

Neutron activation of stable isotopes in soil and groundwater from a radionuclide production facility, South Africa

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ABSTRACT

The neutron activation of stable isotopes in environmental matrices, such as soil and groundwater, is a critical aspect of assessing the impact of radionuclide production facilities on the surrounding ecosystem. The envisioned Low-Energy Radioactive Ion Beams (LERIB) facility at the iThemba LABS, South Africa is anticipated to generate significant sources of ionising radiation. The study investigated the possible repercussions of neutron irradiation stemming from the facility, focusing on the activation of stable isotopic compositions in the environment. The investigation employed a combination of experimental and analytical techniques to characterize the neutron activation products in soil and groundwater samples collected from the vicinity. Samples were collected from designated areas for background radiological measurements and were irradiated with neutrons for a period of 1 h. The induced radioactivity measured by the High Purity Germanium detector included ^{24}Na , ^{22}Na , ^{54}Mn , ^{52}Mn , and ^{46}Sc . The application of Darcy's law for groundwater velocity suggests that radionuclides in groundwater will migrate at an average flow velocity of 0.8 m/day. The isotopes with longer half-lives have count rates at background concentrations; therefore, environmental impacts on the site and surrounding communities might be minimal.

Key words: activation, cyclotron, groundwater, isotope, neutron, radiation

HIGHLIGHTS

- Radionuclides may migrate with groundwater.
- Groundwater flow velocity can determine the likelihood of radioactive material from the source to another point.
- Major isotopes are more prone to neutron activation compared to minor isotopes.
- Sufficient shielding reduces the chances for neutrons to activate matter in the environment.

INTRODUCTION

In the realm of nuclear research facilities and nuclear technologies, the production of radionuclides stands as a crucial endeavour, providing essential tools for medical, industrial, and scientific purposes. However, the inherent risks associated with radionuclide production necessitate a comprehensive understanding of their environmental impact, particularly in the context of neutron irradiation activity. Neutron activation, a phenomenon where stable isotopes absorb neutrons and transform into radioactive isotopes, can significantly alter the isotopic composition of surroundings, such as soil and groundwater. This can be seen in the findings of Pantelias & Volmert (2015) where activation in the concrete walls was produced by the transformation of elements that originally existed as stable isotopes into radionuclides by neutron irradiation during the operation of a nuclear power plant.

The iThemba Laboratory for Accelerator-Based Sciences (iThemba LABS) intends to construct a facility to produce a Low-Energy Radioactive Ion Beam (LERIB). The radioactive ion beam (RIB) from the LERIB facility will be used for nuclear physics research. The Separated Sector Cyclotron (SSC) will be a driver for the isotope separation on-line (ISOL) facility. Beam current up to 50 μA of a 66 MeV proton beam will be delivered by the SSC to produce radioactive beams from a target ion source. The LERIB will be the first stage of the production of radioactive beams at the iThemba LABS. Only low-energy

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radioactive ion beams will be available from the facility. Stage two will be the post-acceleration of the radioactive beams to energies of 4 to 5 MeV per nucleon with a linear accelerator.

The LERIB project is anticipated to generate significant sources of ionising radiation including neutron radiation. The neutron radiation is associated with the ability to activate matter at which it traverses might pose significant risks to the soil and groundwater underlying the LERIB facility. A study by Mantengu (2016), using a programme called FLUKA to calculate the optimal shielding design for the LERIBs project at iThemba LABS predicted that the subsurface at a depth of 2–3 m bgl might be prone to activation when containment is insufficient. Therefore, radioactive isotopes resulting from neutron activation underneath the LERIB facility can be expected due to the ability of neutrons to penetrate any matter they are interacting with.

The induced radioactivity in the subsurface can pose potential environmental risks, particularly in the underlying aquifer. The fluctuating water table, during wet seasons (winter), rises close to the surface and might increase the chances for groundwater to interact with neutrons. The groundwater flow might act as a conduit for the migration of the induced radionuclides, resulting in contamination of groundwater bodies on site and down gradient. Activation of the subsurface will deteriorate the groundwater quality and pose negative effects on groundwater users on site and in communities adjacent to iThemba LABS. For these reasons, it might not only be the radiation-producing facility (iTL) that might face contamination risks associated with ionising radiation but also the nearby surrounding communities. Hence, it is important to sufficiently contain and shield ionising radiation to ensure the safety and protection of the public and the environment.

Samples were undertaken in sandstone geological material and were strategically chosen to capture potential spatial variations in neutron activation products. An existing 30-m borehole drilled in a sandy layer was used for groundwater collection. Three more 15 m boreholes were drilled also in a sandy layer around the facility for the sampling of groundwater (Figure 1). The mud-rotary and odex drilling techniques were used interchangeably for the 15 m borehole drilling process and soil samples were collected from the recovered lithological core at a depth of 2–3 mbgl using 50-ml centrifuge tubes (Figure 1).

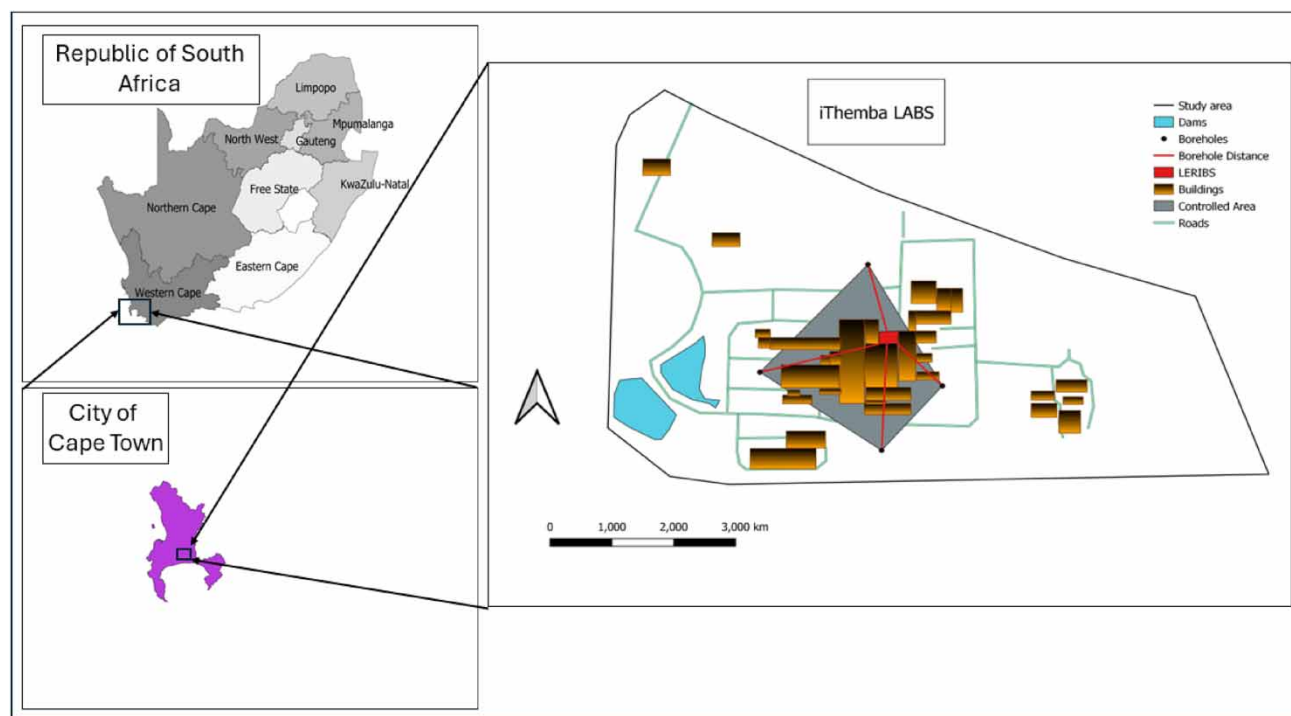


Figure 1 | Digitized topographic map showing the position of the envisioned LERIB facility at the iThemba LABS with sampling points located around the facility. In terms of the climate and weather conditions, Cape Town has a Mediterranean climate, that consists of warm summers with a maximum average of 26 °C. The region experiences cool winters with an average minimum of 7 °C. Regional studies have shown that there is very little rainfall between late spring and early autumn (October–March) (Adelana *et al.*, 2006). However, most of the annual rainfall occurs in the middle of winter (June) with a mean annual precipitation of 619 mm (Olivier & Xu 2019).

