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A Comparative Study of the Radiological Hazard in Sediments Samples from Drinking Water Purification Plants Supplied From Different Sources

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Abstract: *The natural radiation level has been determined for 135 sediment samples from forty-six drinking water purification plants supplied from different sources (Nile River, Ibrahimia Canal and Bahr Yousif Canal) with an aim of evaluating the radiation hazard. The concentration of natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K) has been investigated by using gamma spectrometry (NaI (Tl) 3"x 3") detector. The results showed that the concentrations of average activity in the sediment samples collected from drinking water purification plants supplied from Nile River, Ibrahimia Canal and Bahr Yousif Canal are (29±2, 30±2 and 240±8 Bq kg^{-1}), (47±3, 46±8 and 258±12 Bq kg^{-1}) and (28±2, 27±3 and 219±18 Bq kg^{-1}) for ^{226}Ra, ^{232}Th and ^{40}K, respectively. In general, the distributions of average activity concentrations of samples under investigation are within the world values although some extreme values have been determined. Radiological hazard effects such as: absorbed dose rate (D), outdoor and indoor annual effective dose equivalent (AEDE), radium equivalent activities (Ra_{eq}), hazard indices (H_{ex} and H_{in}), gamma index (I_{γ}), excess lifetime cancer risk (ELCR) and annual gonadal dose equivalent (AGDE) for the corresponding samples were also estimated.*

Key word: *Drinking water purification plants, Sediment, Radiological hazard indices*

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}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 (Aközcan, 2012). Sediment (or deposits of drinking water purification station) plays a predominant role in aquatic radioecology. Sediments are formed when rocks and/or organic materials are broken into small pieces by moving water. Sediment later settles out of the moving water. The radioactivity contents inside the material are normally unaffected because the breaking of a rock into pieces does not change its chemical composition. Sediment plays a role in accumulating and transporting contaminants within the geographic area. Sediment is considered as the environmental host of the waste, which is charged by natural or artificial processes (Suresh et al., 2011a).

Sediments of drinking water purification station are detritus products of rocks and bear the mineralogical properties of the original rock formation. Among the various building materials, sediment (or deposits of drinking water purification station) is one of the most important and major mixing materials for building construction in Egypt, especially in Upper Egypt.

In addition to being the main source of continuous radiation exposure to human, sediment acts as a medium of migration for transfer of radionuclides to the biological systems and hence, it is the basic

indicator of radiological contamination in the environment. Natural radionuclides in river sediment generate a significant component of the background radiation exposure of the population (Suresh et al., 2011b). Therefore, the knowledge of the concentrations and distributions of the natural radionuclides in the deposit samples are of great interest since it provides useful information in monitoring of environmental contamination and associated human health by natural radioactivity.

Man-made or anthropogenic radionuclides are created via human activities, which vary with time and location. Sources of man-made radionuclides are divided into nuclear and non-nuclear industries. Mining (especially uranium and thorium), phosphate fertilizers manufacture, agricultural applications, coal combustion, cement production, street construction and other human activities are non-nuclear industries which have produced and redistributed increasing amounts of radioactive matter leading to a considerable contribution to the radio-ecological pollution (Al-Trabulsy et al., 2011).

Minya governorate is industrial provinces in Egypt. Where there are many industries such as: cement plants black and white, quarries of iron, steel, sand, and marble and factories of oils, carbonated water, sugar, brick sandy, and tile. 4,701 million people drink from forty-six plant to purify drinking water in Minya.

Materials and methods

Sampling and sample preparation

Minya, located in southern Cairo, the present study covered an area in the Minya governorate from (38° 28' 39" N; 30° 83' 32" E) to (38° 37' 34"N; 30° 98' 03" E), about 139 km, as shown in figure (1). 135 samples were collected from 46 drinking water purification stations supplied from three different sources. The first source is the Nile River (NR), it serves twenty-five drinking water purification stations; the second source is the Ibrahimia Canal (IC), it serves thirteen drinking water purification stations and the third source is the Bahr Yousif Canal (BY), it serves eight drinking water purification stations. Samples were collected every month. Deposit samples were oven dried at a temperature of 105° C for 12 h and sieved through a 200 mesh. The dried samples were transferred to polyethylene Marinelli beakers. Each deposit sample was left for at least 4 weeks to reach secular equilibrium between radium and thorium, and their progenies (Kurnaz et al., 2007).

