

# UKSHS Report No. 4

Soil property and radiometric analytical methods









An Agency within the Department of the

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#### Author(s):

Copplestone D., Wood M. D., Tyler A.\*, Crook P.

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Research Contractor: School of Biological Sciences, University of Liverpool Liverpool, L69 3BX, UK Tel: +44(0) 151 7945291 www.liv.ac.uk/biolsci/ In conjunction with \*University of Stirling

Environment Agency's Project Manager: Dr Peter Crook, Block 1, Government Buildings, Burghill Road, Westbury-on-Trym, Bristol, BS10 6BF

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# **Executive Summary**

This report presents abridged versions of the soil property and radiometric analytical methods (both field and laboratory) utilised within the UK Soil and Herbage Survey (UKSHS). The soil properties covered by this report are:

- pH
- soil bulk density
- soil texture (by hand)
- organic carbon/matter.

The radiometric analytical methods are:

- air kerma dose measurements
- laboratory gamma spectrometry
- *in situ* gamma spectrometry.

All analytical methods (except *in situ* gamma spectrometry) were accredited by the United Kingdom Accreditation Service (UKAS) to ISO17025<sup>1</sup> and all methods were peer reviewed by members of the UK Soil and Herbage Survey Project Board.

<sup>&</sup>lt;sup>1</sup> The University of Liverpool is a UKAS accredited testing laboratory (No. 2049). The opinions and interpretations expressed in this report are outside the scope of accreditation.

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# Glossary of terms

Air kerma Base position Effective stack height Gray (Gy)	<ul> <li>Kinetic energy of electrons released per unit mass of air by photon radiation. It is numerically very close to air dose.</li> <li>South west corner of a northerly orientated 20 m x 20m sampling area from which GPS readings and triangulation bearings were taken.</li> <li>The effective stack height is equal to the physical stack height plus the plume rise.</li> <li>Unit of energy absorption from any type of ionising radiation by any type of medium. When the medium is tissue, it is a unit of dose. When the medium is air, it is a unit of exposure. 1 gray (Gy) = 100 rads.</li> </ul>
Industrial Kerma	A site dominated by some form of industry. <u>Kinetic Energy Released in Ma</u> terials. Kerma is the sum of the initial kinetic energies of all the charged particles liberated by uncharged ionizing radiation (neutrons and photons) in a sample of matter, divided by the mass of the sample. Kerma is expressed in gray (or its submultiples), and, unless otherwise specified, it refers to the energy liberated per unit mass.
Rural Quadrat	All other areas not categorised as industrial, urban, semi-urban or semi- rural. Predominantly agricultural land or undeveloped countryside. A quadrat is a square area which defines the field of study. In the case of
Semi-rural	the UKSHS project a quadrat of size 50cm x 50cm was used as standard. Any area within a small town or village. A small town is taken as being 3– 20 km <sup>2</sup> in area and a village being < 3 km <sup>2</sup> in area.
Semi-urban	All areas that abut urban centres and/or are 25 per cent urbanised/built up. Normally up to 3 km outside the urban core. May also be known as the urban fringe.
Undisturbed site	Unploughed land which has not had chemicals (pesticides/herbicides) applied to it. May include common land, meadows, rough pasture, parkland and fields that are infrequently grazed (if at all). Avoids wooded areas where possible.
Urban	An area which is $\geq$ 90 per cent urbanised/built up. A conurbation may be formed when a large town and city merge. Urban areas include large towns (20–50 km <sup>2</sup> in area) and cities (> 50 km <sup>2</sup> in area).

# 1 Introduction

The UK Soil and Herbage Pollutant Survey (UKSHS) is a research project sponsored jointly by:

- Environment Agency
- Department for Environment, Food and Rural Affairs (Defra)
- National Assembly for Wales
- Food Standards Agency
- Food Standards Agency Scotland
- Scottish Environment Protection Agency (SEPA)
- Environment and Heritage Service (Northern Ireland)
- Scotland and Northern Ireland Forum for Environmental Research (SNIFFER).

Dr Peter Crook from the Environment Agency provided overall project management on behalf of the sponsors. A consortium led by the University of Liverpool's School of Biological Sciences was commissioned to undertake the work. The consortium comprised the Environment Agency's National Laboratory Service (NLS), Nottingham Trent University, the University of Stirling and the University of Liverpool (UoL), with additional assistance being provided by Parkman Ltd and the Environmental Advice Centre (EAC) Ltd.

The project's primary objective was to establish a baseline for pollutant levels in soil and herbage in the UK. The UKSHS has involved the collection of soil and herbage samples for chemical and radiometric analysis from industrial, rural and urban sites throughout the UK. Full details of the number of samples/sites visited and sampling techniques used are given elsewhere (UKSHS Report No. 1 and Report No. 2).

The scale of the UKSHS has resulted in a wealth of methodological information and analytical data. This made the presentation of the whole study in one report unwieldy and a series of 14 stand-alone reports has therefore been prepared that users can read individually or as a complete set. This report describes the soil property analysis and radiometric methods used for the UKSHS and is Report No. 4 in the series. Details of the chemical analysis methodologies are presented in UKSHS Report No. 3. Full details of the other reports in the series can be found on the CD-ROM included with UKSHS Report No. 1 or from the Environment Agency website (www.environment-agency.gov.uk).

The soil property analysis methods presented in this report include:

- pH measurement (Section 3.1);
- soil texture assessment (Section 3.2);
- bulk density determination (Section 3.3);
- organic carbon/matter analysis (Section 3.4).

All methods have been accredited by the United Kingdom Accreditation Service (UKAS) to ISO17025<sup>2</sup>.

<sup>&</sup>lt;sup>2</sup> The University of Liverpool is a UKAS accredited testing laboratory (No. 2049). The opinions and interpretations expressed in this report are outside the scope of accreditation.

The radiometric methods used within the UKSHS included:

- gamma ray air kerma dose rate measurements (Section 4.1);
- laboratory-based gamma ray spectrometry of soil and vegetation samples (Section 4.2);
- *in situ* gamma ray spectrometry (Section 4.3).

The University of Liverpool was responsible for the air kerma dose rate measurements and the laboratory-based gamma ray spectrometry. The radiometric methods used have been accredited by UKAS and all personnel involved in these procedures have been trained to the standard required for accreditation.

The University of Stirling was responsible for the development of the *in situ* gamma spectrometry methods.

The study involved a comparison with results from previous surveys (carried out across the whole or parts of the UK) to identify any trends in the data. An important consideration in selecting and developing the analytical methods used in the UKSHS was therefore to ensure that the methods were similar, or related in a defined way, to those methods used previously. However, this was not a limiting factor in the selection and development of methods, which were based on the most current scientific thinking. All UKSHS methods have been peer reviewed by other technical experts in each field (Appendix 1).

# 2 Aims and objectives

The overall aims and objectives of the UKSHS are detailed in the introductory report (UKSHS Report No. 1). Each report in the series addresses one or more of these aims and objectives. This report addresses the specific aim of producing protocols for soil property and radiometric analysis. These protocols have been produced to achieve the following specific objectives:

- to provide protocols for the assessment of soil properties (bulk density, pH, texture and organic carbon content);
- to provide protocols for the use of *in situ* gamma ray spectrometry as a surveying tool;
- to provide protocols for the determination of air kerma measurements;
- to provide protocols for the drying of soil and herbage samples prior to radiometric analysis;
- to provide protocols for the determination of activity concentrations of a range of natural and anthropogenic radionuclides in soil and herbage samples using laboratory-based gamma ray spectrometry.

