Chapter 4

Mapping sediment characteristics through γ-radioactivity on a barrier island, Schiermonnikoog, the Netherlands

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Abstract
The natural γ-radioactivity of sediments (40K, 232Th and 238U) can potentially be used for sedimentology studies. In this chapter we study the variation in radioactivity on the barrier island of Schiermonnikoog (NL) and relate this to sediment properties to investigate possible applications. We measured in-situ radioactivity with two types of detector and analysed sediment samples on natural radionuclide activity concentrations and sediment grain size. The in-situ surveys show that variations in in-situ total count rate are related to geomorphology. From sample analysis, two groups of sediment could be identified on the island: coarse-grained (sand) sediment with low activity concentrations and fine-grained sediment with 2 to 3 times higher concentrations. Sediments consisting of a mixture of the two groups hardly occur. Using the radiometric characteristics (‘fingerprints’) of these groups and correcting for sediment water content, in-situ measurements of γ-radiation on the intertidal flats and beach were successfully converted to maps of sediment grain size. This shows that natural γ-radiation can be used for grain-size mapping in intertidal settings.
4.1 Introduction

Over the past two decades, a number of beaches and dunes of the Frisian Islands have been subject of studies to gain insight into sediment dynamics, using the radiometric fingerprints of sediments (De Meijer et al., 1990; De Meijer and Donoghue, 1995; De Meijer, 1998). The radiometric fingerprint of a sediment component is the representative combination of the activity concentrations of the natural radionuclides $^{40}$K, $^{232}$Th and $^{238}$U of this component (De Meijer et al., 1990; De Meijer and Donoghue, 1995; De Meijer, 1998). The activity concentrations of these natural primordial radionuclides are related to mineral type, provenance and grain size. Initially the studies focused on heavy minerals in beach and dune sands. Based on the difference of radiometric properties, the heavy minerals were found to form regions with similar fingerprints, often separated by rivers or inlets. Similar studies were done along the coasts of e.g. Brazil (Anjos et al., 2006), Albania (Tsabaris et al., 2007), Australia (De Meijer et al., 2001) and South Africa (De Meijer et al., 1997; Macdonald et al., 1997; Philander et al., 1999) and revealed provenances and large-scale transport of sediments along coasts.

The fingerprinting analyses were based on sediment samples measured using laboratory-based $\gamma$-ray detectors. The development of towed seabed $\gamma$-ray detectors, such as the MEDUSA detector (De Meijer et al., 1994; De Meijer, 1998), with enough sensitivity to map natural concentrations of environmental radionuclides in situ opened the possibility to map large areas of the seabed with a high density of data points (Venema and De Meijer, 2001; Van Wijngaarden et al., 2002a). The fingerprints emerging from these studies (typically considering a smaller geographical area than the previous studies) concern fractions based on grain size. Consequently it was possible to map large areas on the occurrence of sand and mud, with a much higher density of data points than was previously possible.

In this chapter we will combine the previous approaches and map sediment $\gamma$-radiation on the Dutch barrier island of Schiermonnikoog. We will determine if, and how, the radiometric technique can be applied in studies on barrier-island sedimentology, using both laboratory-based and in-situ $\gamma$-ray detectors. The beach and part of the dunes of Schiermonnikoog are known to exhibit variation in $\gamma$-radiation (De Meijer et al., 1989) and we will start with extending the measurements to the entire island. Secondly we will establish radiometric fingerprints by relating sediment radioactivity to other sediment properties such as grain size. We will also study the spatial distribution of the anthropogenic radionuclide $^{137}$Cs, which is expected to occur as the result of episodic atmospheric deposition. Because it emits $\gamma$-radiation in the same energy window as the natural radionuclides, it may be necessary to take its presence into account in in-situ measurements. Finally, we will map part of the intertidal flats and beach on in-situ $\gamma$-radiation and use the previously established relations to convert this to sediment properties. The chapter is explorative in nature and we will therefore mainly focus on large-scale patterns.
4.2 Methods

4.2.1 Study site
The study site of this chapter is the island of Schiermonnikoog, described in Chapter 2.

4.2.2 in-situ total radioactivity at the island scale
The total intensity of in-situ $\gamma$-radiation up to 3 MeV, without specifying the source nuclides, was measured using a hand-held Scintrex GIS-5 detector (see Chapter 2). In total 293 measurements were carried out between 2003 and 2006, covering the entire island. The measurement locations were selected to obtain an overall picture of the in-situ intensity variations in $\gamma$-radiation of the island, consisting of single scattered measurements, transects over representative gradients (dune arcs and salt marsh) and measurements on ‘hot spots’. Care was taken to select representative sites by first scanning the surroundings with the detector and avoiding locations that were obviously influenced by man, such as concrete bunkers, artificial dunes and most of the polder area. During measurements, the detector was directly placed on the soil. Counting times were at least 200 seconds. Counts were converted to counts per second (cps) and uncertainties were determined using counting statistics (equation 2.8). The cosmic and detector background were subtracted (section 2.2.5). All measurements were carried out in dry weather and at least three hours after the last rainfall, to avoid the contribution of $\gamma$-ray emitting radon progeny from precipitation. Statistical analyses were carried out with SPSS 14, MS Excel and Matlab.

The locations of the data points were determined with a Garmin GPS or, in a few cases when a GPS was not available, read from a map. In the field each measurement site was assigned to one of the following geomorphology classes: intertidal flats, beach, beach plain, green beach (a complex of vegetated beach, low primary dunes and depressions), dune, salt marsh, washover or polder (Figure 4.1).

The measurements were not all taken within a restricted time period. This might introduce a systematic uncertainty in the measurements, due to temporal variations in cosmic background, ambient temperature (leading to gain drift) and soil moisture content. To estimate the magnitude of this uncertainty, six measurements of 400 s were taken at a fixed site at different times of day in a time span of a few months in 2004. This gave a standard deviation of 9% on an average net total count rate of 28 cps. This uncertainty is larger than the uncertainty (generally less than 1.5%) arising from counting statistics of the individual measurements reported in this chapter. It is not known how representative this uncertainty is and therefore initially only the uncertainties resulting from counting statistics will be reported.

4.2.3 Laboratory measurements: radiometric fingerprinting
The relation between sediment radionuclide activity concentrations and grain-size distribution was derived from analysis of 62 sediment samples, taken at various sites on the island (Figure 4.1). The sample sites are representative for the island geomorphology and extremes in sediment mixtures. Therefore the samples were not taken in
a regular spatial pattern. Relatively many samples were taken at and around the salt marsh, the focus area of this thesis. Four to five sub-samples were randomly taken within 4 m², the depth depending on the depth of the sediment layer of interest. The samples were pooled, collected in plastic bags, sealed and taken to the laboratory where they were put in re-entrance Marinelli sample containers of 0.5 or 1.0 litre (types 133N and 233N). Of the total of 62 samples, 41 samples were sealed and left for three weeks, to re-establish secular equilibrium in the uranium series, and subsequently measured on an HPGe detector (Chapter 2). Five samples were measured as they were (including moisture) and without radon-tight sealing, and the remaining 16 samples were freeze-dried, sealed and then left for three weeks before measuring. The difference in the activity concentrations of the sealed and unsealed samples may only affect the measured $^{238}\text{U}$ concentrations. After the measurements on the HPGe the moist samples were dried and $C_{j,\text{dry}}$ of radionuclide $j$ was calculated following the method discussed in Chapter 3 (equations 3.5 and 3.6).

