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# Distribution and Mobility of $^{137}$Cs in Soil from the Catchment Area of Vefsna

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# Abstract

The primary source of $^{137}$Cs in Norway is the Chernobyl accident, and several decades after the accident, there is still a substantial amount in the environment. The area of Hattfjelldal, within the catchment area of the river Vefsna, is situated in the southern parts of the county Nordland, which was especially affected. In the present work, soil samples were taken from two different sites, Nerlifjellet and Groalia in the area of Hattfjelldal, based on knowledge about the relatively high activity of $^{137}$Cs in this area and the differences in soil composition. The samples from the two soil types were analysed to investigate any differences in distribution of $^{137}$Cs, binding and mobility of $^{137}$Cs as well as stable $^{133}$Cs, and if any differences could be connected to differences in soil components.

The soil was collected using a soil corer and divided based on the top 3 cm and visible layers. For Groalia, the 0-3 cm layer was further divided based on a visible mineral layer (2-3 cm) and an organic layer (0-2cm). The soil layers from Nerlifjellet were characterized with 13-97% organic material, compared to Groalia with 5-40%. The pH was similar dissimilar. A grain size analysis for the layers with an organic matter content of < 20% show that the mineral soil in Nerlifjellet consist of mainly sand (73%) while the mineral soil from Groalia mainly consisted of silt (39-57%).

The activity concentration of $^{137}$Cs was identified to be 200-900 Bq/kg dry soil from Nerlifjellet, depending on the soil layer, while for the soil from Groalia it was 36-1300 Bq/kg. The activity density per cm of $^{137}$Cs in the soil from Nerlifjellet was identified to be 1.0-1.1 Bq/m$^2$ per cm, depending on the soil layer. For the soil from Groalia, the activity density of $^{137}$Cs per cm was 0.19-6 Bq/m$^2$ per cm. For the soil from Nerlifjellet, $^{137}$Cs were more evenly distributed through the soil core than for the soil from Groalia, where $^{137}$Cs were mainly found in the upper 3 cm of the soil core (86%). Based on this, $^{137}$Cs seem to be more mobile in the soil from Nerlifjellet than the soil from Groalia, which was suggested to be connected to the difference in content of organic material.

A sequential extraction of several of the layers from both sites indicate that $^{133}$Cs was bound stronger in the soil with <40% organic material compared to soil with more organic material. This supported the idea that the $^{137}$Cs most likely are leaching in a larger degree from the more organic soil in Nerlifjellet than from the more mineral containing soil in Groalia and thus transported more to underlying layer in the soil.

The activity concentration of $^{137}$Cs in fractions from the grain size analysis was measured and the result showed a higher activity concentration of $^{137}$Cs in the clay fraction than in the other fractions and an activity concentration 9-30 times higher than the bulk sample. In the mineral layers $^{137}$Cs was mainly bound to the silt and clay fractions (52-97%) and in minor to sand (<33%). Comparing the two sites, the proportion of $^{137}$Cs in the clay fraction for the 7.5-13 cm layer from Nerlifjellet was lower than for the 8-13 cm layer from Groalia. This was suggested to be due to a lower amount of clay in the layer from Nerlifjellet. Although a large fraction of sand and silt can be present in the soil, result indicate that a higher amount of clay decreases the likelihood of $^{137}$Cs binding to sand or silt due to the high affinity of $^{137}$Cs onto clays. Results highlight that it is essential to identify the content of organic material and clay content in soil to understand the mobility of $^{137}$Cs in soils and the probability for remobilisation of $^{137}$Cs in runoff.

# Norsk sammendrag

Hovedkilden til $^{137}$Cs i Norge er Tsjernobyl ulykken og selv flere tiår etter finnes det betydelige menger i naturen. Hattfjelldal, som befinner seg i nedbørsfeltet til elven Vefsna, ligger i den sørlige delen av Nordland og ble spesielt berørt. I dette arbeidet ble jordprøver tatt fra to forskjellige lokasjoner, Nerlifjellet og Groalia i Hattfjelldal, basert på kunnskap om relativt høy aktivitet av $^{137}$Cs og forskjeller i jord sammensetning. Prøvene fra de to jordtypene ble analysert for å undersøke om det var noen forskjeller i distribusjon av $^{137}$Cs, binding og mobilitet av $^{133}$Cs i tillegg til $^{137}$Cs og om noen forskjeller kunne knyttes til jord sammensetning.

Jordprøvene ble tatt ved å bruke en jordbor og delt basert på de øverste 3 cm og ellers basert på to synlige jordlag. For jorden fra Groalia ble 0-3 cm laget delt i to basert på et synlig mineral lag (2-3 cm) og et organisk lag (0-2 cm). Jordlagene fra Nerlifjellet hadde 13-97% organisk material, mens jordlagene fra Groalia hadde 5-40%. Det ble også funnet forskjeller i pH. En kornfordelingsanalyse gjennomført for de jordlagene med <20% organisk materialet viste at mineraljord fra Nerlifjellet først og fremst besto av sand (73%) mens mineraljorden fra Groalia i hovedsak besto av silt (39-57%).

Aktivitetskonsentrasjonen til $^{137}$Cs i jord fra Nerlifjellet ble identifisert til å være mellom 200 og 900 Bq/kg tørr jord avhengig av jordlaget, mens for jorden fra Groalia var aktivitetskonsentrasjonen mellom 36 og 1300 Bq/kg. Aktivitetstettheten, per cm, til $^{137}$Cs i jorden fra Nerlifjellet ble identifisert til å være 1.0-1.1 Bq/m$^2$ per cm, mens for jorden fra Nerlifjellet var dette mellom 0.19 Bq/m$^2$ og 6 Bq/m$^2$ per cm. I jorda fra Nerlifjellet var $^{137}$Cs fordelt mer jevnt nedover i jorda enn i jorda fra Groalia, hvor $^{137}$Cs i hovedsak ble funnet i de øverste 3 cm (86%). Basert på dette kan det tyde på at $^{137}$Cs er mer mobil i jorden fra Nerlifjellet enn i jorden fra Groalia, noe som var foreslått at var knyttet opp mot forskjeller i mengde organisk material.

En sekvensiell ekstraksjon av flere av jordlagene fra begge prøvelokasjonene indikerte at $^{133}$Cs var bundet sterkere i jord med <40% organisk materiale sammenlignet med jord med mer organisk materiale. Dette støttet ideen om at $^{137}$Cs lekker i en større grad fra den organiske jorden fra Nerlifjellet enn den mer mineralholdige jorda fra Groalia og derfor blir transportert lenger ned i jorden.

Aktivitetskonsentrasjonen til $^{137}$Cs i jordfraksjonene fra kornfordelingsanalysen ble målt og resultatet viste en større aktivitetskonsentrasjon av $^{137}$Cs i leirefraksjonen enn de andre fraksjonene og en aktivitetskonsentrasjon som var 9-30 ganger høyere enn aktivitetskonsentrasjonen av $^{137}$Cs i den ubearbeidede prøven. I minerallagene var $^{137}$Cs i hovedsak bundet til silt og leirefraksjonene (53-97%) og i mindre grad bundet til sandfraksjonen (<33%). Sammenligning av prøver fra Groalia og Nerlifjellet viste at andelen av $^{137}$Cs i leirefraksjonen var lavere i 7.5-13 cm laget fra Nerlifjellet enn i 8-13 cm laget fra Groalia. Det ble foreslått at dette kunne knyttes til en lavere mengde leire i jorden fra Nerlifjellet. Selv om en det kan være en stor mengde sand og silt i jorda, indikerte resultatene at en større mengde leire påvirker sannsynligheten for binding av $^{137}$Cs til sand og silt negativt. Dette kan være på bakgrunn av leirens høye affinitet for $^{137}$Cs. Resultatene viser at det er essensielt å identifisere mengden av organisk materiale og leire i jorda for å kunne forstå mobiliteten av $^{137}$Cs i jord og sannsynligheten for remobilisering av $^{137}$Cs i avrenning.

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# 1. Introduction

## 1.1 Background

There has been much interest in radionuclides and their behaviour in the environment after the release of several anthropogenic radionuclides due to atomic weapon testing, mining, and accidents at nuclear facilities (Chernobyl and Fukushima) (Choppin et al., 2013). The consequences of exposure to both natural and anthropological radionuclides have been a subject of research, where ionizing radiation has been shown to be able to ionize molecules and create free radicals which can cause cell damage (Sire, 1995) . Ionising radiation can also directly impact the DNA and thus cause cell damage (Choppin et al., 2013; Sire, 1995).

Cesium-137 has been an anthropological radioisotope of interest for several years, being a fission product with a relatively long half-life and emitting relatively high energy β and γ radiation (Ashraf et al., 2014; Dubchak, 2017). Cesium can be easily taken up by biota and thus cause internal radiation dose in addition to the external dose (Karadeniz et al., 2015; Walker, 2012). The Chernobyl accident in 1986 is a substantial source of $^{137}$Cs in Norway (Henriksen, 1987). A study by Backe et al. (1987) estimated that a total of about 2300 TBq of $^{137}$Cs was deposited in Norway and that about 10% of the country received more than 20kBq/m$^2$ of $^{137}$Cs. The study also showed that the mid and central part of the country was especially affected (Gjelsvik & Steinnes, 2013). The Vefsna river and its catchment area is situated in the southern part of Nordland which was especially impacted, see figure 3.1 and thus chosen as the research location for this study (Baranwal et al., 2020; Liland et al., 2001).

It is known that soil accumulates many ground deposited radionuclides with long half-lives (Oughton et al., 1992). Thus, knowledge of their behaviour in the soil is important to be able to understand their behaviour in the ecosystem. Knowledge about the binding, mobility, and sorption of radionuclides in the soil is important for understanding the uptake of radionuclides from soil to plants (Fawaris & Johanson, 1995; Manigandan & Chandar Shekar, 2014). Clay is known to minimize the mobility of $^{137}$Cs and thus somewhat control the behaviour of $^{137}$Cs in the environment (Koarashi et al., 2018). Several other factors have been shown to influence the behaviour of $^{137}$Cs in soil. These include but are not limited to the speciation of $^{137}$Cs, soil texture, mineralogy, the content of organic matter, amount of exchangeable cation, and soil type (Karadeniz et al., 2015).

Dynamic models can be used to assess the long-term impact of a radionuclide. In such a model, there is a need for consideration of the source of the radionuclide,  deposition, factors affecting transport and mobility, ecosystem transfer, biological uptake, and accumulation in sediments, soil, or biota (Salbu, 2016). The Centre for Environmental Radioactivity (CERAD) is making a model of the transport of $^{137}$Cs from catchment areas of rivers, into rivers, and further into the estuarine environment along the coast. An important source of $^{137}$Cs in aquatic systems is wash-off from the soil surface or erosion of Cs- associated particles. This includes both soluble $^{137}$Cs transported through water and wash-off of soil-bound $^{137}$Cs, or transport of particles (Garcia-Sanchez & Konoplev, 2009). Thus, to model the transport of $^{137}$Cs from terrestrial to aquatic environment, it is necessary to take the behaviour of $^{137}$Cs in soil with different soil contents into account.

## 1.2 Main objectives and hypothesis

The goal of this thesis was to determine the activity of $^{137}$Cs in soil from the catchment area of Vefsna and to investigate if soil content has an impact on the mobility, binding, and distribution of $^{137}$Cs in soil. The objectives of this thesis were:

- To characterize the soil by measuring pH, organic matter content, dry matter content and performing a grain size analysis.
- To investigate the distribution of $^{137}$Cs in the two soil types
- To investigate the proportion of $^{137}$Cs found in different soil particle size fractions (sand, silt and clay)
- To investigate the binding and mobility of $^{137}$Cs in soil by preforming a sequential extraction.

The hypothesis is:

H1: Cesium-137 is more bound to soil with a high percentage of organic material than soil types with a lower percentage.

# 2. Theory

## 2.1 Sources of cesium: The Chernobyl accident

Cesium-137, being an anthropogenic radioisotope, has generally been emitted through nuclear weapon testing during the 1950s and 1960s and through accidents in nuclear facilities (Ashraf et al., 2014; Gupta & Walther, 2017; Heldal et al., 2021). Due to the emitted $^{137}$Cs from weapon testing being considered decayed, the Chernobyl accident in 1986 and the Fukushima accident in 2011 are considered the major sources of $^{137}$Cs in the current environment (Ashraf et al., 2014). The Chernobyl accident is regarded as the primary source of $^{137}$Cs in Norway, where south-eastern winds and rain at a critical time after the Chernobyl accident led to the transportation and fallout of about 2300 TBq $^{137}$Cs into the environment of Norway. (Gjelsvik & Steinnes, 2013; Heldal et al., 2021; Henriksen, 1987).

In 1986, one of four reactors at the Chernobyl nuclear power plant in Ukraine exploded and started to burn, which lead to the release of about 1100 PBq of radioactive debris into the atmosphere (Konoplev et al., 2020). Cesium-137 is one of the radionuclides in focus after the accident, and since about 85 PBq was emitted, the Chernobyl accident is considered one of the major sources of $^{137}$Cs into the environment (Ashraf et al., 2014; UNSCEAR, 2011). A large part of $^{137}$Cs occurred in mobile form as condensation particles and was transported a longer distance than other radionuclides emitted (Konoplev, 2020). In Norway, the areas Gudbrandsdalen, Valdres, Trøndelag and Southern parts of Nordland were the areas most affected, see figure 2.1(Liland et al., 2001).

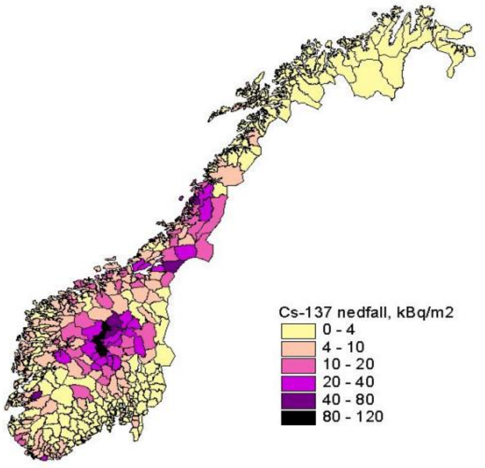

*Figure 2.1. Map showing the measured deposition of $^{137}$Cs in Norway in 1986. (Liland et al., 2001)*

## 2.2 Properties of $^{137}$Cs in soil

Cesium was discovered in 1860 in a Bavarian mineral spring and is the rarest of the group one alkali metals (Avery, 1996). Cesium has 40 known isotopes, where only $^{133}$Cs is stable and occurs naturally in soil (Burger & Lichtscheidl, 2018; Gupta & Walther, 2017; Kim et al., 2016). Two of the radioactive isotopes of cesium, $^{137}$Cs and $^{134}$Cs, has been significant contributors to radiation doses in the environment (Gupta & Walther, 2017). This is due to several characteristics of cesium and the amount produced. Cesium-137 is a high-yield fission product of $^{235}$U, which is a fissile radionuclide used in reactors and weapons, while $^{134}$Cs is an activation product (Dutta & Lodhari, 2018; Gupta & Walther, 2017; Lin et al., 2019). After some time, the significance of $^{137}$Cs compared to $^{134}$Cs increases due to the longer half-life of $^{137}$Cs (30.17 years) (Dubchak, 2017). Cesium-137 emits both $\beta^-$ and $\gamma$ radiation. Disintegration and emission of $\beta^-$ radiation results in $^{137}$Ba and meta stable $^{137m}$Ba from $^{137}$Cs. Barium-137m has a short half-life of about 153 seconds and rapidly decays to $^{137}$Ba through $\gamma$-radiation with an energy of 0.662 MeV (Ashraf et al., 2014; Burger & Lichtscheidl, 2018). Due to the high percentage of decay being $^{137m}$Ba, 95%, the $\gamma$-radiation is used for quantitative and qualitative measurement of $^{137}$Cs (Salih et al., 2006). A decay scheme of $^{137}$Cs is shown in figure 2.2.

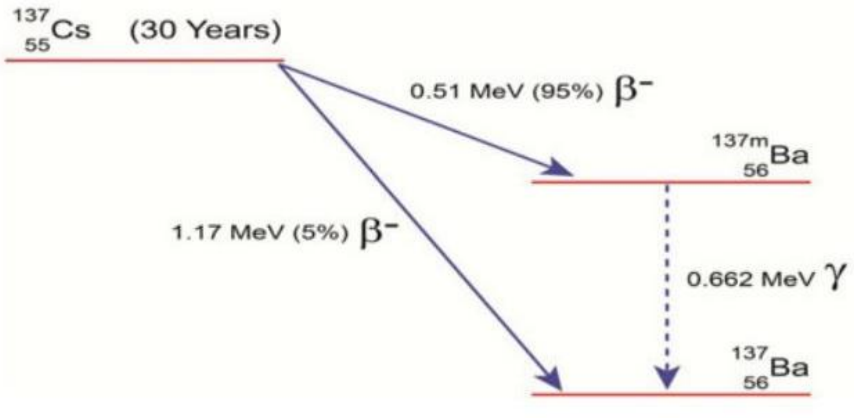

*Figure 2.2 Decay scheme of $^{137}$Cs (Ashraf et al., 2014).*

Cesium is primarily apparent in water and soil as $Cs^+$ and forms few complexes (Lin et al., 2019). Because of its similarity to the essential element potassium, cesium is easily taken up by biota and transported through the environment (Gupta & Walther, 2017). Stable cesium is not toxic in natural concentrations. Still, by uptake of $^{137}$Cs, biota will be exposed to internal radiation in addition to external radiation due to the emission of $\beta^-$-particles of a relatively high energy and $\gamma$-radiation, figure 2.2 (Ashraf et al., 2014; Dubchak, 2017).

## 2.3 Mobility and transport of cesium in terrestrial systems

Several reports have shown that the consequence of $^{137}$Cs being released into the environment highly depends on the movement in soil (Gupta & Walther, 2017). Radiocesium deposited in the environment is distributed between aquatic and terrestrial systems (Cornell, 1993). The distribution depends on the mobility and bioavailability of radiocesium in the environment. High mobility $^{137}$Cs is easily transported through terrestrial systems and into the aquatic systems where it can be transported long distances. The mobility and bioavailability of $^{137}$Cs differ with different chemical speciation (Konoplev, 2020). Chemical speciation is the physico-chemical form of the radionuclide that occurs in the environment (Burger & Lichtscheidl, 2018). Cesium in ion, $Cs^+$, form has a high solubility in water, making it likely to be transported in soluble forms through the ground water into the ecosystems and taken up by plants (Burger & Lichtscheidl, 2018; Cornell, 1993).

Studies by  Hilton et al. (1992) and Konoplev (2020) suggested condensation origin of the particles containing $^{137}$Cs found in areas far from Chernobyl. Hilton et al. (1992)  studied $^{137}$Cs in airborne particulate material in the UK shortly after the Chernobyl accident and found that >85% of $^{137}$Cs were water-soluble and exchangeable. Based on this result, it could be assumed that the radiocesium also has high mobility through the ecosystem and is transported fast both horizontally and vertically through the soil. Still, studies have shown that radioactive cesium is mainly found in the topsoil layer (Dubchak, 2017; Park et al., 2021). The topsoil layer is described to be the top 10-25 cm of the soil (Weil & Brady, 2017b).

The transport of cesium through soils and into surface water is highly dependent on the different sorption mechanisms in soils and thus this has been an extensive area of research (Lofts et al., 2002). After fallout on the soil's surface, radionuclides interact in physicochemical and biological processes resulting in a change of speciation. One of these processes is the sorption to solid particles, where exchangeable forms of cesium fixate and transfer to colloid forms (Konoplev, 2020). The different processes of transport and binding of stable and radioactive cesium is illustrated in figure 2.3.

