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Natural radioactivity of some Egyptian materials used in glasses manufacturing and glass ceramics

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► ***Original article***

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ABSTRACT

Background: The new glasses from harmful environmental waste such as cement dust; limestone phosphate, sand and borax (Genkare) were manufactured. Investigation of the radioactivity present in these materials (Phosphate rock, cement dust, limestone, sand and borax) enables one to assess any possible radiological hazard to humankind by such materials. ***Materials and Methods***: Fifteen samples were collected from five locations. Activity measurements have been performed by gamma-ray spectrometer, employing a high-resolution scintillation detector NaI (Tl) crystal 3 ×3 inch. In addition, the radiological hazards were calculated for the investigated samples. ***Results:*** The average values of activity ranged from 28±2 to 163±12, 2.8±0.7 to 40±3 and from 49±4 to 1337±74 Bq kg-1 for ^{226}R, ^{232}Th and ^{40}K, respectively. The values of absorbed dose rates, radium equivalent activities and annual effective dose due to 226Ra, 232Th and 40K respectively, are ranged from 22.05 to 101.59 nGy h-1, 45.90 to 224.22 Bq kg^{-1} and 27.04 to 124.59 µSv y-1. In addition, the values of external hazard index, internal hazard index and gamma index have been calculated. ***Conclusion:*** According to the obtained results, all materials would not present a significant radiological hazard except phosphate. The results of the study could serve as important baseline radiometric data for future epidemiological studies and monitoring initiatives.

Keywords: *Natural radioactivity, glass ceramics radiological hazards, phosphate rock, cement dust, limestone, sand, borax and glasses.*

INTRODUCTION

Natural radioactivity has always been present and broadly distributed in the earth's crust and the atmosphere [1] and it exists in various geological formations like air, rocks, plants, sand, water and soils. It is found also in our building materials constituting main sources of radiation exposure for human beings [2]. Soil radionuclide activity concentration is one of the main determinants of the natural background radiation [3]. Volcanic geographic structures as well as rocks that are rich in phosphate, granite and salt contain natural radionuclides like ^{238}U, ^{232}Th and ^{40}K [4]. The naturally occurring radionuclides are present in the rock, soil and are easily transported into the environment through plants and water [5, 6]. Long-term exposure to uranium (U) and radium (Ra) through inhalation has several health effects such as chronic lung diseases, acute leucopoenia, anemia, and necrosis of the mouth. Radium causes bone, cranial, and nasal tumors. Thorium exposure can cause lung, pancreas, hepatic, bone, and kidney cancers and leukemia [7]. Hence, humans should be aware of their natural environment with regard to the radiation effects due to the naturally occurring and induced radioactive elements.

The phosphatic rocks were considered as

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natural sources of U, where the U metal of the sea water enriches the sedimentary phosphatic sediments during deposition and diagenesis mentioned that, Sea water is the important source of uranium in marine sediments in the form of dissolved U^{+6} linked to a carbonate complex [8]. Phosphate rocks are the starting materials for the production of all phosphate products and the main source of phosphorus for fertilizers. Phosphate rocks can be of sedimentary, which represent about 85% of the phosphate rocks, was formed mainly from organic residues, the remaining parts of the phosphate rock, are of volcanic origin [9].

Building materials such as sand, soil, bricks and gravel aggregates contain a trace amount of ^{226}Ra which generates radon. However, only a fraction of radon from radioactive material becomes able to escape to the atmosphere that can be transported to an indoor environment through diffusion and convective flow. The amount of activity released per unit surface area per unit time from a material is termed as the exhalation rate. It depends on the radioactive content of the materials, emanation factor and diffusion coefficient of radon in that material, porosity and density of the material [10].

In Egypt, a large amount of limestone dust come into being in Minia and Assuit during quarrying operation. Currently, the block of limestone is extracted via chainsaw, diamond wire and diamond saws from quarries material. The limestone processing includes crushed limestone production. It causes dust, environmental problem and pollutions because of its fine nature. It contaminates the air with the storms in the summer and spring seasons and therefore causes serious health hazards including asthma. The presence of some materials such as phosphate rock, cement dust, limestone, sand and borax in Egypt, push us to care and study the radiological hazards arising from these natural radioactive materials.

