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Assessment of some trace heavy metals and radioactivity concentration in drinking water of Saudi Arabia

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ABSTRACT

Concentration of trace elements and natural gross radioactivity were measured in the drinking water in Jizan region (Saudi Arabia). A preliminary study on trace elements (Zn, Fe, Mn, Ni, Cu, Cr, Co, Se, Sr, V, Ti, Mo, Hg, Cd, Ba, As, Al and Pb) concentrations and natural radioactivity related to gross-α and gross-β radiations in the drinking water were determined. The obtained results showed that, in general, the trace elements concentrations in water did not exceed WHO [1], and GSO [2] guidelines. Generally, heavy metals concentration of the drinking water were found to be in the sequence of Sr > Ti > Fe > Al > Zn > Ba > As > Cu > Mo > Ni > Cr > Co > Se >Hg > Mn, respectively. The results of this study indicated that a general absence of serious pollution in the drinking water used in this region. The results obtained from the radioactivity determination indicate that the drinking water radioactivity concentration of gross-α and gross-β were ranging from 0.06±0.001 to 0.45±0.03 Bq/l and from 0.05±0.006 to 2.95±0.23 Bq/l, respectively. The gross alpha values were found to fall below the GSO and WHO recommended MCL of 0.5 Bq/l while the gross beta values in two samples only exceeds the MCL value of 1 Bq/l, respectively.

Keywords: gross alpha, gross beta, heavy elements, Saudi Arabia

1- INTRODUCTION

Water is very important for our life. It forms 50 to 60% in weight of our body and play an active role in all the vital processes of our body. Therefore, water must be free from organisms that are capable of causing disease and from minerals and organic substances that could produce adverse physiological effects [3].

Environmental contaminant is a major problem being faced by the society. One of the many pollutants in the environment is heavy metals. The presence of heavy metals in atmosphere is due to a natural and an anthropogenic origin. Heavy metals are among the most toxic pollutants present in marine, groundwater and industrial wastewater. The source of heavy metals in environment, and more specifically in water systems has been attributed primarily to man made sources, such as agricultural activities and stack emissions from industrial sources. While the toxic metal compounds turned back to earth arrive to surface waters with river, rain and snow waters, they may be mixed to groundwater by filtering from soil.

Trace elements have important effects in the life processes. The most activities have concentrated some of heavy metals in certain areas up to the dangerous levels of living organism [4]. Trace elements such as lead, mercury, arsenic, copper, zinc and cadmium are toxic for the humans even at very low level of intake and accumulated in living organisms and produce disease and disorders.

Radioactivity in waters is mainly originated from radioactive elements in the earth's crust. Surface water and especially ground water play an important role in the migration and distribution of these elements in the earth's crust [5]. Concentrations of natural radionuclides in water could be related to the physicochemical conditions and the geological formation of the area.

Natural radioactivity arises mainly from the primordial radionuclides, such as ^{40}K, and the radionuclides from the ^{238}U and ^{232}Th and their decay products, which are present at trace levels in all ground formations [6].

The monitoring of drinking water quality is necessary in order to detect pollution and to prevent use of contaminated drinking water for public water supply.

Many standards, both international and national, are available now. These standards recommend maximum permissible limits on several water quality parameters in order to avoid any adverse effect on the health of the population consuming the water.

Saudi Arabia is a desert country with no permanent rivers or lakes and very little rainfall. Water is scarce and extremely valuable, and with the country's rapid growth, the demand for water is increasing. Aquifers are a major source of water in Saudi Arabia. They are vast underground reservoirs of water. In the 1970s, the government undertook a major effort to locate and map such aquifers and estimate their capacity. As a result, it was able to drill tens of thousands of deep tube wells in the most promising areas for both urban and agricultural use.

Many research studies were carried out on the quality of drinking water that consumed in Saudi Arabia at different localities [7-20].

In Jizan, which is located in Southwestern of Saudi Arabia, there are many wells scattered throughout the region are used as the source of drinking water. As a consequence, it is of importance to investigate the state and quality of waters derived from these wells, in addition to the bottle water come from different regions.

The main objectives of this study are to evaluate the drinking water quality in this area and examine their compliance with local and international standards. The knowledge of concentrations and distributions of the heavy metals and gross-alpha and gross-beta radioactivity in water samples are of interest since it provides useful information in the monitoring of environmental contaminations [21].