The grain-size distributions of the sediment samples were determined with a Coulter Malvern Laser-refraction device at NIOO-CEME in Yerseke (NL). Part of each sample was freeze-dried (which preserves grain structure) and sieved through a 1 mm mesh, which removed the occasional root or shell fragment. The analysis involved only a few grams of sample and resulted in the percentages of grains occurring in the mud ($<16 \ \mu\text{m}$ and $<63 \ \mu\text{m}$), very fine sand ($63 - 125 \ \mu\text{m}$), fine sand ($125 - 250 \ \mu\text{m}$) and medium fine sand fractions ($250 - 500 \ \mu\text{m}$). The percentage in the coarse sand fraction ($500 - 1000 \ \mu\text{m}$) could be determined from the fact that the fractions within a sample should sum to unity. The uncertainty in the grain-size fractions was estimated by replicating the measurement of three of the samples a number of times. This did not result in a consistent value of the uncertainty between grain-size classes and
samples, and therefore an uncertainty of 6% was chosen based on the range of the found uncertainties. This value does not include the systematic uncertainty introduced by taking only a small sub-sample of the sediment measured on the HPGe.

In line with earlier work (Greenfield et al., 1989; De Meijer, 1998; Van Wijngaarden et al., 2002b), we will investigate whether the sediment from Schiermonnikoog can be described by a mixture of two sediment components, each containing a characteristic set of radionuclide activity concentrations (i.e. its fingerprint). The activity concentration of sediment would then be given by:

$$C_j = \alpha_j C_{j,1} + (1 - \alpha_j) C_{j,2}.$$  \hspace{1cm} (4.1)

Quantity $\alpha_j$ represents the mass fraction of the sediment component with activity concentrations $C_{j,1}$ of radionuclide $j$, where the second sediment component has characteristic activity concentrations $C_{j,2}$. This expression can be rewritten as:

$$\alpha_j = \frac{C_j - C_{j,2}}{C_{j,1} - C_{j,2}}.$$ \hspace{1cm} (4.2)

The value of $\alpha_j$ is interpreted as the degree of mixing between the two groups, and can be calculated for each radionuclide. Because the value is standardised, the distributions of the radionuclides in the samples can then be easily compared.

### 4.2.4 In-situ radionuclide activity concentrations on the intertidal flats and beach

For the in-situ measurement of sediment activity concentrations, we used the PANDORA detector (Precision Agriculture Needed Detector Of RadioActivity, Van der Graaf et al., 2007), which is described in Chapter 2. The detector receives radiation from up to approximately 50 cm depth (Chapter 3).

The measurements were carried out on the eastern part of the island on the intertidal flats, beach plain and beach. These sites were chosen because they are the dynamic parts of the island and are relatively easy accessible. The detector was towed behind a vehicle (a small 4x4 John Deere diesel 'Gator') and was in continuous contact with the soil surface. Towing speed was 2 – 4 km h$^{-1}$ on the intertidal flats and 6 – 8 km h$^{-1}$ on the beach. The measurements were carried out between 30 March and 2 April 2004 and consisted of one track parallel to and at some distance from the salt-marsh edge and dune foot, and additional tracks on the intertidal flats. The spatial extent of the measurements was limited by the firmness of the substrate to support the vehicle, especially on the intertidal flats.

The spectra were integrated over 20 seconds and analysed using Full-Spectrum Analysis (FSA, see Chapter 2). Firstly, the analysis was carried out with standard spectra representing the response of the detector to an activity concentration of 1 Bq kg$^{-1}$ of a certain radionuclide, which is the traditional application. This analysis resulted in activity concentrations $C_j$. Secondly, in a novel application, the standard spectra were constructed for sediment components (Koomans, 2008). The standard spectra for the environmental radionuclides were created in Monte Carlo simulations.
with MCNPX (Pelowitz, 2005) and calibrated using a stationary PANDORA measurement at the intertidal flats and a sediment sample taken alongside.

To obtain dry activity concentrations, a correction for water content was necessary. Water content ranged from 0.17 to 0.19 in samples taken from the intertidal flats and beach, and therefore a correction was made for $w_a = 0.18$, using equation 3.6. An exception is the eastern sand flat where the water content was found to be only 0.04 and the correction was made accordingly. The uncertainties in the resulting activity concentrations range from 3.5% for $^{40}$K to 6 – 8% for $^{232}$Th and 8 – 10% for $^{238}$U. Within these uncertainties the in-situ activity concentrations from PANDORA agree well with activity concentrations from six sediment samples taken during the survey and subsequently measured on the HPGe detector.

Because of the presence of spectral drift in the electronics due to changes in ambient temperature, the net total count rate was reconstructed from the radionuclides after stabilising the spectra:

$$TC = \sum_{ij} C_j X_j(i). \quad (4.3)$$

Finally, the results were spatially interpolated using Kriging in the software package Surfer (version 8.00, Golden Software, 1999).

4.3 Results

4.3.1 In-situ total radioactivity at the island scale

The spatial pattern of in-situ $\gamma$-radiation, measured as total count rates with the Scintrex GIS-5 detector, was related to geomorphology (Figure 4.2 and Figure 4.3). Lowest count rates (10 – 40 cps) are found on the beaches and beach plains. Intermediate count rates (40 – 55 cps) occur on most dunes, the intertidal flats and on salt-marsh sites where the layer of fine-grained marsh deposits is thin. Highest count rates (55 – 80 cps) occur in some older dune sites landward of the present foredune (north-western dune area, area 1 in Figure 4.2) and on locations on the salt marsh where the fine-grained layer is thick (location 2). The highest count rate was measured locally on the beach spit at the northwest side of the island (location 3), where reddish sand grains indicated local enrichment in heavy minerals. A relatively high count rate from a dune site at the north-eastern part of the island (location 4) seems also to be related to enrichment in heavy minerals, as there was a band of dark sand grains on a recently eroded part of the dune.

The significance of the above differences was tested using multiple Mann-Whitney tests with a Bonferroni correction (e.g. Field, 2005) for the multiple comparison between groups (Table 4.2, polder and washover were excluded because of their very low number of data points). This leads to the following ranking: count rates are lowest on the beach, green beach and beach plains (range of 14 – 78 cps, including enrichment of heavy minerals). Count rates are higher on the intertidal flats (28 – 60 cps),...
Figure 4.2. Total γ-radioactivity on Schiermonnikoog, measured as total count rates with the Scintrex GIS-5 detector. Numbers refer to the discussion in the text.

