Natural radioactivity in groundwater from the south-eastern Arabian Peninsula and environmental implications

A. Murad · X. D. Zhou · P. Yi · D. Alshamsi · A. Aldahan · X. L. Hou · Z. B. Yu

Abstract Groundwater is the most valuable resource in arid regions, and recognizing radiological criteria among other water quality parameters is essential for sustainable use. In the investigation presented here, gross-α and gross-β were measured in groundwater samples collected in the south-eastern Arabian Peninsula, 67 wells in Unite Arab Emirates (UAE), as well as two wells and one spring in Oman. The results show a wide gross-α and gross-β activities range in the groundwater samples that vary at 0.01~19.5 Bq/l and 0.13~6.6 Bq/l, respectively. The data show gross-β and gross-α values below the WHO permissible limits for drinking water in the majority of the investigated samples except those in region 4 (Jabel Hafit and surroundings). No correlation between groundwater pH and the gross-α and gross-β, while high temperatures probably enhance leaching of radionuclides from the aquifer body and thereby increase the radioactivity in the groundwater. This conclusion is also supported by the positive correlation between radioactivity and amount of total dissolved solid. Particular water purification technology and environmental impact assessments are essential for sustainable and secure use of the groundwater in regions that show radioactivity values far above the WHO permissible limit for drinking water.

Keywords Gross alpha · Gross beta · Groundwater · Radioactivity · UAE

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Introduction

Groundwater contains a certain amount of natural radioactivity and measurements of gross alpha and gross beta (will be referred to as gross-α and gross-β throughout the text) are often used as screen method for the evaluation of the quality of the groundwater. The sources of gross-α activity in groundwater is, to a large extent, the decay of uranium and thorium isotopes (^{234}U, ^{235}U,

^{238}U, ^{232}Th) and their progenies (Osmond and Ivanovich 1992). Gross-β activity, however, is mostly related to decay of potassium-40 (^{40}K) and beta radio-nuclides of decay chain of ^{238}U and ^{232}Th, for example ^{228}Ra and ^{210}Pb (Zorer et al. 2013). Anthropogenic isotopes such ^{3}H, ^{90}Sr, ^{137}Cs, and ^{129}I may also contribute to the gross-β, but the contribution is considered negligible in the groundwater investigated here as atmospheric fallout is insignificant in the region and even more remote in the sediment and rocks aquifers.

For practical purposes, determination of gross-α and gross-β is a straightforward and simple way for the estimation of radioactivity level in waters compared to determination of individual radionuclide, which is more complicated and time-consuming (Bonotto et al. 2009). Groundwater represents vital resource in many parts of the world and in particular arid region such as the Arabian Peninsula, where estimates of groundwater radioactivity is mostly missing. This issue is vital for sustainable management of groundwater resources in the region at present and in the future. We here, thus, present the first study of gross-α and gross-β

radioactivity in groundwater from south-eastern region of the Arabian Peninsula, namely the United Arab Emirates (UAE) and Oman (Fig. 1). To our best knowledge, there is no published data on radioactivity level in groundwater for those regions and subsequent assessment of the radioactivity hazards in comparison with the recommended international permissible limits for drinking water and domestic uses.

Sampling and analytical methods

Groundwater samples were collected from 67 different wells located in the UAE and two wells and one spring in Oman (Fig. 1). The water wells are mostly used for agriculture, domestic, and recreational purposes. The sampling area can be divided, topographically, into four major regions. The region 1 is characterized by relatively high mountain ranges and deeply incised valleys filled with wadi deposits and flood terraces. Most of the wells penetrate Triassic aquifers that are composed of carbonate rocks

Fig. 1 Map showing sampling sites of groundwater used in this investigation; 67 samples in UAE and three samples in Oman. *Black dotted line* indicates national borders
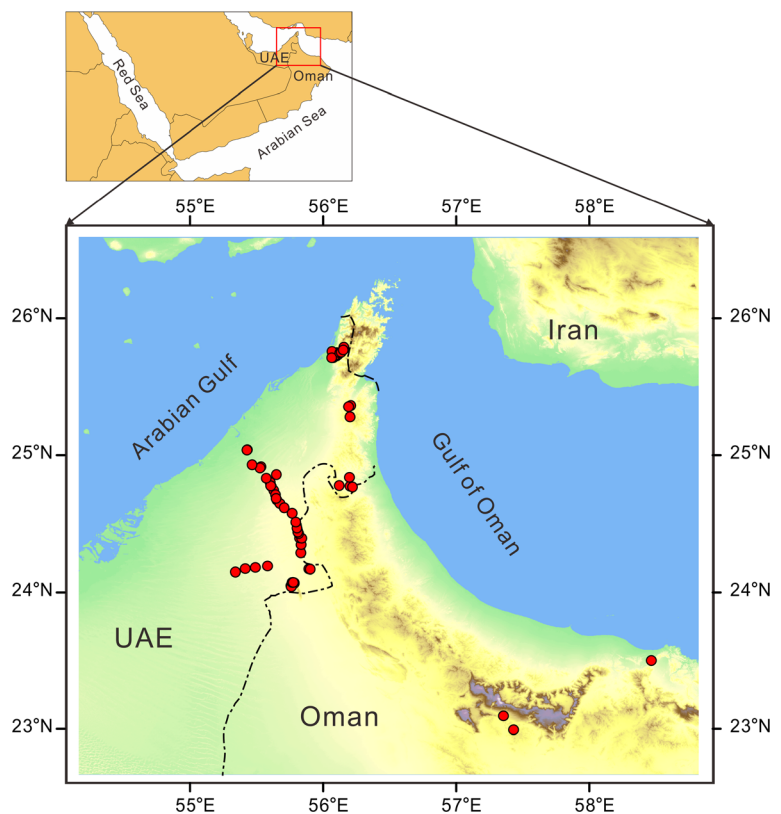
Table 1 Results of gross-α activity, gross-β activity, pH, temperature (T), and total dissolved solids (TDS) in groundwater samples investigated

