

# Radiological Assessment of Weenen farmland Samples, KwaZulu-Natal, South Africa

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**Abstract.** The radiological hazards were evaluated in the farmland of Weenen, in the province of KwaZulu-Natal, South Africa. In this study, gamma spectroscopy was used to measure the activity concentrations of the radionuclides in the field and control soil samples. The overall activity concentrations for <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K were found to be 52±11, 44±4 and 511±56 Bqkg<sup>-1</sup> respectively. The radiological hazard indices calculated from these activity concentrations were lower than the recommended safe limits except for absorbed dose rate which was higher than recommended. In particular, the mean values for the radium equivalent, absorbed dose rate, annual effective dose equivalent (AEDE) and external hazard were 123±24 Bqkg<sup>-1</sup>, 77±17 nGyhr<sup>-1</sup>, 0.101±0.02 mSvy<sup>-1</sup> and 0.33±0.033 respectively. The indices were lower than unity indicating the safety of the farm workers in the area.

## 1. Introduction

Radioactivity is a natural phenomenon that has existed since the formation of earth [1, 2]. The human population is constantly exposed to cosmogenic, primordial and anthropogenic radiations [2, 3, 4]. The natural radionuclides of concern in the environment are mainly <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K [5]. These radionuclides can be classified into primordial, cosmogenic and anthropogenic radionuclides [1]. The cosmogenic radionuclides are formed continuously by the interaction of cosmic rays with matter in space [1, 4] and finally, the anthropogenic radionuclides are a wide variety of radionuclides coming from the activities of man [1, 2, 4]. The primordial radionuclides have the half-lives that stretch to the age of the earth [1, 2, 4]. The <sup>238</sup>U and <sup>232</sup>Th and their decay series are of concern because they produce a lot of ionizing radiation [6].

The agricultural activities also contribute significantly to environmental radioactivity through phosphate fertilizers produced from the phosphate rock that is highly enriched in <sup>238</sup>U and <sup>232</sup>Th series [7, 8, 9, 10]. These radioisotopes made their way into the rock from dissolved uranyl complex in seawater during the geological formation of the phosphate rocks [9]. Consequently, the workers are subjected to an additional source of external radiation exposure [6] in agricultural fields. Fertilizers provide radiation outside the mines where radon gas inhalation is an issue [11], medical applications [12], and fallout from nuclear weapons tests and power plants failures where radiation scatters all over, such as the accident of Chernobyl nuclear power plant on 26<sup>th</sup> of April 1986 in Ukraine, Fukushima Daiichi nuclear power plant on 11<sup>th</sup> of March 2011 in Japan [13] and Three Mile Islands nuclear power plant on 29<sup>th</sup> of March 1979 in USA [14]. The <sup>222</sup>Rn gas contributes hugely in the total background radiation followed by cosmic and terrestrial sources

[15]. The terrestrial radiations come from rocks and soils containing heavy metals of varying concentrations [9, 16]. The radiations are not uniform, but depend on the geographical and geological formation of the underlying rocks [4, 6, 5]. The farm workers are particularly exposed to radiological hazards because they spend lengthy hours working on land.

The anthropogenic activity in agriculture increases environmental radioactivity over time as compared to barren soils where no plantation takes place except natural growths [9]. There is no scientific assessment on the radioactivity profile found for this area. This motivated the present study to be conducted. The objective of the present study is to evaluate the radiological hazards linked with working agricultural land by estimating the absorbed dose rate, the annual effective dose rate, the external hazard index, and radium equivalent [9]. The results will serve as a benchmark and baseline for future assessment studies on monitoring radioactivity in farmland of Weenen, KwaZulu-Natal, South Africa.

