

**THE ACTIVATION OF STABLE ISOTOPES, THEIR MIGRATION IN SOIL AND
GROUNDWATER AT A RADIONUCLIDE PRODUCTION FACILITY: A CASE
STUDY IN FAURE, CAPE TOWN, SOUTH AFRICA**

By

Thandazile Marazula

A DISSERTATION SUBMITTED TO THE CAPE PENINSULA
UNIVERSITY OF TECHNOLOGY IN PARTIAL FULFILMENT OF THE
REQUIREMENTS OF MASTER OF ENVIRONMENTAL
MANAGEMENT IN THE FACULTY OF APPLIED SCIENCES AT THE
CAPE PENINSULA UNIVERSITY OF TECHNOLOGY

Supervisor: Dr N. M. Malaza

Co-Supervisor: Dr J. L. Conradie

Cape Town Campus

Date Submitted: February 2018

CPUT copyright information

The dissertation may not be published either in part (in scholarly, scientific or technical journals), or as a whole (as a monograph), unless permission has been obtained from the University

Declaration

I, Thandazile Marazula, hereby declare that the work contained in this dissertation is my own original work that all sources used or quoted, have been indicated and acknowledged by means of complete references, and that this dissertation was not previously submitted by me or any other person at any other university for a degree awarded.

Signature:.....

Date.....February 2018

Abstract

The iThemba Laboratory for Accelerator Based Sciences (iThemba LABS or iTL), a facility governed by the National Research Foundation (NRF) based in Cape Town has embarked on a project called the South African Isotope Facility (SAIF). Part of the SAIF will be a project called the Low Energy Radioactive Ion Beam (LERIB) for the production of radioactive beams with the Separated Sector Cyclotron (SSC). The LERIB project is anticipated to generate significant prompt sources of ionising radiation such as neutrons displaced by protons during bombardment of targets.

At present, the facility has 3 - 5 m thick concrete walls designed to shield against 200 MeV neutrons. The floor thickness varies from each vault according to the behaviour and characteristics of the produced radiation. The LERIB facility concrete floor is envisioned to be 1 m thick. Insufficient floor shielding might expose the sub-surface strata below the floor with ionising radiation. Some matter in the exposed strata might undergo activation and consequently produce undesirable radioactive isotopes.

Exposure to radioactive isotopes can pose potential risks to the general public and the receiving environment. In the sub-surface, the activated isotopes can spread through the soil and groundwater bodies. Groundwater being mobile in nature might act as conduit for activated isotopes to migrate in the subsurface. The half-lives of the formed radionuclides were used to estimate the distance the radionuclides can migrate in the sub-surface before decay.

The X-Ray Fluorescence (XRF) and Inductive Coupled Plasma - Mass Spectrometry (ICP-MS) were used to quantify the mass fraction of elements from soil and groundwater samples. The Inductive Coupled Plasma - Optical Emission Spectrometry (ICP-OES) was used to measure dissolved metals of major and minor oxides in groundwater samples. These scientific techniques were applied in order to measure mass fractions of elements present in the sub-surface anticipated to be exposed with neutron radiation. Results revealed that the sub-surface in the study site constitutes Si, Al, Na and Fe in abundance. This suggests that, these elements might have a high probability to undergo activation compared to those occurring at

lower concentrations, such as minor and trace elements. However the neutron capture cross-section for these elements varies and hence some elements such as Si and Al were not activated.

To investigate elements measured from the ICP-MS, ICP-OES and XRF which might undergo activation, soil and water samples were irradiated for an hour with neutrons from a Beryllium target from a range of 0 – 66 MeV energy.

The results revealed that activating the sub-surface with neutron radiation will result to the formation of short and long-lived unstable isotopes. However, isotopes with long half-lives such as ^{22}Na , ^{52}Mn , ^{54}Mn and ^{46}Sc only occurred in soil samples and none measured from the water samples except a short-lived isotope ^{24}Na . These results agrees with the reviewed literature that, water being a product of two hydrogens and one oxygen, with ^1H from H_2O considered to be a good neutron moderator should have absorbed the neutrons so as to reduce neutron capture cross-sections. Hence only sodium stable isotope was activated in water as it appeared to be the abundant element.

Due to the short half-life of ^{24}Na (15 hours) in water, it is expected that this radionuclide will decay before migrating a distance escaping the study area. However, the solubility nature of sodium might result in the dissolution of radioactive ^{24}Na and ^{22}Na from soil and through dissolution; radioactivity can be introduced into groundwater bodies. All the long-lived radionuclides were measured at back ground radiation levels, thus insignificant environmental impacts in the sub-surface are anticipated. However, for environmental safety of the sub-surface, measures such as providing a sufficient concrete shielding to attenuate neutron energies are recommended.

Acknowledgements

This Dissertation would not have been of success without the presence of God in my life and for that reason I give glory to Jesus Christ to whom we have our being.

I thank Dr N Malaza, my supervisor who prioritised my thesis from the beginning and proofread my work. You made this work your own.

To Dr J. L Conradie, I thank you for having faith in me and for your willingness to help me whenever I needed assistance. I am deeply so grateful to have worked with you.

My gratitude goes to Mr P Beukes who generously volunteered to help me in ensuring that this thesis is a success and submitted in time.

Thanks to the medical radiation team for ensuring that the radiotherapy vault is set up and ready to provide neutrons for activation purposes.

I would like to thank Dr P Maleka who from the beginning of this Thesis gave advises with the relevant techniques to be applied to ensure that representative results are obtained.

Finally, I would like to thank NRF/iThemba LABS for funding my research.

Dedication

To Mr Jonguxolo Marazula, Ms Buseka Zwelibanzi and Mrs Nosiseko Marazula, my parents whom with the little they had but ensured that I get education.

My brothers and sisters for their support and non-ceasing prayers.

My daughter Yolanda Gaqisa.

Table of Contents

Declaration.....	i
Abstract.....	ii
Acknowledgements.....	iv
Dedication.....	v
List of Figures	x
List of Tables	xii
Glossary.....	xiii
CHAPTER ONE	1
INTRODUCTION.....	1
1.1 Location and description of the study area	1
1.2 Research problem	2
1.3 Research question.....	3
1.4 Aim and Objectives.....	4
1.5 Climate and Vegetation	4
1.6 Geology and Geohydrology	5
1.7 Motivation of research	6
1.7 Research Outline	7
CHAPTER TWO.....	8
BACKGROUND AND LITERATURE REVIEW	8

2.1 Background	8
2.2. Literature Review	10
2.2.1 Isotopes of rock forming elements	10
2.2.1.1 Isotopes of Major elements	11
2.2.1.2 Isotopes of Minor elements and their natural abundances	12
2.2.2 Activation of stable isotopes	13
2.2.3 Types and Sources of Radiation	16
2.4 Soil and Groundwater Processes	18
2.4.1 Soil Processes	18
2.4.2 Groundwater Processes	19
2.5 Radioisotopes Produced at iThemba LABS	21
2.6 The interaction of matter with radiation	23
2.7 The behaviour of radionuclides in the environment	23
2.8 Safety in radionuclides facilities	24
2.9 National and International Radiation Protection Standards	25
CHAPTER THREE	27
RESEARCH DESIGN AND METHODOLOGY	27
3.1 Introduction	27
3.2 Literature review and data collection	27
3.3 Fieldwork	28

3.3.1 Borehole Drilling	28
3.3.2 Borehole Construction	29
3.3.3 Measuring of the Water Table.....	31
3.4 Sampling.....	31
3.4.1 Soil Sampling.....	31
3.4.2 Groundwater Sampling.....	32
3.5 Physiochemical Analysis	33
3.5.1 Titration using a 905 Titrande	33
3.5.2 ICP-OES.....	33
3.5.3 Chemical Auto-Analyser Technique.....	34
3.6 Geochemical Analysis	35
3.6.1 X-Ray Fluorescence Analysis.....	35
3.6.2 Inductively Coupled Plasma – Mass Spectrometer (ICP-MS).....	36
3.6.3 Instrumental Neutron Activation Analysis (INAA).....	37
CHAPTER 4: RESULTS AND DISCUSSIONS	39
4.1 Introduction.....	39
4.2 Geochemistry of soil	40
4.3 Hydrochemistry of groundwater	46
4.4 Activation of stable isotopes in soil and groundwater	50
4.4.1 Measured background radiation in soil and groundwater.....	50

4.4.3 Samples measured one hour post activation	52
4.4.4 Samples measured one day post activation.....	54
4.4.5 Samples measured one week post activation	55
4.5 The migration of activated isotopes in groundwater.....	58
Chapter 5: Conclusion and Recommendations.....	64
5.1 Conclusion.....	64
5.2 Recommendations.....	65
References.....	66
Appendices.....	74

List of Figures

Figure 1.1. iThemba Laboratory for Accelerator Based Sciences site and the surrounding areas.....	1
Figure 2.1. Relative abundance of elements in the earth's upper crust as a function of atomic numbers.....	9
Figure 3.1. Position of the LERIB Facility at iTL and the drilled boreholes.....	28
Figure 3.2. Photographs taken during borehole constructions.....	30
Figure 3.3. Photographs of samples packed and stored in the ERL laboratory.....	32
Figure 3.4. HPGe detector set-up.....	37
Figure 4.1. FLUKA simulations predicting neutrons penetrating the LERIB shielding material.....	40
Figure 4.2. The average percentage of major elements at depths, 2-3 and 3-4 m.....	42
Figure 4.3. The average percentage of minor elements at depths, 2-3 and 3-4 m.....	44
Figure 4.4. The average percentage of trace elements at depths, 2-3 and 3-4 m.....	45
Figure 4.5. Major elements in groundwater samples.....	47

Figure 4.6. Minor elements in groundwater samples.....	48
Figure 4.7. Trace elements in groundwater samples.....	49
Figure 4.8. Background radiation of soil and groundwater samples.....	51
Figure 4.9. Activated soil and groundwater and hour post bombardment.....	52
Figure 4.10. The activated soil and water samples one day post bombardment.....	55
Figure 4.11. Irradiated soil and water samples one week post bombardment.....	58
Figure 4.12. The measured groundwater table and flow direction (winter).....	60
Figure 4.13. The measured groundwater table and flow direction (summer).....	61
Figure 4.14. Conceptual model of groundwater flow.....	62

List of Tables

Table 2.1. Isotopic masses and natural abundances of rock forming major elements.....	11
Table 2.2. Isotopic masses and natural abundances of rock forming minor elements.....	12
Table 2.3. Radionuclides produced at iThemba LABS for a variety of uses.....	22
Table 3.1. Parameters of the drilled six boreholes and existing B4.....	28

Glossary

<u>Abbreviation</u>	<u>Description</u>
CFA	Cape Flats Aquifer
CBD	Central Business District
DWAF	Department of Water Affairs and Forestry
ERL	Environmental Radioactive Laboratory
eV	electron volts
GPS	Global Positioning System
HPGe	High-Purity Germanium
ICP-OES	Inductively Coupled Plasma – Optical Emission Spectrometry
ICPMS	Inductively Coupled Plasma-mass Spectrometry
IAEA	International Atomic Energy Agency
iTL	iThemba LABS
INAA	Instrumental Neutron Activation Analysis
keV	kilo electron Volts
mabsl	meters above sea level
MCA	Multi-channel Analyser
mbgl	meters below ground level
NCRP	National Council on Radiation Protection
MeV	Mega electron Volts

NAC	National Accelerator Centre
NEMA	National Environmental Management Act
NORM	Natural Occurring Radioactive Material
NRF	National Research Foundation
PVC	Polyvinyl Chloride
LERIB	Low Energy Radioactive Ion Beam
SSC	Separated Sector Cyclotron
SPC	Solid pole Cyclotron
SAIF	South African Isotope Facility
TMG	Table Mountain Group
TENORM	Technology Enhanced Natural Occurring Radioactive Material
WHO	World Health Organisation
XRF	X-ray Fluorescence

CHAPTER ONE

INTRODUCTION

1.1 Location and description of the study area

The iThemba Laboratory for Accelerator Based Sciences (iThemba LABS or iTL) is situated in Faure, a small location, about 30 km from the City of Cape Town. The facility has been in operation since the 1960s, using the Van De Graaff machine for the acceleration of sub atomic particles. The study area is bounded by the national and local roads including the N2 to the South, the Baden Powell Drive (R310) to the East, the Old Faure Road to the North and the Spine Road (M32) to the West (Figure 1.1).

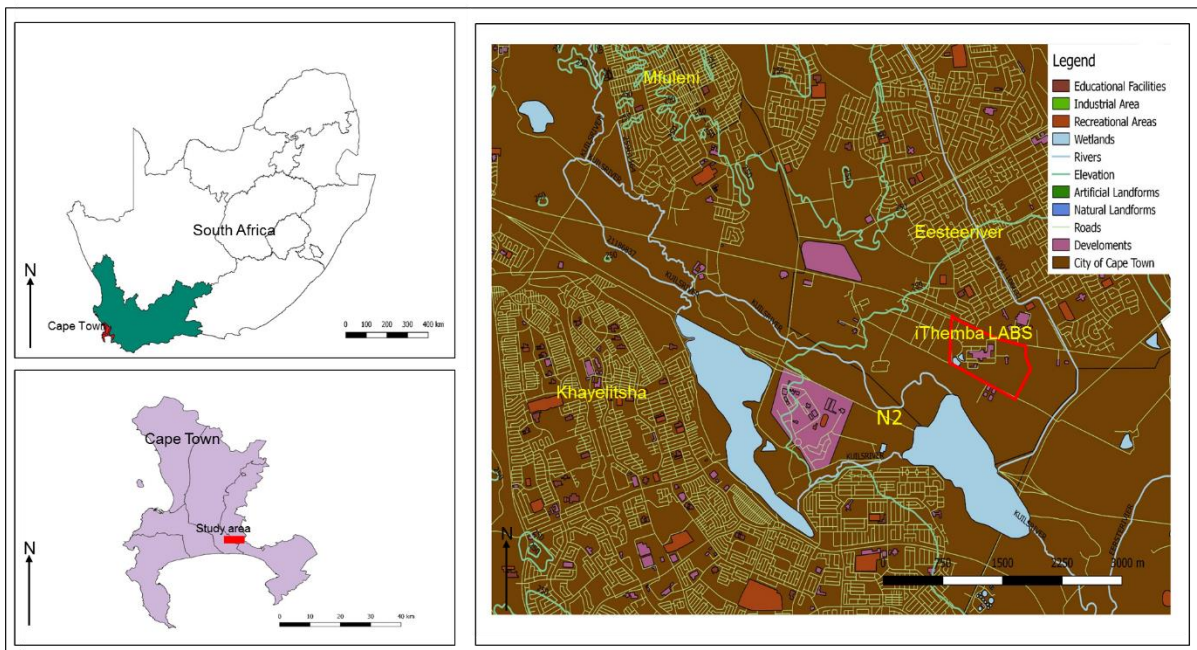


Figure 1.1. The iThemba Laboratory for Accelerator Based Sciences site based in Cape Town.

The study area is characterised by a relatively gentle topography and is densely vegetated over a large area by indigenous and alien plant species such as Port Jackson, Eucalyptus trees and Rooikrans (Hutton-Squire *et al.*, 1992). In the eastern side of iTL in a North-West to South-East direction, the Kuils River System runs

through and discharges into False Bay (South Atlantic Ocean).

The iTL facility provides services for basic and applied research using particle beams, hadron radiotherapy for the treatment of cancer and supply of accelerator-produced radionuclides for nuclear medicine. The iTL facility consists of different facilities responsible for the provision of scientific based services for academics and industries both at national and international level. Such facilities, include a K=200 MeV Separated-Sector Cyclotron (SSC) and a K= 8 MeV Solid-pole Cyclotron (SPC1) as light-ion injector and second injector Cyclotron (SPC2) which is responsible for pre-acceleration of heavy and polarized ions. The accelerators are used for the production of particle beams which are required for radiotherapy, radionuclide production and nuclear physics.

1.2 Research problem

Naturally formed isotopes are found in air, water, soil and the entire environment. The isotopes can be stable or unstable (radioactive) depending on their nuclear structures. Naturally formed isotopes have existed since the formation of the earth and are part of the building blocks of the surrounding environment. Isotopes can be artificially made through accelerator based applications.

In, South Africa, iTL is an example of such facilities and is a group of multi-disciplinary research laboratories administered by the National Research Foundation (NRF). As mentioned from section 1.1, the iTL provides facilities for, basic and applied research using particle beams, supply of accelerator-produced radioactive isotopes for nuclear medicine and research. In order to improve these services and to meet the research and production demands, the iTL has embarked on a project called South African Isotope Facility (SAIF). Part of the SAIF project will be a project called the Low Energy Radioactive Ion Beam (LERIB) facility for the production of radioactive beams with the Separated Sector Cyclotron (SSC).

Through the process of producing radioactive beams and radionuclides, significant sources of ionising radiation will be generated. At present, neutrons of 200 MeV are shielded by 3 - 5 m thick concrete walls from the vaults. The floor thickness varies

from the vaults according to the behaviour and characteristics of the generated radiation. The 1 m thick concrete floor from the LERIB facility might not sufficiently contain the envisaged ionising radiation. The resultant neutrons from ionising radiation can activate matter in the exposed strata of the sub-surface (soil and groundwater) and consequently produce undesirable radio-isotopes.

Inducing radioactivity in the sub-surface environment can pose potential environmental risks, particularly the underlying unconfined aquifer system. The fluctuating water table, particularly during wet seasons where it rises close to surface, might escalate chances for groundwater to interact with neutrons. The groundwater flow might act as conduit for the migration of the induced radionuclides, resulting into contamination of groundwater bodies on site and down gradient. Activation of the sub-surface will deteriorate the groundwater quality and pose negative effects on groundwater uses on site and communities adjacent to iTL.

For these reasons, it might not only be the radiation producing facility (iTTL) that might face contamination risks associated with ionising radiation but also the nearby surrounding environment. Hence it is important to sufficiently contain and shield ionising radiation to ensure the safety and protection of people and the environment, thus the study seeks to investigate the activation of stable isotopes and their migration in soil and groundwater.

1.3 Research question

- Would the neutrons generated from LERIB facility be able to activate stable isotopes in the sub-surface?
- What are the half-lives of the induced radionuclides in the study area?
- Would the activated isotopes in the subsurface migrate with groundwater out of the iTL site?
- Which isotopes of elements will stand more chances to undergo activation from soil and groundwater?
- What is the current radiation background level on-site of iTL?

1.4 Aim and Objectives

The purpose of this work is to assess the activation of stable isotopes and their migration in the sub-surface. For this purpose, the project investigates:

- The activation of stable isotopes beyond containment in groundwater taking into consideration the seasonal fluctuating water table in the confined and unconfined aquifer
- The distance and pathways the induced radionuclides can travel in the sub-surface with regard to their half-lives.
- The environmental risks the induced radionuclides might pose into soil and groundwater.
- Groundwater quality assessment and monitoring from ionising radiation.

1.5 Climate and Vegetation

South African climatic conditions are categorised as relatively dry with long extensive drought periods. The South African mean annual precipitation is estimated to be in the order of 500 mm compared to 860 mm of the world's average rainfall (Wu, 2008). The climatic conditions of the Western Cape Province are largely influenced by the topography (Diamond and Harris, 2000). Due to topographic influences and regional variations, the Cape Peninsula receives 500 mm - 1700 mm, Cape Flats receives 500 mm – 800 mm and the mountains receives 800 mm - 2600 mm per annum (Diamond and Harris, 2000).

The study area is based in Cape Town and the reviewed literature showed that a Mediterranean climate, with an average rainfall between 560 mm and 1400 mm is experienced. This climatic condition is associated with extreme weather periods experienced in summer and winter, with temperatures rising up to 42°C in summer and lower to 1°C in winter seasons (Tadross and Johnston, 2012).

The rainy months are in winter between April and September, when temperatures and evapotranspiration are at minimum. However, in summer, that is not the case,

as significantly higher transpiration than rainfall is experienced. These temperature variations are influenced by the surrounding sea, resulting into temperature increase to be more likely in the inland than towards the coasts (Tadross and Johnston, 2012). Due to the closeness of the study area to the sea, the area is exposed to low level sea breeze patterns, which are influenced by the Peninsula mountain chain and Hottentots-Holland Mountains (Hutton-Squire *et al.*, 1992). The study area is further influenced by the wind vectors which tend to come more from South-East in all seasons except June, July and August when dominant flow is from the west (Tadross and Johnston, 2012).

1.6 Geology and Geohydrology

Cape Town, comprises of four separate landscapes; (1) the sandy Cape Flats that lies in the centre of the city bordered on the western and southern coastal edges by the (2) dune-dominated strandveld, (3) the low shale and granite hills which historically been converted to farmlands that are found in the inland of the Cape Flats (4) the sandstone mountains of the Table Mountain chain and the Hottentots Holland, comprises the southwest and the east respectively (Rebelo *et al.*, 2010).

The larger portion of these landscapes is underlain by the oldest geological formations that belong to the Malmersbury Group, with about 550 Ma when the Peninsula Granite batholith intruded these rocks (Provincial Government of the Western Cape, 2005). The Malmesbury Shale landscapes and intrusive granites of the Cape Granite suites occur below the sandstone belt, in the north-east of the City (Rebelo *et al.*, 2010). The dune landscape of younger, alkaline, aeolian sands is covered with Cape Flats Dune Strandveld along the coastal areas.

The landscapes are further characterised by the geology, consist of rocks having pore spaces and fractures within, which helps in determining the geohydrological makeup of the region. Geological formations with high porosity and permeability are likely to yield exploitable amounts of water. Regions of high porosity and permeability are filled with water and are termed as saturated zone or phreatic zone, whereas regions filled with air and water are termed vadose or unsaturated zone.

According to the (Provincial Government of the Western Cape Department of Environmental Affairs and Planning, 2005) most of the Western Cape's geology comprise of sedimentary rocks that were formed from material that has over years been laid down by wind and water erosion. As a result, the abundant sedimentary rocks in the Western Cape area are shale, limestone, conglomerates and sandstone (Maclear, 1995). The study area consists of sandstones and shales of the Cape Super Group and Malmesbury Group, Cape Granites, younger sand, alluvial deposits and Tertiary Quaternary Formations. The tertiary Quaternary Formations are sandy in nature, containing pedogenic layers of calcrete of variable ferricrete strength (Kantey and Templer, 2008).

At a certain depth below surface, the meta-sedimentary strata of the Malmesbury Group with residual soils is developed over a localised area. The reviewed literature reveals that, the Malmesbury Shale intersect at depth comprising of shattered stiff clayey silt tending to completely highly weathered shattered laminated very soft rock Greywacke. Moreover, below ground level the area consists of weakly to strong cemented slightly sandy clay calcrete with pockets of hardpan calcrete (Kantey and Templer, 2008).

1.7 Motivation of research

Isotopes can be found everywhere in the environment at a stable and unstable state. For this research, the energy for the instability of isotopes is the ionising radiation from the envisioned LERIBLERIB facility, which is anticipated to penetrate through the concrete floor and interact with the soil and groundwater in the sub-surface environment. Isotopes formed in soil are believed to be stagnant at the place of formation due to the immobility nature of the soil. However, activated isotopes in groundwater are envisaged to migrate down gradient as groundwater flows. Thus this study seeks to investigate isotopes prone to activation from soil and groundwater. The isotopes formed from soil and groundwater will be identified so as to find isotopes with likelihood to migrate and not to migrate with groundwater. The migration of activated isotopes would disturb the integrity of groundwater quality on site and down gradient. Through this study, the migration of isotopes on site and distance out of the study area is identified.

1.7 Research Outline

The overall aim of this research was to investigate the activation of stable isotopes and their migration into soil and groundwater from a radionuclide production facility in Cape Town. When formulating this research, chapters containing relevant literature were designed to better understand processes involved in the activation of isotopes and the environmental conditions enabling the activated material to migrate in the sub-surface.

Chapter one provides the background information of the study area, through incorporating literature from the existing geological, geohydrological and climatological data. In chapter two a literature review on different aspects of the research is given. In chapter three, the methods applied to answer the research questions as well achieving the aims and objectives of this research are discussed. The final results and detailed discussion are given in chapter four. The conclusion of the overall study is discussed in chapter five and followed by the recommendations in chapter six. The final chapter are followed by references and appendices.

CHAPTER TWO

BACKGROUND AND LITERATURE REVIEW

2.1 Background

The quality of life for the general public depends on the chemical, physical and biological composition of the surrounding environment (Kabata-Pendias, 2011). It is, therefore important that the natural environment is protected from any form of pollution or contamination, so as to improve the quality environmental conditions. Global laws, policies and regulations have been developed to safeguard the security and sustainability of the natural environment from all kinds of pollutions. South Africa as part of the global community has developed its own regulations to govern the state of the environment through legal frameworks such as the environmental rights. The environmental rights are promoted by the South African Constitution (1996) and National Environmental Management Act (1998).

It is with respect to these legal frameworks that, the activation of stable isotopes is investigated and the migration of activated material in soil and groundwater be understood so as to implement mitigation measures where necessary. However to expand such an investigation, it is important to define the term “environment” as this term is applied in different fields of studies for different subjects. For the purpose of this research, the environment is defined as the surroundings within which humans and animals exist and that are made up of land, water and atmosphere of the earth (NEMA, 1998).

The land, water and atmosphere that make up the natural environment are composed of elements distributed at different concentrations in the earth’s upper crust. The elements that make up the earth’s upper crust are geologically categorised based on their abundances as major, minor, trace, rare earth, and rarest earth elements (Kabata-Pendias, 2013; Haxel *et al.*, 2002). The major oxides of metals (Si, Al, Fe, Mg, Mn, Ca, K, Na and Ti) are present in the earth’s upper continental crust at concentrations exceeding 1%, with minor elements occurring at a range of 0.1 – 1 %, trace, rare and rarest elements are usually <0.1% (Borradaile,

2003). Haxel *et al.* (2002), developed a diagram presenting major, minor, trace, rare earth and rarest earth metals in the basis of abundance (atom fraction) as a function of atomic number (Figure 2.1). Based on Haxel's (2002) diagram, the major and minor elements are the rock forming elements (Figure 2.1).

The soil composition is influenced by the parent material and its geochemical properties are associated to those of the underlying rock material (Kabata-Pendias, 2013). Due to the abundance of the rock forming elements in the upper continental crust, the soil in the study area can be expected to constitute high concentrations of some of these elements.

