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A thorough investigation of the uranium concentration in phosphate mines: a case study of Minjingu phosphate mine, Arusha, United Republic of Tanzania

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Abstract: Using high resolution gamma ray spectrometry, the terrestrial radionuclides from all phosphate rock samples were measured and results are presented. From the measured gamma ray spectra, activity concentrations were determined for ^{226}Ra/^{238}U (range from 122.5±6.9 to 8060±806 Bq kg^{-1}), ^{232}Th (range from 13.2±0.8 to 1201.3±77.3 Bq kg^{-1}), ^{40}K (range from 12.9±1.3 to 1827.6±93.9 Bq kg^{-1}) and ^{137}Cs (range from 0.5±0.017 to 70.58±1.25 Bq kg^{-1}) with mean of 1832.32±00, 447.62±00, 399.46±00 and 22.98±0.43 Bq kg^{-1} respectively. Radium equivalent activity ranges from 219.27±00 to 9127.07±00 Bq kg^{-1} with mean of 2433.56±00 Bq kg^{-1}. The total absorbed dose estimated at 1 metre above the ground varies from 102.1 to 3967.8 nGyh^{-1} with an average of 1055.9 nGy h^{-1}. Investigation of uranium recorded a peak of 650±65ppm, this suggests the existing U concentration is over three times greater than economically viable and production of a cleaner fertiliser is favourable.

Keywords: natural radioactivity; activity concentrations; absorbed dose rate; outdoor annual effective dose; radium equivalent activity; external hazard indices; Minjingu; phosphate.

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1 Introduction

Phosphate is one of the most important industrial materials in the farming activities all over the world. In some of the phosphate-producing localities, uranium is associated with apatite, a mineral which can hold uranium in its composition. This uranium could be extracted as a by-product in the manufacturing of phosphoric acid assuming an average recoverable content of 200 ppm. The use of contaminated standard phosphate fertilisers in farming has been linked to significant raised uranium level in the environment.

Phosphate rocks are materials extensively used, mainly as a source of phosphorus for fertilisers and further for producing phosphoric acid and other special chemicals. Phosphate is also used in animal feed supplements, food preservatives, anti-corrosion agents, cosmetics, fungicides, ceramics, water treatment and metallurgy. Phosphates are typically rich in uranium and as such are one of the sources of Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM), which might increase exposure to man from natural radionuclides (Abbady et al., 2005).

^{238}U is generally found in radioactive equilibrium with its decay products, such as ^{226}Ra. The activity concentrations of ^{232}Th and ^{40}K in sedimentary phosphate rocks are much lower than those of ^{238}U, and comparable to those normally observed in soil. The primary potential environmental radiation problem associated with phosphate rock mining and processing concerns mining spoils and processing waste products. While these materials do not present a direct radiation hazard, problems may be created by their use. Occupational exposures mainly occur during mining, processing and transportation of phosphate rock, as well as during transportation and utilisation of phosphate fertilisers (UNSCEAR, 1989).

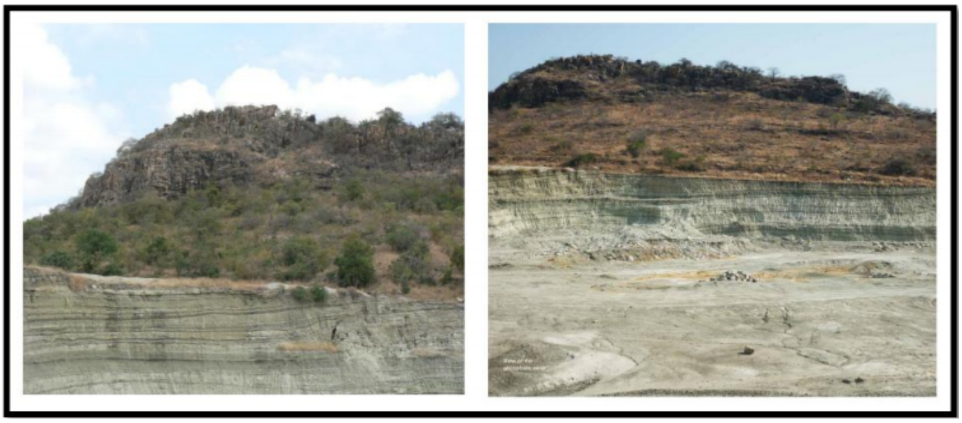
The impacts of crushing, grinding, and sorting at early stages of beneficiation process produce airborne particulates and deteriorate the air quality. Dust emissions during loading, en route, and unloading of the ore equally affect the quality of urban air in the vicinity of the mine area. Radioactive phosphatic dust can be precipitated at and around the mine areas in the form of wet precipitation and/or dry deposition. The radioactive dust can be accumulated on surface soils, become bio-available by animals and plants and enter the food chain. Radon gas produced from phosphate piles and tailings is also of concern and can be considered as a potential hazard (UNSCEAR, 1993; International Atomic Energy Agency [IAEA], 2004).

Adverse health impacts associated with phosphate mining stem from the inhalation of particulate emissions and the intake of heavy metals, metalloids, non-metals and their oxides, either from the mining activities or from the application of the phosphatic fertilisers. Radiation hazards are also of concern as most of the phosphate mines are rich in Naturally Occurring Radioactive Materials (NORM) owing to thorium and uranium decay series ^{232}Th and ^{238}U (Checkoway et al., 1993), and are considered as a radiation health hazard (Abbady et al., 2005; Makweba and Holm, 1993). Air particulates emissions produced during processing and transportation of phosphate ore represent a potential health hazard; they might cause respiratory problems at least to the miners (Khater et al., 2004).

1.1 Background of the study area

The phosphate ore at Minjingu (Figure 1) is mined by the open dry process, a method that can lead to the dispersion of large amounts of dust to the environment. Such emissions may give rise to enhanced exposure to naturally occurring radionuclides through air, soil, leaf vegetation, water and animal products to the population living around the mine (Banzi et al., 2000).

Figure 1 Cross-section of phosphate bedrocks at Minjingu phosphate mine, Arusha



The Minjingu phosphate mine is within the village of Minjingu, which has about 2500 inhabitants. One of the important features of the Minjingu area is its location in a semi-arid zone. The human habitat in this village may be affected by enhanced radiation exposure due to gamma rays, radon and radon progeny, as well as ore dust through exposure pathways.

