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Assessment of radiological risk to workers and members of the public from the operation of a groundwater treatment plant in Jordan

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ABSTRACT

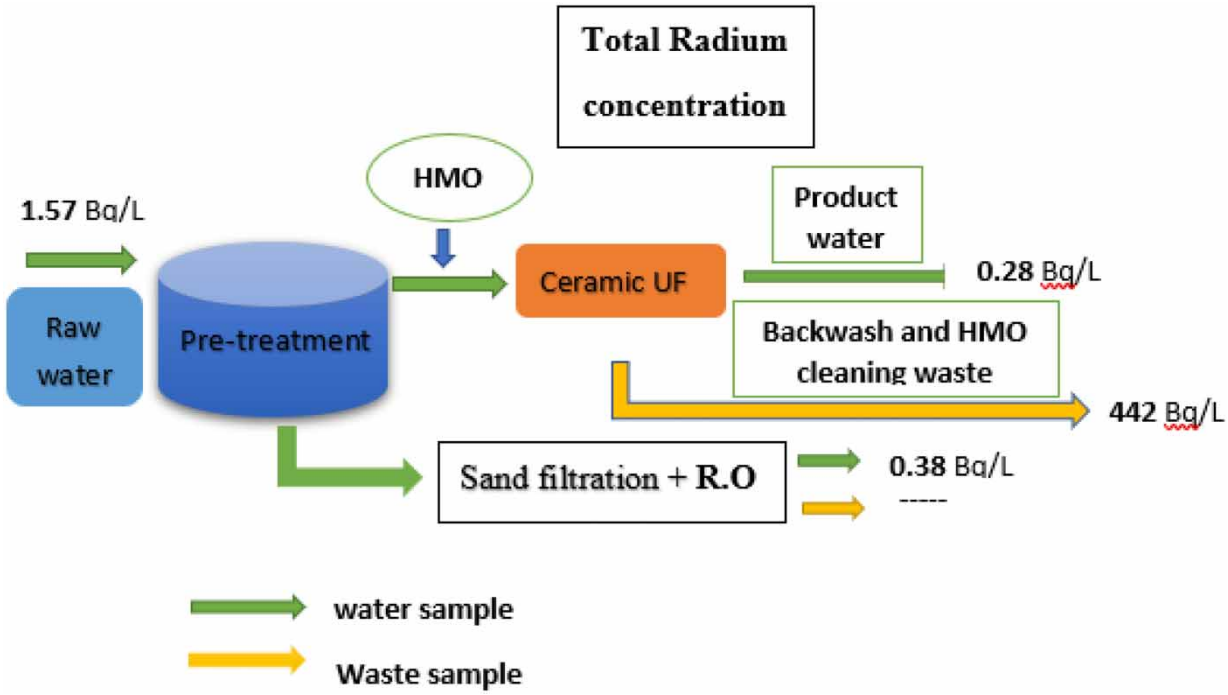
The study aimed to evaluate the radiation doses received by workers at a pilot water treatment plant (WTP) in Jordan. It also examined the concentrations of gross alpha, gross beta, and radium activities in both groundwater and treated water, along with a radiological risk assessment for the waste generated by the treatment process. The radioactivity levels of gross alpha and gross beta in the groundwater were found to exceed the established drinking water limits. In the pilot WTP, two methods were applied for water treatment, namely, ceramic ultra-filtration (CUF) and reverse osmosis (RO), both of which produced treated water that met drinking water quality standards. The annual effective dose from external radiation exposure to the WTP workers was found to be less than 0.007 mSv y^{-1} (during the filters backwash operation). However, the average annual dose from internal radiation due to inhalation of radon released from groundwater reached 3.2 mSv y^{-1}, exceeding the 1 mSv y^{-1} limit. Therefore, monitoring radon levels in workplaces is recommended. Radioisotope concentrations in the waste (sludge) stockpiles exceeded clearance levels, requiring them to be treated as radioactive waste. Overall, the WTP successfully produced drinking water that met quality standards, and the methods used could be replicated in other locations.

Key words: ceramic ultra-filtration, drinking water, effective dose, radioactivity, radiological risk, radium

HIGHLIGHTS

- Water treatment plant achieved the production of water meeting the water quality standards.
- The removal efficiency for radium activity averaged 82% for the CUF unit and 69% for the RO unit.
- Radon concentrations in the CUF and the RO units were below the maximum of 300 Bq m^{-3}.
- Workers received doses of 0.006 mSv y^{-1} in the CUF and the RO units.
- The main contributor to the internal radiation dose in the CUF unit was thoron.

GRAPHICAL ABSTRACT



1. INTRODUCTION

Presently in Jordan, groundwater is the main source of drinking water supplied to the population. Groundwater may contain several undesirable substances. Among these substances, dissolved naturally occurring radioactive materials (NORMs) may occur because of the long-lasting circulation of water in aquifers that facilitates the dissolution of radionuclides from the rocks into pore water. This originates loads of dissolved radionuclides, which are generally higher in groundwater compared with surface waters (Carvalho & Fajgelj 2013).

Radioactivity in water for human consumption is regulated in many countries and common standards have been agreed also at the international level for ensuring the radiological protection of the population (WHO 2011; Carvalho & Fajgelj 2013; USEPA 2018). These standards include limits for gross alpha (0.5 Bq L^{-1}) and gross beta (1 Bq L^{-1}) activity concentrations in drinking water, which would correspond to a radiation dose of 0.1 mSv y^{-1} to the water consumer. Among the radioactive isotopes present in groundwater, the radium isotopes ^{226}Ra ($T_{1/2} = 1{,}600$ years) and ^{228}Ra ($T_{1/2} = 5.75$ years), members of uranium and thorium radioactive decay series, respectively, are of most concern because of their long half-lives and easy absorption into the body (Kitto & Kim 2005). Radium ingested with the water is absorbed into the blood with a high absorption factor. It follows the calcium metabolic pathways and deposits in the bone tissue (Michel 1990; Milvy & Cothern 1990). Both ^{226}Ra and ^{228}Ra isotopes are equally prominent in producing health effects (IAEA 1990).

Radioactivity levels in groundwater may exceed the international and national radiation safety standards. When high concentrations of radionuclides are present, the water meant for human consumption may require treatment to render it safe for human consumption (Michel 1990; WHO 2010). Initially, the water treatment plants (WTPs) were designed to reduce suspended particulate matter, microorganisms, organic materials, and other undesirable chemical substances. Lately, WTPs have been requested to also reduce radionuclide concentrations and total dissolved salts in raw water. Consequently, this water treatment process may lead to the concentration and accumulation of radionuclides, such as radium isotopes, in the waste generated in the process (IAEA 2006). Therefore, water treatment has been identified as one of the industrial sectors with occurrence of NORM, which may require some form of regulatory control by radiation protection authorities (Michel 1990; WHO 2010).

