Gamma activity in some environmental samples in south Egypt

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As a part of national survey to evaluate natural radioactivity in the environment, concentration levels of natural radioactivity have been analyzed from more than 108 environmental samples taken from Wadi Allaqi area, south Egypt. The distribution of naturally occurring radionuclides of 226Ra, 232Th and 40K in the samples (soil, granite, grass and water) have been measured using a high purity germanium detector. The specific activities in these samples, having a radium equivalent activity of less than 370 Bq.kg⁻¹, when evaluated for radiological effects show that all samples meet the external gamma-ray dose limitation of 1.5 mSv.y⁻¹. The data presented here will serve as a baseline survey for primordial radionuclide concentrations in igneous and sediment of the area.

Keywords: Radionuclides, Radioactivity, Dose rate
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1 Introduction

Accurate knowledge of anthropogenic and natural radionuclides distribution pattern is essential in controlling radiation levels and performing accurate assessment of the radiation dose from the radionuclides. In addition to natural radioactivity, man-made radionuclides may be present in the terrestrial environment as a result of nuclear weapon tests, nuclear accidents and unauthorized releases or puffs. Soil is the main reservoir of artificially produced radionuclides from precipitation, and acts as a medium of migration of these radionuclides to the biological systems. For this reason, measurement of the radioactivity concentrations of man-made radionuclides and their activity ratios in soil is essential to assess the radiation exposure to human and total radioactivity contents and eventually contamination from sources.

The consequences from natural radioactivity present in the environmental samples are two-fold: (i) irradiation of the whole body by gamma rays and (ii) irradiation of lung tissue by radon gas and its decay products. The concentration of radon varies strongly with meteorological parameters, while the mean gamma dose rate in environment depends strictly on the sample regions. Thus, the use of various building materials containing above normal levels of natural radioactivity may appreciably increase the radiation dose attained by the population, as compared with the known environmental radioactivity. In general, the abundances of thorium and uranium increase from basic to silicic rocks. Suites of related rocks almost invariably exhibit enrichment of the two elements in the later-forming members. Alkaline rocks are particularly enriched in thorium and uranium, and a distinct correlation of these with potassium has been repeatedly found. Considerable attention is required to be given to the possible exposures of humans to ionizing radiation from external and internal environmental sources. The external radiation exposures from natural radionuclides contribute about half of the average annual dose to humans from all radiation sources.

The present study was undertaken within the framework of our countrywide radiological environmental monitoring programme with the collaboration of other research institutes of the country. The main objective of this programme is to assess the type and amount of natural radioactivity levels in granite, soil, water, food items, and other samples from environmental media.

2 Experimental Details

A total of 108 samples of granite (20), soil (43), grass (30), water (15) were collected from Wadi Allaqi (Aswan) south Egypt. -Wadi Allaqi area 180 km south eastern Aswan city. It is the largest of the numerous wadis, or intermittent stream channels which run through Egypt’s south eastern desert to the shores of lake Nasser extending for more than 400 km south deep into Sudanese territory. The new lake had a considerable impact on the overall physical
environment of the area around it, including that of the Wadi Allqi region. Each igneous, sediment and grass sample was individually dried under laboratory range temperature of 32°C with relative humidity at 70% for 72 hr. The dried samples were grounded to a grain-size of about 100 mesh. Each powdered sample was thoroughly mixed by using electric shaker to ensure the samples are homogeneous. The samples were placed in 40 cc container and stored for four weeks to reach the equilibrium state. Particular care was taken to collect water samples for radon monitoring due to its short half-life. In this study a 1.4 litre polyethylene marinelli beaker were used as sampling and measuring containers. All containers were washed with dilute hydrochloric acid and rinsed with distilled water before use. The containers were filled up to the brim and a tight caps pressed on so that the air is completely removed from it. The samples were taken to the laboratory and stored for a minimum period of one month to allow daughter products to come into radioactive equilibrium with their parents 226Ra and 232Th.

The low level gamma-ray spectrometer used in present measurements consists of high purity germanium detector with its electronic circuits. The gamma detector is a coaxial with closed end, facing window geometry with vertical dipstick in the range 500-800 microns. The detector is housed in a shielded chamber made of plexiglass, copper, lead and cadmium of 10, 30, 100 and 3 mm thickness, respectively. The chamber was so designed as to shield the HPGe detector from cosmic rays, the emitted lead X-rays and also the radioactive impurities due to antimony etc. present in lead.