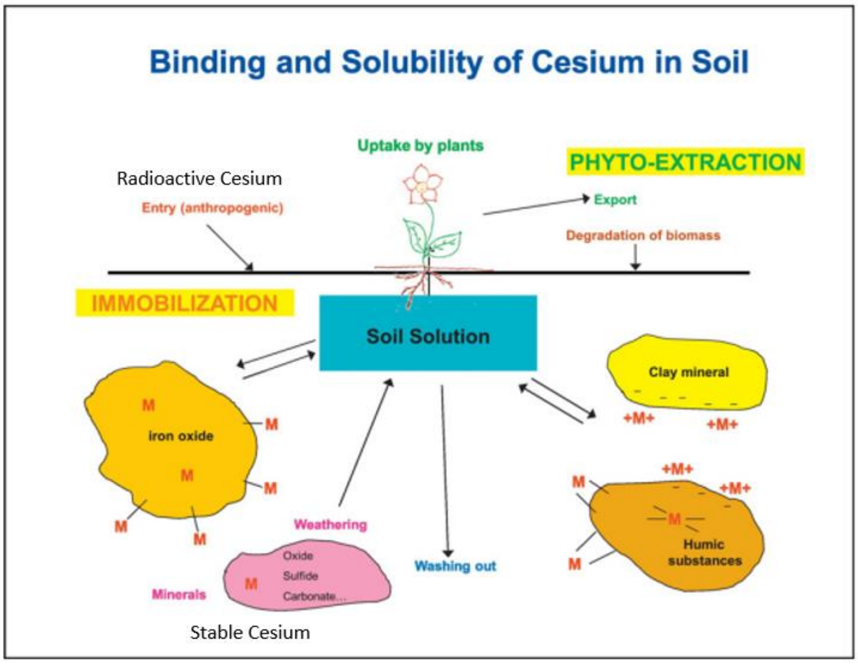

*Figure 2.3: Process of transport and mobility of stable and radioactive cesium in terrestrial environment. Figure is adapted after Burger and Lichtscheidl (2018).*

### 2.3.1 Sorption of cesium onto minerals

Several studies have found that a primary reason for the limitation of mobility and vertical migration rate of $^{137}$Cs is the sorption of cesium on soil minerals (Dumat & Staunton, 1999; Gupta & Walther, 2017; Park et al., 2021). The sorption abilities have been found to depend on the surface area, where the sorption increases with increasing surface areas. The surface area for sand, silt and clay area as follows.: clay > silt > sand, resulting in high sorption abilities for clays (Tan, 2011).

The sorption ability of sand, silt and clay also depend on the minerals present. The mineralogy of clay fraction differs fundamentally from sand and silt and some clay minerals have been observed to have high affinity for cesium (Cornell, 1993; Hillel, 2008). Sand mainly consist of quartz, which generally have a low sorption ability due to it being electrically neural (Barends, 2011). Generally, silt have been regarded to consist of the same minerals as sand (Hillel, 2008). However, clay minerals have been found in sand by Szabó et al. (2012).

Clay minerals contain tetrahedral and octahedral sheets of silicon oxide and aluminium oxide, alternating in different structures. A negative charge can be obtained by Si (IV), or Al (III) being exchanged with ions of comparable size with a lesser charge. This results in a lower net charge, which is compensated by cations associating with the surface of the clay. Cations associated reversibly with surfaces of particles can be exchanged by other cations, like $Cs^+$, in a process called cation exchange (Cornell, 1993; Manahan, 1994). Clays can be separated into two groups based on the order and distribution of the sheets repeating through the clay. 1:1 clays have one tetrahedral and one octahedral sheet alternating, while 2:1 clays have one octahedral sheet between two tetrahedral sheets (Weil & Brady, 2017b). Illite and montmorillonite are examples of 2:1 clays, while kaolinite is an example of a 1:1 clay (Barends, 2011).

The reaction between clay and $Cs^+$ can be through a weak electrostatic attraction between cesium and the surface of the clay or through strong bonds formed when cesium and the oxygen-donor sites on clay material share electrons (Fan et al., 2014). Radioactive cesium can be adsorbed irreversibly and specifically by 1:2 clays, while 1:1 clay has a lower capacity and specificity for cesium adsorption. This is due to the structure of 1:1 clay being stabilized by hydrogen bonds, which only allows partial negative charges on the clay's planar surface, resulting in reversibly bound cesium (Cornell, 1993).

For 1:2 clays, also called micaceous clays, the cations ensuring the stability of the interlayers of the clays can be exchanged by cesium ions. In addition to reversibly bound cesium on the planar surface, this results in the possibility of sorption of $Cs^+$ selectively on the interlayer. Due to low hydration energy, $Cs^+$ can shed its hydration shell when entering the interlayer resulting in dehydration and collapse of the interlayers. This causes irreversibly bound cesium (Cornell, 1993; Park et al., 2021). Weathering of interlayer edges of some clays causes the creation of cesium selective frayed-edges sites (FES). The sorption between FES and cesium is considered to be slowly reversible due to the cation exchange being limited to cations of similar size (Børretzen & Salbu, 2002). The amount adsorbed onto FES or in interlayers depends on the type of micaceous clay (Fan et al., 2014). Figure 2.4 shows an illustration of the bounding of cesium onto 2:1 clay. Depending on the type of and amount clay present in the soil, the result is reversible, irreversible and slowly reversible sorption of cesium.

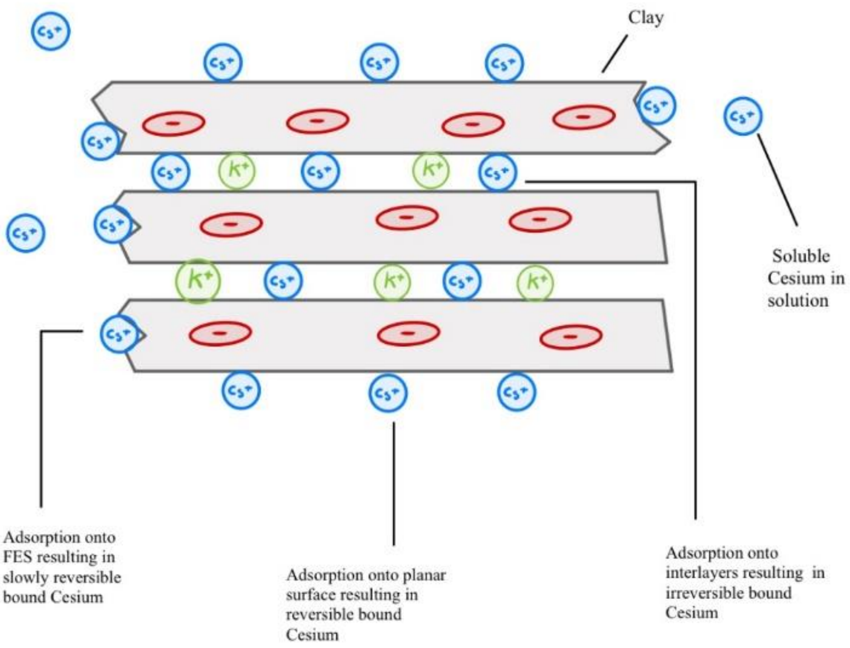

*Figure 2.4: Binding of cesium onto clays. Figure adapted after Børretzen and Salbu (2002).*

Several studies have investigated the effect of time on the sorption of cesium onto inorganic materials. Depending on the minerals, a bulk of the cesium initially sorb within hours, then the rest sorb within days and weeks (Cornell, 1993). However, studies of Fukushima soil showed $^{137}$Cs in water-soluble fractions two years after the release, indicating that the sorption of $^{137}$Cs happens over a longer time (Saito et al., 2014). Reports have also shown that the adsorption of cesium gets stronger over time (Saito et al., 2014).

The impact of the concentration of cesium on the sorption ability of clays has been investigated with a range of concentrations and clay contents(%). Shenber and Eriksson (1993) found that the fraction of cesium adsorbed decreased when increasing cesium concentration regardless of clay content. Other studies showed a linear connection between adsorption of cesium onto minerals and the cesium concentration at lower concentrations ($<10^{-8}$M), while at a higher concentration, no such connection has been observed. $K_d$, which is the distribution coefficient between solid and liquid, can be used to predict the movements of radionuclides in the environment. $K_d$ was observed to decrease for an increased concentration of cesium, indicating higher mobility (Cornell, 1993; Kumar et al., 2020).

In water, cesium is present as $Cs^+$ over a wide pH range. However, pH can affect the exchange properties of the adsorbing substrate. This dependence has been found in several clay minerals. An example is montmorillonite clays, where a higher adsorption was observed with a rising pH (Cornell, 1993). Due to the sorption process being based on an ionic exchange, the adsorption of cesium has been observed to be lower with a higher ionic strength (Cornell, 1993). $Na^+$, $K^+$, $Ca^{2+}$ and $Mg^+$ are the ions which compete the most with cesium for adsorption sites on minerals. The effect has been observed to be depending on the concentration of cesium relative to the concentration of the competing ion. Divalent ions competed stronger for the adsorption sites than univalent ions (Cornell, 1993).

### 2.3.2 Sorption of cesium onto hydrated iron oxides

Several studies have shown that metal ions can be adsorbed on the surface of particles through complexation with hydroxyl groups. Thus, hydrated iron oxides, also called ferrihydrites, have been used to remove heavy metal ions from solutions (Gossuin et al., 2002). The adsorption of cesium ions onto these ferrihydrates has been studied and by some studies found to be negligible (Cornell, 1993). Still, other studies have found significant effects. Kikuchi et al. (2019) found significant adsorption of $Cs^+$ on biogenic hydrated iron oxides, while no adsorption for abiogenic ferrihydrite. Biogenic ferrihydrite is formed by organic means and is found in ground water (Kikuchi et al., 2019).

### 2.3.3 Sorption of cesium onto organic material

The sorption mechanism of clays has been regarded as the main reason for the retention of cesium in soil. Still, some interaction between organic material and cesium has been found. Bonn and Fish (1993) found electrostatic interactions between the group 1 alkali metals $Li^+$, $Na^+$ and $K^+$ and humic acid in humic substances within soils with a pH between 3 and 8. Based on the similarities of structure within the group 1 alkali metals, it was suggested similar interactions for $Cs^+$. Lofts et al. (2002) found a nonspecific binding between the negatively charged dissociated humic acid and $Cs^+$, see figure 2.5, using the Humic Ion Binding Model V.

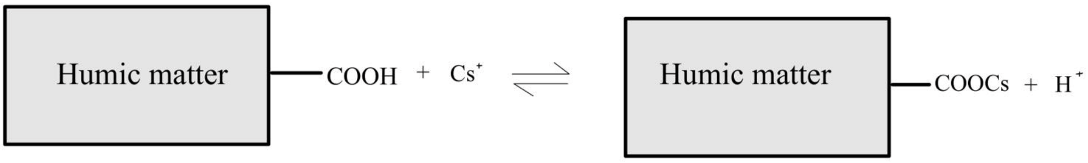

*Figure 2.5: Illustration of how cesium ions can bind to humic matter in organic material based on the Humic ion binding model V (Lofts et al., 2002).*

Due to the high sorption of cesium in clay, it was found that the role of humic acid in the immobilisation of cesium is minor even for soils with a high percentage of organic matter

(Dumat & Staunton, 1999; Lofts et al., 2002; Saito et al., 2014). The adsorption between organic matter and cesium is non-specific and depends on the cation exchange capacity of the organic matter and the amount of cesium compared to other cations (Dumat & Staunton, 1999).

### 2.3.4 Soil properties impacting mobility and distribution of cesium in soil

Cesium is known to sorb to mineral clays in soils, thus reducing radiocesium's mobility in soil. Koarashi et al. (2012) found a positive correlation between clay content and retention rate of $^{137}$Cs in soil from Fukushima after the Fukushima accident, indicating that the amount of clay minerals impacts the distribution and mobility of cesium in soil. The amount of the different types of clay minerals also affects the mobility and distribution of cesium due to differences in sorption between the types of clay minerals. For instance the amount of FES available in the soil will highly impact the selective sorption of cesium onto clays, thus impacting the mobility and distribution of cesium (Park et al., 2019).

In soil with a high percentage of organic matter, cesium has been observed to stay bioavailable over a longer time and being weaker adsorbed to the soil. This can be explained by the lover retention ability of organic material in the soil (Dumat & Staunton, 1999). However, organic matter also seems to impact the sorption ability of clays, by modifying and decreasing the clays' ability to immobilise $^{137}$Cs and thus heighten the bioavailability of $^{137}$Cs in the soil (Staunton et al., 2002).

The mobility of cesium have been observed to be impacted by pH. Giannakopoulou et al. (2007) studied the sorption of added stable cesium in four different types of soil in environments with pH between 5 and 10 using a batch experiment. For all of the soil types, an effect of pH on the sorption was found, with a similar pattern for all soil types. The highest sorption, and thus the lowest mobility of $^{137}$Cs, was observed to be around pH 8. The lowest sorption was around pH 5. It was suggested that an explanation of this was $H^+$ ions competing with $Cs^+$ ions, thus a lager concentration of $H^+$ result in less cesium adsorbing in the soil (Giannakopoulou et al., 2007).

## 2.4 Cesium-133 as a stable analogue for $^{137}$Cs in soil

In radiochemistry, a primary assumption is that radioactive isotopes are chemically identical to their stable isotopes, thus having the same chemical and physical properties. Therefore, uptake and mobility are the same (Choppin et al., 2013). Based on this assumption, $^{133}$Cs could be a stable analogue for $^{137}$Cs and could be used to predict the transfer of radiocesium in the ecosystem. However, $^{133}$Cs have been shown to be differently distributed than $^{137}$Cs in soil (Dubchak, 2017).

Cesium-133 occurs naturally, and the primary natural source is pollucite which is an aluminosilicate mineral and is released due to weathering of rocks materials containing pollucite, while $^{137}$Cs is anthropogenic and added to the environment through deposition and precipitation (Dubchak, 2017; White & Broadley, 2000). Elements entering the terrestrial environment could enter as ions, could be bound to particle surfaces, or deposit as particles. After some time, an equilibrium between the dissolved and bounded fraction of the elements occurs (Tan, 2011). Stable cesium, found naturally fixed in the soil, will have reached this equilibrium. Radioactive cesium will need some time to fixate in the soil before reaching equilibrium, resulting in higher mobility of $^{137}$Cs compared to $^{133}$Cs (Saito et al., 2014). However, after some time, $^{137}$Cs are expected to reach a steady state, and $^{133}$Cs can be used as a stable analogue predicting the long-term movements and behaviour of $^{137}$Cs (Guillén et al., 2014). Measuring stable isotopes and comparing with the radionuclide could give information of the equilibrium state of the radionuclide in the ecosystem (Oughton et al., 1992).

# 3. Material and Method

## 3.1 Site and sampling

The research location for this study is the area of the municipality Hattfjelldal, situated within the catchment area of the river Vefsna. The river Vefsna is situated in the southern part of the county Nordland which was especially impacted by the Chernobyl accident, see figure 3.1.

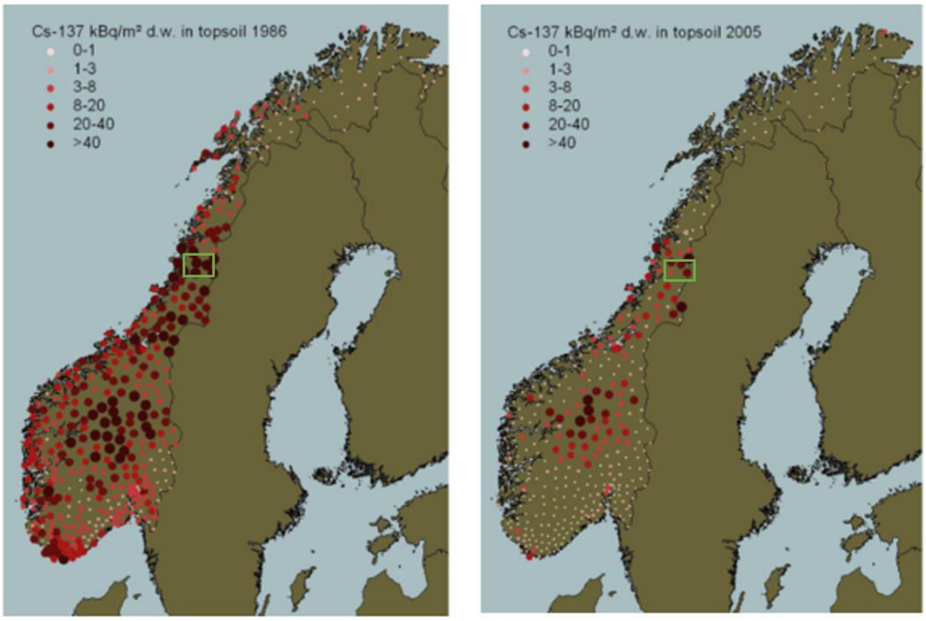

*Figure 3.1: Maps showing the measured activity concentration (kBq/m$^2$) in Norway in 1986 and 2005. The approximate area of the research is marked (Gjelsvik & Steinnes, 2013).*

The river Vefsna is 163 km long and runs into the Vefsnfjord. The catchment area of Vefsna is considered to be 4229 km$^2$, and through simulation, the mean yearly precipitation-runoff between 1961 and 1990 in the area was calculated to be > 4000 mm per year (Beldring et al., 2003; Molvær, 2010). Figure 3.2 shows the catchment area of the river Vefsna with the two research sites marked.

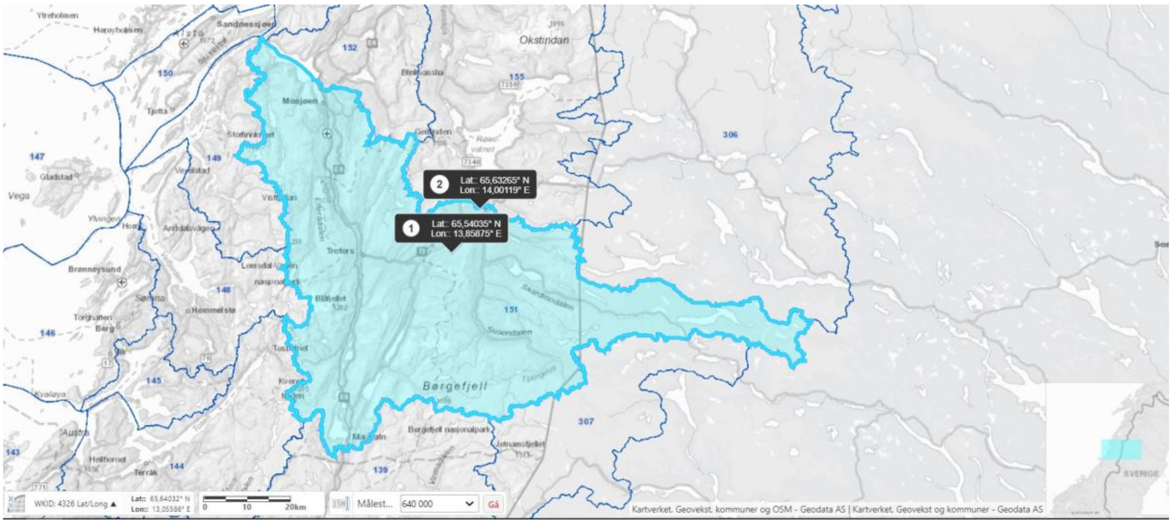

*Figure 3.2: Map showing the coordinates of the two sites within the Catchment area (NVE.atlas.no, u.d).*

In 1986, soil samples from Hattfjelldal showed an activity of 18.77 kBq/m$^{2,}$ while the neighbouring municipalities Grane and Vefsen had activities of 21.13 kBq/m$^2$ and 24.38 kBq/m$^{2,}$ respectively (Backe et al., 1986). In 2014, the Geological Survey of Norway (NGU) measured the $^{137}$Cs activity in the area around Hattfjelldal using helicopters. These measurements showed a heightened activity around the areas Nerlifjellet and Groalia, see figure 3.3 (Baranwal et al., 2020). Nerlifjellet (site 1) and Groalia (site 2) were chosen as the two locations for this study due to the high activity of $^{137}$Cs.

