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Evaluation of Excess Lifetime Cancer Risk Due to Gamma Rays Exposure from Phosphate Fertilisers Used in Saudi Arabia

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ABSTRACT: *Activity concentrations were measured in phosphate fertilisers widely used in Saudi Arabia employing gamma ray spectrometry. The activity concentrations of ^{238}U, ^{232}Th and ^{40}K were used to estimate the excess lifetime cancer risk (ELCR). The obtained data show that ^{238}U and ^{40}K concentrations are higher than the recommended values (35 Bq kg^{-1} and 400 Bq kg^{-1} for ^{238}U and ^{40}K, respectively) in most of phosphate fertilisers. The highest value of ^{238}U was 4122 ± 82 Bq kg^{-1} for diammonium phosphate (DAP) fertiliser. Absorbed dose rate, annual effective dose rate and the ELCR were calculated. The results revealed that the values of outdoor, indoor and total ELCR values were higher than the world averages in all samples except one sample, sample of MAP fertiliser (monoammonium phosphate). The highest value of $ELCR_{tot}$ was for DAP fertiliser which is 50 times higher than the world average value (1.45×10^{-3}). Moreover, the results indicated a strong correlation between uranium in phosphate fertilisers and $ELCR_{tot}$. Based on the obtained results, the direct gamma radiation exposure from phosphate fertilisers is a serious radiological threat to the farmers.*

Keywords: Phosphate fertilisers, activity concentrations, gamma rays, ELCR, cancer risk

1. INTRODUCTION

Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions, and appear at different levels in the soils of each region in the world.[1] One of the sources of radioactivity in soils other than those of natural origin is the extensive use of fertilisers rich in phosphates for agricultural purposes.[2,3] Phosphate fertilisers

are used widely in the reclamation of farmland and increasing agricultural crops. Moreover, phosphate fertilisers are by far the most widely used for agriculture soil due to its cost-effectiveness and its availability in markets.

Sedimentary phosphate ores can be significantly enriched with naturally occurring radionuclides, uranium (^{238}U) and the daughter radionuclides that come from the radioactive decay of ^{238}U.[4] Phosphoric anhydride (P_2O_5) and potassium oxide (K_2O) are important materials used for phosphate fertilisers production. Previous studies have shown that the concentration of uranium and potassium is related to the concentration of P_2O_5 and K_2O in various fertilisers.[3,5] Phosphate ore is typically extracted from mineral deposits in phosphate rock. The phosphate rocks are used as raw materials in phosphate fertiliser industries, which contain radionuclides of the uranium and thorium natural series. Also, one of the important sources of radioactivity is potassium ^{40}K which exists in phosphate rocks.[6]

Kingdom of Saudi Arabia is one of the largest producers and exporter of diammonium phosphate (DAP) and compound phosphate fertilisers worldwide. The phosphate fertiliser factories have been provided with phosphate ore from phosphate mine in northern Saudi Arabia.[3] Before being transported to the factories, the phosphate undergoes the beneficiation process in the plant in order to produce concentrated phosphate ore. Furthermore, several types of imported phosphate fertilisers have been used in agricultural activities. Therefore, concentrations of naturally occurring radionuclides (^{238}U, ^{232}Th and ^{40}K) in phosphate fertilisers in Saudi Arabia were measured, to evaluate the excess lifetime cancer risk (ELCR) for the population due to gamma rays exposure from phosphate fertilisers.

2. EXPERIMENTAL

2.1 Samples Collection and Preparation

Seventeen samples of widely used phosphate fertilisers were collected from local markets, Saudi Arabia (Table 1). Four types of phosphate fertilisers were studied: DAP, monoammonium phosphate (MAP), single super phosphate (SSP) and NPK (nitrogen, phosphor and potassium). Concentrations of P_2O_5 in fertilisers ranging at 5%–61% is shown in Table 1. Samples were grinded, dried and packed in the standard size can and tightly sealed and stored for 28 days to acquire secular equilibrium. Reference material was also packed in the same standard size can for efficiency calibration.[3]

