Journal of Environmental Radioactivity 178-179 (2017) 245–252

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Radiation hazards and lifetime risk assessment of tap water using liquid scintillation counting and high-resolution gamma spectrometry

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ARTICLE INFO

Article history:
Received 28 March 2017
Received in revised form
3 September 2017
Accepted 4 September 2017

Keywords:
Lifetime risk
Annual effective dose
Internal hazard
Gross alpha beta
Natural radioactivity
Radionuclides

ABSTRACT

In this work, two complementary techniques, viz. liquid scintillation counting and high-resolution gamma spectrometry are utilized to analyze radionuclides concentrations in tap water of Irbid governorate, Jordan, and study their correlation. Gross alpha and gross beta concentrations, in the tap water samples collected from the nine districts of Irbid governorate, ranged from <82 to 484 mBq/L with a mean of 295 mBq/L and from <216 to 984 mBq/L with a mean of 611 mBq/L, respectively. Furthermore, gamma spectrometry analysis, for the tap water samples, shows that the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K ranged between <19 and 302 mBq/L, 24 to 119 mBq/L, and <101 to 342 mBq/L, respectively. There was a weak or even no correlation among the identified natural radionuclides with no trace of artificial radioactivity. In addition, the results of both techniques show that storing tap water in drilled wells leads to higher levels of radioactivity concentrations beyond the international permissible limits. Furthermore, the average lifetime risk and annual effective dose received by age-grouped inhabitants due to direct and indirect tap water consumption are evaluated, where most of the received dose is attributed to ^{226}Ra.

1. Introduction

Water is considered one of the most significant human resources; its quality is extremely essential and it is one of the major parameters in environmental studies. Water physical, chemical, and radioactive properties should be well known to evaluate its aptness for human intake. Studying the radioactive content of water has gained a lot of interest in the last decades because of the increasing nuclear activities around the world and water ability to carry on both natural and artificial radioactive pollutants. This has increased the awareness on the effects of natural radioactivity on water quality and its direct contribution to the human ingested radioactive dose.

Depending on its origin, radiation can be mainly divided into two categories, terrestrial and cosmic. The dose due to radiation exposure is mostly (96%) from naturally occurring radioactivity (gamma radiation, beta and alpha particles) or from artificial sources (4%) (Rajeshwari et al., 2014). Weathering process of rocks has a significant contribution on terrestrial water radioactivity, where the degree of weathering and the mineralogical and geochemical composition of rocks determine the possible degree of water radioactivity (Saleh et al., 2015). Most of water radioactive contents is due radionuclides originating from natural decay series of uranium (^{238}U) and Thorium (^{232}Th), in addition to potassium (^{40}K) (Turhan et al., 2013). The natural decay of ^{238}U is a key origin of harmful radioisotopes such as radium (^{226}Ra) and radon (^{222}Rn). ^{222}Rn is a gas at room temperature with reasonable solubility in water that decreases with increasing water temperature. Despite the fact that ^{222}Rn has a relatively short half-life of about 3.82 days; it can be widely distributed due to diffusion through soil, atmospheric convection, or groundwater movement (Ball et al., 1991; Burnett and Dulaiova, 2003). On the other hand, ^{232}Th series contains ^{228}Ra that may become a critical contaminant, as its radiotoxicity is relatively high. Due its low solubility in water; ^{228}Ra mobility is limited and its concentration mainly depends on ^{232}Th concentration (Nuccetelli et al., 2012). ^{40}K is also a widespread gamma/beta emitter of primordial origin. Water sources should be continuously monitored against the contamination levels of these radionuclides that can cause harmful internal exposure to the body as a result of direct water intake or by ingestion through the food chain.

The first step in detection of water radioactivity concentrations

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http://dx.doi.org/10.1016/j.jenvrad.2017.09.005
0265-931X/

is to determine its gross alpha and beta activity levels. This serves as a screening technique that gives a good general indication about the water total alpha and beta radioactivity concentrations (Turhan et al., 2013). However, gross alpha/beta method lacks the ability of determining the responsible radionuclides and, hence, can be followed by gamma spectrometry for radionuclides identification. Heretofore, many researchers investigated the natural radioactivity of drinking water around the world (Akbulut and Taskın, 2015; Calin et al., 2015; Çevik et al., 2006; Chau and Michalec, 2009; Davila Rangel et al., 2002; Desideri et al., 2007; Fatima et al., 2007; Karamanis et al., 2007; Kovács et al., 2004; Palomo et al., 2007; Rožmarić et al., 2012, 2014; Taskin et al., 2012; Turhan et al., 2013). Nevertheless, to the best of our knowledge, only few research works have examined radioactivity levels of drinking water in Jordan so far (Al-Amir et al., 2012). Holding a total population of 1.77 million residents; Irbid is considered the second largest city in Jordan forming around 19% of the total population (Statistics, 2015). In Irbid governorate, tap water is used for drinking, cooking, and irrigating crops and it is primarily provided by near basins and highland aquifers. Consequently, an assessment of up-to-date water radioactivity levels is vital to retain the safe standards. In addition, standard data on the radioactivity of drinking tap water supplies for Irbid governorate is still unavailable and similar investigations are almost vacant; where, up to the moment, there is no systemic study on related possible radiation dose to the public (Al-Amir et al., 2012). In this work, two complementary techniques, viz. liquid scintillation counting and high-resolution gamma spectrometry, are utilized to assess radioactivity levels in water in all of the main districts of Irbid governorate. Existent radionuclides are identified, their concentration levels are determined, and their correlations with respect to each other and with respect to gross alpha/beta activity concentrations are examined. The calculated activity concentrations are used to evaluate the age-dependent annual effective radiation dose with lifetime risk assessment and are compared with the acceptable international limits.