Despite the need for further research, there have been some previous studies of the uranium concentration at Minjingu mine. Bianconi (1987) reported that the Minjingu phosphate deposit has uranium activity with a maximum of 800 ppm U_3O_8. Further study carried out on the phosphate rock at Minjingu (Mustonen and Annanmaki, 1988) has shown that the concentration of ^{238}U in the uppermost phosphate layer is 9550 Bq kg^{-1} and the cumulative ambient radiation level recorded in four weeks is 0.233 mSv (0.35 μSv h^{-1}). Measurements made using a portable dose rate meter indicated 2 μSv h^{-1} (Banzi et al., 2000).

Another similar study (Makweba and Holm, 1993) reported the range of ^{238}U concentration in the soft phosphate of Minjingu to be between 1500 and 11,000 Bq kg^{-1} and an outdoor dose rate in air of 0.4 per month (0.59 μSv h^{-1}) at the offices of the mining company. Both studies have indicated that the Minjingu deposit has a relatively high content of uranium and its daughters when compared with the reported uranium activity levels in phosphate deposits worldwide (UNSCEAR, 2000). According to the radiation protection philosophy, these findings prompt follow-up investigations (Banzi et al., 2000).

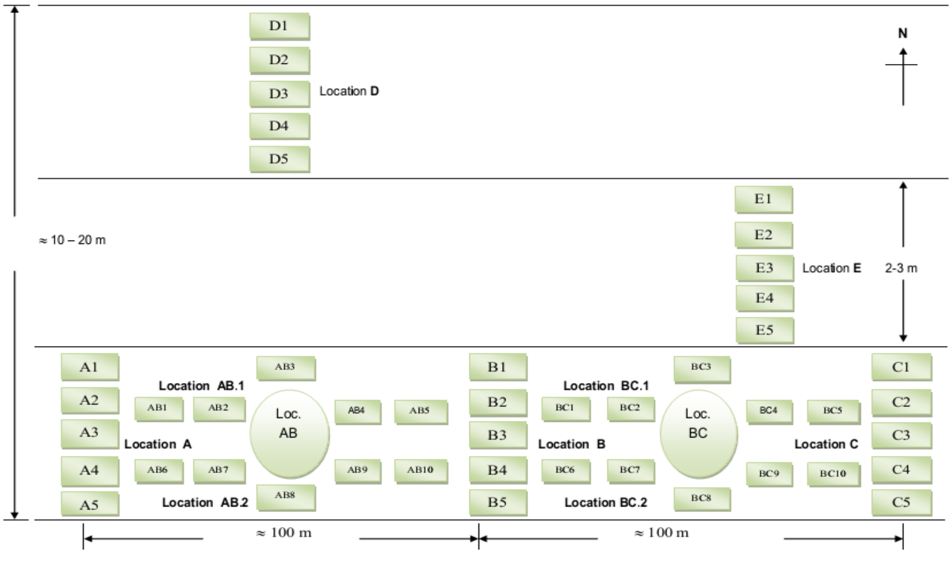
The objective of the present study was to establish radioactivity working levels in the Minjingu phosphate mine by quantifying uranium concentration through measured activities and assessing the radiation hazard from the findings of the experimental results.

2 Materials and method

2.1 Sample collection and preparation

A total of 45 phosphate rock samples contained in overlying and underlying rock beds from surface and sub-surface sections of Minjingu mine were collected and prepared for measurement and analysis. Representative samples extracted from the mine area were mapped and labelled ‘Location A’ (03°42′40″S; 035°54′72″E), ‘Location B’ (03°42′41″S; 035°54′73″E) and ‘Location C’ (03°42′51″S; 035°54′71″E) (Figure 2); samples in these locations were extracted through the channel sampling technique.

Figure 2 A layout showing sample locations in the phosphate rock beds of Minjingu



Source: Authors’ own extract (2013)

Other locations identified were labelled ‘Location AB’ and ‘Location BC’. These were regions between locations A and B and between locations B and C, respectively. In these locations, samples were extracted through the grab and or random sampling technique. The essence was to enable a close approximation especially when calculating mean radioactivity concentrations of the most critical area of the mine.

On the other hand, it was also necessary to extract samples from overburden layers. As such, samples were extracted from the topmost and the second from the top layer which were labelled ‘Location D’ (03°42′31″S; 035°54′80″E) and ‘Location E’ (03°42′26″S; 035°54′92″E), respectively (Figure 2). Samples extracted from both locations D and E were excavated across the bedrocks by using the channel sampling technique.

These samples were dried, crushed and milled to 50 meshes. The mass of the milled samples varied around approximately half a kilogram each. As a requirement for natural radioisotope analysis, samples were packed in plastic containers which were completely and airtightly sealed and kept for at least four weeks to attain a state of secular equilibrium where the rate of decay of the daughters becomes equal to that of the parent (Mustonen and Annanmaki, 1988). This step is necessary to ensure that radon gas is confined within the volume and the decay products also remain in the sample.

2.2 Experimental details and technique

The natural radioactive elements in the samples were determined using a high resolution gamma ray spectroscopy technique (see details below). In a traditional way, environmental radiation is measured to identify the radioactive sources in the environment, quantify their concentration and estimate their dose contribution. Although this is mostly achieved by gamma ray spectrometric technique, the most essential part of such technique remains the methodology associated with the detector's calibration, measurement procedure, analysis and interpretation of data.

In the present study, a high resolution gamma spectrometry system is used for measurement of the energy spectrum of the emitted gamma rays in the energy range of 200 to ~2000 keV. The system consists of high purity germanium (HPGe) detector coupled to a signal processing units including a spectroscopic pulse amplifier and multichannel analyser. Two HPGe detector systems whose specifications are given below were used for sample measurements within a range of 7200–8640 s of counting live time. Detector 1 (D1): a high resolution gamma spectrometer uses *p*-type HPGe detector Model GEM-100210-Plus, Serial No. 38-P40884B, 100% efficiency, and 2.3 keV resolutions at 1.33 MeV Co-60 line. Detector 3 (D3): a high resolution gamma ray spectrometer uses *n*-type HPGe detector Model GMP-100250-S and Serial No. 38-N31278A. The detector has a relative efficiency of 100% and resolution of 2.1 keV at 1.33 MeV of Co-60 line and a peak-to-Compton ratio (P/C) of 64:1.

