that exceeds 5(mSv/y), this means that at the permissible unity of both the hazard indices H_{ex} and H_{in}, the total annual effective dose appears to have more than the value of 6 (mSv/y) which far exceedsthe worldwide average effective dose rate of 1.5 (mSv/y), [8,9].

On another hand, the excess lifetime cancer risk ELCR should be calculated using the total effective dose rate. Although ELCR is calculated using only the external component of the effective dose rate, Equation (8), the values of ELCR for all the studied samples far exceed the worldwide value. From Table (3) only the 0.48 (mSv/y) of the worldwide average of Eext corresponds to the worldwide value of ELCR. The value of ELCR at the worldwide average value of the total effective dose rate of 1.5 (mSv/y) seems to be catastrophic, 5.25×10-3. This indicates an overestimation of ELCR based on an expected overestimation of the fatal risk factor per Sievert, RF. However, because of this uncertainty on health effects at low doses, the Commission judges that it is not appropriate, for the purposes of public health planning, to calculate the hypothetical number of cases of cancer or heritable disease that might be associated with very small radiation doses received by large numbers of people over very long periods of time [15].

It must be recognized that the hazard indices Ra_{eq} and H_{ex} along with the absorbed dose rate Dall evaluate only the external component of the radiation exposure. They are calculated using the same values of the terrestrial radionuclides concentrations. Apart from any contradictions or overestimations, the authors of this study believe that the quantity D is the reliable one sinceit can be compared to the real measured values worldwide with an average of 55 (nG/h) or to that estimated by spectroscopic methods with the comparable average of 60 (nGy/h). Furthermore, D is used to calculate the external effective dose rate. In contrast with the internalhazard index H_{in}, UNSCEAR (2000) established a detailed mathematical approach to calculatethe internal effective dose rate depending on the Ra-226 content in the studied material. Theseeffective doses react directly with the dose limits recommended by the radiation protection regulations.

4. Conclusions

All the samples collected from the chosen tributary at Wadi Nugrus, SED have much higher concentrations of the terrestrial radionuclides; U-238, Th-232 and K-40 compared to the worldwide average concentrations of these radionuclides. The decision about the safety of the studied samples as building materials depending on the calculated values of the hazard indices;Ra_{eq}, H_{ex} and H_{in} represented a contradiction. At the announced safe unity for both H_{ex} and H_{in},the total annual effective dose appears to have more than the value of 6 (mSv/y) which far exceeds the worldwide average effective dose rate of 1.5 (mSv/y). On another hand the value of the excess lifetime cancer risk ELCR at the worldwide average value of the total effective dose rate of 1.5 (mSv/y) is catastrophic, 5.25×10-3 indicating an overestimation compared to the worldwide average of this factor, 1.45×10^{-3}. The effective dose approach established by UNSCEAR is reliable since the effective dose reacts directly with the dose limits recommendedby the radiation protection regulations.

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