#### **ORIGINAL PAPER**



# Radiation hazards and extremophiles bioaccumulation of radionuclides from hypersaline lakes and hot springs

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#### Abstract

Radioactivity measurements for water, sediment, microbial films and the bioaccumulation of radionuclides by extremophiles from hypersaline lakes and hot springs were carried out as scarcity studies in the Siwa Oasis, Egypt. Natural and man-made radionuclides were measured using high-resolution  $\gamma$ -spectrometry. Different radionuclides behaved differently in different environmental samples, while radionuclides were higher in microbial films compared to sediment, but all radionuclide levels except <sup>226</sup>Ra in water were generally low. Microbial films from hypersaline lakes had higher concentrations of <sup>40</sup>K, while microbial films from freshwater hot springs had the highest concentrations of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>137</sup>Cs. The calculated radiological hazard index parameters of radium equivalent activity (Ra<sub>ea</sub>), absorbed dose rate (D), annual effective dose (AED) and external hazard (Hex) in the sediment were within acceptable limits, but were higher in the microbial film samples. Otherwise, the potential cancer risk of the three freshwater springs was  $0.00244 \pm 0.000293$ ,  $0.00135.6 \pm 0.000172$  and  $0.00155.2 \pm 0.000198$ . In addition, the bioaccumulation factor for microbial films indicated that they are good accumulators of radionuclides, especially for <sup>226</sup>Ra and <sup>232</sup>Th, which may contribute to their effectiveness in removing radionuclides from ecosystems.

Keywords Groundwater · Microbial film · Siwa Oasis · Bioaccumulation factor · Radiological hazards

# Introduction

Radioecology is an interdisciplinary science concerned with understanding and quantifying the behaviour of radionuclides in the environment and the processes that determine their transport through ecosystems to many different types of receptors such as plants, animals and humans. A second aspect of this science is the assessment of radiological dose and impact on humans and their environment from the present, past or future. The behaviour of radionuclides in water is central to their migration because water is one of the

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main factors in the movement and distribution of elements on Earth. Therefore, aquatic ecosystems strongly influence and are influenced by the fate of radionuclides (Khater 1998). In particular, the natural decay series of radionuclides  $^{238}$ U,  $^{232}$ Th and  $^{40}$ K are responsible for the radioactivity in groundwater. <sup>226</sup>Ra is one of the most important elements in the <sup>238</sup>U series, indicating the abundance of uranium in the environment. The long half-life of <sup>226</sup>Ra, which decays to radon (<sup>222</sup>Rn), makes <sup>226</sup>Ra a Class A carcinogen and is the second most common cause of lung cancer (EPA 2006). The concentration of radionuclides in groundwater is discovered by the mineralogical composition of the aquifer and by the geochemical and hydrochemical conditions, which may change over time as groundwater is degraded (Schubert et al. 2011). Groundwater increases strongly in radioactivity as it passes through fractures in the bedrock (Canu et al. 2011). Environmental measurements of radioactivity levels in groundwater sources are necessary to protect residents from toxic and high exposure doses through ingestion (Yuce et al. 2009).

Siwa Oasis is a deep natural depression, approximately 23 m below sea level. Agriculture is the most popular activity



in the Siwa Oasis, which is dependent on groundwater from both dug wells and naturally flowing springs (Elkaffas et al. 2013). The lakes of Siwa are the watersheds of the discharged water from the cultivated areas, natural springs and flowing wells (Dahab 2004; El-Sayed et al. 2017). The non-renewable Nubian sandstone aquifer in the Siwa Oasis is the only water resource available for a variety of uses in the area (Ebraheem et al. 2003; Gossel et al. 2004). This aquifer is suffering from water depletion mainly due to excessive extraction from dug wells (Gad et al. 2018). The geochemical properties of the aquifer, especially the toxic metals and radionuclides, are critical in the management of aquifer use and health problems (Baba and Tayfur 2011; Murad et al. 2014; Yehia et al. 2017). Elevated levels of radioactive materials and daughter products may be found in the groundwater of areas rich in radionuclides. The presence of radionuclides with high radiotoxicity in water and the associated health risks requires special attention (Nriagu et al. 2012; Khodashenas et al. 2012). There is a strong need to develop a systematic programme for monitoring radioactive contaminant systems to protect humans and the environment due to the rapidly increasing dependence on groundwater as the sole water supply in such areas (Walencik et al. 2010).

The transfer of radionuclides to the environment must be considered in the initial stages of an environmental impact assessment. Assessment of radionuclide uptake by marine biota depends on the use of concentration factors (Brown et al. 2004). Microorganisms, such as microbial that have witnessed and survived various extreme conditions on Earth over time, could be used to remediate radioactive contamination (Gazso 2001). Microbes in radionuclide environments have mechanisms for their survival and radiation tolerance, focusing on the interactions between bacterial cells and radionuclides, which have immense potential for bioremediation (Shukla et al. 2017). Various algae, including macroalgae, are highly resistant to metals and U, which can be isolated at very high concentrations. As a result, macrophytes and macroalgae have been used as biomonitoring systems for radionuclide contamination in the marine environment (Strezov and Nonova 2005; Al-Masri et al. 2003; Zalewska and Saniewski 2011). In view of this, the study of radioactivity levels in algae is useful as a bioindicator of contamination of ecological communities and the bioremediation process (Garza 2005).

In the extremophilic groundwater, ecosystem of Siwa Oasis grows a special density of cyanobacteria and bacteria embedded in polysaccharide secretions (cyanobacteria–bacteria mat). This extremophilic cyanobacterial–bacterial mat grows in extreme habitats with high temperatures (> 50 °C) and hypersaline water (200%). These films include cyanobacteria, oxygenic photosynthetic bacteria, aerobic heterotrophs and anaerobes, sulphur-reducing bacteria, sulphuroxidising bacteria and methanogenic archaea, among other



biofunctional classes. Adaptations, competition or synergistic cooperation among these organisms result in unique physiological behaviour (El Karim and Goher 2016) and cause them to possess a large number of active sites and more than twenty active functional groups (e.g. hydroxyl, carboxyl, phenol, carboxyl, phosphorus and sulphydryl) that enhance metal adsorption (Al-Qahtani et al. 2021). These extremophilic microbial films efficiently remove Zn<sup>+2</sup>, Fe<sup>+2</sup> and Cd<sup>+2</sup> (Ali et al. 2019), Cu<sup>+2</sup>, Ni<sup>+2</sup> and Pb<sup>+2</sup> (Al-Qahtani et al. 2021) and hexavalent Cr and Mo (Abdelakrim et al., in press) from aqueous solution. The ability of microbial films to accumulate radionuclides has not been investigated in this area. It is, therefore, of particular interest to know the extent to which these microbial mats may be capable of radionuclide uptake and accumulation. Therefore, the aim of this study was to assess the radioactivity levels in the groundwater, sediment and microbial mats, to determine the bioaccumulation factor in order to follow-up the potential use of cyanobacterial-bacterial mats as bioindicators and bioacumulators of radionuclides and, furthermore, to estimate the radiation hazard parameters and cancer risk of this area.