Gamma spectrometric analysis

Activity measurements have been performed by gamma ray spectrometer, employing a scintillation detector (3 × 3 inch). It is hermetically sealed assembly, which includes a NaI (Tl) 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 43200 s. The background spectra were used to correct the net peak area of gamma rays of measured isotopes. A dedicated software program, Genie 2000 from Canberra, was used to carry out the on-line analysis of each measured gamma-ray spectrum. The ^{226}Ra radionuclide was estimated from the 351.9 keV (36.7%) γ-peak of ^{214}Pb and 609.3 keV (46.1%), 1120.3 keV (15%), 1728.6 keV (3.05%) and 1764 keV (15.9%) γ-peaks of ^{214}Bi. The 186 keV photon peak of ^{226}Ra was not used because of the interfering peak of ^{235}U with energy of 185.7 keV. ^{232}Th radionuclide was estimated from the 911.2 keV (29%) γ-peak of ^{228}Ac, 238.6 keV (43.6%) γ-peak of ^{212}Pb and 583.1 kev (84.5) γ-peak of ^{208}Tl. ^{40}K radionuclide was estimated using 1,461 keV (10.7%) γ -peak from ^{40}K itself. All procedures were described in previous publications (Issa et al., 2012).

Results and discussion

Radionuclide activity concentrations

The range and average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the sediment samples from forty-six drinking water purification plants supplied from Nile River, Ibrahimia Canal and Bahr Yousif Canal are presented in Tables 1–3, respectively. For drinking water purification plants supplied from Nile River, the distribution of the detected radionuclides ^{226}Ra, ^{232}Th and ^{40}K in the all sediment samples are shown in Figures 2-4. The activity concentration ranges for ^{226}Ra, ^{232}Th and ^{40}K are found from 10±0.9 to 70±5 Bqkg^{-1}, from 10±0.6 to 94±5 Bqkg^{-1} and from 131±7 to 463±23 Bqkg^{-1}, respectively. The activity concentration are presented in Table 1 vary from site to site, because river bottom can exhibit large variations in chemical, mineralogical properties and rare-earth elements (Krmar et al., 2009).

The world average of ^{226}Ra, ^{232}Th and ^{40}K is 33, 45 and 420 Bq kg^{-1}, respectively (UNSCEAR, 2000). The highest values of ^{226}Ra were found in Matay and Minya drinking water purification plants, while the Bani Mazar, Matay, Samalut and Minya drinking water purification plants are reported the highest values of ^{232}Th. The ^{40}K values are higher than the world average in Matay drinking water purification plants. Bani Mazar, Matay, Samalut and Minya drinking water purification plants are recorded the activity concentration higher than world average values, may be due to the phosphate fertilizer industries located on the Nile bank before drinking water purification plants. The investigation of ^{226}Ra: ^{232}Th activity ratio calculations revealed that ^{226}Ra activity concentrations are seen to be on the average 1.3 times higher than the ^{232}Th activity concentration in the measured sediments collected from drinking water purification plants supplied from Nile River. Fig. 5 shows the different trends of the spatial distribution of ^{226}Ra: ^{232}Th. Maghagha showed the highest ^{226}Ra: ^{232}Th ratio 3.17. This indicated that relatively higher amount (approximately 3 times) of ^{226}Ra (38 Bq kg^{-1}) exist than ^{232}Th (12 Bq kg^{-1}). This is could be due to the presence of the loamy sediments at Maghagha which have relatively high amount of ^{226}Ra (El-Gamal et al., 2007). This is in agreement with Issa (Issa et al., 2013).

For drinking water purification plants supplied from Ibrahimia Canal, The measured activity concentrations of the 38 sediment samples are presented in Table 2. The distribution of the natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in the all sediment samples are shown in Figures 6-8. For ^{226}Ra values are in general higher than the world average value.

Bani Mazar and Dayr Mawas drinking water purification plants are reported the activity concentration of ^{232}Th is higher than the world average value, while the highest value of ^{40}K was found in Bani Mazar Mawas drinking water purification plants. The possible source of radioactivity in Ibrahimia Canal is the authorized release of man-made radionuclides into the system, such as cement and sugar industries. However, in most regions the contribution proceeds from sewage. Fig. 9 shows the different trends of the spatial distribution of ^{226}Ra: ^{232}Th. Abu Qurqs showed the highest ^{226}Ra: ^{232}Th ratio 3.67. This indicated that relatively higher amount (approximately 4 times) of ^{226}Ra (55 Bq kg^{-1}) exist than ^{232}Th (15 Bq kg^{-1}). This is could be due to the presence of the sugar industries waste to Ibrahimia Canal.