The HPGe gamma spectrometer was calibrated for photon energy and efficiency. The energy calibration was carried out by acquiring gamma spectrum from radioactive standards of known energies i.e. 137Cs $E_γ = 662$ keV and 60Co $E_γ = 1332$ keV. For the efficiency calibration, we used a standard source of 131Ba, 137Cs and 60Co. For measurement of background radiation level, a 1.4 litre Marinelli beaker filled with distilled water was used.

2.1 Analytical technique

Radioactivity measurements of 226Ra, and 232Th were carried out with an HPGe gamma-ray spectrometer. The energy resolution of the spectrometer was measured to be 2.23 keV at 1332 keV from 60Co. A PC based multichannel analyzer with Maestro (EG & G ORTIC) software, was used to acquire and analyze the gamma-ray spectra. Each sample was counted for 10 hr. Prior of sample counting, two background spectra were normally taken twice during week ends for 10 hr each and on average of this background was then subtracted from the samples counted during that week.

The specific activity of $^{226}$Ra and $^{232}$Th was found to be dependent upon the peaks of the decayed products in equilibrium with their parent nuclides. The 226Ra content was measured from gamma-rays of 295.21 keV (19.20%), 351.92 keV (35.10%) from 214Pb and 609.32 keV (44.60%) from 214Bi. The concentration of $^{232}$Th was determined from gamma-energies of 238.63 keV (43.50%) from 212Pb, 583.19 keV (30.58%) from 208Tl, 911.16 keV (26.60%) from 228Ac and 968.97 keV (16.23%) from 228Ac, while 40K activity determined from the 1460.7 keV gamma.

2.2 Theoretical calculations

Radiation hazards due to the specified radionuclides of 226Ra, 232Th and 40K were assessed by two different indices. The most widely used radiation hazard index $Ra_{eq}$ is called the radium equivalent activity. The radium equivalent activity is a weighted sum of activities of the above three radionuclides based on the estimation that 370 Bq.kg$^{-1}$ of $^{226}$Ra, 259 Bq.kg$^{-1}$ of $^{232}$Th and 4810 Bq.kg$^{-1}$ of 40K produce the same $γ$-ray dose rates. $Ra_{eq}$ is given by

$$Ra_{eq}=A_{Ra}+(A_{Th} \times 1.43)+(A_{K} \times 0.077) \quad \ldots (1)$$

Another radiation hazard index called-representative level index - $I_γ$, is defined as follows:

$$I_γ=A_{Ra}/150+A_{Th}/100+A_{K}/1500 \quad \ldots (2)$$

where $A_{Ra}$, $A_{Th}$ and $A_{K}$ are the activity concentrations of $^{226}$Ra, $^{232}$Th and 40K, respectively in Bq.kg$^{-1}$.

The total air absorbed dose rate (nGy.h$^{-1}$) due to the mean specific activity concentrations of 238U, 232Th and 40K Bq.kg$^{-1}$ was calculated using the following formula:

$$D = 0.427 S_U + 0.662 S_{Th} + 0.0432 S_K \quad \ldots (3)$$

where $S_U$ is the mean activity concentration of 238U, $S_{Th}$ is the mean activity concentration of 232Th and $S_K$ is the mean activity concentration of 40K in the samples. For calculating the absorbed dose rate in air at a height of 1.0 m above the ground from measured radionuclide concentrations in environmental materials Eq. (3) was applied.
3 Results and Discussion

The distribution of $^{226}$Ra, $^{232}$Th and $^{40}$K through the two area can be compared by taking into account the mean values of the three elements. From the results, it can be seen that in granite samples, $^{226}$Ra content ranges from 10.0±3.5 to 90.0±9.4 Bq.kg$^{-1}$, $^{232}$Th content ranges from 98.0±9.1 to 160.0±15.3 Bq.kg$^{-1}$. While in $^{40}$K the values was 73.0±8 to 102.0 ± 9 Bq.kg$^{-1}$ as shown in Fig. 1. Generally these values of radium for all granite samples 26.1±5 Bq.kg$^{-1}$ is in the normal values. As results to thorium, its average contents in samples were 122.6±12.9 Bq.kg$^{-1}$ which is also higher than the normal range. The measured activity concentration of 89.4±8.6 Bq.kg$^{-1}$ of $^{40}$K exceeds markedly the values of both uranium and thorium, as it is the most abundant radioactive element under consideration. The world averages for $^{232}$Th and $^{226}$Ra are 25 and 25 Bq.kg$^{-1}$, respectively. The variation of natural radioactivity levels at different sampling sites is due to variation in concentrations of these elements in the geological formations.