The present study has been carried out to establish a consistent radiological database for the concentration of the natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in some Egyptian materials used in glasses manufacturing and glass ceramics, to estimate their radiological impacts for populations and worker.

MATERIALS AND METHODS

Samples description

Sand is a granular material made up of fine rock particles [11]. The most common constituent of sand is silica (silicon dioxide, SiO_2), usually in the form of quartz. The composition of sand varies according to local rock sources and conditions. The most common components of sand are listed in table 1.

Limestone (bricks) may be made from clay, shale, soft slate, and calcium silicate. Limestone rocks are sedimentary rocks that are made of the mineral calcite, which comes from the beds of evaporated seas and lakes and from seashells [9]. Large amounts of waste accumulate in every year in all countries which have been running limestone quarries. Waste limestone sawdust (WLS) or dust is abandoned and causes certain serious problems such as environmental problems and health hazards [12]. In Egypt, a large amount of limestone dust comes into being in Minia during quarrying operation. Currently, the block of limestone is extracted via chainsaw, diamond wire and diamond saws from quarries material.

Phosphorites (Phosphate rock) is a general term which refers to rock with a high concentration of phosphate minerals, most commonly of the apatite group {$Ca_5 (PO_4)_3$[F, OH or Cl]}. The phosphate rock is used to manufacture phosphate fertilizers and industrial products and also the only significant global resource of phosphorus used in animal feed supplements, food preservatives, anti-corrosion agents, cosmetics, fungicides, ceramics, water treatment and metallurgy. This supported the phosphorus released from sedimentary phosphate rock containing a high concentration of heavy elements was mainly originating from inorganic phosphorus fraction [13]. The phosphatic rocks can be used in glass manufacturing and porcelain. It enhanced verification reduced the firing temperature and improved the strength of the fired products [14].

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The most common constituents of phosphate rock are listed in table 1.

Cement Kiln Dust (CKD) is a fine powdery material similar in appearance to Portland cement. In Egypt, there are large quantities of produced CKD during the manufacture of cement clinker by the dry process [15, 16]. The CKD contains a mixture of raw feed as well as calcined materials with some volatile salts and oxides. Thus, it could be used as a raw material for glass manufacture. The major constituents of CKD are alumina, silica, metallic oxides, and clay (the primary constituents of cement itself) [17]. Pollution outputs from cement manufacturing plants include process waste, primarily cement kiln dust; air emissions; waste-water; plant maintenance waste, etc CKD is the largest waste stream from cement plants. It is commonly collected in bag houses installed in the grinders and is disposed of as non-hazardous waste [18]. The most common constituent of cement kiln dust is listed in table 1.

Borax, the term "borax" is used to refer both mineral and refined compound with numerous applications. The mineral takes the form of colorless to white soft crystals, which can sometimes be tinged with brown, yellow, or green. When struck against another material, borax leaves a crumbly white streak. The substance is also known as sodium tetraborate or disodium tetraborate, and it has been known to humans for thousands of years. The mineral is a chemical compound of the element boron, and the chemical formula is ($Na_2B_4O_7.10H_2O$) which decomposes by heat into (Na_2O and $2B_2O_3$). Borax and related salts were used in the preservation of both food and mummies, as well as to make pottery glazes and as a cleaning material. The most common constituents of commercial borax are listed in table 1. The chemical composition of the used tailings was analyzed using X-ray fluorescence technique model (JEOL JSX 3222), and their results were listed in table 1.

Table 1. Samples chemical composition using X-ray Fluorescence [19, 20].