In WTPs, a common radiological hazard is the exposure to radon isotopes, ^{222}Rn and ^{220}Rn, released from the water under treatment. Radon (^{222}Rn, $T_{1/2} = 3.8$ days) and thoron (^{220}Rn, $T_{1/2} = 55.6$ sec) are gaseous radium daughters. The release of

gaseous radionuclides into the air usually takes place inside the treatment plant facilities; hence, the WTP workers may be exposed to high occupational radiation doses from the radon and thoron inhalation with the breathed air. Recently, substantial attention has been drawn to the inhalation dose from indoor radon, thoron, and their short-lived progenies (Khandaker *et al.* 2021). Also, the external exposure to gamma radiation in the facilities may provide another contribution to the total dose received by workers in WTPs. Given the likelihood of occurrence of harmful effects on human health resulting from the internal and external exposure to ionizing radiation, the evaluation of radiation exposure and effective doses received by WTP workers is needed (Jokic *et al.* 2016). Other potential radiological hazard related to WTPs arises from radioactivity associated with the waste generated in the plant. The waste resulting from water treatment, such as the sludge, has been for a long time dumped directly into the environment without any specific precaution. Radiological hazards may occur when the sludge is used as a fertilizer in agriculture and as a component of building materials, among several possible uses.

In Southern Jordan, groundwater is used to provide the water for human consumption. Previous analyses of this groundwater have shown elevated levels of naturally occurring radionuclides (Smith *et al.* 1997; Vengosh *et al.* 2009; Moh'd & Powell 2010; Alomari *et al.* 2019a, 2019b, 2019c, 2019d, 2019e). The most important naturally occurring radionuclides identified were two radium isotopes, ^{226}Ra and ^{228}Ra, in activity concentrations up to 2 Bq L^{-1}. Traces of uranium were also reported in the groundwater (Alomari *et al.* 2019a, 2019b, 2019c, 2019d, 2019e). The radioactivity in this groundwater does not meet the national and international standards for drinking water and, hence, water treatment is required to reduce radioactivity. A pilot WTP was built for such purposes near Maan city, South of Jordan (El-Naser *et al.* 2016). If the results from the pilot plant are satisfactory, the construction of other WTPs may follow to ensure the supply of safe drinking water across the country. The current study aimed to assess the radioactivity in raw water and product water and to evaluate the effectiveness of the processes used for radioactivity removal from groundwater. In addition, this study aimed to assess the exposure of WTP workers to ionizing radiation and to assess the radiological hazards to the public and to the environment from the WTP waste disposal. This research is a pioneer study and the first of its kind in Jordan.

2. MATERIALS AND METHODS

2.1. Study area and WTP

The WTP studied is located 100 km south of Maan city, in the southern area of Jordan (29.33° E, 36.02° N), as shown in Figure 1. In this arid area, the summers feature a dry climate, the annual rainfall is less than 50 mm, and no surface water resources are available (Bender 1974). The WTP is fed with groundwater from Disi wells in the Ram-Disi aquifer. The Ram-Disi aquifer was formed some 30,000 years ago and it is non-renewable, thus, it is a fossil groundwater source.

The main geological formation hosting the Ram-Disi aquifer is the Cambrian–Ordovician sandstone, and it mainly consists of white median grained sandstone (El-Naser *et al.* 2016). The wells drilled into this aquifer are considered the most productive in Jordan and this aquifer constitutes the major source of water supply in the country (El-Naser & Gedeon 1996; Al-Zyoud *et al.* 2015).

The WTP studied is fed with groundwater pumped out from the Disi wells with a median flow rate of 30 $m^3 h^{-1}$. The plant treats the groundwater and supplies treated water to Almudawara and other towns in the proximity. The WTP infrastructure contains two lines of water treatment based on different technologies: ceramic ultra-filtration (CUF) and reverse osmosis (RO).

The water treatment line based on CUF processes groundwater that is stored in an interim metal storage tank of 50 m^3 capacity designated as raw water storage tank (RWST). The temporary storage of raw water prior to treatment allows the particulate impurities in water to settle by gravity (Brown *et al.* 2008). Chlorine is injected into the RWST for water disinfection. The disinfection targets microorganisms and aims to cancel biological hazards from raw water. After decantation and disinfection, the water from RWST is allowed to flow by gravity to the CUF unit whose main filtering elements are ceramic membranes made of silicon carbide.

Before water filtration, and for the removal of dissolved radium salts from the water, hydrous manganese oxide (HMO) is injected and mixed with the water. This is a recognized radium treatment method and it is based on the introduction of a mixture of potassium permanganate ($KMnO_4$) and manganous sulphate ($MnSO_4$) into the influent water stream prior to filtration. The $KMnO_4$ and $MnSO_4$ combine creating microparticles of HMO, which adsorb the dissolved radium (and arsenic) from the water. This treatment usually requires three tanks: one for dissolving $MnSO_4$, another for blending the dissolved $MnSO_4$ with the $KMnO_4$, and a third tank from which the freshly produced HMO is injected into the influent water. The