## 2. Materials and Method

### 2.1. The soil sample collection and preparation

Two soil samples were collected on 3rd January 2017, Weenen KwaZulu-Natal. One was collected from a spot that was never fertilized (C17) while the other one was regularly nourished with fertilizers (A17). The samples range from the field samples (A17, A1 A2 and A3) to control samples (C17 and A4) sampled at about 200 - 250 meters away from the field (field A1). The entire field was sampled at a depth of 15 – 20 cm and the soil samples were loaded in 2L containers. In this way, a representative soil sample for each field was obtained. Figure 1 and 2 below are Google Earth views of the sampling spots carried out on the 3<sup>rd</sup> of January 2017 and 9<sup>th</sup> of August 2018.

This study hypothesizes that the field samples have high radioactivity levels [7, 8] as compared to the control samples. The samples were dried by laying them on a metal sheet for two days. After drying, the samples were kept in 2 Litres containers. On the 9<sup>th</sup> of August 2018, a collection of four more representative samples took place on the spots shown on Figure 2 as A1, A2, A3 and A4. On 12<sup>th</sup> June 2018, the crushing and pulverization of samples into powder took place at a milling facility of University of Johannesburg at the Doornfontein campus. They were prepared for analysis at iThemba Labs, Gauteng on Figure 3 below.



Fig 1: Google Earth views of A17 and C17



Fig 2: The fields that were sampled



Fig 3: LN tank, MCA and HPGe.

The homogenized samples were packed in zip-lock plastic bags and taken to iThemba LABS to be analysed. At iThemba LABS, the empty Marinelli beakers were weighed. The difference in mass of the loaded beakers gives the mass of the soil samples contained. The samples were then kept in tightly sealed Marinelli beakers. The samples were kept for 30 days to allow the  $^{222}\text{Rn}$  and its daughters to reach secular radioactive equilibrium at ambient temperature prior to  $\gamma$  - spectroscopy analysis. The background radiation of the  $\gamma$  - spectroscopy was determined

using an empty Marinelli beaker under identical measured conditions. It was negligible as the detector is covered with thick lead castle.

### 2.2. The analysis of soil samples

The concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , for the representative soil samples were measured using High Purity Germanium (HPGe)  $\gamma$  - ray spectroscopy having 10 cm thick lead shielding on all sides with inner Cu and Sn lining, to reduce the background activity to about 95%. The energy resolution (FWHM) of the detector was 1.9 keV at the 1332 keV  $\gamma$  - ray line of  $^{60}\text{Co}$  source. The detector was connected to a data acquisition system applying Genie-2000 analysis software, version 3.3 with both  $\gamma$  - ray energy and radionuclides identification. The prominent  $\gamma$  - ray lines were identified to find  $^{238}\text{U}$  and  $^{232}\text{Th}$  content. Since the analysis was done after the samples had established secular equilibrium, these  $\gamma$  - ray lines 186.20 keV ( $^{226}\text{Ra}$ ), 351.9 keV ( $^{214}\text{Pb}$ ), and 1765 keV ( $^{214}\text{Bi}$ ) were used to estimate  $^{226}\text{Ra}$  concentration. For  $^{232}\text{Th}$ , the  $\gamma$  - lines 583.1 keV ( $^{208}\text{Tl}$ ) and 911.20 keV ( $^{228}\text{Ac}$ ) were used. The singly occurring 1460.80 keV gamma line was used to directly determine the concentration of  $^{40}\text{K}$ . Since the  $^{232}\text{Th}$  content are determined from their progenies, their contents are referred to as thorium equivalent (eTh).

## 3. Numerical Calculations

### 3.1. The absolute efficiency of the detector

The ability of detecting gamma rays from a source depends on the resolution and the efficiency of the detector. The emission probabilities of various radionuclides were obtained from different sources in the literature [17]. The absolute efficiency of the HPGe detector was calculated with the known multisource calibration standards. The efficiency calibration curve of the detector was obtained from the standard sources as well and an empirical formula generated was used to find efficiencies of different signals in the  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ :

$$\varepsilon_{abs} = 4.148E^{-1.009} \quad (1)$$

where  $(E/E_o)$ ,  $E$  (keV) represents the peak energy of a particular radioisotope of interest and  $E_o = 1$  keV. Eqn (1) is in agreement with [18] and produced a curve similar to those on work by [19, 20] which were simulated curves of standard liquid sources from NMISA and CSIR. The linearity and energy resolution of the detector were tested by using signals from these standard sources. For any  $\gamma$  - ray detector, the most important properties are the energy resolution and the detection efficiency of that detector.

