

Comparative Investigation of Airborne and Ground-based Radiometric Survey Techniques

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Declaration

I declare that this Dissertation is my own, unaided work. It is being submitted for the Degree of Master of Science in the University of the Witwatersrand, Johannesburg. It has not been submitted before for any degree or examination in any other University.

A handwritten signature in dark ink, consisting of several loops and a long horizontal stroke, positioned above a dashed horizontal line.

James Larkin

4th day of September 2013

Abstract

An investigation was undertaken to compare airborne and ground-based radiometric survey techniques and to compare and contrast their relative merits and how they can be used in determining the environmental distribution of environmental radioisotopes, particularly those in the decay chains of ^{238}U , ^{232}Th and ^{40}K .

Before a detailed investigation was done of the survey techniques, it was necessary to look at the underlying physical principles of detection of ionizing radiation and the types of detector that are generally used in these different types of survey techniques.

Having looked at the physics of detection, a detailed examination of the potential distribution of these environmental radioisotopes was undertaken.

In these surveys, an assumption is often made, that the daughter isotopes in a decay chain are in secular equilibrium with one another. This assumption was examined and the various possible ways in which secular equilibrium could break down were considered, these included looking at biological, meteorological and chemical processes.

Only after all the influences on assumptions used in these survey processes and the physical limitations on the measurements taken during surveys were considered, was a comparison made of a set of airborne and ground-based measurements taken at a chosen survey site compared. These measurements compared uranium, thorium and potassium activity concentrations, which had been determined by the two survey techniques, and a correlation was found particularly when the uranium measurements were examined.

Dedication

To my Friends, Family, and Colleagues who kept me going when the enthusiasm was lacking

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Nomenclature

EPA	Environmental Protection Agency. A United States of America Federal Agency responsible for the protection of the environment
e-Thorium, e-Th	As it is technically very difficult to directly measure the activity of thorium in an airborne radiometric survey, it is assumed that there is dynamic equilibrium in the ^{232}Th decay chain: It is assumed that the activity of ^{208}Tl which can be directly measured is equivalent to the activity of the thorium in the chain. By convention, thorium activity concentrations are reported as equivalent concentrations.
e-Uranium, e-U	As in the way that thorium is reported, uranium also is reported as an equivalent activity concentration, but in this instance the isotope that is directly measured is ^{214}Bi in airborne radiometric surveys
FWHM	Full width half maximum
GIS	Geographical information system. A software program designed to plot geographical information in a visual manner
Gps	Global positioning system
IAEA	International Atomic Energy Agency
ICRP	International Commission on Radiological Protection
sievert	SI unit of effective dose. Jkg^{-1}
TENORM,	Technologically enhanced naturally occurring radioactive material
UNSCEAR	United Nations Standing Committee of the Effects of Atomic Radiation

CHAPTER 1

INTRODUCTION

The search for financially viable deposits of uranium and thorium has driven the development of various techniques to locate such deposits. The original impetus dates back to the Second World War, when the Allies began looking for deposits of uranium that could be processed to provide the enriched uranium used in the atomic weapons needed to bring the war to a close. The succeeding Cold War, and now the renaissance in the nuclear power generation industry worldwide, provided further impetus. As these techniques have developed, they have successively increased the amount of usable information that can be obtained, to the point where nowadays much can be learnt about the mineralisation of the top layers of earth. In addition, much more can be deduced about the underlying geology and presence of different minerals that have an economic value.

Much less well developed has been the use of radiometric surveys to assess the prevailing radiological conditions in terms of a potential radioactive dose that an individual might receive by staying in a particular area. Such information is of interest to a number of different types of organisation, from those that offer a dosimetry service to organisations that are responsible for regulatory control of any such potential exposures that people might receive either at work or whilst living in a particular area.

In the development of this research project, it became apparent that there is an opportunity to extract dosimetric data from the previously collected airborne radiometric data and use it to develop dose-rate maps etc., which could prove useful to the previously mentioned types of organisation. During the initial stages of the investigation, a number of areas were chosen for which the Council for Geosciences had already carried out airborne radiometric surveys. Comparisons could then be made with ground based radiometric surveys of differing types carried out by the author. These surveys included the collection of a number of soil samples which were then submitted for analysis by high resolution gamma-ray spectroscopy, measurements taken *in situ* using a portable multi-channel

analyser coupled to a sodium iodide detector and a scanning survey looking at total counts as recorded by this detector.

Stemming from the last 50 years of investigation, a number of different techniques have been developed for the determination of the distribution of radioactive materials over the surface of the Earth. There are many reasons for this development including the following: the search for mineral deposits, the detailing of the disposition of radioactivity within the environment (be it of man-made origin or a natural source) and also as a means to measure radioactivity levels for a regulatory purposes or during an accident or emergency. The various *in situ* survey techniques can be broadly split into two groups: airborne radiometric surveys and ground-based radiometric surveys. Each of these types of survey technique makes use of arrays of detectors to measure the intensity and distribution of gamma photon emissions from radioactive materials.

Within this dissertation, the results of different survey techniques from each of the two groups will be compared in a specific case. The feasibility of using a ground-based survey to calibrate an airborne radiometric survey will be examined. The aim is to quantify the airborne radiometric survey so that its relationship to the distribution of the principal radioactive isotopes on the surface and near the surface can be better understood. Many of the original airborne surveys were just a record of the total count rate but it would be desirable to turn this total count into a dose rate. This relationship can then be used for a better understanding of the capabilities and limitations of airborne surveys and to establish how they can be applied for regulatory purposes. To achieve this, the aerial survey should be able to detect these radionuclides at an average activity level below 500 Bqkg^{-1} and preferably to a level of about 80 Bqkg^{-1} over an appropriate area which in terms of a dose rate is equivalent to less than $0.1 \mu\text{Svh}^{-1}$ at ground level. These values are in line with international standards developed by the International Commission on Radiation Protection [ICRP 1990] and implemented by the International Atomic Energy Authority in Basic Safety Series 115 [IAEA 1996] and are those which the National Nuclear Regulator of South Africa currently uses for the unconditional release from further regulatory control of land which was formally contaminated by radioactive material.

The present research can be compared with the MINEO [MINEO 2000] project that has been started in Europe. The emphasis and scope, however, is somewhat different. In the MINEO project, an attempt is being made to develop tools and methods for assessing and monitoring the environmental impact of mining. Radiometric surveys are just one of a number of different techniques that are being utilised. The final aim of the MINEO project is similar though, in that efforts are being made to provide regulators with the necessary information to make informed decisions about areas under regulatory consideration. The scope is far wider in the MINEO project, in that five separate European sites are being surveyed. Those sites are: the Mestersvøj mining area in Greenland, Lahnaslampi talc mine in Finland, the Ezberg mine in Austria, the Kirchheller Scheide coal mine in Germany, the West Cornwall mining district in the United Kingdom and the São Domingos mine in Portugal. These sites have been chosen as being representative of the different types of mining site in Europe. Within this dissertation, however, a more limited approach will be taken, with measurements confined to a single area, although reference, where appropriate, will be made to measurements taken by other groups in other areas. The emphasis will be on an understanding of the details of the relationship between the airborne results and the distribution of the radionuclides on the surface and close to the surface of the earth in a particular target area.

If it is possible to calibrate such airborne radiometric surveys successfully using ground based survey techniques this will greatly facilitate the accumulation of meaningful data concerning the distribution of radioactivity here in Southern Africa. A primary aim of this work is to provide an increased understanding of the relationship between *in situ* radiological measurements and airborne surveys and hence to establish a better interpretation of the airborne data.

However, for completeness, since the measurements were carried out for the present investigation other techniques have matured *e.g.* based on the use of Cs (Na) detectors with full spectrum analysis algorithms [Medusa systems 2011]. Such a system has been used in recent environmental research projects [Talha 2009] and references therein.

1.1. Historical perspective

Airborne radiometric surveys were first flown in South Africa in 1966, at around about the same time as the technique was first used elsewhere in the world [Cook 1952]. At this early stage in their use, radiometric surveys were only considered as a qualitative tool to identify anomalies in the total radioactivity in the search for uranium deposits [Tyler 2008]. The uranium was required for the rapidly developing nuclear power programmes around the world and, additionally, was given extra impetus by the on-going Cold War and the associated arms race that existed between the then super powers of the world, the United States of America and the Union of Soviet Socialist Republics. For example, in Australia, much of the uranium prospecting that was originally undertaken was funded by the United Kingdom and the United States Governments by way of ‘finders fees’ paid to prospectors who located viable mineral deposits. In March 1949, for example, the UK Ministry of Supply offered a guaranteed price of between £1 5s 0d and £1 15s 0d depending on the grade [Nininger, Minerals for Atomic Energy 1954].

During the mid-1970’s, airborne radiometric survey techniques underwent a change. With the advent of better instrumentation, including multi-channel analysers, and more readily available computing power, airborne radiometric surveys started to become quantitative. By the 1980’s and 1990’s with the further development of computing power and its miniaturisation, the survey techniques had become further refined [Neufeld and Ruhter May, 1995].

The development of ground based radiometric surveys has followed a similar path, firstly with the development of more sophisticated detector configurations, and secondly with the development of alternative types of detector, including intrinsic germanium detectors. Most recently, techniques have been developed which have allowed the use of these germanium detectors in the field of electromechanical coolers.

1.2. Requirements of the National Nuclear Regulator Act

The establishment of the distribution of the radioactive nuclides on the Earth’s surface is important for understanding the basic geology as well as the effects of

human activities, in mining for example. It is also important from an environmental perspective. In addition to this it has a legal and regulatory dimension. For example, in South Africa it is a requirement of the National Nuclear Regulator Act. No 47, 1999 [Government of South Africa 1999], which replaced the Nuclear Energy Act No.131, 1993 [Government of South Africa 1993], that all land, which might have become contaminated with radioactive material, should be shown to satisfy rigorous criteria before it is released from further regulatory control. In essence, this means that any site under consideration must meet those conditions as listed in paragraph 5.4 of the Government Gazette 28755 published in April of 2006 [Government of South Africa 2007].

These will include the following:

“5.4.1 A site used in the conduct of an authorised action may be released for unrestricted use provided that it is demonstrated that –

5.4.1.1 that radioactive contamination and radioactive materials which can reasonably be attributed to the authorized action have been removed from the site or, in the case of naturally occurring radioactive nuclides, have activity concentrations below the levels for exclusion specified in section 2.1.1.1 (b),(c) and (d); or

5.4.1.2 where the provisions in section 5.4.1.1 cannot reasonably be achieved, remedial measures have been implemented in accordance with section 4.5.1 to achieve optimisation of protection constrained in accordance with section 4.5.2 such that the annual effective dose received by the average member of the critical group for all feasible future situations, arising from the residual radioactive contamination and radioactive materials which can be reasonably be attributed to the regulated action, does not exceed the dose constraint that was applicable during the operations.

5.4.2 In the event that the release of a site in accordance with the conditions in section 5.4.1.2 can only reasonably be achieved by imposing restrictions on the use of the site, the Regulator may, subject to the conditions in section 5.4.1.2 being met, approve the release of that site for restricted use.”

Much of the land affected by these regulations is former mine land, land that has been used by mining concerns for various purposes, and as a result of which has become contaminated with radioactivity (so-called TENORM, see Sub-section 3.4.4). It has now become highly desirable, indeed necessary, to show that this former mine land, prior to release to the public at large, meets the requirements of both the Act and the Regulator, since such land could have significant financial value as 'brown-field' sites for development purposes. For example, the highly profitable casino which has been developed at Gold Reef City, Johannesburg was developed on former mine land. This land was surveyed and shown to meet the release criteria. Other sites of a similar nature include the Crown City development and the City Deep Container Depot both also in Johannesburg.

National and Local Government too have taken considerable interest in such sites as a possible solution for provision of 'low-cost' affordable housing the so called "RDP" housing designed to alleviate the critical housing shortage. Many of the sites of former mine land that are becoming available for redevelopment are within the boundaries of Greater Johannesburg, and are thus within easy reach of places of potential employment. Sites such as Matholesville, built on former mine land adjacent to the Durban Roodeport Deep gold mine in Roodeport, or Slovoville built on land released by Randfontein Estates in the West Rand, are such examples of former mine land that has been rehabilitated and returned to the public domain for redevelopment.

1.3. Structure of the Dissertation

Starting from a brief review of the global situation the dissertation will progress to a review of the national distribution of naturally occurring radionuclides, and finally concentrate on the local distribution of these isotopes at a specific site. Proceeding from there, consideration will be given to how such a distribution of these radioisotopes can be assessed.

This will encompass airborne surveys, ground-based surveys, and the *in situ* measurement of the isotopes. The analysis of soil samples under laboratory conditions will be compared with the *in situ* measurements of different kinds. Then a comparison will be made between radiometric values of the soil samples

that have been taken and the airborne radiometric data looking for some degree of correlation between the two data sets.

Consideration will be given as to the mechanisms of radioactive decay and how the disturbance of the secular equilibrium in the decay chain affects the measurement of the different isotopes. Environmental aspects will also be considered in terms of their influence on the measurement of such isotopes.

The aim then is, by the detailed examination of a small physical area, to look at in detail aspects of *in situ* radiological assessment and how different influences affect our ability to examine a geographical area and come to some meaningful conclusions regarding the distribution of radioactivity within that area.

This dissertation will take the following progression;

- **Chapter One. Introduction**

Looks at the historical perspective of radiometric surveying in South Africa and the rest of the world, and some of the motivating factors that have driven the development of these techniques over the years

- **Chapter Two. Environmental radiation and its detection**

Looks at the modes of radioactive decay of the isotopes usually found naturally occurring in the environment, with special reference to ^{40}K and the decay chains of ^{238}U and ^{232}Th , and in so doing beta decay both positive and negative, electron capture, alpha decay and gamma radiation are discussed. The chapter then moves on to look at the basic concepts of radiation detection and more specifically the types of detecting systems used in radiometric surveys. Consideration will be given to measurement efficiency and sensitivity.

- **Chapter three. Radiation and its global distribution**

Looks at the distribution of radiation from both artificial sources and natural sources. The review then goes into sources of radiation in South Africa looking at the basic geology and from there looks at the distribution of uranium, thorium and Technologically Enhanced Naturally Occurring Radioactive Material

(TENORM). From there consideration is given to those factors, which could cause disequilibrium in the uranium decay chain, including various physical phenomena such as erosion, chemical fractionation and alpha recoil.

- **Chapter four. Airborne radiometric surveying**

Looks at the fundamentals of airborne radiometric surveys and at the types of flight platforms and detection systems that can be used. From there an examination of the basic spectrum analysis process, calibration of the instrumentation, and process of data manipulation. This includes the effects of height, background and the footprint of the survey.

- **Chapter five. Airborne radiometric surveys in South Africa**

Looks at what was done for the survey at the New Machavie survey site and how this differed from the more generic description of airborne surveys carried out elsewhere.

This chapter also looks at the author's work on the development of a terrestrial dose rate map of South Africa and how it was then used to identify a suitable survey site to a more in-depth comparison of airborne and ground based radiometric survey techniques.

Once the survey area had been selected and the reasons for its selection discussed, the maps of the survey area, developed by the author are presented. These are maps that show total count, uranium window, thorium window and potassium window results

- **Chapter six. Ground survey results, comparison with airborne survey and discussion**

Briefly looks at what is included in a generic ground based survey and then goes onto report what was done at the New Machavie survey site. Reviews the results of the soil samples that were taken during the project and compares them to aerial

measurements taken in the same area, and looks for possible correlations between the two sets of measurements.

Chapter seven. Discussion and conclusions

Discusses what has been achieved in the research work and considers some of the problems encountered during its execution and then considers possible ways forward.

CHAPTER 2

ENVIRONMENTAL RADIATION AND ITS DETECTION

2. Introduction

Radiometric surveys make use of the fact that radioisotopes generally decay by emitting α and β particles and associated gamma rays that have energies unique to a particular isotope. Use is made of this property to then identify the radioisotopes that are present in a sample/environment. By way of example, Fig. 2.1 shows a typical spectrum that might be collected when a soil sample is counted in a high-resolution gamma-ray counting system. The peaks at specific energies are used to identify the isotope, and if the correct calibration has been carried out, a measurement of the activity concentrations of each of the isotopes present can then be made. Nowadays, all of this is done using commercially available computer software.

The general radiometric survey principle is stated above, however, when a closer look is taken at what actually happens matters become significantly more complex. Many of the issues that need to be considered will be covered in this chapter. These include the following:

- The distribution of the radioactive isotopes in the larger environment and
- The type of instrumentation used to take such measurements.

The types of radioisotopes that are going to be measured are naturally occurring radioisotopes, technologically enhanced naturally occurring radioisotopes, cosmogenic radioisotopes, or man-made isotopes. However, in each case the levels, type, and distribution all have an impact of the assessment of their distribution.

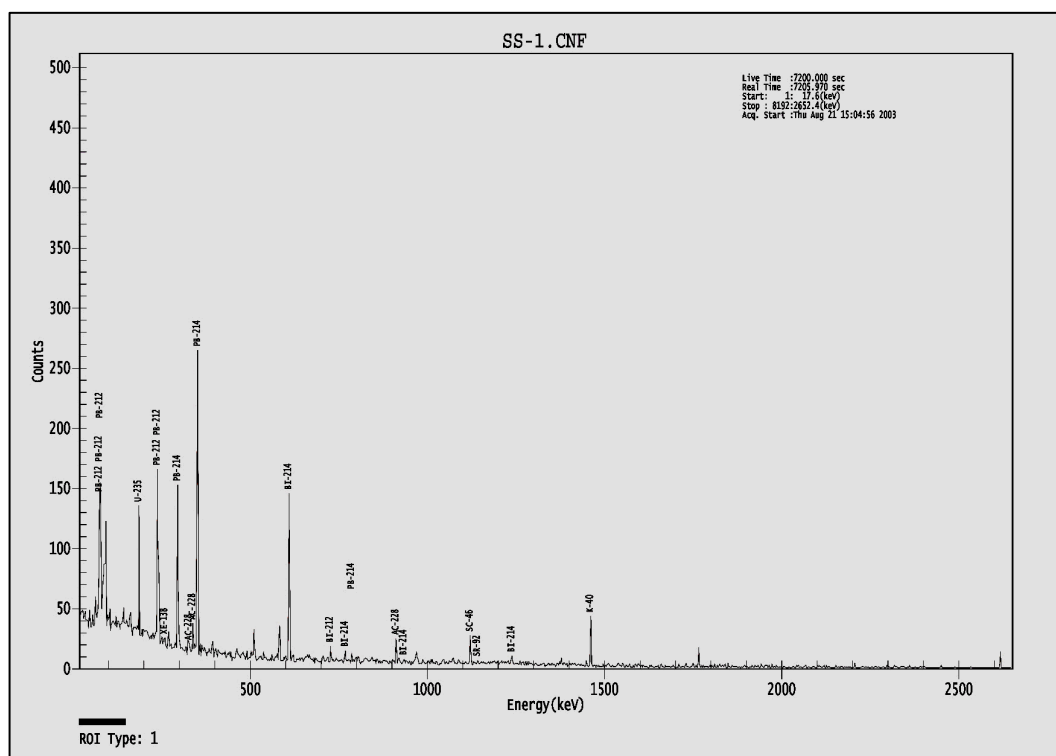


Figure 2-1 A typical gamma-ray spectrum of a soil sample taken (Soil sample 1 – SS1, see Chapter 6) measured using a germanium semiconductor detector. The Applied and Environmental Research Group of the Schonland Research Institute carried out the analysis.

The principle radioisotopes that are examined in this dissertation are those found in the decay scheme of ^{40}K and the decay chains of ^{232}Th , ^{238}U and ^{235}U being the principal decay chains that are used in the measurement of radioactivity by radiometric survey.

The next sections within this chapter will be devoted to coming to an understanding of how a qualitative and quantitative measurement of each of the isotopes is made. Firstly there will be an overview of how these different chains actually decay. Following which some consideration will also be given as to how man-made and technologically enhanced naturally occurring radioactive material is distributed. As can be seen from a later section (Section 3.2) the distribution of these naturally occurring radioisotopes is world wide, though the distribution varies by orders of magnitude depending upon geology, geographic location and a number of other matters addressed later in the chapter.

2.1. Decay Modes of ^{40}K

The only naturally occurring radioactive isotope of potassium is ^{40}K with a half-life of $T_{1/2} = 1.248 \times 10^9$ years being comparable to the current best estimates of the age of the Earth [Dalrymple 2001]. Natural potassium contains 0.01% ^{40}K , which decays in one of the following ways: β^- -decay (89.3%) with the maximum energy of the beta particle being 1.31 MeV, or by electron capture (10.7%) with an associated gamma ray emission of 1.46 MeV or β^+ -decay (0.001%) with the maximum energy of the beta particle of 1.022 MeV (see Fig. 2.2).

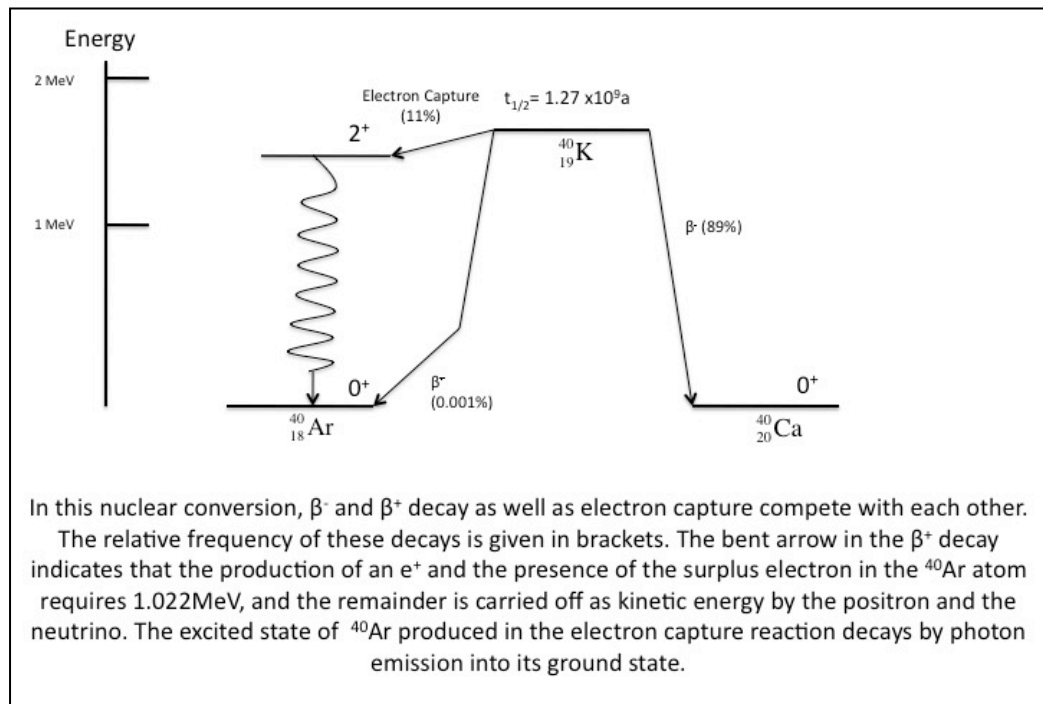


Figure 2-2 Radioactive decay of ^{40}K [Povh 1999] [Ohanian 1987]

2.1.1. Beta minus (β^-) decay of ^{40}K

The underlying process for β^- decay is written as

$$n \rightarrow p + \beta^- + \bar{\nu}_e, \quad \text{Eq. 2.1}$$

where inside a nucleus a neutron spontaneously decays into a proton p , a β^- particle and an anti-electron neutrino $\bar{\nu}_e$.

Anti-electron neutrinos are almost undetectable because of an extremely low probability of interaction with matter. The recoil (daughter) nucleus will appear with very low recoil energy due to its large mass and is not usually detected by conventional detectors. Due to the three-body nature of the decay the β^- -particle or electron has a range of energies.

The β^- -particle will then have a maximum energy or “beta end-point energy” (equivalent to the Q-value) of 1311.07 keV with a continuous range of energies as shown, by way of example, diagrammatically in Fig 2.3.

In the specific instance of ^{40}K the equation is as follows:



2.1.2. Beta plus (β^+) decay of ^{40}K

At a much lower rate ^{40}K has been shown to decay by positron emission [Colgate 1951] [Engelkemeir 1962], which has the general equation



where specifically for ^{40}K the result of the decay is as follows:



Here, inside a nucleus a proton spontaneously decays into a neutron a β^+ -particle (positive electron) and an electron neutrino ν_e . This reaction is energetically impossible for a free proton. However, it is possible for a proton in an unstable nucleus that can supply extra energy for the reaction [Ohanian 1987]. A more general feature of β -decay is that is the electrons or positrons that ejected from the nuclei of a given isotope emerge with a continuous spectrum of energies (see Fig. 2.3). It should be noted that in Fig. 2.3, there is a definite maximum energy, E_{max} , above which no electrons or positrons are observed. It has been shown that the average energy of the electrons $\bar{E}$ is approximately equal to $0.3E_{\text{max}}$ in the case of β^- decay and in the case of β^+ decay $\bar{E} \approx 0.4E_{\text{max}}$ [Ohanian, 1987]

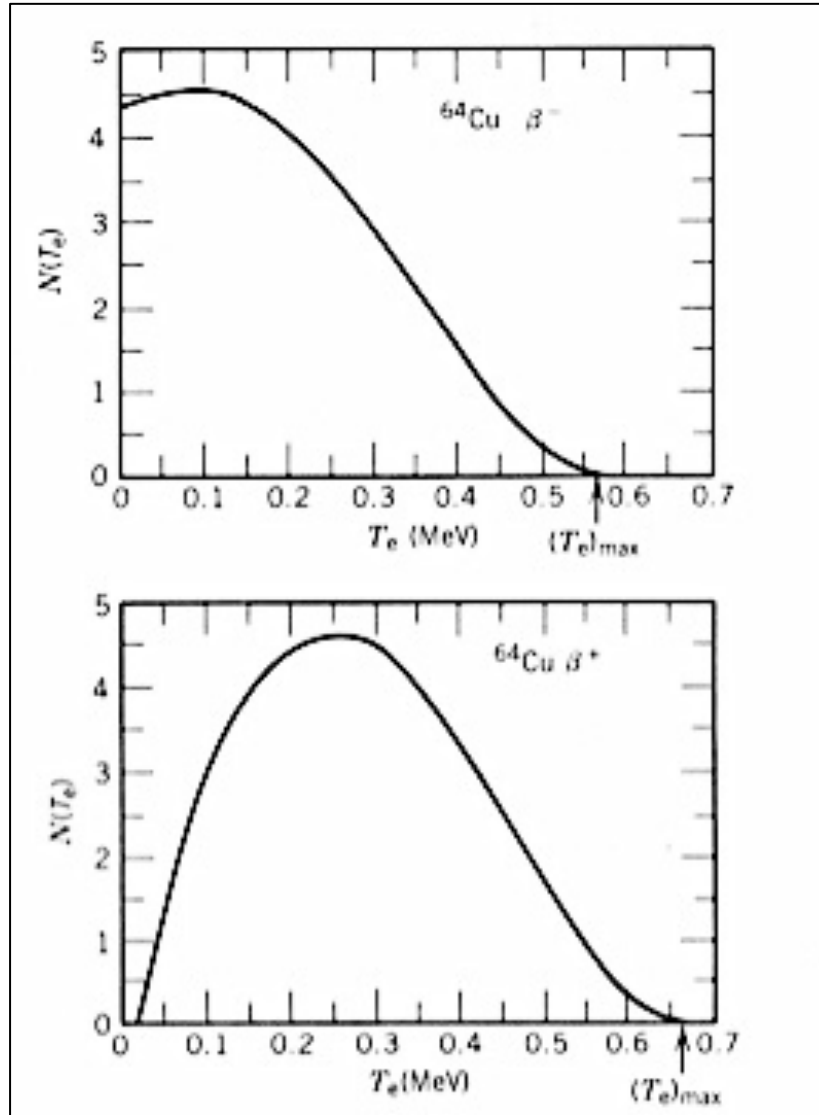


Figure 2-3 Plots of the energies of electrons and positrons emitted from the isotope ^{64}Cu . Note the shift to left in the lower graph arising as a result of Coulomb forces repelling the positron away from the positively charged nucleus. From R.D. Evans, *The Atomic Nucleus* (New York, McGraw-Hill, 1955)

2.1.3. Electron capture in ^{40}K

The general equation for an electron capture reaction is as follows:



where specifically for ^{40}K



Here, a proton in the nucleus, forming a neutron and an electron neutrino, captures one of the K or L-shell electrons. Electron capture occurs because of the wave motion of the orbital electrons can bring them close enough to the unstable nucleus such that the capture of the electron results in the reduction of the number of protons in the nucleus (see Fig. 2.4). As a result, the atomic number and hence the chemical properties are changed but the atomic mass remains the same. In the case of ^{40}K this process leaves the daughter nucleus ^{40}Ar in an excited state, which then de-excites by the emission of a gamma ray with the characteristic energy of 1460.8 keV.

Since K-shell electrons are closest to the nucleus to begin with, most electron capture involves K-capture; however, it is possible for some L-shell electrons to come close enough to be captured (see Fig. 2.5).

In electron capture transformations, Z decreases by 1, N increases by 1 and A the mass number remains the same. However, the capture of an electron from the innermost electron orbitals leaves a vacancy that is immediately filled by electrons from the outer orbitals that are at a higher energy. The process of an electron moving down to a lower energy level or orbital produces characteristic X-rays associated with the daughter nucleus (see Fig. 2.5). On occasion though the X-ray produced does not escape from the atom, but rather interacts with one of the electrons in the outer electron shells, driving this electron out of the electron shell and produces an Auger electron with a characteristic energy associated with it also shown in Fig. 2.5.

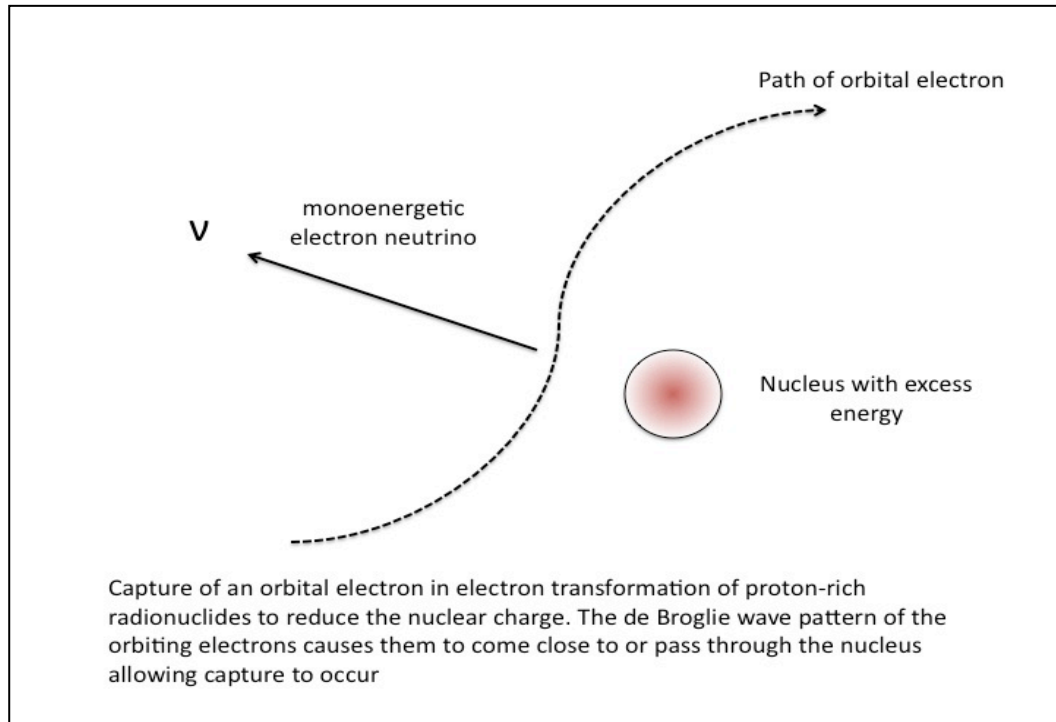


Figure 2-4 Basic process of electron capture [J. Martin 2006 2nd Ed.].

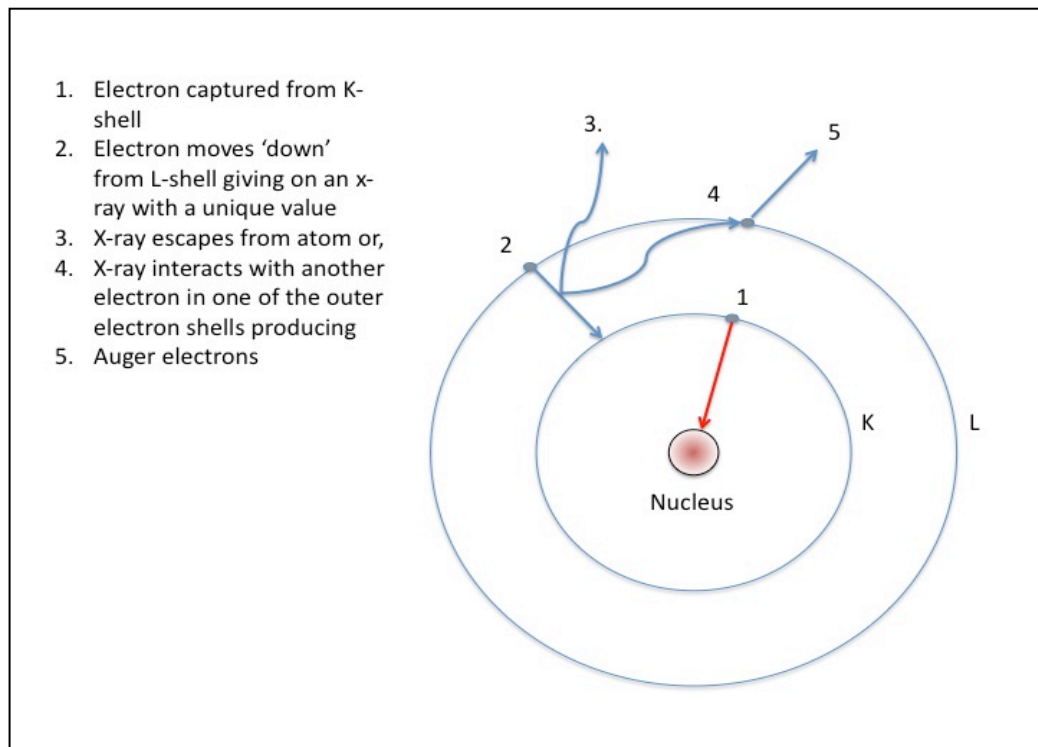


Figure 2-5 Production of characteristic X-rays and Auger electrons [J. Martin 2006, 2nd Ed.].

2.2. Decay modes of thorium and natural uranium

In Figs. 2.6, 2.7 and 2.8 the decay chains of thorium and natural uranium are detailed. From these chains it can be seen that the processes of β^- -decay, α -decay and gamma radiation are all involved before the various radioisotopes reach a stable condition. β^- -decay has already been considered in the previous section when describing the decay of ^{40}K . Attention will now be given to the process of α -decay and the release of the associated gamma radiation.

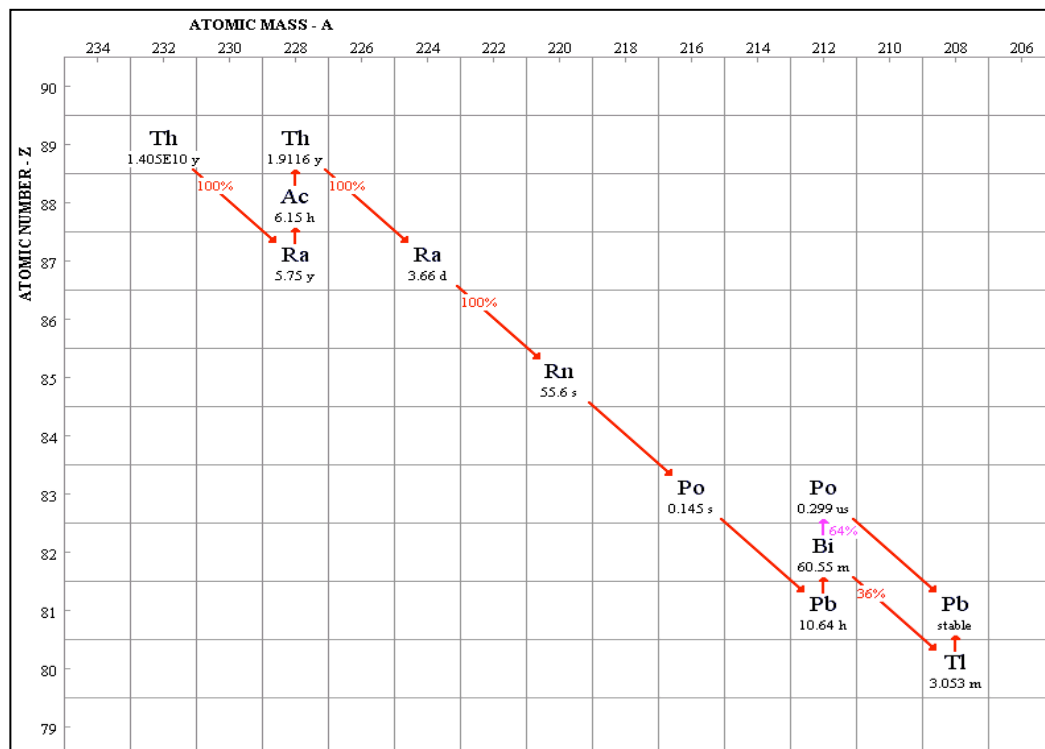


Figure 2-6 ^{232}Th -decay chain (α – decay is shown by the diagonal arrows and the β^- decay by the vertical arrows).

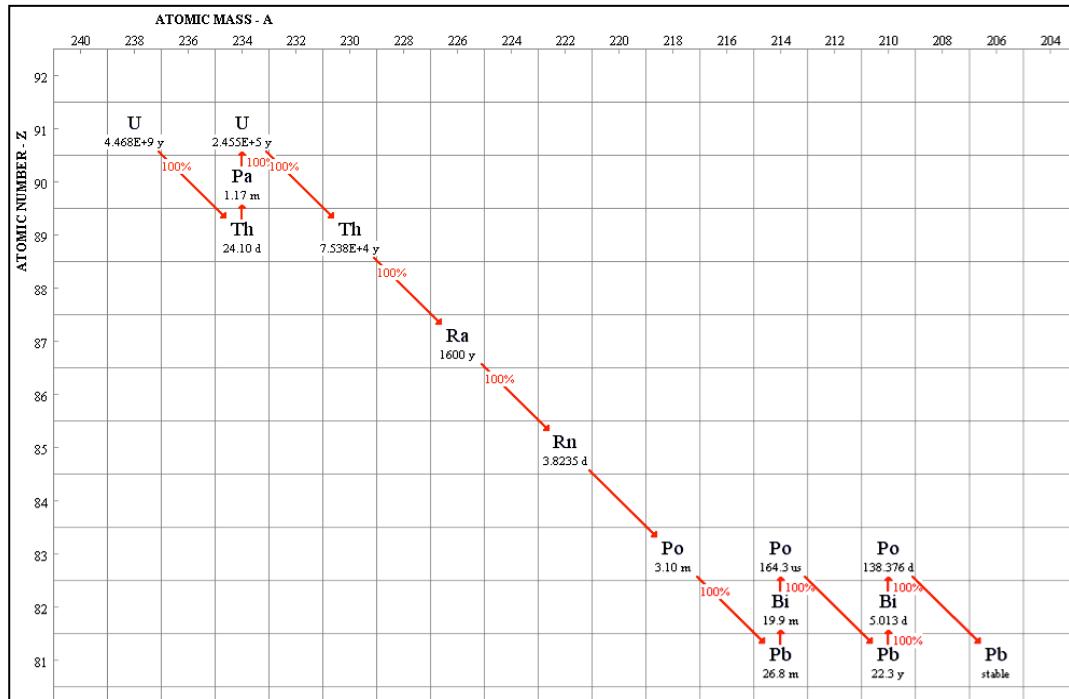


Figure 2-7 ^{238}U Series decay chain (α – decay is shown by the diagonal arrows and the β decay by the vertical arrows).

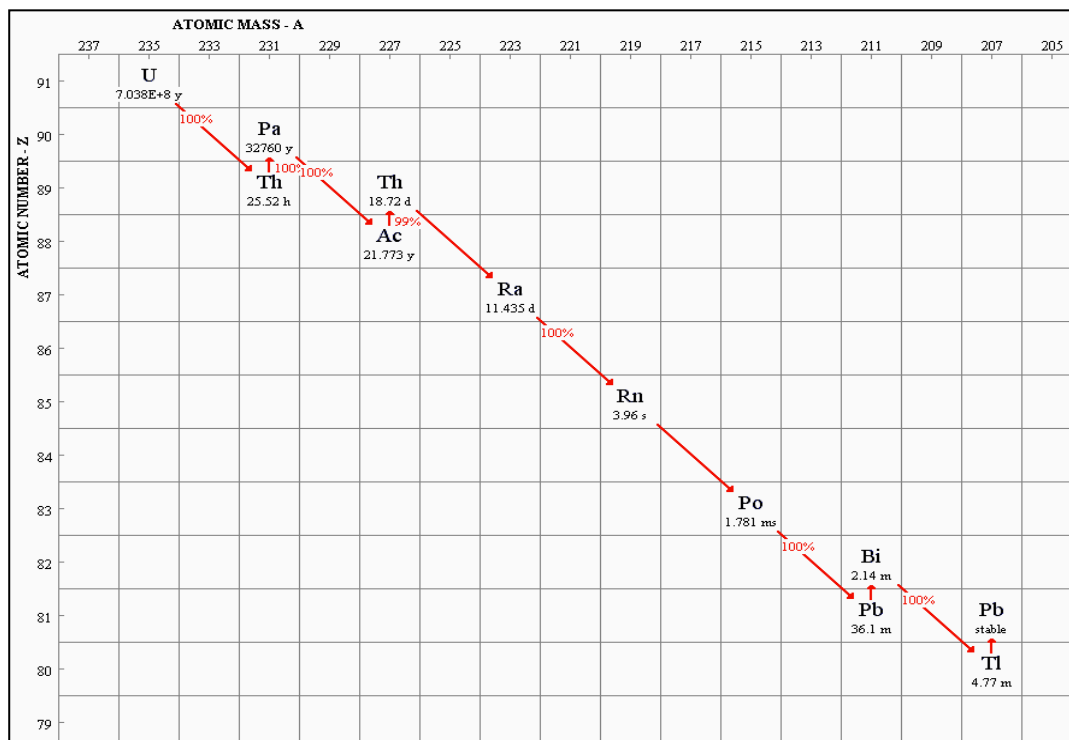


Figure 2-8 ^{235}U decay chain (α – decay is shown by the diagonal arrows and the β decay by the vertical arrows).

2.2.1. α -decay

In order to reach stability the isotopes of thorium and natural uranium radioactively decay to one of the stable isotopes of lead *viz*:

- ^{208}Pb for ^{232}Th (Fig. 2.6)
- ^{206}Pb for ^{238}U (Fig. 2.7)
- ^{207}Pb for ^{235}U (Fig. 2.8).

The most efficient way for these nuclides to reduce both mass and charge is through the emission of several α -particles (helium nuclei), which carry a doubly positive charge $^4\text{He}^{2+}$. This will have the effect of reducing both the proton and neutron numbers by two and the mass number by four. It should be noted that α -particles resulting from a specific nuclear transformation are mono-energetic, and they typically have large kinetic energies, generally above 4 MeV.

Alpha particle transformations cannot be explained by classical physics, [J. Martin 2006, 2nd Ed.] because the α -particle does not have enough energy to penetrate the large potential energy barrier of the heavy parent nucleus. This potential energy barrier can be as much as 30 – 40 MeV, whereas most emitted alpha particles have energies in the range of a few MeV.

It is necessary to consider the problem as a wave mechanical phenomenon. An α -particle confined to a nucleus can be thought of as two protons and two neutrons coming together temporarily to make up the particle [Knoll 2000]. An α -particle of 4 – 5 MeV will have a velocity such that it will bounce back and forth within the potential well of the nucleus approximately 10^3 times per second (see Fig. 2.9). This large number of attacks on the barrier of the nucleus together with the de Broglie wavelength eventually leads to the α -particle “tunnelling through” the barrier. The probability of emission, that is to say the alpha particle “tunnelling” through the potential well, increases with the α -particle energy i.e. going from K to K' .

