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Assessment of Natural Radioactivity in Phosphate Ore, Phosphogypsum and Soil Samples Around a Phosphate Fertilizer Plant in Nigeria

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Abstract The radionuclides present in phosphate ore, phosphogypsum and soil samples in the vicinity of a phosphate fertilizer plants in Nigeria were identified and their activity concentration determined to assess the potential radiation impact on the environment due to fertilizer production. The mean activity concentration of ^{238}U, ^{232}Th, and ^{40}K radionuclides in phosphate ore samples were 616 ± 38.6, BDL (Below Detection Level) and 323.7 ± 57.5 Bq kg^{-1} respectively. For the phosphogypsum, 334.8 ± 8.8, 4.0 ± 1.4, and 199.9 ± 9.3 Bq kg^{-1} respectively and for soil samples range from 20.5 ± 7.3 to 175.7 ± 10.5 Bq kg^{-1} for ^{226}Ra, 15.5 ± 1.5 to 50.4 ± 0.6 Bq kg^{-1} for ^{232}Th and 89.5 ± 8.1 to 316.1 ± 41.3 Bq kg^{-1} for ^{40}K respectively. The mean absorbed dose rate was 71.4 nGy h^{-1}. The mean annual effective dose was 86 μSv.

Keywords Radioactivity · Phosphate ore · Phosphogypsum · Fertilizer plant

Phosphate ores which are the major raw material for phosphate fertilizer contain significant amount of natural radioactivity of mainly Uranium 238 [^{238}U], Thorium 232 [^{232}Th] and Potassium 40 [^{40}K], due to geological reasons (UNSCEAR 1988). Globally 80 %–90 % of mined phosphate rock is employed in the manufacture of phosphate fertilizer (Nehikhare 1987). The processing of the ores into fertilizer products and phosphoric acid results in the generation of phosphogypsum and other waste materials which are discharged as effluents to the environment (World Bank 1996). These effluents have been known to contain large quantities of primordial radionuclides (Erdem et al. 1995; Scholten and Timmermans 2005; Hamdy et al. 2007). The inadvertent exposure of man to these radionuclides through processing of the phosphate ore into fertilizer products, use of the phosphogypsum in building and farming activities constitutes means of radiation exposure (Fisenne et al. 1987; Shiraishi et al. 2000; UNSCEAR 2000).

The phosphate fertilizer plant is located in Kaduna North, Nigeria on latitude 10°31′27″ and on longitude 7°26′34″ East of Greenwich meridian. Effluents from the plant are first made to settle in the holding pond before being discharged into the public drainage.

There has not been any assessment, to the best of our knowledge, of the radiation impact of wastes generated by these plants on their immediate environment. This study aims to assess the environmental radiation dose occasioned by phosphate fertilizer production.

Materials and Methods

Three samples each of phosphate ore and phosphogypsum were collected. Three samples of soil were collected at every sampling point. The sampling point was usually divided into three distinct sampling zones or grids to ensure adequate representation.

The phosphate ore samples were collected from the warehouse at different points in polythene bags, sealed and properly labelled.

The phosphogypsum samples were collected from the effluent channel in the vicinity of the fertilizer plant.

Three soil samples each were collected at definite intervals from the immediate vicinity of the plant to 1 km away

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from the plant. The condition for soil sample collection was that phosphate fertilizer must not have been applied in the land for the past 5 years. The samples were collected at 5 cm depth to exclude dead organic matter (IAEA 1989).

The control soil samples were collected from Kaduna State in an uncultivated land 35 km from the plant area. This was to avoid contamination from phosphate ore dust or effluent discharge. The area was shown from preliminary studies to contain normal background level.

All the samples were air-dried at 33°C to avoid loss of radionuclides (IAEA 1989). The dried samples each were pulverized and made to pass through a 2 mm sieve. The samples were distinctly packed in plastic containers measuring 8.0 cm in diameter by 6.5 cm in height made to fit into the sodium iodide (NaI) gamma spectrometer counting chamber and labelled with codes 1, 2, 3 for each sample. They were left for 28 days for short-lived radionuclides to attain secular equilibrium. The same treatment was applied to the control samples.

The activity counting was carried out using a sodium iodide (NaI) gamma spectroscopy system. The system consists of a 76 × 76 mm NaI (TL) detector [Model 802 series] by Canberra Inc. connected to an Ortec series multichannel analyzer (MCA) through a preamplifier base and coupled to a personal computer. The computer has an uninterrupted power supply connection (UPS) to maintain regular voltage and safeguard the data in the system. There is also an attached printer to the computer.

The detector has a resolution of about 8 % at 662 keV of ^{137}Cs and is capable of distinguishing the gamma ray energies likely to be encountered in the measurements of the samples. The system was calibrated and the quality control carried out using a standard reference material soil IAEA-226 and IAEA-375 whose concentration of natural radioactivity has been certified by the IAEA. The spectral analysis was carried out using MAESTRO Software which identifies the photo peak, deducts the background and gives the total area under the peak.

