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Radioactive uranium and cesium levels in Kuwait Bay seawater

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Abstract
The development of a continuous radioisotope monitoring program has an extreme significance in maintaining the marine environment's radiological safety. This study aimed to evaluate the radiological safety of locally sourced desalinated water and seafood. The establishment of baseline data for observing the nuclear activities already existing in the Gulf region was also targeted. While natural uranium radioisotopes are naturally occurring in the environment, there is a rising concern in Kuwait due to the depleted uranium (DU) presence after the Gulf War in 1991. The anthropogenic radionuclide (^{137}Cs) serves as among the most important indicator of the radioactivity releases. The present work focused on determining the concentration of ^{137}Cs and uranium isotopes in Kuwait Bay (KB) seawater. The concentrations of ^{137}Cs were determined by the ammonium molybdenum phosphate (AMP) co-precipitation method followed by gamma spectrometry measurement, whereas a radiochemical separation using anion exchange chromatography and alpha spectrometry measurement was used for uranium isotope determination. The average concentration levels of ^{235}U, (^{234}U and ^{238}U), and ^{137}Cs were 2.3 mBq l^{-1} ± 0.4, 45.3 mBq l^{-1} ± 3.5, and 1.24 mBq l^{-1} ± 0.5, respectively, and these figures are comparable to those reported worldwide. The concluded isotopic ratio of ^{235}U/^{238}U and ^{234}U/^{238}U indicated that the existing uranium in seawater of KB is only natural and no indication of the DU. The produced radiological data are considered as a beneficial baseline for monitoring the regional marine environment.

Keywords Radioisotopes · Depleted uranium AMP co-precipitation · Anion exchange · Isotopic ratio

Introduction

The continuous monitoring of radionuclides in marine water is a global demand for maintaining the radiological safety of the nearby areas. The periodic measurements of the environmental radioactivity are essential in detecting any variability in the exposure levels from the natural or anthropogenic sources that may cause an elevated radiation dose. The Arabian Gulf deemed to be among the most polluted marine environments around the world. Approximately one-third of the global oil is exported through the Strait of Hormuz (Al-Zamel et al. 2005). Since this sea passage is directly related to the key source of desalinated water in Kuwait, the verification of its radiological safety is significant in terms of economy and the nation's health. In addition, the determination of uranium

concentration in seawater is extremely important for Kuwait marine in order to ensure the absence of the depleted uranium (DU) resulted from weapons used during the Gulf War in 1991.

At present, almost every country in the Gulf area has either a civil nuclear program in progress or a project under consideration, which are expected to supply energy. In particular, the Islamic Republic of Iran launched its Bushehr nuclear power plant (BNPP) in 2011. The pre-operational nuclear power plants and associated radiological surveys were conducted in the UAE and KSA. Barakah NPP in the UAE will be operational by 2020 after some delay, and two nuclear reactors will be constructed by 2021 in KSA, which will increase to sixteen units by 2030 (Kovachev 2012). A number of environmental and research programs will be completed before the initiation and introduction of these nuclear power plants. Therefore, establishing the baseline radioactivity levels of the radionuclides such as ^{137}Cs in seawater can be utilized as an indicator of observing elevated levels caused by the near future nuclear activities in the region.

Different radiological studies in the marine environment have been conducted to assess the impact of different radionuclides in the marine ecology. Since natural and

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anthropogenic radionuclides are widespread in the marine areas, they interact with the marine environment through physical, chemical, and biological processes. Afterwards, they migrate and get concentrated with different levels in the marine ecosystem compartments, such as algae, seafood, and biota through water, food, and sediment. It was reported that the seafood consumption as a common dietary habit in some countries is the major contributor to the ingestion dose (UNSCEAR 1982; Carvalho et al. 2010).

