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Research Article
Risk Assessment of Some Radioactive and Elemental Content from Cement and Phosphate Fertilizer Consumer in Saudi Arabia

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Abstract

Objective: The present study deals with measuring the activities of ^{226}Ra, ^{232}Th and ^{40}K in cement and phosphate fertilizer consumed in Saudi Arabia, using γ-ray spectrometry. From the measured γ-ray spectra, specific activities were determined. **Methodology:** The samples were collected from local markets in Saudi Arabia. The results indicate that the cement and phosphate fertilizer consumed in Saudi Arabia does not pose any significant radiological risk and they are safe for use. **Results:** The obtained results were compared with the reports gotten from similar studies conducted in other countries. The radiation hazard parameters were calculated for all samples and it was shown that the radium equivalent Ra_{eq} and the representative level index, $I_{\gamma r}$ are lower than the acceptable level of 370 Bq kg^{-1} for radium equivalent Ra_{eq}, 1 for level index $I_{\gamma r}$. Thermo Scientific Quant'X EDXRF spectrometer were used also to determine the concentrations of the elements Si, Al, Fe, Ti, Ca, Na, S, Pb, Sn, As and Zn as major, trace and toxic elements. **Conclusion:** The data presented here was useful to those dealing with portland cement, phosphate fertilizer chemistry and related fields.

Key words: Consumed products, portland cement, phosphate fertilizer, γ-ray spectrometry, EDXRF spectrometer

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Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

Human beings are exposed to radiation from different sources in their daily lives. Some of these radiations come from the natural radioactive materials on the earth, such as the natural radioactive series or their products, which can be transferred through air or water to the people in direct or indirect ways. Most of these products are produced from the recycling of materials, which may contain some radioactive isotopes, especially because of the nuclear industries waste used in these products or as a result of the Natural Occurring Radioactive Material (NORM), which are obtained from some oil and gas industries or in water treatment plants[1].

Nowadays, the use of consumer products, which are usually made of natural materials or recycled materials can be contaminated with some level of radioactive or toxic elements. Investigations into the radioactivity of some consumer products have previously been conducted worldwide[2]. The activity of naturally occurring radioisotopes in building materials have also been investigated[3-6].

However, very few studies have investigated both the radioactivity and the toxicity of these products as well as measured the natural radioactivity in the marble used in Saudi Arabia. They investigated the activities of ^{238}U, ^{232}Th and ^{40}K as well as the radium equivalent activity and radiation hazard indices. This study concluded that the majority of the measured commercial samples of marbles are considered safe for construction use[7].

This study deals with the risk assessment of radioactive materials from portland cement and phosphate fertilizers consumer products in Saudi Arabia, using gamma spectroscopy for radioactive analysis and x-ray fluorescence for heavy and toxic elements.

MATERIALS AND METHODS

Sampling and sample preparation: In this study, 18 cement and phosphate fertilizer samples were collected from local markets in Saudi Arabia for the investigation of both radioactive and heavy elements that occur naturally. Gamma spectroscopy was used for radioactivity analysis and Thermo Scientific Quant'X EDXRF spectrometer to determine the heavy elements concentration.

Gamma-ray spectrometry: Each sample about 1 kg in weight was dried in an oven at a temperature of about 110°C to ensure that moisture is completely removed, the samples were crushed, homogenized and sieved through a 200 mm mesh, which is the optimum size to be enriched in heavy minerals. Weighted samples were placed in a polyethylene beaker having a volume of 250 cm^3. The beakers were completely sealed for 4 weeks to reach secular equilibrium, where the rate of decay of the radon daughters become equal to that of the parent. This step is necessary to ensure that radon gas is confined within the volume and the daughters remain in the sample.

