Assessment of the radiological impacts of treated phosphogypsum used as the main constituent of building materials in Jordan

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Introduction

Phosphogypsum (PG) is the major by-product of the phosphate fertilizer industry, and is produced in large quantities by the wet phosphoric acid process.

PG is a major environmental concern in terms of disposal of the large volumes produced. About five tonnes of the phosphogypsum are produced for every tonne of phosphoric acid manufactured

A simplified chemical reaction for the acidulation process is:

$$\underbrace{Ca_{10}(PO_4)_6F_2}_{\{\text{Phosphate rock}\}} + 10H_2SO_4 + 20H_2O -$$
$$--> \underbrace{10CaSO_4 \cdot 2H_2O}_{\{\text{Phosphogypsum}\}} + 6H_3PO_4 + 2HF$$

Through the wet process, some impurities naturally present in the phosphate rock become concentrated in phosphogypsum, although not as high as the concentrations in the source rock.

During the production of phosphoric acid, ^{238}U, ^{230}Th and ^{210}Pb tend to be incorporated in the phosphoric acid and then in the phosphate fertilizer, while ^{226}Ra and ^{210}Po are deposited into the phosphogypsum

Objectives

- **The treatments are designed to remove and leach the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the samples of phosphogypsum, which can be utilized for different purposes (i.e. building materials and cement industry).**

- **The study design also evaluates the radiological impacts of utilizing treated phosphogypsum in the building materials and**
- **Other applications by assessing radium equivalent activity (Ra_{eq}), gamma index (I_γ) alpha index (I_α), absorbed gammadose rate (D_{in}), and corresponding annual effective dose (E_{in}) for public exposure due to the use of treated phosphogypsum in building materials and for other purposes**