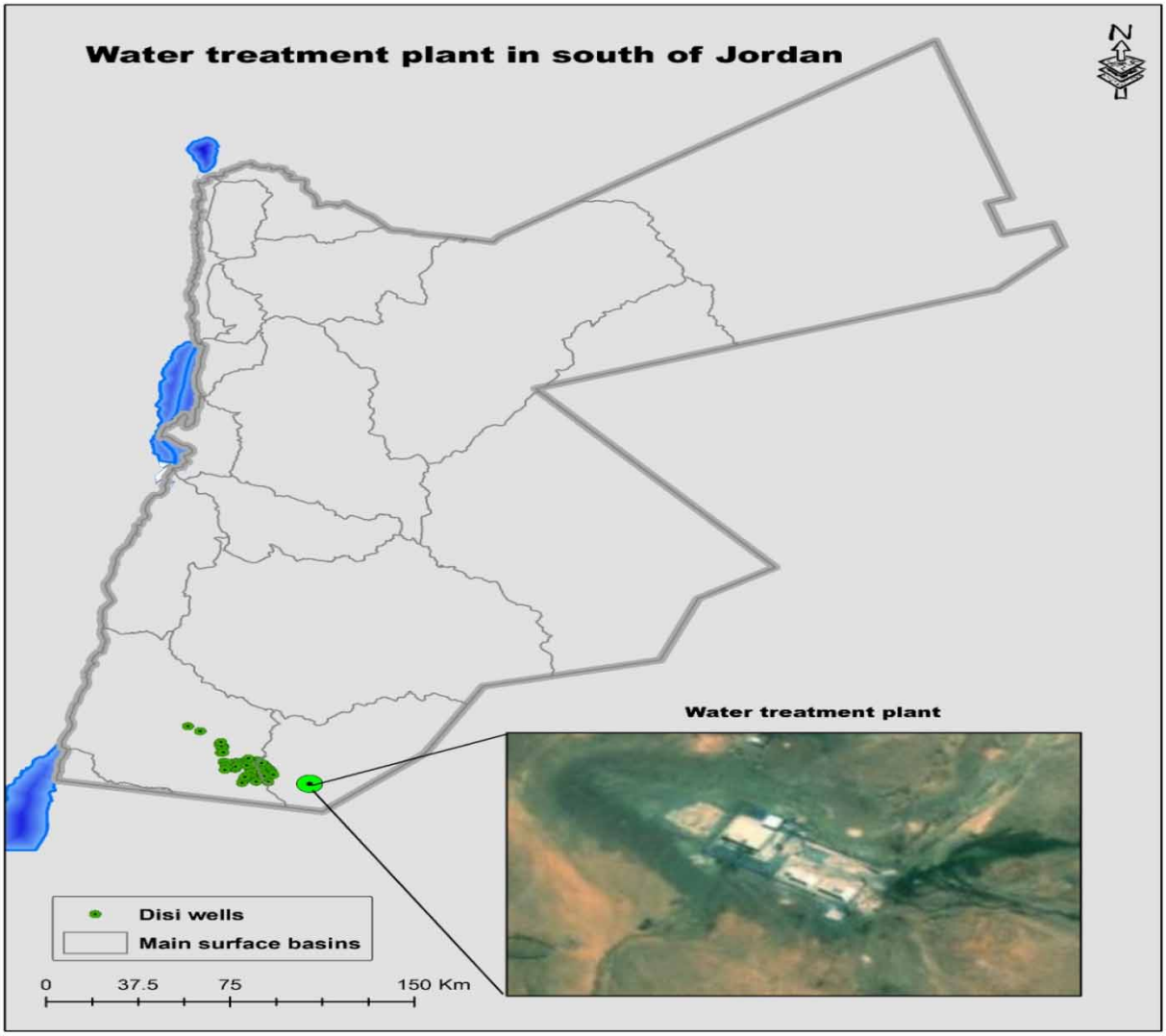


Figure 1 | Map of Jordan indicating the location of the study area and an aerial view of the WTP.

water and HMO slurry are then filtered through the CUF unit, with the HMO microparticles retained by the ceramic filters while purified water is produced. Upon exiting the filtration unit, the purified water is pumped into and collected in the filtered water tank, ready to be consumed.

The CUFs clog rapidly with the HMO microparticles and, in order to maintain the filtration efficiency, every 12 h the filters are backwashed (a reverse flow of water) with pure water. The backwash removes water impurities and the HMO microparticles that were retained on the CUF.

At several stages of this water treatment process, waste materials are produced comprising the sludge from the raw water decantation in the RWST and the water used in the CUF backwash. The waste from the CUF backwash is collected in the waste collection tank and, later, it is rejected with the decantation sludge by pumping both as slurry and discharging in a waste disposal area located 500 m from the WTP. The average cost of installing this plant was about $120,000 USD. Figure 2 shows a schematic diagram of the processes carried out in the CUF unit of the WTP.

The water treatment line based on RO is part of the same WTP, but it was installed in a separate facility that supplies treated water to other neighbourhoods in the region. This treatment line makes use of raw water from the same Disi wells as CUF, but the raw water is filtered first through a sand filter, followed by filtration on a cartridge before the RO. The basic elements of this treatment (sand filtration + RO) are shown in Figure 3. This water treatment line processes the raw water pumped from

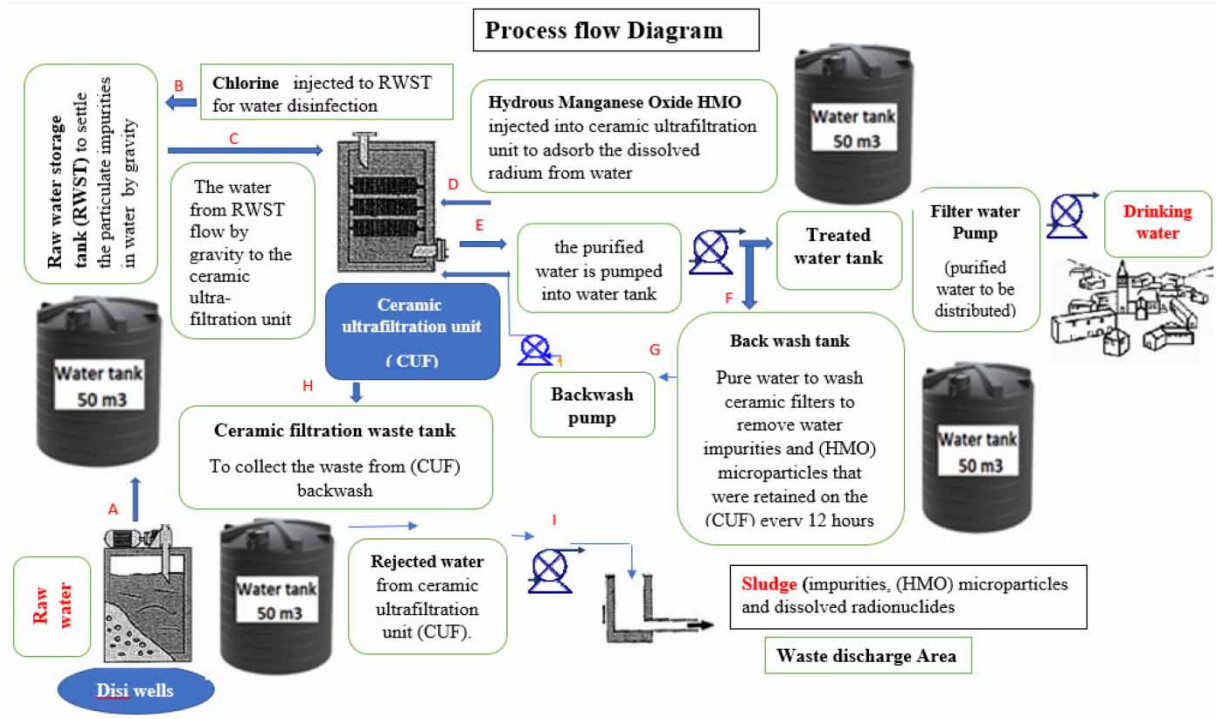


Figure 2 | Schematic diagram of the water treatment process based on CUF carried out in the WTP under study.

the Disi wells into an iron storage tank, with 50 m^3 capacity, for decantation. As in the CUF method, raw water is decanted in a tank to remove suspended particulate matter by gravity. Freshly produced HMO is added to the decanted water to adsorb dissolved radium. Then the water is pumped through two sand filters in order to eliminate the relatively high turbidity of the raw water and obtain low levels of suspended particulates in water. The sand filters consist of several layers of gravel and sand with different grain sizes. The sand filters trap fine colloidal materials and microparticles, including iron and manganese microparticles with adsorbed radium.