2. Materials and methods

2.1. Samples collection and preservation

Twelve water samples were taken from the centers of the nine districts of Irbid governorate as shown in Fig. 1. As illustrated in Table 1, ten of these samples were collected directly from tap water from each district, one sample from a drilled well that stores tap water (Sample ID: S), and another one from a tank that collects rainwater (Sample ID: U).

The samples were collected using clean 20-liter sealed containers. Before filling the containers with water, the containers were washed with HNO_3 acid and rinsed with distilled water few times. The collected water was then transferred directly to Jordan Atomic Energy Commission's (JAEC) chemical preparation laboratory. Instantly, upon arrival to JAEC's chemical laboratory, to preserve the sample and prevent any solvents from precipitating on the containers inside walls, the samples were filtered and their acidity was slightly increased by adding diluted HNO_3 to all studied samples (Saleh et al., 2015).

2.2. The procedure for the measurement of gross alpha and beta

For gross alpha and beta activity measurement, the total evaporation method was applied as follows. A 100 *mL* of each sample was filtered with a filter paper and was evaporated using hotplate to dryness. The heating temperature was maintained to not exceed 80 °C, to prevent alpha & beta emitters from volatilizing and leading to underestimated readings (Jobbagy et al., 2014). In order to maintain the sample homogeneity and efficient heat transfer, all samples were stirred during heating using a magnet capsule. Then, the residues were completely dissolved and converted into a solution by adding 0.1M HNO_3. Reaching a volume of 5 *mL*, the samples were then transferred to 20 *mL* low-potassium glass liquid scintillation counter vials. The solution was then mixed with a 15 *mL* scintillation cocktail "Insta-Gel Plus" using a mixer machine (Maxi-Mix II, type 37600) for five minutes. For bubbles and gas removal from the solution, a (Hettich Rotina-48) centrifuge was used with 2000 RPM for five minutes. Finally, the samples were moved to the liquid scintillation counter (Perkin Elmer Tri-Carb 3170TR/LS) and stored for two hours before counting for 200 min to avoid photoluminescence (Montana et al., 2013).

The efficiency calibration for gross alpha and beta counts was determined by standard solutions that contain pure alpha and beta emitters with different pre-specified activities that were measured separately (Jobbagy et al., 2014). The misclassification study for finding the optimum pulse shape discriminator (PSD) level was conducted by calibrating with a set of standard solutions (L'Annunziata, 2003). The optimal discrimination setting for gross alpha/beta counting, where the percentage spillover of both counts is minimum, was found at the discrimination setting where the percentage spillover of alpha-into-beta and percentage spillover of beta-into-alpha curves intersect. Fig. 2 shows that the optimum discriminator setting value for counting for this study is 130 where both curves intersect.

Gross alpha/beta detection efficiency, at the optimum PSD setting, was calculated using the corresponding standard solution of pure alpha and beta emitters by the following relation:

$$\varepsilon = \frac{C_T - B}{A \times V \times 60} \times 100\% \quad (1)$$

where, ε is gross (alpha or beta) efficiency, C_T is corrected count rate (of alpha or beta in *CPM*), B is background count rate (in *CPM*), A refers to the pre-specified activity of standards (in Bq/L) and V refers to the volume of the standard solution (in L) and 60 is the conversion factor from *Bq* to *CPM*. In this work, alpha particles detection efficiency is $\sim$ 98.45% and beta particles detection efficiency is $\sim$ 97.9%. In addition, the minimum detectable activity (MDA) values at the 95% confidence level for gross alpha and beta measurements were determined using Currie's relation (Currie, 1968). For the current study, the MDA values for gross alpha and beta were found to be 82 and 216 mBq/L, respectively. Finally, gross alpha or beta activity (Bq/L) were calculated for all of the water samples by the following formula:

$$A_{\alpha \text{ or } \beta}(Bq/L) = \frac{\text{Net corrected count rate (CPM)}}{\varepsilon * V * 60} \quad (2)$$

2.3. The procedure of measurement for gamma activity

For gamma spectrometry, first, 10 L samples were filtered to remove the solids and other impurities, then 10 *mL* of (65–70) % HNO_3 were added to each sample and samples were left for evaporation using a hot plate with a constant temperature of 80 °C. The evaporation processes continued until a volume of 490 *mL* is reached. The samples are then transferred to 490 – *mL* Marinelli beakers. These beakers were filled fully and well-sealed with adhesive to prevent radon losses during thirty days storage to make sure that secular equilibrium is achieved between ^{226}Ra and ^{222}Rn. A high-resolution high-purity germanium (HPGe) detector (Ortec, GEM15p4 model) was used for gamma activity measurement with