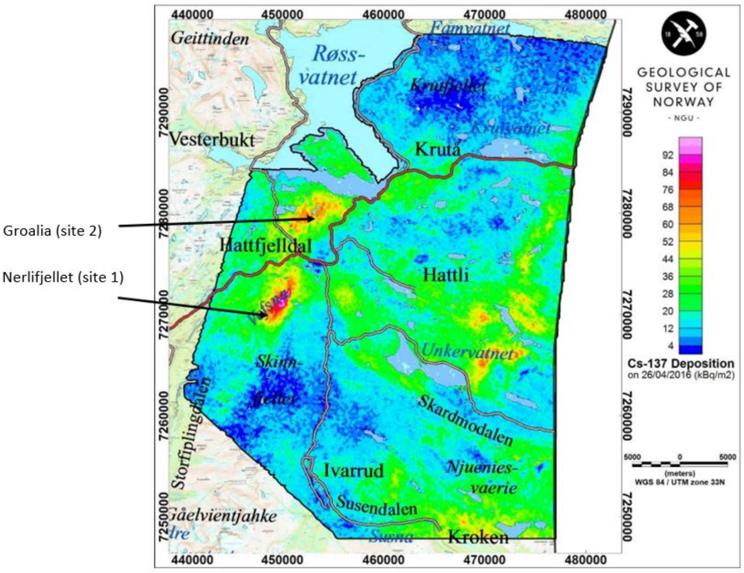

*Figure 3.3: Map showing the measured activity concentration of $^{137}$Cs in the area Hattfjelldal in 2014 (Baranwal et al., 2020) with the sites of sampling being showed.*

### 3.1.1 Site 1: Nerlifjellet

Site 1 was located at 433 meters above sea level in the area Børgefjell at the foot of the westside of Nerlifjellet. The site was an open area with a diameter of about 4 meters, figure 3.4 (A). Most of the trees in the area were spruce, birch, and roe. The soil type in this area was observed to have a clear organic layer on top, a transition layer of hummus and a grey mineral-based layer, figure 3.4 (B). Information about the site is given in table 3.1.

*Figure 3.4: Picture showing the nature of Nerlifjellet (Site 1)(A) and the soil profile (B)*

### 3.1.2 Site 2: Groalia

Site 2 was located 486 meters above sea level in Grolia. This was an opening in the forest with a diameter of about 5 meters. Most of the trees in the area were birch, figure 3.5 (A). The soil type in this area was observed to have a podzol like profile with an organic layer on top, a grey mineral layer, and an iron-based rust coloured mineral layer, see figure 3.5(B & C). Information about the site is given in table 3.1

*Figure 3.5: Picture showing the nature of Grolia (site 2) (A) and the soil sampled (B & C)*

### 3.1.3 Sampling method and equipment

Three soil samples as bore cores were collected from each of the sites. The samples were taken within an area with a radius of approximately 2 meters. The soil samples were taken with a 10.8 cm diameter, 13 cm deep soil sampler corer, see figure 3.6.

*Figure 3.6: The soil sampler corer. The diameter and depth of the corer are shown.*

The collected soil cores were split in situ into three soil samples based on the layers of the core by using a knife. The top 3 cm of the core were separated from the rest of the core for all samples. The rest of the core was divided into two parts based on the two visible soil layers. The soil was collected into zip-lock bags.

For the soil from Groalia, the 0-3 cm layer had a clear 1 cm mineral layer, which was later separated from the sample in the lab. Based on this separation, the soil cores from Groalia were divided into four layers. The layers for the soil cores from Nerlifjellet and Groalia are shown in figure 3.7. A summary of site information with observed characteristics of the soil is given in table 3.1

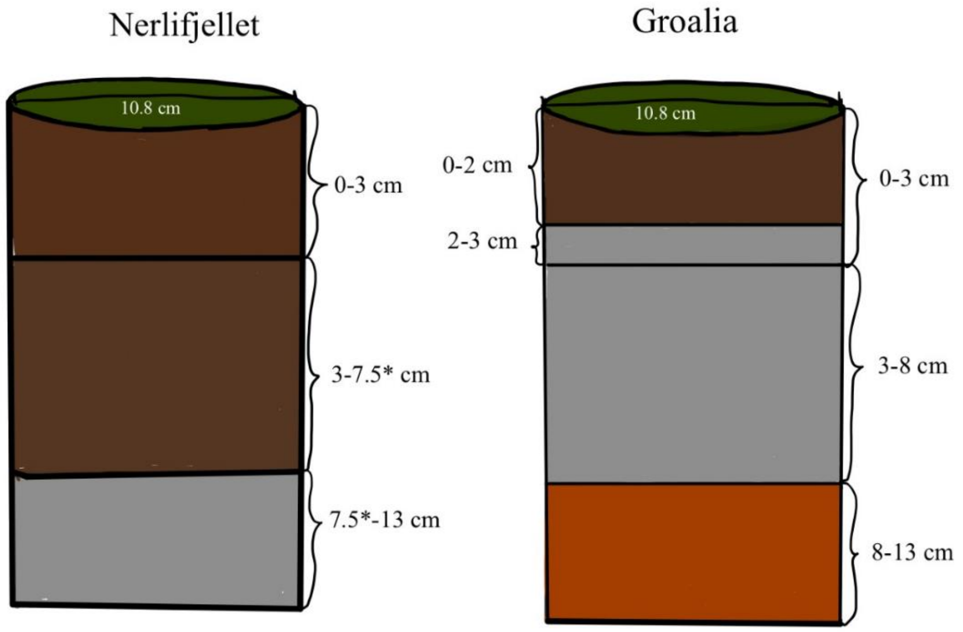

*Figure 3.7: The dividing of the soil cores from Nerlifjellet and Groalia. The brown layers represent layers with a visible large amount of organic material, the grey layer represent the visible mineral layers and the orange layer represent the rust-coloured layer. *Mean value based on the three cores, see table A1 in appendix A*

*Table 3.1: Site and soil profile information. *Mean value based on the three cores, see table A1 in appendix A.*

| Sites | GPS | Height above sea level | Description of the area | Layers | |
|---|---|---|---|---|---|
| | | | | | Observed characteristics |
| Site 1: Nerlifjellet | 65.540353°N 13.858745°E | 433 meters | Open area at the foot of the mountain. Spruce, birch, and roe as the dominating tree types. | 0-3 cm | Organic layer |
| | | | | 3-7.5* cm | Transition hummus layer |
| | | | | 7.5*-12 cm | Grey mineral layer |
| Site 2: Groalia | 65.632647°N, 14.001185°E | 486 meters | Forest area. Birch is the dominating tree type. | 0-2 cm | Organic layer |
| | | | | 2-3 cm | Grey mineral layer |
| | | | | 3-8 cm | Grey mineral layer |
| | | | | 8-12 cm | Rust coloured mineral layer |

## 3.2 Soil characterization

### 3.2.1 Sample preparation

To limit microbiological growth, the soil samples were kept between 0-4 °C in a dark place, and the samples were kept wet and untreated. The soil samples were weighted with the purpose of calculating the activity density (Bq/m$^2$ per cm) of $^{137}$Cs in each layer. Due to the goal of evaluating the binding and mobility of $^{137}$Cs in the soil, only small fractions of the samples were dried. The samples used when measuring $^{137}$Cs in each layer were freeze-dried using Epsilon 2-4 LSC for 72 hours. These samples were also used for grain analysis and pH measurements.

### 3.2.2 pH

The pH of the soil was measured using a protocol based on Kalra (1995). From each of the layers in each of the soil cores from both sites, 5 ml freeze-dried soil and 10 ml Milli-Q water were added to a 50 ml glass beaker. The solution was shaken by hand for 1 minute and kept overnight. The solutions were swirled, and pH was measured using Inolab WTW pH/Ion 735 until the number was stable for over 30 seconds. The pH meter was calibrated using pH 4 and pH 7 buffers.

### 3.2.3 Organic content and dry matter

The dry matter content and organic matter content was determined using the procedure described in Øien and Krogstad (1989). Three samples from each layer from each soil core (N=9) from both sites were analysed. From untreated soil, 2-3 g of soil was weight (M1) into porcelain crucibles and dried at 105°C using overnight using a Termaks drying oven (NLS ISEM No. 1700040). After drying, the crucibles were cooled in an exicator and weighted again (M2). The dry matter content was calculated using formula 1:

$$\%\, Dry\ matter = \frac{M2 \times 100\%}{M1} \tag{1}$$

The crucibles with lids were placed in a Carbolite muffle furnace where the temperature gradually increased to 550°C and was left overnight. The crucibles were cooled in an exicator and weighted (M3). The organic content was calculated using formula 2:

$$\%Organic\ content = \frac{(M2 - M3) \times 100}{M2} \tag{2}$$

### 3.2.4 Grain size characterization

Grain size characterization was only completed for the layers with a lower organic content than 20%. For the 7.5-13 cm layer from Nerlifjellet and the 3-8 cm layer from Groalia, about 10-50 g of freeze-dried soil was separated between the different grain sizes. Due to a lack of material for the 2-3 cm layer from Groalia, a combined sample was prepared by combining about 5-10 g of dried soil from each of the three cores into one sample. Such a combination sample was also obtained from the 8-13 cm layer from the same site to ensure high enough activity to make it possible to determine activity in different fractions later. To consider these combined samples' precision, such samples were also obtained for the 7.5-13 cm layer from Nerlifjellet and the 3-8 cm layer from Groalia.

Before separating in different grain sizes the dried samples were sieved through a 2 mm sieve and weight. Organic matter was removed by adding 20 mL distilled water per 10 g of dried soil while warming at a sand bath at about 75-80°C and adding 30% $H_2O_2$ until all organic matter had reacted, see figure 3.8A.

Thereafter the sand fraction was separated from the clay and silt fraction by wet sieving the solution through a 63μm sieve using just enough water to sieve all the silt and clay fractions through (figure 3.8 B). The clay and silt fractions were collected in a 1 L Nalgene flask. If the clay and silt fraction was more than 1 L, plastic and glass beakers of 1L was used. The sand fraction was collected in pre-weight 100 ml beakers and dried at 105°C overnight using Termaks drying oven (NLS ISEM No. 1700040), cooled in an exicator then weighted.

The collected clay and silt fractions were separated using a method based on stokes law. Stokes law describes the velocity(V) of a particle falling through a fluid. The velocity is proportional to the gravitational force (g), the difference in density of the particle and the fluid ($D_s$-$D_f$), squared off the effective diameter of the particle (d) and inversely proportional to the viscosity of the fluid ($\eta$), see formula 3 (Weil & Brady, 2017b).

$$V = \frac{d^2 g(D_s - D_f)}{18\eta} \tag{3}$$

The gravitational force g is 9.81 N/kg, the density of water ($d_f$) is 1000 kg/m$^3$, the used density for most soil particles ($D_s$) is 2650 kg/m$^3$, and the viscosity of water ($\eta$) at 20°C is 1000 Ns/m$^2$ (Weil & Brady, 2017a; Weil & Brady, 2017b). The smallest silt has an effective diameter (d) of 0.002 mm (Weil & Brady, 2017b).

Using the formula 3, silt is expected to be sedimented 2.6 cm, 5.2 cm and 7.8 cm after two hours, four hours and six hours, respectively. Clay has a lower effective diameter and sedimented a shorter distance in the bottle.

The 1L Nalgene bottles were marked at 7.8 cm (six hours), 5.2 cm (four hours) and 2.6 cm (two hours) and the silt and clay solution were added to the flask up to the mark depending on the sedimented time decided (figure 3.8 C). The content of the flask was stirred vigorously using a glass rod and left for the appropriate amount of time, depending on the chosen sedimentation time. The silt fraction was sedimented at the bottom of the flask, below the zero mark, at about 1 cm above the bottom, while the clay fraction remained in solution. The clay fraction was drained using a peristaltic pump, Watson Malow SCI 323, and collected in 2 L beakers (figure 3.8 D). Water was added to the Nalgene bottles, and the solutions were separated based on sedimentation for 6 hours three times, for 4 hours two times and finally 2 hours 1-3 times. The silt fraction was collected in preweighted 125 ml containers used for measurement using Ge-detector and freeze-dried for 72 hours using Epsilon 2-4 LSC.

The clay fraction collected in the 2 L containers was set to sediment for 6 days after adding 7.5 ml $CaCl_2 \times 2H_2O$ to help with the sediment process (figure 3.8 E). A blank sample made with 7.5 ml $CaCl_2 \times 2H_2O$ was created to measure the weight of the added $CaCl_2$. The clay fraction sedimented to the bottom of the containers, and the water was drained using the peristaltic pump. The clay fraction was collected in weighted 125 ml containers before measurement of $^{137}$Cs and freeze-dried for 72 hours using Epsilon 2-4 LSC.

*Figure 3.8: Illustration of soil grain size characterization A) Removing the organic material using 30%* $H_2O_2$*, B) separating Sand from silt and clay using wet sieving through a 63µm sieve, C) 1L Nagene bottle with markings for separating clay and silt at 2, 4 and 6 hours, D) Draining the clay fraction, E) Clay fraction settling using* $CaCl_2$*.*

All the fractions were weighted, and the percentage distribution was calculated. The calculation did not take into account any possible loss of material.

## 3.3 Sequential extraction of the soil samples

The sequential extractions were performed after a modified procedure from Tessier et al. (1979). Since the activity concentrations of the soil samples were low, there was a need for a larger sample quantity than described in the method (from 1-2 g to 10 g). Samples from all layers from each of the three soil cores were collected and treated separately, except from the 8-13 cm layer from Groalia, which had too low activity. Totally, sequential extraction was done for 18 samples and 2 blank samples. Due to a large quantity of samples, the procedure was done twice over the course of two weeks.

**Step 1:** The first step extracted the water-soluble fraction. About 10 grams of sample was weighted and added to 250 ml centrifuge tubes. Then, 100 ml of Milli-Q water was added, and the samples were on the "Roller table" for 1 hour. Then the samples were centrifuged at 5000 x g for 15 minutes. The supernatant was collected using a 3 ml disposable pipette and filtrated using a funnel with a blue band filter of 0.45 μm for each sample. The filtrate was collected in a 160ml container. The blue band filter for each sample was kept to be used throughout the process. The setup of the filtration process is shown in figure 3.9.

**Step 2:** Due to low pH, below 5, the second step was discarded for all soil samples.

**Step 3:** The third step extracted the reversible fraction. 100 ml of 1M $CH_3COONH_4$ adjusted to pH~5 using $CH_3COOH$ was added to the samples from step 1 in the centrifuge tubes. After that, samples were on the "Roller table" for 2 hours and were centrifuged for 15 minutes. The supernatant was taken off and added to the funnel with the blue band filter used in step 1. Through a washing procedure, 50 ml of Milli-Q water was added to the centrifuge tube with the sample, and the samples were centrifuged for 15 minutes. The supernatant was taken off and filtrated. The washing procedure ensured that as much as possible of the extracted Cs in step three were collected in the filtrate. The filtrate from step 3 and the washing procedure was collected in the same 160 ml container.

**Step 4:** Step four extracted the easily reduced components such as oxides of iron and manganese by adding 100 ml of 0.04M $NH_2OH \cdot HCl$ in 25%(v/v) $CH_3COOH$ to the samples in the centrifuge tubes after step 3. The tubes were in a water bath at 80°C for 6 hours. The samples were centrifuged for 15 minutes, and the supernatant was filtrated before collection. Then the washing procedure mentioned under step three was done. The filtrate was collected.

**Step 5:** Step five extracted the fraction associated with oxidized components like organic material and uranium oxide particles. The samples from step 4 were transferred to an Erlenmeyer flask using 3 x 25 ml 30% $H_2O_2$, with a pH adjusted to ~2 using concentrated $HNO_3$. Then the samples were put in a water bath at 80°C for 5.5 hours. For the samples with high organic matter content, above 20%, the Erlenmeyer flask were instead heated using a sand bath by gradually increasing the temperature for 2 hours and then heating the samples for 3.5 hours at 80°C. The content of the Erlenmeyer flask was transferred back to the centrifuge tube after adding 25 ml 3.2 M $CH_3COONH_4$ in 20% (v/v) $HNO_3$ using 25 ml Milli-Q water. This was added to keep the extracted components in the solution. The samples were centrifuged for 15 min, and the supernatant was filtrated and collected. The washing procedure from step three was performed. The filtrate was collected.

**Step 6:** The sixth step extracted the acid dissolvable fraction by adding 100 ml 7M $HNO_3$ to the sample from step 5 and warming the solution for 6 hours using a water bath at 80°C. The samples were then filtrated directly without any centrifugation. The centrifuge tubes were washed using 50 ml Milli-Q water, which was filtrated, and the filtrate was collected. The filter contained the residue fraction, which represents the non-soluble fraction. The filters were collected and put into separate containers.

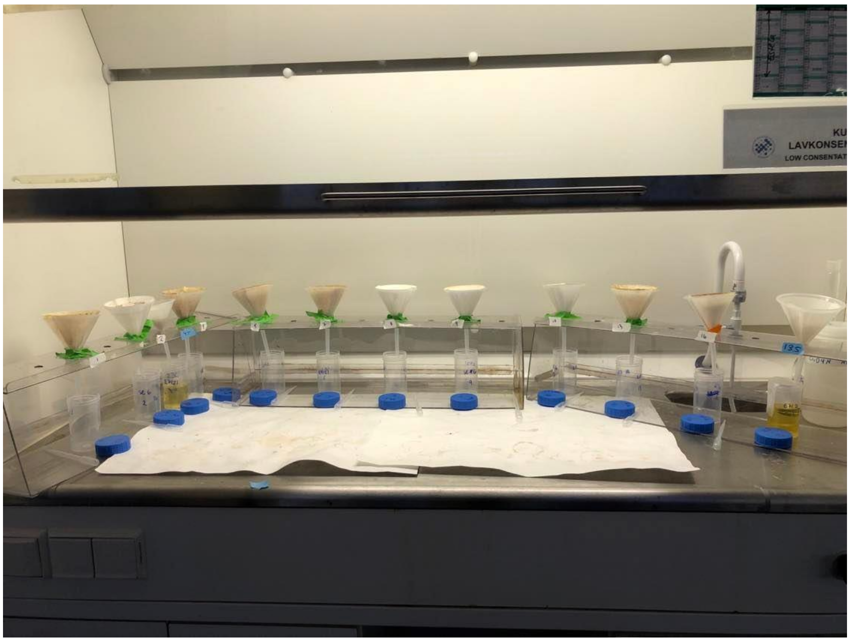

*Figure 3.9 Setup of the filtration process in a fume hood.*

Due to some clogging of several filters during the filtration of the 18 samples at different steps, some supernatants needed to be filtrated overnight, which might be a contributor to some of the filters breaking during the process. The broken filter was collected in the residual container, and a new filter was used for the remaining of the process.