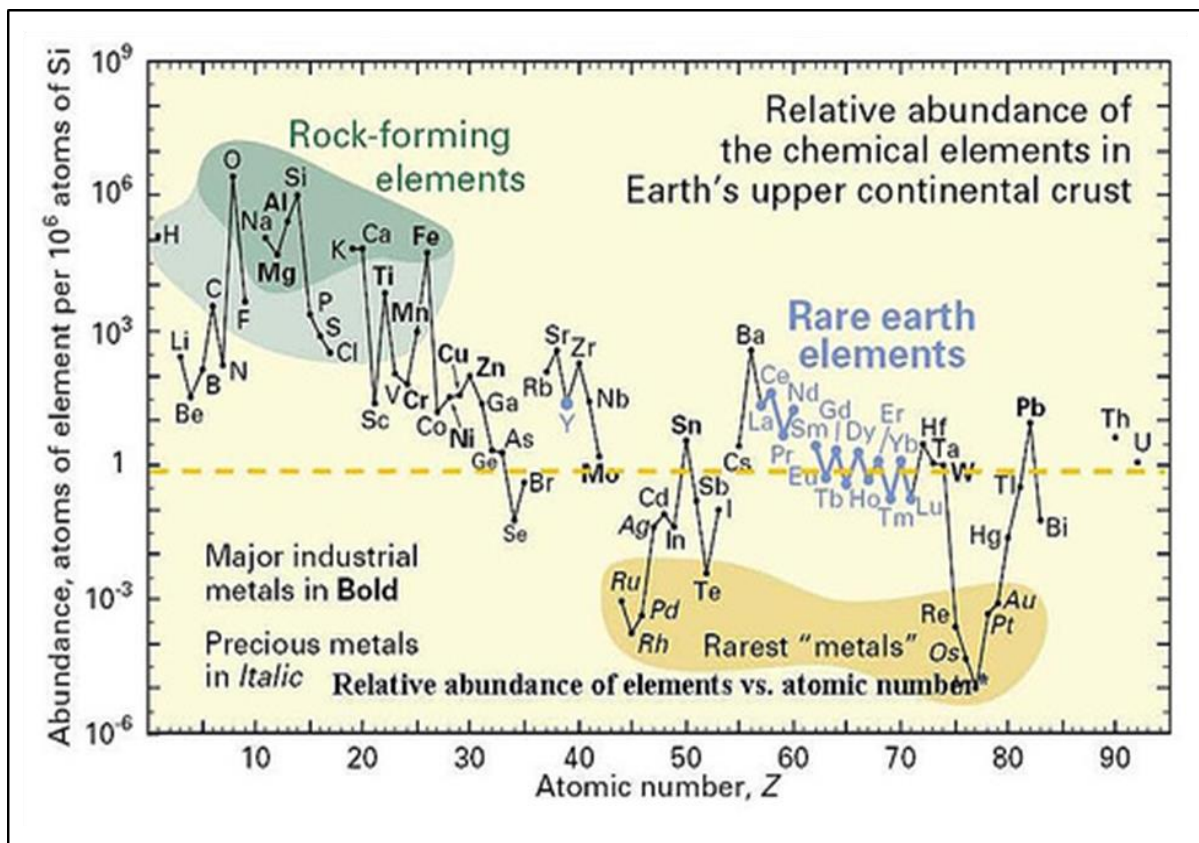


Figure 2.1. Relative abundance of elements in the earth's upper crust as a function of atomic numbers (Haxel *et al.*, 2002).

In the earth's crust, elements of lower atomic number are more abundant than elements with heavy nuclei (Figure 2.1). The elements in the earth's crust are associated with one or more isotopes at stable and unstable state (Firestone, 1996).

The driving force for nuclei stability and instability is the atomic nuclear reaction (Hailu *et al.*, 2012). There are a variety of nuclear factors involved in the induction of stable isotopes to radionuclides. For the purpose of this study the focus is based on the induction of radioactivity through neutron activation process.

2.2. Literature Review

2.2.1 Isotopes of rock forming elements

The isotopes of rock forming elements are largely found from igneous rocks such as peridotite, basalt, andesite, rhyolite and phonolite (Hibstie, 2012). However it is rare to find elements from the earth's crust as stand-alone, as elements sometimes react with one another when conditions favours for physio-chemical and other natural reactions to form minerals or other substances (Hailu *et al.*, 2012). Geological matters resulting from elemental reactions, particularly in clay minerals, are Aluminium Oxide (Al_2O_3) and Silicon Dioxide (SiO_2) formed through the reaction with oxygen (Glascoock *et al.*, 2004). According to Glascock *et al.* (2004), Al_2O_3 and SiO_2 are the main components of clay. On the other hand, the oxides of other elements such as Sodium Oxide (Na_2O), Magnesium Oxide (MgO), Potassium Oxide (K_2O), Calcium Oxide (CaO), Titanium (TiO_2) and Ferric Iron (Fe_2O_3) are found in clay minerals at small concentrations compared to (Al_2O_3) and (SiO_2) (Glascoock *et al.*, 2004).

The major oxides of elements in the earth's crust include: Na_2O , MgO , Al_2O_3 , SiO_2 , K_2O , CaO , TiO_2 and Fe_2O_3 (Haxel *et al.*, 2002). The soil being the product of the underlying geological materials, such as rocks, would be expected to constitute a degree of the percentage of the rock forming elements. The minor elements include: hydrogen, carbon, fluorine, phosphorus, sulphur, chlorine, titanium and manganese (Haxel *et al.*, 2002) (Figure 2.1). The trace, rare and rarest elements are presented in Figure 2.1, and occur in low concentrations in the earth's crust.

2.2.1.1 Isotopes of Major elements

There is a large variety in the isotopic abundance ratios of different elements (Audi *et al.*, 1993). Due to a series of nuclear processes, the isotopic composition of carbon as well as oxygen of calcium carbonate is different for the marine and the freshwater origin; secondly the isotopic composition of oxygen and hydrogen in rainwater varies with latitude, altitude, climate and time of the year (Mook, 2001). This implies that, the variation of isotopic composition from soil can be expected.

The variation of isotopic composition in the environment has also been found among rock forming elements where among major elements such as Sodium and Aluminium have only one naturally occurring stable isotope. The presence of other isotopes of Sodium and Aluminium in any environment beside ^{23}Na and ^{27}Al would be due to anthropogenic factors and such isotopes are expected to be unstable or radioactive. The other stable isotopes of major elements, has more than one natural occurring isotopes (Table 2.1).

Table 2.1. Isotopic masses and natural abundances of rock forming major elements (Audi and Wapstra, 1993; Rosman and Taylor, 1999).

Isotope	Atomic No.	Atomic mass	Natural abundance (%)
Oxygen	8	16	99.76
		17	0.04
		18	0.21
Magnesium	12	24	78.99
		25	10.00
		26	11.01
Silicon	14	28	92.2
		29	4.7
		30	3.1
Potassium	19	39	93.26
		40	0.01

		41	6.7
Calcium	20	40	96.94
		42	0.001
		43	0.14
		44	2.09
		46	0.004
		48	0.19
Iron	26	54	5.85
		56	91.75
		57	2.12
		58	0.28
Titanium	22	46	8.25
	22	47	7.44
	22	48	73.72
	22	49	5.41
	22	50	5.18

2.2.1.2 Isotopes of Minor elements and their natural abundances

Among the rock forming minor elements, Fluorine, Phosphorus and Manganese have only one naturally occurring isotopes. The remaining rock forming minor elements such as Hydrogen, Sulphur, Chlorine and Titanium are made up of more than one stable isotope (Table 2.2). According to Bacchi *et al.* (2002), elements that occur at low concentrations have low neutron absorption cross-sections in soil.

Table 2.2. Isotopic masses and natural abundances of rock forming minor elements (Audi and Wapstra, 1993; Rosman and Taylor, 1999).

Isotope	Atomic No.	Atomic mass	Natural abundance (%)
Hydrogen	1	1	99.9
Deuterium	1	2	0.0115
Tritium	1	3	Unknown
Sulphur	16	32	94.93
	16	33	0.76
	16	34	4.29
	16	36	0.02
Chlorine	17	35	75.78
	17	37	24.22

2.2.2 Activation of stable isotopes

Elements generally consist of atoms with a nucleus surrounded by a cloud of electrons (Van Rooyen, 2006). The atom's nucleus consists of two particles, neutrons and protons, which have about the same mass. These two particles (protons and neutrons) are the main building blocks of the nucleus and are called nucleons (Hailu *et al.*, 2012). Based on elemental nuclei, an element with an atomic nucleus containing different number of neutrons are called isotopes, in which many of the elements have two or more naturally occurring isotopes (Hailu *et al.*, 2012). From natural occurring isotopes, most isotopes of light elements have an equal number of protons and neutrons, while in heavy elements, neutrons exceeds the number of protons (Mook, 2001). In the basis of proton-neutron ratios, isotopic instabilities are greater in heavy elements. As a result, slight excess of neutrons for light elements does not necessarily result in unstable nuclei (Mook, 2001).

Nonetheless, in some instances, an isotope of an atomic mass (A) and atomic number (Z) when placed in a neutron source will, absorb a neutron to become an artificial radio-isotope (Hibstie, 2012). The formed radioisotopes will decay from an unstable state to a stable state by emitting gamma rays (Alfassi, 1990). Most of the

radionuclides formed by neutron activation decay by beta and gamma emission (Alfassi, 1990). When radioisotopes decay radiation will be released in the form of alphas, beta, gamma, neutrons and photons.

Neutron activation analysis is a method of quantitative chemical analysis that is based on nuclear activation of chemical elements present in the sample (Alfassi, 1990). The method can facilitate both quantitative and qualitative identification of elements present in the sample (Glascock, 2016). The activated sample (isotope) emits gamma rays at different energies, thus allowing the determination of nuclear material using attained energy spectroscopy (Clarke *et al.*, 2017). For example the amount of bromine in a sample is measured by the 616 keV gamma emission of ^{80}Br formed from $^{79}\text{Br} (n,\gamma) ^{80}\text{Br}$ (Alfassi, 1990).

The resultant radiation can be in a form of X-rays from the impingement of electrons on matter; gamma radiation from the impingement of ions or neutrons on matter; neutron radiation from the impingement of either electrons and lastly photons or ions on matter and delayed radiation such as beta and gamma rays from the induced radioactivity (NCRP, 1977). Among these types of radiation, the iTL produces radiation which includes alpha particles, beta particles, gamma-rays, X-rays, neutrons, high-speed electrons, high-speed protons and other particles capable of ionising matter.

The radiation type of concern in an environmental point of view is neutron based radiation as it is unlike charged particles, can penetrate through shielding or storage materials due to the lack of electrostatic interaction with the absorbing medium (Pynn, 2009). Neutron radiation exhibits a broad energy spectrum which can be classified as fast (≥ 1 MeV), slow (few eV), epithermal (energy from 0.025 eV) and thermal neutrons with energy distributed according to a Maxwell distribution around $3kT/2$ (0.025 eV at 300K) (Leroy, 2009). Among these neutron energies, thermal neutrons have a different and sometimes much larger effective neutron absorption cross-section for a given nuclide and can be readily absorbed by an atomic nucleus, creating a heavier, often unstable isotope of the chemical element (Byrne, 1994). This process is called neutron activation and through this process, radioactivity is induced.

The rock forming elements might stand more chances to undergo activation compared to trace and rare earth elements due to low concentrations of occurrence in the earth's upper crust (Borradaile, 2003). It is however important to note that the neutron absorption by nuclei depends on the energy of the neutron and the particular target nucleus. Furthermore, the probability of the neutron absorption is measured through the cross section of the reaction that, in general, for most of the elements present in soils, is very low (Bacchi *et al.*, 2002). As a result the neutron absorption occurs only with a few nuclei present in soils such as Ag, Au, In, Fe, Al, and Mn but in most soils these nuclei are present in very low concentrations (Bacchi *et al.*, 2002). These suggestions implies that, not only the neutron energy and nucleus of a target governs the neutron absorption, however the concentration of an element does influence the activation process, where elements of low concentrations will have a low probability to absorb a neutron.

The table of nuclides developed by Firestone (1996), illustrate that the unstable isotopes of major and minor elements formed through a neutron activation process, decay with beta- (β^-) decay mode, with most having very short half-lives (ranging from milliseconds to couple of minutes). Therefore, such nuclides may pose insignificant environmental effects as they may decay before migrating a distance with groundwater. The radionuclides with long half-lives formed from activating stable isotopes of major elements include the following:

- ^{24}Na (15 hrs)
- ^{28}Mg (21 hrs)
- ^{32}Si (152 yrs)
- ^{42}K (12, 4 hrs) ^{43}K (22.3 hrs)
- ^{45}Ca (162, 61 d), ^{47}Ca (4.5 d)
- ^{59}Fe (44.5 d) and ^{60}Fe (1.5×10^6).

These radionuclides would be of environmental concern as some might migrate with groundwater flow for distances exiting the study area.

2.2.3 Types and Sources of Radiation

Radiation has different types and sources. Some types of radiation are associated with damages in biological tissues while others poses insignificant effects (Sherer *et al.*, 2014). Some sources of radiation are considered to be natural due to prolonged existence in the environment, while others are man-made produced for specific purposes (Sherer *et al.*, 2014). Naturally occurring radioactivity is classified as either natural occurring radioactive material (NORM) or technology enhanced NORM (TENORM). NORM are found in varying amounts in soils, water, indoor and outdoor air and even within human bodies and exposure to them is inevitable (WHO, 2011). The NORMs include potassium - 40 and those of the thorium and uranium decay series such as radium – 226, radium – 228, uranium – 238 and lead – 210 (WHO, 2011). These radionuclides can be found in water due to natural or technology enhanced processes involving naturally occurring radioactive materials. The neutron radiation may naturally originate from the interactions of cosmic rays with the atmosphere and soils and from the radioactivity in the earth (Feige *et al.*, 1968). In the earth's crust, the neutrons are generated from (α , n) reactions that mainly involve alpha (α) decay of the uranium and thorium series impinging on the light elements, oxygen, sodium, magnesium and silicon (Feige *et al.*, 1968).

The TENORM may be present in water from several sources such as nuclear fuel cycle facilities, radiation based medicine and radiation released in the past into the environment. The NORM are enhanced through constantly bombarding the earth by high-energy particles (cosmic radiation) originating from the sun and outside the solar system (WHO, 2011). The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (2008), estimated that the global average annual dose per person from all sources of radiation is approximately 3.0 mSv/year. Where 80 % of this dose is due to naturally occurring sources, 19.6 % is from the use of radiation for medical diagnosis and 0.4 % is from other man-made sources.

The resultant radiation from either natural or anthropogenic source is classified into two types namely: ionising and non-ionising radiation (Leroy, 2009; Sherer *et al.*, 2014; Nilsson and Brahme, 2014). However, this study will focus mainly on ionising radiation due to its enough energy to liberate electrons from atoms or nucleus. The

ionizing radiation was discovered in 1895 by a German physicist, Wilhelm Conrad Roentgen and he named then “phenomenon X-rays”. Fairand (2002) described ionising radiation as a phenomenon limited to X-rays, gamma rays and high energy particles which are capable of removing low-energy electrons from atoms and molecules, thereby creating electrically charged atoms and molecules called ions. Based on radiation energy, Kudryashov (2008) argued that, photon radiation with energies 50 eV to 500 keV are termed Roentgen radiation, where radiation higher than 500 keV is considered to be gamma-radiation.

The NCRP (1977) report stated that the artificial radiation can be generated through the application of accelerator based techniques, where radiation is released whenever charged-particle beams are leading out of an evacuated region and interact with the target. A comparison made by the World Health Organisation (WHO, 2011) between man-made and natural occurring radionuclides in terms of risk management revealed that, man-made radionuclides are often controllable at the point of release. Whereas natural occurring radionuclides can potentially enter water bodies at any point resulting to natural occurring radionuclides to be less amenable to control. Both natural and artificial radionuclides undergo transitions which do not occur at the same time but randomly. According to Van Rooyen (2006), every radionuclide undergoes transition at a characteristic average rate, so that the rate at which the number of radionuclides in a given sample decreases is not the same for all radionuclides. The half-life can be used to quantify the rate at which radionuclides can undergo transition (Van Royen, 2006). Furthermore, the half-life of a radionuclide is the average time it takes for one half of the atoms of a collection of a specific radionuclide to undergo radioactive transition. Through the half-life of a radionuclide, the period a radionuclide might spend in the sub-surface before decay can be determined where short-lived radionuclides might live for short periods unlike the long-lived isotopes. Therefore distances for short-lived isotopes might be shorter compared to long-lived.

2.4 Soil and Groundwater Processes

2.4.1 Soil Processes

Soil is a natural body with both mineral and organic components in addition to its physical, chemical and biological properties (Kabata-Pendias, 2011). The soil composition varies horizontally and vertically depending on the region or environment. A study undertaken by Moura and Kroonenberg (1988) used the organic content as a measure for soil variation, where it was suggested that the carbon content decreases with depth. This was due to human activities which are concentrated closer to surface compared to the sub-surface. This implies that the soil layers composing the sub-surface of the study area might constitute a variety of elements increasing or decreasing with depth.

The soil diversity is mainly influenced by natural factors such as climate (temperature, rainfall), vegetation and other biota, parent material (minerals), topography (open or closed systems), time and the anthropogenic activities (degradation, contamination and remediation) (Kabata-Pendias, 2011). Importantly, the status of elements with time in soil will become different due to these factors especially the high practice of anthropogenic activities. As elements are inherited from their parent rock, their distribution within soil profiles and their partitioning between soil components reflect various pedogenic processes as well as the impact of external, especially the anthropogenic factors such as agricultural practices, industrial pollution and etc. Activating soil components by neutrons will form part of the external activities which affects the soil composition.

The soil is composed of three phases namely: solid (mineral and organic), liquid and gaseous phase. These phases significantly impact the distribution of chemical and physical properties in soil. Trace elements are minor components in the solid soil phase, however are important in soil fertility (Bradford *et al.*, 1996).

The soil forming processes are weathering and pedogenesis. The interaction of the lithosphere, atmosphere and hydrosphere within the biosphere powered by solar energy are the main components that drive the weathering process, which forms the

soil. There are driving chemical processes involved in the weathering process, which includes dissolution, hydration, hydrolysis, oxidation, reduction and carbonization are discussed below (Bradford *et al.*, 1996).

- Dissolution – Minerals are soluble in the aquatic phase
- Hydration – Minerals are increased their water content
- Hydrolysis – Reaction of minerals with water, producing new ions or insoluble components
- Oxidation – Incorporation of the oxygen into the chemical components or increase of the elemental potential
- Reduction – Reaction that is the reverse of oxidation reaction
- Carbonization – Alteration of compounds into carbonates due to the incorporation of Carbon Dioxide (CO₂)

These reactions in soil are controlled by chemical equilibrium in each particular earth surface environment. The processes lead to the formation of mineral and chemical components that are relatively stable in particular soil environments. Moreover, the processes involved in weathering together with processes involved in pedogenic lead to the formation of a soil profile.

Based on the chemical processes involved in the weathering of soil such as dissolution, the radionuclides formed in soil might dissolve and partition themselves into soil properties and migrate within the sub-surface through hydrolysis. The mobility of elements during weathering is determined by the stability of the host minerals and the electrochemical properties of elements (Kabata-Pendias, 2011).

2.4.2 Groundwater Processes

The sub-surface consists of an upper zone with soil-water and a lower saturated part with groundwater; these two zones are separated by the groundwater table which sometimes fluctuates. The depth at which groundwater is encountered depends on the regional topography, sub-surface structure and climate. The encountered groundwater is expected to have a specific storage, which can be determined by the porosity nature of an aquifer. However, the ease at which groundwater can flow depends on a combination of interconnection and size of the pores (Mook, 2001).

The water bearing rocks are either consolidated or unconsolidated. The consolidated materials consist of mineral particles of different sizes and shapes that have been brought together by heat, pressure and chemical reactions into solid body of rock (Cunningham and Schalk, 2011). The important water bearing consolidated materials include limestone, dolomite, shale, siltstone, sandstone and conglomerate (Cunningham and Schalk, 2011). This is due to the high porosity and permeability nature found mostly in alluvial sandy deposits and to a lesser extent in sandstone.

Therefore high yields and storage of water in such deposits would be expected. In clay deposits, that is not the case as clay minerals has small pores and consequently the flow resistance is considerable (Mook, 2001). Nonetheless the karstified limestone on the other hand can produce very high permeability due to dissolution features, and can even develop sub-surface streams.

Mudrocks and crystalline rocks are associated with poor aquifer characteristics. However, the weathering, fracturing and jointing can locally produce permeable structures so as to allow an ease for groundwater to be transported (Schalk, 2011). Groundwater can be recharged into the sub-surface through the process of infiltration and can be discharged by the sub-surface drainage through streams. High recharge promotes high storage which later results in a rise of the groundwater table and an increase of the hydraulic gradient, so that the discharge increases (Schalk, 2011).

In shallow drainage systems groundwater can reside between months but thousands of years for deeper systems. Due to such instability of groundwater residence time, the quality and quantity in the sub-surface is unevenly distributed (Cunningham and Schalk, 2011). Groundwater, unlike surface water has a relatively slow movement. Nearly every place on the earth surface is underlain by openings that are large enough to yield water in usable quantities to wells and springs (Heath, 1983). Due to this reason, groundwater can be considered as one of the most widely available natural resource.

The groundwater is likely to be found in saturated zones, stored in soil and rock aquifers in the sub-surface (Abdalla, 2009). Groundwater can occur close to the land

surface (unconfined aquifers) such as in marsh areas. In some instances groundwater can occur hundreds of meters below ground level (m bgl) such as in the arid regions (Abdalla, 2009). This implies that groundwater is not always easily accessible and might not be ready for use without treatment, especially for consumption purposes. Hence bore-hole drilling techniques should be applied when investigating and exploiting groundwater resources.

The difference in groundwater occurrence results to variation in vulnerability for contamination, meaning groundwater occurring in unconfined aquifers (close to surface) has high risks of contamination compared to groundwater occurring in deep confined aquifers (Musekiwa *et al.*, 2011). This is due to the closeness of the unconfined aquifers at surface. The low permeability nature of the confining media in confined aquifers shields the contaminants from seeping through and therefore protects the aquifer from contamination (Musekiwa *et al.*, 2011). Both confined and unconfined aquifers have the water table which is presented either permanent or seasonal, depending with the climatic conditions of the area. The water table can determine the time a contaminant such as radionuclides can travel before being in contact with the aquifer system (Aller *et al.*, 1987).

Radionuclides in the sub-surface can be introduced by the recharging water posing a likelihood of transporting isotopes vertically to the water table and migrate horizontally within the aquifer system (Aller *et al.*, 1987). However, the ability to disperse and dilute the transported and migrating contaminants depends in the quantity of water available in the sub-surface and the radiological properties of the nuclide (Aller *et al.*, 1987). The distribution and movement of groundwater is attributed to the availability of pore spaces and the ease for water to flow in between the geological materials.

2.5 Radioisotopes Produced at iThemba LABS

As discussed in chapter one, the iTL uses particle beams to produce radionuclides for pharmaceutical purposes (Table 2.2). The presence of these isotopes in the sub-surface (i.e soil and groundwater) will not have an impact on the detection or abundance of neutron activated environmental isotopes. However, enhanced

radioactivity is possible in the facility due to gamma-rays from radioactive waste storage facilities, where artificial radionuclides of ^{68}Ge (Germanium-68) and ^{22}Na (Na-22) were detected through the gamma-ray spectra (Hlatshwayo, 2007).

The concentration levels of these radionuclides are considered not to be high enough to result in overexposure of gamma-radiation to people and animals on site (Hlatshwayo, 2007). In 1999 at Brookhaven National Laboratory, ^{22}Na and ^3H were detected in groundwater and the findings showed that, these isotopes originated from the activated soil which was located adjacent the target station. Precipitation infiltrated the activated soil and carried the activated isotopes into the groundwater systems (Douglas *et al.*, 2008). The groundwater flow could transport the activated material for distances in the sub-surface

Table 2.3. Radionuclides produced at iThemba LABS for a variety of uses.

Radionuclide	Half-life	Nuclear Reaction	Main Use
^{18}F	1.83 hours	$^{15}\text{O}(p,n)^{18}\text{F}$	Glucose metabolic studies
^{67}Ga	78.3 hours	$\text{Zn}(p, xn)^{67}\text{Ga}$ $\text{Ge}(p, x)^{67}\text{Ga}$	^{67}Ga Localisation of certain tumours and inflammatory regions.
$^{81}\text{Rb}/^{81\text{m}}\text{Kr}$	4.58 hours	$\text{Kr}(p,xn)^{81}\text{Rb}$	Lung ventilation studies
^{123}I	13.2 hours	$^{127}\text{I}(p,5n)^{123}\text{I}$ $\text{Ag}(p,xn)^{109}\text{Cd}$	Thyroid studies localisation of certain tumours such as neuroblastoma.
$^{68}\text{Ge}/^{68}\text{Ga}$ generator	$^{68}\text{Ge}=271\text{ d}$ $^{68}\text{Ga}=68\text{ min}$	$^{69}\text{Ga}(p, 2n)^{68}\text{Ge}$	Typically detection of neuroendocrine tumours.
^{22}Na	2.602 years	$\text{Mg}(p,n)^{22}\text{Na}$	Used in material sciences-positron inhalation studies
^{82}Sr	25 days	$\text{Rb}(p,xn)^{82}\text{Sr}$	Used to manufacture $^{82}\text{Sr}/^{82}\text{Rb}$ generators.
^{109}Cd	453 days	$\text{Ag}(p, xn)^{109}\text{Cd}$	Non-medical application-calibration sources.

2.6 The interaction of matter with radiation

The discovery of X-rays and radioactivity, led to the understanding that radiation could cause changes in matter. For radiation to cause changes in matter, the composition of the irradiated matter and the amount of energy deposited by the radiation are key factors (Choppin *et al.*, 2013). All forms of radiation (charged particles, photons and neutral particles) interact with matter differently (Knoll, 2010). The actinides have the most hazardous radionuclides which may be released in different steps of nuclear fuel cycle and nuclear waste disposal. This is due to their low solubility and high sorption nature, which makes them move slower than the free groundwater due to the sorption-desorption equilibria (Choppin *et al.*, 2013). Therefore such radionuclides would settle for long time in the released environments. The radionuclides produced from nuclear tests, accidents and normal fuel cycle are mostly short lived, however the longer lived fission and activation products from such process are of concern to the general public if allowed to enter the environment.