Uranium natural radioisotopes exist in the earth's crust and the seawater with long half-lives (hundred thousand to billion years). Uranium exits naturally in the marine environment in the form of uranyl carbonate ion, which is easily soluble in seawater. Soluble uranium is transformed to oceans through river and streams in earth and surface water. It is classified as a high toxic threat to the public health if humans are exposed to it through ingestion (Carter and Burns 1993). Large quantities of DU were used during the Gulf War in 1991; therefore, the campaign of the International Atomic Energy Agency (IAEA) and the Kuwaiti government had evaluated the radiological consequences of the DU and found that there are no radiological health hazards (IAEA 2003), yet it is still a public concern. Therefore, the radiological monitoring of the marine environment will reduce the public fear. Different analytical methods have been applied for the detection of uranium, including laser-induced fluorescence spectrometry (LIFS), inductively coupled plasma (ICP) emission and mass spectrometry, and thermal ionization mass spectrometry (Huang et al. 2016; UNSCEAR 2000).

^{137}Cs is a common fission product in nuclear reactor, and it is regarded as a slightly toxic element decaying by gamma that can pose a significant and persistent threat due to its intermediate half-life of 30 years, while ^{134}Cs is a nuclear activation product with a half-life of only 2 years. Cesium is an electropositive alkaline soluble metal that is capable of contaminating large volumes of water following nuclear accidents. ^{137}Cs and ^{134}Cs are continuously produced by human nuclear activities through major sources, such as Chernobyl disaster in the 1980s, the nuclear test bombs in the North Hemisphere in the 1960s, and solid waste dumping at sea (U.S. National Library of Medicine 2005; Rudjord et al. 2001; Carvalho et al. 2010). The Baltic Sea was contaminated with high activity levels of the anthropogenic radionuclides (IAEA 2005). Up to this day, ^{137}Cs and other radionuclides are still detected in different compartments of the Baltic Sea due to the continuous recirculation from aquatic sediments and rivers input (Carvalho et al. 2010). ^{137}Cs is uptaken by cytoplasm of living cells similar to potassium (Konovalenko et al. 2017), and it accumulates in fishes through ingestion of food and water (Phillips and Russo 1978). For example, ^{137}Cs bio-accumulates in muscle tissues through extraction of ^{137}Cs from water, while seals are exposed to ^{137}Cs mainly through ingestion of prey and slightly through ingesting seawater (Saremi et al. 2018).

A reported persistent ^{137}Cs depositional fluxes were frequently observed in dust fallout in spring and northwestern and southwestern wind periods in Kuwait (Aba et al. 2018; Biegalski et al. 2001). The global atmospheric fallout and the long-range transportation of mainly northwest dust origins primarily condition the presence of ^{137}Cs in Kuwait marine area. Consistent with a similar situation in many countries, the nuclear bomb-test experiments in the North Hemisphere as well as Chernobyl disaster specify ^{137}Cs deposition. High amount of ^{137}Cs-loaded dust was transported to the Gulf region from Europe, Turkey, Syria, and Iraq through the dominant northwestern wind. Aba et al. (2014) reported two distinct peaks related to both nuclear events of ^{137}Cs have been observed in the sediment profile of Kuwait. There are various methods to identify cesium in seawater, where the two most commonly used methods are cesium adsorption of ammonium molybdenum phosphate (AMP) and co-precipitation with various insoluble hexacyanoferrates (II) (Kamenik et al. 2013). These adsorption methods are associated with great promise to timely reveal contamination near nuclear facilities.

The regional Gulf levels of ^{137}Cs concentrations were assessed previously in Kuwait and Qatar. ^{137}Cs was 1.05 mBq $l^{-1} \pm 0.01$ in 2012 and 1.34 mBq $l^{-1} \pm 0.16$ in 2015 in Kuwait marine water (Uddin et al. 2015), and the total average activity in Qatar marine water was 1.59 Bq/m$^3 \pm 0.38$ in both years of 2011 and 2012 (Al-Qaradawi et al. 2015). ^{238}U was 0.040 mBq $l^{-1} \pm 0.005$, and ^{234}U was 0.054 mBq $l^{-1} \pm 0.006$ in one location of KB (Uddin et al. 2012). The significance of this study is related to the shortage in the available radiological data of radionuclide levels in the local seawater.