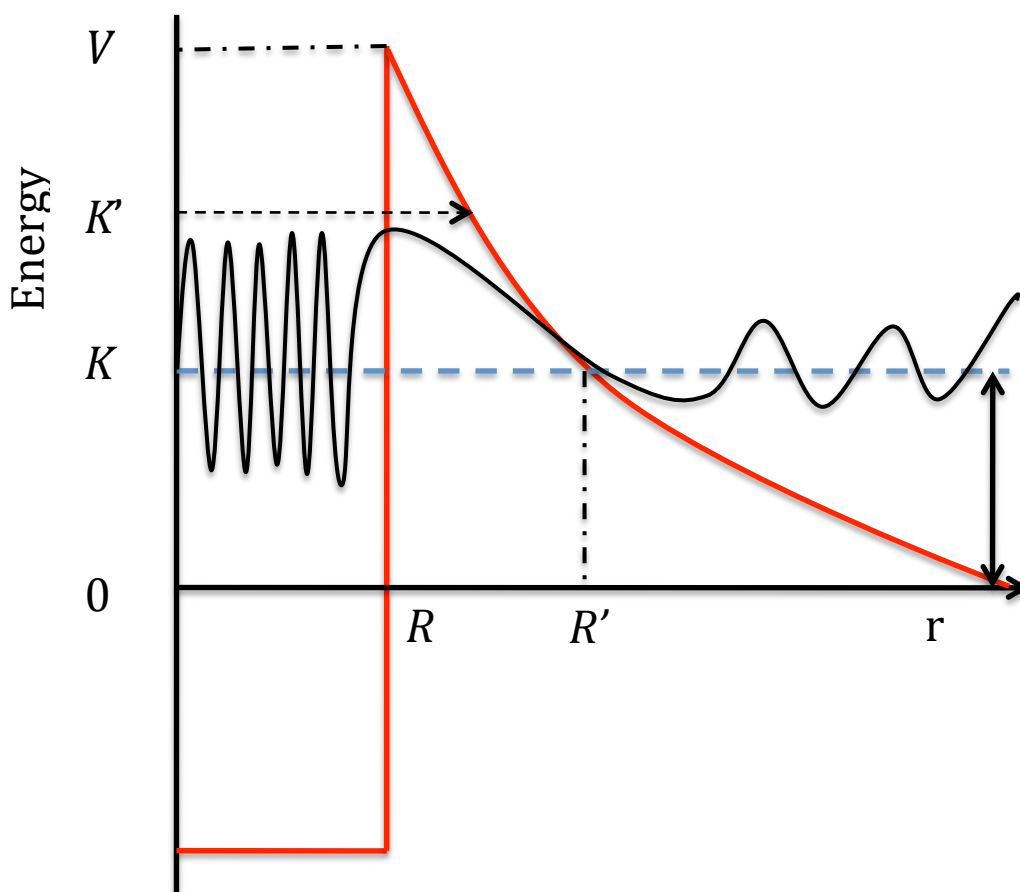


Figure 2-9 Barrier penetration of a heavy nucleus by an alpha particle where V denotes the barrier height and K the kinetic energy of the liberated α -particle.

Plotting the half-lives of various α -emitters against the maximum energy of the emitted alpha-particle results in the Geiger-Nuttall relationship as shown in Fig. 2.10, where it can be seen that the higher the α -particle energy, the shorter the half-life, indicating that the probability of tunnelling is higher. This can be explained by the “thickness” of the potential energy barrier, as indicated by the line K' in Fig. 2.9. The higher energy α -particles interact with the thinner part of the barrier and as a result have a higher probability of tunnelling through whereas the lower energy ones interact with the thicker part of the potential barrier and are less likely to penetrate it. It should be noted though that the daughter nucleus might be left in an excited state as opposed to the ground state thus reducing the maximum energy of the alpha particle emitted.

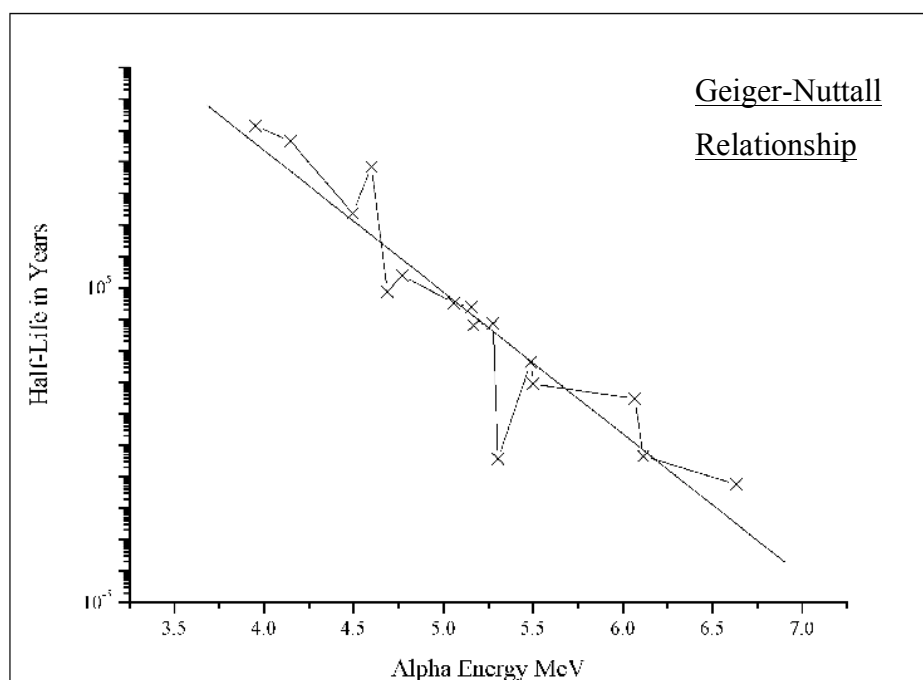


Figure 2-10 Geiger-Nuttall relationship obtained by plotting the half-lives of common alpha-emitting radioisotopes given in Table 2.1 against the maximum energy of the alpha-particle emitted, (plot by author).

Table 2-1 Half-lives of common alpha-emitting radioisotopes

Source	Max. α-Particle Kinetic Energy (MeV)	Half-life (y)
^{232}Th	3.953	1.40×10^{10}
^{238}U	4.149	4.50×10^9
^{236}U	4.494	2.40×10^7
^{235}U	4.598	7.10×10^8
^{230}Th	4.6875	7.70×10^4
^{234}U	4.77	2.50×10^5
^{231}Pa	5.059	3.20×10^4
^{239}Pu	5.1554	2.40×10^4
^{240}Pu	5.1683	6.50×10^3
^{243}Am	5.2754	7.40×10^3
^{210}Po	5.30451	0.37
^{241}Am	5.48574	433
^{238}Pu	5.49921	88
^{244}Cm	6.067	30
^{242}Cm	6.11292	0.446
^{253}Es	6.63273	5.61×10^{-2}

2.3. Gamma Radiation

Along with α and β^- -decays there is an associated emission of gamma photons from the decay series shown in Figs. 2.6, 2.7 and 2.8. This is because radioactive transformation often results in the daughter nucleus being left in an excited state in which the protons and neutrons in the shells of the nucleus are not bound in the tightest state possible [J. Martin 2006, 2nd Ed.]. This ‘excess’ energy will be emitted as electromagnetic radiation when the nuclear components rearrange themselves to the lowest energy state. The shell model for the nucleus suggests discrete energy states for neutrons and protons; it is this difference between energy states that is emitted as a gamma photon when rearrangement takes place.

2.3.1. Gamma ray interactions used in radiation measurements

There are a number of possible ways that gamma rays can interact with matter, but three are of special interest when it comes to detecting gamma radiation, to determine a quantitative measurement of the isotopes present in a sample. These interactions are as follows:

- Photoelectric absorption
- Compton scattering
- Pair production.

All of these processes lead to the partial or complete transfer of the gamma ray photon energy to electron energy [Knoll 2000]. Each of these is dealt with in turn in the following sections.

2.3.1.1. Photoelectric absorption

In the photoelectric absorption process, a photon undergoes an interaction with an absorber atom in which the photon completely disappears. In its place, an energetic photoelectron is produced from one of the electron shells of the absorber atom with a kinetic energy given by the incident photon energy ($h\nu$) minus the binding energy of the electron in its original shell (E_b). This process is shown in Fig. 2.11. For typical gamma-ray energies, the photoelectron is likely to emerge

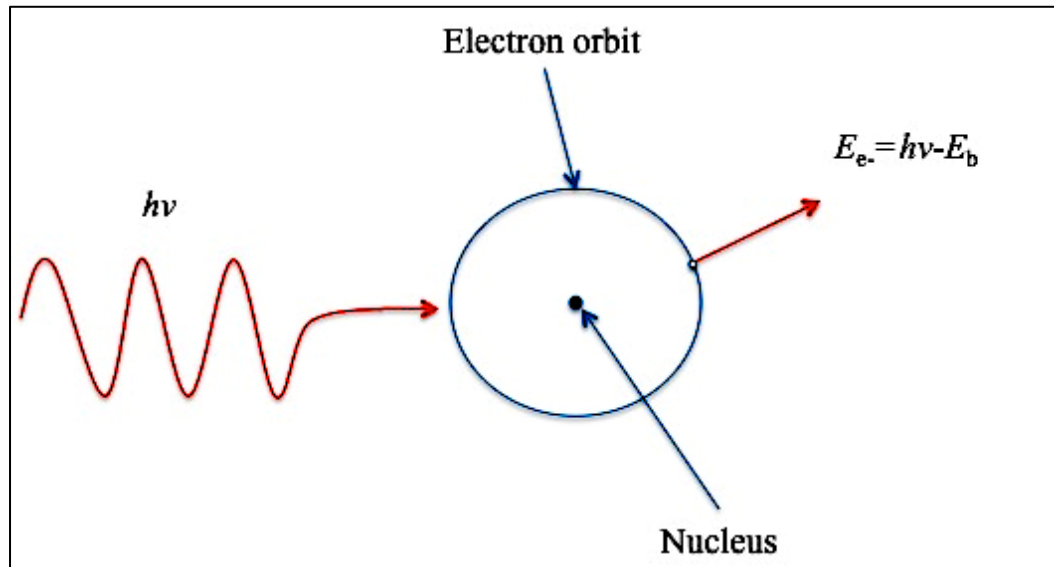


Figure 2-11 Process of photoelectric absorption.

from the K shell [Ohanian 1987], for which binding energies range from a few keV for low-Z materials to tens of keV for materials of higher atomic number. The vacancy that is created by the resultant loss of the photoelectron from one of the electron orbitals is quickly filled by electron rearrangement. During this process of rearrangement of the electrons, so that they are in the lowest energy levels possible, binding energy is liberated either in the form of characteristic X-rays or in some instances as Auger electrons (see Fig. 2.5). Since Auger electrons have very low energies they will only travel a very short distance before they become bound to a free orbital. X-rays may travel several millimetres before they are absorbed by photoelectric interactions with less tightly bound electron shells of the absorber atoms.

The principle result of photoelectric absorption is the liberation of the photoelectron, which carries away most of the gamma ray energy, together with one or more low-energy electrons corresponding to the absorption of the original binding energy of the photoelectron. If this process can then take place completely within a detector set-up, then the sum of the kinetic energies measured must equal the original energy of the gamma ray photon.

2.3.1.2. Compton scattering

The result of a Compton scattering interaction is the creation of a recoil electron and a scattered gamma ray photon, with the division of the energy between the two dependent on the scattering angle.

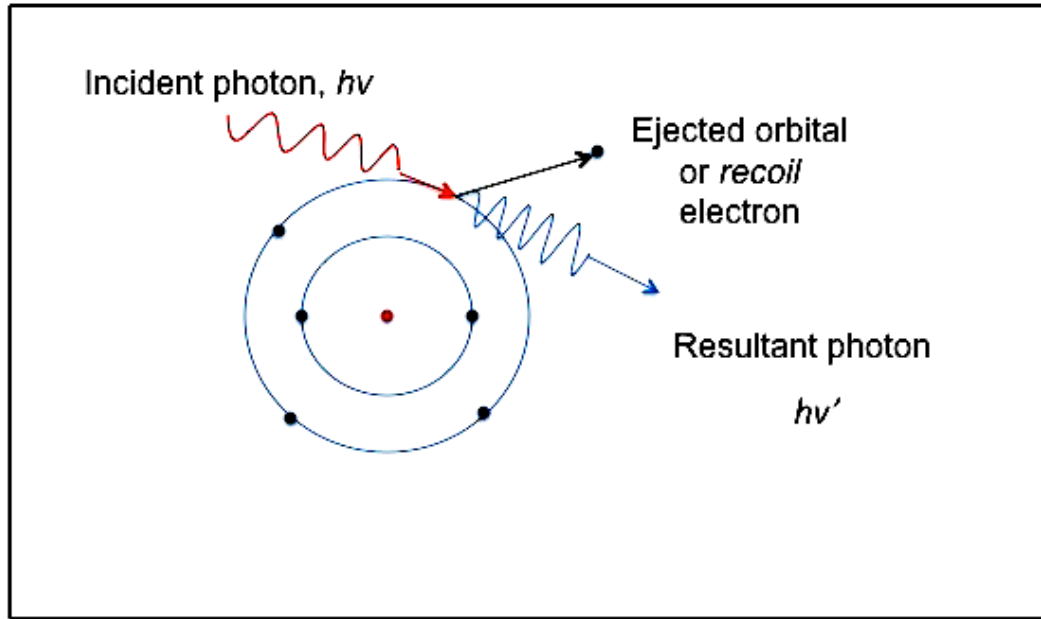


Figure 2-12 Compton scattering

The energy of the scattered photons ($h\nu'$) can be determined using the following equation:

$$h\nu' = \frac{h\nu}{1 + \frac{h\nu}{m_0c^2}} (1 - \cos \theta), \quad \text{Eq. 2.7}$$

where, $h\nu$ is the energy of the incident photon, m_0c^2 is the rest-mass energy of the electron (0.511 MeV) and θ is the angle through which the incident photon is scattered. Plotting Eq. 2.7 yields the graph shown in Fig. 2.13.

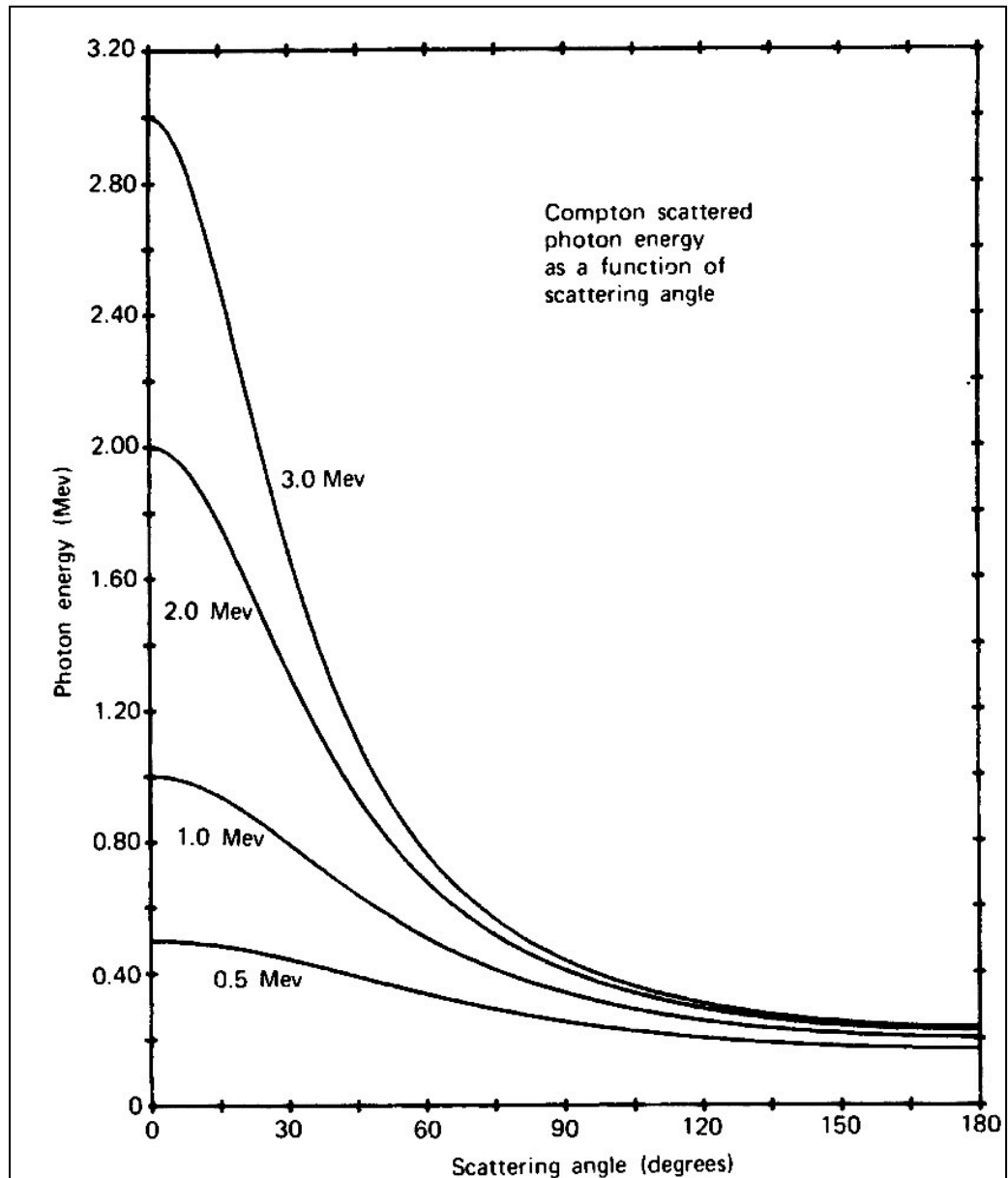


Figure 2-13 Variation of scattered gamma-ray energy with scattering angle [Knoll 2000].

The scattered photon emerges from the collision in a new direction and with a reduced energy and an increased wavelength. The change in the wavelength is commonly referred to as the Compton Shift.

The probability of Compton scattering per atom of the absorber depends on the number of electrons available as scattering targets and therefore increases linearly

with Z . The general dependence on gamma ray energy is shown in Fig. 2.14 and for the case of sodium iodide in Fig. 2.15.

2.3.1.3. Pair Production

This mode of gamma-ray interaction only starts to become important when dealing with high-energy gamma photons (see Fig. 2.14), generally in the region of cosmic gamma photons but it is also important in the detection of high-energy gamma rays from radioactive sources such as ^{208}Tl or ^{226}Ra .

In this interaction, the gamma-ray photon disappears and is replaced by an electron-positron pair. Any additional energy carried in by the incident photon above the 1.02 MeV required (twice the rest mass of the electron) to make the electron-positron pair is shared by this pair as kinetic energy. The positron will then annihilate after slowing down in an absorbing medium producing two annihilation photons. These have the particular energy of 0.511 MeV (rest mass of the electron). This is the physical process that allows the positron emission tomography (PET) scanner to work in the medical world.

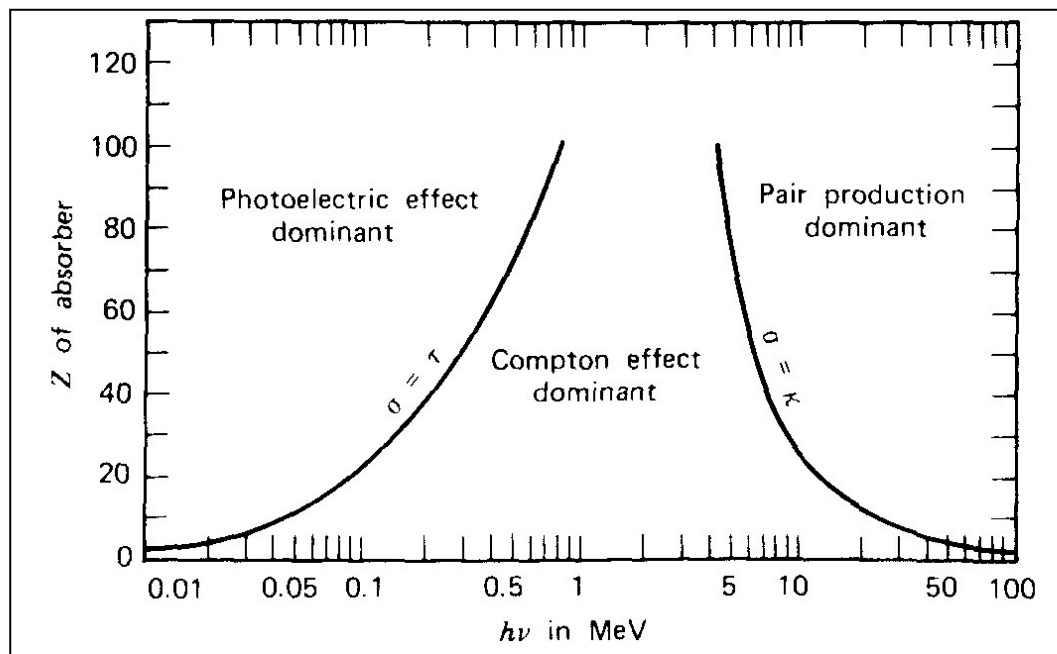


Figure 2-14 the relative importance of the three major types of gamma ray interaction. The lines show the values of Z and $h\nu$ for which the two neighbouring effects are just equal [Evans 1955].

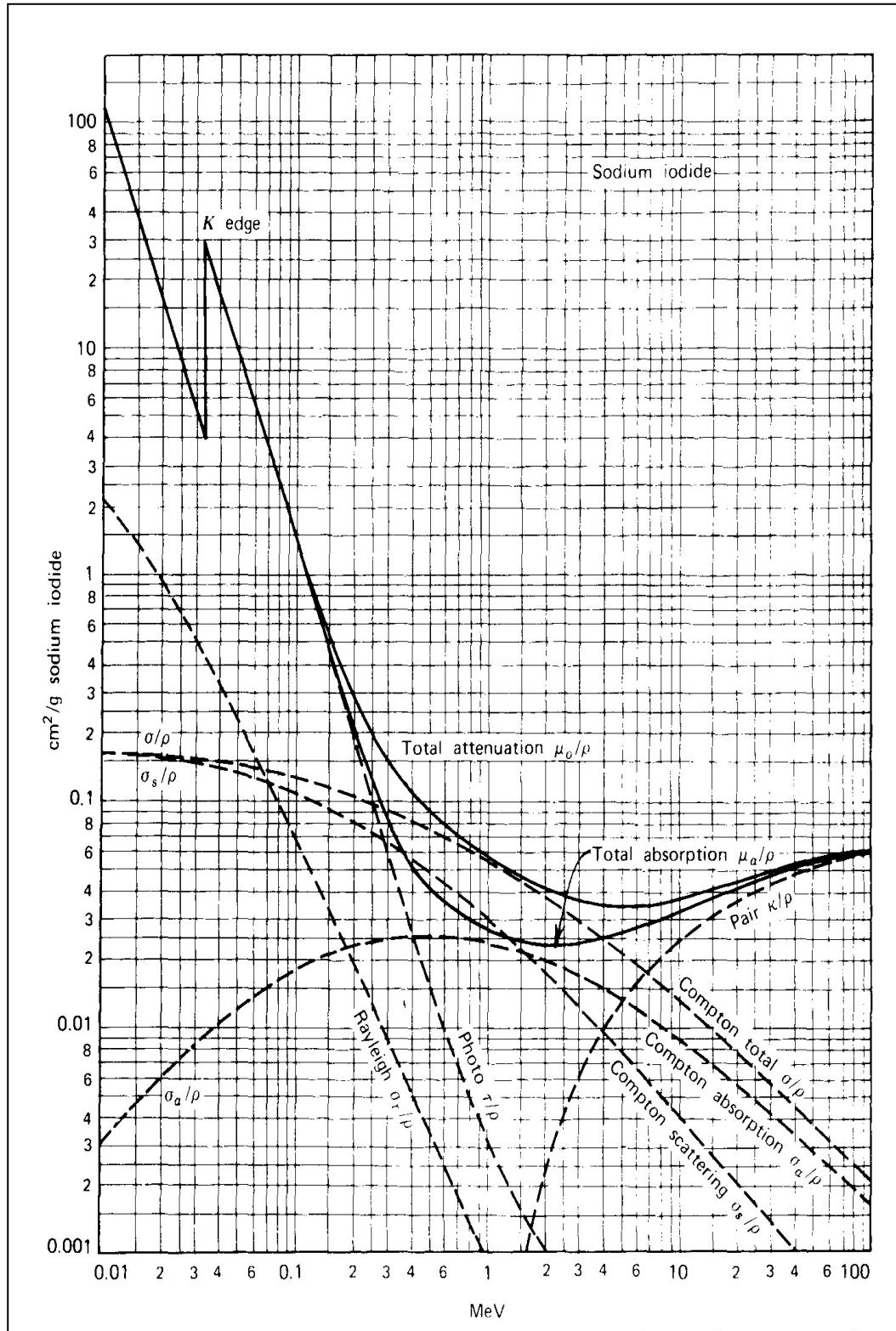


Figure 2-15 Energy dependence of the various gamma ray interaction processes in sodium iodide [Evans 1955] (including a number of types of interaction not relevant to this dissertation).

2.3.2. Thorium decay chain

Thorium and uranium, unlike ^{40}K , decay through a series of intermediate isotopes before they both reach a stable isotope of lead. The detailed decay chain of ^{232}Th is shown in Fig. 2.6, with the decay products that are produced, and the types of radiation that are given off. A detailed list of all the decay products, energies and types of radiation that are produced are detailed in a number of standard texts but one of the most often used is ICRP 38 [ICRP 1983].

2.4. Detection of ionising radiation

In this section the most commonly used detection techniques will be considered that involves the use of a sodium iodide, thallium activated detector. Where appropriate, however, reference will be made to alternative scintillating materials and techniques. A more complete description of how inorganic scintillation detectors work can be found in Knoll [Knoll 2000]. Particular emphasis will be spent on looking at how the thallium activated sodium-iodide scintillator-based detector functions. This detector is almost exclusively used in airborne radiometric surveys.

2.5. Types of detector used in radiometric surveys

2.5.1. Sodium iodide detectors

The basic principle of operation of a thallium-doped sodium iodide detector relies on the scintillation produced (ultra violet (UV) photons) when gamma rays interact with the sodium-iodide crystal (see Fig. 2.16). These UV photons then interact with the second part of the detector, the photocathode, which has the unique property of emitting electrons from its surface when struck by the UV photons produced in the sodium iodide crystal. A positive high-voltage then draws these electrons to a series of curved plates called dynodes arranged in a photomultiplier tube. As an electron strikes a dynode it results in a number of additional electrons being released from that dynode which are then accelerated to towards the next dynode in the photomultiplier tube.

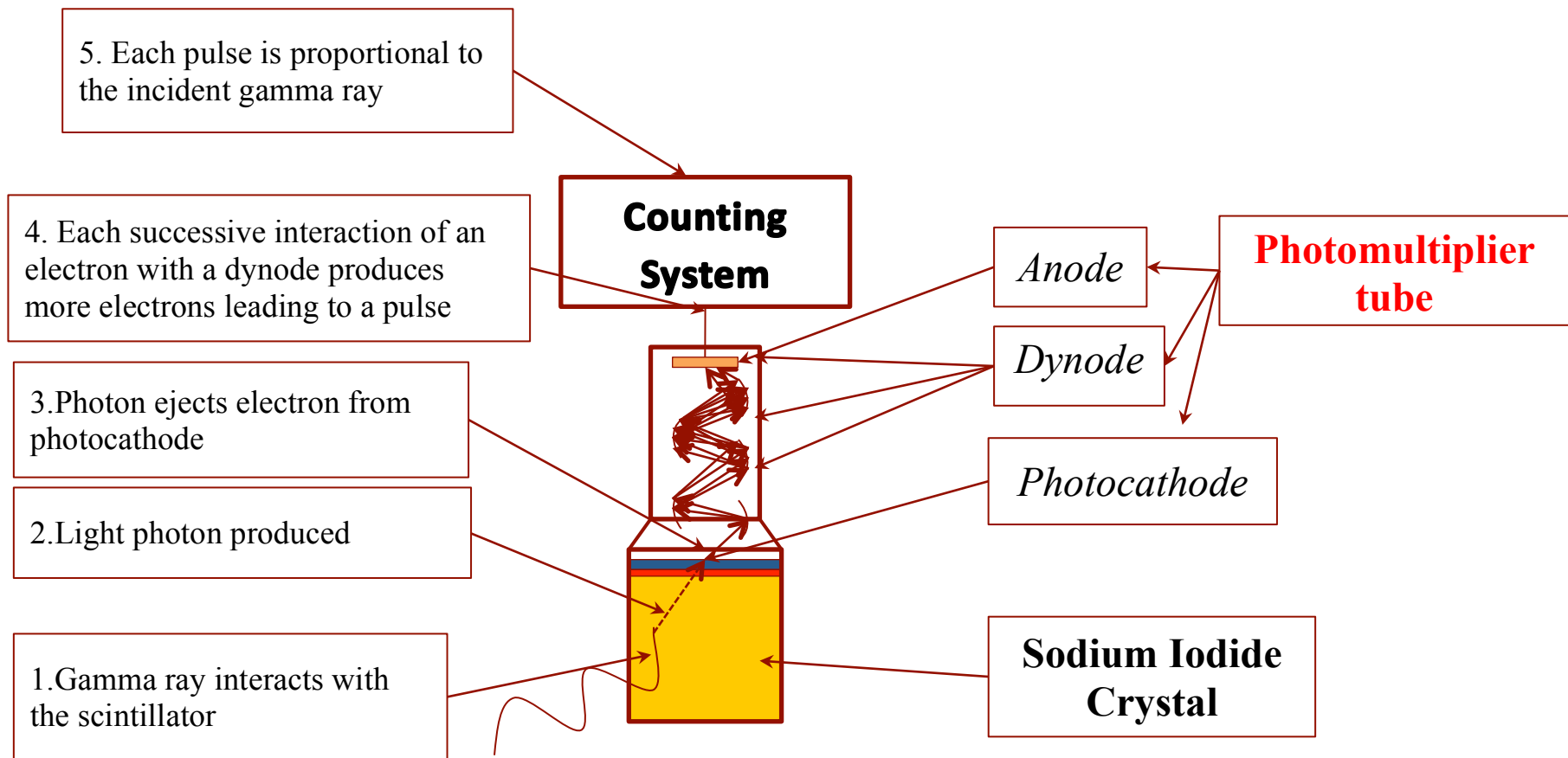


Figure 2-16 Basic operation of a sodium iodide scintillation detector

In this way an initial electron released as a result of a UV photon interaction with the photocathode leads to the production of a measurable current pulse at the anode at the end of the photomultiplier. The current pulse produced is proportional to the energy deposited in the crystal by the initial gamma ray. This is a rather simplified version of what happens; Section 3 in Chapter 10 of Knoll [Knoll 2000] goes into greater detail about the predicted and observed response function of these types of detectors

The one issue that should be borne in mind when looking at experimentally acquired spectra is that not all the gamma photons that interact with the scintillation media, deposit all of their energy in the scintillation crystal (see Fig. 2.17 and 2.18) which adds further complexity to the recorded spectra. Chapter 10 of Knoll [Knoll 2000] goes into the many details that must be taken into account to measure gamma-ray intensity.

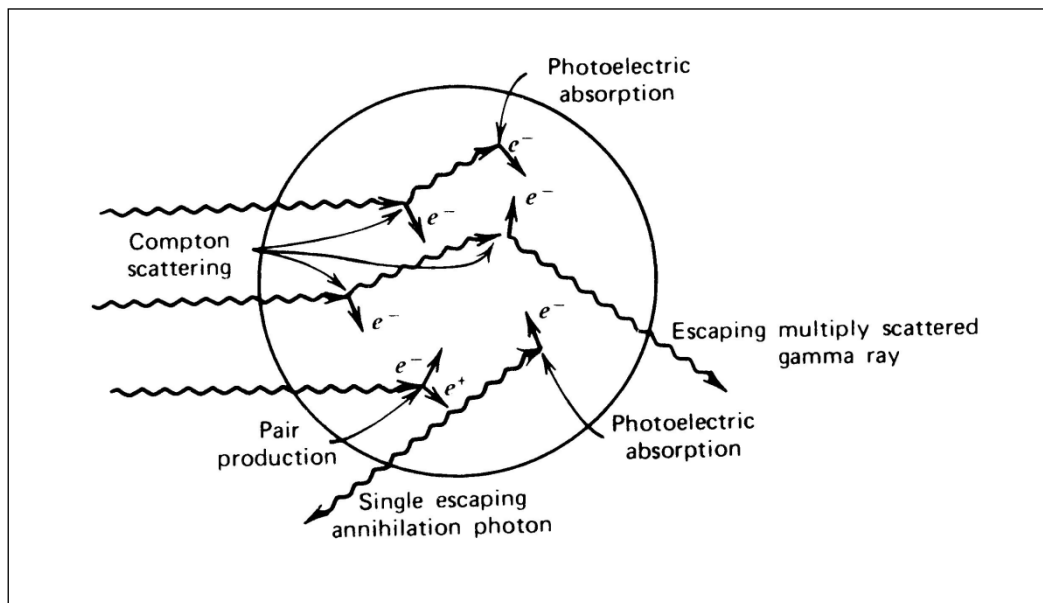


Figure 2-17 "Escape" of annihilation photons ([Knoll 2000] Fig. 10.4).

The broadening of the peaks as can be seen in Fig. 2.19 is due to the statistical nature of the detection process. The Full Width Half Maximum (FWHM) value shown is an indication of this. Rather than a very narrow peak within a spectrum a broad peak is produced.

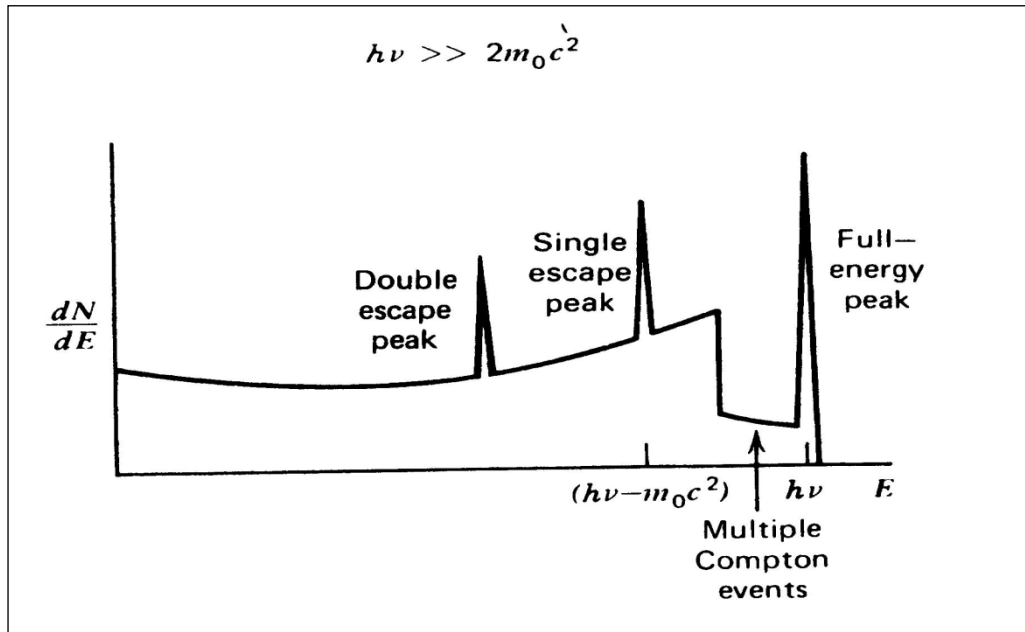


Figure 2-18 Spectrum that shows the escape of both a single and double annihilation photons ([Knoll 2000] Fig.10.4).

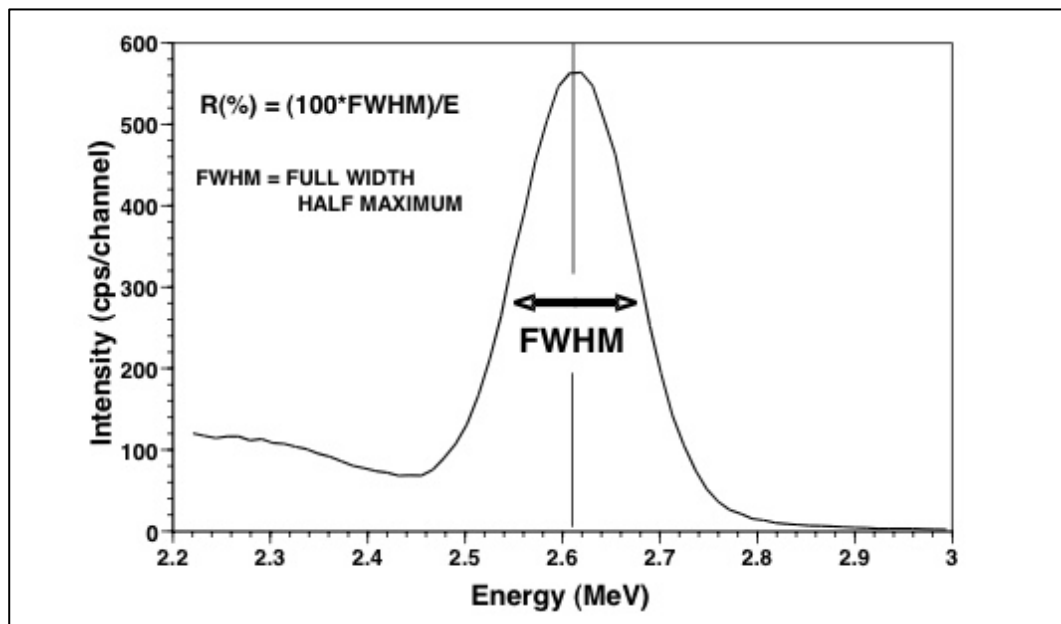


Figure 2-19 Diagram illustrating the energy resolution of a gamma ray spectrometer based on a NaI (Tl) detector [IAEA 1991]

2.5.2. Germanium detectors

These make use of the principle of the migration of electrons and ‘holes’ in a semiconductor material, which results in the measurement of an electronic pulse in a counting system that is proportional to the energy deposited by a gamma

photon that strikes the detection volume. The particular advantage over sodium-iodide based detection systems is the energy resolution of the germanium-based systems. Sodium iodide based systems at 667 keV have at least a 6% resolution (FWHM) associated with the detection system, this represents the very best that the system can do. A disadvantage of germanium systems is the need to cool them to 70 K, so that the thermal energy electrons in the detector volume does not drown out the signal produced by the electrons which have been elevated to the conduction band by the impinging ionising radiation. The details of the operation of these semiconductor-based detectors are covered in Chapters 11 and 12 of Knoll [Knoll 2000].

Germanium-based detectors are available only in much smaller sizes than either NaI or BGO type detectors, discussed in the next section, and therefore have a significantly lower efficiency or sensitivity. This makes them less suitable for airborne surveys.

2.5.3. Bismuth germanate (BGO) crystals

These are among a number of inorganic scintillators that are used as an alternative to thallium activated sodium iodide (NaI (Tl)). Bismuth germanate has a high density and a large atomic number, both of which are desirable characteristics of a scintillation based detector (photoelectric absorption varies approximately with $Z^{4.5}$). The crystals of BGO thus have the largest probability per unit volume of any commercially available scintillation material for photoelectric absorption. The chemical and physical properties of the crystals also make it easy to handle particularly in more rugged conditions. However, a disadvantage is that the light yield of BGO crystals is only between 10 – 20% of that of NaI (Tl) and the crystals also have a high refractive index making the collection of the photoelectrons somewhat more problematic. The timing resolution is also a factor of 2 worse than that of NaI (Tl). These limitations whilst not entirely negating the use of BGO as a detector material do rather restrict its use to some specialised conditions and purposes.

2.6. Measurement sensitivity in detection

As in many other areas of science, it is often desirable to know the actual quantities or concentrations of a particular compound or element that is present within a sample. Such a sample can be one taken for laboratory analysis; often though it is desirable to know what the *in situ* levels of the compound or element are in the sample. The property of radioactive decay that different radioisotopes decay with energies unique to them makes for a convenient method of quantitative and qualitative analysis. Analyses can be done, therefore, in both the laboratory and in the field. The measurement process, however, is not quite as straight forward as is suggested above; because of the stochastic nature of radioactive decay, the electronics used in the systems and the detectors used to measure this decay certain concepts first have to be understood. These include the effect of background radiation on the reported measurement value, the role of the detector in the detection of any radioactivity, the influence of the environment in the detection of radioactivity and the effect that the detection electronics has on any reported measurement values. Before the meaning of any measurement can be understood, it is important examine the influence of these various factors.

2.6.1. Background Radiation

To a greater or lesser extent, background radiation is always present in the environment; even in laboratory conditions it is impossible to reduce levels of background radiation to zero. There are various techniques that have been developed over the years to reduce the effects of background radiation; such a technique is one that relies on anti-coincidence counting using paired detectors, one inside the counting apparatus and one on the outside, so that when they detect a gamma photon simultaneously that particular count is discarded in the final count of activity in the sample ([Knoll 2000], 771). This technique is illustrated in Fig. 2.20.

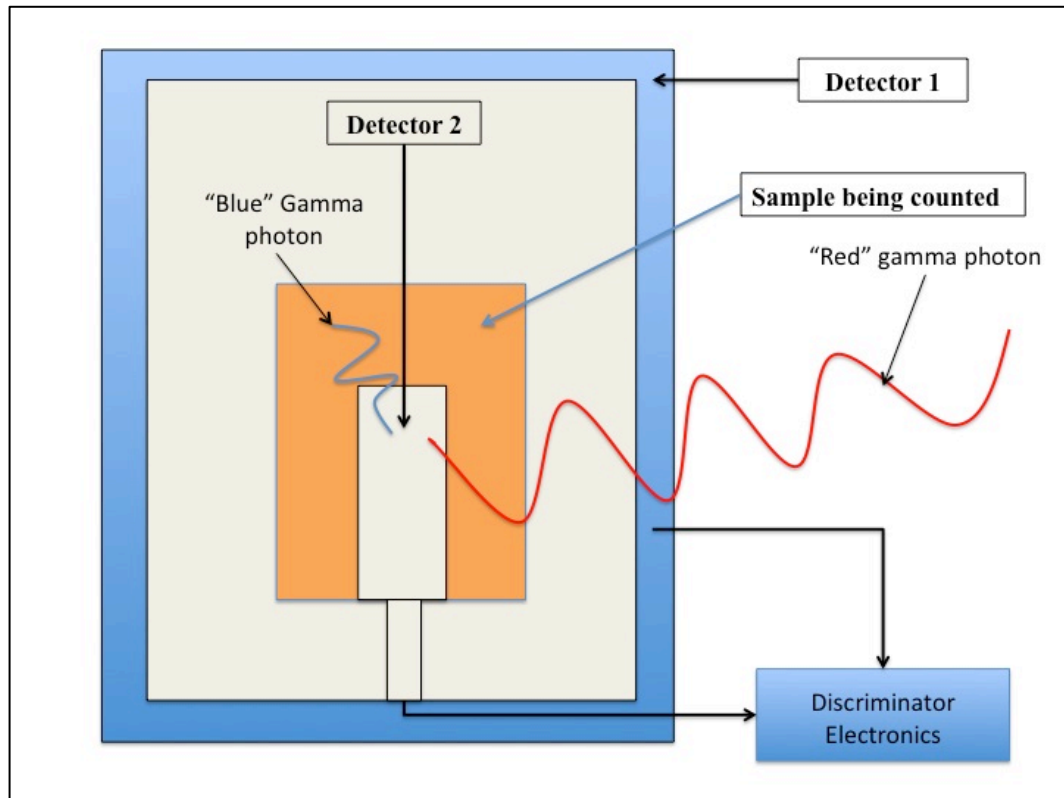


Figure 2-20 Anti-coincidence detection system.

The red photon (Fig. 2.20), a gamma photon from outside the sample, could be recorded by both detectors and is therefore discarded by the discriminator electronics, unlike the blue photon (Fig.2.20) which emanates from within the sample under consideration and is only detected by the second detector, and is counted.

Background radiation is always going to make up a component of a final reported system measurement. This component becomes more and more significant as the quantities of the radionuclide in the sample become smaller and smaller, since proportionately the contribution made by the background part of the measurement becomes larger.

2.6.2. A statistical technique for dealing with the prevailing background count in a sample

After a measurement has been made with a detection system it is necessary to determine whether or not the result can be accurately differentiated from the

background response of the instrument. To understand how this is done it is necessary to introduce the following statistical concepts.

2.6.2.1. Critical Level (L_C)

The critical level (L_C) is the level, in counts, at which there is a statistical probability of incorrectly identifying a measurement system background value as “greater than background”.

The measurement system background is that value that consists of a contribution from background radiation and in addition noise from the amplification circuits of the detection system. If the system background is constant on average a Poisson distribution gives the probability distribution. By way of example see Fig. 2.21 and Section 2.6.3.

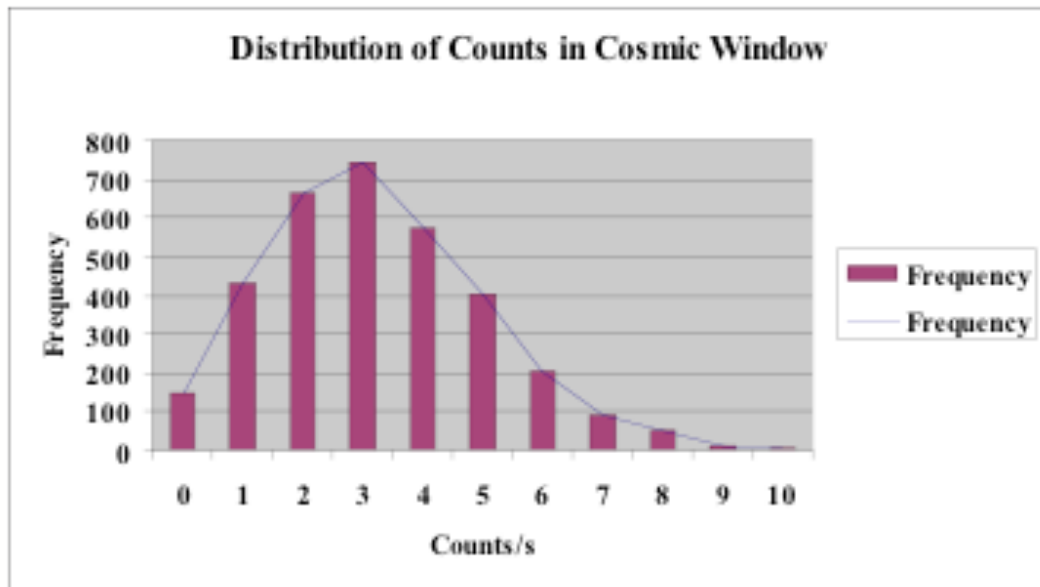


Figure 2-21 Poisson distribution of the Cosmic Radiation as measured at the New Machavie Gold Mine. Plot by author.

2.6.2.2. Detection Limit (L_D)

The detection limit (L_D) is an *a priori* estimate of the detection capabilities of the measurement system for a particular nuclide, for a particular counting time.