Several heavy radionuclides are formed through the process of neutron capture reactions, such as ^{236}U . Nuclides with high activity have active geological and biological behaviour and can present significant radiation concerns to the environment. According to the Canadian Council of Ministers of the Environment (2007) the dispersion of nuclides in the environment is influenced by the texture of sediment and soil (grain size and surface area), biota characteristics (type, organ, age, root distribution) and seasonal changes.

2.7 The behaviour of radionuclides in the environment

The behaviour and migration of radionuclides in the environment can be influenced by many factors, when released to the environment. Because of its mobile nature, groundwater can act as conduit for the channelization and migration of radionuclides. The migration in the biosphere is through water transport path, where migration for long distances may only occur for radionuclides with long half-lives, solubility in groundwater and low sorption along the transport pathway (Choppin *et al.*, 2013).

The findings of Gordon (1992), confirms the conclusions of Choppin *et al.* (2013), where the author argued that uranium migration from the uranium deposits to the biosphere is due to groundwater leaching and physical-chemical conditions. Based on this, the geohydrology might influence the migration of radionuclides, as sandstone and permeable sedimentary rocks are most susceptible to leaching groundwater, due to their high porosity nature. Furthermore, the fractures and faults in impermeable rocks also serve as conduits for groundwater transport; therefore radionuclides can escape through fractures and faults (Choppin *et al.*, 2013). It is important to note that, the decay of radioactive isotope is independent of all external influences such as temperature, and pressure. Moreover, two isotopes of the same element are chemically identical and therefore chemical processes cannot change the ratio of two isotopes of the same element (Šraček and Zeman, 2004).

The water chemistry is sometimes associated with electric charged ions which have a tendency to interact with other electric charge based particles. However, as the neutron carries no net electric charge, there is no Coulomb repulsion to prevent its interaction with nuclei and it is thus able to cross the nuclear boundary even when moving at less than thermal velocity (Hughes, 1957). Based on the findings of Hughes (1957), it can therefore be argued that the electric charges carried by ions will therefore not deter neutrons from activating isotopes in the sub-surface (Bacchi *et al.*, 2002).

2.8 Safety in radionuclides facilities

Neutrons unlike electric charged particles can penetrate through shielding materials due to their lack of electrostatic interaction with the absorbing medium (Pynn, 2009). Depending on the type and source of radiation, the shielding material can be made of lead or thick concrete walls. In the case of concrete shielding, the majority of neutrons reaching the concrete walls can undergo elastic collisions with the nuclei of hydrogen and slow down neutron energies. Mook (2001) argued that the concrete can be considered as a good moderator not a strong absorber of neutrons. As the energetic neutrons may penetrate the insufficient concrete shielding material and contribute for the specific distribution of neutron energy in the receiving environment (Konefal *et al.*, 2012).

A study by Kudryavtsev *et al.* (2008) revealed that, neutrons are more penetrating than γ -rays and are a dangerous background radiation source. It is with this penetrating ability that neutrons can escape the shielding material and interact with matter to form new undesirable radio-isotopes in the receiving environment. Escaping neutrons would result to radiation exposure which is linked to health risks; hence radiation control measures should be implemented and regulated. The dosage above acceptable levels would be harmful to the general public and the entire physical environment hence it is important for people and the environment to be protected from excessive exposure (IAEA, 2004). The effects associated with exposing living tissue or the physical environment with ionising radiation depends on radiation intensity, energy, type and kind of the absorbing material (Lilley, 2001). The effects can be spread throughout the air, water and geohydrological systems. Adequate shielding and containment of sources of radiation are therefore prerequisites and forms part of the operating licencing conditions of authority holders. As a result, to manage and avoid escaping ionising radiation, irradiation facilities globally make use of concrete as means of shielding. However, the concrete thickness should be sufficient enough so as to effectively attenuate neutrons.

2.9 National and International Radiation Protection Standards

Both natural and anthropogenic exposure is controlled by legal regulatory authorities in a state. The National and International radiation protection standards provide legal guidelines in governing the use of sources of radiation and radionuclide sources. Some low level releases of radiation to the environment due to normal operations are allowed by the health authorities, who also monitor the limits or levels. The following are protection standards that are used worldwide:

- The Hazardous Substances Act No.15 of 1973 (Updated 1993).
- DLUG 91-1: Ionizing Radiation Dose Limits and Annual Limits on Intake of Radioactive Materials.
- WSCP 91-1: Code of Practice for the Management and Disposal of Non-Nuclear Radioactive Waste.
- IAEA SS 115: International Basic Safety Standards for Protection against

Ionizing Radiation and for the Safety of Radioactive Sources.

- IAEA TSR-1: Regulations for the Safe Transport of Radioactive Material, 2005 Edition

CHAPTER THREE

RESEARCH DESIGN AND METHODOLOGY

3.1 Introduction

This chapter outlines methods applied to answer the research questions as well as achieving the aim and objectives of this research. The research dealt with the activation of stable isotopes and their migration into soil and groundwater from a radionuclide production facility. The study followed a field and experimental research design approach and incorporated the following main methods:

- Comprehensive literature review and data collection.
- Fieldwork (understanding the geology of the area and borehole drilling).
- Sampling of soil and groundwater.
- Hydrochemical analysis using Titrand, Inductively Coupled Plasma – Optical Emission Spectrometry (ICP-OES) and Auto Analyser.
- Geochemical analysis using X-Ray Fluorescence and Inductively Coupled Plasma – Mass Spectrometer (ICP-MS).
- Instrumental Neutron Activation Analysis (INAA).

3.2 Literature review and data collection

A literature review from different scientific reports has been used in this study to provide relevant information about the activation of stable isotopes in the environment. The collected information has been analysed and incorporated into the study to answer the question of isotopic migration in the sub-surface. Nonetheless, the reviewed literature provided reasons for the need of this study in a radionuclide production facility with regard to the safety of the environment. The desktop study and data collection has involved a review of all relevant available information, including published and unpublished reports, both local and international.

3.3 Fieldwork

3.3.1 Borehole Drilling

The fieldwork was made successful through the drilling of six boreholes on-site. Borehole locations were selected using Google Earth, site layout topography and the Global Positioning System (GPS) Garmin GPS 60 CS to cover the proposed LERIB facility from all directions (Figure 3.1) and (Table 3.1). The drilling of boreholes was completed in a period of 2 weeks (10 March 2017 - 22 March 2017). The authorisation was granted by the Department of Water and Sanitation (Appendix 1).

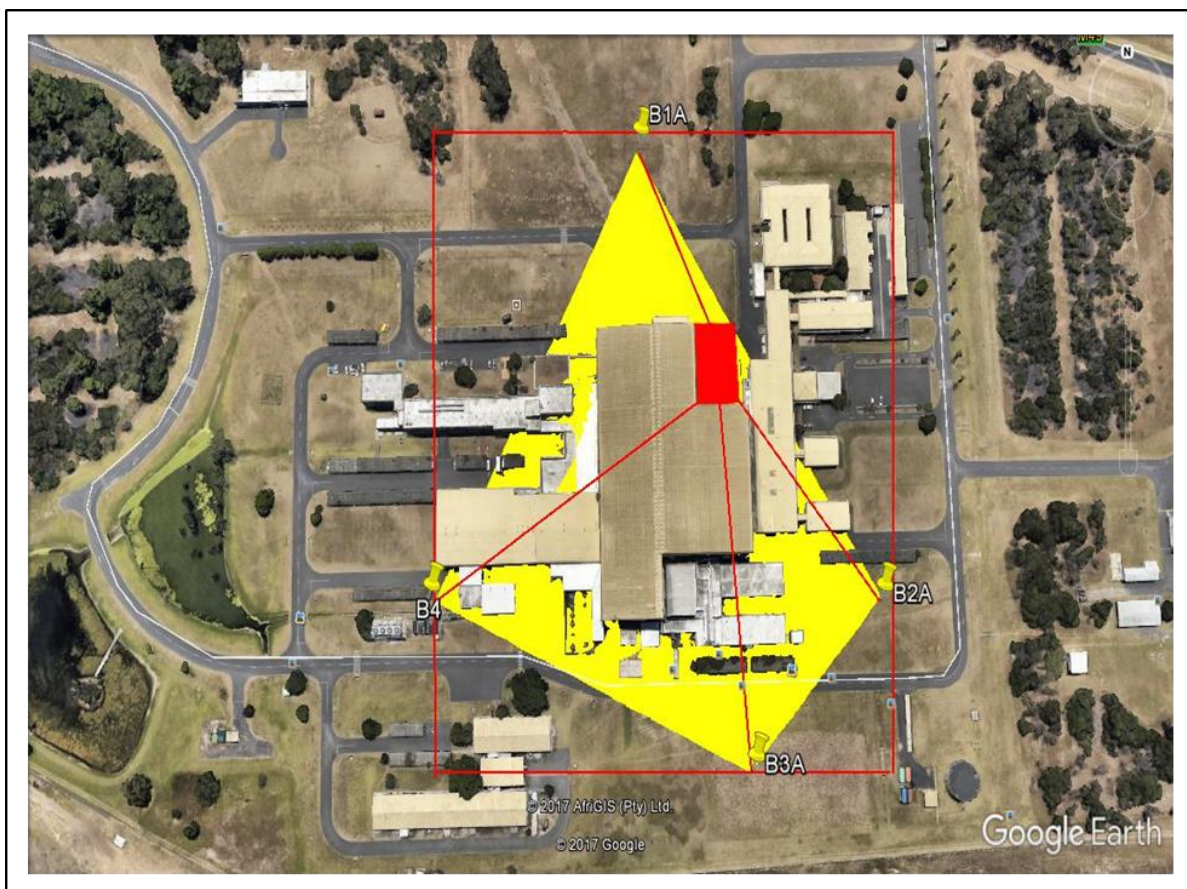


Figure 3.1. Position of the LERIB facility at iTL and the drilled boreholes (sampling points) (Modified from Google Earth, 2017).

Table 3.1. Parameters of the drilled six boreholes and existing B4.

Borehole ID	Coordinates		(Elevation (m))	Screening (m)	Steel Casing(m)
	Latitude	Longitude			
B1A	-34°01'25.5"	18°43'04.1"	15	7-15 m	N/A
B1B	-34°01'25.3"	18°43'03.8"	15	24-48 m	N/A
B2A	-34°01'32.6"	18°43'09.2"	14	7-15 m	N/A
B2B	-34°01'32.8"	18°43'08.8"	14	16-32 m	0-18m
B3A	-34°01'34.8"	18°43'06.2"	14	7-15 m	N/A
B3B	-34°01'35.1"	18°43'05.7"	14	24-48 m	0-33m
B4	-34°01'32.3"	18°42'59.3"	14	N/A	N/A

The depth, diameter, screening and casing of the six boreholes were differentiated. This was done to examine aquifer characteristics in confined and unconfined conditions. Three boreholes (B1A, B2A and B3A) were drilled to a depth of 15 m bgl to assess unconfined aquifer conditions. The other three boreholes (B2B, B3B and B1B) were drilled to a depth of 36 m, 45 m and 45 m bgl respectively for both unconfined and confined aquifer conditions (Table 3.1).

Two methods of drilling, namely mud rotary and odex drilling were applied interchangeable for the deeper (B1B, B2B and B3B) boreholes based on the depth and complexity of the geological conditions of the sub-surface. The mud rotary was only applied for the shallow (B1A, B2A and B3A) boreholes as the underlying geology was not complex to interrupt the process.

3.3.2 Borehole Construction

Borehole B1A, B1B, B2A and B3A were uncased due to their stability to withstand sand, clay and shale materials. A temporary casing of 177 mm diameter was installed into boreholes so as to avoid the walls from collapsing and to allow the drilling to continue smoothly inside the casing (Figure 3.2 (a)). Polyvinyl Chloride (PVC) pipes with a diameter of 125 mm were permanently installed in the boreholes (Figure 3.2). The PVC pipes were half screened and half unscreened so as to allow groundwater to seep through the screened portion on the pipes (Table 3.1).



Figure 3.2. Borehole construction: (a) drilling inside casing, (b) borehole pressurisation, (c) water depth quantification, (d) bore-hole closing, (e) generator to pressurise groundwater and (f) welding a permanent casing.

Only two boreholes (B2B and B3B) were permanently installed with 177 mm diameter steel casing for a depth of 0-18 m and 0-33 m bgl respectively (Table 3.1). Polyvinyl Chloride (PVC) pipes were installed inside steel casing for B2B and B3B, with diameters of 110 mm and 125 mm respectively. The boreholes were tightly

closed with PVC caps so as to avoid exposure from the outside environment (Figure 3.2).

3.3.3 Measuring of the Water Table

After completing borehole construction, groundwater levels were measured using a tape measure and a 2000 OPV Pocket Analogue Multimeter (Major Tech MT 800) (Figure 3.2). The multimeter was extended by an electric rod attached to the tape measure. A 26 cm long piece of steel was attached to the tape measure and an electric rod to create weight when the tape measure is lowered down into the borehole. The tape measure was lowered into the borehole to quantify the depth of groundwater after drilling. The recorded groundwater levels for each borehole were used in quantifying the groundwater table.

3.4 Sampling

3.4.1 Soil Sampling

Samples were collected at a depth of 2 - 3 and 3 - 4 metres from the recovered core of B1A, B2A and B3A, as the interface of the sub-surface with the shielding material is located along this stratum. These depths are anticipated to be the ones prone to activation. For sampling, disposable plastic spoons were used to collect and fill samples into 50 ml centrifuge tubes which were later stored into temperature controlled sample container boxes so as to prevent exposure from different weather conditions which could alter the sample's chemistry (Figure 3.2).

The collected soil samples were 12 in total and were marked according to borehole ID's and depths of collection i.e B1A (2 – 3 m), B1A (3 – 4 m), and etc. The Environmental Radioactive Laboratory (ERL) at iTL was used to store samples at controlled room temperature of 20°C until the drilling was completed. The shipment of samples to the laboratory was made immediately after completion of borehole drilling and construction.

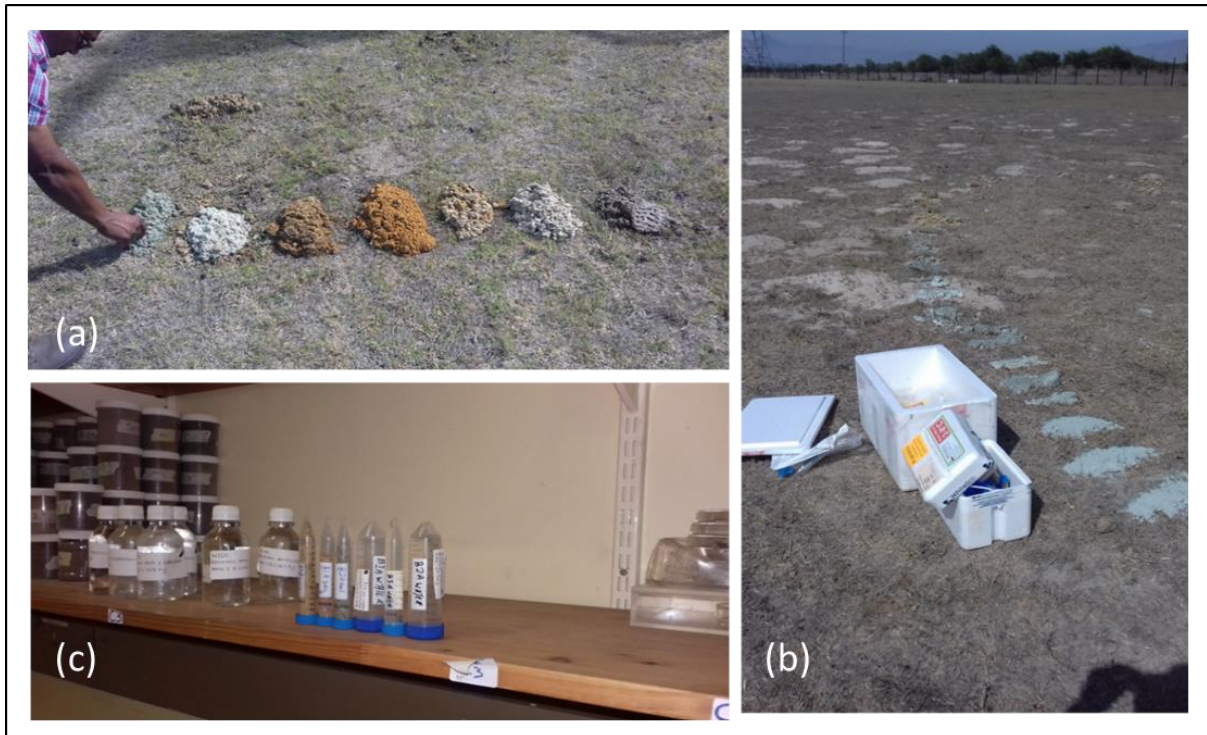


Figure 3.3. Soil and water samples stored at the ERL: (a) recovered lithological core from a drilled borehole, (b) two sample boxes used to store soil and water samples after collection and (c) sample store room at the ERL.

3.4.2 Groundwater Sampling

The mud-rotary and odex drilling techniques both encountered groundwater at depth and through the exerted pressure by the drill bits, the water came out from the drilled holes in large volumes, such that it was impossible to sample while drilling. The groundwater samples were only collected when boreholes were completely drilled and constructed (Figure 3.2 b). Each borehole was highly pressurised by the compressor generator for a period of an hour so as to validate the stability of the casing and to observe consistency of borehole yields. As the boreholes were tested for stability, the compressor was controlled so that the water can come out at controlled velocity. Samples were then collected using well-rinsed polyethylene bottles of 2 l and 50 ml centrifuge tubes without adding any preservatives (Figure 3.3 b). In total, there were 15 samples collected.

The sample containers were marked using black marker according to the borehole ID's (B1A, B2A, and etc). Samples were tightly closed to avoid losses and stored in a temperature controlled box so as to avoid exposure from external weather conditions and were separated from those of soil samples to avoid cross contamination. The collected samples were stored at the ERL until the drilling was completed. Only samples for physiochemical analysis were transported to the laboratory immediately after collection.

3.5 Physiochemical Analysis

Groundwater samples were shipped to the laboratory (Bemlab) on the same day for chemical analysis (Appendix 2, 5, 7 and 9). Three techniques of analysis were applied as discussed below:

3.5.1 Titration using a 905 Titrande

The pH, Electrical Conductivity (EL), Chloride (Cl^-), Total Dissolved Solids (TDS), Bicarbonate (HCO_3^-) were determined using a Titrande at Bemlab. The 100 ml of a sample was pipetted from a volumetric pipet into a 150 mL titration beaker. Thereafter, a magnetic stirrer was placed into the sample, pH and EC was measured. The total alkalinity of water sample was determined volumetrically by titrating it potentiometrically with a standard acid to a fixed pH and end point. For Chloride, 100 ml of sample was measured out into the titration beaker, treated with 5 mL HNO_3 and titrated with 0.02 M AgNO_3 after the first end point.

3.5.2 ICP-OES

The ICP-OES technique was used to measure the major and trace elements from groundwater samples at Bemlab. The measured major and heavy elements included Na^+ , Ca^{2+} , K^+ , Mg^{2+} , Fe, SO_4 and Mn, B, P, Cu, Zn respectively. The samples were analysed without dilution process and samples with higher elemental concentrations were measured. Sample solution was pumped into a high dissolved-solids tolerant nebulizer to produce an aerosol. The aerosol was subsequently transported by argon gas through a spray chamber and torch assembly into an inductively coupled plasma

source where the sample was desolvated, atomized, and the resultant atoms or ions excited. The intensity of light emission that resulted when the excited-state atoms or ions relax to their ground state was directly proportional to the concentration of the emitting species in solution. As a result, concentrations of elements were reported from three replicate determinations.

3.5.3 Chemical Auto-Analyser Technique

The $\text{NH}_4\text{-N}$ and NO_3 were determined using an Auto Analyser method which applies flow analysis (CFA and FIA) and spectrometric detection. For flow injection analysis, the sample was fed into a continuously flowing buffer solution by means of an injection valve or continuous flow analysis and continuously mixed with the buffer. The nitrate in the sample was reduced with metallic cadmium to nitrite. Thereafter, a phosphoric acid reagent solution was continuously added. The nitrite then diazotized sulphaniamide in acid solution to diazonium salt which was then coupled with N- (1-naphthyl) ethylenediamine to form a red dye.

A similar method is applied for $\text{NH}_4\text{-N}$, where in the pH range of most natural waters, ammonia nitrogen exists mainly as NH_4^+ . In water, there exist both unionised ammonia (NH_3) and ionised ammonium (NH_4^+). Both forms were easily converted into the other, with the ratio of ammonia to ammonium largely depending upon pH, salinity and temperature. The ammonium is predominant when the pH is below 8.75 and ammonia is predominant pH is above 9.75. The Total ammonia (TAN) is the sum of ammonium concentrations. The concentration of TAN, NH_4^+ and NH_3 are calculated based on the analytical result (obtained from the Auto Analyser) and the ammonia or ammonium ratio.

3.6 Geochemical Analysis

3.6.1 X-Ray Fluorescence Analysis

Soil samples were analysed at the University of Cape Town (UCT), Department of Geology for major and minor elemental analysis using the X-ray Fluorescence (XRF) technique (Appendix 12). To achieve this objective, a 20 g of each sample was put into ceramic crucibles and heated in a muffle furnace using a ramping temperature program, beginning at 100°C and rising to 500°C over a period of 4 hours. The sample was left at 500°C for 2 hours to ash all organic material. The ashed sample was milled in a carbon steel disk mill to a maximum particle size of approximately 20 µm. The 2 g of the milled sample was weighed out into a previously weighed ceramic crucible and placed in an oven at 110°C for a period of 8-16 hours. Once removed, the sample and crucible were weighed again and the weight loss calculated as a weight percentage “H₂O” (absorbed water weight). The crucible and the sample were placed into a muffle furnace and heated to 800°C for a period of 4 hours, cooled and weighed again. The weight difference was calculated as a percentage and designated as “loss of ignition (LOI)”. The LOI represents the weight percentage lost due to the loss of any structural volatilities (e.g. H₂O, CO₂).

A 0.7 g of a dried and ignited sample powder was mixed with 6 g of a flux composed of lithium metaborate and lithium tetraborate, so as to lower the melting temperature of the sample, allowing it to be melted over a gas burner. An automated fluxer was used to mix and homogenise the sample-flux mixture in a platinum crucible and the molten mixture was poured into a heated platinum mold, which cools slowly into a fusion disk. The fusion disk was used for major element measurements, calibrated against a series of well-characterised rock standards that have also been prepared as fusion disks. The fused disks broke down minerals into an amorphous glass, where matrix effects due to different sample mineralogy was eliminated, resulting into an increase in precision.

The mass of 6 g of an un-ignited sample powder was mixed with a drop of polyvinyl alcohol (as binder) and compacted into a cylindrical disk shape with steel die using

a 10 ton hydraulic press. The pressed pellets were used for minor element measurements by X-ray fluorescence spectrometry.

3.6.2 Inductively Coupled Plasma – Mass Spectrometer (ICP-MS)

Soil and groundwater samples collected in 50 mL centrifuge tubes were analysed at the UCT, Department of Geology for trace elements using the ICP-MS instrument (Appendix 13). The soil samples were ashed and dried as discussed in section 3.6.1. Groundwater samples were filtrated using syringe filters; polyethersulphone (PES) membrane, 0.22 µm pore size, 30 mm diameter. The ICP-MS: Xseries2 Thermo Fisher equipment, with an Argon gas as a carrier was used to measure samples for a period of 10 minutes.

A 50 mg of sample powder was dissolved in a 4:1 HF/HNO₃ acid mixture in sealed Savilex beakers on a hotplate for 48 hours, followed by evaporation to incipient dryness and two treatments of 2 mL concentrated HNO₃. The final dried product was taken up in 5 % HNO₃ solution containing 10 ppb Re, Rh, In and Bi, used as internal standards. Calibration curves were obtained using artificial multi-element standards, from which standard solutions were made.

3.6.3 Instrumental Neutron Activation Analysis (INAA)

Groundwater and soil samples were analysed from the ERL at iTL for the background radiation using the High-Purity Germanium (HPGe) detector (Figure 3.4)

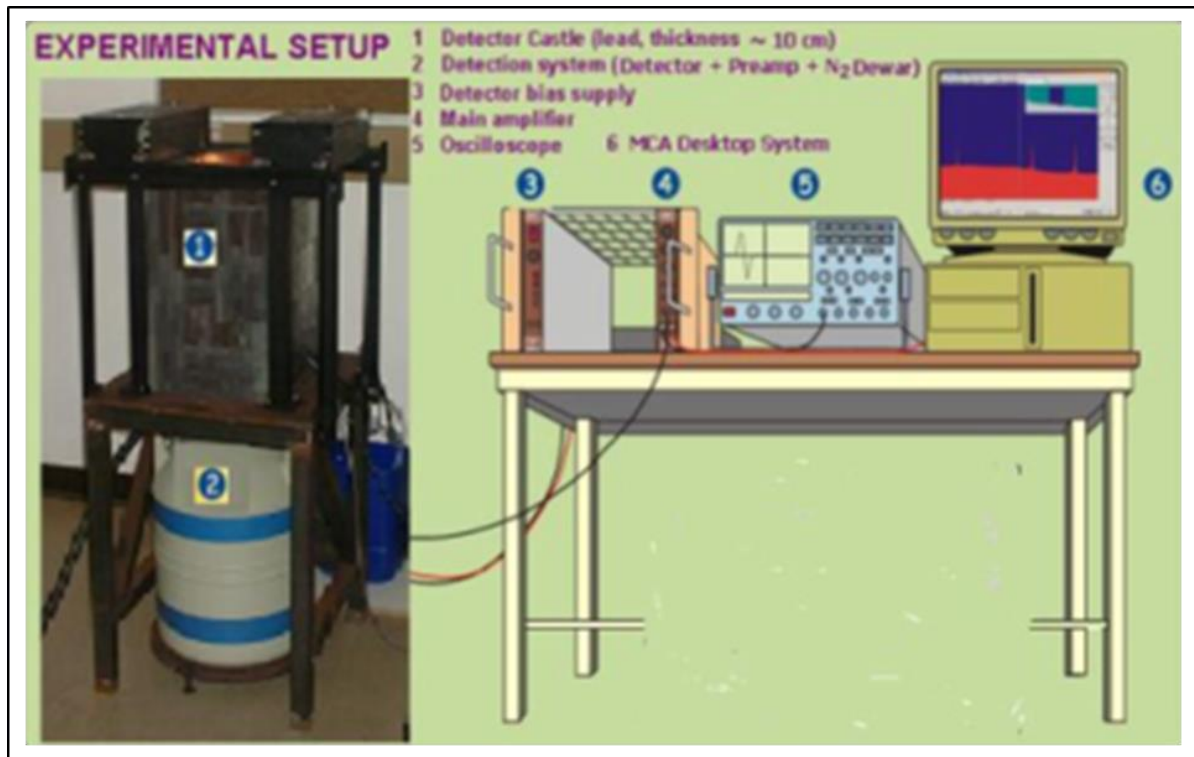


Figure 3.4. HPGe detector set-up with a photo (left) of the lead castle housing the detector and supported on a mechanically rigid cryostat connected to a liquid nitrogen Dewar (Damon, 2005).

