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In-situ gamma-ray mapping of environmental radioactivity at *i*Themba LABS and associated risk assessment

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Abstract. *In-situ* and *ex-situ* measurements of environmental radioactivity were made on the *i*Themba LABS (*i*TL) grounds in South Africa. The MEDUSA and HPGe detector systems were used to make *in-situ* and *ex-situ* measurements, respectively. The MEDUSA was mounted ∼0.5 m above the ground on a 4 × 4 vehicle to traverse [at $\sim 2\,\mathrm{m}\cdot\mathrm{s}^{-1}$] the accessible portions of the *i*TL grounds. Spatial data (via a GPS receiver) were acquired every 1 s, and γ-ray spectra every 2 s. MEDUSA count rate maps were produced to show the spatial distribution of radioactivity on the grounds. The HPGe was used to measure the radioactivity in soil (and also in some grass) samples collected at particular spots on the *i*TL grounds. The sampled spots include six identified high activity spots ("hot spots") and two "calibration spots". The activity concentrations were determined for both the natural and anthropogenic radionuclides. The absorbed and effective doses (from external γ-ray irradiation) were also determined for the natural and anthropogenic radionuclides. The maximum effective dose to humans on the *i*TL grounds as a result of external exposure to natural and anthropogenic radionuclides was found to be well below the regulatory 1 mSv per year per member of public.

1. INTRODUCTION

*i*Themba LABS (*i*TL) is a multidisciplinary research facility managed by the South African National Research Foundation. *i*TL provides facilities for radioisotope production, particle radiotherapy, as well as basic and applied research using particle beams. There are five particle accelerators at *i*TL, namely, the main 200 MeV separated sector cyclotron, two smaller injector solid pole cyclotrons, and two Van de Graaf accelerators. The radioisotopes produced by *i*TL are used for nuclear medicine and other industrial applications. Among the long-lived radioisotopes produced are Iron-55 (^{55}Fe), Sodium-22 (^{22}Na), Cobalt-57 (^{57}Co), and Cerium-139 (^{139}Ce). The production of these radioisotopes results in the controlled release of waste radionuclides into two holding ponds on the *i*TL site. Among the long-lived waste radionuclides produced are Sodium-22 (^{22}Na), Manganese-54 (^{54}Mn), Zinc-65 (^{65}Zn), Cobalt-57 (^{57}Co), Germanium-68 (^{68}Ge), and Tin-113 (^{113}Sn) (Hlatshwayo, 2007).

The water from the ponds is also used to irrigate the *i*TL grounds. Since 2004, the Environmental Radioactivity Laboratory (ERL) of *i*TL has been making *in-situ* and *ex-situ* measurements of environmental radioactivity on the *i*TL site. This is done in order to determine the distribution and quantity of radionuclide contamination due to irrigation, and to assess the radiological risks that the contamination poses to persons on site. This paper focuses on measurements done in June 2004, February 2005, and July 2005.

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2. EXPERIMENTAL METHODS

The MEDUSA *in-situ* gamma-ray spectrometry system (De Meijer, 1997; www.medusa-online.com) and a HPGe detector (low-background) system were used to make *in-situ* and *ex-situ* measurements, respectively. The MEDUSA system comprises a CsI(Na) detector which is interfaced with GPS receiver. The lead-shielded HPGe detector is a Canberra (model GC4520) p-type detector (45% relative efficiency at 1.33 MeV, 2.2 keV FWHM energy resolution at 1332 keV) with crystal diameter 62.5 mm, and length 59.9 mm (Newman, et al., 2008). The MEDUSA detector was mounted ~0.5 m above the ground on the front of a 4 × 4 vehicle to traverse (at $\sim 2\,\mathrm{m \cdot s^{-1}}$) the accessible portions of the *i*TL grounds. Spatial coordinates (with resolution ~3 m) were acquired via the GPS receiver whose antenna was mounted directly above the crystal (see Figure 1(a)). Maps of detector count rate were produced to show the spatial distribution of radioactivity on the grounds. Stationary MEDUSA measurements were made and samples were collected at spots of interest.

The types of samples collected were either soil (~10 cm depth) or grass, but most were soil. A representative sampling strategy, which involved the taking of 5 samples per spot, with a separation of 80 cm between the samples, was used at certain spots of interest (see Figure 1(b)). Each soil sample taken was weighed and oven-dried at 105 °C temperature overnight. Subsequently the sample was sieved through a wire mesh to remove organic materials, stones, and lumps (Joseph, 2005) and then poured into a Marinelli beaker up to the 1 litre mark. The beaker was sealed with a silicone sealant and the sample was kept in the ERL for a minimum of 21 days to allow for secular equilibrium to occur between ^{226}Ra (the parent radionuclide to radon (^{222}Rn)) and its daughter radionuclides in the soil before measuring with the HPGe detector.

