

Radiometry of milk and mineral water from the Western Cape

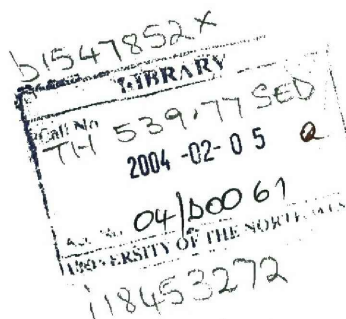
by

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A dissertation submitted in partial fulfilment for the degree of
**Masters of Science in Applied Radiation Science and
Technology at the University of North-West**

June 2003

Supervisor: Dr. R. T. Newman



Declaration

I hereby declare that the work contained in this dissertation is my own work and has not been submitted for a degree or other qualification in any other university or institution.



Lerato Sedumedi

Date: 29/07/03

ACKNOWLEDGEMENTS

I am greatly indebted to the following people who made this thesis possible;

Dr. R. T. Newman, my project supervisor, for guidance and support through the study, Mr. Dawid de Villiers, for his willingness to guide and assist me through the comprehension of the theory behind the study.

The Environmental Radioactivity group; Prof. Robbie Lindsay, Mr. Katse Maphoto, Ms. Tebogo Motlhabane, Mr. Ezekiel Patle Mohajane and Mr. Angelo Joseph for their helpful discussions and valuable suggestions.

I would also like to thank the iThemba MARST coordinator, Mr. Johann van Rooyen, for making it possible for me to acquire the needed computing skills.

Many thanks to the iThemba library staff; Ms. Audrey Sauls, Ms. Joanita Le Kay, Ms. Atline Maluleka and Ms. Wilhelmina Zaal for helping me obtain the background literature.

Dr. Given Mabala, your kind support and proof reading of the thesis are highly appreciated and Mrs Mandy Raman, to whom I am very grateful to for helping me at the end of this thesis.

I am very grateful to my parents and my family for their support and encouragement during the study.

Finally, thanks to God who gave me the strength and faith to make it all possible.

Radiometry of milk and mineral water from the Western Cape

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Abstract

A radiometric analysis of liquid milk and mineral water available at convenience stores in the Western Cape, South Africa was performed. The study, conducted by means of high-resolution γ -ray spectroscopy focussed on determining the activity concentrations of the anthropogenic nuclides ^{137}Cs (half-life 30 years), ^{131}I (half-life 8 days) in milk and natural radionuclides (^{226}Ra , ^{232}Th , ^{238}U and ^{40}K) in mineral water. The measurements were made using a high purity germanium detector (2.6 keV FWHM resolution at 1.33 MeV, 45 % relative efficiency) at the iThemba LABS facility, about 30 km from Cape Town. The full-energy peak absolute detection efficiency as a function of γ -ray energy was determined using a liquid standard source containing ^{152}Eu , ^{137}Cs and ^{60}Co .

Twelve samples of liquid milk (four samples per brand) were analysed. The presence of anthropogenic nuclides was not observed in milk samples. The average ^{137}Cs and ^{131}I activities were found to be consistent with zero to within statistical uncertainties. In view of this, the minimum detectable activities (MDA) of ^{137}Cs and ^{131}I were calculated using a commonly used prescription. The MDA concentrations were found to be 0.06 Bq/l and 0.07 Bq/l, respectively. It was only the 1461 keV line associated with the decay of ^{40}K that was clearly above background in the milk spectra. The average ^{40}K activity concentration was found to be 51.4 ± 0.4 Bq/l.

Six mineral water samples (three of one brand and three of another) were analysed. The samples were measured again after three weeks in order to allow for secular equilibrium to be reached. The average activity concentrations of the natural nuclides ^{226}Ra , ^{232}Th , ^{238}U and ^{40}K were found to be consistent with zero to within statistical uncertainties. The MDA concentrations for these nuclides were found to be 1.9 Bq/l (^{226}Ra), 0.2 Bq/l (^{232}Th), 0.1 Bq/l (^{238}U) and 0.8 Bq/l (^{40}K).

An upper limit to the yearly dose of an adult (older than 17 years) who ingests 37 l of milk and 80 l of mineral water, respectively, per year was calculated using the results from this study in conjunction with published dose conversion factors. In the case of milk the total annual dose due to the ingestion of ^{137}Cs and ^{131}I was found to be less than 0.086 μSv . For mineral water the total annual dose due to the ingestion of ^{226}Ra and ^{232}Th was found to be less than 46.3 μSv .

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Chapter 1

Introduction

Naturally occurring radioactive materials are present in our environment and in our bodies. Most food contains some natural radioactivity. Radionuclides such as ^{40}K , ^{210}Po , ^{226}Ra that occur naturally in the soil are incorporated metabolically into plants and ultimately find their way into food and water [Mer97]. Radionuclides released to the environment as a result of human activities add to the exposure received from natural radionuclides. Once present in the environment, these radionuclides can make their way into the food chain through different pathways.

There are two general pathways of radionuclides in the environment (Figure 1.1): the liquid pathway, which involves entry into the food chain via water [IAE89], these radionuclides move along with water through lakes, rivers and others get deposited on the surrounding soil or rocks. Then plants absorb the radionuclides in water the same way they absorb minerals depending on chemical properties of the nuclides. When the animals drink water some of the radionuclides in the water will concentrate in their bodies. The second pathway through the environment is the gaseous pathway when radionuclides are released into the air by human activities. They later fall back on land and may settle on the surface of the plants. When the animals eat the plants, they ingest the radionuclides on leaves. Eventually the plants and animals will become food for the people and therefore they provide a pathway for radionuclides to the people.

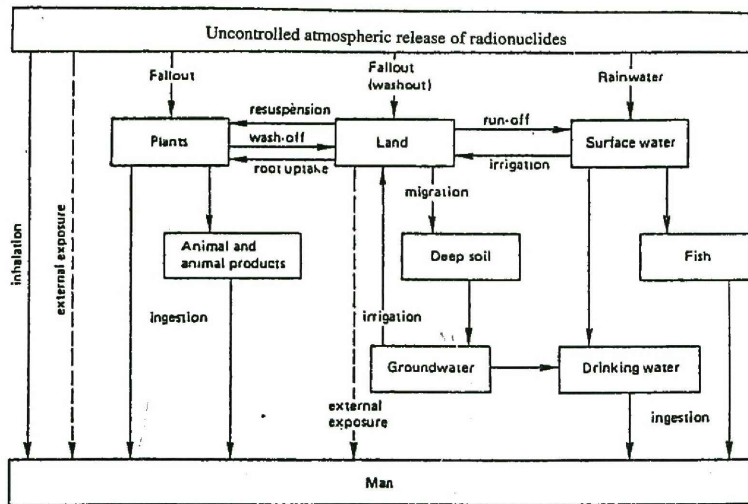


Figure 1.1: Main pathways of radionuclides to humans in the event of an uncontrolled atmospheric release of radioactivity. Taken from [IAE89].

1.1 Types of environmental radioactivity

Radionuclides are found in air, water and soil. Levels of radiation vary in the environment. People living in granite rich areas or on mineralised sands receive more background radiation than others, while people living or working at high altitudes receive more cosmic radiation. The main categories of radiation sources in our natural and working environment are:

- Primordial (formed since the creation of earth),
- Cosmogenic (formed as a result of cosmic radiation interactions with elements in the atmosphere) and
- Anthropogenic (formed due to human actions).

1.1.1 Primordial radionuclides

Primordial radionuclides have been present since the formation of the earth. They have long half-lives, often in the order of hundreds of millions of years (refer to Table 1.1). The most abundant and important of these natural radionuclides are ^{238}U , ^{235}U , ^{232}Th and their decay products, and a non-series nuclide ^{40}K (0.01 % in the body). ^{40}K cause an almost uniform radiation of the whole body, with a dose of 0.17 mSv per year [Poc83]. The primordial nuclei decay slowly and produce several radioactive decay products in their respective decay chains: the uranium, thorium and actinium series. The concentration of these naturally occurring radionuclides vary considerably, depending on the type of rock formation. The three important primordial radionuclides, ^{238}U , ^{232}Th and ^{40}K , are present in higher concentration in the earth's crust than in deeper layers of the planet core. The concentration of these nuclides in seawater is low. The decay chains of these naturally occurring radionuclides are shown in Figures 1.2, 1.3 and 1.4

Table 1.1: Some basic information on common natural radionuclides [WWW01].

Nuclide	Symbol	Half-life	Natural Activity
Uranium 235	U	7.04×10^8 yr	0.72 % of all natural uranium
Uranium 238	U	4.47×10^9 yr	99.3% of all the natural uranium
Thorium 232		1.14×10^{10} yr	1.6-20 ppm in all common rock types
Radium 226	Ra	1.60×10^3 yr	1.6 Bq/kg in limestone, 48 Bq/kg in igneous rocks
Radon 222	Rn	3.28 days	Noble gas
Potassium 40		1.28×10^{10} yr	In the soil ranges from 0.037-1.1 Bq/g

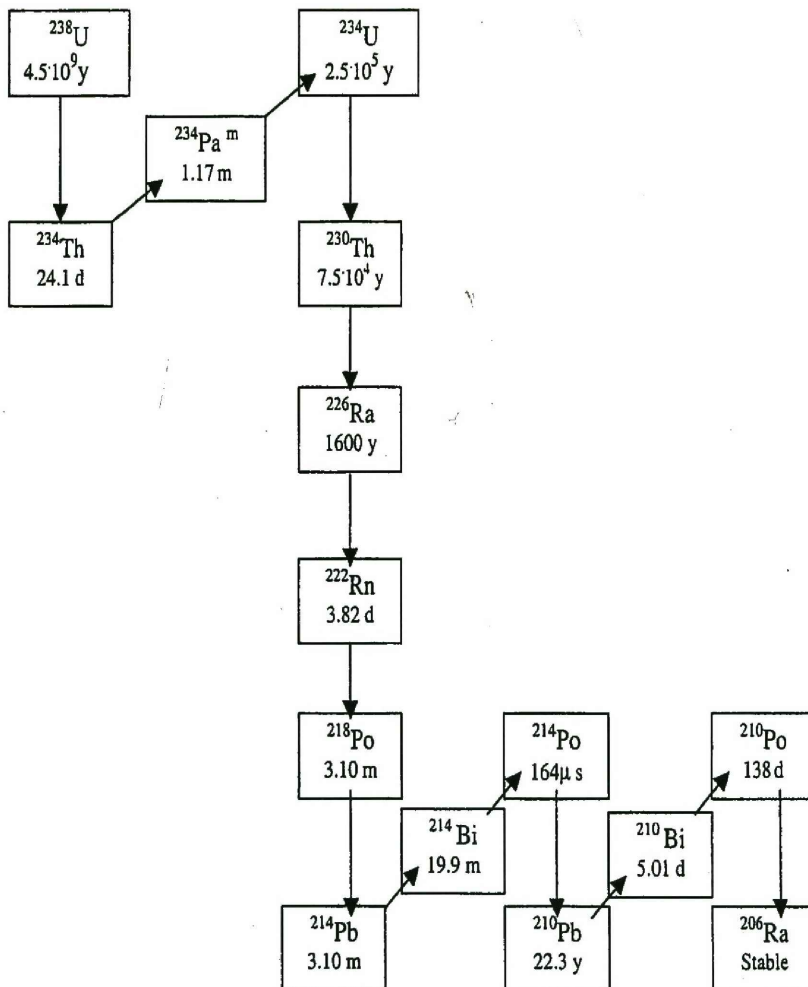


Figure 1.2: ^{238}U decay series [Tyk95].

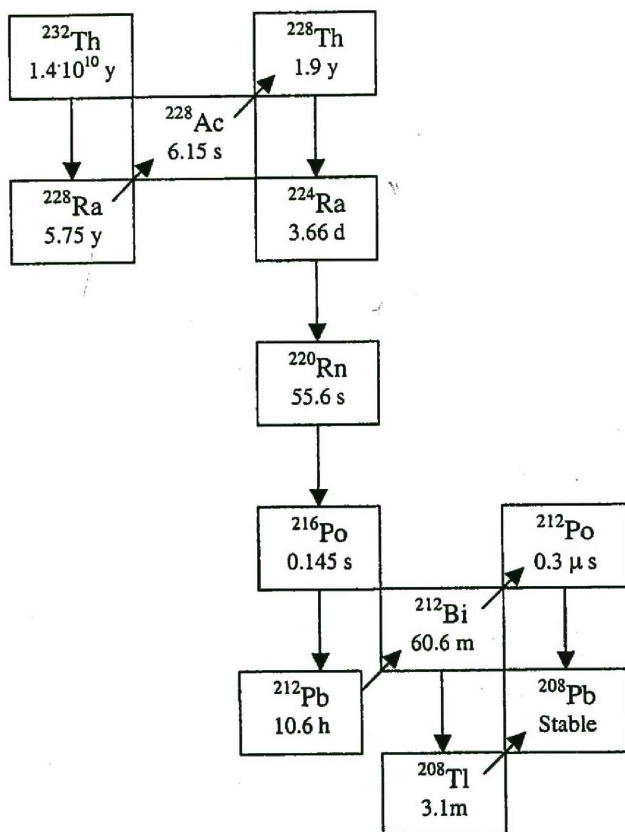


Figure .1.3: ^{232}Th decay series [Tyk95].

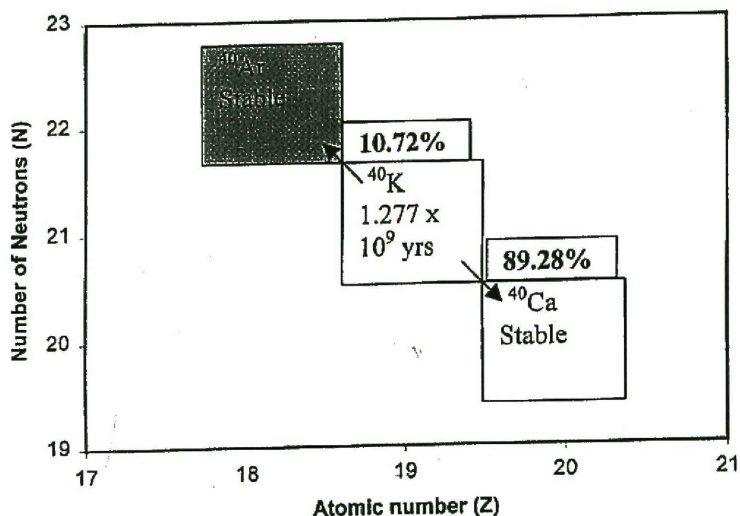


Figure 1.4: Schematic diagram of the decay chain of ^{40}K [Mal01].

1.1.2 Cosmogenic radionuclides

Cosmic radiation reaches the earth from interstellar space and from the sun. It is composed of a wide variety of penetrating radiations such as protons (87 %), α -particles (11 %), heavy ions (1 %) and electrons (1 %), which undergo many types of reactions with the elements they encounter in the atmosphere [Van02]. The atmosphere acts as a shield that reduces the intensity of cosmic radiation that reaches the earth's surface. Cosmogenic radionuclides can have long half-lives, but the majority have shorter half-lives than the primordial nuclides. Examples are ^{14}C (carbon-14) with the half-life of 5730 years and ^3H (tritium-3) with the half-life of 53.28 days. Figure 1.5 shows the annual dose contributed by natural and man-made (anthropogenic) radiation. Natural (primordial and cosmogenic) radionuclides contribute about 88 % of the annual dose. The

radioactive gas radon and its decay products make the largest contribution of about 48 % to the natural radiation dose to the general public [Van02]. On average radiation from natural material internal to the human is responsible for approximately 12 % of the dose to the population, while other contributors are external γ -radiation (14 %), thoron (4 %) and cosmic radiation (10 %). The average annual dose from incorporation of naturally occurring radionuclides in the body is 0.34 mSv, while from cosmic radiation is 0.30 mSv and from external radiation is 0.32 mSv per year [Poc83].

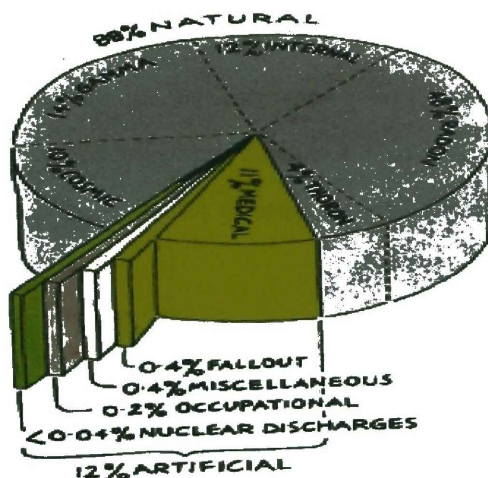


Figure 1.5: An overview of radiation in the environment. Taken from [WWW02].

1.1.3 Anthropogenic radionuclides

Natural (primordial and cosmogenic) radiation is not the only type of dose from ionising radiation that is received by people. Mankind has designed and built many types of apparatus that produce radiation, e.g. nuclear reactors. The radionuclides are released as fission products

(e.g. ^{131}I , ^{137}Cs , ^{90}Sr , ^{95}Zr , ^{144}Ce and ^{140}Ba) from the nuclear reactor fuel or as activation products (e.g. ^{134}Cs , ^{238}Pu , ^{240}Pu and ^{65}Zn) from activation of mobile material in the reactor. These radionuclides are carried into the atmosphere, where they remain for some time and are later dispersed and deposited around the world. The major sources of anthropogenic radiation are radionuclides introduced into the environment by the testing of nuclear weapons, routine operation of nuclear installations and during nuclear accidents.

The major fallout constituents are ^{90}Sr and ^{137}Cs with half-lives of 28 and 30 years respectively. The decay scheme of ^{137}Cs is presented on figure 1.6. From radiological standpoint, they are of concern because of their long half-lives and relatively high uptake by biosystems [Tyk95]. Strontium is chemically similar to calcium, therefore it is taken up in the bone. The chemical behaviour of caesium is similar to that of potassium, which is one of the most important biogenetic elements [Tyk95].