On the right hand side; schematic of the various units used with the HPGe detector to measure and process the detector's signal; photo adapted from (Damon, 2005). Measurements were carried out at a laboratory under low-radiation background conditions achieved by shielding the detector (using lead) from ambient gamma radiation. Nuclear electronics were used to process pulses from the detector, through a multi-channel analyser (MCA) and binned in a gamma-ray spectrum (Figure 3.4). The background counts were measured for a period of 24 hours for each sample.

Thereafter, samples were taken to the Radiotherapy Vault at iTL for irradiation with neutrons. Samples were exposed to high intensity or fast neutrons as these neutrons are associated with larger flux and larger cross-sections (Alfassi, 1990). The activation analysis technique depends on nuclear reaction between projectiles, target nuclei, decay constant of the induced radionuclide, fractional abundance of the particular isotope and activation cross-sections (Alfassi, 1990; Robertson and Carpenter, 1970). Protons at an energy of 66 MeV incident on a Beryllium (^9Be) target (target current 25 μA) generating a spectrum of neutron energies between 0-66 MeV. ^9Be bombarded by protons provides an abundant neutron source (Osipenko *et al.*, 2013). Samples were placed on a 10 cm thick lucite block serving as backscatter medium, covered with a 12 mm thick polycaprolactam (nylon 6) build up material. The samples were exposed for a period of one hour at an average dose rate of 0.35 Gy/min to a total dose of 21 Gy in a reference calibrated field.

The sample activation was quantified by means of gamma ray spectroscopy, where directly after irradiation collective sample dose rate exceeded 1 mSv/h. As a result analysis of samples using gamma ray spectroscopy instrumentation could not be performed, detector dead-time exceeded 90%. Data acquisition for the first sample could only be performed 30 minutes post exposure.

Abundant radioisotopes with short half-lives decayed over this period to levels quantifiable by the detector and analysing equipment. Gamma ray spectrum acquisition time was set at 1 hour detector live time per sample. Sample acquisition time for samples analysed at time points 24, 72 and 168 hours post bombardment were 2 hours each to ensure good counting statistics at these low count rates.

CHAPTER 4: RESULTS AND DISCUSIONS

4.1 Introduction

In this chapter, geochemical, hydrochemical and instrumental neutron activation analysis of soil and groundwater samples is interpreted and discussed to understand possible radionuclides that maybe formed when neutrons interact with the sub-surface. The results are explained by means of tables, graphs and photographs. The geochemical and hydrochemical study of soil and groundwater is a good tool to use prior to the commencement of the LERIB project. The geochemical and hydrochemical results were used to investigate the mass fraction of elements present in the study area. The neutron activation analysis was used in identifying elements that might undergo activation in the sub-surface due to neutrons from the LERIB facility.

For radiological analysis, counts for background radiation were measured using the HPGe detector both from soil and groundwater samples to identify natural and artificial radionuclides present in the study area. After conducting the background radiation measurements, the samples were activated with neutrons for a period of one hour. The background spectra were subtracted from the irradiated spectra, to distinguish nuclear effect of neutrons in the samples. The gamma energies measured from the activated samples were used in identifying the formed radionuclide.

A study by Mantengu (2016) using FLUKA, to calculate the optimal shielding design predicted that, the sub-surface at a depth of 2-3 m bgl would be prone to activation (Figure 4.1). The interaction of neutrons with stable isotopes of major, minor and trace elements at depths of 2-3 m bgl, may result in the formation of radionuclides which might migrate with groundwater flow. According to Mantengu (2016), all neutrons which penetrated the 1 m shielding concrete during the bombardment of a Uranium Carbide target died off at a depth of 3.8 m with a fluence of 1000 neutron/cm²/s (Figure 4.1). This implies that, the neutron flux will decrease with depth, which in turn will reduce neutron energies. Therefore little

activation maybe expected at great depths such as beyond 6 m bgl of the study area (Figure 4.1).

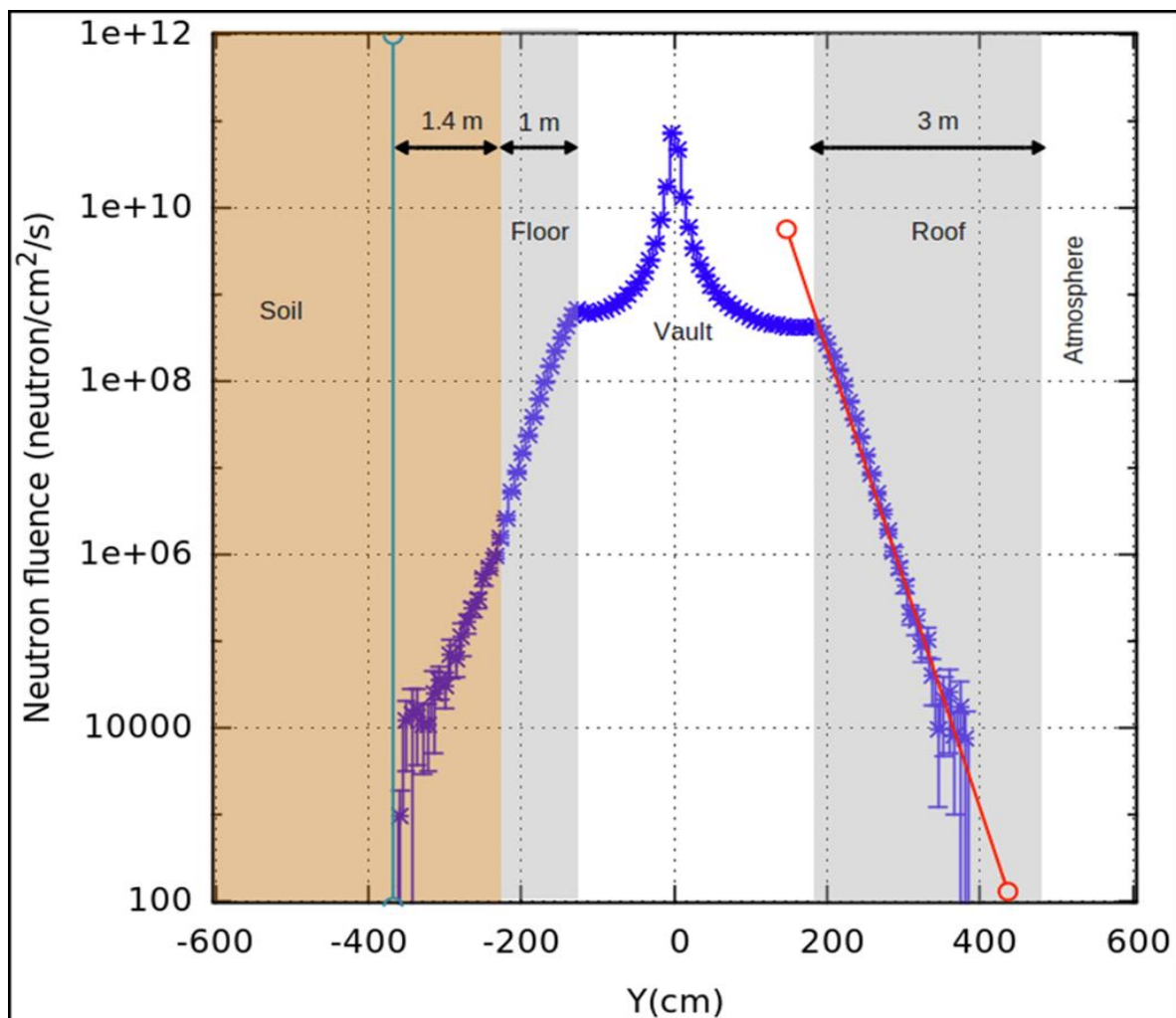


Figure 4.1. FLUKA simulations predicting neutrons penetrating the LERIB shielding material from a depth of 2-3 m bgl (Mantengu, 2016).

4.2 Geochemistry of soil

Using major oxides, minor and trace elements as variables, Haxel *et al.* (2002), established a discriminant diagram for elemental abundance in the earth's crust. In Figures 4.2, 4.3 and 4.2, the Haxel *et al.* (2002), elemental abundance diagram is used to present the abundance of elements in the sub-surface. The results revealed an abundance of silicon dioxide and aluminium oxide when compared with other elements. According to Glascock *et al.* (2004), geological materials formed of high

concentrations of silicon dioxide and aluminium oxide are the main components of clay minerals. The abundance of Al_2O_3 and SiO_2 (Figure 4.2) and less concentrations of other rock forming elements such as Ca, K, Mg and Na from the study area is evident that the soil in the sub-surface is consist of clay materials. The SiO_2 and Al_2O_3 dominated the study area by 84%, with 16 % being shared among other elements. The dominance of these two elements in the sub-surface resulted to most of the major and minor elements not to be in the range discussed by Haxel *et al.* (2002) and Borradaile (2003). Therefore when neutron radiation from the LERIB facility favours conditions for high neutron capture cross-sections, these two elements might stand more chances for neutrons to hit their nuclei due to their abundance (Schulze, 1969).

The Na_2O , K_2O and TiO_2 occurred in the range of 0.1 – 1% throughout the study area. The P_2O_5 , NiO, MnO and SO_3 were in the range of 0.01 – 0.1 %. The major elements occurring at low concentrations such as <0.01 %, are expected to have low for neutron capture and therefore stand less chances to undergo activation compared abundant elements (Bacchi, 2002).

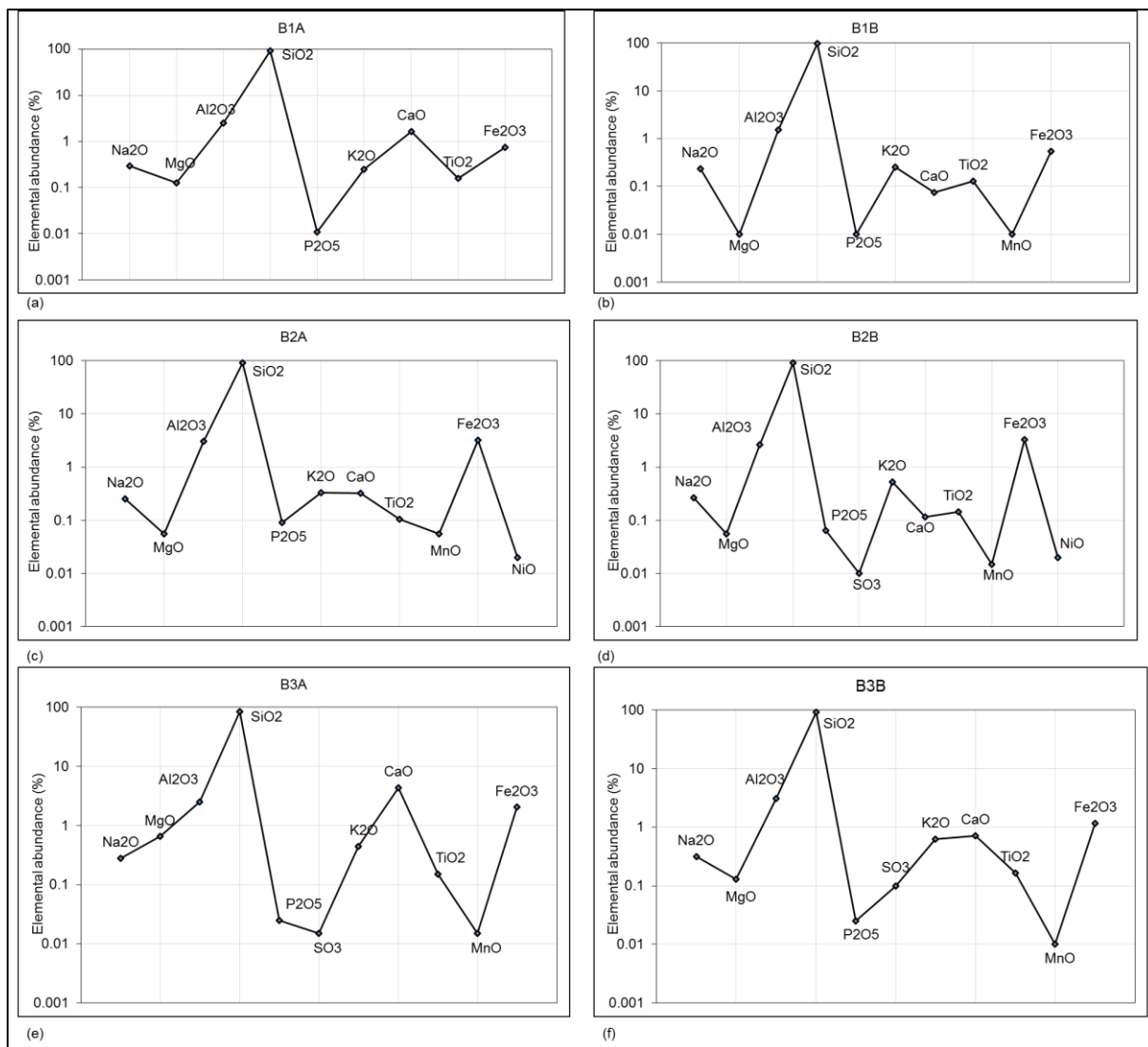


Figure 4.2. The average percentage of major elements at depths, 2-3 and 3-4 m from six boreholes at iThemba LABS. The results present a relative abundance (%) of major oxides of elements in the study area.

The minor elements in the earth's crust are classified as elements occurring at a range of 0.1 – 1% (Borradaile, 2003). The measurements for the soil samples from the laboratory were in parts per million (ppm) (Appendix 13) and had to be converted to percentage (%) units using a conversion factor: 1ppm = 0.0001% (Terrie and Boguski, 2006). The conversion was made so as to present the relative abundance of elements in the study area in a manner applied by Haxel *et al.* (2002). After converting the results into percent units, it was observed that the elements abundant in the range of minor elements were only Chlorine, Fluorine and Sulphur (Figure

4.3). Based on these observations, it is anticipated that when energies of neutrons from the LERIB floor enhances high neutron capture cross-sections in the sub-surface, these three elements might have a high probability to undergo activation (Schulze, 1969). However, if the neutron capture cross-sections are not high enough to induce radioactivity into these three nuclides, there would be a low probability for these nuclides to undergo activation.

The fluorine percentage was constant, as it was at a range of (0.1 - 0.01%) throughout all samples (Figure 4.3). The other elements starting from vanadium to lead were constantly measured at low concentrations (0.001 - 0.0001%), with only manganese and strontium being above 0.001% at B2B and B3A respectively. The concentrations of minor elements occurring at 0.0001 % represent a very low abundance and are expected to have a low probability to undergo activation (Figure 4.3).

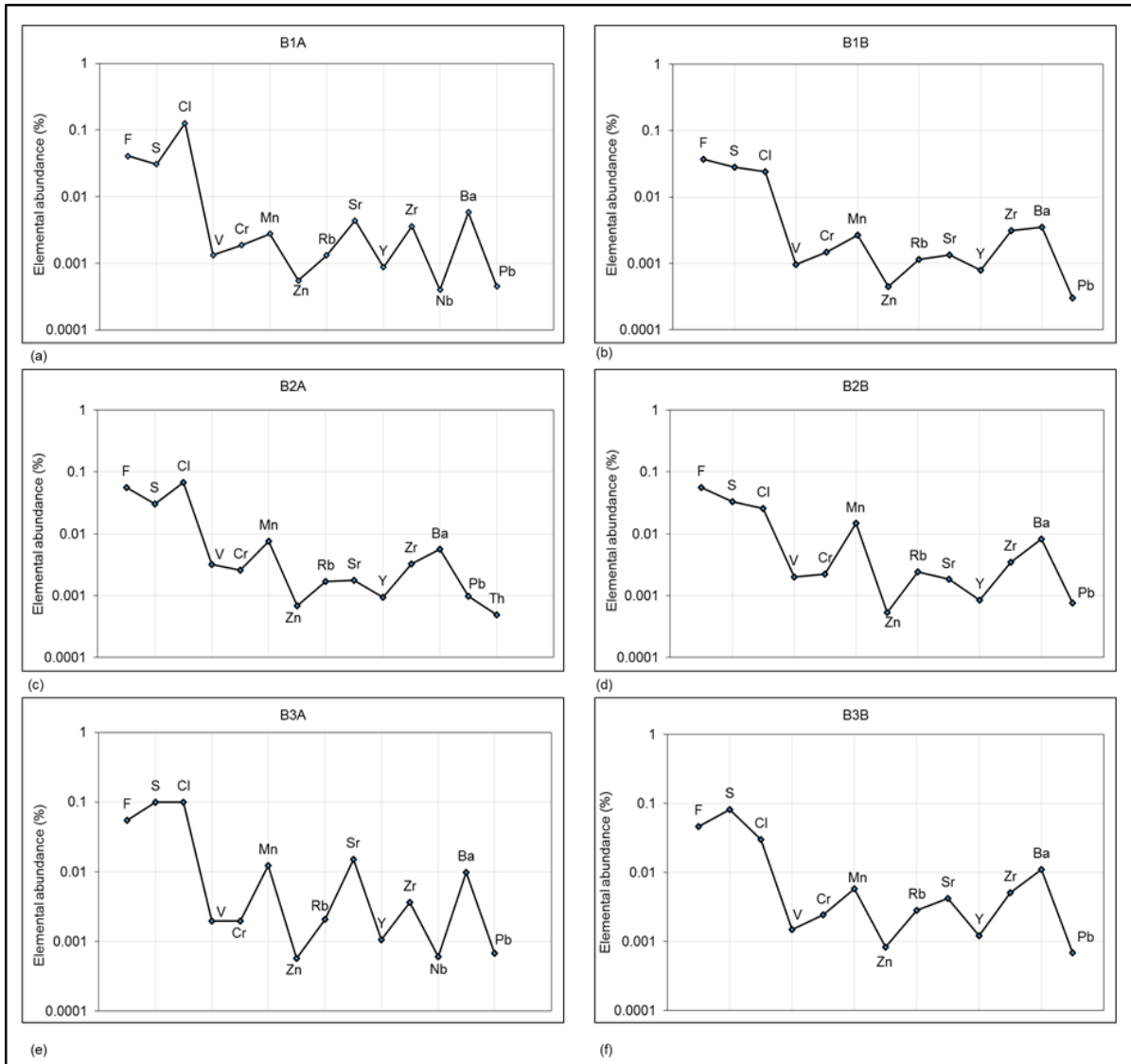


Figure 4.3. The average percentage of minor elements at depths, 2-3 and 3-4 m from six boreholes at iThemba LABS. The results present a relative abundance (%) of minor elements in the study area.

Trace elements are among rare earth geological materials, which occur at low concentrations and have lower (<0.1%) natural abundances compared to major and minor elements (Appendix 14). Among trace elements, there is a majority of heavy metals which when irradiated with neutrons produces intense radiation which can pose significant environmental impacts (Van Rooyen, 2006). However, the results revealed very small concentrations of trace elements and for this reason no activation would be expected to take place in the trace elements (Figure 4.4).

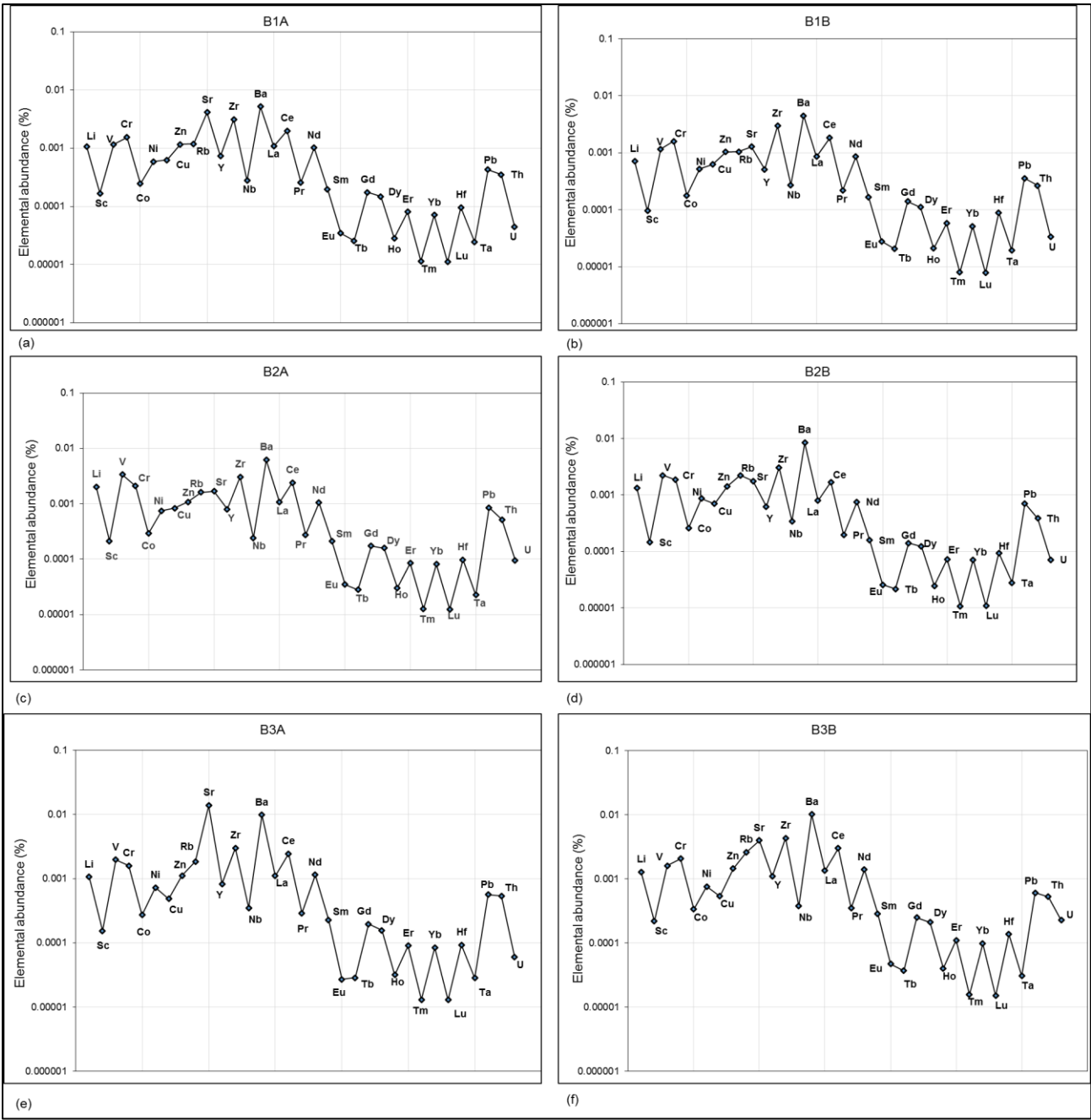


Figure 4.4. The average percentage of trace elements at depths, 2-3 and 3-4 m from six boreholes at iThemba LABS. The results present a relative abundance (%) of trace elements in the study area.

4.3 Hydrochemistry of groundwater

Groundwater contains a wide range of dissolved solids which migrate through aquifers and interacts with aquifer material in the sub-surface environment. As the groundwater moves, it may dissolve, transport and deposit mineral matter. The hydrochemistry of groundwater is controlled by the chemical composition of rain, recharge mechanism, properties of soil and rock in which the groundwater moves, contact time and contact surface between groundwater and geological material along its flow path, rate of geochemical (oxidation/reduction ion exchange, dissolution, evaporation, precipitation) process and microbiological process (Tröjbom *et al.*, 2008). The geochemical processes such as dissolution are important for this study as they can be used to assume radionuclides formed in the soil that might dissolve when in contact with water. For this reason, it is expected that activated isotopes in soil from elements with low solubility characteristics to have low chances of migrating with groundwater bodies. On the other hand, activating soil materials containing elements with high solubility nature might result in the migration of formed radionuclides with groundwater flow.

The hydro-chemical results were validated using the following formula for the ionic balance error (Appelo *et al.*, 1996):

$$\text{Ionic balance error} = \frac{\Sigma\text{cations} - \Sigma\text{anions}}{\Sigma\text{cations} + \Sigma\text{anions}} \times 100$$

The calculated error balance for six boreholes on site was calculated to be in the range of 0.1 to 4.9 % (Appendix 3, 4, 6, 8, 10 and 11). The balance error is at accepted levels and results can be trusted as they fall below the recommended error percentage of 10 % (Appelo *et al.*, 1996).

The groundwater results revealed that, among ions of major elements, the study area is composed of groundwater with high Na⁺ content (Figure 4.5). The abundant elements measured from soil samples using XRF such as SiO₂ and Al₂O₃ were not detected in groundwater samples due to their low dissolution nature in water bodies.. It can therefore be argued that, activated isotopes from these elements will only be stagnant in soil bodies and not migrate with groundwater.

Some of the elements measured from soil samples such as Mg, Fe, K and Ca were detected in groundwater bodies but at low concentrations compared to Na throughout the study area (Figure 4.5). Based on elemental abundance, it can therefore be argued that Na in groundwater bodies has a high probability to undergo activation when the neutron energies from the LERIB facility yield high neutron capture cross-sections. However, when energies of the penetrating neutrons in the sub-surface do not yield cross-sections in favour of activating Na, there will be no Na radionuclide generated regardless of its abundance. The Ca, Mg, Fe and K are expected to have low probabilities to undergo activation in groundwater due to their small elemental abundances (<0.01 %) (Figure 4.5).