## 3.4 Measurements

The activity of $^{137}$Cs in soil samples, grain size fractions and extracts from the sequential extractions were measured using both a NaI(Tl) scintillation detector and two Ge semiconductor detectors.

### 3.4.1 NaI-detector

The NaI(Tl) scintillation detector, PerkinElmer 2480 Automatic Gamma Counter using WIZARD$^2$ software, was used to measure $^{137}$Cs by measuring the activity for energies between 559-735 keV. The counting efficiency was estimated by measuring certified reference material, IAEA 300 at 5.7 Bq (N=4). The energy calibration was done by measuring standard reference material using the energy calibration program on the instrument. Blank samples (N=9) were measured to estimate the background radiation, which was subtracted from the activities measured.

### 3.4.2 Ge-detector

Two different Ge-detectors using Ortec Gammavision software were used, Ortec HPGe coaxial detector (detector 1) and Canberra GL 2020R LEGe detector (detector 2). The data was collected from the peak connected to $^{137}$Cs in the library with an energy of 661.66 keV. Before measurement, an ORTEC calibration file was chosen dependent on the geometry of the samples. Four types of geometries were used: “bluecap bottle” of 180 ml, half of the “bluecap bottle”, “whitecap bottle” of 125 ml, and 20 ml vials. Blank samples for each of the geometries were obtained to estimate the background radiation. The laboratory staff did the energy and efficiency calibration by using certified standard reference material. The accuracy of the measurements was estimated by measuring two IAEA 300 standard reference materials in 20 ml vials and calculating the relative bias.

### 3.4.3 Cesium-137 activity in the different layers

The activity concentration (Bq/kg) and activity density (Bq/m$^2$) of $^{137}$Cs in the different layers of the soil cores for both sites were calculated by measuring the activity of $^{137}$Cs for each of the layers in each of the soil cores and take into account the sampled area or the dry weight of the sample. As not possible to measure the whole collected sample, aliquots were measured. To get as representable samples aliquots as possible, about 100 g or ¼ of the untreated soil from each of the layers from each of the soil cores were homogenised by hand by mixing the content in a plastic bag. Three preweighted 20 ml vials were filled from each layer of each soil sample and measured using a NaI scintillation detector. The mean value of the three measurements was used to calculate Bq/kg and Bq/m2 per cm for each layer from each of the soil cores. The samples were measured for 5400s-7200s with a goal of less than 5% relative uncertainty for each of the samples. Due to low activities, some samples had a lower relative uncertainty than 11%.

To calculate the activity concentration *A* (Bq/kg) of $^{137}$Cs, the 20 ml vials of soil were freeze-dried and weighted. The activity concentration was calculated using formula 4, with *a* being the measured activity of $^{137}$Cs in the soil in the 20 ml vial (Bq) and *m* being the weight of the dried samples.

$$A = a/m \tag{4}$$

The calculation of the activity density *AD* (kBq/m$^2$ per cm) of $^{137}$Cs in each layer of the soil from Nerlifjellet and Groalia was done using formula 5. In the formula, *A* is the calculated activity concentration ((Bq/kg) of $^{137}$Cs in the soil layer, *M* is the measured total weight of the soil layer in the core (kg), *D* is the dry matter content of the soil (%), *d* is the diameter of the soil cores(m), and *t* is the thickness of the layer (cm).

$$AD = \frac{A \times M}{1000 \times d^2 \times t} \tag{5}$$

### 3.4.4 Cesium-137 activity in Sand, Silt and Clay

The sand, silt and clay fractions were measured using Ortec HPGe coaxial detector (detector 1). The silt and clay fractions were measured in a 125 ml container, which was suitable for measurement using Ge-detector and thus did not need any preceding transfer. The sand fraction was dried in glass beakers and was transferred to preweighted 20 ml vials (suitable for measurement of Ge-detector) and weighted. The activity of $^{137}$Cs was measured for all samples with a goal of a relative counting uncertainty of less than 10%. Due to the low activities of some samples, the relative uncertainties varied between 3% and 68%. The activity concentration (Bq/kg) was calculated for all fractions and compared. The total activity of the sample before the grain size fractionation was estimated based on the activity of $^{137}$Cs measured in 3.5.3.

### 3.4.5 Measurement of extracts from sequential extraction

#### *3.4.5.1 Measurement of $^{137}$Cs*

The samples from sequential extraction were measured using Canberra GL 2020R LEGe detector (detector 2). The activity of $^{137}$Cs in the samples was measured before sequential extraction using 20 ml vials using a NaI scintillation detector. The extracts were collected in containers suitable for measurement using Ge-detector (180 ml bluecap bottle and 125 ml whitecap bottle) and did not need any preceding transfer. The samples were measured with a goal of a relative uncertainty of less than 10%, but due to the low activities for some fractions, the relative uncertainties varied between 3% and 25%. All the extracts from step 6 and the filters containing the residual fraction were measured. Due to a lack of time and measurements requiring much time, only some of the extracts from step 1-5 was measured. Still, of the measured extracts, only a few measured above LOD. These were samples with the highest measured total activity of $^{137}$Cs (10-41 Bq), and thus the unmeasured extracts with a much lower total activity of $^{137}$Cs were assumed to measure activities below LOQ. This assumption was made based on the measurement of the activity of $^{137}$Cs before sequential extractions (table D4).

#### *3.4.5.2 Measurement of $^{133}$Cs*

Stable cesium in the extracts from steps 1-6 was measured using ICP-MS (Agilent 8800 triple quad ICP-MS). The extracts from steps 1-5 were diluted 10 times, while samples from step 6 were diluted 50 times prior to measurement. Samples were acidified until 5% (v/v) using 65% ultrapure nitric acid and collected in 15 ml centrifugation tubes. One calibration blank and two calibration standards were made to convert the signal from counts to concentration. The calibration blank was made using milli-Q water and acidified until 5%(v/v) using 65% ultrapure nitric acid. The calibration standards had a concentration of 2.0 µg/l and 0.2 µg/l made by diluting a certified standard solution of $^{133}$Cs using milli-Q water and was acidified in the same way as the calibration blank.

The mass measured was determined using a “relative isotopic abundance table”(AgilentTechnologies, 2008). For $^{133}$Cs, the mass was 133 amu and had 100% abundance, thus having no interfering masses. The samples were measured using $NH_3$ as the reaction gas, and In was added to samples to check for drift, and a drift correction was completed afterwards.

Due to the evaporation of the extracts during the filtration, it was necessary to estimate the volume for each extract from the sequential extraction. The volume for each extract was measured using 100 ml measuring cylinders with an accuracy of ±0.5ml with marks for every 1 ml.

### 3.5 Data handling and statistical analysis

Correction of the measurement due to background radiation was done by subtracting the mean measured background radiation from the measured activity of the sample.

Uncertainties ($\sigma_{counts}$) of the activity measurement from the NaI-detector was calculated using formula 6.

$$\sigma_{counts} = \sqrt{Counts} \tag{6}$$

For the measurement using Ge-detector, the software calculated the uncertainty for the measurement $\sigma_{counts}$. The relative uncertainty (%) $\sigma_{rel}$ was calculated by using formula 7.

$$\sigma_{rel} = \frac{\sigma_{counts}}{cps} \tag{7}$$

When measurements were added or subtracted by, for instance, calculating a mean value or correcting for the background radiation, the following equation was used:

$$\sigma_A \pm \sigma_B = \sqrt{{\sigma_A}^2 + {\sigma_B}^2} \tag{8}$$

When measurements were multiplicated or divided by, for instance, calculating distribution with percentage, the following formula was used.

$$\sigma_A \times \sigma_B = \sqrt{\frac{{\sigma_A}^2}{A} + \frac{{\sigma_B}^2}{B}} \tag{9}$$

For the radiological measurement, the LOD and LOQ were calculated using the mean uncertainties of the background radiation measured, $\sigma_B$. LOD and LOQ were calculated using the equations (Currie, 1968):

$$LOD = 4.65 \times \sigma_B \tag{10}$$

$$LOQ = 14.1\ \times \sigma_B \tag{11}$$

For the measurements using ICP-MS, the LOD and LOQ were calculated based on the standard deviation (σ) on the 12 measured blanks using the equations:

$$LOD = 3 \times \sigma \tag{12}$$

$$LOQ = 10 \times \sigma \tag{13}$$

*Data below LOD or LOQ*

For samples where none of the replicates had a value above LOD or LOQ, the value was replaced by <LOD or <LOQ, and in percentwise distribution, the value was considered 0%. When one or two of the three replicates had a value above LOD or LOQ, the replicate with the value below LOQ or LOD was switched with ½LOD or ½LOQ before the mean value was calculated. When the value was between LOD and LOQ and known, the measured value was used.

*Statistical analysis*

All statistics were preformed using Microsoft Excel and Python. The data being normally distributed was tested using a QQ-plot and a Kolmogorov Smirnov test, where p values>0.05

indicated a normal distribution. Data that was not normally distributed was log-transformed and the distribution was retested. A Pearson correlation test was performed to investigate if there was some correlation between data. Correlations with p-values below 0.05 were considered significant. Linear correlations were studied using linear regression, where $R^2$ show the proportion of the variance which is explained by the model (Løvås, 2015). To investigate if the differences between observations of the two groups were significantly different, a t-test was performed. For dependent samples, a dependent t-test was used. For data that indicated a non-normal distribution, a nonparametric test was used. For dependent groups of samples, a Wilcoxon test was performed. For independent groups of samples, a Mann-Whitney U-test was performed. When it was a difference in sample size, a Welsh's t-test was performed. Differences with p-values below 0.05 were considered significant.

# 4. Results and discussion

## 4.1 Quality

The quality of the measurements using different instruments can be discussed using the calculation of detection limits and accuracy measurements. The result for the NaI-detector, the Ge-detectors and ICP-MS is given in table 4.1

*Table 4.1: Detection and quantification limit and accuracy measurement for NaI-detector, Ge-detectors and ICP-MS. N.a- not analysed, efficiency calculated instead.*

| **Instrument** | | **Limits of detection** | | **Accuracy** | | |
|---|---|---|---|---|---|---|
| | | LOD | LOQ | CRM value | Measured value | Relative bias |
| NaI detector | | 0.15 Bq | 0.46 Bq | N.a | | |
| Germanium detector | | | | | | |
| Detector 1 | 20 ml vial | 0.013 Bq | 0.041 Bq | 5.54 Bq | 5.01 ± 0.04% Bq | 9.56% |
| | ½ 125 ml whitecap bottle | 0.015 Bq | 0.047 Bq | | | |
| Detector 2 | 180 ml bluecap bottle | 0.031 Bq | 0.094 Bq | 5.54 Bq | 5.20 ± 2.69% Bq | 6.13% |
| | ½ 180 ml bluecap bottle | 0.024 Bq | 0.072 Bq | | | |
| ICP-MS | | | | | | |
| Step 1, 3, 5 and 6 | | 0.23 mg/kg | 0.76 mg/kg | 1 µl/l | 1.026 ± 0.0018 µl/l | 2.6% |
| Step 4 | | 1.18 mg/kg | 3.92 mg/kg | | | |

### 4.1.1 Measurements using NaI-detector

The efficiency of the measurement of $^{137}$Cs using the NaI detector was calculated by measuring a certified reference material (IAEA 300). The measurement gave an average efficiency of 19.6% ± 0.28% (n=4), and an efficiency of 19.6% was thus also assumed for all samples and used in calculations of final activity concentrations. The use of a certified reference material to calculate the efficiency ensures accuracy of the measurements. The detection limit (LOD) was 0.15 Bq, while the quantification limit (LOQ) was 0.46 Bq, see table 4.1.

### 4.1.2 Measurements using Ge-detector

The accuracy of the measurements from both detectors was validated by measuring an IAEA certified reference material sediment sample (IAEA 300) with a calculated value of 5.54 Bq in a 20 ml vial. The result is given in table 4.1. The sample was measured with an efficiency calibrated to the geometry of ½ vial, even though the sample filled less than ½ of the vial. This compromise resulted in some overestimation of the efficiency of the measurement and thus activity measured. Generally, compromises are necessary for Ge-detector measurements due to a limited choice of efficiency calibrated geometries. This could result in uncertainty in the measurement of samples using Ge-detector. The measurements of the standardized reference material indicated that this uncertainty is low, with biases of 9.56% and 6.13%.

Table 4.1 shows LOD and LOQ for both detectors with the geometries used. The 20 ml vials measured with detector 1 had a LOD and LOQ of 0.013 Bq and 0.041 Bq. The ½ white cap bottle had a LOD of 0.015 Bq and LOQ of 0.047 Bq. LOD for 180 ml bluecap bottle and ½ bluecap bottle using detector 2 were 0.031 Bq and 0.024 Bq, respectively. The LOQ was 0.094 Bq and 0.07 Bq. Detector 1 had a lower LOD and LOQ than detector 2, suggesting that detector 1 can measure samples with lower quantifiable activity than detector 2. Still, the detection limits for germanium detectors were lower than for NaI-detector. The measurement of certified reference material using detector 1 showed a higher bias than detector 2.

### 4.1.3 Measurements using ICP-MS

The accuracy of the measurement of Cesium-133 using ICP-MS was controlled by measuring a certified reference material 1643 in 5% $HNO_3$. The result is given in table 4.1. The bias was 2.6%. Generally, the blank samples measured low values for $^{133}Cs$, but the blank samples from step 4 showed a higher amount of $^{133}Cs$. Since the blank samples for steps 5 and 6 showed only low values, it was assumed that any contamination only appeared for step 4. Therefore, separate detection limits were calculated for step 4. The result is given in table 4.1. For $^{133}Cs$ in step 4 LOD was 1.18 mg/kg, while LOQ was 3.92 mg/kg. The LOD was 0.23 mg/kg for the rest of the steps, while LOQ was 0.76 mg/kg.

## 4.2 Soil characterization

pH, dry matter content and organic matter content were identified for all layers. Additionally, a grain size analysis was completed for soil layers with less than 20% organic matter content. The result of the characterization is given in table 4.2. The result for each of the soil cores is given in table A2 and A3 in appendix A.

*Table 4.2: Soil characterization for soil from Nerlifjellet and Groalia. Result of measurements of pH, dry matter content, organic content, and grain size analysis. Mean values ± standard deviation. N.a – not analyzed. N- number of parallels.*

| **Parameter** | **Layer** | **Sites** | | |
|---|---|---|---|---|
| | | Nerlifjellet | Groalia | |
| **pH[a]** <br> **(N=3)** | 0-3 cm | 4.11± 0.05 | 4.25± 0.11 | 4.40 ± 0.23 (0-2 cm) |
| | | | | 4.40 ± 0.08 (2-3 cm) |
| | 3-7.5* cm | 4.00± 0.02 | 4.27± 0.05 | |
| | 7.5*-13 cm | 4.12± 0.04 | 4.51± 0.08 | |
| **Dry matter content (%)[b] (N=9)** | 0-3 cm | 21± 2.9 | 57 ± 5.9 | 50 ± 10.5 (0-2 cm) |
| | | | | 70 ± 3.6 (2-3 cm) |
| | 3-7.5* cm | 20 ± 3.6 | 82 ± 1.3 | |
| | 7.5*-13 cm | 60 ± 15.7 | 80 ± 2.2 | |
| **Organic matter content (%)[b] (N=9)** | 0-3 cm | 97.6± 0.9 | 28 ± 7.4 | 40 ± 16 (0-2 cm) |
| | | | | 11 ± 1.7 (2-3 cm) |
| | 3-7.5* cm | 88 ± 6.8 | 3.6 ± 0.7 | |
| | 7.5*-13 cm | 13.6± 5.7 | 5.1± 0.9 | |
| **Sand (%)** | 0-3 cm | N.a | N.a | N.a (0-2 cm) |
| | | | | 48 (2-3 cm) |
| | 3-7.5* cm (N=3)** | N.a | 39 ± 2 | |
| | 7.5*-13 cm (N=3)*** | 73 ± 9 | 39 | |
| **Silt (%)** | 0-3 cm | N.a | N.a | N.a (0-2 cm) |
| | | | | 42 (2-3 cm) |
| | 3-7.5* cm (N=3)** | N.a | 56 ± 3 | |
| | 7.5*-13 cm (N=3)*** | 25 ± 9 | 57 | |
| **Clay(%)** | 0-3 cm | N.a | N.a | N.a (0-2 cm) |
| | | | | 10 (2-3 cm) |
| | 3-7.5* cm (N=3)** | N.a | 5 ± 0.5 | |
| | 7.5*-13 cm (N=3)*** | 2 ± 1 | 4 | |
| **Mass of wet sample (kg) (N=3)** | 0-3 cm | 0.14 ± 0.026 | 0.23 ± 0.036 | 0.18 ± 0.021(0-2 cm) |
| | | | | 0.05 ± 0.016 (2-3 cm) |
| | 3-7.5* cm | 0.4 ± 0.16 | 0.79 ± 0.055 | |
| | 7.5*-13 cm | 0.50 ± 0.084 | 0.28 ± 0.063 | |

**Regarding Groalia, the profile is 3-8 cm and 8-13 cm, ** only for Groalia, ***Only for Nerlifjellet [a] Significant differences between Nerlifjellet and Groalia when comparing all measurements from all layers. [b] Significant differences between Nerlifjellet and Groalia when comparing layer by layer.*

### 4.2.1 Site 1: Nerlifjellet

The pH of the soil ranged between 4.00 and 4.12, being considered in vanLoon and Duffy (2005) to be moderately acidic. The organic matter content was highest for the layer between 0-3 cm (97.6%). While the 3-7.5 cm layer showed slightly lower organic matter content (88%), the bottom layer between 7.5-13 cm had significantly lower values (13.6%) (table A4). A grain size analysis for this layer showed that the main fraction was the sand fraction (73%). The silt fraction was lower than the sand fraction (25%), but the clay fraction was the smallest (2%). See table 4.2. The soil was generally characterised as organic with a high content of sand.

### 4.2.2 Site 2: Groalia

The pH of the soil ranged between 4.25 and 4.51, being considered in vanLoon and Duffy (2005) to be moderately acidic. For the separated samples representing 0-2 cm and 2-3 cm, the pH was higher than the 0-3 cm layer. This and high relative standard deviations suggest some heterogeneity of the soil content or uncertainties associated with the pH measurements. The organic matter content was highest for the upper layer between 0-2 cm (40%), significantly less (table A4) in the underlaying layers with a somewhat decreasing content with increasing depth, 2-3 cm (11%), 3-8 cm (3.6%) and 8-13 cm (5.1%). The organic matter content of the 0-2 cm layer was somewhat higher (nonsignificant) than 0-3 cm (28%). This was expected since the mineral layer between 2-3 cm was excluded.

The three layers where a grain size analysis was completed had comparable results, with silt being the biggest fraction (42-57%). However, the silt fraction might be overestimated due to the uncertainty in the separation of silt and clay using stokes law. The sand fraction was generally smaller than silt (39-48%), but the smallest fraction was clay (4-10%). For the 2-3 cm layer, the main fraction was sand (48%), and the clay content measured (10%) was higher than the clay content in the two other layers (4-5%). This might suggest higher content of clay and sand in the 2-3 cm layer of the soil sample. However, only one sample was measured due to the low amount of material for this layer. Therefore, the precision of the separation and measurement cannot be evaluated, and the measurement can only be used to indicate the distribution. In addition, any differences between the sand, silt and clay content could not be tested for statistical significance. The soil was generally characterised as low in organic material with a high content of silt.

