In situ measurements of radionuclide concentration in soil

An investigation into detector properties and methods

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Abstract

In case of a release of radioactive nuclides into the environment it is necessary to have reliable methods to estimate the potential effect on people and the ecosystem. In this context the total activity deposited, the elemental composition and the depth distribution are of importance.

An efficient in situ method to estimate the average contamination over larger areas using high purity germanium (HPGe) detectors has been developed. The method combines simulation of photon transport with measured detector properties. The total activity of $^{137}$Cs determined from gamma-ray spectra recorded in situ are compared to results from soil sampling.

Another in situ method has been developed to determine the depth distribution of a radionuclide contamination, using an array of small detectors inserted into the ground, as an alternative to the standard procedure of soil sampling. The possibility to use cadmium telluride (CdTe) detectors as well as lanthanum bromide (LaBr$_3$) detectors has been investigated. As a demonstration of the developed method the small-scale variation of the activity distributions in an area covering 350 m$^2$ have been measured and is compared to results obtained by other methods.
This thesis is based on the following papers, which are referred to in the text by their Roman numerals. The author has had the main responsibility for all the papers.


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1. Svensk inledning

Vi utsätts hela tiden för joniserande strålning från vår omgivning. Det finns flera olika typer av joniserande strålning, bland annat alfa-, beta-, gamma- och röntgenstrålning, men alla karakteriseras av förmågan, att direkt eller via kärnreaktioner, frigöra elektroner vid växelverkan med atomer.

Strålningen härstammar till största del från radioaktiva atomkärnor som sönderfaller i vår omgivning, men vi utsätts även för joniserade strålning till exempel när vi besöker tandläkaren eller sjukhuset och genomgår röntgenundersökningar.

De radioaktiva atomkärnorna har olika ursprung, en del nyskapas ständigt i atmosfären genom att högenergetiska partiklar i den kosmiska strålningen växelverkar med atomkärnor i luften. En del radioaktiva atomkärnor (med väldigt långa halveringstider) har funnits i berggrunden sedan jorden blev till. Det radioaktiva sönderfallet av dessa ämnen ger upphov till bland annat varmen i jordens inre men också till radonproblematiken. Andra radioaktiva ämnen som återfinns i naturen skapar vi själva, exempel på detta är bland annat det globala nedfallet från provsprängningarna av kärnvapen som kulminerade i mitten på 1960-talet, men också utsläpp på grund av olyckshändelser, som reaktorhaveriet i Tjernobyl 1986.

I samband med ett utsläpp av radioaktiva ämnen är det viktigt att ha effektiva och pålitliga metoder för att kunna mäta föroreningens omfattning. Detta för att kunna avgöra vilka åtgärder som bör vidtas för att skydda och minimera skador på människa och miljö.

Nedfall av radioaktiva ämnen som har släppts ut i atmosfären sker ofta i samband med nederbörd vilket komplikrar bestämningen av kontaminerings omfattning. När de radioaktiva ämnenas tränger ner i marken, absorberas en del av strålningen i jorden, och mänstrumentet registrerar således enbart en mindre del av den mängd radioaktiva ämnen som finns i området. Detta räcker i och för sig för att bestämma exponeringen av strålning i det akuta skedet, men för att kunna uppskatta hur människa och miljö kommer påverkas över en längre tid behövs kännedom om den totala belägningen av radioaktiva ämnen samt information om ämnenas identitet och deras djupfördelning. Detta är bland annat viktigt för att avgöra om de radioaktiva ämnen kan tas upp av växter och föras vidare uppåt (och ackumuleras) i näringskedjan, till exempel från gräs till ko och vidare till människan (via mjölk och kött).
Den här avhandling handlar om utvecklingen av metoder för att snabbt identifiera de radioaktiva ämnena samt bestämma koncentrationer och djupfördelningar i marken med hjälp av gammastrålning uppmätt direkt på plats.


Detaljerad information om de radioaktiva ämnena djupfördelning bestäms traditionellt genom att ta jordprover som sedan analyseras i laboratoriet, en procedur som är arbetskrävande och tar tid. Även om laboratoriemätningarna kan göras med nästan godtycklig precision så finns det trots det stora osäkerheter i resultaten på grund av ojämnheter i jorden (till exempel stenar och rötter) och deformation av provet vid upptagningen. Till detta kommer att beläggningen ofta uppvisar stora variationer även över små avstånd, vilket ofta kräver att ett stort antal prov tas för att nå ett tillförlitligt resultat.

Målet har varit att undersöka om det är möjligt att ersätta, alternativt komplettera, den tidskrävande jordprovtagningen med en metod där djupfördelningen av de radioaktiva ämnena bestäms direkt genom mätning av gammastrålningen i naturen, med hjälp av små detektorer nedsänkta i marken. Inom ramen för detta har olika detektorers egenskaper studerats i laboratoriet och två fältmätningar utförts där metoden har utvecklats och resultaten jämförts med jordprover. Slutligen har beläggningen av det radioaktiva ämnet cesium kartlagts över ett 350 m² stort område norr om Gävle, som en demonstration av den utvecklade metoden. Det erhållna resultatet står i god överensstämmelse med andra mätningar i området.
2. Introduction

In case of a release of radioactive nuclides into the environment it is necessary to have reliable methods to estimate the potential effects on people and the ecosystem in order to apply relevant countermeasures. A first estimate can be obtained using devices that simply register the number of ionizing particles [1–3]. In order to make a more detailed description of the contamination detectors with the possibility to identify and distinguish between different radionuclides are needed. This information is crucial for the estimation of the future dose [3].

The migration of the radionuclides into the soil complicates the situation because of the shielding effect. This is normally a slow process (∼1-30 mm/year) [4–6], but is accelerated if the deposition occurs with rain. A similar complication occurs if the activity is deposited with or before snow fall [7]. Therefore it is also important to be able to determine the depth distribution [8–10]. The vertical distribution is of large interest, for example, in estimating the possible uptake of radionuclides in the biosphere (plants and mushrooms among others) and also in the decision making of what countermeasures should be applied, for example, removal of the upper layer of soil [11].

The depth distribution is traditionally determined from extracted soil samples. The sampling method is labor intensive and time consuming [1,3,4]: after a careful extraction of the samples in order to minimize cross contamination (between samples) and deformation, the samples have to be transported to the laboratory where the activity is determined by gamma-spectrometry. The soil samples can provide, among other things, an estimate of the total activity deposited and the depth distribution. The results, however, corresponds to local values. For anthropogenic radionuclides the process of deposition, ground properties and topography can lead to large fluctuation even over small distances (a few meters or less) [7,9,12]. Studies [13] have suggested a minimum of 15 soil samples per site for an adequate map of the variations. Therefore the sampling procedure is not optimal, neither in case of an emergency or for routine monitoring, and it is of great importance to develop in situ (on site) methods which make a efficient and reliable determination of the concentration of radionuclides possible.

The thesis consists of three parts. The first part is dealing with the further development of a method regarding the determination of the average radionuclide concentration over a larger area. The concentration is estimated from an in situ measurement of the gamma-ray flux using standard spectroscopy
equipment including a high purity germanium (HPGe) detector. The second part deals with preliminary investigations aiming at developing an in situ method to estimate the vertical activity distribution in detail, a method that could replace or be a complement to the traditional soil sampling. The possibility to use cadmium telluride (CdTe) or lanthanum bromide (LaBr$_3$) detectors is investigated and the resolution and efficiency are compared to that of sodium iodide (NaI) detectors. In the third part field tests are described and as demonstration of the implementation of the developed method a mapping of the small-scale variation of the activity distributions in the Ut-nora marsh (within the high level Chernobyl fallout area around Gävle in Sweden) are presented along with comparisons with results obtained by other methods. Throughout this thesis the emphasis has been on the $^{137}$Cs contamination in Sweden. Cesium is a relative long-lived anthropogenic radionuclide still present in measurable quantities in the nature originating mainly from the Chernobyl nuclear power plant accident and the nuclear weapon tests. The developed methods are however applicable to virtually any gamma-ray emitting nuclide and are not only relevant in case of an emergency, but also for environmental monitoring of radionuclides.
3. Efficient in situ method to estimate the average activity concentration

This chapter is based upon Paper I.

In the process of a radioactive decay a number of gamma-rays (photons) may be emitted. The probabilities for emission as well as the energies of the emitted photons are characteristics of the decaying radionuclide. In principle, therefore, the nuclide can be identified and the activity of the source determined by simply counting the number of photons emitted per unit time and measuring their energies.

In practice this is accomplished by using a gamma-ray detector. The energy distribution (spectrum) due to a $^{137}\text{Cs}$ source recorded with a high purity germanium (HPGe) detector is shown in Figure 3.1 as an example. The general features shown, however, are valid for a wide range of gamma-ray energies. In the decay of a $^{137}\text{Cs}$ nucleus a mono-energetic gamma-ray (662 keV) in addition to some x-rays (discrete energies around 32 keV) are emitted with certain probabilities. These structures (peaks) are the most dominant features in the spectrum, but in between there is a continuum. This feature and also the broadening of the peaks are mainly due to the properties of the detector system. Hence, knowledge about the detector response to a gamma-ray flux, is necessary in order to determine the activity of an unknown source.

The gamma-ray interacts with the detector mainly through photoelectric effect, Compton scattering and pair production and transfers some or all of its energy to the detector material where it can be measured\(^1\). The interaction probabilities depends on the gamma-ray energy and the detector material and as well on the geometry. The full energy peak (photo peak) corresponds to an event where the gamma-ray has deposited all its energy in the detector (by one or several interactions). The continuum, on the other hand, corresponds to events where the gamma-ray only deposits a part of its energy (in one or several interactions) and then leaves the detector\(^2\).

Two important properties in gamma-spectroscopy are the detector resolution and efficiency. The resolution is often quoted as the full width at half maximum (FWHM) of the photo-peak and is important in order to resolve

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\(^1\)Due to the properties of the detector material the energy deposited is not always correctly determined by standard procedures, cf. Chapter 5

\(^2\)For a more detailed discussion see, for example, the book “Radiation Detection and Measurement” by G. F. Knoll [2].
3.1 In situ gamma-ray spectrometry

In situ gamma-ray spectrometry provides a rapid, reliable and relatively simple method to measure the photon flux above a contaminated area. However, the method does not directly give an estimate of the total radionuclide concentration. The reason is that the photon flux at the location of a detector positioned above ground (Figure 3.2) depends both on the amount of radioactive nuclides in the soil and the burial depth. In order to estimate the concentration the effects of burial depth have to be taken into account. There exist several ways to accomplish this (cf. Paper I). The method described here is based upon the so called Peak-to-Valley method [14]. The probability for photons to be scattered or absorbed on the way to the detector increases with increasing burial depth therefore measuring the relative intensity of the photons retaining their full energy (unscattered) to that of photons of lower energy (scattered) it is, in principle, possible to determine both the concentration and burial depth. The method is applicable to virtually all gamma emitting radionuclides.

Figure 3.1: Energy distribution due to a $^{137}$Cs source recorded with a HPGe detector. The laboratory background has been subtracted. The energy distribution (above approximately 40 keV) is an example of the detector response to a mono-energetic photon flux.

closely lying peaks. The efficiency is basically the probability for a photon entering the detector to be detected. There exist several specific efficiencies, for example, the peak efficiency, i.e. the probability for a photon entering the detector to be recorded in the photo-peak, and the total efficiency which gives the probability that the photon interacts at all in the crystal.
however, in this project it has been applied to $^{137}$Cs, a long lived (half life 30 years) anthropogenic radionuclide present in the environment originating mainly from the Chernobyl power plant accident in 1986 and from the nuclear weapon tests in the 1950’s and 1960’s [9].

### 3.2 Description of the method

The basic idea of the method is to deduce the radionuclide concentration by comparing a modeled energy distribution to the distribution measured in situ.

The idealized measurement model is shown in Figure 3.2. A detector is positioned at a height of 1 m above an infinite horizontal soil surface. Further, the concentration of the radionuclide is assumed to depend only on the depth.

