Improving accuracy of *in situ* gamma-ray spectrometry

Jonas Boson

Department of Radiation Sciences,
Radiation Physics
Umeå University, Sweden
2008

Cover: *In situ* gamma-ray spectrometry

Photo by Jonas Boson

ABSTRACT

Gamma-ray spectrometry measurements performed on site, or "*in situ*", is a widely used and powerful method that can be employed both to identify and quantify ground deposited radionuclides. The purpose of this thesis is to improve the calibration of high purity germanium (HPGe) detectors for *in situ* measurements, and calculate the combined uncertainty and potential systematic effects.

An improved semi-empirical calibration method is presented, based on a novel expression for the intrinsic detector efficiency that includes both the energy and angular response of the detector. A three-layer model for the description of the depth distribution of the radionuclide and the soil density is proposed. The combined uncertainty of intrinsic detector efficiency calibrations and *in situ* measurements according to the proposed method was estimated. The uncertainty in the intrinsic detector efficiency was found to be 5.1 and 8.1% (coverage factor k=1, i.e. for a confidence interval of about 68%), for the two detectors calibrated. These numbers were, however, at a later stage reduced to 3.7 and 4.2%, using a revised expression for the intrinsic detector efficiency. For *in situ* measurements, the combined standard uncertainty was found to be 15-20% (k=1), based on the original expression for the intrinsic detector efficiency.

Monte Carlo models of the two detectors were created and Monte Carlo calculated values for intrinsic detector efficiency were compared with experimental data. As a discrepancy was found, a thorough investigation of the detector response was performed. Scanning of the detector surface with a collimated 59.5 keV photon beam revealed the detector response to be highly irregular over the detector surface. It was concluded that the efficiency deficit of the detector could most likely be attributed to an increase in dead layer thickness compared with manufacturer supplied data. The thickness of the dead layer was estimated to be 1.5-1.9 mm, whereas the nominal value was 0.7 mm. Radiographs of the detectors were produced that provided valuable information about the physical dimensions of the germanium crystal, as well as its actual location within the detector housing.

The Monte Carlo models were employed to calculate *in situ* measurement efficiencies for measurements of ^{137}Cs deposition from the Chernobyl fallout. Results from the Monte Carlo simulations were compared both with the semi-empirical method and with soil sample data, and satisfactory agreement was confirmed. It was then proceeded to employ the Monte Carlo model to calculate the effect on *in situ* measurement results by two influencing parameters: ground curvature and activity in trees. Neither of these parameters was found to influence the result by more than about 25%. This deviation is comparable with the measurement uncertainty, and should not deter from measurements in such terrain.

LIST OF PAPERS

This thesis is based on the following five papers, which will be referred to in the text by their Roman numerals:

I.   Jonas Boson, Kenneth Lidström, Torbjörn Nylén, Göran Ågren, Lennart Johansson

   In Situ Gamma-ray Spectrometry for Environmental Monitoring: A Semi Empirical Method. *Radiation Protection Dosimetry* 121, 310-316 (2006)

II.   Jonas Boson, Göran Ågren, Lennart Johansson

   A Detailed Investigation of HPGe Detector Response for Improved Monte Carlo Efficiency Calculations. *Nuclear Instruments and Methods in Physics Research A* 587, 304-314 (2008)

III.   Jonas Boson, Henrik Ramebäck, Göran Ågren, Lennart Johansson

   Uncertainty in HPGe Detector Calibrations for *In Situ* Gamma-ray Spectrometry. *Submitted.*

IV.   Jonas Boson, Agneta Hånell Plamboeck, Henrik Ramebäck, Göran Ågren, Lennart Johansson

   Evaluation of Monte Carlo Based Calibration of HPGe Detectors for *In Situ* gamma-ray Spectrometry. *Submitted.*

V.   Jonas Boson, Torbjörn Nylén, Agneta Hånell Plamboeck, Göran Ågren, Lennart Johansson

   In situ gamma-ray spectrometry: influence of ground curvature and activity in trees. *Manuscript in preparation.*

Papers I and II are reprinted with due permission from Oxford University Press and Elsevier Limited, respectively.

Preliminary reports have been given at:

   -   International Radiation Protection Association, IRPA, 11th International Congress, 23-28 May 2004, Madrid, Spain

   -   The Nordic Society for Radiation Protection, NSFS, 14th Regular Meeting, 27-31 August 2005, Rättvik, Sweden

TABLE OF CONTENTS

INTRODUCTION

Radioactivity is an omnipresent phenomenon in the environment, and naturally occurring radionuclides have always been present in our surroundings. One can distinguish between three different origins of natural radionuclides: primordial, decay products and cosmogenic (Samuelsson, 2001). Primordial radionuclides, like e.g. ^{40}K, have very long half lives (~10^5-10^9 years) and have been present in the crust of the Earth since its creation. Decay products are part of one of the three natural decay-chains: the uranium-, thorium, and actinide-series. Cosmogenic radionuclides are produced by cosmic radiation, through e.g. neutron capture. The most abundant of the cosmogenic radionuclides are ^{14}C and ^{3}H (Samuelsson, 2001).

In the last century, mankind has started to contribute significantly to our radiation environment by the release of anthropogenic radionuclides, primarily from two different sources: atmospheric testing of nuclear explosive devices and releases from nuclear reactors. Radionuclides found in radioactive fallout include for instance ^{131}I, ^{133}I, ^{134}Cs, ^{137}Cs, ^{95}Zr, ^{141}Ce, ^{95}Nb, ^{132}Te (Brune, 2001). In the Chernobyl fallout, the radionuclides contributing most to human radiation exposure are ^{131}I and ^{137}Cs (Brune, 2001). On a worldwide scale the average contribution to human exposure from anthropogenic sources are about one order of magnitude lower than doses from natural radioactivity (Brune, 2001), but on a regional scale doses and dose rates can be very high.

As radioactivity in the environment may have severe implications for human health, accurate methods for assessing activity levels in the environment are essential, both for emergency preparedness purposes and, on a longer timescale, for radioecological surveys. An important technique for determination of ground deposition activity levels is *in situ* gamma-ray spectroscopy. Nowadays, high purity germanium (HPGe) detectors are often the choice for *in situ* measurements, because of their superior energy resolution. This is one of the most effective and accurate methods for measuring the ground deposition of gamma-emitting radionuclides. However, knowledge about the intrinsic detector efficiency as well as of the source depth distribution is necessary in order to obtain an accurate calibration factor for the counting efficiency of the detector (e.g. Beck et al., 1972; Helfer and Miller, 1988; ICRU, 1994). As the intrinsic detector efficiency is empirically determined and the photon fluence at the detector for a particular source distribution is calculated analytically, one often denotes such calibration methods semi-empirical.

Such calibration methods are suitable for reasonably simple geometries and activity depth distributions that can be easily described by mathematical expressions. In order to determine the efficiency for more complex geometries, that are difficult to describe mathematically, one can employ Monte Carlo techniques, which have gained attractiveness with the ongoing increase in computational power. However, in order to validate Monte Carlo models, comparisons with experimental data are essential. Furthermore, in order to facilitate relevant comparisons between different methods, all results must include an estimate of the measurement uncertainty that, at least, include the most significant sources of uncertainty.

Environmental monitoring

Depending on objective and type of deposition to be assessed, a number of different measurement strategies are available, ranging from sampling of food stock, vegetation or soil, through dose-rate measurements, to *in situ* gamma-ray spectrometry.

Sampling of for instance grass or soil are well established methods that are widely used in the field of radioecology. These methods have the advantage of being fairly straightforward, and the sampling scheme can be adapted to a wide variety of objectives and situations. Most sampling, however, is labour intensive, both with regard to the collection of samples and, particularly, concerning laboratory measurements. It is also important to note that sampling will not yield immediate results. All samples will have to be measured in the laboratory before any results are produced. The topic of environmental sampling for radionuclides is dealt with in detail by the International Commission on Radiation Units and Measurements (ICRU) in ICRU report 75 (2006).

Dose rate measurements using hand-held instruments, on the other hand, will give immediate results. Such measurements, however, cannot be used for identification or quantification of individual radionuclides. This means that no prediction of the temporal changes in the dose rate can be made.

In situ gamma-ray spectrometry is a powerful method that can identify and potentially quantify radionuclides directly at the measurement site. It can also be performed as mobile measurements, either on foot or car- or airborne. This allows for the assessment of ground deposition activity levels over large areas. *In situ* gamma-ray spectrometry is however a complex technique that requires both expensive equipment and expert users. The calculation of the necessary calibration factors also introduces a fairly large uncertainty, especially for older depositions where radionuclides have migrated deeper into the soil.

There is of course a significant difference in environmental monitoring for radioecological purposes and for emergency preparedness. The former may require low measurement uncertainties as it might involve detecting small changes over time, but the need for rapid results will possibly be of less importance. For emergency preparedness, speed may be of essence. However, in an initial state, low uncertainties might not be that important. Naturally, there is always a trade-of between speed and simplicity on one hand, and low measurement uncertainty on the other. It is important to acknowledge that each particular scenario will have its own set of parameters and demands, which will govern what compromises will have to be made, and which measurement method(s) is best suited given the actual conditions.

In situ gamma-ray spectrometry

Detectors and equipment

Today, two different types of detectors are predominantly used for *in situ* gamma-ray spectrometry: sodium-iodine, NaI(Tl), and high purity germanium, HPGe.

NaI(Tl) detectors are inorganic scintillators, that work on the principle of incoming radiation causing excitations in the crystal. Subsequent de-excitations produce light photons that are captured on the photo-cathode of a photo-multiplier tube, where they release electrical charge. This electrical charge is then amplified along a dynode chain resulting in an electrical signal for further signal processing. For a more thorough description of scintillation detectors see for instance Knoll (1999). Scintillation detectors are generally impaired by a low energy resolution making nuclide identification difficult, especially if many energy lines are present. The main advantages of NaI(Tl) detectors are high detection efficiency and low cost enabling large detectors. They are therefore suitable for any type of mobile measurements, where the high efficiency allows for shorter measurement times, and thus higher temporal and spatial resolution.

HPGe detectors are, in principle, reverse biased semi-conductor diodes. Incoming radiation produces electron-hole pairs in the semi-conductor crystal, and the released charge is collected at electrodes at the surfaces of the crystal. The number of electron-hole pairs created, and magnitude of the resulting electrical signal, is proportional to the amount of energy deposited in the detector. HPGe detectors have very high energy resolution and are fairly portable, even though they do require cooling, most often provided by liquid nitrogen. The high energy resolution makes HPGe detectors particularly suitable for *in situ* gamma-ray spectrometry as it allows for radionuclide-specific analyses, even in complex radiation fields where many different photon energies are present.

Both types of detectors are coupled to a multichannel analyser (MCA) that processes the signals from the detector. The MCA, in turn, may or may not be connected to a PC. A picture of a typical *in situ* measurement set up, with the detector positioned on a tripod with the centre of the crystal 1 meter above ground, in presented in Figure 1. Included in Figure 1 is also the soil sampling equipment that may be required to asses the activity depth distribution.
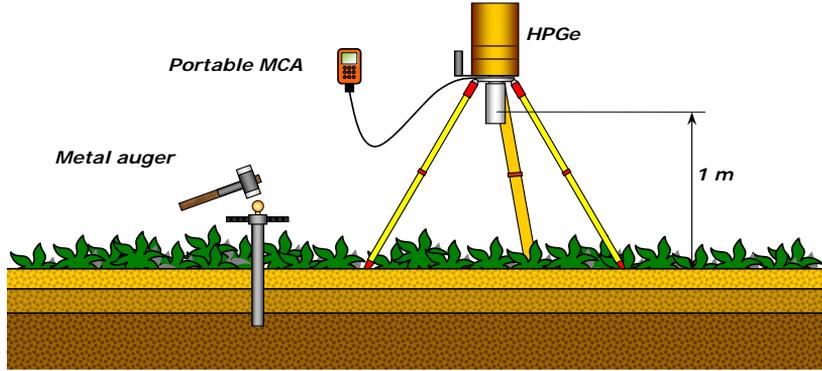
Figure 1. *In situ* gamma-ray spectrometry measurement set-up and accompanying soil sampling equipment.

Efficiency calibrations

The pulse height distribution obtained from the detector and MCA only informs about the distribution of energy depositions in the active volume of the detector. In order to do a radionuclide specific quantification of the activity, an efficiency calibration linking the number of recorded counts in the detector to the ground deposition activity level is required. An expression for the total measurement efficiency was proposed by Beck, DeCampo and Gogolak (1972). This expression has been widely adopted and is recommended by the ICRU (1994).

$$\frac{N}{A_S} = \frac{\dot{N}}{N_0} \times \frac{\dot{N}_0}{\varphi} \times \frac{\varphi}{A_S} \tag{1}$$

In Equation (1) $\dot{N}/A_S$ is the ratio between full-energy peak count rate and ground deposition activity per unit area, i.e. the *in situ* measurement efficiency; N/N_0 is the ratio between the detector response at a specific incident photon field and the response at a parallel photon field incident normal to the detector front face; $\dot{N}_0/\varphi$ is the intrinsic detector efficiency at a parallel photon field incident normal to the detector front face; and φ/A_S is the photon fluence rate at the detector for a specific source distribution.

The intrinsic detector efficiency at different energies is most often experimentally determined using point sources of known activity, and the efficiency function is generally fitted to experimental data on a log-log scale (ICRU, 1994). The angular correction factor can be determined by measuring the relative detector response at different angles of incidence. It should be calculated as the average relative response weighted by the angular distribution of the incident photon field (ICRU, 1994). The final part of the efficiency calibration, the photon fluence rate at the detector for a particular source distribution, is calculated analytically or numerically. A development of the classical calibration method described above is presented in Paper I.