GEOLOGY AND GEOHYDROLOGICAL SETTING

The geological setting of the Western Cape province is largely comprised of the Cape Supergroup deposited in the Cape basin around the southern perimeter of the Kalahari Craton. The findings of [Tankard *et al.* \(1982\)](#) concluded that the Cape Supergroup consists of sediments laid down from early Ordovician to early Carboniferous times, approximately between 500 and 340 million years ago. Moreover, it is worth noting that the Cape Supergroup is divided into three major lithostratigraphic units, namely, the Table Mountain, Bokkeveld and Witteberg Groups ([Tankard *et al.* 1982](#)) ([Figure 2](#)). The Table Mountain Group (TMG) is the lowest in the Cape Supergroup and dominated by quartz-arenites of approximately 400 m thick sequence, in contrast to the feldspathic subgreywackes and shales of the overlying Bokkeveld Group ([Figure 2](#)).

The TMG

The Witteberg sandstones are feldspar-poor, but the finer-grained lithologies contain alkali-feldspar and are very micaceous ([Fourie *et al.* 2011](#)). Moreover, [Rust \(1973\)](#) as cited in ([Fourie *et al.* 2011](#)) suggested that the TMG is characterised by thick packages of medium- to coarse-grained quartz-arenites, conglomerates and subordinate fine-grained lithologies. Whereas on the other side, the boundary of the Bokkeveld Group with the TMG is represented by a marine-flooding surface with quartz-arenites, but conformably grading into shelf and pro-delta shales and mudstones ([Theron 1972](#) as cited in [Fourie *et al.* 2011](#)). The Witteberg Group, which conformably overlies the Bokkeveld Group, comprises quartz-rich sandstones and mica-rich mudstones with intercalated siltstones.

The geology of the TMG where the study area is situated can be further subdivided into six formations, namely the Piekenierskloof Formation, Graafwater Formation, Peninsula Formation, Parkhuis Formation, Cederberg Formation and Nardouw Subgroup ([Table 1](#)). The lowermost unit of the TMG is the Piekenierskloof Formation with a maximum thickness of about 900 m ([Rust, 1967](#)) at Citrusdal and 390 m at Piketberg. The lowest unit of the TMG has a lithology comprised of conglomerate, quartz arenite and minor mudrock that are confined to the West Coast region. The Graafwater Formation is characterized by mud-cracked sandstone, siltstone, and shale beds. The calculations of [Rust \(1967\)](#) show a maximum thickness of 424 m in the area west of Clanwilliam ([Figure 3](#)). The Peninsula Formation is exposed against Table Mountain with a succession of 550 m and comprises a succession of coarse-grained, white quartz arenite with scattered small pebbles and discrete thin beds of a matrix-supported conglomerate ([Rust, 1967](#) as cited in [Duah & Xu 2010](#)). The formation reaches a

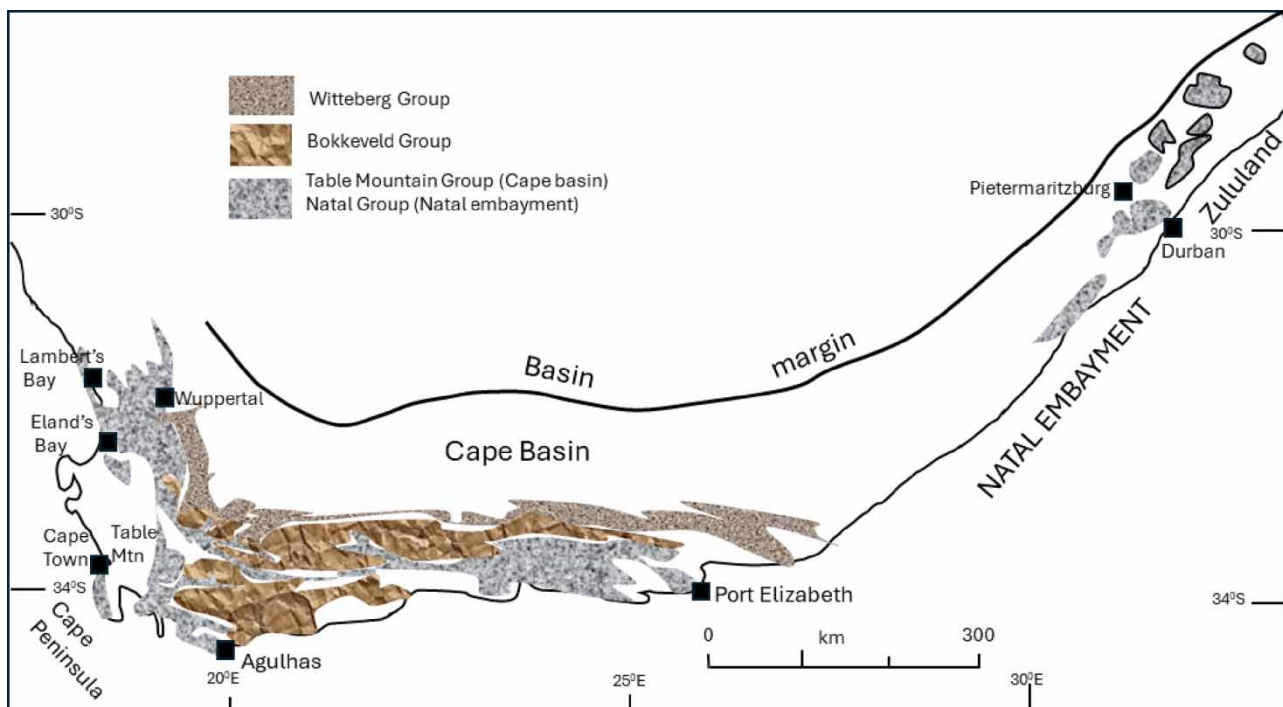


Figure 2 | Geological setting of the Cape Super Group. Source: [Tankard *et al.* 1982](#).

Table 1 | Stratigraphy of the TMG

Group	Subgroup		Bed thickness (m)	Maximum thickness (m)	Lithology
Bokkeveld		Formation	4,000		Siltstones, shales, sandstones
Table Mountain	Nardouw	Rietvlei/Baviaanskloof	0.5–1	280	Feldspathic quartz arenite
		Skurweberg	1–2	390	Quartz arenite
		Goudini	0.3–0.5	230	Silty sandstones, siltstone
		Cedarberg	0.1–0.3	120	Shale, siltstone
		Pakhuis	Variable	40	Diamietite, shale
		Penninsula	1–3	1,800	Quartz arenite
		Graafwater	0.1–0.5	420	Impure sandstone, shale
		Piekenierskloof	0.3–1.5	900	Quartz arenite, conglomerate, shale
Basement		Underlying the TMG are the Malmesbury shales, the Gamtoos and the Kaaimans argillites, comprising moderately to lightly metamorphic sedimentary rocks; and cape granite suite.			