	Sample code	UTM (E)	UTM (N)	Gross-α (Bq/l)	Gross-β (Bq/l)	pH	T (°C)	TDS (mg/l)	^{39}K (mg/l)	^{40}K (µg/l)
Region 1										
1	R-KH01	403,711	2,850,942	0.18	1.08	8.4	35.4	1,510	10.07	1.26
2	R-KH02	403,589	2,849,530	0.13	0.56	7.8	35.7	6,600	5.71	0.71
3	R-KH03	402,635	284,9,717	0.01	0.23	7.1	34.6	6,400	31.09	3.89
4	R-KH04	403,887	2,851,103	0.18	1.08	7.6	35.7	1,330	10.54	1.32
5	R-KH05	404,577	2,851,081	0.16	0.98	7.5	36.2	1,800	11.22	1.40
6	R-KH06	404,555	2,851,588	0.16	1.22	7.2	35.0	1,690	13.31	1.67
7	R-KH07	405,098	2,851,144	0.20	1.00	7.6	36.8	1,596	11.92	1.49
8	R-KH08	408,561	2,853,735	0.08	0.23	8.0	33.4	238	3.45	0.43
9	R-KH10	403,377	2,850,866	0.10	0.71	7.7	36.4	1,268	9.76	1.22
10	R-KH11	403,196	2,851,429	0.12	1.12	7.8	35.6	1,564	11.53	1.44
11	R-KH12	405,935	2,850,976	0.17	0.83	7.1	36.2	1,099	9	1.13
12	R-KH13	405,233	2,851,739	0.10	0.30	7.3	35.2	2,730	18	2.25
13	R-KH14	405,789	2,852,053	0.50	1.40	7.7	36.5	2,200	13.01	1.63
14	R-KH15	406,728	2,853,143	0.08	0.40	8.1	35.2	310	4.95	0.62
15	R-KH16	406,153	2,853,900	0.17	0.99	7.5	33.6	900	10.33	1.29
16	R-KH17	407,252	2,852,160	0.11	0.58	7.8	35.8	414	5.45	0.68
17	R-KH18	409,690	2,854,120	0.04	0.23	7.5	34.1	143	2.88	0.36
18	R-KH19	411,196	2,855,958	0.08	0.16	7.9	38.4	155	3.74	0.47
19	R-KH20	411,668	2,858,249	0.09	0.33	7.9	33.9	229	5.03	0.63
20	R-KH21	402,764	2,854,872	0.08	0.25	7.3	32.4	3,955	18.88	2.36
21	HA-13	415,305	2,744,969	0.08	1.06	8.7	37.5	1,738		
22	HU-1	415,095	2,751,892	0.08	0.22	8.2	35.0	508		
23	MS-1	407,391	2,745,328	0.08	0.26	8.8	29.5	1,320		
24	SH-2	417,350	2,744,096	0.08	0.22	7.7	32.0	7,500		
25	Mf-13	415,654	2,801,252	0.03	0.13	7.9	35.6	380		
26	AS-1	414,737	2,809,750	0.08	0.32	7.1	33.0	283		
27	TA-1	416,423	2,810,602	0.80	0.50	5.6	30.7	2,493		
28	102/72	537,665	2,545,410	0.08	0.13	9.7	35.0	425	2.07	0.26
29	WD-1	530,082	2,556,732	0.04	0.13	7.4	32.4	750	2.1	0.26
30	MO-1	641,343	2,602,779	0.35	0.31	6.9	62.0	890	4.65	0.58
Region 2										
1	Ma-1	375,534	2,711,071	0.09	0.30	8.4	35.0	199	3.12	0.39
2	MO-1	374,768	2,715,691	0.08	0.25	8.5	34.1	251	3.13	0.39
3	HY-1	372,069	2,723,052	0.08	0.62	8.2	33.2	326	4.61	0.58
4	SHB-1	376,819	2,706,480	0.08	0.82	8.1	31.6	481	11.69	1.46
5	EZ-1	376,207	2,707,686	0.08	0.63	8.1	33.6	745	6.96	0.87
6	MK-1	377,423	2,702,654	0.08	0.95	7.7	30.8	1,299	10.39	1.30
7	MK-2	379,070	2,702,337	0.08	0.85	8.0	34.0	1,480	9.45	1.18
8	FO-1	378,277	2,690,607	0.08	1.04	7.8	33.4	2,084	13.13	1.64
9	Gh-1	378,521	2,697,207	0.08	1.02	7.8	32.8	1,689	10.16	1.27
10	Kh-1	366,369	2,727,640	0.09	0.67	8.1	32.1	1,040	17.55	2.20
11	MQ-1	358,665	2,741,848	0.18	2.30	8.4	32.1	1,190	14.18	1.77
12	FQ-1	362,995	2,731,012	0.22	2.13	8.1	33.6	1,230	13.86	1.73
13	US-1	355,953	2,748,498	0.24	2.33	8.1	30.4	1,250	16.75	2.10
14	GS-1	353,131	2,751,866	0.03	0.33	8.3	31.8	1,320	12.92	1.62
15	Yh-1	359,981	2,738,171	0.19	2.22	8.4	32.1	1,500	13.44	1.68
16	MQ-2	356,807	2,745,234	0.22	2.68	8.4	30.8	1,860	25.24	3.16
17	FQ-2	360,970	2,733,806	0.20	3.15	8.1	30.6	2,610	37.2	4.66
18	Mm-1	349,424	2,761,392	0.20	2.85	8.1	30.6	2,830	24.15	3.02
19	LS-1	342,825	2,763,065	0.09	0.71	7.9	32.8	5,840	51.12	6.40
20	Mgm-1	360,766	2,754,718	0.28	3.45	7.8	29.1	5,920	86.09	10.77