Results and Discussion

Table 1 shows the mean activity concentration and radium equivalent of phosphate ore from Nigeria when compared with that from other countries. The table reveals that the radioactivity present in the phosphate ore used in Nigeria is mainly due to ^{238}U and its decay products. This finding is in agreement with studies from other countries (Sam and Holm 1995; Tufail et al. 2006; Uosif and El-Taher 2008). The higher the radioactivity content of the phosphate ore, the higher its radiation impact through mining, processing, use of its products and by-products.

Table 2 shows the mean activity concentrations of ^{226}Ra [^{238}U], ^{232}Th and ^{40}K radionuclides in all the samples collected. The mean activity concentration of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in phosphogypsum from this study were 334.8 ± 8.8, 4.0 ± 1.4 and 199.9 ± 9.3 Bq kg^{-1} respectively. When ^{226}Ra activity concentration in phosphogypsum is compared with that from control soil samples it is about 20 times [control soil is 16.1 Bq kg^{-1}].

The mean activity concentration of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in soil samples from around the holding pond (about 50 meters from the plant) to 1 km along the public drainage channel range from 20.5 ± 7.3 to 175.7 ± 10.5 Bq kg^{-1} for ^{226}Ra, 15.5 ± 1.5 to 50.4 ± 0.6 Bq kg^{-1} for ^{232}Th and 89.5 ± 8.1 to 316.1 ± 41.3 Bq kg^{-1} for ^{40}K respectively. The mean activity concentration of ^{226}Ra radionuclides for the soil samples around the holding pond is more than 10 times that of control soil and more than 5 times the world average activity in normal soil which is 35 Bq kg^{-1} (UNSCEAR 2000). The mean activity concentration of ^{226}Ra radionuclide in soil samples collected along the effluent channel 200 m away was 28.4 ± 3.0 Bq kg^{-1}, 300 m from the fertilizer plant was 21.4 ± 4.7 Bq kg^{-1} and 500 m from the fertilizer plant was 20.5 ± 7.3 Bq kg^{-1}. The mean activity concentration of ^{226}Ra radionuclide in the soil samples collected within public drainage channel 1 km away from the plant is 58.4 ± 5.5 Bq kg^{-1}. This value is greater than the world average activity concentration of ^{226}Ra in soil samples which is 10–50 Bq kg^{-1} (UNSCEAR 2000). This enhanced concentration is probably due to the presence of other waste products in the drainage channel.

The mean activity concentration of ^{226}Ra in the soil samples along the effluent channel was observed to decrease as the sampling distance increased from the fertilizer plant.

Table 1 Comparative activity concentration of ^{226}Ra, ^{232}Th and ^{40}K and corresponding Radium equivalent (Bq kg^{-1}) in phosphate ores from different countries

Country	^{226}Ra	^{232}Th	^{40}K	Ra.eq.	Reference
Abu-Tartor Egypt	287	23.7	21.4	323	
Nigeria	616.5	BDL	323.7	641	This study
Morocco	1,600	20	10	1,629	
Algeria	619	64	22	712	
Sudan (kurun)	555	0.83	23	558	Sam and Holm (1995)
Tanzania (Arusha)	5,022	717	286	6,069	

Table 2 Mean activity concentration of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in samples from super phosphate fertilizer plant, Kaduna and around its effluent (drainage) channel

	Sample	Activity Concentration (Bq kg^{-1} ± SD)		
		^{226}Ra	^{232}Th	^{40}K
1	Phosphate ore	616.5 ± 38.6	BDL	323.7 ± 57.7
2	Phosphogypsum collected 15–20 m away from plant in the effluent channel	334.8 ± 8.8	4.0 ± 1.4	199.9 ± 9.3
3	Soil samples collected from the holding pond in the compd. 50 m from the plant	175.7 ± 10.5	15.7 ± 1.7	218.8 ± 9.8
4	Soil collected in the compd. 100 m from the plant	110.9 ± 15	22.4 ± 7.6	316.1 ± 41.3
5	Soil along urban drainage channel about 200 m from the plant	28.4 ± 3.0	50.4 ± 0.6	155.5 ± 2.7
6	Soil along the urban drainage channel about 300 m from the plant	21.4 ± 4.7	19.6 ± 1.6	238.0 ± 5.1
8	Soil along urban drainage channel about 500 m from the plant	20.5 ± 7.3	47.3 ± 1.0	253.6 ± 9.5
10	Soil samples collected inside the urban drainage channel 1 km away	58.4 ± 5.5	15.5 ± 1.5	89.5 ± 8.1
12	Control soil sample	16.1 ± 1.2	2.3 ± 0.4	169.3 ± 6.0

BDL below detection level

The absorbed dose rate in air at a height of 1 m above the ground is employed when considering radiation risks to humans and other fauna and flora.