In the model the soil column is subdivided into a relevant number of layers. The activity concentration within each layer is approximated by a thin planar source. The photon transport from each individual layer up to the position of the detector is evaluated using Monte Carlo simulations. Hence a library containing the gamma-ray fluxes at the position of the detector has been created.

The consecutive energy deposition in the detector, is not implemented into the simulations, but is instead deduced from a few short and simple measurements using point sources. Thus making the library of spectra applicable to any detector available at the time of the in situ measurement. It is assumed that the detector response is independent of the angle of incidence. This, however, is not strictly the case (cf. section 3.4).

In practice only a limited number of parameters describing the depth distribution may indeed be treated as independent, and therefore a functional form...
3.2 Description of the method

Figure 3.3: Activity depth profiles determined from soil samples extracted in Utnora 2003. Sampling and measurement were performed by the Swedish Defence Research Agency (FOI). The author was present during the soil sampling in the field. The soil cores were divided into two pieces during the transfer from the sampling equipment to cylinders used for the transport. In the laboratory the cylinders were measured separately. The arrows indicate the region where the distributions from the two samples were rejoined. The the statistical uncertainties from the gamma-spectrometry measurements are shown.

of the distribution, \( v(z) \), needs to be postulated. Shortly after a fallout the vertical distribution can be well approximated by an exponential function [8, 15]:

\[
v(z) = a_1 e^{-a_2 z}
\]  

(3.1)

However, studies of \(^{137}\text{Cs}\) concentration in soil samples extracted many years after the fallout [4, 6, 8], have shown that a simple exponential is not any longer representative for the vertical profile. The concentration still approaches zero with increasing depth, but an asymmetric maximum if often present below the soil surface (cf. Figure 3.3). An example of a function, fulfilling these requirements, is the Lorentz function [8]:

\[
v(z) = \frac{b_1}{(z-b_2)^2 + b_3^2}
\]  

(3.2)

In the model the energy distribution corresponding to the distribution measured in situ, is built up by the sum of the individual contributions from each layer weighted with the measured detector response and the activity concentration. The parameters of the distribution function, \( v(z) \) are determined by fitting the model spectrum to the spectrum measured in situ (after an estimated background has been subtracted, cf. Paper I). In this comparison the
Figure 3.4: Gamma-ray spectrum recorded in Utnora (Sweden). The $^{137}$Cs peak is shown along with the valley region used for the estimation of the activity concentration. The energy interval between 614 and 662 keV corresponds to scattering angles between approximately 0 and 20 degrees (assuming a single Compton scattering of a 662 keV photon). In the inset the full energy distribution is shown. The peaks above 662 keV are mainly due to the decay of natural occurring radionuclides.

full energy peak (containing the unscatterad photons) and part of the valley region (containing the scattered photons) are utilized (cf. Figure 3.4).

The total activity can then directly be determined by integrating the activity distribution, $v(z)$. Other properties of the contamination, as the average burial depth, can also easily be deduced. In Paper I the theory behind the method is described in more detail.

3.3 Results

The method has been applied at six different locations in Sweden$^3$. Utilizing the full energy peak and the ratio between the peak and the valley region two independent parameters could be determined, the total activity and the average burial depth. Hence, the activity distributions functions, $v(z)$, had to be modified in order to support only two independent parameters (cf. Paper I).

A comparison regarding the total activity of $^{137}$Cs determined from gamma-ray spectra recorded in situ to results from soil sampling shows good agreement (cf. Figure 3.5). The vertical bars represent the uncertainty in the deter-

$^3$The data from the in situ measurements (spectra and results from analysis of soil samples) were made available by the Swedish Defence Research Agency (FOI). The author was present during the in situ measurements at Utnora 2003
3.4 Angular dependence of the detector efficiency

The approximation in which the detector response is assumed to be independent of the angle of incidence is a relatively poor one. The photo peak efficiency (662 keV) for the detector used in the field measurement varied by 15 per cent and the number of counts in the valley by almost twice as much when changing from vertical to horizontal incidence.

Nir-El and Sima [16] have reported similar values for the peak efficiency and have investigated the possibility to use Monte Carlo simulations to estimate the angular dependence of the detector efficiency. The drawback of such

Figure 3.5: Total activity of $^{137}$Cs in soil samples from six sites in central Sweden compared to the result obtained from in situ spectra. Soil samples were extracted 4 m north, 4 m south, 4 m east, 4 m west, 2 m south-east and 2 m north-west of the detector. The soil samples were approximately 100 mm deep except for site 1 where the soil samples were approximate 200 mm. The vertical bars represent the uncertainty in the measured activity. The dashed bands represent the results and the uncertainty using the method described.

mined activity for each sample. The horizontal bands represent the average activity determined from the spectra measured in situ. (The uncertainty stems partly from the uncertainty in soil density used in order to calculate the activity corresponding to the depth of soil samples, cf. Paper I). The six sites have different surface and soil properties and average activities ranging form 15 to 1000 kBq/m$^2$. The results for the burial depth are more sensitive to assumptions regarding soil composition, but are still in general agreement with the (rather imprecise) results from soil sampling (cf. Paper I).
an approach is the requirement of detailed description of the detector used in the measurements [17].

Instead, as a first order correction, to the developed method, the detector response has been measured at an intermediate angle, approximately corresponding to the average angle of incidence. This correction is fast and simple to implement and does not require any a priori information about the detector. Measuring the detector response at zero degrees (vertical incidence), yields a 20–40 per cent larger value for the total activity due to the underestimation of the efficiency.

3.5 Limitation of the infinite flat surface model

The idealized measurement topography is shown in Figure 3.2. Soil of homogeneous composition with an infinite, horizontal surface and above it air. The real measurement situation is most often quite different (cf. Figure 3.6). The contributing photon flux, in real measurements, comes typically from within 10-30 m radius from the detector [18]. Hence, the activity and depth distribution estimated using the in situ methods with a detector positioned 1 m above ground will be an average over an area of typically 500-1000 m².

The correlation between the average surface activity determined from in situ measurements and the position of the detector has been studied by Boson et al. [19]. Measurements and soil sampling was performed in a cross pattern with 2 m between the positions. The variation in the average surface activity determined from in situ measurements was ±8% as compared to the much larger soil sample variations of ±60%.

However, vegetation, surface roughness and varying soil composition may have a large impact on the photon flux measured above a contaminated area [3, 4, 7, 20], whereas the flux is relatively insensitive to variation of the air density [3]. In the following sections the impact of these parameters on the result will be discussed briefly.

3.5.1 Soil composition

The soil composition may vary not only from site to site, but also within the same site. For energies of interest (i.e. above 100 keV), however, the attenuation of the photon flux varies relatively slowly with the atomic number of common soil elements [3]. Therefore the exact soil composition is not crucial for the estimate of the total activity from the measured photon flux above a contaminated area.

This is supported by an investigation by Jacob and Thummerer [21]. The authors have simulated energy distributions using typical soil and rock com-
3.5 Limitation of the infinite flat surface model

Figure 3.6: In situ measurement of the gamma-ray flux in an alder marsh (Utnora, 2003). A HPGe detector is position 1 m above the ground on a tripod (shown in the middle of the photo).

positions and found the differences to be negligible (given that the source depth is expressed in mass depth$^4$).

The ratio of the flux of unscattered photons to that of scattered photons reaching the detector (peak-to-valley ratio) depends on the probability for Compton scattering which depends upon the electron density in the attenuating material. As discussed in Paper I, this has the consequence that, to first order, a change in soil composition, density or water content does not affect the result for the total activity, but only the estimate of the geometric burial depth.

3.5.2 Surface roughness

Small scale surface irregularities can be described as a drastic lowering of the soil density near the surface and possibly an associated change of composition. Referring to the discussion above and in Paper I regarding the dependence on composition and density, changes in those parameters will not, to first order, have an impact on the determination of the total activity. This is also supported by studies [22] showing that the angular distribution of the primary flux from a contamination on an irregular surface is similar to that produced by a buried contamination.

$^4$Mass depth = geometric depth × density
3.5.3 Vegetation

The screening effect from vegetation is most important prior to the migration of the radionuclides into the soil [20], due to the lower density of vegetation as compared to the density of soil. However, the density above ground can increase over 30 times relative the density of air if the vegetation extends up to a height of 0.5 m. Hence, reducing the primary flux at the detector by 20-50 per cent [20], in addition, to a change in the energy and angular distribution of the scattered photons. These effects are, however, partly canceled during the first time after a fallout, because radioactive particles are deposited on the vegetation thus bringing the activity closer to the detector. A similar, but smaller, effect can be found years after the fallout due to the uptake of radionuclides (especially $^{137}$Cs [11]) from the soil. Studies by Jacob et al. [7] shortly after the fallout from the Chernobyl accident have shown that 5-20 per cent of the deposited cesium was retained on the grass even though the deposit occurred in connection to heavy rain.

It is assumed that vegetation (at least close to the ground) may be treated as a kind of surface roughness, and therefore does not influence on the determination of the total activity, but only the depth profile as discussed previously.

Boson et al. [19] have studied the effects on the in situ measurements due to the presence of trees. The estimated attenuation by trees, determined using simulations of the photon transport, was about 5%. Further, the total deposited activity per unit area was found to be underestimated by about 20% in the scenario where 50% of the deposition was assumed to be located in the trees as compared to a scenario without trees and all activity located in the soil.

3.5.4 Large scale structures

Large scale surface roughness, like hills and large rocks can significantly reduce the gamma-ray flux. Investigations using simulations of the photon transport have shown that the gamma-ray flux can change with ±0-20% due to ground curvature [19].

In the in situ measurement it is, therefore, favorable, if possible, to select a site that is relatively flat and free from large obstacles in the immediate neighborhood of the detector [3, 19, 20]. Approximately 85 per cent of the photon flux (from $^{137}$Cs decay) reaching the detector, originates within a radius of 10 m [18]. Therefore the site requirement can normally be fulfilled by avoiding large structures within this region.

3.6 Conclusions regarding the applicability

Summarizing the discussions above it is believed that the library of simulated spectra is applicable on in situ measurements with large variation in soil composition as long as the total activity is of primary interest. In addition the
method produces an average effective depth profile, but in order to precisely translate this effective depth to geometric depth detailed information regarding soil composition, water content and density is needed. Secondly the surface roughness and vegetation make the definition of a surface, zero point for the geometric depth measurement, ambiguous. It is therefore believed that in cases where detailed information about the depth profile of the contamination is needed other methods are to be preferred.

The implemented peak-to-valley method allows for a simple and reliable determination of the buried contamination from spectra measured in situ. The result corresponds to a weighted average over a larger area. The method has been applied to six sites in Sweden determining, in a relatively short time, \(^{137}\text{Cs}\) contamination corresponding to an average activity as low as 15 kBq/m\(^2\).
4. In situ method to determine depth distribution of buried activity

The measurements using high-resolution germanium detectors, described in the previous chapter, yields reliable estimates for the average activity concentration of various radionuclids even in cases where the activity is buried. However, in order to make predictions concerning the uptake of activity into the biosphere more detailed information concerning the depth distribution is required [11].

Such information is commonly obtained by by taking soil samples and then determine the activity distribution by gamma-ray spectrometry in the laboratory, a both labor intensive and time consuming procedure. Although the laboratory measurements can be conducted with essentially arbitrary precision, the procedure entails large systematic uncertainties due to soil in-homogenties as well as deformation of samples upon extraction, and cross contamination within the sample. In addition, a large number of samples are required due to large variations of the activity even over small distances [13, 23, Paper I and IV].

The aim of this project has been to investigate if it is possible to determine the depth distribution of a radionuclide contamination, in situ, using small detectors inserted into the ground.

For such a method to be relevant, the duration of the individual measurement, should not widely exceed the time needed for extracting a soil sample. A slightly longer duration of the field measurement can, however, be considered as acceptable, as the method does not require any further measurements in the laboratory.

The mean free path of gamma-rays (662 keV) in soil is typically in the order of 0.1 m as compared to 100 m in air (estimated using the soil and air composition in Paper I). Therefore the scattered gamma-ray flux measured by a detector inserted into the ground will originate mainly (90%) from within a radial distance of slightly less than 0.6 m (estimated by assuming a point detector at the surface and a homogeneous activity distribution throughout the ground). This corresponds to a surface area of approximately 1 m$^2$ and should be compared to the typically soil sample cross section area of $\leq 0.01 m^2$ [1,4, 6,8,24,25].