Table 1 (continued)

	Sample code	UTM (E)	UTM (N)	Gross-α (Bq/l)	Gross-β (Bq/l)	pH	T (°C)	TDS (mg/l)	^{39}K (mg/l)	^{40}K (µg/l)
21	LS-2	348,573	2,760,379	0.08	1.12	7.8	30.4	3,470	37.75	4.72
22	MQ-3	356,337	2,745,664	0.12	1.37	8.2	33.4	3,640	42.3	5.29
23	Rw-1	339,214	2,775,281	0.03	1.99	7.4	28.9	4,150	36.43	4.56
24	FQ-3	360,384	2,735,288	0.08	1.35	7.7	32.9	4,540	43.99	5.50
25	Bal-1	339,078	2,775,137	0.34	0.21	8.6	32.5	570	7.59	0.95
26	GWW-Jaw,1	385,251	2,677,000	0.01	0.23	8.8	35.3	354		
27	GWW-Jaw,2	384,216	2,677,310	0.01	0.33	8.5	33.2	247		
Region 3										
1	AD-1	353,302	2,680,076	0.17	2.05	8.6	32.4	1,956	7.71	0.96
2	AD-2	344,134	2,679,008	0.16	3.02	8.0	35.8	5,310	37.4	4.68
3	AD-3	336,559	2,678,041	0.13	2.93	8.2	35.3	4,270	20.71	2.59
4	AD-4	329,139	2,675,454	0.16	2.14	8.3	32.0	4,908	13.49	1.69
Region 4										
1	GWW-58	372,788	2,665,600	19.50	5.23	8.8	46.9	6,080		
2	ADD0911078	373,094	2,665,913	16.50	4.22	8.2	49.0	6,100		
3	ADD0911085	373,142	2,666,352	17.60	6.61	8.8	49.0	6,100		
4	GWW-47	371,506	2,666,511	1.17	3.65	8.1	32.5	6,940		
5	ADD0911076	372,277	2,666,541	4.29	4.12	8.1	34.6	7,040		
6	GWW-F	370,885	2,663,116	5.60	3.84	8.2	33.4	7,200		
7	GWW-F	370,657	2,663,966	10.50	4.88	8.0	34.8	7,300		
8	ADD0911080	372,642	2,665,702	12.80	5.81	8.5	44.9	8,700		
9	GWW-53	372,370	2,666,667	3.76	3.51	8.5	34.5	8,900		

intercalated with evaporites and shales. The average range of precipitation is (100–350 mm/year) which is higher than the other three regions. Annual precipitation in regions 2, 3, and 4 is between 25 and 50 mm. The major difference between regions 2 and 3 is the common occurrence of wadis and alluvial deposits of Quaternary ages that are partly replaced by sand dunes ergs in region 3. Region 4 represents sampling around and in the vicinity of Jabel Hafit (a mountain) that is dominated by Neogene-Paleogene carbonate rock which are also partly intercalated with evaporites and shales.