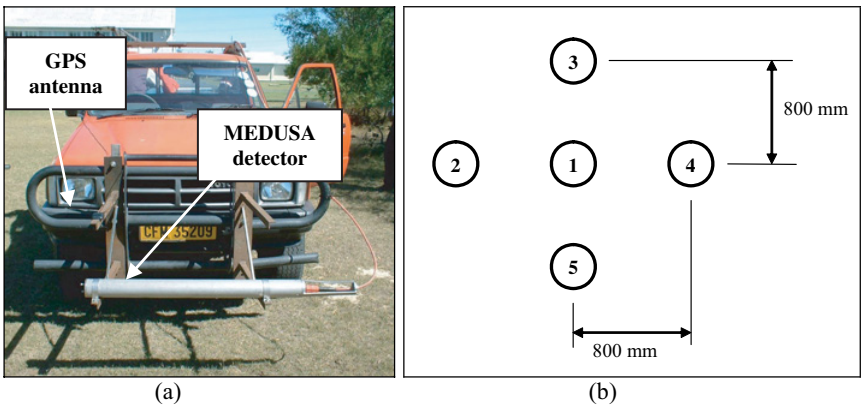

Figure 1. (a) Photograph showing the setup of the MEDUSA detector and GPS antenna on the 4 × 4 vehicle and (b) schematic top-view representation of the sampling strategy used at the spots CS1, NCS1, and HS1. The numbered circles indicate the points of sampling.

3. DATA ANALYSIS AND DOSE MODELLING

The activity concentrations of radionuclides were determined by full spectrum analyses (FSA) and windows analyses of MEDUSA and HPGe spectra, respectively. The FSA technique involves the spectral deconvolution of the entire measured MEDUSA spectra (each acquired over a 2 s period) using so-called standard spectra (generated by Monte Carlo simulations) (Hendriks, et al., 2001). In the windows analysis technique, the activity concentration is determined from the net content of the window around individual energy peaks. From the activity concentration results, the absorbed and effective doses (from external γ-ray irradiation) were determined by using dose conversion factors (for external

exposure to γ-rays associated with NORM radionuclides), the air kerma per unit [anthropogenic] source intensity, the conversion coefficient from absorbed dose in air to effective dose, and the outdoor occupancy factor (UNSCEAR, 2000; Saito and Jacob, 1994; Mohanty, et al., 2004). Effective dose rate is related to activity concentration by:

$$\text{Effective dose rate}\,(\text{mSv}\cdot\text{y}^{-1}) = \text{D}\,(\text{nGy}\cdot\text{h}^{-1}) \times 8760\,\text{h} \times 0.2 \times 0.7\,\text{Sv}\cdot\text{Gy}^{-1} \times 10^{-6} \quad (1)$$

where D = absorbed dose rate, 8760 h = number of hours in a year, 0.2 = outdoor occupancy factor, and $0.7\,\text{Sv}\cdot\text{Gy}^{-1}$ = conversion coefficient from absorbed dose in air to effective dose (UNSCEAR, 2000). The absorbed dose rate (D) was calculated as follows:

$$\text{D}\,(\text{nGy}\,\text{h}^{-1}) = 0.462\text{C}_\text{U} + 0.604\text{C}_\text{Th} + 0.0417\text{C}_\text{K} \quad (2)$$

where 0.462, 0.604, and 0.0417 are dose conversion factors for external exposure to γ-rays associated with the ^{238}U series, ^{232}Th series, and ^{40}K, respectively. C_U, C_Th, and C_K are activity concentrations of the ^{238}U series, ^{232}Th series, and ^{40}K radionuclides, respectively (UNSCEAR, 2000).

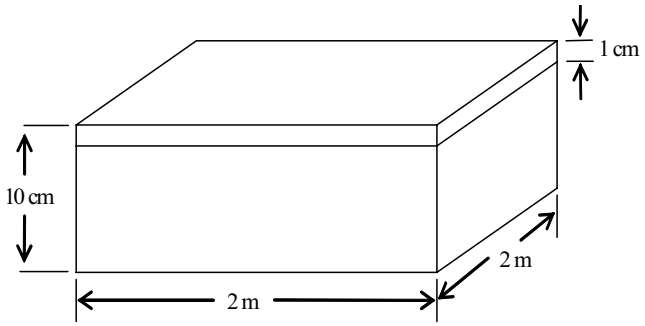

Figure 2. Schematic diagram showing features of the assumed slab of soil. The plane area is 4 m^2, the thickness of the soil slab is 10 cm, and the plane source locations are 1 and 10 cm.