The absorbed dose D (nGy h^{-1}) was calculated using the equation;

$$D = 0.462A_U + 0.604c_{Th} + 0.0417c_K \text{ (UNSCEAR 2000)}$$

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

The dose rate in air due to phosphogypsum was 154.67 nGy h^{-1}. This value is about 2.6 times the world's average dose rate for normal soil (59 nGy h^{-1}) and about 10 times the value for control soil samples for this study (15.55 nGy h^{-1}).

The dose rate in air for the soil samples range from 39.13 to 95.03 nGy h^{-1} with mean of 71.4 nGy h^{-1}.

The absorbed dose rate in air (nGy h^{-1}) at 1 m above the ground does not directly give the radiological health hazard to which the population is exposed (Obed et al. 2005). There are two factors that must be considered, that which converts Gy to Sv accounting for the biological effectiveness of the dose in causing damage in human tissue and the occupancy factor which specifies the total time spent outdoors. UNSCEAR (2000) recommended 0.7 Sv Gy^{-1} and 0.2 as the values for the two factors. A coefficient of 0.7 Sv Gy^{-1} used to convert absorbed dose in air to effective dose for adult (both indoors and outdoors) was based on the analysis in the UNSCEAR (1982) report of experimental and calculated data on environmental exposure to gamma rays of moderate energy. In Nigeria an average person spends more time indoors than outdoors because of weather conditions and as such the occupancy factor of 0.2 was considered appropriate for this work. Obed et al. (2005) adopted these two factors in their study in Nigeria.

Using the mean absorbed dose values in Table 3 and applying the above factors, the effective dose rate due to the activities of the fertilizer plant were calculated thus;

$$\text{Effective dose} = D\ [\text{nGy h}^{-1}] \times 0.7 \times 0.2 \times 8760\ \text{h [Time in hours per year]}$$

The outdoor annual effective dose for soil samples range from 39 to 117 μSv. These values are however less than the world average annual outdoor effective dose of 480 μSv in normal soil (UNSCEAR 2000).

The result revealed that the main radioactivity of the phosphate ore used for fertilizer production was due to ^{238}U. The radioactivity from the phosphate ore was not grossly affected by processing during fertilizer production. They are therefore likely to be transferred to the plants when applied on them. The study is in keeping with other earlier works that implicating phosphate fertilizer as a significant source of radionuclide transfer to the food chain. The mean activity concentration due to ^{238}U for phosphate ore from Nigeria is higher than that from Algeria, Egypt, Sudan and Tunisia. It is 17.6 times average world activity concentration in normal soil and 38 times that of control soil.

The mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in phosphogypsum were 334.8 ± 8.8, 4.0 ± 1.4 and 199.9 ± 9.3 Bq kg^{-1} respectively. This is about 20 times that of control soil and about 9.5 times the world average in normal soil samples. The phosphogypsum generated as by-products are usually either discharged into the environment or employed in plaster and cement industries. Some studies, including this present one, have investigated the influence of phosphogypsum in dose consideration to members of the public and found the absorbed dose rate to be high even when dose considerations from radon gas was not included in the assessment.

Table 3 Absorbed dose rate in air (nGy h^{-1}) and outdoor annual effective dose for the samples

	Sample	Absorbed dose rate in air (nGy h^{-1})	Outdoor annual effective dose E (μSv)
1	Phosphogypsum collected 15–20 m away from plant in the effluent channel	154.67	180
2	Soil samples collected from the holding pond in the compd. 50 m from the plant	95.03	117
3	Soil and mat collected in the compd. 50 m from the holding pond	75.78	93
4	Soil in along urban drainage channel about 200 m from the plant	52.28	64
5	Soil along the urban drainage channel about 300 m from the plant	32.22	39
6	Soil along urban drainage channel about 500 m from the plant	50.94	63
7	Soil samples collected inside the urban drainage channel 1 km away	39.13	48
	Control soil sample	15.55	

The mean activity concentration of radionuclides in the soil samples collected within the plant complex and along the effluent channel range from 20.5 ± 7.3 to 175.7 ± 10.5 Bq kg^{-1} for ^{226}Ra, 15.5 ± 1.5 to 50.4 ± 0.6 Bq kg^{-1} for ^{232}Th and 89.5 ± 8.1 to 916.1 ± 41.3 Bq kg^{-1} for ^{40}K. An interesting finding was that the mean activity concentration of ^{226}Ra in soil samples from the holding pond is about 10 times that of control soil while soil samples in public effluent channel about 1 km away from the plant showed ^{226}Ra mean activity concentration 3.6 times that of control soil samples. The absorbed dose rate range from 32.22 to 95.03 nGy h^{-1} with mean of 71.4 nGy h^{-1}. This is higher than the world average of 59 nGy h^{-1}. This study has shown that the activity of phosphate fertilizer plants impact adversely on the environment.

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