Chapter 2

Methods and study sites

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The Scintrex GIS-5 detector standing on the salt marsh of Schiermonnikoog (left) and the small soil corer (right).
2.1 Introduction

This chapter describes the measurement methods and field sites that are relevant to this thesis. As methods based on the detection of γ-radiation emitted by naturally occurring radionuclides play a major role in this thesis, the chapter starts with the introduction of γ-radiation, the way it interacts with matter and how it is detected and quantified. I describe three types of detector that are used in this thesis and the analysis methods adopted. Monte-Carlo simulations are currently an important tool for quantitatively describing radiation transport and play an important role in data analysis. Hence, their principle and application in this thesis are presented. In the second part of the chapter I describe the non-radiometric measurements of this thesis such as sediment layer thickness, grain-size distribution and soil elevation. The experimental work was carried out at several study sites, which will be introduced in the last part of the chapter.

2.2 Environmental γ-radioactivity and its detection

2.2.1 Radioactivity

An atom consists of a nucleus made up of protons and neutrons, surrounded by a cloud of electrons. The number of protons in the nucleus is expressed in its atomic number $Z$. Mass number $A$ represents the combined number of protons and neutrons. Nuclei are depicted by their element name and mass number, and sometimes atomic number: $^A_ZX$. Some atomic nuclei are unstable and will disintegrate (decay), by the emission of ionising radiation. These radioactive nuclei are called radionuclides. In this thesis, only γ-radiation is of relevance.

In γ-decay, the nucleus is in an excited state and emits a γ-ray (photon) to reach a lower energy state. Gamma-radiation is electromagnetic radiation and does not affect the mass number or atomic number of the atom. The energy of γ-radiation is usually well-defined and corresponds to the energy difference between states of a nucleus. It is therefore typical for a radionuclide and can be used to identify it. Gamma-rays have high penetrating power and for shielding against γ-radiation, thick slabs of material are needed, such as lead or concrete. The unit to express the energy of γ-radiation ($E_\gamma$) is electronvolt (eV), corresponding to $1.6 \cdot 10^{-19}$ J. In this thesis, mostly keV or MeV is used.

The activity $A$ of a radionuclide is the number of decaying nuclei ($N$) per unit time ($t$):

$$A = - \frac{dN}{dt}. \quad (2.1)$$

The unit of activity is becquerel (Bq), defined as the amount of a certain radionuclide in which, on average, one disintegration occurs per second. The rate at which a radionuclide decays is governed by its decay constant $\lambda$ (s⁻¹):
\[
\frac{dN}{dt} = -\lambda N. \tag{2.2}
\]

Every radionuclide has its own decay constant. The number of nuclei of a certain radionuclide at time \( t \), \( N(t) \), compared to the initial number \( N_0 \), is derived after integration of equation 2.2:

\[
N(t) = N_0 e^{-\lambda t}. \tag{2.3}
\]

The decay constant is related to the half-life time \( t_{1/2} \), which is the time it statistically takes until half of the initial amount of a certain radionuclide is left. From equation 2.3 follows that

\[
t_{1/2} = \frac{\ln 2}{\lambda}. \tag{2.4}
\]

The radioactivity of sediments is generally expressed as activity concentration \( C \), which is the activity per unit mass (Bq kg\(^{-1}\)).

Some radionuclides have several decay modes, each with its own decay probability and decay product. Sometimes an unstable atom decays into an unstable decay product. In that case the decay continues until a stable nucleus is reached. Such a chain of decaying nuclei is called a decay series. If the decay products have half-life times shorter than the original nuclides and if no decay products are removed from the system, secular equilibrium develops. This is the situation where for each nuclide of a decay series the number of decaying nuclei equals the number of formed nuclei. From equations 2.1 – 2.3 can be derived that in secular equilibrium the activities of all radionuclides in the decay series are equal. Therefore under these conditions the activity of the parent nuclide can be derived from the activity concentrations of the decay products, even if the parent nuclide itself is not readily detectable.

### 2.2.2 Environmental \( \gamma \)-radiation

As discussed in Chapter 1, there are several radionuclides in the environment that can be used for studies on sediment transport and sedimentology. These are described in this section. Of the anthropogenic radionuclides, \( ^{137}\text{Cs} \) is most relevant to this thesis, as it is relatively abundant and emits \( \gamma \)-radiation in the same range as the natural radionuclides.

\( ^{40}\text{K} \)

The main decay modes of \( ^{40}\text{K} \) are \( \beta^- \)-decay to \( ^{40}\text{Ca} \) and electron capture to an excited state of \( ^{40}\text{Ar} \). The latter decays to its ground state by the emission of a \( \gamma \)-ray of 1.461 MeV, which happens in 10.67% of all decays (Figure 2.1). The half-life time of \( ^{40}\text{K} \) is \( 1.3 \cdot 10^9 \) a.
232Th and 238U

The radionuclides 232Th and 238U have half-life times of 14 · 10^9 and 4.5 · 10^9 a, respectively. These radionuclides do not decay to a stable isotope in one step, but give rise to decay series (Figure 2.2). For 232Th it takes ten steps to reach stable 208Pb, with 346 possible γ-ray emissions. The decay of 238U takes 16 steps to reach 206Pb, with 458 possible γ-rays. The average number of emitted photons per decaying nuclide equals 2.628 for 232Th and 2.197 for 238U. Not all nuclides of the series emit γ-radiation, and the detection of thorium and uranium depends on γ-rays emitted by some of their decay products (Figure 2.2). The most important γ-rays for 232Th are the 0.58 and 2.61 MeV transitions from 208Tl, and for 238U the 0.61, 1.12, 1.74 MeV γ-rays from 214Bi. All decay products have half-life times shorter than 232Th and 238U so that in closed systems secular equilibrium develops. Secular equilibrium in the decay series of 238U can be distorted by the escape of 222Rn. This radionuclide has a half-life time of 3.8 days and is an inert gas, allowing it to move out of the system, for example the soil. Decay products of radon once escaped from the soil can be deposited again by precipitation. These processes affect the activities of the radon decay products in the soil and at the soil surface. The main γ-rays that are used for the detection of 238U are emitted by decay products of 222Rn. Therefore, the possibility of radon escape should be taken into account when detecting 238U. In a laboratory setting the escape can be prevented by sealing samples in radon-proof containers and leaving them for some time, to establish secular equilibrium. Generally three weeks is considered to be sufficient. Another radionuclide that can cause a break in secular equilibrium in the uranium series is 226Ra (t1/2 = 1600 a), which is soluble in water. Radon also appears in the decay series of 232Th, but its isotope, 220Rn, has a half-life time of 55.6 s which is too short for significant escape.