2.2 Experimental Setup

A hyper-pure Germanium detector (HPGe), coaxial type, p-type with relative efficiency of 20% was used. The resolution (full width at half maximum, FWHM) at the peak 122 keV, ^{57}Co, is 0.934 keV and at the peak 1332 keV, ^{60}Co, is 1.79 keV. The detector was shielded with lead shield to reduce the background radioactivity. The software Quantum Gold, Version 4.04.00, was used for analysis and evaluation of the gamma-ray spectra. The HPGe was calibrated for efficiency using the reference material RGU-1 from IAEA. The certified activity of uranium is 400 ppm which is equivalent to 4960 Bq kg^{-1}. The energy transitions of the ^{226}Ra daughters (^{214}Pb and ^{214}Bi) were used to develop the efficiency calibration curve. A fourth-degree polynomial fitting was performed to reach the best R^2 value (≈ 0.983).[3]

Table 1: Description of phosphate fertiliser samples in the present study and their specific activities of ^{238}U, ^{232}Th and ^{40}K (Bq kg^{-1}).

Sample no.	Type of fertiliser	Description	Origin	P_2O_5 %	^{238}U	^{232}Th	^{40}K
1	DAP	Granule, light brown	KSA	46	4122 ± 82	ND*	131 ± 11
2	DAP	Granule, dark brown	KSA	46	3036 ± 61	ND	74.9 ± 3.7
3	SSP	Granule, dark brown	Egypt	20	379 ± 11	41.2 ± 3.3	1213 ± 24
4	MAP	Cristal, white	Emirate	61	41.4 ± 1.6	0.711 ± 0.2	60.0 ± 3.0
5	NPK	Powder, light green	KSA	52	57.4 ± 9.1	ND	1277 ±13
6	NPK	Granule, pink	KSA	40	30.4 ± 3.6	ND	3519 ± 21
7	NPK	Granule, red	Germany	18	827 ± 25	29.1 ± 5.2	5908 ± 59
8	NPK	Granule, red	KSA	5	36.9 ± 7.2	ND	3869 ± 39
9	NPK	Powder, blue	KSA	12	436 ± 8.7	ND	3112 ± 19
10	NPK	Powder, red	KSA	18	904 ± 9.0	ND	492 ± 84
11	NPK	Powder, dark pink	KSA	45	11.4 ± 3.3	ND	2137 ± 17
12	NPK	Granule, black	China	10	67 ± 8	ND	2944 ± 20

(*continued next page*)

Table 1: (*continued*)

Sample no.	Type of fertiliser	Description	Origin	P_2O_5 %	^{238}U	^{232}Th	^{40}K
13	NPK	Granule, light brown	KSA	14	585 ± 41	ND	5029 ± 43
14	NPK	Granule, light blue	KSA	12	507 ± 34	ND	6430 ± 62
15	NPK	Granule, blue	Germany	12	498 ± 45	ND	3801 ± 20
16	NPK	Granule, red	KSA	20	1169 ± 61	ND	1276 ± 51
17	NPK	Granule, blue	KSA	14	794 ± 46	ND	5709 ± 57
Min					11.4 ± 3.3	0.711 ± 0.2	60.0 ± 3.0
Max					4122 ± 82	41.2 ± 3.3	6430 ± 62

Notes: KSA denotes Kingdom of Saudi Arabia; ND denotes not detectable

2.3 Calculations of Activity Concentrations

After achieving secular equilibrium and subtraction of the background, the activity concentrations of ^{238}U, ^{232}Th and ^{40}K were calculated. ^{238}U was measured at the gamma lines 63.29 keV and 92.38 keV for ^{234}Th. For ^{232}Th, the activity concentration was assessed by using energy transition of ^{228}Ac at 93.4 keV and the energy transitions of ^{208}Tl at 583.19 keV and ^{228}Ac at 911.2. The average values were calculated. In the case of ^{40}K, the specific activity was estimated directly by its gamma line of 1460.75 keV.

