Distribution of Gamma-Emitting Radionuclides in Soils around the Centre for Energy Research and Training (CERT) Ahmadu Bello University, Zaria, Zaria-Nigeria

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Abstract: A portable HPGe spectrometer has been employed to characterised, *in-situ* gamma activity concentration from the primordial Radionuclides ^{238}U, ^{232}Th ^{40}K in the soil at 12 monitoring points (MPs) in the environment in and around the Centre for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, Nigeria. The MPs were marked-out using Global Positioning System (GPS) navigation. The measured activity concentrations due to ^{238}U range from 4.8 ± 3.0 to 11.9 ± 2.0 Bq kg^{-1} with an average of 8.3 ± 2.6 Bq kg^{-1}, ^{232}Th range from 15.5 ± 4.3 to 46.4 ± 3.5 Bq kg^{-1} with an average of 34.3 ± 3.4 Bq kg^{-1} and ^{40}K range from 317.2 ± 8.4 to 985.3 ± 7.0 Bq kg^{-1} with an average of 641.8 ± 7.3 Bq kg^{-1}.
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1. Introduction

Ionising radiation and radioactive substances are natural and permanent features of the environment, and thus the risks associated with radiation in all its forms can only be restricted, not eliminated. Moreover, the use of man-made radiation is widespread. The largest contribution to the radiation field is of natural origin: it is due to cosmic rays, the natural radioactivity generated by the decay of unstable, naturally occurring elements and the radioactive decay products of radon in the air (UNSCEAR, 1988). Artificial radioactivity may be released into the environment during the normal operations of nuclear facilities and installations such as nuclear ore processing, uranium enrichment, fuel fabrication, reactor operations, and application of radioisotopes in the fields of nuclear medicine, research, industry and agriculture (Eisenbud, 1987). These emissions are very small in normal operation, although large amounts of radioactivity could be released to the environment through accidents. The total amount of radioactivity in an environment should be accurately known and kept to a level as low as reasonably achievable (ALARA). Exposures from natural radiation are the largest component of all exposures for most people, and form the baseline

upon which exposures from manmade sources are added (UNSCEAR, 2000).

Gamma-ray spectroscopy system provides practical way to characterize dispersed Radionuclides in or on the soil to ascertain possible changes in the environmental radioactivity. Both laboratory and *in-situ* gamma spectroscopy are often used for monitoring and assessment of radioactivity and radiation dose rates in the environment due to both natural and anthropogenic sources. (Beck *et al.,* 1972; Nikl *et al*., 1988; ICRU, 1994; Othman *et al.*, 1994; Tzortzis *et al.*, 2003; Clouvas *et al*., 2004).In large-scale environmental radioactivity measurement, *in-situ* gamma spectroscopy is much favoured compared to laboratory soil analysis because of time and problems associated in cross contamination involved in the laboratory methods. It also gives the opportunity to obtain information not only of the activity concentration but also of the relative contribution of the various nuclides to activity concentration. *In-situ* techniques for measuring the activity concentration, resulting from the gamma radiation and characterizing its sources, with gamma ray spectrometer have been used successfully in the outdoor environment (Beck *et al*., 1972; Clouvas *et al*., 2001; Petalas *et al*., 2005; Auwal,M.M. 2005)

For radiation monitoring near nuclear facilities, baseline data, are indispensable for various purposes: they provide documented reference base; for the assessment of actual or potential consequence of radioactivity on health, and on the environment, due to radioactive materials or radiation fields in the environment from normal operations and accidental releases. The present work has been conducted in the Centre for Energy Research and Training (CERT), and some selected settlements within 2km radius from NIRR-1, using high-resolution portable gamma spectrometry system. The Centre is a nuclear energy (radiation) based research centre, thus, dealing with substantial quantities of artificial radioactive materials, such as neutron generator, Am-Be isotopic neutron source, a nuclear research reactor code named Nigeria Research Reactor-1 (NIRR-1); among others.

The present work puts forward a systematic method for determining environmental activity concentrations levels from *in-situ* measurements, and shows the distribution and intensities of important natural and artificial radionuclides within the studied area. It complements previous studies reported by other authors (Ibeanu *et al.,* 2002) by providing *in situ* gamma spectrometry based, baseline data, for operational and post operational monitoring, of the Centre for Energy Research and Training (CERT).