Source: Duah & Xu 2010.

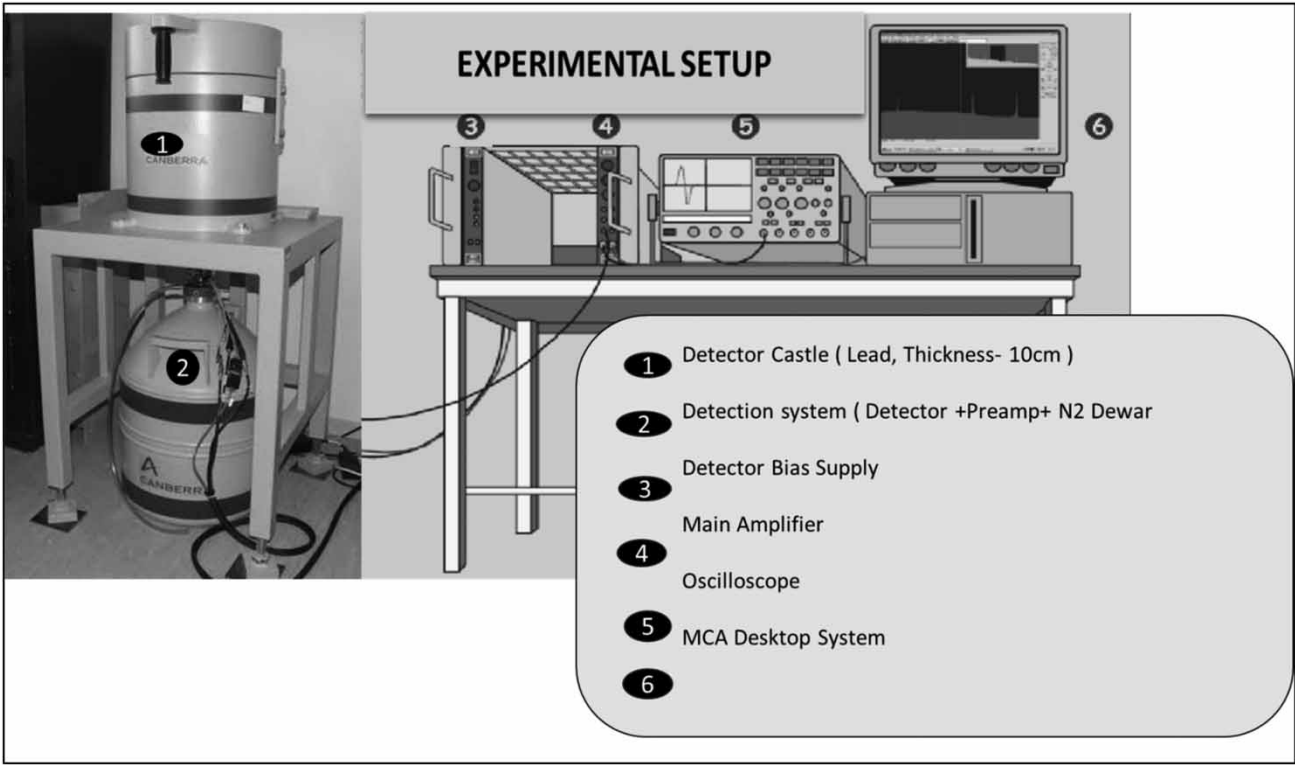


Figure 3 | Photograph of the HPGe detector setup with a lead castle on the left housing the detector and supported on a mechanically rigid cryostat connected to a liquid nitrogen Dewar and a sketch of an experimental setup on the right.

thickness of about 1,800 m at Clanwilliam in the west (Rust, 1967). The Pakhuis Formation occurs above the Peninsula Formation and comprises about 40 m of glacially derived sediments, but is restricted to the southwestern Cape (Rust, 1967; Broquet, 1992).

The Cedarberg Formation has a maximum thickness of 120 m (thin formation) but remarkably continuous unit, consisting of black silty shale at the bottom, grading into brownish siltstone and fine sandstone at the top. It is exposed within the southwestern Cape but continues along the whole length of the southern Cape Fold Belt. The Nardouw Subgroup, with its three subdivisions, the Goudini, Skurweberg and Rietvlei (Baviaanskloof in the Eastern Cape) Formations, is another thick (maximum 1,200 m) unit of sandstone that varies between quartz arenite, silty and feldspathic arenites, accompanied by some very minor inter-bedded conglomerate and shale.

The variation in lithological diversity, textural, grain size and bedding thickness differences of these formations lead to pronounced differences in weathering, structural and hydrogeological characteristics (Duah & Xu 2010). Topping the Cape Supergroup are the recent coastal Cenozoic age deposits which consist of quartzose sands, gravel, clay, lignite, calcareous aeolianites, bioclastic-silicilastic aeolianites. According to the Group A Vegter classification, 1995, the Coastal Cenozoic aquifers of South Africa are all classified as primary aquifers.

The primary Coastal Cenozoic aquifers include the Sandveld group which is divided into four main hydrogeological units (Meyer 2002) as follows:

- (i) Cape Flats Unit – extending from False Bay to Melkbosstrand.
- (ii) Silwerstroom-Witzand is situated in the Atlantis area.
- (iii) Grootwater Unit is situated in the Yzerfontein area.
- (iv) Berg River Unit situated in the Saldanha area:
 - (a) Adamboerskraal Aquifer Unit
 - (b) Langebaan Road Aquifer Unit
 - (c) Elandsfontyn Aquifer Unit

The study area taps from the Cape Flats unit and is associated with groundwater levels generally shallow (2–5 mbgl) throughout and are also vulnerable to contamination (Conrad 2004). Groundwater within the Cape Flats Aquifer flows in a westerly direction towards Zeekoevlei and a southerly direction towards Monwabisi/Mnandi. The groundwater of the Silwerstroom-Witzand Unit, also known as the Atlantis Aquifer System, flows westwards to south-westwards where it discharges along the coast in areas where the aquifer dips below sea level. The Grootwater Unit basically consists of two aquifer systems, an upper and lower aquifer system. The aquifer systems of the Berg River Unit consist of two aquifers, separated by a clay layer. Borehole yields within the Sandveld Group range between 0.1 and 5 l/s with most boreholes yielding 0.5 l/s or less and about 30% yielding 2 l/s and more (Meyer 2002). Below the primary unconsolidated aquifer, the secondary hard rock aquifer is found and consists of Malmesbury shales and intrusive granites (Saayman & Adams 2002).

MATERIALS AND METHODS

Groundwater sampling

Groundwater samples were collected using 50-ml centrifuge tubes. The 50-ml tubes were rinsed in the laboratory using deionised water prior to the collection of groundwater samples to avoid sample cross-contamination. The collected groundwater samples were stored in a temperature-controlled cooler box to avoid exposure to the external environment such as the temperature. The samples were labelled according to borehole numbers, i.e. B1A, B2A, and B3A.