For drinking water purification plants supplied from Bahr Yousif Canal, the concentration of ^{226}Ra, ^{232}Th and ^{40}K are shown in Figures 10-12. As observed in Table 3, the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K are in general lower than the world average values. Average activity concentration of natural radionuclide in sediment samples collected from drinking water purification plants supplied from Nile River, Ibrahimia Canal and Bahr Yousif Canal are lower than the world average values, as listed in Tables 1-3. Sediment samples from drinking water purification plants supplied from Ibrahimia Canal are presented the highest average activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, as shown in Figure 13.

Evaluation of Radiological Hazard Effects

Absorbed dose rate (D)

The absorbed dose rates due to gamma radiations in air at 1 m above the ground surface for the uniform distribution of the naturally occurring radionuclides ^{226}Ra, ^{232}Th and ^{40}K were calculated based on guidelines provided by UNSCEAR (UNSCEAR, 2000). The conversion factors used to compute absorbed gamma dose rate (D) in air per unit activity concentration in Bq kg^{-1} corresponds to 0.462 nGy h^{-1} for ^{226}Ra, 0.604 nGy h^{-1} for ^{232}Th and 0.042 nGy h^{-1} for ^{40}K. Therefore D can be calculated as follows (UNSCEAR, 2000): $D\ (nGy\ h^{-1}) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K$ (1)
where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg $^{-1}$, respectively.

Absorbed dose rate (D) of 74 sediment samples collected from drinking water purification plants supplied from Nile River are presented in Table 4. As could be seen from the table, the absorbed dose rate values in sediment samples NR (16, 17, 18, 31, 32, 33, 34, 35, 36, 38, 40, 41 and 42) are higher than the permissible level corresponding to 55 nGy h^{-1} (UNSCEAR, 2000). The frequency distribution (in percent) of the total absorbed dose rates of the seventy-four measured samples is plotted in Fig. 14. Nineteen samples exhibit dose rates that range from 15 to 30 nGy h^{-1}. Thirty-three samples ranged from 30 to 50 nGy h^{-1} and twenty-two of the samples exhibit values that range from 50 to 165 nGy h^{-1}.

The range and average of absorbed dose rate values in thirty-eight sediment samples from drinking water purification plants supplied from Ibrahimia Canal are presented in Table 5. As observed, the deposit samples collected from Bani Mazar and Dyar Mawas drinking water purification plants are higher than the permissible level corresponding to 55 nGy h^{-1} (UNSCEAR, 2000).

Table 6 shows that, the absorbed dose rate of sediment samples collected from drinking water purification plants supplied from Bahr Yousif Canal. Absorbed dose rate in all sediment are lower than the permissible level corresponding to 55 nGy h^{-1} (UNSCEAR, 2000). The comparative study observed that, the average absorbed dose rate value of sediment from drinking water purification plants supplied from Ibrahimia Canal is highest absorbed dose rate from the other drinking water purification plants supplied from Nile River and Bahr Yousif Canal (see Fig. 13). The frequency of absorbed dose rate in the range of 5-165 nGy h^{-1} for sediment samples collected from drinking water purification plants supplied from Ibrahimia Canal is 12% higher than the frequency of absorbed dose rate in the same range for sediment samples collected from drinking water purification plants supplied from Nile River, and 16% higher than the frequency of absorbed dose rate in the range of 5-165 nGy h^{-1} for sediment samples collected from drinking water purification plants supplied from Bahr Yousif Canal, as shown in Fig. 14.

The Annual Effective Dose Equivalent (AEDE)

Since the present sediments are the main mixing material with cement and cement products for building construction in the area under investigation, the determination of annual effective dose equivalent of each site sample is important. For that, the living style of the people or indoor or outdoor factor of a location is considered (Ramasamy et al., 2011). A typical resident in a location, both male and female, would spend about 8 h of the day in an office or classroom or laboratory, 11 h indoors and the remaining 5 h outdoors. This applies to the greater part of the population in a location who are either office workers or public/students. Hence, 19/24 (0.8) and 5/24 (0.2) is adopted as the indoor (80%) and outdoor occupancy factors (20%), respectively, with a conversion factor of 0.7 Sv Gy^{-1} to convert absorbed dose rate nGy h^{-1} to annual effective dose equivalent μSv y^{-1} for this study (Singh et al., 2009). The annual effective dose is determined using the following equations:

$$\text{AEDE (Outdoor)} = D \times 8760h \times 0.7\ SvGy^{-1} \times 0.2 \times 10^{3} \quad (2)$$

$$\text{AEDE (Indoor)} = D \times 8760h \times 0.7\ SvGy^{-1} \times 0.8 \times 10^{3} \quad (3)$$