The gamma ray energies of radionuclides of interest were pre-defined in the peak analysis library to allow the program to concentrate on the analysis of those elements of interest. The radionuclides were identified using gamma ray spectrum analysis software, ORTEC HPGe. The specific radioactivity of ^{40}K was measured directly by its own gamma ray at 1460.75 keV (10.7%), while specific activity of ^{232}Th was determined from the weighted mean value and average concentrations of ^{212}Pb at 238.63 keV (43%), ^{208}Tl at 583.14 keV (86%),and ^{228}Ac at 911.07 keV (29%) in the samples.

The concentration of ^{226}Ra (^{238}U) was determined from the weighted mean value and average concentrations of the ^{214}Pb at 295.22 keV (19.20%) and at 351.99 keV (37.10%), and ^{214}Bi at 609.30 keV (46.90%), at 1120.28 keV (15.04%) and at 1764.51 keV (15.9) decay products (El-Taher et al., 2007). The values inside the brackets next to energy indicate the absolute gamma ray emission probability. The specific activity (A) in Bq kg^{-1} of the radionuclides in the samples was calculated after decay correction using the equation (1) below:

$$A = \left(\frac{N_p}{T_L P_\gamma \varepsilon M} \right) \tag{1}$$

where M is the mass of dry sample (kg), N_p is the net peak area for the sample in the peak range, P_γ is the gamma emission probability, T_L is the counting live time; and $\mathcal{E}$ is the photo peak efficiency (L'Annunziata, 2003).

2.3 Calculation of radiological effects

The activity levels of natural radiations are represented by a single quantity to estimate the radiological hazard and consequently the environmental impacts of radiations. Among several radioactive nuclides, ^{226}Ra is often chosen in the majority of the published papers concerned with the environmental radiation studies. This is attributed to the fact that the external exposure to the population is mostly by gamma rays, emitted from the two main daughters of radium, namely ^{214}Pb and ^{214}Bi.

It is essential to take note that about 98.5% of the radiological effects of the ^{238}U series are produced by radium and its daughters. Therefore, any disequilibrium between ^{238}U and ^{226}Ra has no effect on the dose estimation from the measurement of ^{226}Ra, and the dose rates derived from ^{226}Ra are usually presented as that of ^{238}U (Jibiri and Bankole, 2006; Beretka and Mathew, 1985).

2.3.1 Outdoor external absorbed dose rate in air (DR)

The outdoor external absorbed dose rate (DR) in air 1 m above the ground surface to the population is calculated from the activities of the Ra, Th and K radionuclides. It is defined as:

$$\mathrm{DR}=\mathrm{C}_{\mathrm{Ra}} \times \mathrm{A}_{\mathrm{Ra}} + \mathrm{C}_{\mathrm{Th}} \times \mathrm{A}_{\mathrm{Th}} + \mathrm{C}_{\mathrm{K}} \times \mathrm{A}_{\mathrm{K}} \qquad (2)$$

where DR is outdoor external absorbed dose rate in nGy h^{-1}, and A_{Ra}, A_{Th} and A_K are the activities (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K, respectively. C_{Ra}, C_{Th} and C_K are the conversation factors in nGy h^{-1} per Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. These factors were taken as means of those from Beck and Planque (1968), Saito and Jacob (1995) and Clouvas et al. (2000). Their mean values are $C_{Ra} = 0.4368$, $C_{Th} = 0.5993$ and $C_K = 0.0417$.

2.3.2 Annual effective dose rate (AEDR)

The Annual Affective Dose Rate (AEDR) is estimated by using conversion coefficient from absorbed dose in air to effective dose (0.7 $SvGy^{-1}$) and the outdoor occupancy factor (0.2) proposed by UNSCEAR (2000). Also, its estimation depends on the assumption that the annual average time for exposure to radiation is 8760 hrs. Accordingly, this index is given in millisievert per year (mSv y^{-1}) and calculated as:

$$\mathrm{AEDR}\left(\mathrm{mSv\,y}^{-1}\right) = \mathrm{dose\,rate}\left(\mathrm{nGy\,h}^{-1}\right) \times 8760 \times 0.2 \times 0.7 \times 10^{-6} \qquad (3)$$

The proposed indoor occupancy factor is 0.8 (this is mainly used to determine exposure in building materials). The recommended values of AEDR are 20 mSv y^{-1} for occupational members and 1 mSv y^{-1} for the public (IAEA, 1996; ICRP, 1990).

2.3.3 Radium equivalent activity (Ra$_{eq}$)

The natural radioactivity contained in phosphate rock samples is determined from ^{226}Ra, ^{232}Th and ^{40}K contents. As radium and its daughter products produce 98.5% of the radiological effects of the uranium series, the contribution from the ^{238}U has been replaced with the decay product ^{226}Ra. Radium equivalent activity is an index that has been introduced to represent the specific activities of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazards associated with them.

However, the distribution of ^{226}Ra, ^{232}Th and ^{40}K in any concerned sample or material is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity expressed in Bq kg^{-1} to compare with the specific activity of samples (materials) containing different amount of Ra, Th and K.

Radium equivalent, therefore, is the weighted sum of activities of the above three radionuclides based on the estimation that 370 Bq kg^{-1} of Ra, 259 Bq kg^{-1} of Th and 4810 Bq kg^{-1} of K produce the same gamma ray dose rates. This quantity index, radium equivalent (Ra$_{eq}$), is hereby defined as:

$$\mathrm{Ra_{eq}} = \mathrm{A_{Ra}} + 1.43\mathrm{A_{Th}} + 0.077\mathrm{A_K} \leq 370\ \mathrm{Bq\ kg^{-1}} \tag{4}$$

where A$_{Ra}$, A$_{Th}$ and A$_K$ are activities (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The maximum value of this index must be <370 Bq kg^{-1}for keeping the external dose <1.5 mGy y^{-1} (Jibiri and Bankole, 2006; Akhtar et al., 2005).

