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ORIGINAL ARTICLE

Heavy elements concentrations, physiochemical characteristics and natural radionuclides levels along the Saudi coastline of the Gulf of Aqaba

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Abstract This paper represents the first work on the concentrations of heavy elements, physiochemical characteristics and activity levels of the naturally occurring radionuclides in the Saudi Arabian coastline of the Gulf of Aqaba. Concentrations of 19 heavy elements were measured, namely: Ag, Al, As, Ba, Be, Cd, Co, Cr, Cu, Fe, Hg, Mn, Mo, Ni, Pb, Sb, Se, V and Zn. The radioactivity levels of ^{238}U, ^{232}Th and ^{40}K were estimated to be: 17 ± 1.7, 22.5 ± 3.7 and 649.6 ± 64.2 Bq kg^{-1}, respectively. The measurements were carried out using inductively coupled plasma-mass spectrometry (ICP-MS). In addition, physiochemical characteristics of 19 sediment samples (i.e., saturation percentage, pH, electrical conductivity, organic matter, cation exchange capacity and content of clay, silt and sand) have been determined. Indications for high correlation between most heavy elements are found. The correlation between heavy elements and the radionuclides ^{238}U, ^{232}Th and ^{40}K was generally significant.

1. Introduction

Most of chemical elements can be tolerable, toxic or lethal, depending on the individual dosage. The term heavy element is widely used and generally refers to any chemical element that has a relatively high density and which is toxic at low concentrations (Ojovan and Lee, 2005). The heavy elements in the human environment can interact, either positively or negatively with the human body (Gomes and Silva, 2007). The selected 19 heavy elements besides being toxic to human beings are of significant impact on the biogeochemical cycles. For instance, Cu, Cd, Hg, Pb and Zn are the heavy elements that

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often cause contamination of soil, water and food chains (Webber, 1981; He et al., 2005).

Limited number of published studies has dealt with concentration levels of heavy elements at the Jordanian coastline of the Gulf of Aqaba (Hulings, 1982; Abu Hilal and Badran, 1990; Wahbeh, 1990; Abu Hilal, 1993). However, only one study was carried out for the distribution of some heavy elements along the Red Sea Saudi coastline, without covering the Saudi coastline of the Gulf of Aqaba, except for one location (Al-Hefne et al., 2005). The present study consists of a survey of concentration levels of heavy elements at the Saudi coastline of the Gulf of Aqaba. The coastline is located in an area contaminated by several sources of pollution emitted from large phosphate fertilizer manufacturing facilities, storage area and loading terminal for potash at the Jordanian side, which is adjacent to the Saudi border. In order to assess the amount of pollution at sites away from the sources of pollution, the measurements were carried out along the coastline of 150 km extent. Our study aims to cover the whole Saudi coastline of the Gulf of Aqaba.

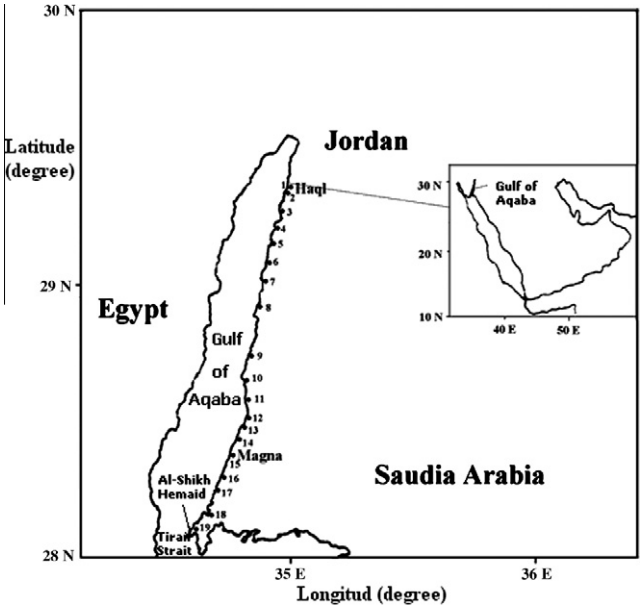

Figure 1 Map of the study area at Gulf of Aqaba, black circles represent the samples sites.