Figure 4.3. Histograms of in-situ total count rates for various geomorphology.
which overlap with the dune group (20 – 73 cps). Finally, the salt marsh has on average highest count rates (28 – 67 cps), but the count rates partly overlap with those of the dune group. The latter overlap can be attributed to the locally high count rates in the older dunes and salt-marsh sites with a thin top layer.

Field observations indicate that for this island, geomorphology and sediment grain-size distribution are closely related. From the correlation between count rates and geomorphology it is therefore expected that sediment grain size will be related to in-situ radioactivity, except for sites with local enrichment of heavy minerals (which we will exclude from further analysis). However, it is possible that spatial variations in e.g. sediment water content affect the level of in-situ radiation (see Chapter 3). To eliminate this effect and define the relation between sediment grain size and γ-radiation, we will use dry activity concentrations from samples.

**Table 4.1.** Weighted average and standard deviation of count rates for the geomorphology units. N is the number of data points.

<table>
<thead>
<tr>
<th>geomorphology unit</th>
<th>N</th>
<th>mean (cps)</th>
<th>standard deviation (cps)</th>
</tr>
</thead>
<tbody>
<tr>
<td>beach plain</td>
<td>6</td>
<td>25</td>
<td>5</td>
</tr>
<tr>
<td>beach</td>
<td>21</td>
<td>25</td>
<td>12</td>
</tr>
<tr>
<td>green beach</td>
<td>8</td>
<td>28</td>
<td>6</td>
</tr>
<tr>
<td>washover</td>
<td>5</td>
<td>34.6</td>
<td>1.2</td>
</tr>
<tr>
<td>intertidal flats</td>
<td>38</td>
<td>36</td>
<td>7</td>
</tr>
<tr>
<td>dune</td>
<td>91</td>
<td>38</td>
<td>10</td>
</tr>
<tr>
<td>polder</td>
<td>3</td>
<td>40</td>
<td>13</td>
</tr>
<tr>
<td>salt marsh</td>
<td>121</td>
<td>42</td>
<td>9</td>
</tr>
<tr>
<td>total</td>
<td>293</td>
<td>37</td>
<td>11</td>
</tr>
</tbody>
</table>

**Table 4.2.** Significance values from non-parametric Mann-Whitney tests for differences between geomorphology units. Values marked with an asterisk are significant at the 0.05 level when corrected for the number of tests.

<table>
<thead>
<tr>
<th>intertidal flats</th>
<th>beach</th>
<th>dunes</th>
<th>salt marsh</th>
</tr>
</thead>
<tbody>
<tr>
<td>intertidal flats</td>
<td>-</td>
<td>&lt; 0.001*</td>
<td>0.202</td>
</tr>
<tr>
<td>beach</td>
<td>&lt; 0.001*</td>
<td>-</td>
<td>&lt; 0.001*</td>
</tr>
<tr>
<td>dunes</td>
<td>0.202</td>
<td>&lt; 0.001*</td>
<td>-</td>
</tr>
<tr>
<td>salt marsh</td>
<td>0.002*</td>
<td>&lt; 0.001*</td>
<td>0.032</td>
</tr>
</tbody>
</table>
4.3.2 Spatial pattern of $^{137}$Cs

The total count rate of the Scintrex GIS-5 detector is the sum of the pulses generated by all environmental radionuclides within a certain time interval. Of these radionuclides, the spatial distribution of the anthropogenic $^{137}$Cs is expected to be related to deposition history rather than to sediment grain size or mineral composition. To determine whether this is the case for Schiermonnikoog, we projected sample activity concentration of $^{137}$Cs on the east – west axis of the island (Figure 4.4). The samples were taken from sites with various geomorphology that have experienced various degrees of sediment dynamics since the deposition of $^{137}$Cs. The highest activity concentrations occur on the middle of the island close to the soil surface. At the eastern and western parts of the island only low activities are found, coinciding with locations on the beach (in general high sediment dynamics) and young marsh (developed after 1986). The overall pattern agrees with the east – west gradient in $^{137}$Cs deposition reported by CCRX (1986) on the basis of precipitation. Hence, we will continue the analysis with only the three natural radionuclides and correct for the presence of $^{137}$Cs if needed.

4.3.3 Radiometric fingerprinting

Correlations

The dry activity concentrations of the natural radionuclides ($^{40}$K, $^{232}$Th and $^{238}$U) from sediment samples are positively correlated having high correlation coefficients (Table 4.3 and Figure 4.5). For values less than 15 Bq kg$^{-1}$, the activity concentrations of $^{232}$Th and $^{238}$U are the same, but for values over 15 Bq kg$^{-1}$ $^{238}$U is lower than $^{232}$Th. A similar but less clear pattern occurs in the relations with $^{40}$K.
Table 4.3. Correlations (Pearson’s R²) between sample radionuclide activity concentrations (N = 62). All correlations are statistically significant at the 0.001 level.

<table>
<thead>
<tr>
<th></th>
<th>40K</th>
<th>232Th</th>
<th>238U</th>
<th>137Cs</th>
</tr>
</thead>
<tbody>
<tr>
<td>40K</td>
<td>1</td>
<td>0.86</td>
<td>0.96</td>
<td>0.21</td>
</tr>
<tr>
<td>232Th</td>
<td>0.86</td>
<td>1</td>
<td>0.92</td>
<td>0.22</td>
</tr>
<tr>
<td>238U</td>
<td>0.69</td>
<td>0.92</td>
<td>1</td>
<td>0.27</td>
</tr>
<tr>
<td>137Cs</td>
<td>0.21</td>
<td>0.22</td>
<td>0.27</td>
<td>1</td>
</tr>
</tbody>
</table>

Figure 4.5. Relations between 40K, 232Th, 238U from sediment samples. In the plot of 232Th and 238U (lower left) the 1:1 relation is given with the dashed line. Lower right panel: 3D plot of radionuclide concentrations indicating the three clusters resulting from cluster analysis based on the radionuclides (N = 62).
with $^{137}\text{Cs}$ are lower, which was expected based on the depositional pattern of $^{137}\text{Cs}$. The good correlation between $^{40}\text{K}$, $^{232}\text{Th}$ and $^{238}\text{U}$ indicates that the activity concentrations of the three radionuclides are not independent. Therefore one parameter such as the total count rate can probably be used for qualitatively describing radionuclide patterns, provided the activity concentration of $^{137}\text{Cs}$ is not playing a large role.

**Cluster analysis**

To further explore the radiometric characteristics of the sediments on Schiermonnikoog, a cluster analysis (average linkage, using the method of Euclidean distances) was applied to the sample activity concentrations of $^{40}\text{K}$, $^{232}\text{Th}$ and $^{238}\text{U}$. This resulted in two meaningful groups and two outliers (Figure 4.5). The first meaningful group of 31 samples has low activity concentrations and about a 1:1 relation between $^{232}\text{Th}$ and $^{238}\text{U}$ activity concentrations. The samples of this group originate from the beach, intertidal flats, dunes and the sand layer underlying the salt marsh, and consist therefore predominantly of coarse-grained (i.e. sandy) sediment. The activity concentrations in the second group are higher and $^{232}\text{Th}$ activity concentrations are on average 1.4 times higher than those of $^{238}\text{U}$. The 29 samples of this group are collected from the top layer of the salt marsh and are therefore predominantly fine-grained sediment. The two outliers were not included in further analysis.