2.6.2.3. Minimum Detectable Concentration (MDC)

The Minimum Detectable Concentration (MDC) is the detection limit L_D multiplied by an appropriate conversion factor and is expressed as the smallest concentration of the particular nuclide that is detectable under the given conditions.

2.6.3. Brief description of the derivation of L_C , L_D and MDC

A brief summary is presented of the often-quoted paper by Currie [Currie 1968] and how the derived formulae should be applied. The two key parameters in a detector system that has a measurement system background response greater than zero are:

- L_C the net response level at which the detector output can be considered ‘above background’ and
- L_D the net response level that can be expected to be seen with a detector with a fixed level of certainty, or “confidence interval”

If it is assumed that a measurement system has a background response, graphically Fig 2.22 can represent the determination of MDC and type 1 and type 2 errors, where

- α , is the probability of a type 1 error, is an error that says that there is something present when in actual fact this is not the case – a “false positive” and
- β , is the probability of a type 2 error, is the type of error that says that there is not a “real” reading when in fact the opposite is the case.

2.6.4. Detector Counting Efficiency [Bicron 1998]

The Detector counting Efficiency (DE) relates the amount of radiation emitted by the source to the amount measured in the detector.

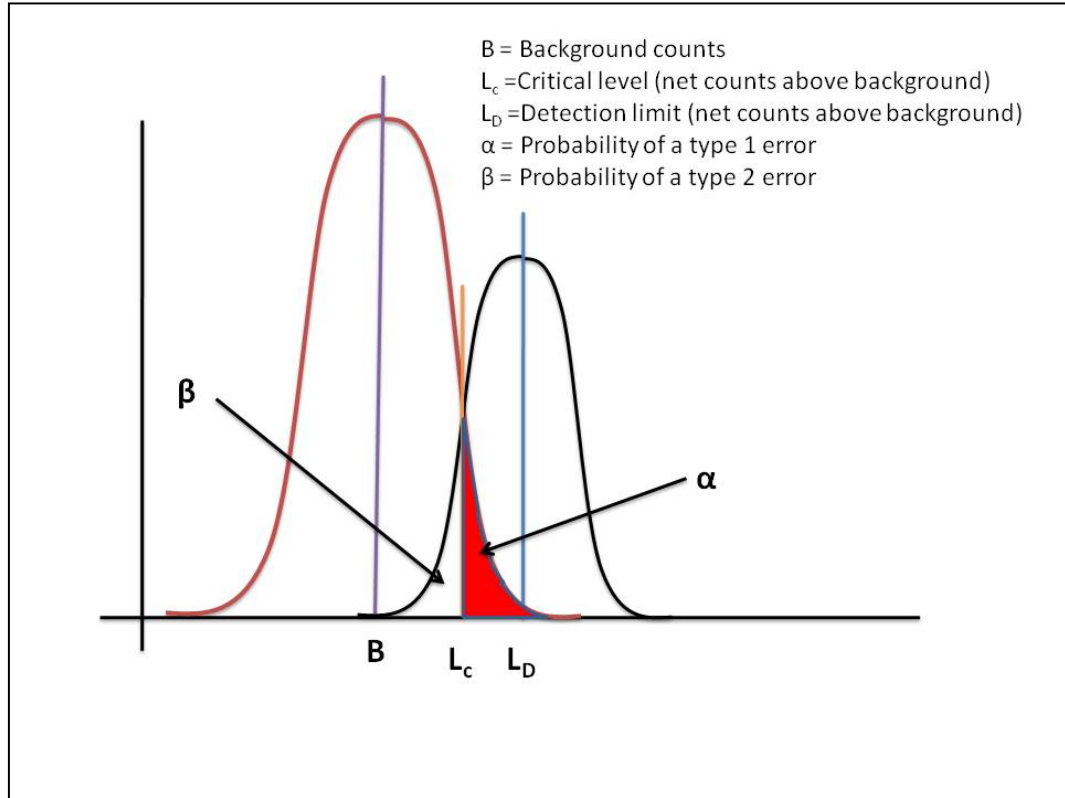


Figure 2-22 Minimum Detectable Concentration (MDC)

Thus, this relationship can be used to calculate the expected count or count rate in the detector if the source strength is independently determined or alternatively if the DE is known, the source strength determined from the reported measurement. The DE is then the ratio of the observed count to the total number of photons emitted in a known counting time and may be written as:

$$DE = \frac{D}{N} , \quad \text{Eq. 2.8}$$

where D is the number of photons counted in the detector and N is the number of photons emitted by the source.

2.6.4.1. Measuring the number of photons in the detector (D)

In general, it is necessary to know the fraction of events in the full energy peak or the peak-to-total ratio R so that the net counts (photofraction) in the photopeak P can be related to the total counts in the detector. It therefore follows that:

$$R = \frac{P}{D} . \quad \text{Eq. 2.9}$$

By rearranging Eq. 2.9 above, D is then determined from the number of counts in the photopeak divided by the peak-to-total ratio. Figure.2.23 shows graphical representations of R calculated for a number of different detectors of varying dimensions.

2.6.4.2. Determining detector efficiency (DE)

There are three factors, G , M , and I that affect the efficient absorption of the photons, N , emitted by the source. Their product is the detector efficiency DE. These factors are defined as follows:

- G is the geometric factor, the fraction of all space around the specimen around the specimen that the detector subtends. Unless the detector completely surrounds the source, the geometrical solid angle factor is less than 1.
- I is the absorption (or transmission) factor, the fraction of the photons transmitted through the intervening materials that reach the detector surface. There are losses due to absorption by material in the path of the photon. Air, detector housing materials and light reflectors around the detector are possible absorbers.
- M is the fraction of the photons absorbed by the detector. The detector material is not always sufficiently thick to stop all the radiation.

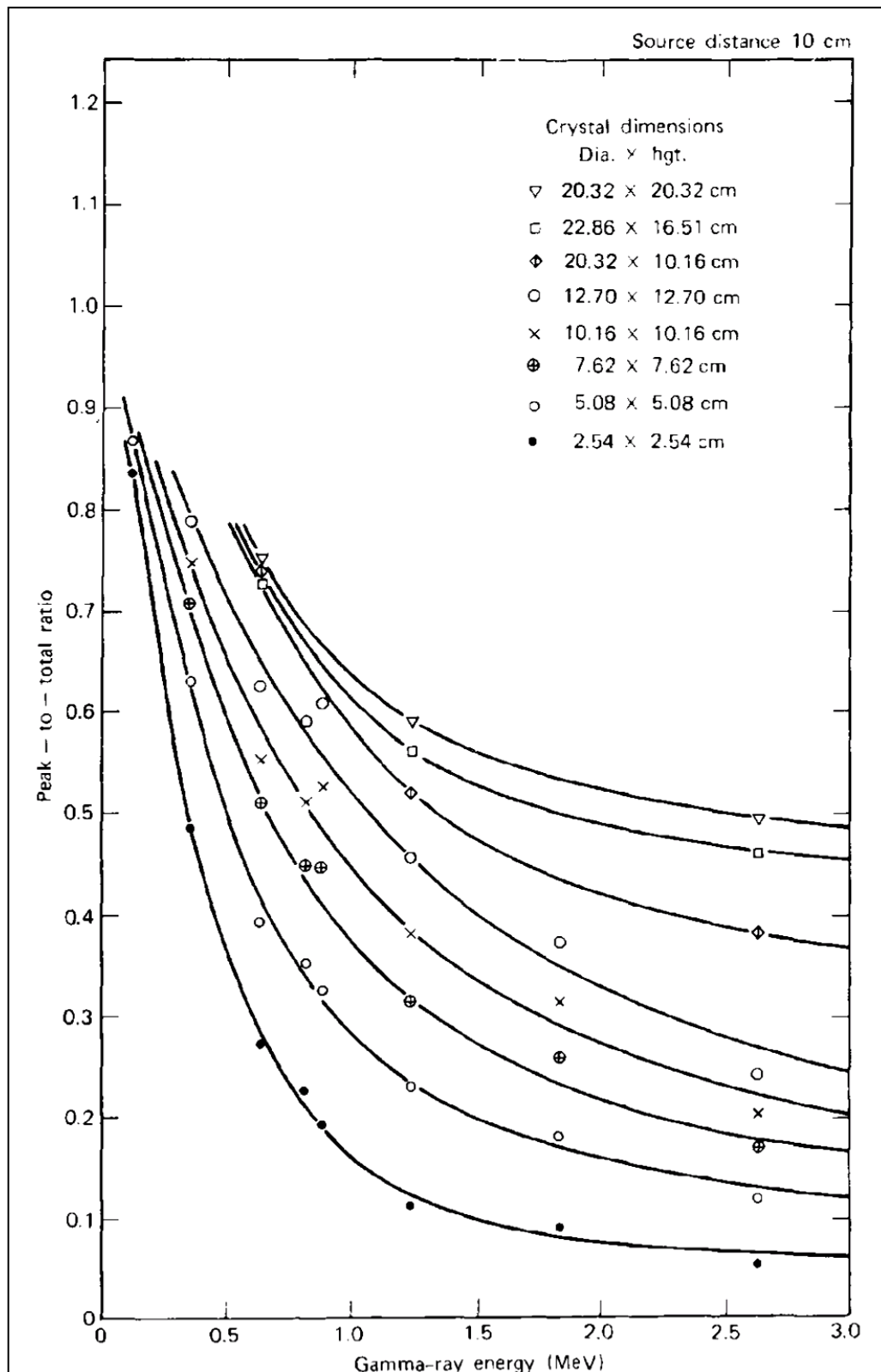


Figure 2-23 Various measured values of r , peak to total ratio for different NaI(Tl) crystal sizes[Bicron 1998].

2.6.4.3. Calculation of G

By way of example, consider a Nanospec® 2" x 2" NaI(Tl) detector that is being used to measure a ^{137}Cs point source 1m away. The geometric factor is determined from

$$G = \frac{\pi r^2}{4\pi R^2}, \quad \text{Eq. 2. 10}$$

where πr^2 is the area of the detector face¹ and $4\pi R^2$ is the area of sphere with a radius equal to the source to detector distance.

Thus, G is determined as

$$G = \frac{\pi \times 0.0508^2}{4 \times \pi \times 1^2} = 6.45 \times 10^{-4},$$

indicating that the detector subtends or intercepts 0.064% of the full sphere at a distance of 1m from the source.

2.6.4.4. Calculation of I

To calculate I , the absorption factor, it is necessary to consider the attenuating effects of 1 m of air and 5.08×10^{-4} m of aluminium (the detector housing) that is there to protect the NaI crystal from the effects of the environment. Thus, I is obtained from

$$I = \exp^{-(\mu_1 \times d_1)} \times \exp^{-(\mu_2 \times d_2)} = 89.5\%, \quad \text{Eq. 2. 11}$$

where

μ_1 = linear attenuation coefficient of air for 662 keV photons, 0.1 m^{-1} [Shleien, Slaback and Birky 1998]

μ_2 = linear attenuation co-efficient of aluminium for 662 keV photons, at 20 m^{-1} [Shleien, Slaback and Birky 1998]

¹ Note that this is an approximation; the numerator should be the area of the spherical section obtained by integrating the area element instead of the area of the flat disc.

d_1 = the distance travelled in air, 1 m

d_2 = the distance travelled in aluminium, 5.08×10^{-4} m.

There is thus an attenuation of approximately 10% of the photons by the intervening air and aluminium. In addition, for a field measurement of radioactivity it would also be necessary to add a component for the linear attenuation of soil, assuming that the isotope of interest was mixed into the soil rather than just lying on the surface. The soil component, however, brings in another order of complexity since various sub-components would go into the calculation of the soil linear attenuation component. These would include, soil density, water content of the soil, depth distribution of the radioactivity and the chemical composition of the soil. This whole complex issue is dealt with at some length in Refs. [Beck, DeCampo and Gogolak September 1972], [Naessens and Xu 1999] and [Benke and Kearfott 2001]. The result of these differing attenuation factors then tends to limit the ability of a detection system such as has been considered here to measure the activity concentrations of the different radioisotopes in the top 60 cm of soil under the detector

2.6.4.5. Calculation of M

The fraction of the photons absorbed by the detector² M , is calculated from

$$M = 1 - \exp^{-(m \times d)} = 0.777, \quad \text{Eq. 2.12}$$

where

m = the attenuation coefficient of NaI of 662 KeV photons 30 m^{-1} [Shleien, Slaback and Birky 1998]

d = the distance of the photons travelled in NaI (TI) detector, 0.0508 m.

² It is assumed here that all photons are traversing the same amount of detector material. A correct procedure takes into account the different paths and their path lengths and sums or integrates to find the total fraction M . This example treats a point source, however, in survey work the source is extended and its necessary to integrate over the extended source.

2.6.4.6. Example of detector efficiency

The detector efficiency (DE) of the Nanospec® for ^{137}Cs at a distance of 1m from the values above is thus determined as:

$$\text{DE} = GIM = (6.45 \times 10^{-4})(8.95 \times 10^{-2})(0.777) = 0.045\%. \quad \text{Eq. 2.13}$$

This means that the Nanospec® at a distance of 1 m from a ^{137}Cs point source detects only 0.045% of the gamma photons emanating from the source. If one were to substitute the 2" x 2" NaI (Tl) detector by a 3" x 3" NaI (Tl) detector then the instrument's DE would then become 0.11% for the same source at a distance of 1 m. This calculation should be considered something of a simplification of the actual situation since it is applied to both ground based and aerial radiometric surveys. A more detailed examination of the airborne situation has been carried out by Billings and Hovgaard [Billings and Hovgaard 1999] and for the ground based situation by Millar [Millar 1997].

2.6.5. Absorption efficiency of the detector material

As can be seen from the calculations above, the photon energies were specified in each calculation. This is necessary because not only does a detector's efficiency vary as a result of its volume, but also with the energy of the photons that the detector is being used to detect. This is illustrated in Fig. 2.24.

2.6.5.1. Energy resolution

When considering the properties of various detectors perhaps one of the more important ones is their ability to differentiate between different peak energies in a spectrum and thus aid in the identification and quantification of isotopes within a sample under consideration. This is known as energy resolution R and is defined as follows:

$$R = \frac{\text{FWHM}}{H_o}, \quad \text{Eq. 2.14}$$

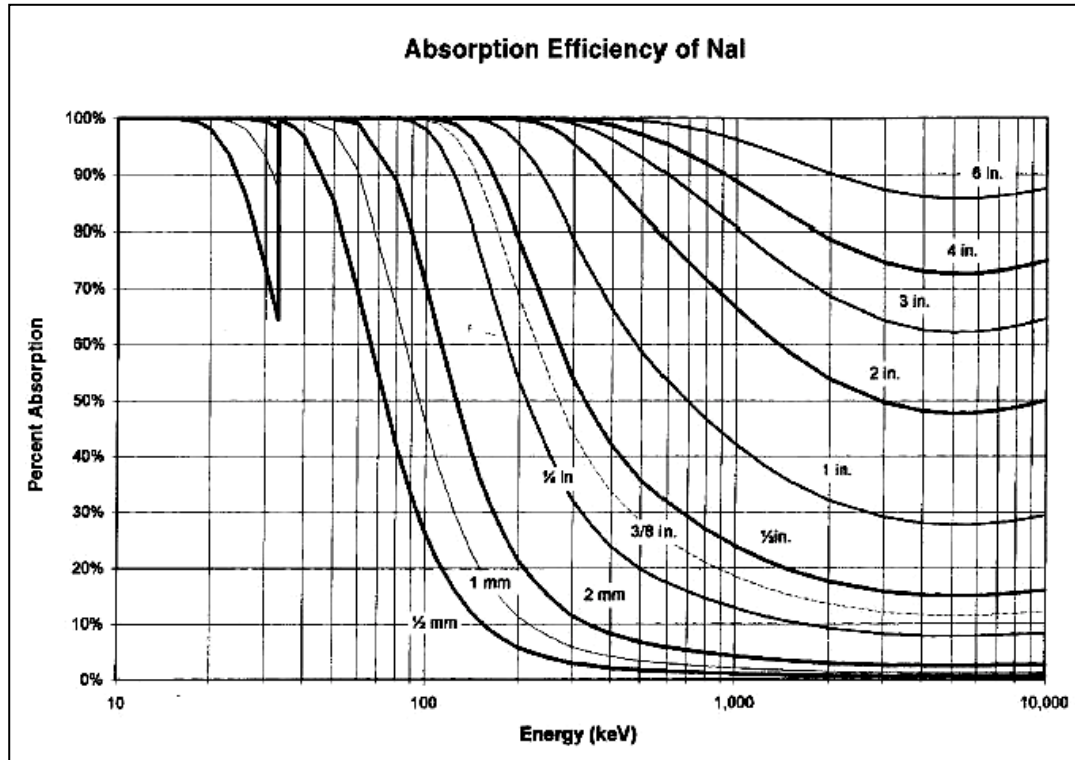


Figure 2-24 Absorption efficiency of NaI (TI) [Bicron 1998]

where

FWHM = full width at half maximum height of the full-energy peak

H_0 = mean pulse height corresponding to the same peak.

There are a number of reasons for this finite energy resolution and which make the scintillation type of detector the worst at resolving peak energies. These include the following:

- Charge collection characteristics, photon collection in the case of scintillators,
- Electronic or thermionic noise,
- Variations in the detector response over its active volume,
- Drifts in operating parameters over the course of a measurement and
- Statistics of the detection process.

Generally, it is the first of these characteristics that results in the greatest ‘broadening’ of the peak and hence loss of resolution. Section 2 (Chapter 10) of Knoll [Knoll 2000] deals with the associated photoelectron statistics.

CHAPTER 3

RADIATION AND ITS GLOBAL DISTRIBUTION

3. Introduction

There are numerous sources of exposure to the human population from radiation and radioactive materials. As can be seen from Table 3.1 the predominant source of exposure is from natural background that will be dealt with in detail later on in this dissertation. However, when consideration is made of the natural background radiation and what specifically is measured, emphasis will be put on the decay chains of ^{238}U , ^{232}Th and ^{40}K as they are the predominant primordial radionuclides that are distributed through the ecosystem and are the ones of principal interest during radiometric surveys both surface and aerial. These are the isotopes that were considered in Chapter two

First though a brief examination will be made of the other sources of radiation that the population at large is exposed to including anthropogenic radionuclides in the environment. Medical isotopes will fall outside the scope of the present.

3.1. Artificial sources of radioactivity

Within Southern Africa, as in any developed country, there are many sources of man-made radionuclides, including many medical sources used for diagnostic work, such as ^{131}I or ^{99}Tc , sources used for various treatment therapies such as ^{60}Co , sources used for irradiation in sterilisation units, again ^{60}Co , and sources used in non-destructive testing including ^{192}Ir and ^{170}Tm . Other sources include accelerators, nuclear reactors and the radioactive products produced during the normal operation of these plants.

Table 3-1 Report to the General Assembly Table 4 Ref. [UNSCEAR 2001]

Annual per caput effective doses in year 2000 from natural and man-made radioactive sources		
Source	Worldwide annual per caput effective dose (mSv)	Range or trend in exposure
Natural background	2.4	Typically ranges from 1-10 mSv, depending on circumstances at particular locations, with sizeable population also at 10-20 mSv.
Diagnostic medical examinations	0.4	Ranges from 0.04-1.0 mSv at lowest and highest levels of health care
Atmospheric nuclear testing	0.005	Has decreased from a maximum of 0.15 mSv in 1963. Higher in northern hemisphere and lower in southern hemisphere
Chernobyl accident	0.002	Has decreased from a maximum of 0.04 mSv in 1986 (average in northern hemisphere). Higher at locations nearer accident site
Nuclear power production	0.0002	Has increased with expansion of programme but decreased with improved practice

3.1.1. Fallout from atmospheric testing of nuclear weapons

Other sources of artificial radioactivity that can be found in the environment include fall-out from the nuclear weapons testing programme that was run by several countries, mostly in the northern hemisphere. (See Fig. 3.1 and 3.2). As can be seen in Fig. 3.3, this contribution is, however, decreasing yearly since airborne testing was stopped in 1963 with the exceptions of France and China who last detonated a weapon in the atmosphere in 1980 [UNSCEAR 2001], after the signature in Moscow on the 5th of August 1963 of the Limited Test Ban

Treaty, which prohibited the detonation of Nuclear Weapons in the atmosphere, under water or in outer space. These figures are taken from the EPA and UNSCEAR and show the effects of the Test Ban Treaty on radioactive doses to the world's population. The Comprehensive Nuclear Test Ban (CNTB) treaty has yet to be ratified by the governments of the world. The world average annual dose from these man-made sources reached a peak in 1963 of 110 μSv and has since fallen to approximately 5 μSv in 2000 [UNSCEAR 2001] the main nuclides that contribute to this dose are ^{14}C , ^{90}Sr and ^{137}Cs .

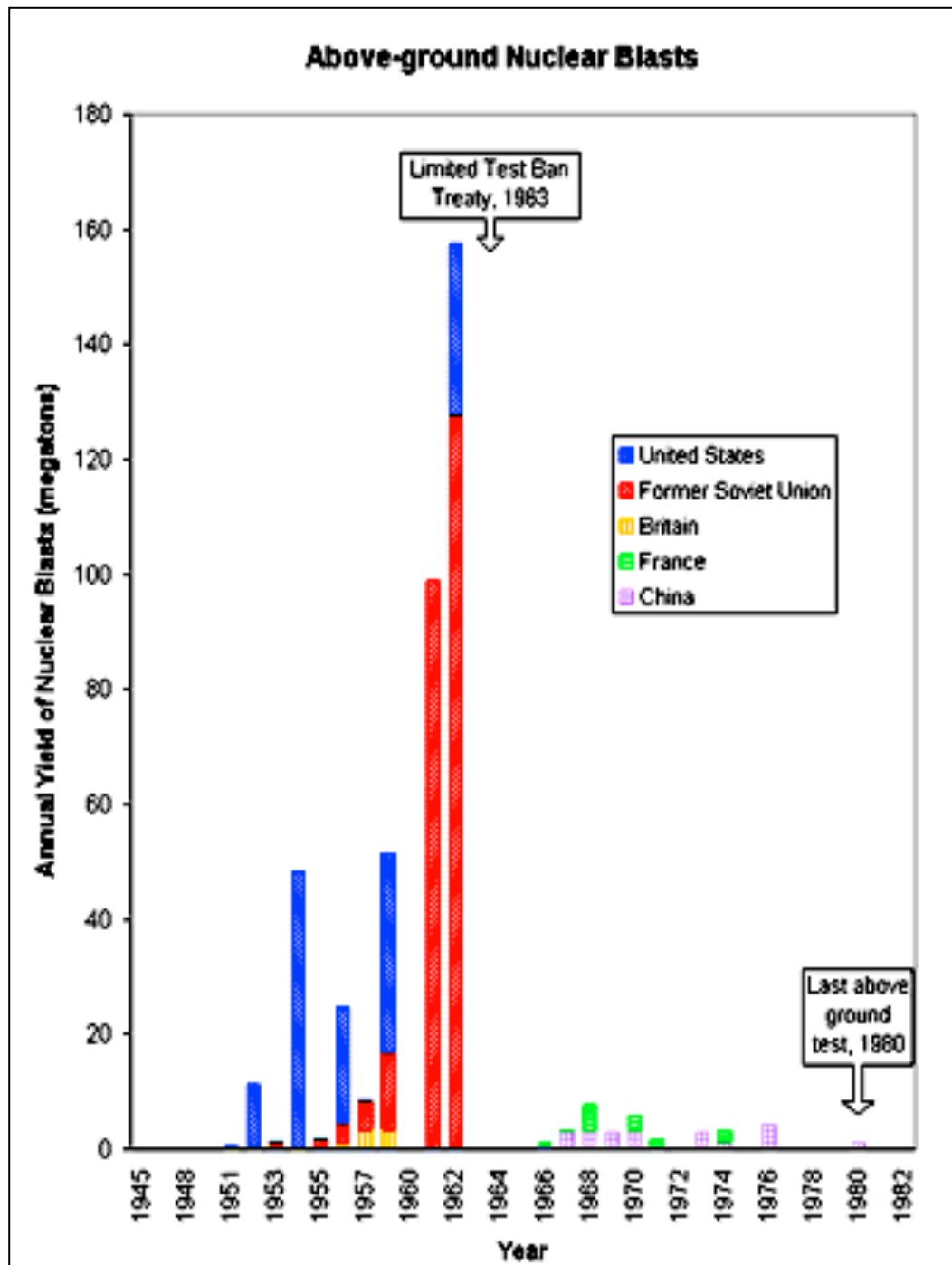


Figure 3-1 Atmospheric Nuclear Weapons Testing, data from Environmental Protection Agency, US

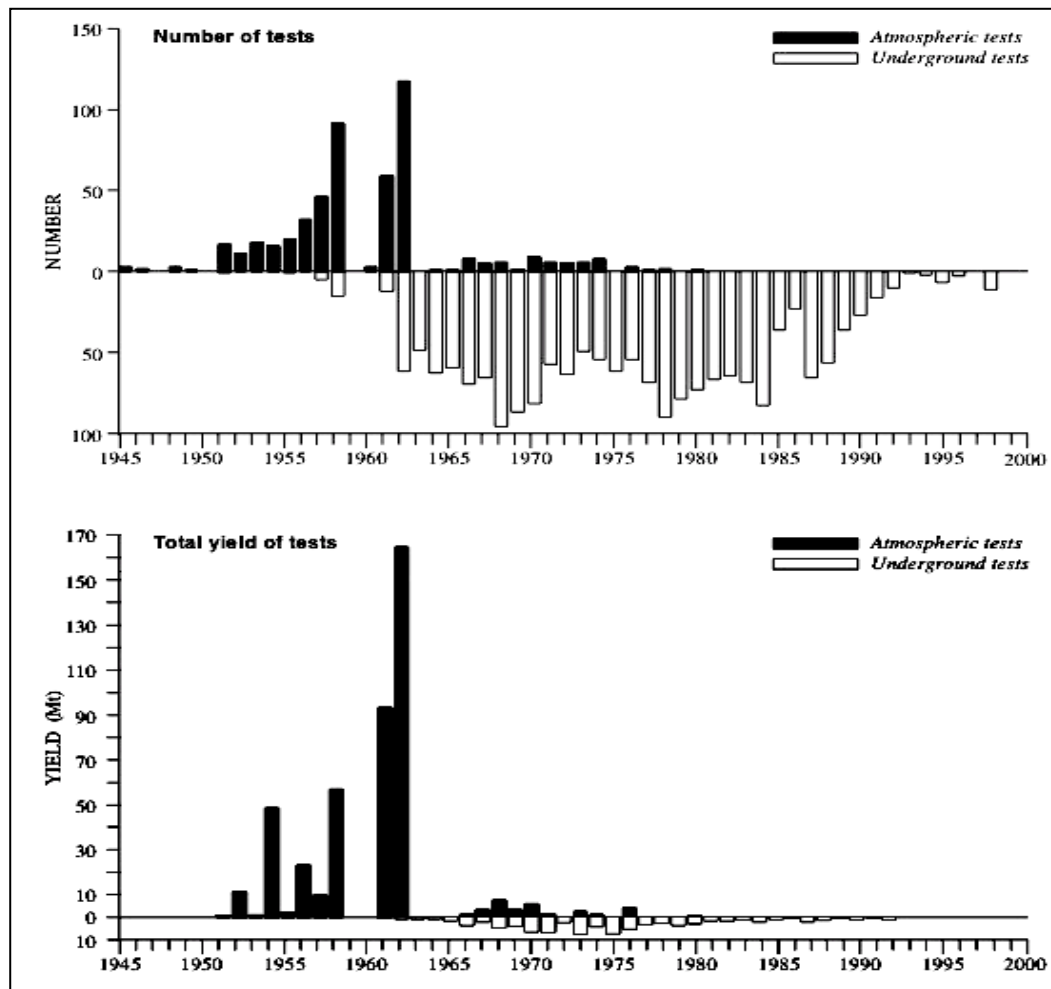


Figure 3-2 Summary of nuclear weapons tested [UNSCEAR 2000].

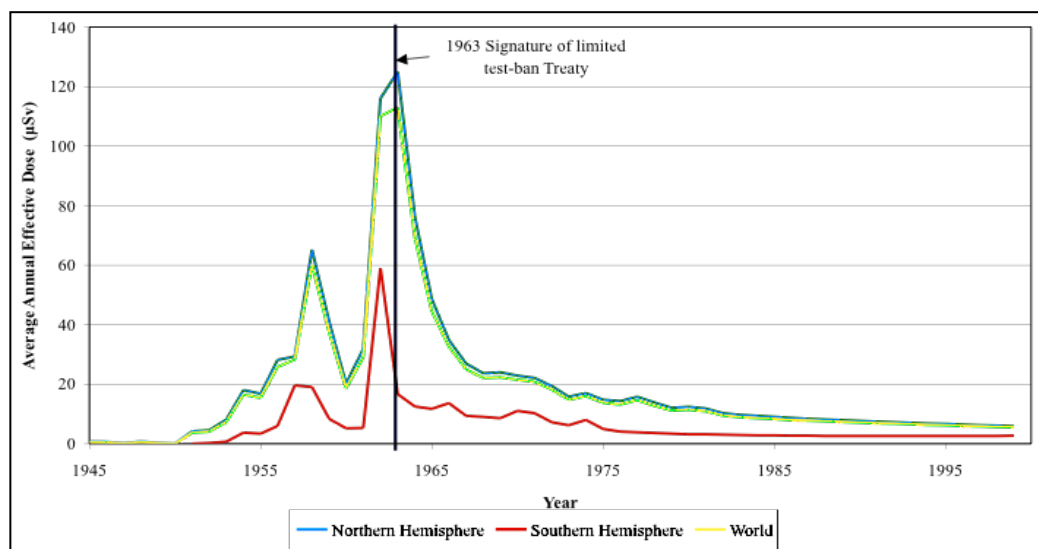


Figure 3-3 Annual effective doses from nuclides produced in atmospheric nuclear testing, [data taken from Table 16 UNSCEAR 2000 Annex C].

3.2. Natural sources of radioactivity

Despite the large number of elements that are known, there are only a few naturally occurring radioisotopes that are found in significant concentrations in the Earth's upper layers and are considered radiologically significant: these are potassium (^{40}K), thorium (^{232}Th) and its daughters and uranium (^{238}U) and its daughters. Table 3.2 lists the average and the range of activity concentrations of these isotopes in selected countries. Many of the reported values are very high resulting in that there are certain parts of the world where it is perfectly possible to receive yearly a radiological dose many times the internationally agreed annual limit for a member of the public merely by living in a particular area.

3.3. Sources of radioactivity in South Africa

In South Africa, there are a number of different sources of radioactivity that have an influence on the measurement of the environmental distribution. Included in this list will be the those radioisotopes potassium, thorium and uranium, sometimes called the 'primordial radionuclides', which because of their long half-lives have a significant portion of their inventory still intact from the time that the Earth's crust was first formed. Also included are TENORM, which are Technologically Enhanced Naturally Occurring Radioactive Materials and cosmogenic radionuclides, which as their name suggests arise as a result of extra-terrestrial bombardment of the Earth's atmosphere. In the next few pages consideration of the distribution and origin of these sources will be considered.

3.4. Basic geology of South Africa

South Africa has a very complex geological history and some of the oldest rocks on earth have been found here. Many geological formations in South Africa are rich in mineral deposits, and some formations have become world renowned, such as the Witwatersrand quartzites (gold), the Bushveld Igneous Complex (platinum) and the coal deposits found in formations of the Karoo sediments as shown in Fig. 3.4 [DEAT 2001].

Table 3-2 Natural Radionuclide content of soil (adapted from [UNSCEAR 2000, Annex B Table 5]).

Country	Population in 1996 x 10 ⁶	Concentration in the soil (Bqkg ⁻¹)					
		⁴⁰ K		²³² Th		²³⁸ U	
		Average	Range	Average	Range	Average	Range
Algeria	28.78	370	66 - 1150	25	2 - 140	30	2 - 110
Argentina	35.22	650	540 - 750				
Bangladesh	120.1	350	130 - 610				
China	1232	40	9 - 1800	41	1 - 360	33	2 - 690
Costa Rica	3.5	140	6 - 380	11	1 - 42	46	11 - 130
Egypt	63.27	320	29 - 650	18	2 - 96	37	6 - 120
India	944.6	400	38 - 760	64	14 - 160	29	7 - 81
United States	269.4	370	100 - 700	35	4 - 130	35	4 - 140

*No information available for South Africa

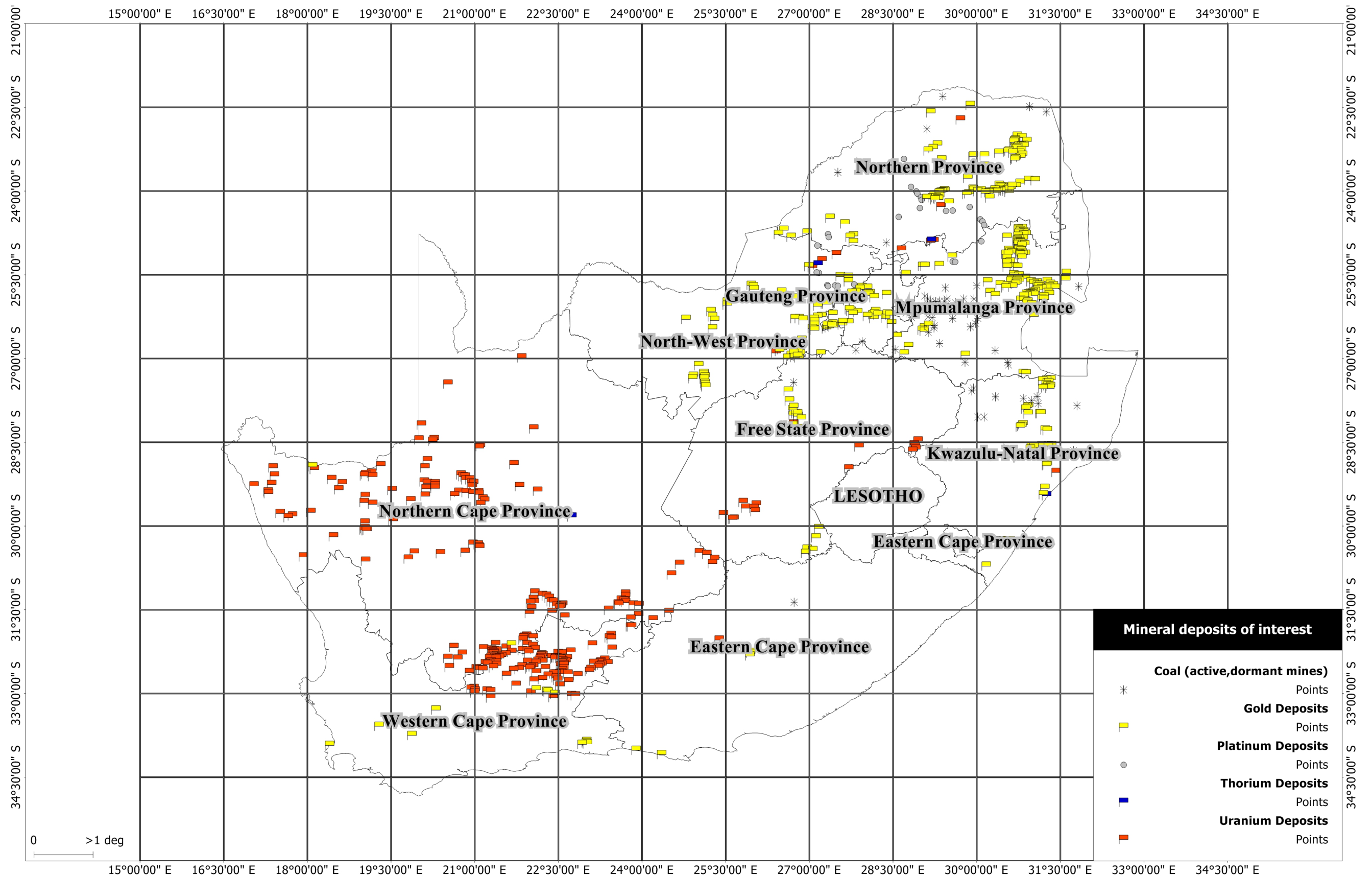


Figure 3-4 Some mineral deposits of South Africa [Council for Geosciences].

3.4.1. Uranium deposits

3.4.1.1. Quartz pebble conglomerate

Quartz pebble conglomerate is the principal gold ore that is mined in the Witwatersrand basin of South Africa. Both gold and uranium are extracted from this ore. It is a mixture of uraninite (uranium oxide UO_2 and U_3O_8), pyrite (iron sulphide FeS_2), chert, silicates, other sulphides and elemental gold. The ore is composed of very hard, white to smoky quartz pebbles cemented by quartz, sericite and chlorite together with dark streaks and seams of pyrite and black carbonaceous material. The gold and uraninite occur in thin seams and between the pebbles in the darker portions of the reefs and can be seen with the naked eye only in the richest specimens as shown in Fig. 3.5 [Nininger, Minerals for atomic energy 1954].



Figure 3-5 Pyritic conglomerate unit of quartz and chert pebbles (photograph from the USGS).

3.4.1.2. Other uranium deposits

In addition to the quartz conglomerates there is also the Karoo basin, a vast area of sandstone uranium deposits (U_3O_8). These tend to be low-grade deposits of the order 0.05–0.4% U_3O_8 . The main uraniferous minerals are uraninite and coffinite.

Additionally, uranium is also found in significant quantities in the coal that is mined in some areas of South Africa, in some instances activity levels up to 100 Bq kg⁻¹ have been found although how widespread is currently unknown. Some unpublished measurements taken by Berkeley Nuclear Services [Berkeley Nuclear Services 2001] on bags of cement produced using fly ash from coal-fired power stations tend to suggest that these levels are indeed quite widespread. Figures 3.6 and 3.7 and Table 3.3 detail the currently known uranium deposits, though this is under constant revision as prospecting continues driven by the world resurgence of nuclear power which has driven the price of uranium oxide up from a long-time stable \$7.00 per pound to values in excess of \$54.00 per pound [Ux Consulting Company 2010].

Another area of uranium deposition is in the Phalaborwa region of the Limpopo Province, where uranium is produced as a by-product of copper mining. Palamin, the company mining at Ba-Phalaborwa, mines ore containing magnetite, vermiculite, apatite, zirconium, titanium and uranium as well as copper. The deposit is hosted in an alkaline igneous complex comprising mainly pyroxenite with occurrences of pegmatites, foskorite and carbonatite.

Table 3-3 Summary of uranium deposit types in South Africa

Type of Deposit	Location and Example
Precambrian gold-bearing quartz-pebble conglomerate	Witwatersrand Basin and Black Reef Formation
Uranothorianite in Alkaline Complexes	Phalaborwa and Pilansberg
Granitic rocks, mostly pegmatites	<i>E.g.</i> Bushveld Complex
Coal and fluvial arenites/argillites	Karoo supergroup

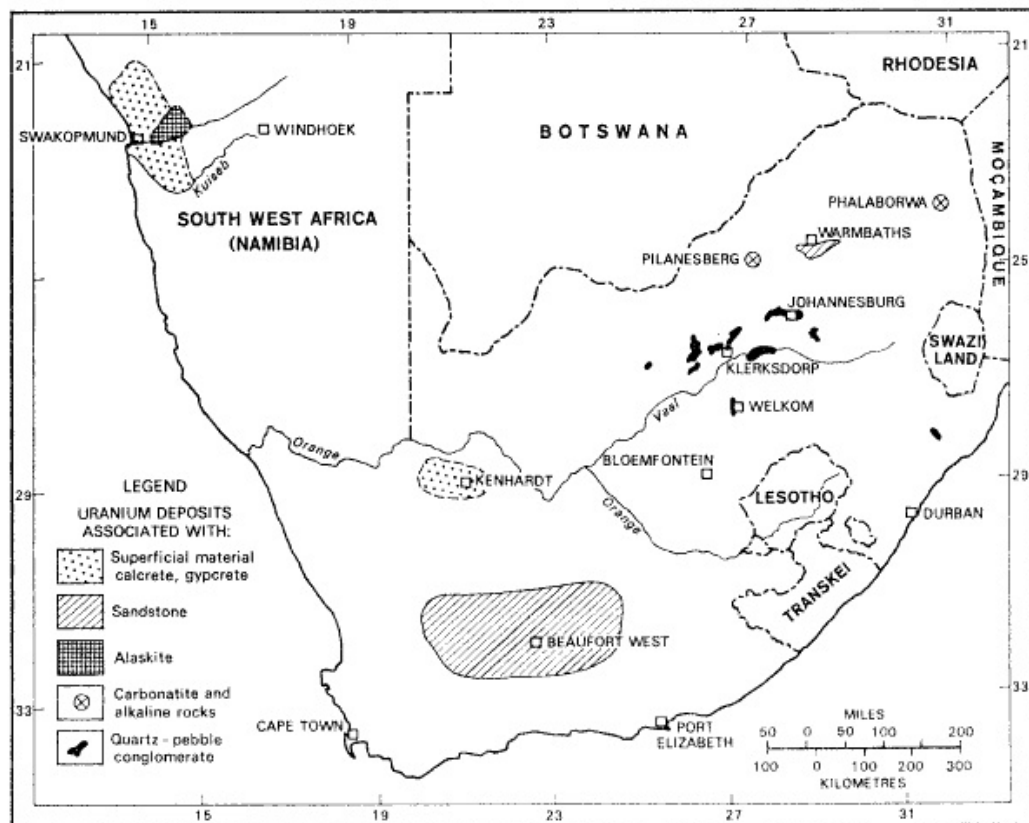


Figure 3-6 Geological features associated with uranium mineralisation in Southern Africa [Von Backstrom 1979].

3.4.2. Thorium deposits

3.4.2.1. Monazite

There are several significant locations within South Africa; the monazite sands on the east coast at Richards Bay north of Durban, and monazite mineral deposits at Steenkampskraal [Andreoli *et al.* 1994] near Vanrhynsdorp in the Northern Cape. The Richards bay location is currently mined and processed to produce rutile, ilmenite and zircon. The Steenkampskraal operation was due to be restarted in April 2002 but is not yet operational, and is designed to extract all the rare earth metals, supplying between 2% and 3% of the world's requirements.

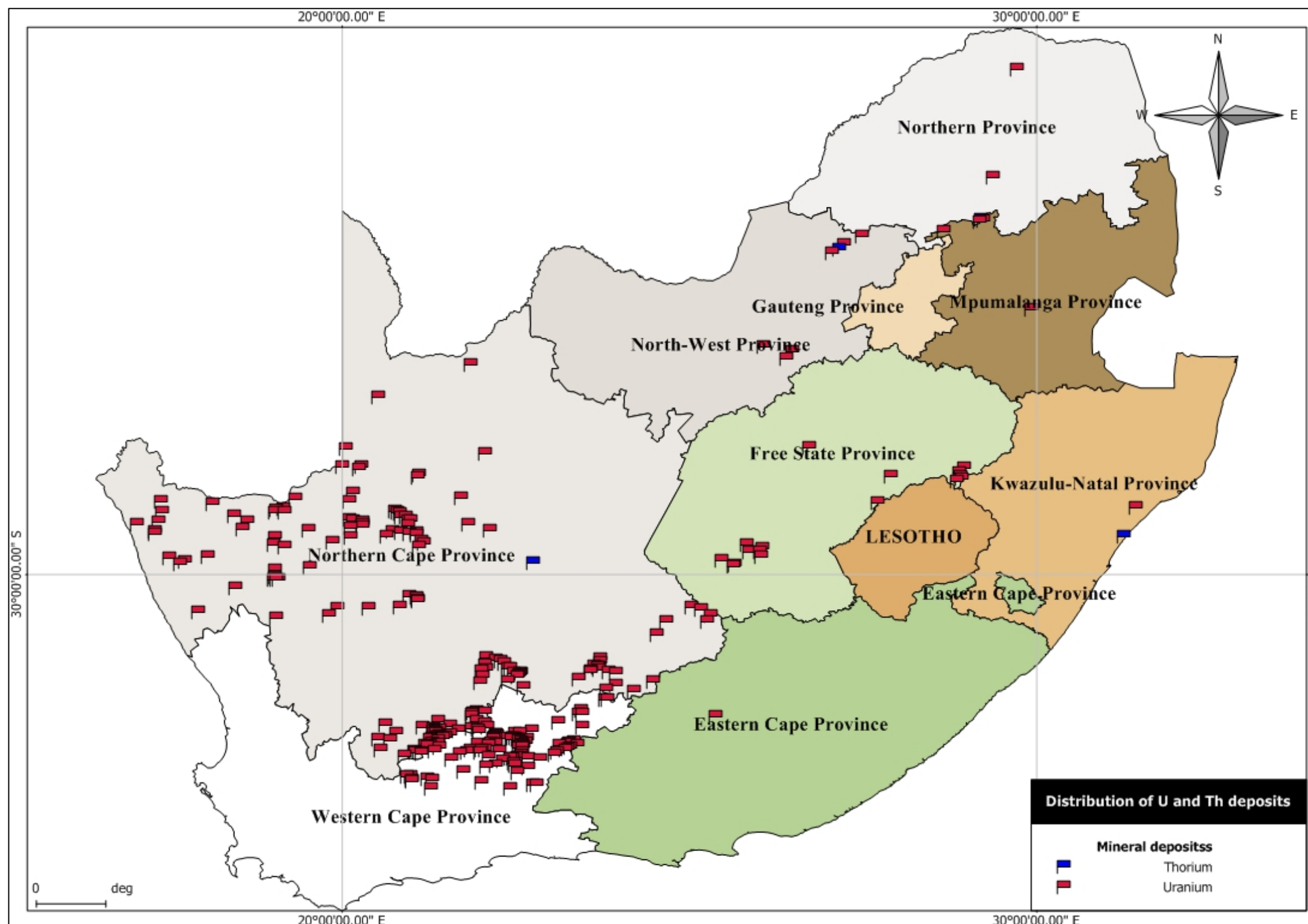


Figure 3-7 Distribution of uranium and thorium deposits, 2001 modified data (Council for Geosciences 2001).