The testing of nuclear weapons in the atmosphere, took place from 1945 until 1980. Several hundred nuclear weapons were tested, mainly by the USA and the USSR (first test in 1949) and also by some other countries such as Great Britain (1952) and more in remote regions in the northern hemisphere [Tyk95]. Each nuclear explosion produced a large amount of different radionuclides. For example ^{90}Sr , ^{103}Ru and ^{137}Cs were produced.

Some of the most challenging environmental issues of today are those associated with the production of electricity. In particular there is a concern that greenhouse gas emissions from power stations (coal and natural gas) and vehicles are contributing to the global warming phenomenon, which eventually could cause sea levels to rise, resulting in communities along

coastal regions being displaced. Nuclear power stations have the big advantage that they contribute negligibly to the greenhouse effect while generating electricity. They do however pose risk to the public as a result of the routine and accidental release of radioactivity into the environment. For example, nuclear power stations produce small amounts of unwanted radioactive waste as a by-product, which is dispersed into the environment, and contributes to the population dose [Van02]. Some of these radionuclides include ^{137}Cs , ^{90}Sr , ^{131}I and a number of isotopes of plutonium.

There have been several accidents involving reactors [Tyk95], which have resulted in radioactive contamination of the site and the wider environment. In the case of a nuclear accident, a major risk to the personnel and the population can arise from breathing air and eating food contaminated by radioactive material released into the environment [Lil01]. Nuclear accidents which were of major significance from an environmental point of view are Windscale (1957), Idaho Falls (1961), Three Mile Island (1979), and in particular Chernobyl (1986) [Tyk95]. The worst accident in the history of nuclear accidents was the explosion of reactor No. 4 of the Chernobyl nuclear power station on 26 April 1986 (in the Ukraine) where radioactive material was released into the atmosphere and distributed to neighbouring countries [Mou00]. The radionuclides released were ^{144}Ce , ^{131}I , ^{239}Np , ^{103}Ru , ^{137}Cs and many others. Ingestion of ^{131}I (see Figure 1.7) was via milk from cows, which had eaten contaminated leafy green vegetables. The pathways of ^{137}Cs were either by deposition on leaves or uptake from soil through roots. Activity concentrations of ^{131}I measured in milk after the explosion of the Chernobyl reactor unit are shown in Table 1.2. These data give a good view of which countries in the world received more

radioactive fallout¹ from Chernobyl than others. In the longer term when ^{131}I was no longer a problem due to its short half-life of 8 days, care still had to be taken in the USSR in terms of ^{137}Cs contamination [Mou00].

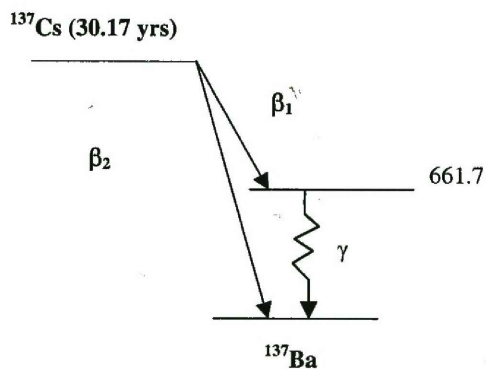


Figure 1.6: The decay scheme of ^{137}Cs .

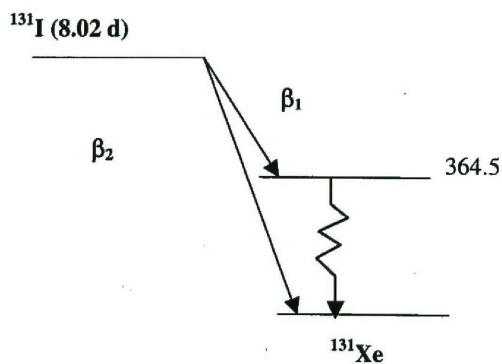


Figure 1.7: The decay scheme of ^{131}I .

¹ airborne radioactive debris.

Table 1.2: Measurements of ^{131}I activity concentrations in milk during May 1986 [Mou00].

Country	Foodstuff	^{131}I concentration (Bq/l)	Date
Albania	Fresh milk	< 800	5 May
Belgium	Farm milk	85 -170	8 May
Hungary	Milk, cows on pasture	100-700	1-2 May
Israel	Cow's milk	Mean of 21	3 May
Japan	Milk	0.4 - 3	4-6 May

1.2 Biological effects of nuclear radiation

Radiation causes ionisations in the molecules of living cells. Any form of radiation (charged or uncharged particles) absorbed in living tissue can cause biological damage. It can interact directly with critical sites in the cells, where the atoms of the site become ionised thus leading to a chain of events that results in a biological change [Hal00]. Radiation may also interact with molecules of water in the cell to produce free radicals that are able to diffuse far enough to reach and damage the critical site.

Most cells that die are of little consequence, the body can replace them. On the other hand cells that change permanently may go on reproducing abnormal cells when they divide. The damaged cells end up not being able to perform their normal functions in the body and as a result this can lead to short or long-term biological effects (refer to Table 1.3). The main concern is that of an increased risk of cancer, which can develop if an irradiated cell is modified rather than killed. The health effects on an individual are related to the dose received by that person. The absorbed dose is defined in terms of the amount of energy deposited per unit mass. The SI unit for dose is

Gray (symbol Gy) where 1 Gy corresponds to 1 Joule being deposited per kg. It was found that the different types of radiation have different abilities to inflict biological damage. To account for this, each type of nuclear radiation is assigned a different quality factor. The absorbed dose is multiplied by the appropriate quality factor(s) to derive the dose equivalent which provides a normalized measure of biological effect. The unit of dose equivalent is the Sievert (Sv) where $\text{Sievert (Sv)} = \text{quality factor} \times \text{Gray}$. In what follows dose equivalent will be referred to as dose or absorbed dose unless otherwise stated. The relationship between levels of radioactivity and dose is complex. For example the dose received depends on whether exposure is external or internal to the body. An internal dose will be received if radionuclides are ingested or inhaled.

At higher doses, radiation above a certain threshold dose kills cells, which plays a crucial role in tissue injury and causes radiation sickness (see Table 1.3). The International Commission on Radiological Protection (ICRP) estimated that the average risk that a dose of 1 Sv to an individual would result in a radiation-induced fatal cancer is about one in ten [Van02], whereas at low doses the risk that fatal cancer would occur is about one in twenty. There is no evidence of a threshold dose, a lower limit at which there is no risk. Deterministic effects are impossible at low doses. Stochastic effects are still possible at low doses because genetic mutations may still occur [Van03].

Table 1.3: Deterministic and Stochastic effects of ionising radiation. Taken from [Van02].

Deterministic effects	Stochastic effects
Caused by death of cells	Caused by changes in cells that do not die
Leads to radiation disease, radiation wounds and blood disorders	There is a risk that cancer may develop after the latency period
No injury if dose is below threshold	No threshold dose for risk
Above threshold, the seriousness of injury is proportional to the magnitude of dose	Seriousness of cancer is independent of the dose, but the probability of cancer is proportional to the dose
Strong dependence on dose rate	Independent of the dose rate

The ICRP recommends a limit of an effective dose of 1 mSv for any combination of external and internal doses, respectively, in one year for a member of the public [Van02]. In special circumstances, an effective dose of up to 5 mSv in a single year, provided that the average dose over five consecutive years does not exceed 1 mSv per year, is allowed [IAE96]. The National Nuclear Regulator (NNR) also recommends these dose limits in South Africa. The effective dose is the sum of all equivalent doses received by different organs in the human body, taking into account the fact that some organs are more sensitive to ionising radiation than others.

1.3 Motivation for this study

Most South Africans consume fresh milk or milk-based products like cheese, yoghurts, condensed milk and powdered milk on a daily basis. If anthropogenic radionuclides should be present in the environment it is in principle possible for these to be transmitted to members of the

public via the soil-grass-cow-milk pathway as illustrated in Figure 1.8. As mentioned above ^{137}Cs and ^{131}I , with a half-life of 30 years and 8 days respectively, have been dispersed around the globe due to the fallout as a result of the testing of nuclear weapons; and routine and accidental releases of nuclear fission products from nuclear reactor plants. Many nuclear technology based analytical techniques involve the use of a radioactive source. In view of its relatively long half-life, ^{137}Cs is often used as a source. It is possible for such sources to be lost or stolen. If such sources should be incinerated it would also contribute to fallout. Prior to this study no published data in the public domain relating to radioactivity concentrations in South African milk could be found.

The Koeberg Nuclear Power Station (KNPS), operated by ESKOM (Pty) Ltd. is South Africa's only electricity producing nuclear reactor. It is located about 40 km from Cape Town along the West Coast. The power station comprises two 900 MW (electrical) pressurised water reactors. ESKOM has established an Environmental Survey Laboratory (ESL) on the KNPS site in order to routinely monitor the presence of anthropogenic nuclides in the environment around the power station. The KNPS has also formulated a Nuclear Emergency Plan that is activated in the event that there is an accidental release of radioactivity into the environment. Radiometric measurements of environmental samples constitute an important component of this plan. In the event that there is a significant release of radioactivity into the environment it is possible that the ESL will be evacuated due to the danger posed to the health of staff. In such an event the emergency plan makes provision for the setting up of an alternative ESL at a location where the radiometric measurements can be safely conducted. At present the alternative ESL is arranged to be the radiometric laboratory of the Department of Health in Bellville, near Cape Town.

The iThemba Laboratory for Accelerator Based Sciences (LABS), a cyclotron facility operated by the National Research Foundation in South Africa, is located at Faure, about 30 km from Cape Town. As a complement to its main focus it recently embarked on a research programme in environmental radioactivity. To this end an Environmental Radioactivity Laboratory (ERL) was established. Presently the ERL is focussed on performing radiometric analyses of environmental samples using high-resolution gamma-ray spectroscopy by means of a germanium detector. Although most samples analysed thus far have been sediments, it was decided to extend research to foodstuffs, in particular milk and mineral water, in order for iThemba LABS to provide an independent assessment of nuclide concentrations and associated public dose estimates. In this way iThemba LABS hopes to contribute to efforts to sustain and promote public health in the Western Cape, especially if there is a significant release of radioactivity into the environment.

In South Africa it is quite fashionable to drink bottled mineral water since it is a "natural" product and therefore considered to be good for one's health. Globally the consumption of mineral water has become popular in recent years. In some parts of the world this is due to necessity, e.g. the absence of safe tap water, rather than choice. Mineral water has its origins in springs and/or wells, which may contain natural radionuclides. The concentration of these nuclides in mineral water depends on, amongst others, the geology of the aquifer from where the water is sourced, the pH of the water at the source of the radionuclide transport, the time the water was pumped, the amount of water pumped, and any water treatment prior to bottling of the water [Kra03]. ^{226}Ra , an alpha particle emitter, is one radionuclide that contributes significantly to the ingestion dose as a result of drinking mineral water. The knowledge of radium concentrations in drinking water is also important since the metabolic behaviour of radium in the body is similar to that of calcium resulting in a fraction of the absorbed radium being deposited

in the bone, with the corresponding contribution to internal dose [UNS82]. While surveys of radioactivity in ground, surface and drinking water have been published [Kie89], similar information about South African mineral water prior to this study could not be found.

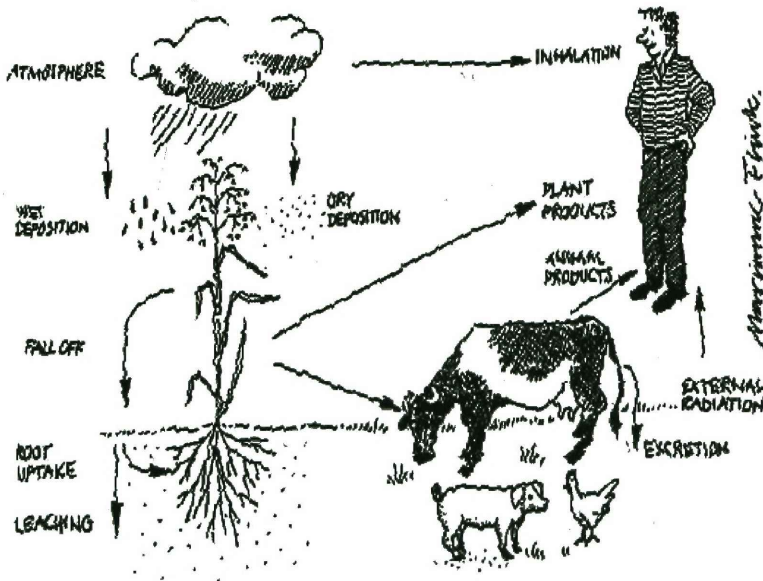


Figure 1.8: Main pathway of radionuclides to man (Drawing by M.Flink, 1996.) Taken from [WWW03].

1.4 Research objectives

In light of the above, the objectives of this study are:

- to use high-resolution gamma-ray spectroscopy to determine the radionuclide concentrations in liquid milk and bottled mineral water available in the Western Cape,

South Africa. In particular we aimed to measure the activity concentrations of the anthropogenic nuclides such as ^{137}Cs and ^{131}I present in three brands of milk readily available at convenience stores, and that of natural nuclides (^{226}Ra , ^{232}Th , ^{238}U and ^{40}K) present in two brands of bottled mineral water available at convenience stores.

- to use the determined activity concentrations along with the published dose conversion factors to estimate the ingestion dose to adult (age > 17 years) members of the public as a result of drinking milk and mineral water.

1.5 Thesis outline

In chapter 2 the techniques for measuring radionuclide concentrations in milk and mineral water, theoretical background of γ -ray spectrometry and similar studies done in measurements of milk and mineral water are discussed. Chapter 3 describes the experimental set-up and methodology used, while the results and interpretation of data are presented in Chapter 4. Finally the conclusion and an outlook are presented in Chapter 5.

Chapter 2

Overview of experimental techniques and literature review

In this chapter γ -ray spectrometry and other techniques used to determine radionuclide concentrations in milk and mineral water, the theoretical background of γ -ray spectrometry and similar radiometric studies performed, will be discussed.

2.1 Techniques for measuring radionuclide concentrations in milk and mineral water

Radio analysis of milk is usually accomplished by γ -spectrometry, α -spectrometry after radiochemical separation or Liquid Scintillation Counting (LSC) [Kat84]. The determination of gross activity is seldom done. Ion exchange methods can be used to remove radionuclides like iodine. Souring or curdling of the milk can render the sample useless. Therefore, the sample must be freezed or refrigerated.

Radium can be detected by α -particle (94 %) or γ -emission (3.5 %) [Mer97]. A number of analytical methods are available for the detection of radium, based on different physical and chemical principles [Köh02]. Another way is the measurement of its progenies where radioactive equilibrium is reached. The α -particle (^{222}Rn , ^{218}Po , ^{214}Po), β -particle (^{214}Pb , ^{214}Bi) and γ -emitters

(^{214}Pb , ^{214}Bi) allow indirect determination of ^{226}Ra . Some more details on these techniques are given below.

2.1.1 Alpha spectrometry after radiochemical separation

The determination is based on spectrometry of ^{226}Ra after chemical separation and purification of radium following quantitative electrodeposition [Alv95]. The analytical procedure described here is developed for the determination of environmental levels of ^{224}Ra and ^{226}Ra in aqueous samples. To an aliquot of 150 ml, concentrated hydrochloric acid (HCl) is added until a pH~1 was obtained. A column is packed with 1g of analytical grade resin and pre-treated with 1 M HCl.

The sample is then poured into a column, followed by 100 ml of 1M HCl. After purification of the radium by a cation exchange, the solution is evaporated to dryness and the residue is dissolved in a solution containing HCl. The sample is electrodeposited onto a stainless steel disc. After 60 minutes the disc containing the analyte is counted on an alpha counter with an efficiency of 34 % [Alv95].

2.1.2 Liquid scintillation counting (LSC)

Liquid Scintillation Counting is another method of determining the concentrations of Ra isotopes (^{226}Ra , ^{228}Ra and ^{224}Ra) in water and aqueous solutions [Cha93]. 10 cm³ of 1 M citric acid, 10 cm³ of 6 M ammonia and 20 cm³ of 0.1 M BaCl₂ is added to the water sample. The sample is heated to boiling, methyl-orange indicator is added and dropped in H₂SO₄ solution until the colour changes. After several hours the supernatant was decanted and the precipitate transferred

into centrifuge vials. The precipitate is centrifuged and rinsed with water until a pH of 7 is reached. Ethylene Diamine Tetracetic Acid (EDTA) and nitric acid are added, and then the samples are heated in a bath. The samples are stirred until the precipitate dissolves. After cooling, Ra and Ba sulphates are precipitated again using glacial acid. The samples are centrifuged and rinsed in water; and the precipitate transferred into scintillation vials. Measurements are performed using a low-background liquid scintillation spectrometer with heavy lead shielding. The low-background conditions allows for high detection sensitivity. The following detection limits for a counting time of 1 hour were achieved 3 Bqm⁻³ (²²⁶Ra), 20 Bqm⁻³ (²²⁸Ra) and 10 Bqm⁻³ (²²⁴Ra) [Cha93].

2.1.3 Gamma-ray spectrometry

Gamma-ray spectrometry is the method which is employed in this study using a high purity germanium (HPGe) detector. Some theoretical background on γ -ray spectroscopy is presented in the next section.

2.2 Theoretical Background on γ -spectrometry

Gamma radiation is electromagnetic radiation produced by transitions between excited states of the atomic nucleus. In the enormous range of energies in the electromagnetic spectrum γ -rays sit at high energies. Unlike α and β decay there is no change in the number or type of nucleons in the nucleus during γ -decay and γ -radiation is usually a by-product of α or β decay.