The complete protocols were produced as a series of UoL Work Instructions that form part of the UKAS<sup>3</sup> accredited quality system operated by the University of Liverpool's School of Biological Sciences. These detailed documents are 'commercial in confidence' and are for restricted readership only; copies are available on request from the Environment Agency (<u>mailto:enquiries@environment-agency.gov.uk</u>). The method descriptions provided in this report are therefore abridged versions of these documents.

<sup>&</sup>lt;sup>3</sup> The University of Liverpool is a UKAS accredited testing laboratory (No. 2049). The opinions and interpretations expressed in this report are outside the scope of accreditation.

# 3 Protocols for soil property analysis

Soil is a combination of mineral and organic matter, water and air. The soil type is determined by:

- the proportions of these components;
- factors such as the underlying geology from which the mineral component is derived;
- the degree of decomposition of the organic matter.

The properties of a particular soil type have a controlling influence on the behaviour and fate of contaminants entering the soil system. For example, the environmental mobility of many metals is influenced by the pH of the soil and there is a well-documented relationship between soil organic matter content and the ability of that soil to act as a sink for organic chemicals such as polychlorinated biphenyls (PCBs) and dioxins.

In order to interpret data from the UKSHS appropriately, it is necessary to understand the predominant soil properties at each sampling site. The collection of samples specifically for the purposes of soil property analysis was therefore built into the sampling protocols (UKSHS Report No. 2). These samples were brought back to UoL where they underwent the following analyses:

- pH
- soil texture
- bulk density
- organic matter.

These four parameters were selected following expert advice from soil scientists.

The techniques used for the analysis of each soil property were reviewed and agreed by Professor P J Loveland from the Soil Survey and Land Research Centre, Cranfield University prior to their accreditation to ISO17025 by UKAS<sup>4</sup> (Appendix 1).

The following sections provide an overview of each technique. Copies of the full procedures are held by the Environment Agency. Table 3.1 gives details of the relevant documents.

#### Table 3.1 – Documents giving full procedures for soil property analysis

Document title	Document number
Determination of pH of soil	WI10/018
Determination of soil texture by hand	WI10/019
Determination of soil bulk density	WI10/020
Determination of organic carbon and/or organic matter in soil	WI10/021
Determination of organic matter/carbon via loss on ignition	WI10/028

<sup>&</sup>lt;sup>4</sup> The University of Liverpool is a UKAS accredited testing laboratory (No. 2049). The opinions and interpretations expressed in this report are outside the scope of accreditation.

# 3.1 pH measurement

Soil pH is one of the most commonly measured parameters when characterising soil. As well as providing an indication of the soil's mineral content and the organic matter decomposition occurring within it, knowledge of the soil pH assists predictions such as the degree to which contaminants will be mobile within that soil system. The degree of contaminant mobility is an important consideration when assessing the likelihood of a contaminant being brought into contact with a receptor.

Measurements of pH were performed on three 5-cm cores collected from each rural and urban site. For industrial sites, one core was collected from each sampling point. Once collected, each core was wrapped in foil, bagged and packed into a cool box for transfer to UoL. On receipt at the laboratory, the samples were dried and homogenised before a soil suspension was prepared for standardised pH determination.

The pH measurement of a 1:5 (volume/volume (v/v)) soil suspension was taken using a Mettler-Toledo MPC227 pH meter, calibrated in accordance with the manufacturer's instructions and fitted with an InLab®413 pH combination polymer electrode.

# 3.2 Soil texture assessment

Soil texture is a property of the mineral fine earth (<2 mm) fraction, which in turn depends on the particle size distribution.

A soil is allocated to a textural class depending on its content of sand, silt and clay sized particles. Sand, silt, clay particles and organic matter impart distinctive qualities to the feel of the soil.

Textural class can be determined subjectively from the feel of a moist soil moulded between the fingers and thumb. An experienced soil scientist will come to know the feel of the textual classes and, with the use of a soil texture flow chart, be able to key out<sup>5</sup> each sample. But because soil texture describes the mineral fraction of the soil (sand, silt and clay), it is not appropriate to determine soil texture by hand on a peat or organic soil. The presence of a high proportion of organic matter in a soil sample tends to impart a 'feel' to the soil during hand texturing which influences the assessor to allocate a 'finer than actual' classification to the soil. Thus, the presence of high organic matter in a sample is noted during the hand texturing evaluation.

The physical and chemical characteristics of soils (e.g. moisture retention, leaching, aeration, cation exchange capacity and anion adsorption) are influenced by whether they are predominantly sandy, silty or clayey in texture.

A soil flow chart/key groups the 12 soil classes recognised in the Hodgson (1974) quantitative texture procedure into four main groups:

- sandy soils
- silty soils
- silty and clayey soils;
- clay soils.

These four classes are used in the interpretation of the overall physical and chemical characteristics of samples.

<sup>&</sup>lt;sup>5</sup> A term used in biology meaning to name/identify/discover/determine/describe.

Determination of soil texture was conducted on unprepared soil samples. Three samples were collected from each rural and urban site, and one from each of the four points around an industrial site. Each soil sample for texture determination consisted of approximately one full trowel of soil placed in a zip lock plastic bag. These samples were returned to the laboratory and stored in plastic containers at <4°C until the soil texture assessment could be carried out.

Soils samples were allowed to reach room temperature before a texture assessment was performed. Internal quality control auditors laid out the samples and included a number of randomly selected duplicates. The assessors then approached the samples in a blind test. The assessors worked the soil in their hands, adding water as necessary until the soil reached the sticky point. The assessors then worked through the soil texture assessment key to determine the appropriate soil texture classification for the soil. Four major classification types and 12 sub-classes were used to describe soil texture (Table 3.2).

Unlike the measurement of pH, soil texture is a more subjective measure that relies on the high level of skill and knowledge of the person undertaking the assessment. As with all other sampling and analysis techniques associated with the UKSHS, all personnel undertaking soil texture assessment had to be specifically trained and assessed to ensure that they could perform the test in line with the standards laid down under the ISO17025 accreditation. The training included an assessment to ensure that the workers could identify a range of soil textures correctly through a series of repeat runs.

Soil texture class	Soil texture sub-class
Sandy textured soils	Sand
	Loamy sand
	Sandy loam
Silty textured soils	Silt loam
	Silt
	Loam
Silty and clayey textured soils	Sandy clay loam
	Silty clay loam
	Clay loam
Clay textured soils	Sandy clay
	Silty clay
	Clay

#### Table 3.2 Soil texture classifications used in the UKSHS

# 3.3 Bulk density

Soil bulk density, also known as apparent density, describes the mass of soil and its total volume (Allen 1989).

Measures of soil bulk density provide an indication of the porosity of the soil. This can be correlated with the drainage properties of the soil, which may influence contaminant movement. Additionally, soil bulk density can provide corroborative evidence for the soil texture assessment and an indication of the compaction of the soil.

The underlying geology can influence the mineral content of the soil and this may affect the measurement, because different minerals will have different densities. Furthermore, high levels of organic matter can also lead to significant variation in bulk density because the density can be heterogeneous over short distances.

The soil bulk density is calculated using equation 1:

Density 
$$(g \text{ cm}^{-3}) = \text{mass } (g)/\text{volume } (\text{cm}^{-3})$$
 (1)

A known volume of soil was collected using a double cylinder corer. The internal cylinder was used to contain the core and the outer cylinder had a sharp cutting edge to minimise soil compression as the corer was driven into the soil.