Oxides	Limestone	Phosphorites	Borax	Sand	Cement dust
CaO	97.81	76.62	6.8	0.22	80.68
B_2O_3	-	-	57.2	-	-
Na_2O	-	-	28.6	-	-
SiO_2	-	4.21	4.3	96.47	9.13
Fe_2O_3	0.079	1.91	-	-	0.33
Al_2O_3	-	-	-	1.17	1.79
P_2O_5	-	12.88	-	-	0
SO_3	-	1.35	-	-	6.24
Cd O	-	0.93	-	-	-
SrO	0.078	0.32	0.3	-	-
Co_3O_4	0.059	-	0	-	-
CuO	0.067	-	0	-	-
RuO_2	0.65	-	0	-	-
Er_2O_3	0.09	-	0	-	-
Lu_2O_3	0.1	-	0	-	-
BaO	-	-	1.3	0.35	-
MgO	-	-	-	-	0.15
Cl	-	-	-	-	1.26
TiO_2	-	-	-	-	0.42

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Samples collection and preparation

Three samples from each material (phosphate rock, cement dust, limestone, sand and borax) were collected from five locations: El -Sibaiya mine (Aswan), Cemex factory (Assiut), a quarry (Minia), South Sinai and a market (Assiut)) respectively, as shown in (figure 1). The collected samples, each is about 300 gm in weight, was dried in an oven at about 110 °C for 24 h to ensure that moisture is completely removed. The samples were crushed, homogenized, and sieved through a 200 μm, which is the optimum size enriched in heavy minerals. Samples were placed in polyethylene beaker of 200 mL each and weighted. The beakers were completely sealed for 4 weeks to reach secular equilibrium when the rate of decay of the daughters becomes equal to that the parent. This step is necessary to ensure that radon gas is confined within the volume and the daughters will also remain in the sample [21].

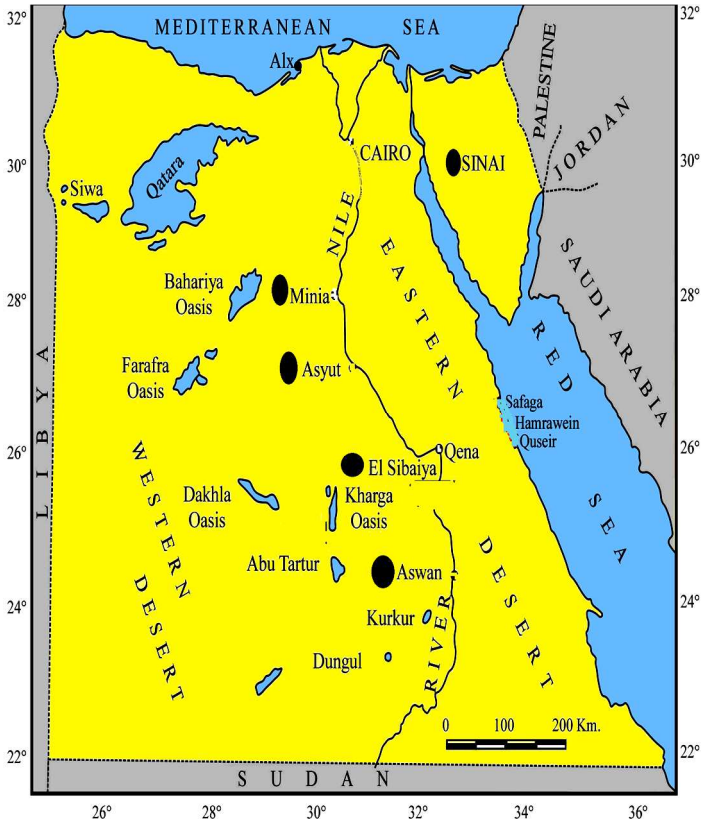


Figure 1. Locations map of the studied samples.

Instrumentations and calibration

The spectrometer system used in this study consists of sodium iodide detector 3×3 inch (NaI (Tl) model (802) (Canberra Co., USA), with a 2048 multichannel analyzer (MCA). Its hermetically sealed assembly which includes a high-resolution NaI (Tl) crystal, photomultiplier tube, an internal magnetic/light shield, aluminum housing and a 14-pin connector, Preamplifier, main amplifier, analogue to digital converter and Canberra Multichannel Analyzer (MCA) with Genie 2000 software. The calculations were done by the Excel program. The applied detector has the following specifications:

(1) Resolution 7.5% specified at the 662 keV peaks of 137Cs.

