Sensitivity of in-situ $\gamma$-ray spectra to soil bulk density and water content

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Abstract
The effects of sediment water content and bulk density on measurements of in-situ environmental $\gamma$-radiation were investigated using Monte-Carlo simulations. The simulations consider a large bismuth-germanate detector in a semi-infinite geometry with sediment compositions based on the range found from sediment samples. The volume contributing radiation to the detector decreases with increasing sediment bulk density, as the detector ‘sees’ a constant sediment mass. For typical soil bulk densities, the effective depth of detection is between 40 and 60 cm and the effective width between 160 and 260 cm. The effect of variations in porosity on $\gamma$-ray spectra is negligible. Variations in water content affect the shape of the spectra only in the region below 0.1 MeV and, more importantly, strongly affect spectral intensity. Separate measurements of sediment water content, when expressed as mass fraction of water, can be used to correct spectra collected on sediments with varying water content. The simulations were further used to describe the effective attenuation of the entire decay series of $^{232}$Th and $^{238}$U in a semi-infinite geometry, which gave effective energies of 0.62 MeV and 0.59 MeV, respectively.


Upper photo: the salt marsh of Schiermonnikoog being partly flooded, which is one of the causes for large variations in water content on the marsh.
Lower photo: dried sediment samples.
3.1 Introduction: salt marshes

In general, sediments in natural settings consist of a mixture of sediment and/or organic material and pores. The pores are filled with water and/or air. Due to natural processes, the water content and bulk density of the soil may vary vertically (e.g. Perrin et al., 2006), in space (e.g. Flemming and Delafontaine, 2000) and time. Most sediments contain γ-ray emitting radionuclides. *In-situ* measurements of the activity concentrations of radionuclides are for instance used for determining dose rates from the anthropogenic $^{137}\text{Cs}$. Another application is the use of $^{40}\text{K}$, $^{232}\text{Th}$ and $^{238}\text{U}$ activity concentrations for characterising sediment and sediment transport (De Meijer, 1998; Anjos et al., 2007).

Gamma-ray emitting radionuclides are mainly present in the mineral phase of sediment. In sedimentological studies, the activity concentrations in these mineral grains (the dry activity concentrations) are the desired quantity. However, part of the radiation from the sediment may interact with the sediment before being detected by an *in-situ* detector. Variations in water content or bulk density may influence the intensity and shape of measured spectra, the resulting activity concentrations and photon-fluence rates (Schimmack et al., 1998; Tyler et al., 2001; Kluson, 2001; Hendriks et al., 2001b; Van Wijngaarden et al., 2002a; Maučec et al., 2004a; Maučec et al., 2004b; Perrin et al., 2006). As a consequence of the varying water content *in-situ* activity concentrations are often reported on basis of wet mass. The question, investigated in this chapter is: how do these changing soil conditions influence *in-situ* γ-ray spectra and the quantities derived from it?

Variations in density and water content also affect the volume of sediment as ‘seen’ by the detector. For the γ-rays of environmental radionuclides the predominant interaction with matter is Compton scattering which is described by the Klein-Nishina formula (Knoll, 2000) and which has per unit of mass (in first order) a Z-independent cross-section (see Chapter 2). Hence, the detector ‘sees’ a certain bulk mass of matter and the corresponding volume is expected to be inversely proportional to the bulk density.

Although Compton scattering contributions have already been used to estimate field moist bulk density from *in-situ* measurements of $^{40}\text{K}$ (Tyler et al., 2001), it remains difficult and time-consuming to experimentally assess the effects of changes in sediment properties on entire γ-ray spectra. Moreover, analytical solutions, in general, only address the unattenuated γ-ray flux. Monte-Carlo simulations can provide spectral information and therefore were deployed in this work to simulate γ-ray spectra. Monte-Carlo simulations have additional advantages over experiments for calibration and spectra generation. The uncertainties in detector resolution and energy calibration do not play a role, all variables can be controlled separately and there is no background radiation present (Fehrenbacher et al., 1996; Helmer et al., 1999; Hendriks et al., 2002). Monte-Carlo simulations were already successfully applied to the creation of γ-ray spectra from *in-situ* measurements and for large volume inorganic scintillator detectors (see e.g. Saito and Jacob, 1995; Thummerer
and Jacob, 1998; Kluson, 2001; Hendriks et al., 2002; Gering et al., 2002; Hendriks, 2003; Maucé et al., 2004a).