Due to the heterogeneous nature of soils in the field, bulk density will vary from place to place – even within the same sampling horizon. Three cores were collected with a bulk density corer from the rural and urban sites, and one core was collected from each sampling point around an industrial site. The cores were extracted, wrapped in aluminium foil and returned to the laboratory for analysis. The water content at the time of sampling was also considered due to the potential for swelling and shrinking of soils.

The mass of the soil was measured in the laboratory. Samples were then dried to constant weight and the dry weight mass recorded. Equation 1 was then applied to obtain the soil bulk density.

# 3.4 Organic matter/carbon content

The organic matter/carbon content of a soil will influence the fate of many of the contaminants that come into contact with it. The tendency for contaminants such as dioxins to be associated with organic matter content was noted above.

Two methods were used to determine the soil organic matter/carbon content:

- a wet oxidation method;
- a loss-on-ignition method (LOI).

The material used for analysis was taken from the cores described in Section 3.1.

Wet oxidation is particularly good for samples with a low organic matter/carbon content, but it is less useful for those where the organic matter/carbon content is high. The wet oxidation method is a titration-based approach and soils containing high level of organic matter quickly cause the titration to go over range. The assessment then has to be repeated on progressively smaller concentrations until a result can be achieved. This can lead to reduced accuracy due to the heterogeneous nature of the soil samples.

By comparison, LOI is more accurate at higher organic matter/carbon concentrations and less accurate when the organic matter/carbon content of the soil is low. Therefore, all samples from the UKSHS were analysed using LOI in the first instance and samples that were found to contain low organic matter/carbon content were re-analysed using the wet oxidation method.

## 3.4.1 Wet oxidation method

The soil sample is transferred to a conical flask. Potassium dichromate and concentrated sulphuric acid are added, the contents mixed and the flask left to stand. Deionised water, orthophosphoric acid and barium diphenylamine sulphonate are then added and the contents mixed again. A burette filled with ferrous sulphate solution is titrated against the remaining potassium dichromate solution in the flask. A blue colour is produced just before the end-point and this then changes to bright green.

The organic matter content is calculated using the equation 2:

Percentage organic matter = 
$$\frac{15(1 - x/b) \times 0.67}{m}$$
 (2)

where:

- *x* is the volume of ferrous sulphate used in titre (ml)
- b is the volume of ferrous sulphate used in standardisation (ml)
- m is weight of sample taken (g).

The organic carbon content is determined using the equation 3:

Percentage organic carbon content = 
$$\frac{15(1 - x/b) \times 0.67}{m \times 1.72}$$
 (3)

#### where:

- *x* is the volume of ferrous sulphate used in titre (ml)
- b is the volume of ferrous sulphate used in standardisation (ml)
- m is weight of sample taken (g)
- 1.72 is a factor used to convert organic matter to organic carbon.

### 3.4.2 LOI method

A known mass of sample is placed in a pre-weighed crucible and dried in an oven at 100°C. The sample is cooled before being placed in an electric muffle furnace for 2 hours at 550°C. The crucible and sample are then cooled and re-weighed.

The organic matter content is calculated using the equation 4:

Percentage organic matter = 
$$\underline{m_3 - m_4 \times 100}_{m_3 - m_c}$$
 (4)

where:

- m<sub>3</sub> is the mass of the crucible and oven dry soil (g)
- m<sub>4</sub> is the mass of the crucible and soil after ignition (g)
- $m_c$  is the mass of the empty crucible (g).

The organic carbon content is calculated using equation 5:

Percentage organic carbon content = 
$$(\underline{m_3 - m_4}) \times 100$$
  
 $(\underline{m_3 - m_c}) \times 1.72$  (5)

where:

m <sub>3</sub>	is the mass of the crucible and oven dry soil (g)
m <sub>4</sub>	is the mass of the crucible and soil after ignition (g)
m <sub>c</sub>	is the mass of the empty crucible (g)
1.72	is a factor used to convert organic matter to organic carbon.

# 4 Protocols for radiometric analysis

The techniques used in the radiometric component of the UKSHS were provided by:

- University of Liverpool air kerma and laboratory-based gamma spectrometry for soil and herbage samples;
- University of Stirling *in situ* gamma spectrometry.

The techniques were peer-reviewed and agreed by Dr Bernie Wilkins from the National Radiological Protection Board (NRPB) (Appendix 1). The techniques used by the University of Liverpool are UKAS-accredited to ISO17025<sup>6</sup>.

The following sections provide an overview of each technique. Copies of the full procedures are held by the Environment Agency. Table 4.1 gives details of the documents.

#### Table 4.1 – Documents giving full procedures for radiometric analysis

Document title	Document number
Determination of the activity concentration of radionuclides in test samples by high resolution gamma ray spectrometry	WI10/001
Measurement and interpretation of gamma ray air kerma rates	WI10/002

# 4.1 Air kerma dose measurements

Air kerma measurements were taken *in situ* by the field team using a Mini-Instruments Environmental Monitor Type 6-80 fitted with a MC71 GM detector and tripod. This was calibrated regularly by NRPB and checked before and after use. Three air kerma dose readings were taken per rural or urban site. Where requested, a single air kerma measurement was made at each sampling point for the industrial sites.

The count time for each measurement was 600 seconds with the detector held vertically at a height of 1 m  $\pm$  0.2 m following the guidance provided by HMIP (1995). Figure 4.1 shows the location of air kerma and *in situ* gamma spectrometry measurements across each site. The readings were taken 30–50 m apart. One reading was centred on the 20 m x 20 m area used for the soil and herbage sampling (UKSHS Report No. 2).

The cores for the radiometric laboratory analysis were collected only after the air kerma and any *in situ* gamma spectrometry measurements had been taken.

The total terrestrial gamma air kerma rate was determined using equation 6:

$$D = \left(\frac{N - P}{K}\right)$$

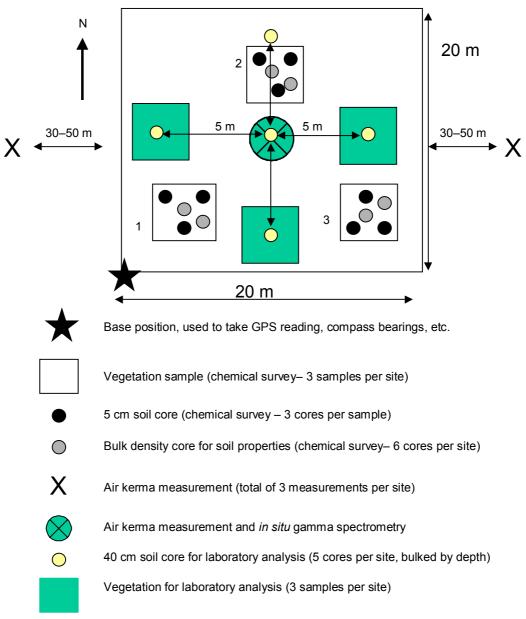
where:

(6)

<sup>&</sup>lt;sup>6</sup> The University of Liverpool is a UKAS accredited testing laboratory (No. 2049). The opinions and interpretations expressed in this report are outside the scope of accreditation.

- D is the calculated air kerma rate ( $\mu$ Gy h<sup>-1</sup>)
- is the observed (<300 s<sup>-1</sup>) or corrected (>300 s<sup>-1</sup>) count rate (s<sup>-1</sup>), the corrected Ν count rate is determined by correcting for dead time if the count rate >300  $s^{-1}$ . Р
- is the sum of cosmic and inherent count rate  $(s^{-1})$ .
- Κ is the response, i.e. the appropriate conversion factor from counts per second to air kerma rate ( $\mu$ Gy h<sup>-1</sup>). This value is stated on the calibration certificate.