In the model used to obtain the absorbed dose from anthropogenic radionuclides it was assumed that the person considered in the radiological risk assessment is a gardener who works 236 shifts of 6 hours each in a year, on the grounds of *i*TL. The outdoor occupancy factor that was calculated is 0.2. It was also assumed that the anthropogenic radionuclide source has a plane geometry as shown in Figure 2. The plane source was assumed to be located at depths of 1 and 10 cm, to determine the upper and lower limits of the absorbed dose, respectively. The air (or tissue) kerma was quoted from reference Saito and Jacob, 1994. The area assumed for the plane was $2\text{m} \times 2\text{m} = 4\text{m}^2$, taking into consideration the dimensions of the representative sampling strategy mentioned before. The thickness assumed for the soil slab was 10 cm, since samples of this approximate depth were considered. Assuming a soil density of $1.5\,\text{g}\cdot\text{cm}^{-3}$, the mass of the slab was found to be 600 kg. The activity of the soil slab was determined by multiplying the mass by the activity concentration (in $\text{Bq}\cdot\text{kg}^{-1}$) of the considered anthropogenic radionuclide. By multiplying the activity of the slab by the branching ratio of the γ-ray used for the radionuclide, the number of photons associated with the plane area was approximated per second. The last step was to multiply the tissue kerma by the number of photons to yield the absorbed dose (in $\text{nGy}\cdot\text{h}^{-1}$) associated with the source.

4. RESULTS AND DISCUSSION

The MEDUSA count-rate maps showed that there were 6 "hot spot" (HS) areas, namely, HS1, HS2, HS3, HS4, HS5, and HS6, where anthropogenic radioactivity was found to be more concentrated.

The locations of these hot spots and other spots of interest are shown in Figure 3 below. The stationary *in-situ* measurements made at these spots of interest produced gamma-ray spectra, such as that shown in Figure 4 for HS1. The radionuclides associated with the identified γ-ray energy lines were determined by referring to Firestone, 1996. Some of these identities were confirmed by observing the decay trends in re-measured samples. The activity concentration results (from ~26000 acquired MEDUSA spectra in 2005) for primordial radionuclides had ranges of 8–39, 6–21, and 30–63 $Bq \cdot kg^{-1}$ for the ^{238}U series, ^{232}Th series, and ^{40}K, respectively. The absorbed and effective dose ranges associated with this natural radioactivity were 10–27 $nGy \cdot h^{-1}$ and 13–33 $\mu Sv \cdot y^{-1}$, respectively. The anthropogenic activity concentrations in soil had the ranges of 1–6534, 0–101, 0–21, 0–1, and 0–193 $Bq \cdot kg^{-1}$ for ^{68}Ga, ^{65}Zn, ^{22}Na, ^{137}Cs, and ^{54}Mn, respectively. The absorbed and effective doses due to these artificial sources were calculated for two plane source depth locations, namely, 1 and 10 cm. The absorbed and effective dose results had ranges of 0–153 $nGy \cdot h^{-1}$ and 0–187 $\mu Sv \cdot y^{-1}$, respectively.

