LEVELS AT THE AL-KHAFJI AND MNEEFA COASTAL AREAS IN SAUDI ARABIA

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ABSTRACT

This work is a part of a project conducted by the Institute of Atomic Energy Research (IAER) at KACST which aims to measure marine radioactivity near the Saudi coasts of the Arabian Gulf and the Red Sea and to establish " Saudi Marine Radioactivity Database" for both naturally occurring as well as man-made radionuclides in water, sediments and biota. This database will be used as a reference to gauge any inputs and transboundary radioactive releases in the future. Sediment samples were collected at depths ranging from 5 up to 25 meters from 10 locations covering about 125 km of the gulf coast at Al-khafji and Mneefa areas. Concentrations of ^{40}K, ^{238}U, ^{235}U, ^{230}Th, ^{226}Ra, ^{232}Th, ^{228}Th, and ^{228}Ra radionuclides in both sediment and water samples have been determined. The ranges of activities observed for ^{238}U and ^{232}Th were 27.78-33.94 Bq/kg and 0.19-0.95 Bq/kg from Al-Khafji and 17.09-39.76 Bq/kg and 0.15-0.82 Bq/kg dry weight from Mneefa respectively. Activity concentrations of ^{226}Ra, ^{228}Ra, and ^{40}K,for both areas, depend on the nature and type of sediment. Concentrations of radionuclides in water samples do not show remarkable variation with sample location. The obtained results are compared with those from coastal waters of other oceans.

Keywords: *marine radioactivity, ^{232}Th, ^{238}U, Al-Khafji, Mneefa, sediment, natural*

الملخص

هذا العمل جزء من مشروع يقوم به معهد بحوث الطاقة الذرية بمدينة الملك عبدالعزيز للعلوم والتقنية ويهـدف إلـى قياس النشاط الإشعاعي البحري القريب من الشواطئ السعودية للخليج العربي والبحر الأحمر ، ويهـدف أيضـا إلـى تأسيس قاعدة بيانات للإشعاعات البحرية للبيئة السعودية للنويدات الطبيعية والصنعية في المياه والرسـوبيات والأحيـاء المائية. هذه القاعدة ستستخدم كمرجع لقياس أي تسربات إشعاعية في المستقبل. وقد تم جمع عينات بأعمـاق تتـراوح مابين ٥ إلى ٢٥ مترا من عشر مناطق تغطي مسافة قدرها ١٢٥ كلم من شاطئ الخليج العربي في كل مـن الخفجـي ومنيفـة. تـم قياس تراكيـز البوتاسـيوم-٤٠، واليورانيـوم-٢٣٨، ٢٣٥، والثوريـوم –٢٣٠، والراديـوم-٢٢٦، والثوريوم-٢٣٢، الثوريوم-٢٢٨ والراديوم-٢٢٨ في الرسوبيات والميـاه . كان النشـاط الإشعاعي لليورانيوم-٢٣٨ والثوريوم-٢٣٢ من ٧٨و٢٧ – ٩٤و٣٣ بيكرل/كجم و من ١٩و. – ٩٥و. بيكرل/كجم على التوالي لمنطقة الخفجي ، وتـراوح في منيفـة من ٠٩و١٧ – ٧٦و٣٩ بيكرل/كجم لليورانيـوم-٢٣٨ و مـن ١٥و٠ – ٨٢و٠ بيكـرل/كجـم للثوريوم-٢٣٢. التراكيز الإشعاعية للراديوم-٢٢٦ والراديوم-٢٢٨ والبوتاسيوم-٤٠ للخفجي ومنيفة تعتمد على طبيعة ونوع الرسوبيات أما تراكيز النويدات في عينات المياه فقد كانت متقاربة. وقد قورنت النتائج التي تم الحصـول عليهـا بنتائج أخرى من المياه الساحلية للمحيطات.