**Radionuclides and sediment grain size**

An independent cluster analysis was carried out on the grain-size fractions of the samples. This resulted in three groups. The average grain-size distribution of the two largest groups is given in Figure 4.6. The first group is identical to the coarse-grained group from the previous cluster analysis. It is relatively well sorted and consists mainly of sand between 125 and 250 µm. The second group of the radionuclide clusters is

![Figure 4.6.](image-url)
subdivided into a fine-grained group (26 samples) and a mixed group (old marsh group, 3 samples). The fine-grained group has a broad grain-size distribution and contains mud (< 63 µm) as well as sand (> 63 µm). The samples of the mixed group were from an old marsh site in the middle of the island, less than 250 m apart, and contain at sight relatively large amounts of plant roots and organic material. These particular samples are considered not representative for the island and are therefore omitted from further analysis.

The combination of the two cluster analyses indicates that apart from local exceptions, sediment-wise Schiermonnikoog can be characterised by a coarse-grained and a fine-grained group. The grain size that marks the boundary between the two sediment groups was derived from a correlation between the cumulative abundance of grains smaller than a certain size in the samples and sample activity concentrations. The correlation coefficient $R^2$ per sediment size fraction for the individual radionuclides is given in Figure 4.7. Highest correlations occur if the sediment is divided into a fine-grained fraction of grains smaller than 125 µm and a coarse-grained fraction of grains larger than 125 µm: if the fraction between 125 and 250 µm is added to the fine-grained fraction, the correlation drops off sharply. The correlations are highest for $^{40}$K, $^{232}$Th and the $^{232}$Th/$^{238}$U ratio, followed by $^{238}$U. The ratios $^{40}$K/$^{232}$Th and $^{40}$K/$^{238}$U correlate poorly with sediment grain size.

The relations between the individual radionuclides and the percentage of fine-grained fraction (< 125 µm) are given in Figure 4.8. Generally, radionuclide activity concentrations increase with the percentage of grains smaller than 125 µm. However, within the two groups from the cluster analyses (indicated by different symbols), the scatter is such that a relation with grain size is extremely weak (for $^{40}$K and $^{232}$Th/$^{238}$U) or absent ($^{232}$Th and $^{238}$U). Therefore, although the existence of a linear relation between radionuclides and grain size cannot be excluded, the natural sorting of the sediment into two groups does not allow reliably establishing such a relation.

![Figure 4.7](image-url)  
**Figure 4.7.** Correlation coefficient $R^2$ (Pearson’s) of radionuclide activity concentrations with cumulative grain-size fractions ($N = 57$).
We will therefore interpret further in-situ measurements in terms of coarse-grained and fine-grained and use the weighted averages of the activity concentrations of the two groups as fingerprints (Table 4.4). The fingerprints differ significantly and amount to a factor 2.0 for $^{40}\text{K}$, 3.4 for $^{232}\text{Th}$ and 2.8 for $^{238}\text{U}$.

**Figure 4.8.** Radionuclide activity concentrations related to abundance of grains smaller than 125 µm in samples ($N = 57$). The coarse-grained and fine-grained groups resulting from cluster analysis are indicated.

**Table 4.4.** Characteristic radionuclide activity concentrations and ratios of the fine-grained and coarse-grained sediment on Schiermonnikoog (weighted averages and standard deviations).

<table>
<thead>
<tr>
<th>group</th>
<th>$N$</th>
<th>$^{40}\text{K}$ (Bq kg$^{-1}$)</th>
<th>$^{232}\text{Th}$ (Bq kg$^{-1}$)</th>
<th>$^{238}\text{U}$ (Bq kg$^{-1}$)</th>
<th>$^{232}\text{Th}/^{238}\text{U}$</th>
<th>$^{238}\text{U}$ from $^{232}\text{Th}$ and $^{232}\text{Th}/^{238}\text{U}$ (Bq kg$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>coarse-grained</td>
<td>31</td>
<td>$224 \pm 22$</td>
<td>$7 \pm 2$</td>
<td>$6 \pm 2$</td>
<td>$1.02 \pm 0.06$</td>
<td>$7 \pm 4$</td>
</tr>
<tr>
<td>fine-grained</td>
<td>26</td>
<td>$458 \pm 61$</td>
<td>$24 \pm 5$</td>
<td>$17 \pm 3$</td>
<td>$1.38 \pm 0.12$</td>
<td>$17 \pm 2$</td>
</tr>
</tbody>
</table>
Mixing coefficient

If sediment is a mixture of two grain-size groups, each characterised by its fingerprint, its activity concentration is given by equation 4.1. Using equation 4.2, we calculated for all samples the degree of mixing of the two groups ($\alpha_j$), using $C_{j,1}$ for the coarse-grained group and $C_{j,2}$ for the fine-grained group (Figure 4.9). Samples consisting mainly of fine-grained sediment then have values around zero and those with mainly coarse-grained sediment group around unity. The results indicate how well the two groups can be discerned on the basis of the radionuclides. The results confirm the results from the cluster analysis: for $^{40}\text{K}$ and $^{232}\text{Th}$, the two groups are clearly separated and sediments with intermediate values are hardly found. The sediment can less well be divided on the basis of $^{238}\text{U}$; the $^{232}\text{Th}/^{238}\text{U}$ ratio yields a better characterisation into.

![Histograms of mixing coefficient $\alpha_j$ in the samples based on $^{40}\text{K}$, $^{232}\text{Th}$, $^{238}\text{U}$ and the ratio $^{232}\text{Th}/^{238}\text{U}$ ($N = 57$).]
two groups. Therefore, we constructed the characteristic activity concentrations of $^{238}\text{U}$ of both groups from $^{232}\text{Th}$ through the $^{232}\text{Th}/^{238}\text{U}$ ratio (Table 4.4) and used these values in further analyses. Because the values of $\alpha_j$ are normalised and dimensionless, the histograms of the individual radionuclides were added up to get the average degree of mixing of the Schiermonnikoog sediments (lower panel of Figure 4.9, the $^{232}\text{Th}/^{238}\text{U}$ ratio was used instead of $^{238}\text{U}$). Again, the two groups are clearly separated and the distributions of both groups are consistent with a Gaussian around the group means. The spread in the fine-grained group is larger than in the coarse-grained group, indicating that there is more variation in the salt-marsh sediment than in the sand.

**Exploring variation within the sediment groups**

The variation within the two sediment groups may be intrinsic or due to external factors such as variations in organic matter content or grain-size independent spatial variations in mineral composition.