Fig. 2 Distribution of gross-α activity in groundwater samples collected in this investigation: *region 1*=mostly mountainous, *region 2*=wadi alluvial with sand dunes, *region 3*=mainly sand dunes, and *region 4*=Jabel Hafit (mountain) and surroundings
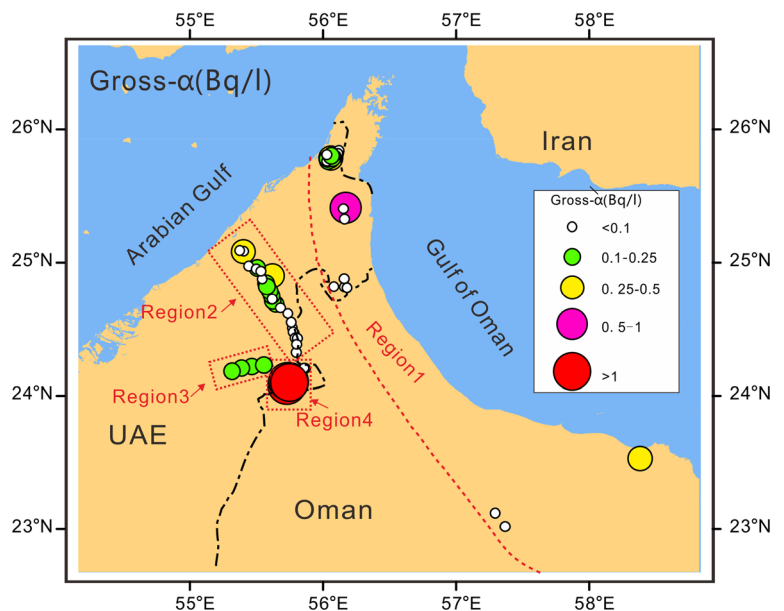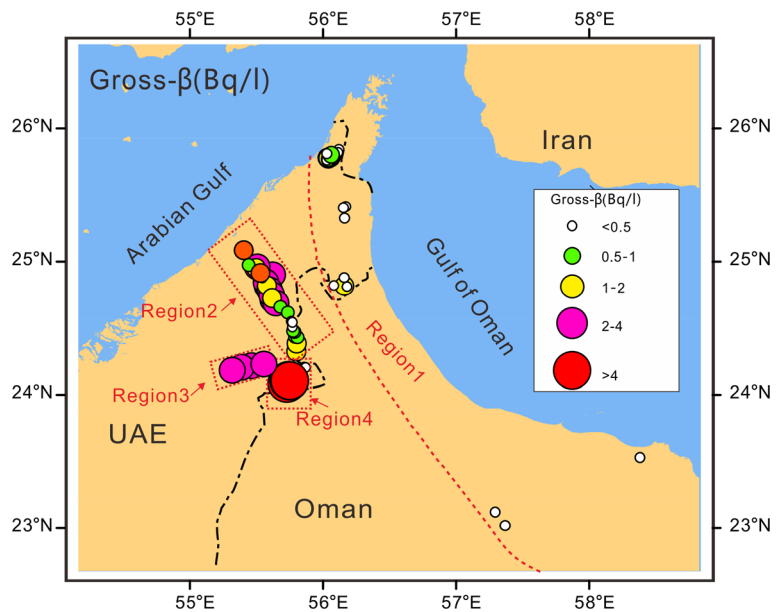
Fig. 3 Distribution of gross-β activity in groundwater samples collected in this investigation: *region 1*=mostly mountainous, *region 2*=wadi alluvial with sand dunes, *region 3*=mainly sand dunes, and *region 4*=Jabel Hafit (mountain) and surroundings

Temperature, electrical conductivity (EC) and pH were measured in the field using WTW-COND-3301 instrument. The total dissolved solids (TDS) in milligrams per liter were calculated from EC using a conversion factor 0.55 and 0.75 at a temperature of 25 °C (Hem 1970). An amount of 100 ml of water was evaporated to near dryness, and the residue was dissolved with water to a final volume of 5–15 ml. A 4 ml of the concentrated solution was taken to a LSC vial, and 16 ml of Ultima Gold LLT scintillation cocktail was added. After mixing and set in dark and cool for 1 h, the sample was measured using Quantulus 1220 liquid scintillation counter for 60 min each sample for 3 cycles using an alpha/beta discrimination function. Detection limits were calculated from the measurement of the procedure blank prepared using deionized water (18.2 MΩ cm^{-2}) to be 0.01 Bq/l for gross-α and 0.03 Bq/l for gross-β.

Results and discussion

The gross-α activity in the collected groundwater samples lies between 0.01 and 19.5 Bq/l, with an average of 1.4 Bq/l and standard deviation of 4.1 Bq/l (Table 1).

Table 2 Range values of the gross-α and gross-β activities in groundwater according to the four regions given in text

	Statistical value	Total	Region 1	Region 2	Region 3	Region 4
Gross-α	Maximum	19.50	0.80	0.34	0.17	19.50
	Minimum	0.01	0.01	0.01	0.13	1.17
	Average	1.43	0.15	0.12	0.16	10.19
	Standard deviation	4.10	0.16	0.08	0.02	6.77
	Above WHO permissible limit (0.5 Bq/l)	10/70	1/30	0/27	0/4	9/9
Gross-β	Maximum	6.61	1.40	3.45	3.02	6.61
	Minimum	0.13	0.13	0.21	2.05	3.51
	Average	1.50	0.57	1.33	2.54	4.65
	Standard deviation	1.52	0.40	0.98	0.51	1.06
	Above WHO permissible limit (1.0 Bq/l)	33/70	6/30	14/27	4/4	9/9

Fig. 4 Diagram showing gross-α activity in groundwater samples of the four regions and the permissible WHO 500 mBq/l limit
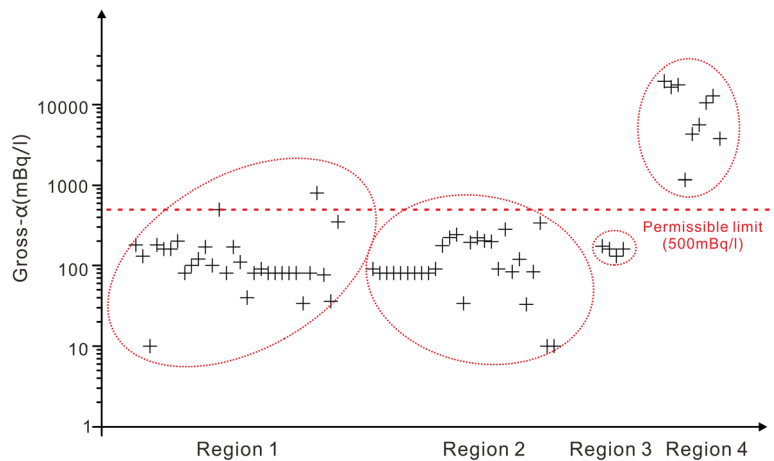
The gross-β activity has a smaller variability range (0.13 to 6.6 Bq/l) and an average of 1.6 Bq/l and a standard deviation value of 1.5 Bq/l. Variability of the gross-α and gross-β within the different regions (Figs. 2 and 3) indicates high values for groundwater from region 4 (average of 10.2 Bq/l for gross-α and 4.7 Bq/l for gross-β) compared to the other three regions. The average activity of gross-α and gross-β in regions 1, 2, and 3 are all below 1 Bq/l.