3.4.3. Cosmogenic radionuclides

The interactions of cosmic-ray particles in the atmosphere produce a number of radionuclides; these include ^3H , ^7Be , ^{14}C and ^{22}Na among others. Production is highest in the upper atmosphere but is also latitude dependent and varies with the 11-year solar cycle [Lal and Peters, Cosmic ray produced radioactivity on the earth 1967] [Lal and Suess, The radioactivity of the atmosphere and the hydrosphere 1968]

Table 3-4 Production rates and concentrations of selected cosmogenic radionuclides in the atmosphere [UNSCEAR 2001].

Radionuclide	Production rate	Global inventory (PBq)	Fractional amount in the troposphere ^a	Concentration in the troposphere ^b (mBqm ⁻³)
	Annual amount ^c (PBqa ⁻¹)			
^3H	72	1275	0.004	1.4
^7Be	1960	413	0.11	12.5
^{14}C	1.54	12750	0.016	56.3
^{32}P	73	4.1	0.24	0.27
^{22}Na	0.12	0.44	0.017	0.0021

^a References [Lal and Peters, Cosmic-ray produced radioactivity on the earth 1967] [Lal and Suess, The radioactivity of the atmosphere and the hydrosphere 1968]

^b Assumes tropospheric volume of $3.62275 \times 10^{18} \text{ m}^3$

^c Assumes surface area of world = $5.1005 \times 10^{14} \text{ m}^2$

3.4.4. Technologically Enhanced Naturally Occurring Radioactive Material (TENORM)

This type of material is certainly the greatest source of concern because of the potential environmental impact it poses and because it is the largest group in terms of quantities involved. The vast majority of the Technologically Enhanced Naturally Occurring Radioactive Material (TENORM) is in the form of slimes dams (see Fig. 3.8) – waste dumps that have been created as a result of mining for various minerals in South Africa, including gold, uranium and phosphate. Figure 3.9 shows the distribution of TENORM in the Gauteng region. It is mapped as total activity in Bqg^{-1} . These dams consist largely of crushed ore that has been processed to remove the desired mineral or metal. Individually, dams contain millions of tons of slimes and there are a large number of them distributed throughout South Africa, usually constructed near the mines that they provide the dumpsite for.



Figure 3-8 Photograph of a slimes dam, east of Johannesburg (photograph taken by author).

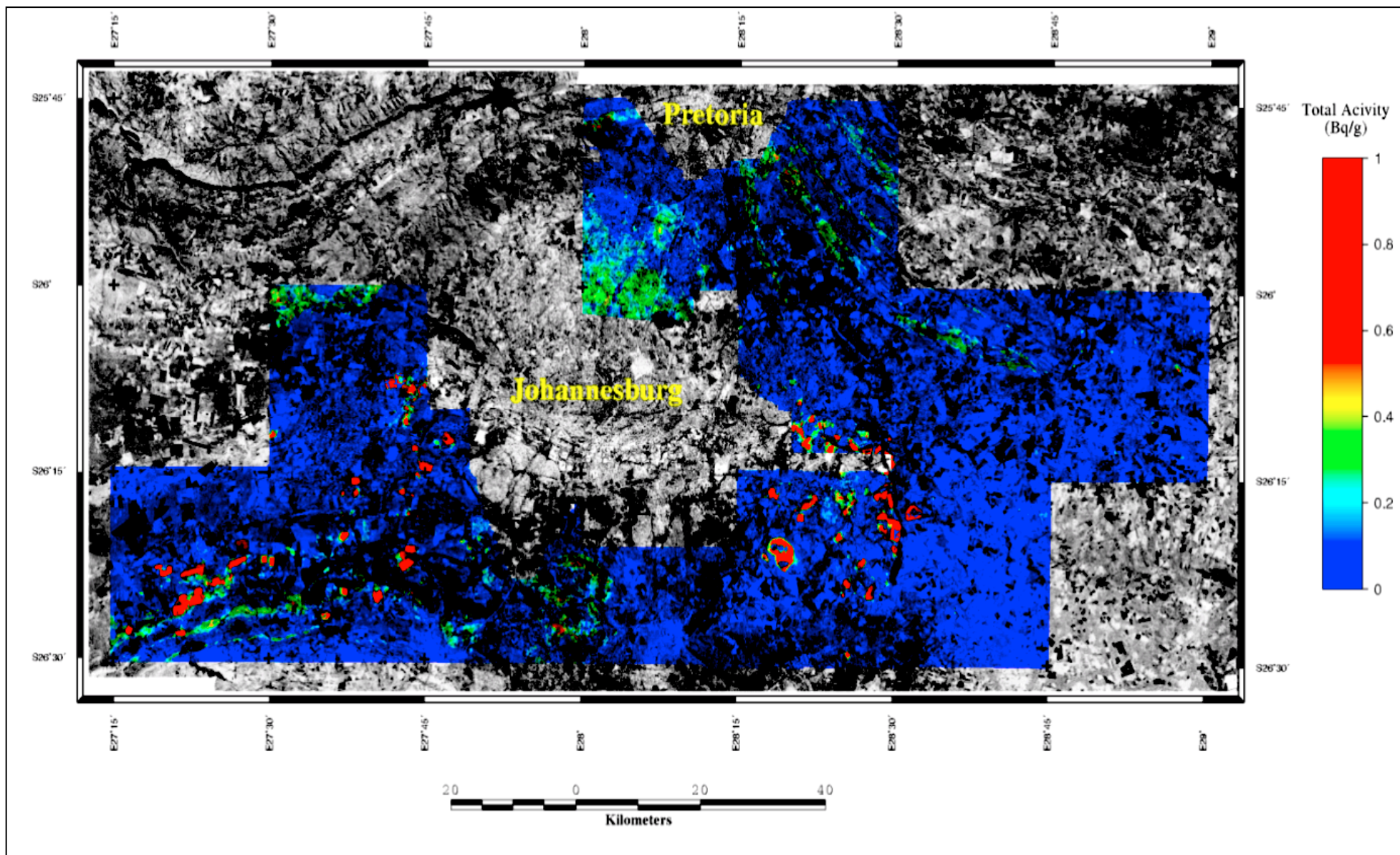


Figure 3-9 Distribution of radioactivity in Gauteng (Council for Geosciences).

3.5. Issues that will affect a survey

In the following sections the various issues that will affect a survey will be discussed.

3.5.1. Disequilibrium in the uranium or thorium decay chains

When radiometric surveys are carried out, it is usually assumed that the isotopes in the ^{238}U and ^{232}Th decay chains are in secular equilibrium, that is to say as the preceding isotope within the decay chain decays into the 'daughter' isotope there is no removal of any of the isotopes by chemical or mechanical means. This assumption is important, since the activity concentrations of the various isotopes in a sample are derived by the measurement of specific isotopes within the decay chains of thorium and uranium. Within the decay chain of ^{238}U , when looking at the ratio of uranium to radium one can check this assumption. If there is equilibrium there should be a value of 2.84×10^6 to 1 [Kirsch 1928].

This is done because of the difficulty of directly measuring the gamma ray energies of many of the daughter products of both decay chains. In airborne radiometric surveys, gamma rays from ^{214}Bi are usually detected and then reported as values in 'equivalent Uranium' or eU units and in the case of ^{232}Th activity concentrations are calculated with reference to ^{208}Tl - these being the isotopes yielding the most prominent gamma-rays in each of the decay chains. The assumption of secular equilibrium is often valid more particularly in the thorium decay chain, however there are situations where this assumption does not hold true. The following few paragraphs (sections 3.62 – 3.7) will deal with some of the reasons why secular equilibrium may not be assumed.

3.5.2. Weathering and erosion processes

These processes are means by which rock is broken down into smaller particles that eventually go to form soil. Figure 3.10 shows a piece of uranium ore that displays some of the typical signs of weathering.



Figure 3-10 Uranium ore (US Geological Survey and the Mineral Institute).

Some of the weathering processes directly affect secular equilibrium [Rashmi, Kant and Sharma 2009]; in addition, the weathering process often has an indirect effect. These indirect processes will be considered first.

Weathering can be regarded as the breakdown or alteration of rock in its natural or original position at or near the Earth's surface through physical, chemical, and biological processes induced or modified by wind, water and climate.

There are several types of physical weathering and are discussed below;

3.5.2.1. Freeze/thaw weathering

Freeze/thaw weathering occurs when the temperature falls below freezing at nighttime and rises above freezing during the day. Any water that is trapped within the cracks and fissures of the rock will expand by approximately 9% when it freezes, thus putting the rock under pressure and further fracturing it [Yoshikawa *et al.* 2006]. This increase in the fracturing of the rock then increases the surface area of the rock speeding up the various chemical-weathering processes. The increased fracturing also increases the rate of radon exhalation

from the rock [Singh *et al.* 2010], which has a significant impact on the equilibrium in the later members of the uranium decay chain.

3.5.2.2. Salt crystallisation

The salt crystallisation process is particularly prevalent in hot deserts and coastal regions. Water enters the fissures and cracks in rock and if it is at all saline as the water evaporates salt crystals are left behind. The increase over time, of these deposited crystals puts the surrounding rock under pressure eventually leading to the rock shattering, that in turn creates larger surface areas for further chemical attacks and more weathering.

3.5.2.3. Thermal expansion or insolation

Thermal expansion or insolation occurs particularly in desert type areas. The process is similar in ways to freeze/thaw weathering. The daytime sun to temperatures in excess of 40oC, causing the rock to expand while as nighttime comes around the rock cools and contracts, heats the outer layers of rock. This continuous cycle of expansion and contraction causes the outer layers of rock to eventually peel away a bit like an onion and then shatter, see Figs. 3.11 and 3.12. [Warke and Smith 1998].

3.5.2.4. Biological weathering

The biological weathering process involves the root systems of plants which grow into the cracks of rock and as the plant system grows, slowly increases the pressure eventually breaking the rock into smaller pieces, increasing the surface area of the rock and allowing the rate of chemical weathering to increase. Figure 3.13 shows a wild fig growing on a rocky outcrop in the Shashi River on the border of South Africa, Botswana and Zimbabwe.



Figure 3-11 Rock showing fracture lines caused by insolation (picture taken by author).



Figure 3-12 Rock showing the progressive breakdown of the material due to insolation (picture taken by author).



Figure 3-13 Wild Fig in rock on the Shashe River Botswana – an example biological weathering (picture taken by the author).

3.5.3. Chemical weathering - an overview

Chemical weathering happens when the minerals are altered in some manner leading to the disintegration of the rock. It tends to affect certain minerals selectively and although there are several different types of chemical weathering, including;

- Oxidation [Thornton 1984],
- Hydration,
- Hydrolysis,
- Carbonation solution chelation and
- Effects of acid.

All of these different forms of 'chemical attack' tend to work in unison. One of the prerequisites for chemical weathering to take place is the presence of water - usually rainwater [Thornton 1984]. Rainwater usually contains low concentrations of dissolved solids, which are mainly derived from the evaporation of seawater,

together with small amounts of atmospheric gas. Some of the chemical processes involved in weathering are as described in the following sections.

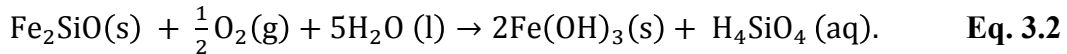
3.5.3.1. Dissolution

This is the simplest chemical weathering process and involves the dissolution of soluble materials, independently of the pH. Water, because of its polar nature is most effective in dissolving solids with ionic bonds such as sodium chloride (NaCl), common salt, and may be represented by:

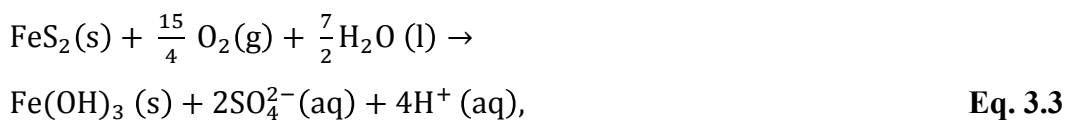


3.5.3.2. Oxidation

Free oxygen is important in the breakdown of rock that contains such reduced substances as ferrous ions (Fe) and sulphur. The oxidation of reduced metals results in the formation of colloidal oxides or hydroids. An example of such a reaction is the following:



An important reaction, particularly in South African gold mines, is the oxidation of sulphides such as pyrite (FeS_2), which produces sulphate and hydrogen ions:

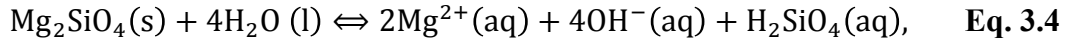


in slimes dams of gold mines, this particular reaction can generate acidic waters with a pH of less than 4, which allows the mobilisation of uranium salts [Li, Victor and Chakrabarti 1980] which can then be deposited some distance from the slimes dam; especially if the integrity of the penstock³ has been ruptured or badly maintained with the resulting leakage of waste water, giving rise to an uncontrolled release of radioactivity into the environment.

³ Barrier around the base of a slimes dam

3.5.3.3. Hydrolysis

This is a very similar process to dissolution, except that the water reacts chemically with the dissolved ions. The reaction of the mineral forsterite, Mg_2SiO_4 , and the sedimentary carbonate mineral calcite CaCO_3 with water are just such examples of hydrolysis:



The two acids produced in these reactions are 'weak' acids, that is to say they do not under normal conditions dissociate much and since the hydroxides of both calcium and magnesium that are formed are strong, well dissociated alkalis the solutions produced are alkaline in nature.

3.5.3.4. Acid hydrolysis

This is a similar process to hydrolysis, only now the water contains hydrogen ions from dissolved acids, which make it a more effective form of chemical weathering. The most important sources of acidity in the environment are from the dissociation of water molecules, the dissolution of carbon dioxide, sulphur dioxide and organic acids of various types. Carbon dioxide dissociates in water into bicarbonate and carbonate ions, thereby supplying hydrogen ions for the chemical weathering reaction.

3.5.4. Radon exhalation

It is only in the ^{238}U decay chain, which includes ^{222}Rn with a half-life of 3.82 days, where consideration needs to be given to the effect on secular equilibrium by losses of gaseous radon. Thoron, the gaseous isotope of radon in the thorium decay chain has a half-life of only 55.5 s after which it decays into another solid element, ^{216}Po , and it is thus rapidly trapped within the matrix of the rock before it has time to escape. Actinon, the gaseous isotope of radon in the actinium series with a half-life of 3.96 s, which also decays into a solid, is even less likely to be able to escape from rock matrices.

3.6. Chemical separation during ore processing and other situations

Due to various chemical reactions during chemical processing of pyrite in the production of oleum (concentrated sulphuric acid) used in the extraction of uranium, radium is preferentially deposited in the joints and behind the rubber lining of various thickener tanks in the process stream. When this rubber is stripped off the tank walls during maintenance, these radium-enriched scales are then collected and dumped on slimes dams as waste. Radium enriched dusts also accumulated in the quench pots used in the production of oleum on various gold and uranium mines in South Africa.

Such a chemical separation is not confined to industrial situations but also happens in the natural environment. An example of this has been given by Vogel and his colleagues for the Uitenhage Aquifer [Vogel *et al.* 1999], here, migration of what is known as an oxygen depletion zone along the aquifer has resulted in the change in the reduction/oxidation conditions causing the change from the U^{6+} valence state which is soluble to the U^{4+} state which is less so. This causes the uranium to precipitate out of solution.

Combining the change of valence as a result of the changing reduction/oxidation conditions and the migration of this so called barrier leaves uranium in various states of secular equilibrium with ^{230}Th its daughter product. This dynamic system then prevents the establishment of secular equilibrium with the various decay chains of uranium in this system.

3.6.1. Chemical fractionation of the uranium and thorium decay chains during weathering

The fractionation of the radionuclides in the uranium and thorium decay chains during weathering processes and the transfer into river waters is largely controlled by the differences in chemical properties of these elements in solution [Chabaux, Riotte and Dequincey 2003]. Here, it should be noted that temperature, pressure, and solution composition (pH, redox potential ionic strength and the occurrence of complex forming ligands) all control how elements behave in the environment.

Thermodynamic data of uranium indicate that in environmental conditions the predominant oxidation states of the uranium ions are either U^{4+} or U^{6+} . Under reducing conditions, U^{6+} is very insoluble and tends to precipitate out of solution as insoluble uraninite. This solubility though increases at low pH in the presence of fluorine by formation of uranous fluoride complexes and above pH 7-8 by complexation of U^{4+} with hydroxyl ions as shown in Fig. 3.14 [Chabaux, Riotte and Dequincey 2003].

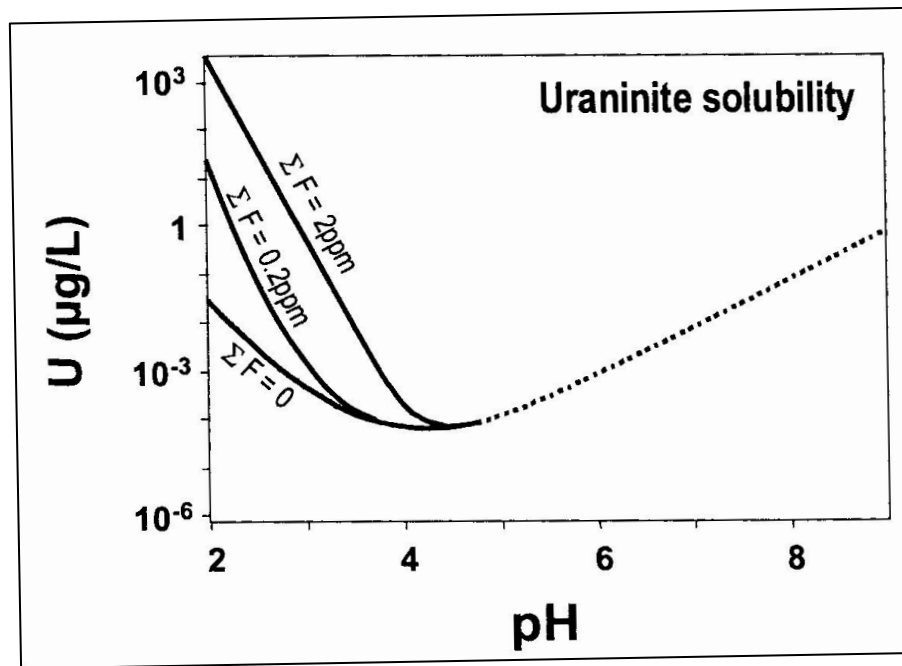


Figure 3-14 Solubility of uraninite as a function of pH at 25°C for different fluoride concentrations. (*Geochim Cosmochim Acta*, Vol. 42, Fig. 4, p. 555).

The thermodynamic properties of the uranium and thorium decay series of nuclides in solution are important parameters to take into account when explaining the U-Th-Ra mobility in surface environments. They are, however, not the only ones controlling radionuclide fractionations in surface waters and weathering profiles. These fractionations and the resulting radioactive disequilibria are also influenced by the adsorption of radionuclides onto mineral surfaces and their reactions with organic matter, microorganisms and colloids [Chabaux, Riotte and Dequincey 2003]. Longmuir [Longmuir 1978] shows that uranium in natural waters usually complexes with a number of different ions including those of hydroxide, phosphate and fluoride and there is a possibility of

silicate being included in the list (see Fig. 3.15). It has been shown that these complexes greatly increase the solubility of uranium minerals and the mobility of uranium in surface and ground waters and as a consequence also greatly increases the possibility of disequilibrium occurring in the uranium decay chain.

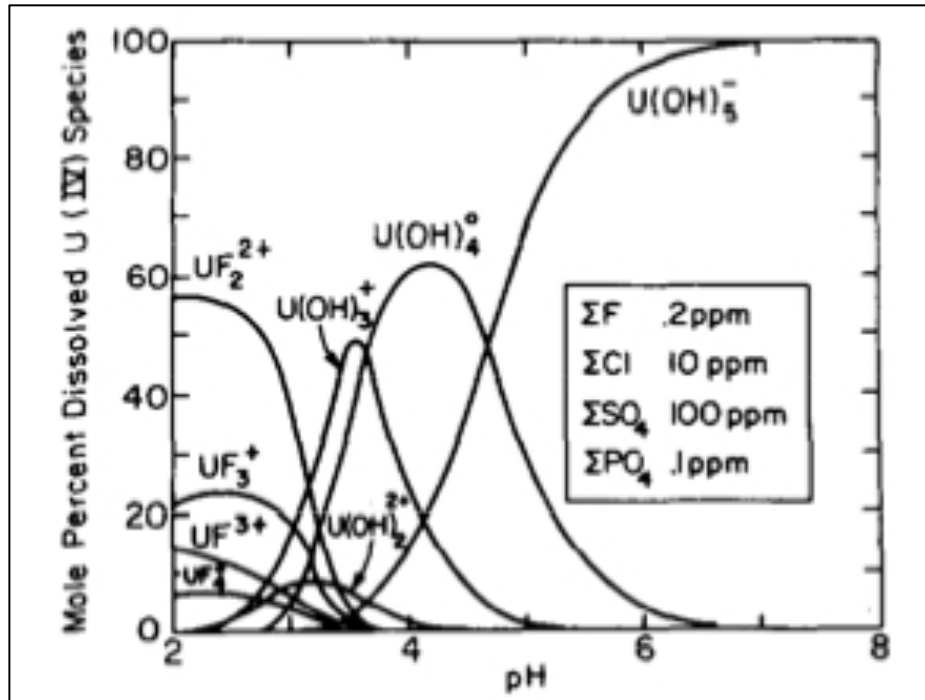


Figure 3-15 Distribution of uranous complexes vs. pH for some typical ligand concentrations in ground water at 25°C [Chabaux, Riotte and Dequincey 2003]

3.6.2. Effects of organic matter, microorganisms and colloids on disequilibrium

A number of studies have been done that show that there is a strong affinity for uranium and thorium by organic matter (see e.g [Chabaux, Riotte and Dequincey 2003]), particularly to the organic acids, citric, fulvic and humic.

There is also a growing body of evidence to show that microbiological activity in soils and waters can also play an important role in the mobility of uranium. In a review it has been shown that there are numerous mechanisms of uranium accumulation by microbes generally sub-divided into metabolism-dependent and metabolism-independent [Suzuki and Banfield 1999]. The details fall outside the scope of this dissertation.

The fixture of radionuclides by colloids is also another important mechanism in the fractionation of the uranium/thorium series nuclides. Chabaux *et al.* [Chabaux, Riotte and Dequincey 2003] detail the various ways in which these colloids interact with the nuclides.

3.6.3. Alpha recoil

This is another mechanism that has been shown to create radioactive disequilibria. The radioactive daughter is mobilised from its initial position by the energy of alpha decay. Besides the displacement of the recoil atom [Kigoshi 1971] of some 20 nm or more (see Fig. 3.16), the alpha-recoil can induce changes in the physical and chemical properties of the recoil atom and damage the host mineral. [B. Minty 1997] Alpha recoil is one of the more important ways of releasing thorium, radium and radon into groundwater [Sun and Semkow 1998] and accounts for the radioactive disequilibria. Damage to the crystal lattice and the oxidation of $^{238}\text{U}^{4+}$ to $^{234}\text{U}^{6+}$ during alpha-decay causes the ^{234}U atom to preferentially go into solution. Compared to the parent nuclide ^{238}U , the recoil atom ^{234}U is loosely held in its mineralogical site [Chabaux, Riotte and Dequincey 2003]. The direct ejection of the recoiling nucleus from the solid phase to the fluid one is another process that accounts for ^{234}U enrichment in waters [Kigoshi 1971].

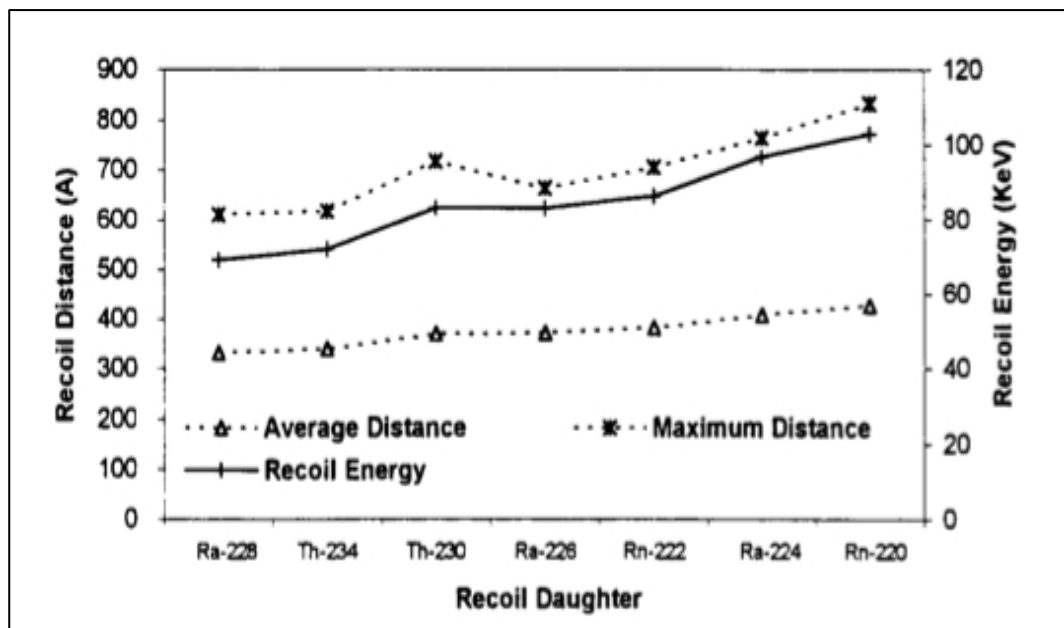


Figure 3-16 Recoil energies plus average and maximum recoil distances in quartz for ^{238}U and ^{232}Th decay chains [Sun and Semkow 1998].

In summary, alpha recoil, thermodynamic properties of the radionuclides in solution, sorption of radionuclides onto mineral surfaces, and their complexation in organic matter in soils and by colloids in surface waters are important processes controlling the mobility of, and hence the fractionations between, the uranium and thorium decay series nuclides within weathering profiles and in river waters [Chabaux, Riotte and Dequincey 2003].

CHAPTER 4

AIRBORNE RADIOMETRIC SURVEYING

4. Introduction

Airborne radiometric surveys have been carried out in different parts of the world from the late 1950's [Martin, et al. 2006] [Zhang, Xiong and Chen 1998] [Galbraith and Saunders 1983] [Smith 1985]. Initially these sorts of surveys were carried out to search for deposits of minerals, which could be mined and used in the nuclear industries and to further the weapons race in the Cold War. More recently, the major emphasis has been on feeding the need for power generation. Nuclear power has once again been included in the world's energy generation mix as it is realised that there are finite quantities of coal and oil to be mined. Additionally, there is the concern about the release of massive quantities of carbon dioxide into the atmosphere and the potential effect it is having on global weather patterns.

4.1. Airborne radiometric surveys instrumentation

Fundamentally, the same basic setup is used for all airborne radiometric surveys: That is a detector, an electronics package that records the data and some sort of storage mechanism, nowadays usually solid state designed to withstand the rigours of flight.

The various components are described in detail below.

4.1.1. Detector

Almost always for airborne work, use is made of detectors based on the scintillation properties of sodium iodide doped with thallium NaI (Tl). As with all detectors there are certain trade-offs to be made. NaI (Tl) based detectors do not have a particularly good resolution (photopeak FWHM of 7-10%). It is, however, possible to grow large volumes of these NaI crystals. Indeed, it is not uncommon to have crystals with volumes between 30 and 50 l [Zhang, Xiong and Chen 1998]. Lithium drifted germanium based detectors have a significantly better

resolution than NaI (Tl) based ones but cannot be made in the same size [Knoll 2000]. The sensitive volume is then much smaller and it has to be said these types of diodes are much more expensive than NaI (Tl) crystals. It is always desirable when carrying out airborne surveys to have as much sensitive volume as is possible to enhance the overall sensitivity of the survey. Though what can be carried is then based upon the aircraft used in the survey. For example, a standard survey set up can be seen in Fig. 4.1.



Figure 4-1 Two RS-500 detector packs fitted in airplane (Picture courtesy of Radiation Solutions Inc.).

The configuration of these detectors is quite interesting; generally a detector system consists of a number of detectors, two as a minimum. The large volume detector is always mounted so that it will detect the gamma photons coming from beneath the detector volume. A secondary detector, usually of a smaller volume, is set up to detect gamma photons coming from above the large detector volume. This detector is used in the calibration of the instrument and to measure 'background' radiation value that is used in post processing of the data. The background is made up of cosmic-ray radiation, radioactivity in the fabric and

construction materials of the aircraft and electronic noise in the detection system itself. However, on a precautionary note a number of aircraft manufacturers have used thorium alloys when manufacturing items like engine casings and the radioactive signal from these parts needs to be excluded from the survey data. The Atlas Cheetah is one such example of an airplane that has small quantities of thorium in its engine casing [Lalkhan 2008]. In addition, the upward facing detector can also be used to give an estimate of radon that might be present, which will potentially affect the calculation of activity in final survey results.

4.1.2. Electronics

Clearly, it is necessary to record and analyse any such measurements that might be made using the detector system. The original systems that were used in the 1950's were considerably different to those that are used nowadays. The original systems consisted of a pre-amplifier mounted on the detector, an amplifier which was fed the signal into a multi-channel analyser with all the output being recorded onto magnetic tape. The results were usually plotted using a line plotter. Nowadays, with the many-fold increase in computing power and data storage, what used to weight possibly 40 or 50 kg can probably now be reduced to about 5 kg. (See Fig. 4.2). The only part of the equipment that has not been reduced in size is the detector package itself.

In addition to the detector package the aircraft needs to be equipped with a number of other instruments. These include a Global Positioning System (GPS) for both navigation and recording the position of the plane during a survey, some sort of precise altimeter (possibly radar based) to give an exact height of the airplane above the ground and above mean sea level. In addition, a barometer and a thermometer provide atmospheric pressure and temperature values respectively, that are necessary for inclusion into the final data algorithm used to plot survey spectra to correct gamma-ray data for the height of the detector above the ground.

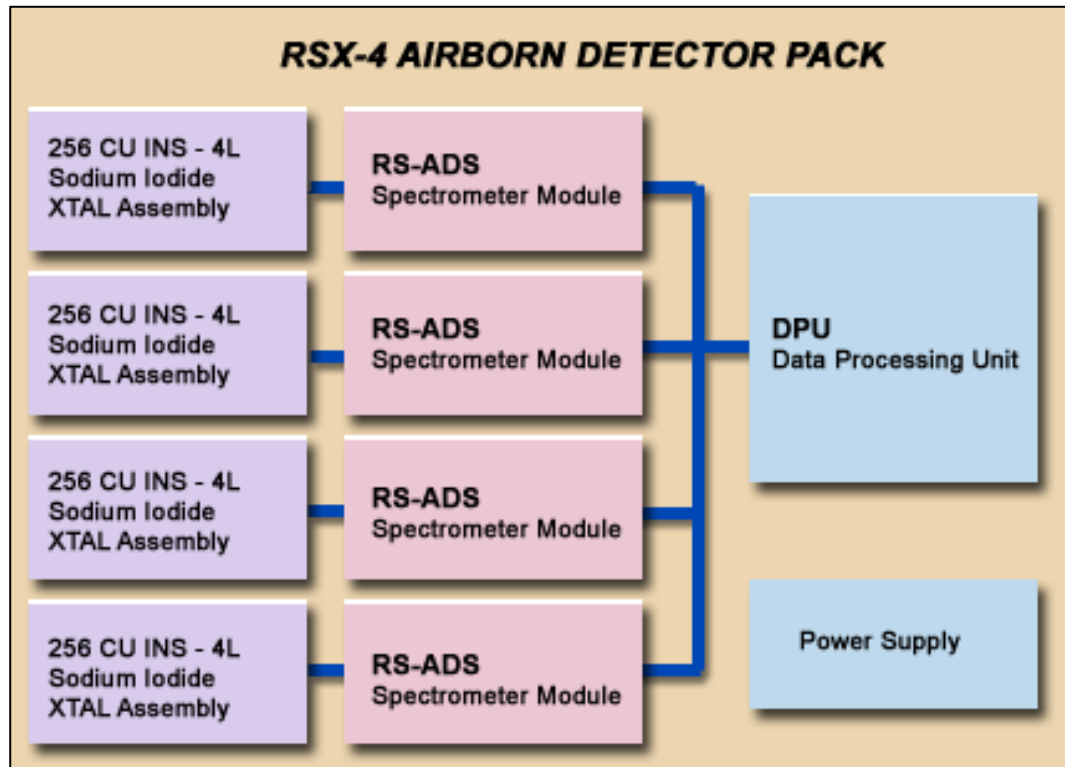


Figure 4-2 A generic "modern" detector system (picture courtesy of Radiation Solutions Inc.).

4.1.3. Flight platform

As the name suggests, there needs to be some sort of platform to carry the instrumentation above the survey area. Nowadays the variety of aircraft available is wide ranging from large fixed wing craft used in investigations like the Mineo project (MINEO 2000), to helicopters and micro-light aircraft, the type recently used by the Council for Geosciences here in South Africa (see Fig. 4.3).

These smaller types of aircraft have the advantage of being able to fly lower and slower than some other more traditional types of aircraft, thereby improving the sensitivity of the survey undertaken. The downside is that the pay load such craft can carry is smaller than larger craft such as the Air Tractor 502 'Data Boss' which can carry a detector volume of at least 66 l (see Fig. 4.4), whereas the Jabiru is limited to a detector volume of 4.2 l.



Figure 4-3 The airplane (Jabiru) used for the survey work (photo courtesy of Council for Geosciences), note the detector housing under slung and the electronics in the cockpit.



Figure 4-4 Air Tractor Data Boss 502 'Data Boss' with under-slung detectors

4.1.4. Data collection and analysis

The principle type of data collected is an energy spectrum of the detected gamma photons sorted into a specific number of analyser channels. These used to be presented in what were known as 'stacked profile plots.' Nowadays though, with the radical improvement in computer power since the 1960's, the results are generally presented in 3-D contour maps, or even ternary type maps which can be used to plot the relationship between the activity concentrations of uranium, thorium and potassium and give insight into the geology in a particular area.

4.2. Basic spectrum analysis process

Irrespective of whether one is using a ground-based or an aerial radiometric survey technique, the fundamentals are almost the same. The intention is to accumulate energy spectra of gamma rays emanating from the ground and then by using these basic data to produce a graphical or digital representation of the accumulated data. It is then possible to make a determination of the activity

concentration of a number of radioisotopes that occur in the decay chains of uranium and thorium. By way of an example, Fig. 4.5 shows a typical spectrum obtained during an airborne survey; on it are also indicated the ‘windows’ that are used to make the measurement of the activity concentrations. These windows listed in Table 4.1 are centred on the highest energy diagnostic photopeak for each of these three radioelements and conform to the standard that is generally accepted for such measurements. This is significant, since only the higher-energy photons are able to reach easily to airborne detector heights, and are also less susceptible to variations in source geometry or the height of the detector in comparison to lower energy photons [B. Minty 1997].

Table 4-1 Standard gamma ray energy windows recommended for natural radioelement mapping [IAEA 1999]. The energy of the photopeak being monitored is shown in brackets after each nuclide.

Window	Nuclide	Energy Range (MeV)
Total Count	-	0.4000 – 2.810
Potassium	⁴⁰ K (1.460 MeV)	1.370 – 1.570
Uranium	²¹⁴ Bi (1.765 MeV)	1.660 – 1.860
Thorium	²⁰⁸ Tl (2.614 MeV)	2.410 – 2.810

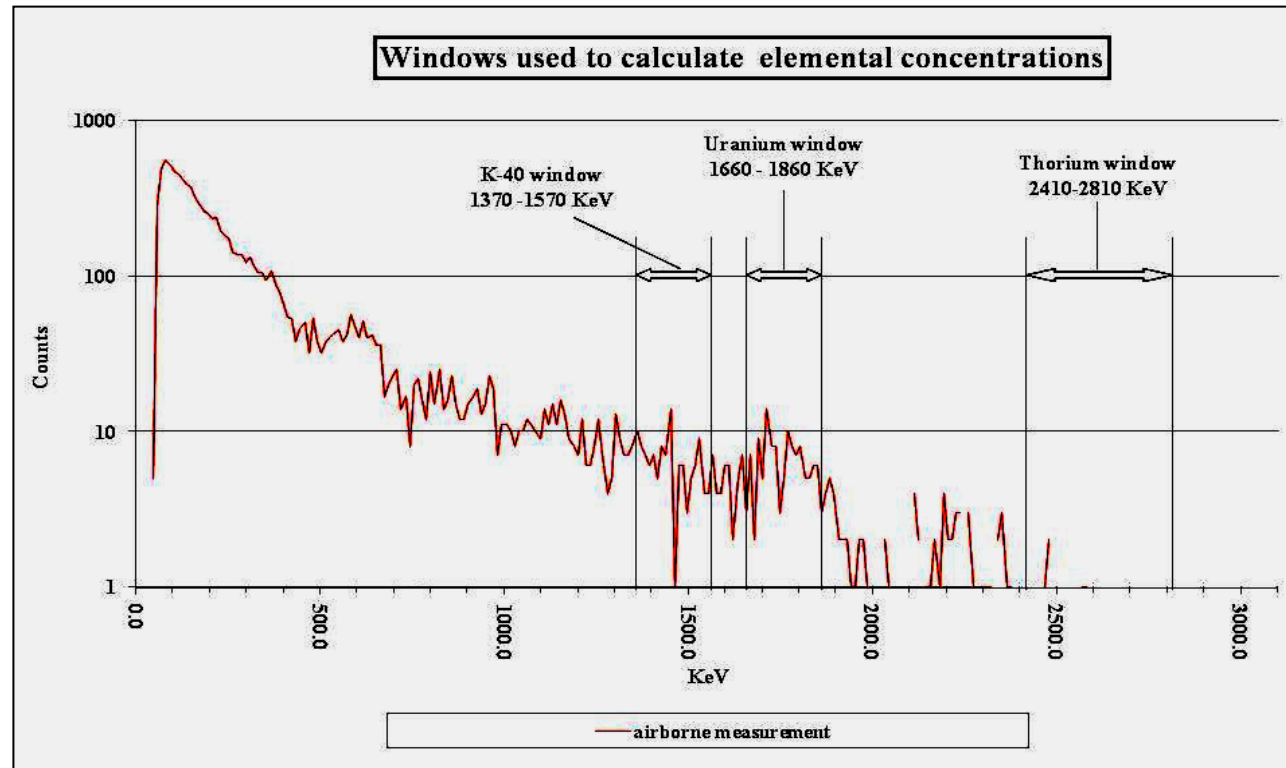


Figure 4-5 The “windows” used to calculate elemental concentrations. The spectra for K, U and Th overlap, and so it’s necessary to remove the influence of each of the isotopes in the other windows. Windows are superimposed on a typical spectrum obtained from one of the soil analyses done in this dissertation.

4.2.1. Measurement of gamma radiation

Modern gamma spectrometers can measure up to 1024 channels but more usually either 256 or 512 channels, generally in the range of approximately 0 – 3.00 MeV. As has been already mentioned in Section 4.2.1 energy resolution for NaI (Tl) based detectors is not particularly good, with values quoted of between 7 and 10 % (FWHM). This perhaps needs to be clarified. The precision with which a NaI (Tl) based detector can measure the energy of a gamma ray is known as the “energy resolution”. This is usually measured as the full width of a photopeak at half the maximum amplitude (FWHM) expressed as a percentage of the photopeak energy [IAEA 1991] and is shown diagrammatically in Fig. 4.6.

The broadening of the photopeaks comes as a result of Compton scattering (detailed earlier in section 2.4.1.2), in the earth carrying the radioactive material, the air layer between the earth and the detector, and within the detector itself (see also Fig. 2.16) [H. Coetzee 2009]. The small angle of Compton scattering brought about by the interaction of the gamma photons with the materials between the point of production and the point of detection will result in a mono-energetic gamma photon from the source producing a peak that is only very slightly less energetic than the original photon. This slightly less energetic photopeak will almost completely overlies the photopeak from an un-scattered photon, which will result in the broadening of the measured photopeak. Using the following formula it is possible to calculate what the effect of a single Compton Scattering event will have on the incident gamma photon energy that is measured by the detector. The energy of the scattered gamma ray photon $h\nu'$ in terms of its scattering angle θ is given by;

$$h\nu' = \frac{h\nu}{1 + \left(\frac{h\nu}{m_0c^2}\right)(1 - \cos \theta)} \quad \text{Eq. 4.1}$$

where m_0c^2 is the rest mass energy of the electron (0.511 MeV). The results of a single Compton scattering event are shown in Table 4.2 below. The results show both the effect of the energy of the incident gamma ray photon and the distance that the scattering event happened from the detector.

Table 4-2 Result of a single Compton scattering event

Distance from detector that Compton scattering occurred, m						
	1	10	20	30	40	50
⁴⁰ K <i>hν</i> MeV	1.462					
<i>hν'</i> MeV	1.2136	1.4587	1.4612	1.4616	1.4618	1.4619
<i>hν'</i> keV	1213.65	1458.67	1461.16	1461.63	1461.79	1461.87
²¹⁴ Bi <i>hν</i> MeV	1.764					
<i>hν'</i> MeV	1.4147	1.7591	1.7628	1.7635	1.7637	1.7638
<i>hν'</i> keV	1414.71	1759.15	1762.78	1763.46	1763.70	1763.81
²⁰⁸ Tl <i>hν</i> MeV	2.614					
<i>hν'</i> MeV	1.9138	2.6034	2.6113	2.6128	2.6133	2.6136
<i>hν'</i> keV	1913.79	2603.36	2611.33	2612.81	2613.33	2613.57

The conventional approach to the accumulation of airborne data particularly with the respect to making measurements of the activity concentrations of the uranium and thorium decay chains, has been to monitor three or four spectral windows as have been listed in Table 4.1. The count rates in the different spectral regions are used to infer the presence of radionuclides on the ground using a calibration procedure and also on the basis of certain assumptions. The higher-energy window (see Fig. 4.5) is mainly responsive to high-energy gamma rays of the 2614 keV peak from the isotope ²⁰⁸Tl in the ²³²Th decay series. It must, however, be corrected for cosmic ray background and small contributions from the uranium decay series. The central window is placed to capture the 1764 keV peak of ²¹⁴Bi as well as other peaks from ²¹⁴Bi in the same region.

The counts in this window are corrected for contributions from the background and the higher energy peaks in the thorium decay series using a set of stripping ratios. Finally, the window around 1462 keV is used to measure the ⁴⁰K contribution directly after corrections for the background as well as contributions from the thorium and uranium decay series. The stripping ratios needed to make the necessary corrections are found by the use of calibration pads [IAEA 2003, 47]. It should be remembered that this procedure measures, for example, ²¹⁴Bi on

the ground. This follows ^{226}Ra and ^{222}Rn in the ^{238}U decay chain. Thus, although airborne survey results are generally reported as an equivalent uranium activity concentration (eU) this is only true if the decay series is in secular equilibrium (this is usually not the case for TENORM). The same consideration applies to the determination of eTh while ^{40}K is of course determined by direct measurement. However, the ^{40}K peak has a relatively low intensity and the spectral errors in its determination can be large. The factors described above that affect all airborne radiometric surveys and their interpretation. How these factors are calculated and controlled will be addressed in the next section.

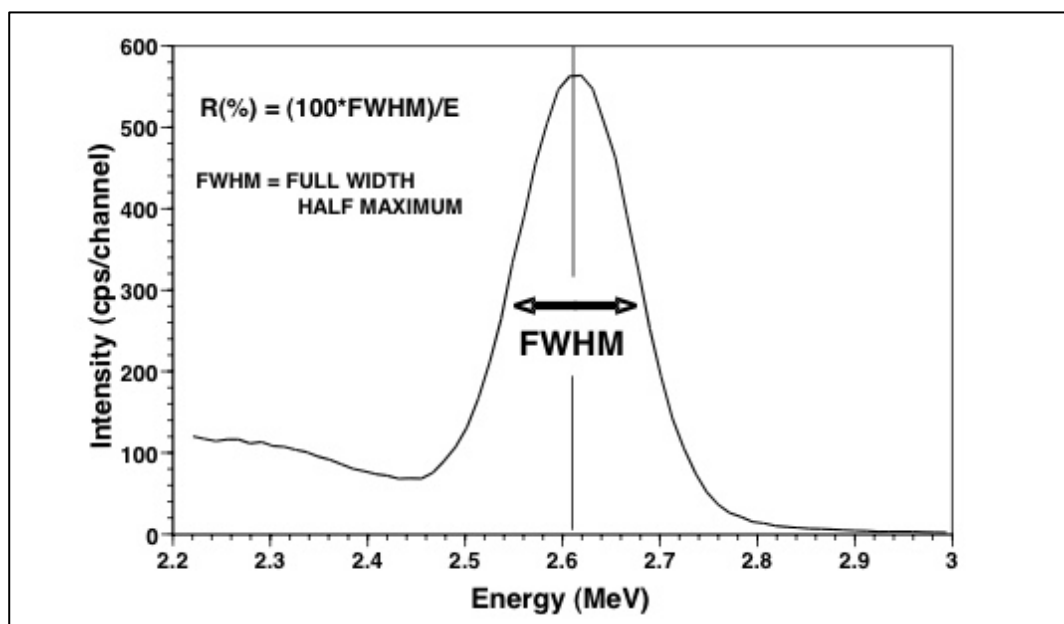


Figure 4-6 Diagram illustrating the energy resolution of a gamma ray spectrometer based on a NaI (Tl) detector [IAEA 1991].

4.3. Calibration of the instrumentation

In this section the calibration of a generic radiometric survey system will be discussed, the specifics of the calibration of the Council for Geosciences survey at the New Machavie site will be discussed in the next chapter.

4.3.1. Calibration data requirements

There are a number of corrections that need to be made to the raw data accumulated by an airborne survey before the values can be reported these include:

- Stripping correction,
- Background correction,
- Height corrections and
- Sensitivity corrections (conversion to elemental concentrations).