Materials and methods

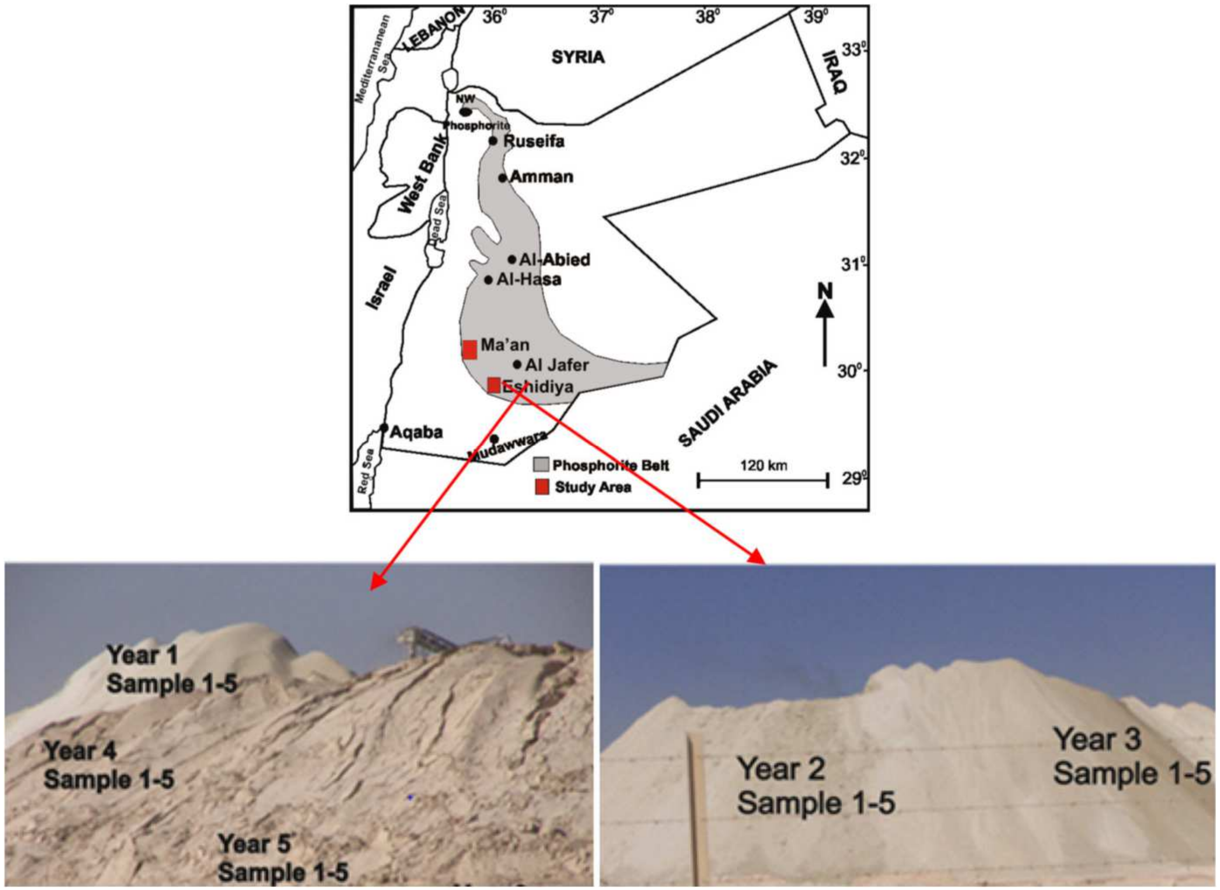

Fig. 1. Location map of the study area and phosphogypsum stacks showing sampling sites

Table 1. The mix proportions were composed of varying percentage of reagent and PG Code PG (g) Reagent Reaction

Code	PG (g)	Reagent	Reaction time (h)	T (°C)	Filtered	Dried (°C)
Phosphogypsum (N = 25)						
PG-1	200	–	–		–	85
PG-2	200	–	–		–	85
PG-3	200	–	–		–	85
PG-4	200	–	–		–	65
PG-5	200	–	–		–	65
Hybrid water treatment (N = 35)						
LW1	200	Lime water + sea water (v/v) (100 ml:700 ml)	24	25	A Büchner funnel	65
LW2	200	Lime water + sea water (v/v) (125 ml:675 ml)	24	25	A Büchner funnel	65
LW3	200	Lime water + sea water (v/v) (150 ml:650 ml)	24	25	A Büchner funnel	65
LW4	200	Lime water + tap water (v/v) (100 ml:700 ml)	24	25	A Büchner funnel	65
LW5	200	Lime water + tap water (v/v) (100 ml:700 ml)	24	25	A Büchner funnel	65
LW6	200	Lime water + distilled water (v/v) (100 ml:700 ml)	24	25	A Büchner funnel	65
LW7	200	Lime water + distilled water (v/v) (1250 ml:675 ml)	24	25	A Büchner funnel	65
Sulphuric acid treatment (N = 20)						
SA1	200	5 % Sulphuric acid + sea water (v/v) (12 ml:88 ml)	24	25	A Büchner funnel	65
SA2	200	5 % Sulphuric acid + tap water (v/v) (12 ml:88 ml)	24	25	A Büchner funnel	65
SA3	200	5 % Sulphuric acid + distilled water (v/v) (12 ml:88 ml)	24	25	A Büchner funnel	65
SA4	200	5 % Sulphuric acid + limewater (v/v) (12 ml:88 ml)	24	25	A Büchner funnel	65
Mixed acid treatment (N = 10)						
AM1	200	300 ml 5 % H_2SO_4 + 4 ml 2 % HNO_3 + distilled water (v/v) (250 ml:750 ml)	24	25	A Büchner funnel	65
AM2	200	300 ml 5 % H_2SO_4 + 4 ml 2 % HNO_3 + distilled water (v/v) (300 ml:700 ml)	24	25	A Büchner funnel	65
Household water treatment (N = 10)						
PW1	200	Tap water (100 ml)	24	25	A Büchner funnel	65
PW2	200	Distilled water (200 ml)	24	25	A Büchner funnel	65
Calcium carbonate powder treatment (N = 25)						
PL1	200	$CaCO_3$ powder (5 g)	–	–	–	–
PL2	200	$CaCO_3$ powder (10 g)	–	–	–	–
PL3	200	$CaCO_3$ powder (25 g)	–	–	–	–
PL4	200	$CaCO_3$ powder (50 g)	–	–	–	–
PL5	200	$CaCO_3$ powder (75 g)	–	–	–	–

N number of sample

Results and discussion

Table 2. Activity concentration values of ^{226}Ra, ^{232}Th and ^{40}K in the phosphogypsum samples from Eshidiya Mine (N = 25)