The response of the detector to both <sup>226</sup>Ra and <sup>137</sup>Cs was determined by NRPB during the detector calibration. The detector response to <sup>226</sup>Ra was used to calculate the air kerma rate for all sites included in the survey. The air kerma rate was also calculated using the <sup>137</sup>Cs response factor, but this was not used in UKSHS Report No. 11.



Note: for in situ gamma spectrometry calibration sites, an additional set of 40 cm cores should be taken at a distance of 8 m from the central point.

# Figure 4.1 – Diagram of a rural or urban site showing where the radiometric field measurements were taken

# 4.2 Laboratory-based gamma spectrometry (University of Liverpool)

Laboratory gamma spectrometry at the University of Liverpool was used to determine the levels of the radionuclides listed in Table 4.2 in samples of soil and herbage collected from rural, urban and industrial sites within the UKSHS. Full details on the number of sites and samples collected and the methods of sample collection are given elsewhere (UKSHS Report No. 1 and Report No. 2).

Isotope	Data reported in:	LOD value <sup>*</sup>
Co-60	kBq m <sup>-2</sup> Bq kg <sup>-1</sup> (dry) Bq kg <sup>-1</sup> (wet) kBq m <sup>-2</sup>	0.2
Cs-137	kBq m <sup>-2</sup> Bq kg <sup>-1</sup> (dry) Bq kg <sup>-1</sup> (wet) kBq m <sup>-2</sup>	0.2
Cs-134	$kBq m^{-2}$ $Bq kg^{-1} (dry)$	0.2
Am-241	Bq kg <sup>-1</sup> (dry) Bq kg <sup>-1</sup> (wet) kBq m <sup>-2</sup> Bq kg <sup>-1</sup> (dry)	0.2
K-40	Bq kg <sup>-1</sup> (dry) Bq kg <sup>-1</sup> (wet) Bq kg <sup>-1</sup> (dry) Bq kg <sup>-1</sup> (wet)	3.2
Th-232 decay series		
Ac-228	Bq kg <sup>-1</sup> (dry) Bq kg <sup>-1</sup> (wet) Bq kg <sup>-1</sup> (dry)	0.6
Ra-224	Ba ka <sup>-1</sup> (wet)	
Pb-212	Bq kg <sup>-1</sup> (dry) Bq kq <sup>-1</sup> (wet)	0.4
Bi-212	Bq kg <sup>−1</sup> (dry)́ Bq kg <sup>−1</sup> (wet)	2.3
TI-208	Bq kg <sup>−1</sup> (dry) Bq kg <sup>−1</sup> (wet)	
U-238 decay series		
Pa-234	Bq kg <sup>−1</sup> (dry) Bq kg <sup>−1</sup> (wet)	
Ra-226	Bq kg <sup><math>-1</math></sup> (dry) Bq kg <sup><math>-1</math></sup> (wet)	4.4
Bi-214	Bq kg <sup><math>-1</math></sup> (dry) Bq kg <sup><math>-1</math></sup> (wet)	0.4
Pb-214	Bq kg <sup><math>-1</math></sup> (dry) Bq kg <sup><math>-1</math></sup> (wet)	0.4
Pb-210	$Bq kg^{-1} (dry)$	

#### Table 4.2 – Radionuclides reported in the UKSHS

	Bq kg <sup>-1</sup> (wet)
U-235 decay series	
U-235	Bq kg <sup>-1</sup> (dry) Bq kg <sup>-1</sup> (wet)

\* LOD calculation based on a sample of 400 g and a live time of 100,000 seconds.

# 4.2.1 Principles of gamma spectrometry

Gamma spectrometry is based on the principle that a proportion of the gamma ray photons incident on a detector interact with atoms within the detector crystal (germanium) in a process known as photoelectric absorption (Krane 1988, Knoll 1989). If the entire energy of the incident gamma ray photon is absorbed within the detector crystal, a photoelectron of a discrete energy equal to that of the incident gamma ray minus the binding energy of the electron in its original shell is produced (Mann *et al.* 1980). Any photoelectrons produced are collected within the crystal and the kinetic energy of the photoelectron produces a peak in the energy spectrum. This peak is proportional to the energy of the incident gamma ray because of the relationship between the photoelectron and the gamma ray. The energy from incident gamma rays on a detector may be retained in the crystal in a number of other ways.

The first of these is by Compton scattering. This occurs as a result of incomplete charge collection within the detector crystal, i.e. only part of the energy of the incident gamma ray is transferred to form a photoelectron within the detector crystal. This photoelectron will have a lower energy than the incident gamma ray because of the incomplete gamma ray absorption.

Incomplete charge collection can occur for a number of reasons. For example, a gamma ray may pass through the detector crystal, thus imparting only a small proportion of its energy to the detector crystal. Alternatively, as the gamma ray passes through the detector, it may be deflected and thus pass out of the crystal again resulting in incomplete charge collection (Knoll 1989). Because this can result in a range of energy peaks in the spectrum, a continuous energy spectrum is produced.

The magnitude and importance of Compton scattering depends, to some extent, upon the count rate of the sample. As an example of the large number of gamma ray/detector interactions that result in the formation of a Compton background, Figure 4.2 shows the true peaks sitting on the Compton background in a typical soil spectrum obtained during this study.

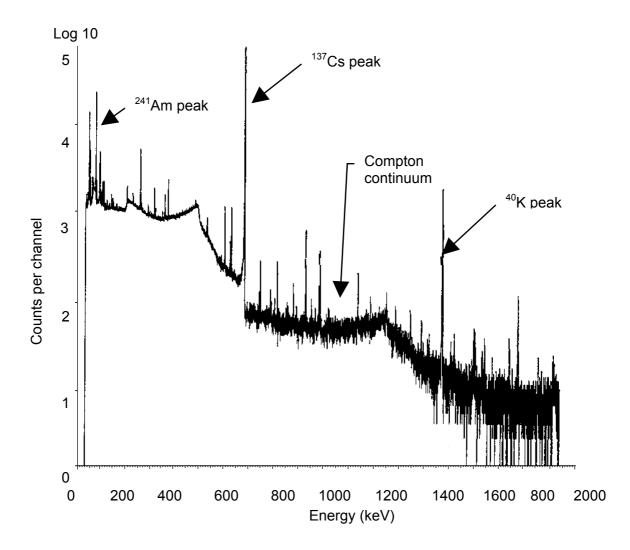
A third significant gamma-ray interaction is pair production. Pair production is the process by which a positron–electron pair is created when a gamma ray photon interacts in the field of a nucleus of the absorbing material. A minimum energy of 1.02 MeV is required to make this interaction energetically viable. Any excess energy over and above 1.02 MeV manifests itself in the form of kinetic energy, which dissipates over a very short distance. As the kinetic energy decreases, the positron becomes unstable and eventually annihilates with the electron. Two annihilation photons of energy 511 keV are then produced, resulting in a 511 keV peak in the energy spectrum (Knoll 1989).

Other processes that occur during sample acquisition involve count summation. There are two forms of summation – random and true coincidence summing (TCS). Random summing results from the chance of any two gamma ray photons from the same or different radionuclides being incident upon the detectors at, or within, the resolving time of the detectors. As a result, random summing is affected by the sample activity. As the sample activity increases, so the probability of two gamma rays being incident at the same time also increases.