Study area

This study involves the KB marine water, a partial small bay of the Arabian Gulf (AG) situated in the eastern side of the country. Figure 1 shows the sampling area in KB presented in the study. The study area lies at the northwest corner of the AG, which forms the Gulf of Kuwait, locally known as the KB and the area nearby Bobiyan and Failaka islands. KB is a marginal sea of the Indian Ocean that lies between the Arabian Peninsula and southwestern Iran. The KB covers an area of 750 km^2, and the water depth varies between 3 and 10 m with an average depth of about 5 m (Al-Shemmari et al. 2002). It hosts the major port, namely, Al-Shuwaikh Port and three power and desalination plants: Doha east, Doha west, and Subiya. The KB and the adjacent area are characterized by a shallow water throughput with sediment movements by intertidal mudflats. Al-Shamal, the dominant northwesterly wind, severely affects the northern area of the AG, shaping the regional dust sources (Khalaf and Al-Hashash 1983). However, the dry land of the southern marshes in Iraq is considered a significant source of dust that founders over the northern part
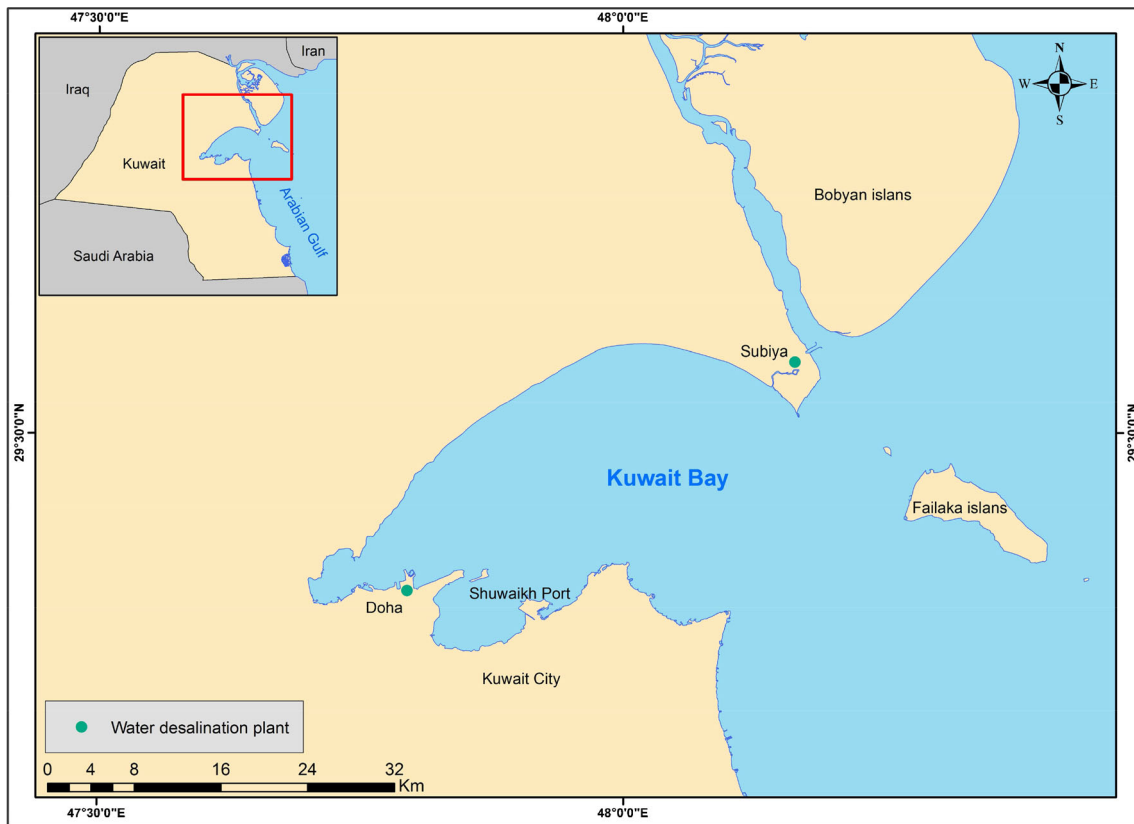
Fig. 1 The Kuwaiti Bay map

of the AG (Al-Ghadban et al. 1999). Two major industries are located closely in KB region, BNPP in Iran and water desalination plant in KB known as the main water source in Kuwait. Distance between the two industrial plants is about 200 km compared with 800 km from the capital of Iran. The significance of ^{137}Cs monitoring in KB is related to the potential hazard linked with any intentional or unintentional radioactive releases from BNPP and their recirculation in the regional biosphere that can reach the water desalination plant in KB.