2.2.1 Interactions of gamma-rays with matter

There are three dominant types of interactions of γ -photons with matter, which must be considered in the detection of photons or measurement of radionuclides emitting photons namely photoelectric absorption, Compton scattering and pair production. In all three cases free electrons are generated, and as these electrons are slowed down on their path through the matter they create electron-ion or electron-hole pairs [Deb88]. Each mechanism transfers energy from a γ -ray to the electron, and in the case of pair production, to the positrons [Gil95].

Photoelectric absorption

Photoelectric absorption arises by interaction of γ -ray photons with one of the bound electrons in the atom [Gil95]. The electron (photoelectron) is ejected from one of its bound shells (Figure 2.1). For γ -rays of sufficient energy, the most probable origin of the photoelectron is the most tightly bound K shell of the atom [Kno79]. The photoelectron appears with energy, E_e , given by:

$$E_e = E_\gamma - E_b \quad (2.1)$$

where E_γ is the γ -ray energy and E_b is the binding energy of the photoelectron in its original shell. This process is dominant at low energies. Photoelectric absorption increases with the atomic number of absorber materials. The probability that a photon will undergo photoelectric absorption can be expressed as a cross section (τ) over all ranges of photon energies (E_γ) and atomic number (Z) by:

$$\tau \propto \frac{Z^n}{E_\gamma^m} \quad (2.2)$$

where n and m are within the range of 3 to 5 depending upon energy [Gil95].

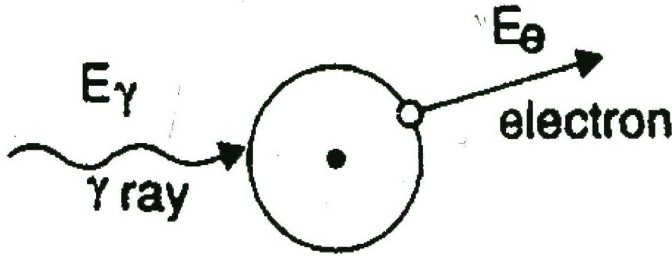


Figure 2.1: The mechanism of photoelectric absorption [Gil95].

Compton scattering

In Compton scattering (Figure 2.2) the interaction is between the incident γ -ray photon and an electron of the absorbing material. The incoming γ -ray photon is deflected through an angle with respect to its original direction [Kno79]. Figure 2.2 shows the direct interaction of the γ -ray with an electron transferring a portion of its γ -ray energy. The photon transfers a portion of its energy to the electron, which is also known as a recoil electron. The energies of outgoing photon and electron can be related to the angles at which they are emitted and the energies can vary from zero to a large fraction of the incident energy. The energy imparted to the recoil electron is given by the following equation:

$$E_e = E_\gamma \left\{ 1 - \frac{1}{\left(1 + \frac{E_\gamma [1 - \cos \theta]}{m_0 c^2} \right)} \right\} \quad (2.3)$$

where E_e is the energy of the recoil electron, E_γ is the incident γ -ray energy, θ is the scattering angle and $m_0 c^2$ is the rest mass of the energy of the electron [Gil95].

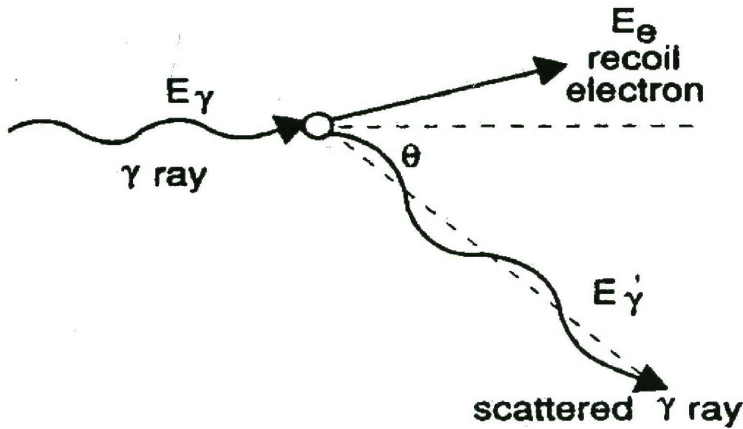


Figure 2.2: The mechanism of Compton scattering (Gil95).

Pair production

This interaction takes place when the γ -ray energies are above a threshold of 1.02 MeV and unlike the other two interactions pair production results from the interaction of the γ -ray with the atom as a whole. In this process the energy is converted in the nuclear Coulomb field to a positron-electron pair [Deb88]. The electron and positron created share the excess energy (above 1.02 MeV) equally and loses it to the detector medium as they are slowed down. Then two 511

keV annihilation photons are produced as secondary products of the interaction. The cross section for the interaction, σ , depends upon E_γ and Z which can be expressed as [Kno79]:

$$\sigma \propto Z^2 f(E_\gamma, Z) \quad (2.4)$$

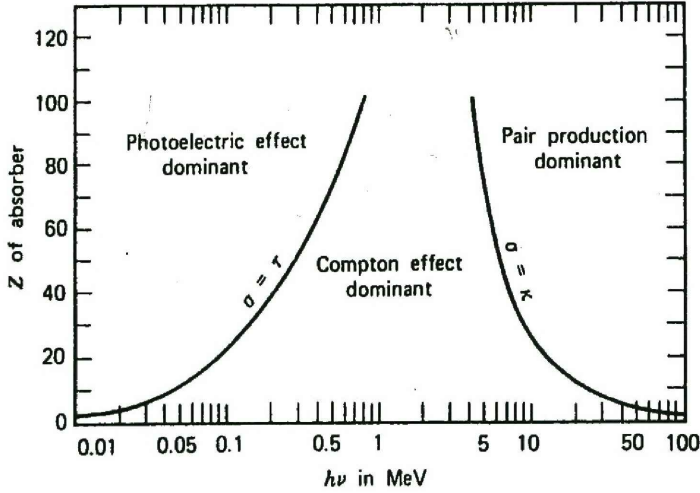


Figure 2.3: An illustration of three dominant areas of photoelectric effect, Compton effect and pair production separated by lines showing the values of Z and photon energy E [Tyk95].

The relative importance of these three types of photon interactions is illustrated in Figure 2.3, which shows the regions of atomic number Z and the photon energy E where each interaction predominates. Compton interactions dominate at medium energies, photoelectric at low energies while pair production dominates only at high energies.

2.2.2 Detection of gamma-rays

It is important that a detector should have a high absorption coefficient for γ -radiation so that there is a high probability of complete absorption. The detector must be capable of absorbing a large fraction of the γ -ray energy. It is better to use a detector of suitable size, or to choose a detector material of suitable high Z . The absorption coefficients for all the significant interaction processes increase with atomic number. Having absorbed the γ -rays and created many charged species the detector material must allow the charge to be collected in some manner and be presented as an electric signal [Gil95].

Semiconductors

A semiconductor is a material that can act as an insulator or a conductor. Silicon (Si) and germanium (Ge) are the most widely used semiconductors. Semiconductors are referred to as solid-state detectors, but also scintillation counters can be thought of as solid-state detectors. Therefore it has come into widespread use to characterize only those devices that are based on electron-hole pair collection as semiconductor media [Kno79]. The most important advantage of semiconductors, compared to other types of radiation counters, is their superior energy resolution [Tso83].

Other advantages are [Tso83]:

- Linear response (pulse height versus particle energy) over a wide energy range,
- Possibility for special geometric configurations,
- Fast pulse rise time (relative to gas counters) and
- Ability to work in vacuum.

For γ -ray detection, germanium is preferred over silicon because of its higher atomic ($Z_{\text{Si}}=14$, $Z_{\text{Ge}}=32$) and its smaller binding energy (E_b). The Ge and Si detectors must be cooled in order to reduce noise to an acceptable level. The treatment of the crystal plays an important role in the operation and performance of the detector. The most common medium for detector cooling is liquid nitrogen (LN_2). In liquid nitrogen cooled detectors, the detector and in some cases the preamplifier components, are housed in a clean vacuum chamber, which may be attached to or inserted in the LN_2 Dewar. In semiconductors, the valence band is always full and the conduction band is always empty and the energy gap between the two bands is very small. The size of the band gap determines whether the material is classified as an insulator or a semiconductor. For insulators the band gap is 5 eV and for semiconductors it is 1 eV [Gli95]. The electron in the material must move from its current energy state to another in order to move from one atom to another. If the electrons can jump into suitable energy levels then an external electric field applied to the material would cause a current to flow. Cooling the material will reduce the number of electrons in the conduction band, thereby reducing leakage current and making it easier to detect extra excitation due to the γ -ray interactions [Gil95].

2.2.3 Attenuation of gamma radiation

When travelling through the sample material some γ -rays are absorbed in the material and others are scattered into the detector. Each of the interaction processes removes a γ -ray photon from the beam thus reducing its intensity. The degree of γ -ray attenuation can be calculated using:

$$I = I_0 e^{-\mu x} \quad (2.5)$$

where I_0 represent the incident intensity, x the thickness of the absorber in units of length [Lil01], and μ is the linear attenuation coefficient, being the sum of probabilities per unit path length that the γ -ray photon is removed from the beam [Kno79], i.e.

$$\mu = \mu_{photoelectric} + \mu_{compton} + \mu_{pair} \quad (2.6)$$

The mass attenuation coefficient (μ_m) is more widely used than the linear attenuation coefficient because the linear attenuation coefficient is limited by the density of the attenuating material [Kno79], even though the attenuating material is the same. The relationship between linear and mass attenuation coefficient is given by:

$$\mu_m = \frac{\mu}{\rho} \quad (2.7)$$

where μ is the linear attenuation coefficient and ρ is the density of the material. The mass attenuation coefficient has the advantage that for a given γ -ray energy the mass attenuation coefficient does not change with the physical state of the given absorber [Kno79].

2.3 Literature review: radiometric studies of beverages

Nigeria is a country of over a 100 million people. Here most of the cows reared are not used for dairy purposes but for meat consumed by the Nigerian population. The milk consumed by the majority of the people is imported from European countries [Osi99]. A study was conducted to identify the type of radionuclides present and to determine the γ -activity levels in the widely consumed milk imported from countries in Europe (United Kingdom, Ireland, Denmark, Switzerland, France and Netherlands) which were once contaminated with radioactive fallout mainly atmospheric from the damaged Chernobyl nuclear power reactor. The samples were

collected from departmental stores located in many metropolitan cities of Nigeria, Lagos, Ibadan, Benin City, Port Harcourt and the others. A total of 9 different makes of instant powder produced in the six European countries were used for this study. It must be noted that the composition of milk is almost identical around the world and it is easy to collect a representative sample that can be analysed in liquid or powdered form. Ten 1-litre Marinelli beakers were filled with known masses of milk powder samples, sealed for days to allow a sufficient time to attain a state of secular equilibrium. For measurements, a Canberra vertical high purity coaxial germanium (HPGe) crystal (model GC2018-7500) enclosed in 100 mm thick lead shield was used. A counting time of 36000 s was used and the same geometry was employed for counting of both milk samples and background. ^{137}Cs was not detected in any of the milk samples in view of the possible transfer through the soil-grass-cow-milk route. It was then assumed that it may be due to the fact that radio caesium has a short biological half-life² of the order of 2-4 weeks. Another possible reason for the absence of anthropogenic radionuclides is the fact that countermeasures were applied during the first few weeks after deposition of long-lived radionuclides following the Chernobyl incident. They only observed naturally occurring radionuclides with activities 23.0 ± 7.8 (^{226}Ra), 4.4 ± 2.1 (^{228}Ra), 831.7 ± 54.8 Bq/kg ^{40}K [Osi99].

As a result of a series of atmospheric nuclear weapon tests conducted by the USA, USSR, UK, France and China, a programme of assessment of fallout radioactive contamination in milk and diet samples was started in India [Shu94]. ^{90}Sr and ^{137}Cs were measured for fallout radioactivity estimation while ^{40}K , ^{210}Po , ^{226}Ra and ^{228}Th were measured for the natural radioactivity assessment [Shu94]. 10 litres of liquid milk samples were collected from the city's major dairy

² the time required for the body to eliminate 50 percent of the dose of any substance by regular process of elimination

centre once every month. For fresh liquid milk the production and the consumption period was about 10 days. Skimmed powder that was produced from the fresh liquid milk is utilised within three months for preparing reconstituted liquid milk. Immediately after the samples are collected, they are ashed on an open flame below 350°C to avoid loss of ^{137}Cs . The ashing process took about 15 days and then the samples are ground to fine powder and transferred to an airtight plastic container and stored for a month to allow ^{222}Rn and ^{220}Rn daughter products to attain radioactive equilibrium with ^{226}Ra and ^{228}Th . The samples were counted for 1000 minutes on a 12.5 cm $\times$ 10 cm NaI (TI) coupled γ -ray spectrometer. After estimation of γ -activity, ^{90}Sr activity was determined. The intake values were calculated based on the assumption that the milk consumption of an adult is 100 ml/d. The average concentration levels of ^{90}Sr and ^{137}Cs in milk ranged from 0.005 to 0.442 Bq/l. The annual effective dose per capita to the population of Bombay ranged between 79 and 177 mSv/y with an average of 110 mSv/y. For the period 1985 - 1990 they found an average ^{40}K activity concentration of 52.0 Bq/l (refer to Table 2.1).

Table 2.1: Annual mean concentrations of fallout and natural radionuclides in milk samples (Bq/l) during the year of collection [Shu94]. NM denotes not measured.

Year	^{90}Sr	^{137}Cs	^{40}K
1985	NM	< 0.007	53
1986	< 0.005	0.420	66
1987	< 0.005	0.120	56
1988	0.16	0.120	33
1989	0.01	0.110	55
1990	0.01	0.130	49

During the year 1997, food including milk, fruit and wine, were collected at several locations surrounding the Hanford site, a nuclear processing site in the south east of Washington State in the USA [WWW04]. The site was used to produce plutonium for nuclear weapons until the 1980s, when it was discovered that it was releasing radioactive and chemical waste into the Columbia River. Samples were collected mostly from locations in the south and east of the site where deposition of airborne wastes could be expected. Others were collected at locations somewhat distant from the site to provide information on background radioactivity. Milk was analysed for ^{90}Sr , ^{129}I and γ -emitters such as ^{137}Cs . These radionuclides have the potential to move through the air-pasture-cow-milk or water-pasture-cow-milk food chains to humans. ^{90}Sr from Hanford was released to the Columbia River through riverbank springs, while ^{137}Cs is present in the atmospheric fallout from weapon testing and it is found in the site radiological waste. They found that the concentration of ^{90}Sr remains near the detection limit (0.7 pCi/l) and have been relatively constant over the past 6 years.

Locally produced red and white wine were analysed for tritium and important γ -emitting radionuclides. The wines were made from grapes grown from individual vineyards near site. Two samples of red and white wine were obtained and analysed from each location. Gamma spectroscopy did not indicate the presence of ^{137}Cs or any γ -emitters in any of the wine samples. They found that the concentration of tritium in 1997 wine samples ranged from 19.4 to 34.4 pCi/l³. There was generally no difference between the variety and locations sampled.

The levels of naturally occurring radioactive elements in bottled mineral water, commercially available in Australia have been assessed [Coo81]. The survey concentrated upon radionuclides like radium (Ra) and lead (Pb), which have a high toxicity. Detectable levels of Ra were found to

³ 1Ci = 3.7×10^{10} Bq

range from 0.02 to 0.32 Bq/l in Australian bottled water and from 0.02 to 0.44 Bq/l in imported brands. The Pb levels were found to be low at 0.08 Bq/l. The radiological health implications of consumption of bottled mineral water were discussed in terms of radiation exposure and increased risk to health. It was concluded that although some brands of water contain radioactivity in excess of drinking water limits recommended by the Australian authorities, the annual radiation dose to an individual would be below the dose limits recommended by the ICRP [Coo81]. Some people who drink water containing ^{226}Ra for many years may have an increased risk of getting cancer.

Mineral water taken from springs in north-eastern Romania is used as table mineral water sources. A study aimed at assessing the radioactive content of these waters and their contribution to the population exposure was done during 1997-2000 [Bot02]. Bottled water and some spring water samples were collected at different locations. In all samples total alpha and beta radioactivity and the concentrations of natural radionuclides like ^{238}U , ^{232}Th , ^{226}Ra and ^{40}K were determined. Dose rates were calculated using ICRP dose conversion coefficients. Neither of the analysed samples exceeds the specific activity allowed in Romania for fresh water in natural radionuclides under investigation except for ^{226}Ra . The results of the study are shown in Table 2.2. The dose calculations, based on the daily intake of 0.5 litre of mineral water, were performed and the resulting annual dose from the intake of natural radionuclides was found to range between 1.5 and 30 $\mu\text{Sv/y}$.

Table 2.2: Activity concentrations (mBq/l) of natural radionuclides in Romanian natural mineral water [Bot02].