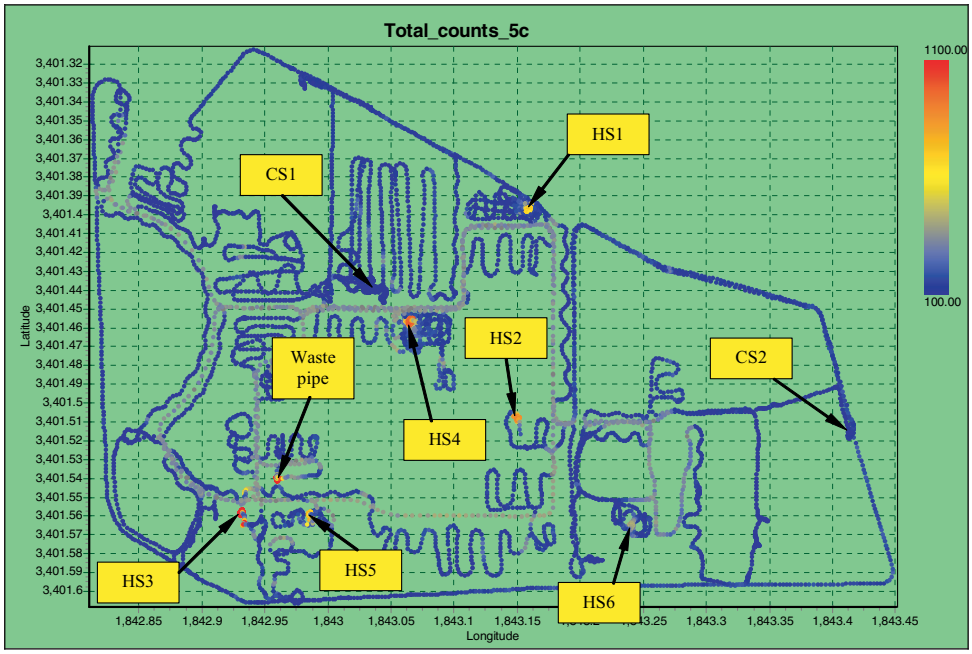

Figure 3. MEDUSA Map showing locations of the six identified "hot spots", the radioactive waste transport pipe, and the two "calibration spots" CS1 and CS2. The map also shows the count rate obtained during the July 2005 survey. The colour-coding range of the count rate is 100–1100 counts per second, and is shown on the top-right corner in the map. (Note: The map shows the Latitude and Longitude coordinates in the format x,xyy · yy instead of the usual xx°yy · yy' where xx and yy · yy correspond to the degrees and minutes, respectively.)

5. CONCLUSION AND OUTLOOK

The radioactivity level due to natural γ-ray emitting radionuclides on the *i*TL grounds was found to be below the world average of 35, 30, and 400 $Bq \cdot kg^{-1}$ for the ^{238}U series, ^{232}Th series, and ^{40}K radionuclides, respectively (UNSCEAR, 2000). The six identified "hot spots" consistently appeared on the MEDUSA count rate maps from June 2004, February 2005, and July 2005 measurements. The radionuclides that seem to contribute to the radiation in these hot spots are summarized in Table 1.

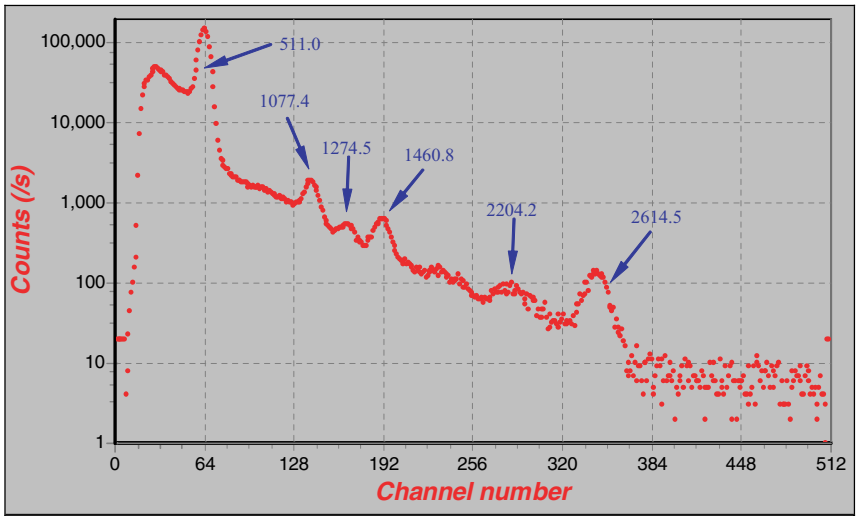

Figure 4. The raw gamma-ray spectrum acquired with MEDUSA in a stationary position over hotspot HS1 in February 2005. The labeled peaks 1077.4, 1274.5, 1460.8, 2204.2, and 2614.5 keV are associated with ^{68}Ga, ^{22}Na, ^{40}K, ^{214}Bi, and ^{208}Bi decay, respectively. The 511.0 keV peak results from the detection of positron-electron annihilation γ-rays associated with the decay of positron sources such as ^{22}Na.

Table 1. Table showing the anthropogenic radionuclides identified at the different hotspots on the site. The underlined markings indicate enhancement in the activity concentration of the particular radionuclide.

Hotspot location	Identified anthropogenic radionuclides				
	^{68}Ga	^{65}Zn	^{22}Na	^{137}Cs	^{54}Mn
HS1	$\underline{\times}$	×	×	×	×
HS2	$\underline{\times}$	×	×	×	×
HS3	$\underline{\times}$	$\underline{\times}$	×	×	$\underline{\times}$
HS4	$\underline{\times}$	×	×	×	×
HS5	$\underline{\times}$	×	×	×	×
HS6	×				

The maximum effective dose to a person on the *i*TL site due to external irradiation by γ-rays emanating from primordial and anthropogenic radionuclides was found to be well below the regulatory 1 mSv per year per member of public (ICRP, 2004; Hlatshwayo, 2007).

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