Activity depth distribution

As previously mentioned the photon fluence rate at the detector for a given ground deposition, φ/A_S, is required in order to determine the total *in situ* measurement efficiency. This ratio will of course depend on how deep into the soil the activity has migrated. There are a variety of different models currently used to describe the activity depth distribution, e.g. exponential function (e.g. Beck et al., 1964; Clouvas et al., 2007; Finck, 1992; ICRU, 1994; Oertel et al., 2004), Convection-diffusion equation (e.g. Almgren and Isaksson, 2006; Krstić et al., 2004), Lorentz function (e.g. Hillmann et al., 1996), and uniform slab (e.g. Sowa, 1989). The calibration method proposed in Paper I uses a model for the source matrix described as three separate layers, each with homogenous activity distribution and density. The exponential model that describes the distribution suggested used by Beck, Condon and Lowder (1964) is presented in Equation (2), where $S(z)$ is the activity at depth z, and α^{-1} is the so called relaxation depth.

$$S(z) = S_0 e^{-\alpha z} \qquad (2)$$

The activity (and density) depth distribution is often determined through soil sampling. Efforts, however, have been made to find methods for estimating the depth distribution based on information in the recorded spectrum. The most common approach seems to be the so called peak-to-valley method (e.g. Benke and Kearfott, 2001, 2002; Gering et al., 1998; Gering et al., 2002; Gutierrez-Villanueva et al., 2006; He and Walling, 2000; Hjerpe and Samuelsson, 2002; Kastlander and Bargholtz, 2005; Likar et al., 2004; Thummerer and Jacob, 1998; Tyler, 1999, 2004). The ratio between scattered and unscattered photon flux is assessed by comparing the full-energy peak count rate with the count rate of the region between the Compton continuum and the full energy peak. From this ratio some information about the depth of the source can be inferred. Alternatively, the depth of the source may be estimated by calculating the relative attenuation of separate gamma-lines from the same radionuclide (e.g. Korun et al., 1991). Of course, this requires a radionuclide having at least two different gamma-lines over a broad energy interval.

Monte Carlo simulations of HPGe detectors

Monte Carlo methods is a collective term used for all techniques that make use of stochastic processes to analyse a system. In the case of radiation transport one particle at a time is followed throughout its lifecycle. The outcome at each event, e.g. creation, scatter or absorption, is determined through sampling of a relevant probability distribution. If a large enough number of particles is simulated, a picture of how the system behaves emerges. A multitude of different Monte Carlo codes are available today. An overview of the main features of some Monte Carlo all-particle transport codes (MCNPX, GEANT4, FLUKA, MARS and PHITS) is given by McKinney and co-authors (2006).

Monte Carlo methods are well suited to handle complex geometries, and they can be used for the calculation of *in situ* measurement efficiency calibration factors (e.g.

Allyson and Sanderson, 1998). Some have used Monte Carlo methods for the calculation of peak-to-valley ratios (e.g. Benke and Kearfott, 2002; Gering et al., 1998; Gering et al., 2002; Gutierrez-Villanueva, 2006; Likar et al., 2004; Thummerer and Jacob, 1998). Calibrations based on Monte Carlo simulations of the measurement setup is tested for two different HPGe detectors in Paper IV, and then employed in Paper V to investigate the influence of different parameters such as ground curvature and activity in trees.

It is important to realize that a Monte Carlo simulation of a detector will give a picture of how an ideal detector would behave. Reality is however rarely ideal, and most often one finds that Monte Carlo simulations overestimate the detection efficiency compared to empirical measurements (e.g. Korun and Vidmar, 1997; Lépy et al., 2001). Such discrepancies were also found for the two detectors used in this thesis and the extent and nature of this efficiency deficit is studied in detail in Paper II.

Measurement uncertainty

According to the "Guide to expression of uncertainty in measurement" (GUM) by the International Organization for Standardization (ISO) (1995), a measurement result presented without a corresponding estimate of the uncertainty is, in practice, useless. Only with an estimate of the involved uncertainties is any comparison between different results relevant.

When discussing measurement uncertainty it is worthwhile to take some time to reflect over the terminology. GUM (ISO, 1995) emphasizes the use of the term measurement uncertainty. An uncertainty evaluated by statistical methods, e.g. the standard deviation of the mean from a set of repeated measurements, is referred to as a Type A uncertainty. Other uncertainties, including e.g. certificates or prior knowledge, are termed Type B uncertainties. Any potential bias in measurement results is referred to as a systematic effect. Known, significant systematic effects should always be compensated by a correction factor, which also is associated with an uncertainty.

However, much of the older terminology lingers in the field, and uncertainty is often described in terms of accuracy and precision (e.g. ICRU, 2006). Accuracy refers to the exactness of the measured value, i.e. how close to the true value it is, whereas precision is related to the reproducibility of the results. It is quite possible to have good precision but poor accuracy in measurements, and vice versa. The concepts are illustrated in Figure 2. In GUM terminology, accuracy is related to (the lack of significant) systematic effects and precision is roughly inversely proportional to Type A uncertainties.
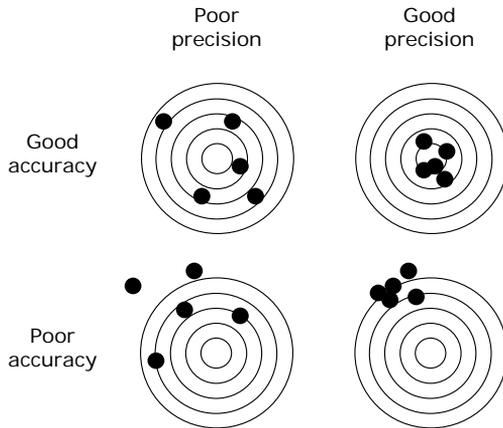
Figure 2. The concepts of accuracy and precision.

In the event of a release of radionuclides into the environment, rapid measurements to quantify the activity may be of essence for radiation protection reasons. Clearly, the measurements need also to be both accurate and with an adequate precision, i.e. a low enough uncertainty. The uncertainty in measurements is sometimes neglected, but it may be of critical importance if measurement results close to a decision limit are reached. Poor precision, or no uncertainty estimation at all, may force an action even though the value is below the action limit, which could potentially result in very high costs for the society. Vice versa, striving to achieve lower uncertainties than what is necessary might be equally costly.

On a longer time scale, uncertainty may be even more important. For any research, e.g. radioecological surveys, detecting changes and variations in ground deposition levels may be hampered by high measurement uncertainties. Especially when investigating natural variability, it is important that the measurement uncertainty is known (and not too large). It is often the case that the natural variability is the dominating factor in the variation between repeated environmental sampling, often accounting for about 80% of the total variability (ICRU, 2006). This fact should however not be used as an excuse not to quantify the measurement uncertainty.

As for the uncertainty of *in situ* measurements, the subject has been studied by Sowa and co-authors (1989). Finck (1992) also presents extensive sensitivity analyses with respect to a large number of parameters. A GUM approach for estimating the uncertainty of *in situ* measurements is presented in Papers III and IV, where the former deals with the uncertainty in the intrinsic detector efficiency calibration, and the latter with uncertainties associated with the source matrix.

Most of the calibration methods used to this date are based on some sort of mathematical model of the source matrix, where the ground surface is described by a perfect plane. In reality, nature is not this simple and more often than not, it is impossible to find a perfectly flat field in which to position the detector, with no trees or

any other potentially interfering structure within, say, a 20 meter distance from the detector. Thus, in order to fully determine the measurement uncertainty one has to investigate what influence such factors will have on the final results. This is dealt with in Paper V.

Fallout of anthropogenic radionuclides in Sweden

About 2400 nuclear weapons test have been performed (Brune, 2001), and the dispersion of radionuclides from such nuclear devices in the atmosphere has given rise to global fallout. In Sweden, the average accumulated ground deposition level of ^{137}Cs from nuclear weapons tests is about 3 kBq m^{-2} (Edvarson, 1991)

On 26 April 1986, reactor 4 at the Chernobyl nuclear power plant suffered a meltdown which resulted in a subsequent large release of radioactivity into the environment. Because of the fire that followed, a significant amount of activity spread over large areas covering large portions of Belarus, Ukraine, Russia, and northern Europe. The ground deposition of ^{137}Cs in Sweden reached levels of up to about 200 kBq m^{-2} (Edvarson, 1991). The deposition levels were strongly correlated to the precipitation at the time of the fallout. In western Sweden, for instance 99 % of the deposition of ^{134,137}Cs, ^{131}I and ^{103,106}Ru was due to wet deposition (Mattsson and Vesanen, 1988).

AIM

The main purpose of this work was to improve the calibrations of *in situ* measurements, and to determine the measurement uncertainty according to GUM. One step towards increased accuracy in measurements is the investigation of various parameters potentially influencing measurement results and, where applicable, the calculation of suitable correction factors.

The strategy to accomplish this followed these steps:

- Commission a semi-empirical model suitable as a benchmark for future Monte Carlo simulations.

- Construct a model of the detector using the MCNP Monte Carlo code that accurately reproduces experimentally determined efficiencies.

- Validate the use of Monte Carlo simulations for calibrations of *in situ* measurements.

- Determine the combined uncertainty of *in situ* measurements

- Investigate influence of ground curvature and activity in trees, and, if necessary, calculate suitable correction factors that can be used to compensate for these factors.

METHODS

Site descriptions

In situ measurements were performed on five different sites in Papers I and IV. Sites 2 and 3 were also chosen as basis for further investigations using Monte Carlo simulations in Paper V. A summary of locations and measurement dates is given in Table 1. The site locations are also indicated on the map in Figure 3 showing the ^{137}Cs surface equivalent deposition density after the Chernobyl accident based on aerial measurements of the fallout performed by the Swedish Geological Company, SGAB (SGAB, 1986). The surface equivalent deposition density is obtained when calibration factors derived for a perfectly flat surface source are used. The actual ground deposition levels can be calculated by multiplying with a factor 1.6 (Edvarson, K., 1991).
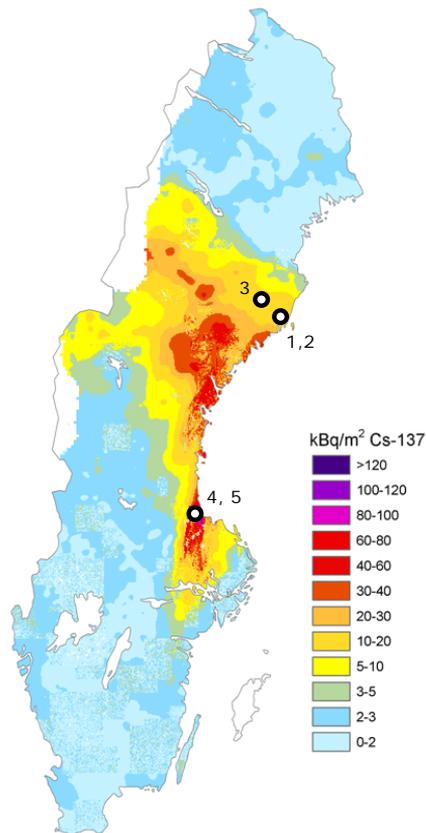
Figure 3. Measurement sites on a map showing ^{137}Cs ground deposition levels in Sweden (reproduced with permission from the Swedish Radiation Protection Authority).

Sites 1 and 2 are both open, relatively flat lawns located within the city of Umeå. Site 3 is located on a pine heath, populated by Scots pine (*Pinus Sylvestris L.*), near Vindeln, about 60 km northwest of Umeå. These sites all received about 10-20 kBq m^{-2} surface equivalent deposition density of ^{137}Cs after the Chernobyl accident (Figure 3). It is worth noting that there was still some snow remaining on Sites 1-3 at the time of the fallout. Sites 4 and 5 are located near Hille, about 20 km north of Gävle, an area that received the highest levels of fallout in Sweden after the Chernobyl accident.

Table 1. Site descriptions.

		Site 1	Site 2	Site 3	Site 4	Site 5
Location		Ersboda, Umeå	FOI area, Umeå	Åheden, Vindeln	Hille, Gävle	Hille, Gävle
Type		Lawn	Lawn	Pine heath	Spruce forest	Alder forest, swamp
GPS coord.	East	20° 19' 11"	20° 19' 56"	19° 46' 57"	17° 16' 14"	17° 16' 28"
	North	63° 51' 23"	63° 51' 1"	64° 13' 54"	60° 46' 2"	60° 46' 19"
Measurement date		25/10/2004	19/9/2006	4/8/2006	16/6/2004	16/6/2004

Soil sampling

On all sites where *in situ* gamma-ray spectrometry was performed a set of soil samples was also collected. The soil samples collected in this work serve three different purposes:

1. Soil samples will provide information about soil density and activity depth distribution necessary for an accurate calibration of *in situ* measurements.
2. The soil samples will also provide estimates of the activity ground deposition levels that can serve as reference values for the validation of *in situ* measurement efficiency calibrations.
3. The collected soil samples will provide a measure of the inter-sample variability for the different source matrix parameters. Knowledge about this variability is essential in order to estimate the *in situ* measurement uncertainty, and will also enable a calculation of a suitable number of samples to be collected for future calibrations.

With all sampling, a sampling scheme should be worked out based on the current objectives (ICRU, 2006). Often the number of samples to be collected is a trade-off between uncertainty in the results and the corresponding workload. The sampling scheme used in papers I and IV is based on a cross pattern suggested by the NATO "Handbook for Sampling and Identification of Radiological Agents" (AEP-49) (2005). However, as the *in situ* gamma-ray spectrometry measurements "sees" a fairly large area, the inter-sample distance was increased from one to two meters, and the number of samples on each arm of the cross was increased to four. Tyler and co-authors (1996) pointed out the importance of matching the area on which soil samples are collected with the field of view of the detector, if relevant comparisons are to be made between the two methods. A central sample was also added, giving a total of 17 soil samples from

each site (Figure 4). The 8 m radius corresponds to an area that contributes to 72-82% of the detector response at all measurement sites in this work.
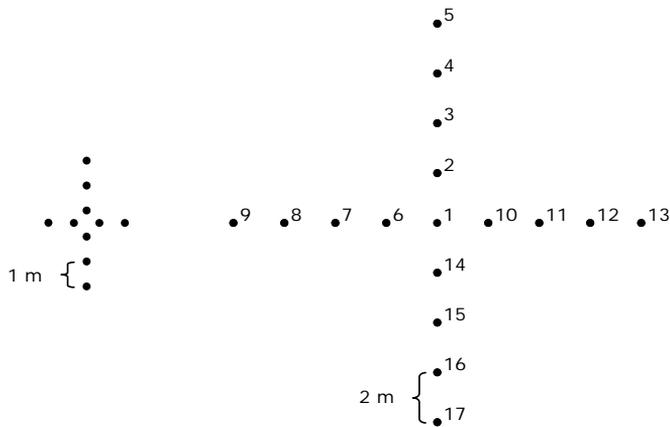

Figure 4. Soil sampling scheme suggested by the NATO Handbook for Sampling and Identification of Radiological Agents (2005) (to the left), and a modified version used in this thesis and included papers (to the right).