### 4.2.3 Comparison of the two sites.

In situ observation of the soil samples suggested differences in the soil profiles from the two sites, Nerlifjellet and Groalia. The characterization of the soil supports this observation. The measured pH of the soil from Groalia was significantly higher than soil samples from Nerlifjellet, see table 4.2. Generally, the organic matter content was higher for soil from Nerlifjellet than for soil from Groalia. For the soil from Groalia, the organic matter was mainly found in the upper 2 cm, while for Nerlifjellet, high content of organic material was found down to 7.5 cm. The organic matter content of the 0-3 cm layer was significantly higher for the soil from Nerlifjellet compared to the soil from Groalia. A significantly higher level of organic matter content for soil from Nerlifjellet was also found when comparing the organic matter content from the two sites for the middle and bottom layers, see table 4.2. The result of the statistical tests comparing the two sites is given in table A4 in appendix A.

The dry matter content showed the opposite trend of the organic matter content for both sites. A Pearson correlation test showed a significant negative correlation between organic matter content and dry matter content (Nerlifjellet: $p<0.001$, $r=-0.92$, Groalia: $p<0.001$. $r=-0.96$), see table A5. A smaller dry matter content means a higher water content, and organic matter has been shown to have a larger volumetric water-holding capacity than mineral matter (Weil & Brady, 2017b).

The grain size analysis suggests some differences between the soil from Nerlifjellet and the soil from Groalia. The mineral layer from Nerlifjellet was dominated by the sand fraction, while the mineral soil from Groalia was mainly dominated by the silt fraction. Due to a low number of measurements for Nerlifjellet, the differences between the two sites could not be tested for statistical significance. Due to the organic matter being mainly found in the top 2 cm of the soil from Groalia, a grain size analysis was possible for the 2-3 cm layer, which indicated more sand and clay than in the layers further down in the soil. Such differences through the core could not be analysed for the soil from Nerlifjellet due to the high organic matter content in the upper 7.5 cm.

## 4.3 Activity of $^{137}$Cs in soil layers

The mean activity concentration (Bq/kg) and activity density (Bq/m$^2$ per cm) in the soil layers from Nerlifjellet and Groalia is presented in table 4.3. The counting uncertainty was below 10% and considerably less than the standard deviations of the measurements. The calculated activity concentrations and activity densities for each of the three cores are given in table B1-B4 in appendix B.

*Table 4.3 The mean activity concentration and activity density per cm of $^{137}$Cs in the soil layers from Nerlifjellet and Groalia are given in Mean value ± standard deviation (Relative standard deviation %)*

| | **Layer** | **Sites** | | |
|---|---|---|---|---|
| | | Nerlifjellet | Groalia | |
| **Activity concentration: Bq/kg (N=9)** | 0-3 cm | 900 ± 150 (17%) | 1300 ± 672 (51%) | 4000 ± 3122 (78%) (0-2 cm) |
| | | | | 900 ± 635 (70%) (2-3 cm) |
| | 3-7.5* cm$^a$ | 500 ± 115 (23%) | 80 ± 31 (38%) | |
| | 7.5*-13 cm$^a$ | 200 ± 134 (67%) | 36 ± 6.6 (18%) | |
| **Activity concentration: kBq/m$^2$ per cm (N=9)** | 0-3 cm | 1.0 ± 0.27 (28%) | 6 ± 3.8 (60%) | 20 ± 18 (94%) (0-2 cm) |
| | | | | 3 ± 2.4 (74%) (0-2 cm) |
| | 3-7.5* cm | 1.0 ± 0.43 (45%) | 1.0 ± 0.54 (56%) | |
| | 7.5*-13 cm | 1.1 ± 0.47 (44%) | 0.19 ± 0.071 (38%) | |

*N- Number of parallels analysed *Regarding Groalia, the profile is 3-8 cm and 8-13 cm. [a]Significant differences between the two sites when comparing layer by layer.*

### 4.3.1 Site 1: Nerlifjellet

The activity concentration (Bq/kg) of $^{137}$Cs was highest for the 0-3 cm layer (900 Bq/kg) and decreased with increasing depth of the soil layers, 3-7.5 cm (500 Bq/kg) and 7.5-13 cm (200 Bq/kg). Still, some activity in the underlying layers indicates that $^{137}$Cs are not completely retained in the surface soil but to some extent transported vertically through the soil. The activity density (kBq/m$^2$ per cm) supports this, showing an almost evenly distribution of $^{137}$Cs through the core. The activity density was around 1 kBq/m$^2$ per cm for all three layers, see table 4.3. The difference between the distribution considering the activity concentration and the activity density is due to the organic-rich soil in the top two layers having a lower density than the mineral-rich soil, resulting in a large difference between the activity concentration and activity distribution. Similar observations have been reported previously (Fujii et al., 2014; Karadeniz & Yaprak, 2008).

The relative standard deviations were relatively high (17-67%), suggesting some inhomogeneity of the $^{137}$Cs in soil. There were differences in activity concentration between cores, but the high relative standard deviations within each of the soil cores suggest some heterogenic content of $^{137}$Cs both within the samples and within the area of sampling (table B1 and B2). These differences could not be tested for significance, due to the small amount of data.

### 4.3.2 Site 2: Groalia

The activity concentration (Bq/kg) and activity density (Bq/m$^2$ per cm) of $^{137}$Cs decreased with increasing depth of the soil layer for soil from Groalia. The activity density of $^{137}$Cs was reduced with a factor of 6.6 from 0-2 cm to 2-3 cm, from 20 kBq/m$^2$ per cm to 3 kBq/m$^2$ per cm, and further with a factor of 3 from 2-3 cm to 3-8 cm, from 3 kBq/m$^2$ per cm to 1 kBq/m$^2$ per cm. This indicates that most of the $^{137}$Cs are retained in the top 0-2 cm layer, but also some retention of $^{137}$Cs in the 2-3 cm layer. The retention in the 2-3 cm layer might be due to high amounts of clay (10%).

The high relative standard deviation (>28%) suggests some inhomogeneity in the distribution of $^{137}$Cs both within the soil cores and the area of the site. The three soil cores, taken within the ~ 13 m$^2$ area, had differences in activity concentration and activity density, with one core having considerably lower activities in the 0-3 cm layer, see table B3 and B4. This suggests a non-homogeneous deposition of $^{137}$Cs within the sampling area of the site. These differences could not be tested for statistical significance due to the small amount of data.

### 4.3.3 Comparison

There were some variations (nonsignificant) in activity concentration between the sample cores from the same site. This could be because of small differences in the deposition. Similar results were found in a study by Andrello and Appoloni (2004) when measuring $^{137}$Cs activity in forest soil in Brazil. The differences in deposition within small areas could be due to variations in the vegetation or the water runoff at the site (Andrello & Appoloni, 2004). Deposition of $^{137}$Cs as particles might explain the high relative standard deviation within the soil cores from both sites.

To compare the distribution of $^{137}$Cs through the cores from Nerlifjellet and Groalia, the percentage distribution of $^{137}$Cs was calculated. This distribution is based on the total content calculated in each layer, and thus the differences in the thickness of the layer need to be considered during the comparison. The results are shown in figure 4.1. The percentage distribution with relative standard deviations is given in table B5.

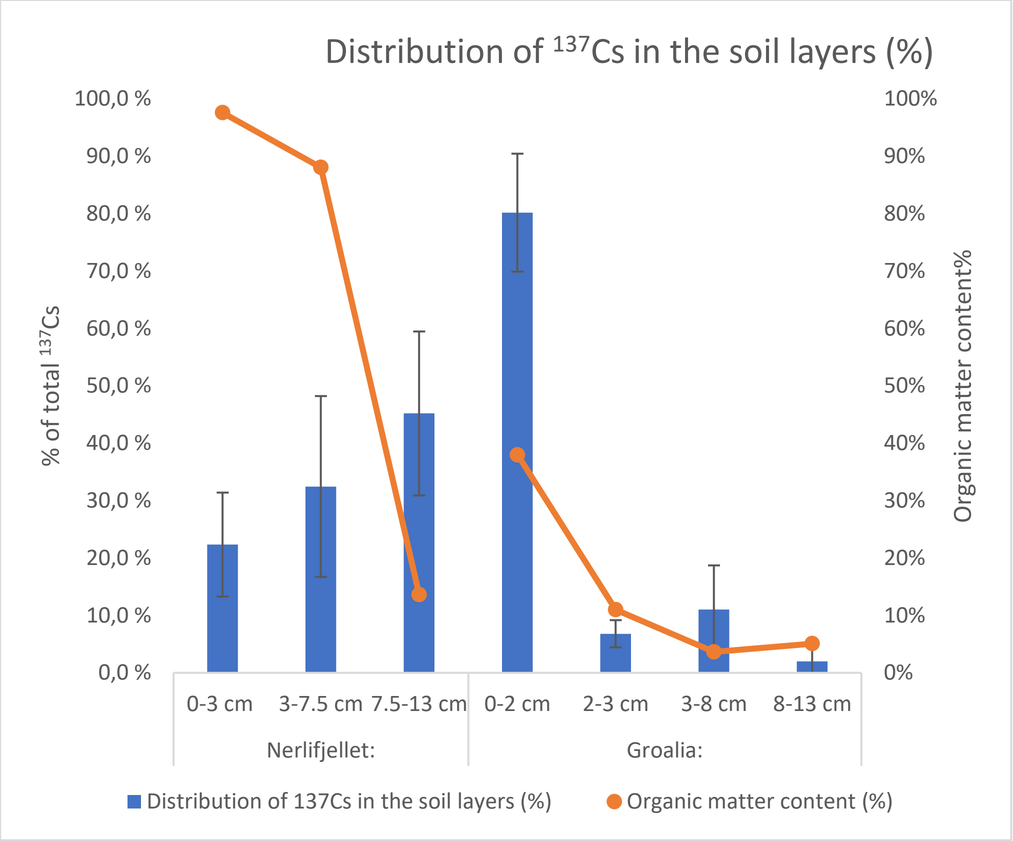

*Figure 4.1: Percentage distribution of $^{137}$Cs in soil cores for Nerlifjellet and Groalia with error bars representing the standard deviation.*

For the soil cores from Nerlifjellet, $^{137}$Cs had a higher vertical migration compared to Groalia and thus seemed to be more mobile. For soil from Nerlifjellet, the layer containing the most $^{137}$Cs was the bottom layer, the 7.5-13 cm layer (42.5%), while the lowest percentage amount was observed for the 0-3 cm layer (22.3%). Still, considering the differences in the thickness of the layers, high relative standard deviations, see appendix B, and the even activity density throughout the core, the distribution might be more even. On the other hand, this differs from the result from Groalia, where most of the $^{137}$Cs retained in the top 0-2 cm (80.2%). The 2-3 cm layer contained 5-10 % of the total measured $^{137}$Cs resulting in the top 0-3 cm containing about 86% of the total measured $^{137}$Cs. The retention of $^{137}$Cs in the upper organic-rich layers has been suggested to be impacted by the content of clay minerals in these layers, and that the retention is higher with increased content of clay (Takahashi et al., 2017). The 0-3 cm layer of soil from Groalia had a lower organic matter content (28%) than the top organic layer (0-3 cm) of soil from Nerlifjellet (97.6%). This might suggest a higher content of minerals, such as clays, in the 0-3 cm layer from Groalia. This could explain a higher retention of $^{137}$Cs within the first 3 cm for soil from Groalia compared to the first 3 cm for soil from Nerlifjellet. The amount of clay (10%) measured in the 2-3 cm layer from Groalia proves a certain amount of clay in the 0-3 cm layer and might indicate some clay in the 0-2 cm layer. Still, there is a need for an analysis of clay content in the 0-3 cm layer of Nerlifjellet and the 0-2 cm layer of Groalia to be able to conclude if the difference in retention in the first 3 cm is due to the difference in the content of clay.

The results from Nerlifjellet showing migration of $^{137}$Cs through the soil layers after deposition on the soil's surface could suggest a higher vertical migration rate of $^{137}$Cs for soil from Nerlifjellet. The layers in the soil profile from Nerlifjellet had a significantly higher amount of organic material compared to the layers in the soil profile from Groalia. The higher organic matter content could be the reason for the high vertical migration of $^{137}$Cs at this site. This is supported by a study of the vertical migration rate of soil samples taken in Sweden, which found that the vertical migration rate of $^{137}$Cs was highest for organic-rich soils (Rosén et al., 1999). Several studies have observed a higher penetration of $^{137}$Cs through the soil profile in organic-rich soil compared to soil with higher mineral content (Graham & Simon, 1996; Koarashi et al., 2012).

There was found no correlation between either the activity concentration, activity density, or percentage distribution of $^{137}$Cs and organic matter content when using data from both sites and all layers. Heterogeneity in the deposition of $^{137}$Cs within the sites and in the distribution of $^{137}$Cs within each soil layer results in a high variance of the results, making it difficult to find correlations. However, For the 0-3 cm layer, a significant negative correlation was found between the percentage distribution of $^{137}$Cs in the three cores from both sites and the measured organic material content, with a p-value of 0.001 and r value of -0.97. Results of statistical tests are given in table B6 and B7 in appendix B.

The grain size analysis results indicate that there are more silt and clay minerals in the soil from Groalia than in the soil from Nerlifjellet, where the soil was more dominated by the sand fraction. The soil from Nerlifjellet also had a smaller fraction of clay. This might be a reason why $^{137}$Cs seem to be more mobile in the soil from Nerlifjellet than in the soil from Groalia.

## 4.4 Activity of $^{137}$Cs in the sand, silt, and clay fractions

The activity of $^{137}$Cs was measured for the different fractions from the grain size analysis. The analysis of $^{137}$Cs in the grain size samples had a relative counting uncertainty between 3% and 68%, with especially high relative counting uncertainty for the low activity sand samples. Results with uncertainties and standard deviations of the measurements are given in table C1 in appendix C. The activity concentrations for each of the fractions are given in figure 4.2. The results showed that the clay fraction had a significantly higher activity concentration than both sand and silt fractions for all samples measured. The activity concentration of the silt fraction was significantly higher than the sand fraction, using dependent t-tests, see table C3. The clay samples had an activity concentration that was 9-30 times higher than the bulk samples, see table C1. Similar results have been found in several studies of soil and support the thought of $^{137}$Cs being easily bound to clay minerals (Livens & Baxter, 1988; Spezzano, 2005; Szabó et al., 2012).

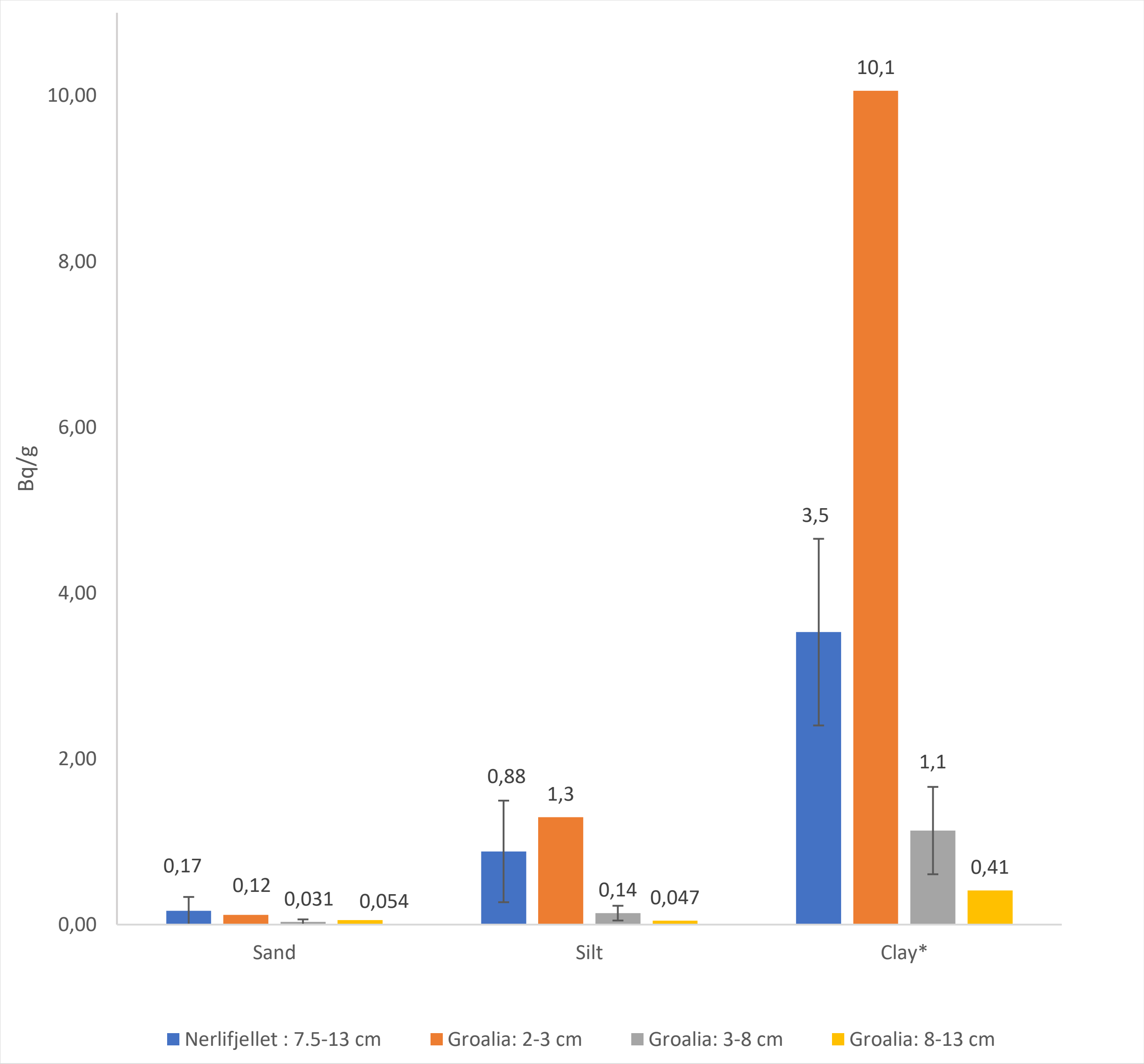

*Figure 4.2: Activity concentration (Bq/g) for sand, silt, and clay fraction separated during the grain size analysis. The error bars represent the standard deviation of the measurements (n=3). * Significant higher activity concentrations, than other fractions*

The proportion of $^{137}$Cs activity (%) associated with each grain size fraction was calculated based on the total activity measured. The result is shown in figure 4.3 and results with relative standard deviation is given in table C2. The measurement did not consider any loss of analyte during the separation process. For the samples where 3 parallels were measured, a relative standard deviation was calculated and was quite high. The high relative standard deviations of the proportion of $^{137}$Cs were mainly due to large relative standard deviations of the activity concentrations of $^{137}$Cs for each layer in each fraction, but also due to large relative standard deviations for the amount (%) of sand, silt and clay found in each layer, see table 4.2. When comparing the combination samples with the 3 replicates for the 7.5-13 cm layer from Nerlifjellet and the 3-8 cm layer from Groalia, there were some differences between the result of the combination samples and the mean value of the replicates. This suggests some uncertainties in the accuracy of the combination samples.