3.2 Methods

First, we will determine the range of sediment conditions that may be encountered in a survey of \textit{in-situ} radioactivity, using samples from some of the measurement areas of this thesis. Next, the effect of density and water content on shape and intensity of natural \(\gamma\)-ray spectra will be studied by Monte-Carlo simulations in a so-called flat-bed situation (with the detector on top of a semi-infinite medium). For that, first the sediment volume contributing to the detector signal is estimated. Secondly, the effects of density and water content for mono-energetic \(\gamma\)-radiation originating from this volume are assessed. Thereafter, spectra of relevant environmental radionuclides are addressed. Finally, we discuss how \textit{in-situ} measurements can be corrected for variations in sediment properties and we use the simulation results to determine the average attenuation behaviour in soil of the complete \(\gamma\)-ray spectrum of the decay series of \(^{232}\text{Th}\) and \(^{238}\text{U}\).

3.2.1 Sediment properties

In radiometry, the main interest is the activity concentration \(C\) (Bq kg\(^{-1}\)) of radionuclide \(j\) in the sediment:

\[
C_j = \frac{A_j}{m}, \quad (3.1)
\]

where \(A_j\) (Bq) is the activity and \(m\) (kg) the mass of a volume unit of sediment. Only the mineral grains within the sediment contain radionuclides. In dry sediment, the activity concentrations are:

\[
C_{j,dry} = \frac{A_j}{m_{dry}}, \quad (3.2)
\]

where \(m_{dry}\) stands for mass of dry solids. If the pores in the sediment contain water, \(A_j\) remains unchanged but the mass per unit volume increases and consequently the activity concentrations in the bulk sediment decrease:

\[
C_{j,wet} = \frac{A_j}{m_{wet}}, \quad (3.3)
\]

where \(m_{wet}\) is the mass of the wet sediment. Consequently the relation between \(C_{j,wet}\) and \(C_{j,dry}\) is:

\[
C_{j,wet} = C_{j,dry} \frac{m_{dry}}{m_{wet}}. \quad (3.4)
\]
The term $m_{\text{dry}} / m_{\text{wet}}$ is the mass of solids with respect to the total mass of the sediment. This is related to the mass of water with respect to the total mass of the sediment, which is the absolute water content $w_a$ (cf. Flemming and Delafontaine, 2000):

$$w_a = \frac{m_{\text{wet}} - m_{\text{dry}}}{m_{\text{wet}}}.$$ (3.5)

The absolute water content can be determined from sediment samples by taking the mass of the sample before ($m_{\text{wet}}$) and after ($m_{\text{dry}}$) drying. $C_{\text{wet}}$ and $C_{\text{dry}}$ are then related through:

$$C_{j,\text{wet}} = (1 - w_a)C_{j,\text{dry}}.$$ (3.6)

The wet bulk density $\rho_{\text{wet}}$ is the combined mass of the solids and water per unit volume

$$\rho_{\text{wet}} = \frac{m_{\text{wet}}}{V},$$ (3.7)

The dry bulk density ($\rho_{\text{dry}}$, the mass of the solids only per unit volume) follows from the water content and wet bulk density:

$$\rho_{\text{dry}} = (1 - w_a)\rho_{\text{wet}}.$$ (3.8)

From this, the porosity ($\varepsilon$, the volume fraction of pores) can be calculated:

$$\varepsilon = \frac{(\rho_s - \rho_{\text{dry}})}{\rho_s},$$ (3.9)

where $\rho_s$ is the density of the solids. In natural sediment mixtures $\rho_s$ is assumed to be 2.65 g cm$^{-3}$. The degree of water saturation of the pores ($s$) follows from:

$$s = \frac{\rho_{\text{wet}} - (1 - \varepsilon)\rho_s}{\varepsilon \rho_w},$$ (3.10)

where $\rho_w$ is the density of water, taken to be 1 g cm$^{-3}$.

The range of sediment conditions that may be encountered in surveys of in-situ radioactivity was derived from 106 sediment samples from the barrier island of Schiermonnikoog (Chapter 2). The samples were taken with a root corer (ø 7.3 cm) from the upper 5 cm of the soil or with pF-rings (ø 5 cm) from various depths. The samples were classified as fine-grained (dominated by silt) or coarse-grained (dominated by sand). Wet bulk density was determined taking untreated sample mass dividing it by sample volume. The samples were subsequently dried for 24 h at 105°C to determine water content, and porosity.