Among the several recommended permissible limits for radioactivity in drinking water, we selected the one established by the World Health Organization (WHO) (WHO 2011) with 0.5 Bq/l for gross-α and 1.0 Bq/l for gross-β activity. There are about 10 out of the 70 groundwater samples that have gross-α activity exceeding the WHO permissible limit of 0.5 Bq/l, and none of the groundwater samples in region 4 is below the

permissible limit (Table 2, Fig. 4). The majority of the samples in regions 1, 2, and 3 lie at 0.5–2 Bq/l for gross-α activity. When it comes to gross-β activity, then 33 out of the 70 samples are above the permissible limit of 1.0 Bq/l, and the majority of groundwater samples that show values below the limit are from region 1 (Table 2, Fig. 5). In regions 2, 3, and 4, there appears a mixture of different levels of gross-β that span from above 4 to<0.5 Bq/l (Figs. 3 and 5).

Correlation between gross-α and gross-β in the groundwater revealed a significant value ($R=0.8$; Fig. 6). This relatively good correlation might indicate that both gross-α and gross-β activities are from the natural radionuclides of decay chains of ^{238}U and ^{232}Th. The gross-α in groundwater is mainly contributed by the decay of uranium isotopes (^{234}U, ^{235}U, ^{238}U) and ^{226}Ra. An important progeny of ^{238}U is radon-222 (^{222}Rn)

Fig. 5 Diagram showing gross-β activity in groundwater samples of the four regions and the permissible WHO 1,000 mBq/l limit
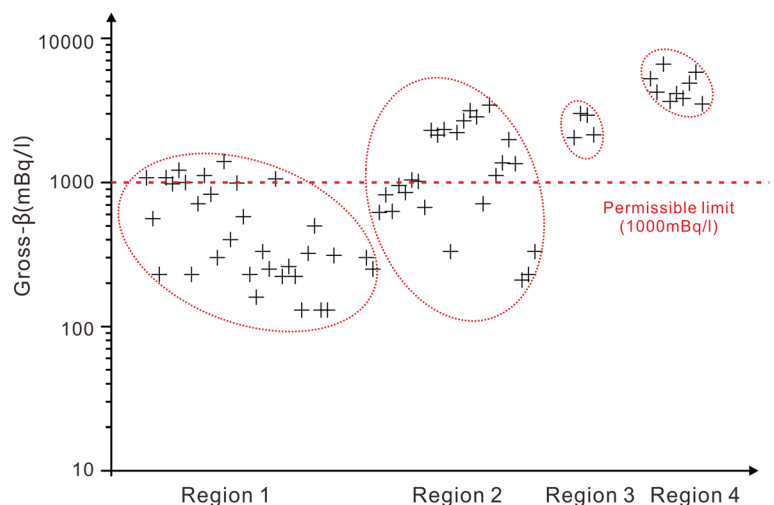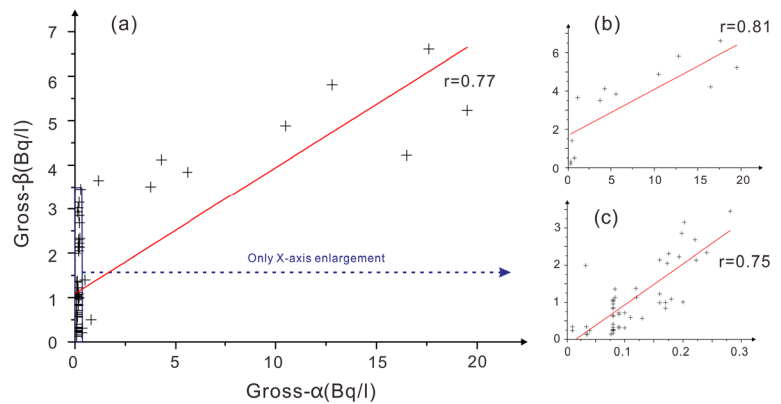
Fig. 6 Diagram showing the relationship between gross-α activity and gross-β groundwater samples. **a** All samples. **b**, **c** Enlargement of the X-axis for samples with gross-α activity at above and below 0.3 Bq/l

which easily escapes from the rock and enters to the groundwater. Because the water samples were evaporated before measurements, activity of ^{222}Rn would be excluded from the gross alpha activity presented here. Data on ^{226}Ra and uranium isotope (Murad et al. 2011; Alshamsi et al. 2013) in some groundwater samples indicated strong correlation ($R=0.8$) with gross-α. This correlation suggests that most of the gross-α activity is related to decay of ^{226}Ra, a progeny produced by ^{238}U decay.