2. Materials and methods

2.1. Study area

The Gulf of Aqaba is about 180 km long and 15–25 km wide. The central part of the Gulf is the narrowest and the deepest. It has an average depth of 800 m and increases to about 1800 m at its maximum. The Gulf of Aqaba is a semi-closed aquatic system, connected only to the Red Sea by 5 km width and 340 m depth at Tiran Strait (Fig. 1). Moreover, the Gulf is located in a very arid area with an extremely low rainfall. This means that evaporation exceeds the rainfall average. So, the salinity is above the ocean's average of 35 g l^{-1} (Al Ouran, 2005).

The coastline of the Gulf of Aqaba is contaminated by several sources of pollution due to several activities occurring at the coastal area. Loading of fertilizer products onto ships and the unloading of some raw material including sulphur and ammonia at the two industrial berths pose a serious risk to the marine environment in case of accidental spills. It is estimated that about 5% of the exported phosphate is lost during transportation and loading. The huge amount of phosphate dust is considered a major source of nutrients entering into the Gulf (Al Ouran, 2005), which could cause pollution reach southwards to Tiran Strait at the Saudi Arabian side of the Gulf.

Table 1 Samples locations along the Saudi Arabian coastline of the Gulf of Aqaba.

Location no.	Location description	Sample code	Latitude N (deg. min.)	Longitude E (deg. min.)	Average bulk density (g cm^{-3})
1	Addurrah	AG 3	29 21.387	34 57.524	1.54
2	Addurrah	AG 4	29 19.974	34 56.975	1.56
3	Haql city	AG 5	29 15.933	34 56.215	1.64
4	6 km south of Haql	AG 6	29 12.914	34 54.702	1.60
5	14 km south of Haql	AG 7	29 08.880	34 53.457	1.52
6	22 km south of Haql	AG 8	29 04.605	34 52.301	1.50
7	Ras Dubur	AG 9	28 59.604	34 51.614	1.60
8	Beer Marsha	AG 10	28 54.813	34 49.777	1.48
9	Ras Swehel	AG 11	28 43.390	34 48.239	1.63
10	9 km north of Taib Isem	AG 12	28 38.619	34 47.247	1.60
11	Taieb Isem	AG 15	28 33.574	34 48.071	1.56
12	7 km north of Magna	AG 16	28 31.066	34 48.024	1.57
13	5 km north of Magna	AG 17	28 28.790	34 46.402	1.63
14	4 km north of Magna	AG 19	28 27.260	34 45.772	1.60
15	Magna town	AG 13	28 25.166	34 44.956	1.53
16	11 km south of Magna	AG 20	28 19.457	34 42.528	1.45
17	17 km south of Magna	AG 21	28 16.071	34 41.020	1.49
18	11 km north of Al-Shikh Hemaid	AG 22	28 09.946	34 39.529	1.57
19	Al-Shikh Hemaid	AG 23	28 05.184	34 34.758	1.57

Table 2 Concentration of heavy elements for sediment samples along Saudi coastline of the Gulf of Aqaba.