INTRODUCTION

The Arabian Gulf constitutes almost entirely closed body of shallow water. It extends northwest about 965 km from the Strait of Hormuz to Shatt Al-Arab by 300 km wide, with an average depth ranges from 35 to 100 m at most. It is one of the most productive plankton water bodies in the world, but it is also regarded as being one of the most fragile and vulnerable marine ecosystems. Its low tidal displacement means that it has little discharge of its water into the Indian Ocean and thus little opportunity to flush out pollutants. Al-Khafji is a small town located about 80 km south of Saudi Kuwaiti borders while Mneefa is a small village located about 125 km south of Al-Kafji on the western bank of the Arabian Gulf. Our world is radioactive and has been since it was created. Over 60 radionuclides can be found in nature, and they can be placed in three general categories: primordial from before the creation of the earth; cosmogenic formed as a result of cosmic ray interactions; human produced - enhanced or formed due to human actions (minor amounts compared to natural). Radionuclides are found naturally in air, water and soil. They are even found in us, being that we are products of our environment. Every day, we ingest and inhale radionuclides with air and food and the water. Natural radioactivity is common in the rocks and soils that make up our planet, in water and oceans, and in our building materials and homes. There is no place on earth that you can not find natural radioactivity.

There is a considerable number of literatures on the concentration of naturally-occurring radionuclides in marine environment, especially marine sediments and bio-organisms from different regions in the world [Joshi, L. U., Ganguly, A. K. 1976, Koide, M. et al., 1973, McCartney, M. et al, 1992, McDonald, P., et al 1991, Moreno J. et al 1997, Nedwell, D., et al, 1993]. However, the vast majority of marine investigations on radioactivity concentrations were conducted on marine environment surrounded by developed countries. It has to be noted that no investigations have been carried out regarding marine environmental radioactivity in the Arabian Gulf.

The Kingdom of Saudi Arabia has more than 750 kilometers of coastline along the Arabian Gulf. This coastline is of great economic, recreational, and tourist values for the country. For water resources, the desalination plants located on the Arabian Gulf play an important rule in water supply for the Kingdom so any contamination especially radioactive one may affect the water quality as well as the people health. It is quite important to keep this gulf free of any radioactive pollutants beside the fact that lack of information concerning marine environmental radioactivity impairs the proper judgment on any possible contamination in the future. Development of national database for marine radioactivity is essential for determining unequivocally any future contamination. Naturally –occurring radioisotopes such as ^{40}K, uranium series namely:^{238}U, ^{235}U, ^{230}Th, ^{226}Ra, and thorium series namely:^{232}Th, ^{228}Th, and ^{228}Ra were studied in Al-Khafji and Mneefa coastal areas.

Fig.1 Map showing the locations of Al-Khafji and Mneefa.

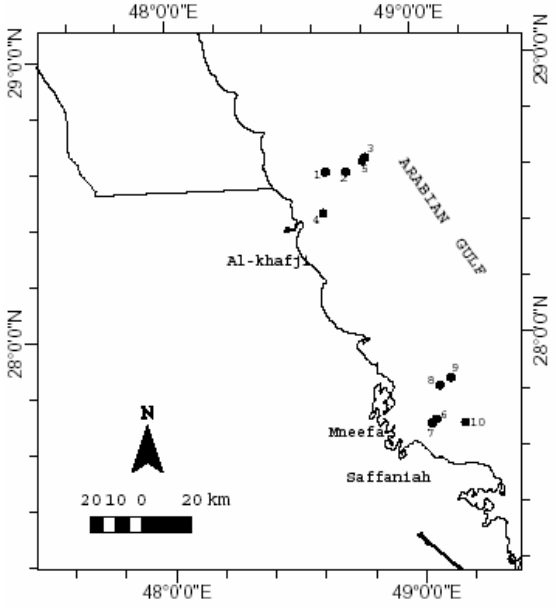

Fig.2. Map showing the Al-Khafji and Mneefa locations on the Arabian Gulf and the locations of the collected samples.