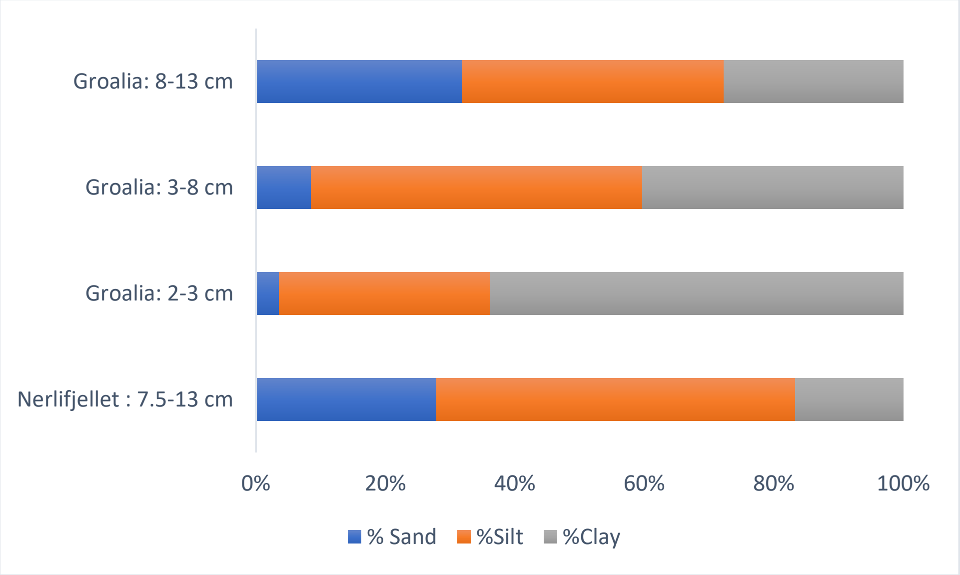

*Figure 4.3: The distribution (%) of $^{137}$Cs activity in sand, silt, and clay.*

Taking into account the grain size distribution and the activity of $^{137}$Cs associated with each size fraction, the $^{137}$Cs were mainly in the clay and silt fractions (68-98%). This is in agreement with several other studies of soil (Saito et al., 2014; Spezzano, 2005; Söderlund et al., 2016).

For the soil from Groalia, there were some differences between each layer. The proportion of $^{137}$Cs in the clay fraction decreased with increasing depth through the 2-3 cm layer (68%), the 3-8 cm layer (40%) and the 8-13 cm layer (28%). This decrease could be explained by differences in clay content for these layers. The amount of clay decreased rapidly from the 2-3 cm layer (10%) to the 3-8 cm layer (5%) and decreased further in the 8-13 cm layer (4%). A significant positive correlation was found between the proportion of $^{137}$Cs found in the clay fraction, see table C4, and the proportion of clay in soil from these layers. As discussed in 4.3, there was some retention of $^{137}$Cs in the 2-3 cm layer of the soil from Groalia. For this layer, the majority of $^{137}$Cs was found in clay (68%), and the activity concentration of $^{137}$Cs in the clay fraction was higher (10.1 Bq/g) than the other layers (0.41-1.1 Bq/g), see figure 4.1. This suggests that the high clay content in the 2-3 cm layer (10%) could explain the retention of $^{137}$Cs in this layer. Still, due to only one sample being measured, the precision of this measurement could not be evaluated. The large standard deviation for samples with multiple parallels and heterogeneity of soil content and distribution discussed in 4.2 and 4.3 could indicate that further analysis is needed to discuss this further.

Comparing the 7.5-13 cm layer from Nerlifjellet to the 8-13 cm layer from Groalia, there were some differences in the distribution of $^{137}$Cs. Still, due to the small number of samples, a test of the significance of the difference could not be completed. Both soil layers had a larger portion of $^{137}$Cs found in the sand fractions compared to the other soil layers from Groalia, with 32% for the 8-13 cm layer from Groalia and 28% for the 7.5-13 cm layer from Nerlifjellet. For the 7.5-13 cm layer from Nerlifjellet, this might be explained by the sand fraction being the main fraction in the soil (72%). A higher amount of sand in the sample might increase the probability of $^{137}$Cs being bound to the sand fraction. However, for the 8-13 cm layer from Groalia, the silt (57%) fraction was the main fraction and had only 39% sand, and thus such a connection was not apparent. Still, due to only one sample of the 8-13 cm layer from Groalia being measured and the fractionation process being prone to errors, there are uncertainties connected to this observation. The sand fraction in the 7.5-13 cm layer from Nerlifjellet had the highest activity concentration (0.17 Bq/g) compared to the other measured sand fractions (0.031-0.12 Bq/g). This fraction could be adsorbing more $^{137}$Cs due to clay minerals present in sand fractions. Szabó et al. (2012) found clay minerals in sand fractions by X-ray diffraction analysis. To discuss if this is the case for this layer, further analysis of the sand is needed.

The proportion of $^{137}$Cs in the clay fraction was lower for the 7.5-13 cm layer from Nerlifjellet (17%) than the 8-13 cm layer from Groalia (28%). This might be explained by the differences in clay content for the 8-13 cm layer from Groalia (4%) and the 7.5-13 cm layer from Nerlifjellet (2%). The activity concentration of $^{137}$Cs being the highest in the clay fraction for all layers, including the 7.5-13 cm layer from Nerlifjellet, support this. However, the organic matter content might also contribute. The layer from Nerlifjellet had a higher mean organic matter content (13.6%) compared to the layer from Groalia (5.1%). Staunton et al. (2002) found that the adsorption of $^{137}$Cs onto clays decreased with an increasing amount of organic material. Similar observations was found in Koarashi et al. (2012). This could indicate some negative impact of organic matter on the probability of sorption of $^{137}$Cs on clays. A negative correlation between organic matter and proportion found in clay was found when comparing all samples measured, see table C4. However, this correlation was not significant, and comparing with the calculated activity of $^{137}$Cs, no significant loss of $^{137}$Cs during the treatment with $H_2O_2$ was found. This suggests low binding of $^{137}$Cs in organic materials and low amounts of $^{137}$Cs being reversibly bound. However, this might also suggest $^{137}$Cs being rebound to sand, silt or clay after being mobilised from the organic matter.

Generally, the results seem to show that the amount of sand, silt and clay fraction impacts the proportion of $^{137}$Cs found in each fraction. This implies that a larger amount of a fraction gives a higher probability of $^{137}$Cs binding to this fraction, which seems to be the case for the clay content for all layers from both sites and for the sand content for the 7.5-13 cm layer from Nerlifjellet. However, the 2-3 cm layer showed the opposite trend for the sand fraction. The sand fraction was the most dominating fraction in this layer (48%) but had the smallest proportion of $^{137}$Cs in the sand fraction (4%) compared to other samples. This layer had a larger proportion of clay (10%), and $^{137}$Cs were mainly found in clay (68%). This indicates that $^{137}$Cs binding to clay had a higher probability than other fractions and that a higher amount of clay decreases the probability of $^{137}$Cs binding to other fractions. Since the 7.5-13 cm layer from Nerlifjellet had a lower amount of clay, the probability of $^{137}$Cs binding to sand might increase due to the low amount of clay present.

## 4.5 Binding and mobility of Cs

The binding and mobility (reversibly, irreversibly and inert fraction) of $^{137}$Cs were studied by performing a sequential extraction and measuring the content of stable($^{133}$Cs) and radioactive cesium ($^{137}$Cs) in the extracts. The fraction extracted from step 1 is considered to be the water-soluble fraction and is available for uptake by biota. The fraction extracted from step 3 is considered to be easily mobilized by ion exchange. The fraction extracted from these steps is considered the reversibly bound Cs. The irreversibly bound fraction consisted of the easily reduced (step 4), organic bound (step 5), and acid dissolvable fractions (step 6) and represents the bound $^{137}$Cs that might be available for uptake by slow oxidation or by a biodegradation process by microorganisms in the soil (Fawaris & Johanson, 1995). Not being impacted by the highly concentrated acid from step 6, the residual fraction is considered inert $^{137}$Cs (Reinoso-Maset et al., 2020).

### 4.5.1 Binding and mobility of $^{133}$Cs

The binding and mobility of $^{133}$Cs were investigated by measuring the content of $^{133}$Cs in the extracts from each step in sequential extraction. The amount in the residual was not measured. The distribution of $^{133}$Cs in percentage was calculated based on the sum of $^{133}$Cs in all fractions. The result is given in figure 4.4. The concentration of $^{133}$Cs in each step and distribution between the steps for all samples are given in table D1 and D2 in appendix D.

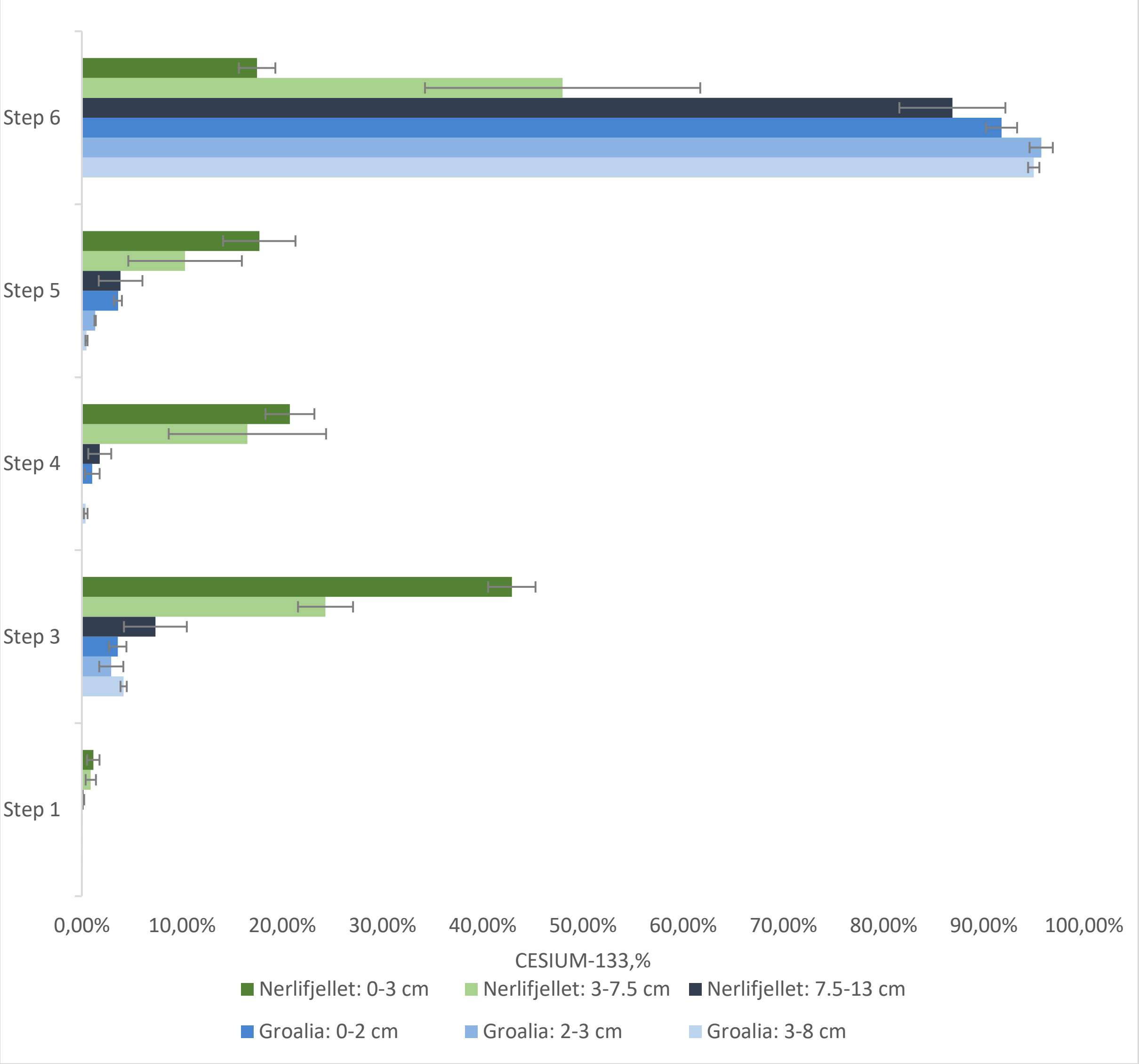

*Figure 4.4: Result of distribution of $^{133}$Cs in extracts from sequential distribution in percentage. Soil with an organic matter content above 40% is coloured in greens, while soil*

*with an organic matter content less than 40% is coloured in blues. The error bars represent the standard deviation of the measurements. Step 2 was not completed and measured due to low soil pH. For the steps and layers where there is no bar, the result was < LOD (table D1 and D2).*

There was a significant difference between the results for the soil with <40% organic matter and the soil with >40% organic matter, see table D9. The soil layers with < 40% organic matter had similar distribution independent of site and pH and a different trend compared to soil with higher organic matter content. In the low organic soil samples (<40%), the 7M $HNO_3$ (step 6) extracted most of the $^{133}$Cs (86.9-95.7%), and significantly smaller amounts of the total were extracted through the other 4 steps. The easily mobilised fraction (step 3) was the second-largest fraction (2.9-7.3%), while the organic bound fraction (step 5) was lower (0.46-3.6%), and the easily reduced fraction was the lowest measured fraction (0.38-1.8%). For the water-soluble fraction (step 1), the measurements were below LOD. The soil layers with > 40% organic matter had a more even distribution of $^{133}$Cs between the extracts, with the highest amounts found for the easily mobilised fraction (step 3) (24.3-42.9%) and the acid dissolvable fraction extracted by 7M $HNO_3$ (step 6) (17.4-47.9%). Results indicate that the samples with low organic content and more minerals (<40% organic matter) had more strongly bound $^{133}$Cs, which needed highly concentrated acid to be mobilized, compared to soil with a higher amount of organic matter (> 40% organic material). A Pearson correlation test was completed to see if there was any correlation between the proportion of $^{133}$Cs extracted from each step of the sequential extraction and the organic matter content in the soil sample. For steps 3-5, a positive correlation with p-values <0.001 for all the steps and R-values >0.89 was found, see table D10. For step 6, there was found a significant (p<0.001) negative correlation with an R-value at -0.93. Linear regression showed linear connections between the amount extracted in each step (3-6) and the organic matter content with $R^2$-values > 0.69, see figure D1- D4. Due to several extracts from step 1 being below LOD, step 1 was not tested for any correlation. The result of the statistical tests suggests a more mobile $^{133}$Cs in more organic soil than in more mineral-rich soil due to the proportion of $^{133}$Cs needing 7M $HNO_3$ to be extracted decreasing with increasing organic matter content.

### 4.5.2 Mobility of $^{137}$Cs

The binding and mobility of $^{137}$Cs were investigated by measuring the activity concentration of $^{137}$Cs in the extracts from sequential extraction. The distribution in percentage was calculated based on the sum of all measurements of the soil sample. For most of the samples, steps 1-5 had activities below LOQ (table D4). The results are given in figure 4.5.

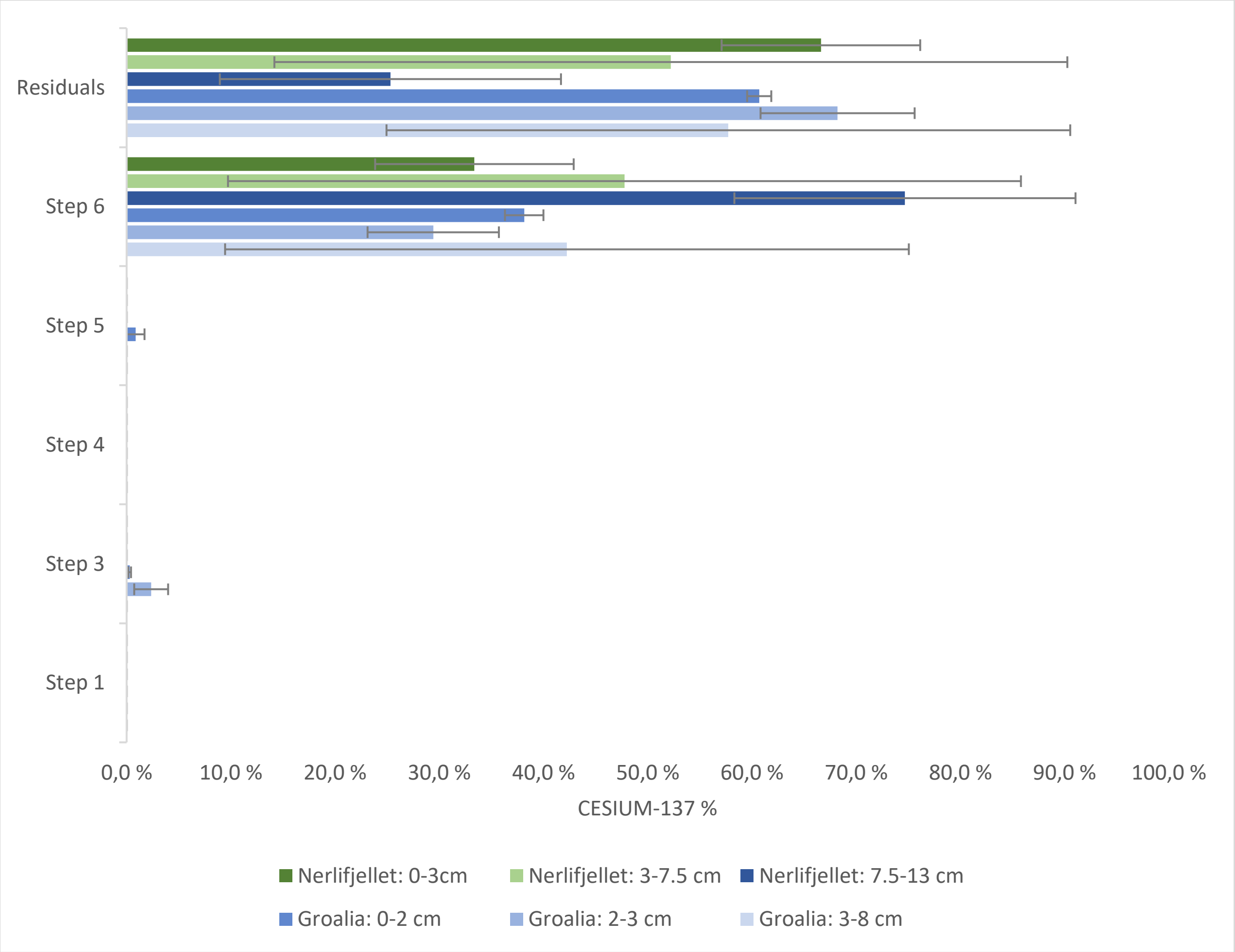

*Figure 4.5: Result of distribution of $^{137}$Cs in extracts from sequential distribution in percentage. Soil with an organic matter content above 40% is coloured in greens, while soil with an organic matter content less than 40% is coloured in blues. The error bars represent the standard deviation of the three measurements for each soil layer measured. For the steps and layers where there is no bar, the result was < LOQ (table D5-D7).*

Generally, the measurements indicate that most of the $^{137}$Cs were extracted in the acid dissolvable fraction (step 6) ( 33-75%) or found in the inert residual fraction (25-68%). This was found for both soil with >40% organic matter and for soil with < 40% organic matter. Cesium-137 has mainly been found in the acid dissolvable or residual fraction in several other studies of chemical fractionation of $^{137}$Cs in soil (Hoffman & Spitz, 2021; Hou et al., 2003; Reinoso-Maset et al., 2020). There were no significant differences between step 6 and the residuals (p=0.295), and a statistical difference in the results from Nerlifjell and Groalia was not found (p-values > 0.18), see table D9. No significant correlation was found between the proportion extracted in step 6 or residuals and the organic matter content, see table D10.