Sample code	Ag (mg kg^{-1})	Al (%)	As (mg kg^{-1})	Ba (mg kg^{-1})	Be (mg kg^{-1})	Cd (mg kg^{-1})	Co (mg kg^{-1})	Cr (mg kg^{-1})	Cu (mg kg^{-1})	Fe (%)	Hg (mg kg^{-1})	Mn (mg kg^{-1})	Mo (mg kg^{-1})	Ni (mg kg^{-1})	Pb (mg kg^{-1})	V (mg kg^{-1})	Zn (mg kg^{-1})
AG 3	0.15	2.19	2.6	190	0.79	0.05	2.4	519	10.3	1.29	< 0.01	218	1.23	11.2	11.5	21	19
AG 4	0.12	2.19	1.9	220	0.79	0.04	2.0	592	9.5	1.37	< 0.01	289	1.30	11.0	10.0	19	14
AG 5	0.10	2.35	1.5	220	0.70	0.02	1.4	506	5.9	0.78	< 0.01	99	1.16	9.1	8.6	8	8
AG 6	0.05	1.83	< 5.0	180	0.43	0.02	0.9	147	3.0	0.33	0.02	48	0.47	1.3	5.5	5	27
AG 7	0.06	4.07	2.1	350	1.30	0.04	1.9	226	4.8	0.93	< 0.01	185	0.82	5.8	8.6	18	14
AG 8	0.07	3.43	2.0	280	1.12	0.05	2.6	437	5.9	1.31	< 0.01	199	1.22	10.4	8.5	26	13
AG 9	0.11	5.88	3.3	700	2.22	0.08	4.6	201	8.5	2.05	0.01	661	1.51	9.3	11.7	30	46
AG 10	0.08	5.93	1.9	280	3.77	0.05	2.2	232	6.2	1.18	< 0.01	310	1.11	7.2	12.4	19	28
AG 11	0.08	5.98	2.8	470	4.17	0.21	3.8	248	7.3	5.74	< 0.01	1980	3.49	7.0	13.2	33	181
AG 12	0.21	7.20	2.7	450	2.72	0.15	14.5	190	22.8	5.88	< 0.01	1400	2.52	15.9	11.3	124	136
AG 15	0.14	5.71	1.2	840	2.09	0.21	3.4	275	8.7	2.24	0.01	815	1.86	8.5	14.3	25	76
AG 16	0.05	5.45	1.0	580	1.55	0.03	3.4	266	9.4	1.15	< 0.01	199	1.01	11.5	9.3	20	26
AG 17	0.08	2.77	1.2	230	0.82	0.08	2.8	519	6.8	1.13	0.01	113	1.27	12.6	8.7	19	20
AG 19	0.05	3.27	7.0	240	0.95	0.04	2.7	159	6.6	0.91	0.02	106	0.70	7.4	9.6	21	16
AG 13	0.08	4.03	1.9	410	0.95	0.02	2.2	352	5.7	0.86	< 0.01	92	1.13	9.3	8.9	13	26
AG 20	0.07	2.27	< 5.0	230	0.55	0.05	1.6	106	4.5	0.42	0.01	68	0.54	2.5	6.1	9	8
AG 21	0.07	2.43	12.0	200	0.78	0.12	2.9	117	6.4	0.81	< 0.01	312	1.18	7.0	8.2	23	18
AG 22	0.05	2.10	1.6	250	0.59	0.02	1.0	250	4.6	0.46	< 0.01	55	0.78	4.9	6.6	7	6
AG 23	0.06	3.01	1.2	290	0.87	0.03	1.3	274	3.6	0.89	< 0.01	86	0.99	4.8	7.7	13	12

Table 3 Activity concentrations of radionuclides and physiochemical characteristics of sediment samples along Saudi coastline of the Gulf of Aqaba.

Sample code	^{238}U (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	^{232}Th/^{238}U	SP (%)	pH	ECe (dS m^{-1})	Clay (%)	Silt (%)	Sand (%)	OM (g kg^{-1})	CEC (C mol kg^{-1})
AG 3	12.40	17.05	306.74	1.38	26.7	8.67	33.5	5	0	95	6.7	3.3
AG 4	12.40	30.86	322.39	2.49	25.0	8.54	19.6	5	0	95	3.3	2.1
AG 5	8.68	8.93	403.77	1.03	23.3	8.28	25.2	5	0	95	6.7	2.4
AG 6	19.84	6.09	338.04	0.31	25.0	8.88	39.6	7	0	93	16.7	4.1
AG 7	11.16	12.18	538.36	1.09	22.7	8.86	48.3	7	0	93	10.0	5.2
AG 8	11.16	15.43	403.77	1.38	24.7	8.68	52.9	7	0	93	10.0	5.6
AG 9	17.36	18.27	873.27	1.05	26.7	8.78	58.0	5	0	95	10.0	6.5
AG 10	22.32	44.25	1136.19	1.98	28.3	8.84	32.5	5	0	95	6.7	3.2
AG 11	37.20	58.87	1067.33	1.58	25.7	8.76	26.6	5	0	95	3.3	2.7
AG 12	17.36	25.17	801.28	1.45	28.3	8.88	29.6	7	0	93	10.0	3.0
AG 15	24.80	54.81	1076.72	2.21	28.3	8.81	36.0	3	0	97	6.7	2.9
AG 16	8.68	11.77	957.78	1.36	24.3	8.75	27.1	3	0	97	3.3	2.7
AG 17	8.68	10.96	456.98	1.26	28.3	8.79	36.5	5	0	95	6.7	3.8
AG 19	23.56	23.95	535.23	1.02	27.7	8.86	51.6	7	8	85	26.7	6.0
AG 13	8.68	8.12	841.97	0.94	27.3	8.77	38.1	5	0	95	10.0	4.1
AG 20	14.88	5.28	435.07	0.35	30.0	8.76	21.9	5	0	95	16.7	2.3
AG 21	22.32	16.24	450.72	0.73	28.3	8.27	43.1	7	10	83	30.0	4.4
AG 22	17.36	18.27	613.48	1.05	27.0	8.36	40.6	5	0	95	10.0	4.2
AG 23	23.56	40.60	782.50	1.72	27.0	8.39	49.6	3	0	97	13.3	5.2