Variations in radionuclides might arise if sediment transport processes are not spatially uniform on the island. The presence of the tidal divide affects the tidal currents at the southern part of the island so that it may affect the sediment characteristics east and west of it. We divided the samples into groups east and west of the line $x = 21,3000$, the approximate location of the tidal divide (Figure 4.1). We restricted the comparison to samples from the salt marsh and intertidal flats as these are most probably directly affected by the tidal divide. We compared the $\alpha_j$’s of the individual radionuclides east and west of the tidal divide, separately for the coarse and the fine-grained group. Based on Student’s $t$-tests, none of the radionuclides is different on both sides of the tidal divide (Table 4.5, the group means are different when $p < 0.05$). This indicates that the position of the present-day tidal divide is not related to radiometric variations in sediment composition.

Organic matter was not removed from the sediment before sample analysis. Organic matter contains only very low quantities of radionuclides and thus effectively dilutes the activity concentrations of the sediment (Van Wijngaarden et al., 2002a; Madsen et al., 2005). For a subset of ten fine-grained salt-marsh samples, the organic matter mass content was determined through Loss On Ignition (3 hours on 500°C). The samples contain between 16% and 33% organic matter of the dry weight. Variations in organic matter content could therefore explain part of the scatter in activity concentrations within the fine-grained group, but this was not pursued further.

**Table 4.5.** Probabilities that the sediment east and west of the tidal divide is identical in radionuclide activity concentrations (expressed in $\alpha_j$), tested with Student’s $t$-test as the data within the groups are normally distributed.

<table>
<thead>
<tr>
<th></th>
<th>$\alpha_{^{40}\text{K}}$</th>
<th>$\alpha_{^{232}\text{Th}}$</th>
<th>$\alpha_{^{238}\text{U}}$</th>
<th>$\alpha_{^{232}\text{Th}/^{238}\text{U}}$</th>
<th>$N$ (west)</th>
<th>$N$ (east)</th>
</tr>
</thead>
<tbody>
<tr>
<td>coarse-grained group</td>
<td>0.07</td>
<td>0.93</td>
<td>0.67</td>
<td>0.14</td>
<td>12</td>
<td>9</td>
</tr>
<tr>
<td>fine-grained group</td>
<td>0.11</td>
<td>0.22</td>
<td>0.16</td>
<td>0.92</td>
<td>13</td>
<td>13</td>
</tr>
</tbody>
</table>
Box 4.1  Schiermonnikoog sediment compared to other marshes in the Wadden Sea

To determine whether the sediment of Schiermonnikoog is comparable with sediment elsewhere in the Dutch Wadden Sea, sediment from other salt marshes was analysed on radionuclide activity concentrations and grain-size distribution (from west to east: Texel: sheltered island marsh; Peazemerlannen: mainland marsh; Dollard: brackish mainland marsh). The activity concentrations of these eleven samples were plotted together with the Schiermonnikoog samples as a function of the percentage of grains < 125 µm (Figure 4.10). The samples from elsewhere contain somewhat more fines than the Schiermonnikoog samples, which is caused by the more sheltered conditions of the marshes from which they were taken. Further the samples follow roughly the same trend as the Schiermonnikoog samples, although the $^{232}$Th/$^{238}$U ratios are on the low side. Therefore we conclude that the radiometric properties, and therefore probably the associated mineralogy, of the fine-grained sediment does not vary notably within the Dutch Wadden Sea. This is consistent with the findings of Van Straaten (1954).

Figure 4.10. Sediment from Schiermonnikoog (open circles) compared to sediment from other locations in the Dutch Wadden Sea (filled circles), based on the relations from Figure 4.8.
4.3.4 in-situ radionuclide activity concentrations on the intertidal flats and beach

Radionuclides

The spectra recorded with the PANDORA detector on the intertidal flats and beach were converted into maps of total count rate and activity concentrations\(^1\), as shown in Figure 4.11. Total count rates (the sum of the contributions of the radionuclides) are lower on the beach and eastern sand flat than on the intertidal flats. On the intertidal flats, count rates are highest close to the salt-marsh edge (especially in the western part of the survey area) and around the tidal divide. There is also a hotspot of radiation in the western point of the survey area, probably related to the presence of notably muddy sediment close to a tidal channel leading to the ferry dam. The spatial distributions of \(^{40}\text{K}, {^{232}\text{Th}} \) and \(^{238}\text{U} \) largely follow the same pattern, but there are some differences between \(^{40}\text{K} \) on the one hand and \(^{232}\text{Th} \) and \(^{238}\text{U} \) on the other. The gradient from the intertidal flats to the salt-marsh edge is stronger developed for \(^{232}\text{Th} \) and \(^{238}\text{U} \) than for \(^{40}\text{K} \): the activity concentrations of \(^{232}\text{Th} \) and \(^{238}\text{U} \) are up to a factor two higher close to the salt-marsh edge than out on the intertidal flats. On the intertidal flats, the area around the tidal divide is notably high in \(^{40}\text{K} \). The area east of the tidal divide, including the eastern sand flat, is higher in \(^{40}\text{K} \) than the area to the west of it. In contrast to the individual radionuclides, the spatial pattern of \(^{232}\text{Th}/^{238}\text{U} \) is dominated by noise.

Spatial pattern of sediment groups

The dataset allows various ways to convert the measured spectra with the PANDORA detector into the mixing coefficient \(\alpha \) of the two sediment groups. First, we will determine \(\alpha \) based on the individual radionuclides from FSA. Secondly, \(\alpha \) will be extracted directly from the raw spectra by assigning a standard spectrum to each sediment group and conducting FSA with these standard spectra.

Although the sediment samples taken in the survey area fall into the coarse-grained sediment group, it is not \(a \text{ priori} \) known whether this is the case for the sediment of the entire survey area. Therefore we consider the sediment initially as a mixture of the two sediment groups, although at least part of the values of \(\alpha \) are expected to range between \(\alpha = 0.6 \) and \(\alpha = 1.3 \), i.e. the distribution around \(\alpha = 1 \) in Figure 4.9.

The distributions and spatial patterns of \(\alpha \) calculated from the individual radionuclides (equation 4.2) varied between the radionuclides (Figure 4.12 A – E). This is not surprising as the spatial patterns of the radionuclides are not identical and the transformation into \(\alpha \) is a linear one. The results from \(^{40}\text{K} \) assign all measured locations to the coarse-grained sediment group, whereas \(^{232}\text{Th} \) and \(^{238}\text{U} \) indicate that part of the