The results from sequential extraction separated into reversibly bound, irreversibly bound and inert $^{137}$Cs are given in figure 4.6. The lowest fraction was the reversibly bound $^{137}$Cs (0.0-2.4%), which mostly consisted of values below detection limits. The irreversibly bound fraction (30-75%) and the inert fraction (25-68%) were the dominating fractions. A dependent t-test showed that the irreversibly bound fraction had lower values than the inert fraction, but the difference was insignificant (p=0.419).

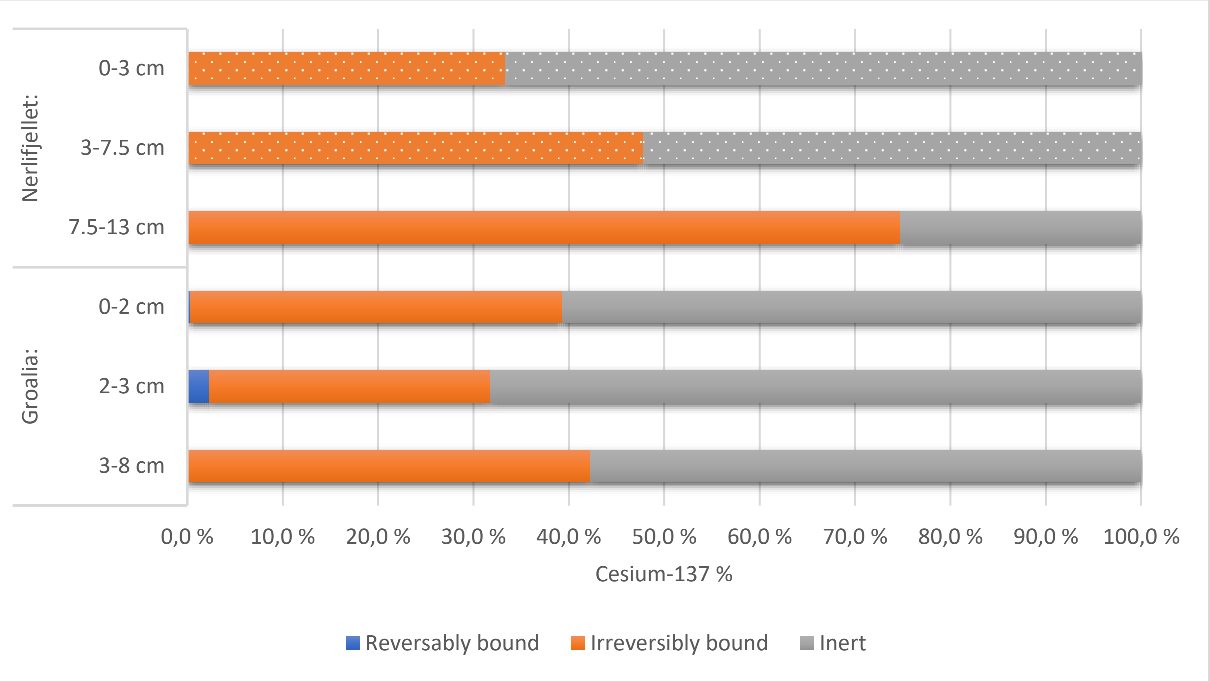

*Figure 4.6: Reversibly bound, irreversibly bound and inert proportion of $^{137}$Cs in the soil samples. The dotted bars represent the results for the samples with > 40% organic material. Results with standard deviations is given in table D8.*

The three parallels for each of the soil layers showed some differences in percentage distribution of the activity of $^{137}$Cs, which could not be tested for statistical significance, due to the small amount of data. These differences resulted in a high relative standard derivation, see table D7. This could be due to a lack of precision in the process of sequential extraction. Still, the standard deviation of the results for $^{133}$Cs was lower, table D3, which indicated that it is more likely that differences in the distribution of $^{137}$Cs in the soil between the three cores, possible differences in soil content between the three cores and possible differences in the speciation of $^{137}$Cs deposited in the soil are the causes of the high relative standard deviation.

The low activity of $^{137}$Cs in some of the extracts measured compared to the detection limits could also increase the uncertainty of the measurements and thus contribute to a higher standard deviation.

Most of the measurements of extracts of soil extraction steps 1-5 were <LOQ, and in figure 4.5 and figure 4.6 are considered 0% of the total $^{137}$Cs. Therefore, the amount of $^{137}$Cs in those steps might be underestimated. The sum of measured $^{137}$Cs from the sequential extraction was compared to the premeasured $^{137}$Cs in the soil samples and showed that for samples with > 40% organic matter, the sum of measured $^{137}$Cs was 12-59% lower than the premeasured $^{137}$Cs. For the samples with < 40% organic matter, this number was 0-32%. See table D4. Differences could indicate loss of analyte during the process of sequential extraction or indicate more $^{137}$Cs in the extracts from steps 1-5. The soil with more organic matter (>40%) had a larger loss (%) than soil with a lower amount of organic matter (<40%), which could indicate that the soil samples with a higher organic matter extracted more $^{137}$Cs in steps 1-5 than in the soil with a lower content of organic matter. Considering $^{133}$Cs to be a stable analogue for $^{137}$Cs, the result of sequential extraction of stable cesium could support this indication. Still, considering the source of $^{133}$Cs is from natural minerals in the soil, while the source of $^{137}$Cs is from anthropogenic airborne particles, the results for $^{133}$Cs might not be directly transferable to the results for $^{137}$Cs. Tsukada et al. (2008) found differences in chemical speciation of $^{137}$Cs and $^{133}$Cs in soils decades after the deposition of $^{137}$Cs, indicating some differences between the two isotopes. Still, the significant greater difference between premeasured $^{137}$Cs and the sum of measured $^{137}$Cs in the fractions from sequential extraction ($p=0.0012$) for the samples with >40% organic material than in the soil with <40% organic material could indicate some difference in the binding of $^{137}$Cs in organic-rich and mineral soils. Fawaris and Johanson (1995) found differences in chemical fractionation of $^{137}$Cs where the proportion of $^{137}$Cs found in the residual fraction and the acid dissolvable fraction was higher for soil with <50% organic matter content than for soil with > 50% organic matter. Shand et al. (1994) found that organic matter slows down the process of retention of cesium to the clay fraction of the soil. This was shown through a sequential extraction where more radiocesium was in the reversibly bound fraction in the organic soil compared to mineral soil. This might be applicable for the soil from Hattfjelldal where the $^{137}$Cs in the organic soil (>40%) could be more mobile than the $^{137}$Cs in the mineral based soil (<40%). Due to low activities in soil extraction steps 1-5, further analysis would be needed. In that case, by analysing  a larger amount of soil for each paralell or by measuring the extracts using a more sensitive detector.

Due to the two groups discussed ( >40% and <40% organic matter) being independent of the sample site and sample layers, the comparison might give some errors because of some possible differences in the speciation of $^{137}$Cs, as a result of vertical migration. Thus, it might be more appropreate to compare each of the soil layers from each site and then consider the organic matter content.

The result of the sequential extraction supports the idea that the $^{137}$Cs are more mobile in the soil from Nerlifjellet than in the soil from Groalia and that the reason for this might be the higher organic matter content of the soil from Nerlifjellet. Shenber and Eriksson (1993) and Koarashi et al. (2012) found that a higher organic matter content compared to the mineral content led to a more mobile $^{137}$Cs.

### 4.5.3 Uncertainties

It is important to be cautious when interpreting the results of sequential extractions due to the fractions being operationally defined (Hilton et al., 1992). In addition, due to a set amount of $H_2O_2$ used, the oxidizing of organic material might not be completed, resulting in an underestimation of organic bound cesium (Riise et al., 1990).

During the process of sequential extraction, several filters broke. The broken filters were measured as part of the residual fraction but contained some material that did not complete the entire sequential extraction. This could result in an overestimation of the inert cesium.

## 4.6 Reflection and further analysis

Several studies have identified $^{137}$Cs in water from the river Vefsna and in sediments and water from the Vefsnfjord. Heldal et al. (2021) found between 159-191 Bq/kg $^{137}$Cs in surface sediments from the Vefsnfjord. Larsen (2021) found 35-152 Bq/kg in surface sediment from the river Vefsna and 3-5 Bq/m$^3$ in the river water. This could indicate some movement of $^{137}$Cs from the terrestrial environment to the aquatic environment. However, the results from this study indicate low movement of $^{137}$Cs downwards in the soil, indicating a low mobility.

Overall, this study has shown some differences in the distribution and mobility of $^{137}$Cs between the two soil types. The distribution of $^{137}$Cs was different for soil from Groalia and soil from Nerlifjellet, which could be connected to organic matter content. However, to be able to discuss this further, an analysis of the vertical migration rate for several types of soil would be beneficial. If the vertical migration rate correlated with the organic matter in the soil, this could support the claim that organic matter contributes to a more mobile $^{137}$Cs.

The samples analysed in this master thesis were chosen based on knowledge about the high activity of $^{137}$Cs in the soil. Still, most of the extracts from step 1-5 of the sequential extraction was expected to be below LOQ, based on the measurements of some of the extracts. However, not every extract was measured and thus, having more time for analysis would contribute to more results for those steps. Still, it would be an advantage to increase the total activity of the samples by analysing a larger quantity of soil in each sample. Using a detector with a higher counting efficiency or a lower detection limit would also make it easier to get quantifiable results for steps 1-5.

In hindsight, there have been several disadvantages in the choices of samples to analyse. For sequential extraction, it would have been an advantage to analyse the 8-13 cm layer from Groalia and thus be able to compare the results to the 7.5-13 cm layer from Nerlifjellet. If there were large differences in the results, this could be connected to the difference in clay content and clay bound $^{137}$Cs. Similarly, more samples of the 8-13 cm layer from Groalia should have been analysed during the grain size analysis, making it possible to consider the standard deviation when comparing with the 7.5-13 cm layer from Nerlifjellet.

The sand fraction being the main fraction found in the sample from Nerlifjellet might explain why $^{137}$Cs seem to be more mobile in the soil from Nerlifjellet than in the soil from Groalia. To conclude this, information about the binding of $^{137}$Cs in the sand fraction compared to the clay fraction is needed. This could be obtained by doing a sequential extraction analysing the different fractions and comparing the results for the sand, silt and clay fractions.

Generally, the results have shown high relative standard deviations of the replicates. Soil is difficult to analyse due to its heterogenic content, which has been apparent in this study.

# 5. Conclusion

The goal of this thesis was to measure the activity of $^{137}$Cs in soil from the catchment area of Vefsna and to investigate if soil type has an impact on the mobility, binding, and distribution of $^{137}$Cs in soil. The soil from Nerlifjellet generally had a higher content of organic matter and a lower pH than the soil from Groalia. The grain size analysis showed that the mineral soil analysed from Nerlifjellet mainly consisted of sand and had the smallest proportion of clay, while the mineral soil from Groalia mainly consisted of silt.

There were large differences in the distribution of $^{137}$Cs through the core, where the $^{137}$Cs were more evenly distributed through the soil core for the soil from Nerlifjellet than the soil from Groalia. In the soil from Groalia, most $^{137}$Cs were retained in the 0-3 cm layer. This indicated more vertical mobility of the $^{137}$Cs in the soil from Nerlifjellet, which was linked to the difference in the organic matter based on the observation of several studies. However, no significant correlation was found between the distribution of $^{137}$Cs and content of organic matter.

Sequential extraction supported the claim that $^{137}$Cs were more mobile in organic-rich soil (>40% organic matter) based on more $^{133}$Cs found in the water dissolvable (step 1) and easily mobilised (step 3) fraction. This was further supported by the greater loss of analyte (%) for the soil samples with >40% organic matter when measuring the $^{137}$Cs content of each extract. This could indicate a greater amount of $^{137}$Cs found in steps 1-5, and thus more mobile $^{137}$Cs, for the soil with > 40% organic matter.

Grain size analysis showed that the clay fraction had a higher activity concentration than the other fractions and thus sorbed more $^{137}$Cs per gram. The clay layer in the 2-3 cm layer contained 68% of the $^{137}$Cs and is thus considered the reason for the retention of $^{137}$Cs in the 2-3 cm layer. The sand fraction of the soil from Nerlifjellet contained the majority of the $^{137}$Cs, and a weaker binding of $^{137}$Cs to the sand fraction than to the clay fraction might explain the higher mobility of $^{137}$Cs in soil from Nerlifjellet. To be able to demonstrate this, an analysis of the binding of $^{137}$Cs to the fractions would be necessary.

Generally, the $^{137}$Cs in the soil from Nerlifjellet seemed to have larger mobility than the $^{137}$Cs in the soil from Groalia. This has been connected to organic matter content due to weaker binding in organic matter and the impact of organic material on the binding of $^{137}$Cs to clay observed by several studies. Result highlight that it is essential to identify the content of organic material and clay content in soil to understand the mobility of $^{137}$Cs in soils and the probability for remobilisation of $^{137}$Cs in runoff.

The hypothesis for this thesis was:

H1: Cesium-137 is more bound to soil with a high percentage of organic material than soil types with a lower percentage.

Through this study, H1 could not be supported. The topsoil layer (0-3 cm) from Nerlifjellet, which mainly consisted of organic matter (98%), contained a smaller proportion (%) of total $^{137}$Cs in the soil core than the corresponding layer from Groalia, which mainly consisted of minerals and had a smaller organic matter content (%). This suggests that $^{137}$Cs were less bound to soil with a higher percentage of organic material than soil types with a lower percentage. The result of the sequential extraction suggesting a more mobile $^{137}$Cs in soil with more organic matter also support this.

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# Appendix

## Appendix A: Soil characterization

*Table A1: Soil layers for different cores.*

| **Layer** | **Nerlifjellet** | | | **Groalia** | | |
|---|---|---|---|---|---|---|
| | **Core 1** | **Core 2** | **Core 3** | **Core 1** | **Core 2** | **Core 3** |
| 1 | 0-3 cm | 0-3 cm | 0-3 cm | 0-3 cm | 0-3 cm | 0-3 cm |
| 2 | 3-10 cm | 3-6 cm | 3-7 cm | 3-8 cm | 3-8 cm | 3-8 cm |
| 3 | 10-13 cm | 6-13 cm | 7-13 cm | 8-12 cm | 8-12 cm | 8-12 cm |

*Table A2: Organic matter content. pH, and dry matter content for each core and soil layer for soil from Nerlifjellet (Nerlifjellet) and Groalia (Groalia). N- number of parallels measured. SD- standard deviation.*

| | Organic matter content (N=3) | SD | Relative SD | pH (N=1) | Dry matter content (N=3) | SD | Relative SD |
|---|---|---|---|---|---|---|---|
| Nerlifjellet: 0-3 cm (1) | 98 % | 0.5 % | 0.5 % | 4.2 | 23 % | 0.9 % | 3.9 % |
| Nerlifjellet: 0-3 cm (2) | 98 % | 1.0 % | 1.0 % | 4.2 | 18 % | 2.4 % | 13.4 % |
| Nerlifjellet: 0-3 cm (3) | 97 % | 0.6 % | 0.6 % | 4.1 | 23 % | 0.2 % | 1.0 % |
| Nerlifjellet: 3-7.5 cm (1) | 86 % | 3.4 % | 3.9 % | 4.0 | 22 % | 1.9 % | 8.9 % |
| Nerlifjellet: 3-7.5 cm (2) | 97 % | 1.2 % | 1.2 % | 4.1 | 15 % | 1.2 % | 8.2 % |
| Nerlifjellet: 3-7.5 cm (3) | 82 % | 3.4 % | 4.2 % | 4.0 | 22 % | 1.2 % | 5.4 % |
| Nerlifjellet: 7.5-13 cm (1) | 14 % | 1.9 % | 14.1 % | 4.2 | 64 % | 2.8 % | 4.4 % |
| Nerlifjellet: 7.5-13 cm (2) | 20 % | 2.1 % | 10.2 % | 4.1 | 37 % | 3.5 % | 9.5 % |
| Nerlifjellet: 7.5-13 cm (3) | 7 % | 2.2 % | 31.4 % | 4.1 | 70 % | 5.4 % | 7.7 % |
| Groalia: 0-3 cm (1) | 28 % | 3.8 % | 13.4 % | 4.1 | 54 % | 3.0 % | 5.5 % |
| Groalia: 0-3 cm (2) | 26 % | 2.1 % | 7.9 % | 4.3 | 62 % | 0.1 % | 0.2 % |
| Groalia: 0-3 cm (3) | 29 % | 13.9 % | 47.4 % | 4.3 | 54 % | 8.1 % | 14.9 % |
| Groalia: 3-8 cm (1) | 4 % | 0.3 % | 8.8 % | 4.3 | 82 % | 1.6 % | 2.0 % |
| Groalia: 3-8 cm (2) | 4 % | 0.2 % | 4.8 % | 4.3 | 81 % | 0.6 % | 0.7 % |
| Groalia: 3-8 cm (3) | 3 % | 0.9 % | 29.5 % | 4.2 | 82 % | 1.5 % | 1.9 % |
| Groalia: 8-13 cm (1) | 6 % | 0.4 % | 6.8 % | 4.4 | 79 % | 0.6 % | 0.7 % |
| Groalia: 8-13 cm (2) | 4 % | 0.2 % | 4.9 % | 4.5 | 82 % | 1.4 % | 1.7 % |
| Groalia: 8-13 cm (3) | 5 % | 0.6 % | 12.5 % | 4.6 | 81 % | 2.6 % | 3.2 % |
| Groalia: 0-2 cm (1) | 41 % | 11.9 % | 29.0 % | 4.2 | 46 % | 4.5 % | 9.7 % |
| Groalia: 0-2 cm (2) | 51 % | 17.0 % | 33.5 % | 4.7 | 44 % | 6.7 % | 15.3 % |
| Groalia: 0-2 cm (3) | 22 % | 8.6 % | 38.5 % | 4.2 | 62 % | 7.6 % | 12.2 % |
| Groalia: 2-3 cm (1) | 13 % | 0.3 % | 2.1 % | 4.4 | 66 % | 0.1 % | 0.2 % |
| Groalia: 2-3 cm (2) | 10 % | 0.3 % | 2.4 % | 4.3 | 70 % | 0.5 % | 0.7 % |
| Groalia: 2-3 cm (3) | 9 % | 0.1 % | 1.2 % | 4.5 | 74 % | 0.2 % | 0.3 % |

*Table A3: Sand. silt, and clay content in grams and proportion of the total (%). cs-combination sample.*

| **Layer** | **Sand (g)** | **Sand (%)** | **Silt (g)** | **Silt (%)** | **Clay (g)** | **Clay (%)** |
|---|---:|---:|---:|---:|---:|---:|
| Nerlifjellet: 7.5-10 cm core 1 | 14.8 | 76 % | 4.5 | 23 % | 0.23 | 1 % |
| Nerlifjellet: 7.5-10 cm core 1 | 6.9 | 84 % | 1.1 | 13 % | 0.24 | 3 % |
| Nerlifjellet: 7.5-10 cm core 2 | 18.0 | 65 % | 9.5 | 34 % | 0.39 | 1 % |
| Nerlifjellet: 7.5-10 cm cs | 6.3 | 67 % | 2.9 | 31 % | 0.14 | 1 % |
| Groalia: 2-3 cm cs | 2.5 | 48 % | 2.1 | 42 % | 0.50 | 10 % |
| Groalia: 3-8 cm core 1 | 12.7 | 40 % | 17.3 | 55 % | 1.49 | 5 % |
| Groalia: 3-8 cm core 2 | 9.8 | 38 % | 14.4 | 56 % | 1.32 | 5 % |
| Groalia: 3-8 cm core 3 | 18.2 | 40 % | 24.4 | 54 % | 2.38 | 5 % |
| Groalia: 3-8 cm cs | 3.4 | 35 % | 5.7 | 60 % | 0.40 | 4 % |
| Groalia: 8-13 cm cs | 3.9 | 39 % | 5.7 | 57 % | 0.44 | 4 % |