1 Water content can be expressed in several ways, and we have chosen one that is most suited to our purpose.
3.2.2 Simulations

The sensitivity assessment was carried out with Monte-Carlo simulations (see section 2.2.5). In the simulations the detector was always positioned on top of a large sediment volume that acted as the source of radioactivity (Figure 3.1). The sediment volume was designed to be large enough to mimic a flat-bed geometry. It was divided into concentric ring-shaped cells, stacked in several layers below the detector. The detector was positioned at the central axis of the cylinders. The simulated detector corresponds to the PANDORA detector of KVI (see section 2.2.6). The model included all parts of the detector crystal and its housing, except for the electronics and photomultiplier tube. The simulated detector was surrounded by air. The sediment was modelled as a mixture of quartz, water and empty pores. Quartz (SiO₂) was used as proxy for naturally occurring sediment mixtures which have approximately the same specific density. Simulations for this geometry and detector were previously validated with experiments by Maucèc et al. (2004a) and Van der Klis (2002).

Adjoint simulations: outlining the contributing sediment volume

The sediment volume that effectively contributes radiation to the detector was estimated from Monte-Carlo simulations of the adjoint type (Briesmeister, 2000). In these calculations the transport direction of the photons is reversed (e.g. from detector to source). The application of adjoint calculations for this specific type of problem is described in Hendriks (2003), and Maucèc et al. (2004b). Since in the simulations the transport direction of the photons is reversed, photons gain in energy during their transport. Consequently the ‘source’ is in the detector and γ-ray fluxes are calculated over cells in the sediment bed. The sediment source volume can then be approximated by the volume the photons in the adjoint calculation reach before they gain the pre-set cut-off energy: 2614 keV, representing the energy of the most energetic environmental γ-ray (emitted by 208Tl from the 232Th-series). Because of the limits imposed by adjoint calculations, the ‘source’ was constructed as a crude approximation of a typical recorded spectrum, in five energy bins from 0 to 3 MeV (Figure 3.2). For each sediment cell, the MCNPX cell flux (F4 tally) was determined and multiplied with the volume of each cell. This volume-flux (gamma-rays cm /starting photon) is equivalent to the importance of each cell for radiation contribution to the detector (Maucèc et al., 2004b).

The adjoint simulations were carried out for two types of sediment, to set the boundaries of the dimensions for further simulations. The two sediment types were based on samples and represent typical high- and low-density sediment from a...
barrier-island (Table 3.1). The extent and shape of the contributing volume were determined in a stepwise approach. The calculations started with a large volume of low-density sediment with large cell sizes, extending beyond the range of the most energetic photons. Subsequently, simulations with smaller cell sizes were used to outline the regions that contribute most radiation to the detector. These results were the basis for setting the size of the volume for the calculations.

Spectra simulations
The effects of density and water content on $\gamma$-ray spectra were determined by simulating the energy-deposition spectra in the crystal of the PANDORA detector. In these simulations the sediment was the source of the photons. The pulse-height spectrum (in counts per starting photon) in the detector crystal was calculated using F8-tallies

![Figure 3.2. Energy distribution of the adjoint source.](image)

### Table 3.1. Material specifications for the adjoint calculations.

<table>
<thead>
<tr>
<th>description</th>
<th>sediment bulk density (g cm$^{-3}$)</th>
<th>material</th>
<th>volume fraction</th>
<th>density (g cm$^{-3}$)</th>
<th>mass fraction</th>
<th>constituting elements</th>
<th>mass fraction elements in sediment</th>
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<tr>
<td>low-density</td>
<td>1.16</td>
<td>sand</td>
<td>0.25</td>
<td>2.65</td>
<td>0.57</td>
<td>Si</td>
<td>0.27</td>
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<td>sediment</td>
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<td>water</td>
<td>0.50</td>
<td>1.00</td>
<td>0.43</td>
<td>H</td>
<td>0.048</td>
</tr>
<tr>
<td></td>
<td></td>
<td>air</td>
<td>0.25</td>
<td>1.2 $\cdot 10^{-3}$</td>
<td>0.00026</td>
<td>negligible</td>
<td></td>
</tr>
<tr>
<td>high-density</td>
<td>2.01</td>
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<td>0.61</td>
<td>2.65</td>
<td>0.81</td>
<td>Si</td>
<td>0.38</td>
</tr>
<tr>
<td>sediment</td>
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<td>0.39</td>
<td>1.00</td>
<td>0.19</td>
<td>H</td>
<td>0.022</td>
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<td></td>
<td></td>
<td>air</td>
<td>-</td>
<td></td>
<td></td>
<td>O</td>
<td>0.30</td>
</tr>
</tbody>
</table>
of MCNPX. To speed up the calculations, a number of variance reduction methods were used (most importantly geometry truncation) and the simulations were split into surface source write/surface source read phases (Briesmeister, 2000; for the specific application used here, see Van der Klis, 2002; Hendriks, 2003). The sediment volume had a radius of 10 m at the surface which decreased stepwise to 120 cm at the maximum depth of 120 cm (Figure 3.1). Adjoint simulations were used to check whether this volume was large enough for all simulated sediment compositions. In each simulation one artificial radionuclide was simulated, emitting γ-rays at one energy, either 0.5, 1.0, 1.5, 2.0 or 2.5 MeV, encompassing the range of environmental γ-ray energies. In this way the energy dependence over the range of possible energies in natural γ-radiation could be assessed. All γ-rays had the same emission probability. The spectra were generated in 10 keV bins to have a detailed basis for the assessment of changes in spectral shape.