District	^{226}Ra	U_{nat}	Th_{nat}	^{40}K	
Suceava	19 ± 16	24.8 ± 8.5	0.61 ± 0.52	220 ± 175	Spring and bottled water
	226 ± 149	16.1 ± 3.7	0.66 ± 0.27	90 ± 51	Bucovina table water
	24 ± 26	27.1 ± 17	0.86 ± 0.74	120 ± 87	Communal springs
Iasi	11.5 ± 6.6	25.6 ± 8.4	1.25 ± 0.48	178 ± 56	Spring and bottled water
	8.7 ± 4.4	20.3 ± 10.4	0.49 ± 0.12	272 ± 81	Communal springs
Neamt	12.1 ± 3.2	19.7 ± 9.1	2.07 ± 0.40	210 ± 70	Spring and bottled water
	15.3 ± 10.2	23.4 ± 7.2	0.67 ± 0.25	78 ± 42	Communal springs

A study aimed at giving a comprehensive overview of the contribution of mineral water to the ingestion pathway was made based on the production data published by the Brazilian Mineral Water Producers Association [God00]. Bottled mineral water samples were analysed for the presence of ^{210}Pb , ^{228}Ra and ^{226}Ra . According to the procedure lead and barium carriers were added to the 1-litre samples. The samples were precipitated. The precipitate was purified by EDTA, filtered and weighed for chemical yield. The samples were allowed to stand for 21 days then measured for its α/γ counting rate using a 10-planchet low-level proportional counter for ^{228}Ra and ^{226}Ra . ^{210}Pb was re-precipitated, the precipitate was filtered, weighed and allowed to stand for one month to be measured for its β -activity. The results obtained for the nuclides are presented in Table 2.3. In regions in Brazil where the uranium content is high in the soil, high dose levels were obtained mainly due to the presence of ^{228}Ra . Examples of these are the northern and south-eastern regions (see Table 2.4). The highest individual doses of 1.15 and 0.87

($\mu\text{Sv/y}$) from northern and south-eastern regions were obtained for a child consuming only a specific type of water from each region.

Table 2.3: Average activities (Bq/l) of ^{226}Ra , ^{228}Ra and ^{210}Pb in Brazilian waters. SE, NE, S and N denote south-east, north-east, south and north respectively. Taken from [God00].

Region	^{226}Ra	^{228}Ra	^{210}Pb
SE	0.029	0.102	0.082
NE	0.020	0.058	0.029
S	0.008	0.013	0.049
N	0.083	0.385	0.102

Table 2.4: Individual effective dose estimates due to ingestion of mineral water in Brazil. SE, NE, S and N denote south-east, north-east, south and north respectively. Taken from [God00].

Region	Average dose ($\mu\text{Sv/y}$)	Min-Max ($\mu\text{Sv/y}$)
SE	49	$6.3\text{--}2.2 \times 10^2$
NE	24	14-31
S	17	8-25
N	129	$14\text{--}2.8 \times 10^2$

Sanchez *et al.* [San99] carried out a study of the radioactive content in commercial bottled mineral water available for human intake in Spain. 43 brands of mineral water with different locations were purchased at different self-service outlets. Firstly, the samples were measured in their own containers to detect the presence of radon isotopes. However, none were detected.

Samples were then dried at 80 °C. The dry residue of each sample was determined by weighing the planchet and compared with the value given on the bottle label. The samples were counted for 1000 minutes with a high purity germanium detector. The activity concentrations of ^{226}Ra ranged between 0.06 and 1.86 Bq/l.

Recently in Austria Kralik *et al.* [Kra03] analysed bottled mineral water available commercially in Austria for natural radioactivity. The main purpose of the study was to assess the current ingestion doses in bottled water due to natural radionuclides. Samples from different origins were analysed with a liquid scintillation counting. Radium and radon were analysed by adding 10 ml of the sample to 10 ml of mineral oil based LSC-Cocktail in a coated counting vial. The samples were stored for five weeks to attain secular equilibrium. The samples were measured again for ^{222}Rn ingrowth with α/β discrimination. ^{210}Pb was determined by chemical separation, stored for five days to allow for ^{214}Bi to grow and counted using α/β discrimination. Concentrations of ^{222}Rn ranged from < 0.12-18 Bq/l, ^{210}Pb activity concentrations ranged from <2-34 mBq/l and ^{226}Ra ranged from < 0.02-0.023 Bq/l. Individual effective doses were calculated for geometric mean and maximum activity concentrations in the mineral waters using dose conversion factors from the EC Basic Safety Standards for different age groups. The annual consumption for infants < 1 year with or without nursing was 55 l and 170 l respectively. Ingestion doses from average activity concentrations in mineral water for different age groups ranged from 0.001-0.034 mSv/y for ^{226}Ra , 0.0003-0.007 mSv/y for ^{210}Pb and 0.0001-0.004 mSv/y for ^{222}Rn .

Chapter 3

Experimental methodology

As mentioned in Chapter 1, the aim of this study is to perform a radiometric analysis of liquid milk and mineral water using γ -ray spectrometry with a view of determining the activity concentrations of anthropogenic and natural radionuclides in these beverages. In this chapter the following are discussed: sample selection and preparation, the high purity germanium detector and associated electronics, the data acquisition system and the sample counting procedure.

3.1 Sample selection and preparation

3.1.1 Milk

Sample selection

The idea was to perform a radiometric analysis of milk, which is directly available to any member of the public. The milk samples used in this study were purchased over the counter at convenience stores in Brackenfell suburb, which is about 20 km from iThemba LABS. Three milk brands denoted by A, B and C were sampled with their respective dairies being based in Cape Town (Durbanville, Athlone) and Worcester, respectively, as indicated in Figure 3.1. All the milk was purchased fresh and in 1 litre plastic satchets.

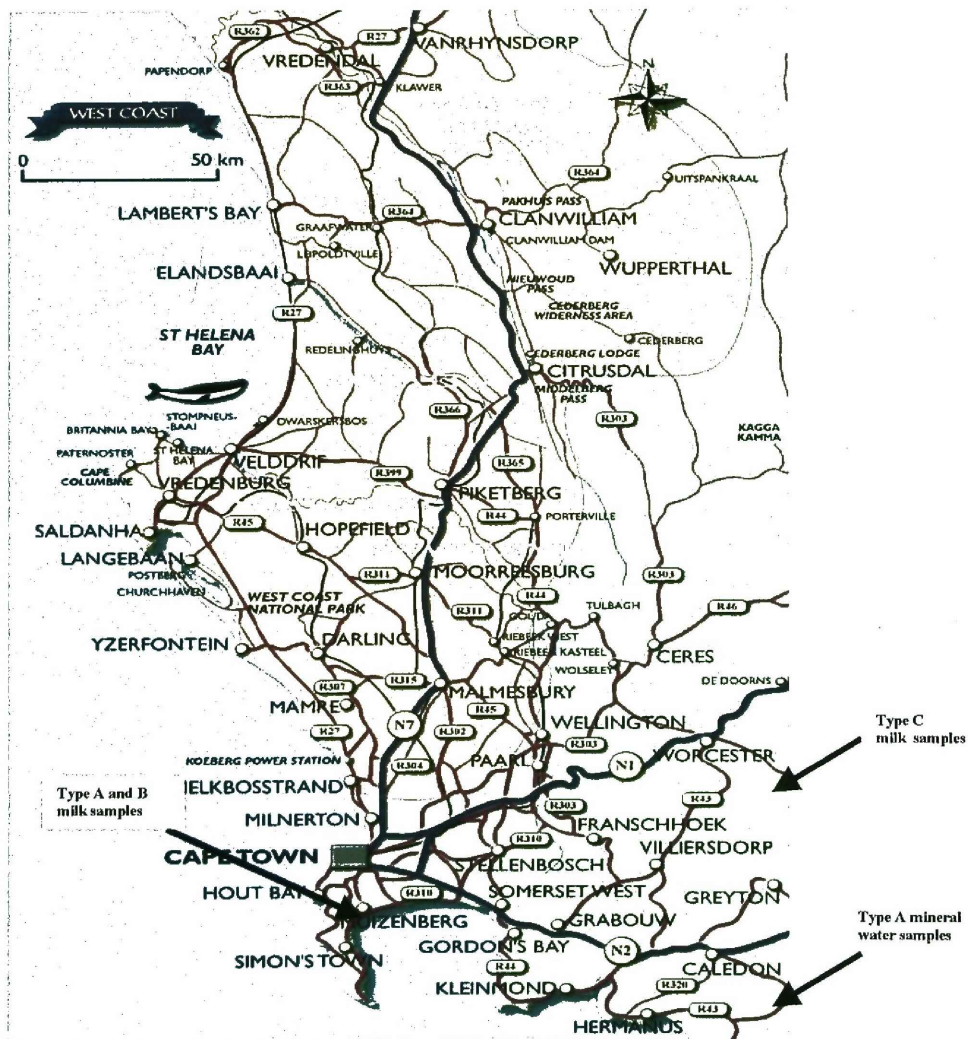


Figure 3.1: A portion of the Western Cape map with arrows indicating where the milk and mineral water samples were packaged [WWW05].

Sampling procedure

The samples were purchased and counted over a three-week period. During the first and second week one sample from each brand was purchased and counted, while two samples from each brand were purchased and counted during the third week. A summary of the samples which were counted is presented in Table 3.1.

Table 3.1: A summary of the sampling procedure used for the milk

Sample no	Brand	Week when measured	Density (g/l) at 15° C	Counting period (hrs)
M1	A	1	1049.9	9
M4	A	2	1046.1	9
M7	A	3	1045.9	9
M10	A	3	1052.5	9
M2	B	1	1055.5	9
M5	B	2	1052.6	9
M8	B	3	1053.0	9
M11	B	3	1055.5	9
M3	C	1	1053.6	9
M12	C	2	1050.6	9
M6	C	3	1061.1	9
M9	C	3	1049.3	9

Sample preparation

The milk was allowed to warm up to room temperature of 15° C inside the Environmental Radioactivity Laboratory (ERL) at iThemba LABS for about 2-3 hours. The Marinelli beakers shown in Figure 3.2 [Ame00] were washed and rinsed with distilled water. Marinelli beakers are re-entrant beakers with good geometry for large volumes of low-activity material. These beakers are made of Polypropylene, optimised to 1 litre, with tight-fitting lids. A Marinelli beaker placed on top of the detector is shown in Figure 3.3. The diameter of the beakers is 85 mm and are manufactured by Amersham [Ame00]. 1 litre of milk was decanted into the Marinelli beaker. The mass of the beaker with the sample was determined on a scale (Sartorius BP2100s model) in order to determine the mass of the milk.

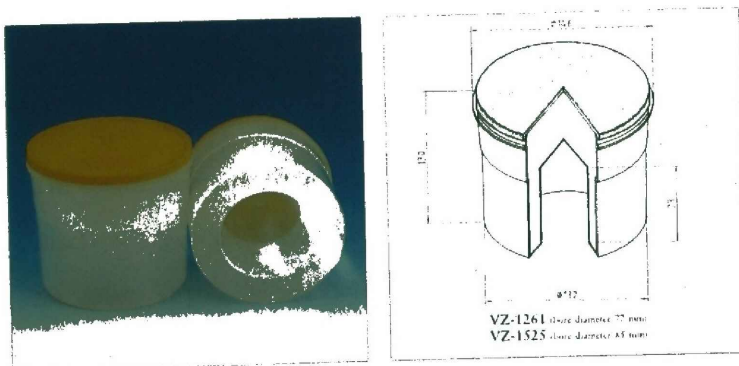


Figure 3.2: A picture of a Marinelli beaker taken from [Ame00] showing its dimensions.

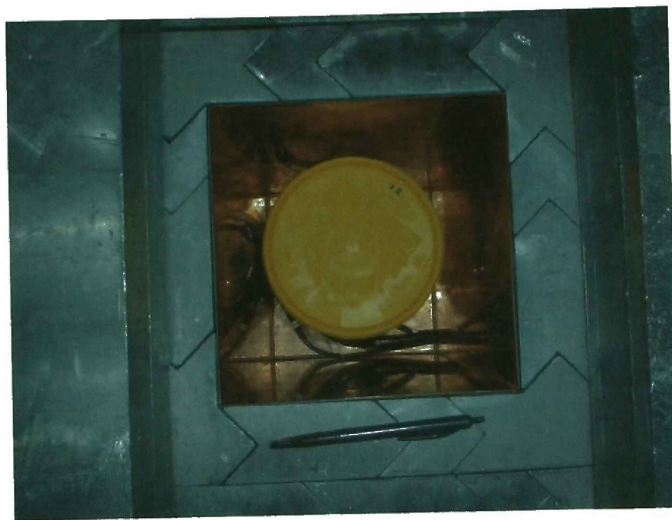


Figure 3.3: A top-view photograph of the Marinelli beaker placed on top of the germanium detector, surrounded by a 10 cm thick lead castle and 2 mm thick copper lining.

3.1.2 Mineral water

Sample selection

Here again the idea was to analyse the samples that are readily available to the public. The bottled mineral water was purchased over the counter at convenience stores also in Brackenfell. Two brands labelled A and B were studied with water having the origin in the vicinity of Caledon and Doornkloof, respectively, as shown in Figures 3.1 and 3.4 respectively.

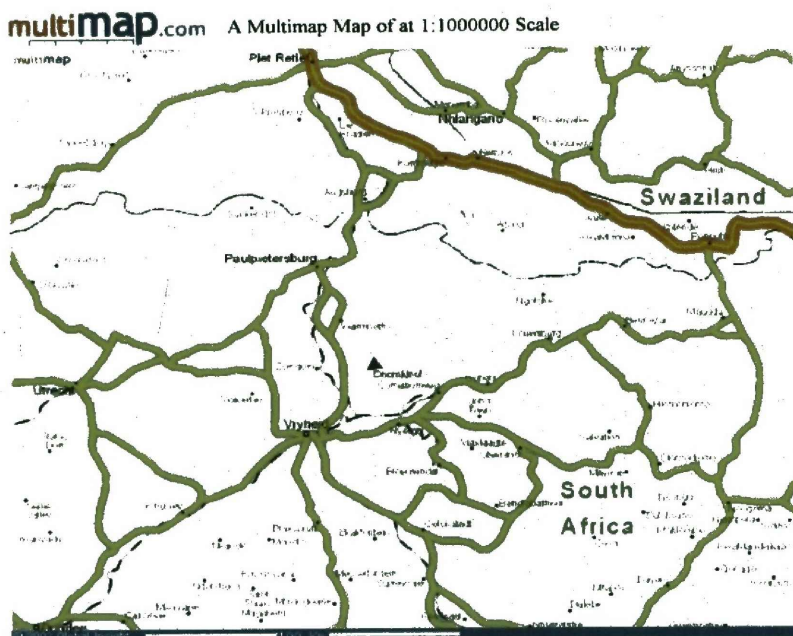


Figure 3.4: A map showing Doornkloof (with a triangle marking) where Type B mineral has its origins [WWW06].

Sampling procedure

The samples were purchased and counted for a period of two weeks, three samples from each brand per week. After a period of three weeks the samples were measured over a two week period again. A detailed summary is presented in Table 3.2 (a) and (b).

Table 3.2: (a) A summary of the sampling procedure of the mineral water for the first set of measurements.

Sample number	Brand	Week when measured	Density (g/l) at 15° C	Counting period (hrs)
MW1	A	1	1012.2	9
MW2	A	1	1010.1	9
MW3	A	1	1015.7	9
MW4	B	2	1010.5	9
MW5	B	2	1008.1	9
MW6	B	2	1007.6	9

Table 3.2: (b) A summary of the sampling procedure of the mineral water for the second set of measurements.

Sample number	Brand	Week when measured	Density (g/l) at 15° C	Counting period (hrs)
MW1a	A	6	1012.1	9
MW2b	A	6	1010.6	9
MW3c	A	6	1015.6	9
MW4a	B	7	1000.5	9
MW5b	B	7	1007.8	9
MW6c	B	7	1007.1	9

Sample preparation

The samples were measured directly without warming them to room temperature of the laboratory. 1-litre Marinelli beakers were washed and rinsed with distilled water. Then 1 litre of mineral water was decanted into the Marinelli beaker. The mass of the beaker and sample was determined on the scale (Sartorius BP2100s model). After the initial counting, samples were sealed and allowed to stand for more than three weeks in order to allow for secular equilibrium in the decay of ^{238}U to be reached. Different Marinelli beakers were used, for the first measurements Marinelli beakers with tight-fitting lids and for the second measurements Marinelli with screw-top lids, which were assumed to provide better sealing [Dev02], were used.

3.2 γ -ray spectrometry

3.2.1 The HPGe detector

For this study a high-purity germanium (HPGe) detector with a vertical orientation (Figure 3.5) was used. It is a Canberra p-type detector with a built-in preamplifier (see Figures 3.6 and 3.7). The detector crystal has a diameter of 62.5 mm and a length of 59.9 mm. The detector has relative photo-peak efficiency of 45 % at 1.33 MeV. The energy resolution at 1.33 MeV was found to be ~ 2.6 keV. To shield the detector from background radiation, a 10 cm thick lead castle with a 2 mm copper lining to absorb some of the X-rays produced by lead surrounds the detector. The detector has a liquid nitrogen (LN_2) dewar as shown in Figures 3.5 and 3.7, which needs to be filled every two weeks in order to cool the crystal to LN_2 temperature. In practice the dewar is filled once a week.



Figure 3.5: A photograph of high-purity germanium detector with accompanying lead castle at iThemba LABS Environmental Radioactivity Laboratory (ERL).

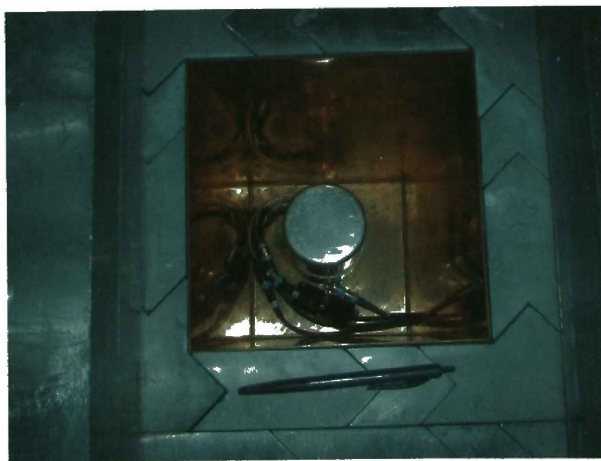


Figure 3.6: A photograph of the top view of the germanium detector with a built-in preamplifier. The lead castle and the copper lining are clearly visible.