137Cs

The anthropogenic radionuclide 137Cs has a half-life time of 30 a and decays to an excited state of 137Ba followed by the emission of 0.662 MeV γ-radiation, with an emission probability of 0.851. As mentioned in Chapter 1, the main sources of 137Cs are releases into the atmosphere by atomic bomb testing around 1960 and the
Chernobyl nuclear-reactor accident in 1986. Subsequently it was predominantly deposited on the Earth's surface by rain. The maximum deposition resulting from atmospheric bomb testing was in 1963. The deposition resulting from the Chernobyl accident was spatially heterogeneous; Figure 2.3 gives the distribution of deposition in the Netherlands. The amount of $^{137}\text{Cs}$ in a certain environment depends on the conditions during deposition and on post-depositional sediment dynamics. For example, a site in the northern hemisphere, that receives continuous sedimentation, will show two $^{137}\text{Cs}$-enriched layers in its depth profile as a result of the two episodic events. Caesium adheres better to clay than to sand particles, so that in sediment that was reworked after $^{137}\text{Cs}$-deposition, a relation with grain size may be present.

**Figure 2.2.** The decay series of $^{238}\text{U}$ and $^{232}\text{Th}$. Half-life times are given in years (a), months (m), days (d), hours (h) and seconds (s); the most relevant $\gamma$-ray emitters are indicated with a bold box (data from Browne and Firestone, 1986).
Detection of $\gamma$-radiation

This section describes the interaction of $\gamma$-radiation with matter and how it is used in $\gamma$-ray detection with various detector types. In this thesis three detectors are deployed, and their specifications, calibration, data analysis and use are described, ending in a comparison between the detectors.

Interaction of $\gamma$-radiation with matter

Gamma-rays interact with matter by transferring all or part of their energy to the electrons in the material. There are a number of ways in which this can take place.

- In the case of photoelectric effect (photoelectric absorption) a $\gamma$-ray transfers all its energy to an electron bound to an atom of the absorber material. The $\gamma$-ray disappears and the energetic electron is ejected from the atom. The ejected electron is subsequently slowed down in the surrounding material and its energy is absorbed. The cross-section ($\sigma$, interaction probability per absorber mass) for the photoelectric effect is proportionally to $Z^n/E_{\gamma}^{3.5}$, with $n = 3 – 4$ and $E_{\gamma}$ the $\gamma$-ray energy (Knoll, 2000).

Figure 2.3. Total deposition of $^{137}$Cs in 1986 in the Netherlands as result of the Chernobyl reactor accident (from CCRX, 1986).
Compton effect or inelastic scattering is the deflection of $\gamma$-rays by electrons. Part of the energy of the $\gamma$-ray is transferred to the electron and the $\gamma$-ray changes direction. The intensity as function of scattering angle decreases with increasing photon energy and for the energy range of the environmental radionuclides the scattering is predominantly directed forwardly. The probability of Compton scattering per unit of mass decreases gradually with increasing $\gamma$-ray energy and is virtually independent of the atomic number of the absorber.

In elastic scattering (or Rayleigh scattering), a $\gamma$-ray is deflected without a change in or transfer of energy. It is mainly of importance for $\gamma$-rays with energies lower than a few hundred keV.

During pair production a $\gamma$-ray decays into an electron-positron pair in the presence of a nucleus. This is only possible if $E_\gamma > 1.022$ MeV, the rest mass of the pair. The surplus energy, $E_\gamma - 1.022$ MeV, is converted into the kinetic energy of the two particles. Subsequently the positron slows down until it is captured by an electron and annihilates in two photons of 0.511 MeV each. The cross-section per unit of mass for pair production increases linearly with increasing $Z$ and increases strongly with increasing energy.

The dominance of the above effects as a function of $\gamma$-ray energy and the atomic number of the absorber is given in Figure 2.4. For the energy range of the natural radionuclides, the Compton effect is most important.

When a beam of $\gamma$-ray photons passes through matter, it is attenuated by the interaction processes in the material. The probability per unit path length that the $\gamma$-ray photon is removed from the beam (Knoll, 2000) is expressed in the linear attenuation coefficient $\mu$ (cm$^{-1}$):

$$\frac{I}{I_0} = e^{-\mu x}, \quad (2.5)$$

where $I$ is the intensity of the beam of photons with energy $E_\gamma$ after attenuation, $I_0$ the initial intensity and $x$ the traversed distance in the material. The linear attenuation coefficient depends on $\gamma$-ray energy and density $\rho$ of the absorber. The attenuation coefficient can also be expressed in the form of the mass-attenuation coefficient $\mu/\rho$ (cm$^2$ g$^{-1}$), which depends on $\gamma$-ray energy but is almost independent of the state of the material. If a material is a mixture of other materials, its mass-attenuation coefficient can be determined from the mass fractions $m_i$ of material $i$:

$$\frac{\mu}{\rho_{\text{mixture}}} = \sum_i m_i \frac{\mu_i}{\rho_i}. \quad (2.6)$$

Gamma-ray spectra
A $\gamma$-ray spectrum is a histogram representing the distribution of $\gamma$-rays from a source as a function of $\gamma$-ray energy, for a particular detector and certain time interval (Figure 2.5). There are several characteristic spectral features. Gamma-rays that undergo total absorption in the detector, either directly through the photoelectric effect or eventu-
ally through multiple Compton interactions, will form a peak in the spectrum: the full-energy peak. When a γ-ray transfers part of its energy through Compton scattering to an electron of the detector material, the other part may escape the detector with the scattered γ-ray of lower energy. The event will then be recorded as the energy of the electron and will show up in a lower energy bin than the original γ-ray energy. These events result in the Compton continuum in the spectrum. Also γ-rays that scatter in surrounding material and enter the detector with lower energy than the original γ-ray, contribute to the Compton continuum. The upper side of the continuum is called the Compton edge. The region between the Compton edge and the full-energy peak represents multiple Compton events that are summed. When a

Figure 2.4. The dominance of macroscopic interaction processes for γ-radiation as a function of γ-ray energy ($h\nu$) and the atomic number $Z$ of the absorber (from Evans, 1969).