The sediment in the simulations consisted of a homogeneous mixture of quartz (SiO₂), water and empty pores. The entire sediment formation acted as a homogeneous source of photons. The radionuclides were considered to be present only in quartz, and not in the air or water in the pores, in a constant amount per unit mass of quartz. The simulated spectra were scaled to represent this situation.

We consider three parameters that may affect in-situ measurements of γ-radiation. These are sediment water content, porosity and γ-ray energy. Bulk density is not considered separately as it is defined by the combination of water content and porosity. Therefore the simulations were separated into the effects of porosity (constant elemental composition) and water content (varying elemental composition).

The effects of porosity and water content on the spectra were analysed for possible differences in spectral shape as well as intensity (c.f. Hendriks et al., 2001b; Hendriks, 2003). The effect of porosity was assessed by modelling sediment without water for increasing sediment porosities of ε = 0, 0.2, 0.4, 0.6 and 0.8. Sediment having a higher porosity without water was considered unrealistic.

For the effect of water content, the sediment geometries with porosity ε = 0.4 and 0.6 were filled up with water in five steps from dry to fully saturated (s = 0, 0.25, 0.50, 0.75 and 1) and the geometry of ε = 0.8 in two steps (s = 0.75 and 1). The simulations span a range of wa from 0 to 0.6 of the total sediment mass. Emitted γ-ray energy was 0.5 or 2.5 MeV.

A final set of simulations with various water contents was run to create realistic γ-ray spectra that meet the requirements for Full-Spectrum Analysis (FSA, see Chapter 2). For that, the shape of the spectra had to be artificially broadened such that the peak widths correspond to the widths in measured spectra. This was achieved by folding the 10 keV bin spectra with an appropriate Gaussian function such that the peak resolutions were characteristic for the PANDORA detector: 14.6% at the 0.662 MeV line of ¹³⁷Cs and 7.3% at the 2.614 MeV line of ²⁰⁸Tl. The γ-ray spectra were simulated for the individual radionuclide sources of ⁴⁰K, ²³²Th, ²³⁸U and ¹³⁷Cs. For ²³²Th and ²³⁸U, the entire decay series were taken into account, including all lines with their emission probabilities and assuming secular equilibrium between all progeny.
3.3 Results & discussion

3.3.1 Barrier-island sediments
The wet bulk density of the barrier-island sediments ranges between 0.63 and 2.08 g cm$^{-3}$ (upper left panel of Figure 3.3), where the bulk density of coarse-grained sediment is higher than that of fine-grained sediment. This is related to the lower porosity in coarse-grained (0.36 – 0.43) than in fine-grained sediment (0.54 – 0.90, lower left panel of Figure 3.3). Consequently the water contents of the coarse-grained samples are relatively low, even if some of the samples are saturated. Overall water contents range between 0.08 and 0.66 (right panel of Figure 3.3). The variations in density, water content and porosity are larger for fine-grained than for coarse-grained sediment. From the differences in water content and porosity between the two sediment types follows that the dominant grain size of the measurement area can already give an indication about the ranges of bulk density and water content encountered during measurements. The effect of variations in these parameters on detected sediment volume will be assessed in the next paragraph.

Figure 3.3. Distribution of sediment properties for coarse-grained ($N = 21$) and fine-grained ($N = 85$) samples from Schiermonnikoog.
3.3.2 Radiation contributing sediment volume

Based on the results of several large-volume adjoint simulations (of which the largest volume was 15 m wide and 1.5 m deep), for further adjoint simulations a sediment volume was chosen that incorporates over 99.5% of the total flux towards the detector (Figure 3.4). The importance of each cell in contributing radiation to the detector was determined from the percentage that a cell contributed to the total flux. Subsequently the percentages were ranked and plotted in a cross-section of the geometry (Figure 3.4). As expected, cells closest to the detector contribute most radiation to the detector. For sediment with $\rho = 1.16 \text{ g cm}^{-3}$, the 95% contributing volume is a half sphere under the detector up to 60 cm depth, plus a stretch up to 260 cm radius of the top layer. The wide extent at the surface is predominantly caused by photons travelling through the air above the sediment. For the high-density sediment, the contributing volume is smaller in both the vertical and horizontal directions. The 95% volume extends up to 40 cm deep and 160 cm radius. Consequently, this is the smallest detail that can be resolved with non-collimated in-situ measurements. Simulations with a smaller cell size in the upper two cell layers indicate that most radiation from further than approximately 1 m from the detector originates in a thin layer of soil of maximum 5 cm deep. Although this is a thin layer with respect to the total depth, the top 5 cm is responsible for almost 40% of the total flux to the detector for low-density sediment, and over 50% for high-density sediment.