## Materials and methods

### Study area

The Siwa Oasis is located in Egypt's Western Desert, 120km east of the Egyptian-Libyan border and 300-km south of the Mediterranean Sea (Fig. 1). The map was created using ArcGIS Pro 3.1® software from Esri. ArcGIS® and ArcMapTM are the intellectual property of Esri and are used here under licence and copyright © Esri. All rights reserved. For more information about Esri® software, visit www.esri.com. The oasis extends approximately 23 m below sea level in an east-west direction (Gindy and El-Askary 1969). The Siwa depression has an abundance of naturally flowing groundwater (Masoud and Koike 2006). There are two main aquifers; the shallowest (upper Siwa) and the deepest (Nubian sandstone). Springs are present in the shallow Middle Miocene carbonates, which are completely isolated from the Nubian sandstone (the deep aquifer) (Masoud and Koike 2006).

#### Sampling and sample preparation

The samples were collected from three freshwater springs and three hypersaline lakes in the Siwa Oasis in January 2018 (Fig. 1). At each site, more than three samples were collected. Water, cyanobacterial mat (microbial film) and sediment samples were collected from hypersaline lakes, and water and microbial biofilm only from springs. The water



Fig. 1 Map showing sampling sites by using ArcGIS Pro 3.1® software by Esri. Copyright © Esri

samples were collected and placed in a plastic bottle (2 L). Surface sediment and microbial film samples were collected by hand. They were packed separately in airtight polyethylene bags. All samples were stored in an icebox until returned to the laboratory. The sediment and microbial film samples were dried in a circulating air oven at 105 °C, crushed and then sieved to a mesh size of 63 mesh.

#### Gamma-ray spectrometry

The natural and anthropogenic radioactivity activity concentrations were analysed using gamma spectrometry. The gamma spectrometer is based on a high-purity germanium (HpGe) detector. The 3×3NaI(Tl) detector, with a resolution of 1.90 keV and a peak/Compton ratio of 69.9:1 at the 1.33-MeV gamma-ray line of <sup>60</sup>Co, has a relative efficiency of about 50%. It is coupled to conventional electronics connected to a multichannel analyser card (MCA) installed in a PC. The detector is shielded by a 10-cm thick lead liner and has a 2-mm copper foil lining inside. The MAESTRO software programme from ORTEC was used for the acquisition of the data and the analysis of the spectra. The spectrometer energy calibration was performed using point sources of <sup>241</sup>Am (59.6 keV), <sup>137</sup>Cs (661.6 keV) and <sup>60</sup>Co (1172 and 1332.3 keV). The calibration of the efficiency of the spectrometer was carried out using three well-known reference materials, namely RGU-1, RGTh-1 and RGK-1 (IAEA 1987; Anjoset al. 2005; El-Aassy et al. 2012). The Marinelli beakers were tightly sealed for at least 4 weeks. This was to ensure that a state of secular equilibrium between radium and its daughters was reached. The time accumulation of the spectra was at least 48 h, depending on the activity of the samples. The activity of <sup>226</sup>Ra (<sup>238</sup>U series) was measured using the 186.1-keV gamma-ray energy transition after subtracting the 185.7 keV of  $^{235}$ U, the 295.2 and 351.9 keV of <sup>214</sup>Pb and the 609.3 and 1120.3 keV of <sup>214</sup>Bi. The gamma energies of 338.4 keV and 911.2 keV for <sup>228</sup>Ac, 583 keV and 2614.4 keV for <sup>208</sup>Tl and 727.3 keV for <sup>212</sup>Bi were taken to represent the concentrations of the <sup>232</sup>Th series. <sup>40</sup>K was measured directly from the 1460.8-keV gamma transition (Chieco et al. 1990). In addition, <sup>137</sup>Cs was measured directly from 661.7 keV. The activity concentration of the radionuclides was calculated according to the formula of EL Afifi et al. (2006). Quality control (QC) is carried out to ensure that the analysis is accurate and to assess the validity and reliability of the data. The precision and accuracy of the analysis were estimated by measuring the radionuclides in the certified reference materials (e.g. IAEA-443, IAEA-446, IAEA-410 and IAEA-312). The measured activity concentrations, with mean deviations and errors not exceeding 3%, were very close to the reported values of the certified materials. The precision of the gamma spectrometer system is obtained by the lowest limits of detection (LLDs), which were determined according to USDOE (1992) and Abid Imtia et al. (2005). The lowest detection limits (LLDs) are 9.35, 0.45, 1.34 and 0.2 Bq kg<sup>-1</sup> for <sup>40</sup>K, <sup>226</sup>Ra, <sup>232</sup>Th and <sup>137</sup>Cs, respectively, for the sediment. Otherwise, the LLD values for water are 0.84, 0.1, 0.1 and 0.02 Bg  $l^{-1}$  for  ${}^{40}$ K, <sup>226</sup>Ra, <sup>232</sup>Th and <sup>137</sup>Cs, respectively.

# Radiation hazard indices and potential health of the radionuclides

It is logical to use various known health risk indices to make a confident statement about human health and the environment. The radiation hazard indices are the absorbed dose rate in air (*D*), the annual effective dose rate (AED), the radium equivalent activity ( $Ra_{eq}$ ), the external hazard index ( $H_{ex}$ ) and the representative level index ( $I\gamma$ ), calculated according to the equations given in Table 1.

The health effects sub-committee recommends that the risks associated with all radium isotopes should be combined to reflect the total risk. The cancer risk associated with ingesting radium isotopes was estimated via EPA 1999; Abdellah and Diab 2012.



Table 1	Radiation	hazard
indices	equations	

Hazard indices	Equation	References
Absorbed dose rate in air (nGyh <sup>-1</sup> ) Annual effective dose rates (mSv y <sup>-1</sup> )	$\begin{split} D &= 0.462 \ C_{Ra} + 0.621 \ C_{Th} + 0.0417 \ C_{K} \\ AED &= D \ (nGy \ h^{-1}) \times 8760 \ (h \ yr^{-1}) \times 0.7 \\ (Sv \ Gy^{-1}) \times 0.2 \times 10^{-6} \end{split}$	UNSCEAR (2000) UNSCEAR (2000)
Radium equivalent activity (Bq kg <sup>-1</sup> ) External hazard index (H <sub>ex</sub> ) Representative level index (Bq kg <sup>-1</sup> )	$\begin{aligned} & \operatorname{Ra}_{eq} = \operatorname{C}_{Ra} + 1.43 \operatorname{C}_{Th} + 0.077 \operatorname{C}_{K} \\ & \operatorname{H}_{ex} = (\operatorname{C}_{Ra}/370 + \operatorname{C}_{Th}/259 + \operatorname{C}_{K}/4810 \le 1 \\ & \operatorname{I}\gamma = \frac{C_{Ra}}{150} + \frac{C_{Th}}{100} + \frac{C_{K}}{1500} \end{aligned}$	Kurnaz et al. (2007) UNSCEAR (2000) (Chad-Umoren and Umoh (2017)

 $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the activity concentrations in Bq kg<sup>-1</sup> of <sup>226</sup>Ra,<sup>232</sup>Th and <sup>40</sup>K, respectively. The conversion coefficient from absorbed dose to effective dose is 0.7 Sv Gy<sup>-1</sup>, and the outdoor occupancy factor is 0.2

Table 2 Conversion factors of  $^{226}$ Ra,  $^{232}$ Th and  $^{40}$ K for different age groups (IAEA 1996; WHO 2003)

Age groups	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K
Adults	$2.8 \times 10^{-7}$	$2.3 \times 10^{-7}$	$6.2 \times 10^{-9}$
Children	$8 \times 10^{-7}$	$2.9 \times 10^{-7}$	$1.3 \times 10^{-8}$
Infants	$9.6 \times 10^{-7}$	$4.5 \times 10^{-7}$	$4.2 \times 10^{-8}$

$$Cancer Risk (CR) = MCL \times RC \times TWI$$
(1)

where CR is lifetime cancer risk corresponding to MCL (unit less), MCL is the maximum contaminant level (Bq l<sup>-1</sup>), RC is the mortality risk coefficient for <sup>226</sup>Ra ( $7.17 \times 10^{-9}$  Bq l<sup>-1</sup>) and <sup>228</sup>Ra ( $2.0 \times 10^{-8}$  Bq l<sup>-1</sup>) and TWI is total water intake (2 L d<sup>-1</sup> × 365.4 d year<sup>-1</sup> × 70 years).