2- METHDOLOGY

Waters have been analyzed at Medical research center Jizan, for 18 elements (Al, As, Ba, Cd, Co, Cr, Cu, Fe, Hg, Mn, Mo, Ni, Pb, Se, Sr, Ti, V, Zn, in addition several parameters such as pH and total dissolved solids are determined.

Eleven brands of plastic bottled water were obtained randomly from 3 supermarkets in Jizan, Saudi Arabia. Three bottles from each brand, each with a different batch number and date of bottling, were purchased. 10 treated drinking groundwater from commercial stations distributed throughout the region. Seven samples from tap water that come from desalinated water plants but most people don't use in drinking water but in other household purposes.

2.1 Chemistry

Samples were collected and stored in PET (polyethylene terephthalate) bottles (0.5 L) with PET caps completely filled, which were previously soaked in 10% nitric acid and thoroughly rinsed with deionized distilled water. The samples for heavy metals analyses were acidified to pH2 with A.R. grade nitric acid in order to minimize the absorption of metals into the wall of the containers and stored approximately at 4 ^{o}C.

Measurements of trace elements drinking waters were done by ICP-MS in addition pH and TDS of water were also determined.

The pH of water samples was determined using a pH meter (CG 817) whereas the total soluble salts were measured using an electrical conductivity (EC) meter (EC) in dS/m at 25 WC (Test kit Model 1500-20, Cole and Parmer)

ICP-MS are by far the most common type of plasma sources used in today's commercial ICP Optical Emission Spectrometry (OES) and ICP-MS instrumentation. In ICP, the first step that takes place is the desolvation of the droplet with the water molecules stripped away; it then becomes a solid particle. As the sample moves further into the plasma, the solid particle changes first into a gaseous form and then into a ground-state atom. The final process of conversion of an atom to an ion is achieved mainly by collisions of energetic argon electrons with the ground state atom. The ion then emerges from the plasma and is directed into the interface on the mass spectrometer. The measurement determines the parts per million (ppm) of elements in the sample.

2.2 Radioactivity

The gas-flow-proportional counter (Eurisys Measure- IN20) eight channel Alpha and Beta counter was used for the measurements of the gross alpha and beta in the portable water samples. Each counter channel has a window thickness of 450 μg/cm^3 and diameter of 0.06 m. The counting gas is an argon-methane mixture in the ratio of 90% to 10%. The counting system incorporates an anti-coincident guard counter used to eliminate interference from high energy cosmic radiation that would enter the measuring environment. The chambers are covered with 0.1 m lead thick to prevent part of ambient gamma rays from entering the measuring environment.

For signal processing purpose, the system is connected to a microprocessor loaded with a spread sheet programme (Quarttro-Pro) and graphic programme (Multiplan). The system can be operated at a bias voltage ~1100V where only alpha particles are detected, referred to as 'alpha only' mode. If the bias voltage is increased to ~1650 V with same gas, the counter will respond to both alpha and beta particles simultaneously. Operation at this higher voltage is referred to as 'simultaneous' or 'alpha + beta' mode. The alpha standards were ^{239}Pu whose half life is 24,200 y. The energy of alpha decay is 5.245 MeV. The beta standards were ^{90}Sr whose half life is 28.8 y and beta decay energy is 0.546 MeV. These standards were certified by CERCA LEA Laboratories in France with certificate numbers CT001/1285/001920-1927 and CT 1271/00/1778–1783, respectively. Plateau test was run with the manufacturer's calibration standards (^{239}Pu and ^{90}Sr) whose activities ranges from 133.4 to 185.7 Bq and 98.4 to 113.8 Bq, respectively in all the three operating modes. This test was run for 1800s for five cycles.

Measured Detector Efficiency: The results of the operational efficiencies of the different channels of the detector in alpha only and beta only modes are presented in Table 1. The result indicated an average efficiency of 34.42±0.92 % for the alpha

counts in alpha-only mode and 53.88±0.76 % for the beta counts in beta-only mode. These are good efficiency values for this type of counting system.

Sample collection and analysis

Samples were collected in clean 0.5 L polyethene containers with tight covers. The containers were rinsed thoroughly with the water. The samples were acidified by adding 20 milliliter (ml) of IN HNO_3 to minimize the lost of radiation to the containers wall. The samples were then analyzed.

Estimation of committed effective dose

Radionuclide may reach the gastrointestinal tract directly by ingestion or indirectly by transfer from the respiratory tract. From small intestine the radionuclide can be absorbed to the body fluids.