Instrumentation and calibration: Gamma spectrometric measurements were performed with NaI (Tl) detector. The measuring time for gamma ray spectra range was 12 h. In order to determine the background distribution, due to naturally occurring radio nuclide in the environment around the detector, an empty polystyrene container was counted in the same manner as the samples. After measurement and subtraction of the background, the activity concentration was calculated[8]. The specific activity of ^{226}Ra was evaluated from gamma-ray lines of ^{214}Bi at 609.3, 1120.3 KeV and ^{214}Pb at 351 KeV, while the specific activity of ^{232}Th was evaluated from gamma-ray lines of ^{228}Ac at 338.4, 911.1 and 968.9 KeV. The specific activity of ^{40}K was determined directly from its 1460.8 KeV gamma-ray line[9,10].

Calculations of count rates for each detected photopeak and radiological concentrations (activity per mass unit or specific activity) of the detected radio nuclides depend on the establishment of secular equilibrium in the samples. The activity concentration in Bq kg^{-1} (A) in the environmental samples was obtained as follows:

$$A = \frac{N_p}{e \times \eta \times m} \tag{1}$$

where, N_p is (cps) sample-(cps) back ground, e is the abundance of the γ-peak in a radio nuclide, η is the measured efficiency for each gamma- ray peak observed for the same number of channels either for the sample or the calibration source and m is the sample mass in kilograms.

Radiation hazard parameters: The natural radioactivity of building materials is usually determined from ^{226}Ra, ^{232}Th and ^{40}K contents. As Ra and its daughter products produce 98.5% of the radiological effects of the U series, the contribution from the ^{238}U has been replaced with the decay product ^{226}Ra. The γ-ray radiation hazards due to the specified radio nuclides were assessed using three different indices[11,12]. Radium equivalent activity is an index that has been introduced to represent the specific activities of ^{226}Ra, ^{232}Th and ^{40}K by

Table 1: Activity levels of ^{226}Ra, ^{232}Th, ^{40}K and radiation hazard parameters of cement consumer products in Saudi Arabia

		Activity concentration ($Bq\ kg^{-1}$)					
Sample No.	Sample type	^{226}Ra	^{232}Th	^{40}K	Ra_{eq} ($Bq\ kg^{-1}$)	Dose rate ($nGy\ h^{-1}$)	$I_{\gamma r}$
C1	Qassim portland	13.33±0.69	14.15±0.74	112.0±6.10	42.17	20.23	0.02
C2	Qassim portland	16.53±0.87	07.44±0.38	195.0±9.70	40.93	20.42	0.03
C3	Riyadh portland	17.72±0.86	18.56±0.97	236.0±11.0	60.80	30.01	0.05
C4	Riyadh portland	13.15±0.92	13.69±0.72	206.0±10.0	47.18	23.55	0.04
C5	Yamama portland	14.75±0.76	16.43±0.86	113.0±5.60	46.17	22.04	0.05
C6	Yamama portland	17.53±0.89	19.63±1.00	132.0±6.60	54.91	26.20	0.06
C7	Hail portland	8.580±0.37	9.330±0.48	90.55±4.50	28.26	13.73	0.01
C8	Hail portland	13.15±0.68	13.69±0.72	206.6±10.0	47.17	23.55	0.02
C9	White cement	13.76±0.70	17.51±0.89	108.6±5.30	46.38	2212	0.05
C10	White cement	11.26±0.57	15.24±0.78	108.0±4.70	39.74	19.00	0.04

a single quantity, which takes into account the radiation hazards associated with them. This first index can be calculated according to Beretka and Mathew[13] as:

$$Ra_{eq} = A_{Ra}+1.43A_{Th}+0.077A_{K} \tag{2}$$

where, A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in $Bq\ kg^{-1}$, respectively. The Ra_{eq} is related to both the external γ-dose and internal dose due to radon and its daughters. The maximum value of Ra_{eq} in building materials must be less than 370 $Bq\ kg^{-1}$ for safe use.