Studies of the vertical distribution of $^{137}$Cs after the Chernobyl accident have shown that the major part of the contamination can be found within the first 0.2 m [4, 6, 8, 12, 26]. Therefore the measurements should cover approx-
imately this region of the soil. However, in a fresh fallout the contamination will be located closer to the soil surface.

4.1 Detector properties

In order to determine the depth distribution of the activity the gamma-ray flux must be measured at more than one depth below the soil surface. Hence, both the interaction position and the energy of the incident gamma-ray are to be determined. This can be accomplished by either using a larger position-sensitive detector or an array of smaller detectors.

The latter device has the disadvantage that distance between the detectors can not be made arbitrary small due to mounting and the requirement of reading out the signal from each unit individually. However, a larger detector is more likely to experience problems due to high count rates (i.e. large dead-time and pulse-height distortions), especially during measurements shortly after a fallout.

Multiple interactions within the detectors may lead to severe distortion of the information obtained about the incident gamma-ray, especially in case of the position-sensitive detector. Additional distortions may arise from scattering between the individual units in an array of detectors. It is, therefore, foreseen that these effects must be further investigated, especially if the valley region of the spectrum is to be used.

The detectors suitable for the project should be small but still yield a reasonably high efficiency, therefore a large stopping power is required. The detectors should also be relatively insensitive to vibrations and mechanical shock during the insertion into the ground. As the ambient temperature can vary drastically during the field measurements, the detector properties should be relatively insensitive to temperature changes (at least within the time frame of a single measurement).

In case of a radioactive fallout, several radionuclides will most probably be present. Therefore not only the time required for the field measurement (detector efficiency) is of great importance but also the possibility to distinguish between relevant nuclides (detector resolution).

4.2 Detector material

The HPGe detector with its excellent energy resolution is well suited for radionuclide identification, but the relative low efficiency for the crystal volumes suitable for the current project together with the requirement of cooling to almost -200°C renders the detector material less suitable.

Sodium iodide (NaI) is a scintillator which has a higher efficiency but relatively poor energy resolution, and is therefore best suited for a situation where
In situ method to determine depth distribution of buried activity

<table>
<thead>
<tr>
<th>Detector type</th>
<th>HPGe</th>
<th>CdTe</th>
<th>NaI</th>
<th>LaBr₃</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density (g/cm³)</td>
<td>5.32</td>
<td>6.06</td>
<td>3.67</td>
<td>5.29</td>
</tr>
<tr>
<td>Effective atomic number</td>
<td>32</td>
<td>50</td>
<td>50</td>
<td>48</td>
</tr>
<tr>
<td>Resolution at 662 keV</td>
<td>&lt;0.5%</td>
<td>~3%</td>
<td>~7%</td>
<td>~3%</td>
</tr>
<tr>
<td>Operating temperature</td>
<td>~178°C</td>
<td>~20°C</td>
<td>~20°C</td>
<td>~20°C</td>
</tr>
</tbody>
</table>

Table 4.1: Detector material properties. Data from Ref. [2, 27]. The effective atomic number, in this thesis, corresponds to the atomic number of a pure element which have approximately the same linear attenuation coefficient for 662 keV gamma-rays as the compound. The effective atomic number has been estimated using Penelope [28].

the gamma flux is totally dominated by a single photon energy (or by widely separated energies). The NaI detectors are relatively cheap and exists in a large variety of geometries.

Cadmium telluride (CdTe) is a semiconducting detector with a large stopping power, but are hampered by poor charge collection, which results in a low resolution for large volume detectors. This can partly be overcome by special designs, for example hemispherical electrode configuration and different types of pulse-processing procedures, and have resulted in good energy resolution.

Lanthanum bromide (LaBr₃) is a relatively new scintillator which combines the high efficiency of the NaI detector and the good energy resolution of the CdTe detector. The properties of the detector materials are summarized in Table 4.1.

### 4.3 Energy distribution after a radioactive fallout

The analysis of the energy distribution measured in field after a radioactive fallout can be quite complex as compared to spectra recorded in laboratory using a single radioactive source. This is due to the presence of large variety of radioactive nuclides often released in, for example, a reactor accident. After a few months the gamma-ray spectra will be dominated by $^{134}$Cs and $^{137}$Cs, but shortly (several days) after a reactor accident $^{131}$I dominates [9, 11]. In Table 4.2 the properties of major contributing isotopes (above 300 keV) are summarized in combination with the activity released in the Chernobyl reactor accident. However, the amount of activity actually deposited depends upon several parameters, for example weather conditions, distance to the source and the altitude of the radioactive cloud [11, 29].
The energy distribution after a radioactive fallout (Figure 4.1) has been estimated using a simplified detector response, defined by single Gaussian. Hence neglecting the Compton continuum and other structures. Further the energy dependence of the resolution and efficiency have been ignored. A detector resolution of 3% and 7% (FWHM) corresponding to the typical resolution at 662 keV for CdTe/LaBr$_3$ and NaI detectors, respectively, were implemented. The ratio between the different isotopes dominating the gamma-ray flux due to the fallout are estimated based upon data from the Chernobyl reactor accident (Table 4.2).

4.4 Conclusions

Based on the considerations above it was decided to investigate in greater detail the performance of an array of small detectors made of either CdTe or LaBr$_3$. 
In situ method to determine depth distribution of buried activity

<table>
<thead>
<tr>
<th>Radioactive nuclide</th>
<th>Energy (keV)</th>
<th>Gamma intensity</th>
<th>Half life</th>
<th>Activity (PBq)</th>
<th>Parent nuclide</th>
</tr>
</thead>
<tbody>
<tr>
<td>99Mo</td>
<td>739.5</td>
<td>0.12</td>
<td>2.7 days</td>
<td>&gt;168</td>
<td></td>
</tr>
<tr>
<td>132Te</td>
<td>-</td>
<td>-</td>
<td>3.2 days</td>
<td>~1150</td>
<td></td>
</tr>
<tr>
<td>132I</td>
<td>505.8</td>
<td>0.05</td>
<td>2.3 h</td>
<td>~0</td>
<td>132Te</td>
</tr>
<tr>
<td></td>
<td>522.7</td>
<td>0.16</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>630.2</td>
<td>0.13</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>667.7</td>
<td>0.99</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>669.8</td>
<td>0.05</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>772.6</td>
<td>0.76</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>812.0</td>
<td>0.06</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>131I</td>
<td>364.5</td>
<td>0.82</td>
<td>8.0 days</td>
<td>~1760</td>
<td></td>
</tr>
<tr>
<td></td>
<td>637.0</td>
<td>0.07</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>140Ba</td>
<td>537.3</td>
<td>0.24</td>
<td>12.8 days</td>
<td>~240</td>
<td></td>
</tr>
<tr>
<td>140La</td>
<td>328.8</td>
<td>0.20</td>
<td>1.68 days</td>
<td>~0</td>
<td>140Ba</td>
</tr>
<tr>
<td></td>
<td>487.0</td>
<td>0.46</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>815.8</td>
<td>0.23</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>867.8</td>
<td>0.06</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>103Ru</td>
<td>497.1</td>
<td>0.91</td>
<td>39.3 days</td>
<td>&gt;73</td>
<td></td>
</tr>
<tr>
<td></td>
<td>557.0</td>
<td>0.87</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>610.3</td>
<td>0.06</td>
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<tr>
<td>95Zr</td>
<td>724.2</td>
<td>0.44</td>
<td>64 days</td>
<td>~196</td>
<td></td>
</tr>
<tr>
<td></td>
<td>756.7</td>
<td>0.54</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>95Nb</td>
<td>765.8</td>
<td>1.00</td>
<td>35 days</td>
<td>~0</td>
<td>95Zr</td>
</tr>
<tr>
<td>134Cs</td>
<td>563.2</td>
<td>0.08</td>
<td>2.06 years</td>
<td>~54</td>
<td></td>
</tr>
<tr>
<td></td>
<td>569.3</td>
<td>0.15</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>604.7</td>
<td>0.98</td>
<td></td>
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<td></td>
</tr>
<tr>
<td></td>
<td>795.9</td>
<td>0.86</td>
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<tr>
<td></td>
<td>802.0</td>
<td>0.09</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>137Cs</td>
<td>661.7</td>
<td>0.85</td>
<td>30.1 years</td>
<td>~85</td>
<td></td>
</tr>
</tbody>
</table>

Table 4.2: Properties of significant radioactive nuclides released in the Chernobyl reactor accident in 1986. Data from Ref. [11, 29]. The table contains only radionuclides emitting gamma-ray with energies in the interval 300 keV to 900 keV with intensities above 5%. (132Te is included because it decays to 132I.)
Figure 4.1: Estimated appearance (cf. text) of the energy distribution after a radioactive fallout from a major reactor accident measured with 3% (full line) and 7% (dashed line) relative detector resolution. The activity of the different radionuclides are based upon data from the Chernobyl power plant accident, cf. text.
5. Investigation into the properties of CdTe-detectors

This chapter is based on Papers II and III.

Cadmium telluride (CdTe) is an interesting candidate for the detector material to be used in the field measurements. CdTe detectors are semiconductors and can be operated at room temperature. The density of cadmium telluride is 6.06 g/cm$^3$ [2] and therefore the crystals can be made relatively small but still yield a reasonable efficiency. Typically CdTe crystal volumes are 50-500 mm$^3$.

The major drawback of CdTe detectors (especially in comparison with HPGe detectors) is the small peak-to-valley ratio, the strongly asymmetric peak shapes and the low photo-peak efficiency. The aim was, therefore, to investigate if it would be possible to improve these properties of the spectrum.

5.1 Charge-carrier transport in CdTe

Gamma radiation with an energy below the threshold for pair production entering a CdTe detector mainly interacts with the electrons in the crystal through photoelectric-effect or Compton scattering resulting in a fast free electron that loses its energy by excitation of electron-hole pairs in proportion to the deposited energy. If an external electric field is applied over the crystal the liberated charges will drift towards the electrodes. The currents persist until the charges reach the electrodes and recombine. The pulses from a CdTe detector therefore comprise the sum of two components, originating form the drift of electrons and holes respectively. The proportion of the pulse generated from each one of these components depends on the distance from the point of interaction in the crystal to the respective electrode. The drift velocity of holes is approximately a tenth of that of electrons, but the trapping time (i.e. the average free time of a charge carrier) is of the same order of magnitude [30]. Hence, the hole contribution to the pulse-height is more severely affected by trapping effects, resulting in a final pulse-height depending on where in the crystal the interaction takes place (Figure. 5.1). This results in a distortion of the pulse-height spectrum resulting in degraded energy resolution, lower peak efficiency and a lower peak-to-valley ratio.
Figure 5.1: Pulse shapes at the preamplifier output from a small planar CdTe detector. All pulses correspond approximately to the same deposited energy. The pulses are the sum of two contributions; one fast component due to electrons and one slow due to holes. The variation in pulse height is (mainly) due to trapping of holes.

For a more detailed discussion about the working principles of a semiconductor and the pulse processing see Ref. [2, 31].

There are several possible methods to improve the resolution and the peak-to-valley ratio, for example by rise time discrimination, i.e. rejecting pulses with long rise times (large hole contributions) [32–34]. This method, however, does not improve the peak efficiency, but merely removes the events falsely recorded at a lower energies due to the pulse-height deficit. Another possibility is to utilize the pulse shapes, instead of only the pulse-height, in order to determine the deposited energy [31, 35]. These latter methods have the drawback that huge amounts of data must be processed and additional pulse processing units and more advanced data acquisitions systems are needed. In the following, two other solutions will be discussed. Firstly, a method that utilizes the correlation between rise time and pulse-height deficit and secondly a hemispheric crystal geometry.