Materials and method

Sampling and sample preparation

A cruise was utilized to collect ten high-volume marine water samples, 60 l each, at 1-m depth from the offshore surface of KB. The samples were collected by pumping seawater into labeled plastic containers. The preservation of samples was ensured by acidifying them with concentrated hydrochloric acid and storage in plastic containers for further processing in the laboratory.

Sample analysis

The determination of uranium by a radiochemistry method includes the adsorption of uranium onto 9-N hydrochloric acid

anion exchanger. The amount of ^{232}U (200 mBq) is added as a radiotracer at the beginning of mineralization. Uranium is directly extracted from resin by passing 0.5-N hydrochloric acid-hydroxyl ammonium chloride solution through the anion exchanger (Monroy-Guzmán 2016). Alpha sample sources are prepared by NdF3 co-precipitation method using 0.1-μm propylene 25-mm-diameter resolve filters. The prepared alpha sources are counted using alpha spectrometry. Figure 2 presents the procedure followed for uranium analysis.

The ^{137}Cs analysis is approached by the adsorption of cesium into ammonium molybdenum phosphate (AMP) (2 g/10 l) and adding some amounts of stable cesium as a carrier, cesium chloride (CsCl), (10 ml of 25 mg/ml solution). Cs-AMP is separated from the sample by gravitational settling and centrifuge of 4 h; then, the precipitation is collected, dried, and weighted followed by ultra-low background gamma spectrometry measurement (Bajoga et al. 2015; Dovelte and Povince 2004). Figure 3 shows the procedure used for cesium analysis.

Instrumentation and measurements

The prepared uranium samples were measured by alpha spectrometry system (Apex, Canberra) equipped with 450-mm^2 passive implanted planar silicon (PIPS) detectors. The detectors' resolution is about 20–24 keV at 4586 keV, and the detection efficiency at 5 mm (2nd shelf) is about 15%. It is well
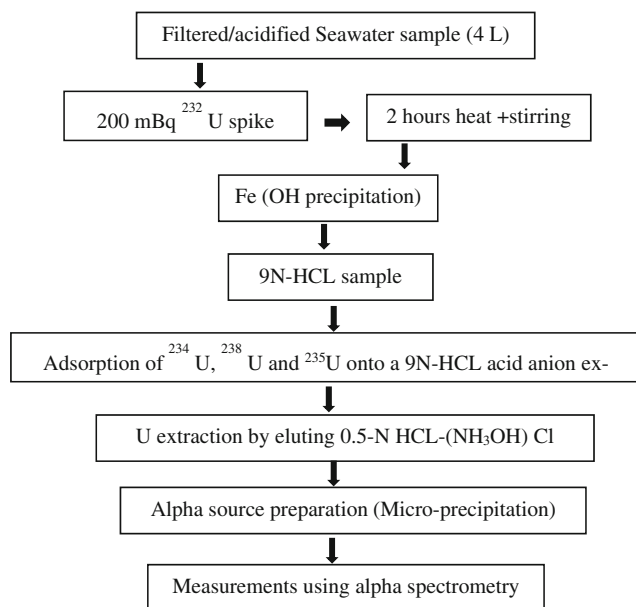
Filtered/acidified Seawater sample (4 L)

200 mBq ^{232}U spike → 2 hours heat +stirring

Fe (OH precipitation)

9N-HCL sample

Adsorption of ^{234}U, ^{238}U and ^{235}U onto a 9N-HCL acid anion ex-

U extraction by eluting 0.5-N HCL-(NH$_3$OH) Cl

Alpha source preparation (Micro-precipitation)