---

\(^1\) Initially the standard spectrum of \(^{137}\text{Cs} \) was included in the analysis, to correct for the presence of this radionuclide if necessary. The spatial pattern of \(^{137}\text{Cs} \) is noisy and dominated by uncertainty (Figure 4.11). Still the weighted average on the intertidal flats (0.57 ± 0.02 Bq kg\(^{-1}\)) is of the same order of magnitude as sample values (0.42 ± 0.04 Bq kg\(^{-1}\)). Leaving \(^{137}\text{Cs} \) out of the analysis does not significantly change the activity concentrations of the other radionuclides. It is therefore not necessary to take into account depositional variations in \(^{137}\text{Cs} \) on the intertidal flats and beach of Schiermonnikoog.
area consists of the fine group. The total count rate gives an intermediate distribution. The distribution of the values from the $^{232}\text{Th}/^{238}\text{U}$ ratio is centred around $\alpha = 1$, but is very broad. Part of the spread in the values of $\alpha_j$ can be attributed to the uncertainty in the activity concentrations from FSA, which is propagated when calculating $\alpha_j$. The uncertainties in $^{40}\text{K}$ are, due to the higher values of these activity concentrations, lower than those in $^{232}\text{Th}$ and $^{238}\text{U}$. This leads to a narrow distribution for $^{40}\text{K}$, wider ones for $^{232}\text{Th}$ and $^{238}\text{U}$ and an even wider one for the ratio $^{232}\text{Th}/^{238}\text{U}$.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Bead</th>
<th>Dunes</th>
<th>Polder</th>
<th>Ferry Dam</th>
<th>Intertidal Flats</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total net count rate</td>
<td>400</td>
<td>360</td>
<td>300</td>
<td>240</td>
<td>200</td>
</tr>
<tr>
<td>$^{40}\text{K}$</td>
<td>275</td>
<td>225</td>
<td>200</td>
<td>175</td>
<td>150</td>
</tr>
<tr>
<td>$^{232}\text{Th}$</td>
<td>25</td>
<td>20</td>
<td>15</td>
<td>10</td>
<td>5</td>
</tr>
<tr>
<td>$^{238}\text{U}$</td>
<td>20</td>
<td>15</td>
<td>10</td>
<td>5</td>
<td>0</td>
</tr>
<tr>
<td>$^{137}\text{Cs}$</td>
<td>2</td>
<td>1</td>
<td>0.5</td>
<td>0.2</td>
<td>0</td>
</tr>
<tr>
<td>$^{232}\text{Th}/^{238}\text{U}$</td>
<td>1.50</td>
<td>1.25</td>
<td>1.00</td>
<td>0.75</td>
<td>0.50</td>
</tr>
</tbody>
</table>

Figure 4.11. Radionuclide activity concentrations on part of the intertidal flats and beach of Schiermonnikoog, measured \textit{in situ} with the PANDORA detector. The thick solid line indicates the transition between intertidal flats and salt marsh at the time of the measurements. Data points (dots) are given in the lower panel, together with the topography of the area.
The second method to determine $\alpha$ from in-situ $\gamma$-ray measurements is based on a method used at Medusa Explorations BV (Koomans, 2008). In this 'two sediment standard spectra' method, a standard spectrum of each group is constructed from the standard spectra from $^{40}$K, $^{232}$Th and $^{238}$U and the fingerprints from sample analysis ($C_j$, Table 4.4). The measured spectra are analysed with these standard spectra, using

$$S(i) = \alpha \cdot \sum_j C_{j,1} X_j(i) + \beta \cdot \sum_j C_{j,2} X_j(i) + Bg(i), \quad (4.4)$$

denoting the fraction of the coarse-grained group, $\alpha$, and that of the fine-grained group, $\beta$. These fractions relate to the wet sediment and were corrected for water content using the water content from sediment samples, as before. The histogram of obtained $\alpha$'s at all measurement points is very broad and does not follow the expected pattern (Figure 4.12 F): from this analysis it would appear as if large parts of the intertidal flats and beach consist of salt-marsh sediment. A possible explanation for this erroneous result is that the standard spectra are very similar in shape, as both contain peaks for $^{40}$K, $^{232}$Th and $^{238}$U in only slightly different ratios. This increases the uncertainty in the analyses results.

To avoid problems with non-orthogonal spectra in least-squares fitting in FSA, equation 4.4 was rewritten in terms of $\beta (= 1 - \alpha)$ and with one variable standard spectrum only:

$$S(i) = (1 - \beta) \cdot \sum_j C_{j,1} X_j(i) + \beta \cdot \sum_j C_{j,2} X_j(i) + Bg(i) \quad (4.5)$$

In FSA the spectrum is analysed with $(\sum C_{j,1} X_j + Bg)$ as a background spectrum and $\sum((C_{j,2} - C_{j,1}) X_j)$ as a standard spectrum with $\beta$ as parameter to be deduced. This approach incorporates the correlation between the radionuclides and effectively reduces the degree of freedom in the fitting. The analysis assigns most data points to the coarse-grained group, as expected, and the histogram and spatial pattern resembles that of the analysis with the total count rates (Figure 4.12 G).

The above results depend strongly on the used analysis technique. The patterns that agree best with field observations are those with $\alpha$ obtained from $TC_{net}$, $^{40}$K and the approach with one sediment standard spectrum, which assign the majority of the sediment in the survey area to the coarse-grained group. These $\alpha$-values are therefore probably most suited to convert into large-scale maps of sediment composition.

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2 The route with $\beta$ is necessary because when using $\alpha$ in the analysis, negative standard spectra would result, which the analysis program MPA cannot handle. Additionally, for computational reasons the correction for water content was included in the standard spectra, using the sample-based water content.
Chapter 4

A radionuclide-based α: $T_{C_{\text{net}}}$

B radionuclide-based α: $^{40}\text{K}$

C radionuclide-based α: $^{232}\text{Th}$

D radionuclide-based α: $^{238}\text{U}$

E radionuclide-based α: $^{232}\text{Th}/^{238}\text{U}$
Conversion into sediment fractions
In sedimentology and ecology, sediment is traditionally characterised in terms of the percentage of sediment smaller than a certain grain size. This distinction is generally taken 63 µm, but the natural distinction between the two groups in the sediment of Schiermonnikoog lies at 125 µm. The percentage of grains smaller than 125 µm of each sediment group (\(x_1\) for the coarse-grained sediment and \(x_2\) for the fine-grained sediment) is known from the grain-size distribution of Figure 4.6, so that the patterns of \(\alpha_j\) can be converted into the percentage of grains smaller than 125 µm (\(x\)) using

\[
x = \alpha_j \cdot x_1 + (1 - \alpha_j) x_2 .
\]

We chose to convert \(\alpha\) of the total count rates (Figure 4.12 A) and the single sediment standard spectrum (Figure 4.12 G) into maps of percentage grains smaller than 125 µm (Figure 4.13). The resulting spatial patterns are almost identical. The difference between the methods is 1 – 3 % on a range of 5 – 50 % grains smaller than 125 µm. Compared to samples taken during the survey, most PANDORA values were between a factor one and five overestimated.

Based on the maps, the eastern sand flat consists only of sand and the beach is predominantly sandy with low percentages of fine-grained sediment. The intertidal

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**Figure 4.12.** Histograms and spatial pattern of sediment mixing coefficient \(\alpha\), based on a number of different analysis methods of spectra, collected *in situ* with the PANDORA detector (see text for explanation). Data points less than 2 m apart were omitted to prevent distortion of the histograms by stationary measurements (\(N = 1250\)).
flats consist of sand with low to intermediate amounts of fine-grained sediment. There is a gradient on the intertidal flats that may be attributed to hydrodynamics: most fine sediment is found close to the salt-marsh edge and especially around the convex part of the salt-marsh edge, where an older part of the marsh is growing southwards. Further, the amount of fine-grained material is relatively high around the tidal divide (c.f. Figure 4.1) and close to the ferry dam in the west of the survey area. The amount of fine-grained sediment is comparable east and west of the tidal divide. Some small-scale variations may be attributed to measurement uncertainty and the fact that the survey was done at four consecutive days, but nevertheless the large-scale pattern is clear.