The effective radiation doses (DRW) resulting from the ingestion of these waters were assessed according to the USEPA (1999) in order to calculate potential health hazards.

$$DRW = A_w \times IR_W \times IDF$$
(2)

where  $A_w$  is the activity concentration of radionuclides (Bq l<sup>-1</sup>), IR<sub>w</sub> is the water intake for a human being for 1 year and IDF is the effective dose in mSv Bq<sup>-1</sup>. For infants, children and adults, doses were calculated using a consumption rate of 150, 350 and 730 L yr<sup>-1</sup>, respectively. The conversion factors for infants, children and adults are given in Table 2.

# Water-to-biota transfer factor (the bioaccumulation factor, BF)

The transfer factor (bioaccumulation factor, BF) was calculated from the concentrations of the radioactive or natural isotope in both water and biota (microbial films) samples to determine the rate of transfer of natural radioisotopes from water to biota in the study area. The bioaccumulation factor is expressed as the concentration of biotic tissue relative to the concentration of water at equilibrium (UNSCEAR 1977).



$$BF = \frac{C_{biota}}{C_{water}}$$
(3)

where  $C_{\text{biota}}$  is radionuclide activity concentration of biota per kg fresh weight, and  $C_{\text{water}}$  is radionuclide activity concentration of water per L.

# **Results and discussion**

#### **Environmental conditions**

The mean temperature values of Siwa lakes ranged between 19.1 and 19.5 °C, and the mean TDS values varied from 178.47 to 198.61 g/l. According to Hassan and Ismail (2018), the limestone, dolomite and evaporite deposits of the Siwa Oasis have fractured and mixed with the trapped ancient seawater, resulting in the high salinity levels. In addition, there are extensive agricultural activities with high evaporation rates and high temperatures that increase TDS levels. Compared to Siwa lakes, spring's water samples are characterised by high water temperature (31.2 °C in spring (1) to 53.2 °C in spring (3)) and very low TDS (0.8 g/l in spring (2) to 1.4 g/l in spring (3)). The studied water is rather alkaline side for lakes and springs. All water samples fall within the recommended range of 6.5–8.5 for irrigation, according to the USDA (2011).

#### **Radioactivity measurement**

#### Water

The activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs in water samples are from hypersaline lakes and freshwater springs. The activity concentrations of <sup>226</sup>Ra and <sup>232</sup>Th in water samples did not differ significantly between hypersaline lakes and freshwater springs (Table 3). The <sup>226</sup>Ra activity concentration of hypersaline lakes ranged from  $0.5 \pm 0.05$  Bq l<sup>-1</sup> in Lake Siwa to  $4.10 \pm 0.49$  Bq l<sup>-1</sup> in Lake Zeitoun. For the springs, it ranged from  $2.01 \pm 0.19$  Bq l<sup>-1</sup>

Table 3 The activity concentration of radionuclides in water samples (Bq/l), sediment and microbial film samples (Bq/kg)

Samples	Туре	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K	<sup>137</sup> Cs
Zeitoun Lake (1)	Water	$4.10 \pm 0.49$	$0.7 \pm 0.08$	$27.00 \pm 0.9$	$0.40 \pm 0.07$
Zeitoun Lake (2)		$3.50 \pm 0.41$	$0.5 \pm 0.04$	$19.00 \pm 0.5$	$0.70 \pm 0.05$
Zeitoun Lake (3)		$3.90 \pm 0.42$	$0.75 \pm 0.07$	$18.00 \pm 0.5$	$0.60 \pm 0.06$
Aghormi Lake (1)		$3.09 \pm 0.40$	$0.7 \pm 0.09$	$71.00 \pm 1.9$	$0.28 \pm 0.1$
Aghormi Lake (2)		$2.70 \pm 0.30$	$0.90 \pm 0.08$	$45.00 \pm 0.9$	$0.31 \pm 0.09$
Aghormi Lake (3)		$1.50 \pm 0.10$	$0.5 \pm 0.08$	$27.00 \pm 1.0$	$0.29 \pm 0.07$
Siwa Lake (1)		$0.50 \pm 0.05$	$0.90 \pm 0.08$	$11.50 \pm 0.9$	$0.30 \pm 0.01$
Siwa Lake (2)		$0.70 \pm 0.06$	$1.20 \pm 0.09$	$14.50 \pm 1.0$	$0.50 \pm 0.02$
Siwa Lake (3)		$0.60 \pm 0.06$	$1.10 \pm 0.09$	$13.50 \pm 0.9$	$0.40 \pm 0.02$
Siwa Lake (4)		$0.70 \pm 0.05$	$0.80 \pm 0.07$	$7.50 \pm 0.4$	$0.35 \pm 0.02$
Mean		$2.13 \pm 0.19$	$0.81 \pm 0.08$	$25.4 \pm 1.08$	$0.41 \pm 0.05$
Spring (1)	Groundwater	$3.59 \pm 0.40$	$1.10 \pm 0.14$	$6.40 \pm 0.4$	$0.40 \pm 0.13$
Spring (2)		$2.01 \pm 0.19$	$0.60 \pm 0.10$	$2.10 \pm 0.13$	$0.23 \pm 0.08$
Spring (3)		$3.12 \pm 0.40$	$0.40 \pm 0.05$	$2.80 \pm 0.2$	$0.24 \pm 0.07$
Mean		$2.91 \pm 0.33$	$0.70 \pm 0.09$	$3.77 \pm 0.24$	$0.29 \pm 0.09$
Worldwide (WHO 2003; IAEA 1996)		1	1	4	10
Zeitoun Lake (1)	Sediment	$22.00 \pm 2.80$	$8.30 \pm 0.60$	$235.40 \pm 3.20$	$1.90 \pm 0.10$
Zeitoun Lake (2)		$41.00 \pm 1.20$	$9.30 \pm 0.90$	$156.00 \pm 2.00$	$2.10 \pm 0.10$
Zeitoun Lake (3)		$19.50 \pm 2.10$	$7.45 \pm 0.50$	$139.00 \pm 1.90$	$2.30 \pm 0.09$
Aghormi Lake (1)		$23.30 \pm 2.30$	$9.70 \pm 0.70$	$245.80 \pm 3.40$	$1.10 \pm 0.10$
Aghormi Lake (2)		$24.50 \pm 2.50$	$7.90 \pm 1.70$	$235.00 \pm 3.10$	$1.40 \pm 0.10$
Aghormi Lake (3)		$28.50 \pm 1.70$	$13.50 \pm 1.80$	$183.50 \pm 2.90$	$1.50 \pm 0.10$
Siwa Lake (1)		$16.90 \pm 1.70$	$19.50 \pm 1.00$	$110.00 \pm 1.50$	$1.90 \pm 0.10$
Siwa Lake (2)		$27.96 \pm 1.40$	$13.50 \pm 1.60$	$168.00 \pm 2.10$	$2.30 \pm 0.10$
Siwa Lake (3)		$19.80 \pm 1.90$	$11.50 \pm 1.10$	$230.40 \pm 2.80$	$1.70 \pm 0.10$
Siwa Lake (4)		$21.50 \pm 2.50$	$14.50 \pm 1.00$	$310.20 \pm 4.10$	$2.80 \pm 0.20$
Mean		$24.50 \pm 2.01$	$11.52 \pm 1.09$	$201.33 \pm 2.7$	$1.9 \pm 0.11$
Worldwide (UNSCEAR 2000)soil		33	35	370	_
Zeitoun Lake	Microbial film	$13.6 \pm 0.9$	$6.2 \pm 0.4$	$296.1 \pm 2.9$	$1.4 \pm 0.1$
Aghormi Lake		$14.4 \pm 1.1$	$7.8 \pm 0.7$	$302.4 \pm 4.5$	$1.1 \pm 0.1$
Siwa Lake		$4.2 \pm 0.5$	$9.2 \pm 0.8$	$350 \pm 4.7$	$1.2 \pm 0.1$
Mean		$10.73 \pm 0.83$	$7.73 \pm 0.63$	$316.17 \pm 4.03$	$1.23 \pm 0.1$
Spring 1	Microbial film	$201.1 \pm 7.2$	$21.9 \pm 1.6$	$56.5 \pm 3.8$	$2 \pm 0.3$
Spring 2		$256.2 \pm 5.3$	$212.5 \pm 3.3$	$606.4 \pm 4.7$	$2.3 \pm 0.3$
Spring 3		$190.5 \pm 3.4$	$156.1 \pm 1.5$	$252.7 \pm 2.2$	$2.2 \pm 0.1$
Mean		$215.93 \pm 5.3$	$130.17 \pm 2.13$	$305.20 \pm 3.57$	$2.17 \pm 0.23$