The spread of uranium and its daughter radium is attributed to the tendency for the uranium to be either precipitated or leached during the deutric stage which characterizes the later history of granitic bodies. In the case of igneous rocks, uranium, thorium and potassium all increase in overall abundance with increasing the degree of crystallization and tend to accumulate in the later differentiates of igneous melts. The relative abundance of the radioactive elements normally remains fairly constant over this crystallization range. This has useful applications in exploration, since anomaloues radioactive element ratios could indicate uranium mineralization. The values of the concentration show that there is a strong coherence between the three elements in the granitic rocks.

In soil samples $^{226}$Ra content ranges from 7.5±0.9 to 38.0±6 Bq.kg$^{-1}$, $^{232}$Th content ranges from 5.4±0.7 to 18.0±4.3 Bq.kg$^{-1}$, while in $^{40}$K the values ranged from 10.0±3 to 198.0±17 Bq.kg$^{-1}$. The average content of specific activity of $^{226}$Ra was 16.0 ± 4 Bq.kg$^{-1}$, $^{232}$Th was 9.5±3.9 Bq.kg$^{-1}$ and for $^{40}$K was 49.0±7.6 Bq.kg$^{-1}$ as shown in Fig. 1. In grass samples, $^{226}$Ra content ranges from 2.5±0.5 to 32.7±5.4 Bq.kg$^{-1}$, $^{232}$Th content ranges from 0.5±0.02 to 21.3±4.3 Bq.kg$^{-1}$, while for $^{40}$K the values ranged from 1.9±0.5 to 21.3±4 Bq.kg$^{-1}$ as shown in Fig. 1. The average content of specific activity of $^{226}$Ra was 11.9±3.5 Bq.kg$^{-1}$, $^{232}$Th was 7.5±2.7 Bq.kg$^{-1}$ and $^{40}$K was 2.9 ± 0.6 Bq.kg$^{-1}$. The $^{137}$Cs radionuclide was not detected, as it is the main residue of the atmospheric nuclear detonation. Many investigations have shown that $^{137}$Cs in the plant is mostly absorbed from the foliar deposition, and only a very small amount is absorbed from the root.

Although the areal variation in $^{226}$Ra concentrations was found, however the variation is not much as the geological structure of the subsoil in this area is generally lime-stone with a low content of natural radionuclides. Thorium is also usually more abundant in the suspended load than in the bottom sediments. This is interesting because the chemical behaviour of thorium in natural water is quite different from that of uranium. Thorium is essentially insoluble in normal surface water, and so thorium transport is within particulate matter rather than in solutions. Potassium enrichment of soil is not possible due to its mobility.

However, slight variation in the radioactivity content in soil may be observed with different locations worldwide mainly due to soil type, formation, and transport process involved. This may be the reason for the variation observed in our results as compared to those of others.

In water samples $^{226}$Ra content ranges from 0.06 to 0.12 pCi.l$^{-1}$ and $^{232}$Th content ranges from 0.02 to 0.08 pCi.l$^{-1}$ as shown in Fig. 1. The average content of specific activity was 0.09 pCi.l$^{-1}$ for $^{226}$Ra and 0.05 pCi.l$^{-1}$ for $^{232}$Th. A large number samples of drinking water were analyzed for $^{226}$Ra content in Salzburg with ionization chamber techniques. The results indicated that the range of $^{226}$Ra was 0.1 to 8.2 pCi.l$^{-1}$ with a mean value of 1.5 pCi.l$^{-1}$. Similar results were those found in Salzburg and Finland. Nearly 700 ground water samples were analyzed which showed a mean concentration of 0.2 pCi.l$^{-1}$, with the maximum found to be 8.5 pCi.l$^{-1}$. Domestic bottled water showed $^{226}$Ra concentration to be less than an average of 2.2 pCi.l$^{-1}$, while imported bottled water showed concentrations as high as 13.5 pCi.l$^{-1}$ (Ref. 16). Gans et al. investigated $^{226}$Ra content of drinking water (tap water and bottled water) in Germany. The results show that average radioactive contamination for tap water was 0.1 pCi.l$^{-1}$ and that of bottled water was about 1 pCi.l$^{-1}$ of $^{226}$Ra. The maximum value found was 3.7 pCi.l$^{-1}$. The average values were found to be somewhat higher in the present investigation.