METHODOLOGY

A total of 10 samples of surface marine sediments as well as surface sea water were collected in May 2001 from 10 locations covering about 125 km of the gulf coast at Alkhafji and Mneefa regions as shown in Fig.1 and Fig.2. Locations of the collected samples were determined using a global positioning system (GARMIN GPS 100 SRVY II) which is a satellite based positioning and navigation system that provides precise position, velocity and time information. The uncertainty is ± 1.8 meter. Thus, for future reference, all collection sites can be predetermined by using the receiver's computed position to navigate to a location whose coordinates have been previously entered. For Al-Khafji and Mneefa, A sediment sample was collected at a water depth ranging from 0 to 0.75 meter while other samples at depths ranging from 5 up to 25 meters. Due to the limited depth near the coast, which is a characteristic feature of the Arabian Gulf, samples were collected up to 7 kilometers from the shore. In tidal areas with shallow water sediment sampling was conducted by hand with assistance of a semi-cylindrical metallic spade, while deep-water bottom sediment samples were collected using 7 liters grap sampler. Sediment samples were collected in polyethylene bags and transported to the laboratory at IAER in Riyadh. Samples were air-dried in porcelain or aluminum dishes at air temperature ranging fom 32 up to 42 ◦C and then oven dried at 95◦C. The coarse grained sediments and sponge debris were screened by a 2-mm mesh sieve. The fine grained sediment samples were packed in 0.5 liter marinelli beaker which was hermetically sealed and left for four weeks to attain secular equilibrium of the daughter ^{214}Bi and ^{214} Po radionuclides with their ^{226} Ra grand parent. After that, samples were subjected to activity measurements.

Concentrations of ^{238}U, ^{235}U, ^{226} Ra , ^{228} Ra, and ^{40}K radionuclides in both sediment and water samples have been determined using a high resolution gamma-ray spectrometers based on a hyperpure germanium detector with relative efficiency ranging from 20 up to 40% and 1.90 KeV energy resolution for the 1332 KeV gamma ray line of ^{60}Co. Measured concentrations of these radionuclides in sediment samples show wide variations starting from less than Becquerel/kg dry weight (about 0.05 Bq/kg) up to few hundreds of Becquerel/kg depending on the radionuclide as well as on the nature and type of the sediment. Concentrations of radionuclides in water samples do not show remarkable variation with the sample location.

Energy and efficiency calibration of the three used hyperpure germanium spectrometers in the energy range from 60 up to 1500 KeV were determined using a set of standard sources which are ^{133}Ba, ^{152}Eu, ^{154}Eu, ^{241}Am, and ^{226}Ra in the same geometrical configuration in which all sediments and water samples were prepared. All these sources were distributed homogeneously in the same 0.5 liter marinelli beakers.

The given concentrations were calculated using the gamma ray spectra emitted from the studied sediment samples, which were collected for a time period of about 24 hours. The activity of ^{40}K was determined using 1460.8 KeV photo peak . The 295.21, 351.92, and

609.31, KeV photopeaks resulting from the decay of daughter ^{214}Pb and ^{214}Bi radionuclides were used to determine the activity of ^{226}Ra. While the activity of ^{228}Ra were calculated from 338.40 and 911.07 KeV photopeaks respectively. For ^{238}U, a completely isolated 63.29 KeV photopeak, which comes from the decay of ^{234}Th, was used to find the concentrations throughout the collected samples. The 185.7 KeV gamma-ray line resulting from the decay of ^{235}U was not used to determine the ^{235}U activity since it is not resolved from the 186.21 KeV photo peak resulting from the decay of ^{226}Ra to ^{222}Rn, and the 143 KeV gamma line was used for ^{235}U activity determination despite the fact that it has a relatively low intensity. The relative error of the measured concentrations ranged from 5 to 18% for different radionuclides and different concentrations with tendency to increase as the concentration decreases.