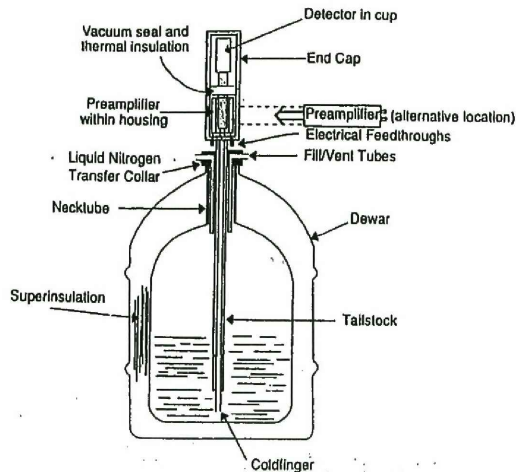


Figure 3.7: A cross section diagram showing various parts of the germanium detector with liquid nitrogen reservoir. Taken from [Gil95].

3.2.2 The electronic set-up

The electronic set-up (see Figure 3.8) used to perform measurements in the ERL consists of a preamplifier, bias supply, amplifier, and the multi-channel analyser. The function of the electronic set-up is to collect the charge produced by γ -ray interactions with the detector, measure the amount and store the information [Gil95] in the form of a voltage level also called a pulse height. A brief description of the electronic modules is given below:

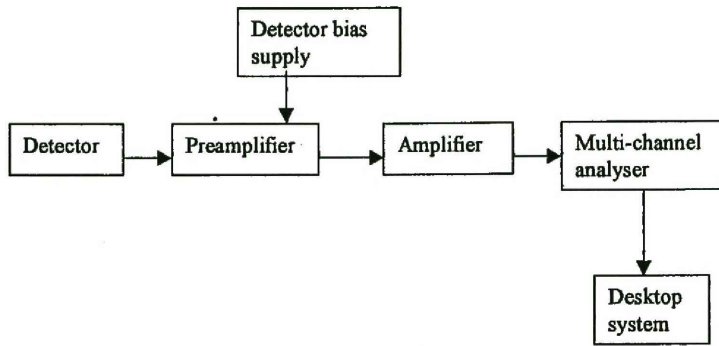


Figure 3.8: Schematic diagram of the electronic set-up used in conjunction with the HPGe detector.

- Preamplifier

The charge created within the detector by interaction with the γ -radiation is collected by the preamplifier [Gil95]. The charge is then converted to a voltage pulse. The other purpose of the preamplifier is to minimize any sources of noise, which will be transmitted along with the pulse and thus reduces the energy resolution of the system [McG83]. In modern systems like the one used in this study a charge-sensitive preamplifier comes as an integral part of the detector assembly. This is advantageous, since that the preamplifier will be cooled whenever the detector system is cooled thus reducing thermal noise contributions. Preamplifiers have various modes of operation such as current-sensitive and voltage-sensitive.

- Detector bias supply

The detector bias supply (SILENA model 7716) supplies the detector with the external voltage needed for a proper operation. The bias supply used for this study is 3.50 kV. The bias supply is delivered via the preamplifier and it provides the electrical field to sweep the electron-hole pairs

out of the detector, which are then collected by the preamplifier. This is the least critical unit in the electronic system for a semiconductor γ -ray detector

- **Amplifier**

The output signal from the amplifier (ORTEC 572 model) is in a range of few millivolts. As a result it cannot travel very far without losing information or itself being lost in the noise. The amplifier plays two roles of amplifying and shaping the signal [Tso83]. The signal needs to be shaped to reduce the signal-to-noise ratio. Choosing an appropriate shaping time can minimize the noise contribution. The longer the shaping time the better the resolution for the peaks. The shaping time used for the pulse was 6 μ sec. The gain drift of the amplifier is caused by small changes in the characteristics of the resistors, capacitors, etc., as a result of temperature changes. To obtain quality results the gain stability of the amplifier should be monitored.

- **Multi-channel analyser (MCA)**

The multi-channel analyser (OXFord-Win) sorts the pulses by pulse height (the pulse height is proportional to the γ energy deposited in the detector) and counts the number of pulses within individual pulse height intervals. The pulse heights are in analog form from the amplifier and they are converted to digital quantities by the ADC (Analog-to-Digital Converter), which is one of the components of the MCA and stored as counts in channels [Mal00].

3.2.3 Energy and FWHM calibration

The main objective of the energy calibration is to derive the relationship between pulse height and channel number in the MCA spectrum and the corresponding γ -ray energy. The calibration is obtained by determining the centroids of the γ -lines with known energies, and by fitting a polynomial function to the curve of centroid versus γ -ray energy. In this way the parameters

specifying the polynomial are determined. The polynomial function normally has linear or quadratic form. The energy calibration of the detector system was routinely performed before the milk and mineral water samples were counted. A liquid standard source prepared by the National Metrology Laboratory of the Council of Scientific and Industrial Research (CSIR) was used for this purpose. The standard solution was in a Marinelli beaker similar to that used for milk and mineral water samples (section 3.1.1) spiked with ^{152}Eu , ^{137}Cs and ^{60}Co (see Figure 3.9). Further details of the standard source are summarised in Table 3.3. A spectrum of the standard source acquired over a period of two hours is shown in Figure 3.10.

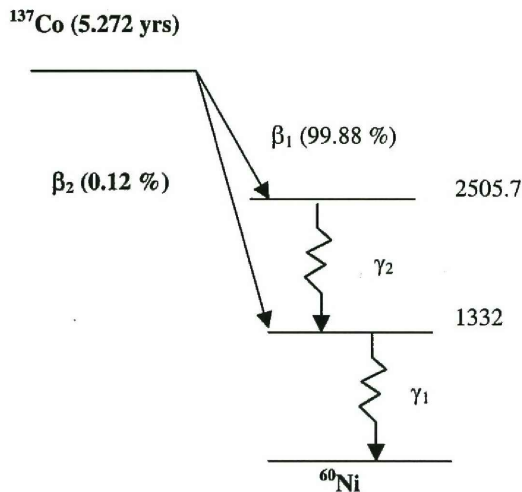


Figure 3.9: The decay scheme of ^{60}Co .

Table 3.3: Details of the liquid standard source used for this study. The activities quoted were present at the time the source was prepared.

Radionuclide	Activity (kBq)	Uncertainty (1σ)	Half-life (yrs)
^{152}Eu	6.550	$\pm 3.0\%$	13.506
^{137}Cs	0.661	$\pm 2.0\%$	30.018
^{60}Co	1.907	$\pm 2.0\%$	5.271

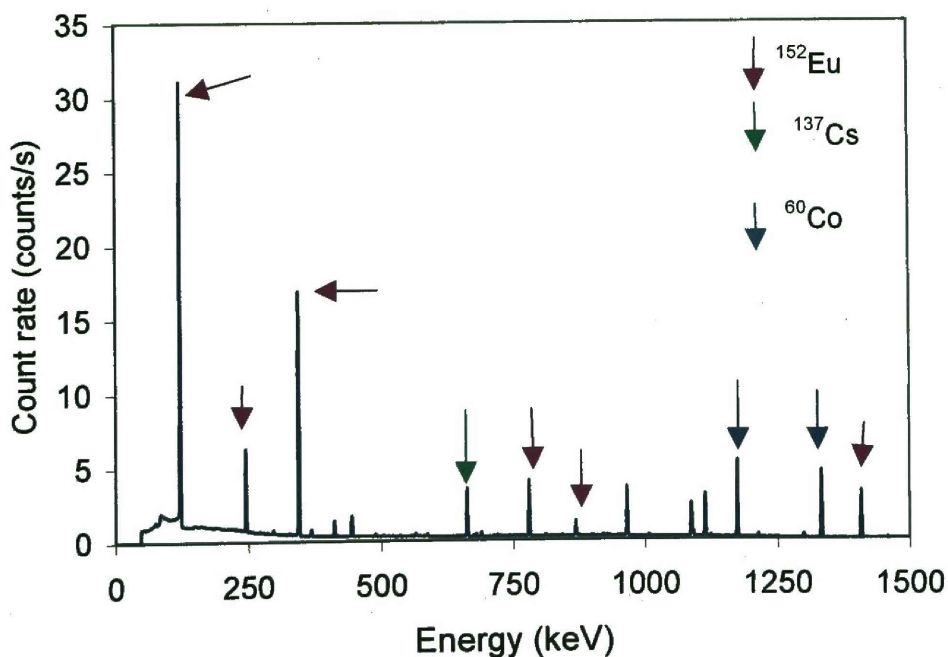


Figure 3.10: A spectrum of the liquid standard source with peaks of interest shown.

The energy calibration was performed using at least 9 lines present in the spectrum. A plot of the centroid of these lines versus γ -ray energy is shown in Figure 3.11. The linear function fitted to the data is also shown.

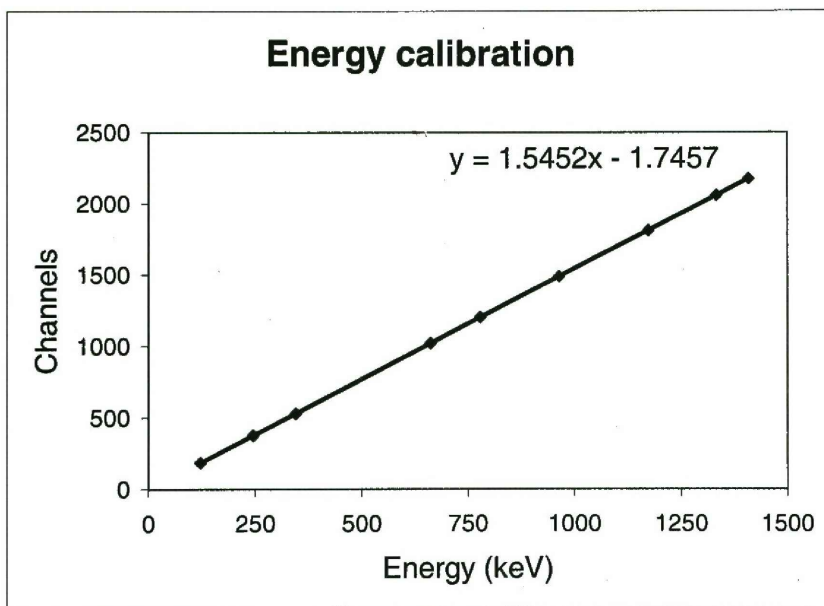


Figure 3.11: Plot of the energy calibration showing a linear relationship between centroid and γ -ray energy.

The energy resolution of the HPGe detector at a specific energy is determined by measuring the full-width-at-half-maximum (FWHM) of the peak centre at that energy. By measuring the FWHM at a number of energies it is possible to parameterise the dependence of FWHM on the energy by again fitting a polynomial (normally a linear function) to the data. A plot of FWHM versus the energy lines used from the standard source is shown in Figure 3.12.

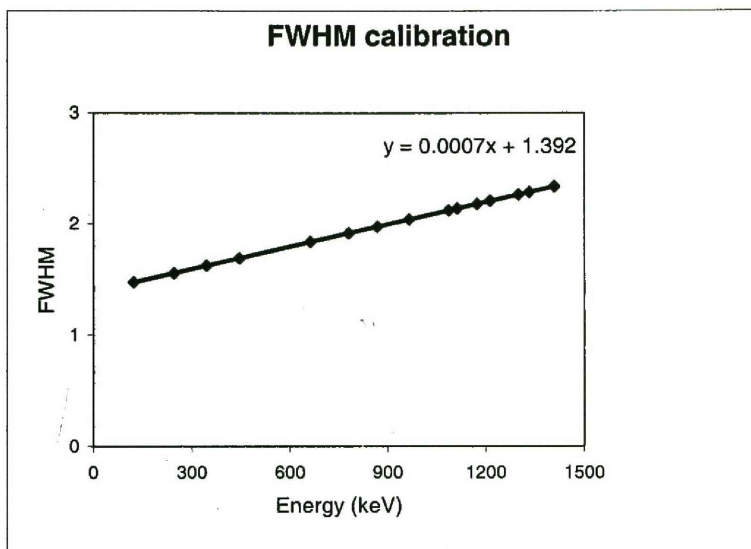


Figure 3.12: A plot of the FWHM versus energy for peaks indicated in Figure 3.10.

3.3 Sample counting

3.3.1 Milk

The samples were each counted for a period of nine hours (refer to Table 3.1). The dead time while measuring each sample was 0 % meaning that the detector was active all the time when the sample was counted. A typical spectrum of a milk sample is shown in Figure 3.13.

3.3.2 Mineral water

The mineral water samples were each counted for a period of nine hours for both the first and second measurements shown in Tables 3.2 (a) and (b). The dead time while measuring the samples was 0 %. A typical spectrum of a mineral water sample is shown in Figure 3.14.

3.3.3 Background: tap water

Shielding was provided around the detector to reduce background due to sources in the laboratory, cosmic radiation and building. Despite this, it is important to measure the background so that it can be subtracted from the sample counts. In the spectrum sample the background counts are unavoidably included in the overall calculated peak areas [Gil95]. A Marinelli beaker filled with 1 litre tap water was counted as a background for a period of 34 hrs. The dead time while measuring the sample was 0 %. Instead of using an empty Marinelli beaker as background water, tap water was used to normalize for the attenuation factor in both the background and the sample. A typical spectrum of the background is shown in Figure 3.15 and the corresponding spectrum for an empty Marinelli beaker is shown in Figure 3.16.

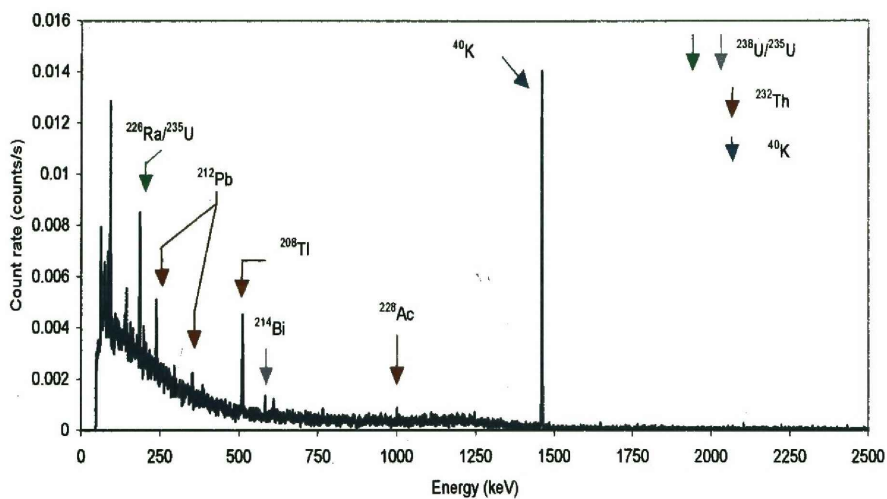


Figure 3.13: A typical spectrum of a milk sample with the location of natural lines from the ^{238}U , ^{232}Th and ^{40}K decay series shown.

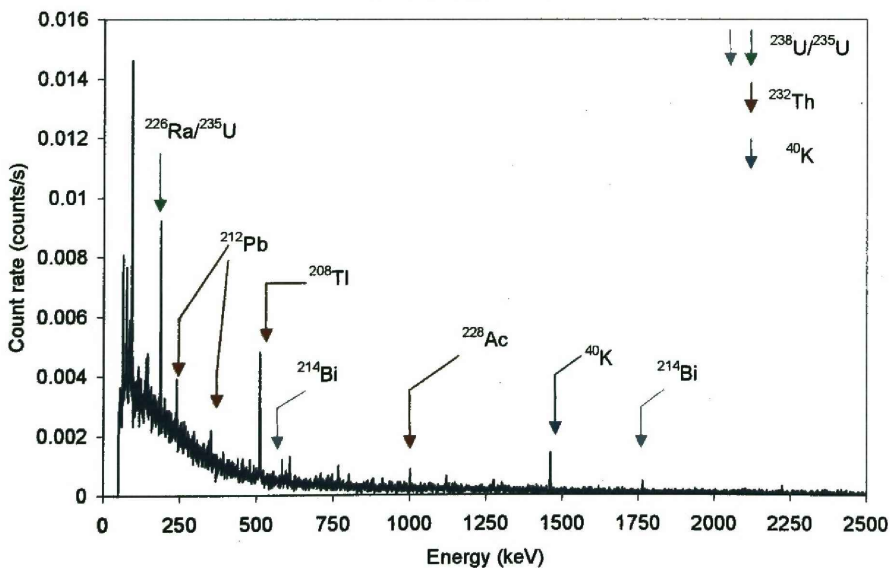


Figure 3.14: A typical spectrum of a mineral water sample with the location of natural lines from the ^{238}U , ^{232}Th and ^{40}K decay series shown.

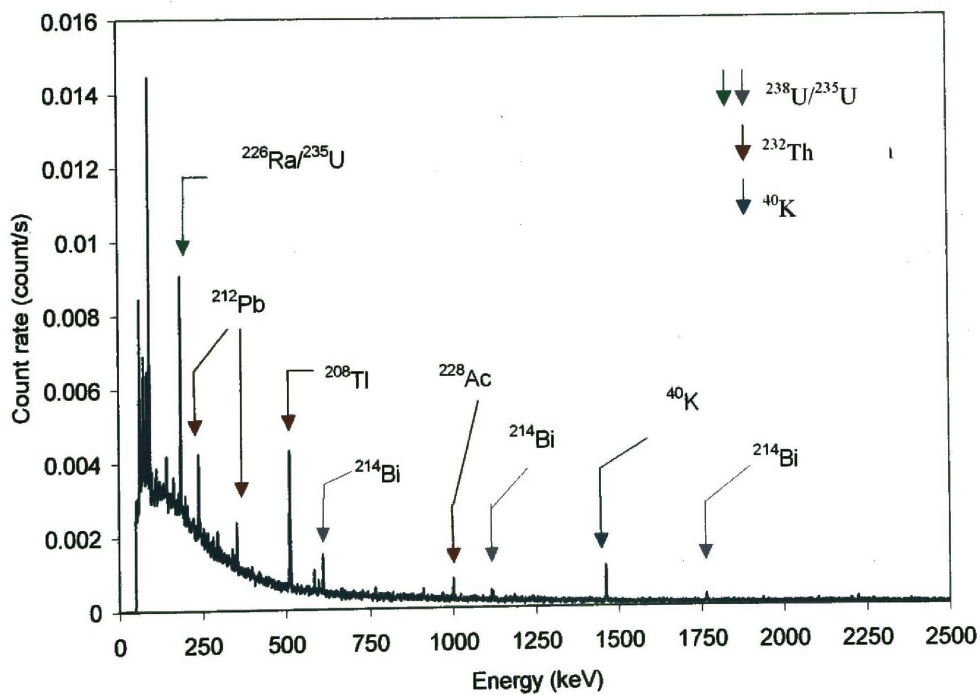


Figure 3.15: The background spectrum measured using a Marinelli filled with tap water.