in spring (2) to  $3.59 \pm 0.40$  Bq l<sup>-1</sup> in spring (1). The high <sup>226</sup>Ra concentrations in water samples indicate that the area of the aquifer matrix from which the water originates is enriched in <sup>238</sup>U activity concentrations; <sup>226</sup>Ra activities in groundwater are mainly controlled by the primary <sup>238</sup>U content of the aquifer system (Lauria et al. 2004). Whereas; radium in groundwater can be derived from a number of sources, including (1) the decay of uranium or thorium parent nuclei dissolved in solution, (2) decay from formation minerals, (3) alpha-recoil release of radium from the decay of parent nuclei in the aquifer, (4) radium adsorbed on the surface layer, clays and oxides and (5) co-precipitation with

the secondary minerals (Porcelli and Swarzenski 2003). In addition, the high <sup>226</sup>Ra activity in hypersaline lakes could be the result of water runoff from local agriculture containing phosphate fertilisers. Hussein and Ahmed (1997) report an increase in <sup>226</sup>Ra concentrations in the lakes as a result of the use of phosphogypsum and superphosphate fertilisers in agriculture. Although the mean activity concentration of <sup>226</sup>Ra exceeded the internationally permissible value (1 Bq l<sup>-1</sup>) in the springs and hypersaline lakes, except for Lake Siwa, it was much lower than the value found by Allam (2019) and lower than the mean value found by Abd El-Mageed et al. (2013) in the hot springs and groundwater in



Yemen. The elevated radium activity concentration observed in this study is consistent with previously published radium data for groundwater from the Nubian aquifer in the eastern desert of Egypt (Sherif & Sturchio 2018), the Nubian aquifer in the Western Desert of Egypt (Sherif & Sturchio 2021), the Sinai Peninsula in Egypt (Sherif et al. 2018) and the Disi aquifer in Jordan (Vengosh et al. 2009). The distribution and behaviour of the Ra isotopes in the groundwater is determined by the coupled effects on various geochemical processes (Sherif & Sturchio 2021). Compared to other sandstone aquifers worldwide, radium activity in ancient groundwaters in the Middle East and northeastern Africa is generally much higher than in other sandstone aquifers (Sherif & Sturchio 2021).

The levels of <sup>232</sup>Th concentration are low in most samples, with the exception of spring (1) and Lake Siwa. The activity concentrations of <sup>232</sup>Th ranged from  $0.40 \pm 0.05$  Bq l<sup>-1</sup> in spring (3) to  $1.20 \pm 0.09$  Bql<sup>-1</sup> in Lake Siwa. The mean activity concentrations of <sup>232</sup>Th in the hypersaline lakes and springs were lower than the international allowable value (1 Bq l<sup>-1</sup>). These values were lower than the upper limits found by Allam (2019) in the Siwa region and comparable to those found by Mathuthu et al. (2021) in Namibian groundwater. Furthermore, the <sup>232</sup>Th concentrations in freshwater wells in this study were much lower than the levels found by Yehia et al. (2017) in thirty wells and springs in the Western Desert, Egypt. <sup>232</sup>Th is virtually immobile in groundwater, has not migrated elsewhere and is strongly adsorbed to iron hydroxides (Murphy et al. 1999). The lower values of the <sup>232</sup>Th/<sup>226</sup>Ra ratio (0.13–0.33) in all samples, except those in Lake Siwa (1.14–1.83), indicate that radium in the Nubian sandstone aquifer is derived exclusively from interactions with rocks in the U decay series. In the same context, the activity concentrations of <sup>40</sup>K are higher than the worldwide value in the hypersaline lakes and spring (1), as shown in Fig. 2. The activity concentrations of <sup>40</sup>K ranged from  $2.10 \pm 0.13$  Bql<sup>-1</sup> at the spring (2) to  $71.1 \pm 1.90$  Bql<sup>-1</sup> at Lake Aghormi. The higher <sup>40</sup>K activity observed in water samples from hypersaline lakes (Zeitoun, Aghormi and Siwa) was generally explained by the use of potassium fertiliser in agricultural activities. The <sup>40</sup>K activity concentration in the springs ranged from  $2.10\pm0.13$  to  $6.40\pm0.4$  Bql<sup>-1</sup>. which was significantly lower than the values reported by Adetunji et al (2018) in Nigeria and Allam (2019) in Siwa area for underground water.