True coincidence summing, on the other hand, occurs when gamma ray photons emitted from the different nucleus energy levels of the same radionuclide follow one another very rapidly

(usually within a few milliseconds). When this occurs, there is a chance that the two gamma ray photons will be incident upon the germanium crystal and be measured together. This is independent of the count rate and can, therefore, be a significant source of error at all sample activities.

In both cases, summing results in a detected event with the combined energy of the two gamma rays (Gilmore and Hemingway 1995). This reduces the number of counts that should be present in the true nuclide peak and, unless steps are taken to correct the problem, will result in an underestimation of the true sample activity.



# Figure 4.2 – Typical spectrum obtained after counting a soil sample showing <sup>40</sup>K, <sup>137</sup>Cs and <sup>241</sup>Am peaks sitting on the Compton continuum

Little can be done to reduce the effect of random summing, apart from increasing the distance of the sample from the detector for samples of very high activity. Since gamma ray emission occurs in all directions, increasing the distance of the sample from the detector effectively reduces the detector area for interactions. The result is a decrease in the number of gamma ray photons incident upon the detector and hence a reduction in the chance of two incident photons.

Fortunately, random summing is only important at very high sample activities – much greater than the environmental levels found within the UKSHS.

True coincidence summing can be corrected by using single isotope calibration sources and counting them at different heights above the detector. This allows correction factors to be calculated for individual radionuclides. During this survey, TCS corrections were applied through the analysis software.

## 4.2.2 Sample counting

Soil and herbage samples were oven dried at 60°C to constant dry weight before being homogenised in a rotary mill or commercial blender for soil and herbage, respectively. The homogenised material was then transferred to a 330 ml volume Marinelli beaker, which was sealed with wax. The samples were then left for three weeks before being counted on n-type high purity germanium (HPGe) detectors. Table 4.3 summarises the main properties of the nine detectors used during the survey. Herbage samples were counted for a minimum of 24 hours and soil samples for a minimum of eight hours.

Parameter	Specification
Detector type	n-type HPGe coaxial
Manufacturer	EG&G Ortec
FWHM at 1.33 MeV	<1.90 to 2.50 keV
Relative efficiency	20–70 per cent
Arrangement	Horizontal and vertical

Table 4.3	Gamma detector specifications
-----------	-------------------------------

Each detector was surrounded by a 100 mm thick cylinder of lead with two inner rings of cadmium and then copper, both between 2 and 3 mm thick. These reduced the background radiation incident on the detectors. The detectors were cooled with liquid nitrogen to reduce thermal-induced leakage current and to improve the energy resolution of the detector. This cooling to a temperature of approximately 77K was achieved through the use of an insulated Dewar containing a reservoir of liquid nitrogen, which was kept in thermal contact with the detector crystal via a cryostat.

## 4.2.3 Detector calibration – energy calibration

Energy calibration relates the multi-channel analyser (MCA) channel number to the specific energy of an incident gamma ray photon. This is necessary to allow correct identification of the peaks present within a spectrum. An energy calibration was performed weekly using a certified multi-nuclide source, which was counted for 900 seconds (15 minutes).

Efficiency calibrations consist of two components. The first is related to the proportion of counts registered per gamma ray incident on the detector. This relates to the emission energy of the gamma ray. The second is a component related to the geometry of the sample. This measures the proportion of gamma rays incident on the detector from each radioactive decay event within the sample matrix.

Efficiency calibrations were performed by counting a standard of known radionuclide content and activity and with a fixed and standardised geometry on each detector. Count times for the

standards were for 1,500 seconds (i.e. 25 minutes). Efficiency calibrations were carried out monthly.

# 4.2.4 Measurement of background gamma radiation within the detector chamber

Cosmic radiation and natural terrestrial radiation present in the environment form a continuous background signal to all gamma radiation detectors. The magnitude of the signal depends upon the location of the radiometric laboratory, detector types and size and the degree of shielding placed around the detector crystal. Some of the natural radionuclides contributing to this type of background radiation are ubiquitous within the environment and are often incorporated, albeit at very low levels, within the materials used to manufacture both the detector casing and the shielding which surrounds it.

Surrounding the detector with lead shielding can effectively reduce the background signal from natural radiation. Lead is a common shielding material because it is very effective at stopping the passage of gamma rays. In this case, the lead shielding was supplemented by a cadmium layer and then a copper layer to form a graded shield. Graded shields are employed because X-rays are produced by irradiation of the lead by both the sample and by external background radiation. The cadmium and copper layers absorb these lead X-rays and themselves emit only low energy or weakly penetrating X-rays, which do not interact significantly with the detector.

Background measurements were taken at monthly intervals by counting an unused Marinelli beaker for 220,000 seconds (i.e. 60 hours).

## 4.2.5 Data analysis and activity calculations

Sample spectra were saved to disk using EG&G Ortec GammaVision®-32 software. The spectra were then interrogated and analysed using this software. Typically, this involved locating peaks and recording information concerning the peak area, channel centroid and fill width half maximum (FWHM) (Figure 4.3) in a logbook.

Having set regions of interest, automatic routines within GammaVision-32 were used to calculate and subtract the Compton background from the peaks. The software selected an optimum number of channels on either side of the region of interest and then assessed the Compton background component before its subtraction.

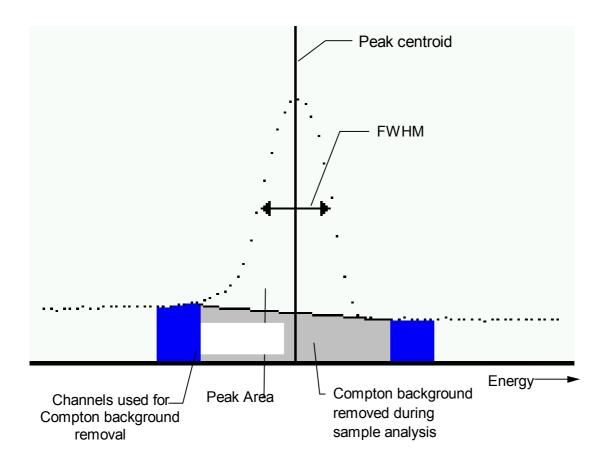
The specific activity of each radionuclide were calculated using equation 7:

Activity (A) = 
$$\underline{G-B}$$
 (7)  
DE × RA × Mass × LT

where:

А	=	activity of the sample in Bq kg <sup>-1</sup> or Bq g <sup>-1</sup>
G	=	gross counts in peak
В	=	background counts in peak (true peak background counts)
DE	=	fractional detector counting efficiency
RA	=	relative abundance of a gamma ray at a specific energy (keV) as a fraction
Mass	=	mass of the sample in kg or g
LT	=	live time in seconds.
DE RA Mass	= = =	fractional detector counting efficiency relative abundance of a gamma ray at a specific energy (keV) as a fraction mass of the sample in kg or g

The background count in the peak (B) was determined by multiplying the live time of the sample count by the relevant background count rate (cps) as determined from a background count. Peak background counts were determined by counting empty Marinelli beakers. Data were analysed using GammaVision-32.



### Figure 4.3 – Stylised spectrum peak

# 4.3 *In situ* gamma spectrometry (University of Stirling)

### 4.3.1 Introduction

The *in situ* gamma spectrometry method was agreed before it was applied to the UKSHS.

The method used for gamma dose rate determination was checked and validated through a European intercomparison exercise organised by the European Radiation Dosimetry Group (EURADOS) in September 2002.