On average, adults are considered to consume two and half liters of water per day which corresponds to 913 L/y. The committed quantities, because of small effective half-lives, are practically realized within one year after intake [22]. In this work, the effective dose over one year was calculated using the following relation.

$$E = I\,AC \times 365$$

Where I is the daily water consumption in l/day, A is the activity/l; C is a dose conversion factor in Sv/Bq. Dose conversion factors used to calculate the internal radiation exposure by ingestion of radionuclide is 2.8×10^{-7} [23].

Table 1. Channel Efficiencies for different modes of measurement

Detector Channel No	Eff. alpha only mode %	Eff. beta only mode %
01	34.22±0.95	56.48±0.41
02	34.49±0.90	55.89±0.88
03	34.3±0.91	56.33±0.63
04	34.76±0.87	50.20±0.87
05	34.2±0.95	52.44±0.81
06	34.09±0.90	54.48±0.84
07	34.79±0.99	53.58±0.79
08	34.53±0.90	51.67±0.90
Average	34.42±0.92	53.88±0.76

3- RESULTS AND DISCUSSION

3.1 Chemical Characteristics

pH value: The pH of the bottled waters varied between 6.7 and 7.8 whereas that of treated waters was in the range of 7-8.1 and for tap water 7.1-8.3. The WHO guidelines and the GSO standards recommended 6.5-8.5 pH values for waters for drinking purposes (Table 2). These limits were respected in all types of waters.

Total dissolved solids (TDS)

The bottled waters TDS values varied between 110 and 175 mg/l whereas those of the treated ranged from 127 to 187 mg/l and from 125 to 210 mg/l for tap waters. However, dissolved solids in all samples were very much within the WHO and GSO drinking water standards (Table 2).

Trace metals

A total of 18 trace elements were analyzed and the results are shown in Table 2 whereas WHO and GSO standards have been presented in Table 3. The average values of the trace elements for all samples are given in Table 2.

Aluminum (Al): The Al level varied between 22 to 148 µg/l in the bottled samples and ranged between 78 and 188 µg/l in the treated waters while from 55 to 110 µg/l for tap waters. None of the types had values that exceeded the GSO (100 µg/l) and WHO guidelines of 200 µg/l.

Barium (Ba): In all water samples, the concentration values ranged between 0.151 and 12.07µg/l. None of the water samples had values that exceeded the maximum GSO and WHO limits 700 µg/l.

Table 2: Summary of the whole parameters measured in the Saudi drinking waters

Element	Bottled			Tap			Treated		
	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
pH	6.7	7.8	7.3	7.1	8.3	7.5	7.0	8.1	7.6
TDS	110	175	127	125	210	145	127	187	139
Al	22	148	98	55	110	83	78	188	124
As	0.235	4.08	1.51	0.45	3.65	1.87	0.788	2.878	1.585
Ba	0.151	4.723	0.934	0.26	2.56	1.59	1.733	12.07	5.305
Cd	ND			ND			ND		
Co	0.020	3.017	0.336	0.046	0.872	0.324	0.062	0.253	0.143
Cr	ND			ND			0.025	1.128	0.274

Cu	0.208	1.935	0.845	0.322	3.35	2.087	0.272	8.15	1.933
Fe	0.89	14.56	7.56	1.36	19.5	12.8	3.56	22.7	16.9
Hg	0.007	0.083	0.026	0.017	0.17	0.036	0.017	0.25	0.098
Mn	0.001	0.129	0.023	ND			0.011	0.069	0.024
Mo	0.032	0.641	0.075	0.062	1.28	0.404	0.008	1.35	0.50
Ni	0.152	1.032	0.301	ND			ND		
Pb	ND			ND			ND		
Se	-	-	0.0679	ND			-	-	0.114
Sr	1.104	199.4	66.77	10.12	55.3	33.9	12.36	364.2	164.2
Ti	11.64	68.64	24.93	1.23	25.12	6.853	13.15	43.83	25.96
V	ND			ND			ND		
Zn	ND			0.82	11.36	1.13	0.561	19.53	9.09

Chromium (Cr): The solubility of Cr in most water is low; however, with decreasing pH the solubility increases. None of the bottled waters brands had Cr above the detection limit of 0.08 µg/l whereas the treated waters had values ranged from 0.025 to 1.128 µg/l. All Cr levels were much below the maximum concentrations of 50 µg/l allowed by GSO and WHO standards.

Lead (Pb): None of the water samples were found to contain Lead in concentrations above the detection limit of 1.0 µg/l. None of the samples had significantly high Pb concentrations.