3 Results and discussion

Table 1 (and Figure 3) summarises the results of measurements of naturally occurring radionuclide (^{238}U/^{226}Ra, ^{232}Th and ^{40}K) concentrations in the vertical orientation of the collected phosphate rock samples of Minjingu mine. The summary of results from Table 1, and at least through the calculated radium equivalent activity, shows that, in general, the average and ranges of activity concentration of ^{238}U, ^{232}Th and their decay products and ^{40}K in phosphate rocks of Minjingu are higher than the world figures reported in UNSCEAR (2000). Table 2 (and Figure 4) shows activity concentrations in lateral orientation of the extracted samples.

The range of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K and ^{137}Cs in the phosphate rock samples from the studied locations of the mine varies from 122.5±6.9 Bq kg^{-1} (D3) to 8060.0±806 Bq kg^{-1} (C1), 13.2±0.8 Bq kg^{-1} (D4) to 1201.3±77.3 Bq kg^{-1} (E4) and 12.9±0.7 Bq kg^{-1} (BC9) to 1827.6±93.9 Bq kg^{-1} (D5) and 0.2±0.01 Bq kg^{-1} to 70.6±4.8 Bq kg^{-1} for ^{137}Cs with arithmetic mean values of 1832.3±91.6 Bq kg^{-1}, 447.6±22.4 Bq kg^{-1}, 399.5±19.9 Bq kg^{-1} and 22.9±1.2 Bq kg^{-1}, respectively.

Observation of high concentrations of ^{226}Ra and ^{232}Th in phosphate rock samples relative to the world average may be correlated with the presence of high concentration of uranium in the phosphate rocks at Minjingu mine. Some related findings were also reported in earlier studies (Makweba and Holm, 1993; Banzi et al., 2000). On the other hand, an account of traces of ^{137}Cs concentration may suggest that radioactivity fallouts especially from airborne radiation which are mainly associated with nuclear weapons tests carried out in the 1950s and 1960s. Also, uncontrolled releases of radionuclides from nuclear and radiological accidents are probably the source of such environmental contamination.

Moreover, Table 3 (and Figure 5) fairly summarises the experimental findings and shows that the Minjingu phosphate deposit has uranium activity with a maximum of 650±65 ppm U_3O_8 (an equivalent of 8060±806 Bq kg^{-1}). These findings could be approximately compared to those obtained by Bianconi (1987) who reported the activity concentration of uranium at the maximum of 800 ppm U_3O_8 (an equivalent of 9920 Bq kg^{-1}). Another study carried out at Minjingu (Mustonen and Annanmaki, 1988) had shown that the concentration of ^{238}U in the uppermost phosphate layer was 9550 Bq kg^{-1}.

Figure 3 Concentration of terrestrial radionuclides for samples extracted in vertical orientation

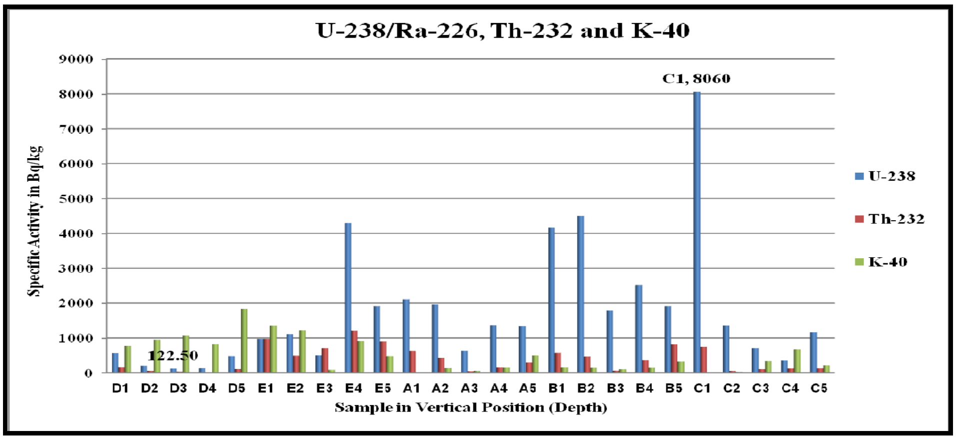


Figure 4 Concentration of terrestrial radionuclides for samples extracted in lateral orientation

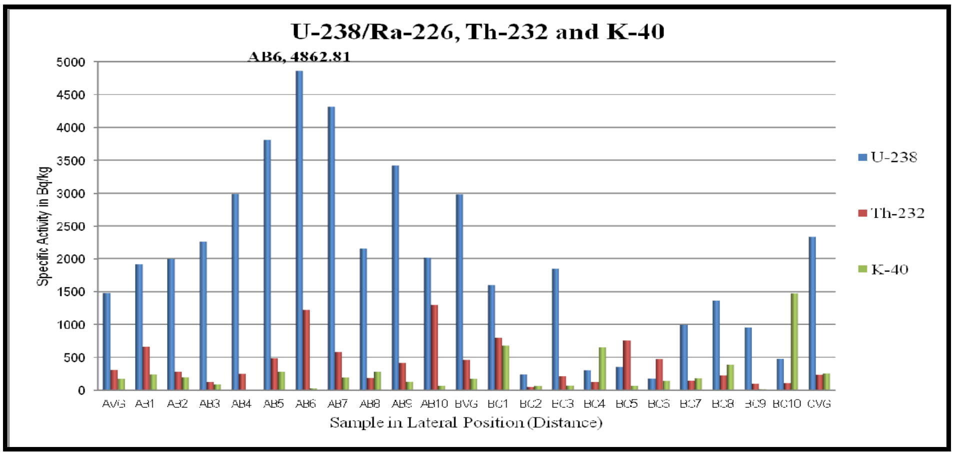


Table 1 Activity concentration of terrestrial radionuclides $^{238}U/^{226}Ra$, ^{232}Th and ^{40}K and radiation indices for samples in vertical position