Measurements using alpha spectrometry

Fig. 2 Procedure followed for the uranium analysis

known that ICP-MS is more accurate and sensitive in measuring low radioactivity levels of ^{235}U (Rusconi et al. 2004; Zheng et al. 2006). Thus, samples were counted for a long time (4–5 days) in order to obtain less statistical error of small net peak area of ^{235}U, where it reached less than 20% after subtracting the background. The uncertainty of the acquired uranium ratios was calculated based on the error propagation of ^{235}U, ^{234}U, and ^{238}U with taking into consideration the efficiency, uncertainty of measurements, and chemical recovery. The calculated minimal detectable activity limit for uranium was approximately 0.3 mBq l^{-1}.
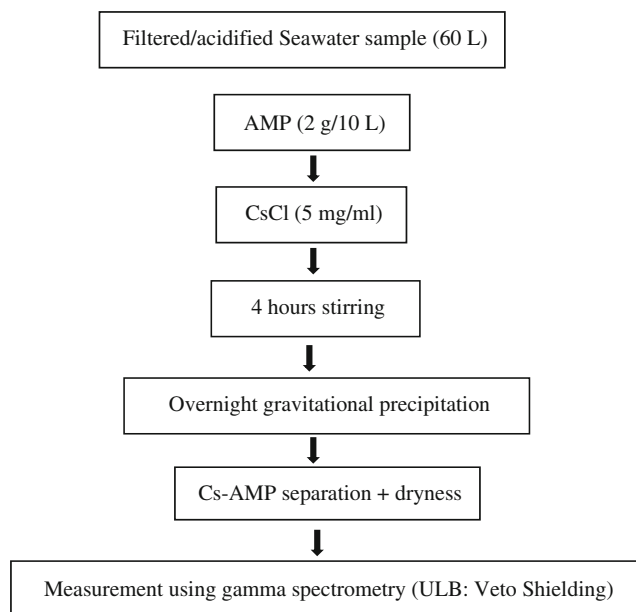
Filtered/acidified Seawater sample (60 L)

AMP (2 g/10 L)

CsCl (5 mg/ml)

4 hours stirring

Overnight gravitational precipitation

Cs-AMP separation + dryness

Measurement using gamma spectrometry (ULB: Veto Shielding)

Fig. 3 Procedure followed for the cesium analysis

The ultra-low background gamma spectrometry system, supplied with a high-purity germanium detector and broad energy type, was employed to determine the radioactivity of ^{137}Cs. The system is linked to an anti-cosmic active Veto shielding to reduce the background by increasing signal to noise ratio revealing very low-level concentration of ^{137}Cs. A standard solution containing a known amount of ^{137}Cs radionuclide was similarly prepared for efficiency calibration. The precipitate was filled in a small petri-dish—5 cm in diameter. The prepared samples were counted for 3 days to reach good statistics area at 661.6 keV (uncertainty less than 10%). The gamma spectra were analyzed by Genie 2000 software (Canberra Inc.) after background subtraction, and the calculated detection limit for ^{137}Cs was about 0.3 mBq l^{-1}.

Activity concentrations and chemical recovery determination

A blank sample was prepared with the same co-precipitation and separation methods for each radionuclide. This step was critical in considering the background contribution in the measured samples. The blank samples of each radionuclide were used as the background and subtracted from the sample's spectra. The specific activity concentration of gamma radionuclide ^{137}Cs was calculated using Eq. (1) (Dovelte and Povince 2004).

$$A = \frac{C_n}{\varepsilon_f P_\gamma t_s m} \tag{1}$$

where A is the activity concentration of a radionuclide in becquerel unit, C_n is net count (background subtracted) of the corresponding full energy peak areas, ε_f is the full energy peak detection efficiency, P_γ is the emission probability per decay corresponding to the specific gamma-ray energy, t_s is the counting time in term of seconds, and m is the mass of water samples in liter (l). Specific activity concentration of alpha radionuclides (^{235}U, ^{234}U, and ^{238}U) was calculated using Eq. (2) (IAEA 2009):

$$A = \frac{a_T . m_T . r_n}{Vs . r_{nt}} f_T \tag{2}$$

where A is the activity concentration of a natural uranium radionuclide in becquerel per liter unit, a_T is the specific activity of a tracer (Bq g^{-1}), m_T is the mass of the tracer (g), r_n is the net count rate of the natural radionuclide (background subtracted), Vs is the volume of the sample (l); r_{nt} is the net count rate of the tracer (background subtracted), and f_T is the decay correction factor of a tracer between calibration date and measurement date. The decay of ^{235}U, ^{234}U, and ^{238}U between separation date and measurement date is negligible because of the long half-lives of the three radioisotopes. The associated uncertainty (u) of the activity concentration of radionuclides was calculated using the error propagation Eq. (3):