Each of these corrections is done empirically and the way they are done will be looked at in the following sections. Where these techniques vary from what has been done in the specific situation at the New Machavie site will be addressed in the following chapter.

4.3.1.1. Stripping correction

The calibration of gamma ray spectrometers is done so that a relationship between the measured instrument count rate and radionuclide concentration is established. This relationship is then used to calculate radioisotope activity concentrations in any given spectrum that has been accumulated by the detector system. The detailed process of the calculation of these stripping ratios is addressed in the IAEA's Tecdoc 1363 [IAEA 2003]. Examples of calibration pads that have been used in the past for aerial surveys in South Africa are the pads that existed at Lanseria Airport Johannesburg, Gauteng. These calibration pads (discussed later in Section 5.2), are unfortunately now lost to the scientific community as they were removed during the expansion of the airport facilities.

4.3.1.2. Background correction

There are several components to what can be considered 'background noise' or 'interference'. These include the following:

4.3.1.3. Noise and error propagation

Statistical noise is the greatest source of error in gamma ray spectrometry. The numbers of counts accumulated in any one channel over the usual count time of 1 s are generally very low, and this leads to large errors in the estimation of the activity of any isotope. However, using some standard techniques an estimation of the errors associated with the determination of the activity concentration can be made with a good deal of confidence in the result, but with the caveat and understanding of how the absolute errors do propagate [Dickson 2001].

4.3.1.4. Aircraft and cosmic background corrections

The contribution to any accumulated spectrum from the aircraft is constant. This can be recorded in a series of increasing altitude flights, generally carried out over water where the atmospheric conditions create a low radon concentration in the air. The result can then be subtracted from the overall spectrum as part of the developed spectrum analysis algorithm. The calibration process needed to take cognisance of these issues is detailed in the IAEA's document Tecdoc 1363 [IAEA 2003] but in concise terms consists of creating a normalized cosmic-ray spectrum together with the aircraft component, which is then used to amend a recorded spectrum. In southern Africa such flights are often carried out off the coast of southern Namibia. Included in these measurements are gamma rays with energy greater than 3 MeV, most often recorded in a separate channel of the detection system nowadays.

4.3.1.5. Radon background corrections

The process of correcting accumulated spectra for the effects of atmospheric radon is comprehensively described in Ref. [IAEA 2003]. Briefly though, there are three recognised techniques that have been developed over the years. These include the following; the spectral ratio method [B. Minty 1998], the full spectrum method and the upward-looking Detector Method [IAEA 1991].

4.3.1.6. Height correction

As the aircraft carries out the airborne survey, its height and thus the height of the detector above the ground varies and this needs to be allowed for and the recorded data then amended to a nominal survey height. The counts in each of the channels or 'windows' vary approximately exponentially depending upon height.

Corrections to the data can be made allowing for the variation in detector height, using the following equation:

$$n = n_o e^{-\mu(H-h)} \quad \text{Eq. 4.2}$$

where

μ = the window attenuation coefficient (m^{-1})

n_o = the observed count rate at the STP height, h , and

n = the corrected count rate for the nominal survey terrain clearance, H .

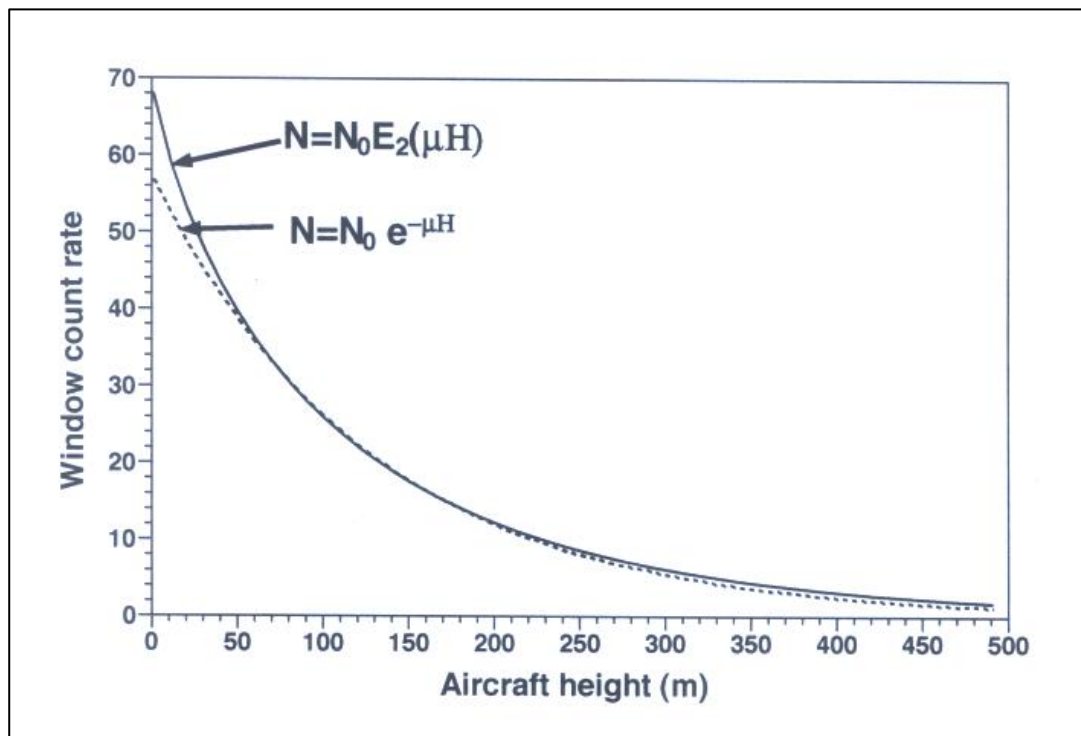


Figure 4-7 Exponential functions used to model the theoretical fall-off of gamma radiation with height. [IAEA 2003]

This algorithm is adequate for infinite sources and subdued topography with survey heights in the range 50 -250 m. In mountainous regions more accurate algorithms may be needed. The true fall-off of radiation with height for infinite sources more closely approximates that for an exponential integral rather than the simple exponential given in Eq. 4.2, though the difference between the two over the range of heights 5- 250 m is small. (See Fig. 4.7).

Equation 4.2 however must be corrected to STP, standard temperature (273.15 K) and pressure (101.325 kPa) because both of these parameters affect the density of the air that in turn affects the attenuating and shielding properties of air.

The following equation is used to make the correction to STP:

$$h_{\text{stp}} = \frac{273.15 \times P \times h_{\text{obs}}}{(T + 273.15) \times (101.325)} , \quad \text{Eq. 4.3}$$

where

h_{obs} = observed height above ground level (m)

h_{STP} = equivalent height at STP (m)

T = air temperature (°C)

P = barometric pressure (kPa).

4.4. The ‘footprint’ swept out by a survey

Modelling work on the footprint of a detector was originally carried out by Duval *et al.* [Duval, Cook and Adams 1971]. This was based on models using ‘circles of investigation’ and the area swept out by a moving detector. This model was refined by Grasty *et al.* [Grasty, Kosanke and Foote 1979] on realising that the concept of circles of investigation needed to be modified by the addition of factors that took into account the movement of the detector. Their work showed that the width of the strip that produces a fixed percentage of the detected radiation is significantly less than the diameter of the circle contributing the same percentage of the radiation detected by a stationary gamma-ray detector. Billings and

Hovgaard then further updated this model some twenty years later, with a model that also took account of the geometry of the detector volume. An extensive discussion on this problem can be found in [Billings and Hovgaard 1999].

In essence though, the footprint of a single measurement increases with flying height, whilst the count level decreases [Duval, Cook and Adams 1971] thus decreasing both spatial and spectral resolution. Airborne surveys then become a fine balancing act that on one hand has altitude of the flight and the concomitant safety risks of low flying, and on the other hand the loss of data and resolution with increasing height of the detector.

CHAPTER 5

AIRBORNE RADIOMETRIC SURVEYS IN SOUTH AFRICA

5. Introduction

Airborne surveys in South Africa have mirrored what has happened elsewhere in the world with regard to how and when airborne surveys have been carried out. Thus, surveys were originally carried out to search for uranium anomalies that could then be further surveyed by ground-based techniques [Cook 1952]. As the instrumentation and more particularly the computing and data storage abilities have dramatically improved, so survey techniques have gone from being a gross count process to multichannel techniques that are routinely used nowadays.

One of the particular features of the South African programme of aerial surveys was the fact that there were locally built calibration pads available to the scientific and minerals prospecting communities of Southern Africa. These were the pads constructed at Lanseria airport.

Another result of all the aerial radiometric-survey work was to allow the compilation of a national terrestrial dose rate map, which has applications in a number of diverse fields from radiation protection to minerals prospecting [Coetzee and Larkin, 2009]. Such information is useful in a radiation protection situation as, for example, it will allow an RP expert to determine whether the dose that has been recorded on a thermo-luminescent badge has come from some action an individual has taken, or that it has arisen as a result of the prevailing ambient dose rates in an area. In the case of minerals research, elevated doserates can indicate the presence of certain economically interesting mineral deposits.

This work was particularly useful because it then let the author try to ‘home-in’ on a smaller area for a comparative investigation of airborne and ground-based radiometric survey techniques.

5.1. National radiometric survey

The national radiometric survey map was compiled along with the national aeromagnetic survey map. It was carried out in two stages: the Karoo survey which was undertaken between 1976 and 1982 by Geosearch (Pty Ltd) with the remainder of the country being, largely done by various contractors over a period of 30 years starting in 1958 and finishing in 1997. Not all of the surveys were radiometric and magnetic, so there are still significant gaps in the radiometric data. Figure 5.1 shows the survey blocks as flown by the various contractors involved in the project.

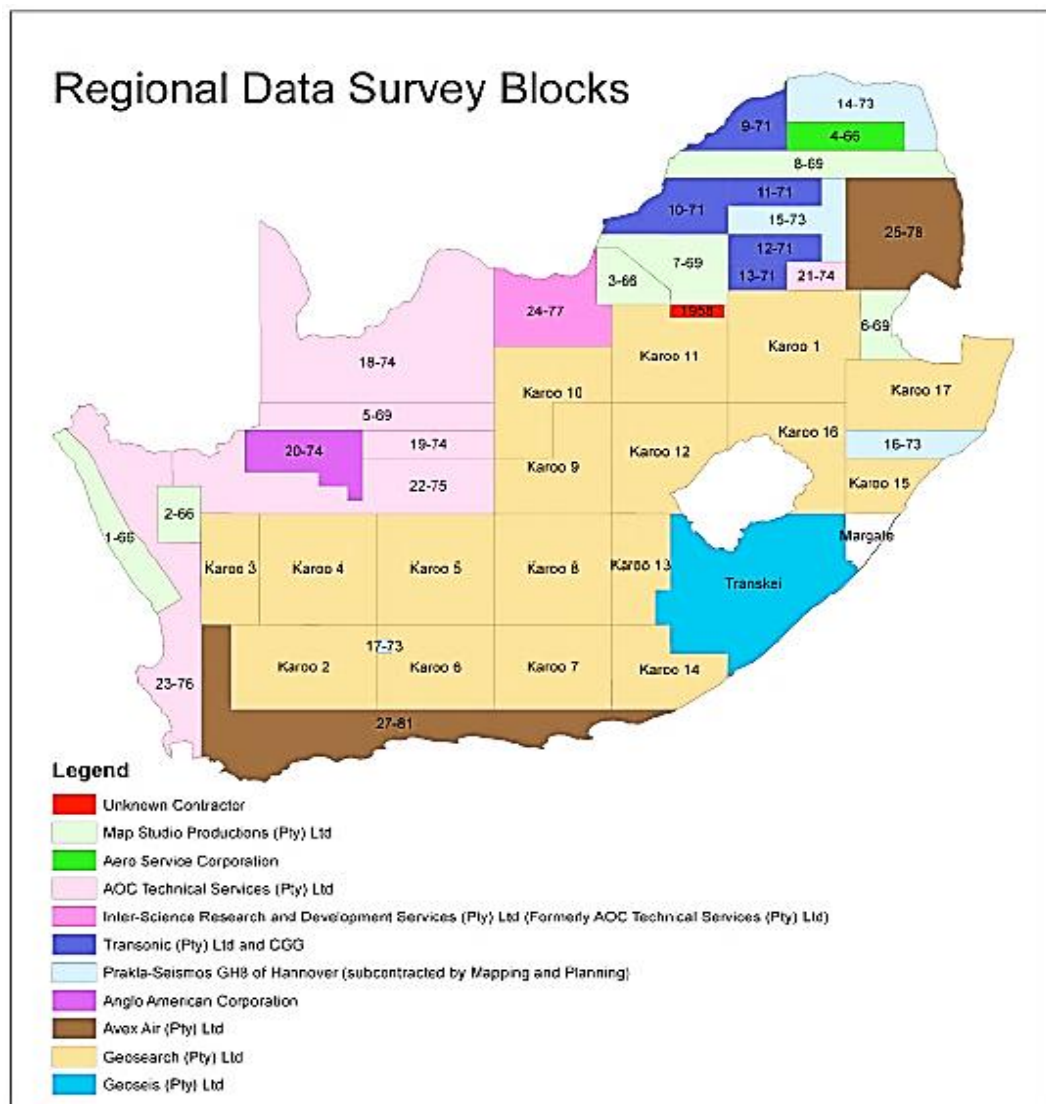


Figure 5-1 Regional survey blocks of the national radiometric/aeromagnetic survey [Ledwaba, *et al.* 2009].

5.2. Development of the calibration pads at Lanseria airport

The Lanseria airport calibration pads were constructed in early 1981 to cater for the needs of airborne and truck mounted multichannel spectrometry systems. Lanseria airport was chosen for the location as it was both close to Pelindaba, the then Nuclear Development Corporation of South Africa (Pty) Ltd, and Johannesburg where a number of private survey companies were based.

The pads consisted of four 8 m diameter by 0.35 m thick concrete structures sunk into the asphalt of a disused runway (see Fig. 5.2). Three of these pads were doped with known activity concentrations of potassium, uranium and thorium. The concrete in the pads was made from identical aggregates so as to try and remove extraneous sources of the above mentioned radioisotopes which can frequently be found in many such aggregates, particularly if they are sourced from gold mines which regularly sell the waste rock for inclusion into concrete and tarmac.

The dopants for the calibration pads were sourced from the following locations:

- **Uranium pad** - Ten tonnes of Bird Reef ore (Central Rand Group of the Witwatersrand Supergroup) was crushed and included in controlled aliquots into the cement. The activity concentrations were 200 ppm U_3O_8 and 20 ppm ThO_2 . A number of tests showed that the material was in secular equilibrium.
- **Thorium pad** - Monazite sand from the previously mentioned Steenkampskraal mine (Nama sequence) containing 10 000 ppm ThO_2 and 650 ppm U_3O_8 was used to dope this pad.
- **Potassium pad** - Pure microcline purchased from Ceramic Minerals Ltd was used for the potassium source. Tests showed radioelement concentrations of 12% K_2O and less than 1 ppm for both U_3O_8 and ThO_2 .

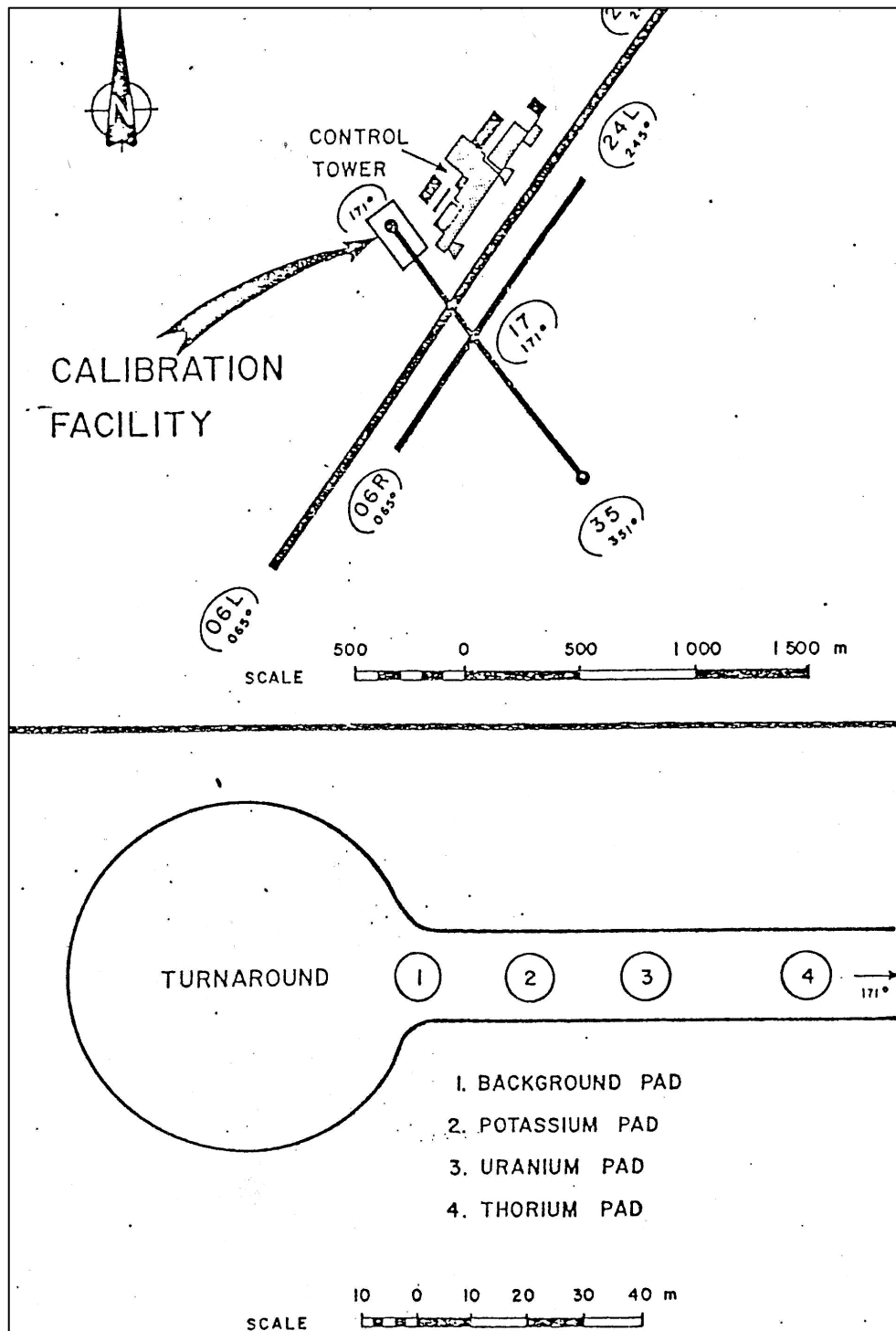


Figure 5-2 Layout of calibration pads at Lanseria airport (now removed) [Corner and Smit, 1983]

These dopants were incorporated into the concrete mix in a carefully controlled process ensuring to the greatest extent possible that the dopants were homogeneously distributed throughout the concrete pad, so as to try and prevent the development of ‘hotspots’ within a pad that could potentially degrade any subsequent calibrations, which were carried out using the site. The fourth pad, the background pad was constructed in exactly the same manner as the other three pads but was not doped with any specific isotope. During the pouring of the concrete, a large number of grab-samples were taken of the wet concrete that were then analysed to ensure the continued homogeneity of the concrete mix. After the concrete was set, and it was possible to walk on the pads, the pads were then again surveyed to confirm that there was no breakdown in the pouring/spreading process of the mix that had lead to a heterogeneous distribution of the dopants in each of the pads. Table 5.1 lists the final activity concentrations that are used in calibration calculations when using the Lanseria pads.

Table 5-1 Activity concentrations of the various calibration pads at Lanseria airport [Corner and Smit, 1983].

Source	K ₂ O (%)	U ₃ O ₈ (ppm)	ThO ₂ (ppm)
Background	0.21 +/-0.03	1.3 +/- 0.1	1.9 +/- 0.2
Potassium	6.1 +/- 0.2	0.9 +/- 0.2	1.9 +/- 0.2
Uranium	0.24 +/- 0.05	57.0 +/- 3.0	7.4 +/- 0.6
Uranium (radiometric assay)		67.0 +/- 4.0*	
Thorium	0.26 +/- 0.06	4.3 +/- 0.3	158 +/- 7

The mean uranium radiometric assay is reported separately in view of the fact that it falls outside the limits of the standard deviation quoted for the mean of the other laboratories. This value (67+/- 4 ppm U₃O₈ is recommended for all calibrations).

During the calibration process, airplanes would taxi down the runway as shown in Fig. 5.2 and would stop, in turn, over each calibration pad where a spectrum

would then be collected. These spectra would then be used to create the algorithm detailed previously in Section 4.4.1.1

Unfortunately due to a programme of expansion at Lanseria Airport that started in 2000 the site on which the pads were constructed was needed for aircraft hangers and despite efforts to relocate the pads to an alternative site at the airport this facility is no longer available to the scientific community as the thorium pad was irreparably damaged during the relocation exercise.

5.3. Construction of background terrestrial doserates map

The author and co-workers looked at trying to accumulate all the existing information of background radiation; this was originally done as part of a project for the National Nuclear Regulator of South Africa. One of the results was a database of South African airborne radiometric surveys (see [Ledwaba *et al.* 2009]) Using all the aerial survey work carried in South Africa has led to the opportunity to develop a radiological dose rate map of South Africa, [Larkin *et al.* 2004]. A compilation of the surveys that were included into the overall radiometric data used to construct the terrestrial doserate map is given:

Table 5-2 Surveys used to construct terrestrial dose-rate map of South Africa.
[Ledwaba, *et al.* 2009]

Survey companies	Area surveyed - see Fig. 5.1	Date	Type of detector
Map Studio Productions (Pty) Ltd	Blocks 1/66 & 2/66	1966	3 NaI (Tl) crystals each 15 cm diameter by 10 cm deep, with a total volume of 5.3 l.
	Blocks 6/69, 7/69 & 8/69	1976	
Aero Service Corporation, Philadelphia, USA	4/66	1966	3 NaI (Tl) crystals each 15 cm diameter by 10 cm deep, with a total volume of 5.3 l.
Joint venture between Map Studio and Aero service	3/66	1966	
A.O.C Technical services (Pty) Ltd	Block 5/69	1970	Exploranium Spectrometer fitted with a pair of 6 by 4 inch NaI (Tl) crystals.
	Blocks 18/74, 19/74, 21/74 & 22/75	1976	An exploranium DiGRS 3001 4 channel differential gamma ray spectrometer with nine NaI (Tl) detecting crystals each 15.2 cm in diameter and 10.2 cm thick giving a total crystal volume of 16.68 l.
	Block 23/76	1977	An Exploranium DiGRS 3001 four channel differential gamma ray spectrometer with nine NaI (Tl) detecting crystals each 15.2 cm in diameter and 10.2 cm thick giving a total crystal volume of 16.68 l was used.
	Block 24/77	1977	An Exploranium DiGRS 3001 four channel differential gamma ray spectrometer with nine NaI (Tl) detecting crystals each 15.2 cm in diameter and 10.2 cm thick giving a total crystal volume of 16,68 l was used.

Survey companies	Area surveyed - see Fig. 5.1	Date	Type of detector
Transonic (Pty) Ltd	Blocks 9/71, 10/71, 11/71, 12/71 & 13/71	1971/2	A CRC 4 channel gamma ray spectrometer and 3 NaI(Tl) crystals, each 20 cm in diameter by 10 cm long were used
Planning and Mapping Pty (Ltd) (PRAKLA-SEISMOS GMBH)	Blocks 14/73, 15/73, 16/73 & 17/73	1974	The gamma spectrometer had 1 detector unit consisting of 5 NaI (Tl) crystals of 6-inch diameters and 4-inch heights with a total crystal volume of 565 cubic inches.
Anglo American Corporation	20/74	1972	A Geometrics GR 800 multichannel spectrometer with GAX-1000 extended square array crystal pack with a volume of 16.78 l.
Geosearch (Pty) Ltd	Karoo sequence	1976	A Geometrics GR 800 multichannel spectrometer with GAX-1000 extended square array crystal pack with a volume of 16.78 l.
Avex Air (Pty) Ltd	25/78		McPhar Spectra II spectrometer and a slab sensor of 16.4 l crystal volume.
Geoseis (Pty) Ltd	Transkei and Margate Blocks	1995 - 1997	Geometrics GR 820 with 2 crystals (total of 16 l).

5.3.1. Calibration of airborne radiometric databases

Before the terrestrial doserate map could be constructed it was necessary to calculate *in situ* doserates for much of South Africa because at the time this was not known. What was known from work done by the Council for Geoscience was that a dataset of *in situ* activity concentrations for ^{40}K , eTh (as ThO_2) and eU (as U_3O_8) existed, since these had been measured on a road trip around South Africa. (see Fig. 5.3 for the points at which measurements were taken).

These measurements were taken at 10 km intervals, 50 -100 m off the road using a portable Geofyzika-Brno GS 265 MCA with a 3" NaI (Tl) crystal. The measurements were recorded as counts s^{-1} and were subsequently converted into Bq kg^{-1} values using a calibration derived from pads at Lanseria.

Using these known activity concentrations it was then possible to calculate the doserates that each of these isotopes would contribute to the overall absorbed dose and plot this against a known total count rate as measured by airborne survey that had been measured at the same point that the ground activity measurements had been taken. Figure 5.4 is the graphical representation of these results.

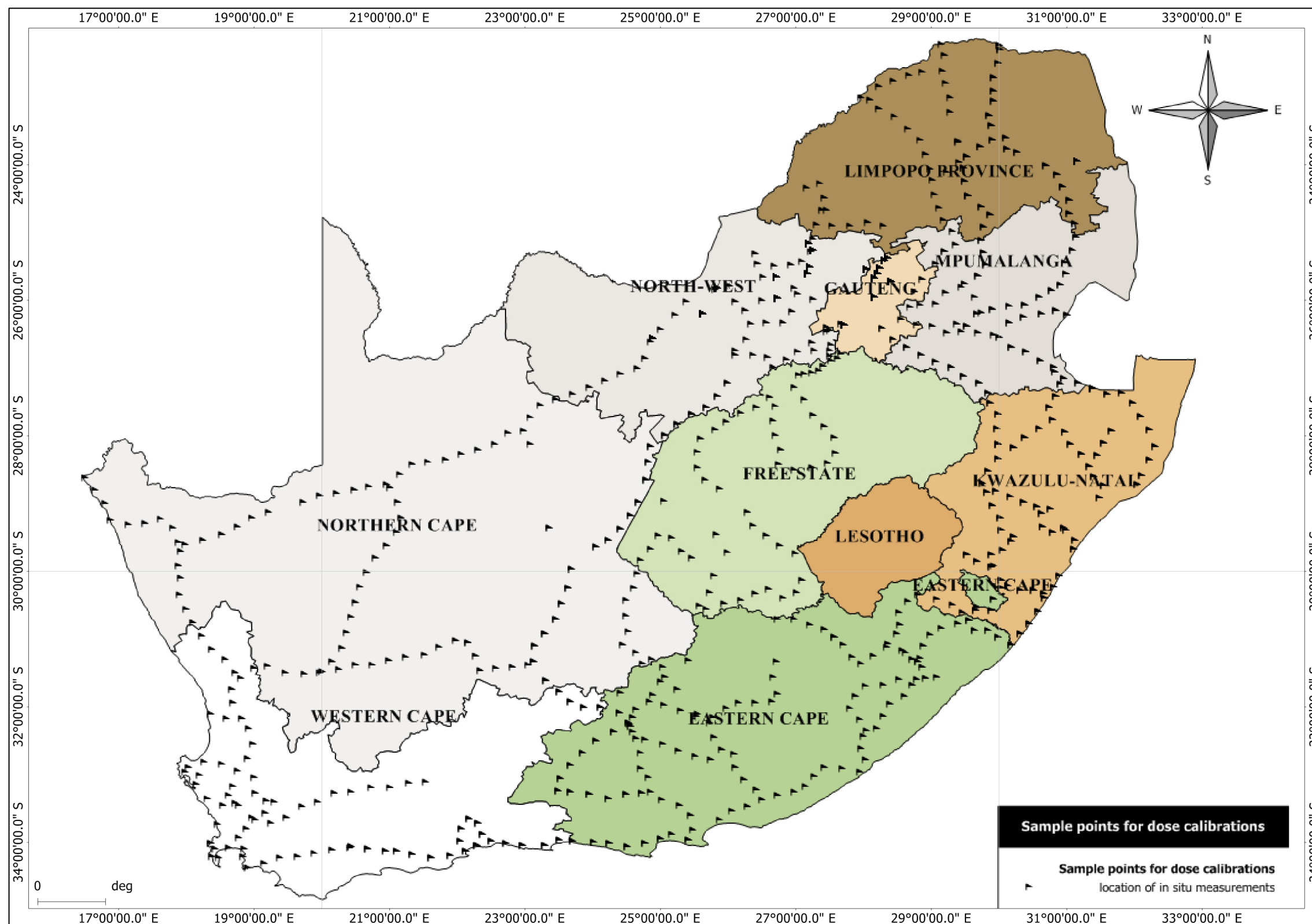


Figure 5-3 Location of soil sample points used to produce calibration (see Section 5.4.1).

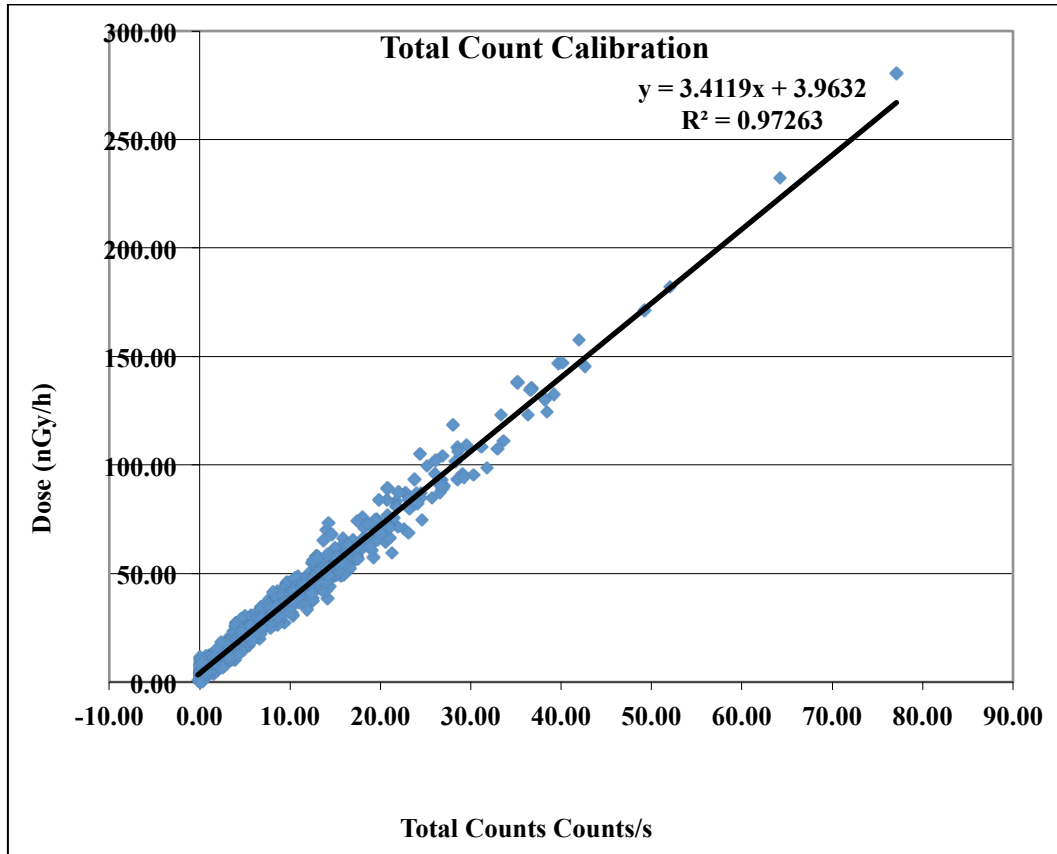


Figure 5-4 Total count calibration used to create terrestrial doserate map.

Using the conversion factor determined from Fig. 5.4 it was then possible to calculate an *in situ* dose rate from total count results that had been accumulated over the many years of airborne surveys. This is a very useful advance in the process of evaluation of the ambient dose to the population and the control of that exposure. The next stage in the process of the map construction was to include a correction to take account of the cosmic ray background that always exists; this was calculated based on altitude measurements gained from topological information. The cosmic ray dose rates in nanogreys/hr (nGyh⁻¹) were based on the following relationship from Ref. [UNSCEAR 2001]. Following the reference,

$$E_1(Z) = E_1(0)[0.21e^{-1.694Z} + 0.79e^{0.4528Z}], \quad \text{Eq. 5.1}$$

where $E_1(0)$ is the dose rate at sea-level and, Z is the altitude, (km). Once the total cosmic ray doserate had been calculated, it could be combined with the calculated *in situ* values to produce a map such as can be seen in Fig. 5.5.

The author plotted the maps using a GIS programme called Manifold version 8.0, [Manifold Software Limited 2011] (unless specifically mentioned otherwise). Manifold version 8.0 has extensive plotting abilities of all of the various geographical files generally in use. In addition, the programme has extensive database manipulation abilities, which has allowed the production of the various different contour and doserate maps included in this dissertation.

5.4. Doserate map of South Africa

After the summing of the contribution to the dose rates of the directly ionizing and the component of indirect component cosmic radiation, and the doserates calculated from the airborne radiometric surveys it is then possible to develop a terrestrial doserate map of South Africa (see Fig. 5.5.). The values are presented as an absorbed dose as it makes no assumptions as to the effects on the human body.

There are a number of white areas on the map because there were no available airborne radiometric data at the time that it was compiled either because no survey had ever been carried out, or if a survey had been carried out, it was done on behalf of a client who wished to keep the findings confidential. This is often done in the case of mineral exploration since the findings of a survey can have profound effects on the viability of a mineral mining company.

On closer examination, the results of the map reveal a number of areas within the country that have significantly elevated terrestrial doserates. Two such areas have been previously mentioned *viz.* Steenkampskraal thorium and rare earth mine in the Northern Cape and the gold mining area of the West Rand, Gauteng. There are many other areas but these are well documented and show the use of the overall map.

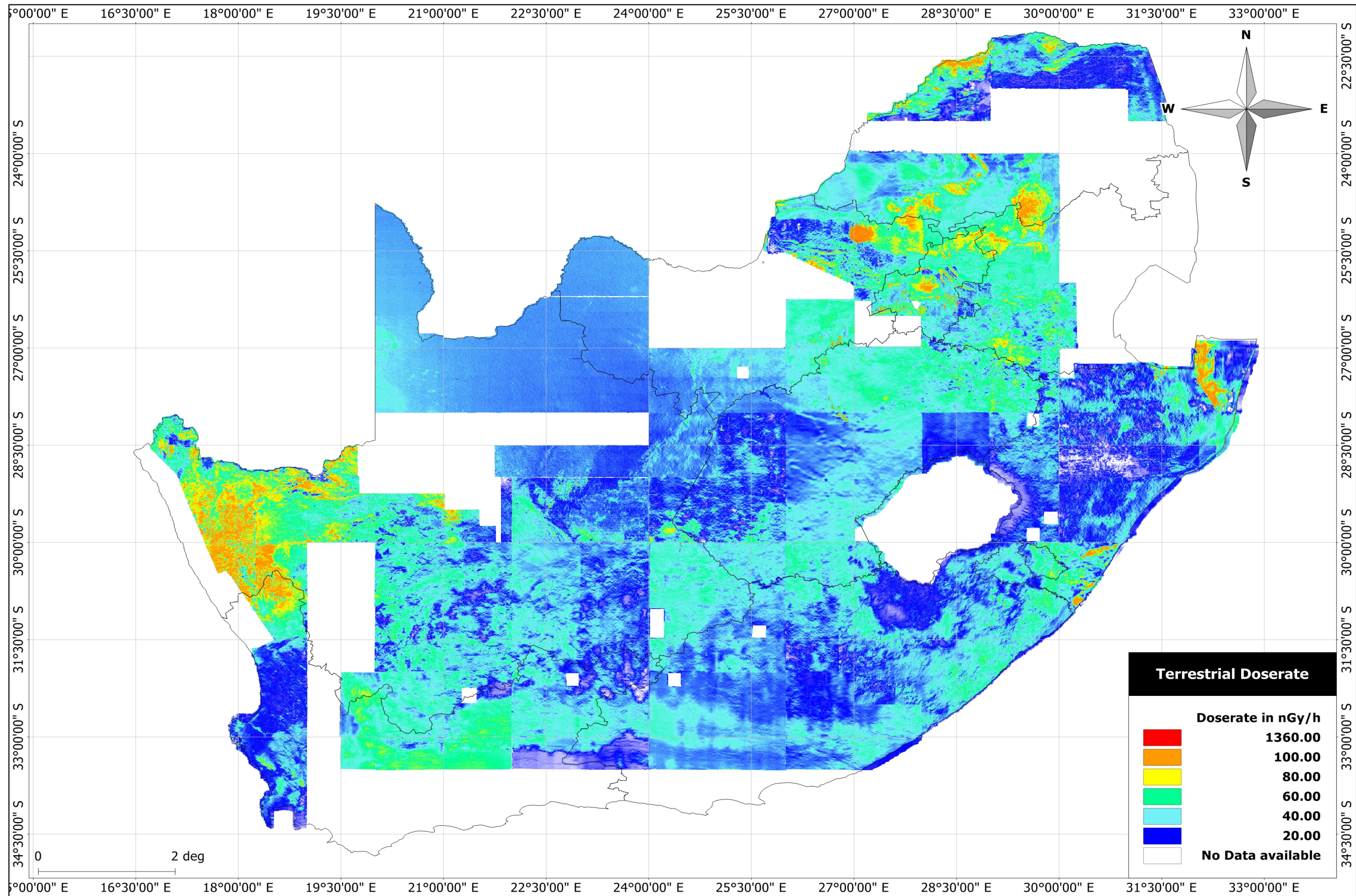


Figure 5-5 Terrestrial background doserates of South Africa [Larkin *et al.* 2004].

5.4.1. Steenkampskraal thorium and rare earth mine

There are a large numbers of areas around the mine site that show elevated doserates up to 1 mGyh^{-1} . Two particular reasons for this can be identified; firstly, the prevailing geology of the area is rich in thorium-bearing minerals and many of these are very close to the surface (Fig.5.6 shows a photograph of the mine environs). These areas are to the west of the mine. The second reason is as a result of the past mining activities at the mine; from Fig. 5.7 it can be seen that there are large areas of elevated doserates all around the working areas of the mine, probably as a result of the spread of radioactive dust produced when the minerals were extracted [Great Western Minerals Group Limited 2010]. These areas are largely to the north and a lesser extent the south of the mineshaft. This is shown when the doserate map is over laid onto a satellite image (see Fig. 5.8). Given the topography of the site there is a strong suggestion that this radioactive mineral dust has been spread away from the site by the action of rainwater runoff, which has allowed patches of material to accumulate downhill from the excavation point.



Figure 5-6 Environs of the mine at Steenkampskraal [Great Western Minerals Group Ltd. 2010].

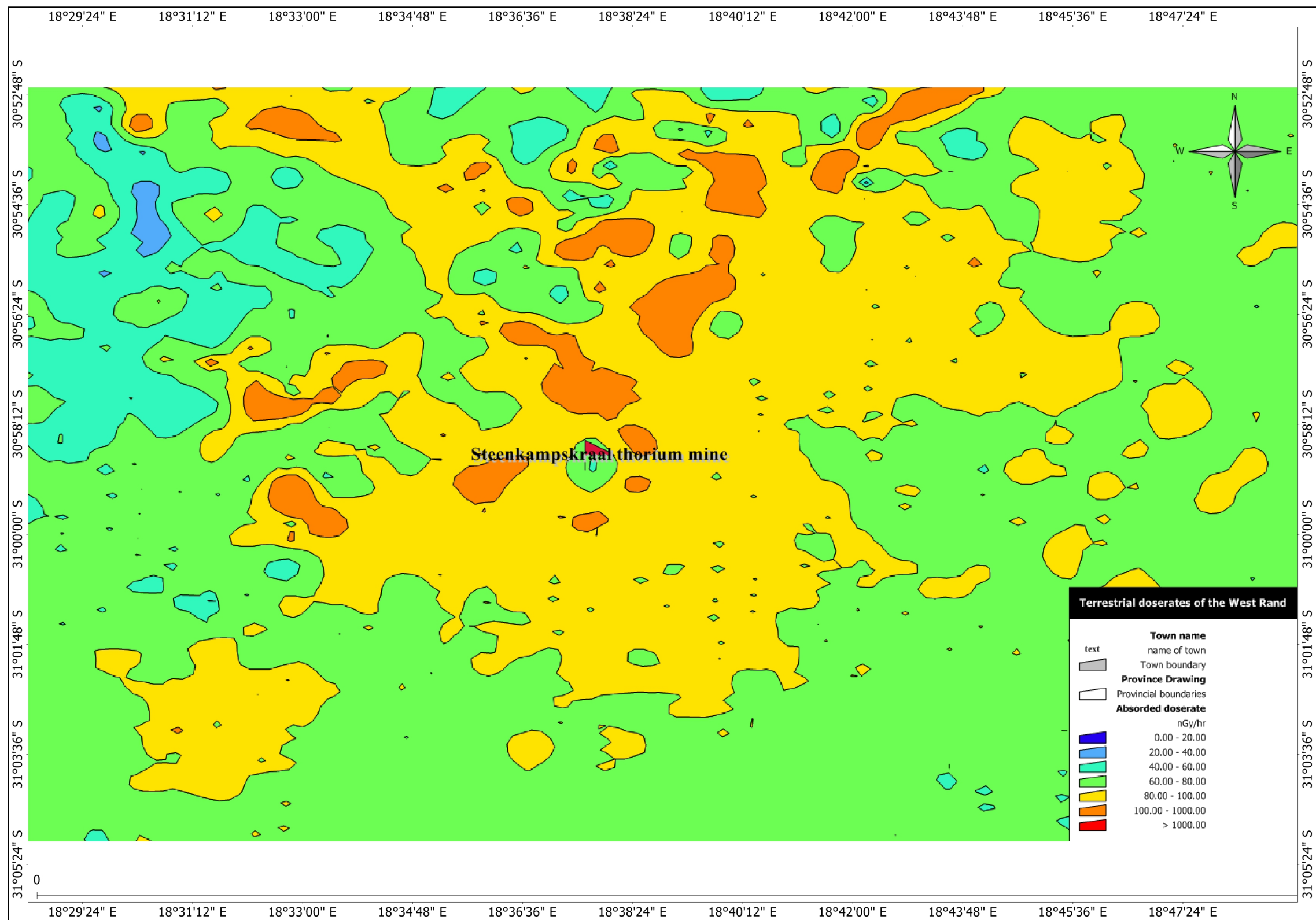


Figure 5-7 Steenkampskraal thorium and rare earth mine, Northern Cape, South Africa.

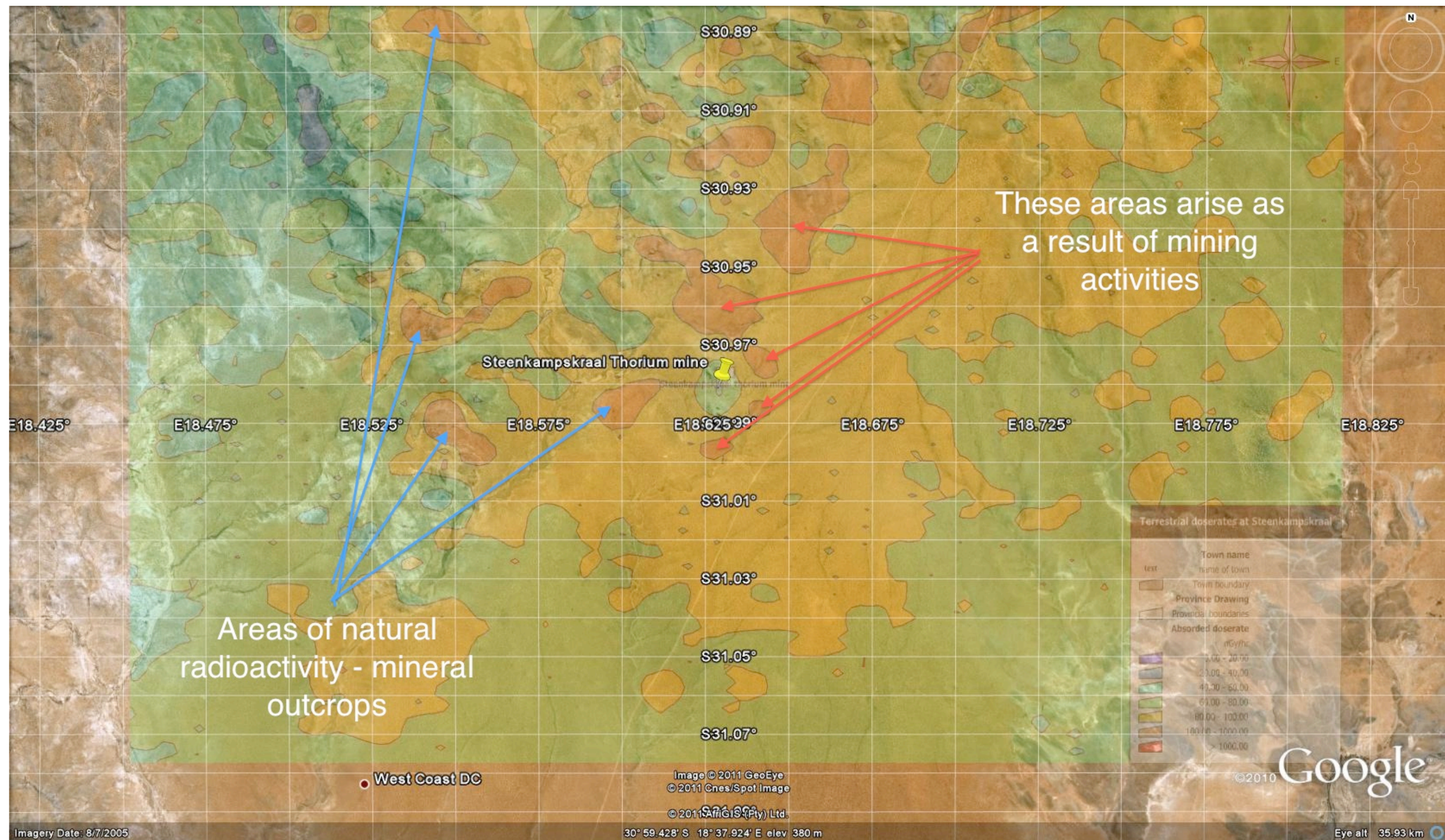


Figure 5-8 Overlay of terrestrial dose rate on Google Earth image at the Steenkampskraal mine.