2.2. Sampling

During February 2008, 19 sedimentary samples were collected from the coastline of the Gulf of Aqaba, starting from Addurah beach at the Saudi-Jordanian border going southward to Al-Shikh Hemaid at Tiran Strait through Haql city and Magna town. Sampling sites were selected to cover the shore area as uniformly as possible despite the access to some beaches being difficult topography-wise. These locations are on the average 8 km apart. The samples were collected by core sampler at most depth of 10 cm. Each sample was obtained from five sub-samples from the same location within 10 m distance. The five sub-samples were homogenized in situ and kept in thick plastic bag. Sampling locations are shown in Table 1.

2.3. Experimental measurements

All samples were brought to the laboratory at King Saud University, Riyadh City, weighed and dried for 48 h in an oven at 70 °C. Then stones, leaves and other foreign particles were removed. The sediment samples were screened with a 2.0 mm laboratory test sieve. The samples were then analyzed in ALS-Chemex Laboratories (Vancouver, Canada) using Perkin–Elmer Elan 9000 ICP-MS to determine the heavy metals as well as the radionuclides; U, Th and K concentrations. The conversions used for the specific radioactivity of the three radionuclides were: 1 mg kg^{-1} U = 12.4 Bq kg^{-1} ^{238}U, 1 mg kg^{-1} Th = 4.06 Bq kg^{-1} ^{232}Th and 1% K = 313 Bq kg^{-1} ^{40}K. The samples were also examined to measure their physiochemical characteristics including: saturation percentage (SP), pH and electrical conductivity (ECe) in saturation paste, cation exchange capacity (CEC) and contents of clay, silt and sand (Klute, 1986). In order to determine the organic matter (OM) in a sample, 30 g of it was ashed at 500 °C for 24 h and the content of OM was determined.

3. Results and discussion

Table 2 shows the concentrations of the heavy elements found at the Saudi coastline of the Gulf of Aqaba, while Table 3

Table 4 Basic statistics of heavy elements content of sediment samples along Saudi coastline of the Gulf of Aqaba.