(2) Window aluminum 0.5 mm thick, density 147 mg cm^{-3}

(3) REFLECTOR- oxide; 1.6 mm thick; density 88 mg cm-3.

(4) Magnetic/light shield-conetic lined steel.

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The detection array was energy calibrated using ^{60}Co (1173.2 and 1332.5 keV), ^{133}Ba (356.1 keV), ^{137}Cs (661.9 keV) standard sources. The efficiency calibration curve was made using IAEA-314 reference material where the specific activity was known, which containing three radionuclides: ^{226}Ra, ^{232}Th and ^{238}U. To determine the background distribution in the environment around the detector, an empty sealed beaker was counted in the same manner and in the same geometry as the samples. The measurement time of activity or background was 43200 s. The background spectra were used to correct the net peak area of gamma-ray of measured isotopes. The offline analysis of each measured γ-ray spectrum has been carried out by a dedicated software program genie 2000 [22]. The ^{226}Ra radionuclide was estimated from the 351.9 keV γ-peak of ^{214}Pb, and 609.3 keV, 1120.3 keV, 1728.6 keV and 1764 keV γ-peak of ^{214}Bi. The 186 keV photon peak of ^{226}Ra was not used because of the interfering peak of ^{235}U, with the energy of 185.7 keV. The ^{232}Th radionuclide was estimated from the 911.2 keV γ-peak of ^{228}Ac and the 238.6 keV γ-peak of ^{212}Pb. The ^{40}K radionuclide was estimated using the 1461 keV γ-peak from ^{40}K itself [7].

Derivation of Radiation Hazard Indices

Radium equivalent (Ra_{eq})

The radium equivalent activities (Ra_{eq}) have been calculated on the estimation that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th and 4810 Bq kg^{-1} of ^{40}K produce the same gamma-ray dose rate [23]. Therefore, the Ra_{eq} was estimated by equation (1) [24]:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_k\backslash \quad (1)$$

Where, A_{Ra}, A_{Th} and A_K are the activities of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1}), respectively.

Absorbed gamma dose rate (D)

The absorbed dose rates (D) due to gamma radiations in the air at 1 m above the ground surface for the uniform distribution of the naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were calculated based on guidelines provided by UNSCEAR [25]. The absorbed dose rates can be calculated using equation (2) [26]:

$$D = 0.462A_{Ra}+0.604A_{Th}+0.0417A_K \quad (2)$$

Annual effective dose

Annual estimated average effective dose equivalent (AED) received by an individual was calculated using a conversion factor of 0.7 Sv Gy^{-1}, which was used to convert the absorbed rate to the human effective dose equivalent [27]. The annual effective dose was evaluated using equation [3]:

$$AED = D*T*F \quad (3)$$

T is the occupancy time and F is the conversion factor (0.7 Sv Gy-1).

Gamma radiation hazard index

The gamma index (Iγ) is defined by equation (4) [28]:

$$I\gamma = 0.0067\ A_{Ra} +0.01\ A_{Th}+0.00067\ A_K \quad (4)$$

External hazard index

The external hazard index (H_{ex}) was determined from criterion formula (5) [29]:

$$Hex = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4180} \leq 1 \quad (5)$$

Where, C_{Ra}, C_{Th} and C_K are activities of ^{226}Ra, ^{232}Th and ^{40}K respectively in Bq Kg^{-1}

Internal hazard index

On the other hand, the internal hazard index (H_{in}) gives the internal exposure to carcinogenic radon and its short-lived progeny [30], and it is given by equation (6) [31, 32]:

$$H_{in} = (A_{Ra}/185 + A_{Th}/259 + A_K/4810) \leq 1 \quad (6)$$

RESULTS AND CONCLUSION

The results of the average activity, as well as the uncertainty of ^{226}Ra, ^{232}Th and ^{40}K of different samples, were presented in figure 2. It could be seen that, the values of activity concentrations in the studied samples varied from 28±2 to 163±12, 2.8±0.7 to 40±3 and from 49±4 to 1337±74 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The variation of radionuclides concentration in studied samples may be due to