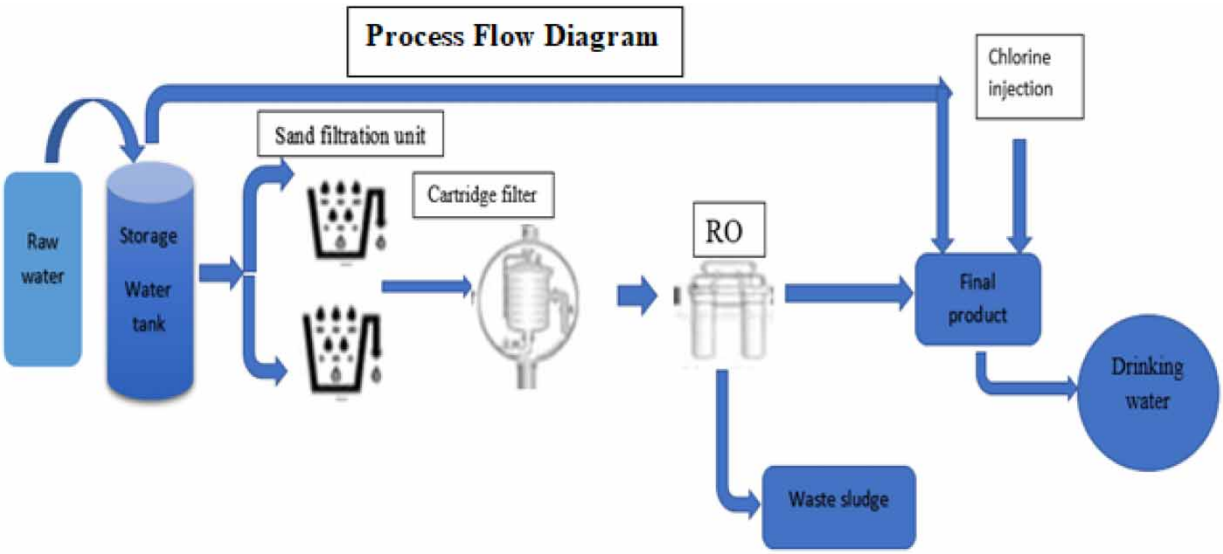


Figure 3 | Schematic diagram of the water treatment process carried out with sand filters and reverse osmosis (sand filtration + RO).

Water is then filtered through a cartridge filter to remove the remaining suspended particles. After this filtration the water is pumped through an RO unit. In the RO unit, the water moves through a semi-permeable membrane from the solution with higher salts concentration to the lower concentration solution using applied pressure (70 atmospheres). In this way the water is separated from the dissolved mineral salts and the radionuclides, and high purity water is obtained.

The treated water is collected in a ground tank with a capacity of 150 m^3. Before distribution, a small portion of the raw water (without treatment) is added to the treated water in the 150 m^3 ground tank in order to provide an appropriate concentration of minerals. The mix of treated water with raw water is disinfected with chlorine injected into this tank. Finally, this chlorine treated water is pumped from the tank, injected into the water distribution network, and supplied to the public as drinking water.

2.2. Water and sludge sampling and sample preparation

For the analysis of ^{226}Ra and ^{228}Ra radionuclides, replicate water samples (8 L each) were collected from raw groundwater, product drinking water, and wastewater from the filters backwash. These water samples were collected and prepared according to APHA standard methods (APHA 2012) and described in detail in previous reports (Alomari *et al.* 2019a, 2019b, 2019c, 2019d, 2019e). Additional 1-L water samples collected in polyethylene bottles were used for determination of gross alpha and gross beta activities. Analytical results were similar in the replicate water samples and averaged.

Sludge samples were collected from the sludge stockpile in the waste deposition area. The samples were obtained using a soil Auger sampler and the layers 0–5, 5–10, 10–20, 20–25, and 25–40 cm were sliced for radiometric analyses. To account for radionuclide background concentrations in the local top soil, one soil sample (0–25 cm depth) was taken in the area of Almudawara town. The sludge and soil samples were collected according to the methods recommended by the International Atomic Energy Agency (IAEA 2004), and the samples were prepared according to IAEA's guidebook on the measurement of radionuclides in food and environmental materials (IAEA 1989). A detailed description on the preparation of sludge samples to be measured using gamma spectroscopy system was written and can be found in previous reports (Alomari *et al.* 2019a, 2019b, 2019c, 2019d, 2019e).

2.3. Activity concentration measurements using liquid scintillation and gamma spectrometry

The gross alpha and gross beta radioactivity measurements were performed by liquid scintillation counting (LSC) using a LSC Tri-Carb 3110 (Perkin Elmer) and according to methods previously used (Alomari *et al.* 2019a, 2019b, 2019c, 2019d, 2019e). Water samples, collected in 1-L polyethylene bottles, were preserved at the laboratory by adding 2 ml of concentrated HCl (35%) to bring water pH level below 2. Water samples for gross alpha and gross beta measurements were prepared by the gentle evaporation method and 10 ml of concentrated water was mixed with 10 ml Ultima Gold LLT (Perkin Elmer) scintillation cocktail in glass vials for LSC.

The efficiency calibration of the LS counter was carried out by using certified standards of ^{90}Sr and ^{241}Am. The alpha/beta discrimination was set up by counting separately standards of ^{90}Sr (as pure beta emitter) and ^{241}Am (as pure alpha emitter). The background of the counting system was determined by preparing an LSC vial with 10 ml of Ultima Gold LLT scintillation cocktail and 10 ml of bi-distilled water.

The activity concentrations of radium isotopes in water and sludge samples were determined by gamma ray spectrometry using a high-purity germanium (HPGe) detector with a relative efficiency of 50% (Canberra). The energy resolution of the detector was 2.2 keV at the 1,332 keV gamma ray from ^{60}Co. The Genie2000 (Canberra) software was used to analyse the spectrometric data.

Energy and efficiency calibration were performed using a customized multistandard source (containing certified activities of radionuclides ^{139}Ce, ^{203}Hg, ^{109}Cd, ^{57}Co, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{60}Co, and ^{88}Y) in a 1-L volume Marinelli beaker (Eckert and Ziegler Isotope Products). A Marinelli beaker full with distilled water was counted to strip the background from the gamma spectra of samples. The activity of samples was measured for counting times of 20 h after formation of radioactive secular equilibrium in 1-month stored samples. ^{226}Ra from the uranium series was determined using the weighted mean activities of the three ^{214}Bi photopeaks (609.3, 1,120.3, and 1,764.5 keV) in secular radioactive equilibrium. ^{228}Ra from the thorium series was determined from the gamma peak of ^{228}Ac at 911.2 keV energy (Gilmore 2008).

Certified pure standards of ^{226}Ra and ^{228}Ra (Water ERA company) were used for quality control. The minimum detectable activity (MDA) of the measuring system for each radionuclide was calculated using Currie's method. The value of the MDA was 0.09 and 0.08 Bq L^{-1} for ^{226}Ra and ^{228}Ra, respectively. The method used was adopted from APHA (2012).

2.4. Assessment of the effective dose due to external radiation dose measurement

In order to monitor the dose rates of external radiation to workers, monitoring points were selected in the WTP to account for all work places. At each monitoring point, gamma dose rate (GDR) measurements were carried out in the air, 1 m above the ground (Alomari *et al.* 2019a, 2019b, 2019c, 2019d, 2019e).

The Inspector (S.E. International, USA) radiation survey meter was employed as a main gamma detector for *in situ* measurements of external GDR. Conversion of the GDR readings to effective dose rates was made from $\mu R\,h^{-1}$ to $nGy\,h^{-1}$ ($1\,\mu R\,h^{-1} \approx 8.7\,nGy\,h^{-1}$ was employed as the conversion factor). A second gamma detector, the RADIAGEM 2000 survey meter, which has a built-in Geiger–Mueller counter, was used. The Jordan Atomic Energy Commission Calibration Facility, a recognized Secondary Standard Dosimetry Laboratory (SSDL), was used for calibrating the instruments.