Soil cores were collected using metal augers, with radii 2.15 or 2.4 cm. All samples were sectioned on site into three layers in order to provide some information about the activity and density depth distribution. At the laboratory, all samples were weighed, dried at 70ºC for a few days (until constant weight), and then weighed again to determine dry weight and water content. The ^{137}Cs content in samples were then measured using HPGe detectors (Papers I and IV).

An important aspect of the sampling procedure is how a soil core is divided into sections. Whenever distinctive horizons were present, the intersections were preferably made at these. This was done to produce as homogeneous aliquots as possible. The humus layer, in particular, often has noticeably lower density than mineral soil.

Activity and density depth distributions with higher resolution

In order to obtain an example of activity and depth distributions with higher resolution, four soil cores were collected at Site 3 that were not sectioned in accordance with the aforementioned method. Two of the samples were sliced in 2 centimetre sections in order to assess the density depth function. An exception was made for the top layer, as it was obvious that the top 3 cm represented a layer of soil with high content of organic matter and distinctly lower density. Two samples were kept in plastic tubes with diameters matching the ones of the metal augers. The tubes were subsequently moved in front of a detector shielded by a 50 mm thick lead collimator with a 5 mm slit, in accordance with a method described by Finck (1992). Measurements of the relative ^{137}Cs activity content were performed for each 5 mm interval. Live times of 27000

seconds were used for measurements of each 5 mm interval in the top 12 and 14 cm of sample 1 and 2, respectively. This resulted in uncertainties due to counting statistics of 10 – 20%. The lower sections, containing no significant activity, were measured using 5400 seconds live time. Spectra were evaluated using GammaVision 6.01 (Ortec Inc., USA).

Due to a short source-to-detector distance (chosen to provide reasonable measurement times) and the width of the collimator slit, there was significant contribution to each measurement result from adjacent sections of the sample. As suggested by Finck (1992) a de-convolution was therefore performed based on the relative response function found when moving a point source in front of the collimator. A matrix A_{ij} consisting of coefficients representing the relative contribution to measurement result i from position j, can be used to define the equation system

$$Y_i = \sum_j A_{ij} X_j \qquad (3)$$

where Y_i and X_i are the measured value and true relative activity content for position i, respectively. The true de-convoluted activity distribution X can be found by solving the equation

$$A^T Y = A^T A X \qquad (4)$$

However, as the resulting equation system for the de-convolution turned out to be ill-posed, a Tikhonov-regularisation of the system was performed. A regularisation term, $\alpha = aI$, was added to $A^T A$, where I is the identity matrix and a is a real number. The constant a is to be chosen as small as possible without causing "noise" in the solution. The final solution to the system is thus

$$X = (aI + A^T A)^{-1} A^T Y \qquad (5)$$

In situ measurements

Two detectors were used for the *in situ* measurements in Papers I and III, referred to as Detector 1 and Detector 2. Both detectors are of p-type and were manufactured by Ortec Inc. (USA). Their efficiencies, measured relative a 3"x3" NaI detector at 1.33 MeV, were 36% for Detector 1 and 57% for Detector 2. For the *in situ* measurements the detectors were mounted on tripods so that the centre of the germanium crystal was positioned about 1.0 m above ground. Spectra were recorded on a DigiDART (Ortec Inc, USA) portable MCA and measurement times (live time) between 1000 and 1800 seconds were used at all sites. Spectra were evaluated using GammaVision 6.01 (Ortec Inc., USA).

Figure 5. One of the HPGe detectors used (Detector 2, 57% efficiency)
and the DigiDART portable MCA.

Semi-empirical calibration method

The semi-empirical method for the efficiency calibration of HPGe detectors is described in detail in Paper I. It differs from the method described by ICRU (1994) in the fact that the two parameters that describe intrinsic detector efficiency, i.e. the efficiency for a photon beam incident parallel to the detector symmetry axis, N_0/φ, and the angular correction factor, $\dot{N}/\dot{N}_0$, are integrated into a single detector efficiency parameter, $\varepsilon(E,\theta)$. Furthermore, the activity depth distribution is described as three separate layers with homogenous activity content and density.

Intrinsic detector efficiency

The intrinsic detector efficiency was determined for the two detectors in Paper I. In Paper III an additional detector, Detector 3, was calibrated and used in the development of a new expression for intrinsic detector efficiency. This detector was of p-type, manufactured by Ortec Inc. (USA), and had an efficiency of 120%. This detector was, however, not used for *in situ* measurements. The detector was placed so that the centre of the germanium crystal coincided with the centre of a rotating bow on which the calibration sources could be placed (Figure 6). The purpose of the rotation of the bow is to cancel out any variations in detector efficiency in the azimuth plane. Two different calibration sources were used: [60]Co and [152]Eu. The live times of the acquisitions were 1000 s. The detector efficiency was determined for a set of 11 energies (ranging from 244.7 to 1408 keV) and 19 angles (between 0 and 90 degrees, with 5 degree increments). In Paper III, six of the energies were utilized for the calculation of the uncertainty in the calibration.
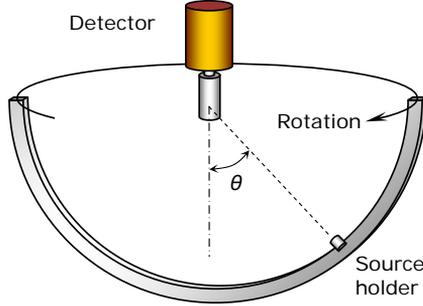
Figure 6. Schematic picture of the detector and rotating bow, used for the efficiency calibration.

An empirical expression for the intrinsic detector efficiency was proposed in Paper I. The parameters in the expression were determined by regression analysis of empirical data.

$$\varepsilon(E,\theta) = \left(\frac{k_1\,\theta + m_1}{E} \right)^{(k_2\,\theta + m_2)} \qquad (6)$$

In Paper III, a new empirical expression for the detector efficiency function was proposed in order to reduce the uncertainty of the fit to empirical data.

$$\ln\varepsilon(E,\theta) = C_0 + C_1\theta^4 E + C_2\theta^3 + C_3\theta^2 + C_4\theta + C_5 E + C_6 E^{-1} + \\ + C_7\cos(x\theta / E^z - y) + C_8\ln E + C_9(\ln E)^2 \qquad (7)$$

The combined uncertainty in calibration measurements were estimated in Paper III in accordance with GUM (ISO, 1995). The sources of uncertainty contributing to the combined uncertainty were: standard uncertainty of calibration source activities, 1.5%; counting statistics, <2.5% for all full-energy peaks used; uncertainty in the attenuation coefficient of air, 2%; standard uncertainty in gamma-ray intensities, ≤0.25%; and the uncertainty in the source-to-detector distance, 0.8%.

Total measurement efficiency

Once the intrinsic detector efficiency is determined, the total *in situ* measurements efficiency can be calculated. An expression for the efficiency from a volume element d*V* was postulated, and the total efficiency is found through integration over the total volume of the source. A schematic picture of the measurement setup indicating the integration parameters is shown in Figure 7. The source matrix is as previously mentioned described as three (or fewer) separate homogeneous layers.
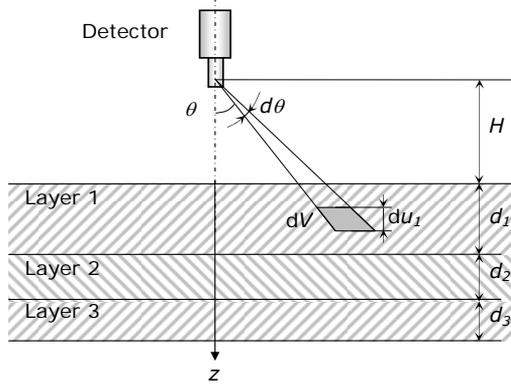
Figure 7. Geometry for the calculation of measurement efficiency from a volume element dV. H is the height of the detector above ground; d_i and u_i are the thickness of, and depth in, layer i, respectively; θ is the angle of incidence relative the symmetry axis of the detector; and z is depth in soil.

The total expression for the measurement efficiency, i.e. full-energy peak count rate $\dot{N}$ per activity per unit area A_s, is

$$\frac{\dot{N}}{A_S} = \sum_i \frac{I_\gamma}{2d_i} a_i \iint \varepsilon(E,\theta)\tan(\theta) C_{tot}^i \, du_i d\theta \qquad (8)$$

where

- I_γ is the gamma-ray intensity for a specific energy;
- a_i is the relative activity content in layer i;
- d_i is the thickness of layer i;
- C_{tot}^i is the total attenuation, in air and soil; and
- u_i is the depth in layer i.

The total attenuation in air and all soil layers is given by:

$$C_{tot}^i = \exp\left(-\mu_{air}\left(\frac{H}{\cos(\theta)}\right)\right)\exp\left(-\mu_{soil}\frac{u_i}{\cos(\theta)}\right)\prod_{n=1}^{i-1}\exp\left(-\mu_{soil}\frac{d_i}{\cos(\theta)}\right) \ (9)$$

where H is the height of the detector above ground; and μ_{air} and μ_{soil} are linear attenuation coefficients of air and soil respectively. A complete derivation of the equations is presented in Paper I. The integral in Equation (8) is calculated numerically. Values for the source parameters, i.e. layer thicknesses, densities and relative activity contents, are based on information from soil samples.

The combined uncertainty of *in situ* measurements was addressed in Paper IV. Uncertainty in the intrinsic detector efficiency was established in Paper III, and uncertainties in all soil sample parameters were calculated the standard deviation of the mean. Unfortunately, the rather complex expression for the *in situ* measurement

16

efficiency makes propagation of the uncertainty contributions difficult. Instead, an alternative approach was adopted, were the combined uncertainty for a set of discrete angles were calculated. The combined standard uncertainty of the *in situ* measurement was then calculated as the average of these results. For the assessments of combined uncertainties and relative contributions from the various parameters GUM Workbench (Metrodata GmbH, Weil-am-Rhein, Germany) was used.

A potential systematic effect is introduced by the model used to describe the activity depth distribution. If the true distribution is decreasing with depth, the assumption of a homogeneous activity distribution within layers will result in an underestimation of the measurement efficiency, and correspondingly, to an overestimation of the ground deposition activity level. To account for this systematic effect, a correction factor was introduced. The correction factor was assumed to have the value 1, but was associated with an uncertainty. The magnitude of the uncertainty is determined by the size of the systematic effect. This was investigated in Paper IV, by the use of Monte Carlo simulations. Two alternative distributions, expected to yield as different results as realistically possible in comparison with the three-layer model, were used to determine the possible range of the systematic effect.

Monte Carlo modelling of detectors

Monte Carlo models of the two detectors used for *in situ* measurements in this thesis were constructed using the MCNP5 1.40 code (Los Alamos National Laboratory, USA) (Figure 8).
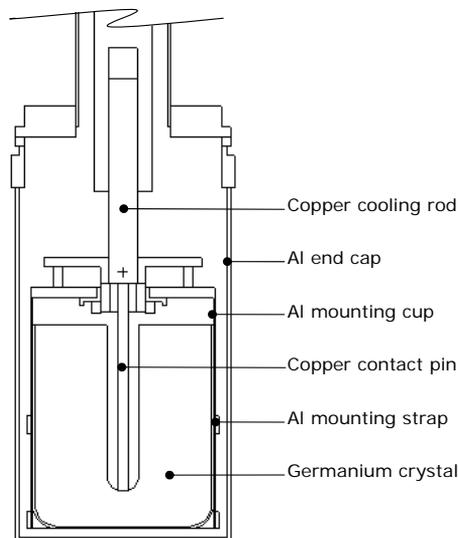

Figure 8. Monte Carlo model of the detector (cryostat not included).

The models were initially based entirely on detector dimensions supplied by the manufacturer. However, several studies (e.g. Korun and Vidmar, 1997; Lépy et al., 2001) have reported a discrepancy between Monte Carlo-calculated and empirically determined detector efficiencies. Such discrepancies were also found in some initial detector simulations. A thorough investigation of the detector response was therefore performed in Paper II, with the objective to determine the size of this discrepancy and, if possible, its cause.

In short, the following investigations were performed on Detector 1:

1. The detector was X-rayed in order to confirm the physical dimensions of the germanium crystal.

2. The homogeneity of the detector response over the crystal surface was investigated by scanning the detector with collimated photon beams of low energies.

3. The thickness of the dead layer at the n$^+$-contact of the detector was assessed by measurement of the relative efficiency at different incidence angles for a collimated low energy photon beam.

4. Properties of the electrical field have been suggested as a cause of the decrease in detector efficiency. This was investigated by examining the detector efficiency as a function of bias voltage.

For the scanning of the detector an 11.5 mm thick collimator, with a 2 mm radius hole was used. The collimator and source was moved along four parallel lines along the lateral surface and two orthogonal lines across the front face of the detector. The relative detector response was recorded at 2 mm intervals. Two different photon energies were utilized: 59.5 keV from ^{241}Am and 244.7 keV from ^{152}Eu. The scanning process was also modelled in Monte Carlo simulations.

In order to assess the thickness of the dead layer, a collimator was constructed that allowed for two different angles of photon incidence on the detector surface, as suggested by Dryak and Kovar (2006) and Van Riper, Metzger and Kearfott (2002). From the relative detector response at the different angles, the thickness of attenuating layers can be calculated. 59.5 keV photons from ^{241}Am were used for these measurements.

Monte Carlo based calibrations for *in situ* gamma-ray spectrometry

Monte Carlo simulations were used to obtain *in situ* measurement efficiency calibration factors in Paper IV. As mentioned in the previous section, the Monte Carlo model of the detector overestimates the intrinsic detector efficiency. It was opted to adjust calculated detector efficiencies by a correction factor, rather than by tweaking detector parameters such as the thickness of the dead layer. The same source model was used as for semi-empirical calibrations, i.e. three layers, each with homogenous density and activity content. Layer thicknesses, densities and relative activity contents were based on

results from soil sampling at each site. The chemical composition in mineral soil was taken from Finck (1992), whereas the composition of humus was based on data from Site 3 (Plamboeck et al., 2006).

As mentioned in the section about the semi-empirical calibration method, the influence on *in situ* measurement efficiency by assuming radically different activity depth distributions were investigated in order to obtain a measure of the potential systematic effect of the three-layer model. Two different distributions were tested: one exponential, and one with a linearly decreasing activity.