Location (age)	Sample	Activity concentration (in Bq kg^{-1}+1σ)		
		^{226}Ra	^{232}Th	^{40}K
Year-1	1	328.9 ± 24.1	3.6 ± 0.3	112.6 ± 8.4
	2	329.2 ± 23.9	3.5 ± 0.2	114.1 ± 8.2
	3	330.7 ± 22.6	3.3 ± 0.4	113.9 ± 7.7
	4	327.3 ± 20.2	3.7 ± 0.3	111.9 ± 8.1
	5	328.1 ± 24.0	3.6 ± 0.3	112.6 ± 9.1
	Mean + STD	328.8 ± 1.3	3.5 ± 0.2	113.0 ± 0.9
Year-2	1	335.7 ± 27.7	3.7 ± 0.2	115.0 ± 8.6
	2	336.2 ± 26.2	4.1 ± 0.1	116.0 ± 9.2
	3	337.5 ± 22.8	3.4 ± 0.5	114.2 ± 8.4
	4	335.1 ± 24.2	3.5 ± 0.3	115.2 ± 7.6
	5	335.7 ± 22.7	3.9 ± 0.2	114.3 ± 8.3
	Mean + STD	336.0 ± 0.9	3.7 ± 0.3	114.9 ± 0.5
Year-3	1	338.1 ± 23.9	3.7 ± 0.4	115.8 ± 8.7
	2	337.4 ± 27.2	3.4 ± 0.3	116.1 ± 7.7
	3	339.1 ± 26.9	4.1 ± 0.5	115.1 ± 8.2
	4	337.6 ± 25.7	2.9 ± 0.1	114.8 ± 9.2
	5	338.2 ± 23.3	4.2 ± 0.5	115.6 ± 6.9
	Mean + STD	338.1 ± 0.7	3.7 ± 0.5	115.5 ± 0.7
Year-4	1	356.1 ± 20.0	3.9 ± 0.2	122.0 ± 8.2
	2	357.1 ± 24.2	4.2 ± 0.4	122.2 ± 9.6
	3	354.1 ± 23.3	3.2 ± 0.2	121.6 ± 6.2
	4	354.5 ± 22.1	2.9 ± 0.2	123.2 ± 7.8
	5	356.1 ± 25.0	4.2 ± 0.4	124.1 ± 7.6
	Mean + STD	355.6 ± 1.2	3.7 ± 0.6	122.6 ± 1.0
Year-5	1	342.8 ± 28.5	3.8 ± 0.3	117.4 ± 8.8
	2	340.2 ± 21.4	3.3 ± 0.2	115.1 ± 7.8
	3	342.3 ± 22.5	3.6 ± 0.3	114.6 ± 8.6
	4	341.7 ± 26.1	4.1 ± 0.5	117.2 ± 7.6
	5	342.2 ± 23.3	4.2 ± 0.5	118.1 ± 9.8
	Mean + STD	341.8 ± 1.0	3.8 ± 0.4	116.5 ± 1.5

STD standard deviation

Table 3. Descriptive statistical analysis of activity concentration and radiological hazard indices in untreated phosphgypsum compared with treated phosphogypsum materials