Soil sampling

The soil samples were collected at a depth of 2–3 and 3–4 m from the recovered lithological core of B1A, B2A and B3A. The layer at which soil samples were collected is the interface of the subsurface where the shielding material is anticipated to be located with a thickness of 2–4 m stratum. The disposable plastic spoons were used to collect and fill samples into 50 ml centrifuge tubes. A total of 12 soil samples were collected and marked according to the borehole IDs and depths of collection such as B1A (2–3 m), B1A (3–4 m). The Environmental Radioactive Laboratory (ERL) based at iTL was used to store samples at a controlled room temperature of 20 °C until the borehole drilling and sample collection were completed.

Soil and groundwater neutron activation

The soil and groundwater samples were analysed for background radiation counts for a period of 24 h using the High-Purity Germanium (HPGe) detector (Figure 2). The background radiation measurements were carried out at the iThemba LABS in the Environmental Radiation Laboratory under low-radiation background conditions achieved by shielding the detector (using lead) from ambient gamma radiation. The nuclear electronics were used to process pulses from the detector, through a multi-channel analyser (MCA) and binned in a gamma-ray spectrum (Figure 3).

The neutron activation technique was conducted to qualitatively ascertain the different radioisotope species that would typically form should the soil and groundwater material be exposed to ionising radiation such as neutrons. The soil and groundwater samples were irradiated by neutrons travelling in the same direction as the proton beam resulting in a higher

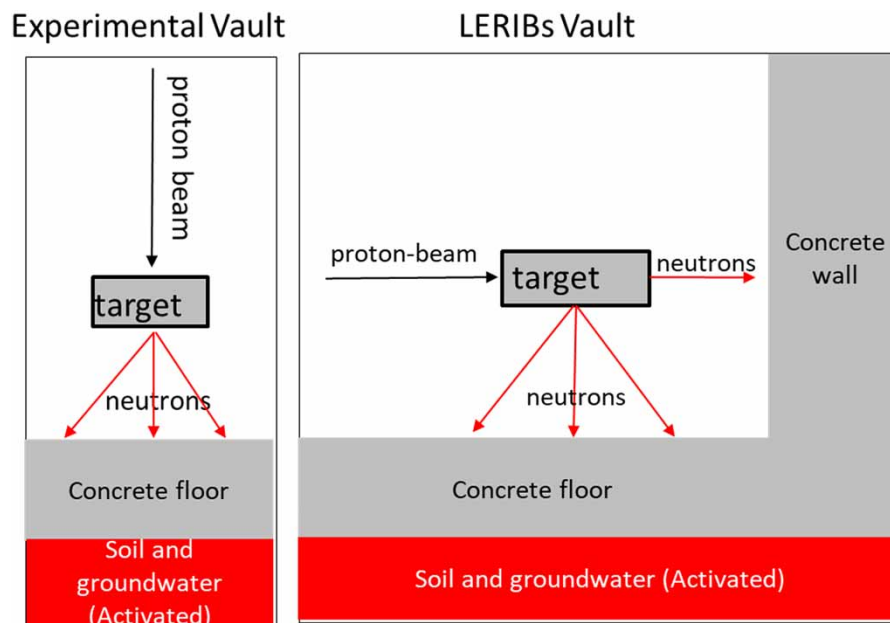


Figure 4 | Sketch diagram depicting an incident of a proton beam and resultant neutrons from the experimental vault and the anticipated LERIBs vault.

neutron dose (Figure 4). By contrast, in the LERIB facility, the neutrons incident on the soil and groundwater will be traveling at 90 degrees to the proton beam as a result, a lower neutron flux is expected (Figure 4).

The protons at 66 MeV energy incident on a Beryllium (^9Be) target (target current 25 μA) generate a spectrum of neutron energies between 0 and 66 MeV. The bombardment of ^9Be by protons can provide abundant neutrons which then extend the probability of activating matter in the receiving environment (Osipenko *et al.* 2013). According to Hughes (1957), neutrons of extremely wide energy range such as 0–66 MeV can interact effectively with nuclei. Moreover, in terms of neutron energy range, a study by Yashima *et al.* (2004) generated neutrons from a lithium target bombarded by proton energies of 30, 35, 40, 50, 60 and 70 MeV and revealed that neutrons generated from these energies effectively interact with atomic nuclei. Thus, the neutron energy of 0–66 MeV range is deemed adequate to be used in activating samples for the purpose of this study. Samples were placed on a 10 cm thick lucite block serving as a backscatter medium, covered with a 12 mm thick polycaprolactam (nylon 6) build-up material.

The soil and groundwater samples activation were quantified by means of gamma-ray spectroscopy, where directly after irradiation the collective sample dose rate exceeded 1 mSv/h. The neutron activation analysis using gamma-ray spectroscopy instrumentation could not be performed immediately after activation, as the detector dead-time exceeded 90%. Data acquisition for the first soil sample could only be performed 30 min post-activation.

Abundant radioisotopes with short half-lives decayed over the 30-min period to levels quantifiable by the detector and analysing equipment. Gamma-ray spectrum acquisition time was set at 1-h detector live time per sample. The sample acquisition time for samples analysed at time points 24, 72 and 168 h post bombardment was 2 h each to ensure good counting statistics for the low count rates. To distinguish between isotopes resulting from neutron activation and naturally occurring stable isotopes in the study area, the detected isotopes from the background radiation were subtracted from the activated samples. This was to dissociate the current background radiation from the anticipated neutron activity that might be induced by the LERIB project in the subsurface environment.

RESULTS AND DISCUSSIONS

Background radiation

The background radiation was measured from the soil and groundwater samples to determine the current environmental radiological conditions of the study area. The measured background radiation is believed to be from the Naturally Occurring

Radiation Materials (NORM) as there were no detected Technologically Enhanced Naturally Occurring Radiation Materials (TENORM) from the soil and groundwater samples (Figure 5). Natural occurring radioactive isotopes such as ^{40}K and ^{208}Bi were measured at gamma energies 1,460 and 2,614 keV, respectively, both from soil and groundwater samples (Figure 5). Based on the attained background spectra, it was evident that the study area had no presence of artificial radionuclides.

Weir 2004 argued that ionizing radiation can be mutagenic, and exposure increases the risk of cancer. The gamma radiation, which the activated isotopes in the study area emit when decaying, is associated with penetrating ability and can