### 3.2. The activity concentration in soil samples

The background count was determined by counting an empty Marinelli beaker of the same dimension as those containing the samples and subtracting from the gross count. Each sample was counted for 28800 seconds to reduce the statistical uncertainty. The count rates were then used to compute the activity concentrations for the natural radionuclides in the measured samples [3, 21, 22, 23]:

$$A = \frac{C_{net}}{I_{\gamma} \times \varepsilon \times m} \quad (2)$$

where  $A$  represents the activity concentration of an isotope of concern in a particular sample in  $\text{Bqkg}^{-1}$ ,  $C_{net}$  is the net peak count rate of the sample corrected for background, then  $\varepsilon$  is the absolute detector efficiency of the specific  $\gamma$  - ray,  $I_{\gamma}$  is the emission probability of a specific energy photo peak.

### 3.3. Evaluation of radiation hazards

The radium equivalent ( $Ra_{Eq}$ ) quantity was developed to express the gamma yield from the mixture of the radionuclides in a soil sample and it also represents the activity levels due to  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  [2, 3, 23, 24]:

$$Ra_{Eq} = C_{Ra} + 1.43 \times C_{Th} + 0.077 \times C_K \quad (3)$$

where  $Ra_{Eq}$  is the radium equivalent,  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the sample. The concentration of  $^{238}\text{U}$  is sometimes replaced by that of  $^{226}\text{Ra}$  if the secular equilibrium is established. Eqn (03) assumes that 370 Bq.kg<sup>-1</sup> of  $^{226}\text{Ra}$ , 259 Bq.kg<sup>-1</sup> of  $^{232}\text{Th}$  and 4810 Bq.kg<sup>-1</sup> of  $^{40}\text{K}$  produce the same gamma dose rate and its maximum value should not exceed 370 Bq.kg<sup>-1</sup>. The coefficient of  $C_{Th}$  and  $C_K$  are found by a ratio of expected radium concentration to those of  $C_{Th}$  and  $C_K$ . From the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ , the total absorbed dose rate due to natural occurring radioactive materials (NORM) in air 1 metre above the ground is calculated using the following formula [2, 25, 26]:

$$D_R = 0.429 \times C_{Ra} + 0.604 \times C_{Th} + 0.042 \times C_K \quad (4)$$

where D is the absorbed dose rate, 0.429 for  $^{226}\text{Ra}$  series, 0.604 for  $^{232}\text{Th}$  series and 0.042 for  $^{40}\text{K}$ , are dose conversion factors in units determined from the ratio of absorbed exposure in air to the activity concentration in the soil [27]. The estimate the annual effective dose was estimated using the following equation. So using the calculated dose rate in Eqn (04), the estimates of effective dose rates per annum can be made from this relation [23]:

$$D_{Eff} = D_R \times T \times F \quad (5)$$

where  $D_R$  is the absorbed dose rate in (nGyh<sup>-1</sup>), T is (365×24×0,2) and F is conversion coefficient equivalent to (0,7×10<sup>3</sup> mSv/10<sup>9</sup>nGy). The UNSCEAR reports used 0,7 Svy<sup>-1</sup> for the conversion coefficient from an absorbed dose in air to effective dose received by adults, and 0.2 for the outdoor occupancy factor [27, 23, 15] and the annual effective dose rate should be less than 1 mSvy<sup>-1</sup> [28, 29]. The external radiation exposure is usually associated with gamma radiation emitted by radionuclides of concern. The external hazard index ( $H_{Ex}$ ) is obtained from  $Ra_{Eq}$  expression [2, 4, 30]:

$$H_{Eff} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (6)$$

where  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are as defined above. For safer limits, this index should be small than a unity.