Conversion factors to transform specific activities A_K, A_{Ra} and A_{Th} of K, Ra and Th, respectively into absorbed dose rate at 1 m above the ground (in $nGy\ h^{-1}$ by $Bq\ kg^{-1}$) are calculated by the Monte Carlo method and the values are UNSCEAR[14]:

$$D\ (nGy\ h^{-1}) = 0.0417A_K+0.462A_{Ra}+0.604\ A_{Th} \tag{3}$$

Another radiation hazard index called the representative level index, $I_{\gamma r}$ is defined as follows NEA-OECD[15]:

$$I_{\gamma r} = \left(\frac{A_{Ra}}{150\ Bq\ kg^{-1}}+\frac{A_{Th}}{100\ Bq\ kg^{-1}}+\frac{A_K}{1500\ Bq\ kg^{-1}}\right) \tag{4}$$

where, A_{Ra}, A_{Th} and A_K have the same meaning as in Eq. 2.

EDXRF spectrometer: The XRF data presented here are quantitative, in that they are derived from "Filtered" intensity values which are ratios to the appropriate x-ray continuum regions, through a least squares fitting formula rather than plotting the proportions of the net intensities in a ternary system[16]. All analysis for this study were conducted on a Thermo Scientific Quant'X EDXRF spectrometer, located in the Department of Physics, Qassim University. It is equipped with a thermo-electrically Peltier cooled solid-state Si (Li) x-ray detector, with a 50 KeV, 50 W, ultra-high-flux end window bremsstrahlung, Rh target x-ray tube and a 76 μm beryllium (Be) window (air cooled), that runs on a power supply operating at 4-50 KeV/0.02-1.0 mA at 0.02 increments. The spectrometer is equipped with Edwards's vacuum pump, allowing for the analysis of lower-atomic-weight elements between sodium (Na) and titanium (Ti). Data acquisition is accomplished with a pulse processor and an analogue-to-digital converter. Elemental composition is identified with digital filter background removal, least squares empirical peak deconvolution, gross peak intensities and net peak intensities above background. The samples were ground into a fine powder using an agate motor. All powder samples were dried at a temperature of 105°C and stored in desiccators, until they were analysed. Fill up ¾ parts of the cup with the sample analyzed. Then, cover the cup with the film. Fix the slim ring by sliding it down and fix the thick ring at the top of the cup. When the cups are prepared, place them on the sample stage inside the spectrometer chamber with the side to be analyzed facing down toward the x-ray tube).

RESULTS AND DISCUSSION

Radioactivity concentrations: The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K and radiation hazard parameters in the consumer products used in Saudi Arabia have been determined and divided into two categories as follows.

Cement: Table 1 shows the specific activities of ^{226}Ra, ^{232}Th and ^{40}K and the radiation hazard parameters of portland and white cement from different factories in Saudi Arabia, Qassim, University Riyadh, Hail and Yamamah were consumed in Saudi Arabia. The activity concentration of ^{226}Ra ranged between 8.58±0.37 $Bq\ kg^{-1}$ from Hail to 17.72±0.86 $Bq\ kg^{-1}$ from Riyadh with an average value of 14.34 $Bq\ kg^{-1}$. While the activity concentration of ^{232}Th ranged between 7.44±0.038 $Bq\ kg^{-1}$ from Qassim to 19.63±1.00 $Bq\ kg^{-1}$

Table 2: Activity levels of ^{226}Ra, ^{232}Th, ^{40}K and radiation hazard parameters of phosphate fertilizer consumer products in Saudi Arabia

		Activity concentration (Bq kg^{-1})			
Sample No.	Sample type	^{226}Ra	^{232}Th	^{40}K	Ra_{eq} (Bq kg^{-1})
F1	SSP fertilizer	51.6±3.60	9.2±1.50	462.6±21.0	100s
F2	SSP fertilizer	53.8±3.60	8.1±1.40	580.8±13.0	110
F3	TSP fertilizer	106.0±7.00	6.1±2.40	560.3±19.0	157
F4	TSP fertilizer	101.4±8.20	6.5±1.20	567.9±23.0	154
F5	DAP fertilizer	12.1±1.30	30.4±1.26	51.6±4.38	55
F6	DAP fertilizer	9.1±1.30	36.4±1.79	45.6±3.27	63
F7	MAP fertilizer	17.3±1.86	42.7±3.22	78.6±4.38	83
F8	MAP fertilizer	15.7±1.10	34.5±2.67	71.2±4.17	75