Potassium-40 is expected to be an important source of gross-β activity. The concentration of ^{40}K in the groundwater was estimated from the concentration of ^{39}K (Table 1) by using the naturally occurring isotopic abundance of potassium as ^{39}K (93.258 %), ^{40}K (0.0116 %), and ^{41}K (6.730 %) (Al-Amir et al. 2012; Turhan et al. 2013). The estimated concentration varies

from about 0.2 to 11 μg/l. The occurrence of variable concentrations of potassium in the sampled groundwater could produce variation in gross-β activity. The positive and relatively good correlation between ^{40}K and gross-β ($R=0.57$, Fig. 7) suggests that part of the groundwater gross-β production can be related to ^{40}K in addition to other naturally occurred beta emitter, mainly decay products of ^{238}U and ^{232}Th. Other possible minor sources of gross-β are ^{228}Ra and ^{210}Pb which are not measured here.

The effect of groundwater pH on the gross-α and gross-β activities seems to be insignificant. The pH values range at 5.6–9.7 (Table 1), and there are rather weak correlation between gross-α, gross-β, and pH ($R=0.3$, Fig. 8). The pH value is strongly related to the water chemistry and in particular availability of chlorine and carboxyl ions. As most of the investigated

Fig. 7 Diagram showing the relationship between ^{40}K and gross-β activity in groundwater samples investigated
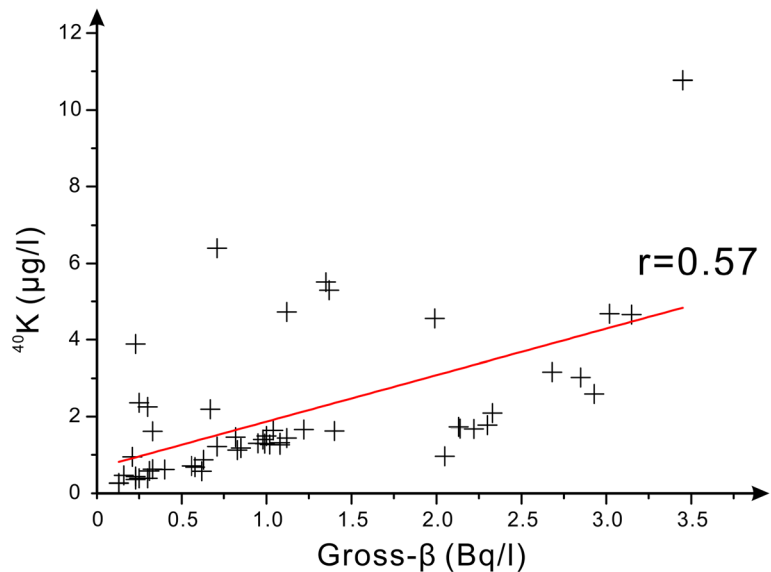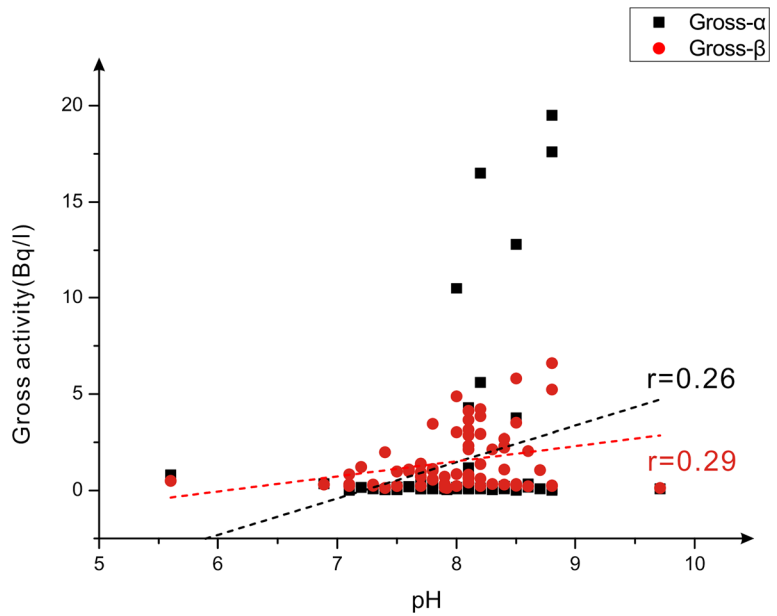
Fig. 8 Diagram showing the relationship between gross-α activity and pH (*black line*) and gross-β activity and pH (*red line*) in groundwater samples investigated

groundwater are carbonate-rich or occur in carbonate aquifers, uranium may convert into carbonate–uranium complexes at alkaline pH. These can in turn affect the solubility of radionuclides in groundwater. Uranium becomes more soluble in alkaline system in the presence of carbonate to produce $(UO_2(CO_3)_2^{2-})$ ions among many other soluble complexes and thus is expected to stay in solution at pH 6–8 (Yu et al. 2007). This is not the case for thorium which is more soluble in acidic

conditions. In general, it is expected that alkaline pH will increase the solubility of uranium and thus increase the radioactivity in water compared to the solid phase; the rather complicated behavior of the radionuclides may mask the correlation with pH.