2. Materials and Methods

2.1 ***Site***

The study site is located at 007°38.523'–007°40.822'E and 11°07.830'–11°09.790' N within the Zaria sheet 102.The number of monitoring points (MPs) includes; eight locations within the Centre for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, and four other locations at selected settlements around CERT within 2Km radius. Table 1 gives the description of the monitoring points (MP) with respect to the Nigeria Research Reactor 1 (NIRR1) as reference point. The chosen sites were undisturbed with little or no surface features and modest vegetation. These sites were marked-out using Global Positioning System (GPS).

2.2 ***Gamma-ray Detection system***

The experimental set-up is a stand-alone high-resolution spectroscopic system used for the in-situ measurement of the energy spectrum of the emitted gamma rays in the energy range between 50 keV and 3000 keV. The system consist of a high-purity germanium (HPGe) detector EG & G ORTEC® coaxial cylinder of crystal length of 46.8 mm and diameter 55.0 mm, with an efficiency of 23.5% 27 relative to a 7.6 x 7.6 cm^2 NaI(Tl) crystal. In addition, energy resolution (FWHM) of 2.0 keV was achieved, all for a ^{60}Co emission point source at 25 cm for 1.33 MeV. This type of detector can sustain warm up when not in use, which is a convenient feature during extended field trips. The detector was mounted on a portable (hand-held) 10 litres liquid nitrogen Dewar that features an all attitude capability. Liquid nitrogen was used for cooling the detector during operation. After filling the portable Dewar with liquid nitrogen, it requires a 6 hour cool-down time before becoming operational with 24 hours nominal holding time. The detector assembly was mounted on a 1 m tripod with the crystal end cap facing down towards the ground and the Dewar above. This orientation maximizes the flux that will be intercepted and registered by the detector (Kelvin, 1997). The detector unit was connected to a battery powered EG & G ORTEC® "Normad Plus" portable computer based spectroscopy system. High voltage and preamplifier power were supplied to the detector by the system. An advanced multi-channel analyzer (MCA) emulsion software (MAEASTRO-32) was used for data acquisition, storage display and analysis, of the acquired gamma-spectra.

Figure 1: Typical *in situ* g-ray detection system at one of the monitoring points (MP)

2.3 In-situ measurement and analysis

The current study was conducted in the months of May and June 2005. The field measurement of terrestrial gamma radiation was based on the assumption that there exist laterally uniform distribution of natural Radionuclides in the environment and that the vertical contribution from the soil is limited to the first horizon (≈ 10cm to 30cm). Measurements were performed over flat terrain; that allow source geometry to be represented as an infinite half-space; that is 2π geometry in terms of solid angle subtended by the source.

The source measured was soil sample and counting statistics for a given spectral absorption peak were obtained in a fraction of the time required for counting a small collected sample. Measurements of spectra in the field were made for a period of 5000 s. However, series of random short readings of about 600 sec were first taken, to ensure that, there is approximate uniformity, consequently, the desired counting statistics. The measuring system was routinely checked with ^{137}Cs and ^{60}Co standard sources.

A computer based multi-channel analyzer system with emulsion software (MAESTRO–32) was used for spectra acquisition. Based on two-point energy calibration as set for the operation, prominent peaks were identified in a benchmark spectrum (fig.1d) and the appropriate regions of interest were set up. These peaks, which are characteristics of typical environmental spectra, are:

- the 295, 352, 609, 1120 and 1765 keV peaks in the ^{238}U series,
- the 238, 510, 583, 911, 965 and 2615 keV peaks in the ^{232}Th series,
- the 1460 keV peak of ^{40}K.

The set energy bands define the peaks where the left and right channel markers are representative of the Compton continuum. Detector's specific calibration factors (measured efficiency) determined in an earlier experiment (Auwal, 2005), were applied to convert from net peak count rate to activity concentration. Only peaks with reasonable gamma-ray emission probabilities were considered.