Table 3: Activity concentrations of consumed products in Saudi Arabia compared with those of other countries

Countries	Sample type	^{226}Ra	^{232}Th	^{40}K	References
China	Portland cement	118.7	36.1	444	Lu *et al.*[19]
Egypt	Portland cement	35.60	43.17	82.08	El-Taher[20]
Malaysia	Portland cement	34.7	32.9	190.6	Majid *et al.*[21]
Nigeria	Portland cement	30.2	24.6	251.3	Agbalagba *et al.*[22]
Pakistan	Portland cement	37	28	200	Rahman *et al.*[4]
Austria	Portland cement	27	14	210	Trevisi *et al.*[23]
Nigeria	White cement	41.9	30.1	340.2	Agbalagba *et al.*[22]
Iraq	White cement	31.0	3.8	10.2	Ali[24]
Turkey	White cement	12.5	2.7	1141.9	Baykara *et al.*[25]
Saudi Arabia	Portland cement	14.34	14.11	161.25	Present study
Brazil	SSP	375	100	871	Favaro *et al.*[26]
Pakistan	SSP	221	49.7	556	Khan *et al.*[27]
Nigeria	NPK	143	9	4729	Jibiri and Fasae[28]
Germany	NPK	520	15	720	Khan *et al.*[27]
Brazil	NPK	647.6	753.9	603	Becegato *et al.*[29]
Saudi Arabia	SSP	52.7	8.65	521	Present study
Saudi Arabia	TSP	103.5	6.3	564	Present study
Saudi Arabia	DAP	10.6	33.5	48.6	Present study
Saudi Arabia	MAP	16.5	38.6	77.9	Present study

from Yamama, with an average value of 14.11 Bq kg^{-1}. However, the activity concentration of ^{40}K ranged between 90.55±4.50 Bq kg^{-1} from Hail to 236±11 Bq kg^{-1} from Riyadh with an average value of 161.25 Bq kg^{-1}. The radionuclide concentrations of cement products consumed in Saudi Arabia are below the world averages for building materials 35 (Bq kg^{-1}), 50 (Bq kg^{-1}) and 500 (Bq kg^{-1}) for ^{226}Ra, ^{232}Th and ^{40}K, respectively[17]. These materials do not pose a significant radiological hazard when used for building construction radium equivalent (Ra_{eq}) ranged between (28.60-60.80 Bq kg^{-1}) for portland cement and from (39.74-46.38 Bq kg^{-1}) for white cement. The absorbed dose rate, the external hazard index, Hex ranged between (13.74-30.01 Bq kg^{-1}) and (0.01-0.06 nGy h^{-1}), respectively. The average values of the radiation hazard parameters, for all cement products under investigation are lower than the acceptable level 370 Bq kg^{-1} for radium equivalent (Ra_{eq}) and 59 (nGy h^{-1}) of the absorbed dose rate and ≤ 1 for the external hazard index[15].

Phosphate fertilizer: Table 2 shows the specific activities of ^{226}Ra, ^{232}Th and ^{40}K and the radiation hazard parameters in DAP, MAP, Single Super Phosphate (SSP) and triple super phosphate fertilizers consumed in Saudi Arabia. The concentration of ^{226}Ra and ^{40}K in single and triple super phosphate is higher than that (the concentration) in DAP and MAP. While, the concentration of ^{232}Th in single and triple super phosphate is lower than that in DAP and MAP. The radium equivalent (Ra_{eq}), for phosphate fertilizer ranged between (55-157 Bq kg^{-1}). The average values of the radiation hazard parameters for all phosphate fertilizers under investigation, are lower than the acceptable level, 370 Bq kg^{-1} for radium equivalent (Ra_{eq})[15]. The variations in the NOR specific activities in phosphate fertilizers from different brands is due to the different sources of phosphate ore, different geological origin of the phosphate ore and/or chemical processing of the ore during fertilizers manufacture.