To assess the effective dose from external radiation to the workers inside WTP facilities, the absorbed gamma dose rates, GDR ($Gy\,h^{-1}$), measured in the air were converted to effective dose rates ($Sv\,y^{-1}$). The external annual effective dose (AED_{ext}) was calculated from the absorbed GDRs measured in the air, as follows (UNSCEAR 2000):

$$AED_{ext}\ (mSv) = GDR(nGy\,h^{-1}) \times 8,760(h) \times C_f \times 0.7(Sv\,Gy^{-1}) \times 10^{-6} \quad (1)$$

where GDR is the absorbed gamma dose rate in the air; 8,760 is the number of hours in 1 year; C_f is the occupation factor ($C_f = 0.2$ and $C_f = 0.8$ for outdoor and indoor occupancy, respectively); and $0.7\ Sv \cdot Gy^{-1}$ is the conversion coefficient from the absorbed GDR in the air to the effective dose received by adults. The indoor occupancy factor corresponds to the worker spending only 0.16 h per day indoors for implementing the filter backwash operation. In this study, it was assumed that the worker stands very close to the CUF for the whole duration of the backwash operation.

2.5. Assessment of the effective dose from exposure to radon and thoron through inhalation

The determination of radon (^{222}Rn) and thoron (^{220}Rn) activity concentrations in the indoor atmosphere of WTP facilities was carried out using an active radon analyser (RTM 2200, SARAD, Germany). This instrument measures the concentration of radon gas in the air pumped into a chamber containing a semiconductor detector. The detector counts the short-lived daughter products resulting from the radon decay inside the chamber. Following the radon radioactive decay, the resulting ^{218}Po nuclei become positively charged for a short period of time, and the ^{218}Po ions are collected by the electrical field of the detector. The number of collected ^{218}Po ions is proportional to the concentration of radon inside the chamber. For thoron, the activity concentration is determined through counting the thoron daughters, ^{216}Po ions. The uncertainty of radon and thoron activity concentrations in the air depends mainly on the number of counts recorded during the counting period (Janik & Bossew 2016). The radon and thoron contents were measured at several monitoring points to determine the exposure at all workplaces in the WTP.

The internal annual effective dose (AED_{int}) received by the WTP workers, from inhalation of radon and thoron at the workplace, was calculated using the following equation (UNSCEAR 1993):

$$AED_{int}(mSv) = C_{Rn} \times F \times T \times D_f \quad (2)$$

where C_{Rn} is the measured indoor radon or thoron concentration in $Bq\,m^{-3}$, and F is an adjustment factor representing the degree of equilibrium between the radon and radon progeny. An F value of 1 represents full radioactive equilibrium between radon and its airborne short-lived progeny. For the indoor air measurements, this factor was assumed at 0.4, an average value adopted by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and the International Commission on Radiological Protection (ICRP) (WHO 2010). Furthermore, in the abovementioned equation, T represents the worker's exposure time in hours during a whole year, and D_f is a concentration-to-dose conversion factor that is equal to $(1.2 \times 10^{-5}\ mSv\,m^3\,Bq^{-1}\,h^{-1})$ and $(1.2 \times 10^{-4}\ mSv\,m^3\,Bq^{-1}\,h^{-1})$ for radon and thoron, respectively (ICRP 2017). The worker's exposure time was calculated based on actual working hours for the whole year in the filters' backwash operation, i.e., 25 h exposure per year.

2.6. Efficiency of water treatment methods in the removal of gross radioactivity and radium isotopes

The water treatment processes implemented at the WTP aim at removing radionuclides from the water and converting the raw water into potable drinking water considering the radioactivity parameters. In this study, the efficiency of treatments

was calculated as a measure of total radioactivity and radium isotopes removal by the water treatment. The removal efficiency (RE) of the CUF and the sand filtration + RO methods was calculated by comparing activity concentration values in the influent (raw) with those in the effluent (treated) waters of the process using the following equation (Montaña *et al.* 2013):

$$RE(\%) = \frac{Activity\,influent - Activity\,effluent}{Activity\,influent} \times 100 \tag{3}$$

2.7. Assessment of the radiological risk from reuse of WTP sludge

Concerning the sludge generated in the WTP, the current study assumed its possible reuse as a construction material. The radiological risk associated with this use was evaluated through the activity concentration index (I), as follows:

$$I = \frac{C_{Ra226}}{300\,Bq \cdot kg^{-1}} + \frac{C_{Th232}}{200\,Bq \cdot kg^{-1}} + \frac{C_{K40}}{3,000\,Bq \cdot kg^{-1}} \tag{4}$$

where C_{Ra226}, $C_{Th232,}$ and C_{K40} are the concentrations in Bq kg^{-1} (dry weight) of these radionuclides in the sludge (UNSCEAR 2000). The concentrations of ^{232}Th in the sludge were not determined directly by the gamma spectrometric method used. However, assuming radioactive equilibrium between ^{232}Th and ^{226}Ra in the sludge, the ^{232}Th concentration can be considered equal to the ^{228}Ra concentration. The assumption is reasonable because ^{228}Ra is a radioactive progeny of ^{232}Th and if there has been rupture of radioactive equilibrium that would be because of radium dissolution and removal by water and, in such case, this would lead to an underestimation of ^{232}Th concentration. Therefore, this is a conservative approach.

The I index is an estimate of the gamma radiation dose for a member of the public exposed to the radiation from construction materials made with the sludge from the water treatment. An I index value exceeding unity indicates a radiation exposure exceeding the dose limit of 1 mSv y^{-1} for radiation added to the natural radiation background (UNSCEAR 2000; IAEA 2018).

3. RESULTS AND DISCUSSION

3.1. Radiological dose assessment

3.1.1. Annual effective dose due to the external radiation

The results of external effective dose rates (nGy h^{-1}) based on the *in situ* GDR measurements are shown in Figure 4.

The average background effective dose rate for the Almudawarah area was 70 nGy h^{-1}, which is slightly lower than the world's average external effective dose rate of 84 nGy h^{-1} (UNSCEAR 2000).

The average effective dose rate by the water storage tank was determined to be 85 nGy h^{-1}, which is comparable with the world's average value. In the waste deposition area, by the discharge of the filter backwash water and sludge from the raw water decantation, the average effective dose rate was determined to be 208 nGy h^{-1}, and at the CUF unit, the dose rate reached 356 nGy h^{-1}, closely followed by the dose rate at the sand filtration + RO unit. Inside the WTP, the highest gamma dose rates were consistently measured in the CUF unit during the ceramic filter backwash operation. The dose rate values measured at the waste deposition area and at the CUF unit were approximately 2 and 5 times higher than the world's average value, respectively.

As shown in Figure 5, the average value of AED_{ext} in the area of Almudawara town was determined as 0.08 mSv y^{-1}, which is a typical natural background value and close to the world's average of 0.07 mSv y^{-1} (UNSCEAR 2000).

Inside the WTP facilities, at the CUF unit, the dose rate received by workers during the filter washing process (the radiation exposure worst-case scenario, but with a limited exposure time) corresponds to an annual effective dose of 0.007 mSv.

To assess the annual effective dose risk to workers by the sand filtration + RO unit, the results obtained during the washing process of the filter were of 321 nGy h^{-1}, which corresponds to an annual effective dose of 0.006 mSv y^{-1}, similar to the effective dose in the CUF unit. The effective dose received by the workers during filters' backwash in the RO unit and the dose received by workers in the CUF unit were similar. For comparison, the AED_{int} value is 0.41 mSv y^{-1} (UNSCEAR 2000).