$$u(A) = \sqrt{\left(\frac{u(a_T)}{a_T}\right)^2 + \left(\frac{u(r_n)}{r_n}\right)^2 + \left(\frac{u(r_{nt})}{r_{nt}}\right)^2 + \left(\frac{u(V_s)}{V_s}\right)^2 + \left(\frac{u(m_t)}{m_t}\right)^2 + \left(\frac{u(f_T)}{f_T}\right)^2} \tag{3}$$

The minimum detectable activity (MDA)

$$MDA = \frac{\frac{2.71}{T_B} + 4.65\sqrt{R_B}}{E.V.I_\gamma} \tag{4}$$

The MDA was calculated by using the Curri formula, where T_B is the time of background counting, R_B is the count rate of spectroscopy background, E is the detector counting efficiency percentage, V is the sample volume, and I_γ is the gamma emission intensity percentage. Chemical recovery (CR%) was calculated for both analytical procedures to ensure the minimal loss of tracer/carrier activity through the analysis.

$$CR\% = \frac{netcps/E}{dpsadded}.100 \tag{5}$$

$$CR\% = \frac{amountofAMPrecoveredafterseperation}{originalamountofAMPadded}.100 \tag{6}$$

Equation (5) was applied in calculating ^{232}U tracer chemical recovery percentage, where *net cps* is the net count per seconds in the spectrum at peak of 5320 keV, *dps* is the disintegration per second of the added tracer activity, and E is the detection efficiency of the spectrometry (Fadzilah Yousf et al. 2016). CR% of uranium analysis was 95–100% indicating that most of the uranium was excreted from the sample. Equation (6) was applied in calculating cesium-AMP chemical rec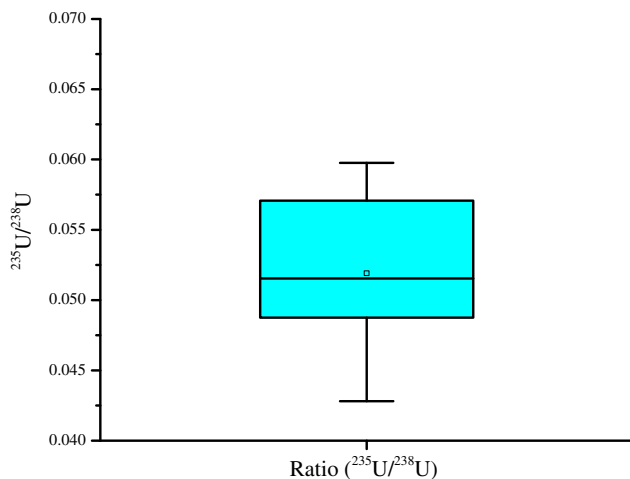overy percentage. A repeated co-precipitation was carried out to confirm the completion of ^{137}Cs scavenging from the large seawater samples, and the sample was measured to indicate any remaining ^{137}Cs. The test showed that almost 100% of ^{137}Cs was removed from the sample solution indicating the full extraction of ^{137}Cs with minimal loss.

Quality assurance

The certified reference material prepared by IAEA containing known amounts of uranium and ^{137}Cs, coded as IAEA-Irish Sea water-443, was repeatedly analyzed along with the samples to maintain the quality of the analytical results and to check the performance of the spectrometry. The calculated results of reference material analysis were in an analogous agreement with the certified values.