Naturally occurring ^{232}Th and its grand daughter ^{228}Th as well as ^{230}Th, arising from the uranium series, were determined using radiochemical separation and a silicon surface barrier detector for alpha spectrometry. For the digestion of thorium, three grams (in 6 portions of 0.5 gm each) of the ashed soil samples were digested using $HCl/HNO_3/HF$ digesting mixture. The digested portions were combined, spiked with ^{229}Th radiotracers and evaporated to dryness. The residue was converted to nitrate form by adding 5 ml of conc. nitric acid and evaporated to dryness. The later process is repeated 2 times to ensure complete conversion to nitrate form. The sample was dissolved in 8 M HNO_3 and filtered using glass filler filters to be prepared for analytical procedure. Water samples were evaporated to dryness and the residue is converted to nitrate form and treated as in soil samples. For separation of thorium, the sample in 8 M HNO_3 solution was loaded into a small column (10 ml from Bio-Rad) and packed with (AG 1-8Y) anion exchanger, Cl-form and preconditioned with 20 ml of 8M HNO_3 to separate thorium. Possible Th was stripped with 20 ml of 1M HNO_3 solution. For thorium source separation to the strip solution, 100 μl of 70% $NaHSO_4$ was added and the solution was evaporated to dryness. HNO_3 was added and the sample solution was evaporated again to destroy any organic materials that may be eluted from the resin. The residue is taken up in 20 ml of 1M HNO_3 and thorium was precipitated as fluoride on a membrane filter and counted using high resolution α-spectrometer.

The specific activity (SA) was calculated according to the following equation

$$S.A.\ (Bq/kg) = C / \eta \,.\, f \,.\, w$$

Where

C : the net counts per second under the photopeak area of interest.

η : The photopeak efficiency in the beaker-detector fixed configuration.

f : a fraction representing number of photons with the certain energy per one decay.

w : the sample dry weight in kg

RESULTS AND DISCUSSION

The activity concentrations of uranium series radionuclides, namely ^{238}U, ^{235}U, ^{230}Th, and ^{226}Ra of the Al-Khafji and Mneefa (in Bq/kg) are presented in Table 1. Most of specific activities were rounded to two digits. Latitude and longitude of each sample location as well as each sample depth are indicated in the same table. Absolute statistical and systematical errors are also shown for each measured concentration. Samples number 1,2,3,4 and 5 were collected from Al-Khafji while samples 6,7,8,9,10 were collected from Mneefa. The activity concentrations of ^{238}U for Al-Khafji were 27.78- 33.94 Bq/kg with an overall mean of 30.33 ± 2.61 Bq/kg whereas, the ^{238}U activity concentrations for Mneefa were 17.09-39.76 Bq/kg with an overall mean of 30.51 ± 9.91 Bq/kg. The proximity of uranium concentrations of both areas can be attributed to the same nature of sediments, in other words, the physiochemical composition of samples collected are almost the same. The activity concentrations of ^{226}Ra for Al-Khafji and Mneefa were 1.99-4.52 Bq/kg and 3.94-9.46 Bq/kg respectively with an average mean of 2.84 ± 0.98 and 6.77 ± 2.43 Bq/kg respectively. For grand daughter of uranium,^{230}Th, the activity fall within the range of 0.71-1.83 Bq/kg and 0.42- 1.72 Bq/kg for Al-Khafji and Mneefa respectively. Uranium activity concentrations of all samples listed in Table 1 are accepted and fall within the international limits.

The concentrations of ^{40}K as well as thorium series radionuclides namely ^{232}Th, , ^{228}Th, and ^{228}Ra at Al-Khafji and Mneefa coastal areas are presented in Table 2. The activity concentrations of ^{232}Th were 0.19-0.95 Bq/kg and 0.15-0.82 Bq/kg for Al-Khafji and Mneefa respectively. The activity concentrations of ^{40}K were 23.69 – 52.16 and 113.9 – 253.3 Bq/kg for Al-Khafji and Mneefa coastal areas respectively which fall within the acceptable limits and are considered to be fairly low if compared with the range of 23.7 – 515 Bq/kg of the Red Sea while the activity concentrations of ^{228}Ra fall within the range of 1.25-2.17 Bq/kg.

The obtained concentrations of ^{238}U, ^{235}U, and ^{40}K are comparable with those values obtained for the fringing reefs of Port Sudan on the western coast of the red sea [Sam, A. K., et al , 1998] and with the activities obtained for the marine sediments from Ghazaouet Bay on the western coast of Algeria and from other world marine areas [Noureddine, A. and Baggoura, B., 1997].