5.4.2. The West Rand Gauteng

In the West Rand there are a large number of areas that show elevated terrestrial doserates. This is as a result of the gold mining activity that has gone on in the region for the last 150 years. All the elevated areas coincide with the slimes dams that have been constructed in the area. These slimes dams contain significant quantities of the isotopes of the uranium decay series (refer to Section 3.4.4). Figures 5.9 and Fig. 5.10 shows how the elevated terrestrial doserates in the West Rand coincide with where the slimes dams are seen on the satellite image.

5.5. Selection of an area for study

Using the results of the construction of a terrestrial doserate map of South Africa it was possible to refine the search for a suitable radiometric anomaly that was amenable to further investigation. In discussion with Dr Henk Coetzee, a choice was made to look at just such an anomaly at the New Machavie Gold mine. The Council for Geosciences chose this site since there had just recently been a high-resolution survey of the area and who were prepared to make the data set available. Dr Coetzee had done the work as part of an investigation into the use of ultra-light aircraft as an instrument platform [H. Coetzee, Acquisition, processing and enhancement of multichannel radioelement data collected with ultra-light aircraft mounted detectors 2008]. There was at the time ready vehicular access to the site that made the task of taking *in situ* measurements and soil samples considerably easier.

5.6. The New Machavie Gold Mine

The survey site lies immediately adjacent to the abandoned gold mine known as New Machavie the location of which is shown in Fig. 5.11. This site was chosen for a number of reasons, including those mentioned above. In addition, the site was in reasonably close reach from the University meaning a day's work could be fitted in between travel to and from the site. There were a number of clearly measureable radioactive anomalies on the site but the background levels were reasonably low so as to not swamp the instrumentation with excessive levels of radiation.

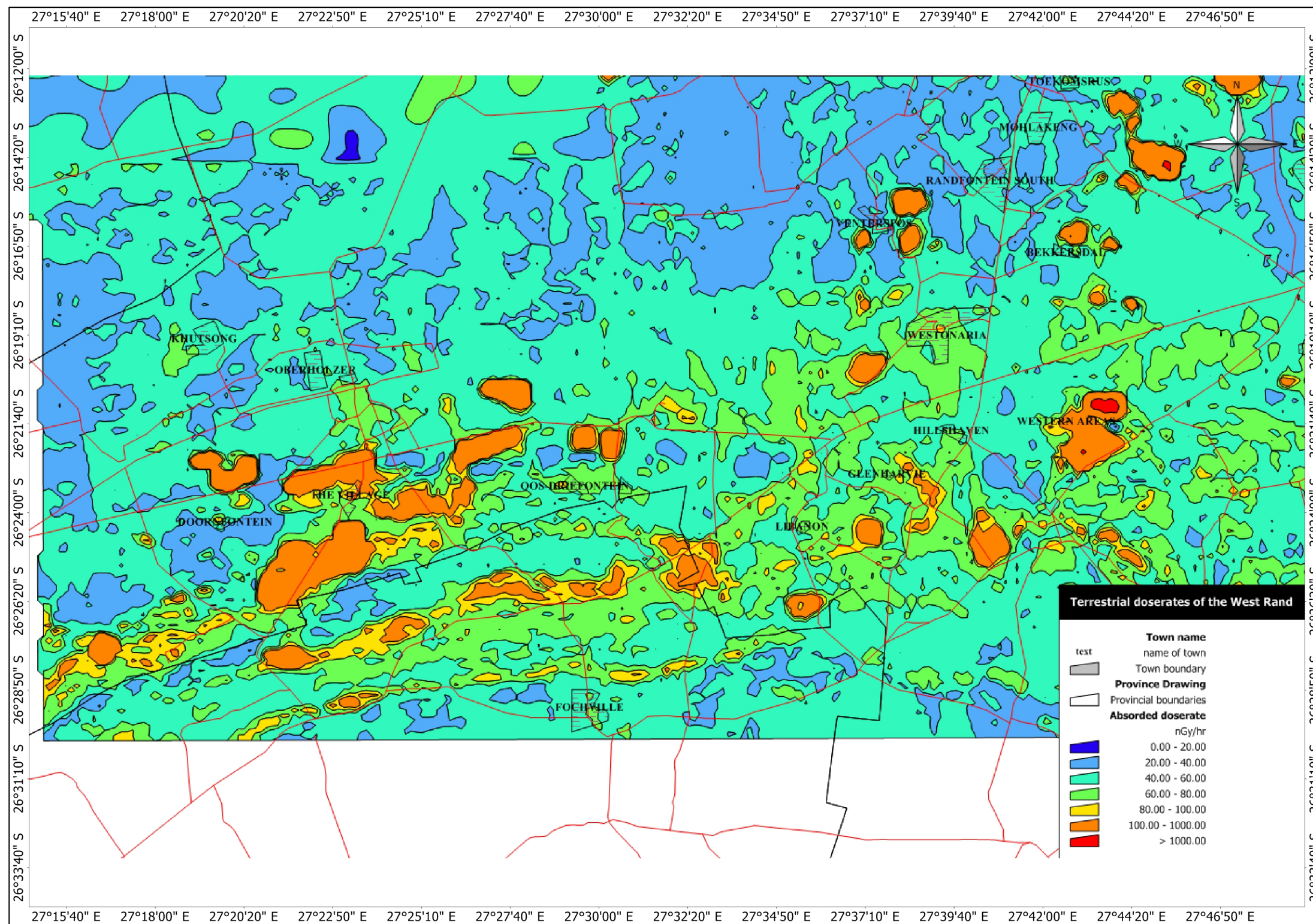


Figure 5-9 West Rand, North West Province, South Africa, showing a large number of regions with doserates greater than 100 nGy h⁻¹ and one region close to Westonaria with a doserate > 1000 nGyh⁻¹ (compared to ambient background)

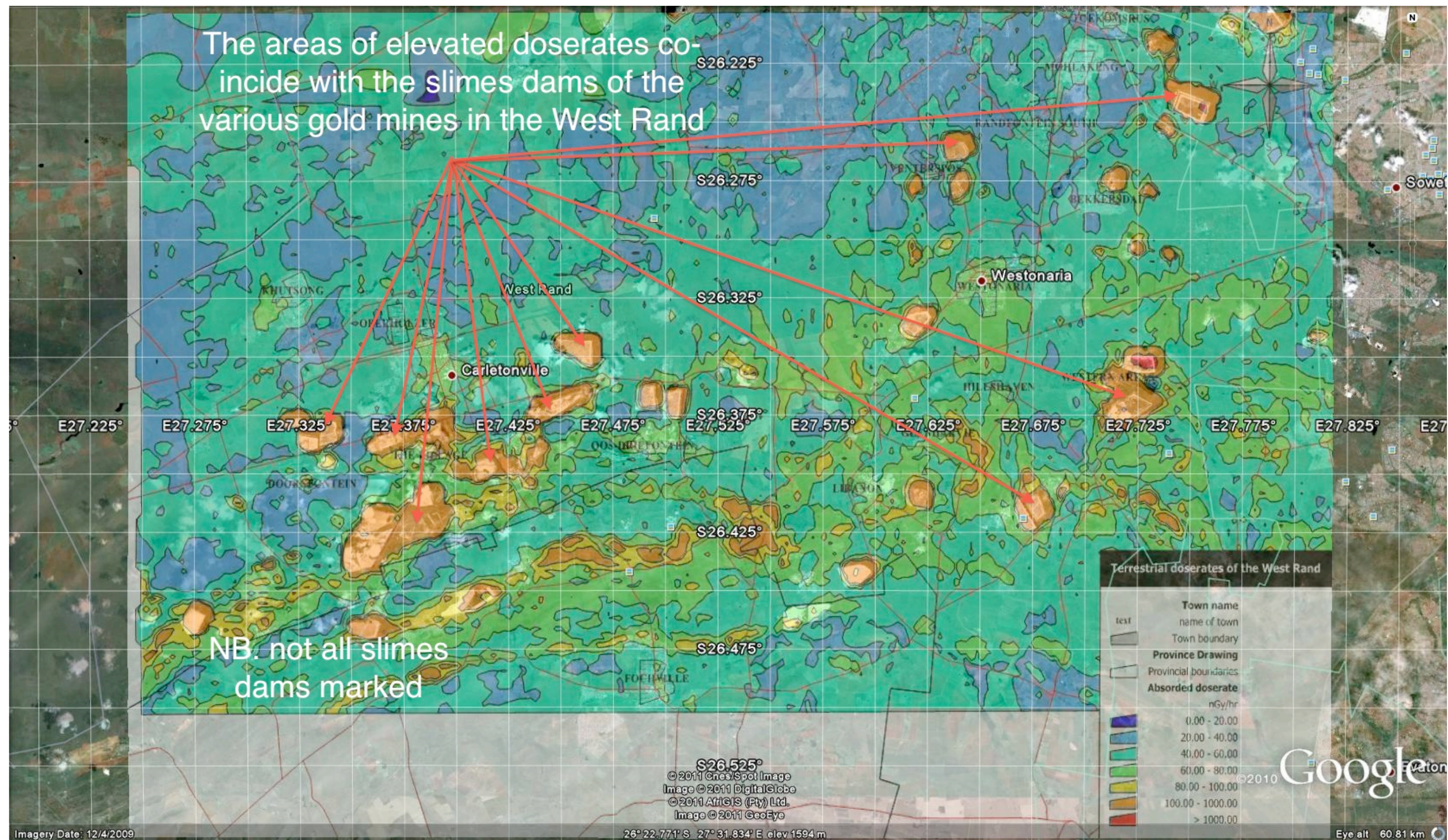


Figure 5-10 Terrestrial doserates overlaid onto a satellite photograph of the West Rand.

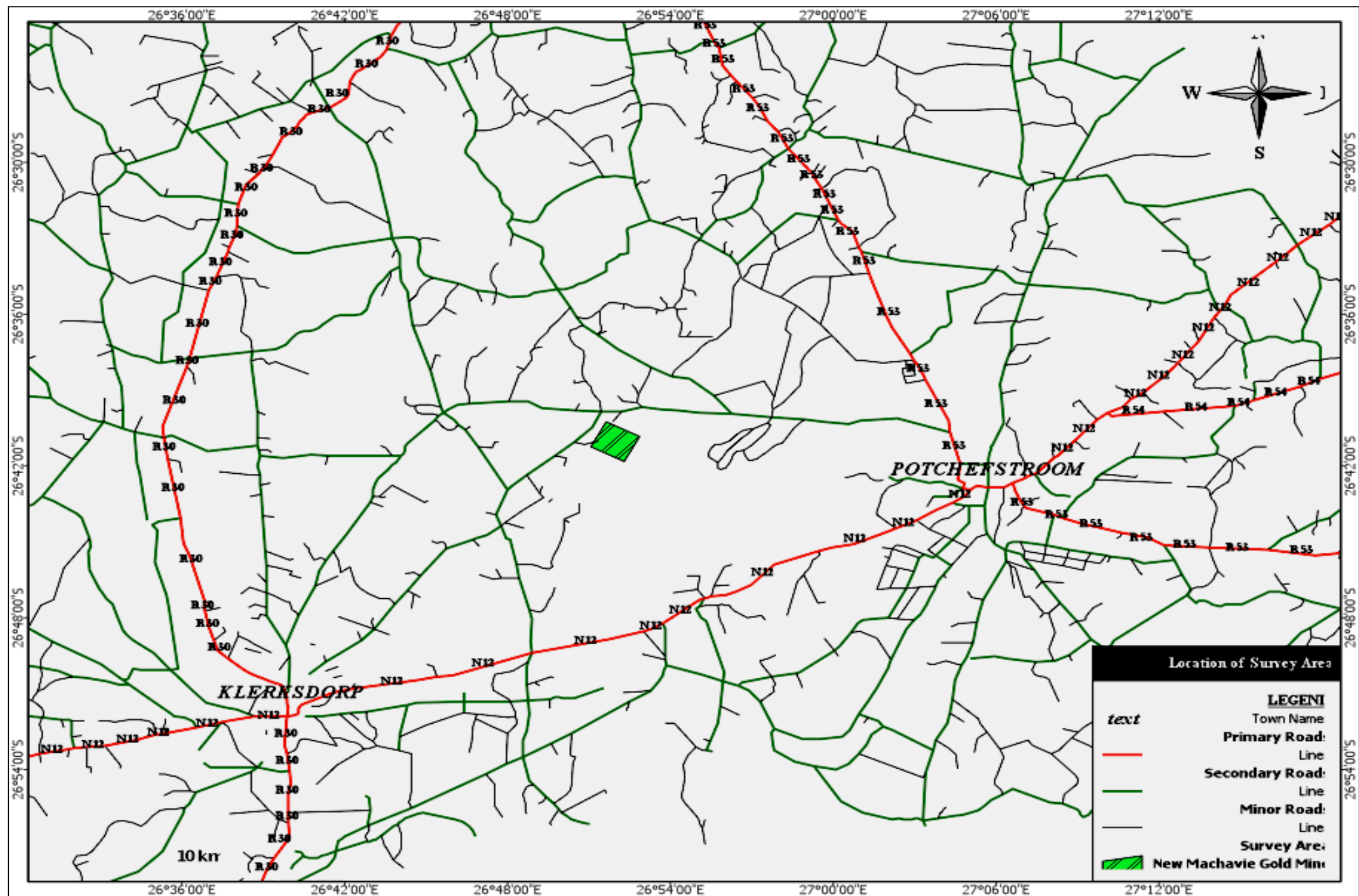


Figure 5-11 Location of the New Machavie mine.

5.6.1. Site description

The site is farmland used for grazing cattle (see Fig. 5.12 and 5.13). There had been no intensive agriculture carried out on the land and there was no evidence of any ‘pasture improvement’. As such, the land has not been ploughed which could easily upset the secular equilibrium that exists in the decay chains of both uranium and thorium. In addition, there has not been any spreading of phosphate based artificial fertiliser or sewage sludge – all potential sources of radioactivity. The mine has a tailings dam, (see Fig. 5.14), waste rock dump and a known pollution plume, contaminated with uranium-enriched material (see Fig. 5.15). The ore body mined at this mine was in the Black Reef Formation, which is not part of the Witwatersrand Supergroup [H. Coetzee 2008]. This formation has however been reported as containing elevated uranium concentrations. The underlying geology is of dolomite, conglomerate and quartzite.

The topography of the site was relatively simple with a slight elevation of approximately 30 m at the western end of the survey area. Running from northeast and heading south, southwest there is a watercourse, which only runs in the rainy part of the year. There is, however, evidence of run-off from the slimes dams that is outside of the survey area (see Fig. 5.15). It is highly likely that there has been some migration of the slimes and associated radioactivity that is slowly moving down the watercourse and moving elevated levels of radioactivity into the survey area. From the terrestrial doserate map, it can be seen there is a radiometric anomaly within the survey area, but not of a magnitude that it would overload the detector electronics increasing the dead-time to a point that the detector system would become virtually inoperable.



Figure 5-12 New Machavie survey area



Figure 5-13 New Machavie aerial survey area. Note detector on tailgate of pickup



Figure 5-14 Slimes dam at New Machavie gold mine.

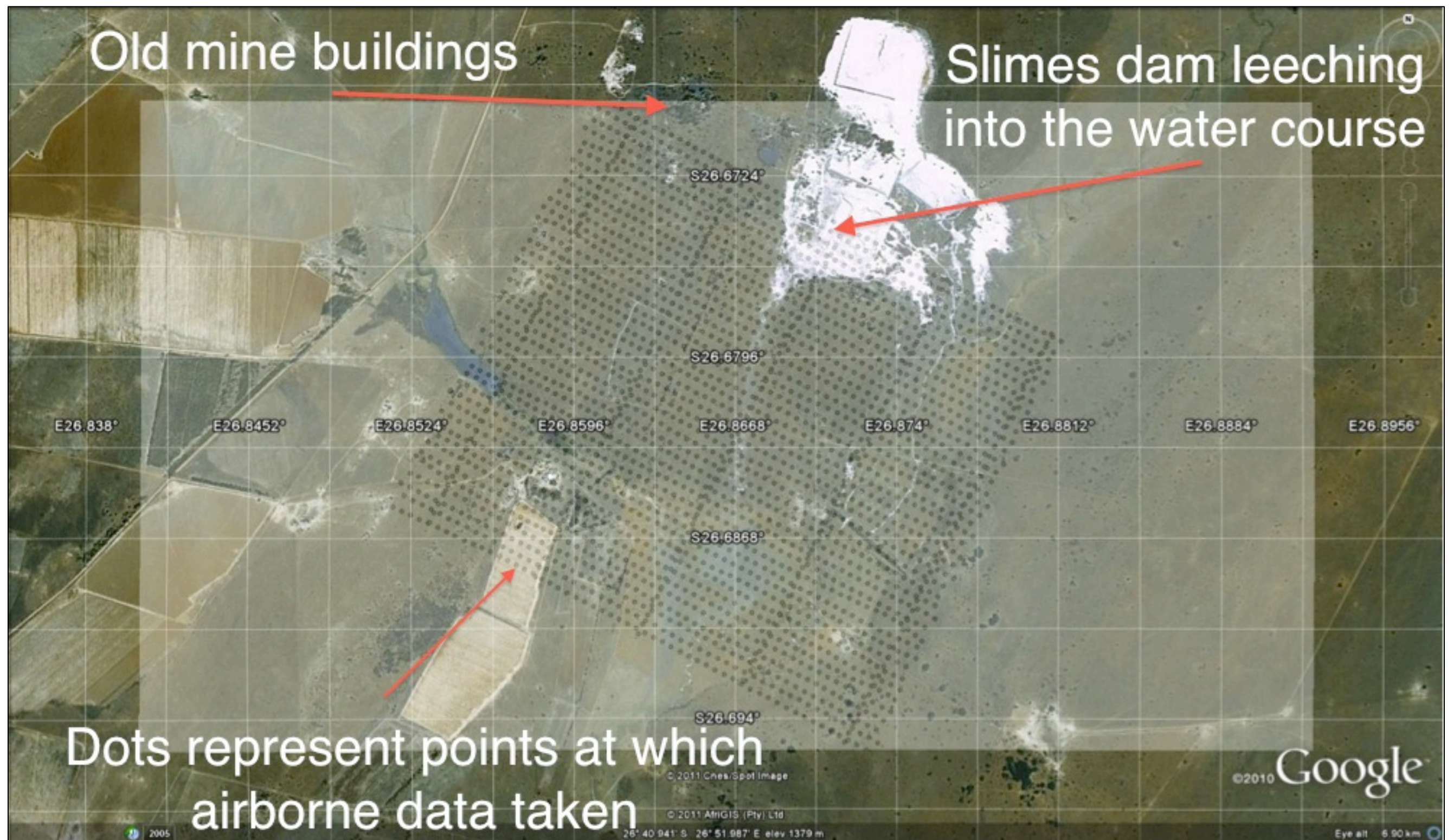


Figure 5-15 Survey area at the New Machavie mine. Note the white slimes material moving onto the survey from a northerly direction.

5.7. High resolution airborne survey of the New Machavie Gold Mine flown by the Council for Geosciences

In July 2002 the Council for Geosciences flew a high-resolution airborne survey over the area known as the New Machavie gold mine. This one was done using one of the Council's ultra-light aircraft, specifically the Jabiru (see Fig. 4.3). This was equipped with a 4.2 l (10.2 cm x 10.2 cm x 40.8 cm) Na (Tl) crystal coupled to a 3.5" photomultiplier tube in a 'ruggedized' system provided by Geotron Systems of Potchefstroom, South Africa. A laser altimeter (Riedl Laser Altimeter) was used to record height above the ground. Nominally, this was 50 m. A differential GPS (Staloc Airstar GPS) was used to plot the location of the aircraft. The survey data was accumulated by a PC 104 bus computer that made use of solid-state memory, since conventional hard drives are particularly susceptible to the vibrations that exist in a plane during flight. In addition to the radiometric data recorded, total magnetic-field intensity was recorded as well.

5.8. Methodology used for the airborne radiometric survey of the New Machavie survey site

Because the New Machavie site was surveyed using the Jabiru ultra-light aircraft, there are a number of issues associated with the data set that might not have normally arisen had the survey been flown with a more conventional setup. Mostly the issues are to do with the airplane's operational parameters including range, flight ceiling and lift capacity. Firstly range: the plane could not travel to the one area in Southern Africa, Henties Bay in Namibia where it is possible to fly over a homogeneous calibration range with known ground activity concentrations [Coetzee 2008]. Such a calibration can then be used to derive an algorithm, which can be back fitted to the data that then modifies recorded values in light of the variations in flight height above the ground and the overall height above sea level. This was resolved by the development of an empirical method using the laser altimeter to record height above ground and known altitude above sea level to modify the recorded results.

The next issue peculiar to the ultra-light aircraft is its inability to fly at altitudes sufficiently high enough to be able to eliminate ground effects from measurements used to determine background and radon influences on recorded data, especially when the survey work is being done at the altitude prevalent on the South African Highveld plateau. To overcome this problem background corrections were applied using survey data that were accumulated from areas of a survey known not to be geologically radioactive and parts of surveys that were flown over inland water-ways (ultra-light aircraft not generally being permitted to fly over the sea at all). In addition to this the low carrying capacity of the ultra-light aircraft works against the survey process here because it can only carry a 4.2 l detector that needs at least 120 minutes of high-level flight to accumulate sufficient data for a proper background correction, this is compared to the 15 minutes a detector pack of 33.6 l takes.

The next problem associated with the airplane's limitations is how to deal with the presence of radon during the survey process. Under normal circumstances, high altitude flights together with flights over water, also employing an upwards-facing detector, give enough data to allow the contribution from radon to be removed from the final data. However, as the ultra-light Jabiru does not carry an upwards-facing detector and since it cannot fly in the necessary manner as usually required an alternative technique has to be used. In such circumstances the tie lines that are flown at right angles to the rest of the survey can be used to largely remove the effects of radon. Sections 5.4 to 5.6 in Dr Coetzee's doctoral thesis [H. Coetzee, Acquisition, processing and enhancement of multichannel radioelement data collected with ultra-light aircraft mounted detectors 2008] goes into the details of how each of these issues are dealt with. Fundamentally, a NASVD (Noise Adjusted Singular Value Decomposition) filter [Hovgaard and Grasty 1997] and continuum removal technique was devised which allowed the extraction of the data [Debertin and Helmer 1988]. Details of the New Machavie flight survey are given in Table 5.3 (towards the end of the chapter), following which Figs. 5.16 to 5.24 are presented sequentially for ease of comparison. Here it should be noted that the results presented in maps 5.16, 5.17, 5.18 are Council for Geosciences airborne data formatted, or in effect re-analysed by the author. Figure 5.20 is

again basic Geosciences data, which has been formatted by the author and then combined with the national dose rate map as presented in Ref [Larkin *et al.* 2004]. In addition Figs. 5.21 – 5.24 are again Geosciences data interpreted by the author. As it can be seen in Fig. 5.16 the flight lines are not all exactly uniformly placed, and in the process of data analysis this needs to be taken into account. There are numerous ways in which this data can now be manipulated and presented graphically. Fig. 5.17 shows a very basic way in which the total counts data could be presented – assigning a colour range for the range of values of total counts recorded.

5.9. Airborne survey results

In the following, each of the count-rate maps reports the values in “stripped counts” (only the gamma-rays from the isotopes centred on the previously discussed windows are counted, see Table 5.1, Section 5.3), with the exception of the total count map. It should be noted that at this stage that no assumptions about secular equilibrium need be made. This also simplifies the mapping of the results since convenient intervals can be chosen when selecting the intervals on the maps.

The aerial survey points are interpolated by a process known as Kriging [Journel Huijbregts, 1978], being a geospatial interpolation based on the assumption that the parameter being interpolated can be treated as a regionalized variable. Which in this case is count rate. It is one of the functions built into the GIS software Manifold V 8.0. In obtaining the maps the exponential Kriging method was used based on the nearest 10 neighbours. Those areas that fall outside of the survey area, as delineated by the aerial survey points, need to be treated with a certain amount of circumspection, as they are largely artefacts of the interpolation process.

5.9.1. Topography maps

The basic topography of the survey area is shown in Fig. 5.18, which is overlaid on a satellite image in Fig. 5.19. Both figures have the airborne survey points also included. The highest point at just over 1410 m is toward the western side of the site, the lowest point on the site is the southern most area of the survey site at just

below 1370 m. The bulk of the survey area slopes down in a south easterly direction. The Kromdraaispruit perennial river runs across the site from northeast to south, from the satellite image overlay it can be seen that the abandoned slimes dam is starting to encroach onto the survey area through the eroding actions of the weather. Figure 5.20 compares the results of the terrestrial doserate map developed by Larkin *et al.* with the airborne survey carried out by the Council for Geosciences.

5.9.2. Total count map

Figure 5.21 shows the total counts accumulated in a 1 s period of flight mapped to a particular aerial survey point. The survey results show that there is a clearly recorded radioactive anomaly in the eastern region of the map with a peak count rate of in excess of 1900 counts s⁻¹, the majority of the area has a count-rate of 400 counts s⁻¹. There is a second anomaly to the west of the first area but with a count rate an order of magnitude lower.

5.9.3. Uranium window map

As a point of reference, the total-count contour map is shown in Fig. 5.21. Figure 5.22 shows the distribution of the counts which have been accumulated in the uranium window but have the counts from isotopes outside of the window “stripped out”, leaving only the counts from gamma-rays originating from ²¹⁴Pb that is distributed within the top 60 cm of soil. Here, it can be seen that there is a pronounced anomaly to the west of the site with a peak count rate of more than 100 counts s⁻¹ coming from ²¹⁴Pb. Further to the west of the major anomaly there is an area of slightly elevated count rate with a count rate of about 40 counts s⁻¹, where the general count rate in the U-window is less than 20 counts s⁻¹. The major anomaly coincides with the anomaly shown in the total count map (see Fig. 5.21) suggesting that the anomaly is due, in large part, to the uranium present.

5.9.4. Thorium window map

Generally, as can be seen in Fig. 5.23, the counts in the thorium-based window for the isotope ²⁰⁸Tl are low compared to both the uranium window and the potassium

window, the highest count rate being in the region of 20-30 counts s^{-1} . Much of the area is at about 10-15 counts s^{-1} .

5.9.5. Potassium window map

In the potassium window map given in Fig. 5.24, there are two areas of elevated count rates. The highest area, in the eastern portion of the survey area, closely coincides with the areas of elevated uranium counts. At this point the count rate is in excess of 140 counts s^{-1} . The second area of elevated count rate falls in the western section of the survey area where the count rate gets to between 80 and 100 counts s^{-1} , and again this area is in a similar location to the areas of elevated uranium counts. The general count rate is 20 – 40 counts s^{-1} .

Overall, the major contributors to the total counts recorded in the aerial surveys come from the uranium and potassium windows. However, in the total count map because there are many more isotopes and many more gamma rays of different energies than are recorded in each of the windows, there are quite a number more counts s^{-1} than would be suggested by the simple summation of the contributions in the uranium and potassium windows.

Table 5-3 Details of the survey flight at the New Machavie survey site

Terrain:	Undulating.	Its necessary to record the type of terrain as this can have a significant impact on the recorded signal
Flying Height:	50 m	This is the aimed for height above ground level that the pilot tries to fly his aircraft at. The actual height above the ground needs to be known so that allowances in the activity concentration algorithm can be made for fluctuations in the height
Line Spacing:	50m	The chosen line spacing represents a compromise between the cost of flying each line and the data that are accumulated – as can be seen from the Fig. 5.16 there is significant overlap of each of the flight lines thus trying to ensure as far as is possible, that no significant anomalies are missed in the survey process
Tie line spacing:	500m	These are flown perpendicular to the survey flight lines to give a degree of quality assurance to the measurements taken, see Fig. 5.16
Flying Speed:	140 kmh⁻¹	This is dictated to a large degree by the aerodynamics of the chosen aircraft. The Council for Geosciences made use of the Jabiru as it is an economical plane to fly, it also has the ability to be flown at a relatively slow speed, which helps improve the sample statistics as has already been discussed in Section 4.6
Radon effect:	Present	It is necessary to take in to consideration whether or not atmospheric radon is present during the survey process. If it is, then it is necessary to reflect this in the data processing algorithm.
Datum:	CAPE	
Projection:	Transverse Mercator Necessary for the correct plotting of the data	
Central Meridian	27 East	
Weather:	Moderate with winds, the atmospheric conditions can affect things like radon exhalation from the soil, and as a result the overall survey results.	

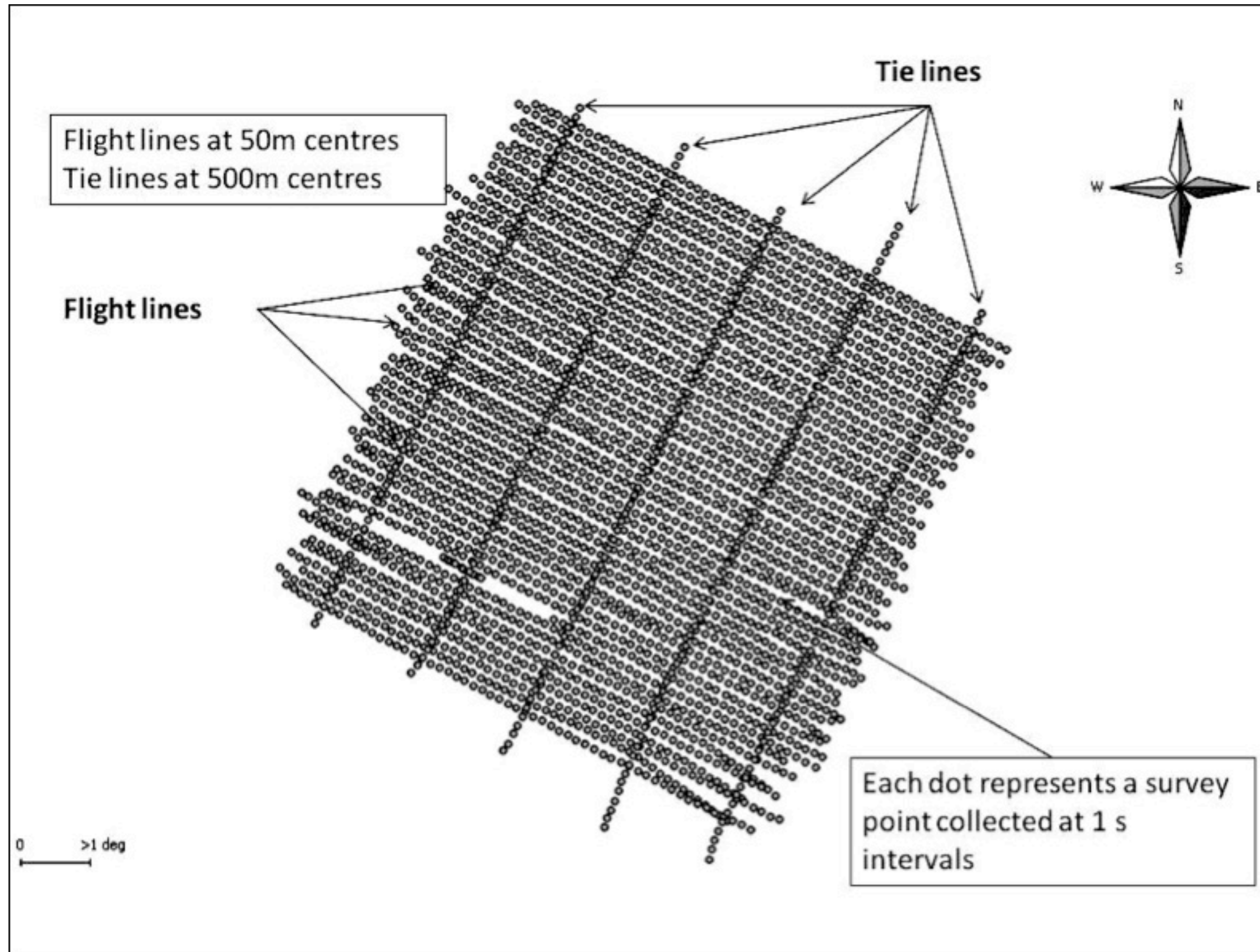


Figure 5-16 Basic survey pattern flown at the New Machavie survey site (the co-ordinates have been omitted for the sake of clarity).

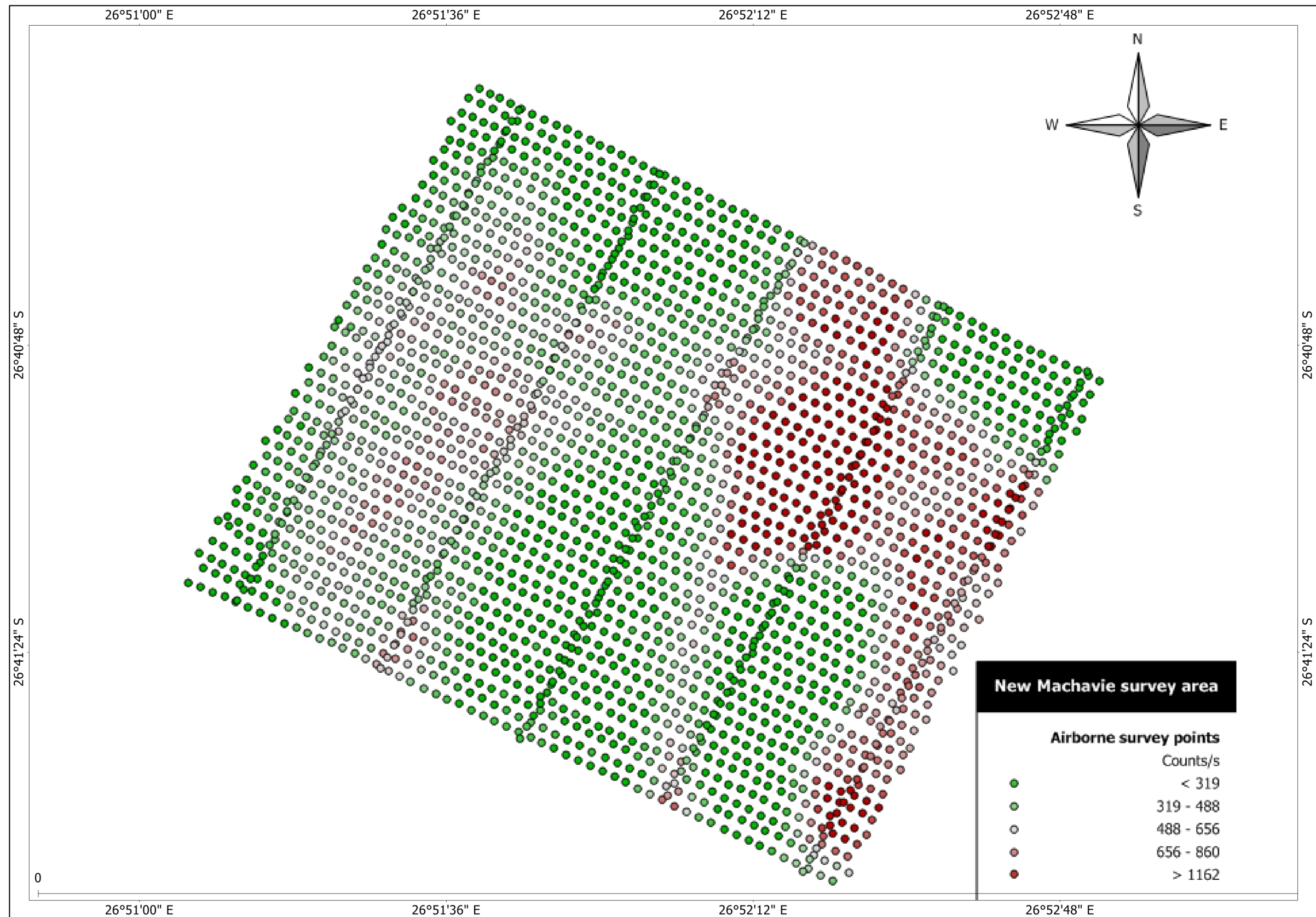


Figure 5-17 Basic presentation of airborne survey data at the New Machavie survey site.

The values outside of the flown area are generated as a reasonable extrapolation of the flight data, bound by the GIS routine outside of the flown area. This is pertinent to the following figures. Fig 5.18, 5.21, 5.22, 5.23 and 5.24

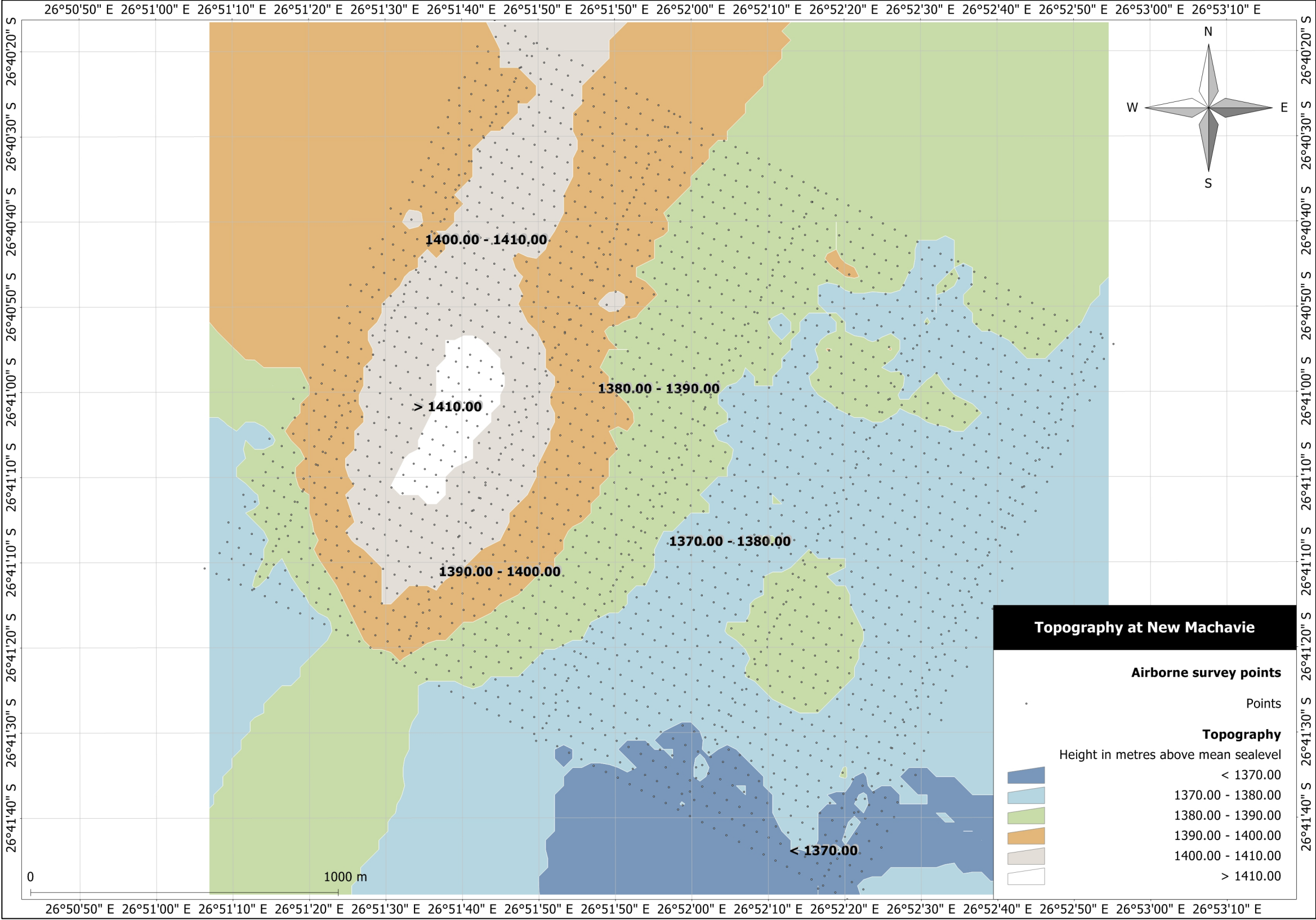


Figure 5-18 Topography and aerial survey points at the New Machavie survey site

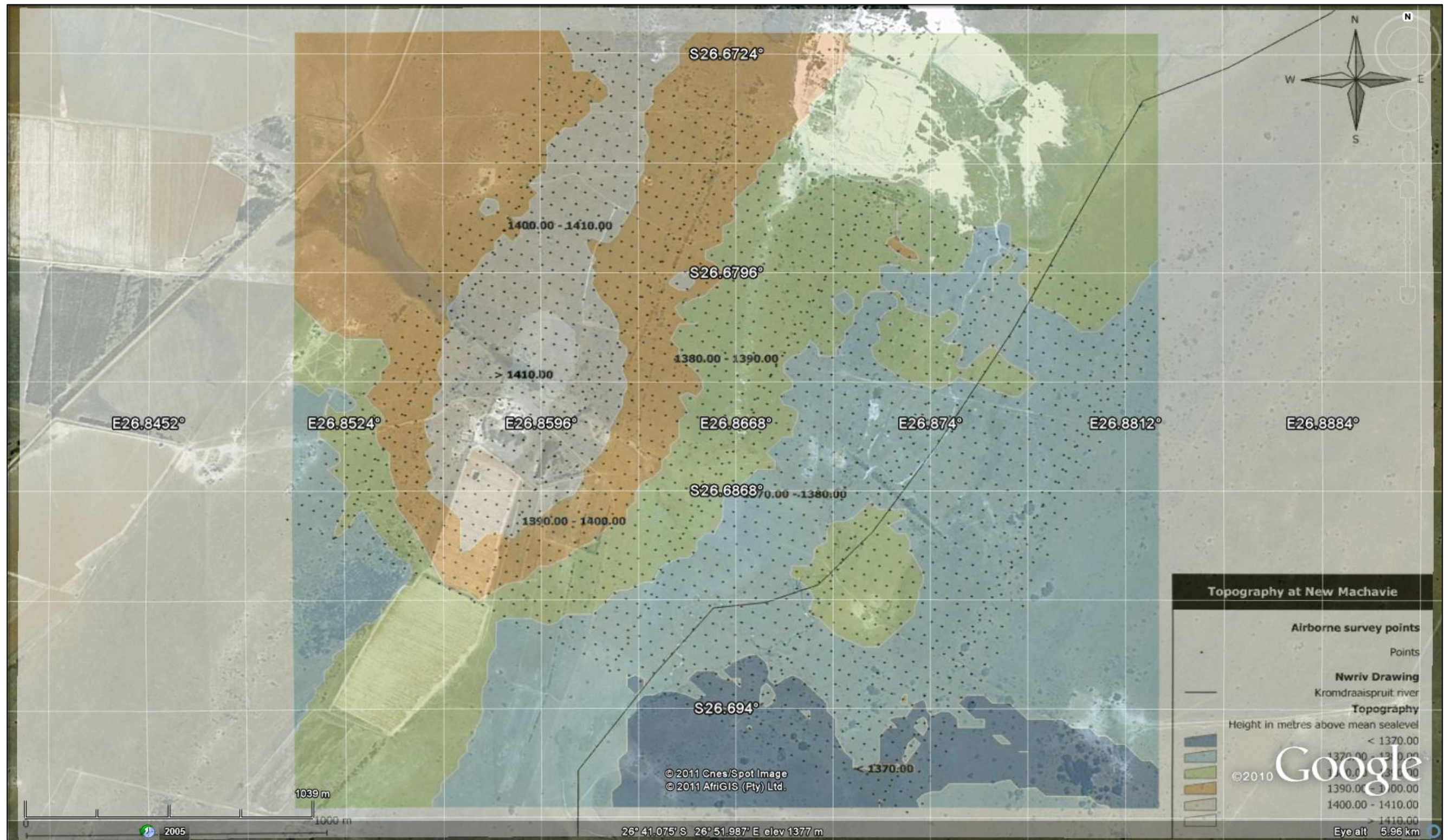


Figure 5-19 Topography at the New Machavie survey site over-laid on to a satellite image.