Sample code	Age	Activity concentration (mean + STD)			Radiological hazard indices				
		^{226}Ra (Bq kg^{-1})	^{232}Th	^{40}K	Ra_{eq}	I_γ	I_α	D_{in} (nGy h^{-1})	E_{in} (mSv y^{-1})
PG ($N = 25$)									
PG-1	Year-1	328.8 ± 1.3	3.5 ± 0.2	113.0 ± 0.9	342.60	1.10	1.60	315.50	1.50
PG-2	Year-2	336.0 ± 0.9	3.7 ± 0.3	114.9 ± 0.5	350.20	1.20	1.70	322.40	1.60
PG-3	Year-3	338.1 ± 0.7	3.7 ± 0.5	115.5 ± 0.7	352.20	1.20	1.70	324.30	1.60
PG-4	Year-4	355.6 ± 1.2	3.7 ± 0.6	122.6 ± 1.0	370.30	1.20	1.80	341.00	1.70
PG-5	Year-5	341.8 ± 1.0	3.8 ± 0.4	116.5 ± 1.5	356.20	1.20	1.70	328.00	1.60
Hybrid water ($N = 35$)									
LW1	Year (1–5)	48.4 ± 11.3	12.5 ± 0.6	127.1 ± 10.6	76.06	0.27	0.24	68.45	0.34
LW2	Year (1–5)	47.8 ± 6.5	12.3 ± 0.6	121.1 ± 10.1	74.71	0.26	0.24	67.19	0.33
LW3	Year (1–5)	44.5 ± 5.8	10.9 ± 0.6	105.5 ± 8.8	68.21	0.24	0.22	61.37	0.30
LW4	Year (1–5)	46.3 ± 5.9	12.3 ± 0.6	115.4 ± 9.7	72.77	0.25	0.23	65.36	0.32
LW5	Year (1–5)	43.9 ± 6.5	12.1 ± 0.6	125.4 ± 10.5	70.86	0.25	0.22	63.73	0.31
LW6	Year (1–5)	49.1 ± 8.9	13.1 ± 0.7	134.5 ± 11.3	78.19	0.27	0.25	70.34	0.35
LW7	Year (1–5)	37.7 ± 9.6	11.3 ± 0.6	110.1 ± 9.9	62.34	0.22	0.19	55.92	0.27
Sulphuric acid ($N = 20$)									
SA1	Year (1–5)	39.5 ± 5.0	12.6 ± 0.6	146.5 ± 10.6	68.80	0.24	0.20	65.90	0.30
SA2	Year (1–5)	45.4 ± 7.7	12.2 ± 0.6	118.6 ± 9.3	71.98	0.25	0.23	61.92	0.32
SA3	Year (1–5)	42.8 ± 6.2	12.1 ± 0.6	122.5 ± 10.2	69.54	0.24	0.21	64.68	0.31
SA4	Year (1–5)	40.9 ± 5.9	12.8 ± 0.7	120.1 ± 10.4	68.45	0.24	0.20	62.49	0.30
Mixed acid ($N = 10$)									
AM1	Year (1–5)	46.7 ± 5.5	13.2 ± 0.7	129.7 ± 10.8	75.56	0.25	0.23	61.32	0.33
AM2	Year (1–5)	45.2 ± 5.5	12.8 ± 0.7	127.9 ± 10.8	73.35	0.26	0.23	65.90	0.32
Household water ($N = 10$)									
PW1	Year (1–5)	51.7 ± 6.7	12.4 ± 0.7	118.2 ± 9.9	78.53	0.27	0.26	70.66	0.35
PW2	Year (1–5)	51.3 ± 6.7	13.1 ± 0.7	126.7 ± 10.6	79.79	0.28	0.26	71.74	0.35
Calcium carbonate powder ($N = 25$)									
PL1	Year (1–5)	48.5 ± 6.3	11.9 ± 0.6	119.2 ± 10.0	74.70	0.26	0.24	67.25	0.33
PL2	Year (1–5)	47.2 ± 5.7	12.3 ± 0.6	121.9 ± 10.2	74.18	0.26	0.24	66.71	0.33
PL3	Year (1–5)	53.8 ± 8.8	11.7 ± 0.6	115.41 ± 9.7	79.42	0.28	0.27	71.60	0.35
PL4	Year (1–5)	43.6 ± 4.9	11.6 ± 0.6	112.6 ± 9.4	68.86	0.24	0.22	61.88	0.30
PL5	Year (1–5)	45.5 ± 6.6	11.9 ± 0.7	113.7 ± 9.9	71.27	0.25	0.23	64.05	0.31