In addition, the working calibration was compared with measurements made during an international comparison exercise, Resumé 2002 (Sanderson *et al.* 2002), conducted in the Dumfries and Galloway region of Scotland as part of the ECCOMAGS (European Calibration and Co-ordination of Mobile and Airborne Gamma Spectrometry) EU thematic network project (<u>www.gla.ac.uk/ECCOMAGS/</u>). This enabled a direct comparison of the working calibration derived for the UKSHS with other national and international measurements of specific radioactivity concentrations, inventories and gamma dose rates.

*In situ* gamma ray spectrometry surveys are well-established techniques for monitoring and mapping of anthropogenic and naturally occurring radionuclides (e.g. Beck *et al.* 1972, ICRU 1994). However, comparison of estimates of environmental radioactivity derived from soil samples with those from *in situ* have been hindered by issues of:

- spatial variability (IAEA 1976);
- variations in the vertical activity distribution within the soil profile (ICRU 1994).

Sampling strategies that account for spatial variability and enable effective comparisons with the spatial response of gamma detectors have been reported by Tyler *et al.* (1996a); these are discussed below. However, the vertical activity distribution in the environment may not be the same as observed at the calibration point resulting in a substantial reduction in the accuracy of the *in situ* measurement (Tyler *et al.* 1996b).

A solution to this problem has been developed whereby a spectrally derived coefficient, *Q*, is used to estimate changes in the vertical radionuclide distribution and thereby permit the use of an appropriate calibration coefficient. This procedure was applied by Zombori (1992a,b) to estimate changes in Chernobyl-derived <sup>137</sup>Cs inventory with depth in terrestrial environments; these were typically characterised by exponential decreases in activity concentration with depth.

Tyler *et al.* (1996b) applied a similar approach to salt marsh environments, which exhibited significant variations in subsurface maxima of <sup>137</sup>Cs specific activity. The approach has also been used to estimate sediment accretion rates in salt marsh environments (Tyler, 1999). Similar approaches have been applied to ploughed field systems (Tyler *et al.* 2001).

## 4.3.2 Soil coring for detector calibration

Spatial variability is an important aspect of the distribution of radionuclides and subsequent sampling errors (Tyler *et al.* 1996a). An *in situ* detector at 1 m above the ground has a field of view of about 10 m radius ( $314 \text{ m}^2$ ) at 662 keV for a typical exponential decrease in specific activity with depth. This translates to an effective sample of about 100 m<sup>3</sup> or 130,000 kg. In contrast, a soil sample typically represents about 2.6 x  $10^{-3} \text{ m}^3$  or 3 kg.

When comparing two measurement techniques with very different spatial sensitivities, it is therefore essential to use an effective sampling methodology to enable the two approaches to be statistically comparable for calibration and validation purposes in environments that are typically heterogeneous.

In order to calibrate the *in situ* gamma spectroscopy system for varying soil types across the UK, 10 locations across the UK were selected and surveyed (Table 4.4). These sites were preselected to cover:

- a range of geological settings and thus primordial radionuclide concentration;
- soil types;
- different contamination histories (e.g. from weapons testing and Chernobyl).

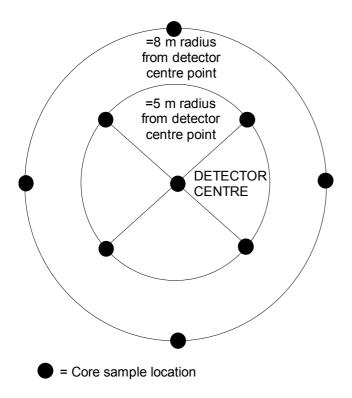
Site no.	Site location	Soil type	Likely natural background	Likely <sup>137</sup> Cs inventory
2	Margate, SE England	Brown soil	200–500 Bq kg <sup>-1</sup>	<2 kBq m <sup>-2</sup>
5	Penrith, W England	Surface water gley	200–500 Bq kg <sup>-1</sup>	2–10 kBq m <sup>-2</sup>
7	Windermere, NW England	Podzolic soil	200–500 Bq kg <sup>-1</sup>	>2 kBq m <sup>-2</sup>
14	Brigg, E England	Surface water gley	200–500 Bq kg <sup>-1</sup>	<2 kBq m <sup>-2</sup>
16	Oldham, W England	Surface water gley	200–500 Bq kg <sup>-1</sup>	<10 kBq m <sup>-2</sup>
32	Evesham, Central England	Pelosols	200–500 Bq kg <sup>-1</sup>	<2 kBq m <sup>-2</sup>
53	Penzance, Cornwall	Podzolic soil	>500 Bq kg <sup>-1</sup>	<2 kBq m <sup>-2</sup>
164*	Sorbie, SW Scotland	Brown soil	200–500 Bq kg <sup>-1</sup>	>10 kBq m <sup>-2</sup>
171	Eaglesham, W Scotland	Groundwater gley	<200 Bq kg <sup>-1</sup>	>2 kBq m <sup>-2</sup>
185	Aviemore, Scotland	Peat soil	<500 Bq kg <sup>-1</sup>	2–10 kBq m <sup>-2</sup>

 Table 4.4
 Calibration site locations

\*Intercomparison site with ECCOMAGS RESUMÉ 2002 project (Sanderson et al. 2002).

At each calibration location, nine soil cores were sampled (Figure 4.4) following the protocol given in UKSHS Report No. 2:

- one at the centre of the location;
- four at a distance of 5 m from the centre;
- an additional four at 8 m from the centre.





The cores were sectioned into the following depth increments: 0–5 cm; 5–10 cm; 10–15 cm; 15–20 cm; 20–30 cm; and 30–40 cm.

Soil moisture and bulk density for each slice were recorded in the laboratory. The specific radioactivity concentration and associated inventories from each core were determined (see Section 4.2) and the depth distributions characterised in terms of mass per unit area. This takes into consideration the field moist bulk density (i.e. the bulk density of the soil including moisture content at the time of sampling).

The results were spatially integrated to match the spatial response of the *in situ* detector for calibration purposes as described by Tyler *et al.* (1996a).

## 4.3.3 *In situ* gamma spectrometry approach

### Equipment

The University of Stirling operates an Ortec 35 per cent relative efficiency n-type HPGe detector. The detector crystal is cooled with liquid nitrogen using a portable 5 litre Dewar, providing up to 72-hours of operation time before a refill is required. The detector and Dewar are collectively called the 'gamma gauge'. The gamma gauge is supported from a purpose-built tripod, providing a downward-looking configuration and the possibility of support at a variety of heights.

Normally the crystal is supported at 1 m above the ground, providing a nominal field of view of 10 m radius (Tyler *et al.* 1996a). The detector is powered by an EG&G Nomad Plus portable spectrometry system. This system provides the high voltage (HV) power supply to the detector for up to 8 hours per day. The Nomad Plus system incorporates a high quality amplifier, a multi-channel analyser and a laptop computer running GammaVision-32. This is also used to analyse the spectra collected and to control the detector and electronics.

### Detector performance monitoring pre-survey

Before equipment was used in the field, the detector performance was characterised in the laboratory. This included:

- a pulse zero check;
- energy calibration using a National Physical Laboratory (NPL) mixed gamma source;
- a spectral resolution check.

A background count was also acquired within 10 cm thick lead shielding. Further background checks were undertaken at the end of the UKSHS at the Physikalisch-Technische Bundesanstalt (PTB) in Germany, as part of a EURADOS intercomparison exercise.