3.1.2. Annual effective dose due to radon and thoron inhalation

As mentioned previously, one of the radiation exposure situations in the WTP is the exposure to radon degassed from the raw water inside closed rooms during water filtration and backwash operations in CUF and sand filtration + RO units. The results of measurements of radon and thoron concentrations inside the facilities of both units are provided in Table 1.

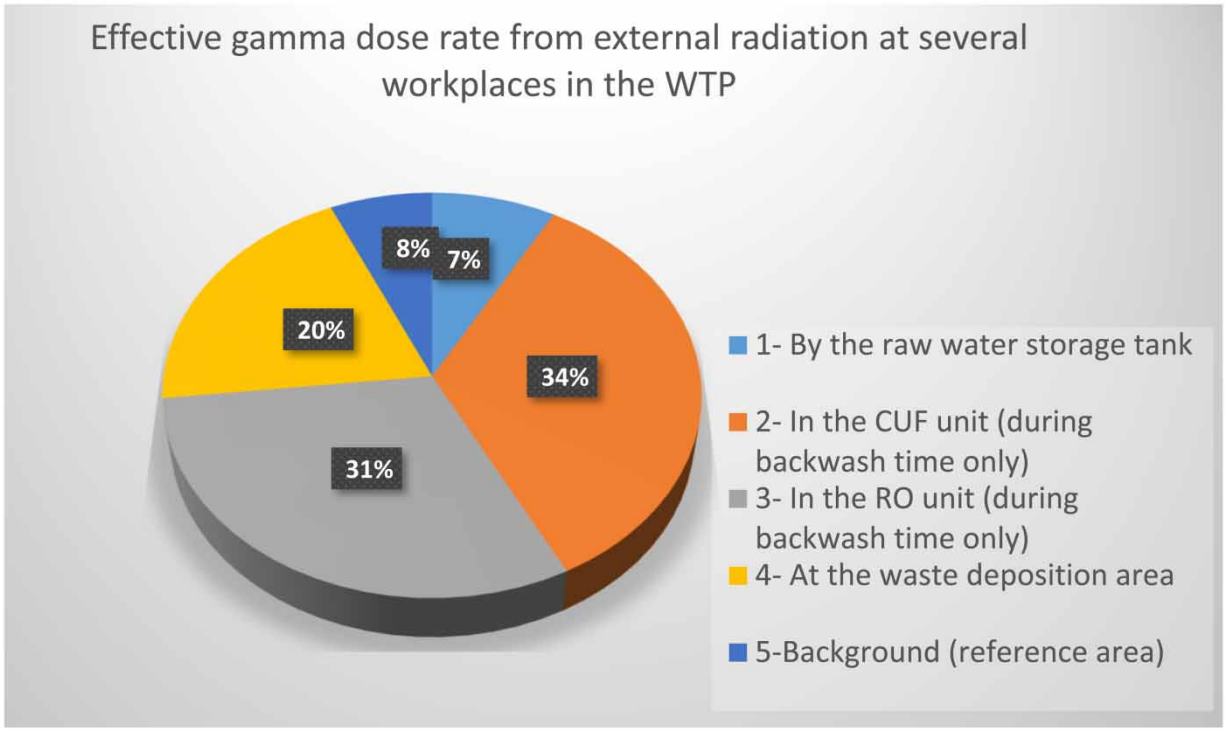


Figure 4 | Effective gamma dose rate from external radiation at several workplaces in the WTP.

The average values of radon (^{222}Rn) concentrations in the indoor air during water filtration and backwash operations in CUF and sand filtration + RO units were below the maximum of 300 Bq m^{-3} adopted by the international standards (Table 1). Thoron (^{220}Rn) was the main contributor, with 97% and above, to the internal radiation dose received by workers through inhalation. The radiation dose from inhaled radon isotopes was higher during the filters' backwash operation.

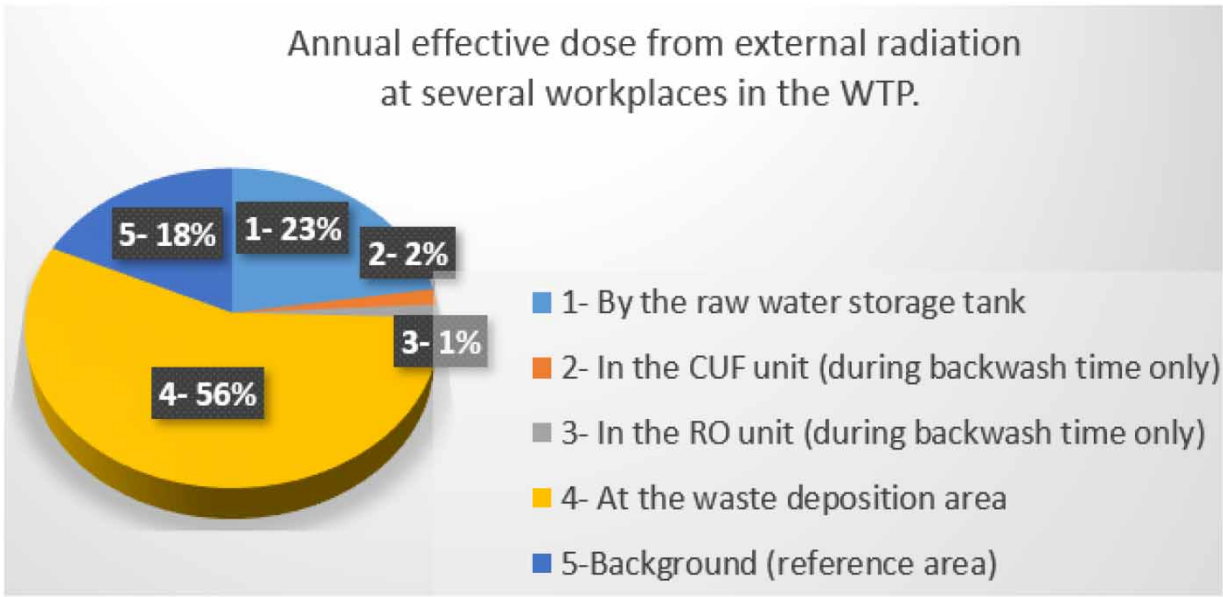


Figure 5 | Annual effective dose from external radiation at several workplaces in the WTP.

Table 1 | Annual effective dose due to the inhalation of radon and thoron and their progenies in the WTP facilities during water filtration and filters' backwash

	Indoors in CUF unit			Indoors in sand filters + RO unit		
	Concentration in the air (Bq m^{-3})			Concentration in the air (Bq m^{-3})		
Radionuclide	Backwash	Filtration	Annual effective dose (mSv y^{-1})	Backwash	Filtration	Annual effective dose (mSv y^{-1})
Radon (Rn222)	115 ± 6	61 ± 3	0.02	108 ± 5	55 ± 5	0.03
Thoron (Rn220)	1,141 ± 50	86 ± 4	3.2	450 ± 25	183 ± 9	1.10
Total	–	–	3.22	–	–	1.13

Since workers' presence during the washing process does not exceed 10 min each time and this is repeated 150 times in 1 year, the annual radiation dose that workers receive during this operation (25 h per year) from radon-222 and radon-220 inhalation were calculated accordingly (Table 1). The AED_{int} values due to exposure to radon and thoron and their short-lived progenies for indoor workplaces were in both units mostly due to thoron. The AED from exposure to radon and thoron was significantly higher in the CUF unit than in the sand filtration + RO unit (Table 1).

