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### **Assessment of natural radionuclide content in deposits from drinking water purification station and excess lifetime cancer risk due to gamma radioactivity.**

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#### **ABSTRACT**

The concentrations of natural radionuclide in deposits samples taken from Thirty-six drinking water purification stations have been measured and determined using gamma-ray spectrometry system using (sodium iodide NaI (Tl) detector). Knowledge of radioactivity present in deposits of drinking water purification station enables one to assess any possible radiological hazard to humankind by the use of such materials. The natural radionuclide ( $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ ) contents have been analyzed for the deposits samples with an aim of evaluating the radiation hazard nature. The Absorbed dose rate, The annual effective dose equivalent, Radium equivalent activities, Hazard indices ( $H_{\text{ex}}$  and  $H_{\text{in}}$ ), Gamma index, Excess lifetime cancer risk and Annual gonadal dose equivalent were calculated for investigated area. Results of the study could serve as an important baseline radiometric data for future epidemiological studies and monitoring initiatives in the study area.

***Keywords: Natural radionuclide, drinking water purification stations and radiological implications.***

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#### **INTRODUCTION**

Natural radionuclides have been components of earth since its existence. There are many naturally occurring radionuclides in environment, containing uranium and thorium series radioisotopes and natural  $^{40}\text{K}$ . Natural radionuclides are widely spread in earth's environment, they exist in soil, sediment, water, plants and air. Natural environmental radioactivity and associated external exposure due to gamma radiation depend primarily on the geological conditions and soil and sediment formations of each region in the

world. The main objective of this study was to determine natural radionuclide activity concentrations in sediment samples from some drinking water purification stations. For control and monitoring of radioactivity levels in El-Mynia governorate concentrations of natural ( $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ ) radionuclides were measured in sediment using gamma-ray spectrometry [1].

## **MATERIALS AND METHODS**

### ***2.1. Study area***

The present study area covers drinking water purification stations in Eledwa, Samalot, Matti, Dermwas, Maghaha, Beni Mazar and Abu Korqas city as shown in figure (1). Minia governorate is one of the important agricultural and industrial regions in Egypt. Minia is mainly an agricultural governorate, as it has around 6% of the total agricultural lands in Egypt, producing cotton, wheat, corn and potatoes. In addition, it encompasses several industrial activities including textile and weaving, packing and freezing vegetables, fish farms and several other activities. Therefore, the Nile River is the lifeline of Minia governorate.

### ***2.2. Sample collection preparation***

The samples in present study area, from which seven city and 36 locations were selected. The samples were collected May 2010 - June 2011, the recently deposited sediment samples were manually collected with the help of a plastic spade in polyethylene bags. Sediment samples were oven dried at a temperature of  $110\text{ }^{\circ}\text{C}$  for 12 h and sieved through a 200 mesh. The dried samples were transferred to polyethylene Marinelli beakers of  $350\text{ cm}^3$  capacity. Each sediment sample was left for at least 4 weeks to reach secular equilibrium between radium and thorium, and their progenies [2, 3].

### ***2.3. Gamma spectrometric analysis***

Activity measurements have been performed by gamma ray spectrometer, employing a scintillation detector ( $3 \times 3$  inch). It is hermetically sealed assembly, which includes a NaI (TI) crystal, coupled to PC-MCA Canberra Accuspec. To reduce gamma ray background, a cylindrical lead shield (100 mm thick) with a fixed bottom and movable cover shielded the detector. The lead shield contained an inner concentric cylinder of copper (0.3 mm thick) in order to absorb X-rays generated in the lead. In order to determine the background distribution in the environment around the detector an empty sealed beaker was counted in the same manner and in the same geometry as the

samples. The measurement time of activity or background was 43200s. The background spectra were used to correct the net peak area of gamma rays of measured isotopes. A dedicated software program [4].



**Fig. (1): Samples location.**

The  $^{226}\text{Ra}$  radionuclide was estimated from the 609.3 keV (46.1%)  $\gamma$ -peak of  $^{214}\text{Bi}$ , 351.9 keV (36.7%), 1120.3 keV (15%), 1728.6 keV (3.05%) and 1764 keV (15.9%)  $\gamma$ -peak of  $^{214}\text{Pb}$ . The 186 keV photon peak of  $^{226}\text{Ra}$  was not used because of the interfering peak of  $^{235}\text{U}$  with energy of 185.7 keV.  $^{232}\text{Th}$  radionuclide was estimated from the 911.2 keV (29%)  $\gamma$ -peak of  $^{228}\text{Ac}$  and

238.6 keV (43.6%)  $\gamma$  -peak of  $^{212}\text{Pb}$ .  $^{40}\text{K}$  radionuclide was estimated using 1,461 keV (10.7%)  $\gamma$  -peak from  $^{40}\text{K}$  itself. The below detectable limit (BDL) were 25.2 Bq kg<sup>-1</sup> for  $^{40}\text{K}$ , 6.5 Bq kg<sup>-1</sup> for  $^{226}\text{Ra}$  and 5.7 Bq kg<sup>-1</sup> for  $^{232}\text{Th}$ . All procedures were described in previous publications [5].