4.4 Discussion

4.4.1 Sediment characterisation
The patterns of in-situ radioactivity that we measured with the Scintrex GIS-5 detector are largely consistent with the surveys done by De Meijer et al. (1989) on the beach and parts of the dunes of Schiermonnikoog. The latter were done 15 years earlier, indicating the robustness of the method.
The grain-size patterns from the survey with the PANDORA detector coincide well with maps based on sediment samples from the intertidal flats, as far as the used parameters (< 16 µm or < 63 µm vs. our 125 µm), sample depth (max. 10 cm vs. our ~ 50 cm) and sample density of these surveys allow (Rijkswaterstaat RIKZ Haren, 1998; Zwarts, 2004). The difference between the results from the samples and in-situ measurements is not clear. However, the mud percentages were generally low, so that uncertainties are relatively large.

The relation between grain size and radionuclide activity concentrations did not follow the expected linear relation (cf. Van Wijngaarden et al., 2002b). The fact that the fine-grained group contains small amounts of sand indicates that a linear relation may be present, but the absence of sediment with intermediate characteristics only allows the identification of two radiometric distinctive sediment groups. Surprisingly, the radionuclides were related to the fraction < 125 µm rather than the more commonly used < 63 µm. In the Schiermonnikoog sediment the abundance of the fractions of < 16 µm, 16 – 63 µm and 63 – 125 µm are positively correlated (whereas the correlation of these fractions is negative with the fractions > 125 µm), probably as a result from local transport processes. Any distinction based on regression will then be naturally assigned to a division at 125 µm. For a better understanding of the relation between radionuclides and grain size, more detailed measurements such as sieving and mineral identification would be necessary. In addition, removing organic matter before analysis may improve the relation.

To place the radiometric fingerprints from Schiermonnikoog into a broader geographical context, we compared them with radiometric analysis of samples of previous work from the Dutch marine environment (Table 4.6). The coarse-grained group corresponds well with earlier samples from Schiermonnikoog, sediment from the Western Scheldt (SW Netherlands) and sand from dredge spoil dump site Loswal (in the North Sea near the Hague). The activity concentrations of the coarse-grained group are higher than in the light minerals and sand from Bergen (Dutch west coast) and the Dutch Frisian island of Texel, and lower than in sand from Haringvliet (a former estuary in SW Netherlands). The activity concentrations are clearly different from the samples from the adjacent island of Ameland. Overall, the sand shows variation of a factor two to three in natural radionuclide content. The fine-grained group of Schiermonnikoog has lower characteristic activity concentrations than mud from Loswal and Haringvliet. This may be related to differences between sediment from marine and fluvial origin and/or the difference in analysis methods and grain size boundaries (< 63 µm) compared to our study. Within the area of the Wadden Sea the fine-grained sediment has similar radiometric properties (Box 4.1). This is in line with the observed uniform mineral composition of the mud fraction along the entire Dutch coast (Van Straaten, 1954).
Table 4.6. Literature overview of radiometric fingerprints of fine and coarse fractions of sediment in the Dutch marine environment. Schiermonnikoog, Ameland and Texel are Dutch barrier islands; Bergen is a site on the mainland coast; the sediment from dumpsite Loswal originates from dredge spoil from the harbour of Rotterdam and the seafloor close to The Hague; Haringvliet is a former estuary and the Western Scheldt an actual estuary, both in SW Netherlands.

<table>
<thead>
<tr>
<th>Source</th>
<th>location</th>
<th>method</th>
<th>fraction</th>
<th>(40\text{K} ) (Bq kg(^{-1}))</th>
<th>(232\text{Th} ) (Bq kg(^{-1}))</th>
<th>(238\text{U} ) (Bq kg(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>coarse-grained sediment</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Venema and De Meijer, 2001</td>
<td>Loswal</td>
<td>sieve and subsequently HPGe fraction analysis</td>
<td>sand (63 - 2000 µm)</td>
<td>213 ± 13</td>
<td>5.8 ± 0.2</td>
<td>5.2 ± 0.2</td>
</tr>
<tr>
<td>De Meijer, 2001</td>
<td>Malvern &amp; HPGe sand (63 - 2000 µm)</td>
<td>254 ± 6</td>
<td>5.9 ± 0.4</td>
<td>6.5 ± 0.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nederbragt and Koomans, 2005</td>
<td>Western Scheldt–Oostgat</td>
<td>unknown sand</td>
<td>sand sample</td>
<td>212</td>
<td>5</td>
<td>6</td>
</tr>
<tr>
<td>Van Wijngaarden et al., 2002b</td>
<td>Haringvliet</td>
<td>Malvern &amp; HPGe fraction analysis</td>
<td>sand (&gt;63 µm)</td>
<td>-</td>
<td>9.7 ± 0.9</td>
<td>9.3 ± 0.9</td>
</tr>
<tr>
<td>De Meijer and Donoghue, 1995</td>
<td>Schiermonnikoog</td>
<td>HPGe</td>
<td>sand sample (current coarse-grained group)</td>
<td>212 ± 6</td>
<td>5.2 ± 0.3</td>
<td>5.0 ± 0.2</td>
</tr>
<tr>
<td></td>
<td>Ameland</td>
<td>HPGe</td>
<td>sand sample (current coarse-grained group)</td>
<td>172.5 ± 1.3</td>
<td>9.4 ± 0.5</td>
<td>9.3 ± 0.2</td>
</tr>
<tr>
<td>Oost, 1998</td>
<td>Schiermonnikoog</td>
<td>bromoform + HPGe</td>
<td>light minerals in sand</td>
<td>210 – 220</td>
<td>appr. 2</td>
<td>appr. 2</td>
</tr>
<tr>
<td>De Meijer et al., 1990</td>
<td>Texel</td>
<td>bromoform + HPGe</td>
<td>light minerals in sand</td>
<td>148.4 ± 1.2</td>
<td>3.15 ± 0.18</td>
<td>2.87 ± 0.10</td>
</tr>
<tr>
<td></td>
<td>Bergen</td>
<td>bromoform + HPGe</td>
<td>light minerals in sand</td>
<td>79.5 ± 1.7</td>
<td>3.4 ± 0.2</td>
<td>3.4 ± 0.1</td>
</tr>
<tr>
<td>current</td>
<td>Schiermonnikoog</td>
<td>HPGe + Malvern, 2 groups</td>
<td>coarse (&gt;125 µm)</td>
<td>220 ± 20</td>
<td>7 ± 2</td>
<td>6 ± 2</td>
</tr>
<tr>
<td>fine-grained sediment</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Venema and De Meijer, 2001</td>
<td>Loswal</td>
<td>sieve and subsequently HPGe fraction analysis</td>
<td>clay (&lt;16 µm)</td>
<td>540 ± 30</td>
<td>32.4 ± 1.1</td>
<td>31.2 ± 1.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>silt (16 - 63 µm)</td>
<td>340 ± 20</td>
<td>28.5 ± 1.0</td>
<td>29.7 ± 0.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>mud (&lt;63 µm)</td>
<td>440 ± 30</td>
<td>29.7 ± 1.0</td>
<td>33.5 ± 1.1</td>
</tr>
<tr>
<td>Van Wijngaarden et al., 2002b</td>
<td>Haringvliet</td>
<td>Malvern &amp; HPGe fraction analysis</td>
<td>mud (&lt;63 µm)</td>
<td>-</td>
<td>45.6 ± 1.9</td>
<td>46.2 ± 1.9</td>
</tr>
<tr>
<td>current</td>
<td>Schiermonnikoog</td>
<td>HPGe + Malvern, 2 groups</td>
<td>fine (&lt;125 µm)</td>
<td>460 ± 60</td>
<td>24 ± 5</td>
<td>17 ± 2</td>
</tr>
</tbody>
</table>
4.4.2 Application