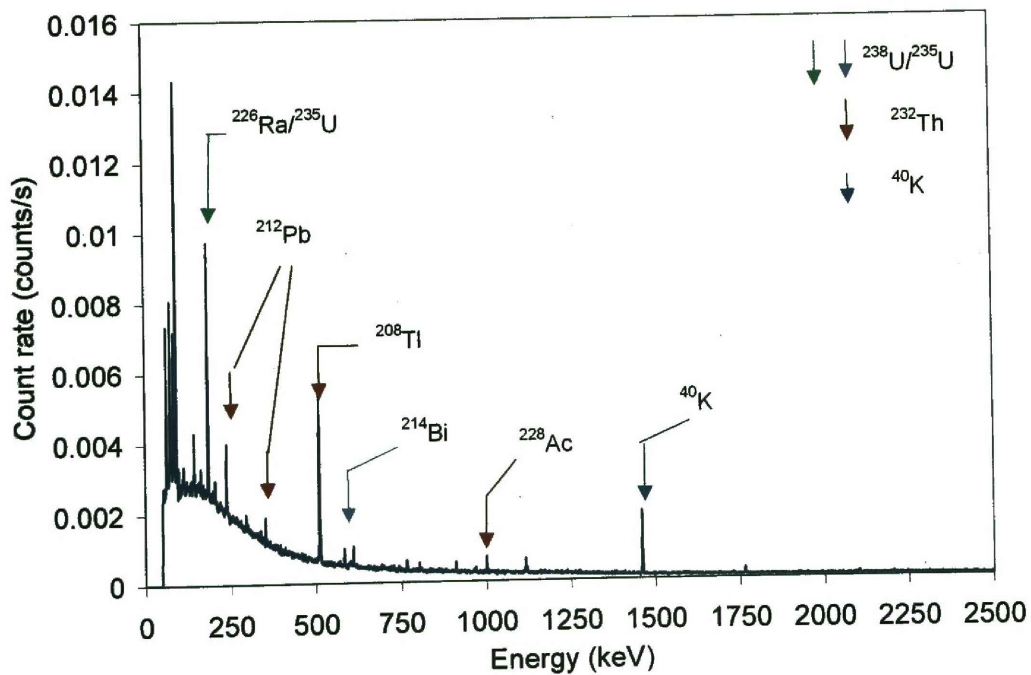


Figure 3.16: The spectrum measured using an empty Marinelli beaker.

Chapter 4

Data analysis and results

In this chapter, the calculation of activities, the determination of the full-energy peak detection efficiency, the minimum detectable activity concentrations of nuclides found in milk and mineral water samples and the estimation of the dose rate will be discussed.

4.1 Calculation of activity concentration

The activity concentration of the sample can be determined by counting the number of γ -rays (or any other particle) detected by the detector in a fixed period of time. The activity, A, can be calculated using:

$$A \text{ (Bq/l)} = \frac{C_b}{L_t * B_r * \epsilon * V} \quad (4.1)$$

where ϵ = detection efficiency, the ratio of the number of detected photons to the number emitted by the source,

C_b = net counts, after appropriate background subtraction,

L_t = live time, the time during which the system is available for processing a pulse,

A = activity of the sample,

V = volume of the sample and,

B_r = branching ratio, is the statistical chance that a particular γ -ray is emitted per decaying nucleus.

The net counts, C , for a particular full-energy peak from the spectrum is determined by manually setting what is called a region of interest (ROI) around it. The MCA software then uses an algorithm to automatically calculate the gross counts and net counts (i.e. those above background in the sample spectrum) associated with the ROI. As mentioned in chapter 3 the background was determined by means of a tap water measurement. The acquired background spectrum (see Figure 3.16) was analysed.

The appropriate peak areas were determined (Table 4.1 shows ROI data extracted from the background spectrum) and were subtracted from the sample peak areas, C , in order to determine C_b , the net peak area after subtracting the tap water background. To account for the difference in live times for sample and the background measurement, a ratio of the live times, r_T (for sample and background), was obtained. Therefore the net peak area, C_b , was calculated using the equation:

$$C_b = (C - B) \times r_T \quad (4.2)$$

where C is the peak area in the sample spectrum, B is the corresponding peak area in the tap water spectrum, and r_T is the sample live time divided by the tap water live time.

Table 4.1: A region of interest (ROI) report for the tap water background spectrum. The live time for the measurement was measured for 34.8 hours.

Decay	Nuclide	Energy (keV)	Gross (cnts)	Net (cnts)
$^{238}\text{U}/^{235}\text{U}$	$^{226}\text{Ra}/^{235}\text{U}$	185.6	5766 ± 76	2106 ± 161
Fallout	^{131}I	364.5	521 ± 23	47 ± 60
Fallout	^{137}Cs	661.7	229 ± 15	28 ± 39
^{232}Th	^{228}Ac	910.3	460 ± 21	180 ± 43
^{238}U	^{214}Pb	351.9	1510 ± 39	258 ± 81
^{40}K	^{40}K	1460.8	713 ± 27	512 ± 45

4.2 Calibration of γ -ray detection efficiency

As can be seen from equation 4.1 the detection efficiency calibration is required in order to determine activity concentrations. The detection efficiency is dependent on the geometry, density and chemical composition of a sample [Kno89]. The same standard liquid source solution used for the energy calibration (see chapter 3) was used for performing an efficiency calibration. The detector efficiency [Gil95] was calculated using the formula below:

$$\varepsilon = \frac{C}{L_i * B_r * A * V} \quad (4.3)$$

where the symbols have the same meaning as discussed with relation to equation 4.1. The volume of the sample was 1 litre and branching ratios [Fir96] associated with the γ -ray lines used in the analysis are tabulated in Table 4.2. The location of peaks in the standard

spectrum used to calculate the efficiency are shown in Figure 4.1. The activity of each nuclide was corrected for decay since preparation of the source. The calculated photopeak efficiencies are given in Table 4.2 and plotted in Figure 4.2. In order to parameterise the dependence of efficiency on the γ -ray energy a polynomial of a form $Y = ax^b$, with x representing γ -ray energy, a and b being free parameters, was fitted to the data in Table 4.1 using a least-squares fitting procedure. The resulting best-fit curve through the data is also shown in Figure 4.2 corresponding to $a = 0.935$ and $b = 0.636$.

Table 4.2: γ -ray energy lines from the standard source with associated branching ratios.

The efficiency quoted was obtained using equation 4.2.

Nuclide	γ -ray energy (keV)	Branching ratio	Efficiency (ϵ)
^{152}Eu	121.8	0.284	0.040
^{152}Eu	244.7	0.075	0.031
^{152}Eu	344.3	0.266	0.026
^{137}Cs	661.7	0.852	0.017
^{152}Eu	779.0	0.13	0.014
^{152}Eu	867.4	0.042	0.012
^{60}Co	1173.5	0.999	0.011
^{60}Co	1332.8	1.0	0.010
^{152}Eu	1408.4	0.208	0.010

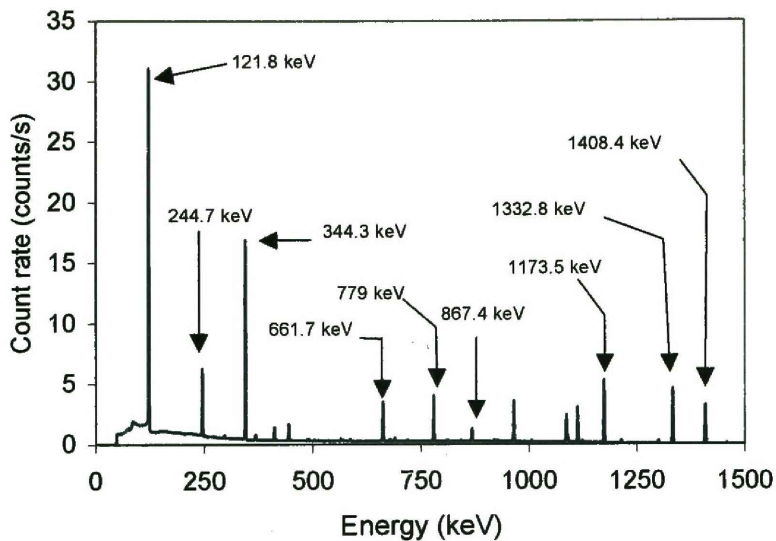


Figure 4.1: Location of peaks used in the efficiency calibration.

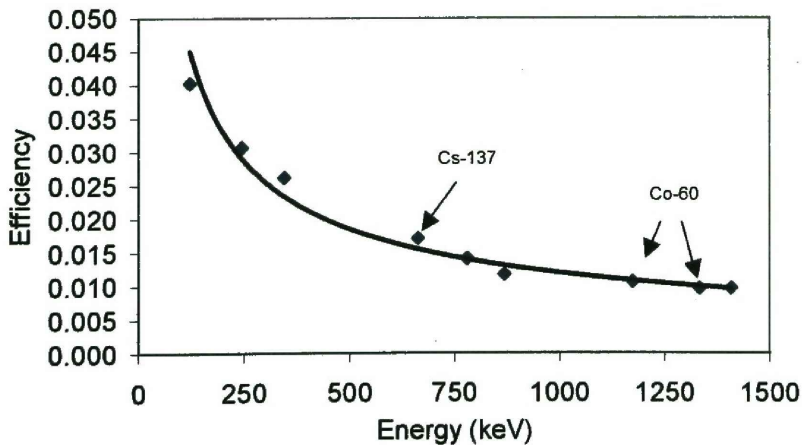


Figure 4.2: The efficiency curve fitted through efficiency values measured using the liquid standard source. The data points associated with ^{137}Cs and ^{60}Co are shown. The remaining data points are associated with ^{152}Eu decay.

Although ^{152}Eu is a convenient radionuclide to use for efficiency calibrations in view of its many associated decay γ -rays, the use of this nuclide in a volume geometry (as we use here) can lead to a distortion of the true efficiency due to an effect called coincident summing. Coincident summing occurs when a number of γ -rays are released from a decaying nucleus and they are detected at the same time. If this happens a pulse height, which represents the sum of energies of two individual energy lines, can be recorded [Gil95]. ^{152}Eu (see decay scheme in Figure 4.3) is a typical example of a nucleus that emits more than one γ -ray.

There was no peak of 466.06 keV (sum peak) observed, but peaks at 366.5, 1123.2 and 1529.8 keV (see Figure 4.4) were observed which indicate summing between 121.8 keV + 244.7 keV, 344.3 keV + 778.9 keV and 121.8 keV + 1408 keV [Gil95] γ -rays. Although the sum peaks are relatively small, they represent counts lost from the main peaks [Boy03], which are used in the calibration. Although algorithms exist to correct for coincidence summing [Def02] this was not attempted in this work.

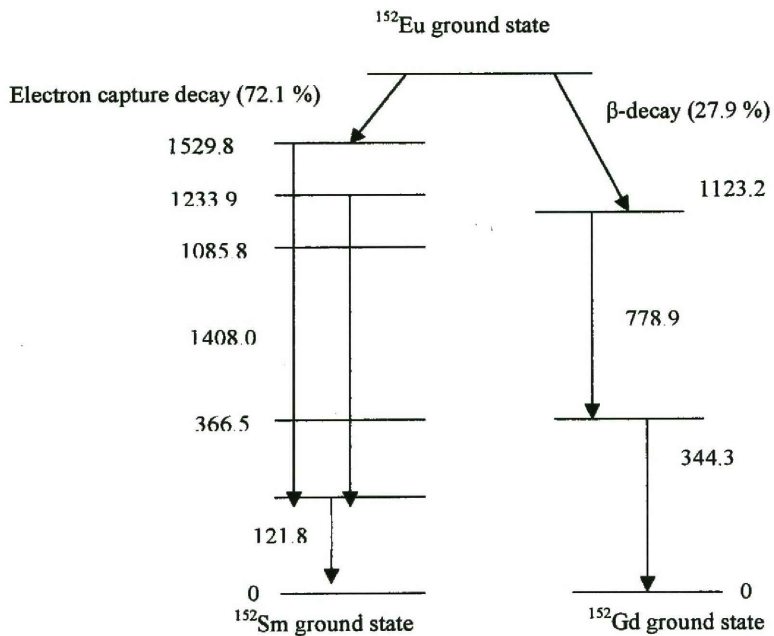


Figure 4.3: A decay scheme for ^{152}Eu .

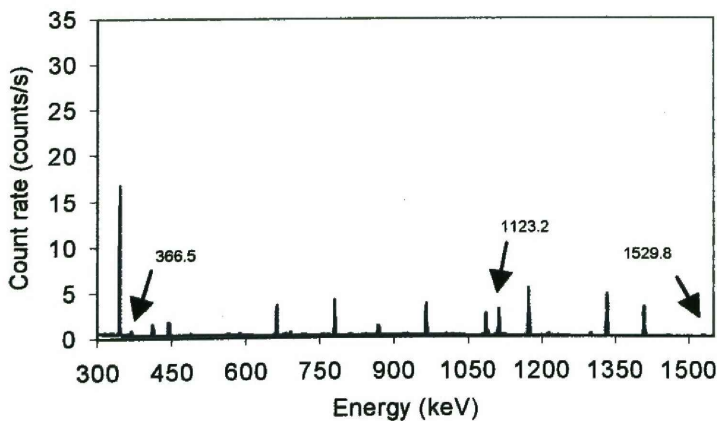


Figure 4.4: Sum peaks at 366.5, 1123.2 and 1529.8 keV in the liquid standard source spectrum due to coincidence summing effects.

4.3 Calculation of activities and results

4.3.1 Milk

A typical γ -ray spectrum for a milk sample with the acquired tap water background spectrum overlaid, for brand A, B, and C are shown in Figures 4.5 (a), 4.5 (b) and 4.5 (c), respectively. Regions from the spectra shown in Figures 4.5 (a-c) where γ -ray lines associated with ^{131}I (365 keV) and ^{137}Cs (662 keV) are expected, are shown separately in Figures 4.6, 4.7 and 4.8. In all milk spectra studied it is only the 1461 keV ^{40}K line, which is clearly distinguishable above the background. Despite this each milk sample spectrum was analysed using regions of interest (ROIs) centred on the γ -ray energies associated with the decay of ^{137}Cs , ^{131}I and ^{40}K . The ROI for the ^{137}Cs line was set using the observable peak in the standard source spectrum (Figure 4.9). The ROI for the ^{40}K line was set using the observed peak in the tap water background spectrum. The ROI for the ^{131}I line was estimated using the parameterisation of the variation of FWHM with energy. The ROIs, γ -ray energies and associated branching ratios [Fir96] used for this analysis are given in Table 4.3.

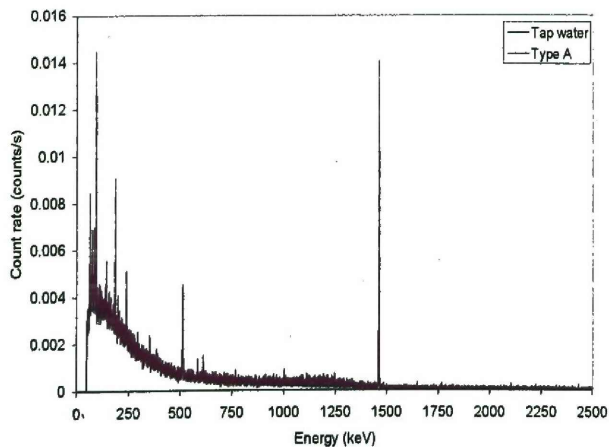


Figure 4.5 (a): Type A milk sample with tap water.

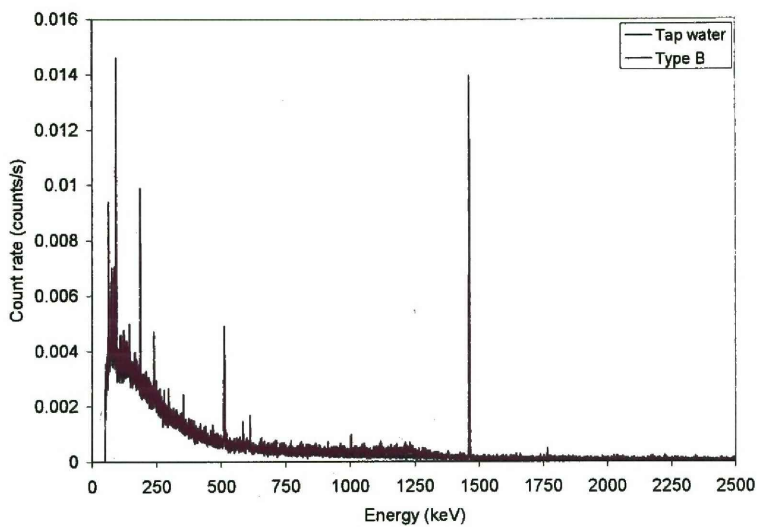


Figure 4.5 (b): Type B milk sample with tap water spectrum overlaid.