STD standard deviation

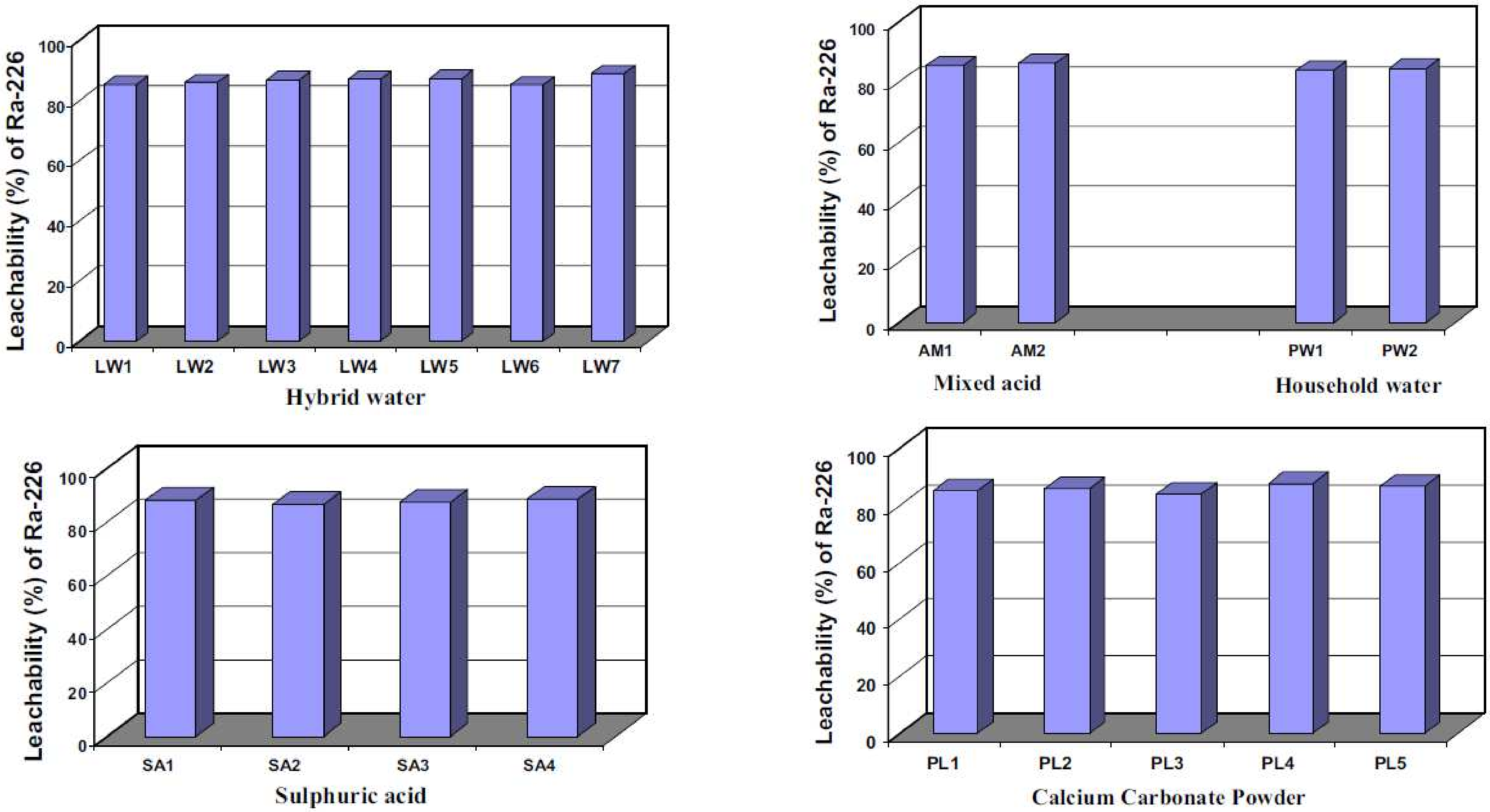

Fig. 2. Effect of hybrid water, sulphuric acid, mixed acid, and calcium carbonate powder on the leachability of Radium-226 content in PG waste

Conclusions

The calculated values of the radium equivalent ctivity (Ra_{eq}), gamma index, and the alpha index for all the treated phosphogypsum samples are significantly below the recommended upper level of unity.

The measured mean value of the indoor absorbed gamma dose rate is about 20 % lower than the population-weighted average value of 84 nGy h^{-1} for the indoor absorbed dose rate

The evaluated values of the indoor annual effective dose resulting from use of all the treated phosphogypsum samples are significantly below the recommended upper level of 1 mSv. The mean value of the indoor annual effective dose is about 70 % lower than upper level of 1 mSv.

In the overall assessment, concluded that the possibility of using Eshidiya-treated PG in building materials and other applications in proportions up to 100 % will be safe from the radiation protection point of view.

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