In contrast to the volume, the total mass as seen by the detector does not significantly depend on bulk density. These results are in line with those of Maucvec et al. (2004b), who did similar simulations.

Figure 3.4. The cumulative relative contribution of cells to the γ-ray flux in the detector, for low-density sediment of 1.16 g cm$^{-3}$ (upper panel) and high-density sediment of 2.0 g cm$^{-3}$ (lower panel). All cells together contribute 100% γ-flux to the detector. The figure is a cross-section of half of the sediment body. The shaded sphere depicts the detector (not to scale) and the dashed line gives the line of symmetry.
3.3.3 Effect of porosity

The simulated spectra reproduce the expected features well, such as the full-energy peak, Compton continuum and escape peaks associated with pair production (Figure 3.5). The low intensity peak just below the full energy peak is due to escape of X-rays after emission of the photo electron (Debertin and Helmer, 1988). The shape variations between spectra are better presented by the ratio between a bin content in the spectrum with a given \( \varepsilon \) over the content of the same bin in the spectrum for \( \varepsilon = 0 \) (Figure 3.6). The predominantly horizontal lines indicate that porosity does not influence the shape of the \( \gamma \)-ray spectra, except when sediment porosity increases to \( \varepsilon = 0.8 \). The overall intensity of the spectra with \( \varepsilon = 0.8 \) decreases, where the decrease is a factor two higher in the lower channels than in the higher channels. However, these effects are most likely artefacts caused by geometry truncation. The sediment density for \( \varepsilon = 0.8 \) is very low and additional adjoint simulations show that in that case the simulation volume is too small: it is missing at least 6 % of the total contribution from a semi-infinite sediment source (in contrast, the simulations up to \( \varepsilon = 0.6 \) include over 99.3% of the total contribution). The stronger decrease in the lower-energy channels indicates that \( \gamma \)-radiation from the – missing – faraway regions contributes mostly to the low-energy part of the spectrum. This is because the probability that a \( \gamma \)-ray interacts with the sediment and loses part of its energy increases if the photon travels a large distance before reaching the detector. Because the spectra for \( \varepsilon = 0.8 \) are underestimating the detector response, these simulations are left out from further analysis.

Because the shape of the spectra does not significantly change, the total spectral contents can be compared (Figure 3.7). The spectral content increases with increasing \( \gamma \)-ray energy. This reflects the effect of the energy-dependence of the mass-attenuation coefficient. The effect of porosity on the spectral contents is very small: maximum 1.3% for the case where \( \varepsilon = 0.6 \). This is negligible compared to typical measurement uncertainties in in-situ radiometric surveys.

Figure 3.5. Spectra of simulations with a source emitting only 2.5 MeV \( \gamma \)-rays for sediment porosities of \( \varepsilon = 0, 0.2, 0.4, 0.6 \) and 0.8.
Sensitivity of in-situ $\gamma$-ray spectra

**Figure 3.6.** The relative spectra bin content for a number of artificial mono-energetic spectra for sediment porosities of $\varepsilon = 0$, 0.2, 0.4, 0.6 and 0.8. The content is calculated relative to the case $\varepsilon = 0$. The lower line always represents $\varepsilon = 0.8$. The spectral bins are 10 keV wide and the error bars represent statistical uncertainties in the simulations, given every 100 keV.

**Figure 3.7.** Spectral content (per starting photon per kg) as a function of porosity. Statistical uncertainties in the simulations are smaller than the marker size.
3.3.4 Effect of water content

General

The effect of water content on spectra was investigated from sediments with porosities of 0.4, 0.6 and 0.8 that were stepwise filled up with water. The resulting spectra for the artificial, mono-energetic $\gamma$-ray sources with $E_\gamma = 0.5$ and 2.5 MeV are presented relative to the spectrum at $\varepsilon = 0$ in Figure 3.8. Only for $E_\gamma < 0.1$ MeV a change in the spectra is observed which is consistent with a smaller attenuation coefficient for water compared to quartz in this energy region (a factor 1.4 at 0.5 MeV, Berger et al., 2007). With increasing water content, the scattered $\gamma$-rays with energies < 0.1 MeV therefore, undergo less attenuation than those with higher energies.