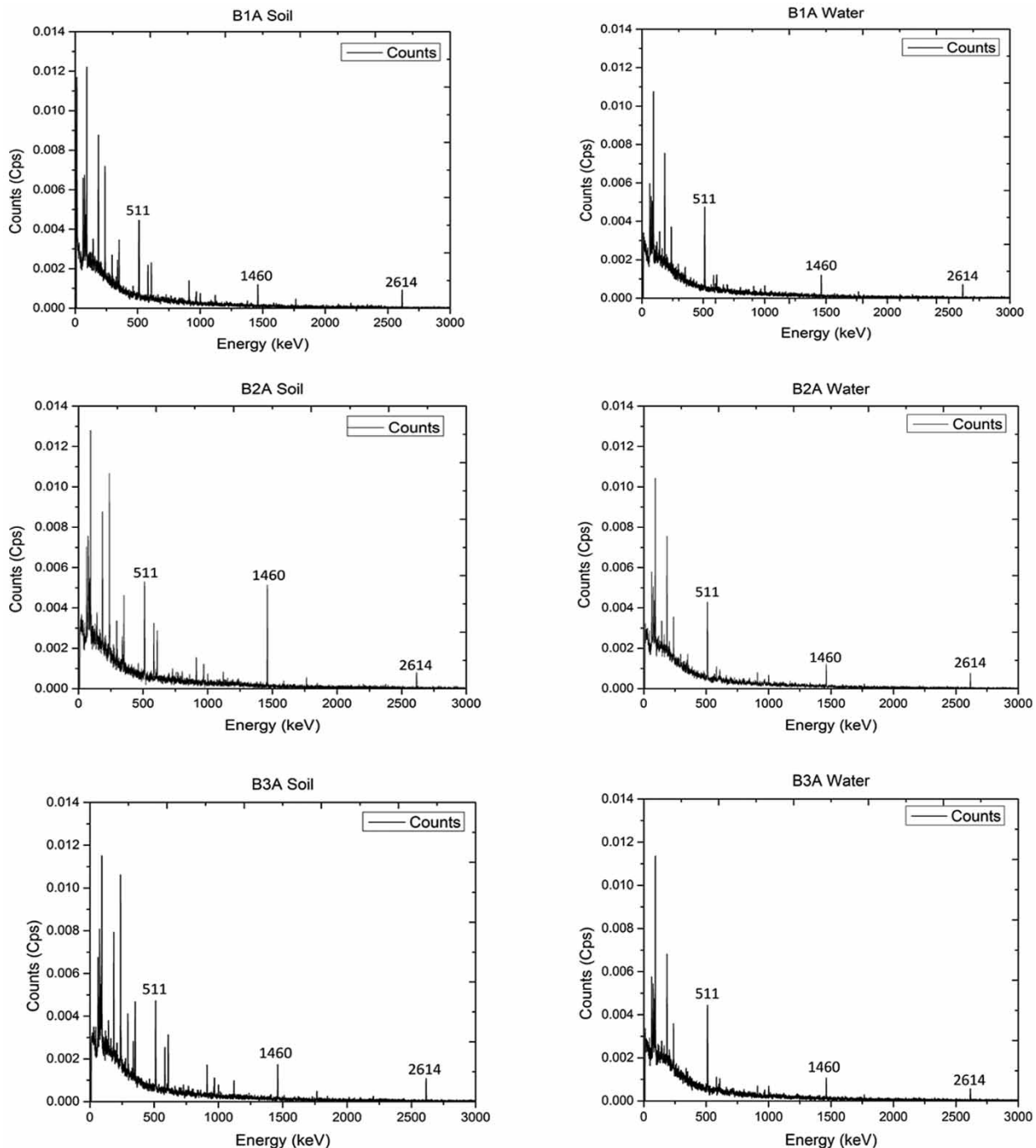


Figure 5 | Background radiation of soil and groundwater sampled from borehole numbers B1A, B2A, and B3A.

cause severe cell damage and mutagenesis even when the source is not taken internally (Campbell 2009). It is for this reason that the proper shielding of neutron radiation is recommended.

1-h post activation

The measured soil and groundwater samples at 1-h post activation revealed the presence of an isotope of sodium-24 (^{24}Na) (Figure 5). According to Gritzay *et al.* (2002), sodium-24 might be formed by activating a stable isotope of ^{23}Na through a nuclear reaction $^{23}\text{Na}(n,p)^{24}\text{Na}$. The gamma energies representing ^{24}Na are 1,368 and 2,754 keV (Chu *et al.* 1999). Both gamma energies of ^{24}Na were detected in soil and groundwater samples (Figure 6).

When comparing the ^{24}Na counts from both soil and groundwater samples, it was observed that the soil samples contained high concentrations compared to groundwater samples (Figure 6). The variation in concentrations can be attributed to the fact that water being a product of two hydrogens and one oxygen to form H_2O , with ^1H from H_2O considered to be a

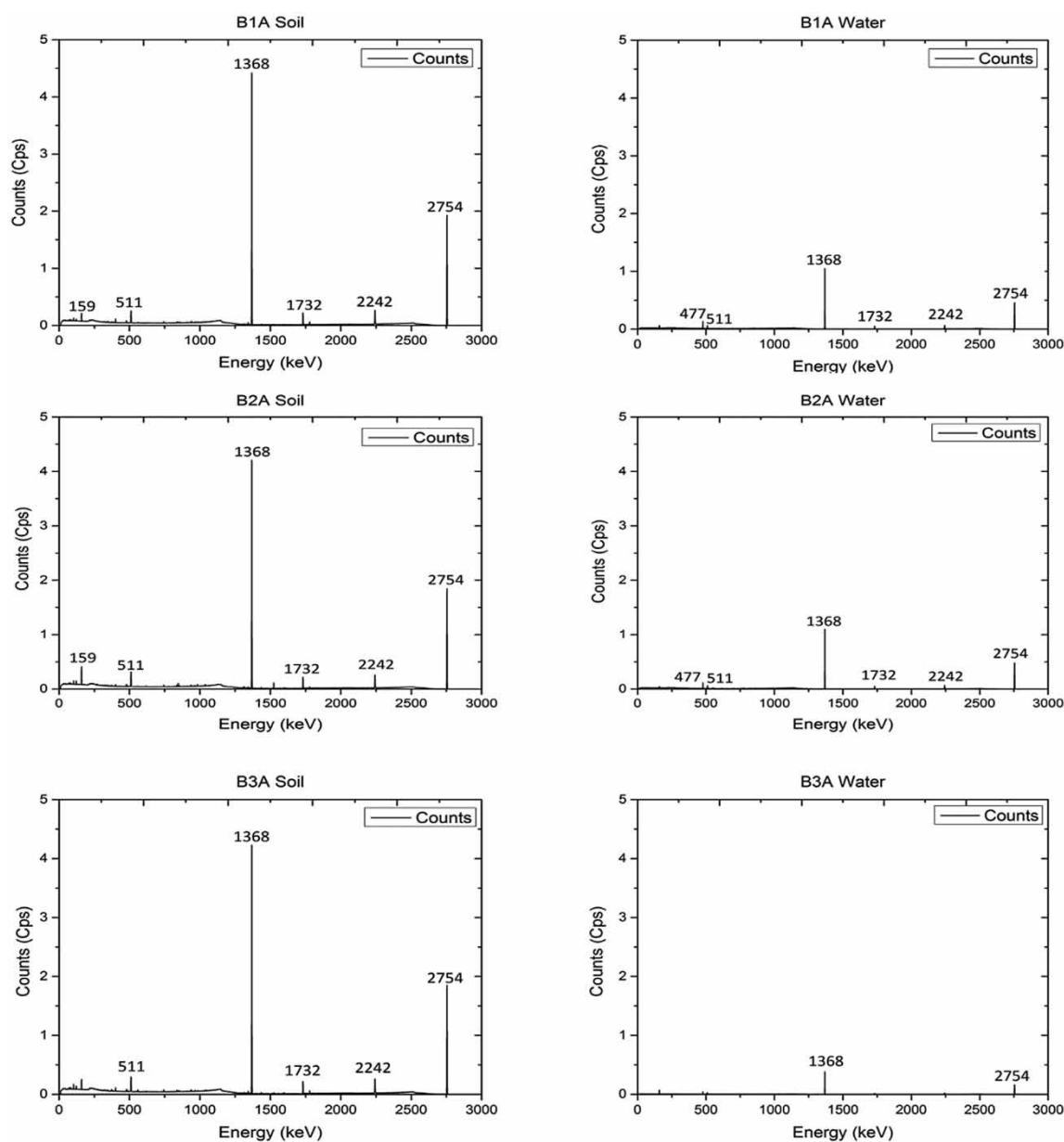


Figure 6 | Irradiated soil and water samples analysed 1-h post activation for the three boreholes.

good neutron moderator might have retarded neutrons and lowered the energy to reduce the radiation activity (Bacchi *et al.* 2002).

