Radiological Assessment of Weenen Agricultural Fields Samples

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Abstract. The radiological hazards were evaluated in the agricultural fields of Weenen, province of KwaZulu-Natal, South Africa. In this study, gamma spectroscopy was used to measure the activity concentrations of the radionuclides in field representative soil samples and two representative soil samples from the control area. The mean equivalent activity concentrations for field samples of ^{238}U, ^{232}Th, and ^{40}K were found to be 23.2±3.9, 44.5±3.5 and 552.5±21.4 Bq.kg-1, respectively. On the other hand, the mean activity concentrations for ^{238}U, ^{232}Th, and ^{40}K from the control area were found to be 19.5±0.9 Bq.kg-1, 39.4±2.9 Bq.kg-1 and 427±30 Bq.kg-1, respectively. Radiological hazard indices calculated from these activity concentrations were lower than recommended safe limits. In particular, calculated mean values for the radium equivalent (Raeq), absorbed dose (D_R), external hazard (H_{Ex}), and Annual effective dose equivalent (D_{Eff}), respectively. All these values were lower than unity, posing a lower health risk to 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 ^{238}U, ^{232}Th, and ^{40}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 decay of ^{238}U and ^{232}Th and their series is 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 ^{238}U and ^{232}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 exposed 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], fallout from nuclear weapon tests and power plants failures where radiation scatters all over, such as Chernobyl in 1986, and Fukushima Daiichi nuclear power

station on 11 March 2011 [13] and Three Mile Islands [14]. The ^{222}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 soils were loaded in 2L containers. In this way, a representative soil sample for each field was obtained. Figure 1 and 3 are Google Earth views of the sampling spots on the 3^{rd} of January 2017 and 9^{th} 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^{th} of August 2018, a collection of four more representative samples took place on the spots shown on Figure 3 as A1, A2, A3 and A4. On 12^{th} 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. The homogenized samples

Figure 1: Left: Google Earth views of A17 and C17, middle: The fields that were sampled, and right: LN tank, MCA and HPGe.

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}Rn and its daughters to reach secular radioactive equilibrium at ambient temperature prior to γ-spectroscopy analysis. The background radiation of the γ-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 ^{238}U, ^{232}Th and ^{40}K, for the representative soil samples were measured using High Purity Germanium (HPGe) γ-ray spectrometer 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 γ-ray line of ^{60}Co source. The detector was connected to a data acquisition system applying Genie-2000 analysis software, version 3.3 with both γ-ray energy and radionuclide identification. The prominent -ray lines were identified to find of ^{238}U and ^{232}Th content. Since the analysis was done at after the samples had established secular equilibrium, these γ-ray lines 186.20 keV (^{226}Ra), 351.9 keV (^{214}Pb), and 1765 keV (^{214}Bi) were used to estimate ^{238}U concentration. For ^{232}Th, the γ-lines 583.1 keV (^{208}Tl) and 911.20 keV (^{228}Ac) were used. The singly occurring 1460.80 keV gamma line was used to directly determine the concentration of ^{40}K. Since the ^{238}U and ^{232}Th content are determined from their progenies, their contents are referred to as uranium equivalent (eU) and thorium equivalent (eTh) respectively.

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 ^{238}U, ^{232}Th and ^{40}K and others.

$$\varepsilon_{abs} = 4.148E^{-1.009} \tag{1}$$

where E represents the peak energy of a particular isotope of interest. 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 γ-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 activity concentrations for the natural radionuclides in the measured samples were computed using the following relation [3, 21, 22, 23].

$$A = \frac{C_{net}}{I_\gamma \varepsilon m} \tag{2}$$

where A represents the activity concentration of an isotope of concern in a particular sample in Bq.kg^{-1}, C_{net} is the net peak count rate of the sample corrected for background, then ε is the absolute detector efficiency of the specific γ-ray, I_γ 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 ^{238}U, ^{232}Th and ^{40}K [2, 3, 23, 24]:

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

where Ra_{Eq} is the radium equivalent, C_U, C_{Th} and C_K are the concentrations of ^{238}U, ^{232}Th and ^{40}K in the sample. The concentration of ^{238}U is sometimes replaced by that of ^{226}Ra assuming that there exists equilibrium. Eqn (03) assumes that 370 Bqkg^{-1} of ^{238}U, 259 Bqkg^{-1} of ^{232}Th and 4810 Bqkg^{-1} of ^{40}K produce the same gamma dose rate and its maximum value should not exceed 370 Bqkg^{-1}. 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 ^{238}U, ^{232}Th, and ^{40}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]:

$$D_R = 0.429C_U + 0.666C_{Th} + 0.042C_K \quad (4)$$

where D is the absorbed dose rate, 0.429 for ^{238}U series, 0.666 for ^{232}Th series and 0.042 for ^{40}K, are dose conversion factors in units determined from the ratio of absorbed exposure in air to the activity concentration in the soil [27][7]. 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]:

$$D_{Eff}(mSvy^{-1}) = D_R TF \quad (5)$$

where D is the absorbed (nGyh^{-1}), T is (36524h0,2) and F is conversion coefficient equivalent to (0,7103mSv/109nGy). The UNSCEAR reports used 0,7 Svy^{-1} 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 effective dose rate per annum should be less than 1 mSvy^{-1} [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]:

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

where C_U, 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}Pb/^{214}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}Pb to ^{214}Bi. And the ratio of ^{228}Ac to ^{208}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 radionuclide is the same that is, $A_p = A_d$

The distribution of activity concentrations of the daughter isotopes of the primordial radionuclide ^{238}U, ^{232}Th and a singly occurring ^{40}K in the samples is presented in Table 1. The ^{238}U concentration was determined through the ^{226}Ra decay products average concentrations, that is, ^{214}Pb (351.9 KeV) and ^{214}Bi (1764.5 KeV) decay products in the sample. The concentration of ^{232}Th was determined from the average concentrations of ^{212}Pb (583.19 KeV) and ^{228}Ac (911.20 KeV) in the samples. The singly occurring 1460.6 KeV gamma ray signal was used to determine the concentration of ^{40}K in samples. In quantifying ^{226}Ra it was difficult to use 186.2 keV due to the interfering 185.7 keV from ^{235}U [31]. The Activity concentration (Bq kg^{-1}), Radium equivalent (Ra_{Eq}), Dose rate (D_R), Annual effective dose equivalent (D_{Eff}) and External hazard (H_{Ex}) in soil samples. The activity concentrations of ^{238}U, ^{232}Th and ^{40}K in soil

Sample ID			Activity Concentration (Bq/kg)				Radiation Hazard Indices			
Sample		Parameters	^{238}U	^{232}Th	^{40}K	^{226}Ra	Ra_{Eq} ($Bqkg^{-1}$)	D_R ($nGyh^{-1}$)	D_{Eff} ($mSvy^{-1}$)	H_{Ex}
Control	C17	Min	15±4	39±3						
		Max	68±5	44±2						
		Ave ± σ	**18.8±4**	**41.4±3.8**	**430±20**	**67,5±4,8**	**160±8,2**	**69±5,8**	**0,084±0,01**	**0,300±0,07**
	A4	Min	19±6	35±3						
		Max	29±4	39±2						
		Ave ± σ	**20.1±6.4**	**37.3±3.7**	**424±22**	**29.0±4,1**	**115±10**	**66±6,8**	**0,081±0,01**	**0,288±0,97**
Field	A17	Min	20±6	41±3						
		Max	49±4	49±2						
		Ave ± σ	**20.8±6.4**	**45.0±3.8**	**578±24**	**49,1±4**	**158±9,9**	**83±7,2**	**0,102±0,01**	**0,352±0,11**
	A1	Min	19±6	40±2						
		Max	58±4	41±3						
		Ave ± σ	**20.2±6.1**	**40.4±3.7**	**526±23**	**58,4±4,3**	**157±9,8**	**76±6,9**	**0,093±0,01**	**0,322±0,10**
	A2	Min	19±6	48±4						
		Max	42±5	49±2						
		Ave ± σ	**23.0±6.6**	**48.8±4.1**	**556±25**	**41,7±4,9**	**154±11**	**85±7,5**	**0,105±0,01**	**0,369±0,11**
	A3	Min	28±7	42±2						
		Max	60±4	45±3						
		Ave ± σ	**28.8±6.8**	**43.4±3.9**	**550±24**	**60,3±4,4**	**165±11**	**84±7,4**	**0,103±0,01**	**0,362±0,09**
Overall	M ± σ	Min	15±4	35±3	424±22	29.0±4,1	115±10	66±6,8	0,081±0,01	0,288±0,97
		Max	68±5	49±4	578±24	67,5±4,8	160±8,2	85±7,5	0,105±0,01	0,369±0,11
		Ave ± σ	**22±4**	**43±4**	**511±67**	**51,0±14**	**123±18**	**77±8**	**0,095±0,01**	**0,33±0.033**