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the different origins of the raw material. The highest values of ^{226}Ra and ^{232}Th activity concentrations were found in Phosphate rock sample, while the highest value of ^{40}K activity concentration was found in CKD sample as shown in (figure 2). The mean concentrations of ^{226}Ra and ^{232}Th for cement dust, limestone, sand and borax samples were lower than the world averages for building materials 50 and 50 (Bq kg^{-1}), respectively [28]. On the other hand, the mean concentrations of ^{40}K were lower than the world averages for building materials (370 Bq kg^{-1}) except for cement dust samples. For phosphate rock samples the mean concentrations of ^{232}Th and ^{40}K were lower than the permissible activity levels [33]. While, the mean concentrations of ^{226}Ra were higher than the permissible activity levels only for phosphate rock, this is may be due to the reported elsewhere [34] That the phosphate rock used as raw material in the Brazilian phosphate industry presents concentration of radionuclides of the U and Th series ranging from 10 to 1200 Bq kg^{-1}.

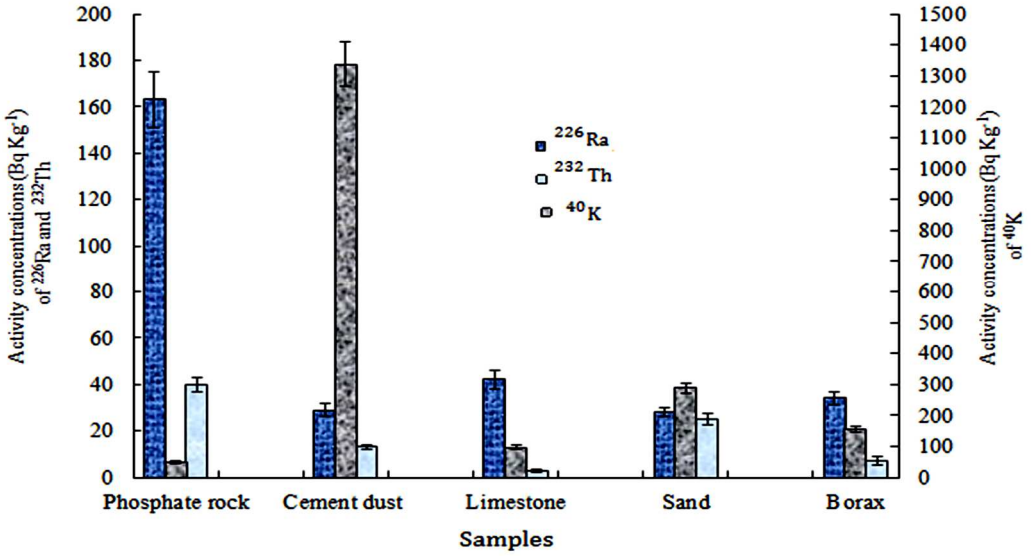


Figure 2. Activity concentration average values of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1} for the studied samples.

Ra_{eq}, dose rate, AED and other radiation hazard indices (H_{ex}, H_{in} and I_γ) were estimated using equations (1) to (6) and their results are presented in tables 2 and 3, respectively. From table 2 the values of radium equivalent in samples are lower than the allowed maximum value [31] of 370 Bq kg^{-1} and fluctuate from 45.90 Bq kg^{-1} in borax to 224.22 Bq kg^{-1} in phosphate rock. The absorbed dose rates due to ^{226}Ra, ^{232}Th and ^{40}K are ranged from 22.05 to 101.59 nGy h^{-1}. As shown in table 2 the values of absorbed dose rates in samples are lower than the allowed maximum value [27] of 59 nGy h^{-1}, except for phosphate rock and CKD. The annual effective dose rate varies from 27.04 to 124.59 μSv y^{-1}, these values are lower than the world average values [32] of 70 mSv y^{-1} as shown in table 2 except for phosphate rock and CKD.