As could be seen from the table 4, the outdoor annual effective dose of deposits samples from drinking water purification plant (DWPP) supplied from Nile River are lower than the world average values 70 μSv y^{-1} (Orgun et al., 2007), except for sediment samples collected from Bani Mazar, Matay, Samalut and Minya DWPPs. While the indoor annual effective dose values are in general lower than world average values 450 μSv y^{-1} (Orgun et al., 2007), except in site numbers NR16, NR 31 and NR 41 the results are higher than world average values. Outdoor and indoor annual effective dose of deposits samples from DWPP supplied from Ibrahimia Canal are listed in Table 5. Sediment samples from Bani Mazar and Dayr Mawas DWPPs are higher than world average values for outdoor and indoor annual effective dose (Orgun et al., 2007). Indoor and outdoor annual effective dose values of sediment samples from DWPP supplied from Bahr Yousif Canal are lower than world average values (Orgun et al., 2007), see Table 6.

Radium equivalent activities (Ra_{eq})

Radium equivalent (Ra_{eq}) index in Bq kg^{-1} is a widely used radiological hazard index. It is a convenient index to compare the specific activities of samples containing different concentrations of ^{226}Ra, ^{232}Th and ^{40}K. This index can be calculated according to Beretka and Mathew (Beretka and Mathew, 1985):

$Ra_{eq} = A_{Ra} + 1{:}43A_{Th} + 0{:}077A_K$ (4)

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K K in Bq kg^{-1}, respectively.

Tables 4-6 show that, the average radium equivalent values are 90, 133, 84 Bq kg^{-1} for sediment samples collected from DWPPs supplied from Nile River, Ibrahimia Canal and Bahr Yousif Canal, respectively. The average values are lower than the limit of 370 Bq kg^{-1} adopted by the Organization for Economic Cooperation and Development (Beretka and Mathew, 1985), but the average radium equivalent of sediment collected from DWPPs supplied from Ibrahimia Canal is highest the average radium equivalent of sediment collected from DWPPs supplied from Nile River, and Bahr Yousif Canal, as shown in Fig. 13.

Hazard indices (Hex and Hin)

In order to measure the hazards one can define radiation hazard indices (Ahmad and Matiullah Hussain, 1988): the external radiation hazard (H_{ex}), and internal radiation hazard (H_{in}). The external hazard index is another criterion to assess the radiological suitability of a material. It is defined as follows:

$H_{ex} = A_{Ra}/370 + A_{Th}/258 + A_K/4810$ (5)

where A_{Ra}, A_{Th} and A_K are the activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq kg^{-1}. The values of the indices should be < 1. The internal hazard index is a criterion for index radiation hazard. In addition to gamma rays, ^{222}Rn plays an important role for internal exposure in a room. Effectively, the radio toxicity of ^{238}U is increased by a factor of two to allow for the contribution from ^{222}Rn and its short lived progeny. The internal exposure due to radon and its daughter products is quantified by the internal hazard index H_{in} (Ravisankar et al., 2012), which has been defined as shown below:

$H_{ex} = A_{Ra}/185 + A_{Th}/258 + A_K/4810$ (6)

The internal hazard index is defined so as to reduce the acceptable maximum concentration of ^{238}U to half the value appropriate to external exposures alone. For the safe use of materials in the construction the following criterion was proposed by Krieger (Krieger, 1981) $H_{in} \leq 1$. In general, the external and internal hazard indices of deposit samples collected from DWPPs supplied from Nile River, Ibrahimia Canla and Bahr Youfis Canal are less than the unity, as listed in Tables 4-6, respectively.

As could be seen from Fig. 15, the external and internal hazard indices of deposit samples from DWPPs supplied from Ibrahimia Canla are highest the external and internal hazard indices of deposit samples from DWPPs supplied from Nile River and Bahr Youfis Canal.