## RESULTS AND DISCUSSION

### 3.1. Radioactivity analysis

The distribution of the detected radionuclides  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the sediment samples are shown in Table. (1). The average activity concentrations ranged from 37 to 95, from 17 to 62 and from 200 to 314 Bqkg<sup>-1</sup> for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , Respectively.

**Table (1): Average activity concentration of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in sediment samples.**

Location	$^{226}\text{Ra}$ BqKg <sup>-1</sup>	$^{232}\text{Th}$ BqKg <sup>-1</sup>	$^{40}\text{K}$ BqKg <sup>-1</sup>
Eledwa	48±7	22±1	200±10
Samalot	52±3	49±3	252±12
Matti	62±3	17±1	314±14
Dermwas	95±5	62±4	246±13
Maghagha	37±2	32±2	233±12
Beni mazar	38±2	54±3	282±14
Abu korqas	55±3	41±3	232±12

Table (1) shows that, the average activity concentrations vary from location to location, because river bottom can exhibit large variations in chemical, mineralogical properties and rare-earth elements [6]. The  $^{40}\text{K}$  activity concentration dominates over  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  elemental activities like what normally happens in soil.

### 3.2. Evaluation of radiological hazard effects

#### 3.2.1. Absorbed dose rate

It is the first major step for evaluating the health risk. With regard to biological effects, the radiological and clinical effects are directly related to the absorbed dose rate [6]. The measured activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  are converted into doses by applying the conversion factors 0.462, 0.604 and 0.0417 for uranium, thorium and potassium, respectively [7]. These factors are used to calculate the total doserate (D) (nGy/h) using the following equation:

$$D = 0.462C_{\text{Ra}} + 0.604C_{\text{Th}} + 0.0417C_{\text{K}} \quad (\text{nGy}^{-1})$$

where  $C_R$ ,  $C_{Th}$  and  $C_{K}$  are the activity concentrations (Bq/kg) of  $^{226}Ra$ ,  $^{232}Th$  and  $^{40}K$  in river sediments, respectively. The calculated values for samples are presented in Table (2). The ranges of Average absorbed dose rate is from 43.8 to 91.6 (nGy/h) in locations Eledwa, Samalot, Matti, Dermwas, Maghagha, Beni mazar and Abu korqas, respectively. Average absorbed dose rate for sediment samples in Eledwa, Matti and Maghagha is lower than the world average value (57 nGy/h) [7]. While the average absorbed dose rate for sediment samples in Samalot, Dermwas, Beni mazar and Abu korqas is higher than the world average value (57 nGy/h) [7]. Table (2) reported that, the highest average value of dose rate was found in Dermwas.

**Table (2):** Average dose rates, AEDE (indoor and outdoor) and  $Ra_{eq}$ .

Location	Dose rates	AEDE( $\mu$ Sv/y)		$Ra_{eq}$
	(nGy/h)	Outdoor	Indoor	Bqkg <sup>-1</sup>
Eledwa	43.8	53.2	214.9	94.9
Samalot	64.1	77.8	314.6	141.5
Matti	52.0	63.1	255.1	110.5
Dermwas	91.6	111.2	449.3	202.6
Maghagha	46.1	56.0	226.3	100.7
Beni mazar	61.9	75.2	303.8	136.9
Abu korqas	59.8	72.6	293.6	131.5

### 3.2.2. The annual effective dose equivalent (AEDE)

Annual estimated average effective dose equivalent received by a member was calculated using a conversion factor of  $0.7 \text{ Sv Gy}^{-1}$ , which was used to convert the absorbed rate to human effective dose equivalent with an outdoor occupancy of 20% and 80% for indoors [8]. The annual effective dose is determined using the following equations:

$$\text{AEDE (Outdoor) } (\mu\text{Sv/y}) = \text{Absorbed dose (nGy/h)} \times 8760\text{h} \times 0.7\text{Sv/Gy} \times 0.2 \times 10^{-3}$$

$$\text{AEDE (Indoor) } (\mu\text{Sv/y}) = \text{Absorbed dose (nGy/h)} \times 8760\text{h} \times 0.7\text{Sv/Gy} \times 0.8 \times 10^{-3}$$

Table (2) observed the calculated minimum and maximum value of annual effective dose equivalent for investigated locations. The average value indoor AEDE for sediment samples are lower than the world average values  $450 \mu\text{Sv/y}$  [9]. The average values outdoor annual effective dose equivalent for sediment samples in Samalot, Dermwas, Beni mazar and Abu korqas are higher than the world average values  $70 \mu\text{Sv/y}$  [9] where the outdoor AEDE do exceed the world average due to the presence of high activity concentration of  $^{232}Th$  and  $^{40}K$ .