Figure 2.5. Example of a simulated spectrum for a mono-energetic 2.5 MeV γ-ray source, observed with a hypothetical high-resolution bismuth-germanate detector. The full-energy peak has been truncated.
\( \gamma \)-ray undergoes pair production, it is possible that one or two of the annihilation \( \gamma \)-rays escape from the detector. This gives rise to peaks that are 0.511 and 1.022 MeV below the full-energy peak, the Single Escape (SE) and Double Escape (DE) peaks, respectively. The low-intensity peak just below the full energy peak is due to escape of X-rays after emission of the photo electron.

The resolution of a detector indicates how well spectral peaks from that detector can be distinguished. The narrower a peak, the better the resolution is. Resolution \(( R \)\) is a function of \( \gamma \)-ray energy and is often expressed as the ratio of the Full Width at Half Maximum \(( \text{FWHM} \)\) and the centroid energy of the peak \(( E_0 \)\):

\[
R = \frac{\text{FWHM}}{E_0} \cdot 100\% .
\]

Measured spectra generally include contributions from background radiation, which is basically the radiation from all sources other than the investigated sample. In the detection of environmental \( \gamma \)-radioactivity the background can constitute a considerable part of the total signal, because the activity concentrations of the target elements are generally low. The background has several sources:

- *Electronic and microphonic noise* mainly contributes to the lower channels.
- *Intrinsic detector radioactivity* stems from radionuclides present in the detector crystal, housing and photomultiplier.
- *Gamma-rays from natural radioactivity* are the studied quantity in this thesis, but also materials surrounding a detector which are not of interest can contribute radiation (concrete walls, bricks, etc).
- So-called *cosmic radiation* generally dominates the total background spectrum. Particles with a large range of energies bombard the Earth. Their interaction with the atmosphere produces secondary particles that give rise to this background component. The cosmic background is variable in time and latitude.

The background of a detector is by definition a recording obtained without a source. In practice a background measurement can be rather complex.

**Gamma-ray detectors**

Gamma-ray detectors are devices that transform \( \gamma \)-radiation into a measurable electrical current. There are various detector types with different characteristics, depending on detector material. As a response to incident \( \gamma \)-rays, a detector material generates a current, voltage or light pulse. The intensity of the pulse is proportional to the energy deposited by the \( \gamma \)-ray in the detector, and so a spectrum emerges.

The amount of pulses recorded for a given number of incident \( \gamma \)-rays is expressed in detection efficiency. The higher the efficiency of detection, the shorter the recording time can be. In general, the efficiency of a detector increases with an increase in mass and atomic number of the detector material. This overview is limited to detector types that are deployed in this thesis.

The functioning of a *semi-conductor diode detector* is based on the dislodging of electrons by \( \gamma \)-rays so that the material, normally an isolator, becomes temporarily
conductive and a voltage drop occurs. The resolution of semi-conductor detectors is generally very good. Drawbacks are that this type of detector has to be cooled with e.g. liquid nitrogen, and that crystal sizes are rather limited. This results in relatively long acquisition times. An example of this type of detector is an HPGe (High-Purity Germanium) detector.

Inorganic scintillation detectors consist of translucent material. When absorbing $\gamma$-radiation, a light pulse is emitted which is detected by a photomultiplier tube (PMT) attached to the detector crystal, which converts the light into an electric signal. Scintillators have a poorer resolution than semi-conductor detectors. However, the average atomic number of inorganic scintillators is generally higher and the crystal sizes can be larger than that of semi-conductor detectors, leading to higher detection efficiencies and relatively more counts in the full-energy peak. Examples of inorganic scintillators are NaI(Tl), CsI(Tl) and BGO (bismuth germanate or Bi$_4$Ge$_3$O$_{12}$).

### 2.2.4 Spectral analysis

Data recorded with a $\gamma$-ray detector are given in the form of total count rates or spectra. Total count rates represent the total number of detected $\gamma$-rays per unit time $t$ in a specified part of the energy spectrum, and are generally expressed in counts per second (cps). As radioactivity is a statistical process that follows a Poisson distribution, the uncertainty in the count rate is proportional to the square root of the number of recorded events $N$:

$$\text{count rate} = \frac{N}{t} \pm \frac{\sqrt{N}}{t}. \quad (2.8)$$

There are several methods available for converting measured spectra into activity concentrations. Which method is most suitable depends amongst others on what type of detector was used for the measurement. For the detectors used in this thesis, either Peak-Content Analysis or Full-Spectrum Analysis is used.

**Peak-Content Analysis** converts the net counts in the spectral full-energy peaks into activity concentrations. This method is used for detectors that have a good energy resolution. The user selects the desired peaks and sets the regions of interest (ROI) around those peaks, or alternatively a computer algorithm determines the ROIs. The net peak content is determined by subtracting the spectral background (extrapolated continuum) from the full-energy peak, assuming a linear relation between continuum parts at both sides of the peak. Peak-content analysis is used for the HPGe detector employed in this thesis.

The method of **Full-Spectrum Analysis** (FSA, e.g. Knoll, 2000) uses the entire energy spectrum for the determination of activity concentrations, including all structural features. It is generally used for spectra from detectors that have a relatively poor resolution. A requirement for this analysis is that the radionuclides that are present in the source are known, which is generally the case in environmental applications. The method considers a spectrum of a certain source as the sum of the individual spectra of all radionuclides in that source and background (Figure 2.6). The shape of each
contribution is given by its standard spectrum and the magnitude by its activity concentration. Standard Spectra represent the detector response to a source containing 1 Bq kg\(^{-1}\) of a given radionuclide in a particular geometry. The contribution in each energy bin \(i\) of the measured spectrum \(S\) is then given by:

\[
S(i) = \sum_j C_j X_j(i) + Bg(i),
\]

in which \(C_j\) (Bq kg\(^{-1}\)) is the activity concentration for nuclide \(j\), \(X_j\) is the standard spectrum of nuclide \(j\) and \(Bg\) is the background spectrum. Activity concentrations \(C_j\) are determined by proportionally scaling the standard spectra and minimising the sum of squares, \(\chi^2\):