3. RESULTS AND DISCUSSION

3.1 Activity Concentrations in Phosphate Fertilisers

The activity concentrations of ^{238}U, ^{232}Th and ^{40}K in phosphate fertilisers were measured as shown in Table 1. The activity concentration of ^{238}U ranges from 11.4 ± 3.3 Bq kg^{-1} to 4122 ± 82 Bq kg^{-1}. The activity concentration of ^{232}Th varies from 0.711 ± 0.2 Bq kg^{-1} to 41.2 ± 3.3 Bq kg^{-1}. However, 14 samples do not contain ^{232}Th as shown in Table 1. The value of ^{40}K ranges from 60.0 ± 3.0 Bq kg^{-1} to 6430 ± 62 Bq kg^{-1}. From these results, it can be observed that the activity concentrations of uranium in 15 phosphate fertiliser samples are higher than permissible limit in soil (35 Bq kg^{-1}).[7] The highest value was 4122 ± 82 Bq kg^{-1} for DAP with 46% of P_2O_5 which is 114 times higher than the permissible limit. Uranium content in DAP

sample was higher than the values in the other phosphate fertiliser samples. High concentration of uranium in DAP fertiliser (KSA) may be due to the use of phosphate rocks as raw materials for manufacturing phosphate fertilisers.[8,9] Similarly, the activity concentrations of ^{40}K in all fertiliser samples were higher than the recommended value (400 Bq kg^{-1}), except of DAP and MAP phosphate fertilisers.[7] The high concentration of potassium may be related to the concentration of K_2O.[3] The result indicates that the large use of phosphate fertilisers leads to increase the radioactivity of soil, especially for DAP fertiliser. The elevated levels of ^{238}U may due to wet phosphate processing where phosphate ore is firstly attacked by sulfuric acid to produce the phosphoric acid where uranium will be mainly concentrated in the phosphoric acid, while radium, thorium and other insoluble radionuclides will be precipitated as sulphate salts and concentrated in the phosphogypsum by product.[10] In 2000, Mazzilli et al. found that 90% of ^{226}Ra and 80% of ^{232}Th fractionate into phosphogypsum, while ^{238}U was being predominantly incorporated in phosphoric acid.[11] Uranium and its progenies are usually in secular equilibrium in the different geological formation, while secular equilibrium could be disturbed during chemical treatment of phosphate ore to produce phosphoric acid.[10]

3.2 ELCR

There is always a chance of 33% (or 0.33% per person) that an individual will be diagnosed with cancer at some stage of life due to normal exposure to natural radioactivity and or a toxic substance.[12] The Department of Environmental Quality (DEQ) considers a chance of 1 in 100,000 persons. That means 1×10^{-5} is allowable limit. ELCR is an additional possibility that a person may develop cancer if that person is exposed to additional cancer-causing materials for a longer time. In order to evaluate the ELCR of phosphate fertilisers, some radiological parameters were calculated (Table 2). The outdoor absorbed dose rate (D_{out}) in air 1 m above the ground surface from the gamma radiation from ^{232}Th, ^{238}U and ^{40}K was calculated.[7] The conversion factors used to compute the outdoor external dose rate (D_{out}) in air per unit activity concentration in 1 Bq kg^{-1} correspond to 0.604 nGy h^{-1} for ^{232}Th, 0.462 nGy h^{-1} for ^{238}U and 0.0417 nGy h^{-1} for ^{40}K. Therefore, D_{out} could be calculated from the following equation:[6,13]

$$D_{out}\ (nGy\ h^{-1}) = 0.462\ A_U + 0.604A_{Th} + 0.0417\ A_K \quad (1)$$

The indoor gamma ray dose (D_{in}) from the above radionuclides was calculated using Equation 2:[13]

$$D_{in}\ (nGy\ h^{-1}) = 0.92\ A_U + 1.1\ A_{Th} + 0.081\ A_K \quad (2)$$

where A_U, A_{Th} and A_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K (Bq kg^{-1}), respectively.

As shown in Table 2, The outdoor and indoor absorbed dose rates (D_{out} and D_{in}) in air 1 m above the ground surface in phosphate fertilisers ranged from 22.1–1910 nGy h^{-1} and 43.7–3803 nGy h^{-1}, respectively. The outdoor and indoor absorbed dose rates in 16 phosphate samples, except MAP, are higher than the world averages for outdoor and indoor exposure due to terrestrial gamma radiation (51 nGy h^{-1} and 84 nGy h^{-1}, respectively).[7] The results indicate the direct connection between the outdoor and indoor absorbed dose rates in air and the specific activity of uranium and potassium, where the greatest part of gamma radiation comes from them.