Gamma index (Iγ)

A number of indices dealing with the assessment of the excess gamma radiation arising from building materials such as external and internal hazard indices and gamma-concentration indices have been proposed by several investigators (Beretka and Mathew, 1985; Krieger, 1981). In this study, the gamma-index (I_γ) was calculated as shown below:

$I_\gamma = C_{Ra}/300 + C_{Th}/200 + C_K/3000$ (7)

where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq.kg^{-1}, respectively. The value of gamma index depends on the dose criterion and the mode and quantity of the materials used in a building. As is suggested by the European commission for the materials used in bulk amounts, the exemption dose criterion (0.3 mSv y^{-1}) corresponds to $I_g \leq 0.5$, whereas the dose criterion 1 mSv/y corresponds to $I\gamma \leq 1$ (EC, 1999). On the other hand, for superficial and other

materials, the corresponding values of I_γ should be between 2 and 6. From the calculated values (see Tables 4-6), the ranges of I_γ are 0.1–0.8, 0.2-1.3 and 0.2-0.4 of sediment samples from DWPPs supplied from Nile River, Ibrahimia Canal and Bahr Yousif Canal, respectively. No exceeding of the recommended upper limit is noted, but the I_γ of sediment samples from DWPPs supplied from Nile River is highest the I_γ of sediment samples from DWPPs supplied from Ibrahimia Canal and Bahr Yousif Canal, as shown in Fig. 15.

Excess lifetime cancer risk (ELCR)

Excess lifetime cancer risk (ELCR) is calculated using the following equation:

$$ELCR = AEDE \times DL \times RF \quad (8)$$

where AEDE, DL and RF is the annual effective dose equivalent, duration of life (70 years) and risk factor (Sv^{-1}), fatal cancer risk per sievert. For stochastic effects, ICRP 60 uses values of 0.05 for the public (Taskin et al., 2009). Excess lifetime cancer risk of sediment samples collected from DWPPs supplied from Nile River are presented in Table 4. The calculated range of ELCR is (0.8-4.3) × 10^{-4} with an average of 1.8 × 10^{-4}, the average value of ELCR are less than the world average 2.9 × 10^{-4}. Bani Mazar, Matay, Samalut and Minya DWPPs have higher ELCR values.

Table 5 presents the excess lifetime cancer risk of sediment samples collected from DWPPs supplied from Ibrahimia Canal. The calculated range of ELCR is (1.2-7.1) × 10^{-4} with an average of 2.6 × 10^{-4}, The average value of ELCR are less than the world average 2.9 × 10^{-4} (UNSCEAR, 2000). Calculated excess lifetime cancer risk of sediment samples collected from DWPPs supplied from Bahr Yousif Canal are listed in Table 6. As could be seen, the range of (1.1–2.3) × 10^{-4} and an average of 1.7 × 10^{-4} are lower than the world average 2.9 × 10^{-4} (UNSCEAR, 2000). Excess lifetime cancer risk of sediment samples collected from DWPPs supplied from Ibrahimia Canal is 31% higher than the Excess lifetime cancer risk of sediment samples collected from DWPPs supplied from Nile River, and is 34% higher than Excess lifetime cancer risk of sediment samples collected from DWPPs supplied from Bahr Yousif Canal.

Annual gonadal dose equivalent (AGDE)

The annual gonadal dose equivalent (AGDE) due to the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K was calculated using the following formula (Mamont-Ciesla *et al.*, 1982):

$$AGDE = 3.09C_{Ra} + 4.18C_{Th} + 0.314C_K \quad (9)$$

Annual gonadal dose equivalent (AGDE), their standard error and average, standard deviation for deposits from DWPPs supplied from Nile River (NR), Ibrahimia Canal and Bahr Yousif Canal are presented in Tables 4-6, respectively.

From the calculated values, the ranges of AGDE are 0.13–0.69 mSv yr^{-1} with average of 0.29 mSv yr^{-1}, 0.19-1.13 mSv yr-1 with average of 0.42 mSv yr^{-1} and 0.19-0.38 mSv yr^{-1} with average of 0.27 mSv yr^{-1} of sediment samples from DWPPs supplied from Nile River, Ibrahimia Canal and Bahr Yousif Canal, respectively. The average calculated value of sediment samples from DWPPs supplied from Ibrahimia is the highest, as shown in Fig. 13.

Conclusion

The activity concentrations of deposits in DWPPs supplied from Nile River, Ibrahimia Canal and Bahr Yousif Canal are within the world values although some extreme values have been determined. Averages of all the calculated radiological indices in all the DWPPs are within the recommended level. Deposits in DWPPs supplied from Ibrahimia Canal have the highest averages of the calculated radiological indices, which may be due to the authorized release of man-made radionuclides into the system, such as cement, oil and sugar industries. However, in most regions the contribution proceeds from sewage. According to Radiation Protection 112 (EC, 1999) and UNSCEAR (UNSCEAR, 2000) report, some calculated radiation hazard indices of deposits in DWPPs supplied from Ibrahimia Canal are higher than the recommended level. Therefore, the sediments in DWPPs supplied from Ibrahimia Canal have posed significant radiological threat to the population when they are used as a building material.

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