Results and discussion

The obtained results of ^{235}U and (^{234}U, ^{238}U) average activity concentrations were assessed to be 2.3 mBq $l^{-1} \pm 0.4$ and 45.3 mBq $l^{-1} \pm 3.5$. ^{235}U value was almost the same as the standard average value reported in the oceans (2 mBq l^{-1} and 1.8 mBq l^{-1}) (Kusakabe 2017; Harrison 2001). Whereas ^{234}U and ^{238}U average concentrations are comparable to the reported average content of the worldwide seawater (42 mBq l^{-1} ^{238}U, 40 mBq l^{-1} ^{238}U, and 50 mBq l^{-1} ^{234}U) (Monty 2001; Harrison 2001). These values are also in an agreement with
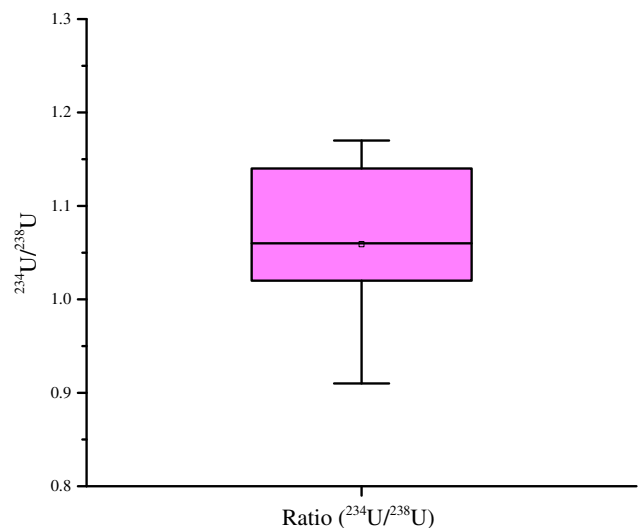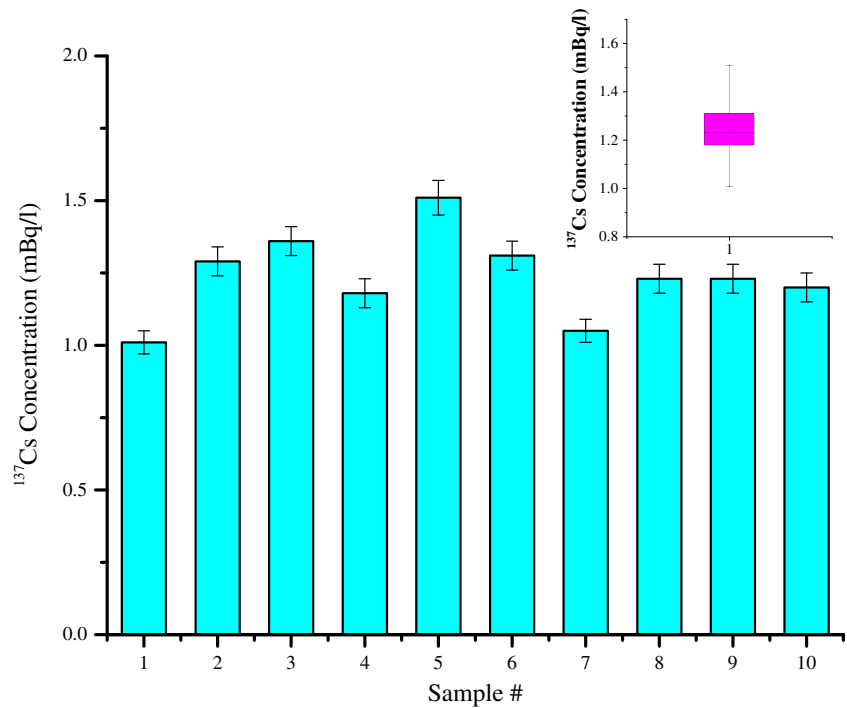

Fig. 4 ^{235}U/^{238}U isotopic ratio

Fig. 5 ^{234}U/^{238}U isotopic ratio

Fig. 6 ^{137}Cs average activity concentration (mBq l^{-1}) in KB water

uranium concentration ranges in eastern Mediterranean sea-water in Lebanon and Syria, and they were 44 to 68 mBq l^{-1} and 36.9 to 44.5 mBq l^{-1} respectively (ElSamad 2010; Mamish et al. 2015).

^{235}U/^{238}U isotopic ratio has been calculated to investigate the presence of DU in Kuwait marine water, and it was found to be 0.054 ± 0.03 as shown in Fig. 4. This ratio is comparable to the natural isotopic ratio of ^{235}U/^{238}U in seawater (0.047 ±

0.013 and 0.046), and it is extremely distant from the depleted uranium ratio (0.013) (Rusconi et al. 2004; Steele et al. 2009).