	Average	World average*	Min.	Max.	Median	Mode	SD	SE
Ag (mg kg^{-1})	0.09	0.08	0.05	0.21	0.08	0.05	0.04	0.01
Al (%)	3.79	8.4	1.83	7.2	3.27	2.19	1.7	0.39
As (mg kg^{-1})	2.52	9.36	0	12	1.9	1.9	2.74	0.63
Ba (mg kg^{-1})	347.9	340	180	840	280	220	185.4	42.5
Be (mg kg^{-1})	1.43	1.9	0.43	4.17	0.95	0.79	1.09	0.25
Cd (mg kg^{-1})	0.07	0.06	0.02	0.21	0.05	0.05	0.06	0.01
Co (mg kg^{-1})	3.03	10.4	0.9	14.5	2.4	2.2	2.94	0.67
Cr (mg kg^{-1})	295.6	20.2	106	592	250	519	148.9	34.2
Cu (mg kg^{-1})	7.39	20	3	22.8	6.4	5.9	4.25	0.97
Fe (%)	1.56	5.2	0.33	5.88	1.13	–	1.57	0.36
Hg (mg kg^{-1})	0.004	0.03	0	0.02	0.01	0.01	0.01	0.001
Mn (mg kg^{-1})	380.8	1100	48	1980	199	199	512.3	117.5
Mo (mg kg^{-1})	1.28	1–5	0.47	3.49	1.16	–	0.71	0.16
Ni (mg kg^{-1})	8.25	40	1.3	15.9	8.5	9.3	3.55	0.82
Pb (mg kg^{-1})	9.51	10.15	5.5	14.3	8.9	8.6	2.38	0.55
Sb (mg kg^{-1})	0.25	0.2	0.16	0.4	0.24	0.18	0.07	0.02
Se (mg kg^{-1})	2.16	0.2	1	5	2	2	1.01	0.23
V (mg kg^{-1})	23.8	190	5	124	19	19	25.4	5.8
Zn (mg kg^{-1})	36.5	10.3	6	181	19	14	46.5	10.7

* Cox (1989), James and Lord (1992), and He et al. (2005).

Table 5 Basic statistics of activity concentrations of radionuclides and physiochemical characteristics.

	Average	Min.	Max.	Median	Mode	SD	SE
^{238}U (Bq kg^{-1})	17.0	8.7	37.2	17.4	8.7	7.5	1.72
^{232}Th (Bq kg^{-1})	22.5	5.3	58.9	17.1	18.3	16.2	3.71
^{40}K (Bq kg^{-1})	649.6	306.7	1136.2	538.4	403.8	279.7	64.2
$^{232}Th/^{238}U$	1.3	0.3	2.5	1.3	1.1	0.56	0.13
SP (%)	26.6	22.7	30.0	27.0	28.3	2.0	0.45
pH	8.7	8.3	8.9	8.8	8.9	0.2	0.05
ECe (dS m^{-1})	37.4	19.6	58.0	36.5	–	11.1	2.50
OM (g kg^{-1})	10.9	3.3	30.0	10.0	10.0	7.3	1.70
CEC (C mol kg^{-1})	3.9	2.1	6.5	3.8	–	1.3	0.30
Clay (%)	5.3	3.0	7.0	5.0	5.0	1.4	0.32
Silt (%)	0.9	0.0	10.0	0.0	0.0	2.9	0.66
Sand (%)	93.7	83.0	97.0	95.0	95.0	3.7	0.84

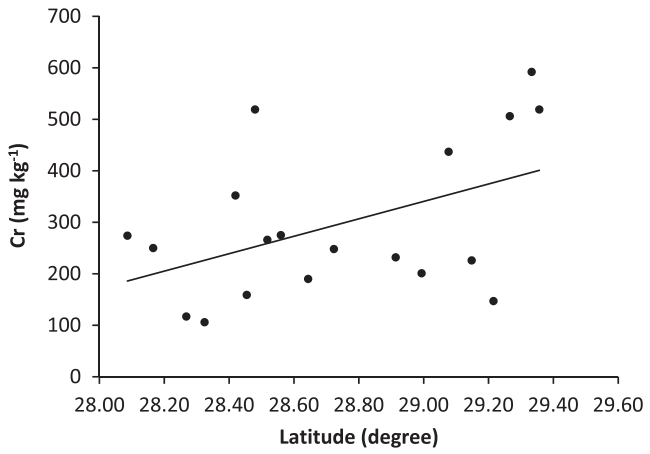

Figure 2 Trend line for concentrations of Cr versus latitude of samples along Saudi coastline of the Gulf of Aqaba.

shows the recovered activity concentrations amounts of the radionuclides ^{238}U, ^{232}Th, ^{40}K and the ratio ^{232}Th/^{238}U as well as the physiochemical characteristics of the sediment samples. Basic statistics of the heavy elements concentrations, including minimum and maximum values, average, world average, median, mode, standard deviation (SD) and standard error (SE) are given in Table 4. The element concentrations were found to widely vary as a result of the physiochemical and geochemical properties and their related environment (El Mamoney and Khater, 2004).