\[
\chi^2 = \frac{1}{n-m} \sum_{i=1}^{n} \left[ S(i) - \sum_j C_j X_j(i) + Bg(i) \right]^2 / w(i)
\]

where \(i\) is channel (up to \(n\)), \(w(i)\) is a weight factor, which is usually set equal to \(S(i)\) and \(m\) is the number of extracted parameters (activity concentrations, Hendriks et al., 2001a). The value of \(\chi^2\) is indicative for the quality of the fit. If the detector under consideration is temperature-sensitive, the resulting spectral drift can be corrected by ‘stabilising’ the spectra in the least-squares analysis. A disadvantage of FSA is that the derived activity concentrations are influenced by the covariances between the standard spectra due to overlap in shape. FSA is used for the BGO detector employed in this thesis. Standard spectra are generated through calibration measurements or (like in this thesis) derived from Monte-Carlo simulations.
2.2.5 Monte-Carlo simulations

In the description of radiation transport and detection, Monte-Carlo simulations play an important role. This is because experiments are not always representative or efficient enough to obtain detector response (e.g. standard spectra) for in-situ geometries. 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 for example for sensitivity analyses 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 for reproducing $\gamma$-ray spectra from in-situ measurements and 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; Mauçec et al., 2004a).

Monte-Carlo simulations determine the average outcome of a process by repeatedly random sampling all possible pathways of that process. For every step in the process, the probabilities associated with that step are sampled from a probability distribution. For the transport of $\gamma$-radiation a large number of single events (photons and electrons) are simulated from creation to termination. For each event the starting energy, starting location and initial direction are sampled, and subsequently every interaction type, location, remaining energy and new direction. The probability distributions for each step are based on physics measurements and modelling and are associated with the type of event and absorber material under consideration. The steps generally continue until the source event and all its secondary electrons and $\gamma$-rays have deposited all of their energy or have moved outside of the defined geometry. After running a large number of ‘histories’ (a lifespan of an event plus its progeny from creation to termination) the average behaviour of the radiation can be extracted. The final result can for instance be the pulse-height spectrum of deposited energy in a detector volume, or the $\gamma$-ray flux over part of the source volume (see Figure 2.5).

In this thesis we use the package MCNPX from Los Alamos National Laboratory (Monte Carlo N-Particle eXtended, a general-purpose Monte Carlo radiation transport code, Pelowitz, 2005). The calculations were performed at the High-Performance Cluster of the University of Groningen. In MCNPX all parts of the geometry, including detector and source, are represented by cells, which can consist of various materials. MCNPX provides several calculation types, called tallies. This thesis applies pulse-height tallies (F8) for the creation of standard spectra and F4-tallies (the ‘particle’ flux in a cell) for the determination of source volumes. MCNPX provides the relative uncertainty with the output.

Generally, Monte-Carlo simulations are carried out as ‘forward’ calculations, in which a $\gamma$-ray or electron, like in reality, loses energy by interactions as it proceeds. It is also possible to reverse the transport and make particles gain energy during transport. These reversed calculations are called adjoint calculations and are for instance used to infer contributing volumes in situations where the source is much larger than the detector (Briesmeister, 2000).
Because a large number of events need to be created to generate statistically reliable results, computer times can be very long (days to weeks). To reduce computer times, in forward calculations Variance Reduction Techniques can be applied. A number of these are very suitable for calculations regarding BGO detectors in an environmental setting (Hendriks et al., 2002; Hendriks, 2003) and were therefore used in simulations in this thesis:

- **Geometry truncation** omits parts of the source volume that do not contribute significantly to the detected signal, e.g. far away from the detector.
- **Biasing of source location** and direction were applied to sample the source as effectively as possible.
- **Secondary electrons** with energies below 0.01 MeV are cut-off, as tracking these particles takes too much time, whereas they do not contribute to the relevant part of the spectrum.

Additionally, the run-time reduction technique of Surface Source Write/Surface Source Read (SSW/SSR) is used. It is based on the notion that electrons require many interactions to deposit all their energy, but do not penetrate very far. Tracking a large number of electrons far away from a detector is therefore unnecessary. Hence, the calculations are split in two parts: the first part tracks only γ-rays in a volume further away from the detector and the second part incorporates both electron and γ-ray transport in a volume close to the detector. The boundary between the two volumes is a surface. All γ-rays that cross the surface in the first step are stored (written) on the surface (SSW) and are used (read) as input (‘source’) for the second step (SSR). The surface should be positioned further from the detector than the maximum traversed distance of electrons. The difference in outcome with ‘normal’ calculations is negligible, whereas for borehole configurations computer times can be reduced by a factor 20 (Hendriks, 2003).

### 2.2.6 Detectors in this thesis

**Portable scintillation rate detector: Scintrex GIS-5 detector**

In this thesis, three detectors with specific characteristics and applications are applied. The Scintrex GIS-5 detector is a detector for measuring total γ-ray intensity up to 3 MeV. It is a hand-held detector with a count-rate display, containing a thallium-activated sodium iodide crystal (NaI(Tl)) of $4.05 \times 4.05 \times 4.85$ cm³ (Appendix 2A, Table 2.1). The detector was originally designed for field exploration of uranium deposits, but in this thesis it is used as an instrument measuring a summed intensity of γ-radiation, without specifying the source nuclides. The instrument displays the total counts for a specified time interval, which have to be recorded manually. Cosmic and detector background are included in the total count rate. The combined intrinsic and cosmic background of the Scintrex detector was determined from an experiment in which detector was placed in the open air, while shielded from radiation from the soil and the sides by 10 cm lead. This resulted in a background of 13.19 (± 0.09) cps, which was subtracted from all measurements. The scintillation output of a sodium iodide crystal
is temperature-dependent. However, in the field it was not possible to do regular calibrations. It has been stated that under normal field conditions, the uncertainties induced by temperature variations are smaller than three percent (Stapel et al., 1992).