The activity concentrations of  $^{137}$ Cs ranged from  $0.23 \pm 0.08$  Bq l<sup>-1</sup> at spring (2) to  $0.70 \pm 0.05$  Bq l<sup>-1</sup> in Zeitoun Lake. The activity concentrations of  $^{137}$ Cs are lower than the worldwide range (WHO 2003; IAEA 1996) but were slightly higher than the values conducted by Zgorelec et al. (2021) in the acidic soil of northwestern Croatia. The fallout radionuclide  $^{137}$ Cs was detected in all the water samples (hypersaline and springs), which could be due to  $^{137}$ Cs falling from the atmosphere to the earth's surface, where it



Fig. 2 The distribution of <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs in water samples of Siwa Oasis

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could enter the groundwater circulation system. The advection and dispersion model was used to try to understand the mechanism of migration of Cs-137 into groundwater. It was assumed that the pore water in the uppermost 0.25 m of the soil was equilibrated with the Cs-137 fallout at the ground surface (GEO-SLOPE 1950), and the dissolved <sup>137</sup>Cs migrated in the soil and reached aquifers at different depths (Shizuma et al. 2018). Furthermore, the vertical migration of <sup>137</sup>Cs in aquifers is relatively weak and depends on the contaminant levels (fallout) and their forms (particles or dust) (Ashraf et al. 2014). As a result, there are small amounts of contamination distributed through the groundwater circulation system.

#### Sediment

The mean value of activity concentration of <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs in sediment samples was found to be  $24.5 \pm 2.01$ ,  $11.52 \pm 1.09$ ,  $201.33 \pm 2.7$  and  $1.9 \pm 0.11$  Bq kg<sup>-1</sup> (Table 3), respectively. Figure 3 shows the spatial distribution of activity concentrations for <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs in sediment sample of hypersaline lakes that indicate levels below the permissible level of worldwide (UNSCEAR 2000). The results show that the order  ${}^{137}Cs < {}^{232}Th < {}^{226}Ra < {}^{40}K$  is the mean activity concentration for all sampling sites. The <sup>232</sup>Th concentration is significantly lower than that of  $^{226}$ Ra and  $^{40}$ K. The elevated activity concentration of <sup>226</sup>Ra in Lake Zeitoun could be due to the discharge of agricultural drainage water containing chemical fertilisers. It is assumed that the original materials making up the various geological formations of the area under study are the cause of the variance and frequency of radionuclide concentrations. These results suggest that atmospheric deposition in this area could be an explanation for the discovery of the <sup>137</sup>Cs radionuclide in hypersaline lakes (Zeitoun, Aghormi and Siwa). The <sup>137</sup>Cs radionuclide forms associations with mud minerals in the upper soil layer (Bourcier et al. 2011), and clay minerals can absorb caesium (Fan et al. 2014). In addition, most caesium constituents have a chemical affinity comparable to that of potassium (K) and are soluble in water (UNSCEAR 1982). Timofeeva-Resovskaya (1963) classified <sup>137</sup>Cs as a pedotope, which means that it belongs to the group of elements deposited by bottom sediments in 95-100% of cases. Darwish et al. (2013) found that the mean activity concentration of <sup>226</sup>Ra and <sup>232</sup>Th of sediment in the saline Oarun Lake was consistent with this study and lower than the recommended value of soil (UNSCEAR 2000). Otherwise, Lui et al. (2021) reported high activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in the bottom



Fig. 3 The distribution of <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs in sediment samples of Siwa Oasis



sediments of the Nansha Sea and South China Sea that were 14.6-38.51, 15.81-49.21 and 149.7-569.35 Bq kg<sup>-1</sup>, respectively.

#### **Microbial films**

The mean activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and  ${}^{137}$ Cs were  $10.73 \pm 0.83$ ,  $7.73 \pm 0.63$ ,  $316.17 \pm 4.03$ and  $1.23 \pm 0.1$  Bq kg<sup>-1</sup>, respectively, in microbial films from hypersaline lakes (Table 3 and Fig. 4). In contrast, they were  $215.93 \pm 5.3$ ,  $130.17 \pm 2.13$ ,  $305.20 \pm 3.57$  and  $2.17 \pm 0.23$  Bq kg<sup>-1</sup> in microbial films from hot springs, respectively. In the samples from the springs, the microbial films were 100 times more radioactive than the surrounding groundwater. The cyanobacterial community of the Siwa hot springs could serve as a biosorbent for radionuclides. These results indicate that <sup>226</sup>Ra and <sup>232</sup>Th are present at high concentrations in the microbial film samples from springs. This may be due to the fact that radium, a substance that readily penetrates plants due to its barrierless nature (Titaeva 2000), is also a unique biologically essential element (Vanov 1994). Furthermore, this suggests that <sup>226</sup>Ra can adsorb to the capsule surfaces of cyanobacteria, cocci and bacilli in microbial mats, leading to the formation of extracellular polymer capsules and slime layers around the cells that protect against radionuclides and other metals in hot springs. It is conceivable that the ability of a particular bacterium to immobilise heavy metals could be used to mitigate the terrible effects of radioactive pollution (Tazaki 2009). Based on their similar chemical properties, it could be predicted that the two radionuclides <sup>226</sup>Ra and <sup>232</sup>Th behave similarly during the processes of precipitation and mineral formation (Lazareva et al. 2011). Furthermore, <sup>40</sup>K concentrations varied within a narrow range in microbial film samples from hypersaline lakes, whereas they were widely separated in spring samples with the highest activity concentration in the spring (2). <sup>137</sup>Cs concentrations were higher in microbial films of freshwater springs than from hypersaline lakes. In the same context, Lee et al. (2019) found that the uptake efficiency of <sup>137</sup>Cs by Haematococcus pluvialis and Chlorella vulgaris was 95% and 90%, respectively. The movement and accumulation of Cs-137 in the air depends on the radionuclide particle size, climatic conditions and earth science. Bioaccumulation of radiocaesium in higher organisms is significantly influenced by several factors (e.g. pH, K<sup>+</sup> and organic matter) (Avery 1995). Caesium is known to be transported into cells as a potassium analogue; consequently, the presence of potassium in the media interferes with <sup>137</sup>Cs



Fig. 4 The distribution of <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs in microbial films samples of Siwa Oasis

absorption (Whicker and Schultz 1982; Bystrzejewska-Piotrowska and Urban 2003). It has been shown to be taken up by plant and algal cells via potassium transport channels due to chemical similarities (Lee et al. 2019).

Lee et al. 2019 also showed that each microalgal species can selectively remove different elements; which explains why the extremophilic microbial film of Siwa Oasis, composed of different groups and many species, can remove different radionuclides from the surrounding media, which is more difficult than the remediation of a single form of contaminant, as reported by Thakare et al. (2021). Microbial films, especially those from freshwater sources, appear to be good accumulators of various radionuclides, which are several times higher in freshwater microbial films than in the surrounding media. In the global search by Shin-va et al. (2014), the highly active strains for radionuclide removal were all freshwater strains. However, none of the marine strains could show high radionuclide removal efficiency because competing elements in seawater such as K and Ca would reduce the ability of cells to absorb or accumulate radionuclides (Shin-ya et al. 2014). The high content of metals in seawater results in lower accumulation of radionuclides by marine biota (Chebotina and Kulikov 1998) due to competition of salts with radionuclides for adsorption sites (Strand 1998).