Based on the presence of ^{24}Na from the activated soil and groundwater samples, it can be argued that the soil and groundwater samples contained a stable isotope of ^{23}Na which when interacting with the anticipated neutron flux from the LERIBS facility will form the radioactive ^{24}Na . According to Firestone (1996), ^{24}Na has a half-life of 15 h. The ^{24}Na peaks in groundwater samples decreased drastically with time, as a result when measuring the B3A water sample, the concentration decreased from >1 count per second (Cps) (sample B1A and B2A water) to <1 Cps (Figure 6). The measured groundwater velocity suggests that ^{24}Na will undergo full decay before migrating a distance exiting the facility of iTL. Therefore, minimum environmental and health effects from ^{24}Na within the study area and down gradient can be expected.

One-day post activation

A decrease in count rates of all the measured isotopes in Figure 5 from both soil and groundwater was observed 1-day post activation. The results revealed a significant decrease in count rates from groundwater samples when compared with soil samples. The decrease in count rates can be attributed to the fact that at the beginning of the analysis (Figure 6), soil samples had a high presence of activated isotopes. The samples measured 1-day post activation showed no presence or formation of other isotopes except the isotopes measured 1-h post activation (Figure 6 and Figure 7).

One-week post activation

The last sample analysis was done 1 week post the neutron activation process to observe radionuclides with longer half-lives. Furthermore, the analysis was for the examination of whether the peaks representing short half-life isotopes re-occurred (Figure 6 and 7). The radionuclides detected in Figures 6 and 7 are isotopes with short half-lives and have undergone a full decay 1 week post-activation (Figure 8). The results showed the formation of new isotopes 1-week post-activation. The newly formed isotopes were only measured from soil samples. The groundwater samples showed no activated material 1-week post activation (Figure 8). The isotope of ^{24}Na was the only nuclide observed at 1 h and 1-day post activation from groundwater samples and results reveal that the nuclide had undergone full decay 1-week post activation.

The spectra analysed 1-week post activation revealed the presence of isotopes with longer half-lives (Figure 8). The radionuclides included manganese-54 (^{54}Mn), manganese-52 (^{52}Mn), sodium-22 (^{22}Na) and scandium-46 (^{46}Sc) (Figure 8). The radionuclides were only measured from soil samples.

Manganese-54

The isotope ^{54}Mn was measured at gamma energy 834 keV and can be formed through activating a stable isotope of ^{54}Fe , by a nuclear reaction $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ (Gritzay *et al.* 2002) (Figure 8). Iron is among the abundant major elements in the earth's crust and consists of numerous natural isotopes such as ^{56}Fe , ^{57}Fe , and ^{58}Fe . It can be noted that the iron isotopes will undergo activation at different conditions. The evidence is in the fact that only ^{54}Fe underwent activation when bombarded with neutron flux. It is therefore expected that, among the iron isotopes, only ^{54}Fe will be prone to activation once the anticipated neutrons from the LERIBs interact with the soil in the subsurface. The ^{54}Mn has a half-life of 312.3 days (Firestone 1996) and has a likelihood of exiting the study.

Manganese-52

The isotope of ^{52}Mn was measured at 744, 935 and 1,434 keV gamma energies (Figure 7). Through neutron activation, ^{52}Mn is formed by a nuclear reaction $^{54}\text{Fe}(n,t)^{52}\text{Mn}$ (Sublet *et al.* 1990). A study showed that bombarding a ^9Be target with 40 MeV (for 2.0 h of irradiation) produced neutrons that activated ^{54}Fe to form ^{52}Mn in soil. The findings of the study suggest that soil samples containing ^{52}Fe might be activated when interacting with neutrons to form a radioactive isotope ^{52}Mn with tritium being released in the reaction (Sublet *et al.* 1990). For this reason, the formation of ^{52}Mn due to neutron radiation from the LERIBs facility can be expected in soil material (Figure 8). The ^{52}Mn has a half-life of 5.6 days (Firestone 1996). According to the measured groundwater velocity, ^{52}Mn will undergo full decay before migrating a distance to any of the boreholes down the gradient of the LERIB facility.

Sodium-22

The unstable isotope, ^{22}Na was measured from soil samples at energy 1,274 keV (Figure 8). The radioactive ^{22}Na , through the neutron activation process, is formed by activating the stable isotope ^{23}Na following a nuclear reaction $^{23}\text{Na}(n,2n)^{22}\text{Na}$ and

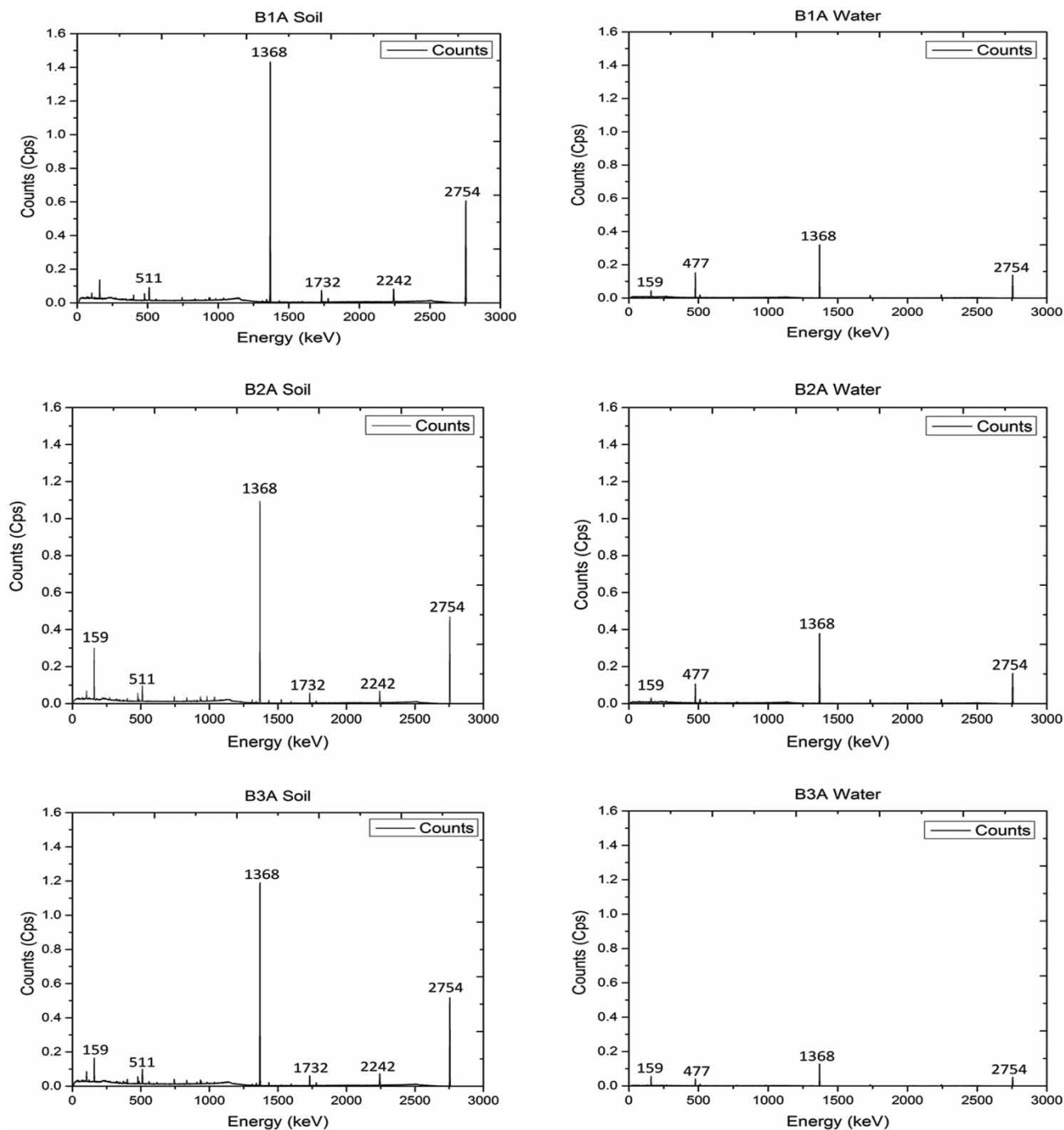


Figure 7 | Irradiated soil and water samples analysed 1-day post activation for the three boreholes.