Simulations were also made using the activity and density depth distributions with higher resolution determined at Site 3. However, as the data for these distributions were based on only two samples each, they can not be assumed to be representative of the conditions at the site. Thus, no quantitative conclusions can be inferred by comparing these results with ones obtained using data from the 17 samples sectioned in accordance with the three-layer model.

Simulations of ground curvature and activity in trees

In Paper V the influences on *in situ* measurement results by ground curvature and activity in trees were investigated by Monte Carlo simulations. In order to estimate the effect of ground curvature on *in situ* measurement results, the Monte Carlo model was modified so that the ground surface was described as a section of a sphere. The radius of the sphere was chosen to create the desired depth or height at the centre, where the detector was positioned. In addition to activity and density depth distribution based on soil samples, i.e. for approximately 20-year old fallout, simulations were also performed for the case of fresh fallout. The source distribution for fresh fallout was modelled as a 2 cm thick slab of 500 kg m^{-3} density having homogeneous activity content (Lidström and Nylén, 1998).

The modelling of the trees was based on biomass data (Plamboeck et al., 2000) and tree dimensions (Plamboeck and Nylén, 2008) from Site 3. Figure 9 shows a section of a row of trees. By performing separate simulations with the source located in tree crowns, stems, or in soil, it was possible to calculate total measurement efficiencies for different distributions of the activity within the system. As with the case of ground curvature, simulations were performed for both fresh and 20-year old fallout.
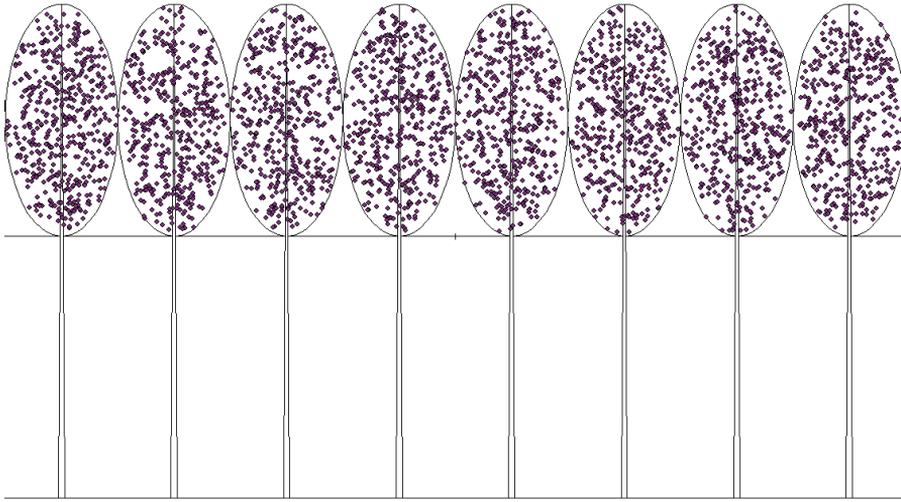
Figure 9. Monte Carlo model used for simulations of activity in trees. The points indicate starting location for photons in Monte Carlo simulation (the plane below tree crowns is included in order to simplify the description of the source geometry in Monte Carlo simulations).

RESULTS AND DISCUSSION

Soil sampling

Results from soil sampling performed at the different measurement sites in Papers I and IV illustrate the high inter-sample variability often encountered in any form of environmental samples. Sources of this variability include measurement uncertainties and variations in the sampling procedure, as well as natural variability (ICRU, 2006). The relative standard deviation of ^{137}Cs activity content in soil samples ranges from 31% at Site 4 to 60% at Site 5 (Figure 10). As mentioned in Paper IV, the particularly high inter-sample variation found at Site 5 is probably an effect of activity redistribution due to regular flooding of the area and deposition of sediments.

Uncertainties in collection, handling and measurements of soil samples were not considered, and the standard deviation of the mean was used as an estimate of the combined uncertainty in all soil sample parameters. This measure does not include Type B uncertainties (ISO, 1995), such as the uncertainty in the efficiency calibration. However, as long as Type B uncertainties are small compared to Type A uncertainties and natural variability, this will have negligible implications for the combined uncertainty. The main Type B uncertainty is the one in the detector efficiency calibration, which is about 2.5% (coverage factor k=1, i.e. for a confidence interval of about 68%). Combined Type A uncertainties in soil sample handling and measurements are expected to be in the order of 10% (Lettner et al., 2000). Adding the natural variability to this, it is easy to see that Type B uncertainties will not contribute significantly to the combined uncertainty in soil sample parameters.

A systematic effect that has been neglected in this work is the density and self-absorption correction in sample measurements. A lower density in a sample compared to the density of the calibration standard, will result in an increase in measurement efficiency and, consequently, in an overestimation of the activity content. The uncertainty introduced by this systematic effect was not expected to be more than a few percent.

The number of soil samples collected at each site was considered to be a good compromise between achieved uncertainty and total workload. The resulting standard deviation for the various soil parameters, i.e. densities and activity contents, ranges from 6.4 to 150%. The largest and smallest variations were found in the activity content and in the density of the third layer, respectively. As each sample is sectioned into three layers, 17 ground cores collected become 51 samples to analyse in the laboratory. A larger number of cores would result in an overwhelming laboratory workload. Unfortunately, the 17 samples collected are too few to determine the true distributions of the various soil sample parameters with reasonable significance. ICRU concludes that activity content in soil samples is often found to be log-normally distributed, if properly investigated. However, the ICRU recommends the use of normal distributions when presenting results, as this is the most common practice. The ICRU recommendation was followed in this thesis.
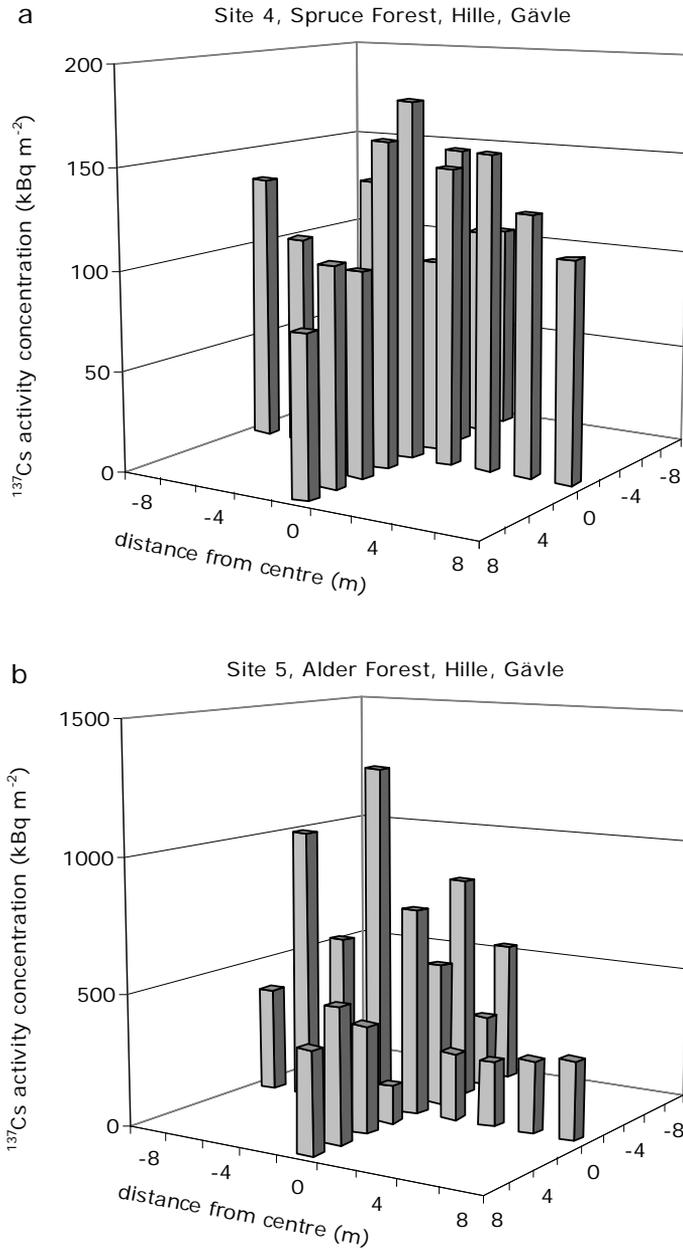
a

Site 4, Spruce Forest, Hille, Gävle

b

Site 5, Alder Forest, Hille, Gävle

Figure 10. [137]Cs content in soil samples collected at (a) Site 4 and (b) Site 5, recalculated to the corresponding ground deposition activity level in kBq m[-2].

Looking at the distribution of activity content in individual soil samples collected at Site 5 (Figure 10b), one might suspect that the *in situ* measurement result could be correlated to the positioning of the detector. To determine that this was not the case, *in situ* measurements were performed directly above all 17 individual soil sampling points. These measurements were performed on 4 October 2005, i.e. one year after the initial measurements on the site. Uncertainties due to counting statistics were at sub-percent level for all measurements. The resulting variation in these 17 *in situ* measurements was found to be 7.7% (one standard deviation). This is certainly much less than the variability in soil samples, and is a testament to the averaging power of the *in situ* measurement technique.

Activity and density depth distributions

The activity depth distributions found at the five sites is presented in Figure 11. The densities were found to be somewhat lower in the first layer, particularly at Sites 3 and 4, but fairly constant in the under-laying layers (Paper IV).

The three-layer model has three distinct merits. First, it is easily compatible with the sampling procedure since it does not require any calculations to fit an expression for depth distribution to sampling results. Secondly, it is immediately clear from examination of soil cores that the density of the soil may change with discrete intervals. Finally, the three-layer model makes no assumption about the general shape of the distribution, and can easily be fitted to a wide variety of depth distributions.

That the density of soil is varying with depth has also been reported by e.g. Isaksson and Erlandsson (1995), that also points out that homogeneous soil density is often assumed in the exponential activity depth distribution model.
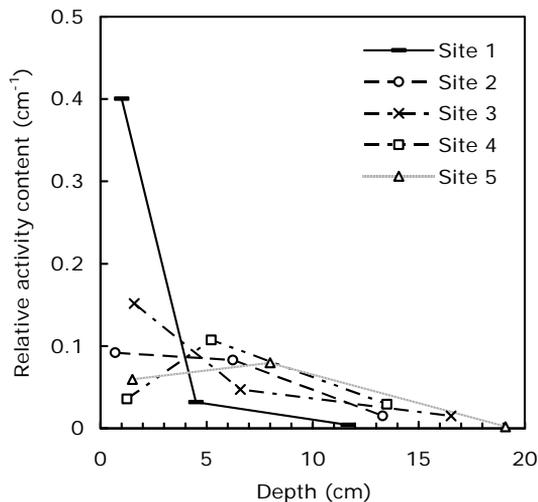

Figure 11. Relative activity depth distributions, as determined at the five sampling sites.

The shapes of the activity distributions presented in Figure 11 are markedly different at different sites. That the shape of the activity depth distribution may vary considerably with location is also evident upon comparisons of previously reported results. For instance, Hillmann and co-authors (1996) conclude that an exponential function is well suited to describe fresh fallout, but less so for older depositions. Clouvas and colleagues (2007), on the other hand, finds that a double exponential gave good results for the period 1987-94, but an exponential function was better for measurements on older fallout (about 20 years old). Several authors (e.g. Almgren and Isaksson, 2006; Hillman et al., 1996; Krstić et al., 2004; MacDonald et al., 1996; Owens and Walling, 1996; Rosén et al., 1999) report depth distributions with a distinct peak in the activity distribution at a certain depth, in which case an exponential model is clearly unrealistic.

Depth distributions with higher resolution

As previously mentioned, the response function of the scanning equipment must be determined in order to enable a de-convolution of measurement results to find the "true" activity depth distribution. The response function of the detector system was determined by moving a point source in front of the collimated detector. The recorded detector response function is shown in Figure 12. This response function was then used to construct the response matrix A in Equation 3, which in turn was used to de-convolute the results from the scanning of two of the un-sectioned soil samples from Site 3. The resulting relative activity depth distributions for the two samples and their average are presented in Figure 13a. Activity data was pooled into 1 cm-layers. The corresponding density data is presented in Figure 13b.
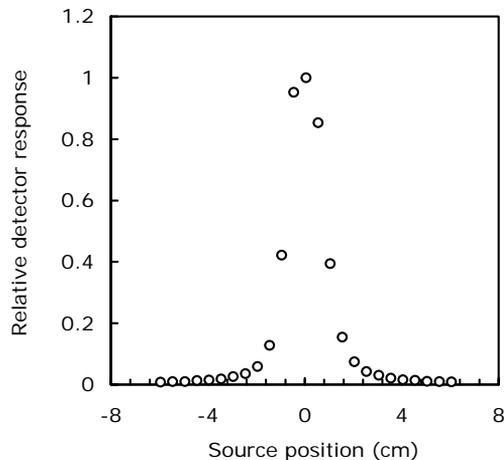

Figure 12. Normalized detector response plotted against position of source relative centre of collimator
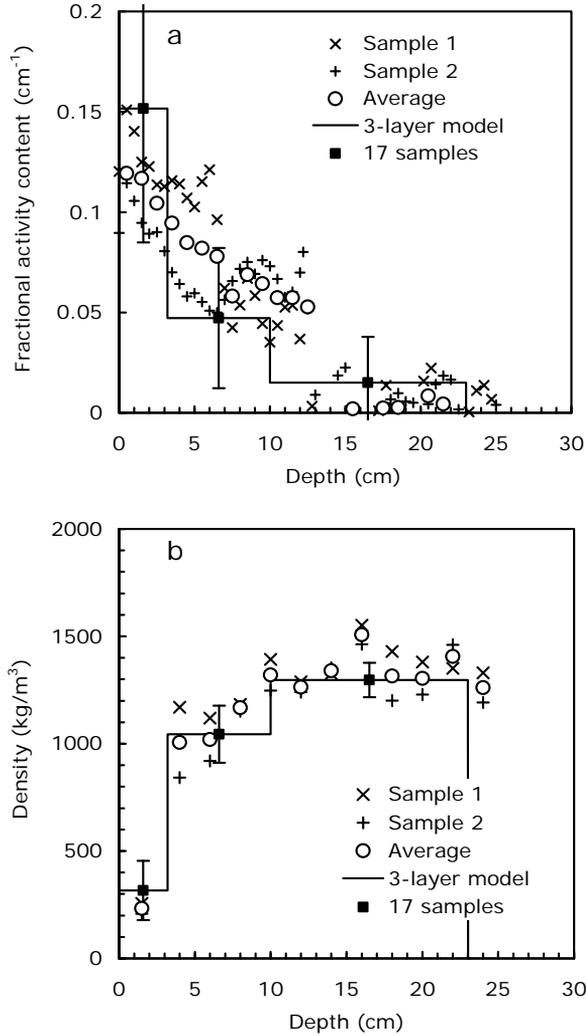
Figure 13. [137]Cs activity (a) and density (b) depth distributions from soil samples collected at Site 3. Individual data points represent values from samples analysed with higher linear resolution and the solid squares and curves represent average values from the 17 samples, and the corresponding three-layer model. Uncertainties represent one standard deviation.