5.2 Small volume CdTe detector

A simple method to improve the peak-to-valley ratio is described in Paper II. Briefly the method is implemented as follows: the preamplified pulse from a CdTe detector (Figure 5.1) is feed into two standard spectroscopy amplifiers
Investigation into the properties of CdTe-detectors

Figure 5.2: Scatter plot of pulse heights from a small planar CdTe detector for $^{137}$Cs using amplifiers with different shaping times (0.5 µs and 3.0 µs respectively). The time constants were selected based upon the sampled pulse-shapes (Figure 5.1).

with different shaping times\(^1\). The resulting scatter plot is shown in Figure 5.2. In this representation the resolution is much better than in the one dimensional case (i.e. the pulse-height distribution recovered by projecting the scatterplot upon the y-axis). Events with a large contribution from electrons (short rise time) will be located on (or close to) the diagonal. On the other hand, events with a large contribution from holes (long rise time) will be located below the diagonal. In Figure 5.2 the full energy events (all gamma-ray energy deposited within the crystal) are clearly separated from those belonging to the Compton continuum (partly deposition of the gamma-ray energy within the crystal). The ratio between the pulse-heights after the two amplifiers is in principle a measure of the interaction depth, and this can be used to calculate a correction to the pulse-height [36].

There exist two possible paths to continue, either working in the 2-dimensional representation or use the 2-dimensional representation to establish an improved 1-dimensional energy distribution for which conventional methods of analysis can be directly applied. The method

\(^1\)The shaping time is, in principle, the time interval over which the amplifier will process the preamplified pulse. A short shaping time ($\lesssim 0.5$ µs, cf. Figure 5.1) will result in a pulse height after the amplifier that almost entirely depend on the electron contribution to the preamplified pulse. A long shaping time ($\gtrsim 0.5$ µs, cf. Figure 5.1), on the other hand, will result in a pulse height after the amplifier that depend on both the electron and hole contribution to the preamplified pulse.
5.3 Large volume CdTe detectors

Implemented in Paper II utilizes the latter approach. The algorithm has been applied to a small planar CdTe detector (5x5x2 mm$^3$) resulting in an increase in the peak-to-valley ratio (peak efficiency) by more than a factor of six (four) without rejecting any events (Figure 5.3). The energy resolution (FWHM) of the full energy peak for gamma-ray energies of 662 keV is approximately 2%, however, a remaining drawback is a certain broadening of the peak at the base. This effect can be reduced, if a loss of efficiency is acceptable, by rejecting events occurring at large interaction depth (large hole contribution).

5.3 Large volume CdTe detectors

Inspired by the radical improvement in case of the small CdTe crystal the investigation was continued with crystals of larger volume (500 mm$^3$) in order to increase the efficiency and thus reduce the required measuring time in field.

5.3.1 Description of the detectors

Typical pulse-height distributions from two large volume CdTe detectors, one planar and one hemispheric both manufactured by Eurorad [37], are shown in Figure 5.4. Notice how the pulse-height distribution from the large planar detector is similar to that of the small planar detector (Figure 5.3), but the distribution from the hemispheric detector is quite different.

The major differences in the distributions can be understood by studying the crystal geometries and the electric field configurations for the two detectors. The detector referred to as hemispheric is actually a quasi-hemispheric detector [37]. Both the quasi-hemispheric and the planar detector have rectangular dimensions of 10x10x5 mm$^3$. On the hemispheric detector all surfaces except one are held at negative potential. The remaining surface has a positive electrode at its center. On the planar detector one surface acts as a positive electrode and one as a negative electrode (cf. Paper III). The planar detector has (ideally) a uniform electric field and both charge carries contribute to the pulse-height. The pulse-height distribution is distorted mainly due to incomplete hole collection (Section 5.1). The hemispheric detector on the other hand has an electric field that is (ideally) radial and therefore much stronger near the positive electrode. This field configuration accelerates the collection of electrons and slows down the velocity of holes. As a result, the device works (in principle) in a single charge collection mode.

5.3.2 Experimental motivation

As a first approach the rise time dependency has, as before, been studied using two amplifiers with different shaping times. However, the scatter plots
Investigation into the properties of CdTe-detectors

Figure 5.3: The corrected (uncorrected) energy distribution for a $^{137}$Cs source recorded using a small planar CdTe detector is shown in blue (red).

Figure 5.4: Pulse-height distribution (uncorrected) for $^{137}$Cs for the large planar (blue) and the quasi-hemispheric (red) CdTe detector (at room temperature) both of 500 mm$^3$ volume.
5.3 Large volume CdTe detectors

Figure 5.5: Schematic drawing of experimental setup. A $^{137}$Cs source is positioned behind a lead collimator and the photon flux is directed towards the CdTe detector, the scattered photons are detected in the HPGe detector. Due to a significant contribution in the energy region of interest from photons back-scattered in the concrete floor the source was shielded below the collimator using 20 mm lead plates (not shown in the figure).

of the pulse-height distributions (cf. Section 5.3.4) did not reveal a full energy ridge well separated from the Compton continuum, as in the case for the small planar detector (Figure 5.2). Therefore it was decided to continue the investigation by recording the pulse-height distribution for samples of mono-energetic deposition in the CdTe crystal. In addition the pulse-shapes for both detectors were recorded with the aim to achieve a better understanding of the pulse-height distributions.

5.3.3 Experimental setup

Mono-energetic samples were created using Compton scattering of mono-energetic photons and measuring the energy of the scattered photon (Figure 5.5). The CdTe detector was irradiated by a collimated photon flux of 662 keV and a second detector (HPGe) was positioned at a given angle and carefully shielded from the radioactive source.

Events that correspond to a photon scattering in the CdTe detector at an angle such that it can be detected in the HPGe detector can be selected by requiring coincidence between the two detector signals. Due to the extended size of both crystal and the close distance between the detectors a range of scattering angles (and therefore a range of deposited energies) are covered in one position. Hence, by selecting a certain energy in the HPGe detector (Figure 5.6), correspondingly, a certain deposited energy in the CdTe crystal can be selected (Figure 5.7).
Investigation into the properties of CdTe-detectors

Figure 5.6: Scatter plot for coincidence events of the energy deposited in the HPGe and the planar CdTe detector. The arrow indicates the position of the energy interval selected as a requirement for the spectrum in Figure 5.7.

Figure 5.7: Pulse-height distribution for planar the CdTe detector in coincidence with events depositing between 211.3 and 213.0 keV in the HPGe detector. The low-energy tail is due to incomplete charge collection.
The geometric limitations due to the extended size of the detectors and the requirement to shield the HPGe detector from the source results in a maximum scattering angle of approximately 130 degrees, corresponding to a deposited energy in the CdTe detector of approximately 450 keV.

Studies [38–40] (see Paper III for an overview) have shown that the noise can be considerably reduced and therefore the energy resolution improved by cooling the CdTe detector. Therefore a Peltier-element was mounted in thermal contact with the housing of the detector, allowing for measurements at sub-zero temperatures.

5.3.4 Results

For both the planar and the hemispheric detector the energy deposition was studied in coincidence and single. In Paper III the measurements are discussed in greater detail, in this section, however, the results are summarized with focus on the properties important for the field measurements.

**Large planar detector**

In the pulse height distribution recorded using the planar detector (Figure 5.8) the majority of the events are located close to the diagonal as opposed to the case for the small planar detector (Figure 5.2) where the events depositing the same energy are (mainly) located on ridges extending well below the diagonal. Studies of the pulse shapes (Paper III) have revealed that the hole contribution to the total pulse height is small, probably due to trapping of charge carriers. Due to the small difference in rise time, the pulse shapes after the main amplifiers will have almost the same amplitude independent of the shaping times, which causes the events to be located close to the diagonal in the scatter plot.

Reversing the polarity of the high voltage has only marginal effect. Increasing the applied high voltage, hence decreasing the collection time for holes (and electrons) increases the separation of the events from the diagonal but not to such an extent that the simple pulse-height correction algorithm (section 5.2) can be successfully applied. Similar results were obtained also after cooling the detector to sub-zero temperatures.

**Large hemispheric detector**

In the scatter plot of pulse-height spectrum for the hemispheric detector (Figure 5.9) the separation of events from the diagonal is also much less pronounced as compared to the small planar detector. However, there exists two distinct regions (tails) extending below the diagonal. For the first tail (closest to the diagonal) pulse-shape studies and geometric consideration reveal a phenomenon similar to what was seen for the events belonging to the full energy ridge in case of the small planar detector. The second, more pronounced, tail, extending below both the full energy peak and the
first tail, is missing in the results from the coincidence measurements (cf. Paper III) which indicates that these events are not produced in a single Compton scattering interaction (which dominates the events recorded in the coincidence measurements). This assumption is further supported by studies of the pulse-shapes (cf. Paper III). It is believed that the events are the result of multiple interactions within the crystal.

Considering the scatter plot in Figure 5.9 it is obvious that the pulse-height correction algorithm developed for the small planar CdTe detector (Paper II) will not be applicable. An unambiguous relationship between the rise time (deviation from the diagonal) and the pulse height deficit cannot be defined because the tails corresponding to different energy depositions overlap in the scatter plot. In principle it would be possible to use more detailed information concerning the pulse shapes in order to distinguish between different type of events and correct for the amplitude deficit. As an alternative, awaiting further investigation into the explicit exploitation of pulse-shapes, the scatter plot can be used for rise time discrimination of the recorded pulse-height distribution. This can be achieved by including only data from the diagonal region and would improve the peak-to-valley ratio and the resolution, but would also lead to a loss of efficiency.

The final result for the quasi-hemispheric detector at room temperature (cooled to -7.5°C and rise-time discriminated) is a resolution of 7% (4%) at 662 keV and a peak-to-valley ratio of 10 (92). Utilizing the rise-time discrimination the peak efficiency was lowered by 25%. Hence, the peak efficiency (fraction of the number of incoming photons to those registered in the full energy peak), rise-time discriminated and at subzero temperature, was $2.3 \pm 0.1\%$.

### 5.4 Conclusions

A simple method to correct for the incomplete charge collection has successfully been implemented for a small planar CdTe detector but was found not to be applicable for the large volume detectors. The conclusion is that the resolution and peak-to-valley ratio of the large planar detector cannot be substantially improved, and therefore the detector is not well suited for gamma-ray spectrometry. The pulse-height distribution from the hemispheric detector, however, could be significantly improved by cooling the detector. In order to fully exploit its properties rise time discrimination should be implemented. The conclusion is that the hemispheric detector is an interesting candidate for the detector array to be used for in situ gamma-ray spectrometry. Additional investigations into methods for correcting the pulse-height deficit utilizing sampled pulse-shapes should be undertaken in order to determine if the efficiency and resolution can be further improved.
Figure 5.8: Scatter plot of pulse-height spectrum of events in the large planar CdTe detector using two amplifiers with different shaping times. The arrow indicates a small deviation from the diagonal. (The energy calibration is approximate).

Figure 5.9: Scatter plot of pulse-height spectrum of events in the hemispheric CdTe detector using two amplifiers with different shaping times. The arrows indicate the two distinct tails.
6. Investigation into the properties of LaBr$_3$-detectors

Lanthanum bromide (LaBr$_3$) is a novel scintillator, discovered at Delft and Bern Universities in 2001 [27]. It has received large attention in the field of gamma- and x-ray detection. A light yield of 61,000 photons/MeV (162% as compared to NaI), a resolution better than 3% (FWHM) at 662 keV and a decay time of 26 ns (as compared to 240 ns for NaI) have been reported in the literature [27, 41–43]. In addition, the light output and the resolution are very stable in the temperature range of -25 to 50°C, ±1% and ±4% respectively [43]. These properties in combination with a density of 5.29 g/cm$^3$ [27] and room temperature operation render the detector a highly interesting candidate for in situ measurements of gamma-radiation. However, a drawback is the presence of intrinsic radioactivity (cf. Section 6.3).

A gamma-ray interacting within a scintillation crystal (LaBr$_3$ and NaI) results in an excitation of crystal molecules and during the deexcitation process visible light is emitted (the intensity is, ideally, proportional to the deposited energy). As opposed to semiconductor detectors (CdTe and HPGe) where the interaction within the crystal directly yields an electrical signal, ideally, proportional to the deposited energy (cf. Chapter 5), the emitted light has to be converted into a electrical signal. This can be achieved by using a light sensitive device, most commonly used are photomultiplier-tubes (PMT) but other devices can be used as well, for example avalanche-photo-diodes (APD) or silicon-drift-detectors (SDD). The two first possibilities to read out the light from the scintillator will be discussed in the next section, the possibility to use SDDs are discussed in Ref. [44, 45].