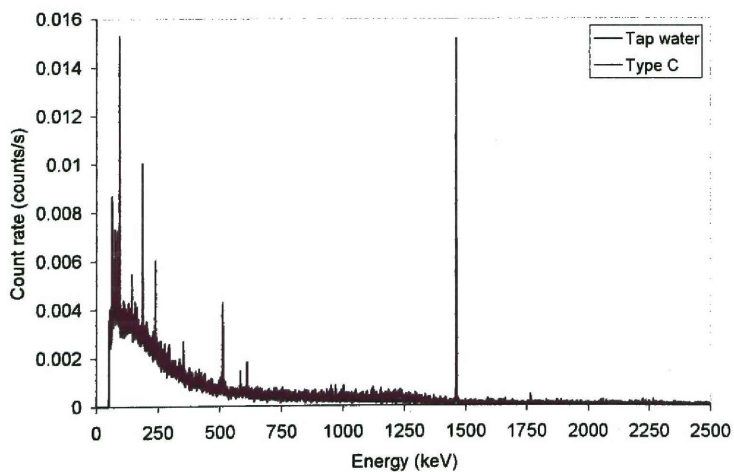


Figure 4.5 (c): Type C milk spectrum with tap water spectrum overlaid.

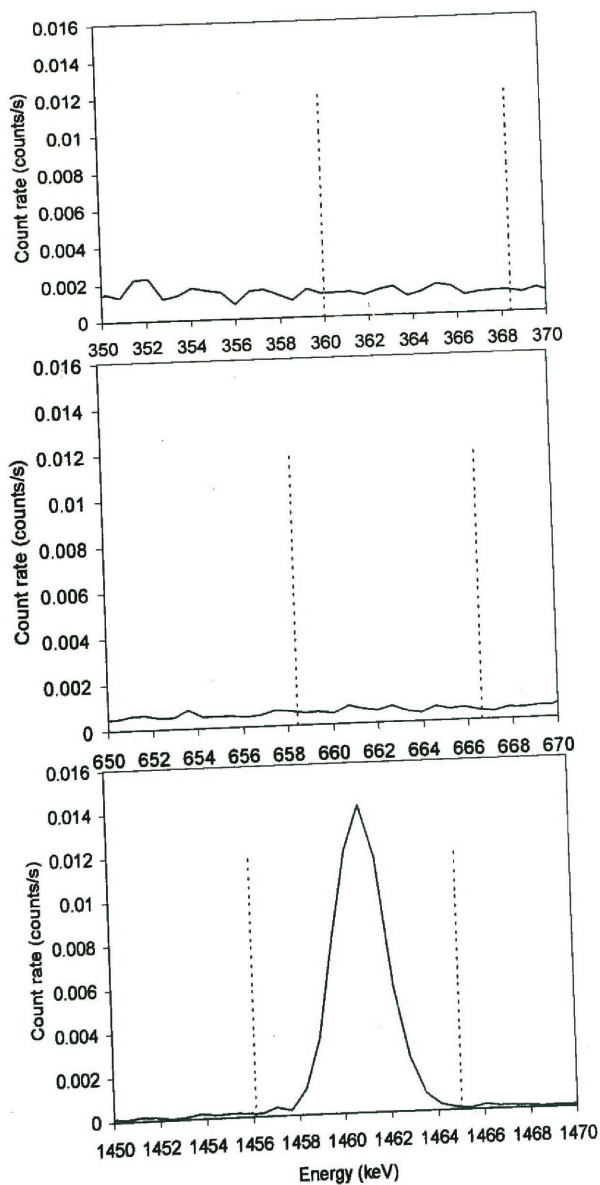


Figure 4.6: Type A milk sample showing expanded regions centred on 365, 662 and 1460 keV lines; the regions of interest used are shown (in Table 4.4) by dashed lines.

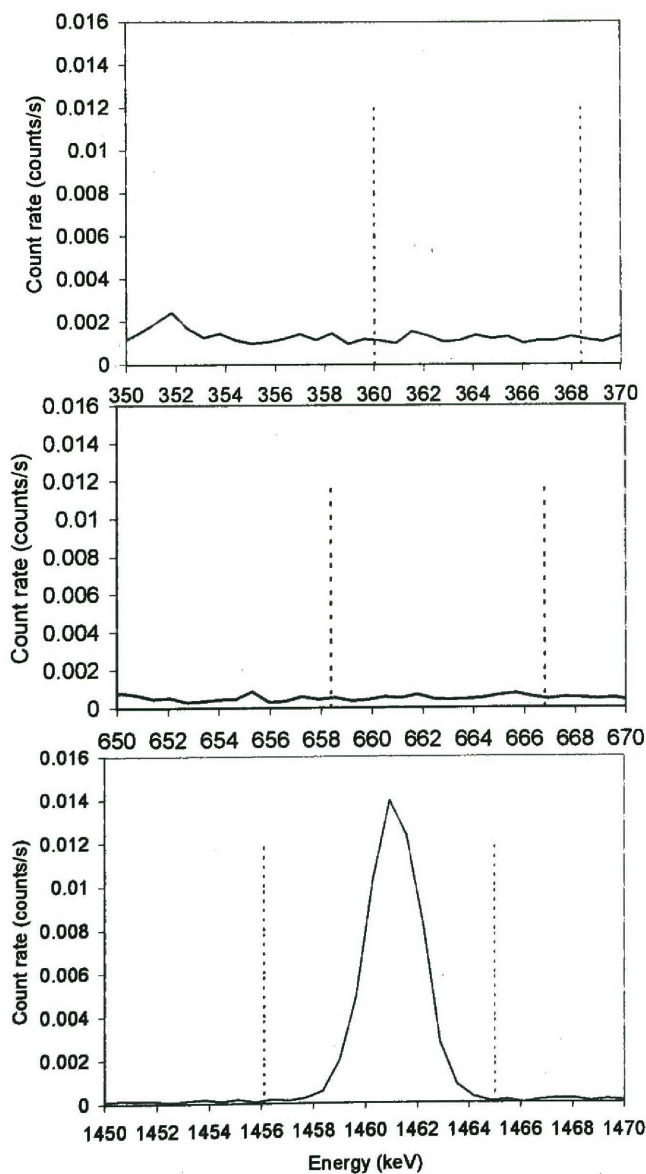


Figure 4.7: Type B milk sample showing expanded regions centred on 365, 662 and 1460 keV; the regions of interest used are shown in (Table 4.4) by dashed lines.

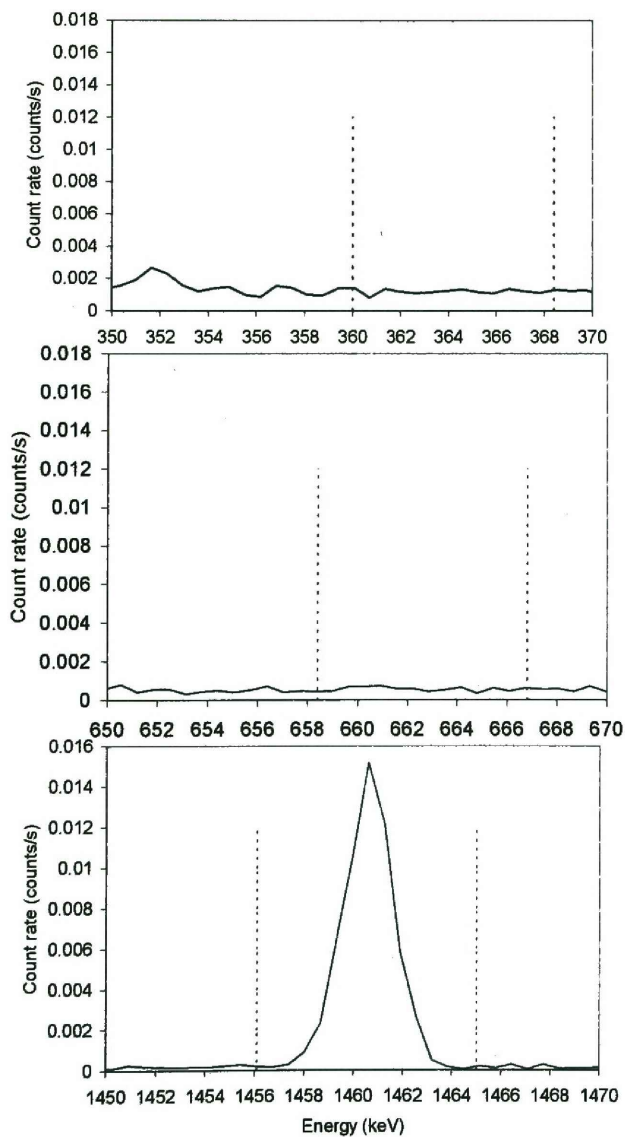


Figure 4.8: Type C milk sample showing expanded regions centred on 365, 662 and 1460 keV; the regions of interest used are shown in (Table 4.4) by dashed lines.

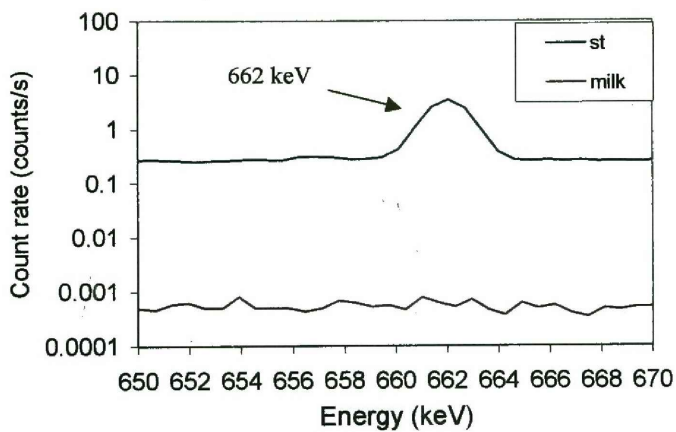


Figure 4.9: Portions of spectra of a sample (bottom) and liquid standard source (top).
The ^{137}Cs in the standard source sample spectrum is clearly visible.

Table 4.3: γ -ray energies and associated branching ratios [Eml97] used in the analysis
of milk sample spectra.

Decay series	Nuclide	Energy (keV)	Branching ratio
Fallout	^{131}I	364.3	0.812
Fallout	^{137}Cs	662.5	0.852
^{40}K	^{40}K	1460.6	0.107

Table 4.4: Regions of interest used in the analysis of milk sample spectra.

γ -ray energy	Start of ROI		End of ROI	
	Channels	KeV	Channels	keV
364.3	557	360	569	368.6
662.5	1018	658.3	1030	666.6
1460.6	2251	1456	2263	1464.3

Activity concentrations of ^{40}K , ^{137}Cs and ^{131}I present in the milk were calculated using equation 4.1. These calculated concentrations and associated uncertainties due to counting statistics are given in Table 4.5 (a) for each milk sample analysed, while average concentrations are given in Table 4.5 (b). Only the ^{40}K activity concentrations were found to differ from zero when taking the statistical uncertainties into account. The average ^{40}K activity concentrations in milk brands A and C were found to be the same within statistical uncertainties, while that for brand B was found to be 3 % lower. The average ^{40}K activity concentration obtained by averaging over all samples was found to be 51.4 ± 0.4 Bq/l. As mentioned in chapter 2, Shukla et al. measured the activity concentrations of fallout and natural radionuclides in Indian milk [Shu94]. For the period 1985 - 1990 they found an average ^{40}K activity concentration of 52.0 ± 4.4 Bq/l which is in agreement with the results from this work. This agreement indicates that the concentration of potassium in milk is similar in South Africa and India.

Table 4.5 (a): Activity concentrations (Bq/l) of nuclides of interest in milk sample.

Sample #	Milk brand	^{137}Cs	^{131}I	^{40}K
M1	A	0.028 ± 0.091	0.08 ± 0.10	54.1 ± 1.5
M4	A	-0.04 ± 0.11	0.07 ± 0.10	52.2 ± 1.5
M7	A	0.03 ± 0.10	0.09 ± 0.11	50.3 ± 1.6
M10	A	-0.04 ± 0.11	0.00 ± 0.11	51.6 ± 1.5
M2	B	-0.03 ± 0.09	0.00 ± 0.11	53.4 ± 1.4
M5	B	0.06 ± 0.09	0.00 ± 0.11	48.1 ± 1.5
M8	B	-0.04 ± 0.11	0.00 ± 0.11	47.8 ± 1.6
M11	B	0.04 ± 0.10	0.00 ± 0.11	52.1 ± 1.5
M3	C	0.01 ± 0.10	0.00 ± 0.12	49.5 ± 1.6
M6	C	-0.009 ± 0.066	0.00 ± 0.11	51.5 ± 1.7
M9	C	0.042 ± 0.095	0.00 ± 0.11	53.2 ± 1.5
M12	C	0.044 ± 0.095	0.071 ± 0.106	52.8 ± 1.5

Table 4.5 (b): The average activity concentrations (Bq/l) in milk brands A, B and C.

Milk brand	^{137}Cs	^{131}I	^{40}K
A	0.00 ± 0.05	0.06 ± 0.05	52.1 ± 0.8
B	0.01 ± 0.05	0.00 ± 0.06	50.4 ± 0.8
C	0.02 ± 0.05	0.02 ± 0.05	51.8 ± 0.8
A, B, C	0.01 ± 0.03	0.03 ± 0.29	51.4 ± 0.4

As can be seen from Table 4.5 (a) and (b), the activity concentrations of ^{137}Cs and ^{131}I were found to be consistent with zero. This means that the activities of these nuclides in milk are smaller than the minimum detectable activity (MDA) associated with the experimental configuration used. In view of this the MDA for ^{137}Cs and ^{131}I was calculated.

The minimum detectable activity is defined, as the minimum amount of radioactive nuclide, which can be determined given circumstances of that particular measurement, and is dependent on the quantity, type of sample, geometry of sample, nuclide identity, detector environment, counting time, and the confidence level assumed [Gil95]. The MDA can be calculated using the relation:

$$MDA = \frac{L_D}{\epsilon * B_r * L_T} \quad (4.3)$$

where L_D is the lower limit of detection and other symbols have the same meaning as for equation 4.1. L_D refers to the ability of counting system to detect a minimum number of counts in the sample and is statistically based. According to an early and still widely used method, the L_D is considered to be the count equal to two standard deviations of the background counts [Kat84]. In this way the L_D is determined at the 90 % confidence level, that is a sample containing an amount of activity equal to the L_D would be detected 90 % of the time. The calculated L_D and MDA concentration for the lines of interest in milk are given in Table 4.6. The background, B_M , used for the MDA calculation was taken to be the gross area (corrected for the live times) in the region of interest on the tap

water spectrum. The standard deviation, σ_b , of the background was taken to be equal to

$$\sqrt{B_M}.$$

Table 4.6: Minimum detectable activity concentrations of various nuclides.

Nuclide	γ - energy (keV)	Background (B_M) (counts)	σ_b (counts)	LD = $2\sigma_b$ (counts)	MDA (Bq/l)
^{40}K	1461	184	13.6	27.1	0.8
^{137}Cs	662	174	13.2	26.4	0.06
^{131}I	365	453	21.3	42.6	0.07
^{226}Ra (^{238}U series)	186	1497	38.6	77.1	1.9
^{228}Ac (^{232}Th series)	911	119	10.9	6.6	0.2
^{214}Pb (^{238}U)	352	390	19.7	39.5	0.1

The MDA concentrations of ^{137}Cs and ^{131}I were found to be 0.06 and 0.07 Bq/l, respectively. As expected these values exceed the measured average ^{137}Cs and ^{131}I activity concentrations (Table 4.5 (b)). The activity concentration of ^{137}Cs in milk in 1985 (prior to the Chernobyl incident) was found to be < 0.007 Bq/l in India [Shu94]. During the year of the Chernobyl explosion it was found to be 0.420 Bq/l, that is, sixty times higher than the year before. The average ^{137}Cs concentration in milk over the years 1987 - 1990 was found to be 0.12 Bq/l, which is twice the MDA concentration found in this study. Although we have not managed to detect any ^{137}Cs and ^{131}I in the milk

samples, we have managed to set upper limits of 0.06 and 0.07 Bq/l on the activity concentrations of ^{137}Cs and ^{131}I in the milk analysed respectively. These values will serve as a useful benchmark against which future results for the Western Cape can be compared.

4.3.2 Mineral water

Typical mineral water spectra for brands A and B, shown with the tap water background spectrum overlaid, are shown in Figures 4.10 (a) and 4.10 (b). The nuclides, γ -ray energies and associated branching ratios [Eml97] used in the analysis of the mineral water spectra are given in Table 4.7, while the regions of interest used for the spectral analysis are given in Table 4.8. The ROI were determined from the tap water background spectrum. As can be seen from Table 4.7 the focus here is on the natural nuclides ^{226}Ra , ^{238}U , ^{232}Th and ^{40}K . The activity concentration of ^{238}U was determined using the 352 keV line associated with the decay of ^{214}Pb , a daughter formed in the decay of ^{238}U . The activity concentration of ^{232}Th was determined using the 911 keV line associated with the decay of ^{228}Ac , a daughter formed in the decay of ^{232}Th . The branching ratio for the 186 keV line associated with the ^{226}Ra decay (given in Table 4.7) was adjusted to take into account the fact that ^{235}U emits a 185.7 keV γ -ray during its decay.

Activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K present in the mineral water were calculated using equation 4.1. These calculated concentrations and associated uncertainties due to counting statistics are given in Table 4.9 (a) (before secular equilibrium was reached in the ^{238}U decay) and 4.10 (a) (after secular equilibrium was reached in the decay of ^{238}U) for each mineral water sample analysed, while average

concentrations are given in Table 4.9 (b) (before equilibrium) and 4.10 (b) (after equilibrium). All but one activity concentration extracted from the first set of measurements was found to be consistent with zero. The exception was the activity concentration of ^{238}U , which was found to be 0.53 ± 0.17 Bq/l for sample Mw3 (brand A). The measurement of the same sample after storing it in a sealed Marinelli beaker for a period of at least 3 weeks yielded an activity concentration of -0.12 ± 0.17 Bq/l. This points to the possibility that there were systematic errors associated with some of the measurements. The MDA concentrations (refer to Table 4.6) for ^{226}Ra , ^{232}Th , ^{238}U and ^{40}K associated with our measurement were found to be 1.9, 0.2, 0.1 and 0.8 Bq/l, respectively. All extracted average activity concentrations from the second set of measurements were found to be below the MDA concentrations.