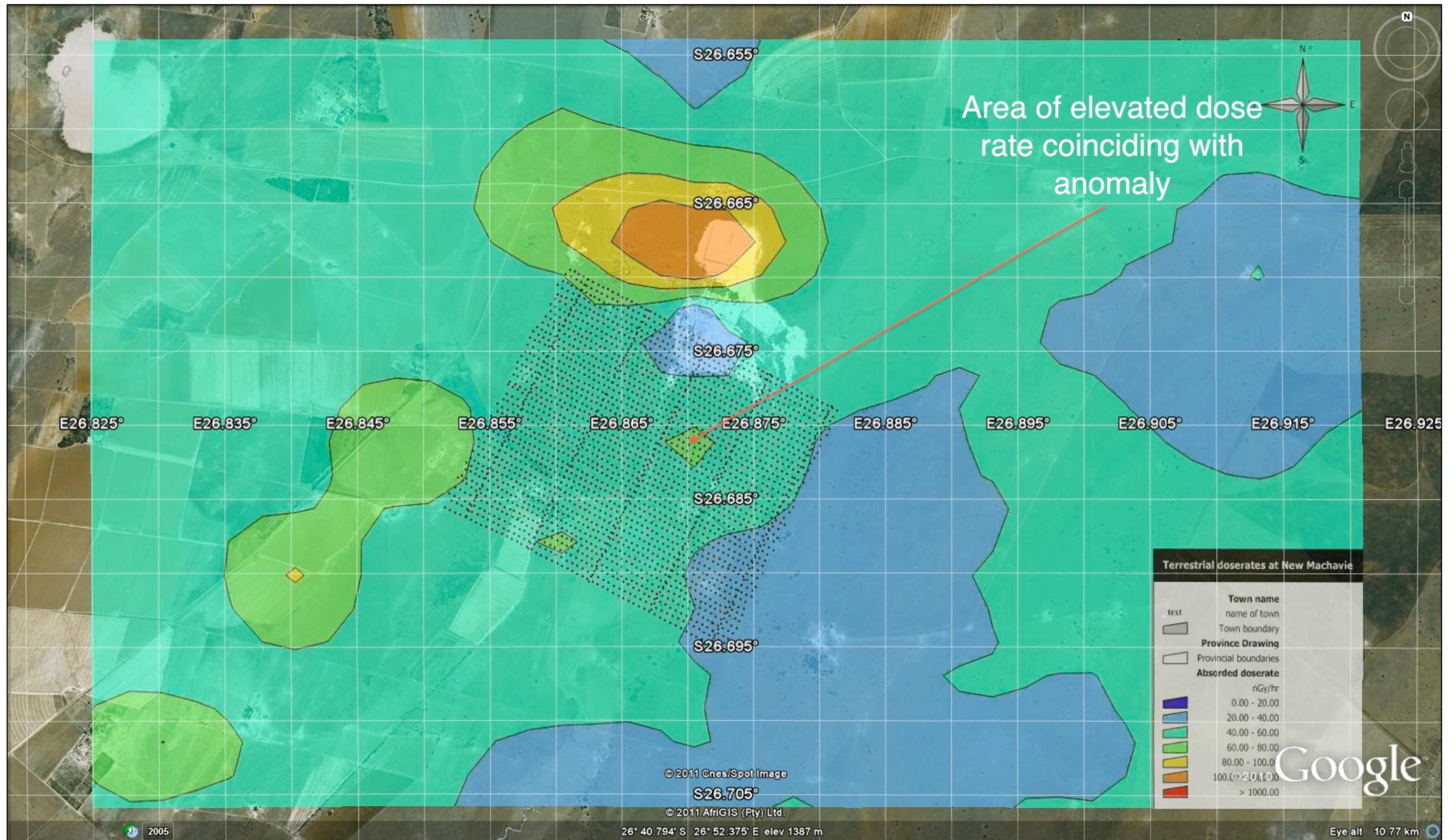


Figure 5-20 Coincidence of total count anomaly from ultra-light aircraft survey with point of elevated dose rate from countrywide survey project [Larkin *et al.* 2008].

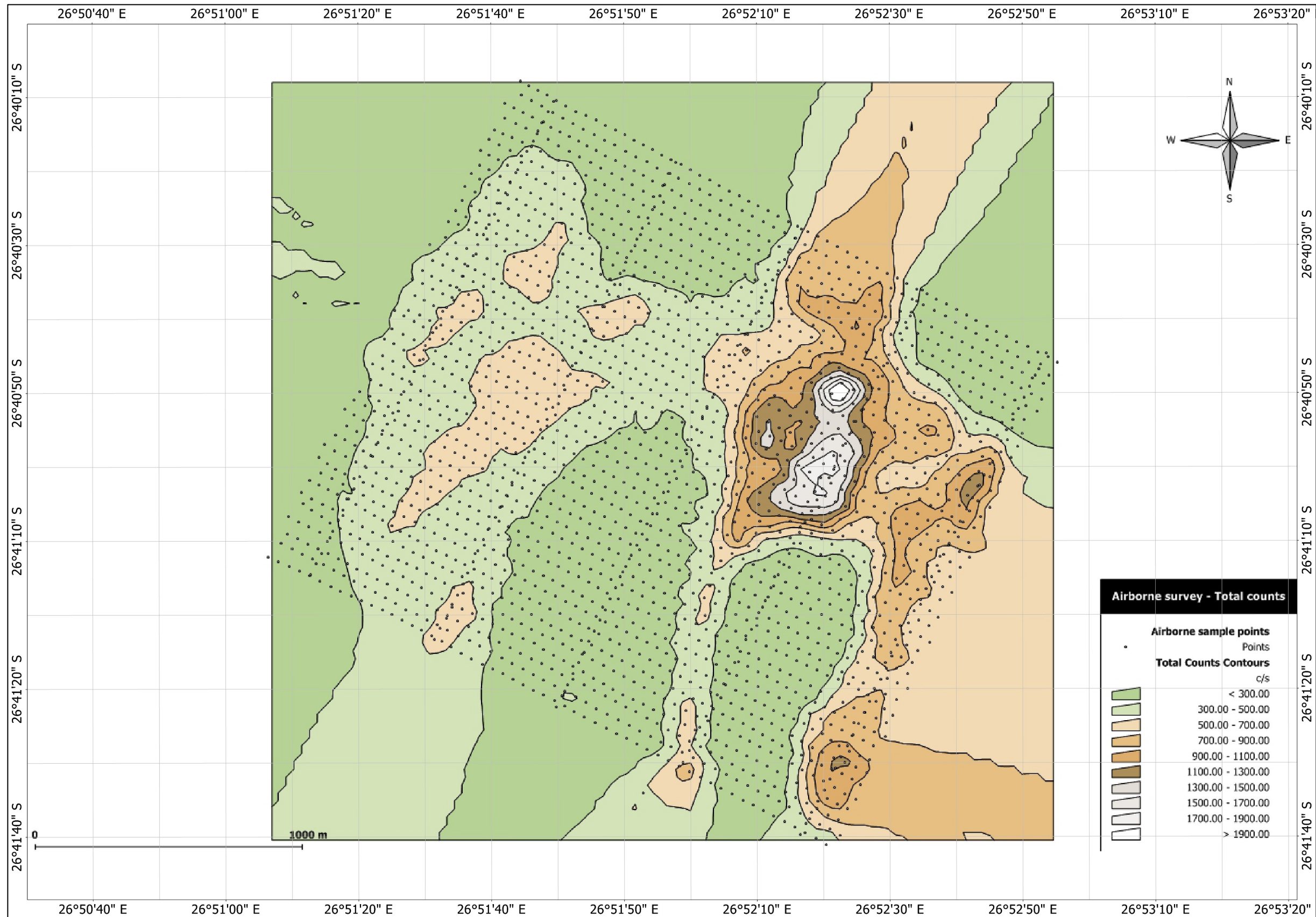


Figure 5-21 Total count contours at the New Machavie survey site

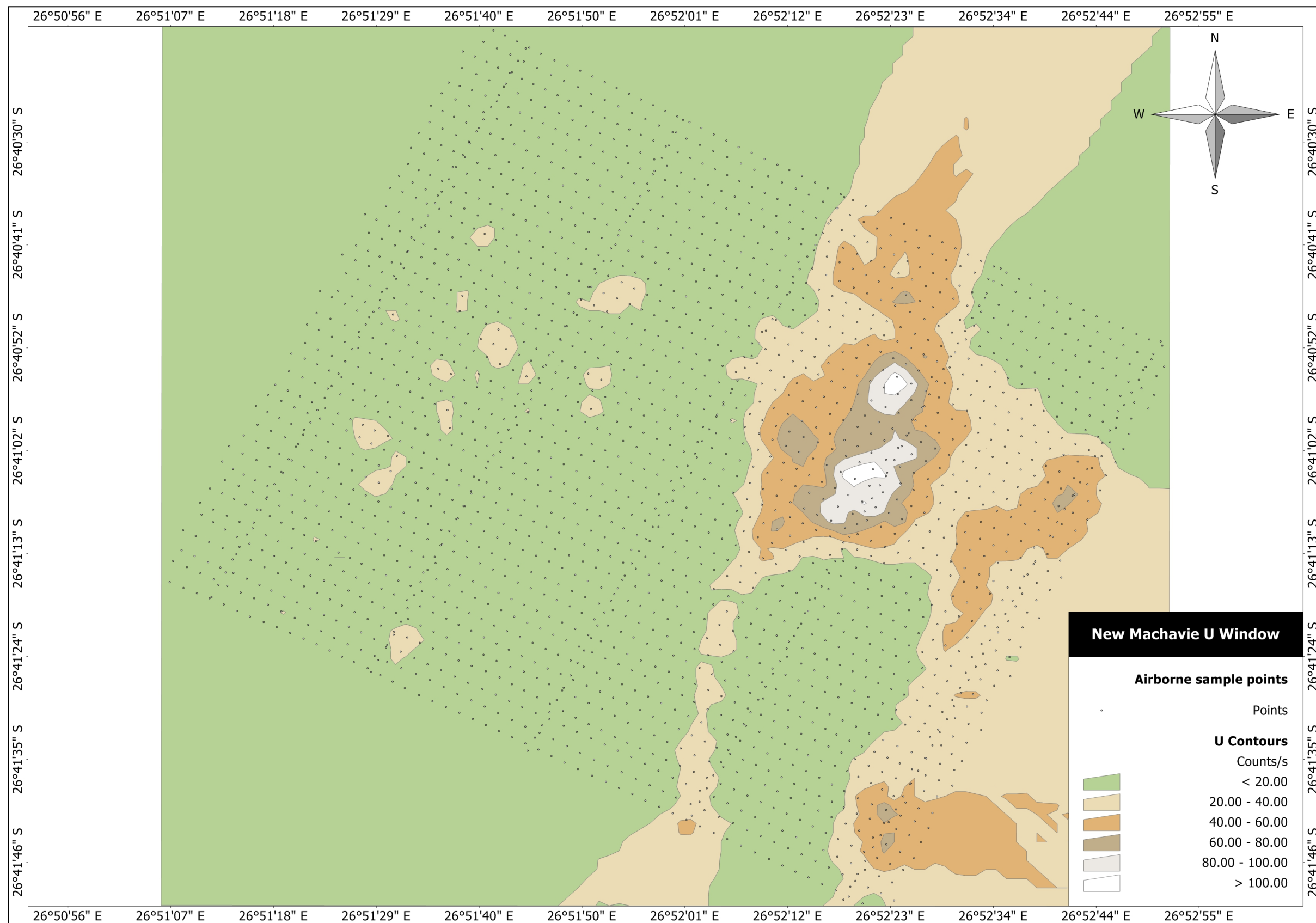


Figure 5-22 Distribution of count contours in U window at the New Machavie survey site.

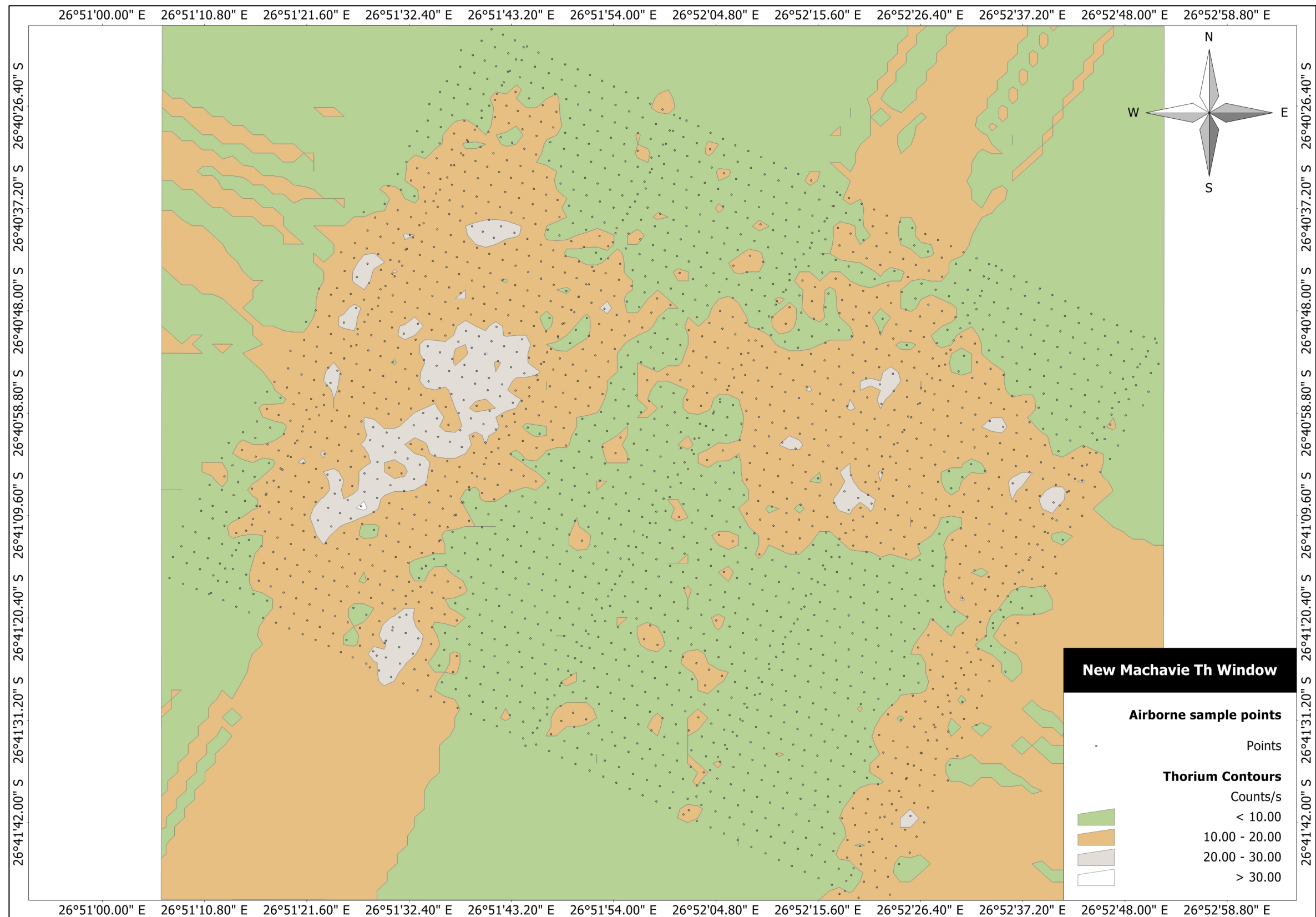


Figure 5-23 Distribution of count contours in the Th window at the New Machavie survey site.

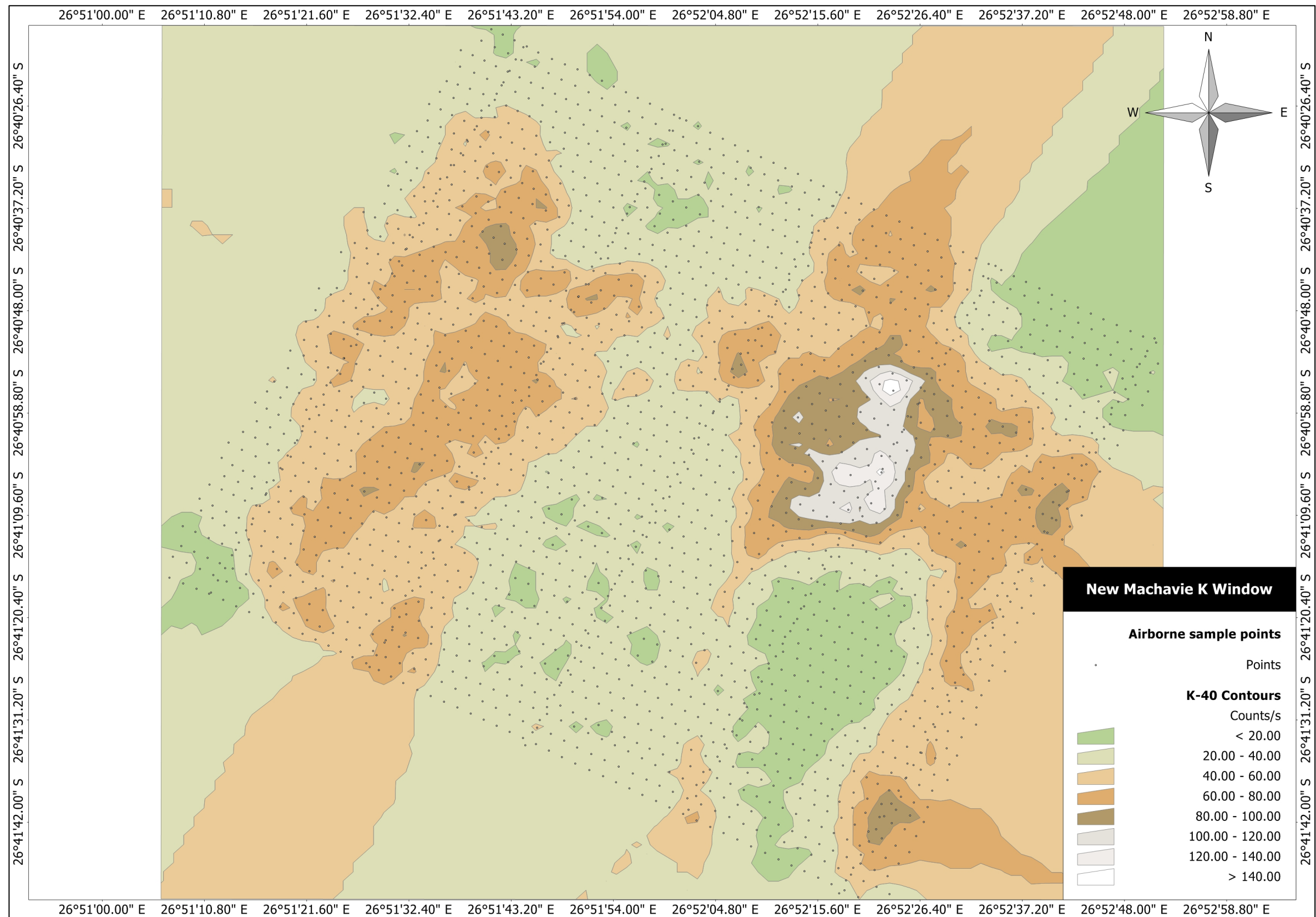


Figure 5-24 Distribution of count contours in the ^{40}K window at the New Machavie survey site.

CHAPTER 6

GROUND SURVEY RESULTS, COMPARISON WITH AIRBORNE SURVEY AND DISCUSSION

6. Introduction

Usually ground-based surveys are carried out on small areas seldom larger than 10 hectares in size as the work is very labour intensive. Hand-held NaI (TI), detectors will only ‘see’ a radius of about 12 m at maximum [Strydom 2003] which means if a scanning survey is to be carried out that gives full coverage of an area, survey lines at a maximum of 10 m need to be walked. These scanning surveys are then also combined with the collection of a number of soil samples, which cover the range of radiological conditions that exist on the site, and together are then used to develop an overall radiological picture of the survey area.

At the New Machavie site it was decided not to do a scanning survey both because the resources to carry one out were not available and because the airborne survey could be used as the scanning survey of the area. It was, therefore, decided to collect a number of representative soil samples of the area.

6.1. Soil Sampling

Twenty-two soil samples for laboratory analysis were collected. Each sample point was chosen based on the total count-rate map and was located on the survey area using a hand-held GPS unit and is indicated in Figure 6.1. Each soil sample taken, details of which are given in Table 6.1 represents a soil profile of the top 20 cm of soil. The soil samples were double-bagged to prevent possible leaks and cross contamination, and were labelled and recorded in a field data log. The samples were then submitted to the Applied and Environmental Research Group of the Schonland Research Institute for Nuclear Sciences for analysis, where they were analysed by the resident technician and the results reported. The calibration

of the equipment was done in line with the Group's standard operating procedures.

The basics of the analysis process are as follows;

- All organic matter was manually removed from the sample,
- Soil was passed through a series of sieves to produce a material, that was as homogeneous as possible,
- The sample was then put into a Marinelli beaker of a constant volume and then into a 'lead castle' and counted for two hours using a germanium diode [Dryad, Kovar and Suran 2001] and,
- Finally activity concentrations were reported along with an associated counting error and confidence level indicating the actual presence of the various radioactive isotopes being investigated. The reported activity concentrations were then converted, in the case of uranium and thorium, into parts per million values and in the case of ^{40}K a percentage by a simple mathematical calculation. This allowed direct comparisons with the reported airborne survey values.

These samples were then analysed using a high purity intrinsic germanium diode coupled to Genie 2000 software. The software will also report data necessary for the quality assurance of the samples, including live time, real time and time when the sample count started and finished. Figures 6.1 is the energy calibration curve used in the analysis of the soil samples and Fig. 6.2 is the efficiency curve used in the sample analysis. Fig. 6.3 is an example of the spectrum accumulated from the soil samples. This spectrum shows a number of peaks that have been identified and labelled by the Genie 2000 software including those from ^{40}K and ^{214}Bi , peaks that are used in airborne radiometric surveys

These samples were then compared to the airborne survey results for the same location in the survey area. Samples were taken to represent the whole spectrum of count rates recorded by the aerial survey.

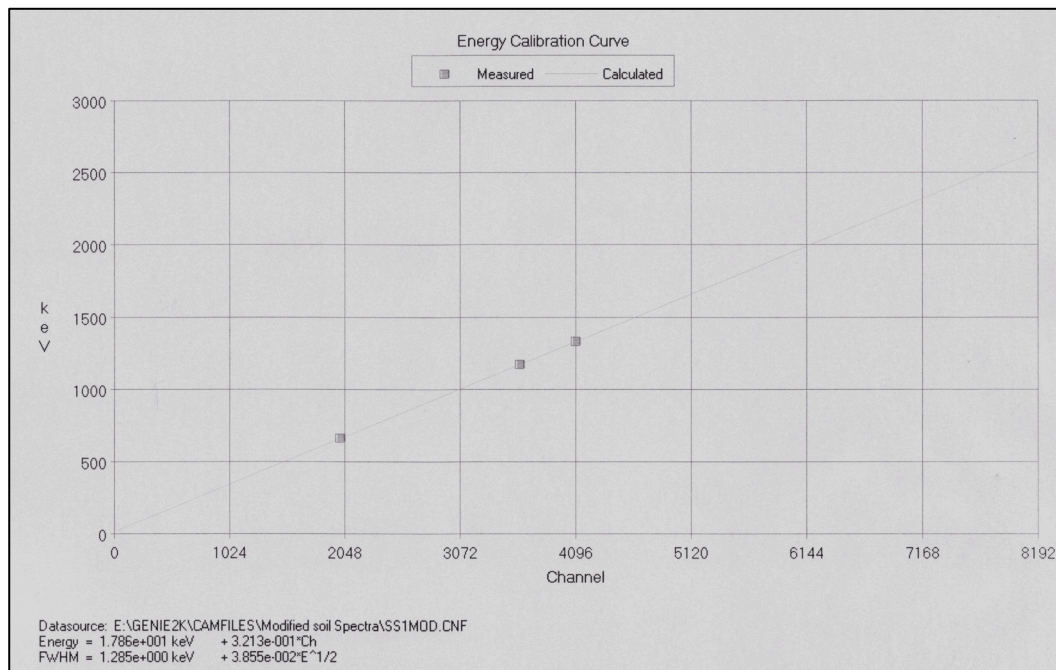


Figure 6-1 Energy calibration of the Genie 2000 counting system.

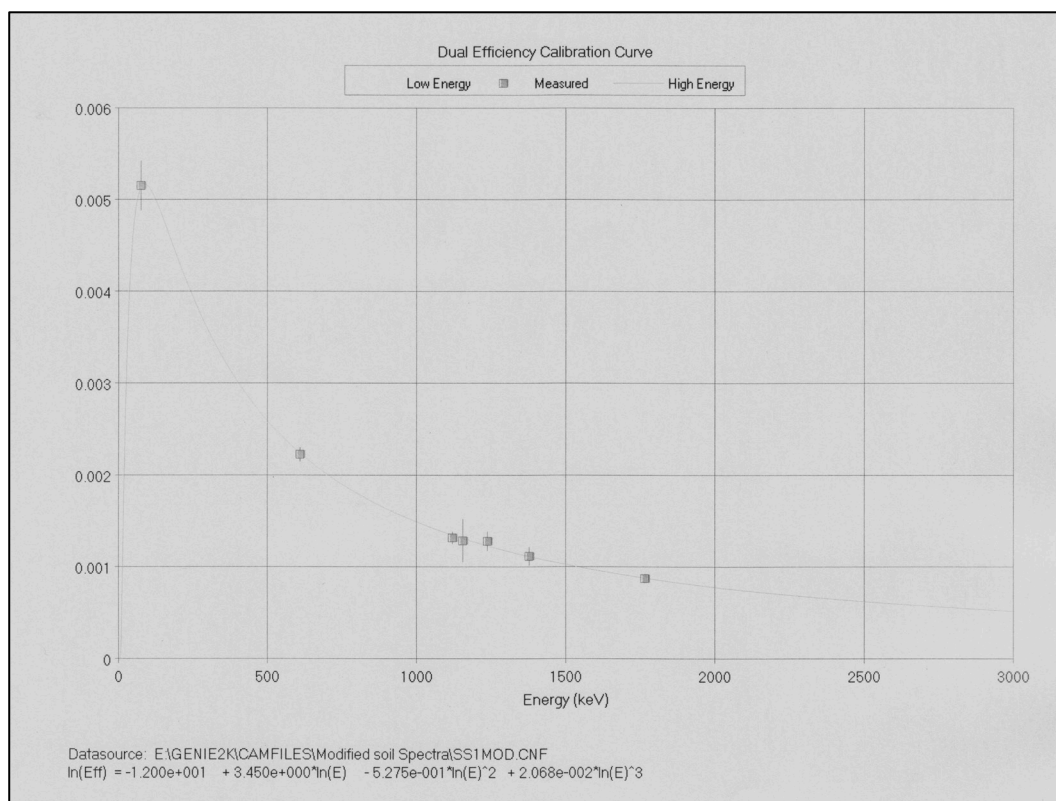


Figure 6-2 Efficiency calibration of Genie 2000 counting system.

Additionally a few *in situ* measurements were taken with a multichannel analyser (MCA) but these were qualitative in nature rather than quantitative, as the

operating software of the MCA did not allow for a quantitative calibration of the instrument.

6.1.1. *In Situ* measurements

In addition to the soil samples that were taken, *in situ* gamma spectra were accumulated at four of the sample points. These spectra were qualitative in nature since the operating software would not allow for a quantitative calibration. (Discussed later in Section 6.7)

Table 6-1 Masses of soil samples collected and analysed

Samples			
Sample Number	Mass (g)	Sample Number	Mass (g)
SS 1	413	SS 12	530
SS 2	333	SS 13	487
SS 3	465	SS 14	427
SS 4	431	SS 15	430
SS 5	371	SS 16	344
SS 6	293	SS 17	472
SS 7	334	SS 18	507
SS 8	314	SS 19	504
SS 9	330	SS 20	492
SS 10	488	SS 21	605
SS 11	530	SS 22	448

Density of the dried soil samples were in the range 1.1 – 1.6 gcm⁻³

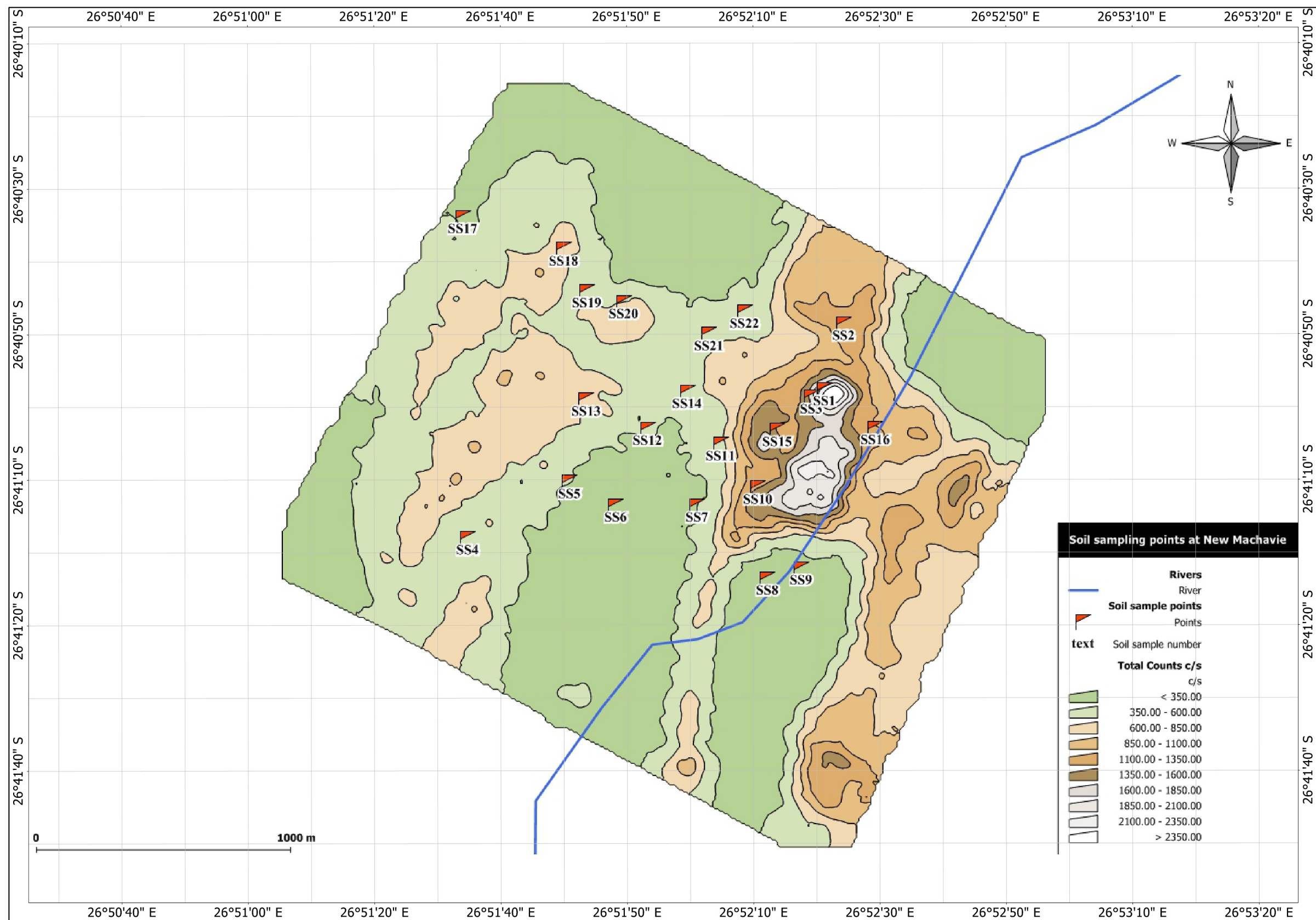
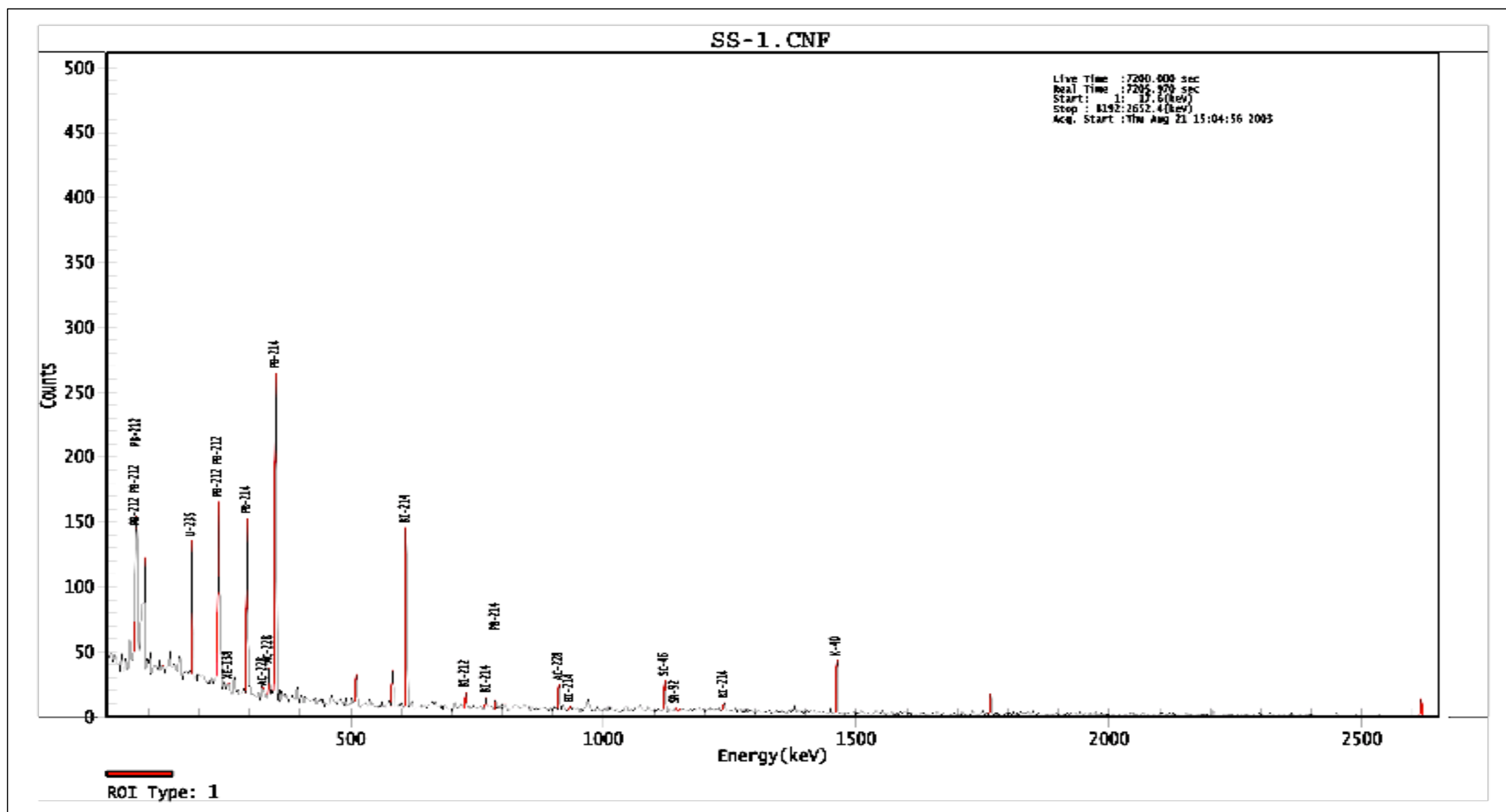


Figure 6-3 Soil sampling points on topographical layout at the New Machavie survey site.



6.2. Ground-based survey results and inter-comparison with airborne survey

An inter-comparison will be made between the airborne radiometric survey data and soil activity concentrations from the samples that were taken at the same point as the airborne data were recorded. In addition, the *in situ* results taken with the MCA are included here for completeness. The soil sample analysis results are reported in parts per million (ppm) which is the preferred unit for geophysical surveys. In the case of ^{40}K , percentages are used. This is to facilitate a comparison with the airborne survey where results are routinely reported in either ppm or percentage in the case of ^{40}K .

No measurement error was reported with the airborne radiometric survey data, so it was decided, on a conservative basis, to use standard error when plotting the data on the following graphs. In the case of soil samples, the errors associated with the plots are those reported by the spectrum analysis software, modified appropriately in those cases when average activity concentrations are recorded. Note, for consistency and readability a table of results for ground-based and airborne surveys followed by the associated figures is presented at the end of each of the sections below.

6.2.1. Uranium inter-comparison

The uranium soil sample activity concentrations are based on the 1765 keV gamma ray peculiar to ^{214}Bi and the 351 keV gamma ray of ^{214}Pb and the airborne survey results are based on the stripped counts in the U-window (1765 keV gamma ray energy of ^{214}Bi). A number of interesting things come out of the inter-comparison of the soil samples and the airborne measurements taken in the same place.

Firstly, in Fig. 6.5 it can be seen there are several points that seem to fall right outside of any obvious relationship, the specific points being samples 8, 9, 14 and 16. These samples seem to have very high soil-sample activity concentrations as compared to the measured airborne activity concentration.

Here the belief is that in the period between when the airborne survey was carried out and when the soil samples were collected, there was significant 'run-off' from the slimes dam to the northeast and just outside of the survey area leading to an alteration of the levels of radioactivity of the survey area. If the soil sample/aerial survey results are re-plotted omitting points 8, 9, 14 and 16 which all fall on or very near to the various watercourses Fig. 6.6 is produced. In this plot one can see there is a linear relationship between the results from the soil and aerial measurements particularly when various sources of error are taken into consideration. The aerial survey data represent a 33 m² "footprint" and as such is an indication of the activity concentrations of the isotopes of interest within the footprint. This is justified by looking at Fig. 6.7, where it can be seen that white material (slimes known to contain elevated levels of uranium) that has moved down the various low-lying areas of the survey area. The blue arrows on the Fig. 6.7 indicate the seasonal watercourses that cross the survey area. As these seasonal watercourses dry-up, they have deposited the slimes across the survey area leaving elevated levels of uranium that were subsequently sampled.

The relationship is better shown when a number of transects (see Figs. 6.8 and 6.9 with the corresponding values given in Tables 6.3 and 6.4) are taken across the survey site; particularly Transect 2 which runs northeast to southwest across the survey site.

Table 6-2 Uranium soil sample and aerial sample point comparison

	Uranium		
	Soil samples		Airborne survey
Sample	U average (ppm)	Associated error (ppm)	U (ppm)
Sample 19	1.67	0.35	5.17
Sample 5	2.54	0.61	1.04
Sample 21	2.90	0.44	6.06
Sample 18	2.96	0.61	2.83
Sample 4	2.98	0.55	2.07
Sample 11	2.99	0.52	2.36
Sample 6	3.12	0.73	0.78
Sample 17	3.41	0.49	3.83
Sample 13	3.44	0.70	1.45
Sample 12	3.70	0.50	1.2
Sample 7	3.71	0.70	3.59
Sample 20	3.73	0.50	3.57
Sample 10	3.93	0.56	1.48
Sample 22	4.46	0.62	5.85
Sample 15	6.97	0.66	17.06
Sample 2	10.62	0.76	34.63
Sample 9	12.70	2.07	0.84
Sample 1	15.18	1.02	39.71
Sample 3	20.77	1.00	42.95
Sample 14	26.25	1.47	1.84
Sample 8	27.69	1.40	1.71
Sample 16	68.00	1.88	11.49

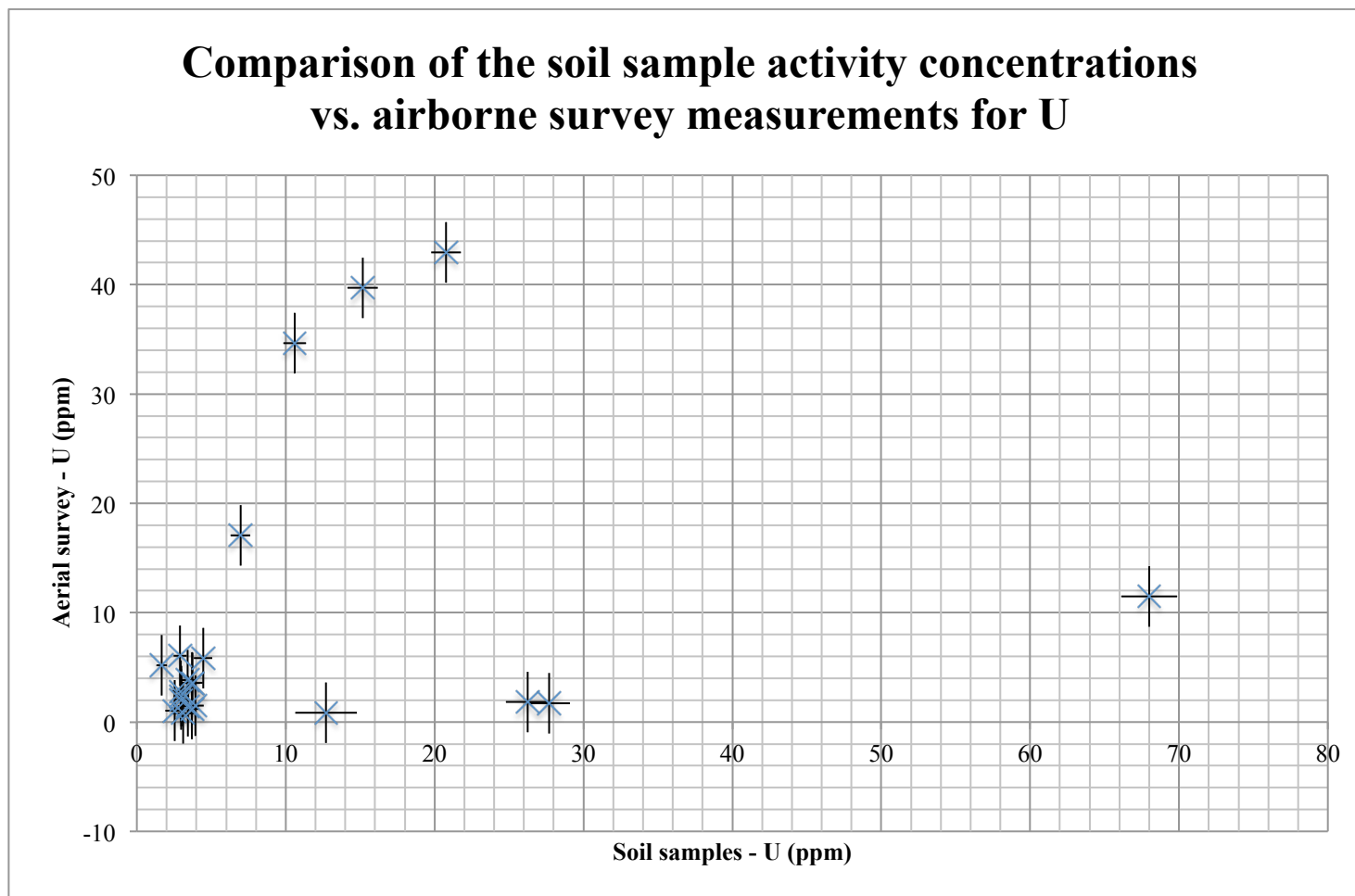


Figure 6-5 Inter-comparison of soil sample and aerial results for uranium

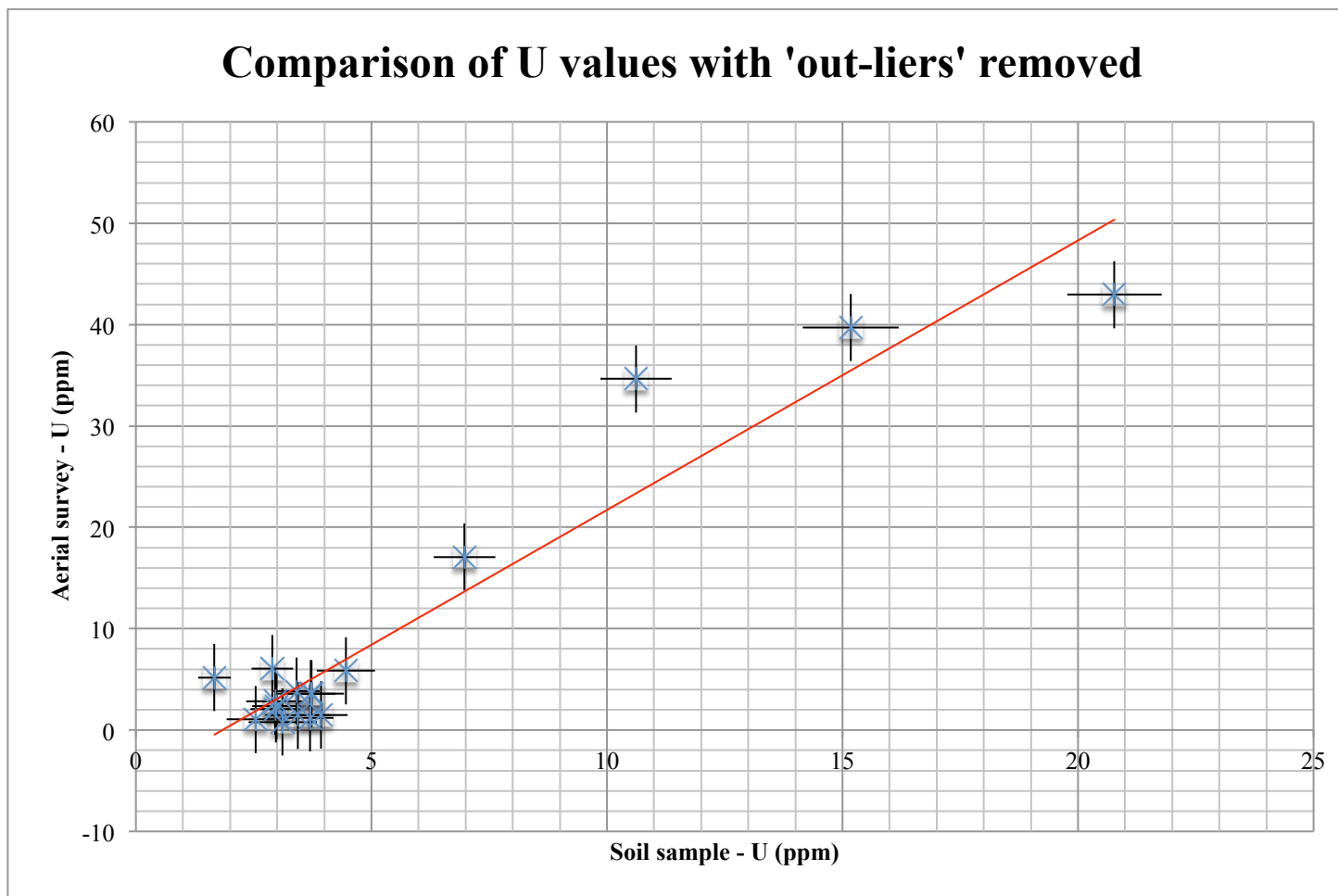


Figure 6-6 Uranium inter-comparison with a number of the obvious outliers removed

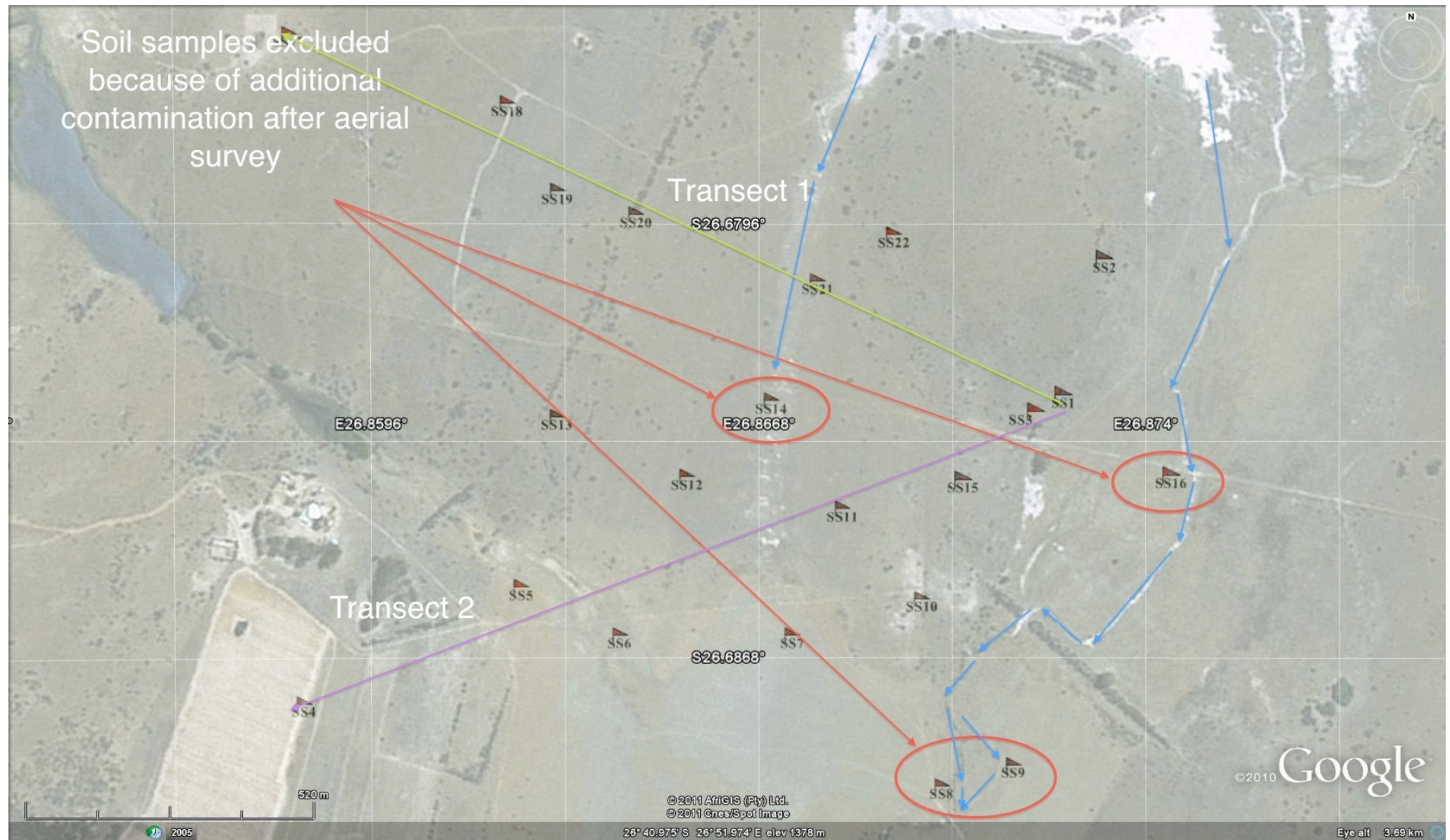


Figure 6-7 Soil sample points excluded because of addition contamination after aerial survey. Blue arrows show direction of seasonal run off from slimes dam.