3.2. Gross alpha, gross beta, and radium isotopes activity concentrations in water and the radioactivity RE

The activity concentrations of gross alpha, gross beta, ^{226}Ra, and ^{228}Ra were determined in raw water, treated water, and wastewater. The origin of gross alpha and gross beta activities in water from the Disi aquifer had been investigated previously and it was established that gross alpha activity was related to ^{226}Ra while gross beta activity was largely due to ^{228}Ra content (Alomari *et al.* 2019a, 2019b, 2019c, 2019d, 2019e). The gross alpha, gross beta, and radium RE of both water treatment methods were calculated from the concentration measurements. The results for the water treatment with the CUF method are presented in Table 2, and for the water treatment with sand filtration + RO, the results are provided in Table 3.

The gross alpha and gross beta activity concentrations in raw water averaged 2.2 and 1.7 Bq L^{-1}, respectively (Table 2). Both values were higher than the limits set by the Jordanian standards for drinking water quality at 0.5 Bq L^{-1} for gross alpha and 1 Bq L^{-1} for gross beta, in line with the World Health Organization (WHO) recommendations (JISM 2008).

Table 2 | Activity concentrations of gross alpha, gross beta, ^{226}Ra, ^{228}Ra, and total radium in water samples and radioactivity RE of the CUF treatment method

	Gross alpha (Bq L^{-1})	Gross beta (Bq L^{-1})	^{226}Ra (Bq L^{-1})	^{228}Ra (Bq L^{-1})	Total Ra (Bq L^{-1})
Raw water	2.2 ± 0.1	1.7 ± .08	0.17 ± 0.01	1.40 ± 0.01	1.57
Product water	0.17 ± 0.01	0.3 ± 0.01	0.08 ± 0.01	0.2 ± 0.001	0.28
RE (%)	92	82	53	86	82
Wastewater	–	–	71 ± 0.06	371 ± 0.15	442
WHO limit (Bq L^{-1})	0.5	1	1	0.1	–

Table 3 | Activity concentrations of gross alpha, gross beta, ^{226}Ra, ^{228}Ra, and total radium, and the radium RE through the water treatment process using the sand filtration + RO method

Treatment stage	Gross alpha (Bq L^{-1})	Gross beta (Bq L^{-1})	^{226}Ra (Bq L^{-1})	^{228}Ra (Bq L^{-1})	Total Ra (Bq L^{-1})
Raw water	2.2 ± 0.1	1.7 ± .08	0.17 ± 0.01	1.4 ± 0.01	1.57
Product water	0.03 ± 0.001	0.05 ± 0.001	0.10 ± 0.01	0.28 ± 0.01	0.38
RE (%)	> 99	> 97	41	80	69
WHO limit	0.5	1	1	0.1	–

In raw water, the mean gross alpha activity concentration was about four times the national regulatory limit, which was in agreement with previous reports (Vengosh *et al.* 2009; El-Naser *et al.* 2016; Alomari *et al.* 2019a, 2019b, 2019c, 2019d, 2019e). The mean gross beta activity concentration of raw water was one and half times the limit set by the Jordanian standards (JISM 2008). These results confirmed that groundwater would not be suitable for human consumption without previous treatment to abate radioactivity.

With the construction of the WTP, the average gross alpha and gross beta activity concentrations in product (treated) water were 0.17 and 0.3 Bq L^{-1}, respectively. These values for treated water were below the limits set by the Jordanian standards and the WHO standards for drinking water. The efficiency of removal of gross alpha and gross beta activity concentrations by the CUF method was of 92 and 82%, respectively (Table 2). These results show that raw water treatment by the CUF unit decreased the water gross alpha and gross beta radioactivity to acceptable values.

The analysis indicated also that the average activity concentration of ^{226}Ra isotope in raw water was 0.17 $\pm$ 0.01 Bq L^{-1}, but after CUF treatment, the average activity concentration of ^{226}Ra in treated water was reduced to 0.08 $\pm$ 0.01 Bq L^{-1}, which was much lower than the specific limit for ^{226}Ra recommended by the WHO for drinking water, 1 Bq L^{-1} (WHO 2006). The RE for ^{226}Ra by the CUF treatment method was calculated at 53% (Table 2). It is worth mentioning that the high ^{226}Ra concentrations in groundwater are not of anthropogenic origin. The high ^{226}Ra activity concentrations are indicative of the ^{238}U content of the aquifer containing rocks and are mainly controlled by the ^{238}U content (Lauria *et al.* 2004).

The average ^{228}Ra activity concentration in raw water was determined at 1.4 $\pm$ 0.01 Bq L^{-1}, a value that is 14 times higher than the limit of 0.1 Bq L^{-1} set by the WHO specifically for this radioisotope (WHO 2006). However, the water filtration process by the CUF method decreased the ^{228}Ra activity to 0.200 $\pm$ 0.001 Bq L^{-1} in treated water, which is only slightly higher than the limit recommended by the WHO. The RE of ^{228}Ra activity concentration was calculated at 86% (Table 2). The CUF treatment removed radium (the two isotopes) from raw water with an average efficiency of 82% (Table 2).

According to previous reports, the highest mean activity concentration of ^{228}Ra in the groundwater of Jordan was measured in water samples from the Disi Aquifer (1.441 $\pm$ 0.024 Bq L^{-1}) (Alomari *et al.* 2019a, 2019b, 2019c, 2019d, 2019e). This elevated ^{228}Ra concentration gives an indication of the existence of high ^{232}Th levels in the aquifer rock matrix. Some authors suggested that high ^{228}Ra activity can be expected when the groundwater has an extended residence time in the sandstone aquifer (Weaver & Bahr 1991). Furthermore, within the sandstone rock of the Disi aquifer, there are thorium-enriched abundant nodules (mineral aggregations) (El-Naser *et al.* 2016). Such aggregations could be the source of radioactive isotopes, and particularly of ^{228}Ra.

In 2016, the Jordanian Ministry of Water supported an investigation to measure radioactive isotopes in water from Disi wells. One of the most prominent results of that investigation was the finding of high radium concentrations in groundwater, which supported the raising in the Jordanian water standards of the radiation dose limit from the ingestion of drinking water from 0.1 to 0.5 mSv y^{-1} (thus committing 50% of the international dose limit for members of the public to drinking water, justified with the absolute need to provide water to the population). The current study indicates that with the installation of the WTP, the internal radiation dose from radium isotopes in drinking water to the consumer is now 0.2 mSv y^{-1}, which is significantly lower than the limit of 0.5 mSv y^{-1} adopted in the Jordanian water standard. Therefore, the consumption of product water from the WTP does not originate a radiation exposure exceeding the legal limit and meets the radiation safety standards.