	Activity concentration in Bq kg^{-1}				*Radiation hazard indices*					
Sample	$^{238}U/^{226}Ra \pm E$	$^{232}Th \pm E$	$^{40}K \pm E$	$^{137}Cs \pm E$	Ra_{eq}	H_{ex}	H_{in}	*DR nGy h^{-1}*	*AEDR mSv y^{-1}*	*Activity index*
D1	570.4 ± 57.0	164.0 ± 16.4	778.2 ± 77.8	–	864.8	2.3	4.1	379.9	0.5	5.9
D2	194.6 ± 11.2	58.3 ± 3.8	945.5 ± 48.7	2.1 ± 0.16	350.7	0.9	4.0	159.3	0.2	2.5
D3	122.5 ± 6.9	30.6 ± 1.9	1073.6 ± 55.1	0.7 ± 0.04	248.9	0.7	4.4	116.6	0.1	1.8
D4	137.3 ± 7.6	13.2 ± 0.8	818.9 ± 42.2	13.2 ± 0.7	219.3	0.6	3.3	102.1	0.1	1.6
D5	480.9 ± 27.3	106.2 ± 6.8	1827.6 ± 93.9	6.4 ± 0.5	773.5	2.1	7.8	349.9	0.4	5.5
E1	973.5 ± 56	975.6 ± 63.4	1354.9 ± 69.8	13.9 ± 1	2472.9	6.7	11.0	1066.4	1.3	17.2
E2	1100.6 ± 63.3	487.3 ± 31.8	1214.6 ± 62.6	16.5 ± 1.2	1890.9	5.1	7.7	823.4	1.0	13.0
E3	496.1 ± 27.7	701.6 ± 45.4	85.3 ± 4.4	0.5 ± 0.03	1506.4	4.1	4.4	640.9	0.8	10.4
E4	4293.2 ± 244	1201.3 ± 77.3	907.9 ± 46.7	64.4 ± 4.5	6083.0	16.4	11.3	2633.9	3.2	41.3
E5	1909.6 ± 190.9	893.8 ± 89.4	479.9 ± 47.9	–	3224.7	8.7	7.4	1389.8	1.7	21.9
A1	2108.0 ± 210.8	627.3 ± 62.7	<DL	–	3005.0	8.1	4.0	1296.7	1.6	20.3
A2	1961.8 ± 110.4	425.3 ± 27	152.8 ± 7.8	28.3 ± 1.9	2580.7	6.9	3.4	1117.6	1.4	17.4
A3	631 ± 35.6	45.2 ± 2.7	69.9 ± 3.5	8.9 ± 0.6	700.9	1.9	0.6	305.5	0.4	4.7
A4	1367.1 ± 77.1	158.6 ± 10.9	168.9 ± 8.6	18.5 ± 1.3	1606.1	4.3	1.8	698.8	0.9	10.8
A5	1336.5 ± 76.9	290.29 ± 19.03	502.9 ± 25.9	12.7 ± 0.9	1789.4	4.8	3.8	778.2	0.9	12.1

Table 1 Activity concentration of terrestrial radionuclides $^{238}U/^{226}Ra$, ^{232}Th and ^{40}K and radiation indices for samples in vertical position (continued)

	Activity concentration in Bq kg^{-1}				*Radiation hazard indices*					
Sample	*$^{238}U/^{226}Ra \pm E$*	*$^{232}Th \pm E$*	*$^{40}K \pm E$*	*$^{137}Cs \pm E$*	*Ra_{eq}*	*H_{ex}*	*H_{in}*	*DR nGy h^{-1}*	*AEDR mSv y^{-1}*	*Activity index*
B1	4158.4 ± 239.7	572.9 ± 38.9	160.1 ± 8.2	59.1 ± 4.3	4990.0	13.5	4.8	2166.4	2.7	33.6
B2	4500.7 ± 254.1	470.7 ± 30.6	151.6 ± 7.8	70.6 ± 4.8	5185.5	14.0	4.2	2254.3	2.8	34.8
B3	1793.97 ± 101.3	64.2 ± 3.8	100.3 ± 5.2	6.3 ± 0.4	1893.6	5.1	1.1	826.3	1.0	12.7
B4	2514.3 ± 143.2	367.5 ± 24.1	152.1 ± 7.8	36.9 ± 2.6	3051.5	8.3	3.2	1324.8	1.6	20.5
B5	1909.6 ± 190.9	815.9 ± 81.6	326.8 ± 32.7	–	3101.3	8.4	6.3	1336.6	1.6	21.1
C1	8060.0 ± 806	746.2 ± 74.6	<DL	–	9127.1	24.7	5.9	3967.8	4.9	61.2
C2	1357.7 ± 76.8	56.9 ± 3.37	25.7 ± 1.3	8.7 ± 0.5	1441.0	3.9	0.7	628.2	0.8	9.6
C3	703.4 ± 39.9	102.7 ± 6.7	338.1 ± 17.4	9.2 ± 0.6	876.3	2.4	2.0	382.9	0.5	5.9
C4	359.6 ± 20.5	128.3 ± 7.9	676.7 ± 34.9	11.3 ± 0.6	595.1	1.6	3.4	262.2	0.3	4.1
C5	1166.6 ± 66.1	131.6 ± 8.2	212.6 ± 10.9	5.3 ± 0.4	1371.1	3.7	1.8	597.3	0.7	9.2

Table 2 Activity concentration of terrestrial radionuclides $^{238}U/^{226}Ra$, ^{232}Th and ^{40}K and radiation indices for samples in lateral position

	Activity concentration in Bq kg^{-1}				*Radiation hazard indices*					
Sample	$^{238}U/^{226}Ra \pm E$	$^{232}Th \pm E$	*40K ± E*	$^{137}Cs \pm E$	Ra_{eq}	H_{ex}	H_{in}	*DR nGy h^{-1}*	*AEDR mSv y^{-1}*	*Activity index*
AVG	1481 ± 74.1	309.3 ± 15.5	169.3 ± 8.5	–	1936.4	5.2	2.7	839.4	1.0	13.1
AB1	1920.8 ± 108.2	660.9 ± 38	236.9 ± 12.1	29.6 ± 2	2884.2	7.8	5.1	1244.9	1.5	19.6
AB2	1993.8 ± 114.7	284.4 ± 18.6	197.7 ± 10.2	<DL	2415.7	6.5	2.8	1049.6	1.3	16.3
AB3	2261.5 ± 128.4	124.2 ± 7.7	90.4 ± 4.7	<DL	2446.1	6.6	1.5	1066.0	1.3	16.4
AB4	2988.4 ± 298.8	246.0 ± 24.6	<DL	–	3340.2	9.0	2.0	1452.8	1.8	22.4
AB5	3808.6 ± 214.9	481.5 ± 31.2	282.4 ± 14.5	55.9 ± 3.8	4518.8	12.2	4.6	1963.9	2.4	30.4
AB6	4862.8 ± 278.9	512.2 ± 33.9	24.1 ± 1.2	5.7 ± 0.4	6609.1	17.9	8.1	2856.2	3.5	44.6
AB7	4315.4 ± 246.6	577.3 ± 38.2	194.7 ± 9.9	66.9 ± 4.7	5155.9	13.9	4.9	2239.0	2.7	34.7
AB8	2157.6 ± 215.8	188.6 ± 18.9	274.9 ± 27.5	–	2448.5	6.6	2.0	1066.9	1.3	16.5
AB9	3424.3 ± 194.6	416.8 ± 26.7	124.8 ± 6.4	9.0 ± 0.5	4029.9	10.9	3.6	1750.7	2.1	27.1
AB10	2011.7 ± 120	493.1 ± 31.2	63.6 ± 3.5	25.4 ± 2.10	3871.4	10.5	8.1	1658.7	2.0	26.4