### 3.2.3. Radium equivalent activities ( $Ra_{eq}$ )

The results were evaluated in terms of the radiation hazard by means of the radium equivalent activity ( $Ra_{eq}$ ). Radium equivalent activity is a widely used hazard index and it is calculated through the relation given by [10]. It is assumed that 370 Bq/kg of  $^{226}\text{Ra}$ , 259 Bq/kg of  $^{232}\text{Th}$  and 4810 Bq/kg of  $^{40}\text{K}$  produce the same gamma-ray dose rate

$$Ra_{eq}(\text{Bq/kg}) = C_{Ra} + 1.43C_{Th} + 0.077C_K$$

where  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in Bq/kg, respectively. The range of  $Ra_{eq}$  is estimated for the collected samples and is given in Tables (2). The estimated average values are lower than the recommended maximum value of 370 Bq/kg for the safe use of materials in the construction of buildings [7], because the leaching of heavy minerals by continuous flow of water in the river [6].

### 3.2.4. Hazard indices ( $H_{ex}$ and $H_{in}$ )

Beretka and Mathew [10] defined two indices that represent external and internal radiation hazards. The prime objective of these indices is to limit the radiation dose to a dose equivalent limit of 1mSv/y. The external hazard index ( $H_{ex}$ ) is calculated using the given equation

$$H_{ex} = (C_{Ra}/370 + C_{Th}/259 + C_K/4810) \leq 1$$

where,  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in Bq/kg, respectively.  $H_{ex}$  must not exceed the limit of unity for the radiation hazard to be negligible. On the other hand, the internal hazard index ( $H_{in}$ ) gives the internal exposure to carcinogenic radon and its short-lived progeny [6] and is given by the following formula [9, 10]

$$H_{in} = (C_{Ra}/185 + C_{Th}/259 + C_K/4810) < 1$$

where,  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in Bq/kg, respectively. The value of  $H_{in}$  must also be less than unity to have negligible hazardous effects of radon and its short-lived progeny to the respiratory organs [7]. Table (3) shows that, the calculated average values of hazard indices for all samples are less than unity.

**Table (3): Average hazard indices ( $H_{ex}$ ,  $H_{in}$ ,  $I_\gamma$ , ELCR and AGDE) for investigated samples.**

Location	Hazard indices				
	$H_{ex}$	$H_{in}$	$I_\gamma$ (mSv/y)	ELCR $\times 10^{-4}$	AGDE
Eledwa	0.3	0.4	0.3	1.9E-04	303.1
Samalot	0.4	0.5	0.5	2.7E-04	444.6
Matti	0.3	0.5	0.4	2.2E-04	361.2
Dermwas	0.5	0.8	0.7	3.9E-04	630.0
Maghagha	0.3	0.4	0.4	2.0E-04	321.3
Beni mazar	0.4	0.5	0.5	2.6E-04	431.7
Abu korgas	0.4	0.5	0.5	2.5E-04	414.2

### 3.2.5. Gamma index ( $I_\gamma$ )

Another radiation hazard, called the gamma activity concentration index,  $I_\gamma$ , has been defined by the European Commission [11, 12] and it is given below:

$$I_\gamma = (C_{Ra}/300 + C_{Th}/200 + C_K/3000)$$

where,  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in Bq/kg, respectively. The index  $I_\gamma$  is correlated with the annual dose rate due to excess external gamma radiation caused by superficial material. Values of index  $I_\gamma \leq 2$  corresponds to a dose rate criterion of  $0.3 \text{ mSv/y}^{-1}$ , whereas  $I_\gamma \leq 6$  corresponds to a criterion of  $1 \text{ mSv/y}^{-1}$  [11, 13]. Thus, the activity concentration index should be used only as screening tool for identifying materials that might be of concern to be used as construction materials, though materials with  $I_\gamma > 6$  should be avoided [14] since these values corresponds to dose rates higher than  $1 \text{ mSv/y}^{-1}$  [11], which is the highest value of dose rates recommended for population [7].