6.1 Readout devices

6.1.1 Photomultiplier tube (PMT)

The photomultiplier tube consists of a chamber, usually made out of glass, needed to preserve vacuum conditions inside, which is required for an efficient transport of low-energy electrons. The two major structures inside the chamber are the photocathode which converts the incoming light to an electron current via the photo-electric effect and the electron multiplier structure. The latter amplifies the weak current sufficiently for it to serve as a signal. The amplification occurs in several steps, first the liberated electrons at the pho-
tocathode are collected and accelerated towards the first dynode, upon hitting the dynode the electrons interact with the dynode material and a few electrons are liberated for each incoming electron which are then accelerated towards the second dynode. This process is repeated in several steps until the electron current is large enough for further analysis. The transport of electrons within the photomultiplier is very sensitive to external magnetic fields (even the earth’s magnetic field can affect the trajectories), and therefore the PMT is usually shielded using a mu-metal foil. For a more detailed discussion about the working principles of the photomultiplier see Ref. [2]

In this study a $\Omega 25 \times 50$ mm PMT (9111B) manufactured by Electron tubes has been used. The active area of the photocathode has a diameter of $\Omega 22$ mm and the PMT was connected to a base of height 60 mm, yielding a total length of 110 mm. The quantum efficiency of the photocathode is 25% at at the emission wavelength of LaBr$_3$ (380 nm) according to the manufacturer. Optical grease was used between the crystal and the window of the PMT. Since the scintillation detector system is extremely photosensitive it must be encapsulated to avoid exposure to ambient light. In the present study this has been solved by covering the PMT and crystal with a plastic cup attached snugly against the base.

6.1.2 Avalanche photo diode (APD)

The avalanche photo diode (APD) is a semiconductor used to convert the scintillation light into an electric signal. The working principle is as follows: a photon enters the APD and interacts somewhere within the active part of the device. In this interaction electron-hole pairs are created (in a similar manner as for the gamma-ray interaction within the CdTe-crystal, Chapter 5). The liberated charges are then accelerated by an external applied electric field. The field is so high that the charge carriers are accelerated enough between collisions to generate additional electron-hole pairs along the transport path. Hence, this creates an avalanche of charge-carriers and generates a signal large enough for further amplification in the preamplifier. However, the gain achieved is very sensitive to both temperature and applied voltage [2].

Although the APD is considerably smaller than a PM-tube, it has the drawback of several orders of magnitude lower amplification [2], possibly resulting in a worse energy resolution. This effect is, however, partly compensated by a quantum efficiency of 70% (at 380 nm according to the manufacturer), roughly three times larger than for a typical PMT [2, 46].

In this study a non-cooled large area blue silicon avalanche photo diode (SD 630-70-74-500) manufactured by Advanced Photonix Inc. was used. The APD had a outer diameter of 30 mm, with an active area 16 mm in diameter. The total height of the unit was 36 mm including contact. In order to achieve optical contact between the crystals and the APD a $\Omega 16$ mm and 2 mm thick plastic light guide was used, because laboratory tests have shown that a light
Investigation into the properties of LaBr$_3$-detectors

<table>
<thead>
<tr>
<th>Crystal</th>
<th>APD resolution</th>
<th>PMT resolution</th>
<th>Passport resolution</th>
</tr>
</thead>
<tbody>
<tr>
<td>K586</td>
<td>4.9%</td>
<td>4.6%</td>
<td>3.7%</td>
</tr>
<tr>
<td>L016</td>
<td>4.7%</td>
<td>4.4%</td>
<td>3.2%</td>
</tr>
</tbody>
</table>

Table 6.1: *Energy resolution (FWHM) at 662 keV for the two LaBr$_3$ crystals using APD and PMT for the read out. Passport resolution refers to the resolution measured by the manufacture prior to delivery.*

guide results in better resolution and light yield as compared to a 2 mm air gap otherwise present due to the mismatch in geometry of the crystal and the APD. Optical grease was used on both side of the light-guide. The APD and the LaBr$_3$ crystal was encapsulated within a plastic cup in order to avoid ambient light.

6.2 Results from laboratory tests

Two LaBr$_3$ crystals (BrilLanCe-380), with dimensions $\varnothing 17 \times 10$ mm$^3$, manufactured by Saint-Gobain Crystals have been tested in the laboratory. The APD was operated at 1700 V without experiencing any problems with the linearity in the spectrum. However, for the PMT readout the energy scale was highly non-linear when using the recommended high voltage of 1000 V. The linearity (up to approximately 700 keV) was restored when the voltage was reduced to 620 V. This effect has also been seen in other studies [47, 48]. The problem occurs because of the large light yield in combination with the very short decay time. Others studies have used PMTs where the dynode chain has been modified which have resulted in an improved linearity [47,48]. It is more favorably to use a higher voltage and fewer multiplier steps (than the opposite) because the main contribution to the resolution comes from the first (few) amplification steps [2].

A typical spectrum recorded using the LaBr$_3$ crystal with PMT read out is shown in Figure 6.1, the corresponding spectrum for readout using the APD is similar. In Table 6.1 the achieved resolution for the two crystals read out with PMT and APD, respectively, are presented. The peak-to-valley ratio is approximately 40 for both crystals. The peak efficiency (i.e. the fraction of all photons entering the detector, giving rise to an event in the full-energy peak) is $5.7 \pm 0.1\%$. 
6.3 Internal activity

The internal activity is due to the presence of the long lived radioactive isotope $^{138}$La (lifetime in order of $10^{11}$ years) and a contamination with the radioactive element $^{227}$Ac, which is the chemical homologue of lanthanum. The energy distribution due to internal activity recorded in a low-background-laboratory (see Appendix A) is shown in Figure 6.2. The intrinsic activity of LaBr$_3$ have been studied more thoroughly in Refs. [42, 49, 50].

In the studied crystals the total background counting rate is less than 1 count/s in the energy interval 0.3-3 MeV. However, the alpha contamination was found to be approximately three times higher in one of the crystals. In studies where the gamma flux is very low this background may be destructively interfering with the measurement. However, in the type of field measurements this project is dealing with the count rate due to the contamination in the environment will generate one or more orders of magnitude higher counting rates. If this is not the case the measuring times required in field will become too long, and the traditional soil sampling approach will have to be used. The largest destructive interference is foreseen in measurements of low activities of $^{40}$K or other gamma decays with similar energies.

6.4 Angular dependence of the efficiency

In Chapter 3.4 the importance of the angular dependence of the efficiency in the HPGe measurement is discussed. It is foreseen that this effect will have a large impact on the measurement with a detector submerged in the soil as well. Therefore the angular dependence of the efficiency has been measured for the LaBr$_3$ crystal coupled to the APD and the PMT respectively. The results are compared to the efficiency for a larger NaI-crystal (Figure 6.3). The deviation of the results for the APD and PMT readouts below 90° is believed to be due a slight relative misalignment in the distance to the source. The increased deviation at larger angles is due to the screening of the APD crystal and the metal holder. At even larger angles the PMT screens the photon flux and the efficiency also falls off for the PMT readout. The smaller change in efficiency for the NaI crystal is probably due to the larger PMT. The faster decrease in efficiency in the beginning is due to the pronounced asymmetry of the NaI crystal ($\varnothing 38.1 \times 12.7$ mm$^3$) in combination with a thinner front window. These measurements illustrate the need for correcting for the variation in efficiency, when the photon flux impinges on the detector from a large range of angles.

6.5 Conclusion

The studied LaBr$_3$ crystals yielded a resolution of approximately 4.5% (FWHM) at 662 keV which is slightly worse than the resolution stated in the
**Figure 6.1:** A typical energy distribution recorded with the LaBr$_3$ crystal coupled to a PMT using a $^{137}$Cs source placed at a distance of 16 cm. The laboratory background has been subtracted. The typical spectrum recorded using the APD is similar.

**Figure 6.2:** The energy distribution of the internal activity (including laboratory background activity) of the LaBr$_3$ crystal recorded in a low-background-environment. The spectral structures are identified by Ref. [49].
6.5 Conclusion

![Figure 6.3: The angular dependency of the peak efficiency (662 keV) of the LaBr$_3$ crystal coupled to the APD (○) and the PMT (▼) respectively (distance to source \(\sim 160\) mm). As a comparison the efficiency of a NaI (□) crystal (\(\varnothing 38.1 \times 12.7\) mm$^3$) coupled to a \(\varnothing 50.8\) mm PMT (distance to source \(\sim 190\) mm) is shown. Uncertainties are within the size of the points. A relative peak efficiency of 1 for the LaBr$_3$ (NaI) detector corresponds to \((4.00 \pm 0.02) \cdot 10^{-3}\) (\((16.76 \pm 0.04) \cdot 10^{-3}\)) percent of all photons emitted in the decay, giving rise to to an event in the full energy peak.]

Passports, and almost two times worse than the best reported values in the literature (crystal sizes are not identical) [27, 49]. The degraded resolution is probably partly due to the non-optimal PMT dynode-configuration and the smaller active area of the APD as compared to the crystal surface (such effects have also been seen in Ref. [46, 51]). The main drawbacks of the PMT-solution are the sensitivity to magnetic fields, size, power consumption, weight, and the fragility [2, 51]. The main drawback of the APD-solution is the strong sensitivity in gain due to ambient temperature and small changes in applied voltage. In our study the APD operation was also hampered by large noise contributions and somewhat worse resolution as compared to the PMT-readout. The size of the detector system for reading out the light using APDs is less than a third of that required for the PMT-solution, which is also an important parameter in the construction of a detector array.

Studies [44, 51] have shown that it is possible to achieve a resolution equally good with APDs and PMTs. Further, Flamanc and Rosza [51] have obtained a resolution of 3% and very good linearity up to 6 MeV using APD as readout device. These studies suggests that it is possible to achieve good linearity and better resolution than obtained using the present equipment. In the laboratory both solutions were working well, and the conclusion is that the LaBr$_3$-detector system is a good candidate for in situ measurements.
In Chapter 5 and 6 the properties of CdTe and LaBr$_3$ detectors have been investigated. In this chapter other comparative studies of detector properties are briefly described and conclusions about a suitable detector for the in situ measurements are drawn.

7.1 Other studies of detector properties

Dorenbos et al. [48] have made an extensive experimental comparison of a LaBr$_3$ and a NaI crystal both of dimensions $\varnothing 19 \times 19$ mm$^3$. They conclude that the LaBr$_3$ is superior to the NaI for gamma spectroscopy for energies above 100 keV. The LaBr$_3$ shows both better energy resolution (by a factor of two) and better linearity in light yield ($\pm 1\%$ as compared to $\pm 10\%$ for NaI in the energy range of 0.2-6 MeV). They have shown that the photoelectric attenuation coefficient is very similar for the two detectors, but that the Compton scattering coefficient is 25% larger for the LaBr$_3$ detector, leading to a significantly larger count rate in the full energy peak for large sized crystals (for energies around and above 1 MeV).

Milbrath et al. [52] have compared a LaBr$_3$ ($\varnothing 38.1 \times 38.1$ mm$^3$) with a NaI ($\varnothing 38.1 \times 55.9$ mm$^3$) detector. They have experimentally investigated the possibility to identify radionuclides typically found in cargo and special nuclear materials (both shielded and unshielded sources). The measurements were conducted in short time frames. Their aim was to investigate if the LaBr$_3$ is suitable to use in hand-held radioisotopes identification devices. They conclude that the LaBr$_3$ detector is superior to the NaI detector (the measuring times needed for the same level of significance was usual 2-3 times shorter and more peaks are identified). They also note the exception of $^{40}$K, where the NaI detector was better due to the destructive interference of the internal activity within the LaBr$_3$ detector.