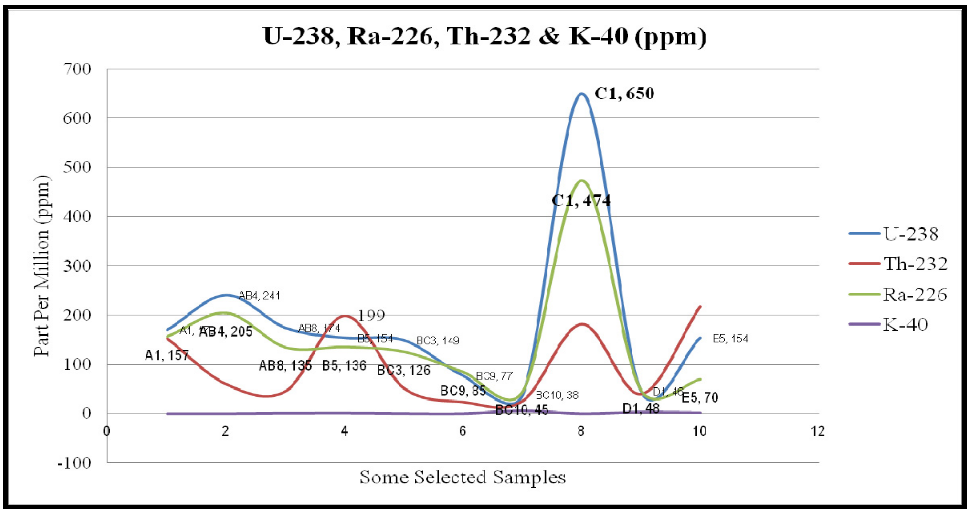
Table 2 Activity concentration of terrestrial radionuclides ^{238}U/^{226}Ra, ^{232}Th and ^{40}K and radiation indices for samples in lateral position (continued)

	Activity concentration in Bq kg^{-1}				*Radiation hazard indices*					
Sample	*$^{238}U/^{226}Ra \pm E$*	*$^{232}Th \pm E$*	*40K ± E*	*$^{137}Cs \pm E$*	*Ra_{eq}*	*H_{ex}*	*H_{in}*	*DR nGy h^{-1}*	*AEDR mSv y^{-1}*	*Activity index*
BVG	2975.4 ± 149	458.2 ± 23	177.6 ± 8.9	–	3644.4	9.8	3.9	1581.7	1.9	24.5
BC1	1602.7 ± 96.5	794.9 ± 57.5	470.4 ± 24.8	<DL	2791.3	7.5	7.5	1204.7	1.5	19.1
BC2	237.9 ± 14.1	45.9 ± 2.9	317.5 ± 15.9	<DL	307.9	0.8	0.5	133.8	0.2	2.1
BC3	1847.6 ± 184.8	213.2 ± 21.3	72.6 ± 7.3	–	2158.1	5.8	1.9	937.8	1.2	14.5
BC4	303.6 ± 16.4	123.6 ± 7.7	464.0 ± 27.5	<DL	530.6	1.4	3.3	233.9	0.3	3.7
BC5	352.2 ± 21.4	756.9 ± 50.4	2147.2 ± 107.5	<DL	1439.4	3.9	4.7	610.1	0.7	9.9
BC6	179.4 ± 10.8	184.3 ± 11.5	138.6 ± 7.1	0.2 ± 0.02	862.6	2.3	3.3	365.9	0.4	5.9
BC7	991.8 ± 56.9	140.4 ± 9.3	182.7 ± 9.4	<DL	1206.5	3.3	1.7	524.9	0.6	8.1
BC8	1360.5 ± 77.9	223.6 ± 14.8	391.4 ± 20.1	19.9 ± 1.41	1710.3	4.6	3.1	744.6	0.9	11.6
BC9	954.8 ± 95.5	98.4 ± 9.8	12.9 ± 1.3	–	1096.5	2.9	0.8	476.6	0.6	7.4
BC10	471.2 ± 47.1	106.6 ± 10.7	1475.9 ± 147.6	–	737.3	1.9	6.4	331.3	0.4	5.2
CVG	2329.4 ± 116.5	233.1 ± 11.7	250.6 ± 12.6	–	2682.1	7.2	2.8	1167.7	1.4	18.0

Table 3 Terrestrial radionuclides concentrations expressed in parts *per* million (ppm)