The results showed that the distribution of <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs in water, sediment and microbial films was not consistent. The radioactivity varied significantly depending on the different sources and the geography of the region. The findings suggest that water–rock interaction in sandstone aquifers can potentially mobilise radionuclides from the U and Th series, which may explain the higher levels of <sup>226</sup>Ra and <sup>232</sup>Th in the springs water and microbial film samples. Furthermore, freshwater microbial films indicate that they are good accumulators of different radionuclides. The distribution of radionuclides <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs in hypersaline lakes also showed the influence of agriculture in this area. Using a global comparison, the activity concentration of radionuclides in groundwater, microbial films and sediment of Siwa Oasis is compared with the studies across different countries (Table 4). The mean activity concentration of <sup>226</sup>Ra and <sup>232</sup>Th of groundwater in this study agreed with the Marsa Alam-Shalateen area, Egypt (Arafat et al. 2017), and Siwa Oasis, Egypt (Saleh et al. 2016), and lower than Anarak-Khour a desertic area, Iran (Ehsanpour et al. 2014), and Al-Hurrah city in Najaf, Iraq (Alaboodi et al. 2020), and the activity concentration of <sup>226</sup>Ra series was higher than the values obtained in Phra Nakhon Si Ayutthaya Province, Thailand (Kodcharin et al. 2018) and Jordan (Alomari et al. 2019). Also, the activity concentration of <sup>40</sup>K was lower than Al-Hurrah city in Najaf, Iraq (Alaboodi et al. 2020), and higher than in Phra Nakhon Si Ayutthaya Province, Thailand (Kodcharin et al. 2018), Anarak-Khour a desertic area, Iran (Ehsanpour et al. 2014), Marsa Alam-Shalateen area, Egypt (Arafat et al. 2017), and Siwa Oasis, Egypt (Saleh et al. 2016). The activities of <sup>226</sup>Ra and <sup>232</sup>Th of microbial films in Siwa were higher than Gulf of Suez, Egypt (Diab et al. 2019), plants in Siwa Oasis, Egypt (El Begawy et al. 2019), and moss in the Marmara region, Turkey (Belivermis and Cotuk 2010). Furthermore, the activity concentration of <sup>40</sup>K was lower than Gulf of Suez, Egypt (Diab et al. 2019), plant in Siwa Oasis, Egypt (El Begawy et al. 2019), and Iskenderun Bay, Turkey (Varinlioğlu et al. 1997), but higher than moss in Marmara region, Turkey

**Table 4** Comparison of the activity concentrations of  ${}^{226}$ Ra series,  ${}^{232}$ Th series and  ${}^{40}$ K of groundwater (Bq/l), plant and sediment samples (Bq/ kg) from different countries

Locations	<sup>226</sup> Ra series	<sup>232</sup> Th series	<sup>40</sup> K	Note	References
Siwa Oasis	2.31±0.26	$0.79 \pm 0.08$	$20.41 \pm 0.74$	Groundwater	Present study
	$113.33 \pm 3.07$	$68.95 \pm 1.38$	$310.68 \pm 3.8$	Biofilm	
	$24.5 \pm 2.01$	$11.52 \pm 1.09$	$201.33 \pm 2.7$	Sediment	
Phra Nakhon Si Ayutthaya Province, Thailand	$0.77 \pm 0.13$	$1.03 \pm 0.19$	$15.56 \pm 1.28$	Groundwater	Kodcharin et al. (2018)
Anarak-Khour a desertic area, Iran	$\leq 0.0005 - 9.7$	$\leq 0.0002 - 28.22$	MDA—10.3	Groundwater	Ehsanpour et al. (2014)
Al-Hurrah city in Najaf, Iraq	$7.15 \pm 1.88$	$2.19 \pm 0.44$	$40.89 \pm 8.93$	Groundwater	Alaboodi et al. (2020)
Marsa Alam-Shalateen area, Red Sea coast, Egypt	< 0.7–7.6	< 0.6	<3-32.84	Groundwater	Arafat et al. (2017)
Jordan	$0.293 \pm 0.005$	$0.508 \pm 0.009$	-	Groundwater	Alomari et al. (2019)
Siwa Oasis, Egypt	0.08-7.47	0.472-4.1	0.025-5.33	Groundwater	Saleh et al. (2016)
Gulf of Suez, Egypt	$9.5 \pm 0.7$	$7.4 \pm 0.7$	$609.3 \pm 10.9$	Algae	Diab et al. (2019)
Siwa Oasis, Egypt	35	27	338-2102	Plant	El Begawy et al. (2019)
	< 0.7-104	< 0.6-67	82-1969	Soil	
Iskenderun Bay, Turkey	_	5.98-36.02	550-1690	Algae	Varinlioğlu et al. (1997)
Marmara region, Turkey	0.87-6.70	1.51-6.17	17.1–181.1	Moss	Belivermis and Çotuk, (2010)
Lake Nasser	$26 \pm 1.6$	19±1.5	$255.6 \pm 7.9$	Sediment	Imam et al. (2020)



(Belivermis and Çotuk 2010). The activity concentrations of  $^{226}$ Ra,  $^{232}$ Th and  $^{40}$ K in the sediment were lower than in Lake Nasser (Imam et al. 2020).

# Environmental hazard indices and risk assessment

The radiological hazards indices are radium equivalent activity (Raea), absorbed dose rates (D), annual effective dose (AED), external hazard  $(H_{av})$  and the representative gamma index  $(I\gamma)$  in microbial films, and sediment was determined and listed in Table 5. The mean radium equivalent activity (Ra<sub>ea</sub>), absorbed dose rates (D), annual effective dose (AEDs), external hazard ( $H_{ex}$ ) and the representative gamma index ( $I\gamma$ ) are 54.84 ± 3.76 Bq kg<sup>-1</sup>, 26.68 ± 1.7 nGyh<sup>-1</sup>,  $0.03 \pm 0.002 \text{ mSvy}^{-1}$ ,  $0.15 \pm 0.01 \text{ and } 0.41 \pm 0.03$ , respectively. These radiation indices of sediment are less than the reference maximum levels provided by the UNSCEAR (2000). This indicates that although the hypersaline lakes' sediment being contaminated with chemical fertiliser of the agriculture activity in this area, the results of the measurements are still within acceptable ranges. Similarly, the findings of environmental hazard of the microbial films showed that the  $Ra_{eq}$  values varied from 41.19 to 602.6 Bq kg<sup>-1</sup> with mean value of  $233 \pm 5.3$  Bqkg<sup>-1</sup>, and the mean absorbed dose rate (D) and annual effective dose (AEDs) outdoors were  $107.45 \pm 2.39$  nGyh<sup>-1</sup> and  $0.132 \pm 0.003$  mSvy<sup>-1</sup>, respectively, with the highest value in spring (2). The calculated value of the external hazard indices of the microbial films due to natural radioactivity <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K was lower than unity at all sites except at springs (2) and (3). In the same context, the representative level index (*I* $\gamma$ ) ranged from 0.35 to 4.24, with a mean value of 1.65 that higher than the recommended value (<1). Radium equivalent activities, annual outdoor effective doses (AEDs) and external hazard index H<sub>ex</sub> in microbial films were lower than the UNSCEAR (2000) permissible levels.

Otherwise, the absorbed dose rate (D) and the representative level index  $(I\gamma)$  in microbial films were higher than the recommended values of the UNSCEAR (2000) (18-93)  $nGyh^{-1}$  for (D) and less than unity for (I $\gamma$ ), respectively. This could be due to the fact that the calculated absorbed dose of the microbial films is used as an equivalent for the dose absorbed by living tissue. According to Lloyd and Lovley (2001), Shukla et al. (2017), Lopez-Fernandez et al. (2020) and Pande et al. (2022), the mechanisms of microbial tolerance to radiotoxicity from radionuclides are primarily realised through various forms of interaction between microbes and radionuclides, such as, e.g. biomineralisation, biosorption, bioaccumulation and biotransformation. In the technique of biosorption, radioactive cations are taken up by bacteria through the cell scaffold and then adsorbed onto binding sites such as amine, carboxyl, hydroxyl and other groups in the cell walls. Adsorption has been found to help many types of organisms reduce the harmful effects of a variety of radionuclides (Shukla et al 2017; Lopez-Fernandez et al 2020).