The distribution of values of the index  $I_\gamma$  for Nile River sediment, which used as building materials analyzed in this work, is presented in Table 3. The average gamma index  $I_\gamma$  in sediment samples varied between 0.3 and 0.7. All the values of  $I_\gamma$  are  $< 1$ . Therefore, the annual effective dose delivered by sediment samples is smaller than the annual effective dose constraint of  $1 \text{ mSv/y}^{-1}$ . Hence, these building materials can be exempted from all the restriction concerning radioactivity.

### 3.2.6 Excess lifetime cancer risk (ELCR)

Excess lifetime cancer risk (ELCR) was calculated using the following equation and presented in Table 3.

$$\text{ELCR} = \text{AEDE} \times \text{DL} \times \text{RF}$$

where AEDE, DL and RF is the annual effective dose equivalent, duration of life(70 years)and risk factor ( $\text{Sv}^{-1}$ ), fatal cancer risk per sievert. For stochastic effects, ICRP60 uses values of 0.05 for the public [6]. The calculated average values of ELCR showed that, the highest average values were found in Mattay. These average values of ELCR are less than the world average  $0.29 \times 10^{-3}$  [7] for sediment sample in Minia, Egypt.

### 3.2.7 Annual gonadal dose equivalent (AGDE)

The activity bone marrow and the bone surface cells are considered as the organs of interest by [15]. Therefore, the AGDE due to the specific activities of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  was calculated using the following formula [16]:

$$\text{AGDE } (\mu\text{Sv/y}) = 3.09 C_{\text{Ra}} + 4.18 C_{\text{Th}} + 0.314 C_{\text{K}}$$

The obtained values of AGDE are listed in Tables 3. The average values of AGDE varied from 303.1 to 630  $\mu\text{Sv/year}$ . Table 3 shows that, the highest average value of AGDE was found to be 630  $\mu\text{Sv/year}$  in Dermwas.

## CONCLUSIONS

The activity levels and distribution of natural terrestrial radionuclides of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  were measured by gamma-ray spectrometry system for deposit of some drinking water purification stations collected from Minia in Egypt. From the measured values, the average values of absorbed dose rate in air (D), radium equivalent activity ( $\text{Ra}_{\text{eq}}$ ), Hazard indices ( $\text{H}_{\text{ex}}$  and  $\text{H}_{\text{in}}$ ), annual gonadal dose equivalent (AGDE) and annual effective dose equivalent (AEDE) (outdoor and indoor) were calculated. The Radium equivalent activity ( $\text{Ra}_{\text{eq}}$ ) and Hazard indices ( $\text{H}_{\text{ex}}$  and  $\text{H}_{\text{in}}$ ) and annual gonadal dose equivalent (AGDE) are calculated to assess the radiological hazard of sand mixed with sediment since sand is used as construction materials in this region. This study can be used as a baseline for future investigations and the data obtained in this study may be useful for natural radioactivity mapping. The results may also be used as a reference data for monitoring possible radioactivity pollutions in future.

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## المؤتمر الدولي الثالث للعلوم الإشعاعية وتطبيقاتها

١٢ - ١٦ نوفمبر ٢٠١٢ - الغردقة - مصر

### تقييم محتوى النويدات المشعة طبيعيا في رواسب محطات معالجة مياه الشرب وخطر الإصابة بمرض السرطان بسبب النشاط الإشعاعي لأشعة جاما

تم قياس تركيزات النويدات المشعة الطبيعية في عينات أخذت من ستة وثلاثين محطة لتنقية مياه الشرب باستخدام مطياف أشعة جاما (كاشف يوديد الصوديوم). معرفة النشاط الإشعاعي الموجودة في محطات تنقية مياه الشرب تمكننا من تقييم أي خطر إشعاعي محتمل للجنس البشري عن طريق استخدام مثل هذه المواد. لقد تم قياس النويدات المشعة الطبيعية ( $^{226}\text{Ra}$ ) طبيعيا في محطات تنقية مياه الشرب وذلك بهدف تقييم المخاطر الإشعاعية الطبيعية لذا تم حساب الاتي:-

- ١- معدل الجرعة الممتصة.
  - ٢- الجرعة السنوية الفعالة.
  - ٣- المكافئ الريديومي.
  - ٤- مؤشرات الخطر الإشعاعي الطبيعي الداخلي والخارجية.
  - ٥- مؤشر جاما.
  - ٦- احتمالية الإصابة بمرض السرطان.
  - ٧- احتمالية إصابة الغدد التناسلية باى خطر اشعاعى.
- تعتبر نتائج الدراسة بمثابة بيانات أساسية وهامة للنشاط الإشعاعي الطبيعي فى حالة الحاجة للدراسات الوبائية في المستقبل، ومبادرات الرصد في منطقة الدراسة.