For most of the elements the average concentrations of the elements in the samples are found to be smaller compared to the world average (i.e., a factor of 2.2 for Al, 3.7 for As, 3.2 for Co, 2.7 for Cu, 7.5 for Hg, 2.9 for Mn, 4.8 for Ni, and 8 for V). Whereas, concentrations of some other elements, such as: Ag, Ba, Mo, Cd, Pb and Sb were close to the world average (Cox, 1989; James and Lord, 1992; He et al., 2005). The average concentrations of Cr, Se and Zn exceeded the world average. The concentration of Se ranges between 1 and 5 mg kg^{-1}, with an average value of 2.2 mg kg^{-1} and mode 2 mg kg^{-1}, and for Cr ranged between 106 and 592 mg kg^{-1}, with an average value of 296 mg kg^{-1} and mode of 519 mg kg^{-1}, indicating high content of Se and Cr in all samples compared to the world average of 0.2 and 20 mg kg^{-1}, respectively. This high content can be explained as due to geological formation and the parent rocks from which they were derived (Veiga et al., 2006; Dragovic et al., 2008).

Plotting the concentration of each heavy element with latitude using linear regression, the trend line shows decrease of concentrations southwards, i.e., away from the sources of pollution for most elements, as shown in Fig. 2 for Cr as example.

In Table 5, the average saturation percentage (SP) of the samples was 26.6%; the average electrical conductivity (ECe) was 37.4 dS m^{-1} and the average of pH was 8.7. The particle size distribution in the samples of the studied locations indicated that the sand is the main constituent in all samples, with an average of 93.7%. The silt percentage was zero in all samples, except in locations AG 19 and AG 21, which have loamy-sand texture. The average percentage of clay was 5.3%; the average cation exchange capacity (CEC) was 3.9 C mol kg^{-1}. The organic matter content (OM) average value was 10.9 g kg^{-1}, which was low in all analyzed samples. The organic matter is a component of great importance because it tends to form soluble or insoluble complexes with the heavy elements, which is why they can migrate throughout the profile, (Schnitzer and Khan, 1989), or be retained in the soil (Vega et al., 2004).

Table 5 also indicates that the activity concentration of ^{238}U and ^{232}Th ranged from 8.7 to 37.2 Bq kg^{-1} with an average of 17 Bq kg^{-1} and from 5.3 to 58.9 Bq kg^{-1} with an average of 22.5 Bq kg^{-1}, respectively. These average values are lower than the global average of 25 Bq kg^{-1} (UNSCEAR, 1988). The activity of ^{40}K is found to be greater by a factor of 1.8 than the global average of 370 Bq kg^{-1} (UNSCEAR, 1988). The concentration activity ratio of ^{232}Th/^{238}U was computed for all samples and the values varied from 0.3 to 2.5, with an average value of 1.28. It is slightly above the world's average ratio, which is equal to unity (UNSCEAR, 1988), while the average Th/U concentration ratio was 3.9 which is close to reported global Th/U ratio of 3.5 (Adams, 1962).

Relationships between heavy elements provide information on those elements sources and pathways (Dragovic et al., 2008). Pearson's correlation coefficients show a high correlation between most elements, mostly positive correlation. However, the elements such as As, Cr and Sb show low correlation with most elements, but show high correlation between each other. The correlation between CEC and clay content is positive, with $r = 0.36$, and also positive with OM ($r = 0.47$), because both organic matter and clay hold cations. In contrast, CEC correlation was inversely related with sand content. There was a positive low correlation between CEC and pH. On the other hand, the correlation was also positive between clay content and OM. The correlation between heavy elements and the radionuclides ^{238}U, ^{232}Th and ^{40}K was generally significant, except for Ag, As, Co, Cu, Sb and V (Table 6).

There is a positive correlation between activity concentrations results of radionuclides of ^{238}U, ^{232}Th and ^{40}K. Fig. 3 shows the corresponding best linear fit. The correlation is positive with $r = 0.77$ between ^{238}U and ^{232}Th while the correlation coefficient $r = 0.65$ between ^{232}Th and ^{40}K. The activity concentrations between ^{238}U and ^{40}K exhibit a correlation of coefficient of 0.47. Activity concentrations of radionuclides are directly proportional to the density of the sediments samples (Schuiling et al., 1986; Ligero et al., 2001), and inversely proportional to that of clay.