**BGO detector: PANDORA detector**

The PANDORA detector (Precision Agriculture Needed Detector Of RadioActivity Van der Graaf et al., 2007) is a large-volume BGO detector, manufactured by Scionix (Bunnik, the Netherlands). It was designed for the fast *in-situ* surveying of large areas, e.g. for creating maps of activity concentrations for separate radionuclides. The PANDORA detector is a larger version of the MEDUSA detector, which has already proven its mapping qualities in e.g. underwater mapping of the sediment in an estuary and monitoring sediment transport at a sludge disposal at the seafloor (Venema and De Meijer, 2001; Van Wijngaarden et al., 2002a).

The detector consists of a cylindrical BGO crystal of 15.0 cm long and 12.7 cm diameter. The crystal, a photomultiplier and additional electronics are housed in an aluminium cylindrical casing (Figure 2.7). The entire system is approximately 1 m in length and 16 cm in diameter, weighing 35 kg. Batteries, additional hardware, laptop and GPS are housed in a separate box, connected to the detector by a cable. The main output of the PANDORA system consists of time, total count rate and a γ-ray spectrum set at a range between 0.02 and 3 MeV. During measurements, the output of the PANDORA system and GPS receiver (Global Positioning System; in this case a Garmin GPS 76) are logged simultaneously to a laptop with the Medusa Data Logger programme (Feenstra, 2000). Spectra are generally logged every 2 seconds. Further specifications can be found in Appendix 2A, Table 2.2. A performance test of the detector is described in Van der Klis (2002) and performance assessments can be found in Maučec et al. (2004b) and Van der Graaf et al. (2007).

Measurements with the PANDORA detector were carried out with the detector lying on, or towed over, the ground with an arbitrary part of the cylinder wall facing the soil. The detector was not collimated.

![Figure 2.7. Components and dimensions (in mm) of the PANDORA detector.](image)
From spectra, created by the PANDORA detector, radionuclide activity concentrations are extracted by Full-Spectrum Analysis in the MEDUSA Post Analysis software (Limburg, 2003). The analysis requires individual standard spectra for $^{40}$K, $^{232}$Th, $^{238}$U and $^{137}$Cs. Before fitting the standard spectra, the contributions of the intrinsic and cosmic background were subtracted. The background spectra were taken from earlier measurements in which the detector was slightly submersed in a deep freshwater lake (Van der Klis, 2002). The analysis results in the activity concentrations of $^{40}$K, $^{232}$Th, $^{238}$U and $^{137}$Cs and their standard deviations, and the $\chi^2$ of the analysis. In Chapter 5, the net count rate was determined by stabilising the spectra (i.e. correcting for spectral drift) and subsequently summing spectral content over the range 0.2 – 2.8 MeV and subtracting the contribution of the background. In Chapter 4 the large amount of obtained spectra made this method too labour-intensive and the net count rate was approximated by summing the activity concentration times the counts in the standard spectrum (0.2 – 2.8 MeV) for $^{40}$K, $^{232}$Th and $^{238}$U. The energy region of interest is chosen because the lower channels often contain electronic and other noise (Maučec et al., 2004a).

The standard spectra for $^{40}$K, $^{232}$Th, $^{238}$U and $^{137}$Cs were created using Monte-Carlo simulations in MCNPX (Pelowitz, 2005), for a situation in which the radionuclides are distributed homogeneously within the soil and the decay series of $^{232}$Th and $^{238}$U are in secular equilibrium. The standard spectra were calibrated by comparing the derived concentrations with those measured from soil samples, analysed in the laboratory with a hyper-pure germanium detector at the Kernfysisch Versneller Institute (KVI) in Groningen (Hendriks et al., 2002; Maučec et al., 2004a). A description of the simulations is given in Chapter 3.

**HPGe detector**

The detector used in this thesis with the highest resolution is the laboratory-based HPGe detector from Ortec (EG&G Ortec p-type; model GEM-45200-S, see Table 2.3 in Appendix 2A). It was used for measuring radionuclide activity concentrations in sediment samples. The detector is shielded by 10 cm of old lead and a copper lining to create a low-background environment and is cooled with liquid nitrogen. Samples were measured in Marinelli beakers or small sample containers, depending on sample size. Two calibrated Marinelli-beaker geometries are used (233N of 1.0 l and 133N of 0.5 l) and smaller samples are measured in cylindrical plastic containers of 100 ml. The detector is calibrated using certified radionuclide mixtures. Activity concentrations are derived from Peak Content Analysis, taking into account measurement time, branching ratio, detector efficiency and sample mass. The procedure is carried out at KVI according to the Dutch National Standards (NEN 5623, Nederlans Normalisatie-instituut, 2002). Before measuring, most of the sediment samples were sealed and left for at least three weeks to ensure secular equilibrium in the uranium series.

**Wet and dry activity concentrations**

In case the samples contained a certain amount of water, the measurements provide
activity concentrations of the wet sediment, $C_{j,wet}$. Water in itself is not radioactive and its presence dilutes the sediment activity concentrations. Therefore, after measurement on the HPGe, part of each sample was oven-dried for 24 hours at 105°C, so that the absolute water content $w_a$ (weight fraction of total mass, $0 < w_a < 1$) of the sample could be determined. From that, the dry activity concentrations $C_{j,dry}$ of only the mineral and organic part could be derived. This relation is worked out in more detail in Chapter 3. Because the samples were later used for calibration with in-situ measurements, organic material was not removed.

Detector application

The choice of detector in a specific survey usually is a trade-off between detector output type, required counting times and how well the detector can be transported in the field. The Scintrex GIS-5 detector is most suitable for obtaining a quick-scan of spatial variations in radioactivity in a certain area. The HPGe detector is used for detailed point investigations of sediment samples and calibration of the other detectors. Its lead shielding and the requirement of liquid nitrogen make the present detector not suitable for field application. The PANDORA detector is capable of non-destructive in-situ mapping of individual radionuclides with continuous logging. It is a relatively new detector and has not been applied on salt marshes before.

2.3 Non-radiometric measurements

This part of the chapter gives an overview of the non-radiometric methods used in this thesis. The methods are related to the geomorphology of salt marshes and properties of salt-marsh sediment. The measurement layouts are discussed in the individual chapters.