Syntfeld et al. [53] have compared a LaBr$_3$ ($\varnothing 25.4 \times 25.4$ mm$^3$) with a large volume CdZnTe detector ($30 \times 15 \times 12.1$ mm$^3$). Their aim has also been to investigate the possibility to replace NaI crystals in hand-held radioisotope identification devices by detectors with better performance. The CdZnTe detector differs from the CdTe detectors studied in Chapter 5, not only by the addition of zinc (Zn), but also in the electrode configuration (a coplanar grid was implemented, described in Ref. [54], instead of the quasi-hemispheric electrode configuration). The detector was also fitted with a thermoelectric cooler.
for temperature stabilization. The LaBr$_3$ detector in their study shows better energy resolution and slightly higher peak efficiency. However, the CdZnTe is less affected by temperature changes (in the interval -10 to 40°C). The instability found for the LaBr$_3$ detector was mainly due to PMT instabilities. They conclude that both the large volume CdZnTe and the LaBr$_3$ detectors are a more suitable choice than NaI for radioisotope identification but, that LaBr$_3$ is superior to the CdZnTe detector. However, they also point out that there exist smaller CdZnTe detectors (volume of 2.25 cm$^3$) with energy resolution superior to that achieved by the tested LaBr$_3$ detector. The electrode configuration used in this study has the drawback of requiring two separate amplification chains and additional electronics for the pulse-height analysis, which can pose a problem due to the limited size available for the detector array in the current project.

Alexiev et al. [55] compared the energy resolution and detection efficiency of a LaBr$_3$ (12.9 cm$^3$), a CdZnTe (12.9 cm$^3$) and a NaI (21.2 cm$^3$) detector. The LaBr$_3$ detector yielded a better resolution than the NaI detector and the CdZnTe detector had better resolution than the LaBr$_3$ detector, but the CdZnTe detector was hampered by asymmetric peaks shapes (as is the case for CdTe-detectors, cf. Chapter 5). They concluded that LaBr$_3$ is a very good alternative to NaI detectors, and in the cases where moderate energy resolution is sufficient but high efficiency is of importance it is also to be preferred over CdZnTe.

7.2 Discussion and conclusion

The conclusion from the laboratory investigations and the literature review above was to continue field investigations using LaBr$_3$ detectors. The CdTe detectors studied in this thesis was rendered less suited for the present project, due to the need for cooling to subzero temperatures and rise time discrimination to reach its optimal performance. CdTe detectors, in general, are hampered by the limited crystal sizes and require additional units for signal processing to reach optimal performance. The NaI-detector is cheaper than LaBr$_3$ and can be made in a larger variety of sizes, but the poor energy resolution renders it less suitable in the case of several gamma energies closely spaced. Today, more than 20 years after the Chernobyl accident, the activity concentrations of $^{137}$Cs are relatively low, and a high efficiency is of large importance in order to minimize the measuring time in field. However, shortly after a fallout the need for efficiency is lower and the smaller size of the CdTe crystals (including the small overhead to read out the signals) may be more favorable as the major part of the activity most likely will be located close to the ground surface.
8. Field tests of detectors and method

This chapter is based on Paper IV.

In situ measurements were conducted approximately 200 km north of Stockholm close to the village Utöra (Figure 8.1) in order to test the method and detectors. This region of Sweden received a relatively large radioactive contamination due to the Chernobyl power plant accident in April 1986 and air surveys have located a local maximum of activity in a marsh within this region [25].

![Map showing the positions of the in situ measurements, in the region close to the village Utöra, discussed in this thesis.](image)

Figure 8.1: Map showing the positions of the in situ measurements, in the region close to the village Utöra, discussed in this thesis.

1The deposition occured mainly in connection to rain and the initial deposition density was approximatly 100 times larger than the most common values in Sweden (0.2-5 kBq/m²) [9, 25]
8.1 Description of the method

The idealized measurement (for one detector) is shown in Figure 8.2. The detector is positioned at a fixed depth below the surface in a hole in the ground. In the soil there is a certain radionuclide emitting gamma-rays with a given energy. The emitted gamma-rays are attenuated in the soil due to scattering and absorption, which yields a complex distribution of photon energies.

![Figure 8.2: Cross-section view of the in situ gamma-ray measurement, one detector submerged into the ground (idealized).](image)

In order to simplify the model, the detector is approximated by a point detector, and only the unscattered (full energy) gamma-rays are considered. The effects of this over-simplification are small for the present detector size and can to a large extent be compensated for by defining an operational detector efficiency as the ratio of the number of photons emitted from a point source at a given position (relative to the detector) and the number of photons detected in the full-energy peak. Further the soil is assumed to be homogeneous and the radionuclide concentration is assumed to depend only on the vertical distance from the surface, hence the model is cylinder symmetric.

To be able to determine the activity distribution with a limited number of detectors the depth dependence of the concentration is approximated by an analytical function, based upon transport and interaction mechanisms of radionuclids in soil [56]:

\[
C(z) = J_0 e^{-\lambda t} \left\{ \frac{1}{\sqrt{\pi Dt}} e^{-(z-vt)^2/(4Dt)} - \frac{v}{2D} e^{vz/D} \text{erfc} \left( \frac{v}{2\sqrt{D}} + \frac{z}{2\sqrt{Dt}} \right) \right\} \quad (8.1)
\]
The implemented function (8.1) has five parameters: the time of the fallout \((T)\), the decay constant of the radionuclide \((\lambda)\), which both are treated as constants in this approach, the total surface deposition \(J_0\), assuming the deposition to be momentary, the effective convection velocity \((v)\) and the effective dispersion \((D)\). The parameters of the distribution function are determined by fitting the model to the count rate measured in situ. Once the parameters are determined the total activity and mean depth can be deduced from the postulated distribution function. The model is described in more detail in Paper IV.

8.2 Exploratory measurements using NaI-detector

An exploratory field excursion was conducted using a NaI-detector \((\varnothing38.1 \times 12.7 \text{ mm}^3)\) coupled to a PMT in June 2007. The measurements were conducted in an alder marsh north of Gävle (Figure 8.1). The area, including vegetation, has been described in a study by Stark et al. [24]. Two sites were chosen approximately 1.6 m apart. One of the positions was located close to a local maximum in the photon flux above ground, identified by scanning the area using a Geiger-Müller counter. A large tent was set up above the measuring location to shield the detector and the electronics from moisture and direct sun light. A portable generator was used to power the equipment.

8.2.1 Soil sampling

Prior to each in situ measurement a soil sample was extracted (cf. Figure 8.3 and Paper IV). The extracted soil core was divided into approximately 1 cm thick slices, weighted and stored in plastic boxes (Figure 8.4). The activity concentration of the soil samples was later determined in a low-background-laboratory at the department by gamma-ray spectroscopy (Appendix A). Measurements of the depth of the holes left after soil samples had been extracted and the length of the soil cores differed by approximately 15-30%. This discrepancy makes the interpretation of data from the measurements on the soil samples uncertain, an uncertainty that will be addressed below.

8.2.2 In situ measurements

Standard laboratory electronics were used. The in situ measurement is illustrated in Figure 8.3. At each site gamma-ray spectra were recorded for every centimeter down to a depth of 13 cm, each for a duration of 900 s. In addition to the submerged measurements the area surrounding the sites was scanned using the NaI detector directly on the ground, in order to study the variations in the count rates in the immediate area surrounding the sites. (Figure 8.5).
8.2 Exploratory measurements using NaI-detector

Figure 8.3: In the schematic drawings and the photos above, the apparatus for extracting soil samples and detector support are shown. In Figures (a) and (d) the apparatus has been inserted into the ground. Figures (b) and (e) shows the outer and inner (aluminum) cylinder containing the soil sample after the upper part of the apparatus has been removed. In Figure (f) the soil sample has been removed and only the outer cylinder remains in the hole, stabilizing the soil walls. Figures (c) and (g) shows the detector inserted into the ground.

The absorption coefficient of the soil was measured at both sites by inserting a point source approximately 8 cm from the center of the detector at a depth of 7 cm (Figure 8.3). Positioning the NaI detector at the same depth, the absorption in the soil was estimated.

8.2.3 Results

The absorption coefficients of the soil at the two sites were identical within the limits of uncertainty. Gain variation of 30% due to incomplete shielding of the earth magnetic field and changes in the ambient temperature were noted during the measurements and an energy resolution of 11% (FWHM) at 662 keV was achieved, which is slightly worse than in the laboratory.

The activity distributions were derived by fitting the model to the measured number of registrations in the full energy peak. However, the comparison of the results for the activity distribution derived from the in situ measurements and the extracted soil samples is not straightforward, due to the difference
Figure 8.4: In field handling of soil samples. The aluminum cylinder containing the soil sample has been taken apart and the soil core is divided into approximately 10 mm thick slices and thereafter stored in plastics boxes (seen on the right).

in length of the extracted sample and the hole left in the ground. In order to overcome this ambiguity, two alternative and limiting assumptions regarding the extracted samples have been made:

a) The extracted part of the soil cores has not been compressed or altered. Instead the lower part of the soil column was compressed underneath and to some extent around the steel cylinder. Therefore not all activity originally contained within the hole has been extracted.

b) The extracted soil sample corresponds to the complete volume inside the sampler. The length difference is due to compression of the soil column, hence all activity that once was located within the hole has been extracted. In addition a constant density prior to sampling is assumed.

Using the second assumption above, the total activity for the two sites determined from the in situ measurements (soil samples) was $788\pm47$ ($736\pm53$) kBq/m$^2$ and $1582\pm97$ ($1207\pm87$) kBq/m$^2$, respectively. The mean depth of the activity, for both soil samples and in situ measurements, was estimated to be between 5 and 7 cm for both sites (cf. Paper IV). Uncertainties quoted correspond to statistical uncertainties from the fit and an estimated $\pm5$ mm uncertainty in the definition of the soil surface, that dominates the uncertainty in
8.2 Exploratory measurements using NaI-detector

Figure 8.5: Variation in the photon flux (number of counts per second in the $^{137}$Cs peak), in the region close to the extracted soil samples, measured using the NaI detector positioned on the ground.

the mean depth. The first assumption (above) yields a somewhat smaller values for the total activity (difference less than 6%) and a somewhat shallower activity distribution. The distributions determined from the in situ measurements and the soil samples, respectively, are shown in Paper IV.

8.2.4 Conclusions and discussion

The agreement between the measured count rate in the full energy peak and the values predicted by the model were excellent at both sites. However, at one site results from analysis of soil sample and in situ measurement agreed within the limits of uncertainty, while at the other site a significant difference between the results was found. This discrepancy, however, is not larger than what could be expected due to the large variation in the activity between the two sites and taking into account that 50% of the photons registered in the full energy peak originate more than 0.25 m from the center of the soil sample. In addition, the presence of soil inhomogeneities, like roots and stones, may partly block the passage of soil into the sampler as it is inserted into the ground. This will lead to an additional uncertainty of how much of the activity original contained in the soil core is transferred to the extracted sample. Even if these inhomogeneities will also effect the in situ measurements, these effect will be much less important due to the much larger volume sampled by the in situ measurement as compered to the volume occupied by the extracted soil core. The adequacy of the model chosen and the procedure of the analysis is illustrated by the close fit obtained to the measured intensities, and the overall agreement with the analysis from the extracted soil samples. The discrepancy between the soil samples and the actives measured in situ in combination with the deformation of the soil cores raises additional questions regarding how reliable the details actually are, that can be determined from the extracted soil samples. A study by Lettner et al. [57] estimates the uncertainty in the determined activity due to the soil sampling procedure (including the measurement) to be in the order of 10%.

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8.3 Field tests of a LaBr$_3$-detector

The second field test was conducted using a LaBr$_3$ crystal coupled to an APD and a PMT, respectively. However, in October 2008 the alder marsh was partly covered with water and the measurements had to be conducted a few hundred meters south of the previous site in a spruce forest close to Verkmyr-stream (Figure 8.1). A dosimeter was used to scan the region along the stream to find another local maximum. The dose rate at the chosen site was approximately 4-5 times lower than at site in the alder marsh. The aim of the excursion was to test the LaBr$_3$ and the readout devices under field conditions and to determine the minimum number of detectors needed in the array to determine the activity distribution.