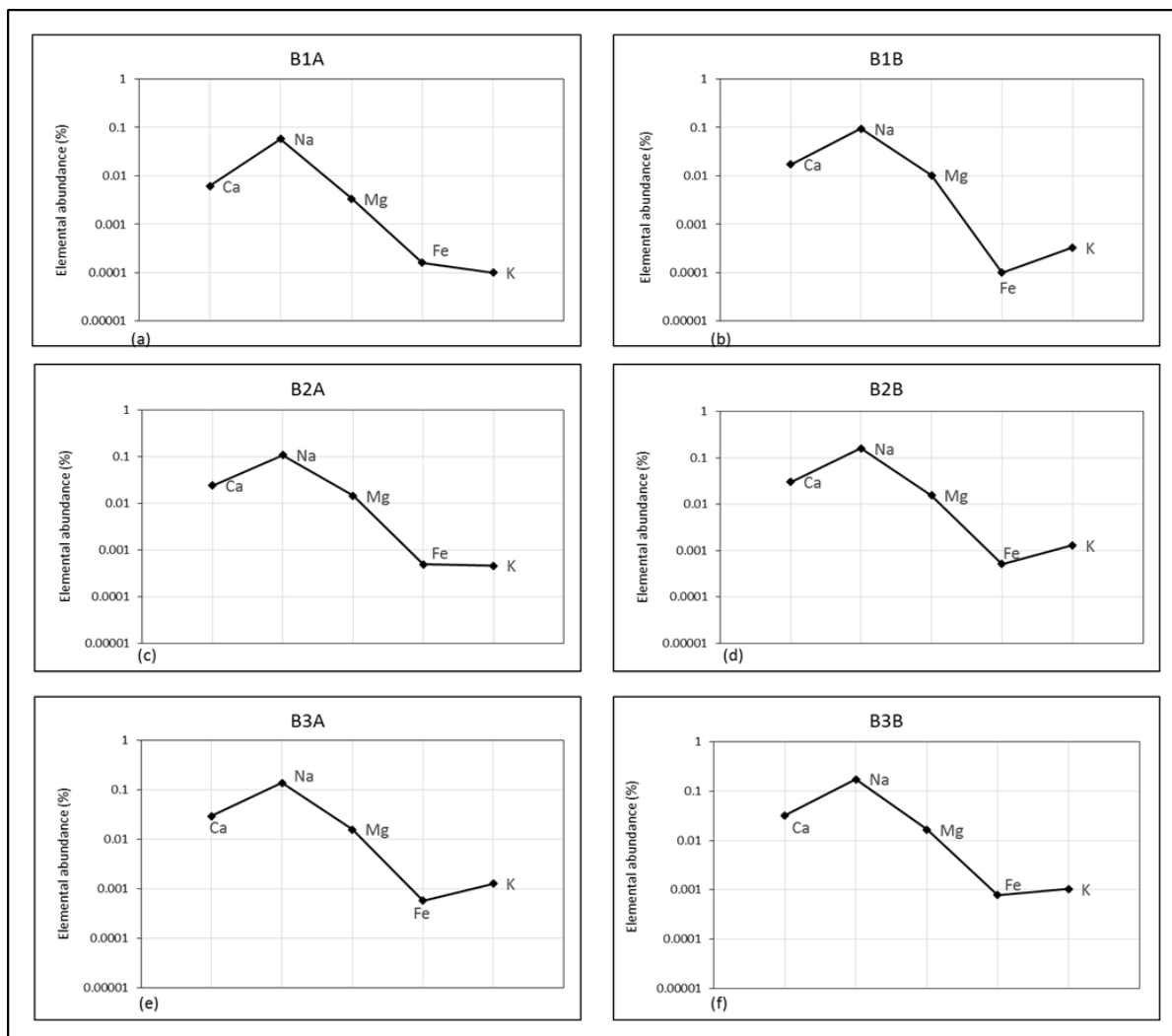


Figure 4.5. Major elements in groundwater samples collected from the six boreholes at iThemba LABS.

The minor elements from groundwater samples occurred at low concentrations except Chlorine which was measured at a range of minor elements (0.1 - 1 %) (Figure 4.6). The Manganese was below detection levels from the shallow borehole (B1A) (Appendix 9) hence not present in Figure 4.6 (a). The concentrations of F, Mn and P were very low throughout the study area and therefore chances for these elements to undergo activation are expected to be limited.

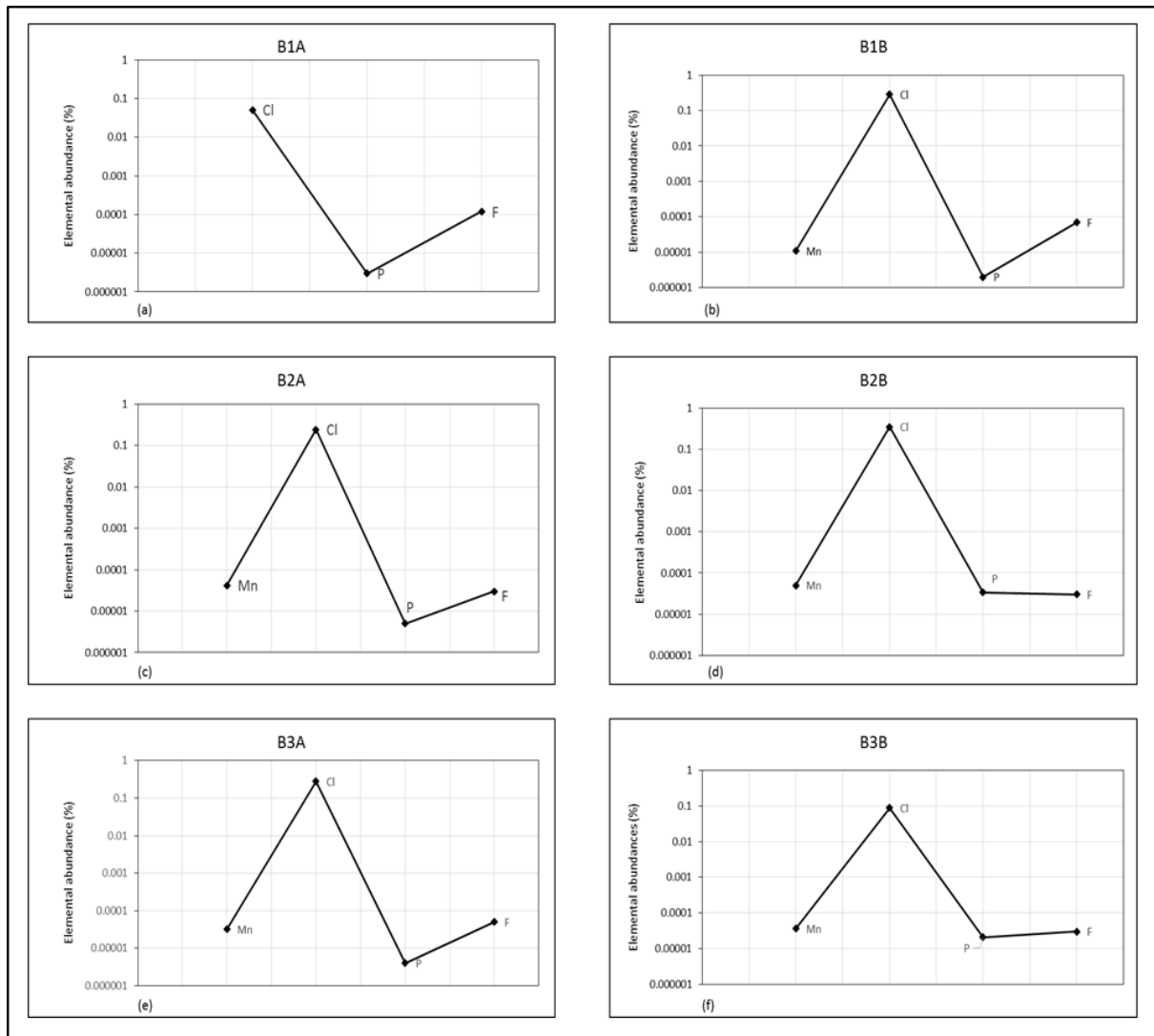


Figure 4.6. Minor elements in groundwater samples from six boreholes at iThemba LABS.

The trace elements in the earth's crust normally occur at very low concentrations when compared to major and minor elements. From soil samples, the most abundant element was at 0.01 % (Figure 4.4 (d, e and f)). A decrease in abundance of trace

elements in groundwater samples was observed where a dominant element was in a range of 0.0001 – 0.001 %. These are very low concentrations and such elements when exposed to neutrons might stand limited chances to undergo activation Bacchi *et al.*, (2002).

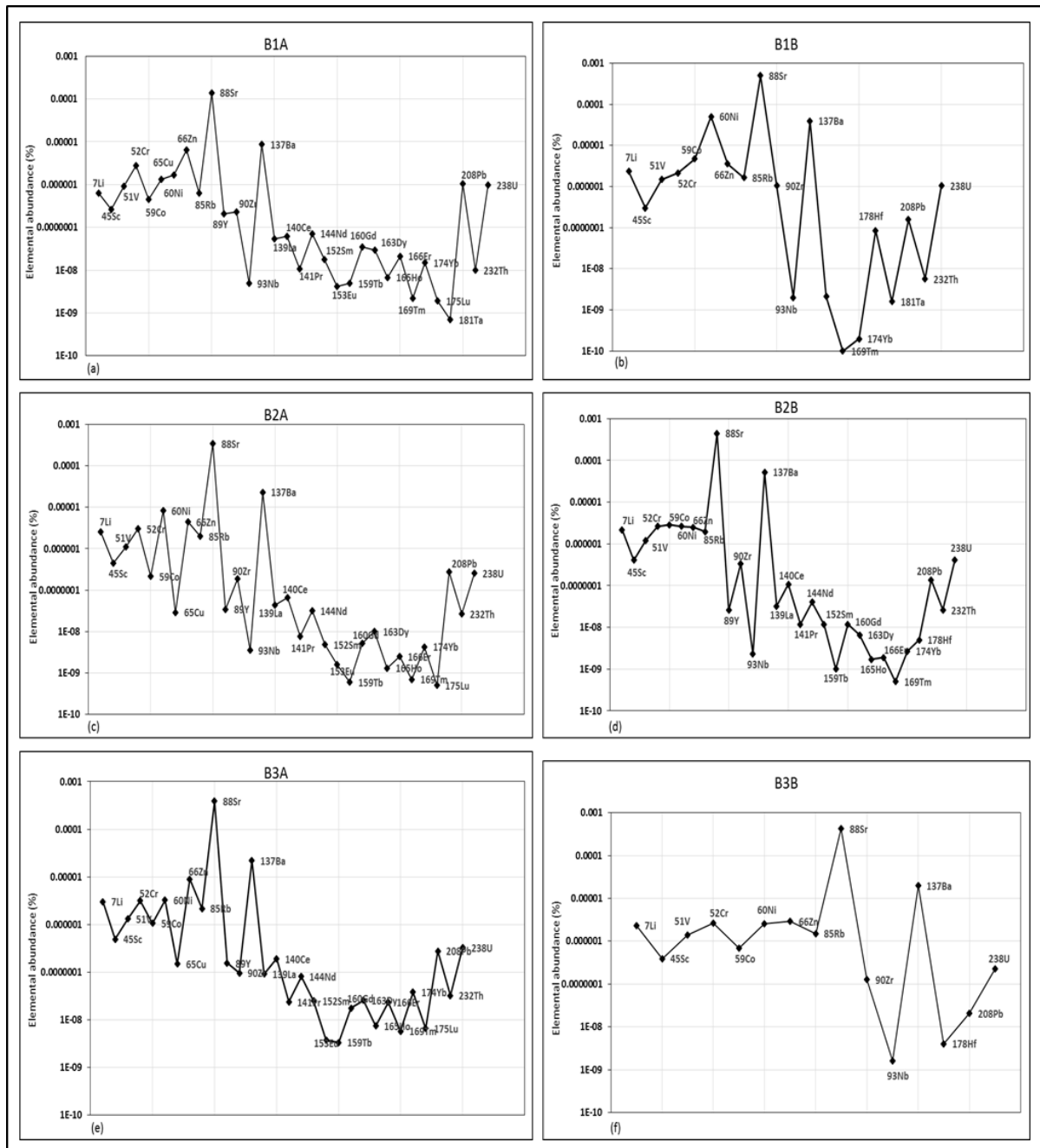


Figure 4.7. Trace elements in groundwater samples from the six boreholes at iThemba LABS.

4.4 Activation of stable isotopes in soil and groundwater

4.4.1 Measured background radiation in soil and groundwater

The neutron activation analysis was undertaken from soil and groundwater samples collected only from shallow boreholes (B1A, B2A and B3A), as these boreholes are designated for radiation monitoring for present and future operations at the iTL. The geochemical and hydrochemical investigations were undertaken in the sub-surface both from deep and shallow aquifers so as to understand environmental conditions underneath the proposed area for the construction of LERIB facility. The background radiation was measured from the samples so as to indicate current radiological conditions of the soil and groundwater onsite. The measured background radiation is believed to be from natural and artificial factors, where man-made radiation can be expected from the facility like iTL (Hlatshwayo, 2007).

The background measurements showed the presence of natural radioactive isotopes, ^{40}K and ^{208}Bi represented by gamma energies of 1460 keV and 2614 keV respectively (Figure 4.8). The measured ^{208}Bi in soil and water samples occurred consistently throughout the site. The counts for ^{40}K varied from borehole to borehole, particularly in soil samples where ^{40}K ranged from 0.001 – 0.005 counts/sec. Based on the spectra attained, it was evident that the locations, at which the samples were collected, had no presence of artificial radionuclides (Figure 4.8).

In order to distinguish between neutron induced and naturally occurring radioactivity, the detected radionuclides from the background radiation were subtracted from the irradiated samples. This was conducted so as to dissociate the anticipated activity induced by the LERIB project from the current background radiation in the study area.

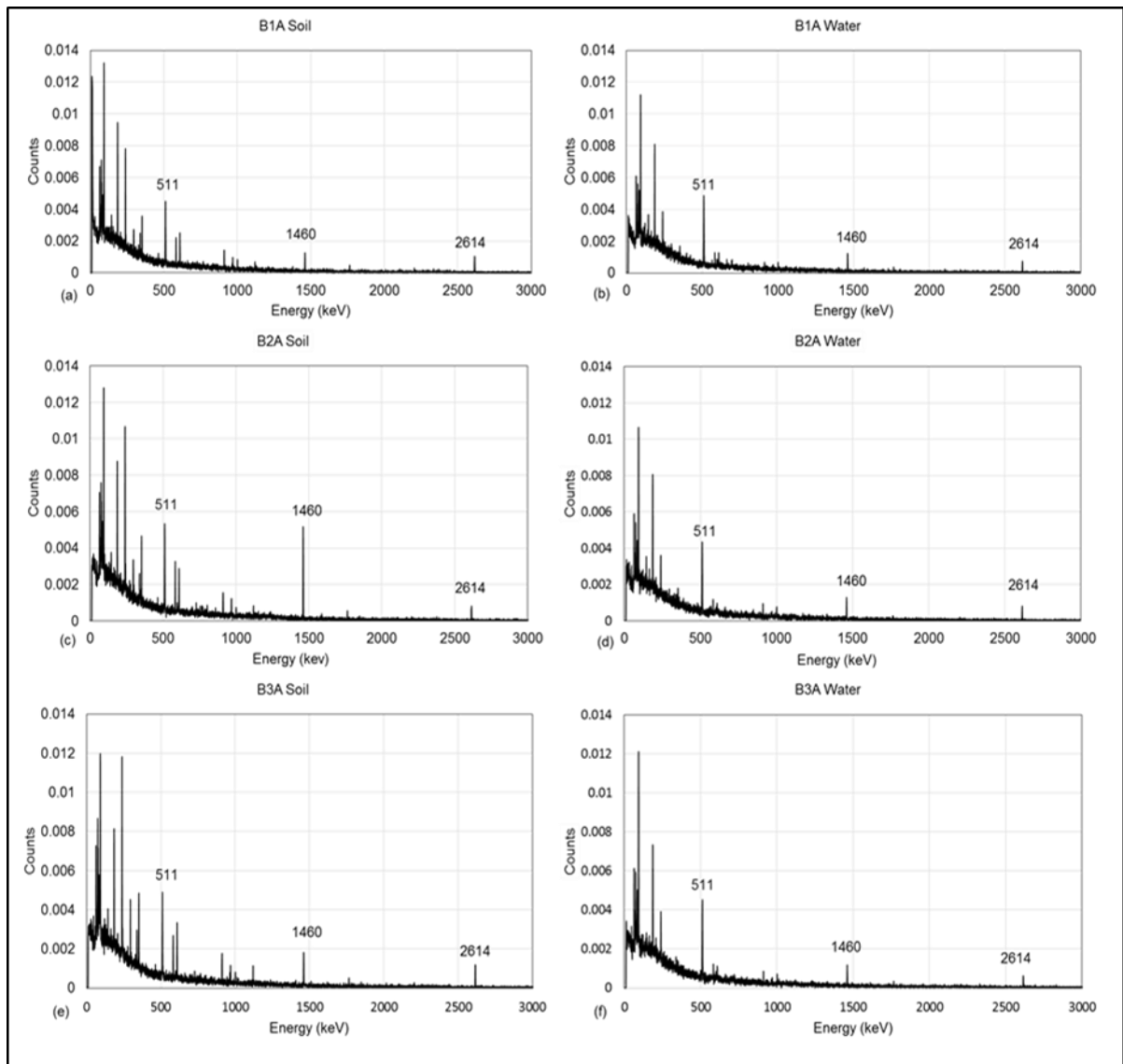


Figure 4.8. Background radiation of soil and groundwater samples from B1A, B2A and B3A.

4.4.2 Activated soil and groundwater samples

The samples were activated by neutrons generated from a Beryllium target at an energy of 0 - 66 MeV (mean energy of 29 MeV). According to Hughes (1957) neutrons of extremely wide energy range such as 0 - 66 MeV are able to interact effectively with nuclei and widen the energy range as they interact effectively with elemental nuclei. Furthermore, in terms of neutron energy ranges, a study by Yashima *et al.* (2004), produced neutrons from a Lithium target bombarded by proton energies of 30, 35, 40, 50, 60 and 70 MeV and revealed that widen neutron

energies effectively interact with an atomic nuclei. Thus the neutron energy range of 0 - 66 MeV deemed adequate to be used in activating samples for the purpose of this study.

After the irradiation of samples, the gamma rays emitted from the irradiated samples were measured with a high-purity germanium (HPGe) detector. Gamma spectroscopy spectra were captured one hour, one day and one week post bombardment to identify activated elements existing within these periods. The attained spectra showed energies of gamma rays through peaks counted per second corresponding to radionuclides formed by neutron activation technique (Figures 4.9, 10 and 11). It is however important to note that, not all of the peaks present in the spectra represent gamma energies. This is due to the fact that, the photon radiation with energies 50 eV to 500 keV is termed Roentgen radiation, where radiation higher than 500 keV is considered to be gamma-radiation (Kudryashov, 2008). For this reason only energies of gamma rays above 500 keV will be considered when identifying isotopes.

The gamma energy 511 keV represent the annihilation peak which was only measured from soil samples (Figure 4.9 - 4.11). According to the reviewed literature, the annihilation peak is a common feature in gamma spectra, which is much enhanced when β^+ nuclides are present (Gilmore, 2008). Gilmore (2008) further explains that the 511 peak is formed when the positron and electron disappear and two photons (annihilation pair) are produced with energy comparable to the electron mass of 511 keV. Saha (2001) argued that the β^+ particles are annihilated to produce two 511 keV photons, which appear as photopeaks in the gamma-ray spectrum. The results, showed the annihilation peak to be decreasing with respect to the decrease of nuclide concentrations (Figure 4.9 – 4.11).

4.4.3 Samples measured one hour post activation

The measured soil and water samples at one hour post activation revealed the presence of radionuclide Na-24 (^{24}Na) (Figure 4.9). According to Gritzay *et al.* (2002), ^{24}Na might be formed by activating a stable isotope Na - 23 (^{23}Na) through a nuclear reaction $^{23}\text{Na}(n,p)^{24}\text{Na}$. The gamma energies representing Na are 1368 and

2754 keV (Chu *et al.*, 1999). These gamma energies were measured in soil and water samples, thus confirms the formation of ^{24}Na in the samples (Figure 4.6).

When comparing the ^{24}Na counts from the samples, it was observed that, soil samples contained more of ^{24}Na compared to that of water samples (Figure 4.9). This variation in dominance 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 good neutron moderator might have absorbed the neutrons and lowered the energy to reduce the activity (Bacchi *et al.*, 2002).

Based on the presence of ^{24}Na from the activated soil and water samples, it can be argued that the soil and water samples contained stable isotope of ^{23}Na which when interacting with excessive neutrons forms unstable isotope ^{24}Na . The formed radionuclide ^{24}Na has a half-life of 15 hours (Firestone, 1996).

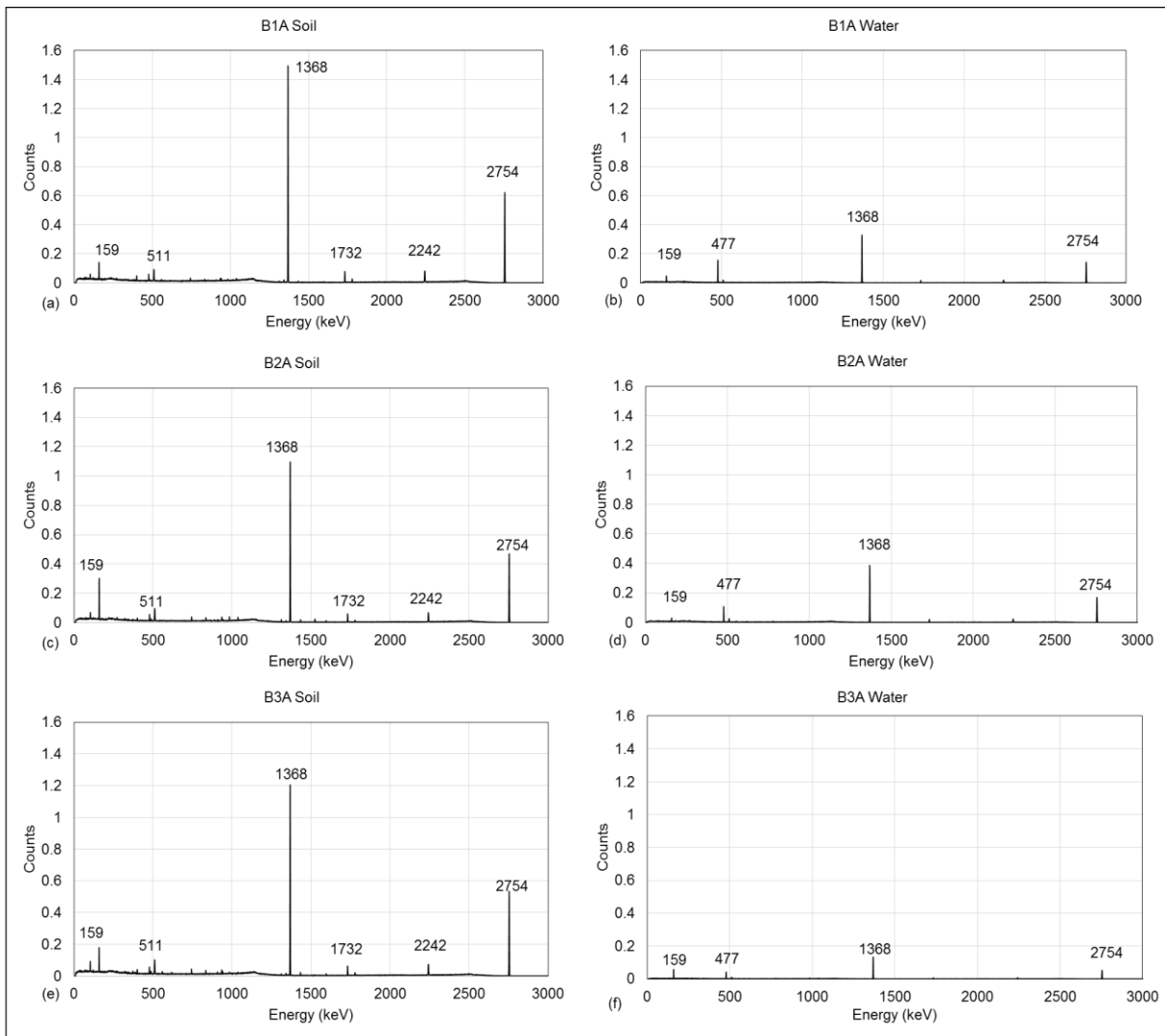


Figure 4.9. Activated soil and groundwater an hour post bombardment.

4.4.4 Samples measured one day post activation

Activity measurements were further conducted one day post activation and a significant decrease in count rates was observed (Figure 4.10). In water samples, the decrease of counts was immense when compared to soil samples. This can be associated with the fact that, soil samples had more counts from the beginning (1 hour post activation) of the measurements (Figure 4.9). The samples measured one day post activation showed no presence of other isotopes except the isotopes measured one hour post activation (Figure 4.9 and Figure 4.10).