No uncertainty measure for the detailed distributions have been included in Figures 3a and b, as uncertainty in measurements will be trivial in comparison with the large uncertainty resulting from natural variability and the low number of samples (*n*=2). Therefore no quantitative comparisons between the three-layer distributions and the distributions with higher resolution can be made. However, this was not the intent with the acquisition of the more detailed distributions. The purpose was only to acquire an example of a realistic "true" distribution, and compare the calibration factors calculated for this distribution with the ones based on the 3-layer model.

Semi-empirical calibrations

Intrinsic detector calibrations

Three detectors were calibrated in accordance with the method described in Paper I. The recorded detector efficiency at different photon energies and angles of incidence for Detector 1 is shown in Figure 14. Included in the graph are also fitted detector efficiency functions, both according to the original expression in Paper I (Equation 6) and the new expression proposed in Paper III (Equation 7). It is apparent that Equation 7 is better suited to describe the intrinsic detector efficiency. Particularly Detector 3 displayed a pronounced periodicity in the detector response as a function of photon angle of incidence (Paper III). Equation 7 proved to be more apt at reproducing this periodicity.

Uncertainties of individual data points in Figure 14, i.e. for a specific photon energy and angle of incidence, are about 3% (k=1). The major contributions to the combined uncertainty come from full-energy peak count rate, source activity, and source-to-detector distance. As shown in Paper III, reducing the uncertainty in only one of these sources will not reduce the combined standard uncertainty to any large extent. Uncertainty in the fitted efficiency function (Equation 6) was found to be significantly higher than the uncertainty in individual data points: 8.2 and 5.1% (k=1) for Detectors 1 and 2, respectively. This indicates room for improvement in the proposed expression. Using Equation 7 these uncertainties are reduced to 4.2 and 3.7%, which approaches the uncertainty in individual data points.

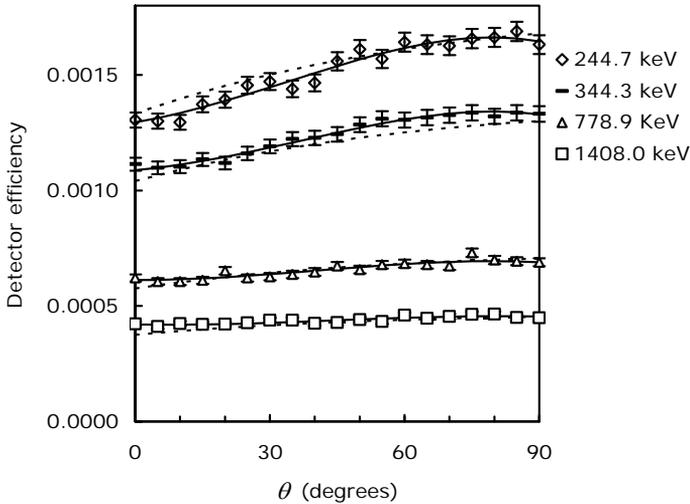**Figure 14.** Detector efficiency for Detector 1 as a function of angle of incidence. Data points represent empirical data, dashed lines the fitted efficiency function using the expression suggested in Paper I (Equation 6), and solid lines the fitted efficiency function using the new expression proposed in Paper III (Equation 7). Uncertainties represent one standard deviation due to counting statistics.

26

One of the main advantages with the calibration method presented in Paper I is the inclusion of the angular response of the detector into the detector efficiency function. This eliminates the potential systematic effect of using a fixed angular correction factor (in Equation 1) for different depth distributions. As previously mentioned, the average angular correction factor should be calculated by weighting with the angular distribution of the photon field from the particular depth distribution. However, in practice this is not always done. Helfer and Miller (1988) suggest the use of a surface source for shallow distributions, and a source distributed uniformly with the depth in the cases where radionuclides have migrated to a greater depth, while for medium depth distributions the median value may be used. MacDonald and co-authors (1998) estimate that the maximum systematic effect caused by such simplifications amounts to about 5%. This could be considered an insignificant contribution to the combined standard uncertainty of *in situ* gamma-ray spectrometry measurements. However, since the angular response of the detector still has to be determined, no extra measurements are required with the method proposed in Paper I, and, as stated by GUM (ISO, 1995), any known significant systematic effects should preferably be eliminated or compensated for. It is also advantageous to obtain a general expression for the detector response that can be applied to a wide variety of measurement geometries.

Paper III points out a potential systematic effect in the intrinsic detector efficiency calibrations, namely the displacement of the virtual centre of the detector. When calculating the detector efficiency one must consider the distance from the source to the detector. Most often this distance is calculated to the centre of the detector. This will be a good approximation for higher photon energies, where the photons will interact with the detector material fairly evenly throughout the active volume. Lower photon energies, on the other hand, will not penetrate to the same depth in the detector and interactions will preferably take place closer to the surface. Consequently, there will be a displacement of the virtual centre of the detector towards the source. This effect is illustrated in Figure 15 (see Figure 8 for a description of some of the detector components), which shows interaction points for photons from a calibration source for a couple of energies (244.7 and 661.7 keV) and angles of incidence (0 and 40 degrees). For 244.7 keV photons there seems to be a displacement of the interaction centre of about 1.5 cm. This would result in a difference in the calculated intrinsic efficiency of about 5%. This is clearly not a negligible effect for lower energies, and would be worthwhile to consider in future work. Undoubtedly, this is one of the advantages with using Monte Carlo-based calibrations, as this effect then is not an issue.
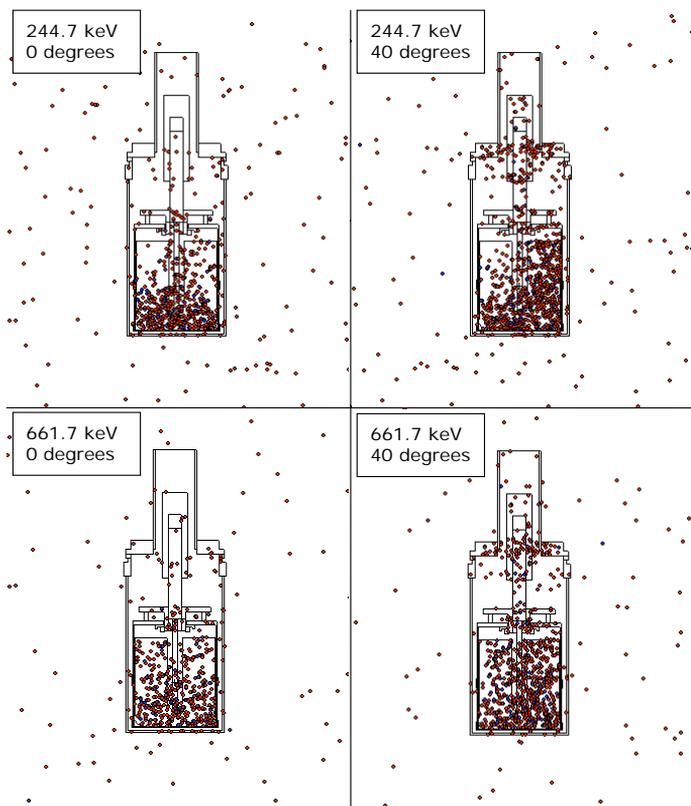
Figure 15. Photon interactions inside and outside the detector for a set of different photon energies and angles of incidence (indicated in the figure). The source-to-detector distance was 63.5 cm. The detector crystals are facing downwards in the picture (the nitrogen dewar is not included in the model).

Since the empirical expression for the soil attenuation coefficient used in the semi-empirical *in situ* calibrations is not valid below 300 keV, the intrinsic detector calibration was not extended below 244.7 keV. The inclusion of lower photon energies in the calibration would most likely be a complex task as these energies are much more sensitive to changes in thickness of any attenuating layer. The intrinsic detector efficiency may vary with ±50% with changing angle of incidence for a photon energy of 42 keV (Finck, 1992). The scanning of the detector in Paper II with a collimated 59.5 keV photon beam also revealed a highly irregular detector response over the front surface of the detector (Figure 17b). This fact, coupled with the fact that the detector efficiency function is known to display a distinct "knee" at about 100 keV, means that an expression for the detector efficiency function, valid also for lower energies, will most likely be highly complex.

In situ calibrations

The empirically determined expression for intrinsic detector efficiency was used in the numerical integration of Equation 8 (Paper I). The combined standard uncertainty of *in situ* measurements was assessed in Paper IV, and found to be 15-20% (k=1). Parameters contributing the most to the combined standard uncertainty were intrinsic detector efficiency, relative activity content in layers 1 and 2, and, to a lesser extent, soil densities. A minor reduction of *in situ* measurement uncertainty is expected if the new expression for the intrinsic efficiency is implemented.

The uncertainty in the correction factor for the three-layer model was, as previously mentioned, assessed by determining (with Monte Carlo simulations) the potential variation in *in situ* measurement results using different models for the activity depth distributions. The uncertainty of the correction factor was estimated to be 8.7%. This result is discussed further in the following section on Monte Carlo simulations of *in situ* measurements

In particular, assuming homogeneous activity distribution in the third layer (to the complete sampling depth) may introduce further bias in the measurement efficiency and a corresponding overestimation of the ground deposition activity level. It is, for instance, implied by Figure 13a that the ^{137}Cs activity does not extend below ~ 13 cm. The magnitude of this bias is easily assessed by correcting the thickness of the third layer and recalculating the measurement efficiency using the semi-empirical method. The resulting difference in *in situ* measurement efficiency was found to be about 2%, which can be considered negligible. The bias is expected to be equally insignificant for all measurement sites, due to low relative activity content and high attenuation of photons from the third layer, which means that the contribution to the full-energy peak count rate in the detector from this layer is often insignificant.

The total *in situ* measurement efficiency for Detector 1 on Sites 1, 2, 4 and 5 varies with a factor of about two. This is an indication of the uncertainties involved in mobile measurements, where a constant calibration factor is used (i.e. no use of, for instance, peak-to-valley corrections). MacDonald and colleagues (1996) estimate the uncertainty in *in situ* measurement of ^{137}Cs by guessing the depth distribution to be within a factor of four (about 10 years after the fallout).

For fresh fallout the activity depth distribution will of course be much more shallow than the distributions of ^{137}Cs encountered in Sweden today, more than 20 years after the Chernobyl fallout. Laedermann reports relaxation depths α^{-1} (according to an exponential model, Equation 2) of 0.3 cm for dry deposition and 1 cm for wet deposition. At such shallow depths, the precise shape of the activity depth distribution will not be as important. Sowa and co-workers (1989) compare calibration factors calculated for exponential and homogeneous slab depth distributions (α^{-1}=0.33 and d=0.54 cm; α^{-1}=1.0 and d=1.46 cm; α^{-1}=3.3 and d=4.3 cm), and finds a maximum difference of 6.4%. Furthermore, the actual depth distribution will be difficult to assess, as the resolution required is difficult to attain. For these reasons, activity depth distributions used for the calculation of *in situ* measurement efficiency for fresh fallout situations are often based on some assumption. In this thesis a 2 cm thick homogenous slab with density 500 kg·m^{-3} was used to describe the source matrix for fresh fallout (Lidström and Nylén, 1998). Sowa and co-workers (1989) calculate the maximum

difference in *in situ* measurement efficiency if the relaxation depth is changed by a factor of two. For relaxation depths of 0.33 and 1 cm, the differences range from 13 to 34%. This gives an indication of the uncertainties associated by such an assumption.

Monte Carlo modelling of detectors

Investigations of intrinsic detector efficiency

It is clear that the empirically determined efficiency of the detector differs significantly from the values obtained in MCNP simulations (Paper II). Such discrepancies have previously been reported by a number of other authors (e.g. Korun and Vidmar, 1997; Lépy et al., 2001). Ratios between Monte Carlo-calculated and empirical efficiencies for 661.7 keV photons were found to be 1.18 and 1.08 for Detector 1 and 2, respectively. The uncertainty in both correction factors were about 2.7% (k=1) (Paper IV). The correction factors are constant with respect to photon angle of incidence, but do vary with photon energy (Paper II).

Most often, this efficiency deficit is attributed to an increased dead layer thickness (Ashrafi et al., 1999; Bochud et al., 2006; Clouvas and Xanthos, 1998; Ewa et al., 2001; Gilmore and Hemingway, 1995; Hardy et al., 2002; Helmer et al., 2003; Herold and Kouzes, 1991; Hurtado et al., 2004; Huy et al., 2007; Kamboj and Kahn, 1996; Karamanis et al., 2002; Korun and Vidmar, 1997; Laborie et al., 2002; Lépy et al., 2001; Maleka and Maučec, 2005; Peyres et al., 2007; Ródenas et al, 2003; Sima et al., 2004; Vidmar et al., 2001; Wang et al., 2002). The reported thicknesses, necessary to compensate for the efficiency discrepancy, in the referenced papers match well with the measured thickness in Paper II, i.e. 1.5-1.9 mm. Recently, Huy, Bihn, and An (2007) have presented data indicating that the dead layer thickness may increase with time. It would certainly be interesting to redo the intrinsic detector efficiency calibrations and dead layer thickness measurements on the detectors used in this work in a few years time, to see if an increase in dead layer, and corresponding efficiency decrease, can be verified.