The maximum allowed value for radium concentrations in drinking water is 5 pCi.l$^{-1}$ (Ref. 18). The radium concentration in our investigated samples
(which are used for drinking) is found to be below this permissible limit of 5 pCi.l⁻¹. Generally, measures should be taken if the activity of the radioactive contaminates detected in water exceeds the permissible levels. In coastal waters, it can be assumed that the mineral fraction of the sediment has uranium and thorium contents similar to those of terrestrial rocks¹⁹.

In order to compare the radiological effects with the environmental samples containing $^{226}$Ra, $^{232}$Th and $^{40}$K, a common index called the radium equivalent ($R_{aeq}$) has been introduced.

The radium equivalent activities of samples under investigation were calculated on the basis of the permissible levels and are given in Table 1. There are considerable variations in the $R_{aeq}$ of the different type of samples originating from different areas. This fact is important from the point of view of selecting suitable materials for use in building and construction, especially concerning those, which have large variations in their activities.

Large variations in radium equilibrium activities may suggest that it is advisable to monitor the radioactivity levels of materials from a new source before adopting it for use as a building material²⁰. It may be noted that $^{238}$U has been replaced by $^{226}$Ra, a decay product of $^{238}$U as there may be dis-equilibrium between $^{238}$U and $^{226}$Ra (Ref. 21).

The value of level index must be less than unity for the radiation hazard to be negligible. i.e. the radiation exposure due to radioactivity in construction material must be limited to 1.5 mGy.y⁻¹ For the maximum value of level index to be less than unity, the maximum value of $R_{aeq}$ must be less than 370 Bq.kg⁻¹ (Ref. 21). The calculated values of level index for samples studied in this work range from 0.2 to 1.5
with an average of 0.63 Bq.kg\(^{-1}\).

The estimated external gamma doses due to natural gamma emitters present in the samples were 92.2 nGy.h\(^{-1}\) (granite), 15.5 nGy.h\(^{-1}\) (soil), 10.3 nGy.h\(^{-1}\) (grass), while in water the value was 0.07 nGy.h\(^{-1}\). These values were lower than the estimate of average global terrestrial radiation of 55 nGy.h\(^{-1}\) except in granite samples.

4 Conclusion

From the results, it can be seen that: \(^{226}\)Ra and \(^{232}\)Th have measurable values in most samples. The \(^{40}\)K concentration varies in different samples and do not show any observable trends and \(^{137}\)Cs remained to be in undetected in all samples.

Based on these results, it could be said that there are no restrictions on the use of the granite of these location to be used in masonry building purposes. As compared to other environmental samples, very low concentrations of all the detected radionuclides were found in water samples.

\(\text{Ra}_{\text{eq}}, \text{I}_{\gamma r}\) values and gamma absorbed dose rates of granite samples vary appreciably from sample to another due to the variation of radium, thorium and potassium contents. The average \(\text{Ra}_{\text{eq}}\) values for granite rocks in the studied areas are below the internationally accepted values 370 Bq.kg\(^{-1}\). The radiation dose to which workers are subjected is not negligible, although depending on dust inhalation. The values of gamma absorbed dose rates in air of the samples are comparable to the average global terrestrial radiation of 55 nGy.h\(^{-1}\). For samples investigated, the mean value of absorbed dose rates fluctuates from 10.3 to 92.0 nGy.h\(^{-1}\), with average of 39.2 nGy.h\(^{-1}\).

References

5. Allaqi project working paper No.26, Unit of Environmental studies and development, Faculty of Science, Aswan (1995).