Despite the large variability in the temperature of the groundwater (range 29–49 °C with only one sample above 60 °C, Table 1), the correlation is poor with gross-β ($R=0.3$) but is relatively better with gross-α

Fig. 9 Diagram showing the relationship between gross-α activity and temperature (*black line*) and gross-β activity and temperature (*red line*) in groundwater samples investigated
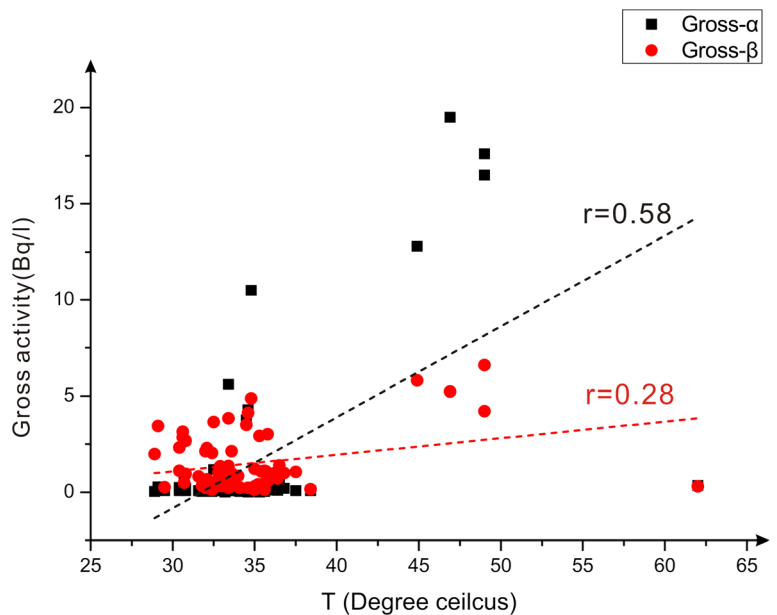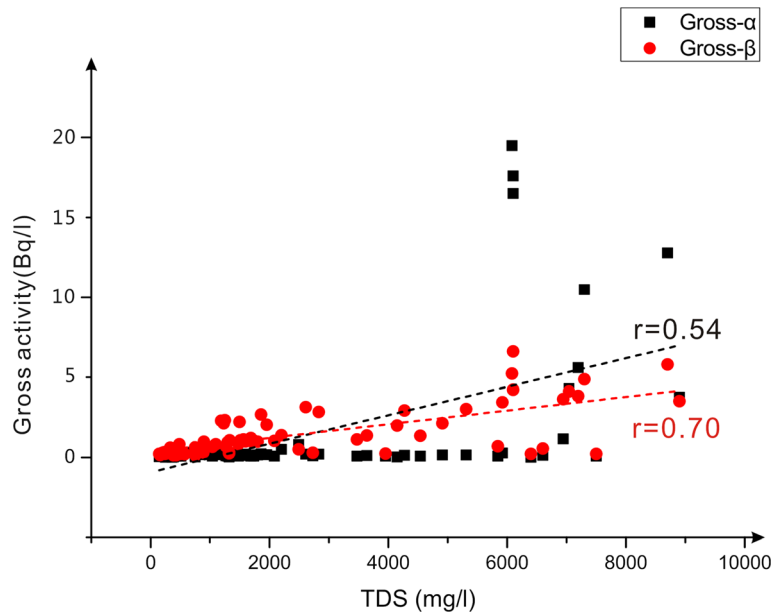
Fig. 10 Diagram showing the relationship between gross-α activity and total dissolved solids (TDS) (*black line*) and gross-β activity and (TDS) (*red line*) in groundwater samples investigated

($R = 0.6$) (Fig. 9). The effect of temperature on radionuclide distribution is difficult to evaluate. It is possible that temperature probably being a factor enhances removal of radionuclides from the aquifer body into the water phase and thus implies more emission of gross-α. It was found that in the presences of carbonate, the solubility of uranium increases with temperature in the temperature range at 15–50 °C (Yu et al. 2007).

The idea of enhanced radionuclide leaching from the aquifer body with increasing temperature is supported by the groundwater total dissolved solids (TDS). There is a relatively good correlation of TDS with both gross-α and gross-β activities at $R = 0.5$ and $R = 0.7$ (Fig. 10), respectively. The TDS values measured here

span a wide range of concentration (143–8,900 mg/l), and the positive correlation strongly suggests that radionuclide leaching from the aquifer rocks is proportional to TDS, which was also shown by studies in other areas (Duenas et al. 1997).

The level of gross radioactivity of the investigated regions here was plotted together with selected regions where gross-α and gross-β activities were determined in groundwater, bottle water, thermal water, lake water, and tap water (Figs. 11 and 12). The distribution diagrams show that groundwater of the south-eastern Arabian Peninsula is characterized by relatively higher values than the other regions (Bonotto et al. 2009; Damla et al. 2009; Zorer et al. 2009, 2013) with respect