1.png

Figure 2: Activity concentration (Bq kg-1), Radium equivalent (RaEq), Dose rate (DR), Annual effective dose equivalent (DEff) and External hazard (HEx) in soil samples

samples were measured. ^{40}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 $Bqkg^{-1}$ [24, 27] ranging from 115±10 $Bqkg^{-1}$ to 160±8.2 $Bqkg^{-1}$ in control soil samples to 154±11 $Bqkg^{-1}$ - 165±11 $Bqkg^{-1}$ in the field soil samples. The mean equivalent activity concentrations for ^{238}U, ^{232}Th, and single occurring ^{40}K from the field samples were found to be 23.2±3.9, 44.5±3.5 and 552.5±21.4 $Bqkg^{-1}$, respectively. The mean activity concentrations for ^{238}U, ^{232}Th, and single occurring ^{40}K from the control samples were found to be 19.5±0.9, 39.4±2.9 and 427±30 $Bqkg^{-1}$, respectively.

For the field samples, the mean radium equivalent was 158.5±14.5 $Bq.kg^{-1}$ and whereas for the control samples, the radium equivalent was 137.4±12.9 $Bqkg^{-1}$. 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±8 $nGyh^{-1}$, which is higher than a recommended value of 55-60 $nGyh^{-1}$ [5, 27]. The mean annual effective dose rate for the representative field samples was 0.101±0.049 $mSvy^{-1}, avaluelesserthan1mSvy^{-1}$ 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 0.3330.03.

5. Conclusion

The soil samples from different parts of a farm 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 equivalent ^{238}U and ^{232}Th were 23.2±3.9 $Bqkg^{-1}$ and 44.5±3.5 $Bqkg^{-1}$ and the activity of the singly occurring ^{40}K peak was 552.5±21.4 $Bqkg^{-1}$ all are presented in Table 1. The absorbed dose rate is higher than the value of 55 $nGyh^{-1}$ .as it was found to be 77±8 $nGyh^{-1}$. The average annual effective dose rate is 0.101±0.0049 $mSvy^{-1}$ that is a value lesser than 1 $mSvy^{-1}$ recommended by ICRP and UNSCEAR for public radiation exposure control.

Again, the control samples C17 and A4 display 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. The results, however, indicate that the radiological dose from the soil is within safe limits. Thus, the application of fertilizers in these agricultural fields that were sampled poses no significant radiological hazard to the farm workers. This claim is primarily due to that the external hazard index and the average annual effective

dose both fall within a unity of their measurements. According to the data on UNSCEAR 2000, the concentrations of ^{238}U, ^{232}Th and ^{40}K from this study are within world average.

6. Acknowledgements

The author would like to acknowledge with great appreciation the service rendered at iThemba LABS, Gauteng by Mr A. Kwelilanga for whose knowledge of gamma spectrometric analysis is outstanding. Thank you again for your eye-opening insights, sir. Dr LP Masiteng, my supervisor, is acknowledged for helping me out.

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