### Daily survey checks

During the survey, the detector was checked every morning and *an in situ* detector quality assurance sheet was completed daily. During the survey period, 141 such sheets were completed. Each sheet provided a check on:

- energy resolution (FWHM) at 662 keV (nominally 1.45 keV);
- the time required for the detector to stabilise to reach the required energy resolution;

- a measure of relative efficiency from an International Atomic Energy Agency (IAEA) 375 reference standard;
- peak location (energy calibration);
- detector dead time (around 2 per cent).

These measures provided:

- a quality control on the detector output;
- the possibility of identifying the onset of problems enabling early rectification, e.g. increasing time required for detector stabilisation following the application of the HV.

Each spectrum collected was saved and two copies were backed up onto floppy disks.

The in situ detector measurement sheet included;

- a cross-reference to each daily in situ detector quality assurance sheet;
- details about the site, e.g. a description of the weather conditions;
- a check on detector resolution (at 662 keV and 1,460 keV) from each spectrum collected;
- peak location;
- dead time.

The acquisition time for each measurement was set as a standard at 7,200 seconds.

### Detector transportation

A specially modified Ford Transit van was used to transport liquid nitrogen (around 90 litres) around the survey project sites. The van was designed to comply with Health and Safety Executive (HSE) regulations. In addition, the vehicle carried dangerous goods notes for the transportation of liquid nitrogen on ferries for survey work taking place on islands.

### Quantification of radionuclide depth distributions

The following text is adapted from Tyler et al. (1996b).

### Exponential depth distributions

Beck *et al.* (1972) introduced the approximation of the exponential decrease of activity with depth, typically observed in <sup>137</sup>Cs fallout from atmospheric weapons testing. The parameter,  $\alpha$  (cm<sup>-1</sup>), was defined as the reciprocal of the relaxation length of the exponential distribution such that:

$$A = A_o \cdot e^{-\alpha z} \tag{8}$$

where:

z is linear depth (cm)

 $A_o$  is the specific activity measured at the surface (Bq kg<sup>-1</sup>).

However, gamma photon flux rates in soil are a function of the soil density ( $\rho$ ) in addition to source depth, *z*. Thus, the preference may be to quantify source burial in terms of mass depth or mass per unit area (g cm<sup>-2</sup>), which is given by  $\rho z$  (ICRU 1994). Given that the soil density varies with depth and the depth intervals selected also tend to vary, then the combined effect will lead to further variations in  $\rho z$  within the soil column.

The function mass per unit area ( $\xi$ ) is given by equation 9:

$$\xi = \int_{0}^{z} \rho(z') dz'$$
(9)

where:

ξ

is the total mass depth or effective mass per unit area (g  $cm^{-2}$ ).

The relaxation mass per unit area or mean mass depth is defined as ß (g cm<sup>-2</sup>), which is equivalent to  $\rho/\alpha$ .

From equation 9 and as defined by the International Commission on Radiological Units (ICRU 1994), the specific activity distribution with depth  $A(\xi)$  (Bq kg<sup>-1</sup>) is given by equation 10:

$$A(\xi) = A_o \cdot e^{\left(\frac{-\xi}{\beta}\right)}$$
(10)

Such exponential distributions have mean values of  $\alpha^{-1}$  (equation 7) and ß (equation 10).

#### Depth distributions exhibiting a subsurface maxima

The assumption of the exponential model is a generalisation, which is more often than not inaccurate. This is particularly true for aged deposits of longer-lived radionuclides such as <sup>137</sup>Cs, where post-depositional processes or multiple deposition events can lead to the development of subsurface activity maxima within the soil or sediment depth profile.

For such non-exponential cases, it is suggested that the mean mass per unit area (ß) can also be a useful measure of source self-shielding in a general case. In this case, ß should be obtained using equation 11 by evaluating the mean ratio from equation 10 (Tyler *et al.* 1996b, Tyler 1999):

$$\beta = \frac{\int_{0}^{\infty} \xi \cdot A(\xi) d\xi}{\int_{0}^{\infty} A(\xi) d\xi}$$
(11)

where:

 $\xi$  is the mass depth (g cm<sup>-2</sup>) A( $\xi$ ) is the activity per unit mass (Bq g<sup>-1</sup>).

The  $^{137}$ Cs depth distribution profiles for the UKSHS were estimated by plotting specific activities (Bq kg<sup>-1</sup>) with depth.

ß was estimated by re-integrating the mass depth profile into equal intervals (1 g cm<sup>-2</sup>) and completing the numerical integration from equation 12:

$$\beta = \frac{\sum_{0}^{\xi} A(\xi) \cdot \Delta \xi}{\sum_{0}^{\xi} A(\xi)}$$
(12)

where:

 $\begin{array}{ll} A(\xi) & \text{is the specific activity (Bq kg^{-1})} \\ \Delta \xi & \text{are equal mass depth intervals, of the order of 1 g cm}^{-2}. \end{array}$ 

### In situ detector calibration

#### Working calibration

A working calibration was determined for use as the UKSHS progressed to check the data being obtained. The working calibration was based on empirical measurements made at calibration sites encompassing a range of environmental conditions from the ECCOMAGS project, Resumé 2002 (Sanderson *et al.* 2002). The sites encompass a range of <sup>137</sup>Cs deposition settings, including:

- weapons testing aged <sup>137</sup>Cs;
- Chernobyl fallout;
- <sup>137</sup>Cs derived from marine discharges from BNFL Sellafield on the Cumbrian coast.

The primordial radionuclide concentrations also vary across these sites.

Each ECCOMAGS calibration site encompassed an expanding hexagonal sampling plan. This was designed to provide sufficient data to calibrate both *in situ* and airborne gamma spectrometry systems, which operate on very different spatial scales (Tyler *et al.* 1996a). The specific activity concentrations from each site were derived from 10 different gamma spectrometry laboratories, including the Universities of Liverpool and Stirling. The analysis included quality assurance tests to determine the reliability of the data. The samples within the field of view of the detector were weighted spatially to match the spatial response of the detector.

### In situ efficiency calibration

From a single calibration site (Inch Farm), full energy peaks for the natural radionuclide series were used to calculate an energy-dependent efficiency calibration to derive specific activity concentrations (Bq kg<sup>-1</sup>) for the *in situ* detector. This calibration was then tested against the other two ECCOMAGS sites to provide specific activity concentrations (Table 4.5).

This enabled the GammaVision-32 operating software to determine specific activity concentrations for  ${}^{7}\text{Be}$ ,  ${}^{41}\text{Ar}$ ,  ${}^{40}\text{K}$ ,  ${}^{60}\text{Co}$ ,  ${}^{134}\text{Cs}$ ,  ${}^{208}\text{TI}$ ,  ${}^{208}\text{Pb}$ ,  ${}^{212}\text{Pb}$ ,  ${}^{214}\text{Pb}$ ,  ${}^{212}\text{Bi}$ ,  ${}^{214}\text{Bi}$ ,  ${}^{224}\text{Ra}$ ,  ${}^{226}\text{Ra}$ ,  ${}^{228}\text{Ac}$ ,  ${}^{234}\text{Th}$ ,  ${}^{234}\text{Pa}$ ,  ${}^{235}\text{U}$  and  ${}^{241}\text{Am}$ .

Table 4.5 demonstrates a robust calibration for specific activity concentrations.