has a half-life of 2.6 years (Firestone 1996; Gritzay *et al.* 2002). Due to the high dissolution nature of sodium, it is anticipated that ^{22}Na might dissolve with water and migrate at the speed of groundwater. The half-life of ^{22}Na suggests that the isotope will migrate a distance exiting the study area.

Scandium-46

The soil samples were measured to contain a radionuclide of scandium – ^{46}Sc at gamma energies 889 and 1,120 keV (Figure 8). The ^{46}Sc is produced by activating a stable isotope of ^{46}Ti with a nuclear reaction $^{46}\text{Ti} (n, p)^{46}\text{Sc}$ and has

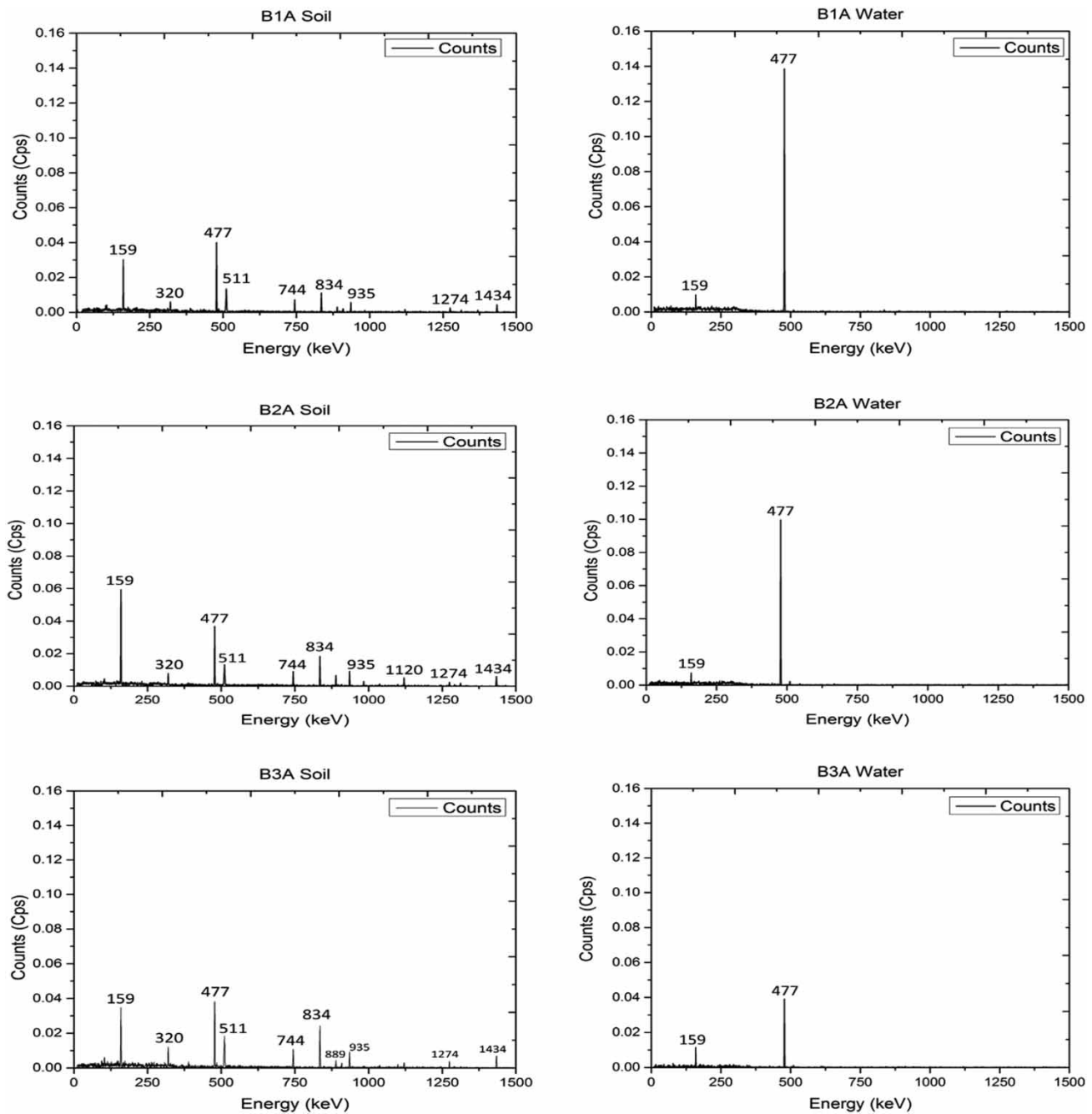


Figure 8 | Irradiated soil and water samples analysed 1-week post bombardment for the three boreholes. The graphs are skewed.

a half-life of 83.8 days (Gritzay *et al.* 2002; Firestone 1996). The formation of radioactive ^{46}Sc indicates the presence of a titanium element in the soil samples. Titanium has 5 naturally occurring isotopes (Audi & Wapstra 1993). The formation of ^{46}Sc distinguishes which of the titanium isotopes is present in the study area and prone to neutron activation. The results suggest that the neutrons from the envisioned LERIBs demonstrator might activate a stable isotope of ^{46}Ti to form ^{46}Sc .

MIGRATION OF ACTIVATED ISOTOPES WITH GROUNDWATER FLOW

The soil and groundwater containing radioactive material from iThemba LABS can be eroded and migrate to the nearby Kuilsriver and Eersterivier basins. A study by Valković (2019) suggests that water serves as the major transport medium of

the U-series nuclides. The measured short half-life and long half-life radionuclides in the study area can be expected to be transported through migration in groundwater as is the case with the U-series nuclides. The physical soil properties such as porosity and permeability might allow activated matter to seep through and travel distances with flowing groundwater. The groundwater flow can be explained using Darcy's law. Darcy's law states that the velocity is directly proportional to the permeability of the aquifer and the hydraulic gradient (Todd *et al.* 1976). Typical groundwater flow velocities fall in the range of 1.5 m per year to 1.5 m a day depending on the permeability of the aquifer (Todd *et al.* 1976).

Darcy's Law was used to describe the flow of groundwater flow as follows:

The first step for the calculations would be to find the hydraulic gradient, which represents the slope of the water table and was calculated as follows:

$$i = \Delta h / \Delta L \quad (1)$$

where h = difference in water elevation between two wells; L = distance between the two wells (Table 2).

The calculations for the hydraulic gradient were as follows:

$$\begin{aligned} \text{Hydraulic gradient } (i) &= -\Delta h / \Delta L \\ &= -\frac{h_2 - h_1}{L} \\ &= -\frac{12.24 \text{ m} - 12.8 \text{ m}}{233.15 \text{ m}} \\ &= -0.0024 \end{aligned}$$

Based on the measured hydraulic gradient in the study area, it was observed that the water flows from the northern side (high hydraulic head) to the southern side of iThemba LABS facility (low hydraulic head) with a hydraulic gradient of -0.0024 (Figure 9). The measurements correspond to those of (Meyer 2002).