Outdoor annual effective dose rate, E_{out} (mSv y^{-1}), received by the population is calculated using the following equation:[3,7]

$$E_{out}\ (mSv\ y^{-1}) = D_{out}\ (nGy\ h^{-1}) \times 8760\ (h\ y^{-1}) \times 0.2 \times 0.7\ (Sv\ Gy^{-1}) \times 10^{-6} \quad (3)$$

where,

D_{out} = the outdoor absorbed dose rate in air (nGy h^{-1});

8760 = the time for one year (h);

0.7 = the conversion factor (Sv Gy^{-1}), which converts the absorbed dose rate in air to human effective dose; and

0.2 = the outdoor occupancy factor.[7]

Outdoor annual effective dose rate, E_{out}, ranged at 0.03–2.34 mSv y^{-1}. It can be observed that the outdoor annual effective dose rate for all samples is higher than the world average value from the outdoor exposure (0.07 mSv y^{-1}).[3,7]

Similarly, the indoor annual effective dose rate in air was calculated using the formula as follows: [3,7]

$$E_{in}\ (mSv\ y^{-1}) = D_{in}\ (nGy\ h^{-1}) \times 8760\ (\ h\ y^{-1}) \times 0.7\ (Sv\ Gy^{-1}) \times 10^{-6} \times 0.8 \quad (4)$$

where,

D_{in} = the indoor gamma ray dose;

0.8 = the indoor occupancy factor;

0.7×10^{-6} = the conversion factor from absorbed dose rate in air to effective dose received by the person; and

8760 = the time per year (h).

The annual effective dose rate for indoor ranged from 0.21 mSv y^{-1} to 18.7 mSv y^{-1}. The result indicates that the values of indoor annual effective dose rate for all samples, except MAP fertiliser, were higher than the world average of 0.41 mSv y^{-1} from indoor exposure.[7]

The highest values of E_{out} and E_{in} are 1910 mSv y^{-1} and 18.7 mSv y^{-1}, respectively, in Sample 1 for DAP phosphate fertiliser. Therefore, the agricultural uses of DAP fertilisers could lead to significant radiation exposure. The annual effective dose rate for outdoor and indoor exposure must be less than 0,07 mSv y^{-1} and 0.41 mSv y^{-1}, respectively, to keep the radiation hazard insignificant.

The ELCR was calculated to estimate the probability of cancer incidence in a population of individuals. The indoor and outdoor ELCR were calculated using the following equations:[12,14]

$$ELCR_{out} = E_{out}\ (mSv\ y^{-1}) \times DL\ (y) \times RF\ (Sv^{-1}) \quad (5)$$

$$ELCR_{in} = E_{in}\ (mSv\ y^{-1}) \times DL\ (y) \times RF\ (Sv^{-1}) \quad (6)$$

where E_{in}, E_{out}, DL and RF are indoor and outdoor annual effective dose, duration of life (70 y) and risk factor, 0.05, for the public, respectively.[15]

From Table 2, the calculated $ELCR_{out}$ ranged from 0.09×10^{-3} to 8.20×10^{-3} for outdoor exposure. These values, except the value for MAP, were higher than the worldwide recommended value of 0.29×10^{-3}.[7] The highest value is 8.20×10^{-3} for DAP fertiliser which is greater than the world average value. The indoor $ELCR_{in}$ was calculated to estimate the total ELCR. The indoor $ELCR_{in}$ varied from 0.75×10^{-3} to 65.3×10^{-3}. The $ELCR_{tot}$ total for all samples was computed as shown in Table 2. The values of $ELCR_{tot}$ ranged at 0.84×10^{-3}–73.5×10^{-3}. The $ELCR_{tot}$ total for all samples, expect MAP, are greater than the word average value 1.45×10^{-3}.[16] The highest value was in DAP sample which is 50 times higher than the world average value, 1.45×10^{-3}. The variations of ELCR can be seen clearly as shown in Figure 1, where the highest value of ELCR was in DAP fertiliser and the lowest value was in MAP fertiliser.