Radiographing the detector is important to confirm the physical dimensions of the crystal, as well as the actual position of the crystal within the detector housing (Aaltonen et al., 1994; Friedman et al., 2001; Hurtado et al., 2004; Nir-El and Sima, 2001). In Paper II, X-ray imaging of the two detectors used for *in situ* measurements in this thesis were performed (Figure 16). Whereas both lengths and diameters of the crystals were found to match the values supplied by the manufacturer, the radius of the bulletizing was found to be substantially larger than the size indicated on the drawing of the detector provided by the manufacturer. The radius of the bulletizing in the Monte Carlo model was adjusted accordingly. More recently, manufacturers have started to supply a value for this parameter, but this was not the case for the two older detectors used in this thesis (Detectors 1 and 2) The radiography also revealed a displacement of the crystal within the detector housing for one of the detectors. This is of less concern for *in situ* measurements, where the source-to-detector distance is large in comparison with the displacement. For measurements of samples or other close geometries it may, however, be of great significance.
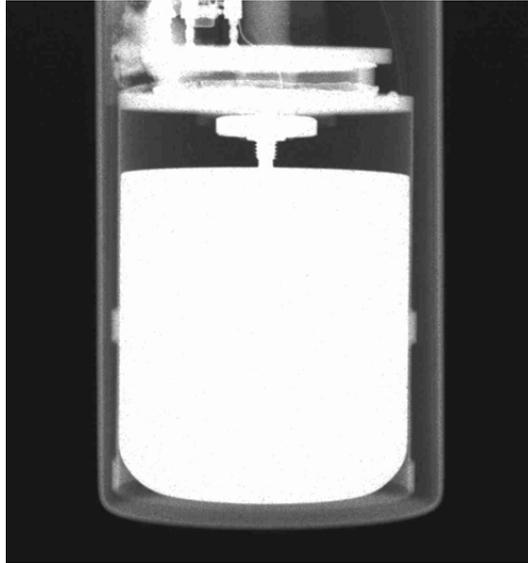
Figure 16. X-ray image of Detector 1.

The X-ray images do not, however, provide any information about the dimensions of the central hole in the crystal. Dryak and Kovar (2006) have in their work used radiation from both ^{137}Cs and ^{192}Ir to radiograph a detector. With these higher energy photons it is possible to produce radiographic images in which the hole can be seen.

The X-ray images revealed straps used for holding the crystal in place inside the mounting cup, which were not indicated on detector drawings. These straps will have an effect on low energy photons, as shown in Figure 17a where the relative detector response for a 59.5 keV collimated photon beam displays a small dip at about 4 cm from the front of the end cap. Some effect is observed even for 244.7 keV (Figure 18a), but for higher energies the effect of the mounting straps is negligible.

Furthermore, it is possible, albeit slightly difficult due to the rounded projection of the front of the detector, to confirm the crystal-to-end cap distance. In close geometries the crystal-to-end cap distance will naturally be of great importance; e.g. Aaltonen and colleagues (1994) and Karamanis and co-workers (2002) adjust this value in their model to achieve better agreement between Monte Carlo simulations and experimental data. The thickness of this gap can also be assessed with an ultra-sound probe, as done by Nir-El and Sima (2001). For larger source-to-detector distances, such as for *in situ* measurements, the crystal-to-end cap distance will not be important.

The scanning of the detector with collimated, low energy photon beams revealed that the response of the detector is far from homogeneous. In particular, a rather considerable decrease in detector response for 59.5 keV photons was found at the centre of the front surface (Figure 17b). As previously mentioned, the effects of the mounting straps can be seen in Figures 17a and 18a. Debertin and Grosswendt (1982), Keyser (2004) and Schläger (2007) have previously reported results from similar scannings of HPGe detectors. The results of Keyser (2004) and Schläger (2007) also

show the influence of the aluminium mounting straps. The fact that the measured response of the detector differs so radically from the results of the Monte Carlo simulations, indicates that very careful validation of any Monte Carlo detector model is required before it is employed at such low energies.
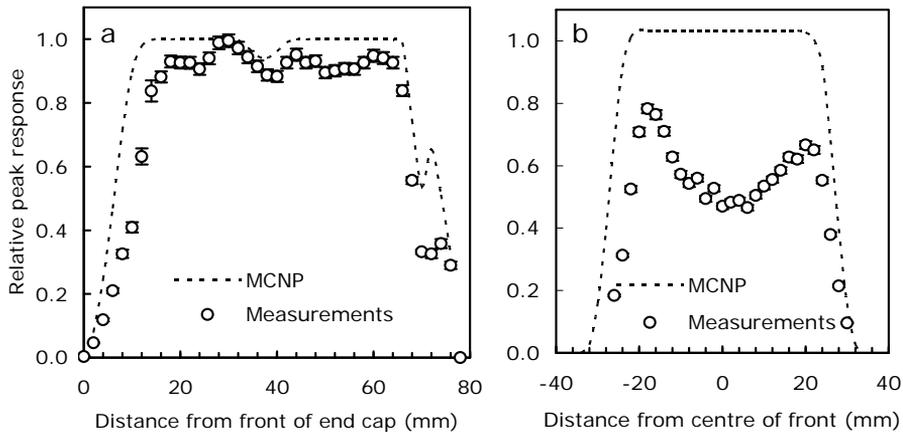

Figure 17. Results from scanning of the detector surface with a collimated 59.5 keV photon beam (a) along the lateral surface and (b) across the front face of the detector. Uncertainties represent one standard deviation due to counting statistics. Data is normalised to the highest value registered on the side of the detector.

Figure 18. Results from scanning of the detector surface with a collimated 244.7 keV photon beam (a) along the lateral surface and (b) across the front face of the detector. Uncertainties represent one standard deviation due to counting statistics. Data is normalised to the highest value registered on the side of the detector.
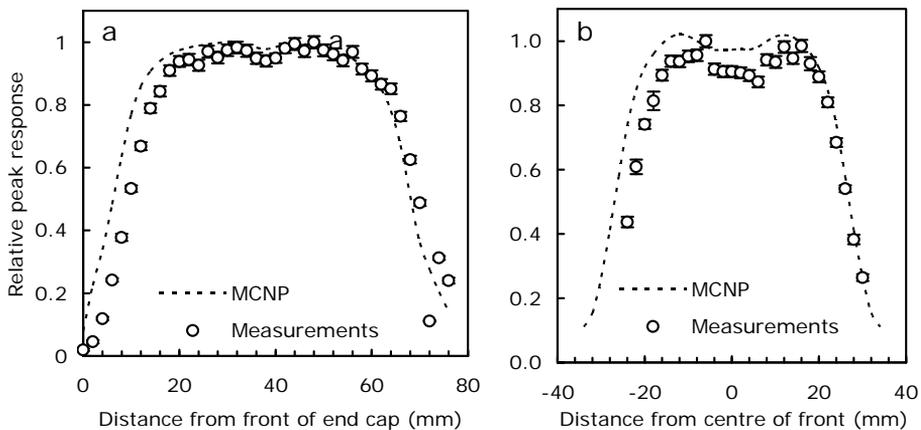
It was suggested (Friedman et al, 2001; Hernandez and El-Daoushy, 2003) that the irregular electrical field in the germanium crystal might have regions where the field is low enough to cause incomplete charge collection. These regions are thus considered by the authors to be inactive, i.e. photon interactions in these regions do not contribute to the full-energy peak. To investigate if properties of the electrical field might be the cause of the efficiency deficit found for Detector 1, the detector efficiency as a function of bias voltage was determined (Paper II). However, no decrease in efficiency was apparent until the depletion voltage was reached. Furthermore, incomplete charge collection will lead to tailing on the low energy side of the peak (Knoll, 2000), and not necessarily to a decrease in the full-energy peak area. It was thus concluded that the electrical field is most likely not to blame for the decreased efficiency of Detector 1.

The efficiency discrepancy could possibly be due to inaccuracies in the Monte Carlo code. An inter-comparison between seven codes is presented by Vidmar and co-authors (2008). The authors report discrepancies at lower photon energies of up to 10% between different codes. As no comparisons with experimental data are made, it is not possible to make any conclusions as to which codes produce the most correct results. For energies above 120 keV, discrepancies between different codes seems to be below 2%. This is not enough to account for the difference between empirical data and Monte Carlo simulations.

It was opted in Papers IV and V to use a correction factor to compensate for the higher efficiency of the Monte Carlo model, rather than adjusting the model to fit experimental data. This approach has the advantage, as concluded in paper II, that one does not have to assume any cause of the detector efficiency deficit. The task of fine-tuning the model may also prove to be quite laborious. This is especially true if lower energies are to be considered, as demonstrated by the scanning of the detector with a collimated 59.5 keV photon beam. It is important to notice, however, that the use of correction factors depend on the fact that the efficiency discrepancy is constant with the angle of photon incidence. If this is not the case, a bias will be introduced if the correction factor is applied to a measurement geometry with different angular distribution of the incident photon field than the one for which the correction factor was determined.

Simulations of in situ measurements

In Paper IV Monte Carlo simulations were used to calculate *in situ* measurement calibration factors, and the uncertainty in such calibrations was assessed. Results from Monte Carlo simulations were found to be in good agreement both with results from the semi-empirical calibration method and with ground deposition activity levels assessed by soil samples. The combined uncertainty of Monte Carlo-based calibrations for the five measurement sites was found to be 14-19% (*k*=1), which can be considered to be equal to the uncertainty of semi-empirical calibrations. It was concluded that it was safe to proceed using Monte Carlo methods to determine the influence on *in situ* measurements of various factors that cannot be handled by the semi-empirical calibration method.

An uncertainty component specific for Monte Carlo-based calibrations is the counting statistical uncertainty in Monte Carlo simulations of the *in situ* measurement setup. Because of the large areas that contribute to the detector response, and the correspondingly low measurement efficiency, large number of particles have to be

simulated in order to keep the statistical uncertainties reasonably low. In this thesis the statistical uncertainty in Monte Carlo simulations of *in situ* measurements was below 5% (*k*=1). This uncertainty could have been reduced, but as the counting statistical uncertainty was not found to be the major contributor to the combined calibration uncertainty, this was not considered necessary.

The uncertainty in the three-layer correction factor was determined in Paper IV by altering the source distribution in the Monte Carlo model, and recording the corresponding difference in measurement efficiency. As previously mentioned, two alternative models were fitted to experimental data and used to describe the depth distribution at Site 1: one exponential and one linearly decreasing with depth. The different models are shown in Figure 19. The resulting differences in *in situ* measurement efficiency were found to be within 15%. It was assumed that the alternative distributions are as different from the three-layer model as realistically possible. Consequently, one can assume the possible range of the systematic effect caused by the three-layer model to be rectangularly distributed with a maximum value of 15%. The corresponding standard uncertainty of the three-layer correction factor then becomes 8.6%. As this method for the determination of the systematic effect of the three-layer model is fairly crude and only based on data from one site, there is certainly room for improvement. However, as the three-layer correction is not the main contributor to the combined uncertainty of *in situ* measurements the effect of an improvement will be small, and the method was considered to be fit-for-purpose.
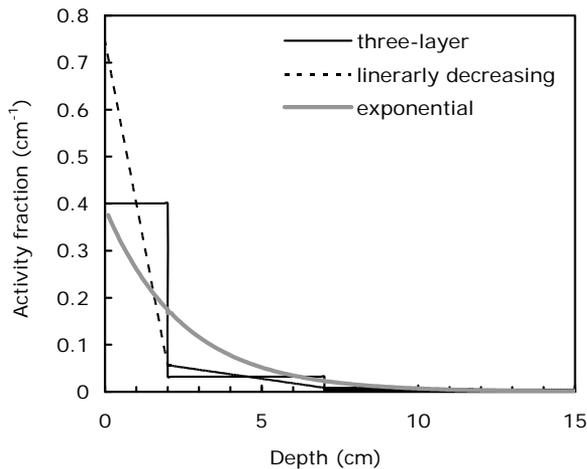

Figure 19. Different models for the depth distribution at Site 1, used to assess the magnitude of the systematic effect caused by the three-layer activity depth distribution model.

As previously mentioned, a Monte Carlo simulation of the *in situ* measurement was performed using the density and activity depth distributions from Site 3 that was determined with a higher resolution. In fact, no significant difference (less than 1%) was found in the resulting *in situ* measurement efficiency using this source term or using three-layer data from the 17 "standard" soil samples.

Comparisons between different methods

Initial field tests of the semi-empirical calibration method were performed and presented in Paper I. Good agreement with soil samples results were indicated, but no real conclusions could be made as an uncertainty estimate for the *in situ* measurements was still missing. Further validations of semi-empirical and Monte Carlo-based calibrations were performed in paper IV. Estimates of the ^{137}Cs ground deposition activity levels at the five sites based on soil sampling and *in situ* measurements, using both semi-empirical and Monte Carlo-based calibrations, are presented in Figure 20.
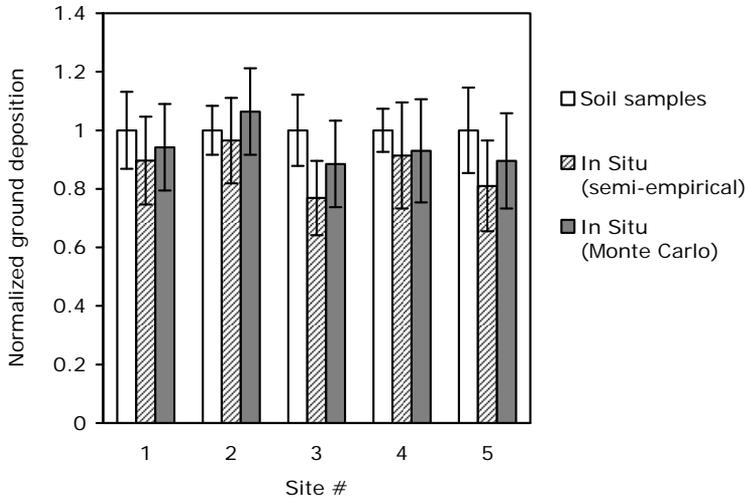

Figure 20. Results for ^{137}Cs ground deposition activity at the five sites from soil sampling and *in situ* measurements using either semi-empirical or Monte Carlo based calibrations. All data are normalized to the soil sample results for the corresponding site. Uncertainties are presented with a coverage factor of *k*=1.

Results for all methods agree within measurement uncertainties, indicating that estimates of measurement uncertainties are reasonable, and that no significant systematic effects are unaccounted for. The difference between soil sample results and *in situ* measurements using the semi-empirical method was on average 13%. However, according to ICRU (2006), the activity content in environmental samples is often found to be log-normally distributed. The average difference between the geometric mean of soil samples and *in situ* measurements is only about 2%. The average difference between the semi-empirical method and Monte Carlo-based calibrations was 8.3%. Uncertainties in *in situ* measurements were found to be somewhat higher then the standard deviation of the mean of the activity concentration found in the 17 soil samples at each site (16 samples at Site 4).