 Table 5
 The radiological hazard parameters in microbial film, sediment and water samples

Sample	Туре	Ra <sub>eq</sub> (Bqkg <sup>-1</sup> )	$D (nGyh^{-1})$	AEDs (mSv y <sup>-1</sup> )	H <sub>ex</sub>	$I_{\gamma r}$
Zeitoun Lake	Microbial films	$43.2 \pm 1.7$	$22.7 \pm 0.8$	$0.030 \pm 0.001$	$0.12 \pm 0.005$	$0.35 \pm 0.01$
Aghormi Lake		$46.6 \pm 2.3$	$24.3 \pm 1.1$	$0.030 \pm 0.001$	$0.13 \pm 0.006$	$0.38 \pm 0.02$
Siwa Lake		$41.19 \pm 1.97$	$22.97 \pm 0.95$	$0.028 \pm 0.001$	$0.12\pm0.005$	$0.35 \pm 0.01$
Spring 1		$236.4 \pm 9.7$	$102.9 \pm 4.3$	$0.126 \pm 0.005$	$0.64 \pm 0.026$	$1.60 \pm 0.07$
Spring 2		$602.6 \pm 10.4$	$276.2 \pm 4.7$	$0.339 \pm 0.006$	$1.64 \pm 0.028$	$4.24 \pm 0.07$
Spring 3		$431.4 \pm 5.7$	$195.6 \pm 2.5$	$0.240 \pm 0.003$	$1.17 \pm 0.015$	$3.00 \pm 0.04$
Zeitoun Lake	Sediment	$51.82 \pm 3.15$	$24.94 \pm 1.41$	$0.031 \pm 0.002$	$0.14 \pm 0.009$	$0.38 \pm 0.02$
Aghormi Lake		$55.76 \pm 4.39$	$27.28 \pm 1.99$	$0.033 \pm 0.002$	$0.15 \pm 0.01$	$0.42 \pm 0.03$
Siwa Lake		$56.96 \pm 3.74$	$27.83 \pm 1.70$	$0.034 \pm 0.002$	$0.16 \pm 0.01$	$0.43 \pm 0.03$
Worldwide (UNSCI	EAR 2000) soil	<370	57	0.5	1	1

The annual effective doses and cancer risk associated with water consumption

Samples		Total dose (mS	Cancer risk (CR) $\times 10^{-5}$		
		Infants	Children	Adults	
Spring 1	Groundwater	$0.63 \pm 0.07$	$1.14 \pm 0.12$	$0.94 \pm 0.1$	$244 \pm 29.3$
Spring 2		$0.34 \pm 0.04$	$0.63 \pm 0.06$	$0.52 \pm 0.06$	$135.6 \pm 17.2$
Spring 3		$0.49 \pm 0.06$	$0.92 \pm 0.12$	$0.72 \pm 0.09$	$155.2 \pm 19.8$
Mean		$0.49 \pm 0.05$	$0.91 \pm 0.12$	$0.73 \pm 0.08$	$178.1 \pm 22.2$
Worldwide (WHO 2003; IAEA 1996) <sup>a</sup> (USEPA 2000) <sup>b</sup>		0.26 <sup>a</sup>	$0.2^{a}$	0.1 <sup>a</sup>	10 <sup>b</sup>



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Under the above circumstances, the effective dose was estimated for different age groups, infants, children and adults, considering the intake of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in Siwa groundwater (see Table 5 and Fig. 5). The mean annual effective doses (DRW) of radiation during water consumption for all wells/springs in this study area for infants, children and adults were 0.49, 0.91 and 0.73 mSv/year, respectively. The annual effective dose (DRW) for infants, children and adults was below the minimum risk level (MRL) of 1 mSv/year as reported by ICRP 2000. But also, it was about two, five and seven times higher, for infants, children and adults, respectively, than reported in the WHO 2003. The high values of effective dose in the springs samples could be due to the high radium concentration. These results of the effective dose are consistent with those obtained by Saleh et al. (2016) in groundwater of the Siwa Oasis, lower than those obtained by El-Mageed et al. (2013) from ground and hot spring water in Yemen and higher than those obtained by Faanu et al. (2016) from underground water in the open areas for gold mining and processing in Ghana. The cancer risk associated with lifetime (70 years) intake of radium isotopes in water samples (<sup>226</sup>Ra and <sup>232</sup>Th) ranged from  $(135.6 \pm 17.2) \times 10^{-5}$  to  $(244 \pm 29.3) \times 10^{-5}$  with an average value of  $(178.1 \pm 22.2) \times 10^{-5}$ , which is slightly higher than the USEPA (2000) range  $(1 \times 10^{-4} - 1 \times 10^{-6})$ . Natural springs, shallow and deep wells and numerous springs are distributed across Siwa Oasis. These springs are used for drinking, irrigation and medical tourism (medical treatment).

We selected three springs for our study: Spring (1) is used for irrigation, spring (2) is utilised for medical tourism (spa) and spring (3) is used for drinking, irrigation and feeding a mineral water company. The mean values of the estimated effective dose from ingesting  $^{226}$ Ra,  $^{232}$ Th and  $^{40}$ K and the excess lifetime cancer risk in groundwater from this study were higher than the globally acceptable value. This is indicated that groundwater has to be treated to reduce the levels of radioactive contaminants before being utilised for drinking and other domestic purposes. Although there are no immediate public health effects at this time, it is likely that there will be long-term, accumulative health effects in this range. This suggests the need for environmental investigations and monitoring of wells and springs for the distribution of minerals and radionuclides in this region.

# **Bioaccumulation factor**

The bioaccumulative properties of microbial films toward <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs contamination may be very effective for potential application in monitoring and assessment of extreme habitats, but only in a short term, since the corresponding turnover rates are quite short. The other rationale for using microbial films for radionuclide monitoring purposes is their ease of sampling, continuous growth processes, rapid response to the contamination and the consideration as a unique biological system that can grow in such extreme habitats. Microbial films are good indicators and accumulators of radionuclides in Siwa extreme habitats, considering the very short lifespan of microbial films (from days to a few weeks) compared to the long lifespan of marine macroalgae. Moreover, these results may indicate the further ability of these microbial films to bioremediate radionuclides contaminated.