2.3.1 Top-layer thickness

The thickness and composition of the salt-marsh sediment were described from soil cores taken with a small soil corer of 1 cm diameter and 53 cm length. This relatively simple method can be repeated often. The core was cut in length and examined for grain-size transitions. The thickness of each individual homogeneous layer, if thicker than 0.5 cm, was measured with a ruler to the nearest 0.5 cm (left panel of Figure 2.8). For simplicity, all sediment where cohesive properties dominated and where most grains were not individually visible was classified as ‘fine-grained’. This encompasses all sediment with considerable amounts of lutum, silt and possibly organic material. Sediments that showed no clear cohesive properties and where most grains were individually visible were classified as ‘sand’. The top layer was defined as the sum of all fine and coarse layers above a continuous layer of sand, the base layer.

Additionally, from all cores the transition between top and base layer was classified. If the transition was abrupt, i.e. there was no mixing of fine-grained material and sand, the transition was classified as ‘sharp’ (right panel of Figure 2.8). If the transition
graded along several cm from sand to fines (fining upward, often including all inter-
mediate grain sizes), it was referred to as ‘gradual’. Transitions grading within 1 cm
were referred to as ‘intermediate’. The transition types give an indication about
the conditions during the early stages of salt-marsh formation (see Chapter 6) and about
the accuracy with which the transition between the two layers could be determined.
The latter is given in classes, representing 0.5, 1.0 and 2.0 cm, corresponding to the
sharp, intermediate and gradual transitions.
In case the small soil corer was not long enough, the position of the transition was
based on the results of the combination of an Edelman soil sampler and a longer soil
corer. These measurements have an estimated uncertainty of 2 cm.

2.3.2 Soil elevation
Soil elevation was measured with respect to local ordnance datum. In the case of the
Netherlands that is Normaal Amsterdams Peil (Dutch Ordnance Level, NAP), which
represents average sea level. For locations in Denmark the Danish equivalent DNN
was used (DNN = NAP - 0.16 m, Lassen, 1989). Using local tide conditions, the soil
elevations were converted to elevation relative to Mean High Tide (MHT). Two types
of levelling equipment were used: a Trimble Spectra Precision laser LL500 and a tradi-
tional levelling instrument. The precision of the measurements was 1 mm and the
accuracy depended on the survey and ranged from several mm’s to several cm’s.
Soil elevation minus top-layer thickness gave the elevation of the underlying sand
surface, which is referred to as base elevation.

2.3.3 Geographical coordinates
The geographical locations of the data are given in the Dutch RD grid
(Rijksdriehoekstelsel, http://www.rdnap.nl/). Geographical coordinates were deter-
mined from GPS readings (using various Garmin GPS receivers) when measurement
spacing was more than 25 m. For smaller distances a combination of measuring tape
and GPS recordings was used. If necessary conversions between various geographical

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**Figure 2.8.** Classification of marsh top layer from soil cores. Dark colours represent fine-
grained sediment and light colours sand.
projections were carried out using the program PCTrans (Royal Netherlands Navy Hydrographic Service, 2003). The horizontal accuracy of the measurement location depended on the spacing used in each specific measurement layout. Points with distances of less than 10 m had spatial accuracies of approximately 10 cm, whereas on longer distances the estimated uncertainty was in the order of 10 m.

2.3.4 Bulk density, porosity and water content
The wet bulk density, porosity and water content of sediments were determined from soil samples, if necessary with known volume. The relevant equations can be found in Chapter 3, where the method is discussed in more detail.

2.3.5 Grain-size distribution
For the determination of sediment texture, a few grams of the untreated samples were freeze-dried (which preserves grain structure) and analyzed on a Coulter Malvern Laser-refraction device at the NIOO-CEME in Yerseke (NL). The analysis resulted in the percentages of the grains in the mud, very fine sand, fine sand and medium fine sand fractions, the median grain size and the specific surface area of the grains.

2.3.6 Salt-marsh age
On Schiermonnikoog, salt-marsh age was determined from the age of the vegetated surface, based on aerial photographs and historical maps (Kers et al., 1998). As the sedimentation of fine-grained material is directly related to the presence of vegetation, the establishment of vegetation is effectively the same as the beginning of marsh development. The assigned year of development is the average year between the maps or photographs with and without vegetation. All locations that were classified as being salt marsh during surveys and were not on the age map of 1998, were considered to have developed after the map was produced, and were assigned to the year 2000. Due to small errors in the boundaries on the age map, this year class contains also older locations that are incorrectly classified.

For Terschelling and Skallingen, marsh age was estimated from island descriptions, maps and aerial photographs (in Roozen and Westhoff, 1985; Aagaard et al., 1995; Schoorl, 2000; Nielsen and Nielsen, 2006a; Ten Haaf and Buijs, 2008).

2.4 Study sites
The study sites in this thesis are all temperate tidal salt marshes in Northwest Europe. The main study site is the barrier island Schiermonnikoog (NL). Because one island is not enough to draw general conclusions about European barrier-island salt marshes, part of the measurements were repeated on the island of Terschelling and the peninsula of Skallingen (DK).
2.4.1 Wadden Sea and island marshes
The Wadden Sea stretches from Denmark in the north, via Germany to the Netherlands in the southwest (Figure 2.9). It forms a mixed-energy barrier system that consists of tidal basins between barrier islands and the mainland coasts, harbouring large intertidal areas that emerge during every tidal cycle. Several estuaries merge into the Wadden Sea, of which the Elbe estuary is the largest. The Wadden Sea acts as a sink for sand and mud. The main sources of the sand are the erosion of the North Sea floor, the North Sea coast of the barrier islands and the Holland coast. For the Dutch part of the Wadden Sea, the sources of the mud are predominantly the Rhine and the North Sea floor. Through the import of sediment, the system has been keeping up with the rising sea level. The Wadden Sea evolved in the Holocene and is in that respect a geologically young area (Oost and De Boer, 1994; Vos and Van Kesteren, 2000). It is a very dynamic system in which islands emerge, grow, move and erode, and where shoals and channels frequently shift. Waves and currents sort the sediment, and morphological units within the Wadden Sea are connected through flows of water and sediment. Through the processes of scour lag (Postma, 1967) and settling lag (Van Straaten and Kuenen, 1958; Postma, 1961) the average grain size decreases with distance from the inlets. At locations where flow velocities are low enough for the smallest sediment particles to settle, salt marshes can develop, as described in Chapter 1.
Human influence has reduced the dynamics of the Wadden Sea. Seawalls were built to cut off sea arms and reclaim salt marshes into cultivated fields. Sand dikes were constructed on many islands to stabilise the dunes and salt marshes. Land reclamation from the past centuries still affect the morphodynamics of the Wadden Sea and limit the possibilities for the Wadden Sea to grow with the rising sea level (Dijkema, 1987a; Ehlers, 1988b).