8.3.1 In situ measurements

A series of measurements similar to those described in the previous section including extraction of a soil sample was performed at a single site. The in situ measurements were repeated twice, first using the APD and then the PMT. Measurements were done in steps of 10 mm from the surface down to 170 mm. At each position the gamma-ray spectrum was recorded during 900 s. In addition, the soil absorption was measured at two depths (48 mm and 96 mm below the surface). The equipment for soil sampling was identical to the one used during the previous excursion, while the upper steel cylinder had been modified to allow simpler adjustment of the detector position. Representative spectra from the in situ measurements are shown in Figure 8.6.

8.3.2 Results

The absorption coefficient of the soil was found to be the same at both depths within the limits of uncertainty, and in the further analysis the average of the measured coefficients was used.

The overall drift of the $^{137}$Cs peak position in the spectra using the PMT was very small, in the order of 1%. For the APD the drift was larger (10%) but the gain was monotonously increasing at a slow rate. In spite of the drift, the energy resolution was somewhat better in the field than found previously in the laboratory, probably due to the lower ambient temperature.

The analysis of the measurements was in principle carried out as for the NaI measurements in the previous section and the results were essentially identical for both APD and PMT. In Figure 8.7 the deduced activity distribution is compared to the activity distribution in the corresponding soil sample. The determined total activity and mean depth using all available data, using the second assumption in Section 8.2 (the soil core is compressed and had a constant density prior to sampling), was 121±6 (95±7) kBq/m$^2$ and 6.3±0.5 (6.8±0.5) cm, respectively, where the results from the soil sampling are given within the parentheses. Using the first assumption regarding the soil sample, the result
8.3 Field tests of a LaBr$_3$-detector

Figure 8.6: Typical pulse-height spectra from the in situ measurements using the LaBr$_3$ detector. The crystal center was positioned approximately 25 mm and 165 mm below the surface, respectively. The large photon intensity below approximately 300 keV is due to multiple scattering in the soil [58]. The peak at approximately 30 keV is (mainly) due to x-rays from the decay of $^{138}$La (internal activity, cf. Chapter 6.3).

for the total activity determined in situ will be 4% lower. The result for the total activity is in agreement with other studies in the same region [24,25,59].

8.3.3 Conclusions and discussion

Due to the better energy resolution of the LaBr$_3$ detector as compared to the NaI-detector used the previous year, the determination of the intensity of the cesium peak in the spectra was rendered much simpler in spite of the lower statistics. However, it should be noted that the activities determined from the soil samples are in both excursions somewhat lower than the activities determined from the in situ measurements.

In order to estimate the minimum number of detectors needed to determine the activity distribution analysis was made of subsets of three to six measurements including the top and bottom position and one to four evenly distributed intermediate points. The analysis showed that using only three detectors (the minimum required in order to use the analytical function postulated) the results were sensitive to the precise choice of detector positions. The results using four to six detector positions was more stable and close to identical with the results given above with slightly larger uncertainties. The conclusion is that a minimum of four detectors, evenly distributed over the range corre-
Field tests of detectors and method

**Figure 8.7:** Activity distribution deduced from in situ measurements (shaded area) compared to results determined from soil samples (points). Distribution derived under assumption a) and b) (cf. Section 8.2) are represented by circles (○) and squares (■) respectively. Measured intensity along with the result from the fit is shown in the inset.

Corresponding to the main part of the contamination, is required. The results using PMTs and APDs are in practice identical, hence, in order to minimize the size of the detector array, APDs are to be preferred as read-out devices.

In the present measurements the drift in the amplification did not significantly affect the results. In case of longer measuring times (needed for lower activity concentrations) or the presence of several radionuclides emitting photons in the same energy region, the drift may cause problems. For a fresh fallout the measuring times will most likely be shorter than the currently used, but on the other hand several several gamma-emitting radionuclides may be present.

There exist several possible ways to correct for or avoid the problems due to gain shifts, such as measuring shorter times and add spectra together after the measurement (and simultaneously correct for the drift). Kataoka et al. [60] have designed a system using a temperature sensor close to the APD to determine the required change in operating voltage in order to maintain the gain. Another solution is to use a peak in the spectrum and monitor the count rate, and if the rate in the region changes, let the electronics change the applied high voltage to counteract the gain shift. In case of a fresh fallout, there will exists peaks with high enough count rates to implement this method, in other situations an artificial peak could be added using for example a light emitting diode. An implementation of such an idea has been made by Pausch et al. [61].

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The system developed also reduces distortions due to pile up and noise, which can become important when having a fresh fallout with high count rates or a situation with low count rates and closely spaced peaks in the spectrum.

8.4 Mapping of activity distribution in Utnora marsh

In order to demonstrate the applicability of in situ measurements to determine, in detail, the depth distribution of activity, the $^{137}$Cs contamination in an area covering approximately 350 m$^2$ in the south-west corner of Utnora marsh has been mapped in the autumn of 2009.

8.4.1 In situ measurements

Due to delivery problems of the APDs the measurements were carried out using two LaBr$_3$ crystals (Ø17 mm) coupled to small PMTs (Ø13 mm). In order to emulate four detectors the two available units were fitted inside a cylinder which could be positioned at two depths, yielding in total four detector positions (Figure 8.8).

Prior to selecting the area for which the activity distribution should be mapped, a rough scan over the region was conducted using a dosimeter. The maximum dose rate was found to coincide with an area surrounding the measuring sites from 2003 and 2007, indicating that the south-west corner of the marsh constitutes a local hot-spot in the region. This conclusion is also supported by a larger mapping of the activity in the region conducted in 2001 by Nylén et al. [25]. The dose rate (0.5 m above the ground) where the HPGe detector was positioned in 2003 was 0.7 $\mu$Sv h$^{-1}$, while the dose rate measured approximately 75 m to the south-west was a factor of two smaller.

The field equipment used in the mapping of the activity was in principle the same as in the previous field experiments, with two exceptions, first, two detectors are used and read out simultaneously, secondly, the steel cylinder used to insert the detector array into the ground was narrower and did not support soil sampling (Figure 8.8). The electronics was stationed in a large tent next to the area mapped and cables of approximately 30 m length were used in between the electronics and the detectors. Due to the relative large output signal from the PMTs the preamplifiers could be located in the working tent, in contrast to the output signal from an APD which is much smaller and would have required the preamplifier to be located as close as possible to the detector. The PMTs were operated at a high voltage of -600 V, which is lower than the recommended voltage, in order to restore linearity up to approximately 700 keV (cf. Chapter 6). The resolution was 10% worse than in the previous field measurements, mainly due to the mismatch in size between the crystals and PMTs.
Figure 8.8: Schematic drawing of apparatus for mapping the activity distribution using multiple detectors. In figures a) and b) the two alternative positions of the detectors are shown. An in situ measurement with the detectors inserted into the soil is shown in c), where the handle has been removed.

The activity concentration was scheduled to be determined at 100 sites, with 2±0.3 m between adjacent positions distributed in a regular grid covering an area of 18×18 m² (cf. Appendix C). Out of these sites 11 were not measured because it was not possible to insert the detector array within 30 cm of the scheduled position due to environmental conditions, for example ground flooded with water or nearby trees and roots. At each of the remaining 89 sites four spectra were recorded, two at a time, for a duration of 450 s. In Figure 8.9 the mapped area is shown along with the working tent including the electronics and data acquisition system. In order to minimize vibrations and shocks to the detector array, a hole was made using a heavy steel rod (with a smaller diameter than the detector array) prior to the insertion of the detector array.

8.4.2 Results

The soil absorption was measured at 15 sites (yielding 60 measurements), in principle as described in Section 8.2. The point source position was positioned at a depth of 67 mm. The spread of the result for the absorption coefficient within the same site was of the same order as the spread between sites. Therefore the average was used in the calculations of the activity distributions.

A maximum drift of 15% in the peak position for the 662 keV line of $^{137}$Cs was seen during the excursion. The ambient temperature varied from 7 to 15°C and the peak positions for both detectors were found to be strongly correlated to the temperature. The main contribution to the drift was probably due to the temperature response of the PMTs.
8.4 Mapping of activity distribution in Utnora marsh

Figure 8.9: The major part of the investigated area is shown in the upper left hand corner including the working tent. The strings shown were used to create a grid to determine the position of each site. (A hole is prepared using a steel rod). In the upper right hand corner the detector array is moved to a new position and inserted into the ground. In the lower left hand corner the power generator is shown. The working tent including the electronics and the data acquisition system is shown in the lower right hand corner.

The model (described in Section 8.1) was fitted to the measured count rates at each site and the activity distribution was determined. A map of the distribution of activity and mean depth are shown in Figures 8.10 and 8.11, respectively. The determined activity varies from 380 to 2230 kBq/m² while the uncertainty of the individual measurements is on the order of 10 kBq/m², hence the spread in activity between different sites is much larger than the uncertainty due to statistics and model uncertainties for the individual sites. This is also true for the mean depth of the activity, varying from 4 to almost 12 cm, with an individual uncertainty of 0.5 cm (dominated by the uncertainty in the definition of the position of the surface).

The average of the determined activity and mean depth is presented in Table 8.1 along with a comparison with the results from the excursion 2003 (Chapter 3) corrected for the intervening time. The results for the activity
Field tests of detectors and method

<table>
<thead>
<tr>
<th>Measurement</th>
<th>Activity (kBq/m²)</th>
<th>Mean depth (cm)</th>
<th>Year</th>
<th>Number of sample(s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Extracted soil samples</td>
<td>895±46&lt;sup&gt;a&lt;/sup&gt;</td>
<td>6.3±0.5&lt;sup&gt;b,c&lt;/sup&gt;</td>
<td>2003</td>
<td>6</td>
</tr>
<tr>
<td>HPGe in situ (above ground)</td>
<td>921±53&lt;sup&gt;a&lt;/sup&gt;</td>
<td>7.9±1.6&lt;sup&gt;b&lt;/sup&gt;</td>
<td>2003</td>
<td>1</td>
</tr>
<tr>
<td>LaBr&lt;sub&gt;3&lt;/sub&gt; in situ (submerged)</td>
<td>893±47&lt;sup&gt;d&lt;/sup&gt;</td>
<td>8.0±0.5&lt;sup&gt;d&lt;/sup&gt;</td>
<td>2009</td>
<td>89</td>
</tr>
</tbody>
</table>

<sup>a</sup> The result has been corrected for decay to the date of the measurement 2009.

<sup>b</sup> The result has been corrected for vertical migration (only approximate).

<sup>c</sup> The result is not corrected for soil compression and is therefore likely to be an under estimate.

<sup>d</sup> The result presented here is based on the constant density assumption, cf. text.

Table 8.1: The average results from the excursion 2009 compared with results from 2003 (corrected for decay and migration to the date of the measurement 2009). The activity for the individual soil samples from 2003 are presented in Figure 3.5 and should be multiplied with 0.87 to correspond to the autumn of 2009. The uncertainties reported here for the soil samples 2003 and the in situ measurements 2009 are the standard deviations of the mean for the respective populations, cf. text.

distribution agrees well for the two determinations. The results are also in qualitative agreement with other studies in the region [24, 25, 59]. However, the very large local fluctuations render any detailed comparison to the latter measurements impossible.

Stark et al. [24] concluded, from their studies, that until 2003 a regular redistribution of activity occurred from a nearby lake by the outlet stream passing through the wetland thereby slowing down the decrease of activity in the marsh. The results from the mapping of the activity distribution compared to the measurements using the HPGe detector in 2003 indicates that in the southern part of the marsh (approximately 20-40 m from the Verkmyr-stream) no significant additional accumulation has taken place since 2003.

8.4.3 Discussion and conclusion

In contrast to the previous measurements (in 2007 and 2008) there are no soil samples extracted prior to the in situ measurements, and therefore the soil volume corresponding to the steel cylinder holding the detector array must be redistributed upon insertion, whereas in the previous measurements only a minor volume of soil that did not fit into the sampler was redistributed. This results in a compression of the soil close to the detector array, however, this process is thought to affect the results much less than the compression problems discussed earlier regarding the soil sampling procedure, mainly because the volume of soil contributing to the measured flux in situ is much larger than the volume of the soil sample (and disturbed soil column). However, a complication, is the the possibility of increased activity (due to a compressed soil layer) close to the detector. In the model this has been taken into account
by adding an layer with increased density (both of mass and activity) around the steel cylinder. The compressibility of the soil is estimated from the soil samples extracted in the previous excursions. For simplicity a homogeneous layer large enough to contain both the original and redistributed soil, due to the insertion of the detector array, is modeled around the steel cylinder. The exact assumption is not crucial for the derived results, but if this correction is neglected the result for the activity will be over estimated by approximately 10%.