To verify this finding, the isotopic ratio of ^{234}U/^{238}U was calculated and found to be 1.07 ± 0.1 as shown in Fig. 5. It confirms the ratio of the natural uranium in Kuwait marine water, which is 0.935, while DU isotopic ratio is 0.185 (WHO 2001; Rusconi et al. 2004). It should be noted that average isotopic ratio ^{234}U/^{238}U is slightly higher than a unity due to the radionuclide

Table 1 ^{137}Cs worldwide average values in marine water

Location	^{137}Cs average concentration (mBq l^{-1})	Reference	Year
Kuwait Bay	1.24 ± 0.5	Current study	
Kuwait Bay	1.34 ± 0.16	Uddin et al. (2015)	2015
Kuwait Bay	1.05 ± 0.1	Uddin et al. (2015)	2012
Qatar coast	1.6 ± 0.38	Al-Qaradawi et al. (2015)	2015
Arabian Sea	1.6 ± 0.3	IAEA (2005)	2000
North Pacific	2.4 ± 0.3	IAEA (2005)	2000
Equatorial Pacific	2.1 ± 0.3	IAEA (2005)	2000
South Pacific	1.3 ± 0.5	IAEA (2005)	2000
Sea of Japan	2.8 ± 0.5	IAEA (2005)	2000
Indian Ocean	2.1 ± 0.3	IAEA (2005)	2000
Southern Ocean	1.0 ± 0.6	IAEA (2005)	2000
Mediterranean Sea	2.6 ± 0.4	IAEA (2005)	2000
North Atlantic	1.7 ± 0.8	IAEA (2005)	2000
Central Atlantic	1.4 ± 0.2	IAEA (2005)	2000
South Atlantic	0.6 ± 0.1	IAEA (2005)	2000
Brazilian Seawater		Cunha et al. (1993)	1993
Location 1	1.7		
Location 2	1.9		

recoil process during alpha decay of ^{238}U (Chabaux et al. 2001). This near unity value of the ^{234}U/^{238}U ratio specifies the radioactive equilibrium of uranium that is consistent with the average ^{234}U/^{238}U ratio recorded of seawater for 1.144 ± 0.002 (Steele et al. 2009), and the one found in Western North Pacific (1.09 ± 0.05) (Miyake et al. 1966). Clearly, both ^{235}U/^{238}U and ^{234}U/^{238}U ratios demonstrate that there is no anthropogenic source other than the naturally provided, which reflects the negligibility of the DU impact.

^{137}Cs calculated radioactivity in KB seawater varied between 1.0 and 1.51 mBq l^{-1} with an average value of 1.24 mBq l^{-1} ± 0.5 as presented in Fig. 6. The obtained results indicate the location, deposition rate, and sources of cesium precipitated in the area. This range of ^{137}Cs concentration is comparable to those reported in different locations of worldwide marine (Table 1). It should be noted that ^{134}Cs was not detected, and it declined below the MDA due to its short half-life. Since there is an absence of the regional nuclear incidents, the low data of ^{137}Cs in KB can be attributed to the indirect impact of ^{137}Cs fallout and its hypothetical deposition in the Gulf area that is resulted from the high release of the Northern Hemisphere bomb test (UNSCEAR 2000).

Conclusion

This study investigated the specific activity concentrations of radioactive uranium and cesium in Kuwaiti marine water. It was concluded that the obtained specific activity concentrations were relatively low and equal to the levels of provincial marine environment. The calculated ^{235}U/^{238}U and ^{234}U/^{238}U isotopic ratios confirm that the impact of the Gulf war is insignificant and considered negligible. It was determined that the major source of ^{137}Cs found in KB water is the transported ^{137}Cs-loaded dust through the northwesterly wind. Ultimately, the overall generated low-level data for uranium and cesium reflect the low radiological risk of Kuwait marine environment. The present data can be utilized as a baseline within the national continuous marine radiation-monitoring program of KB to observe the temporal radiation levels. Further studies will be applied to investigate selected endemic biota known as ^{137}Cs bio-accumulators.

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