Table 2: ELCR, absorbed dose rate (D) and annual effective dose (E) for phosphate fertiliser samples.

Sample no.	Type	D_{out} (nGy h^{-1})	D_{in} (nGy h^{-1})	E_{out} (mSv y^{-1})	E_{in} (mSv y^{-1})	$ELCR_{out}$ $\times 10^{-3}$	$ELCR_{in}$ $\times 10^{-3}$	$ELCR_{tot}$ $\times 10^{-3}$
1	DAP	1910	3803	2.34	18.7	8.20	65.3	73.5
2	DAP	1406	2799	1.72	13.7	6.03	48.1	54.1
3	SSP	251	492	0.31	2.41	1.08	8.45	9.53
4	MAP	22.1	43.7	0.03	0.21	0.09	0.75	0.84
5	NPK	79.8	156	0.10	0.77	0.34	2.68	3.02
6	NPK	161	313	0.20	1.54	0.69	5.37	6.06
7	NPK	628	1239	0.77	6.08	2.70	21.3	24.0
8	NPK	178	347	0.22	1.70	0.77	5.96	6.73
9	NPK	331	653	0.41	3.20	1.42	11.2	12.6
10	NPK	438	872	0.54	4.28	1.88	15.0	16.8
11	NPK	94.4	184	0.12	0.90	0.41	3.15	3.56
12	NPK	154	300	0.19	1.47	0.66	5.15	5.81
13	NPK	480	946	0.59	4.64	2.06	16.2	18.3
14	NPK	502	987	0.62	4.84	2.16	17.0	19.1
15	NPK	389	766	0.48	3.76	1.67	13.1	14.8
16	NPK	593	1179	0.73	5.78	2.55	20.2	22.8
17	NPK	605	1193	0.74	5.85	2.60	20.5	23.1
Min		22.1	43.7	0.03	0.21	0.09	0.75	0.84
Max		1910	3803	2.34	18.7	8.20	65.3	73.5
World average values.[7,16]		59	84	0.07	0.41	0.29	1.16	1.45

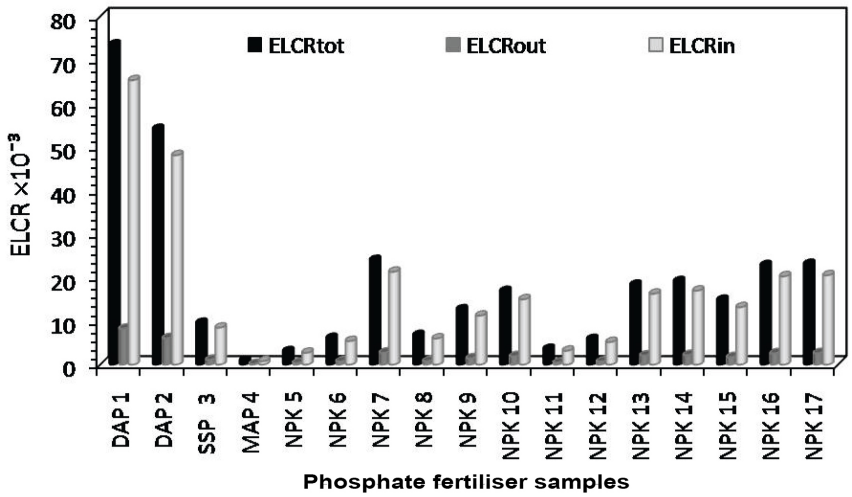

Figure 1: Variations of ELCR in phosphate fertiliser samples.

Figures 2 and 3 show the correlation of $ELCR_{tot}$ versus uranium and potassium. A strong correlation has been observed between $ELCR_{tot}$ and uranium with a correlation coefficient of R = 0.973 (Figure 2). The correlation of $ELCR_{tot}$ versus potassium shows a poor positive correlation with a correlation coefficient of R = 0.051. This result revealed that the high values of the excess lifetime cancer risk are attributed to elevated levels of uranium in the analysed phosphate fertiliser samples.