The mapped area is not truly flat (as assumed in the model), large scale gradients up to 10 cm per meter have been noticed, along with local depressions in the ground. In this study no correlation study between the topography and the activity distribution has been made. However, a study by Haugen [23] suggests that the hot-spots can be formed due to local depressions in the ground. In an other study by Stark et al. [24] the main factors influencing the accumulation of $^{137}$Cs are summarized, including the effects of topography but also other factors, like the vegetation, that can act as a filter and retain cesium.
The in situ measurements have demonstrated that inserting an array of detectors into the ground gives a reliable determination of the distribution of activity in a relatively short time without the need for time consuming laboratory analysis. The total measuring time including moving the detector array was less than 20 minutes per site, however, with four detectors the time could almost be cut in half. In addition, using crystals somewhat larger than the ones used in the current measurements would increase the efficiency and further reduced the required measuring time. The amount of time is an important parameter in the competition with soil sampling. As a comparison it takes approximately 5-10 minutes to extract a soil sample, including transfer to storage for the transport to the laboratory and cleaning the sampler.

Soil samples may yield a more detailed information about the radionuclide concentration and distribution by gamma-ray spectrometry in the laboratory, but may also be less representative for the true activity distribution in the field due to the large deformation of the small sample volume compared to the in situ measurements. An additional advantage compared to traditional soil
8.4 Mapping of activity distribution in Utnora marsh

sampling is that the activity distribution can be directly monitored during the in situ measurement. This in situ monitoring gives the possibility to adjust the number of sites measured if large variations are found.
9. Conclusions and Outlook

9.1 Conclusions

In this thesis methods and detectors have been investigated with the aim to improve already existing methods and to develop new efficient ways to quantify the contamination by radionuclides in the environment, both for routine measurements but also as possible tools in an emergency.

Using a high-resolution gamma detector (HPGe) the average surface activity and average burial depth over a larger area can be determined in an efficient way. Utilizing the measured detector response and the stand-alone simulated photon transport library the method can be applied to any HPGe detector available at the time of the measurement. The method has been tested with good results at six different sites in Sweden with activities ranging from 15 kBq/m² to over 1000 kBq/m² and measuring times not exceeding one hour.

In the second part of the thesis detector studies have been reported where two relatively new detector materials, CdTe (semiconductor) and LaBr₃ (scintillator) have been investigated with the aim to see if they were suitable to use for in situ measurements. Finally a method was developed to offer an alternative to the traditional soil sampling, using an array of four LaBr₃ detectors inserted into the ground. By the implemented method the activity distribution can be determined over a small area in the same time required to extract a soil sample.

The activity distribution in an alder marsh north of Gävle has been studied using the HPGe in situ measurements above ground (2003) and using the detector array and measuring at 89 sites surrounding the HPGe detector position. The results from the two measurements are in agreement within narrow limits of uncertainty. These studies have shown that the two developed methods to determine the radionuclide contamination in the environment are efficient and reliable. The HPGe measurements give the average activity concentration and buried depth. The detector array, on the other hand, can study the depth profiles in more detail, in principle it provides the same information as the soil samples without the time consuming laboratory analysis.
9.2 Outlook

A further development of the method for in situ measurements using a detector above ground could utilize a larger part of the spectrum in order to determine more than two parameters of the distribution of activity. The limits of applicability of the method in cases where the in situ measurements are conducted in locations with large scale structures (i.e. many trees, dense vegetation, large rocks, large topographical variations) should be further studied along with the limitation of the method in case there are several strong gamma-emitting radionuclides present in the contamination.

The properties of the LaBr$_3$ detector coupled to either a PMT or an APD are very good both in the laboratory and in the field. The available size of the crystals are increasing. A future project would be to study if it is possible to replace the HPGe detector used for the in situ measurements above ground with a large LaBr$_3$ detector, which would have the advantages of larger efficiency (shorter measuring times) and room temperature operation.

In the simple model developed for the detector array submerged into the ground the soil absorption has to be measured separately. In the field measurements presented in this thesis this was not a real problem, because the activity of the point source dominated over the contamination distributed within the soil. In a fresh fallout, however, it may be hard to measure the absorption directly. Instead the possibility to deduce the absorption in the soil from the distribution of scattered photons should be investigated. Scattered photons could also be included in the analysis in order to further reduce the required measuring time. In both cases, the effect of scattering within the detector array would also have to be studied.

In this thesis the $^{137}$Cs distribution in the soil due to a fallout occurring more than 20 years ago has been studied and conclusions regarding the number of detectors and their relative positions have been drawn. In case of a fresh fallout the activity will be located closer to the surface and a non-negligible amount may be deposited on the vegetation above ground. In addition, an older contamination can in some places significantly contribute to the photon flux, especially at larger depths. A future project would be to investigate this situation more in detail, and try to deduce if four detectors are sufficient, and how the optimal position of the detectors should be. In this scenario the use of CdTe detectors may be more favorable as they can be stacked closer together, and the need for high efficiency is lower. Additional investigations into methods for correcting the pulse-height deficit of CdTe detectors utilizing sampled pulse-shapes could be undertaken in order to determine if the efficiency and resolution can be further improved. The optimal solution may even be a mix of the two detector materials, several CdTe crystals close to the surface and more spacious located LaBr$_3$ crystals at greater depths.
Appendices
A. Analysis of soil samples

After returning to the department the extracted soil samples were prepared for the analysis. First the samples were repacked. The procedure included homogenizing the samples, refilling them into smaller plastic boxes and compressing them (Figure A.1). Thereafter the activities of the soil samples were measured in the low-level-laboratory using a HPGe detector. The samples were inserted as close as possible in front of the detector.

![Figure A.1: Preparation of soil samples. To the left an unhomogenized sample from the field and to the right a homogenized sample, repacked, and ready to be brought to the the low-level-laboratory for analyses by gamma-ray spectroscopy.](image)

A.1 Calculation of activity in a soil sample

The activity of the soil samples at the time of the extraction can be calculated using Equation A.1. This yields the total activity of a single slice of the soil core. To calculate the surface activity and the concentration the activity of the slice has to be divided by the cross section area of the soil sampler and volume occupied by the slice (prior to extraction), respectively. The total activity for the soil core is calculated by summing the results for the individual slices. In these calculation the inner diameter of the soil sampler was used to define the cross section area of the soil core sampled (cf. Chapter 8 and Paper IV).

\[
\text{Activity} = \frac{N \cdot e^{\lambda \cdot \Delta T}}{\beta \cdot T \cdot e \cdot C_{abs}} \quad [Bq]
\]
where

\[ N \] is the area of \(^{137}\)Cs peak
\[ \beta \] is the gamma intensity (0.85 for \(^{137}\)Cs)
\[ T \] is the measuring time [s]
\[ \varepsilon \] is the detector efficiency
\[ C_{abs} \] is the self absorption correction
\[ \Delta T \] is the time since the extraction [years]
\[ \lambda \] is the the decay constant [years\(^{-1}\)]

The detector efficiency was measured using an extended \(^{137}\)Cs source of known activity. The calculation of the self absorption was conducted in two steps, first the total absorption of a soil slice was measured using a \(^{137}\)Cs source behind the sample. This result was used as an input parameter in order to calculate, using the simulation package Penelope [28], the self absorption due to the homogeneous distribution of activity throughout the soil sample. The correction for self absorption was less than 10% for all samples.

**A.2 Low-level laboratory**

The laboratory is located on the ground floor of the AlbaNova University Center in Stockholm. The laboratory has a floor area of \(6.6 \times 3.3\) m\(^2\) and is shielded by 0.6 m low-level concrete on the inside covered with 50 mm thick iron plates. The incoming air is filtered to remove radon daughters. The laboratory is described in more detail in Ref. [62]. In order to further reduce the environmental background the HPGe gamma detector is placed inside a lead shield, built from 5 mm steel, 100 mm lead and 1 mm copper (Figure A.2).

On top of the lead shield an active muon shield is located. Muons are, highly penetrative, charged elementary particles mainly created in the earth’s atmosphere by the cosmic radiation. The muon contribution to the background measured with the HPGe detector is two fold, firstly a continuum due to direct muon interaction with the HPGe crystal and, secondly, due to muon interaction with the lead shielding, resulting in 511 keV photons (from annihilation of positrons created via pair production) and lead x-rays (Figure A.3).

The task of the muon shield is to detect the passage of the muons. The HPGe detector is operated in anti-coincidence with the muon shield (cf. Figure A.4). The muon shield reduces the background count rate with more than 50% in the energy range from 100 keV to 3 MeV. The muon shield is further described in Ref. [63].

As a contribution to the data base describing the properties of the low-level laboratory a determination was made of the thermal neutron flux inside the lead shield. For that purpose a set of of gold objects (18 carat) with a total
mass of approximately 18 g were activated by the naturally occurring neutron flux at ground level with negligible over burden outside Stockholm for several weeks. A neutron captured results in the instable isotope $^{198}\text{Au}$, which has a half life of 2.7 days and decays to an excited state in $^{198}\text{Hg}$ which in turn de excites and emits a 411.8 keV gamma-ray.

The measurements were started two hours after the gold objects were placed inside the lead shield in order to minimize the contribution from radon daughters. Gamma-ray spectra were saved every 15 hours in order to deduce the intensity change of the 411.8 keV line, for a total of more than 500 h. Because of the large absorption of both neutrons and gamma-rays the effective number of activated nuclei as well as the self absorption of gamma quanta was determined by extensive simulations.

The neutron flux as well as the cross section are energy dependent and therefore the results is given by the integral of the product which corresponds to the probability for a given gold atom to be activated per second:

$$\langle \sigma \Phi \rangle = \int_0^\infty \sigma(E) \frac{d\Phi}{dE} dE \quad (A.2)$$

In this notation the result at ground level and outdoor is $\langle \sigma \Phi \rangle = (4.9 \pm 0.9) \cdot 10^{-25} \text{ s}^{-1}$. In the lead cave in the low-level laboratory the probability is $\langle \sigma \Phi \rangle = (2.5 \pm 4.5) \cdot 10^{-26} \text{ s}^{-1}$. The thermal neutron flux inside the lead shield placed in the low-level laboratory is thus at least a factor of five below (90% confidence) the outdoor flux at ground level.
A.2 Low-level laboratory

Figure A.3: Spectrum recorded by the HPGe detector in coincidence with the muon shield.

Figure A.4: Spectrum recorded by the HPGe detector in anti-coincidence with the muon shield.
B. Results from soil sampling in Stockholm

A single soil sample was extracted in December 2007 at Brunnsviken (Stockholm), without making any in situ measurements. The soil sampling procedure as well as the analysis were identical to the procedure used for the previous excursions (Chapter 8 and Appendix A). The total activity was $1.1 \pm 0.1$ kBq/m$^2$ and the depth distribution is shown in Figure B.1. The $^{137}$Cs content is the sum of the fallout from weapon testing (mainly deposited in the 1960’s and 1970’s) and the fallout from the nuclear power plant accident in Chernobyl (1986). In contrast to the measurement north of Gävle, presented in Chapter 8, the two contributions in Stockholm are of the same order of magnitude [9].

Figure B.1: Soil sampling in Stockholm. The activity distribution is determined by gamma-spectrometry in the laboratory. The two profiles corresponds to different assumptions regarding the deformation of the soil core during extraction (cf. Chapter 8).
Map of the region in the alder marsh (200 km north of Stockholm) where the activity distribution was measured in the autumn 2009. The HPGe measurements (2003) along with the soil samples (2003) are discussed in Chapter 3 and the measurements using the NaI-detector 2007 (positions A and B) are discussed in Chapter 8.
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