Influencing parameters

Effects on *in situ* measurement results by two potentially influencing parameters, ground curvature and activity in trees, were examined in Paper V. The measurement efficiency for measurements on a convex surface, i.e. on a hill, are decreased compared with measurements on a perfectly flat surface, as the curvature of the surface increases the distance in soil traversed by photons incident on the detector. Inversely, the efficiency is increased for measurements on a concave surface, i.e. in a depression. The resulting effects on *in situ* measurement effiency, as calculated in Paper V using Monte Carlo simulations, are illustrated in Figure 21.
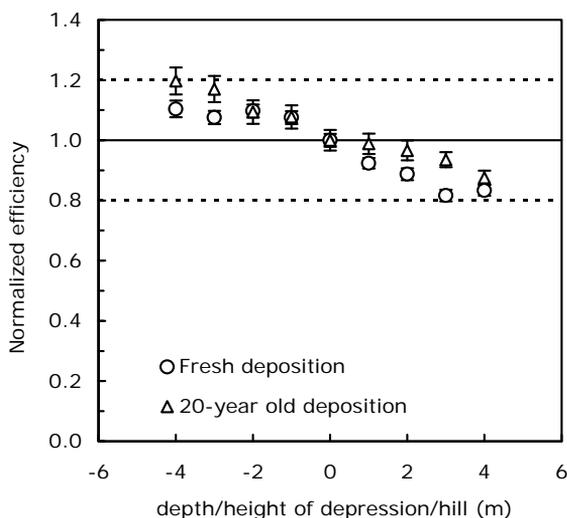

Figure 21. Relative change in *in situ* measurement efficiency with varying ground curvature for fresh and for 20-year old fallout of [137]Cs.

The measurement efficiency for source locations in tree crowns or stems were calculated in Paper V. These results, and results for source locations in soil with activity depth distributions representing either fresh or 20-year old fallout, are presented in Figure 22. It should be noted that the trees on Site 3 used as reference for the Monte Carlo tree model were fairly small, with an average height of 13.2 m and an average diameter of 14 cm, at a height of 1.3 m. The effects of attenuation in trees were found to be about 5%. According to Nylén (1996), shortly after the Chernobyl accident, up to 50% of the total activity deposition per unit area, was located in trees. The total *in situ* measurement efficiency for this situation is 78% of the efficiency without any trees present and all of the activity located in soil. Data from 1996 (Plamboeck et al., 2000) indicate that only 5% of the total activity per unit area were located in trees 10 years after the Chernobyl accident. In this case, the implications for *in situ* measurement efficiencies are certainly negligible.
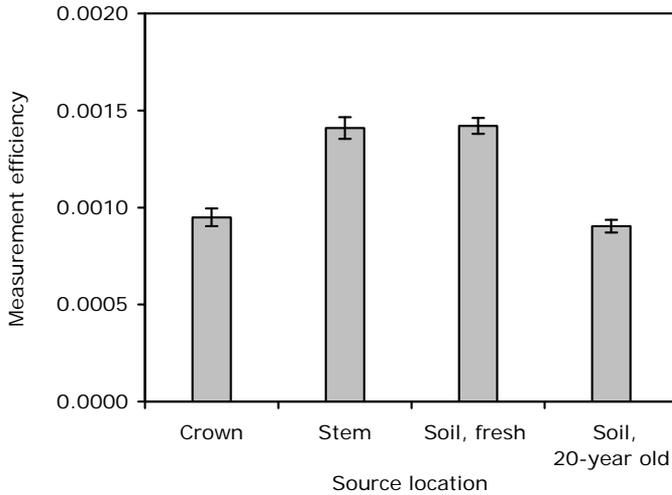
Figure 22. Detection efficiency for ^{137}Cs at various source locations: in tree crowns or stems; in soil assuming depth distribution of fresh fallout; and in soil with activity depth distribution of the Chernobyl fallout, 20 years after the accident. Tree and soil sample data from Site 3. Uncertainties represent one standard deviation.

Most notably, none of the investigated parameters seem to influence the results to more than about 25%. This is not a dramatic change and it was considered unnecessary to calculate specific correction factors. Furthermore, the previous assumption that *in situ* measurements should always be performed on an open flat surface for the calibration to be valid is not necessarily correct. In fact, always choosing a particular terrain type for the measurements may introduce further bias in results depending on the type of deposition. It is also important to remember that whereas the area that contributes to, for instance, 95% of the detector response may be very large, the majority of the contribution comes from a relatively small area. A 5 m radius is generally sufficient, for a 20-year old deposition, to encompass 60-70% of the contribution to the detector response. Thus, even if one wishes to make a measurement in an open area, free of interfering parameters, the radius of the area need not necessarily be that large.

CONCLUSIONS AND FUTURE PROSPECTS

A semi-empirical method for the calibration of *in situ* measurements is presented in Paper I. In contrast to the classic calibration method suggested by Beck, DeCampo and Gogolak (1972), both the efficiency and angular response of the detector is incorporated into a single efficiency parameter for the intrinsic detector efficiency. The intrinsic detector efficiency is experimentally determined and an empirical expression for the efficiency function is fitted to the data. Furthermore, a three-layer model was proposed for the description of activity and density depth distributions. This model is well-suited to the soil sampling procedure and adaptable to a wide range of activity and density depth distributions. Two detectors were calibrated according to the proposed method for a photon energy range of 244.7-1408 keV. In Paper IV, ground deposition levels assessed by the semi-empirical method and results from soil sample measurements were found to be in good agreement.

The uncertainty in intrinsic detector efficiency calibrations, performed in accordance with the semi-empirical method, is estimated according to a GUM approach in Paper III. The combined uncertainty was found to be 5.2 and 8.1% (k=1) for the two calibrated detectors, with the main contribution originating from uncertainty in the fit of the efficiency function to empirical data. Because of this, a new expression for the intrinsic detector efficiency was proposed, which reduced the uncertainties in the two detector calibrations to 3.7 and 4.2% (k=1). The new expression, however, has yet to be implemented in the semi-empirical calibration method.

In Paper V the combined standard uncertainty of *in situ* measurements performed on the ^{137}Cs deposition from the Chernobyl accident, about 20 years after the initial fallout, was estimated. The uncertainty was found to be 15-20% (k=1), with the main contributions coming from the intrinsic detector efficiency and the distribution of the activity content in the two top layers. For certain conditions the uncertainty in density of the top layer also contributes significantly. Although the uncertainty in the intrinsic detector efficiency contributes to the combined uncertainty of *in situ* measurements, it is not the dominating factor. Hence, the reduction in *in situ* measurement uncertainty obtained when implementing the new expression for intrinsic detector efficiency is expected to be fairly small. However, for fresh fallout with a more shallow activity distribution, it is reasonable to assume that the intrinsic detector efficiency contributes to the *in situ* measurement uncertainty to a larger extent. In this case, the benefits of the new expression may be more substantial.

The work in this thesis was mainly based on measurements of ^{137}Cs from the Chernobyl fallout, which has migrated to some depth in soil. The uncertainty of *in situ* measurements of fresh fallout according to the proposed semi-empirical method is a question that remains to be addressed.

Furthermore, the resulting *in situ* measurement uncertainty as a function of the number of soil samples collected to obtain information about the activity and density depth distributions should be calculated. This will facilitate the determination of a suitable number of soil samples to be collected for future *in situ* measurement calibrations. It would also be interesting to extend the detector calibrations to a wider range of photon energies. However, the incorporation of lower photon energies may

prove to be a complicated task. As indicated by the scanning of the detector surface the detector response for low photon energies may be highly inhomogeneous. It is also clear that the uncertainty in measurements will most likely increase for lower energies, as they are more sensitive to attenuation.

Some important conclusions can be drawn from the investigations of the detector and detector response performed in Paper II. As a first step towards a more accurate Monte Carlo model of the detector, radiographing of the detector should be performed in order to verify the physical dimensions of the crystal and detector housing. Results from the scanning of the detector surface with a collimated low energy photon beam further emphasize the need for careful validation of the model. For one of the detectors used in this study the dead layer thickness was estimated to be about twice the nominal value. A thicker dead layer will affect both higher and lower photon energies, but for slightly different reasons. Lower energies will be sensitive to the thickness of any attenuating layer. Higher energies, for which the attenuation in the dead layer may be negligible, will still be affected since an increase in dead layer thickness will result in a corresponding decrease in active volume of the detector.

In Paper IV, Monte Carlo simulations were used to calculate *in situ* measurement efficiencies, based on the three-layer activity depth distribution model used for the semi-empirical calibrations. The Monte Carlo-based calibrations were found to yield similar results and uncertainties as the semi-empirical model. Good agreement with soil sample results was also achieved.

The Monte Carlo detector models were subsequently employed to assess the magnitude of potential systematic effects on *in situ* measurement results. Initial studies of the effects of ground curvature and activity in trees are presented in Paper V. Results indicate that both ground curvature and activity in trees can potentially influence the results from *in situ* measurements with up to 20-25%. This is, however, not a substantially high figure if one considers measurement uncertainty and natural variability. Particularly, for older fallout, where only a minor fraction of the activity is located in trees, the influence on *in situ* measurements is negligible. In fact, it is not unreasonable to suspect that the consequent positioning of the detector on a large open and flat field, as the current paradigm of *in situ* measurements suggests in order for the calibration to be as accurate as possible, may in fact lead to systematic effects larger than the ones caused influencing parameters potentially interfering parameters such as trees.

ACKNOWLEDGEMENTS

REFERENCES

Aaltonen, H., Klemola, S., Ugletveit, F., 1994. Nuclear Instruments and Methods in Physics Research A 339, 87-91.

Allyson, J. D., Sanderson, D. C. W., 1998. Monte Carlo Simulation of Environmental Airborne Gamma-Spectrometry. Journal of Environmental Radioactivity 38, 259-282.

Almgren, S., Isaksson, M., 2006. Vertical migration studies of ^{137}Cs from nuclear weapons fallout and the Chernobyl accident. Journal of Environmental Radioactivity 91, 90-102.

Ashrafi. S., Likar, A., Vidmar, T., 1999. Precise modeling of a coaxial HPGe detector. Nuclear Instruments and Methods in Physics Research A 438, 421-428.

Beck, H. L., Condon, W. J., Lowder, W. M., 1964. Spectrometric techniques for measuring environmental gamma radiation. USAEC Report HASL-150.

Beck H.L., DeCampo J. and Gogolak C., 1972. In situ Ge(Li) and NaI(Tl) gamma-ray spectrometry for the measurement of environmental radiation. USAEC Report HASL-258.

Benke, R. R., Kearfott, K. J., 2001. An improved in situ method for determining depth distributions of gamma-emitting radionuclides. Nuclear Instruments and Methods in Physics Research A 463, 393-412.

Benke, R. R., Kearfott, K. J., 2002. Demonstration of a collimated in situ method for determining depth distributions using γ-ray spectrometry. Nuclear Instruments and Methods in Physics Research A 482, 814-831.

Bochud, F., Bailat, C. J., Buchillier, T., Byrde, F., Schmid, E., Laedermann, J.-P., 2006. Simple Monte-Carlo method to calibrate well-type HPGe detectors. Nuclear Instruments and Methods in Physics Research A 569, 790-795.

Boson, J., Lidström, K., Nylén, T., Ågren, G. and Johansson, L., 2006. In Situ Gamma-ray Spectrometry for Environmental Monitoring: a Semi Empirical Calibration Method. Radiation Protection Dosimetry 121, 310-316.

Boson, J., Ågren, G. and Johansson, L., 2008. A detailed investigation of HPGe detector response for improved Monte Carlo efficiency calculations. Nuclear Instruments and Methods in Physics Research A 587, 304-314.

Brune, D., 2001. Nuclear Technology. In: Brune, D., Hellborg, R., Persson, B. R. R., Pääkkönen, R., Eds. Radiation at home, outdoors and in the workplace. Scandinavian Science Publisher, Oslo, Norway, 166-186. ISBN: 82-91833-02-8
Clouvas, A., Xanthos, S., Antonopoulos-Domis, M., Silva, J., 1998. Monte Carlo based method for conversion of in-situ gamma ray spectra obtained with a portable Ge detector to an incident photon flux energy distribution. Health Physics 74, 216-230.

Clouvas, A., Xanthos, S., Takoudis, G., Antonopoulos-Domis, M., Zinoviadis, G., Vidmar, T., Likar, A., 2007. Twenty-year follow-up study of radiocesium migration in soil. Radiation Protection Dosimetry 124, 372-377.

Debertin, K, Grosswendt, B., 1982. Efficiency calibration of semiconductor detectors by primary standard sources and Monte Carlo calculations. Instruments and Methods 203, 343-352.

Dryak, P., Kovar, P., 2006. Experimental and MC determination of HPGe detector efficiency in the 40-2754 keV energy range for measuring point source geometry with the source-to-detector distance of 25 cm. Applied Radiation and Isotopes 64, 1346-1349.

Edvarson, K., 1991. Fallout over Sweden from the Chernobyl accident. In: Moberg, L. ed. The Chernobyl fallout in Sweden: Results from a research programme on environmental radiology. Stockholm: Swedish Radiation Protection Institute, 47-65.

Ewa, I.O.B., Bodizs, D., Czifrus, Sz., Molnar, Zs., 2001. Monte Carlo determination of full energy peak efficiency for a HPGe detector. Applied Radiation and Isotopes 55, 103-108.

Finck, R. R., 1992. High resolution field gamma spectrometry and its application to problems in environmental radiology. (Thesis) Lund: Department of Radiation Physics, Lund University. ISBN 91-628-0739-0.

Friedman, R. J., Reichard, M. C., Blue, T. E., Brown, A. S., 2001. Evaluation of scatter contribution from shielding materials used in scatter measurements for calibration range characterization. Health Physics 80, 54-61.

Gering, F., Hillmann, U., Jacob, P., Fehrenbacher, G., 1998. In situ gamma-spectrometry several years after deposition of radiiocesium, II. Peak-to-valley method. Radiation Environmental Biophysics 37, 283-291.

Gering, F., Kiefer, P., Fesenko, S., Voigt, G., 2002. In situ gamma-spectrometry in forests: determination of Kerma rate in air from 137Cs. Journal of Environmental Radioactivity 61, 75-89.