Table 6 Pearson's correlation coefficients between heavy elements and activity concentration of radionuclides.

	Ag	Al	As	Ba	Be	Cd	Co	Cr	Cu	Fe	Hg	Mn	Mo	Ni	Pb	Sb	Se	V	Zn
^{238}U	−0.03	0.32	0.31	0.21	0.58	0.63	0.09	−0.51	−0.03	0.48	0.46	0.62	0.52	−0.36	0.39	−0.08	0.76	0.13	0.61
^{232}Th	0.24	0.51	−0.11	0.42	0.71	0.66	0.15	−0.05	0.15	0.54	−0.20	0.64	0.64	0.03	0.72	0.02	0.63	0.19	0.60
^{40}K	0.10	0.84	−0.24	0.74	0.79	0.48	0.29	−0.34	0.19	0.47	−0.49	0.54	0.50	0.10	0.63	−0.35	0.46	0.25	0.55

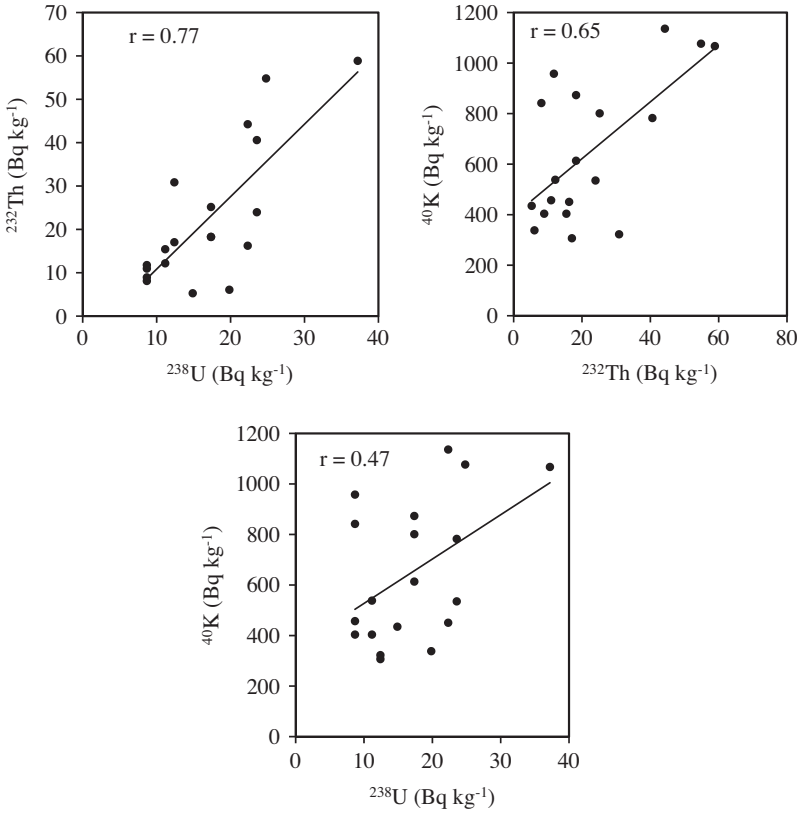

Figure 3 Correlation between activity concentrations of ^{238}U, ^{232}Th and ^{40}K in Bq kg^{-1}.

4. Conclusion

Nineteen heavy elements, radionuclides of ^{238}U, ^{232}Th and ^{40}K and physiochemical characteristics have been measured comprehensively for whole coastline of Saudi Arabian Gulf of Aqaba for the first time. The study indicated that the coastline is contaminated due to the pollution from the industrial zone in Jordanian side. Nevertheless, the concentration of toxic elements and the activity concentration of radionuclides are low compared to the world average. The authors believe that the further extensive study of radionuclides and heavy elements distribution and their relations with physiochemical characteristics are needed for the beach of Addurah and Haql city which are exactly adjacent to the Jordanian industrial region.

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