## 4. Results and discussion

The isotopic ratio of daughter radionuclides  $^{214}\text{Pb}/^{214}\text{Bi}$  was used to define the level of radioactive secular equilibrium achieved in the samples analysed. These values are 1.51±0.40, 1.11±0.36, 1.16±0.38, 1.37±0.46, 1.04±0.25 and 1.08±0.36 for the ratio of  $^{214}\text{Pb}$  to  $^{214}\text{Bi}$ . Whereas the ratio of  $^{228}\text{Ac}$  to  $^{208}\text{Tl}$  0.899±0.082, 0.823±0.074, 1.10±0.096, 0.978±0.085, 1.06±0.093 and 0.876±0.088. When the secular equilibrium is established, the decay and production rate of a radionuclides is the same that is,  $A_p = A_d$ .

The distribution of activity concentrations of the daughter isotopes of the primordial radionuclides  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and a singly occurring  $^{40}\text{K}$  found in the samples is presented in Table 1 below. The  $^{226}\text{Ra}$  concentration was determined through its progeny, that is,  $^{214}\text{Pb}$  (351.9 KeV) and  $^{214}\text{Bi}$  (1764.5 KeV) decay products in the sample. The concentration of  $^{232}\text{Th}$  was determined from the average concentrations of  $^{212}\text{Pb}$  (583.19 KeV) and  $^{228}\text{Ac}$  (911.20 KeV) in the samples. The singly occurring 1460.6 KeV gamma ray signal was used to determine the

concentration of  $^{40}\text{K}$  in samples. In quantifying  $^{226}\text{Ra}$  it was difficult to use 186.2 keV due to the interfering 185.7 keV from  $^{235}\text{U}$  [31]. The Activity concentration, Radium equivalent ( $Ra_{Eq}$ ), Dose rate ( $D_R$ ), Annual effective dose equivalent ( $D_{Eff}$ ) and External hazard ( $H_{Ex}$ ) in soil samples.

**Table 1:** Activity concentration, Radium equivalent, Dose rate, Annual effective dose rate and External hazard index in soil samples.

Activity Concentrations in soil samples and Radiation Hazard Indices									
Sample	Sample ID	Parameter	$^{226}\text{Ra}$	$^{232}\text{Th}$	$^{40}\text{K}$	Radium Equiv (Bq/kg)	Dose Rate (nGy/h)	AEDE (mSv/y)	External Hazard
			(Bq/kg)	(Bq/kg)	(Bq/kg)				
Control	C17	Ave $\pm\sigma$	68 $\pm$ 5	41 $\pm$ 4	430 $\pm$ 20	160 $\pm$ 8	69 $\pm$ 6	0.084 $\pm$ 0.01	0.30 $\pm$ 0.07
	A4	Ave $\pm\sigma$	29 $\pm$ 4	37 $\pm$ 4	424 $\pm$ 22	115 $\pm$ 10	66 $\pm$ 7	0.084 $\pm$ 0.01	0.28 $\pm$ 0.9
Field	A17	Ave $\pm\sigma$	49 $\pm$ 4	45 $\pm$ 4	578 $\pm$ 24	158 $\pm$ 10	83 $\pm$ 7	0.102 $\pm$ 0.01	0.35 $\pm$ 0.1
	A1	Ave $\pm\sigma$	58 $\pm$ 4	40 $\pm$ 4	526 $\pm$ 23	157 $\pm$ 10	76 $\pm$ 7	0.093 $\pm$ 0.01	0.32 $\pm$ 0.1
	A2	Ave $\pm\sigma$	48 $\pm$ 5	49 $\pm$ 4	556 $\pm$ 25	154 $\pm$ 11	85 $\pm$ 8	0.105 $\pm$ 0.01	0.37 $\pm$ 0.1
	A3	Ave $\pm\sigma$	60 $\pm$ 4	43 $\pm$ 4	550 $\pm$ 24	165 $\pm$ 11	84 $\pm$ 7	0.103 $\pm$ 0.01	0.36 $\pm$ 0.1
Overall	Min		29 $\pm$ 4	35 $\pm$ 3	424 $\pm$ 22	115 $\pm$ 10	66 $\pm$ 7	0.081 $\pm$ 0.01	0.29 $\pm$ 0.9
	Max		68 $\pm$ 5	49 $\pm$ 4	578 $\pm$ 24	160 $\pm$ 8	85 $\pm$ 8	0.105 $\pm$ 0.01	0.37 $\pm$ 0.1
	Ave $\pm\sigma$		52 $\pm$ 11	43 $\pm$ 10	511 $\pm$ 56	123 $\pm$ 24	77 $\pm$ 17	0.095 $\pm$ 0.02	0.33 $\pm$ 0.9

The activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in soil samples were measured. The  $^{40}\text{K}$  was higher in field soil samples (A17, A1, A2, and A3) and was a bit lower for the control samples C17 and A4. The calculated radium equivalent for all the activity was lower than the reference value of  $\text{Bqkg}^{-1}$  [24, 27] ranging from  $115\pm 10 \text{ Bqkg}^{-1}$  to  $160\pm 8 \text{ Bqkg}^{-1}$  in control soil samples to  $154\pm 11 \text{ Bqkg}^{-1}$  to  $165\pm 11 \text{ Bqkg}^{-1}$  in the field soil samples. The mean equivalent activity concentrations for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and single occurring  $^{40}\text{K}$  from the field samples were found to be  $23\pm 4$ ,  $44\pm 4$  and  $552\pm 21 \text{ Bqkg}^{-1}$ , respectively. The mean equivalent activity concentrations for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and single occurring  $^{40}\text{K}$  from the control samples were found to be  $20\pm 1$ ,  $39\pm 3$  and  $427\pm 30 \text{ Bqkg}^{-1}$ , respectively.

For the field samples, the mean radium equivalent was  $158\pm 21 \text{ Bqkg}^{-1}$  and whereas for the control samples, the radium equivalent was  $138\pm 12 \text{ Bqkg}^{-1}$ . The external radiation hazard indices were found to be less than unity, which is within a permissible limit. The overall mean dose rate of the samples is  $77\pm 17 \text{ nGyh}^{-1}$ , which is higher than a recommended safe value of  $55 - 60 \text{ nGyh}^{-1}$  [5, 27]. The mean annual effective dose rate for the representative field samples was  $0.101\pm 0.02 \text{ mSvy}^{-1}$ , a value lesser than  $1 \text{ mSvy}^{-1}$  recommended by ICRP and UNSCEAR [5] as the limit for the public radiation exposure control. The external hazard index fell below a unity with an overall average of  $0.33\pm 0.03$ .

## 5. Conclusion

The soil samples from different parts of a farmland were analysed for natural radionuclides emanating from the soil and continuous application of fertilizers. Only naturally occurring radionuclides were detected in the samples. On average, the overall  $^{226}\text{Ra}$  and the equivalent  $^{232}\text{Th}$  were  $52\pm 11 \text{ Bqkg}^{-1}$  and  $43\pm 10 \text{ Bqkg}^{-1}$  and the activity of the singly occurring  $^{40}\text{K}$  peak was  $511\pm 56 \text{ Bqkg}^{-1}$  all as presented in Table 1 above. The absorbed dose rate is higher than the value of  $55 \text{ nGyh}^{-1}$  as it was found to be  $77\pm 17 \text{ nGyh}^{-1}$ . The average annual effective dose rate is  $0.101\pm 0.02 \text{ mSvy}^{-1}$  below the value recommended by ICRP and UNSCEAR for radiation exposure control.

The samples C17 and A4 show a slightly lower potassium concentration in agreement with each other. A conclusion can be drawn that the higher potassium concentration in the field samples is due to the application of fertilisers. Thus, the application of fertilizers in the assessed fields, poses no significant radiological hazard to the farm workers. According to the data on UNSCEAR 2000, the concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  from this study are within world average. This is supported by both external hazard index and the average annual effective dose falling within a unity of their measurements.

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