2.4.2 Schiermonnikoog

Schiermonnikoog (53°30´N, 6°10´E) is one of the West Frisian barrier islands in the Dutch Wadden Sea (Figure 2.9 and Figure 2.10). The island is approximately 15 km long and 0.5 – 3 km wide. Local tidal amplitude is 2.3 m (Oost and De Boer, 1994) and MHT is around 1.00 m above NAP. The island is orientated roughly east-west and consists of dunes, beaches including green beaches (a complex of vegetated beach, low primary dunes and depressions), beach plains, a polder, salt marshes and intertidal flats.

The island has migrated several kilometres in south-eastern direction since the Middle Ages, caused by changes in the tidal inlets and a dominantly eastward directed longshore drift. Meanwhile, the western side eroded (Bakker, 1989; Oost and De Boer, 1994). At present, the western part of the island is stable and the eastern end is still extending, resulting in the gradual formation of new beach plains, dunes and salt marshes.

The main salt marsh complex of Schiermonnikoog is located at the eastern part of the island and is 8 km in length and 0.5 – 1.5 km in width (Figure 2.10). The average net surface elevation change ranges from 0.1 to 0.5 cm a\(^{-1}\) (with extremes of 1.1 cm a\(^{-1}\) in the pioneer zone) and depends on marsh age and base elevation (Van Wijnen and Bakker, 2001). The marsh is drained by several large creeks, oriented roughly north – south (Figure 2.10). Part of these creeks are connected to overwash channels (Oost and De Boer, 1994; Ten Haaf and Buijs, 2008). The tidal divide\(^1\) lies south of the salt marsh. The vegetation of the low marsh is dominated by *Salicornia* spp., *Suaeda maritima*, *Limonium vulgare* and *Puccinellia maritima*. Higher marsh elevations feature *Festuca rubra*, *Atriplex portulacoides*, *Elytrigia atherica* and *Artemisia maritima*. At present, the oldest part of the salt marsh is grazed by cattle, which strongly affects the vegetation.

As a result of the island dynamics, the marsh exhibits a chronosequence: the marsh age decreases from approximately 200 year-old in the middle of the island towards very young at the eastern end and the average top-layer thickness varies accordingly (Olff et al., 1997, Figure 2.10).

In 1959 a sand dike was built that cut off a large area of beach plain from the North Sea. The sand dike extended 5.5 km from west to east and strongly facilitated the

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\(^1\) The tidal divide is the line in the back-barrier area where the tidal waters coming from two sides from the open sea converge. Here, average flow velocities are low and the elevation of the intertidal flats is relatively high.
growth of the salt marsh south of it. Part of this area (between the 3rd and 4th creek) was already vegetated and consisted of green beach, dunes, washovers and salt-marsh areas (De Leeuw et al., 2008). Currently the eastern part of the sand dike is not main
tained anymore (Rijkswaterstaat Directie Friesland and Rijkswaterstaat Meetkundige dienst, 1991) and has been partly breached by overwash complexes (Oost and De Boer, 1994). Aerial photographs indicate that since 1970 especially the eastern part of the sand dike has been strongly reshaped by wind and water. By 1989, virtually the entire sand dike east of the 4th creek had been reworked. New dunes have developed around the location of the previous artificial sand dike. The salt marshes have now extended beyond the previous limit of the sand dike, in the shelter of new natural dunes. Additionally to the eastwards growth, the majority of the marsh is laterally accreting southwards. Only the oldest marshes close to the polder and ferry dam are eroding at a rate of maximum 0.8 m a⁻¹ (Ehlers, 1988a).

Figure 2.10. Age of salt marsh (as beginning of vegetation development) on Schiermonnikoog (from Kers et al., 1998). The original location of the artificial sand dike on Schiermonnikoog is given by the dashed line, the part that is still present in dark grey and the by now reworked part in light grey. The numbering of the creeks is indicated.
The largest human influences on sedimentation processes have been the construction of a seawall in 1860 that converted marsh into polder, local dune reinforcements and the previously mentioned artificial sand dike (Reitsma and Bakker, 1986). Additionally, the many seawalls that were built in the eastern part of the Dutch Wadden Sea during the past centuries have affected the back-barrier area around Schiermonnikoog and altered the behaviour of the tidal channels and shoals around the island. Especially the closure of the Lauwerszee has had a noticeable impact (Ehlers, 1988a; Oost, 1995). In contrast to other Dutch Frisian islands, no sand nourishments with offshore sand have been carried out on Schiermonnikoog.

2.4.3 Terschelling
The second study area is the Dutch Wadden Sea island of Terschelling (53°26’N, 5°28’E), west of Schiermonnikoog. The tidal range is 2 m and MHT is 0.87 m + NAP. The salt marshes are located at the eastern part of the island. It consists of two older parts (de Grie and de Groede, see Schoorl, 2000) and the salt marsh on the Boschplaat, on which most of our measurements are situated. The marsh on the Boschplaat covers the largest area and formed after an artificial sand dike was built between 1931 and 1938, closing off the connection with the North Sea (Roozen and Westhoff, 1985). It is of more uniform age than the marsh on Schiermonnikoog and the eastern part is presently eroding. Otherwise the salt marsh is comparable to the one on Schiermonnikoog.

2.4.4 Skallingen
The third site is the peninsula of Skallingen in Denmark (55°30’N, 8°20’E), the barrier spit that forms the northern end of the international Wadden Sea. The tidal range is on average 1.5 m (Bartholdy et al., 2004; Christiansen et al., 2004) and MHT lies around 0.75 m + DNN. The salt marsh started to develop between 1900 and the 1950s, in two phases (Bartholdy and Madsen, 1985; Aagaard et al., 1995). Close to the location of our measurement sites, an artificial sand dike was constructed in 1933, sheltering the marsh from the North Sea and blocking washover channels (Aagaard et al., 1995; Nielsen and Nielsen, 2006a). The marsh appears to contain more organic material than the marshes on Schiermonnikoog and the base material is coarser (personal observation).