Manganese (Mn) : Manganese was found below the ICP detection limit in 81% of the bottled waters and 40% of the treated waters. However, all values were much below the WHO maximum limits of 400 µg/l.

Cobalt (Co): The minimum and maximum Co concentrations for bottled waters were 0.02 and 3.017 µg/l, respectively. The treated waters had values between 0.062 and 0.253 µg/l while vary from 0.046 to 0.872 µg/l for tap water. According to GSO, as well as other international organizations, there is no maximum admissible concentration for cobalt.

Copper (Cu): The concentration of Cu in bottled water varied from 0.208 to 1.935µg/l and from 0.27 to 8.15 µg/l for treated waters while from 0.332 to 3.35 µg/l for tap water. According to GSO, Cu concentration should not exceed 1000 µg/l.

Iron (Fe): Iron was present in all samples with levels ranging between minimum and 22.7 µg/l. only three bottled and 1 treated samples had concentrations below the detection limit (0.08 µg/l). None of the samples had concentrations above the GSO and WHO limit of 300 µg/l.

Molybdenum (Mo): The Mo concentration in all samples in the range of minimum to 1.35 µg/l. According to GSO and WHO standards, Mo concentration should not exceed 70 µg/l.

Selenium (Se): only one treated sample and one bottle sample were found to contain Se concentration. None of the samples exceeded the maximum limit of 10 µg/l set by GSO and WHO standards.

Zinc (Zn): Zinc was found below the ICP detection limit in all bottled water. The obtained value in the treated water was found to range between 0.561 to 19.53 µg/l. According to WHO, Zn concentration should not exceed 3000 µg/l.

Titanium (Ti): None of the water samples were found to contain Titanium in concentrations above the detection limit ICP. According to GSO, as well as other international organizations, there is no maximum admissible concentration for titanium.

Nickel (Ni): None of the bottled waters were found to contain Nickel in concentrations above the detection limit. Only 36% from the treated water had Ni above the ICP detection limit. According to WHO, Ni concentration should not exceed 70 µg/l.

Vanadium (V): Although there are no data on V oral toxicity, none of the water samples were found had V values above the ICP detection limit.

Cadmium (Cd): Humans are exposed to Cd as a result of its ingestion from food or water, with the major contribution coming from food. The Cd level in the bottled water was found below the ICP detection limit while one treated water sample was found above the detection limit. According to GSO and WHO limits, Cd concentration should not exceed 3 µg/l.

Mercury (Hg): Mercury is one of the earth's rarest elements. In its natural state it occurs mainly in combination with sulfur. Only 17 % of the bottled water while 20% from the treated waters were found values below the ICP detection limit. The values ranged from BDL to 0.25 µg /l. According to WHO, Hg concentration should not exceed 6 µg/l.

Table 3: WHO guideline values and GSO normative limits for drinking water (µg/l).

Parameter	GSO, 2009	WHO, 2011
pH	6.5-8.5	6.5-8.5
TDS	100-600 mg/l	1000mg/l
Al(µg/L)	100	200
AS	10	10
Ba	700	700
Cd	3	3
Co	-	-
Cr	50	50
Cu	1000	2000
Fe	300	300
Hg	1	6
Mn	10	400
Mo	70	70
Ni	20	70
Pb	20	10
Se	10	10
Sr	-	-
Zn	100	3000
V	-	-
Ti	-	-

Arsenic (As): Arsenic is widely and evenly distributed in solids and water in low concentrations. Generally, the earth crust contains an average of 2 mg kg^{-1} or less of arsenic. Most of the arsenic in water occurs naturally from erosion of rock surfaces. Where arsenic concentrations are abnormally high, the source is usually industrial. Arsenic (As) concentration in all samples was ranged between 0.235 to 4.08 µg/l. According to GSO and WHO, Arsenic concentration should not exceed 10 µg/l.

Strontium (Sr): The Sr in naturally occurring element and the guideline value (4200 µg/L) for Sr is very high, compared to all trace elements.

The range of Sr in bottled water varied from 1.104 to 199.4 µg/l, 12.36 to 364.3 µg/l in treated water while from 10.12 to 55.3 µg/l in tap water. However, the recorded values in all three water types did not exceed the guideline value.

Results show that the drinking water quality in Jizan generally meets the international standards. Toxic elements such as Cd, Cr, Ni, V, and Pb were either present in minute quantities or not detectable at all, and were below the WHO and GSO guideline values. As such, human consumption of treated drinking water or bottled drinking water in Jizan should not lead to adverse health effects typically associated with these elements. The levels of trace elements in the bottled and treated drinking water samples do not suggest any implications for human health, based on international/national guideline values (Table 3).