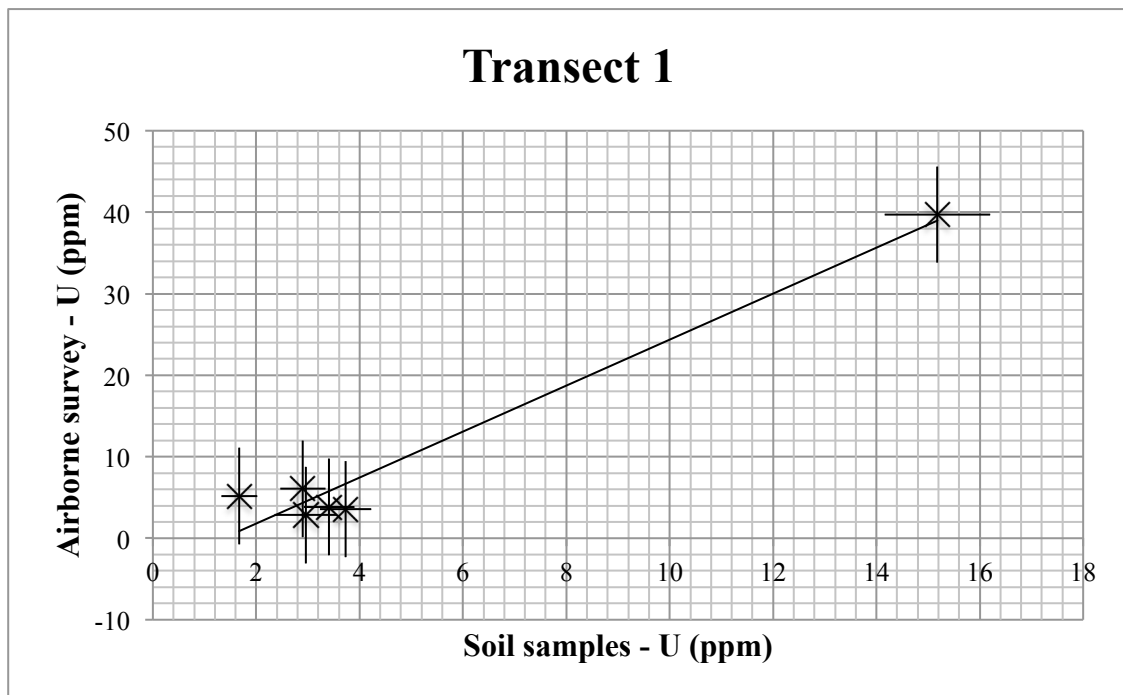


Figure 6-8 Soil sample and airborne survey results for Transect 1 across New Machavie survey site

Table 6-3 Soil sample and airborne survey results across Transect 1

	Uranium		
	Soil samples		Airborne survey
Sample	U average (ppm)	Associated error (ppm)	U (ppm)
Sample 19	1.67	0.35	5.17
Sample 21	2.90	0.44	6.06
Sample 18	2.96	0.61	2.83
Sample 17	3.41	0.49	3.83
Sample 20	3.73	0.50	3.57
Sample 1	15.18	1.02	39.71

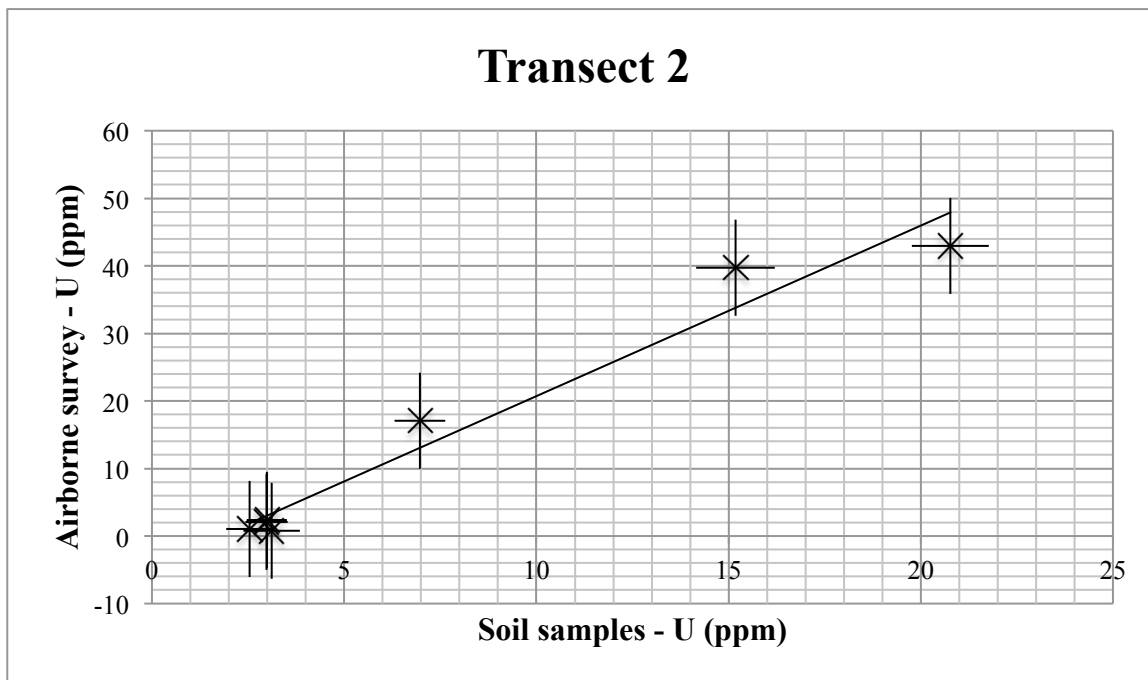


Figure 6-9 Soil sample and airborne survey results for Transect 2 across New Machavie survey site

Table 6-4 Soil sample and airborne survey results across transect 2.

	Uranium		
	Soil samples		Airborne survey
Sample	U average (ppm)	Associated error (ppm)	U (ppm)
Sample 5	2.54	0.61	1.04
Sample 4	2.98	0.55	2.07
Sample 11	2.99	0.52	2.36
Sample 6	3.12	0.73	0.78
Sample 15	6.97	0.66	17.06
Sample 1	15.18	1.02	39.71
Sample 3	20.77	1.00	42.95

6.2.2. Thorium inter-comparison

The thorium activity measurements in the soil samples are based on the averaged measured activity concentrations of the ^{228}Ac peaks in the sample analyses. This isotope was chosen rather than the ^{208}Tl peak used in airborne surveys because the counting statistics are better. The peaks of ^{228}Ac can be readily identified in most of the soil spectra whereas the ^{208}Tl can often be lost in the “noise” associated with these types of measurement techniques particularly at the low levels of activity found at the survey site. As such, the thorium activity concentrations are given in Table 6.5.

Despite the choice of ^{228}Ac as the isotope to measure the activity concentrations in the soil samples, the number of counts was very low and has been impossible to definitively ascertain activity concentrations within the soil samples. In almost every measurement the associated error on the measurement has been in excess of 50%. When an attempt to plot the soil sample *versus* airborne radiometric data is carried out, an inconclusive graph, showing no correlation between the two survey techniques is the result, as can be seen in Fig. 6.10.

Table 6-5 Thorium soil sample and aerial survey comparison

	Thorium		
	Soil samples		Airborne survey
	Th average activity concentration (ppm)	Associated error (ppm)	Activity concentration (ppm)
Sample 10	37.1	20.8	3.09
Sample 19	63.7	22.0	8.02
Sample 15	91.5	40.9	5.22
Sample 5	97.2	41.2	3.89
Sample 11	97.4	29.3	7.9
Sample 20	106.8	35.3	6.22
Sample 21	107.1	30.7	3.02
Sample 18	113.0	29.8	8.65
Sample 13	115.6	42.3	5.86
Sample 1	119.6	61.8	8.56
Sample 22	123.4	40.9	6.79
Sample 2	128.2	53.4	9.03
Sample 6	132.0	56.1	4.95
Sample 4	133.1	40.7	11.14
Sample 12	150.4	41.9	2.65
Sample 3	155.6	49.6	2.71
Sample 17	157.4	40.9	6.88
Sample 7	168.4	50.3	5.4
Sample 9	193.7	55.6	3.21
Sample 14	202.0	55.2	4.82
Sample 8	244.8	79.8	3.87
Sample 16	274.7	68.3	6.35

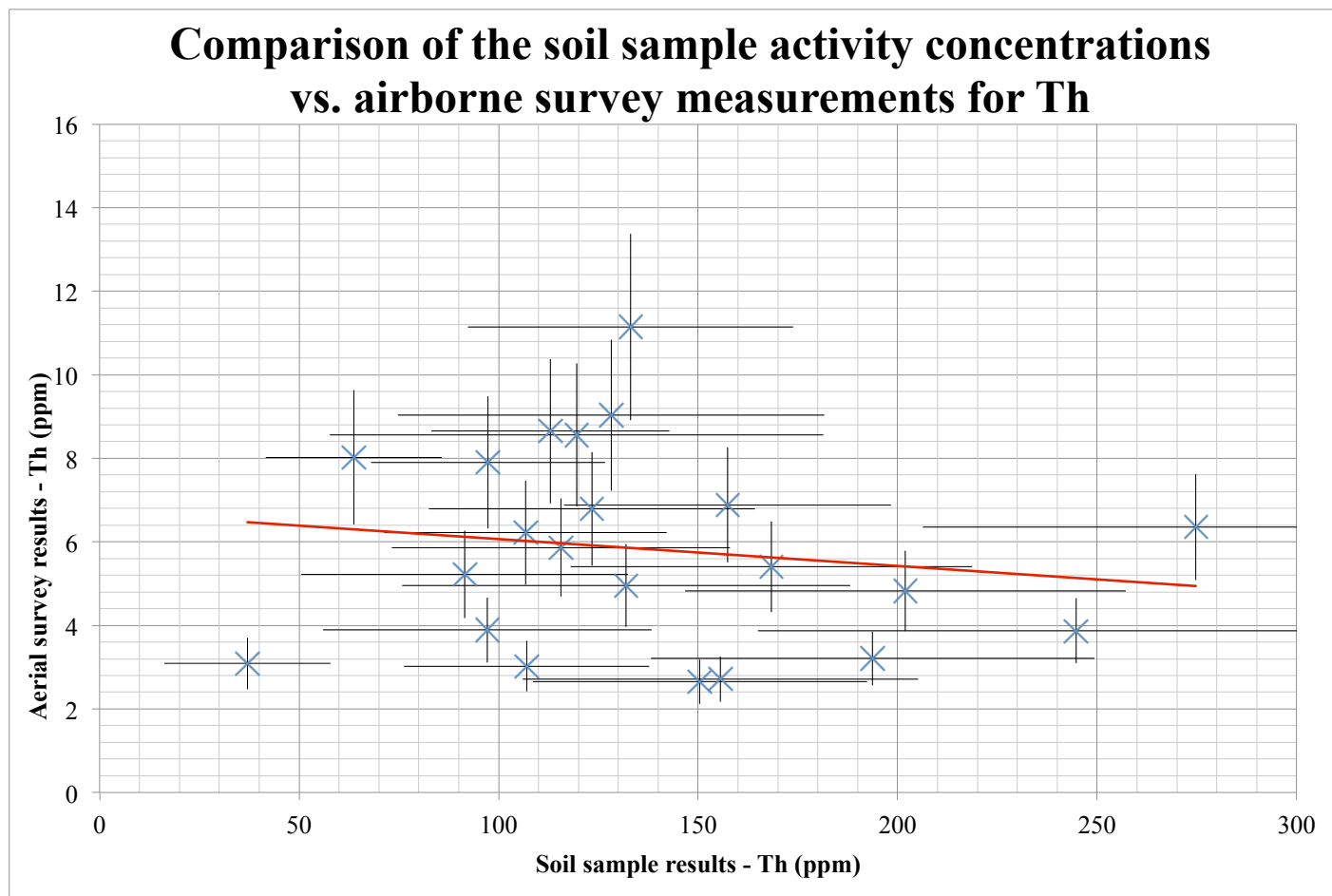


Figure 6-10 Comparison of the soil samples and airborne for thorium.

6.3. Potassium results

Again as in the case with the thorium results, the relationship between the airborne radiometric data and the soil sample results for ^{40}K , no clear relationship emerges when the data given in Table 6.6 is plotted (see Fig. 6.9). This might be due to a number of possibilities; the first is that the potassium is distributed in feldspars and these do not have a uniform distribution across the site [pers conv. Dr H. Coetzee], and the second perhaps more concerning reason is that there is an error in the calculation of the stripping ratios used to develop the airborne radiometric data and this has led to cross talk between the potassium and uranium channels. The result of this cross talk could be the generation of false positives, suggestion the presence of one of the isotopes when actually no exists. In this case possibly counts from the uranium window have been recorded in the potassium window.

Table 6-6 Comparison of soil samples measuring percentage of ^{40}K in a soil sample and the ^{40}K activity (as a percentage) as derived from the airborne survey measurement taken in the same position.

	Soil samples		Airborne survey results
	^{40}K (%)	Associated error (%)	^{40}K (%)
Sample 19	0.81	0.10	1.27
Sample 21	2.05	0.14	1.1
Sample 11	2.25	0.24	0.78
Sample 10	2.42	0.17	0.32
Sample 22	2.54	0.19	1.82
Sample 18	2.78	0.17	1.37
Sample 12	2.84	0.18	0.56
Sample 20	2.85	0.18	0.83
Sample 15	2.87	0.19	1.84
Sample 13	3.07	0.20	0.26
Sample 9	3.08	0.29	0.27
Sample 5	3.16	0.23	0.31
Sample 14	3.36	0.22	0.58
Sample 1	3.42	0.22	1.02
Sample 4	3.54	0.21	1.61
Sample 7	3.62	0.25	0.42
Sample 2	3.73	0.25	2.27
Sample 3	4.01	0.22	0.22
Sample 17	4.16	0.22	1.04
Sample 6	4.27	0.29	0.66
Sample 8	4.46	0.34	0.49
Sample 16	4.57	0.30	1.21

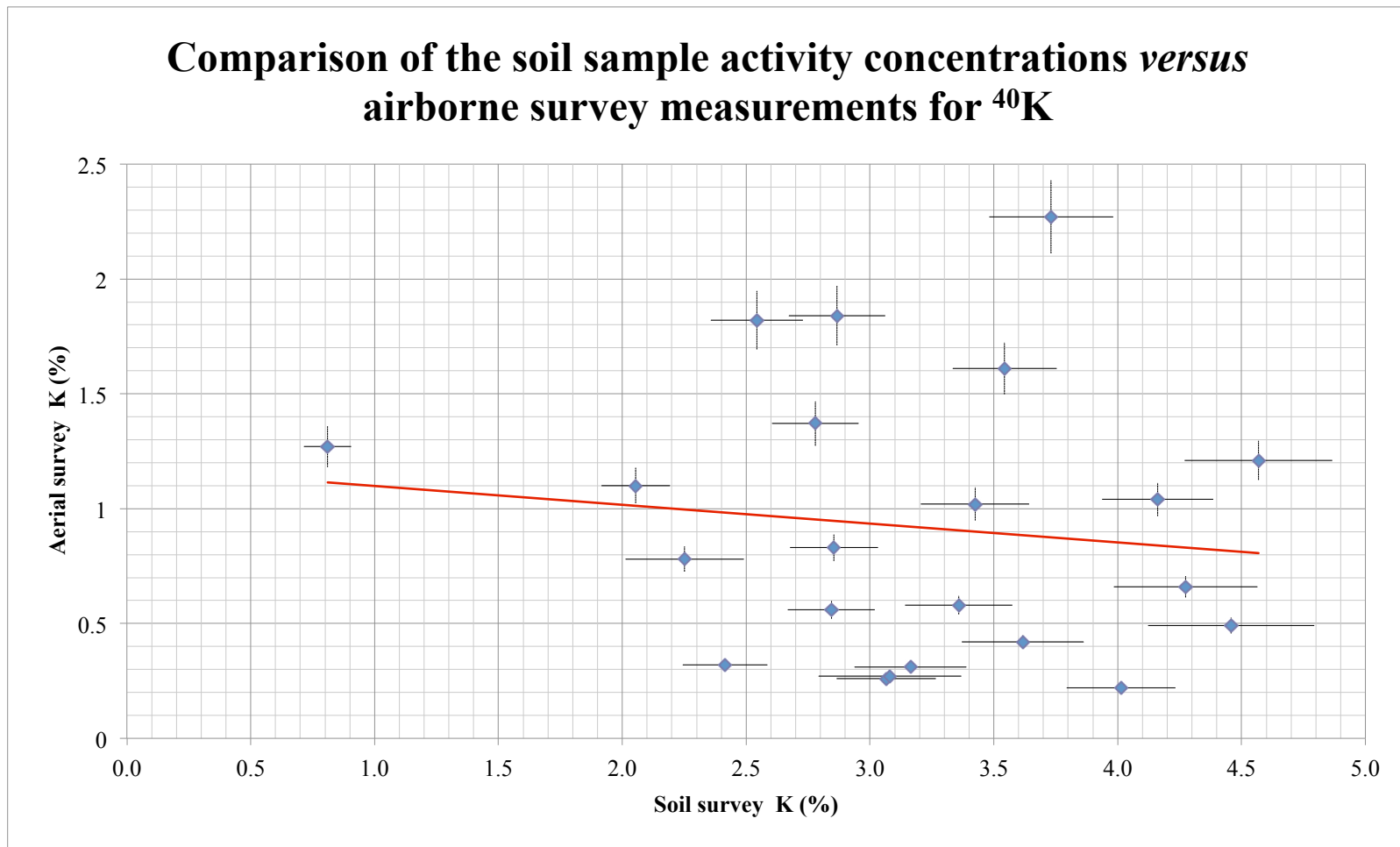


Figure 6-11 Inter-comparison of the soil sample activity concentrations *versus* airborne survey measurements for ^{40}K

6.4. *In situ* measurement of radioactivity

At four soil sample sites, SS1, SS2, SS3 and SS4, *in situ* measurements of the radioactivity were measured using a portable multichannel analyser that was based on a 2" x 2" NaI (Tl) detector. This was held at approximately 1 m above the ground and a measurement was made for a period of 10 minutes for each sample position. Unfortunately since the operating software did not allow for a quantitative calibration, it was not possible to determine activity concentrations but rather only the presence or absence of particular isotopes. It might have been possible to do a retrospective calibration but this was not possible since the calibration pads at Lanseria had been damaged in removal. The original spectra are shown in Fig. 6.10 and 6.11.

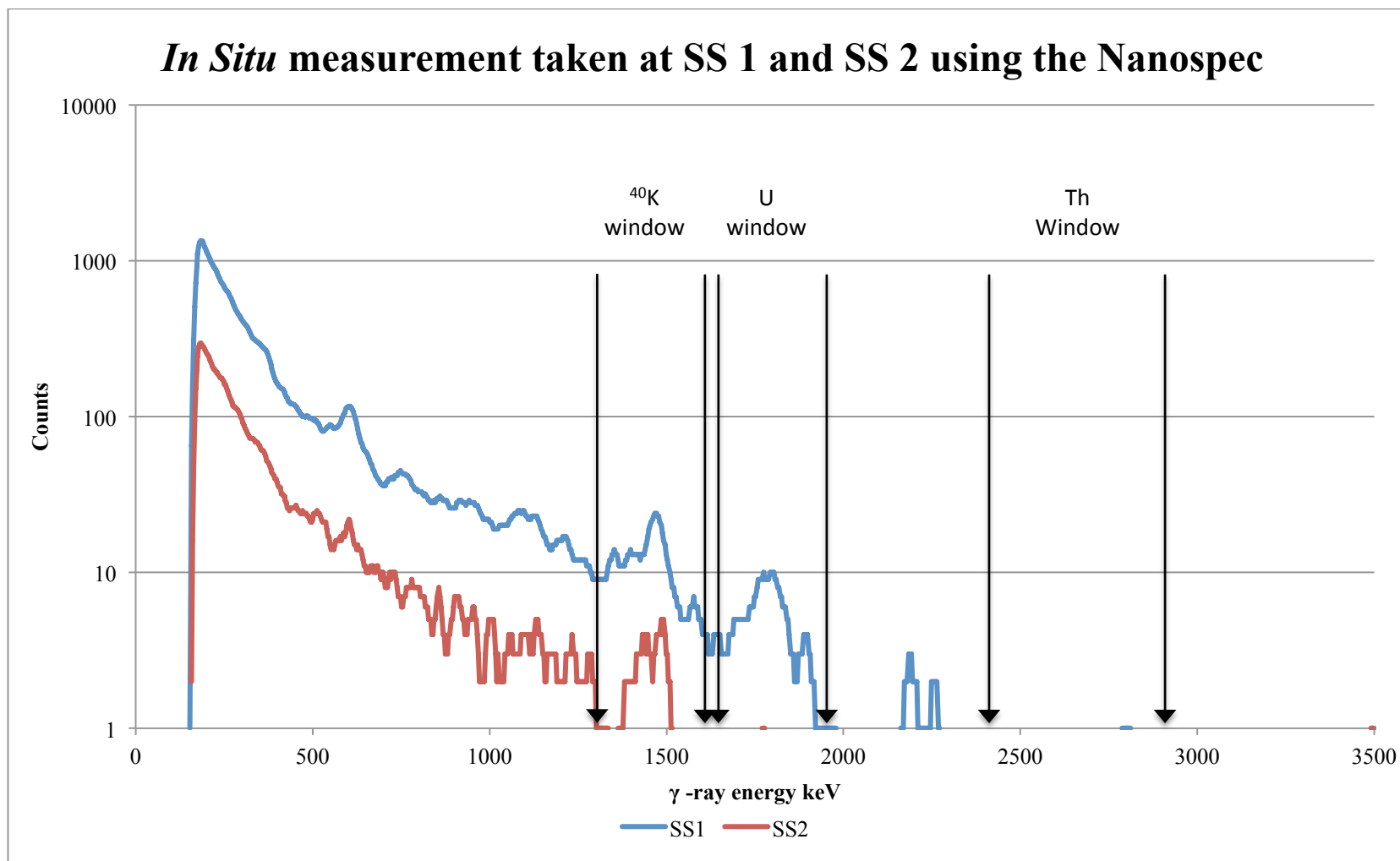


Figure 6-12 *In situ* spectra accumulated on site at New Machavie SS 1 and SS 2

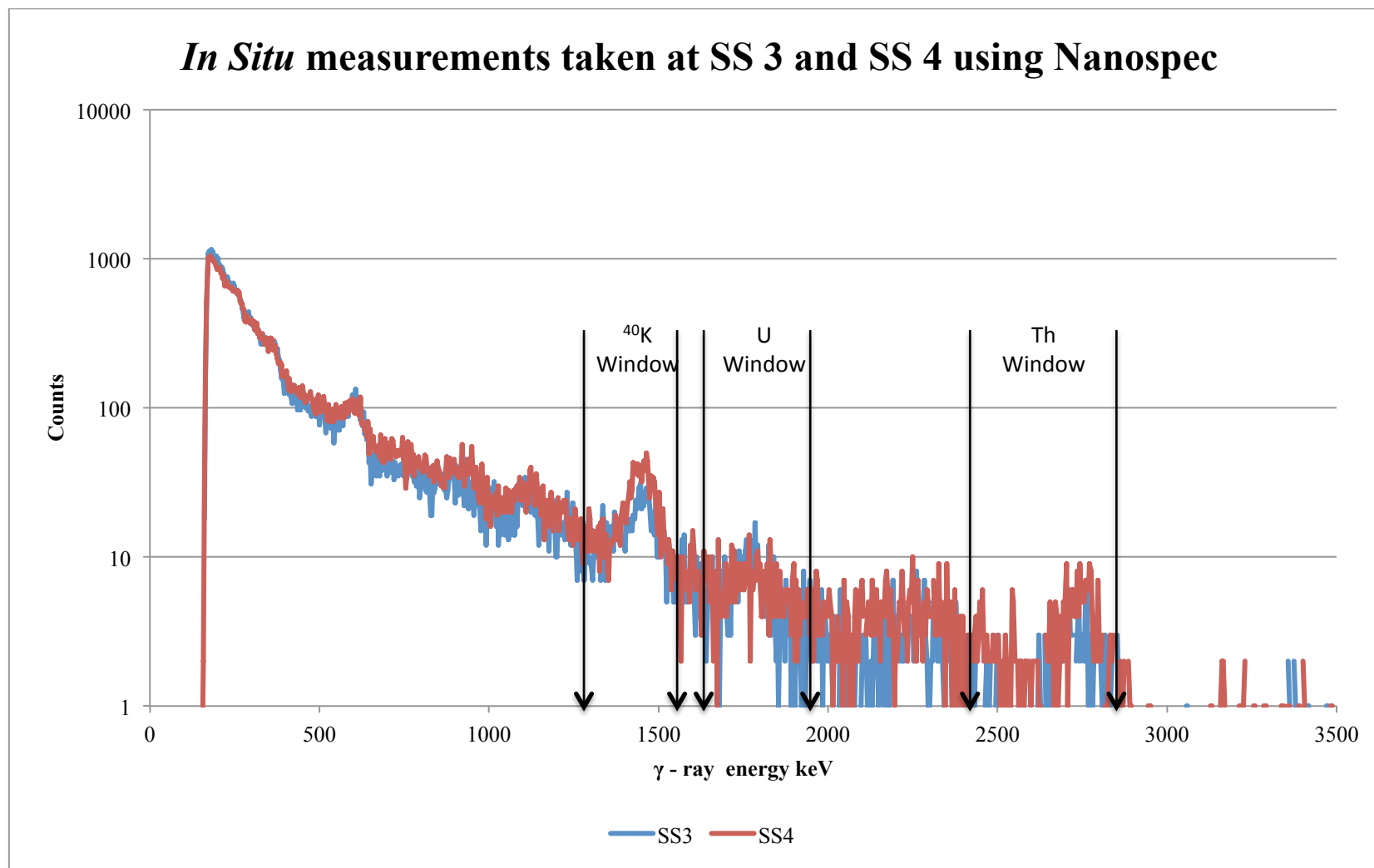


Figure 6-13 *In situ* spectra accumulated on site at New Machavie SS 3 and SS 4

CHAPTER 7

SUMMARY AND CONCLUSIONS

7. General

This investigation set out to compare different radiometric survey techniques that are in general use today and to examine the strengths and weaknesses of the two techniques, and how these techniques can be exploited when determining the distribution of radioactivity within the environment.

Airborne radiometric surveys have the advantage of allowing large areas of land to be surveyed relatively quickly but with the distinct disadvantage of being a costly exercise both in the cost of the detector packs and the flight platform: though for a minerals exploration company the cost/benefit ratio can soon be justified if geological indicators suggest the presence of mineral deposits in commercial quantities. On the other-hand, ground-based surveys are almost the opposite of airborne surveys. They are labour intensive and only allow small areas of land to be examined in any period of time compared to aerial surveys but at a comparatively much lower cost. The distinct advantage though of the ground-based survey is the ability to make statistically significant measurements of the *in situ* radioisotopes, and to be able to plot with a high degree of accuracy the distribution of these radioisotopes in the top 60 cm of soil.

It should, however, be noted that there is a limitation that both aerial and ground based surveys suffer from; they are both unable to determine activity concentrations in the soil to a depth greater than approximately 60 cm. This is because of the shielding offered by the soil in which the radioactivity is distributed [Beck, DeCampo and Gogolak September 1972]. This depth can vary somewhat based on soil type, density and water content.

7.1. Airborne radiometric surveys

When looking at the terrestrial doserate map derived from low resolution general airborne surveys and comparing it to the subsequent high-resolution airborne

survey map that was created as a result of the Council for Geosciences work there is a good correlation between the two. This is despite the poor quality of the data used to produce the terrestrial dose rate map, which was based on pixels measuring $250 \times 250 \text{ m}^2$ (considered very poor resolution nowadays). Based on a flight speed of 140 kmh^{-1} , the high-resolution data from the Council for Geosciences would be on centres of 39 m and the line spacing between flight lines was 50 m, so giving a pixel size of $39 \times 50 \text{ m}^2$, in essence a pixel area some 32 times smaller than the original data set. This has allowed mapping in much more detail. Comparing Fig. 5.20, an example of the terrestrial dose rate map over the New Machavie survey area that shows an area of increased dose rate and Fig. 5.21, which is a map of the same area. This time in 'high resolution' where it is possible not only to see an area of increased count rate but also to make an identification of the isotopes that are responsible for the count rate and the increased ambient dose rate.

One of the disadvantages of using the ultra-light airplane for the high-resolution survey work, certainly for this particular project, was that the effect of atmospheric radon on the results could not be determined. Whilst it is accepted that because the duration of the survey flight over the area was short and the area surveyed was relatively small, there was going to be very little significant change in the atmospheric conditions leading to a change in radon concentration during the flight, the proposed techniques for removing its influence were not altogether satisfactory.

Another disadvantage, certainly when compared to more conventional flight platforms, was the lift capacity of the Jabiru. This meant that it could only carry a single 4.2 l NaI (Tl) detector and the associated electronics, which reduced the sensitivity and increased the signal/noise ratio in the detector system [H. Coetzee, Acquisition, processing and enhancement of multichannel radioelement data collected with ultra-light aircraft mounted detectors, 2008]. This was to some extent offset by the ability of the aircraft to fly lower and slower over the survey area, so reducing the shielding effect of the atmosphere and accumulating more counts per unit distance travelled along the survey axis (see Section 4.5).

7.2. Disequilibrium in the thorium and uranium decay chains

One of the key concerns of any airborne radiometric survey, or *in situ* measurement, is the following “is the uranium decay chain in secular equilibrium?” at the survey site under consideration. If there is significant radon emanation from the site, then any surveys that are undertaken using ^{214}Bi as the ‘indicator’ isotope used to calculate the concentrations of uranium present is going to underestimate these values. To make an empirical correction to derive the ^{226}Ra concentration is an extremely complex process based on the radon emanation rates at the site. Atmospheric pressure, temperature, wind speed, soil moisture, soil type and vegetative cover in turn affect these rates. The only really effective way to solve this particular problem is by taking soil samples and analysing them in a laboratory. The conventional approach is to process the samples in the manner detailed in Section 6.2 and then seal the samples in radon gas-tight containers and leave the samples for two and a half weeks to reach secular equilibrium. However, based on work done [Tsela 1997] the suggestion is that there is comparatively little radon emanation and so the samples can be counted as soon as they have been processed into suitable counting vessels, such as Marinelli beakers. This was the approach taken in this project.

Disequilibrium in the thorium decay series is not usually considered a problem because the only gaseous isotope in the decay chain, thoron or ^{220}Rn has a half-life of $T_{1/2} = 55.5\text{ s}$ which will limit the possibilities of the thoron from escaping the matrix in which it is held. Since it does not, it will not then affect the secular equilibrium within the decay chain.

7.2.1. Calibration facilities

As a result of the loss of the calibration facilities at Lanseria airport to the scientific community before the high resolution airborne survey, matters were further complicated; both because it was not possible to calibrate the detector system in the ultra-light airplane in a conventional manner, nor was it then possible to calibrate the MCA used to take a number of *in situ* radiological measurements. With regard to the calibration of the detector system in the ultra-

light airplane, the process of H. Coetzee (NASVD and continuum removal)[Coetzee, 2008] is quite effective but is hampered in those situations where the signal to noise ratio is small. Unfortunately this was the situation at New Machavie especially in the thorium and potassium windows. The result of this particular problem is shown in Figs. 6.8 and Fig. 6.9 where no correlation with one another can be seen. In the case of thorium because the levels actually present on the site, as shown by the soil sample analyses, are low, and in the case of potassium because the difference between the peaks and the lows were relatively small allowing the noise to smother any signal.

7.3. Change in the radiological conditions at the survey site

A particular point of concern was the suspicion that the prevailing radiological conditions at the site had changed between the times the airborne survey was flown and when access to the site for the series of soil samples was gained. See Fig. 7.1 where there is evidence from the satellite image of the movement of slimes.

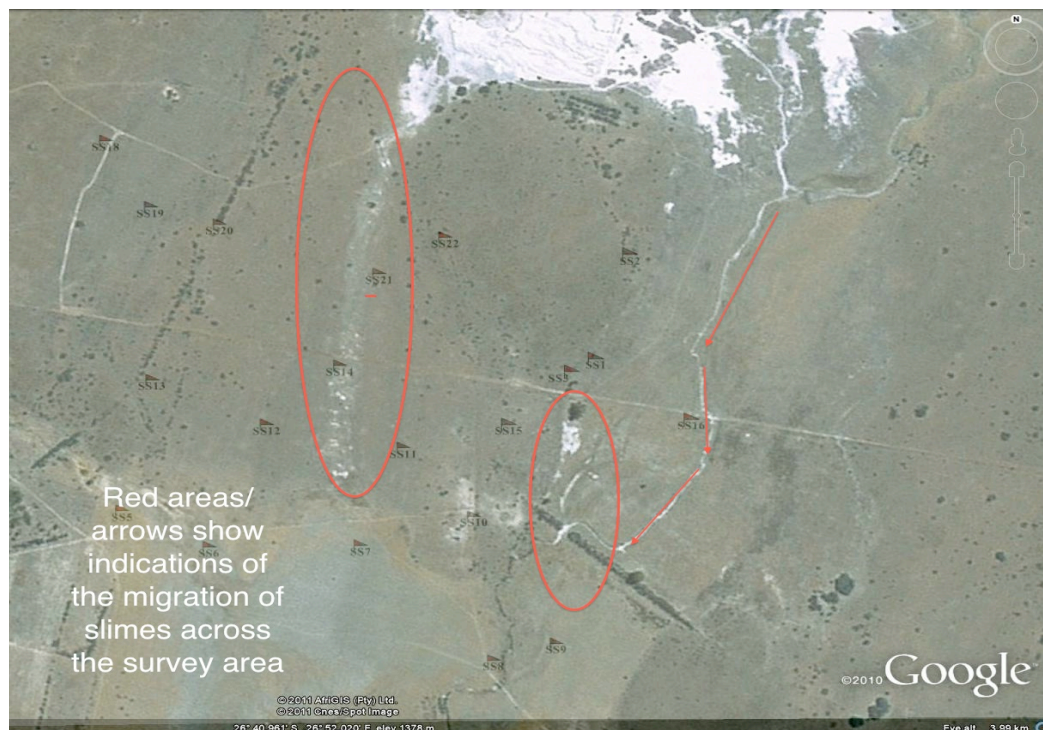


Figure 7-1 Migration of slimes across the survey area

This migration of material is unfortunate since it then renders some of the soil samples less useful, if not entirely valueless. Based on H Coetzee's work at New Machavie, there is good reason to believe that the slimes, which have moved across the site, contain some significant levels of radioactivity. On this basis it was decided to plot two transects across the survey area looking at samples that were less obviously contaminated by slimes. What the results do show is that there is a need to investigate any anomalies that are shown up by the airborne survey by a detailed ground-based survey. The ground-based survey will then be able to establish the types and quantities of radionuclides that are present and from there it will be possible to relate these findings to the airborne survey and identify any shortcomings in assumptions and algorithms that have been used to deconvolute the airborne spectra that have been collected.

7.4. Soil sampling

As was described in Section 6.2, soil samples were taken at various points on the survey site. The selection of these points was based on the total counts map and they were selected to try and cover the range of count rates from highest to lowest. The actual sample points were located in the field by using a hand held GPS unit. One concern was that the unit would not take the person to the correct spot. All of these units have a limit of accuracy that at best is ± 5 m but is always varying. In a simple test a GPS unit was held in a stationary position and was polled every second for 890 s to determine the variation in reported position. As can be seen in Fig. 7.2 the variation in the reported position can be anything up to 25 m. Understanding how the GPS could possibly take the sampler to the 'wrong spot' on the survey area could easily mean that the sample gathered isn't part of the contribution to the airborne count associated to the map reference.

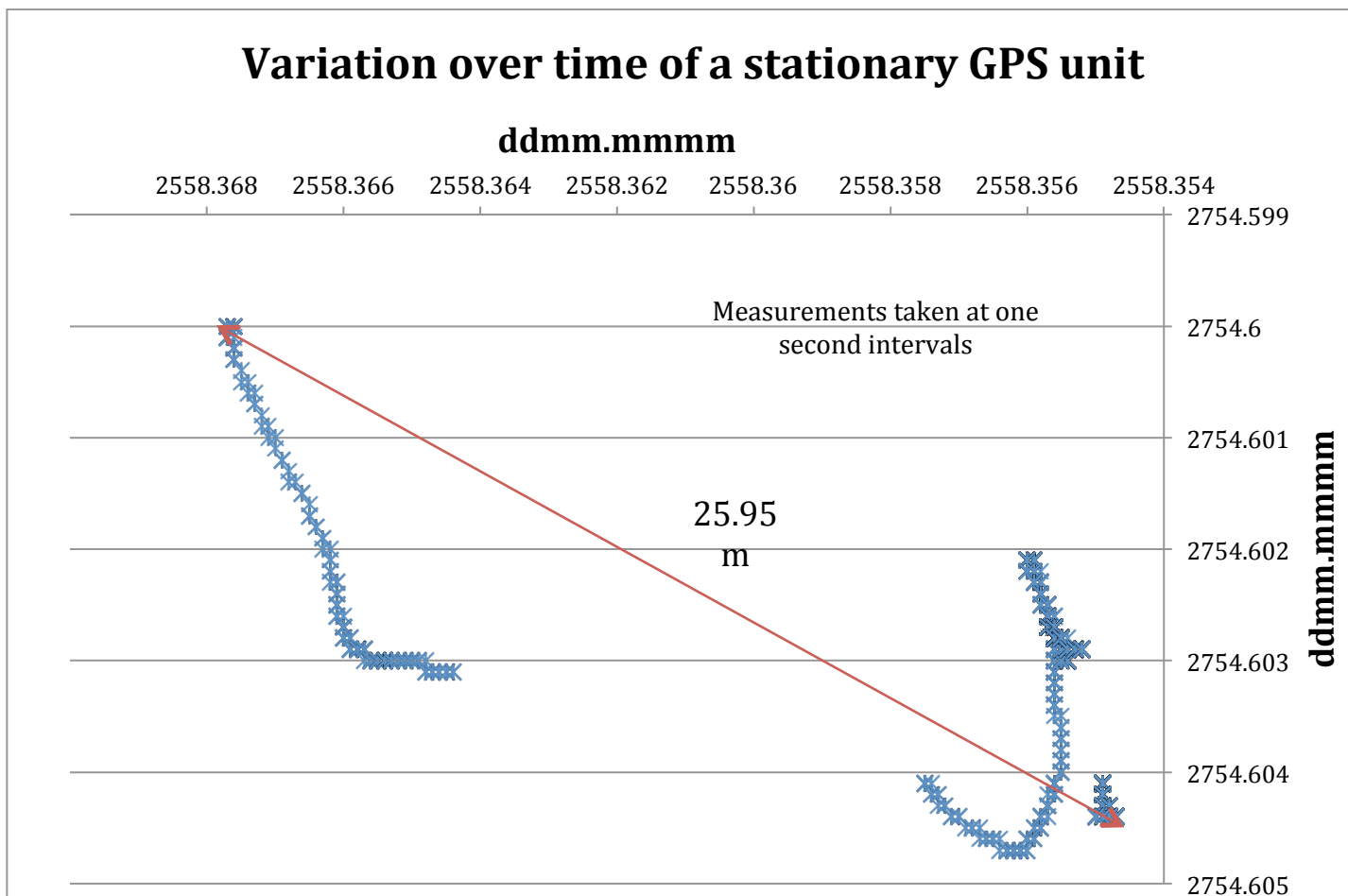


Figure 7-2 Variation over time of a stationary GPS unit.

7.5. Conclusions

The two survey techniques, airborne and ground-based, provide very useful tools for the determination of the distribution of radioactive material within a given area. That being said it is important to realize the limitations placed on each of these methods by technological constraints of the equipment and the physical constraints of the environment in which these survey systems operate. These survey techniques should be considered as part of a whole toolkit of processes which complement and to some extent overlap one another, but the choice of selection of which tool to use will be determined by a host of other considerations.

Airborne surveys are excellent tools for covering large areas of ground in short periods of time which can then give a broad overview of the prevailing radiological conditions. But then to get a more detailed examination of the distribution of radioactive isotopes within an area, it is necessary to use ground-based survey technique which overcomes the limitations of the airborne detection systems previously mentioned.

The demonstration of the correlation between the airborne survey measurements and the ground based soil activity concentrations was non-existent in the case of two of the isotopes considered. However, in the case of uranium there did in fact appear to be a good relationship between the soil samples collected and the airborne measurement taken in the same location, with the exception of the anomaly illustrating subsequent pollution of the site by the slimes dam.

In the case of thorium, this was probably because there was simply not enough of the isotope in the ground, which made any measurements subject to large errors. In which case any correlation between the airborne and ground based surveys that might exist would be completely obscured by the errors associated with the measurements. To determine if such a correlation does exist, it would be interesting to repeat this investigation in an area that has high levels of thorium in the ground, such as Richards Bay in KwaZulu-Natal.

The situation with regard to potassium is somewhat more complicated. In Fig. 6.9 it would appear that there is no correlation between the ground-based survey and

the airborne survey. The possibility of contamination produced by potassium cyanide, a chemical used in the extraction of gold from its ore should also be investigated

Going forward more use should be made of the newer technologies that have become available at a much-reduced cost. Differential GPS units would be used to eliminate the “spoofing” built into the gps signal that handheld units use. Spoofing is a deliberate inaccuracy built into the algorithm used by GPS units to determine their location. This was done at the behest of the American military.

More use of *in situ* multichannel analysers should be made particularly so since the electromechanically cooled intrinsic germanium detection systems that have now come onto the market. This would eliminate the need for assumptions of secular equilibrium as these types of units have significantly improved resolution over the NaI (Tl) based units, which thus far have been the standard tool for these types of surveys. The germanium-based instruments would be able to determine activity concentrations of the entire set of gamma emitting radioisotopes in the uranium and thorium decay chains.

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