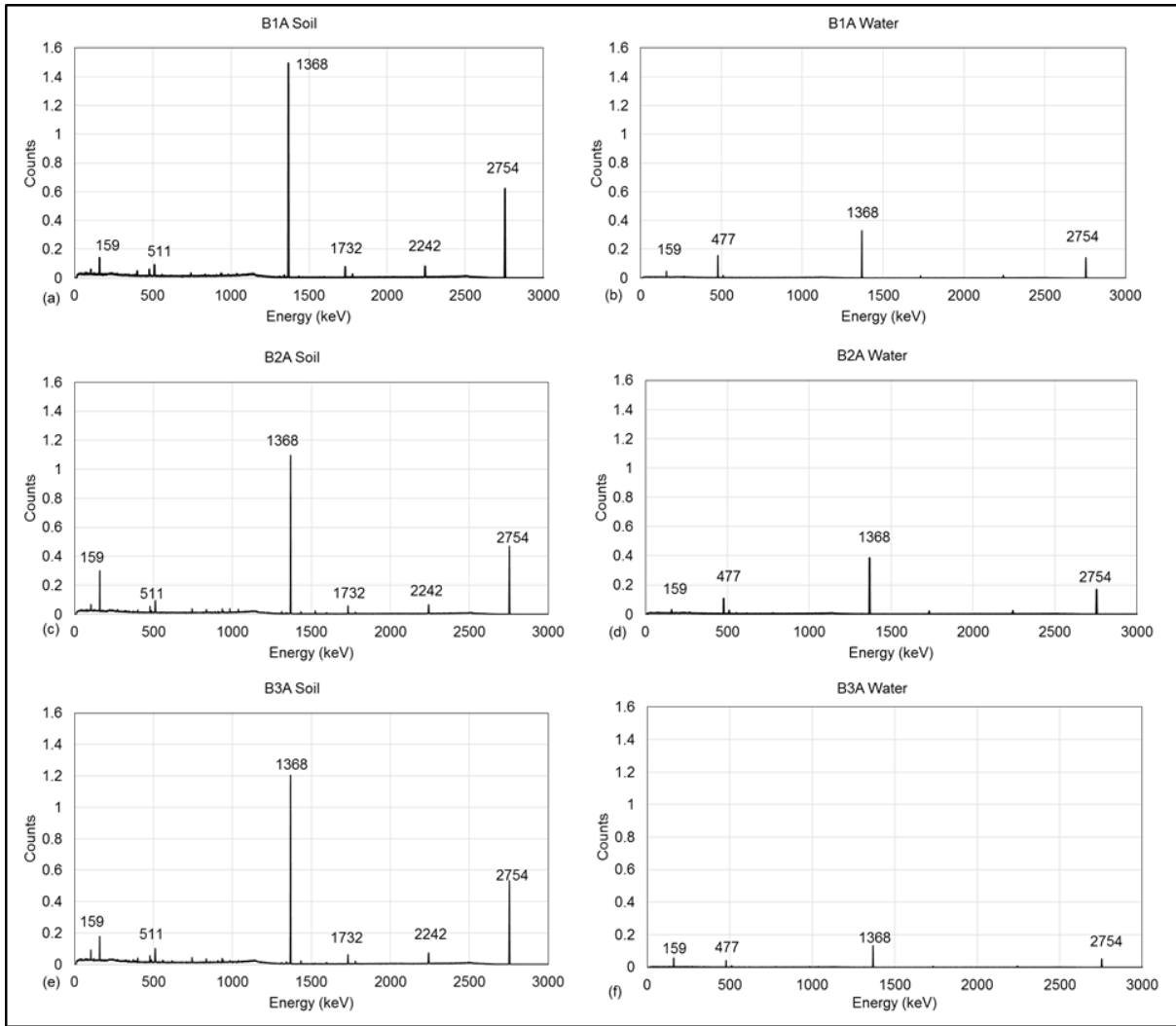


Figure 4.10. The activated soil and water samples one day post bombardment. Energies of gamma rays (keV) are represented by peaks counted at counts/second.

4.4.5 Samples measured one week post activation

Samples were measured one week post activation to observe nuclides with longer half-lives and to examine if peaks representing the presence of different radionuclides from those measured in Figure 4.9 and 4.10 appeared. The gamma energies measured from Figure 4.9 and 4.10 did not appear from the samples measured one week post activation (Figure 4.11). Instead a number of peaks which were different from the previously measured spectra were counted. However the gamma rays were only measured from soil samples and there were no gamma rays measured from water samples (Figure 4.11).

^{24}Na was the only nuclide observed one hour and one day post activation from water samples and disappeared one week post activation. The disappearance of ^{24}Na peaks one week post activation can be associated with its half-life of 15 hours. A nuclide with a half-life of 15 hours would be expected to undergo a full decay within a one week. The measured average distance of the three boreholes located down gradient from the envisioned LERIB facility is 167.23 m and the ^{24}Na would undergo a full decay before reaching any boreholes. Therefore, it can be argued that ^{24}Na formed in soil and groundwater bodies might not migrate a distance exiting the study area.

The spectra measured one week post activation revealed the presence of isotopes with longer half-lives ((Figure 4.11 (a), (c) and (e)). The observed radionuclides with long half-lives included manganese - 54 (^{54}Mn), manganese - 52 (^{52}Mn), Na - 22 (^{22}Na) and scandium - 46 (^{46}Sc) (Figure 4.11).

4.4.5.1 Manganese - 54

The ^{54}Mn was measured at gamma energy 834 keV (Figure 4.11 (a, c and e)). The ^{54}Mn is formed through activating a stable isotope ^{54}Fe , by a nuclear reaction $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ (Gritzay *et al.*, 2002). The iron was among abundant elements measured from the soil samples (Figure 4.2). In water samples, Fe was measured at low concentrations and as a result none of the Fe isotopes were activated in water. For this reason, the ^{54}Mn is anticipated not to be present in groundwater bodies.

Due to numerous stable isotopes of iron present in the earth's crust, such as ^{56}Fe , ^{57}Fe and ^{58}Fe , it was not clear which stable isotope of iron formed part of the soil in the study area. Measuring ^{54}Mn from the soil samples was an indication of the presence of ^{54}Fe , which has a natural abundance of 5.9 % in the crust (Audi *et al.*, 1993). Therefore having soil containing ^{54}Fe which is prone to activation from excessive neutrons might result to the sub-surface underneath the envisioned LERIB facility to constitute concentrations of radioactive ^{54}Mn . The ^{54}Mn has a half-life of 312.3 days (Firestone, 1996).

4.4.5.2 Manganese - 52

Another radionuclide of manganese measured from the soil samples was ^{52}Mn , at 744, 935 and 1434 keV gamma energies (Figure 4.11 (a, c and e)). 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 by Klix *et al.* (2014), showed that bombarding a Be target with 40 MeV and 10 μA deuterons beam (for 2.0 hours of irradiation) produced neutrons that activated ^{54}Fe to form ^{52}Mn . Tritium was also formed in the reaction. These findings implies that, soil samples containing ^{52}Fe might activate when interacting with neutrons form a radioactive isotope ^{52}Mn with tritium being released in the reaction (Sublet *et al.*, 1990). The ^{52}Mn maybe expected to be among radionuclides formed due to exposure of neutrons from the LERIB facility (Figure 4.11). The ^{52}Mn has a half-life of 5.6 days (Firestone, 1996).

4.4.5.3 Sodium - 22

The unstable isotope, ^{22}Na was measured from soil samples at energy 1274 keV (Figure 4.11 (a), (c) and (e)). The radioactive ^{22}Na , through neutron activation process is formed by activating the stable isotope of ^{23}Na following a nuclear reaction $^{23}\text{Na}(n, 2n)^{22}\text{Na}$ (Gritzay *et al.*, 2002). The radionuclide ^{22}Na has a half-life of 2.6 years (Firestone, 1996). This implies that ^{22}Na might migrate a distance exiting the study area due to its half-life. The high activity concentrations of ^{22}Na in the sub-surface might result to the health risks when groundwater is used for domestic purpose.

4.4.5.4 Scandium - 46

The soil samples were measured to contain a radionuclide, scandium – 46 (^{46}Sc) at gamma energies 889 and 1120 keV (figure 4.11, (a, c and e)). The ^{46}Sc is produced through activating a stable isotope of ^{46}Ti with a nuclear reaction $^{46}\text{Ti}(n, p)^{46}\text{Sc}$ and has 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 samples as seen from Figure 4.2 (a-f). Titanium has 5 naturally occurring isotopes (Audi *et al.*, 1993; Audi *et al.*, 1995). The formation of ^{46}Sc distinguishes which Titanium isotope

is present in the study area and prone to activation. Based on these results, it can therefore be argued that the neutrons from the envisioned LERIB facility might activate a stable isotope of ^{46}Ti .

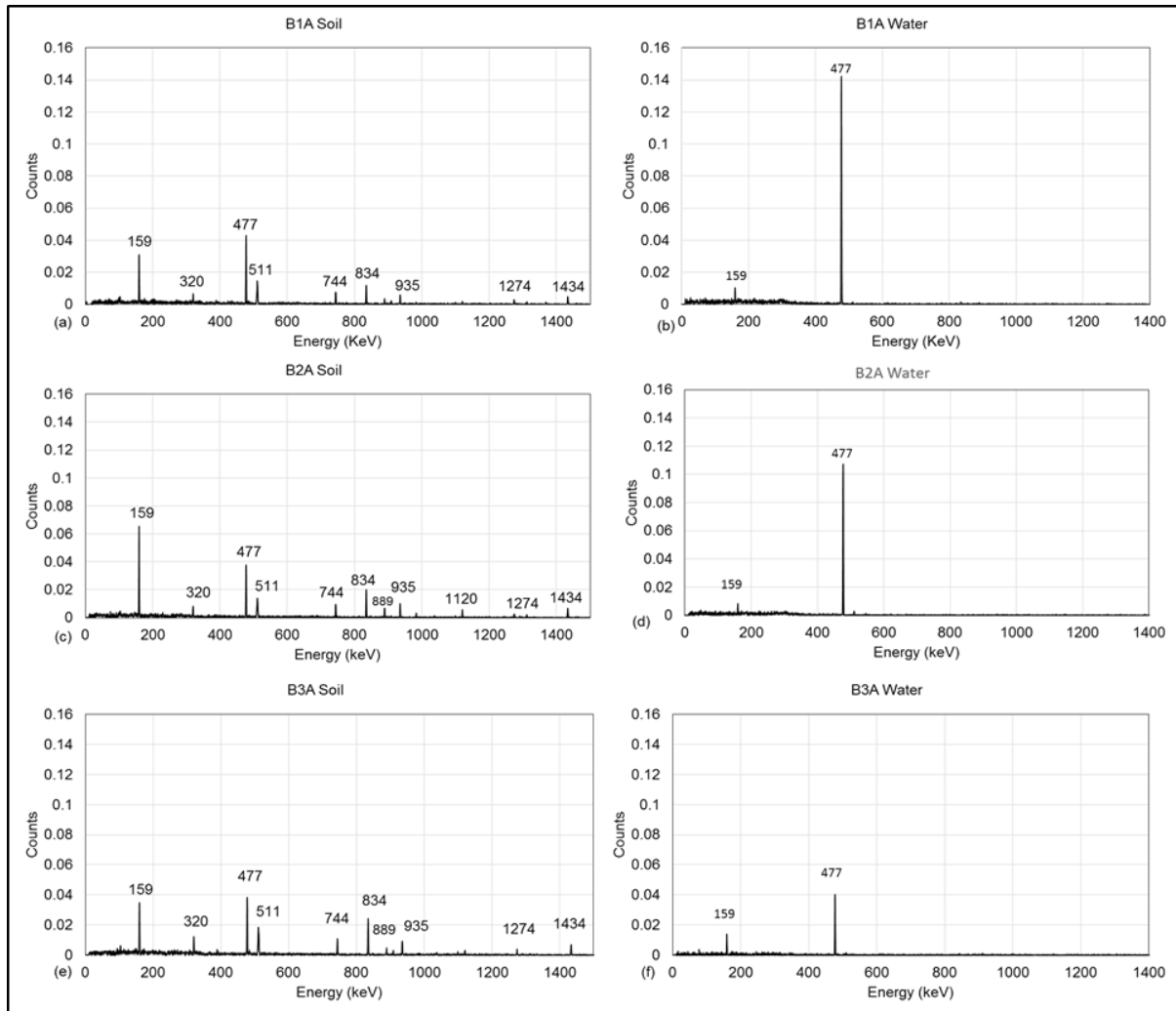


Figure 4.11. Irradiated soil and water samples analysed one week post bombardment.

4.5 The migration of activated isotopes in groundwater

The physical soil properties such as texture and grain size might allow radionuclides to seep through and travel distances within the subsurface. The solid (mineral and organic), liquid and gaseous phases governs the movement of matter in soil components (Bradford *et al.*, 1996). These three stages might allow for the migration of radionuclides in soil as well as for groundwater. Most of groundwater flows in

accordance with Darcy's Law, which 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 nature of the underlying aquifer (Todd *et al.*, 1976). Todd *et al.* (1976) further argued that, within confined aquifers, water movement is roughly horizontal due to the negligible vertical movement within the confining strata. This implies that, minimal or no activated isotopes from the unconfined aquifer will move vertically to the confined aquifer and as a result, the unconfined aquifer will therefore be primarily exposed to ionising radiation.

The groundwater flow measurements were conducted at summer and winter seasons, where the water table was measured and observe to vary at both seasons. During the winter season, the water table was high when compared to the summer season (Figures 4.12 and 4.13). The water table in winter was measured to be at an average depth of 13.6 metres above sea level (m abs) and 12.4 m abs at summer (Figures 4.12 and 4.13). In both seasons, the water table did not drop to a depth less than the layer anticipated to be exposed to neutron radiation (Figures 4.12 and 4.13). This implies that the iTL is underlain by a high water table environment with little seasonal variations and chances for neutrons to easily interact with groundwater bodies will be high.

To investigate the local groundwater flow and direction, modelling was done using the Surfer 14 program with an understanding that, groundwater flows from regions of higher hydraulic head to regions of lower hydraulic head (Barackman and Brusseau, 2002). However, due to the gentle slope of the study area and the short distance (~100 m) between the boreholes, the hydraulic head in the study area was measured to be higher towards the False Bay Ocean (region of lower hydraulic head) (Figure 4.12 and 4.13). This suggests that, the groundwater flow velocity in the study area towards False Bay would be relatively low.

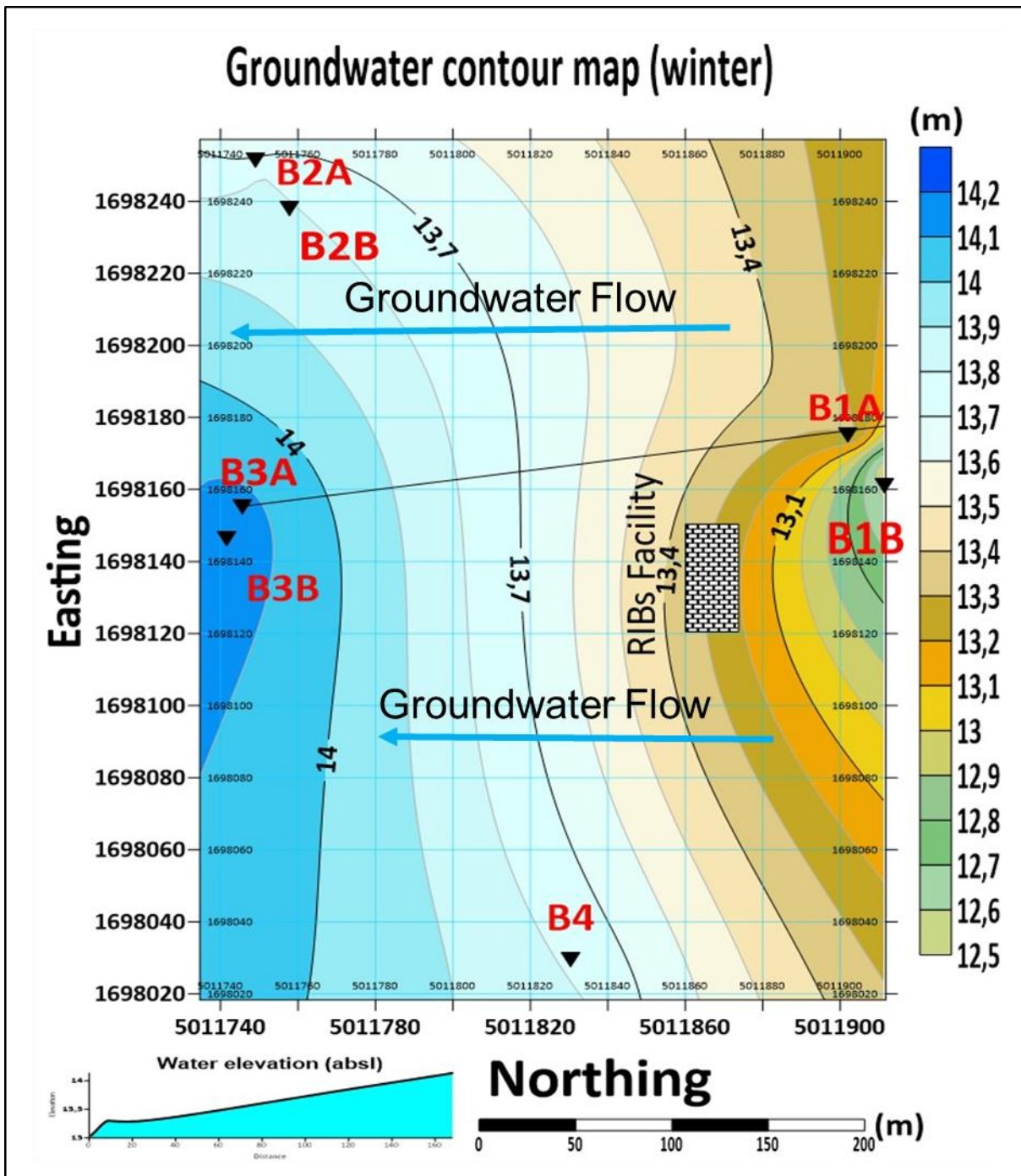


Figure 4.12. The measured groundwater table and flow direction from six boreholes at winter.

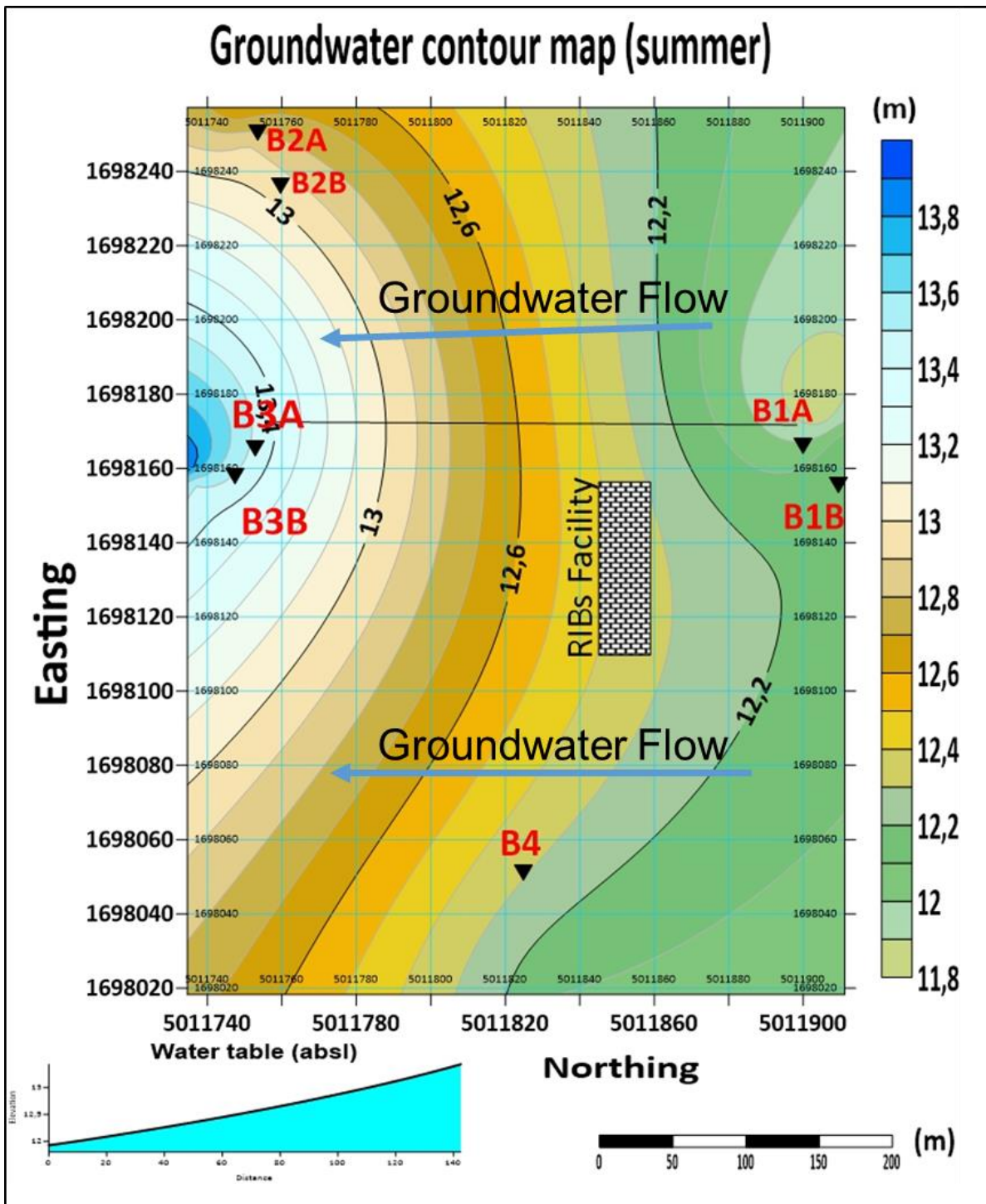


Figure 4.13. The measured groundwater table and flow direction from six boreholes at summer.

Based on the groundwater occurrence in the study area, it is evident that the water table drops slowly and may result to the interaction with ionising radiation for longer periods compared to water occurring in the confined aquifer (Figure 4.14). The

radionuclide activity concentrations are expected to be high in shallow groundwater compared to deep groundwater bodies (Figure 4.14). Some of the induced nuclides might settle in groundwater bodies and follow a vertical movement within the aquifer. It will therefore be the nuclides moving vertical within the aquifer that might introduce radioactivity in water occurring at great depth. The three boreholes located adjacent the LERIB facility were designed to monitor any activation of radionuclides that will follow a horizontal movement within the study area towards the south direction (down gradient) (Figure 4.14).

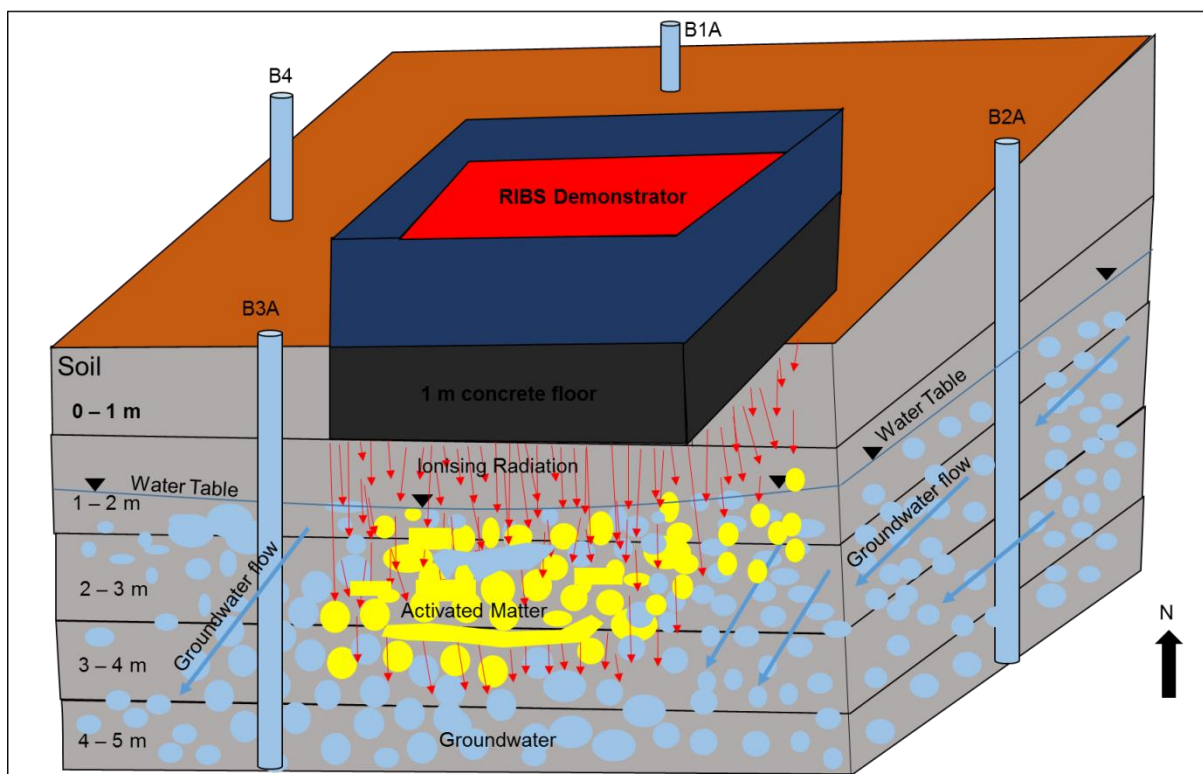


Figure 4.14. Conceptual model of groundwater flow in the envisioned LERIB facility at the iThemba LABS. The red arrows represent the ionising radiation through a 1 m thick concrete floor. The yellow circles represent possible activated matter in the subsurface either in water or soil. The blue circles and arrows represents groundwater and groundwater flow directions respectively (not to scale).

The groundwater velocity was calculated using Darcy's law, which describes the flow of fluid through a porous medium as:

$$V = K \left\{ \frac{h_2 - h_1}{L} \right\} \dots\dots\dots(4.1)$$

Where K = hydraulic conductivity (m/s)

h = hydraulic head (m)

L = distance (m)

The velocity of groundwater was calculated to be 1.04 m/day. Based on groundwater velocity, the activated isotopes in water bodies are anticipated to migrate at a velocity of 1.04 m/day. The three boreholes located down gradient from the LERIB facility will monitor radionuclides with long half-lives which might when present in water migrate at groundwater velocity a distance outside the study area (Figure 4.12).

The only radionuclide formed in water, was ^{24}Na and due to its short half-life it is expected to decay before migrating a distance outside the study area. According to the groundwater hydrochemistry results in Figure 4.5 it was evident that among major elements, Na highly dissolves in water as was measured at high concentrations from the study area. For this reason, it was argued that although ^{22}Na was not formed in water samples but formed in soil samples, the solubility nature of Na might result to ^{22}Na in soil to dissolve when in contact with groundwater and concurrently migrate with groundwater flow. This follows the findings of Šraček and Zeman (2004) from which it was argued that two isotopes of the same element are chemically identical and therefore if ^{23}Na dissolves in water, ^{22}Na and ^{24}Na are expected to undergo the same reaction when in contact with water.

Nonetheless, it is important to note that all radionuclides measured one week post activation including ^{22}Na were within the background radiation concentrations. This is evident when comparing the background spectra in Figure 4.8 and Figure 4.11, where the highest counts from the background radiation were less than 0.014 and less than 0.08 counts per second one week post activation. The activity concentration of all radionuclides measured one week post activation can be neglected regardless of their long half-lives as their impact in soil and groundwater bodies might be insignificant.