The ^{235}U concentrations vary from 1.30 – 1.65 and 0.88-1.45 Bq/kg with an overall mean of 1.44 ± 0.174 and 1.16 ± 0.21 Bq/kg respectively. From the obtained results, one can not draw any conclusion concerning the change of activity concentration as a function of sediment depth within the too limited depths included in this study, since the sediment depth at the western coast of the gulf is generally too limited, even at large distances from the shore. It has to be mentioned that comparison between ^{235}U and ^{238}U average concentrations for both The Al-Khafji and Mneefa areas indicates good agreement with natural relative abundance of these two isotopes belonging to two different radioactive series. The concentrations of ^{226}Ra

and ^{228}Ra are higher for all locations of Mneefa region in comparison with those of the Khafji region. Moreover, it should be mentioned that there is a clear disequilibrium between ^{226}Ra and its grand parent ^{238}U for both Al-Khafji and Mneefa coastal areas.

Table 1 Activity concentrations of uranium series (Bq/kg) of Al-Khafji and Mneefa

Sample #	Lat. & Long.	Depth, m	^{238}U	^{235}U	^{230}Th	^{226}Ra
1	28 25 625N 48 32 388E	S	31.59 ± 2.09	1.60 ± 0.20	1.30 ± 0.15	2.79 ± 0.18
2	28 25 625N 48 32 688E	8	33.94 ± 2.33	1.65 ± 0.21	0.71 ± 0.14	2.61 ± 0.20
3	28 25 798N 48 32 977E	12	30.50 ± 2.31	1.33 ± 0.20	1.83 ± 0.13	4.52 ± 0.23
4	28 26 035N 48 33 286E	18	27.84 ± 2.50	1.30 ± 0.20	1.79 ± 0.14	2.31 ± 0.21
5	28 25 754N 48 33 872E	25	27.78 ± 2.50	1.30 ± 0.20	0.76 ± 0.11	1.99 ± 0.19
6	27 35 412N 48 54 574E	S	17.09 ± 1.99	0.88 ± 0.17	1.16 ± 0.15	3.94 ± 0.22
7	27 35 364N 48 54 519E	5	34.59 ± 2.33	1.20 ± 0.20	1.72 ± 0.14	7.53 ± 0.31
8	27 35 850N 48 54 638E	10	23.11 ± 2.11	1.05 ± 0.19	0.94 ± 0.12	4.52 ± 0.23
9	27 35 939N 48 54 800E	14	39.76 ± 2.86	1.23 ± 0.20	1.70 ± 0.15	9.46 ± 0.35
10	27 36 297N 48 54 996E	15	37.98 ± 2.74	1.45 ± 0.20	0.42 ± 0.08	8.42 ± 0.33

The activity concentrations of ^{232}Th, and ^{228}Th for Al-Khafji were 0.19 – 0.95 and 0.23 – 1.10 Bq/kg respectively while the activity concentrations of ^{232}Th, and ^{228}Th for Mneefa were 0.15 – 0.82 and 0.24 – 1.15 Bq/kg respectively as presented in Table 2. There is good agreement and equilibrium in all measured samples of ^{232}Th, and ^{228}Th for both areas of Al-Khafji and Mneefa while the measured samples of the ^{228}Ra show a disequilibrium when compared with the grand parent ^{232}Th. It has to be noticed that activity concentrations of ^{232}Th and its grand daughter ^{228}Th are much less than those values published from other different coastal marine sediments such as the such as the Bay of Bengal [Sharif, A. K. M., et al , 1994] and north west Pacific ocean [Yang, H. S., et al , 1986]. The spatial distribution of activity concentrations of ^{226}Ra, ^{228}Ra, and ^{40}K seems to be heterogeneous which could be attributed to the nature of sediments. There is no direct or inverse proportionality of sample depth with respect to the measured concentrations of radionuclides in this study.