Our data have been described as the activity concentration for the dry weight, multiplied by 0.16 (Kulikov and Molchanova 1975), to be expressed as the fresh weight. In







general, the highest transfer factor was observed for <sup>232</sup>Th, followed by <sup>40</sup>K, and <sup>226</sup>Ra (Table 6). The transfer factors of <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs calculated for the microbial film samples collected from Siwa Lakes were 0.87, 1.59, 2.68 and 0.5, respectively. For the freshwater springs, the transfer factors of <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs calculated for the microbial film were 13.04, 40.77, 20.69 and 1.29, respectively. It is obvious that the highest transfer factor in hypersaline lakes was <sup>40</sup>K, while in springs, it was <sup>232</sup>Th. It is normal that the transfer factor of <sup>40</sup>K is high in lakes, while potassium is more abundant in hypersaline lakes as it is an essential element in soils and plants and is used as phosphate fertiliser. Otherwise, the transfer factor of <sup>232</sup>Th is high due to the low solubility of thorium in water, which explains the complete transfer of thorium from water into biota (microbial films). Similarly, the transfer factor of <sup>226</sup>Ra is lower than that of <sup>232</sup>Th, which is due to the fact that part of the radium is lost as a dissolved and precipitated substance in groundwater that some of the radium is loss as dissolved and precipitates in groundwater. However, the Siwa Oasis microbial systems can sequester <sup>226</sup>Ra (<sup>238</sup>U series), <sup>232</sup>Th series and <sup>40</sup>K at concentrations higher than those found in the surrounding water, making algae superior to other aquatic biotas for radionuclide monitoring, especially in systems where such biotic elements are lacking. Extremophilic microbial films can remove different radionuclides from surrounding media because each group/species selectively removes specific elements.

The accumulation of radionuclides in biota depends on the biological availability of the absorbed radionuclides, differences in biota size and lifespan, food chain variations and vertical distribution in the water column (Mito et al. 1999). Even though the life span of microbial films is short (days or weeks), they tend to accumulate high concentrations of radioactive nuclides in comparison with sediments. Microbial films are a consortium of cyanobacteria and bacteria embedded in gelatinous polysaccharide secretions. Such consortium has been reported to efficiently adsorb heavy metals (Ali et al. 2019).

The efficiency of radionuclide removal is influenced by the amount of viscous polysaccharide molecules covering the cell surface (Hill et al., 1997; Tamaru et al., 2005); these extracellular polysaccharides are capable of adsorbing U (Kalina et al., 2005). Most plants, algae and bacteria have negatively charged polysaccharides and carbohydrates on the surface of their cell walls (Myers et al. 1975; Lobban and Wynne 1981). Zn, Cu, Al and U are attracted to the negatively charged groups (Marques et al. 1990; Barker et al. 1998). Many additional side groups, such as amino, carboxyl, hydroxyl and sulphide, found in the polysaccharide backbone of cell walls, provide an additional net negative charge to the cell. However, certain groups have a positive charge and can adsorb complex anionic metal species. The number of cations and anions complexed is determined by the mixture of these groups on the cell surface. For a representative phytoplankter, the possible number of high-affinity surface binding sites is estimated to be about 107-108 per cell (Morel and Hudson 1985). On the bacterium Shewanella putrefaciens, the number of ligand sites for U(VI) was determined to be  $2.0 \times 10^{19}$  carboxyl sites,  $5.5 \times 10^{18}$ phosphoryl sites and  $2.3 \times 10^{19}$  amine sites per gram of bacteria (Haas et al. 2001). Mycobacterium smegmatis cell walls could probably adsorb about 100-mg U per gram of dry cell weight, according to Andres et al. (1994).

# Conclusion

Groundwater is the only source of water that sustains life in the Siwa Oasis. The increasing use of groundwater for drinking and agricultural purposes requires careful assessment of radioactive substances and their radiological risks to public and environmental health, and the transfer of radionuclides to biota must be considered in the initial stages of an environmental impact assessment. The results showed that the elements of the radionuclides in different environmental samples (water, sediment and microbial films) vary considerably, depending on the different

Sample no.	TF (BF)						
	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K	<sup>137</sup> Cs			
Zeitoun Lake	$0.57 \pm 0.33$	$1.53 \pm 1.01$	$2.22 \pm 0.73$	$0.40 \pm 0.27$			
Aghormi Lake	$0.95 \pm 0.66$	$1.78 \pm 1.34$	$1.06 \pm 0.57$	$0.60 \pm 0.18$			
Siwa Lake	$1.08 \pm 1.45$	$1.47 \pm 1.55$	$4.77 \pm 0.94$	$0.50 \pm 0.91$			
Mean	0.87	1.59	2.68	0.50			
Spring 1	$8.96 \pm 2.88$	$3.19 \pm 1.83$	$1.42 \pm 1.52$	$0.80 \pm 0.37$			
Spring 2	$20.39 \pm 4.46$	$56.67 \pm 5.28$	$46.2 \pm 5.78$	$1.60 \pm 0.60$			
Spring 3	9.77 ± 1.36	$62.44 \pm 4.80$	$14.44 \pm 1.76$	$1.47 \pm 0.23$			
Mean	13.04	40.77	20.69	1.29			
Worldwide (IAEA,1978)	100	1000	_	10			

 Table 6
 Transfer factor

 (bioaccumulation factor) water biota (microbial films) for

 radionuclides (l kg<sup>-1</sup>)
 biota (l kg<sup>-1</sup>)



sources and on the geography of the region. The mean activity concentrations of <sup>232</sup>Th and <sup>137</sup>Cs in the groundwater were in agreement with the recommended reference values, while the concentrations of <sup>226</sup>Ra and <sup>40</sup>K were higher than the reference value. Furthermore, the mean concentrations of radionuclides in microbial films and sediment were within the global value, except for <sup>226</sup>Ra and <sup>232</sup>Th in microbial film samples. Furthermore, the microbial films from the freshwater suggest that they are good accumulators of various radionuclides. The influence of agriculture in this area was also shown by the distribution of the radionuclides <sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs in hypersaline lakes. Regarding radiation hazards, the radium equivalent activity, external hazard and annual effective dose of the microbial films and sediment samples were below the permissible limits. Otherwise, the absorbed dose rates (D) and the representative level index ( $I\gamma$ ) were higher than the recommended value, which could represent the ability of the microbial films to absorb the absorbed dose into living tissue. However, the annual effective dose to infants, children and adults was below the ICRP 2000, MRLs of 1 mSv/year. The annual cancer risk from radionuclides in the Siwa groundwater varied within a narrow range with an average value of  $178.1 \times 10^{-5}$ , unacceptable when compared with the reference value.

The bioaccumulation factors of the radionuclide elements calculated for the microbial film samples collected from the freshwater springs are higher than the transfer factors of the radionuclide elements calculated from the hypersaline lakes. The microbial films studied can absorb <sup>226</sup>Ra, <sup>40</sup>K and <sup>137</sup>Cs from their living media. Furthermore, these microbial films have a high capacity for the uptake of <sup>232</sup>Th, in spite of the low solubility in water. The bioaccumulation efficiency of microbial films in the extreme habitats of Siwa Oasis toward radionuclides is very useful for potential applications in monitoring, assessing and remediating these habitats. This study has shown that the microbial films in the Siwa Oasis are able to tolerate not only high salinity and high temperature, but also high levels of radioactivity. The rationale for using these microbial films is simple: They are abundant, easily accessible and grow continuously. Most importantly, microbial films are the only ones living in these extreme habitats. For further bioindicators and bioremediation purposes, we need to gather basic information on the physiology and growth requirements of microbial films and their bioaccumulation properties toward radionuclides or heavy metals.

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**Data availability** The datasets and materials used during the current study are available from the corresponding author on reasonable request. All data generated or analysed during this study are included in this published article.

#### Declarations

Conflict of interest The authors declare no competing interests.

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