The results of the in-situ survey on the intertidal flats show that, apart from local enrichments with heavy minerals, the sediment on Schiermonnikoog can be characterised using just one parameter (total count rates or mixing coefficient \( \alpha \) from FSA). The reason for that is the overall good correlation between the natural radionuclides. There are slight differences between the spatial patterns of the radionuclides on the intertidal flats, but the correlation dominates on the spatial scale and sensitivity level of this survey, so that small variations in the content of fine-grained sediment can be detected.

In this study we applied two types of detector for in-situ measurements. The Scintrex GIS-5 detector is lightweight and easy to handle, so that rough terrain can be surveyed. However, it does not have logging capabilities and although it allows selecting a couple of energy windows, it is not capable of distinguishing between all environmental radionuclides. In contrast, the PANDORA detector is capable of measuring the activity concentrations of the individual radionuclides. Its weight (the entire detector system plus power supply weighs 80 kg) limits its use to terrain that is accessible with a vehicle or boat and does not suffer damage from that. Due to the presence of vegetation and the generally soft soil, surveying the entire salt marsh of Schiermonnikoog with this detector was therefore not feasible. Which detector is most suitable for a specific survey depends on the requirements of the survey: do total count rates give enough information or is information on the individual radionuclides necessary? This is preferably determined from a pilot study, involving sample analysis on the correlation between radionuclides and the variation in the presence of \(^{137}\)Cs. Recently e.g. Paridaens (2006) and Plamboeck et al. (2006) introduced detectors that can be carried, automatically logged and that are capable of distinguishing between radionuclides. Using such detectors, would be a valuable improvement for studies on salt marshes if their sensitivity is sufficient.

Sediment water content on Schiermonnikoog is variable in time and space, which will affect measured in-situ radioactivity. The fine-grained sediment with relatively high radioactivity has in general large pore volumes and the coarse-grained low-activity sediment smaller pore volumes (see Chapter 3). In case the sediment is saturated with water, the reduction in detector response will be larger for the fine-grained sediment than the coarse-grained sediment, thereby reducing the difference between the two groups. In case the in-situ water content is known, this effect can be corrected. The agreement between in-situ PANDORA measurements and sample values found in this chapter shows that this correction is effective in practice.

It was not possible to correct the Scintrex measurements, because for these measurements sediment water content was largely unknown. The magnitude of the obscuring of total count rates by variable water contents can be inferred from the wet activity concentrations of the samples used for fingerprinting (Figure 4.14). In wet conditions, the two sediment groups can only be partly distinguished based on \(^{232}\)Th, and hardly on \(^{40}\)K and \(^{238}\)U. Only the \(^{232}\)Th/\(^{238}\)U ratio does not change. The previously reported 9 % uncertainty in Scintrex measurements over time may partly be
introduced by variations in water content. Therefore, care should be taken when surveying areas that contain both sediment groups and are subject to large spatial and temporal variations in water content. Nevertheless a pattern can be identified in radiometry correlated to geomorphology, indicating that the effect of the variations in water content is slightly smaller than that of variations in sediment radioactivity.

**Technical improvements of the radiometric method**

The experiments from this chapter led to some practical improvements for working with a PANDORA-type detector. Firstly, the Monte-Carlo derived spectra needed very little correction for intensity, provided the standard spectra represented activity concentrations in the wet sediment. If such standard spectra are used, in principle only one set of standard spectra is necessary as long as the geometry does not change. The analysis then yields wet activity concentrations \( C_{j,wet} \). Afterwards a correction can be made for the right sediment water content to derive the dry activity concentrations \( C_{j,dry} \), equations 3.6 and 3.11), which should in principle be applicable with various detector types.

In Full-Spectrum Analysis, standard spectra are regarded as orthogonal eigenfunctions. If this assumption is not fulfilled, the analysis results are less reliable. Unfortunately, this is the case for the environmental radionuclides. Reducing the degrees of freedom greatly improved the quality of the analysis results. In the first
place, this was done by using a constant cosmic background spectrum, as opposed to a scalable one as done in earlier studies (Van der Klis, 2002; Van der Graaf et al., 2007). This optimisation step was not explicitly shown in the methods, but was a choice made during data analysis. Secondly, adapting the analysis involving standard spectra representing sediment groups, strongly improved the analysis results.

4.5 Summary and conclusions

*In-situ* and laboratory measurements of sediment γ-radiation revealed that spatial variations in environmental γ-radiation on the barrier island of Schiermonnikoog amount to a factor eight. The variations in the natural radionuclides ($^{40}$K, $^{232}$Th and $^{238}$U) are strongly positively correlated and are largely related to sediment grain size. Variations in $^{137}$Cs are related to depositional patterns, modified by sedimentation history. Consequently, only the natural radionuclides are suited for characterising sediment type. Total count rates can be used as an indicator for sediment composition as long as the spatial variations in $^{137}$Cs are not too large.

Apart from some local deviations, the sediment on Schiermonnikoog falls into two groups: fine-grained sediment with high natural radioactivity and sand with low radioactivity. The distinction between the groups lies at a grain size of 125 µm. Mixing of the two groups occurs, but intermediate values are seldom found.

The *in-situ* radioactivity related to the upper 50 cm of sediment on the intertidal flats and beach could be successfully converted into sediment grain size. This was aided by the correction of *in-situ* measurements for sediment water content, established in Chapter 3. In this process we found that uncertainties in radionuclide concentrations and sediment parameters resulting from non-orthogonal standard spectra in Full-Spectrum Analysis can be effectively reduced by reducing the degrees of freedom in the analysis.

In case sediment water content was not known, the radiometric difference between the two sediment groups was partly obscured. Still, variations in γ-radiation are mostly large enough to obtain an estimation of grain-size patterns with simple total counts measurements.

In the next chapter we will apply the radiometric method on the salt marsh, where the two sediment groups occur in a layered configuration. Using the radiometric difference between the groups, our aim is to determine sediment layer thickness from *in-situ* radioactivity.