Chapter 5: Conclusion and Recommendations

5.1 Conclusion

The current study was about the investigation of activating stable isotopes in the subsurface by neutron radiation from the envisioned LERIB facility, where the ICP-MS and XRF methods were used to analyse the geochemical composition of the sub-surface underlying the study area. The INAA method was used to investigate isotopes of elements measured from ICP-MS and XRF that might be prone to activation when interacting with neutron radiation. The discussed results revealed that the stable isotopes in soil and groundwater will undergo activation when excessive neutrons from the LERIB facility interact with the sub-surface. However, only isotopes of major elements undergone activation in soil, where none of the stable isotopes for minor and trace elements undergone activation due to their low abundances. Among all the isotopes of major elements measured from soil samples, only ^{23}Na , ^{54}Fe , ^{52}Fe and ^{46}Ti undergone activation. The activation of only few elements is due to low cross-sections for neutron absorption in soil. Based on neutron absorption cross-sections, some of the elements which occurred in soil at high concentration such as Silicon and Aluminium did not undergo activation due to their low cross-sections. Nonetheless, due to the ability of water to shield neutrons, only ^{23}Na was activated in water samples. For these reasons, it is anticipated that, isotopes of major elements in the subsurface are prone to activation as a result short and long-lived radionuclides can be expected. However, the distance short-lived isotope can migrate is shorter, such that they will not migrate outside the parameters of the study area. The long-lived isotope can migrate distances exiting the study area; however their activity concentrations are at background levels, therefore insignificant environmental concerns on site and down gradient can be expected.

5.2 Recommendations

Based on the results outlined in 5.1, the following key recommendations were made, that the groundwater be monitored for radiological quality on a monthly basis so as to see whether would there be changes on the water quality after the installation or operation of the LERIB facility. The radiation monitoring should be conducted in the three 15 m deep boreholes located adjacent the proposed location of the envisioned LERIB facility. However, for safety practises, a thick shielding concrete should be built so as to ensure that only less energetic neutrons or zero neutrons passes through the shielding to activate matter in the sub-surface.

References

- Abdalla, S. A. T. 2009. Measurements and applications of radon in South African aquifer and river waters. P.hD Thesis, University if the Western Cape, South Africa (unpublished).
- Aller, L., Lehr, J. H., Petty, R. 1987. DRASTIC: A standardized system for evaluating ground water pollution potential using hydrogeologic settings: NWWA/EPA Series, EPA-600/2-87-035, U.S Environmental Protection Agency, Washington.
- Appelo, C., A., J, and Postma, D.,1996: Geochemistry, groundwater and pollution : Third corrected print, A.A. Balkema Publishers, Netherlands.
- Alfassi, Z. B. 1990. Activation Analysis. Volume I. CRC Press. Boca Raton, Florida.
- Aris, A. Z., Lim, W. Y. and Looi, L. J. 2015. Natural and anthropogenic determinants of freshwater ecosystem deterioration: An environmental forensic study of the Langat River Basin, Malaysia. Springer International Publishing, Switzerland.
- Audi, G. and Wapstra, A. H. 1993. Nucl. Phys A, 565 1 – 65. International Atomic Energy Agency. Austria.
- Bacchi, O. S., Reichardt, K., Clvache, M. and Timm, L., C. 2002 Neutron and gamma probes: Their use in agronomy. Vienna, International Atomic Energy Agency (<http://indico.ictp.it/event/a0261/session/43/contribution/30/material/0/0.pdf>) (cited 22 November 2017).
- Barackman, M and Brusseau, M., L. 2002. Groundwater Sampling. In: Artola, J. F., (edts), Environmental Monitoring and Characterization. Elsevier Inc. U.S.A, pp 121-139.
- Borradaile, G. J. 2003. Statistics of earth science data: Their distribution in space, time and orientation 1st Edition. Springer-Verlag Berlin Heidelberg.

Bradford, G. R., Change, A. C., Page, A. L., Bakhtar, D., Frampton, J. A. and Wright, H. 1996. Background concentrations of trace and major elements in California soils. Kearney Foundation Special Report. California.

Byrne, J. 1994. Neutrons, Nuclei and Matter: An exploration of the Physics of slow neutrons. Institute of Physics, London.

Canadian Council of Ministers of the Environment. 2007. Canadian soil quality for uranium: Environmental and Human Health. Scientific Supporting Document, PN 1371. Canadian Council of Ministers of the Environment, Canada.

Choppin, G., Liljezin, J. O., Rydberg, J. and Ekberg, C. 2013. Behaviour of Radionuclides in the Environment. Radiochemistry and Nuclear Chemistry (Fourth Edition). Academic Press. USA.

Chu, S. Y. F., Ekstrom, L. P. and Firestone, R. B. 1999. The Lund/LBNL nuclear data search, Version 2. (<http://nucleardata.nuclear.lu.se/toi/abouttoi.htm>) (cited 08/01/2018).

Clarke, S. D., Hamel, M. C., Di Fulvio, A. and Pozzi, S. A. 2017. Neutron and gamma-ray energy reconstruction for characterization of special nuclear material. Korean Nuclear Society, Elsevier. Korea.

Cunningham, W. L. and Schalk, C. W. 2011. Groundwater Technical Procedures of the U.S. Geological Survey: U.S. Geological Survey Techniques and Methods, 1-A1, 151 p. Reston, Virginia.

Damon, R. W. 2005. Determination of the photopeak detection efficiency of a HPGe detector, for volume sources, via Monte Carlo simulations. Masters Thesis, University of the Western Cape, South Africa (unpublished).

Department of Water Affairs and Forestry. 1996. South African Water Quality Guidelines. Volume 1, Domestic Use. Pretoria, South Africa.

Diamond, R., E. and Harris, C. 2002. The thermal springs of the Table Mountain Group: A stable isotope study. In: Pietersen, K and Parsons, R (eds.), A synthesis of

the hydrogeology of the Table Mountain Group – formation of a research strategy. Water Research Commission (WRC) Report, TT 158/01. South Africa. Douglas, E., Paquette, P. G., Chek, B. N. and Penny, G. 2008. Soil activation and groundwater contamination at Brookhaven National Laboratory, Upton, New York – 8080. WM2008 Conference, Phoenix, AZ. New York.

Feige, Y., Oltman, B. G. and Kastner, J. 1968. Production rates of neutrons in soils due to natural radioactivity. American Geophysical Union. United States of America.

Firestone, R. B. 1996. Table of Isotopes. Version 1.0 8th Edition. Wiley Interscience. United State of America.

Gilmore, G. R. 2008. Practical gamma-ray spectrometry 2nd Edition. John Wiley and Sons, Ltd, Chichester, England.

Gluscock, M. D. 2016. Overview of Neutron Activation Analysis. http://archaeometry.missouri.edu/naa_overview.html (cited 07 November 2017).

Gordon, S. 1992. Link between ore bodies and biosphere concentrations of uranium. AERCB Project No. 5.140.1. Atomic Energy Control Board, Ottawa, Canada.

Gritzay, O., Vlasov, M., Chervonna, L., Klimova, N. and Kolota, G. 2002. In: Wagemans, J., Abderrahim, H., A., D'hondt, P and De Raedt, C (edts), Reactor Dosimetry in the 21st Century. Proceedings of the 11th International Symposium on Reactor Dosimetry. World Scientific. Brussels, Belgium.

Hailu, A. T., Chaubey, A. K., Mamo, D. A. and Hibstie, A. Y. 2012. Application of instrumental neutron activation analysis for the elemental analysis of various rocks from areas around Debre Birhan City, Ethiopia. International Journal of Research in Agricultural Sciences. India.

Haxel, G. B., Hedrick, J. B. and Orris, G. J. 2002. Rare Earth Elements-Critical Resources for High Technology. U.S. Geological Survey Fact Sheet 08702. United States of America.

Heath, R. C. 1983. Basic ground-water hydrology: U. S. Geological Survey Water-Supply Paper 2220, 86 p.

Hibstie, A. Y. 2012. Elemental analysis of geological samples using neutron activation technique. P.HD Theis, University of South Africa (unpublished).

Hlatshwayo, I. N. 2007. In-situ gamma-ray mapping of environmental radioactivity at iThemba LABS and associated risk assessment. Masters Thesis, University of Zulu Land, South Africa (unpublished).

Hughes, D. J. 1957. Neutron cross sections. Pergamon Press. London.

Hutton-Squire, M., Madell, C. and Pudifin, C. 1992. Driftsands and environs technical report. MLH Architects and Planners, Cape Town. South Africa.

International Atomic Energy Agency. 2004. Radiation, People and the Environment. International Atomic Energy Report, IAEA/PI/A.75 / 04-00391, Austria.

Kabata-Pendias, A. 2011. Trace Elements in Soils and Plants. Taylor and Francis Group, LLC. United States of America.

Kantey and Templer. 2008. Report on geotechnical investigations for the proposed Cape Town Film Studios, Eesteriver. K & T Project Reference: 9671GG. (unpublished).

Kudryavtsev, V. A., Pandola, L. and Tomasello, V. 2008. Neutron- and muon-induced background in underground physics Experiments. Springer-Verlag. Italy.

Kudryashov, Y. B. 2008. Radiation biophysics (ionizing radiation). Nova Science Publishers, Inc. New York.

Knoll, G. F. 2010. Radiation Detection and Measurement, 4th ed. John Wiley and Sons, Inc. United States of America.

Konefal, A., Orlef, A., Łaciak, M., Ciba, A. and Szewczuk, M. 2012. Thermal and resonance neutrons generated by various electron and X-ray therapeutic beams

from medical linacs installed in polish oncological centres. Reports of practical oncology and radiotherapy. Poland.

Leroy, C. 2009. Radiation interaction in matter and principles of detection. American Institute of Physics Conference Proceedings. United States of America.

Lilley, J. S. 2001. Nuclear Physics: Principles and Applications, Wiley. United States of America.

Maclear, L. G. A .1995. Cape Town Needs Groundwater: A Note on the Potential of the Cape Flats Aquifer Unit to Supply Groundwater for Domestic Use in the Cape Town Metropolitan Area. Government Print, South Africa.

Magill, J., Pfennig, G. and Galy, J. 2006. Chart of nuclides, 7th edition. European Communities. Germany.

Mantengu, N. R. 2016. Radiation Shielding Calculations using the FLUKA transport code for a Radioactive-ion Beam Facility at iThemba LABS. Masters Thesis, University of Zulu Land, South Africa (unpublished).

Mook, W. G. 2001. Environmental Isotopes in the Hydrological Cycle Principles and Applications. IHP-V Technical Documents in Hydrology, No 39. UNESCO – IAEA.

Moura, M. L. and Kroonenberg, S. B. W. 1988. Major and minor elements geochemistry and mineralogy of four soil profiles from Araracuara, Colombian Amazons. Vol. 15. Catena Verlag. W. Germany.

Musekiwa, C. and Majola, K. 2011. Groundwater vulnerability map for South Africa. Council of Geoscience (Western Cape Unit), South Africa.

National Council on Radiation Protection. 1977. Radiation protection guidelines for 0.1-100 MeV particle accelerator facilities. MD: NCRP; NCRP Report No. 51. Bethesda.

Nilsson, B. and Brahme, A. 2014. Interaction of ionising radiation with matter. In: Brahme, A. and Budinger, T. F (eds). Comprehensive biomedical physics: Nuclear medicine and molecular imaging, Vol. 1. Elsevier, USA.

Osipenko, M., Ripani, M., Alba, R., Ricco, G., Schillaci, M., Barbagallo, M., Boccaccio, P., Celentano, A., Colonna, N., Cosentino, L., Del Zoppo, A., Di Pietro, A., Esposito, J., Figuera, P., Finocchiaro, P., Kostyukov, A., Maiolino, C., Santonocito, D., Scuderi, V. and Viberti, C. M. 2013. Comprehensive measurement of neutron yield produced by 62 MeV protons on Beryllium target. Cornell University Press. New York.

Provincial Government of the Western Cape. 2005. Western Cape Provincial Spatial Development Framework: The Western Cape Province Today. https://www.westerncape.gov.za/text/2005/12/1-3_cfgdtdc_pages_1-50_web.pdf (cited: 20/03/2017).

Pynn, R. 2009. Introduction to neutron scattering. Springer Science+Business Media LLC, New York.

Ralph, C. H. 2004. Basic Groundwater Hydrology. U.S Geology Survey Water-Supply Paper 2220. United States Of America.

Rebelo, A. G., Holmes, P. M., Dorse, C. and Wood, J. 2010. Impacts of urbanization in a biodiversity hotspot: Conservation challenges in Metropolitan Cape Town. South African Journal of Botany. South Africa.

Robertson, D. E. and Carpenter, R. 1970. Neutron Activation Techniques for the Measurement of Trace Metals in Environmental Samples. National Information Centre, United States Atomic Energy Commission. United State of America.

Rosman, K. J. R. and Taylor, P. D. P. 1999. Pure Appl. Chem. 71, 1593 – 1607. European Commision. Belgium.

Saha, G. B. 2001. Physics and radiology of nuclear medicine. Springer Science and Business Media, New York.

Schulze, W. 1969. Activation analysis: Some basic principles. In: Lenihan, J. M. A. and Thomson, S. J (eds). Advances in activation analysis, Vol. 1. Academic Press, London.

Sherer, M. A. S., Visconti, P. J., Ritenour, E. R. and Haynes, K. W. 2014. Radiation protection in medical radiography 8th edition. Elsevier, Missouri.

Šrámek, O and Zeman, J. 2004. Introduction to environmental hydrochemistry. MASARYK UNIVERSITY IN BRNO Faculty of Science. ISBN 80-210-3586-2. Czech Republic.

Sublet, J. C., Mann, F. M. and Goddard, A. J. H. 1990. Proceedings of a specialists meeting on neutron activation cross section for fission and fusion energy applications: A-one group averaged cross-section benchmark for fusion activation studies. Organisation for Economic Co-operation and Development, Paris, France.

Tadross and Johnson, M. and Johnston, P. 2012. Sub-Saharan African Cities: A five-City Network to Pioneer Climate Adaptation through Participatory Research & Local Action: Climate Change Projections for Cape Town: Adding value through downscaling. Local Governments for Sustainability. South Africa.

Terrie, K. and Boguski, P. E. 2006. Understanding units of measurements. Issue 2. Centre of Hazardous Substance Research. New York.

Todd, D. K., Tinlin, R. M., Schmidt, K., D. and Everett, L. G. 1976. Monitoring groundwater quality: Monitoring methodology. National Technical Information Service, Las Vegas, Nevada.

Tröjbom, M., Söderbäck, M. B., Kalinowski, B. and Kärnbränslehantering, A. A. B. 2008. Hydrochemistry of surface water and shallow groundwater: Site descriptive modelling SDM-Site Laxemar. SKB Rapport R-08-46. Stockholm.

United Nations Scientific Committee on the Effects of Atomic Radiation. 2008. Sources, effects and risks of ionizing radiation. Vol. I. Sources. New York. http://www.unscear.org/unscear/en/publications/2008_1.html. (cited: 31/01/2018).

Van Rooyen, J. 2006. The science of radiation protection. South African Nuclear Energy Corporation. South Africa.








World Health Organisation. 2011. Guidelines for drinking-water quality, 4th Ed. <http://www.who.int>. (cited: 30/01/2018).

Wu, C. 2008. Groundwater occurrence in the Table Mountain area of Cape Town, South Africa. Masters Thesis, University of the Western Cape (unpublished).

Yashima, H., Terunuma, K., Nakamura, T., Hagiwara, M., Kawata, N. and Baba, M. 2004. Measurements of neutron activation cross sections for major elements of water, air and soil between 30 and 70 MeV. Journal of Nuclear Science and Technology, Japan.

Appendices

Appendix 1. Letter of approval from the Department of Water and Sanitation.

	water & sanitation Department: Water and Sanitation REPUBLIC OF SOUTH AFRICA
WESTERN CAPE Private Bag X16, Sanlamhof, 7532, 52 Voortrekker Road, Bellville, 7530	
 021 941 6185	 Mr. Warren Dreyer
 086 556 9985	 27/2/1/G200/A11
 dreyerw@dws.gov.za	
Attention: Thandazile Marazula	
iThemba LABS Old Faure Road Faure Cape Town 7131	
Dear Sir	
WATER USE QUERY: PROPOSED USE OF BOREHOLES AT Ithemba LABS FOR THE RESEARCH PURPOSES, ERF 41967, FAURE, CAPE RD	
The Department has perused the information provided. Your study, which is based on the investigation of activation of stable isotopes in soil and groundwater from a radionuclide production facility will require you to sample soil and groundwater once for the analysis using the XRF and ICPMS techniques. Samples will be taken from eight (8) on the iThemba LABS property. The Department confirms that the proposed water use does not require water use authorization in terms of the National Water Act, 1998 (Act No. 36 of 1998).	
All applicable sections of the National Water Act, 1998 (Act No. 36 of 1998) must be adhered to at all times.	
Should you have any further questions in this regard you are welcome to contact the above mentioned official.	
Yours faithfully	
	
ACTING CEO: BERG-OLIFANTS PROTO CMA Letter signed by: Mrs. H. Lyons Designation: Control Industrial Technician: Berg-Olifants Proto CMA Date: 7/4/2017	

Appendix 2: Hydro-chemical results from Bemlab for two deep boreholes (B2B and B3B).



16 Van der Berg Crescent
Gant's Centre
Strand

Tel. (021) 853-1490
Fax (021) 853-1423

E-Mail admin@bemlab.co.za

P O Box 684
Somerset Mall,
7137

Vat Reg. Nr. 4200161414

CERTIFICATE OF ANALYSES

Report Nr.: **WT005832_b.DOC** (Supplement to Test Report No.: WT005832_a.DOC)

Thandazile Marazula
iThemba Labs
Old Faure Road
Faure
7131

Date received: **20-04-2017**
Order nr.: **NPO-0048332**

Sampled by client

Water Analyses Report

Drinking water evaluation

Origin	Lab. Nr.	pH @ 25°C	EC @ 25°C mS/m	Na mg/l	K mg/l	Ca mg/l	Mg mg/l	Fe mg/l	Cl mg/l	CO ₃ ²⁻ mg/l	HCO ₃ ⁻ mg/l	SO ₄ mg/l	B mg/l	Mn mg/l	Cu mg/l	Zn mg/l	P mg/l	NH ₄ -N mg/l	NO ₃ -N mg/l	*F mg/l
B2B	5832	7.0	1052	1605.3	13.0	303.2	154.3	5.1	3485.3	0.0	329.8	111	0.19	0.50	<0.02	<0.03	0.34	0.40	<0.36	0.3
B3B	5833	6.9	1067	1728.0	10.3	320.0	165.2	7.9	3556.6	0.0	321.7	152	0.19	0.36	0.03	<0.03	0.21	0.52	<0.36	0.3
Norm		≥5.0-≤9.7	≤170	≤200.0				≤2.0	≤300.0			≤500	≤2.40	≤0.40	≤2.00	≤5.00		≤1.50	≤11.00	≤1.5

Origin	Lab. Nr.	*TDS mg/l	Date Tested	Date Sampled	Temperature at reception (°C)	*Langelier index
B2B	5832	6300.0	21-04-2017	20/04/2017	23.0	0.4
B3B	5833	6380.0	21-04-2017	20/04/2017	23.4	0.4
Norm		≤1200.0				≥-0.5-≤0.5

* = Not SANAS Accredited

Norms according to SANS 241-1:2015.

Microbiological analyses

Origin	Lab. Nr.	Total Bacteria cfu/1ml	Total Coliforms cfu/100 ml	E. coli cfu/100ml	Date Tested	Date Sampled	Temperature at reception (°C)
B2B	5832	1250	>2420	4	20/04/2017	20/04/2017	23.0
B3B	5833	450	>2420	<1	20/04/2017	20/04/2017	23.4
Norm		≤1000	≤10	<1			

Norms for [drinking water](#) can vary depending on the origin.
Norms according to SANS 241-1:2015.


Appendix 3: Calculated ionic balance and percentage error for deep borehole (B2B).


B2B							
Cations	Concentration (mg/L)	Molar Mass (g/mol)	Valence (meq/L)	Anions	Concentration (mg/L)	Molar Mass (g/mol)	Valence (meq/L)
Na ⁺	1605.3	23	69.79565217	Cl ⁻	3485.3	36	96.81388889
K ⁺	13	40	0.325	HCO ₃ ⁻	329.8	61	5.406557377
Ca ⁺⁺	303.2	20	15.16	SO ₄ ⁻	111	96	1.15625
Mg ⁺⁺	154.3	12	12.85833333				
Fe ⁺⁺	5.1	28	0.182142857				
Total			98.32112836				103.3766963
Ionic balance error (%)							-2.506505913


Appendix 4: Calculated ionic balance and percentage error for deep borehole (B3B).

B3B							
Cations	Concentration (mg/L)	Molar Mass (g/mol)	Valence (meq/L)	Anions	Concentration (mg/L)	Molar Mass (g/mol)	Valence (meq/L)
Na ⁺	1728	23	75.13043478	Cl ⁻	3556.6	36	98.79444444
K ⁺	10.3	40	0.2575	HCO ₃ ⁻	321.7	61	5.273770492
Ca ⁺⁺	320	20	16	SO ₄ ⁻	152	96	1.583333333
Mg ⁺⁺	165.2	12	13.76666667				
Fe ⁺⁺	7.9	28	0.282142857				
Total			105.4367443				105.6515483
Ionic balance error							-0.101760245

Appendix 5: Hydro-chemical results from Bemlab for shallow borehole (B3A).



Part of 



T0654

16 Van der Berg Crescent
Gant's Centre
Strand

P O Box 684
Somerset Mall,
7137

Tel. (021) 853-1490
Fax (021) 853-1423
E-Mail admin@bemlab.co.za

Vat Reg. Nr. 4200161414

CERTIFICATE OF ANALYSES

Report Nr.: WT006158.DOC

Thandazile Marazula
iThemba Labs
Old Faure Road
Faure
7131

Date received: 25-04-2017
Order nr.: B3A

Sampled by client

Water Analyses Report

Irrigation evaluation

Origin	Lab. Nr.	pH @ 25°C	EC @ 25°C mS/m	Na mg/l	K mg/l	Ca mg/l	Mg mg/l	Fe mg/l	Cl mg/l	CO ₃ ²⁻ mg/l	HCO ₃ ⁻ mg/l	SO ₄ mg/l	B mg/l	Mn mg/l	Cu mg/l	Zn mg/l	P mg/l	NH ₄ -N mg/l	NO ₃ -N mg/l	*F mg/l
B3A	6158	7.1	951.6	1400.0	12.7	293.8	159.2	5.8	2810.0		357.0	184	0.21	0.32	0.03	0.08	0.04	<0.28	1.88	0.5
Norm		≥5.5-≤9.0	≤270.0	≤70.0				≤1.5	≤100.0		≤150.0		≤1.00	≤10.00	≤1.00	≤1.00				

Origin	Lab. Nr.	*TDS mg/l	Date Tested	Date Sampled	*OP (kPa)	*Adj. SAR	*Langelier index	*Class	Temperature at reception (°C)
B3A	6158	6090.0	26-04-2017	25/04/2017	342.6	20.3	0.5	C4-S4	25.3
Norm							≥-0.5-≤0.5		

* = Not SANAS Accredited

See [analyses interpretation](#) for assistance with interpreting the report and relevance of each analyses.

The classification of the water has the following meaning:
 C4: Very high salinity water - Not suitable for irrigation under normal conditions.
 S4: Very high sodium water - Generally unsuitable for irrigation except at C1 and perhaps C2, where gypsum can be applied.

Comments:
 With sodium concentrations greater than 200 mg/l severe problems with sodium toxicity is expected. The water is not suitable for irrigation.
 With chloride concentrations greater than 200 mg/l severe problems with Cl toxicity is expected. The water is not suitable for irrigation.
 A positive value for the Langelier index indicate a tendency for calcium carbonate to precipitate, whereas a negative value indicate that the water will dissolve calcium carbonate. The latter water is regarded as aggressive and corrosive. If there is a tendency for calcium to precipitate it will result in a higher SAR value than the value shown in the table above and the sodium hazard with use of this water will therefore be greater.

\\172.17.48.240\bemlab\bem\lms\reports\2017\water\word\wt006158.doc

This Laboratory participate in the Agrilisa proficiency and SABS water testing scheme

Page 1 of 3

Appendix 6: Calculated ionic balance and percentage error for shallow borehole (B3A).

B3A							
Cations	Concentration (mg/L)	Molar Mass (g/mol)	Valence (meq/L)	Anions	Concentration (mg/L)	Molar Mass (g/mol)	Valence (meq/L)
Na ⁺	1400	23	60.86956522	Cl ⁻	2810	36	78.05555556
K ⁺	12.7	40	0.3175	HCO ₃ ⁻	357	61	5.852459016
Ca ⁺⁺	293.8	20	14.69	SO ₄ ⁻	184	96	1.916666667
Mg ⁺⁺	159.2	12	13.26666667				
Fe ⁺⁺	12.7	28	0.453571429				
Total			89.59730331				85.82468124
Ionic balance error							2.150598218

Appendix 7: Hydro-chemical results from Bemlab for deep borehole (B1B).



16 Van der Berg Crescent
Gant's Centre
Strand

Tel. (021) 853-1490
Fax (021) 853-1423

E-Mail admin@bemlab.co.za

P O Box 684
Somerset Mall,
7137

Vat Reg. Nr. 4200161414

CERTIFICATE OF ANALYSES

Report Nr.: **WT007134_b.DOC** (Supplement to Test Report No.: WT007134_a.DOC)

Thandazile Marazula
iThemba Labs
Old Faure Road
Faure
7131

Date received: **16-05-2017**
Time received: **00:00**
Order nr.: **NPO-0048332**

Sampled by client

Water Analyses Report

Irrigation evaluation

Origin	Lab. Nr.	pH @ 25°C	EC @ 25°C mS/m	Na mg/l	K mg/l	Ca mg/l	Mg mg/l	Fe mg/l	Cl mg/l	CO ₃ ²⁻ mg/l	HCO ₃ ⁻ mg/l	SO ₄ mg/l	B mg/l	Mn mg/l	Cu mg/l	Zn mg/l	P mg/l	NH ₄ -N mg/l	NO ₃ -N mg/l	*F mg/l
B1B	7134	7.2	482	642.9	5.1	140.7	102.7	0.35	1160.0		889.0	137	0.43	0.05	<0.02	0.24	<0.01	<0.28	<0.36	0.3
Norm		≥5.5-≤9.0	≤270	≤70.0				≤1.50	≤100.0		≤150.0		≤1.00	≤0.10	≤1.00	≤1.00				

Origin	Lab. Nr.	*TDS mg/l	Date Tested	Date Sampled	*OP (kPa)	*Adj. SAR	*Langelier index	*Class	Temperature at reception (°C)
B1B	7134	3085.0	17-05-2017	16/05/2017	173.6	12.5	0.9	C4-S4	24.0
Norm							≥-0.5-≤0.5		

* = Not SANAS Accredited

See [analyses interpretation](#) for assistance with interpreting the report and relevance of each analyses.