Fig. 11 Diagram showing the distribution of gross-α activity in groundwater samples investigated here and water samples from other areas (Bonotto et al. 2009; Damla et al. 2009; Zorer et al. 2009, 2013)
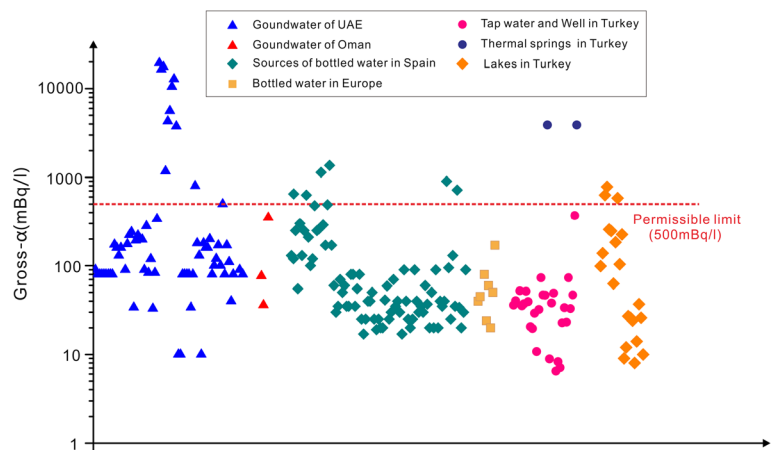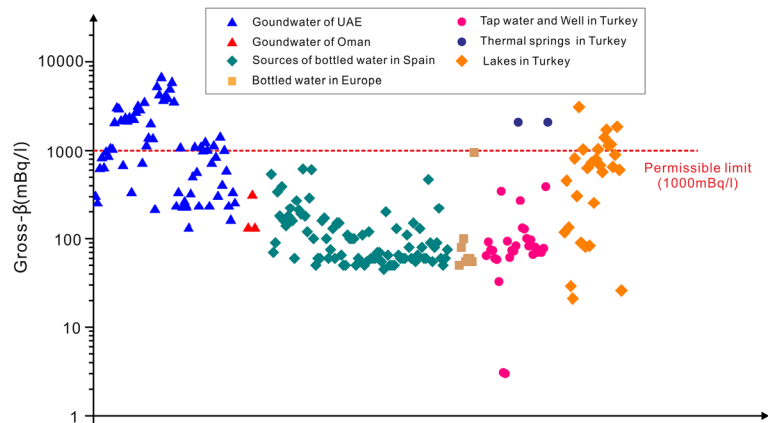
Fig. 12 Diagram showing the distribution of gross-β activity in groundwater samples investigated here and water samples from other areas (Bonotto et al. 2009; Damla et al. 2009; Zorer et al. 2009, 2013)

to gross-α and gross-β activities. This feature may relate to the climate conditions of the region which is characterized by arid to semi-arid compared to the other regions which occur in a temperate climatic zone. Investigation of uranium isotopes distribution in samples taken from carbonate aquifers in the investigated region here (Alshamsi et al. 2013) indicated a rather clear negative correlation between uranium concentrations and precipitation rates. The effect of higher precipitation rate may be attributed to dilution of radioactivity in groundwater with the associated higher recharge rate of aquifers.

Although the groundwater investigated here may not be used for human drinking purposes, it may be drunk by animals and is also heavily used for agricultural and domestic purposes. Rapid measurements of gross-α and gross-β activities in one tap water sample and two bottled water samples from the UAE indicated values <200 mBq/l comparable to the data given in Figs. 11 and 12. These results and data in Figs. 11 and 12 suggest that conventional water purification technology used is not entirely capable of removing all radioactivity from the water. Accordingly, new technology for removing radioactivity is needed, most likely a combination of different ion-exchange columns and nanoparticle filters. At present, conventional water purification system can provide, however, reduction in total radioactivity, but additional specific radioactivity purification system is important in case the groundwater will be used for drinking purposes. However, inspection should be given to the purification system and waste filters as more radioactivity will be accumulated and become a possible environmental hazard in the disposal program.

Since most of the investigated groundwater is used for agricultural activity and landscaping, this means that

radioactivity has been and will be accumulating in the surface soil and can even penetrate into deeper layers of the soil and contaminate again the groundwater or absorbed by plants. Plants absorb uranium, particularly in carbonate-rich soil where uranium can be more available in soluble forms. The environmental impact assessments of using the groundwater for agriculture and other urban activities are therefore essential to evaluate the accumulated radioactivity in such areas. This issue is vital for the environmentally sustainable future development and in particular in arid regions were groundwater and cultivated lands are valuable. Furthermore, climate change models indicate that arid regions will be most sensitive to precipitation and desertification, and thus, use and quality control of groundwater should be considered with great care for future water security and land use. To acquire a comprehensive picture about the supported radioactivity in groundwater, it is essential that the analysis of as much as possible radionuclides be conducted. This task is, however, cannot be easily performed and may be applied to those groundwater that show relatively high value of gross activity.

Conclusions

Results of gross-α and gross-β in groundwater from south-eastern Arabian Peninsula reveal the following:

1. Majority of the groundwater contain gross-α and gross-β activities below the WHO permissible limits for drinking water. Groundwater in region 4 shows radioactivity values above the WHO permissible limit.

2. Most of the gross-α activity is related to decay of uranium and thorium isotopes and in particular ^{226}Ra, a progeny produced by ^{238}U decay.

3. Main sources of the gross-β in the groundwater are attributed to ^{40}K.

4. The groundwater pH imposes little or no effect on the gross-α and gross-β, while high temperatures probably enhance leaching of radionuclides from the aquifer body and thereby increase the radioactivity in the groundwater. This conclusion is also supported by the positive correlation between radioactivity and TDS.

5. Particular water purification technology and environmental impact assessments are essential for sustainable and secure use of the groundwater in regions that show radioactivity values far above the WHO permissible limit for drinking water.

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