Gilmore, G., Hemingway, J.D., 1995. Practical Gamma-ray Spectrometry, Wiley, Chichester. ISBN 0-471-95150-1.

Gutierrez-Villanueva, J. L., Martín-Martín, A., Peña, V., García-Talavera, M., Gómez, C., Pardos, R., 2006. Monte Carlo calibration of a portable HPGe detector to determine 137Cs inventories in soils. Applied Radiation and Isotopes 64, 1333.

Hardy, J.C., Iacob, V.E., Sanchez-Vega, M., Effinger, R.T., Lipnik, P., Mayes, V.E., Willis, D.K., Helmer, R.G., 2002. Precise efficiency calibration of an HPGe detector: source measurements and Monte Carlo calculation with sub-percent precision. Applied Radiation and Isotopes 56, 65-69.

He, Q., Walling, D. E., 2000. Calibration of a field-portable gamma detector to obtain in situ measurements of the ^{137}Cs inventories of cultivated soils and floodplain sediments. Applied radiation and Isotopes 52, 865-872.

Helfer, I.K., Miller, K.M., 1988. Calibration Factors for Ge Detectors Used for Field Spectrometry. Health Physics 55, 15-29.

Helmer, R.G., Hardy, J.C., Iacob, V.E., Sanchez-Vega, M., Neilson, R.G., Nelson, J., 2003. The use of Monte Carlo calculations in the determination of a Ge detector efficiency curve. Nuclear Instruments and Methods in Physics Research A 511, 360-381.

Hernandez, F., El-Daoushy, F., 2003. Accounting for incomplete charge collection in Monte Carlo simulations of the efficiency of well-type Ge-detectors. Nuclear Instruments and Methods in Physics Research A 498, 340-351.

Herold, L.K., Kouzes, R.T., 1991. Intrinsic Germanium Detector Efficiency Calculations. IEEE Transactions on Nuclear Science 38, 231-238.

Hillmann, U., Schimmack, W., Jacob, P., Bunzl, K., 1996. In situ γ-spectrometry several years after deposition of radiocesium. Radiation Environmental Biophysics 35, 297-303.

Hjerpe, T., Samuelsson, C., 2002. Accounting for the depth distribution of ^{137}Cs in on-line mobile gamma spectrometry through primary and forward-scattered photons. Radiation Environmental Biophysics 41, 225-230.

Hurtado, S., García-León, M., García-Tenorio, R., 2004. Monte Carlo simulations of the response of a germanium detector for low-level spectrometry measurements using GEANT4. Applied Radiation and Isotopes 61, 139-143.

Huy, N.Q., Bihn, D.Q., An, V.X., 2007. Study on the increase of inactive germanium layer in a high purity germanium detector after a long time operation applying MCNP code. Nuclear Instruments and Methods in Physics Research A 573, 384-388.

ICRU, 1994. International Commission on Radiation Units and Measurements, Gamma-Ray Spectrometry in the Environment. ICRU Report 53, Bethesda, MD, USA.

ICRU, 2006. International Commission on Radiation Units and Measurements, Sampling for Radionuclides in the Environment. ICRU Report 75, Oxford University Press.

Isaksson, M., Erlandsson, B., 1995. Experimental Determination of the Vertical and Horizontal Distribution of ^{137}Cs in the ground. Journal of Environmental Radioactivity 27, 141-160.

Kamboj, S., Kahn, B., 1996. Evaluation of Monte Carlo simulation of photon counting efficiency for germanium detectors. Health Physics 70, 512-519.

Karamanis, D., Lacoste, V., Andriamonje, S., Barreau, G., Petit, M., 2002. Experimental and simulated efficiency of a HPGe detector with point-like and extended sources. Nuclear Instruments and Methods in Physics Research A 487, 477-487.

Kastlander, J., Bargholtz, C., 2005. Efficient in situ method to determine radionuclide concentration in soil. Nuclear Instruments and Methods in Physics Research A 547, 400-410.

Keyser, R.M., 2004. Resolution and Sensitivity as a Function of Energy and Incident Geometry for Germanium Detectors. Nuclear Instruments and Methods in Physics Research B 213, 236-240.

Knoll, G.F., 2000. Radiation Detection and Measurement, third ed. John Wiley & Sons, New York.

Korun, M., Martinčič, R., Pucelj, B., 1991. In-situ measurements of the radioactive fallout deposit. Nuclear Instruments and Methods in Physics Research A 300, 611-615.

Korun, M., Vidmar, T., 1997. Monte Carlo calculation of the spatial dependence of the coaxial HPGe detector efficiency for point sources. Nuclear Instruments and Methods in Physics Research A 390, 203-208.

Krstić, D., Nikezić, D., Stevanović, N., Jelić, M., 2004. Vertical profile of ^{137}Cs in soil. Applied Radiation and Isotopes 61, 1487-1492.

Laborie, J.-M., Le Petit, G., Abt, D., Girard, M., 2002. Monte Carlo calculation of the efficiency response of a low-background well-type HPGe detector. Nuclear Instruments and Methods in Physics Research A 479, 618-630.

Lépy, M.C., Altzitzoglou, T., Arnold, D., Bronson, F., Capote Noy, R., Décombaz, M., De Corte, F., Edelmaier, R., Herrera Peraza, E., Klemola, S., Korun, M., Kralik, M., Neder, H., Plagnard, J., Pommé, S., de Sanoit, J., Sima, O., Ugletveit, F., Van Velzen, L., Vidmar, T., 2001. Intercomparison of efficiency transfer software for gamma-ray spectrometry. Applied Radiation and Isotopes 55, 493-503.

Lettner, H., Bossew, P., Hubmer, A. K., 2000. Spatial variability of fallout Caesium-137 in Austrian alpine regions. Journal of Environmental Radioactivity 47, 71-82.

Lidström, K., Nylén, T., 1998. Beredskapsmetoder för mätning av radioaktivt nedfall. FOA-R--98-00956-861—SE, FOA, Umeå, Sweden.

Likar, A., Vidmar, T., Lipoglavšek, M., Omahen, G., 2004. Monte Carlo calculation of entire in situ gamma-ray spectra. Journal of Environmental Radioactivity 72, 163-168.

Losana, M. C., Magnoni, M., Righino, F., 2001. Comparison of Different Methods for the Assessment of the Environmental Gamma Dose, Radiation Protection Dosimetry, 97 (4), 333-336.

Maleka, P.P., Maučec, M., 2005. Monte Carlo uncertainty analysis of germanium detector response to γ-rays with energies below 1 MeV. Nuclear Instruments and Methods in Physics Research A 538, 631-639.

Mattsson, S., Vesanen, R., 1988. Patterns of Chernobyl fallout in relation to local weather conditions. Environment International 14, 177-180.

MacDonald, J., Smith, P. H., Assinder, D. J., 1996. The development and use of an *in situ* gamma-ray spectrometry system in North Wales. Journal of Radiological Protection 16, 115-127.

MacDonald, J., Gibson, C. J., Fish, P. J., Assinder, D. J., 1998. The significance of variations in the angular correction factor in in situ gamma spectrometry. Journal of Radiological Protection 18, 37-42.

McKinney, G., Durkee, J., Hendricks, J., James, M., Pelowitz, D., Waters, L., 2006. Review of Monte Carlo All-Particle Transport Codes and Overview of Recent MCNPX Features. International Workshop on Fast Neutron Detectors, University of Cape Town, South Africa, April 3-6.

NATO, 2005. NATO Handbook for Sampling and Identification of Radiological Agents (SIRA), Volume 1 – Operational, AEP-49.

Nir-El, Y., Sima, O., 2001. Angular response of a HPGe detector to gamma-rays in in situ measurements. Applied Radiation and Isotopes 55, 1-7.

Nylén, T., 1996. Uptake, turnover and transport of radiocaesium in boreal forest ecosystems. (Thesis) Uppsala: Department of Radioecology, Swedish University of Agricultural Sciences. ISBN 91-576-5149-3.

Oertel C. P., Giles J. R., Thompson K. C., Wells R. P., 2004. In situ depth profiling of 137Cs contamination in soils at the Idaho NationalEngineering and Environmental Laboratory. Health Physics 87, 664-669.

Owens, P. N., Walling, D. E., 1996. Spatial Variability of Caesium-137 Inventories at Reference Sites: an Example from Two Contrasting Sites in England and Zimbabwe. Applied Radiation and Isotopes 47, 699-707.

Peyres, V., García-Toraño, E., Efficiency calibration of an extended-range Ge detector by a detailed Monte Carlo simulation. Nuclear Instruments and Methods in Physics Research A 580, 296-298.

Plamboeck, A. H., Nylén, T., Grip, H., 2000. Uptake of cations under two different water regimes in a boreal Scots pine forest. The Science of the Total Environment 256, 175-183.

Plamboeck, A. H., Nylén, T., Ågren, G., 2006. Comparative estimations of ^{137}Cs distribution in a boreal forest in northern Sweden using a traditional sampling approach and a portable NaI detector. Journal of Environmental Radioactivity 90, 100-109.

Ródenas, J., Pascual, A., Zarza, I., Serradell, V., Ortiz, J., Ballesteros, L., 2003. Analysis of the influence of germanium dead layer on detector calibration simulation for environmental radioactive samples using the Monte Carlo method. Nuclear Instruments and Methods in Physics research A 496, 390-399.

Samuelsson, C., 2001. The Earth. In: Brune, D., Hellborg, R., Persson, B. R. R., Pääkkönen, R., Eds. Radiation at home, outdoors and in the workplace. Scandinavian Science Publisher, Oslo, Norway, 166-186. ISBN 82-91833-02-8.

Schläger, M., 2007. Precise modelling of coaxial germanium detectors in preparation for a mathematical calibration. Nuclear Instruments and Methods in Physics research A 580, 137-140.

SGAB, 1986. Cs-137 ground deposition map over Sweden. Map 1:2000000

Sima, O., Cazan, I.L., Dinescu, L., Arnold, D., 2004. Efficiency calibration of high volume samples using the GESPECOR software. Applied Radiation and Isotopes 61, 123-127.

Sowa, W., Martini, E., Gehrcke, K., Marschner, P., Naziry, M. J., 1989. Uncertainties of in situ gamma spectrometry for environmental monitoring. Radiation Protection Dosimetry 27, 93-101.

Thummerer, S., Jacob, P., 1998. Determination of depth distributions of natural radionuclides with in situ gamma-ray spectrometry. Nuclear Instruments and Methods in Physics Research A 416, 161-178.

Tyler, N., 1999. Monitoring anthropogenic radioactivity in salt march environments through in situ gamma-ray spectrometry. Journal of Environmental Radioactivity 45, 235-252.

Tyler, N., 2004. High accuracy in situ radiometric mapping. Journal of Environmental Radioactivity 72, 195-202.

Tyler, A. N., Sanderson, D. C. W., Scott, E. M., Allyson, J. D., 1996. Accounting for Spatial Variability and Fields of View in Environmental Gamma Ray Spectrometry, Journal of Environmental Radioactivity 33, 213-235.

Van Riper, K. A., Metzger, R. L., Kearfott, K. J., 2002. Radionuclide Depth Distribution by Collimated Spectroscopy. Proceedings of the Topical Meeting on Radiation Serving Society, Santa Fe, NM, April 2002, on CD-ROM, American Nuclear Society, LaGrange Park, IL.

Vidmar, T., Korun, M., Likar, A., Martinčič, R., 2001. A semi-empirical model of the efficiency curve for extended sources in gamma-ray spectrometry. Nuclear Instruments and Methods in Physics Research A 470, 533-547.

Vidmar, T., Aubineau-Laniece, I., Anagnostakis, M. J., Arnold, D., Brettner-Messler, R., Budjas, D., Capogni, M., Dias, M.S., De Geer, L-E., Fazio, A., Gasparro, J., Hult, M., Hurtado, S., Jurado Vargas, M., Laubenstein, M., Lee, K.B., Lee, Y-K., Lepy, M-C., Maringer, F-J., Medina Peyres, V., Mille, M., Moralles, M., Nour, S., Plenteda, R., Rubio Montero, M. P., Sima, O., Tomei, C., Vidmar, G., 2008. An intercomparison of Monte Carlo codes used in gamma-ray spectrometry. Applied Radiation and Isotopes 66, 764-768.

Wang, Z., Kahn, B., Valentine, J.D., 2002. Efficiency Calculation and Coincidence Summing Correction for Germanium Detectors by Monte Carlo Simulation. IEEE Transactions on Nuclear Science 49, 1925-1931.

TACK

Nu när avhandlingen är färdigskriven skulle jag vilja tacka alla som på olika sätt bidragit till mitt arbete eller gjort de senaste fem åren till vad de blev. En komplett lista är såklart omöjlig, men jag vill rikta ett särskilt tack till:

- Mina handledare Lennart Johansson och Göran Ågren för allt stöd under min tid som doktorand.

- Kenneth Lidström för många givande diskussioner om mätteknik och annat.

- Agneta Hånell Plamboeck och Torbjörn Nylén för alla givande diskussioner om statistik, radioekologi och provtagningsstrategier, och för all hjälp med provtagning och mätningar.

- Erik Johansson – du har varit en ypperlig "room mate" under tiden på FOI.

- Henrik Ramebäck för allt engagemang du visat för mitt arbete, för värdefulla kommentarer på avhandlingen och för att du dragit ner mig i osäkerhetsträsket – det har varit jobbigt men givande!

- Björn Sandström för nitisk korrekturläsning av avhandlingen och ingående arbeten.

- Daniela Stricklin för kommentarer på språket i de första artiklarna.

- Alla de andra medlemmarna av R-gruppen.

- Karin Lindh och Catharina Söderström för hjälpen med skanningen av jordprover.

- Leif Persson för att du tålmodigt svarat på diverse statistiska spörsmål.

- Alla de övriga kollegorna på FOI som verkligen fått mig att trivas, för alla trevliga fikastunder, innebandymatcher, fester och pubar.

- Doktorandkollegorna på Radiofysik: Jörgen Olofsson och Tufve Nyholm. Tack för trevliga sammankomster och för att ni gjorde vistelserna i Norge uthärdliga.

Jag vill också passa att tacka till mina föräldrar som alltid stöttat, hjälpt och skämt bort mig.

Slutligen, ett jättestort tack min älskade Klara för att du finns och för allt stöd under jobbet med avhandlingen. Och, när jag tänker efter, för att du släpade med mig till Umeå – annars hade ju denna avhandling aldrig blivit till!