# Table 4.5 – ECCOMAGS Resumé 2002 project: calibration site data (Sanderson *et al.* 2002) and *in situ* comparisons\*

Nuclide	Wigtown Merse		Inch Farm† (Site 164)		Castle Kennedy	
	Soil core	In situ	Soil core	In situ	Soil core	In situ
<sup>40</sup> K (Bq kg <sup>-1</sup> )	409 ± 12	416 ± 10	337 ± 18	335 ± 9.7	272 ± 16	262 ±8.6
<sup>214</sup> Bi (Bq kg <sup>-1</sup> )	15.2 ± 0.4	14.5 ± 1.0	19.9 ± 0.7	20.9 ± 0.7	15.4 ± 1.3	18.5 ± 1.1
<sup>208</sup> TI (Bq kg <sup>-1</sup> )	7.8 ± 0.6	7.5 ± 0.7	9.5 ± 1.2	8.4 ± 0.46	$6.0 \pm 0.6$	$6.2 \pm 0.76$
<sup>137</sup> Cs Bq kg <sup>-1</sup> (0–15 cm)	350 ± 50 <sup>#</sup>	338 ± 6.76	$130 \pm 30^{\#}$	150 ± 3	$30 \pm 10^{\#}$	31.4 ± 1.0
<sup>137</sup> Cs (Bq m <sup>-2</sup> )	225 ± 11	47 ± 2.6	20.8 ± 1.0	20.8 ± 1.5	6.3 ± 0.35	$4.3 \pm 0.3$

\* Errors quoted at 1 sigma.

# Estimated from <sup>137</sup>Cs soil profiles.

† Calibration site

To determine <sup>137</sup>Cs inventories, it is normal practice at Stirling to use a peak to forward Compton scattering (Q) approach to compensate for variations in the vertical activity distribution (Tyler *et al.* 1996b, Tyler 1999, Tyler *et al.* 2001). Without the data derived from the 10 calibration sites from the UKSHS, a working <sup>137</sup>Cs-inventory calibration was derived using this information.

However, the inflexibility of this approach can be demonstrated by comparing the <sup>137</sup>Cs inventories across the three sites in Table 4.5. The reason for the observed differences is related to how the <sup>137</sup>Cs deposition occurred. For example, Inch Farm is typical of a mineral-rich pasture site dominated by Chernobyl fallout (April–May 1986) and where  $\beta = 8 \pm 1.0$  g cm<sup>-2</sup>. In contrast Wigtown Merse has a deeply buried <sup>137</sup>Cs depth profile as a result of the past discharges to the marine environment from BNFL Sellafield, which peaked around 1973, and where  $\beta = 20.5 \pm 0.2$  g cm<sup>-2</sup> (Tyler 1999). Castle Kennedy, however, is dominated by nuclear weapons testing fallout, which peaked around 1963, and has a slightly older and deeper <sup>137</sup>Cs depth profile where  $\beta = 11.4 \pm 0.4$  g cm<sup>-2</sup>. The deeper <sup>137</sup>Cs profiles observed at Wigtown and Castle Kennedy resulted in greater attenuation of the primary gamma photon flux, leading to an underestimate of the <sup>137</sup>Cs inventories at these sites. Hence, it was necessary to implement the calibration correction procedure utilising the forward scattering parameter *Q*.

Regression analysis was then used to compare the working calibration with the remaining nine calibration sites from across England, Scotland and Wales. A series of calibrations was produced, modified for wet weight and dry weight specific activity concentrations, for each soil type.

#### **Radionuclide inventories**

At each calibration site, the calibration coefficient  $C_N$  for a given radionuclide N is given by:

$$C_N = \frac{A_N}{I_N} \tag{13}$$

where:

- $A_N$  is the full energy peak area for radionuclide N
- $I_N$  is the spatially weighted mean inventory and specific activity for radionuclide N.

In this way, a preliminary real time working calibration can be derived from a single calibration site.

#### In situ calibration correction and Q

Some radionuclides, principally those derived from atmospheric fallout such as  $^{137}$ Cs, are characterised by significantly different vertical depth distributions.  $C_N$  must then be calculated for the range of possible site characteristics.

*Q* can be calculated from the spectrum collected at each site and regression analysis undertaken to enable the appropriate  $C_N$  to be derived for an appropriate *in situ* calibration. Examples of this procedure can be found in Tyler *et al.* (1996b), Tyler (1999) and Tyler *et al.* (2001).

This same approach can be used to compare Q and the vertical depth distribution ß such that, at each point, ß can be estimated from *in situ* measurements.

Q is given by equation 14:

$$Q = \frac{A}{B_T}$$

(14)

where:

- *A* is the area under the full energy peak
- $B_{T}$  is the height of the step (Figure 4.5).

The error on Q is derived from the counting error on A and the error on  $B_T$ . The error  $B_T$  is derived from the precision with which the height of the continuum either side of the full energy peak can be estimated.

#### Stirling in situ software

A working version of *in situ* software has been developed at the University of Stirling. This enables real-time estimates of *in situ* radionuclide specific activity concentrations and radionuclide inventories through the implementation of the forward scattering methodology.

The software was not used during the survey period, but was used to derive estimates of Q though post-processing. These estimates were checked by manual calculations using Microsoft® Excel.

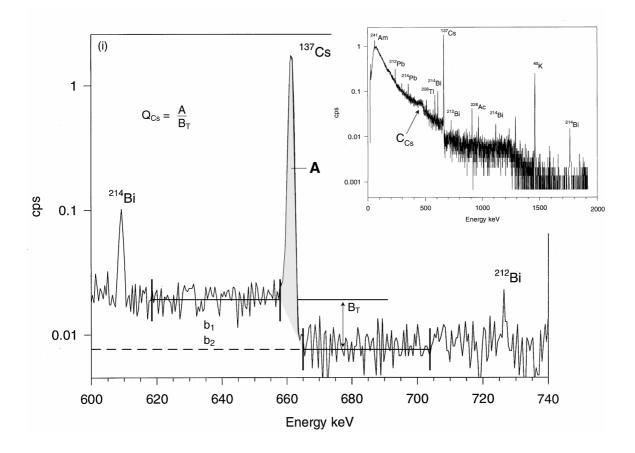


Figure 4.5 – Derivation of *Q* for <sup>137</sup>Cs from Tyler (1999)

### 4.3.4 Air kerma estimation

Having derived the specific activity concentrations and inventories in the soil, air kerma rate conversions from ICRU (1994) were used to derive the terrestrial air kerma dose rate. These calibrations were checked through a EURADOS intercomparison exercise at the Physikalisch-Technische Bundesanstalt carried out immediately following the end of the survey in September 2002. The air kerma contributions from each gamma-emitting source can be estimated from this approach.

# List of abbreviations and acronyms

FWHM	full width half maximum (the full width of the peak in keV at half the maximum height of a peak in a spectrum)			
HPGe	high purity germanium			
HV	high voltage			
LOI	loss-on-ignition			
IAEA	International Atomic Energy Agency			
ICRU	International Commission on Radiological Units and Measurements			
TCS	total coincidence summing			
UKAS	United Kingdom Accreditation Service			
UKSHS	UK Soil and Herbage Pollutant Survey			
UoL	University of Liverpool			

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# Appendix 1: External peer review

The methods for the UK Soil and Herbage Pollutant Survey were developed and modified through a consultative process with further refinement through peer review.

### Internal peer review

The internal peer reviewers were:

- Dr Paul Dale, Scottish Environment Protection Agency
- Dr Rob Allott, Environment Agency
- Dr Jane Rowe, Environment Agency.

#### External peer review

The external peer reviewers were:

- Professor P J Loveland, PhD, Professor of Applied Soil Chemistry, Soil Survey and Land Research Centre, Cranfield University
- •
- Dr Bernie T. Wilkins, Environmental Assessments Department, National Radiological Protection Board.

Copies of the letters from Professor Loveland and Dr Wilkins are available on request.

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