The activity concentrations of gross alpha, gross beta, ^{226}Ra, and ^{228}Ra in water treated by the sand filters and RO process were measured in raw water and treated water (Table 3). The radium RE of the water treatment method was calculated from these concentration measurements. The results showed that the radioactivity RE by the sand filter and RO filtration unit was very high for gross alpha and gross beta activities and better than those obtained by the CUF unit. Nevertheless, in the product water, the concentrations of radium isotopes were lower in the CUF method than by the sand filtration + RO.

It is interesting to note that in raw water ^{226}Ra accounted for nearly 8% of the gross alpha activity, which means that there are other alpha emitting radionuclides in the groundwater (may be ^{210}Po and ^{224}Ra), while ^{228}Ra in raw water accounted for 82% of the gross beta activity (Table 3).

3.3. Radionuclides in the sludge at the waste disposal site

Ceramic filters and sand filtration + RO are backwashed every few hours to remove trapped radionuclides and microparticles to maintain the filters' efficiency. The wastewater resulting from this backwash process is dumped into the waste area with the

sludge from the raw water decantation, as described previously. The access to the RO waste area was not possible, and the study of radioactive waste was performed only in the waste area from the CUF unit.

The activity concentrations of the radionuclides in the sludge samples collected from the waste disposal area are reported in Table 4. The sludge contained higher radioactivity concentrations than a normal soil from the region. The enhanced radionuclide concentrations of the sludge resulted from the transfer of naturally occurring radionuclides from the raw water to the solid waste operated by the water treatment process. For this reason, this waste is often referred to as technologically enhanced naturally occurring radioactive material (TENORM).

Potassium (^{40}K) in the waste (sludge) samples displayed concentrations ranging from 540 $\pm$ 11 to 1,075 $\pm$ 21 Bq kg^{-1} and those concentrations increased with depth. The highest ^{226}Ra and ^{228}Ra activity concentrations in the sludge layers were 481 $\pm$ 5 and 1,596 $\pm$ 9 Bq kg^{-1}, respectively, and both occurred in the 5–10 cm layer below the surface. This difference in distribution pattern of radionuclides with depth suggests that potassium is easily dissolved by the wastewater and transported downwards by percolation, while radium, which has a high partitioning coefficient (Kd $= 7.4 \times 10^3$), remained adsorbed by the sludge particles and displayed a slow downward migration in the waste stockpile.

Environmental impact assessments often include the assessment of radionuclide migration through the subsoil. In the context of water treatment, the potential migration of radium in soil would be a major concern because of high concentrations and toxicity of radium. However, the migration of radium through the layers of sludge and soil beneath the sludge piles might be a slow process.

3.4. Radiological risk from the reuse of sludge

The radiological risk of reusing sludge was assessed through the gamma index (I). To keep the radiation exposure of the public at negligible levels, the I value should not exceed unity, corresponding to the dose limit of 1 mSv y^{-1} (IAEA 2018). The calculated gamma index for the sludge varied from 1.2 to 2.8 based, respectively, on the lower and higher concentrations determined in sludge layers (Table 4) and, thus, it was consistently above the dose limit. Therefore, if sludge is used as a construction material, it may pose a significant radiation hazard to the public because of the exposure to ionizing radiation emitted by radionuclides in the sludge and because of the inhalation of radon isotopes released by that material.

The radiological risk of the sludge can also be assessed by comparison of radionuclide concentration values with the clearance and exemption levels (IAEA 2014). For the natural radionuclides, the clearance levels recommended are 10 kBq kg^{-1} for ^{40}K and 1 kBq kg^{-1} for all other radionuclides. Furthermore, the exemption level recommended for ^{40}K is 100 kBq kg^{-1} and 10 kBq kg^{-1} for all other radionuclides (IAEA 2014). This means that the activity concentrations of ^{40}K and ^{226}Ra in the sludge were lower than the clearance levels recommended by the IAEA, while the activity concentration of ^{228}Ra was higher than the respective clearance level.

Therefore, the residues from groundwater treatment, including the sludge from water decantation and wastewater from filters' backwash, contain significant levels of radioactivity. The direct discharge of these wastes in the environment without any treatment would enhance environmental radioactivity levels and could represent a radiological hazard to the population and to the non-human biota. Recently, high attention was given by the Water Authority of Jordan to the disposal of radioactive waste materials generated as a result of the groundwater treatment. It was advised by the Authority that such waste must be collected in a large tank and afterwards disposed by land-spreading in a restricted area of the desert.

Table 4 | Activity concentrations of radionuclides (Bq kg^{-1} dry weight) in sludge layers from the waste stockpile and in a background soil sample

Sludge profile and background soil sample	^{40}K	^{226}Ra	^{228}Ra
Layer 1 (0–5 cm)	540 $\pm$ 1	55 $\pm$ 1	173 $\pm$ 1
Layer 2 (5–10 cm)	756 $\pm$ 17	481 $\pm$ 5	1,596 $\pm$ 9
Layer 3 (10–20 cm)	925 $\pm$ 27	191 $\pm$ 2	501 $\pm$ 2
Layer 4 (20–25 cm)	950 $\pm$ 29	174 $\pm$ 2	479 $\pm$ 3
Layer 5 (25–40 cm)	1,075 $\pm$ 21	115 $\pm$ 3	313 $\pm$ 4
Background soil (0–25 cm)	440 $\pm$ 3	25 $\pm$ 1	33 $\pm$ 1

4. CONCLUSIONS

The levels of gross alpha, beta, ^{226}Ra, and ^{228}Ra radioactivity in water from the Disi wells were measured at 2.2 ± 0.1, 1.7 ± 0.08, 0.17 ± 0.01, and 1.4 ± 0.01 Bq L^{-1}, respectively, all surpassing the recommended limits. Due to this high radioactivity, treatment is necessary before the water can be safely consumed. Two methods were used at the WTP: CUF and a combination of sand filtration and RO. CUF reduced gross alpha and beta levels to 0.17 ± 0.01 and 0.3 ± 0.01 Bq L^{-1}, while sand filtration + RO achieved further reductions to 0.03 ± 0.001 and 0.05 ± 0.001 Bq L^{-1}. Sand filtration + RO had higher RE, eliminating over 99% of alpha and 97% of beta activity, outperforming CUF's efficiency of 92 and 82%, respectively. Additionally, the groundwater showed higher concentrations of ^{228}Ra compared with ^{226}Ra, with CUF removing 82% of radium and sand filtration + RO removing over 69%. Both methods successfully produced water that complied with WHO guidelines and Jordan's water quality standards. A radiation dose assessment for workers at the WTP revealed low external gamma exposure during filter cleaning. However, inhalation of radon, especially thoron (^{220}Rn), posed a significant risk, potentially exceeding the 1 mSv y^{-1} limit for non-radiation workers, emphasizing the need for workplace radon monitoring. The treatment process requires proper management under radioactive waste regulations. Improper disposal could lead to environmental contamination and public radiation exposure. Reusing WTP sludge in construction materials was found to pose a radiological hazard. Therefore, the sludge is unsuitable for construction use and must be managed as radioactive waste. Despite this, the water treatment methods effectively produced safe drinking water, and these techniques could be successfully implemented in other areas.

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DISCLOSURE STATEMENT

All authors have read and agreed to the published version of the paper.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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