In Figure 3.9 the total content of the spectra (given as the total spectral content, in counts per starting photon per kg of dry quartz) is presented. The results show a linearly decreasing spectral content with increasing water content. By introducing more material between a point source and a detector, the attenuation of the unscattered radiation scales exponentially with the mass of the material. In addition, also the amount of scattered radiation (build up) impinging on the detector will increase. For a semi-infinite geometry, the results in Figure 3.9 show that the net effect on the total spectral contents (scattered and unscattered $\gamma$-rays) turns out to be linear. Adding water to the sediment, the activity concentrations in the detected mass decreases since the activity remains the same whereas the mass per unit volume increases.

Consequently, in a good approximation, the wet activity concentrations ($C_{wet}$) can be calculated from the dry activity concentrations ($C_{dry}$) using equation 3.6. The main effect of adding water is, according to equation 3.6, a nuclide-independent linear decrease in intensity with $w_a$, depicted by the straight lines in Figure 3.9. However, a secondary effect is present caused by differences in the mass-attenuation coefficients for water and quartz (see Box 3.1). The net effect of this is that the addition of water introduces a small extra decrease in spectral content. This extra decrease is linearly related to water content and depends additionally on the spectral region of interest and initial $\gamma$-ray energy. For the cases $w_a = 0.6$ it reaches a maximum of 3.5% for the $\gamma$-ray energy of 0.5 MeV and 5.5% for 2.5 MeV. This extra decrease can be accounted for by adding an extra factor $\theta$ on the right hand side of equation 3.6:

$$C_{j, wet} = (1 - (1 + \theta)w_a)C_{j, dry},$$

(3.11)

where $\theta$ is a radionuclide specific constant that can be derived from the slope of the lines in Figure 3.9. It is $0.0214 \pm 0.0014$ for $\gamma$-rays of 0.5 MeV and $0.035 \pm 0.005$ for 2.5 MeV, for a spectral region of interest of $0 - 3$ MeV. We like to point out that the absolute magnitude of the spectral content increases with increasing energy of the emitted $\gamma$-rays. This effect is already observed and discussed in section 3.3.3.
Sensitivity of in-situ $\gamma$-ray spectra

Figure 3.8. The relative $\gamma$-ray spectra for various water contents and two artificial, mono-energetic $\gamma$-ray energies. The simulations represent a case where sediment with a constant dry activity concentration is filled up with water. The bin content of the spectra is taken relative to the content of the corresponding bin of the spectrum with $\varepsilon = w_a = 0$. Bin width is 10 keV and the error bars represent statistical uncertainties in the simulations.

Figure 3.9. Content of the $\gamma$-ray spectra as a function of water content for artificial, mono-energetic $\gamma$-ray sources of 0.5 (filled circles) and 2.5 MeV (open triangles), per emitted $\gamma$-ray per kg dry sediment. The lines represent the expected spectral content according to equation 3.6. Statistical uncertainties in the simulations are smaller than the marker size.
Chapter 3

Box 3.1 Extra attenuation by water compared to quartz

The extra attenuation resulting from water can be explained as follows, schematised for a one-dimensional situation.

In general, the attenuation of $\gamma$-radiation by a medium follows the relation

$$I_x \propto e^{-\mu x} \, dx,$$

where $I_x$ is the intensity measured after radiation is attenuated by material with thickness $x$ and linear attenuation coefficient $\mu$. The linear attenuation coefficient is the product of the mass-attenuation coefficient $\mu/\rho$ and material bulk density $\rho$. In a homogeneous flat-bed geometry, the radiation reaching the detector is the integration of the contribution from layers with thickness $dx$ from the soil surface ($x = 0$) to infinite depth and is therefore expected to be proportional to the inverse of the linear attenuation coefficient of the bulk sediment:

$$I_{detector} \propto \int_0^{\infty} e^{-\mu x} \, dx = \left[ \frac{1}{-\mu} \, e^{-\mu x} \right]_{0}^{\infty} = \frac{1}{\mu} = \frac{1}{\rho \cdot \mu}.$$  (3.13)

When water is added to the sediment, the mass per unit volume (bulk density) of the sediment increases and consequently the initial intensity of the emitted radiation decreases (c.f. equation 3.3). Firstly, this leads to a change in detected radiation proportional to the change in water content (equation 3.6). Secondly, the attenuation characteristics of the sediment change, because the energy-dependent mass-attenuation coefficients of quartz differ slightly from those of water. For $E_\gamma < 0.1$ MeV, the mass-attenuation coefficient of quartz is higher than that of water (a factor 1.4 at 0.5 MeV, Berger et al., 2007) and between 0.1 and 3 MeV it is lower (factor 1.1 at 1.5 MeV). The net effect of these changes on spectral content is given in equation 3.11 (the ‘extra attenuation’). The extra attenuation is linear because attenuation is related to the mass of the medium. A twofold increase in the mass of water therefore means a twofold increase in the effect of changed attenuation coefficients.