It is noteworthy that none of the measured elements in drinking water samples in this study exceeded the international guideline values. Therefore, one can conclude that the drinking water quality in Jizan region complies with international guidelines.

3.2 Radioactivity

From the laboratory results of the gross alpha and gross beta activities, indicate the gross alpha activity ranges from 0.06 ± 0.001 Bq/l – 0.45± 0.03 Bq/l, with an average of 0.19 ± 0.033 Bq/l. The gross beta activity ranges from 0.05 ± 0.006 Bq/l – 2.95 ± 0.23 Bq/l, with an average of 0.54±0.05 Bq/l (Fig. 1). However, a comparison to the world standard values of the values obtained from the laboratory results for the gross alpha and beta activities are presented.

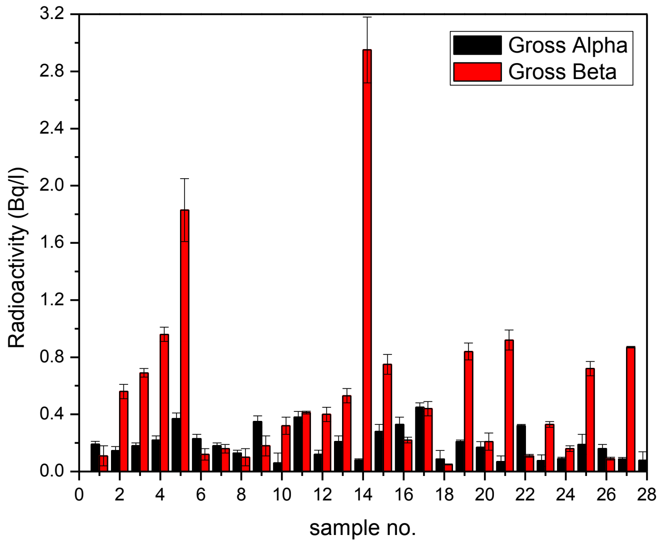

Fig 1: The gross alpha and beta activities in portable water

The study results reveals that; all the values of the gross alpha activity concentrations are within the recommended upper limit value of 0.5 Bq/l while all the values of the gross beta activity concentrations are lower than recommended upper limit value of 1Bq/l except two samples. Hence, the drinking water of the study area (Jizan) is radioactively safe to use. The gross alpha and beta activity concentrations obtained compare well with other values reported in literatures as presented in Table 4

Table 4. Comparison of the gross α and β concentrations determined in portable drinking water samples with those reported in other countries

Country	Gross alpha (mBq/l)	Gross Beta (mBq/l)	Reference
Jordan	29-3146	0-5014	[24]
Nigeria	216-1299	64-582	[25]
Greece	82	283	[26]
Brazil	1-400	120-860	[27]
Nigeria	5.8-174	14.7-222.5	[28]
Turkey	192	579	[29]
Saudi Arabia	194	540	This study

The estimated committed effective dose values are presented in Table 5. The committed effective dose due to the gross alpha activity concentrations within Jizan region ranged from 0.015 mSv/y to 0.1 mSv/y with a mean value of 0.05 ± 0.008 mSv/y (0.015-0.097, 0.018-0.1 and 0.019-0.08 mSv/y) for bottled, treated and tap waters, respectively. The estimated mean committed effective dose are within 0.1 mSv/y reference dose level [30]. This study therefore shows that the consumption of drinking water does not pose any health burden to the population.

A comparison of the level of dose intake by population from the three water sources show that the dose intake from treated water > dose intake from bottled water > dose intake from tap water.

CONCLUSIONS

The study clearly indicates the most water of Jizan region to be free from any obvious pollution (according to national and international standards).

The gross alpha and beta activity concentrations in portable drinking water samples collected from the different areas in Jizan region were determined to investigate the radiological burden to the population. The data obtained can be used as a baseline for ascertaining possible changes in radioactivity concentrations in portable drinking water samples in this region. From the present work, it can be inferred that the drinking water samples from the study area have low radioactivity and all the results measured are below the World Health Organization, drinking water guideline values of 1.0 Bq/l for the gross beta radioactivity, and value of 0.5 Bq/l for gross alpha activity. Hence, the drinking water of the study area (Jizan, KSA.) is not radioactively contaminated, or rather is radioactively safe to use.

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