	Sample	^{238}U		^{232}Th		^{226}Ra		^{40}K	
	ID no.	*ppm*	*Bq kg^{-1}*	*ppm*	*Bq kg^{-1}*	*ppm*	*Bq kg^{-1}*	*ppm*	*Bq kg^{-1}*
Selected locations	A1	170 ± 17	2108 ± 210.8	153 ± 15.3	627.3 ± 62.7	157 ± 15.7	1946.8 ± 194.7	<DL	<DL
	AB4	241 ± 24.1	2988.4 ± 298.8	60 ± 6	246 ± 24.6	205 ± 20.5	2542 ± 254.2	<DL	<DL
	AB8	174 ± 17.4	2157.6 ± 215.8	46 ± 4.6	188.6 ± 18.9	135 ± 13.5	1674 ± 167.4	1.06 ± 0.11	274.9 ± 27.5
	B5	154 ± 15.4	1909.6 ± 190.9	199 ± 19.9	815.9 ± 81.6	136 ± 13.6	1686.4 ± 168.6	1.26 ± 0.13	326.8 ± 32.7
	BC3	149 ± 14.9	1847.6 ± 184.8	52 ± 5.2	213.2 ± 21.3	126 ± 12.6	1562.4 ± 156.2	0.28 ± 0.03	72.6 ± 7.3
	BC9	77 ± 7.7	954.8 ± 95.5	24 ± 2.4	98.4 ± 9.8	85 ± 8.5	1054 ± 105.4	0.05 ± 0.00	12.9 ± 1.3
	BC10	38 ± 4.0	471.2 ± 47.1	26 ± 2.6	106.6 ± 10.7	45 ± 4.5	558 ± 55.8	5.69 ± 0.57	1475.9 ± 147.6
	C1	650 ± 65	8060 ± 806	182 ± 18.2	746.2 ± 74.6	474 ± 47.4	5877.6 ± 587.8	<DL	<DL
	D1	46 ± 5.0	570.4 ± 57.0	40 ± 4.0	164 ± 16.4	48 ± 4.8	595.2 ± 59.5	3 ± 0.3	778.2 ± 77.8
	E5	154 ± 15.4	1909.6 ± 190.9	218 ± 21.8	893.8 ± 89.4	70 ± 7.0	868 ± 86.8	1.85 ± 0.19	479.9 ± 47.9

Figure 5 Terrestrial radionuclides material concentration expressed in parts *per* million (ppm)



Also, Figures 6 and 7 show how external absorbed dose rate between samples extracted from vertical (102.1 to 3967.8 nGy h^{-1}) and lateral (133.8 to 2856.2 nGy h^{-1}) orientation vary in magnitude. Observation shows that most of the vertically extracted samples show high activity concentration and so do the corresponding radiation hazard indices. Similarly, Figures 8 and 9 show how annual effective dose rate varies between samples extracted from vertical (0.1 to 4.9 mSv y^{-1}) and lateral (0.2 to 3.5 mSv y^{-1}) orientation.

Figure 6 Outdoor external absorbed dose rate for samples extracted in vertical orientation (see online version for colours)

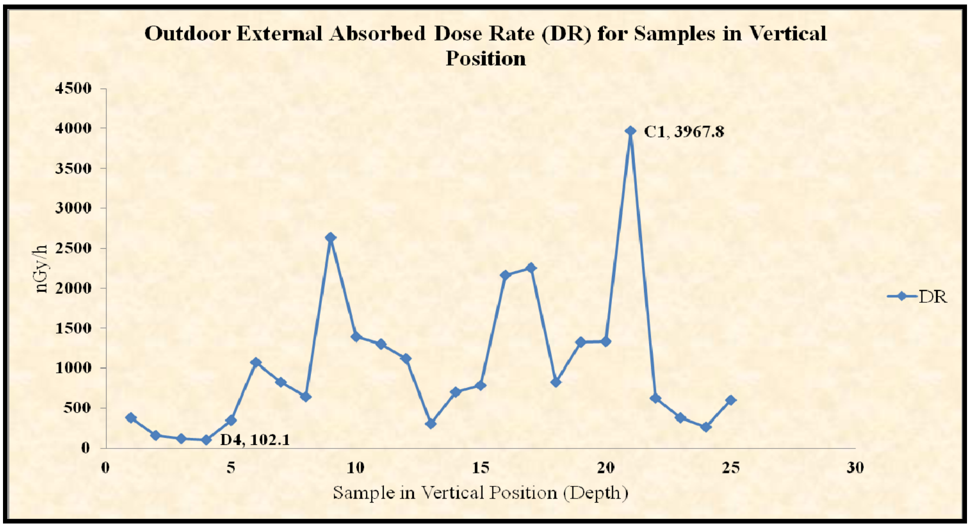


Figure 7 Outdoor external absorbed dose rate for samples extracted in lateral orientation (see online version for colours)

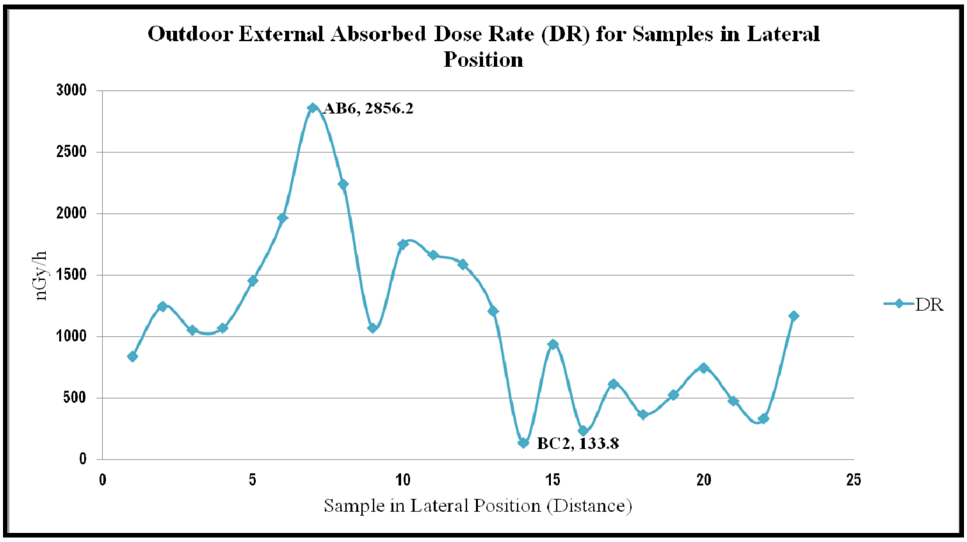
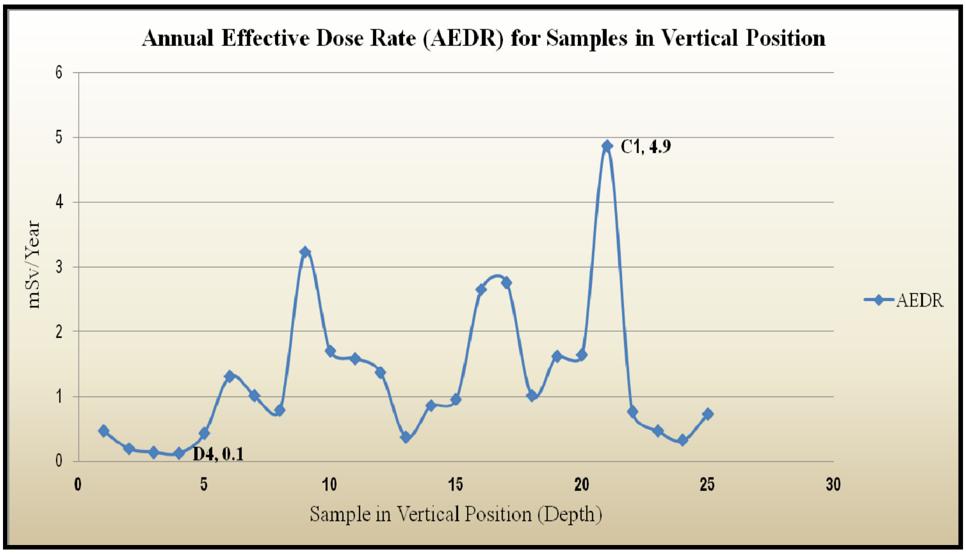