The above equation can be adapted for a three-dimensional situation, but the resulting equations are not straightforward to solve for a single $\gamma$-ray, let alone for an entire spectrum. This again advocates the use of Monte-Carlo simulations for the type of assessments as done in this chapter.
Environmental spectra

The previous spectra were all for artificial, mono-energetic radionuclides. For practical application, spectra for $^{40}$K, $^{232}$Th, $^{238}$U and $^{137}$Cs were simulated for the PANDORA detector, including its broadening characteristics. Again, sediments were filled stepwise with water. The spectra were normalised to represent counts per 1 Bq kg$^{-1}$ in the quartz.$^2$

As expected the shape of the spectra did not change for various water contents, except for the region $< 0.1$ MeV. For practical purposes, the change in spectral shape below 0.1 MeV is not important. The region of interest (ROI) used in Full-Spectrum analysis for the PANDORA detector is 0.2 – 2.8 MeV, as lower-energy channels are unreliable due to a combination of electronic and other noise (Maučec et al., 2004a).

Spectral intensity was also calculated for the PANDORA ROI of 0.2 – 2.8 MeV. Also for the environmental radionuclides the effect of adding water is a linear decrease in intensity with a small extra effect due to the difference in mass-attenuation coefficients (see Figure 3.10). For the used region of interest, the small extra effect (correction factor $\theta$ from equation 3.11) is the same for the four radionuclides within their uncertainties, with an average value of 0.038 ± 0.002. The conclusion of this is that water content of sediment has a considerable influence on spectral intensity, but not on the shape. When sediment water content is known, a correction can be made.

![Figure 3.10. Spectral content of spectra (0.2 - 2.8 MeV) resulting from 1 Bq kg$^{-1}$ dry quartz, for four environmental radionuclides as a function of the mass fraction of water. The lines represent the expected spectral content according to equation 3.6. Statistical uncertainties in the simulations are smaller than the marker size.](image)

$^2$ The MCNPX results are given in counts per starting photon. To convert this to Bq, the results were multiplied by the average number of emitted $\gamma$-rays per decaying mother nuclide.
Handling water content in *in-situ* measurements

The simulation results show that the spectral intensity observed from sediment with constant dry activity concentrations largely follows the relation of equation 3.6, which describes the dilution of the activity concentrations. This means that any uncorrected detector response essentially scales with the wet activity concentration of the sediment.

When the water content of the measured sediment is known, the dry activity concentrations can be calculated using equation 3.6 or 3.11. Water content can be obtained from samples from the measurement site, by weighing them before and after drying (equation 3.5). The correction of equation 3.6 is valid for measurements irrespective of ROI and for all environmental radionuclides. It does however not take into account the ‘extra’ attenuation by water introduced by the difference in mass-attenuation coefficients of water and quartz. This introduces a systematic error in the dry activity concentrations of at maximum 7% for $w_d = 0.66$, the highest water content found in the samples. Equation 3.11 does take into account the extra attenuation, but the correction factor $\theta$ depends on the spectral region of interest and radionuclide, and may also depend on the type of minerals of which the sediment constitutes.

For some agricultural fields (Schimmack et al., 1998), water content varied between wilting point (condition in which plants cannot take up sufficient water anymore) and field capacity (water content when excess water has drained away), resulting in uncertainties in activity concentrations of only 10%. However, the observed range in water contents on Schiermonnikoog is much larger. If *in-situ* water content is unknown, the systematic error in dry activity concentrations on the island can amount to a factor three.

The effect of variations in water content within the view of the detector on the detector response could not be inferred from the simulations. Therefore the corrections should only be used for sediment with homogeneous water content. In the special case of underwater measurements, the sediment is always saturated with water. Bulk density and water content are then directly related and density can be used as proxy for water content (Venema and De Meijer, 2001; Van Wijngaarden et al., 2002a).

In this thesis, we will obtain activity concentrations by calibrating the standard spectra for the PANDORA detector to represent wet sediment activity concentrations. All measurement results are therefore initially given as $C_{j,wet}$. If water content is known and reasonably homogeneous, subsequently the values are corrected for water content in the sediment using equation 3.6.