3. Results and discussion

The activity concentrations for ^{232}Th, ^{238}U and ^{40}K with standard error of the measured locations are presented in table 2, while table 3, show the statistical analysis of the results and percentage contribution is shown in figure 3. The activity concentrations were calculated by the net peak areas of 511 keV (^{208}Tl) and 911 keV (^{208}Ac) for ^{232}Th, 609 keV (^{214}Bi) and 352 keV (^{214}Pb) for ^{238}U, and 1460 keV for^{40}K. The concentrations range from 15.5 ± 4.3 to 46.4 ± 3.5 Bq kg^{-1}(34.3 ± 3.4) for ^{232}Th, 4.8 ± 3 to 11.9± 2 Bq kg^{-1} (8.3 ± 2.6) for ^{238}U and 317.2 ± 8.4 to 985.3 ± 7 Bq kg^{-1} (684 ± 7.3) for ^{40}K with mean values enclosed in the brackets. The relative contribution of the different natural gamma emitters (Thorium series, Uranium series, and ^{40}K) to the total activity concentration vary among the monitoring point. This range from

3.3% to 8.3% for ^{232}Th, 0.7% to 1.7% for ^{238}U and 90.2% to 95.5% for ^{40}K .Of all the locations MP004 and MP011 appear to have the highest concentration of ^{232}Th, also MP004 exhibits the highest concentration of ^{238}U. MP012 appear to have much higher concentration of ^{40}K when compared with the concentrations at other locations, with value of 985.3 ± 7 Bq kg^{-1}. In addition, the ^{232}Th/^{238}U ratios for the locations range from 2.8 to 6.4. This shows uranium is less than thorium by a factor of three.

Table 1Site identification by Global Positioning System (GPS), and description of Monitoring Points

Site ID	Location		Description
	Latitude	Longitude	
MP001	11°08.439'N	07°39.840'E	CRMP07 about 5m from NIRR-1
MP002	11°08.426'N	07°39.844'E	CRMP08 about 105m from NIRR-1
MP003	11°08.476'N	07°39.901'E	CRMP05 about 141m from NIRR-1
MP004	11°08.497'N	07°39.851'E	CRMP06 about 100m from NIRR-1
MP011	11°08.415'N	07°39.776'E	CRMP03 about 140m from NIRR-1
MP012	11°08.358'N	07°39.912'E	CRMP04 about 220m from NIRR-1
MP013	11°08.505'N	07°39.963'E	CRMP01 about 280m from NIRR-1
MP014	11°08.549'N	07°39.823'E	CRMP02 about 220m from NIRR-1
MP111	11°07.830'N	07°39.447'E	Beside ABU Dam
MP112	11°08.205'N	07°40.186'E	LEA Unguwam Jema'a a nearby settlement
MP113	11°08.749'N	07°40.822'E	Aviation site ii
MP114	11°09.790'N	07°38.524'E	LEA Samaru a nearby settlement

Comparing the activity concentrations of the present work with the Ibeanu *et al*., (2002), in which a laboratory based NaI(Tl) detector was employed to measure the radionuclide concentrations in dry soil samples from the same measurement points, the *in situ* measurement results for ^{232}Th and ^{238}U were generally significantly lower than the laboratory data, except for some of the ^{238}U that were reported as below detection limit. On the other hand, ^{40}K has exhibited certain degree agreement between the two results, with a variation of approximately ±5% at some points.

Table 2Activity concentration of ^{232}Th, ^{238}U and ^{40}K

Concentration ±Stat. Error (Bq kg^{-1})								
Site ID	**^{232}Th**	**Contr. (%)**	**^{238}U**	**Contr. (%)**	**^{40}K**	**Contr. (%)**	**^{232}Th/^{238}U**	**Total**
MP001	21.1±2.5	3.3	7.5±2.8	1.2	609.6±5.7	95.5	2.8	638.2±9.6
MP002	45±3.5	5.5	11.9±2.0	1.5	759.9±10	93.0	3.8	816.8±15.5
MP003	37.5±2.4	4.1	10±1.8	1.1	868.4±6	94.8	3.8	915.8±10.2
MP004	46.4±3.5	5.3	11.9±2.0	1.4	809.9±10	93.2	3.9	868.1±15.5