Figure 8 Annual effective dose rate for samples extracted in vertical orientation (see online version for colours)



Further analysis of the trends of uranium activity concentration still shows that vertically extracted samples lead followed by laterally extracted ones. Figure 10 suggests, through radium equivalent activity, that its value in terms of vertical orientation varies from 217.3 to 9127.1 Bq kg^{-1}, while laterally radium activity concentration varies from 307.9 to 6609.1 Bq kg^{-1}. Also, such variation in concentration may be associated with washing away of uranium concentration from the ground surface when it rains and such concentration being infiltrated into the deep ground through weak points such as cracks or fracture on the ground. Similar effect occurs due to running underground water which sweeps away concentration and deposits it in lateral orientation. It is worth noting that uranium was originally deposited on the ground surfaces by the action of volcanic eruption which took place several years ago (Nash, 2010).

Figure 9 Annual effective dose rate for samples extracted in lateral orientation (see online version for colours)

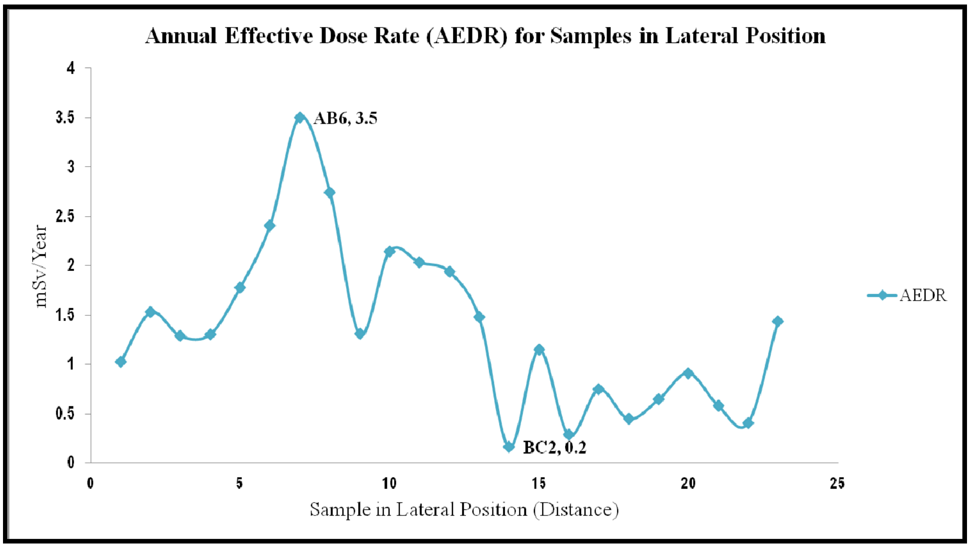
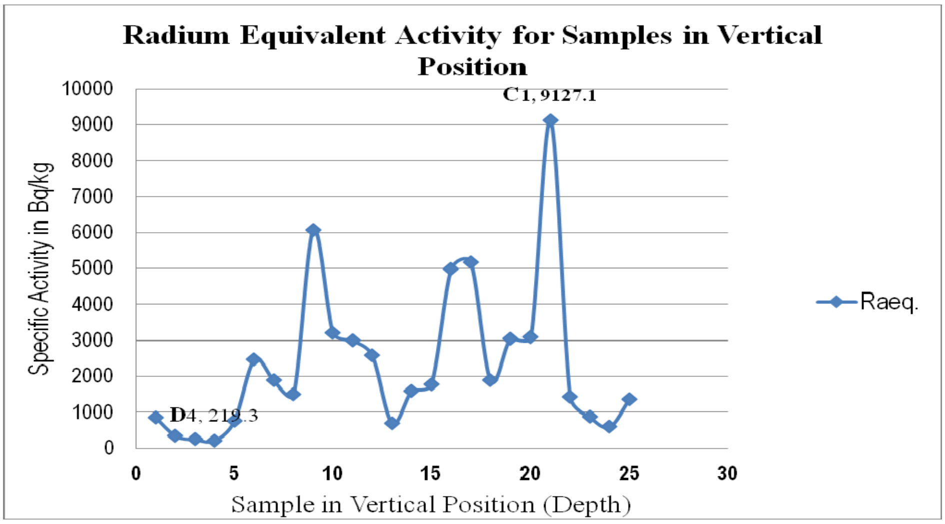


Figure 10 Radium activity concentration for samples extracted in lateral orientation (see online version for colours)



4 Conclusion

A thorough investigation of the uranium concentration in phosphate mines, a case study of Minjingu mine, has been presented. The authors are convinced that much of uranium concentration in the phosphate mines could be expected from vertical location across the phosphate bedrocks. Besides, radioactivity working levels and the related radiation hazards indices have been established, updated and discussed in details. As uranium concentration seems to be more than three times (650±65 ppm) higher than the minimum economically viable activity concentration (200 ppm), the direct application of raw phosphate rock as fertiliser is highly discouraged as it is likely to exhaust agricultural land instead of increasing the expected soil fertility.

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