The following step was to determine the flow velocity of groundwater in the area. The groundwater velocity will assist in anticipating the distance of the activated matter likely to migrate in the subsurface. A geological report conducted by Kantey & Templer 2008 suggested that the study area is composed of coarse sand material. According to the order of magnitude of K (hydraulic conductivity) for different kinds of rocks (Bouwer 1978), the coarse sand material has a K with an order of $2 \times 10^1 - 10^2 \text{ m/day}$.

To calculate the groundwater velocity, the porosity (n) and hydraulic flux (q) were considered using the following formulas:

$$\begin{aligned} \text{Hydraulic flux } (q) &= -K_i \\ &= -100 \text{ m/day} * (-0.0024) \\ &= 0.24 \text{ m/day} \end{aligned} \quad (2)$$

and

$$\begin{aligned} v &= q/n \\ &= 0.24/0.3 \\ &= 0.8 \text{ m/day} \end{aligned} \quad (3)$$

Table 2 | Water levels measured from two wells at iThemba LABS

	B1B	B3B
Water level (Δh)	12.8 m	12.24 m
Horizontal distance (ΔL)	233.15 m	

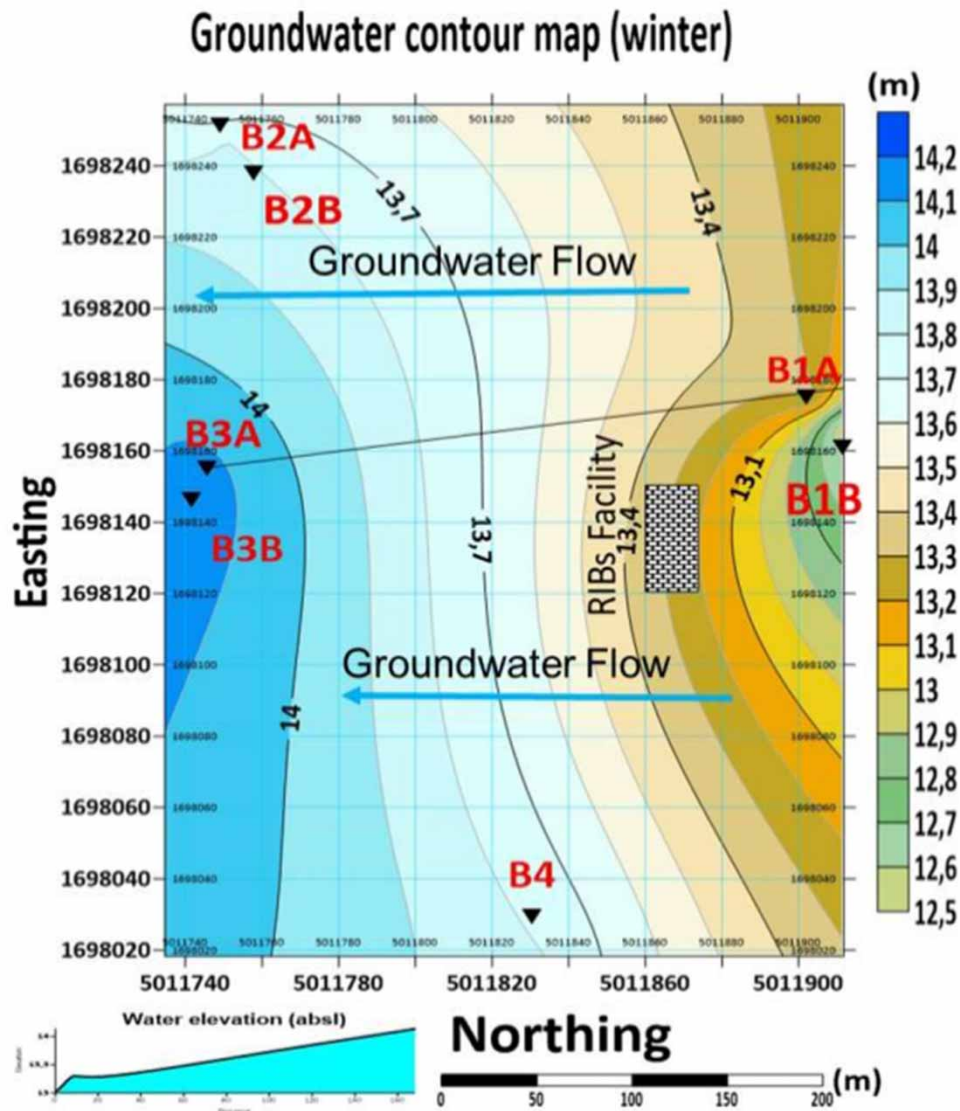


Figure 9 | Groundwater contour map showing the groundwater flow at iThemba LABS.

Based on the calculations using Equation (4), it can be concluded that the velocity of groundwater in the study area is around 0.8 m/day. The measured average distance of the monitoring boreholes located down gradient from the envisioned LERIB facility was measured to be about 167 m. Based on these measurements, it is anticipated that the radionuclide formed in water, which is ^{24}Na , will not migrate a distance exiting the study area. This is because ^{24}Na has a half-life of 15 h and therefore will decay before exiting the study area. Therefore, the facility should only put measures to protect the groundwater from irradiation within the site of iThemba LABS in compliance with section 24 of the Constitution and the National Environmental Management Act 108 of 1998. All radionuclides measured 1-week post activation were within the background radiation concentrations.

CONCLUSION

The soil and groundwater samples were activated in the radiotherapy vault at iThemba LABS with neutrons at an energy of 0–66 MeV. The HPGe detector was used to measure the gamma spectroscopy of the activated isotopes in the soil and groundwater samples. The results revealed that some of the stable isotopes will undergo activation when interacting with neutron radiation from the envisioned LERIB facility. However, only the isotopes of major elements were activated from both soil

and groundwater samples. The activated isotopes in soil samples included ^{22}Na , ^{24}Na , ^{52}Mn , ^{54}Mn , and ^{46}Sc . Only ^{24}Na occurred in groundwater.

It is therefore anticipated that isotopes of major elements in the subsurface are prone to activation from which short and long-lived radionuclides can be expected. The coarse sand material in the subsurface reduces the hydraulic flux resulting in slow groundwater velocity of 0.8 m/day. Based on the groundwater velocity, it is anticipated that the only radionuclide formed in water, which is ^{24}Na , will not migrate a distance exiting the study area. The ^{24}Na radionuclide will decay before exiting the study area. Therefore, the facility should put measures to protect the groundwater from irradiation within the site of iThemba LABS facility, as no impacts on down gradient can be expected. The long-lived isotopes in soil samples have shown background concentration readings. Due to the long half-lives of such radionuclides, they can be expected to migrate and exit the facility. However, these radionuclides might pose minimal threats to the soil and groundwater as they are occurring at background concentration levels. Due to the dangers of exposure to ionising radiation, it is recommended that proper shielding from neutron radiation be considered. The results of this study contributed valuable information to the understanding of the environmental impact of radionuclide production facilities. The findings have implications for risk assessment, environmental monitoring, and the development of remediation strategies. For environmental safety practises it is recommended that a thick shielding concrete should be built to minimise the amount of neutron radiation that passes through the shielding material.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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