The classification of the water has the following meaning:

C4: Very high salinity water - Not suitable for irrigation under normal conditions.

S4: Very high sodium water - Generally unsuitable for irrigation except at C1 and perhaps C2, where gypsum can be applied.

Comments:

With sodium concentrations greater than 200 mg/l severe problems with sodium toxicity is expected. The water is not suitable for irrigation.

With chloride concentrations greater than 200 mg/l severe problems with Cl toxicity is expected. The water is not suitable for irrigation.

A positive value for the Langelier index indicate a tendency for calcium carbonate to precipitate, whereas a negative value indicate that the water will dissolve calcium carbonate. The latter water is regarded as aggressive and corrosive. If there is a tendency for calcium to precipitate it will result in a higher SAR value than the value shown in the table above and the sodium hazard with use of this water will therefore be greater.

Microbiological analyses

\\172.17.63.10\bemlab\bemlms\reports\2017\water\word\wt007134_b.doc


This Laboratory participate in the Agrilasa proficiency and SABS water testing scheme


Page1of3


Appendix 8: Calculated ionic balance and percentage error for deep borehole (B1B).

B1B							
Cations	Concentration (mg/L)	Molar Mass (g/mol)	Valence (meq/L)	Anions	Concentration (mg/L)	Molar Mass (g/mol)	Valence (meq/L)
Na ⁺	642.9	23	27.95217391	Cl ⁻	1160	36	32.22222222
K ⁺	5.1	40	0.1275	HCO ₃ ⁻	889	61	14.57377049
Ca ⁺⁺	140.7	20	7.035	SO ₄ ⁻	137	96	1.427083333
Mg ⁺⁺	102.7	12	8.558333333				
Fe ⁺⁺	0.35	28	0.0125				
Total			43.68550725				48.22307605
Ionic balance error (%)							-4.937045745

Appendix 9: Hydro-chemical results from Bemlab for shallow boreholes (B1A and B2A).



www.bemlab.co.za
Part of 



T0654

18 Van der Berg Crescent
Gant's Centre
Strand

Tel. (021) 853-1490
Fax (021) 853-1423
E-Mail admin@bemlab.co.za

P O Box 684
Somerset Mall,
7137

Vat Reg. Nr. 4200161414

CERTIFICATE OF ANALYSES

Report Nr.: WT006408.DOC

Thandazile Marazula
iThemba Labs
Old Faure Road
Faure
7131

Date received: 02-05-2017
Order nr.: B1A B2A

Sampled by client

Water Analyses Report

Origin	Lab. Nr.	pH @ 25°C	EC @ 25°C mS/m	Na mg/l	K mg/l	Ca mg/l	Mg mg/l	Fe mg/l	Cl mg/l	CO ₃ ²⁻ mg/l	HCO ₃ ⁻ mg/l	SO ₄ mg/l	B mg/l	Mn mg/l	Cu mg/l	Zn mg/l	P mg/l	NH ₄ -N mg/l	NO ₂ -N mg/l	*F mg/l
B1A	6408	7.5	312.4	583.1	1.0	61.4	33.6	1.6	510.0		1014.0	139	0.33	<0.03	<0.02	0.08	0.03	<0.28	<0.36	1.2
B2A	6408	7.1	828.4	1097.3	4.6	241.3	145.3	4.9	2420.0		440.0	146	0.17	0.42	<0.02	0.10	0.05	<0.28	1.01	0.3

Origin	Lab. Nr.	*TDS mg/l	Date Tested	Date Sampled	Temperature at reception (°C)
B1A	6408	1999.0	03-05-2017	02/05/2017	22.8
B2A	6408	5302.0	03-05-2017	02/05/2017	22.5

* = Not SANAS Accredited

Origin	Lab. Nr.	Total Bacteria cfu/1ml	Total Coliforms cfu/100 ml	E. coli cfu/100ml	Date Tested	Date Sampled	Temperature at reception (°C)
B1A	6408	920	37	<1	02/05/2017	02/05/2017	22.8
B2A	6408	1100	20	<1	02/05/2017	02/05/2017	22.5

Statement: The reported results may be applied only to samples received. Any recommendations included with this report are based on the assumption that the samples were representative of the source from which they were taken.

Notes:
 To ensure sample integrity, samples are stored only for seven days after release of the report. Thereafter it is disposed of and a fresh sample will be required if additional analyses are requested.
 Results marked with "Not SANAS Accredited" in this report are not included in the SANAS Schedule of Accreditation for this laboratory. These results relate to the items tested. This test report shall not be reproduced except in full, without written approval of the laboratory.
 CO₃²⁻ analysis only applicable in case of pH > 7.5.
 Refer to [website](#) for uncertainty of measurement and referenced methods.
Sample condition: Water sample temperatures were higher than 10°C, but was delivered at Bemlab within the 6 hour timeframe.

\\172.17.48.240\bemlab\bem\lms\reports\2017\water\word\wt006408.doc

This Laboratory participate in the Agrilasa proficiency and SABS water testing scheme

Page 1 of 2

Appendix 10: Calculated ionic balance and percentage error for shallow borehole (B1A).

B1A							
Cations	Concentration (mg/L)	Molar Mass (g/mol)	Valence (meq/L)	Anions	Concentration (mg/L)	Molar Mass (g/mol)	Valence (meq/L)
Na ⁺	583.1	23	25.35217391	Cl ⁻	510	36	14.16666667
K ⁺	1	40	0.025	HCO ₃ ⁻	1014	61	16.62295082
Ca ⁺⁺	122.8	20	6.14	SO ₄ ⁻	139	96	1.447916667
Mg ⁺⁺	33.6	12	2.8				
Fe ⁺⁺	1.6	28	0.057142857				
Total			34.37431677				32.23753415
Ionic Balance error (%)							3.207811504

Appendix 11: Calculated ionic balance and percentage error for shallow borehole (B2A).

B2A							
Cations	Concentration (mg/L)	Molar mass	Valence (meq/L)	Anions	Concentration (mg/L)	Molar Mass (g/mol)	Valence (meq/L)
Na ⁺	1097.3	23	47.70869565	Cl ⁻	2420	36	67.22222222
K ⁺	4.6	40	0.115	HCO ₃ ⁻	440	61	7.213114754
Ca ⁺⁺	241.3	20	12.065	SO ₄ ⁻	146	96	1.520833333
Mg ⁺⁺	145.3	12	12.10833333				
Fe ⁺⁺	4.9	28	0.175				
Total			72.17202899				75.95617031
Ionic balance error (%)							-2.554639388

Appendix 12: Geochemical results for major elements from soil samples analysed from the University of Cape Town, Department of Geology for B1A, B1B, B2A, B2B, B3A and B3.

Sample	SiO2	TiO2	Al2O3	Fe2O3	MnO	MgO	CaO	Na2O	K2O	P2O5	SO3	NiO	H2O-	LOI	Total
	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%
B1A 3m	89.82	0.18	3.54	1.06	b.d.	0.21	1.88	0.34	0.34	0.01	b.d.	b.d.	0.01	1.81	99.21
B1A 4m	94.10	0.14	1.40	0.44	b.d.	0.04	1.38	0.24	0.15	0.01	b.d.	b.d.	0.02	1.21	99.14
B2A 3m	90.45	0.09	2.26	5.66	0.01	0.04	0.27	0.21	0.18	0.16	0.01	0.02	-0.04	0.70	100.02
B2A 4m	92.51	0.12	3.78	0.79	b.d.	0.07	0.37	0.30	0.48	0.02	b.d.	b.d.	0.02	0.77	99.24
B3A 3m	76.35	0.20	3.63	1.48	0.02	1.22	8.07	0.33	0.61	0.02	0.02	b.d.	0.00	7.25	99.19
B3A 4m	93.11	0.10	1.41	2.60	0.01	0.08	0.71	0.23	0.27	0.03	0.01	b.d.	0.10	0.80	99.47
B1B 3m	96.02	0.13	1.63	0.58	0.01	0.01	0.05	0.24	0.26	0.01	b.d.	b.d.	0.00	0.34	99.26
B1B 4m	96.63	0.13	1.41	0.51	b.d.	b.d.	0.10	0.23	0.25	0.01	b.d.	b.d.	0.05	0.26	99.59
B2B 3m	91.30	0.14	2.57	3.68	0.01	0.04	0.14	0.26	0.49	0.08	0.01	0.02	-0.06	0.64	99.34
B2B 4m	91.96	0.15	2.71	2.86	0.02	0.07	0.09	0.27	0.57	0.05	b.d.	b.d.	0.03	0.51	99.29
B3B 3m	92.92	0.16	2.85	1.13	0.01	0.05	0.49	0.27	0.58	0.03	0.10	b.d.	-0.04	0.81	99.37
B3B 4m	91.07	0.17	3.24	1.20	0.01	0.21	0.94	0.36	0.66	0.02	0.10	b.d.	0.03	1.14	99.15

Appendix 13: Geochemical results for minor elements from soil samples analysed from the University of Cape Town, Department of Geology for B1A, B1B, B2A, B2B, B3A and B3B.

	Zn	Nb	Zr	Y	Sr	Rb	Th	Pb	Mn	Cr	V	F	S	Cl	Ba
	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm
B1A 5m	4.701	4	40.48	9.396	36.45	7.93	<5	4	31.1	12.839	5.892	335.1	287.9	668.6	41.9
B2A 5m	7.877	<4	42.371	10.104	21.77	23.8	4.87	10.8	44.074	30.465	10.968	470.7	330.5	867.9	84
B3A 5m	4.669	<4	37.203	10.188	35.05	12.31	<5	5	108.053	16.452	18.83	486.5	287.7	563.3	61.4
B1B 5m	4.602	<4	34.047	8.274	13.87	11.28	<6	3	31.859	16.216	8.258	339.5	282.5	270.4	31.8
B2B 5m	5.779	<4	33.834	9.126	19.07	26.09	<7	7	162.333	25.281	16.648	489.2	357.1	224.9	83.3
B3B 5m	9.873	<4	59.729	11.934	45.5	29.78	<8	6	62.28	26.215	14.491	497	782.1	341.3	119.7
B1A 4m	6.355	<4	31.971	8.118	50.89	18.43	<9	5	24.237	24.757	20.612	483.1	326.7	1863.8	75
B2A 4m	5.852	<4	22.762	8.67	13.72	10.1	<10	8.95	107.857	20.906	52.843	660.1	281.7	482.8	27.9
B3A 4m	6.678	6.07	35.297	10.83	266.86	28.85	<11	8.45	136.591	22.547	20.354	612.3	339.9	1426.2	134.1
B1B 4m	4.362	<4	28.637	7.392	12.93	11.58	<12	<5	21.71	13.571	10.876	394.2	276.4	208.2	38.2
B2B 4m	4.966	<4	35.65	7.806	17.46	22.26	<13	8.27	132.545	19.181	23.511	636.3	305.8	291.4	82.5
B3B 4m	6.597	<4	40.969	12.234	38.09	27.08	<14	7.81	52.954	21.909	15.158	422.3	833.7	260.7	100.1

Appendix 14 (a): Geochemical results for trace elements in soil samples from 7 Li to 93 Nb analysed from the University of Cape Town, Department of Geology for B1A, B1B, B2A, B2B, B3A and B3B.

	7Li	45Sc	51V	52Cr	59Co	60Ni	65Cu	66Zn	85Rb	88Sr	89Y	90Zr	93Nb
	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm
	TPB 6/22/2017 2:01:33 PM												
x	0.036	0.003	0.015	0.139	0.008	0.099	0.079	0.399	-0.002	0.01	0.005	0.144	0.038
	BHV02 6/22/2017 2:06:48 PM												
x	4.414	35.76	290.1	279.0	45.08	117.7	142.0	118.1	8.515	389.7	26.79	185.0	18.56
%RSD	0.362	0.157	0.618	0.498	0.583	0.565	0.720	0.152	0.674	0.825	0.483	0.128	0.616
	B1A 6/22/2017 2:12:00 PM												
x	7.782	0.822	6.466	12.66	1.415	4.873	5.233	8.447	7.720	34.16	6.082	34.87	2.441
%RSD	0.619	0.923	0.222	0.812	0.399	0.437	0.852	0.898	0.686	0.214	0.625	0.938	1.076
	B1B 6/22/2017 2:17:13 PM												
x	6.999	0.868	10.07	19.33	1.590	5.224	4.957	8.535	10.49	13.33	4.639	31.23	2.987
%RSD	0.632	1.456	0.867	0.783	0.946	0.439	0.935	0.154	1.001	0.345	0.815	0.897	0.557

	B1A-4m 6/22/2017 2:22:26 PM												
x	13.57	2.529	16.63	18.08	3.490	6.809	7.263	14.85	16.03	48.38	8.449	27.75	3.092
%RSD	0.567	0.682	0.911	0.492	0.603	0.871	1.029	0.565	0.390	0.080	0.622	0.277	0.138
	B1B-4m 6/22/2017 2:27:39 PM												
x	7.113	1.052	13.25	12.43	1.954	5.222	7.446	12.12	10.38	12.22	5.577	27.77	2.448
%RSD	0.840	0.396	0.924	0.142	0.283	1.037	0.556	1.427	1.243	0.904	0.806	0.120	0.585
	B2A 6/22/2017 2:32:52 PM												
x	23.09	2.473	14.11	24.66	1.951	7.920	12.96	15.06	21.72	19.99	7.938	39.42	2.745
%RSD	1.018	1.179	0.364	0.081	0.975	0.945	0.991	1.083	0.384	0.206	0.692	0.076	0.876
	B2B 6/22/2017 2:38:06 PM												
x	13.27	1.474	21.11	23.25	2.748	11.56	9.543	17.55	24.26	18.52	7.064	31.03	3.691
%RSD	1.139	0.461	0.871	1.349	0.481	0.750	0.857	0.951	0.368	0.812	0.984	1.196	0.347
	B2B-4m 6/22/2017 2:43:20 PM												
x	13.49	1.453	23.20	14.13	2.432	5.739	4.494	11.09	20.48	16.69	5.439	29.99	3.148
%RSD	0.450	1.288	1.364	0.575	0.724	1.127	0.384	0.531	1.004	0.752	0.505	0.418	0.164

	B3A-4m 6/22/2017 2:48:34 PM												
x	13.64	2.226	16.74	15.99	3.174	6.521	4.606	11.90	26.34	243.6	9.683	28.96	4.578
%RSD	0.372	0.346	1.046	0.285	0.797	0.614	0.692	1.610	0.673	0.946	0.097	0.507	0.811
	B3B-4m 6/22/2017 2:53:49 PM												
x	11.71	2.096	16.31	19.27	3.161	6.735	4.554	14.72	25.09	38.27	12.46	35.20	3.789
%RSD	0.805	1.241	0.370	0.531	0.670	1.279	1.246	1.348	0.171	0.341	0.730	0.635	0.842
	B3A-5m 6/22/2017 2:59:04 PM												
x	7.734	0.861	22.68	16.00	2.283	7.994	5.107	10.44	10.76	33.13	6.844	31.18	2.465
%RSD	0.148	1.124	1.085	0.357	0.393	1.265	1.070	1.283	0.534	0.727	0.690	0.677	0.047
	B3B-5m 6/22/2017 3:04:19 PM												
x	13.89	2.272	16.01	22.25	3.599	8.485	6.168	14.29	27.00	41.62	9.446	51.60	3.762
%RSD	0.941	0.603	0.942	0.868	1.024	0.786	0.217	0.526	0.999	0.584	0.968	0.796	0.571
	TPB 8/1/2017 11:52:47 AM												
x	0.004	0.001	0.011	0.086	0	0.023	-0.001	0.222	0	0.001	0	-0.003	0.001
	BHV02 8/1/2017												

	11:58:01 AM												
x	4.442	29.63	312.3	277.8	44.65	117.1	139.3	117.1	8.951	388.7	23.23	156.6	16.52
%RSD	1.072	0.825	0.786	0.806	0.74	0.61	1.175	0.854	0.757	0.222	0.648	0.338	0.985
	B2A 4m 8/1/2017 12:03:13 PM												
x	17.42	1.769	53.08	17.26	3.831	6.972	3.665	6.588	10.46	13.74	8.044	21.41	2.078
%RSD	1.236	1.138	1.153	0.764	0.256	0.230	1.537	0.137	0.790	1.231	0.489	0.257	0.235

Appendix 14 (b): Geochemical results for trace elements in soil samples continuing from 93 Nb to 174 Yb analysed from the University of Cape Town, Department of Geology for B1A, B1B, B2A, B2B, B3A and B3B.

137Ba	139La	140Ce	141Pr	144Nd	152Sm	153Eu	159Tb	160Gd	163Dy	165Ho	166Er	169Tm	174Yb
ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm
0.034	0.006	0.007	0.006	0.006	0.006	0.006	0.006	0.007	0.006	0.006	0.006	0.006	0.006
127.0	14.19	35.13	4.951	23.62	5.835	2.018	0.904	6.185	5.119	0.953	2.473	0.313	1.929
0.406	0.628	0.776	0.509	0.447	0.404	0.684	0.372	1.269	1.445	0.604	1.768	0.718	0.813
38.63	8.932	15.73	2.145	8.398	1.646	0.286	0.219	1.472	1.255	0.238	0.672	0.097	0.606
0.653	0.731	0.792	1.021	0.835	0.983	0.344	0.607	0.228	0.571	1.508	0.639	1.031	1.157
46.46	9.033	20.01	2.261	8.762	1.654	0.262	0.191	1.355	1.033	0.192	0.523	0.073	0.466
0.528	0.987	0.237	0.338	0.344	0.974	1.306	0.337	0.600	0.787	1.207	0.573	0.535	1.069
64.72	12.54	23.96	3.022	11.79	2.280	0.404	0.289	1.998	1.685	0.328	0.925	0.132	0.811
0.845	0.886	0.702	0.376	0.514	1.150	1.520	2.689	1.390	0.345	0.085	0.810	0.896	0.926
43.04	8.055	16.63	2.103	8.315	1.657	0.298	0.219	1.451	1.175	0.227	0.636	0.088	0.568
0.992	0.711	0.495	0.485	0.626	0.489	1.832	0.162	0.784	1.055	0.528	0.750	1.052	0.447
76.24	13.91	31.79	3.498	13.28	2.543	0.481	0.320	2.006	1.662	0.304	0.798	0.113	0.712
0.441	0.326	0.348	0.278	0.281	1.564	1.547	0.796	0.099	1.205	0.182	0.726	1.693	0.525
88.84	9.279	19.47	2.204	8.465	1.727	0.268	0.246	1.558	1.330	0.272	0.781	0.114	0.763
0.555	0.798	0.550	0.315	0.762	1.268	1.160	1.395	1.100	1.666	0.150	1.933	1.609	1.508

79.91	6.658	14.33	1.720	6.677	1.421	0.248	0.186	1.252	1.112	0.220	0.683	0.101	0.659
0.764	0.977	1.207	0.516	0.239	0.900	1.181	1.054	0.655	1.003	0.199	0.514	0.499	1.583
136.7	16.56	35.56	4.208	16.58	3.188	0.347	0.360	2.630	1.854	0.373	1.013	0.143	0.896
0.584	0.632	1.384	1.088	1.374	0.621	1.527	2.191	1.260	0.911	1.283	1.273	0.546	1.621
100.6	15.30	35.01	4.015	16.07	3.272	0.533	0.424	2.841	2.426	0.467	1.269	0.176	1.118
0.252	0.230	0.574	0.262	0.187	0.091	0.893	1.598	1.259	1.213	1.523	1.659	0.700	0.701
60.61	5.678	13.51	1.588	6.395	1.385	0.195	0.212	1.310	1.301	0.264	0.800	0.118	0.801
0.814	0.244	1.038	1.334	0.759	1.803	0.195	0.374	0.579	1.296	0.695	0.562	0.754	0.736
102.7	11.64	25.36	2.983	11.97	2.461	0.407	0.319	2.158	1.809	0.339	0.953	0.137	0.849
0.963	1.020	0.642	1.061	1.089	0.752	0.225	2.153	0.036	0.612	0.166	1.212	1.659	0.874
0.001	0.001	0	0	0	0	0	0	0	0	0	0	0	0
127.4	14.54	36.26	5.164	23.97	5.896	1.946	0.894	6.001	5.015	0.933	2.456	0.320	1.954
0.283	0.469	0.489	0.875	0.784	1.102	0.679	0.183	0.948	0.119	0.267	0.242	1.696	1.069
47.37	7.323	16.00	1.973	7.637	1.690	0.217	0.237	1.440	1.494	0.302	0.893	0.138	0.912
0.527	0.774	0.611	0.577	0.319	1.097	1.706	0.870	1.277	0.536	0.454	1.349	2.055	0.440

Appendix 14 (c): Geochemical results for trace elements in soil samples continuing from 174 Yb to 238 U analysed from the University of Cape Town, Department of Geology for B1A, B1B, B2A, B2B, B3A and B3B.

175Lu	178Hf	181Ta	208Pb	232Th	238U
ppm	ppm	ppm	ppm	ppm	ppm
0.007	0.009	0.009	0.011	0.006	0.011
0.291	4.251	1.075	1.927	1.222	0.392
2.315	0.513	1.108	0.299	1.169	1.610
0.096	1.057	0.220	3.387	2.459	0.421
0.219	1.165	0.383	0.264	0.723	0.587
0.073	0.968	0.189	3.245	2.931	0.365
0.453	0.132	0.817	0.266	0.516	0.238
0.126	0.859	0.263	5.103	4.558	0.459
0.332	0.275	0.749	0.553	0.507	0.079
0.084	0.810	0.202	3.856	2.380	0.304
1.266	0.096	0.572	0.279	0.963	0.924
0.106	1.268	0.232	9.595	5.446	0.970

0.238	0.938	0.594	0.604	0.409	0.356
0.110	0.959	0.309	6.823	4.018	0.666
1.542	0.837	0.748	0.503	0.415	0.296
0.106	0.923	0.248	7.264	3.672	0.766
0.194	0.071	0.318	0.496	0.538	0.405
0.137	0.889	0.384	6.794	7.726	0.562
1.113	0.945	1.367	0.558	0.193	0.959
0.171	1.096	0.322	6.871	5.919	2.439
1.612	0.809	0.485	1.286	0.186	1.074
0.123	0.969	0.187	4.500	3.007	0.635
0.195	0.693	0.667	0.779	0.421	0.057
0.130	1.646	0.295	5.296	4.678	2.092
1.266	0.882	0.213	0.742	0.245	0.721
0	0	0.001	0.003	0	0
0.294	4.188	1.286	1.429	1.164	0.401
1.517	0.229	0.909	1.085	1.251	0.899
0.142	0.682	0.221	7.312	4.862	0.927
0.667	0.697	0.414	0.023	0.407	2.013

Appendix 15 (a): Hydrochemical results of trace elements in water samples from 7 Li to 93 Nb analysed from the University of Cape Town, Department of Geology for B1A, B1B, B2A, B2B, B3A and B3B.

	7Li	45Sc	51V	52Cr	59Co	60Ni	65Cu	66Zn	85Rb	88Sr	89Y	90Zr	93Nb
	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppm	ppb	ppb	ppb
B1-A	6.268	2.626	9.189	28.01	4.514	13.42	16.84	63.64	6.423	1.403	2.068	2.295	0.049
B1B	23.42	3.001	14.68	21.42	46.94	502.1	< d.l.	35.30	16.36	4.933	< d.l.	10.43	0.020
B2A	25.78	4.527	11.15	30.12	2.132	82.82	0.291	43.85	19.81	3.459	0.345	1.859	0.035
B2B	21.71	4.055	12.02	26.31	28.91	26.33	< d.l.	25.19	19.63	4.491	0.258	3.376	0.023
B3A	30.22	4.902	13.10	31.41	10.63	32.71	1.476	89.02	21.50	3.889	1.539	0.960	< d.l.
B3B	23.27	3.926	13.83	26.59	6.850	26.03	< d.l.	29.12	15.09	4.259	< d.l.	1.272	0.016

Appendix 15 (b): Hydrochemical results of trace elements in water samples continuing from 93 Nb to 174 Yb analysed from the University of Cape Town, Department of Geology for B1A, B1B, B2A, B2B, B3A and B3B.

137Ba	139La	140Ce	141Pr	144Nd	152Sm	153Eu	159Tb	160Gd	163Dy	165Ho	166Er	169Tm	174Yb
ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb
87.00	0.534	0.611	0.108	0.717	0.180	0.042	0.049	0.348	0.292	0.068	0.212	0.022	0.150
393.9	0.021	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	0.001	0.002
231.2	0.438	0.666	0.076	0.314	0.048	0.016	0.006	0.052	0.101	0.013	0.025	0.007	0.043
522.2	0.324	1.071	0.118	0.406	0.116	< d.l.	0.010	0.118	0.066	0.017	0.019	0.005	0.027
220.9	0.913	1.904	0.236	0.814	0.248	0.037	0.033	0.175	0.257	0.074	0.234	0.057	0.381
197.9	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.

Appendix 15 (c): Hydrochemical results of trace elements in water samples continuing from 174 Yb to 238 U analysed from the University of Cape Town, Department of Geology for B1A, B1B, B2A, B2B, B3A and B3B.

175Lu	178Hf	181Ta	208Pb	232Th	238U
ppb	ppb	ppb	ppb	ppb	ppb
0.019	< d.l.	0.007	10.64	0.1	9.781
< d.l.	0.859	0.016	1.566	0.057	10.40
0.005	< d.l.	< d.l.	2.724	0.265	2.556
< d.l.	0.050	< d.l.	1.365	0.260	4.180
0.066	< d.l.	< d.l.	2.746	0.321	3.245
< d.l.	0.040	< d.l.	0.204	< d.l.	2.274