3.3.5 Effective energy and $\gamma$-ray attenuation

The simulations in this thesis give the opportunity to compare the behaviour of the natural radionuclides within the soil. From these radionuclides, $^{40}\text{K}$ and $^{137}\text{Cs}$ each emit mono-energetic $\gamma$-rays, whereas the decay series from $^{232}\text{Th}$ and $^{238}\text{U}$ give rise to a complex spectrum. Attenuation is energy-dependent and therefore it is generally difficult to estimate the effective attenuation for a decay series. Because the shape of the spectra is virtually independent of porosity and water content, we can consider the
spectra as unit shapes of which only the intensity varies. Consequently, we determine the effective energy $E_{\text{eff}}$, which is a mono-energetic $\gamma$-line that has the same overall attenuation characteristics as the complete spectrum in a semi-infinite geometry. To derive $E_{\text{eff}}$, simulation results from the decay series are compared with those from the mono-energetic sources.

In Figure 3.11 the spectral contents of spectra from mono-energetic sources (0.5 – 2.5 MeV and $^{40}$K and $^{137}$Cs) and pure quartz sediment are plotted as a function of $\gamma$-ray energy (ROI = 0 – 3 MeV, non-broadened spectra and sediment with $\varepsilon = 0$ and $\rho = 2.65$ g cm$^{-3}$). The relation is nonlinear and was therefore described by the function $TC = aE_\gamma^b$, which secures zero spectral content at $E_\gamma = 0$. This gave $a = 4.341 \pm 0.006$ and $b = 0.355 \pm 0.002$. The effective energies for the decay series of $^{232}$Th and $^{238}$U were subsequently determined from their spectral content and this function (Figure 3.11). The effective energy is 0.62 MeV for the decay series of $^{232}$Th and 0.59 MeV for that of $^{238}$U. This is lower than the average $\gamma$-ray energy of each series when weighted by the branching ratio of every line (which is 0.78 for $^{232}$Th and 0.80 for $^{238}$U). The cause for that is that the mass-attenuation coefficients increase rapidly below 0.5 MeV (Berger et al., 2007). Lower energies are attenuated much stronger than higher energies, giving the lower energies relatively more weight. Comparing the effective energies of the environmental radionuclides shows that $^{40}$K emits on average the ‘hardest’ radiation ($E_\gamma = 1.46$ MeV).

Figure 3.11. Spectral content (per emitted $\gamma$-ray per kg sediment) as a function of $\gamma$-ray energy, fitted with the relation $TC = 4.341 \cdot E_\gamma^{0.355}$. The spectral contents are from simulations in which the sediment was modelled as pure quartz. Simulation uncertainties are less than 0.5%. The straight lines represent the spectral contents of the simulations with $^{232}$Th (solid line) and $^{238}$U (dashed line) and the corresponding effective energies.
3.5 Summary and conclusions

In this chapter we investigated the effect of porosity and water content on the activity concentrations derived from *in-situ* γ-ray spectra with Monte-Carlo simulations. We took into account the possible effect of γ-ray energy and based the simulated sediment on the range of conditions found in samples from a barrier island.

Simulations in the adjoint mode showed that a detector in first order ‘sees’ a constant mass and hence the detected sediment volume depends on soil bulk density. For typical soil bulk densities, the effective depth of detection is between 40 and 60 cm and the effective width between 160 and 260 cm. Additionally, this type of simulations prove to be a useful tool for avoiding erroneous effects in simulated spectra, by using adjoint simulations as a check on the required geometry size.

The simulations in the regular mode of MCNPX of the spectra for various values of porosity showed that spectral shape and intensity do not change. Thus the effect of sediment porosity alone on detected spectra is negligible.

Simulations for various values of water content reveal that the shape of the spectra does not significantly change as a function of water content. In contrast, spectral content decreases linearly proportionally with absolute water content. There is a small additional effect due to the difference in γ-ray attenuation coefficients in water and quartz. For the larger effect, a straightforward correction can be made if the absolute water content is known, which can be derived from samples. The correction is the same for all environmental radionuclides. In case water content is not known, uncertainties in dry activity concentrations may amount to a factor three for the observed range of sediment water contents.

From the spectral contents it was found that complex γ-ray spectra of $^{232}$Th and $^{238}$U show behaviour for absorption that is similar to mono-energetic γ-radiation with energies of 0.62 and 0.59 MeV, respectively. Despite the presence of higher energies in the spectra of $^{232}$Th and $^{238}$U, $^{40}$K is the source of the relatively most penetrating γ-radiation of the natural radionuclides.

The results from this chapter are used in following chapters of this thesis. In Chapter 4, the correction for water content will be applied to *in-situ* measurements of environmental γ-radiation. The effective attenuation characteristics and size of the contributing volume will be used in Chapter 5 for the determination of top-layer thickness from *in-situ* measurements.
Sensitivity of in-situ γ-ray spectra