MP011	46.2±2.9	6.8	10.5±2.5	1.5	626.2±9.1	91.7	4.4	682.9±14.5
MP012	45.9±3.6	4.4	7.2±1.8	0.7	985.3±7	94.9	6.4	1038.5±12.4
MP013	32.1±5.3	6.1	7.2±3.8	1.4	487.9±5.5	92.6	4.5	527.1±14.6
MP014	30.9±3.3	5.9	6.4±2.5	1.2	487.9±4.1	92.9	4.8	525.2±9.9
MP111	31.1±3.1	6.5	8.34±2.9	1.7	442±7.1	91.8	3.7	481.5±13.1
MP112	36.8±2.9	4.4	9.6±2.4	1.2	783.5±9.9	94.4	3.8	829.9±16.2
MP113	15.5±4.3	4.6	4.8±3.0	1.4	317±8.4	94.0	3.2	337.2±15.7
MP114	41±3.5	8.3	7.5±2.9	1.5	444±9.1	90.2	5.5	492±14.5

The reason for the difference may be mostly coming from the type of detector used in each case, the accuracy of the response at the energies used in the analyses, and the method of calibration. Moreover, soil moisture variation or actual variation in the dry concentration of radionuclides with depth is among the limiting factors for establishing comparability between in-*situ* and soil sample measurement.

Table 3Statistical data for activity concentration (Bq kg^{-1})

Nuclide	**Mean**	**Confid. ±95.000%**		**Median**	**Range**		**Std.Dev.**	**Standard error**
^{232}Th	34.3	26.8	41.9	34.5	15.5	46.4	10.5	3.3
^{238}U	8.3	6.8	9.9	7.9	4.8	11.9	2.1	0.7
^{40}K	641.8	488.8	794.8	617.9	317.0	985.3	213.9	67.6
^{232}Th/^{238}U	4.1	3.4	4.8	3.9	2.8	6.4	1.0	0.3
Total	684.4	524.9	844.0	660.6	337.2	1038.5	223.1	70.5

It may be noted that in the *in-situ* measurement, the detector samples the photon flux from a volume of soil out to a radius of approximately 10 m and down to a depth of about 30 cm, depending upon the photon energy (Kelvin, 1997). Fig. 1.c shows a pictorial representation of the relative ground area contributions to the primary (uncollided) flux at a height of 1m for a medium energy (662keV) source with typical exponential depth profile in the soil. Thus, the activity that was closer to the soil surface tends to produce a wider field of view. In effect, a field spectrum samples an area of several hundred square metres: averaging out the local inhomogenities in the distribution of the Radionuclides.

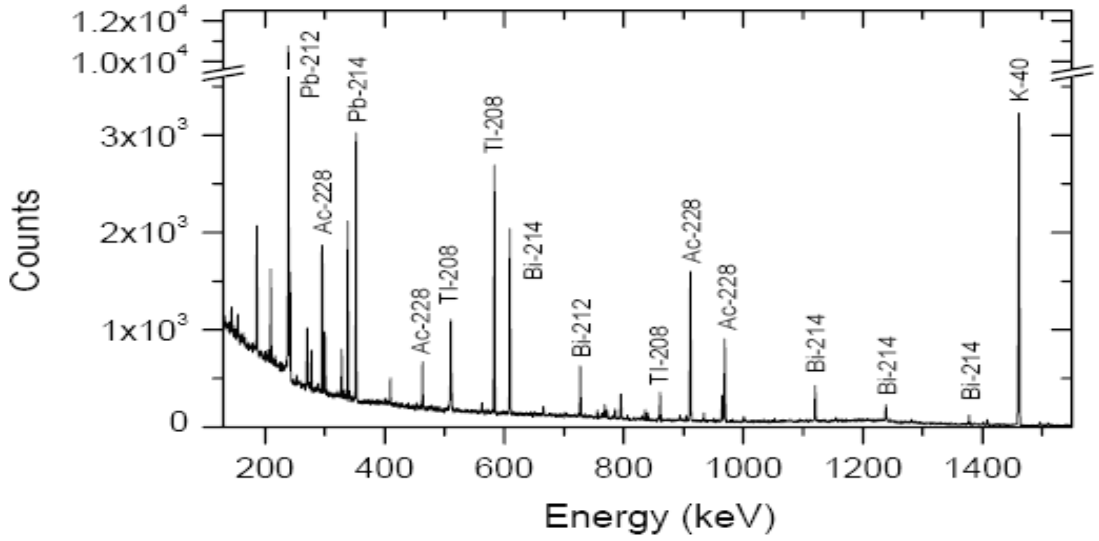

Figure 2: A typical gamma-ray spectrum showing important identified photo-peaks and their associated Radionuclides