The calculated I_γ and H_{ex} values for the samples under investigation are given in table 3. It is clear that I_γ and H_{ex} values are lower than unity [25, 27], which does not cause any harm to the populations. Also, table 3 shows that the calculated values of H_{in} for all samples which are less than unity except for phosphate rock samples.

The average activity concentrations (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K, in Limestone, Phosphate rock, Cement dust, Sand, and Borax samples of the present work compared with other studies are presented in table 4, where we can see, the mean values of ^{226}Ra, ^{232}Th and ^{40}K in samples in the range or less than the corresponding values in the listed countries.

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Table 2. The equivalent radium (Raeq), dose rate and annual effective dose (AED) for samples.

Locations	Samples	Ra_{eq} (Bq Kg^{-1})	Dose rate (nGy h^{-1})	(AED) (μSv y^{-1})
El-Sibaiya mine (Aswan)	Phosphate rock	*224.22	101.59	124.59
Cemex Assiut (Assiut)	Cement dust	132.15	68.31	83.78
Quarry (Minia)	Limestone	49.82	23.54	28.88
South Sinai	Sand	86.72	40.19	49.29
Market (Assiut)	Borax	45.90	22.05	27.04

*Mean value (for three reparations)

Table 3. External hazard (H_{ex}) Internal hazard (H_{in}) and γ radiation hazard index ($I_{γr}$) for the studied samples.

Locations	Samples	Hazards		
		H_{ex}	H_{in}	$I_γ$
El-Sibaiya mine (Aswan)	Phosphate rock	*0.61	1.05	0.76
Cemex Assiut (Assiut)	Cement dust	0.36	0.16	0.43
Quarry (Minia)	Limestone	0.13	0.23	0.17
South Sinai	Sand	0.23	0.25	0.29
Market (Assiut)	Borax	0.12	0.18	0.15

Table 4. Comparison of the average activity concentrations in the present study with other different studies

Country	Samples	Activity(BqKg^{-1})			Ref.
		^{226}Ra	^{232}Th	^{40}K	
Egypt (Minia)	Limestone	42	2.8	97	Present work
Egypt		24.50	43.01	63.2	(35)
Egypt		121	55	194	(36)
India		73.9	-----	64.6	(37)
Brazil		24.3	7.0	205	(38)
Egypt (South Sinai)	Sand	28	25	289	Present work
Egypt		23	21	618	(39)
Saudi Arabia (Ad-Dahna Desert)		23.4	29.7	380	(40)
China		22	39	859	(41)
Egypt (Aswan)	Phosphate rock	163	40	49	Present work
Brazil		257	284	45>	(34)
Sudan (Kurun)		554	0.83	23	(42)
Egypt (El-Mahamid)		567	217.3	217.3	(21)
Egypt (W. El-Mashash)		666	329.4	329.4	
Egypt (Assiut)	Cement dust	29	13	1337	Present work
Egypt	Cement waste	57.1	12.2	1606.2	(43)
Slovakia	Cement	11.8	18.4	156.5	(44)
Egypt	Cement	25.0	12.0	498.0	(36)

DISCUSSION

The activity levels natural radionuclides, ^{232} Th, and ^{40}K and related radiation hazards in the material under investigation were assessed by γ-ray spectrometry. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K of samples are lower than the world average (35, 35 and 400 Bq kg^{-1}) respectively except ^{226}Ra and ^{232}Th in phosphate rock are higher (163 and 40 Bq kg^{-1}) and ^{40}K (1337 Bq kg^{-1}) in cement dust samples. The results indicate that the concentration of

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radioactive elements depends on local geological conditions. The concentration of ^{40}K is the most compared to ^{232}Th and ^{226}Ra. This study can be used as a baseline for future investigations and the data obtained in this study may be useful for natural radioactivity mapping. Finally, it could be concluded that all materials would not present a significant radiological hazard except phosphate.

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Conflicts of interest*: *Declared none.

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