Once again we have only managed to set upper limits of 1.9, 0.2, 0.1 and 0.8 Bq/l on the activity concentrations of ^{226}Ra , ^{232}Th , ^{238}U and ^{40}K , respectively, in the mineral water analysed. These values will serve as a useful benchmark against which future results for the Western Cape can be compared.

As mentioned in Chapter 2, Botezatu et al. conducted a study of natural nuclide activity concentrations in Romanian mineral water [Bot02]. They found ^{226}Ra activity concentrations ranging from 0.009 to 0.226 Bq/l and ^{40}K concentrations ranging from 0.078 to 0.220 Bq/l. In the study by [God00] et al. of Brazilian mineral waters ^{226}Ra activity concentrations were found to vary between 0.008 and 0.083 Bq/l. It is clear that if the activity concentrations in the mineral water studied here are similar to that in Romania and Brazil, the measurement technique employed in this study will not allow the detection of at least ^{226}Ra and ^{40}K . In a recent tabulation of ^{226}Ra activity

concentrations of mineral water [Kra03] the highest concentration given is that of 2.185 Bq/l for mineral water having its origins in Portugal. If the activity concentrations in the water studied was as high as this we would have been able to measure it.

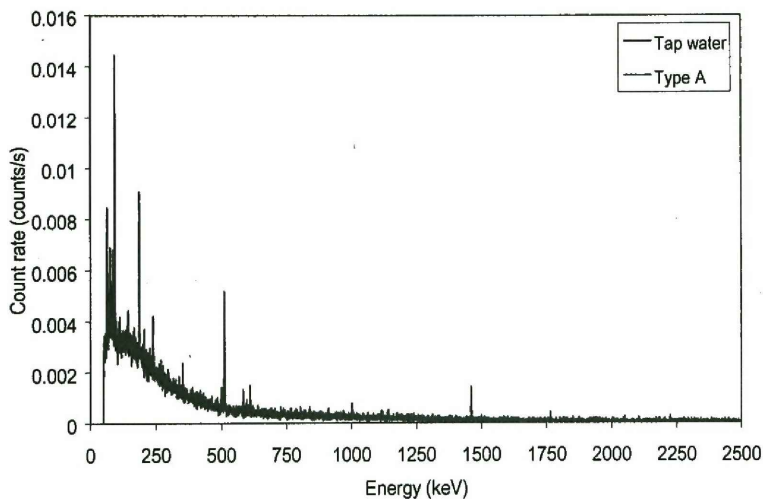


Figure 4.10 (a): Type A mineral water spectrum with tap water overlaid.

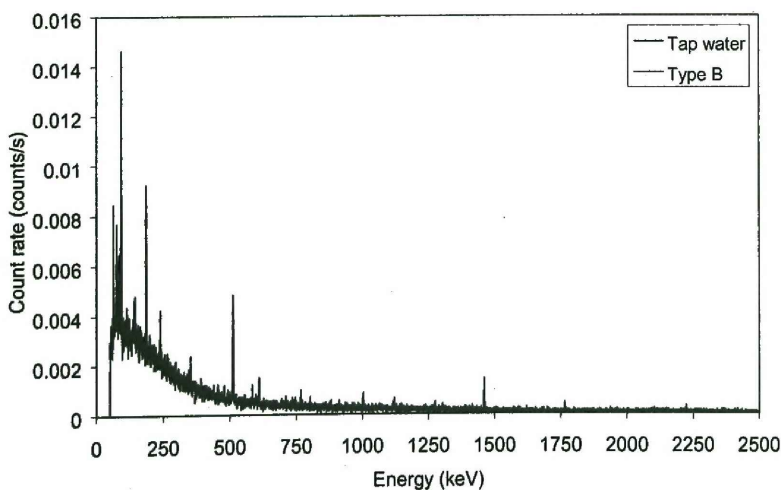


Figure 4.10 (b): Type B mineral water spectrum with tap water overlaid.

Table 4.7: γ -ray energies and associated branching ratios [Em197] used in the analysis of mineral water.

Decay series	Nuclide	Energy (keV)	Branching ratio
$^{238}\text{U}/^{235}\text{U}$	^{226}Ra	186.1	0.0579
^{232}Th	^{228}Ac	911.2	0.29
^{238}U	^{214}Pb	352	0.317
^{40}K	^{40}K	1460.8	0.107

Table 4.8: Regions of interest used in the analysis of mineral water samples.

	Start		End	
Nuclide	Channels	KeV	Channels	keV
^{226}Ra	284	187.7	294	188.8
^{228}Ac	1405	907.3	1414	913.6
^{238}U	542	348.8	549	353.8
^{40}K	2253	1456.2	2265	1464.4

*Table 4.9 (a): Activity concentrations (Bq/l) determined from first set measurements
(directly after sealing) of the mineral water samples.*

Sample #	Brand	^{226}Ra	^{40}K	^{232}Th	^{238}U
Mw1	A	-1.3 ± 1.5	0.03 ± 0.81	-0.17 ± 0.2	0.21 ± 0.16
Mw2	A	0.6 ± 1.4	0.10 ± 0.78	0.17 ± 0.18	0.24 ± 0.37
Mw3	A	1.4 ± 1.5	-0.8 ± 0.76	-0.01 ± 0.23	0.53 ± 0.17
Mw4	B	-1.0 ± 1.4	-0.34 ± 0.55	0.02 ± 0.21	0.04 ± 0.16
Mw5	B	-1.9 ± 1.5	-0.58 ± 0.78	-0.06 ± 0.21	0.04 ± 0.16
Mw6	B	1.3 ± 1.4	0.36 ± 0.76	-0.36 ± 0.23	-0.15 ± 0.17

*Table 4.9 (b): Average activity concentrations (Bq/l) in mineral water measured directly
after sealing.*

Brand	^{226}Ra	^{40}K	^{232}Th	^{238}U
A	0.24 ± 0.83	-0.23 ± 0.78	0.00 ± 0.19	0.32 ± 0.15
B	-0.56 ± 0.83	-0.2 ± 0.68	-0.02 ± 0.13	-0.02 ± 0.09
A+B	-0.16 ± 0.59	-0.2 ± 0.52	-0.01 ± 0.11	0.15 ± 0.09

Table 4.10 (a): Activity concentrations (Bq/l) from the second set measurements (after secular equilibrium was reached) in mineral water samples.

Sample no	Brand	^{226}Ra	^{40}K	^{232}Th	^{238}U
Mw1	A	-0.3 ± 1.4	-0.19 ± 0.68	-0.07 ± 0.21	0.13 ± 0.16
Mw2	A	-0.5 ± 1.4	1.22 ± 0.68	-0.07 ± 0.21	0.13 ± 0.16
Mw3	A	2.1 ± 1.4	1.34 ± 0.65	-0.12 ± 0.21	-0.12 ± 0.17
Mw4	B	1.0 ± 1.4	0.21 ± 0.78	-0.39 ± 0.23	0.05 ± 0.16
Mw5	B	-1.0 ± 1.4	-0.82 ± 0.84	-0.21 ± 0.23	-0.02 ± 0.16
Mw6	B	-2.7 ± 1.4	0.70 ± 0.60	0.18 ± 0.19	0.1 ± 0.15

Table 4.10 (b): Average activity concentrations (Bq/l) in mineral water after sealing.

Brand	^{226}Ra	^{40}K	^{232}Th	^{238}U
A	0.40 ± 0.82	0.79 ± 0.39	0.00 ± 0.18	0.05 ± 0.09
B	-0.89 ± 0.81	0.03 ± 0.43	-0.24 ± 0.13	0.04 ± 0.09
A+B	-0.25 ± 0.57	0.41 ± 0.29	-0.16 ± 0.11	0.05 ± 0.07

4.4 Ingestion dose estimates: results and discussion

The annual ingestion dose rate for an adult was calculated using activities and dose conversion factors [IAE96] by means of the equation:

$$\text{dose rate (mSv/y)} = A \times \text{DCF} \quad (4.4)$$

where A is the annual activity ingested by an adult (older than 17 years) and DCF is the dose conversion factor. The DCFs give the dose per unit intake by ingestion or inhalation for different radionuclides for adults and children. Consider the case where there are two nuclides, a and b, that are ingested. In this case the total dose is calculated using the relation:

$$\text{dose rate (mSv/y)} = A_a \times \text{DCF}_a + A_b \times \text{DCF}_b \quad (4.5)$$

where the subscripts a and b denote the respective nuclides. In the same way it is possible to adapt equation 4.4 to the case where there are n nuclides ingested.

4.4.1 Milk

In the case of milk the aim is to calculate the ingestion dose due to the anthropogenic nuclides ^{137}Cs and ^{131}I . Although the activity concentrations of ^{40}K in milk could be measured the ingestion dose contribution from this nuclide is normally not calculated since potassium is under homeostatic control in the body [Shuk94]. An average adult receives a dose of about 190 microSv/y from ^{40}K present in the body [Shu94]. The dose calculations for milk are based on the assumption used by Shukla *et al.* [Shuk94] that an adult consumes 37 l (about 100 ml a day) annually. Since the activity concentrations of ^{137}Cs and ^{131}I were found to be consistent with zero to within uncertainties the dose

calculations were made using the minimum detectable activities (MDAs) (see Table 4.6) for these nuclides. This means that the dose calculated here represents an upper limit. The dose conversion factors associated with ingestion of ^{137}Cs and ^{131}I by an adult are given Table 4.11. Equation 4.4 was used to calculate the maximum dose rate from ^{137}Cs and ^{131}I ingestion, respectively. The results are given in table 4.12.

Table 4.11: The dose conversion factors associated with the ingestion of radionuclides analysed both in milk and mineral water samples. Taken from [IAE96] for adults (older than 17 years).

Nuclide	DCF (Sv/Bq)
^{137}Cs	1.3×10^{-8}
^{131}I	2.2×10^{-8}
^{226}Ra	2.8×10^{-7}
^{232}Th	2.3×10^{-7}

Table 4.12: The maximum dose to adults (older than 17 years) from fallout radionuclides through the intake of milk assuming a consumption of 37 l per year.

Nuclide (s)	Max. activity ingested (Bq/y)	Max. dose rate (microSv/y)
^{137}Cs	2.22	0.029
^{131}I	2.59	0.057
^{137}Cs and ^{131}I	4.81	0.086

The average annual per capita dose received by the population of Bombay by intake of fallout and natural radionuclides through milk and diet was 92 microSv/y over the period 1987 - 1990, with ^{137}Cs from the ingestion of milk contributing 0.06 microSv/y [Shuk94]. Shukla et al. did not measure ^{131}I activities. The maximum annual per capita dose rate for ^{137}Cs we found is about one third of the corresponding (average) value for Bombay in the period 1987 - 1990. The global average per capita ingestion dose as determined in 1988 is 316 microSv/y (including ^{40}K) [Shuk94]. The maximum total dose due to ^{137}Cs and ^{131}I determined in this study is only 0.03 % of this average.

4.4.2. Mineral water

In the case of mineral water the aim is to calculate the ingestion dose due to the natural nuclides. Here only the contribution from ^{226}Ra and ^{232}Th is considered. The dose calculations for mineral water are based on the assumption used recently by Kralik *et al.* [Kra03] in an Austrian study that an adult consumes 80 l of mineral water annually (about 200 ml a day). Since the activity concentrations of ^{226}Ra and ^{232}Th were found to be consistent with zero to within uncertainties the dose calculations were made using the minimum detectable activities (MDAs) (see Table 4.6) for these nuclides. This means that the dose calculated here again represents only an upper limit. The dose conversion factors associated with ingestion of ^{226}Ra and ^{232}Th by an adult are given table 4.11. Once again equation 4.4 was used to calculate the maximum dose rate from ^{226}Ra and ^{232}Th ingestion, respectively. The results are given in table 4.13.

*Table 4.13: The maximum dose to adults (older than 17 years)
through the intake of mineral water assuming a consumption of 80 l
per year.*

Nuclide(s)	Max. activity ingested (Bq/y)	Max. dose rate (microSv/y)
²²⁶ Ra	152	42.6
²³² Th	16	3.7
²²⁶ Ra and ²³² Th	168	46.3

Kralik *et al.* found the maximum ingestion dose associated with ²²⁶Ra to be 8 microSv/y for adults corresponding to a measured maximum ²²⁶Ra concentration of 0.23 Bq/l. The reason for the big difference between their value and that from this study, i.e. 43 microSv/y, is that we used the estimated MDA concentration value of 1.9 Bq/l for ²²⁶Ra. The average annual ingestion dose (excluding ⁴⁰K) received from food and water in Austria for all groups of the population, except infants < 1 year, is 50 microSv/y. It is clear that the experimental technique used in this work does not allow for the determination of an accurate enough maximum dose rate associated with the ingestion of mineral water.

Chapter 5

Summary and outlook

5.1 Summary

The partial aim of the study was to determine the radioactivity levels of anthropogenic radionuclides, in particular ^{137}Cs and ^{131}I , in milk in view of the possible transfer to the public via the soil-grass-cow-milk pathway, and natural radionuclides in bottled mineral water. The results obtained by means of high-resolution γ -ray spectrometry, showed that the activities obtained were found to be consistent with zero to within statistical uncertainties in the milk samples. Since the peaks of the anthropogenic radionuclides, ^{137}Cs and ^{131}I were not observed, their minimum detectable activities (MDA) were estimated and were found to be 0.06 and 0.07 Bq/l for ^{137}Cs and ^{131}I , respectively for counting 1 litre of milk for a 9 hr period. The only natural radionuclide observed above background was ^{40}K , with the average activity of 51.4 ± 0.4 Bq/l. The activity concentration of ^{40}K was found to be consistent with the other values obtained in literature [Shu94]. For the dose rate estimation, IAEA dose conversion factors were used. The annual dose to an adult (older than 17 years) who consumes 37 l of milk per year was found to be less than $0.086 \mu\text{Sv}$, which is less than the 1 mSv annual dose limit received by a member of the public.

The average activity concentrations of natural radionuclides in mineral water were found to be consistent with zero to within statistical uncertainties and below the MDA. For

radionuclides below the background, their MDAs were used to calculate the dose as in mineral water. The annual dose estimated to an adult (older than 17 years) who consumes 80 l of mineral water per year was less than 46.3 μSv , which is below the annual dose limit of 1 mSv for a member of the public.

5.2 Outlook

The method employed to determine the activity concentrations of anthropogenic and natural nuclides in liquid milk and mineral water involved no elaborate sample preparation. In future this will allow for rapid determination of activity concentrations for example in liquid milk. The capability could be of value in the event that there is a need to rapidly screen the milk following a major radiation accident or fallout from future nuclear weapon explosions.

In order to improve the accuracy of the analytical method used in this study, it would be necessary to introduce corrections for the coincidence summing effect when calculating the full-energy peak detection efficiency. As mentioned above, the effect was observed in this work due to the nature of the ^{152}Eu decay. Alternatively, additional standard sources of ^{40}K , ^{137}Cs , ^{131}I and ^{238}U in Marinelli geometry will have to be obtained and used to calculate the detection efficiency response.

In this work only ^{40}K was detected above the background level in milk samples. The activity concentration of ^{137}Cs , ^{131}I , ^{238}U and ^{232}Th were found to be less than the minimum detectable activity (MDA). In order to determine the activities of these nuclides in milk and mineral water, ways to increase the concentration of nuclides in the sample

measured will have to be considered. One possibility in this regard is the use of evaporation lamps to eliminate the problem of spattering, ashing the samples to be able to count large volumes and using other methods like liquid scintillation counting.

APPENDIX

Uncertainty propagation

In radioactivity measurements, there may be several sources of uncertainty (peak measurement, source preparation, standard calibration and efficiency estimation), which have to be taken into account. When calculating the uncertainties one cannot associate the standard deviation σ with the square root of any quantity which is not a directly measured number of counts [Kno79]. In the case where the calculation is a sum or difference of counts the following is applied:

If

$$u = x + y \text{ or } u = x - y \quad (\text{A1})$$

then

$$\sigma_u = \sqrt{\sigma_x^2 + \sigma_y^2} \quad (\text{A2})$$

This is applied mostly in cases where the counts due to a radioactive source must be corrected by subtracting an appropriate background count. Since x and y are both directly measured numbers of counts, the expected standard deviation of each is known to be its own square root [Kno79], therefore the objective is to deduce the expected standard deviation of the net counts as a derived number. In the case where one has a multiplication or division of counts the following is applied:

$$u = xy \text{ or } u = \frac{x}{y} \quad (\text{A3})$$

then

$$\left(\frac{\sigma_u}{u}\right)^2 = \left(\frac{\sigma_x}{x}\right)^2 + \left(\frac{\sigma_y}{y}\right)^2 \quad (\text{A4})$$

For either $u = xy$ or $u = \frac{x}{y}$ the fractional uncertainty in x and y combine in a quadrature sum to give a fractional error in u . In order to calculate average activities, f , the following equation was applied:

$$f = \frac{a+b+c}{3} \quad (\text{A5})$$

where a , b and c are three measured activities and the uncertainty, $\Delta \bar{f}$, associated with the average activity can be calculated using the expression:

$$\Delta \bar{f} = \frac{\sqrt{\Delta a^2 + \Delta b^2 + \Delta c^2}}{3} \quad (\text{A6})$$

where Δa , Δb and Δc are uncertainties of the measured activities.

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