The results indicate that the excessive use of phosphate fertilisers in agriculture soil can lead to increase the radiation risk for public, especially for DAP. Further, the obtained values of absorbed dose rate, annual effective dose rate and the excess lifetime cancer risk in phosphate fertilisers show that the agricultural uses of phosphate fertilisers could lead to increase the probability of cancer incidence in a population of individuals, with the exception of MAP fertiliser.

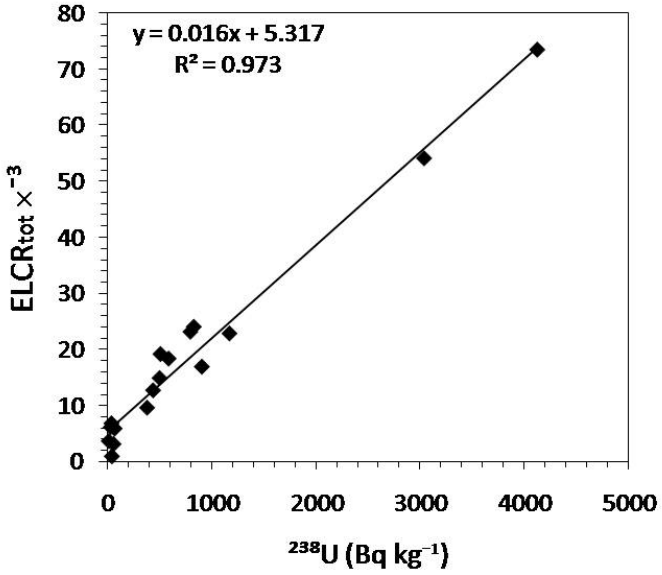

Figure 2: Correlation between uranium concentration and $ELCR_{tot}$.

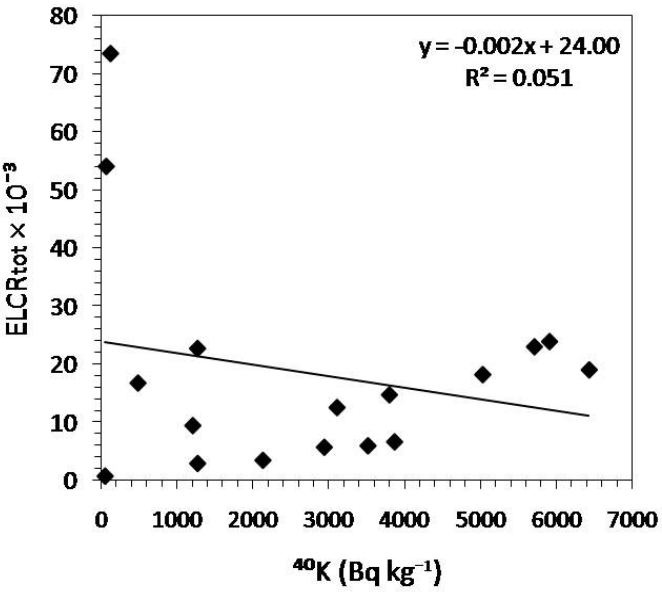

Figure 3: Correlation between potassium concentration and $ELCR_{tot}$ in phosphate fertilisers.

4. CONCLUSION

The activity concentrations of ^{238}U, ^{232}Th and ^{40}K in local and imported phosphate fertiliser samples were measured using gamma ray spectrometry technique. The data show that the activity concentrations of ^{238}U in 15 phosphate samples were higher than the world average value (35 Bq kg^{-1}), while ^{40}K activity was higher than the world average value (400 Bq kg^{-1}) in all samples except two samples (DAP and MAP fertilisers). However, the activity concentrations of ^{232}Th were found to be lower than the recommended value (30 Bq kg^{-1}) in all phosphate samples. The absorbed dose rate and annual effective dose rate were calculated to evaluate the ELCR for the analysed phosphate fertiliser samples. The outdoor, indoor and ELCR values were higher than the world average values. This result reveals that the direct exposure to the phosphate fertilisers may increase the probability of developing cancer for farmers, especially for DAP fertilisers due to the elevated levels of uranium. Moreover, a strong correlation has been observed between ELCR and uranium. Based on this study, the current data indicate to the need of new rules to improve and develop the regulations that can control the import and production of phosphate fertilisers in Saudi Arabia.

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