The ^{228}Th:^{232}Th ratios were found to be slightly above unity which indicates clearly that the sampled sediments are old surficial sediments as a results of ^{228}Ra migration and decay of initial excess ^{228}Th [Koide, M. et al., 1973]. For young surficial sediments, the ^{228}Th/^{232}Th ratio would be much higher than unity. The sedimentation rates in coastal regions are known to be two to three orders of magnitude higher than in the open sea so it is not uncommon for ^{228}Th to appear in considerable excess of the parent ^{232}Th in coastal sediments. The ^{228}Th:^{232}Th activity ratio has therefore been used as a geochronometer in dating coastal marine sediments deposited during the last decade or so, providing that the sedimentation rate is rapid enough to allow the delineation of decreasing excess ^{228}Th with depth [Sam, A. K., et al , 2000].

Table 2 Activity concentrations (Bq/kg) of ^{40}K and thorium series of Al-Khafji and Mneefa

Sample #	Lat. & Long.	Depth, m	^{232}Th	^{228}Th	^{228}Ra	^{40}K
1	28 25 625N 48 32 388E	S	0.45 ± 0.12	0.54 ± 0.09	2.17 ± 0.26	52.16 ± 2.05
2	28 25 625N 48 32 688E	8	0.19 ± 0.05	0.23 ± 0.07	1.90 ± 0.27	27.04 ± 1.73
3	28 25 798N 48 32 977E	12	0.95 ± 0.09	1.10 ± 0.11	1.51 ± 0.26	24.22 ± 1.68
4	28 26 035N 48 33 286E	18	0.82 ± 0.12	1.01 ± 0.13	1.53 ± 0.27	28.91 ± 1.93
5	28 25 754N 48 33 872E	25	0.42 ± 0.14	0.55 ± 0.11	1.25 ± 0.23	23.69 ± 1.60
6	27 35 412N 48 54 574E	S	0.45 ± 0.10	0.52 ± 0.08	3.19 ± 0.31	113.9 ± 2.99
7	27 35 364N 48 54 519E	5	0.82 ± 0.08	1.02 ± 0.13	7.53 ± 0.48	204.2 ± 4.4
8	27 35 850N 48 54 638E	10	0.23 ± 0.06	0.29 ± 0.07	3.96 ± 0.34	152.2 ± 3.41
9	27 35 939N 48 54 800E	14	0.80 ± 0.08	1.15 ± 0.11	10.26 ± 0.56	253.3 ± 4.4
10	27 36 297N 48 54 996E	15	0.15 ± 0.04	0.24 ± 0.05	8.74 ± 0.51	219.6 ± 4.3

The activity concentrations of ^{238} U and ^{232}Th radionuclides of different areas in the world are shown in Table 3. The obtained concentrations were in agreement with those concentrations of different coastal oceans. The average activity concentration of ^{238} U was 53 times higher than that of ^{232}Th in Al-Khafji area and 62 times higher than that of ^{232}Th in Mneefa area. This excess of ^{238} U is probably attributed to the organic and mineral biogenic material, which is

always associated with the sediments in biologically productive shallow-waters . This is because in a such highly productive waters, sedimentation rates are usually high and reducing conditions lead to the transformation of uranium to the more insoluble uranus form[Sam, A.K., et al , 2000].

Table 3 Uranium and thorium activity concentrations of different areas in the world

Location	^{238}U (Bq/kg)	^{232}Th (Bq/kg)
Irish Sea[3]	1.8 - 6.5	4.1 - 12.1
United Kingdom[4]	3.6 - 32.3	2.6 - 24.2
North Sea[5]	3 - 11	-
Red Sea[9]	6.5 - 53	0.22 – 19.29
Al-Khafji	27.78 – 33.94	0.19 – 0.95
Mneefa	17.09 – 39.76	0.15 – 0.82

CONCLUSION

It is concluded from the preliminary measurements and investigation of naturally occurring radionuclides for sediment samples collected from both the Al-Khafji and Mneefa areas that the activities are within the international limits. It has been found that the activity concentrations of all studied natural radionuclides are independent of samples depths. It is highly recommended that more samples and living organisms should be collected along the coast of the gulf especially at Hormoz strait which is considered to be the open gate of the closed body of the Arabian Gulf.

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