The calculated activity mean values from the locations were 34.3, 8.3 and 641.8 Bq kg^{-1} for ^{232}Th, ^{238}U and ^{40}K respectively. Correspondingly the revised median values worldwide (UNSCEAR, 2000) are 30, 35, and 400 Bq kq^{-1} respectively. This reveals that the mean concentration level measured in the selected areas of Zaria for ^{232}Th is slightly higher than the world median value while that of ^{238}U is lower than the corresponding world value; thus ^{238}U is less than the world value by a factor of three. However, ^{40}K concentration appeared to be higher than the world median value at all the points, except MP113 with value of 317 ± 8.4 Bq kg^{-1}.

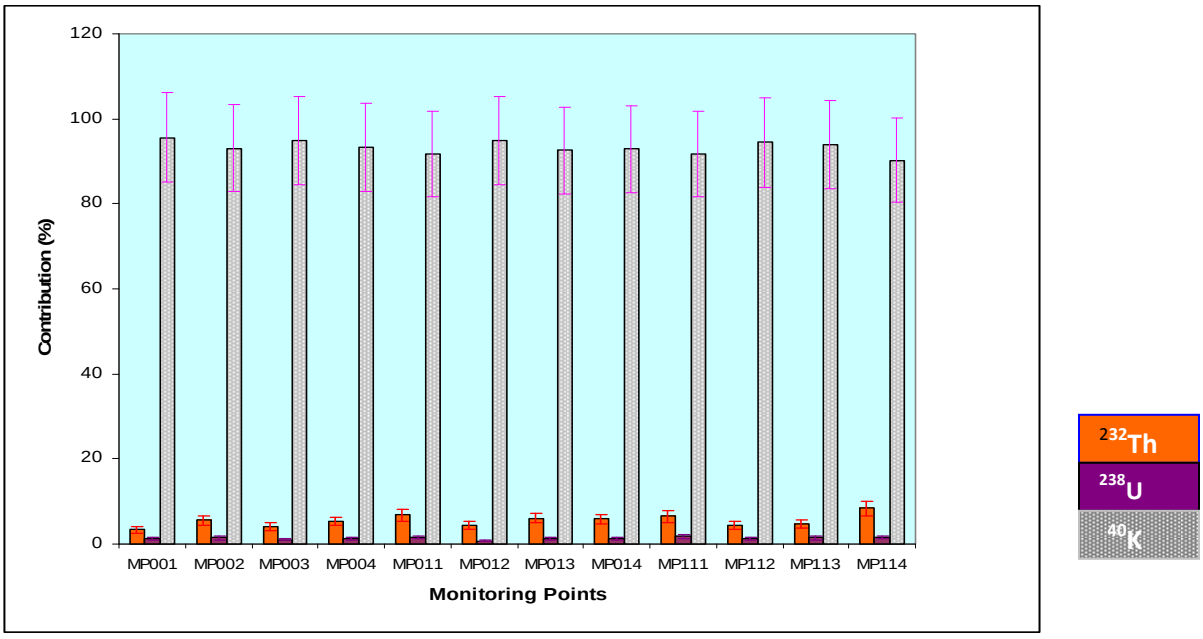

Figure 3: Percentage contribution of activity concentration

4 Conclusion

High-resolution gamma ray spectrometry was exploited to determine distribution of gamma emitting Radionuclides in soils around the Centre for Energy Research and Training, Ahmadu Bello University, Zaria. Natural radionuclide concentrations determined for ^{232}Th, and ^{238}U were significantly lower when compared with those determined by laboratory based NaI(Tl) detector system but in general agreement for ^{40}K by both techniques. The mean activity concentrations were higher than the revised world median values for ^{232}Th, and ^{40}K and lower for ^{238}U. This technique is useful for operational and post operational monitoring for the Centre, which will serve as reference for present and future assessment. The assessment of the radionuclide level of the area did not detect the presence of any artificial radionuclide and thus no significant impact of the extensive usage of radioactive materials within and around the Centre the on the radiation burden of the environment.

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