2.4.5 Westerhever
A small number of measurements were done on the Westerhever salt marsh (54°22’N, 8°38’E), located on the mainland coast of Germany. The marsh is built towards and on top of a sand flat and consequently consists of fine-grained sediment on top of sand, and is in that respect comparable to the island marshes.
## Appendix 2A. Detector specifications

### Table 2.1. Specifications of the Scintrex GIS-5 detector (from Scintrex Limited).

<table>
<thead>
<tr>
<th>Specification</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>Manufacturer</td>
<td>Scintrex Limited</td>
</tr>
<tr>
<td>Overall dimensions</td>
<td>250 × 190 × 95 mm³</td>
</tr>
<tr>
<td></td>
<td>2.8 kg including batteries</td>
</tr>
<tr>
<td></td>
<td>magnetically shielded and waterproof</td>
</tr>
<tr>
<td>NaI (Tl) crystal</td>
<td>40.5 × 40.5 × 48.5 mm³</td>
</tr>
<tr>
<td>High voltage supply</td>
<td>internal converter, 750 V</td>
</tr>
<tr>
<td>Energy range</td>
<td>TC setting: 0.05 – 3.0 MeV</td>
</tr>
<tr>
<td>Counting periods</td>
<td>1, 3, 10, 30, 100 seconds, switch selectable</td>
</tr>
<tr>
<td>Calibration</td>
<td>supplied ThO₂ calibration source</td>
</tr>
<tr>
<td>Audio output</td>
<td>response time 0.25 s, the frequency of the output is proportional to the</td>
</tr>
<tr>
<td></td>
<td>excess count-rate over the threshold, which is continuously variable</td>
</tr>
<tr>
<td>Visual output</td>
<td>digital liquid crystal display, output max 19 999</td>
</tr>
<tr>
<td>Power supply</td>
<td>4 ‘D’ cells</td>
</tr>
<tr>
<td>Sensitivity in TC mode</td>
<td>15 cps per ppm uranium equivalent (for infinite half-space sources)</td>
</tr>
<tr>
<td>Temperature range</td>
<td>technical specifications met from –20° to + 55°C</td>
</tr>
</tbody>
</table>
### Table 2.2. Specifications of the PANDORA detector.

<table>
<thead>
<tr>
<th>Manufacturer</th>
<th>Scionix</th>
</tr>
</thead>
<tbody>
<tr>
<td>Overall dimensions</td>
<td>cylinder of 1.25 m length and 17 cm diameter, 35 kg box containing external hardware: $42 \times 56 \times 37 \text{ cm}^3$, 37 kg</td>
</tr>
<tr>
<td>Crystal</td>
<td>bismuth germanate: Bi$_4$Ge$<em>3$O$</em>{12}$ (BGO) diameter of 127 mm, length 150 mm</td>
</tr>
<tr>
<td>Crystal coating</td>
<td>- 0.3 mm teflon reflector coating (to prevent loss of scintillation light); - 2.4 mm layer of silicon rubber (to protect the crystal from shocks)</td>
</tr>
<tr>
<td>Photomultiplier</td>
<td>type ETL 9390, diameter 130 mm</td>
</tr>
<tr>
<td>Housing</td>
<td>detector: 0.8 mm stainless steel housing, watertight aluminium cylindrical casing: 125 cm length and 17 cm diameter, watertight</td>
</tr>
<tr>
<td>Counting periods</td>
<td>1, 2 or 10 seconds</td>
</tr>
<tr>
<td>Logging parameters</td>
<td>- time - total count rate (raw) - $\gamma$-ray spectrum of 512 channels, approximately 0 - 3 MeV (depending on gain drift) - temperature within detector housing - sound - GPS coordinates (external)</td>
</tr>
<tr>
<td>Calibration</td>
<td>indirect by Monte-Carlo simulations and benchmark experiment</td>
</tr>
<tr>
<td>Power supply</td>
<td>12 V (external)</td>
</tr>
<tr>
<td>Additional sensors</td>
<td>internal: AD590 temperature sensor and microphone external: Garmin GPS 76: a differential-ready 12 parallel channel receiver, logging every second. Accuracy is better than 15 meters RMS 95% typical, depending on satellite reception, at best 5 - 6 m</td>
</tr>
<tr>
<td>External hardware</td>
<td>- power supply (car battery) - ALADIN telemetry board, ANTARES Datensysteme GmbH, Stuhr, Germany - connecting cable - laptop</td>
</tr>
</tbody>
</table>
### Table 2.3. Specifications of the HPGe detector.

<table>
<thead>
<tr>
<th>Specification</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>Manufacturer</td>
<td>Ortec</td>
</tr>
<tr>
<td>Detector type</td>
<td>EGG Ortec p-type; model GEM-45200-S</td>
</tr>
<tr>
<td>Crystal</td>
<td>High-Purity Germanium diameter 60.4 mm, height 78.3 mm</td>
</tr>
<tr>
<td>Counting periods</td>
<td>between 8 and 24 hours for the desired precision (depending on sample activity and mass)</td>
</tr>
<tr>
<td>Calibration</td>
<td>Several times per year</td>
</tr>
<tr>
<td>Shielding</td>
<td>shield of 10 cm 'old' lead, lined internally with 2 mm of copper</td>
</tr>
<tr>
<td>Geometry</td>
<td>- Marinelli beaker of 0.5 or 1 l, and calibrated for intermediate volumes</td>
</tr>
<tr>
<td></td>
<td>- cylindrical plastic sample containers (0.1 l)</td>
</tr>
<tr>
<td>Cooling</td>
<td>Liquid nitrogen</td>
</tr>
<tr>
<td>Output</td>
<td>Activity concentrations of $^{40}$K, $^{238}$U, $^{226}$Ra, $^{234}$Pa, $^{232}$Th, $^{235}$U, $^{134}$Cs, $^{137}$Cs, $^{60}$Co, $^7$Be</td>
</tr>
<tr>
<td>Software</td>
<td>- Acquisition: PCA3 (Personal Computer Analyzer)</td>
</tr>
</tbody>
</table>