Table 3 shows a comparison between the consumed products in Saudi Arabia with those from other countries. With respect to cement, the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K for portland cement consumed in Saudi Arabia is comparable with the corresponding values of other countries. The radioactivity in portland cement varies from one country

Table 4: Major, minor and toxic elements content in cement and phosphate fertilizer consumed in Saudi Arabia

Sample type	Si (%)	Al (%)	Fe (%)	Ti (%)	Ca (%)	Na (%)	S (%)	Pb (ppm)	Sn (ppm)	As (ppm)	Zn (ppm)
Yamamah cement	13.64	9.84	4.26	0.289	64.70	-	1.85	0.001	0.011	-	-
Riyadh cement	15.41	9.59	3.86	0.09	61.59	-	1.74	0.005	0.013	0.012	0.015
Qassim cement	44.95	18.44	4.71	1.38	3.300	-	-	-	0.009	0.020	-
Hail cement	12.70	9.80	1.69	0.095	33.16	24.97	0.90	0.004	0.004	0.008	-
Super phosphate	8.89	0.008	1.73	0.08	41.53	39.87	-	-	0.012	-	0.018
DAP	-	-	-	12.91	1.650	53.57	-	8.430	0.840	11.08	-
MAP	-	-	7.19	0.013	0.134	23.59	16.97	-	0.016	0.110	0.154

to another because of different materials that are used in the cement manufacture. The radioactivity in portland cement varies from one country to another because of different materials that are used in the cement manufacture[18]. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for the investigated phosphate fertilizers were compared with similar investigations in other countries and presented in Table 3.

The radioactivity in fertilizers varies from one country to another. It is important to point out that these values are not representative values for countries mentioned, but are representative of the region from where the samples were collected[30].

Heavy elements concentration: Cement and phosphate fertilizer samples purchased from markets in Saudi Arabia were tested, using XRF analysis. This was performed using a cost-effective XRF system, assembled at the University of Qassim. The major elements that form the elemental contents of cement and phosphate fertilizer samples are summarized in Table 4. The uncertainties shown in the results are estimates of experimental errors contributed by calibration and the peak height of the respective spectrum. The values quoted are the average of two analyses (repeat) of the same sample. The Si, Al, Fe, Ti, Ca, Na and S were measured as major elements. The Pb, Sn, As, Zn as trace and toxic elements. It is important to point out that the different values presented in Table 4 due to the location and the regions from where the samples were collected.

CONCLUSION

In this study, activity levels of 18 cement and phosphate fertilizer samples, which were collected from local markets in Saudi Arabia have been measured, using gamma-ray spectrometer. The radium equivalent activity Ra_{eq} and the representative level index, $I_{\gamma r}$ were evaluated to assess the potential radiological hazard associated with these materials. The results show that the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in all the samples do not pose any significant radiological risk and they are safe for use. The average Ra_{eq} values of the studied samples ranged from 11.5-228 Bq kg^{-1}, which are below the internationally accepted values (370 Bq kg^{-1}) and the representative level index, $I_{\gamma r}$ are less than unity. No risk assessments were found. In conclusion, this study shows that, most of these materials do not pose any significant threat of radiation hazard and are safe for use in the construction of dwellings. Thermo Scientific Quant'X EDXRF spectrometer was used to determine the concentrations of Si, Al, Fe, Ti, Ca, Na, S, Pb, Sn, As and Zn for the same samples as major, trace and toxic elements.

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