*Table A4: Test of significant differences for soil characterization. Significan p-values is marked green*

| Test | | Matrix1 | Matrix2 | P-value |
|---|---|---|---|---|
| t-test | Organic matter content | Nerlifjellet 0-3 cm | Nerlifjellet 3-7.5 cm | 0.003 |
| Mann-whitney U-test | | Nerlifjellet 3-7.5 cm | Nerlifjellet 7.5-13 cm | 0.007 |
| t-test | | Nerlifjellet 7.5-13 cm | Nerlifjellet 0-3 cm | <0.001 |
| | | Groalia: 0-3 cm | Groalia: 3-8 cm | <0.001 |
| | | Groalia: 3-8 cm | Groalia: 8-13 cm | 0.021 |
| | | Groalia: 0-3 cm | Groalia: 8-13 cm | <0.001 |
| | | Groalia 0-2 cm | Groalia: 2-3 cm | <0.001 |
| | | Groalia 0-2 cm | Groalia: 3-8 cm | <0.001 |
| | | Groalia 0-2 cm | Groalia 8-13 cm | <0.001 |
| | | Groalia: 2-3 cm | Groalia: 3-8 cm | <0.001 |
| | | Groalia: 2-3 cm | Groalia: 8-13 cm | <0.001 |
| | | Groalia 0-3 cm | Groalia 0-2 cm | 0.11 |
| Mann-Whitney U-test | | Nerlifjellet: 0-3 cm | Groalia: 0-3 cm | <0.001 |
| | | Nerlifjellet: 3-7.5 cm | Groalia: 3-8 cm | <0.001 |
| | | Nerlifjellet: 7.5-13 cm | Groalia: 8-13 cm | 0.002 |
| t-test | pH | Nerlifjellet | Groalia | 0.002 |
| t-test | Dry matter content | Nerlifjellet 0-3 cm | Nerlifjellet 3-7.5 cm | 0.011 |
| | | Nerlifjellet 3-7.5 cm | Nerlifjellet 7.5-13 cm | <0.001 |
| | | Nerlifjellet 7.5-13 cm | Nerlifjellet 0-3 cm | <0.001 |
| | | Groalia: 0-3 cm | Groalia: 3-8 cm | <0.001 |
| | | Groalia: 3-8 cm | Groalia: 8-13 cm | 0.291 |
| | | Groalia: 0-3 cm | Groalia: 8-13 cm | <0.001 |
| Mann-Whitney U-test | | Nerlifjellet: 0-3 cm | Groalia: 0-3 cm | <0.001 |
| | | Nerlifjellet: 3-7.5 cm | Groalia: 3-8 cm | <0.001 |
| | | Nerlifjellet: 7.5-13 cm | Groalia: 8-13 cm | <0.001 |

*Table A5: Test of significant correlation between two soil parameters. Green celles have significant correlation.*

| Test | Matrix 1 | Matrix 2 | p-value | r-value |
|---|---|---|---|---|
| Pearson correlation test | All dry matter content | All organic matter content | <0.001 | -0.95 |
| | Site 1 dry matter content | Site 1 organic matter content | <0.001 | -0.92 |
| | Layrer 1 dry matter content | Layrer 1 organic matter content | <0.001 | -0.99 |
| | Layrer 2 dry matter content | Layrer 2 organic matter content | <0.001 | -1 |
| | Layrer 3 dry matter content | Layrer 3 organic matter content | <0.001 | -0.95 |
| | Site 2 dry matter content | Site 2 organic matter content | <0.001 | -0.96 |

## Appendix B: Activity of $^{137}$Cs in different soil layers

*Table B1: Mean activity concentration of $^{137}$Cs and standard deviation for soil core 1. 2, and 3 from Nerlifjellet: Nerlifjellet. N-number of parallels measured.*

| | **Core 1** | | | **Core 2** | | | **Core 3** | | |
|---|---|---|---|---|---|---|---|---|---|
| | Bq/kg (N=3) | SD | Relative SD | Bq/kg (N=3) | SD | Relative SD | Bq/kg (N=3) | SD | Relative SD |
| **0-3 cm** | 990 | 61 | 7 % | 700 | 101 | 14 % | 1000 | 158 | 16% |
| **3-7.5 cm** | 480 | 92 | 19 % | 640 | 63 | 10 % | 412 | 2.9 | 0,7 % |
| **7.5-13 cm** | 123 | 3.7 | 3 % | 390 | 42 | 11 % | 134 | 2.5 | 1,9 % |

*Table B2: Mean activity density of $^{137}$Cs and standard deviation for soil cores 1. 2 and 3 from Nerlifjellet: Nerlifjellet. N-number of parallels measured. SD-standard deviation.*

| | **Core 1** | | | **Core 2** | | | **Core 3** | | |
|---|---|---|---|---|---|---|---|---|---|
| | kBq/m$^2$ per cm (N=3) | SD | Relative SD | kBq/m$^2$ per cm (N=3) | SD | Relative SD | kBq/m$^2$ per cm (N=3) | SD | Relative SD |
| **0-3 cm** | 0.9 | 0.09 | 11 % | 0.8 | 0.06 | 8 % | 1.3 | 0.21 | 16 % |
| **3-7.5 cm** | 0.9 | 0.19 | 21% | 1.5 | 0.021 | 1.4% | 0.6 | 0.016 | 2.6 % |
| **7.5-13 cm** | 1.2 | 0.03 | 2.5 % | 1.2 | 0.03 | 2.5 % | 0.9 | 0.08 | 8.8 % |

*Table B3: Mean activity concentration of $^{137}$Cs and standard deviation for soil core 1. 2 and 3 from Nerlifjellet: Groalia. N-number of parallels measured. SD-standard deviation.*

| | **Core 1** | | | **Core 2** | | | **Core 3** | | |
|---|---|---|---|---|---|---|---|---|---|
| | Bq/kg (N=3) | SD | Relative SD | Bq/kg (N=3) | SD | Relative SD | Bq/kg (N=3) | SD | Relative SD |
| **0-3 cm** | 1967 | 236 | 12 % | 1455.0 | 61.0 | 4 % | 467 | 58.6 | 13 % |
| **3-8 cm** | 69 | 1.3 | 2 % | 113 | 7.3 | 6 % | 44 | 6.9 | 16 % |
| **8-13 cm** | 44 | 2.3 | 5 % | 37.1 | 0.9 | 2 % | 29 | 3.5 | 12 % |

*Table B4: Mean activity density of $^{137}$Cs and standard deviation for soil core 1. 2, and 3 from Nerlifjellet: Groalia. N-number of parallels measured. SD-standard deviation.*

| | **Core 1** | | | **Core 2** | | | **Core 3** | | |
|---|---|---|---|---|---|---|---|---|---|
| | kBq/m$^2$ (N=3) | SD | Relative SD | kBq/m$^2$ (N=3) | SD | Relative SD | kBq/m$^2$ (N=3) | SD | Relative SD |
| **0-3 cm** | 8 | 0.6 | 8 % | 9 | 0.4 | 4 % | 2.0 | 0.17 | 9 % |
| **3-8 cm** | 0.94 | 0.007 | 1 % | 1.5 | 0.09 | 6 % | 0.4 | 0.10 | 23 % |
| **8-13 cm** | 0.27 | 0.015 | 5 % | 0.16 | 0.007 | 4 % | 0.13 | 0.012 | 9 % |

*Table B5: Percentage distribution of $^{137}$Cs with standard deviations. N-number of parallels measured. SD- standard deviation. R SD- Relative standard deviation.*

| **Site** | **Layer** | **Distribution of $^{137}$Cs in the soil layers (%) (N=9)** | **SD** | **R SD** |
|---|---|---|---|---|
| **Nerlifjellet:** | 0-3 cm | 22.3 % | 9 % | 40.5 % |
| | 3-7.5 cm | 32.5 % | 16 % | 48.5 % |
| | 7.5-13 cm | 45.2 % | 14 % | 31.6 % |
| **Groalia:** | 0-2 cm | 80.2 % | 10.3 % | 12.8 % |
| | 2-3 cm | 6.8 % | 2.4 % | 34.7 % |
| | 3-8 cm | 11.0 % | 7.7 % | 69.5 % |
| | 8-13 cm | 2.0 % | 3.0 % | 153.4 % |

*Table B6: Test of significant differences for activity concentration of $^{137}$Cs between cores and layers. Marked p values- significant.*

| Test | Parameter | Matrix 1 | Matrix 2 | P-value |
|---|---|---|---|---|
| Dependent t-test | Bq/kg | Nerlifjellet: 0-3 cm | Nerlifjellet: 3-7.5 cm | 0.002 |
| | | Nerlifjellet: 3-7.5 cm | Nerlifjellet: 7.5-13 cm | <0.001 |
| | | Nerlifjellet: 0-3 cm | Nerlifjellet: 7.5-13 cm | <0.001 |
| | | Groalia: 0-3cm | Groalia: 3-8 cm | 0.001 |
| | | Groalia: 3-8 cm | Groalia: 8-13 cm | 0.004 |
| | | Groalia: 8-13 cm | Groalia: 0-3 cm | <0.001 |
| | | Groalia: 0-2 cm | Groalia: 2-3 cm | 0.016 |
| | | Groalia: 2-3 cm | Groalia: 3-8 cm | 0.0035 |
| T-test | | Nerlifjellet: 0-3 cm | Groalia: 0-3 cm | 0.099 |
| | | Nerlifjellet: 3-8 cm | Groalia: 3-8 cm | <0.001 |
| | | Nerlifjellet: 8-13 cm | Groalia: 8-13 cm | 0.004 |

*Table B7: Correlation tests between activity concentration. activity density, and %distribution of* $^{137}$*Cs and organic matter content and pH. Marked p values- significant.*

| Test | Matrix 1 | Matrix 2 | P-value | r-value |
|---|---|---|---|---|
| Pearson correlation | Nerlifjellet: Bq/kg | Organic matter | 0.004 | 0.84 |
| | Nerlifjellet: kBq/m$^2$ | Organic matter | 0.139 | -0.53 |
| | Nerlifjellet: % distribution | Organic matter | 0.112 | -0.57 |
| | Groalia: Bq/kg | Organic matter | 0.006 | 0.83 |
| | Groalia: kBq/m$^2$ | Organic matter | 0.019 | 0.76 |
| | Groalia: % distribution | Organic matter | <0.001 | 0.95 |
| | Both sites: Bq/kg | Organic matter | 0.082 | 0.42 |
| | Both sites: kBq/m$^2$ | Organic matter | 0.633 | -0.12 |
| | Both sites: % distribution | Organic matter | 0.973 | 0.01 |
| | Both sites 0-3 cm % distribution | Organic matter | 0.001 | -0.97 |
| | Both sites 3-7.5/8 cm % distribution | Organic matter | 0.291 | 0.54 |
| | Both sites 7.5/8-13cm % distribution | Organic matter | 0.09 | 0.76 |
| | Nerlifjellet: Bq/kg | pH | 0.661 | 0.17 |
| | Nerlifjellet: kBq/m$^2$ | pH | 0.403 | -0.32 |
| | Nerlifjellet: % distribution | pH | 0.278 | -0.41 |
| | Groalia: Bq/kg | pH | 0.093 | -0.59 |
| | Groalia: kBq/m$^2$ | pH | 0.105 | -0.58 |
| | Groalia: % distribution | pH | 0.051 | -0.66 |
| | Both sites: Bq/kg | pH | 0.137 | -0.36 |
| | Both sites: kBq/m$^2$ | pH | 0.494 | -0.17 |
| | Both sites: % distribution | pH | 0.07 | -0.44 |
| | Both sites 0-3 cm % distribution | pH | 0.651 | 0.24 |
| | Both sites 3-7.5/8 cm % distribution | pH | 0.225 | -0.58 |
| | Both sites 7.5/8-13cm % distribution | pH | 0.01 | 0.92 |

## Appendix C: Activity of $^{137}$Cs in the fractions

*Table C1: Activity concentration and proportion of* $^{137}$*Cs in sand, silt, and clay. The proportion of* $^{137}$*Cs in silt and clay fraction. Calculated total activity of* $^{137}$*Cs and sum of the measured activity of* $^{137}$*Cs. % of calculated gives the percentage of calculated activity measured. Numbers above 100% due to uncertainties of measurement and uncertainties of calculated activity of the sample. RU- relative uncertainty of the measurement of activity.*

| Layer | Sand Bq/g | RU % | Sand % of total | Silt Bq/g | RU % | Silt % of total | Clay Bq/g | RU % | Clay %activity of total | Bq/g Bulk | Clay (Bq/g) compared to total (Bq/g) (Bq/g clay /bq/g bulk) | Silt+Clay % | Calculated Bq | Sum measured Bq | % of calculated |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Nerlifjellet: 7.5-10 cm core 1** | 0.068 | 9.9 % | 25 % | 0.564 | 6.1 % | 64 % | 1.968 | 8.7 % | 11 % | 0.12 | 16.0 | 75 % | 3.58 | 3.98 | 111 % |
| **Nerlifjellet: 7.5-10 cm core 1** | 0.035 | 5.0 % | 9 % | 0.502 | 5.6 % | 68 % | 4.099 | 3.3 % | 23 % | 0.13 | 30.7 | 91 % | 5.87 | 6.97 | 119 % |
| **Nerlifjellet: 7.5-10 cm core 2** | 0.405 | 5.1 % | 50 % | 1.796 | 6.0 % | 35 % | 3.495 | 12.7 % | 15 % | 0.39 | 8.9 | 50 % | 4.82 | 5.61 | 116 % |
| **Nerlifjellet: 7.5-10 cm cs** | 0.156 | 4.4 % | 27 % | 0.670 | 5.2 % | 55 % | 4.550 | 12.4 % | 18 % | 0.22 | 21.0 | 73 % | 2.98 | 3.56 | 119 % |
| **Groalia: 2-3 cm cs** | 0.116 | 8.0 % | 4 % | 1.296 | 9.9 % | 33 % | 10.060 | 6.7 % | 64 % | 1.05 | 9.6 | 96 % | 8.43 | 7.89 | 94 % |
| **Groalia: 3-8 cm core 1** | 0.020 | 42.6 % | 7 % | 0.094 | 9.0 % | 48 % | 1.018 | 9.7 % | 45 % | 0.07 | 14.8 | 93 % | 2.79 | 3.39 | 122 % |
| **Groalia: 3-8 cm core 2** | 0.016 | 67.6 % | 3 % | 0.239 | 4.4 % | 59 % | 1.680 | 5.5 % | 38 % | 0.11 | 14.9 | 97 % | 4.83 | 5.81 | 120 % |
| **Groalia: 3-8 cm core 3** | 0.012 | 38.1 % | 10 % | 0.039 | 9.0 % | 42 % | 0.456 | 10.0 % | 48 % | 0.04 | 10.3 | 90 % | 2.07 | 2.25 | 109 % |
| **Groalia: 3-8 cm cs** | 0.078 | 35.4 % | 14 % | 0.181 | 4.5 % | 56 % | 1.383 | 17.1 % | 30 % | 0.08 | 18.3 | 86 % | 1.57 | 1.86 | 119 % |
| **Groalia: 8-13 cm cs** | 0.054 | 45.2 % | 32 % | 0.047 | 12.8 % | 40 % | 0.413 | 15.9 % | 28 % | 0.04 | 11.8 | 68 % | 0.57 | 0.66 | 116 % |

*Table C2: Activity concentration with standard deviation and proportion of $^{137}$Cs in sand, silt, and clay. SD- standard deviation. n-number of parallels measured. R SD- Relative standard deviation*

| | **Sand Bq/g** | **SD** | **R SD** | **Sand % of total** | **SD** | **R SD** | **Silt Bq/g** | **SD** | **R SD** | **Silt % of total** | **SD** | **R SD** | **Clay Bq/g** | **SD** | **R SD** | **Clay %activity of total** | **SD** | **R SD** |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Nerlifjellet: 7.5-13 cm** | 0.2 | 0.167 | 83% | 28 % | 16.6 % | 59% | 0.9 | 0.61 | 69 % | 55 % | 14.5 % | 26.1 % | 4 | 1.126 | 32 % | 17 % | 4.9 % | 29.3 % |
| **Groalia: 2-3 cm** | 0.12 | | | 4 % | | | 1.3 | | | 33 % | | | 10.1 | | | 64 % | | |
| **Groalia: 3-8 cm** | 0.03 | 0.031 | 99 % | 9 % | 4.8 % | 53% | 0.14 | 0.089 | 64 % | 51 % | 7.8 % | 15.3 % | 1.1 | 0.527 | 46 % | 40 % | 8.1 % | 20.1 % |
| **Groalia: 8-13 cm** | 0.054 | | | 32 % | | | 0.05 | | | 40 % | | | 0.41 | | | 28 % | | |

*Table C3: Test of significant differences for activity concentration and proportions of $^{137}$Cs between grain sizes. Marked p values- significant.*

| Test: | Matrix 1 | Matrix 2 | p-value |
|---|---|---|---|
| **Dependent t-test** | Sand (% of total activity) | Silt( % of total activity) | 0.001 |
| | Silt( % of total activity) | Clay (% of total activity) | 0.158 |
| | Sand (% of total activity) | Clay (% of total activity) | 0.168 |
| | Sand Bq/g | Silt Bq/g | 0.018 |
| | Silt Bq/g | Clay Bq/g | 0.016 |
| | Clay Bq/g | Sand Bq/g | 0.013 |

*Table C4: Correlation tests between activity proportion and proportion for sand, silt, and clay, between the proportion of $^{137}$Cs in sand. silt and clay and pH and between the proportion of $^{137}$Cs in sand. silt and clay Organic matter content. Marked p values- significant.*

| Test: | Matrix 1 | Matrix 2 | p-value | r-value |
|---|---|---|---|---|
| **Pearson correlation test** | Sand (% of total activity) | Sand (% in sample) | 0.043 | 0.65 |
| | Silt( % of total activity) | Silt (% in sample) | 0.918 | -0.04 |
| | Clay (% of total activity) | Clay(% in sample) | <0.001 | 0.92 |
| | Sand (% of total activity) | pH | 0.462 | -0.26 |
| | Silt( % of total activity) | pH | 0.257 | -0.4 |
| | Clay (% of total activity) | pH | 0.111 | 0.54 |
| | Sand (% of total activity) | Organic matter content | 0.015 | 0.74 |
| | Silt( % of total activity) | Organic matter content | 0.643 | -0.17 |
| | Clay (% of total activity) | Organic matter content | 0.105 | -0.54 |

## Appendix D: Sequential extraction

*Table D1: Result of measurement of $^{133}$Cs in the fractions from sequential extraction and sum of all measured. (1). (2) and (3) give the core number.*

| Sample | Step 1 mg/kg | Step 3 mg/kg | Step 4 mg/kg | Step 5 mg/kg | Step 6 mg/kg | Sum mg/kg |
|---|---|---|---|---|---|---|
| Nerlifjellet: 0-3 cm(1) | <LOD | 10.18 | 5.16 | 3.25 | 4.07 | 22.64 |
| Nerlifjellet: 0-3 cm(2) | <LOD | 2.80 | <LOQ | 1.11 | 0.99 | 4.90 |
| Nerlifjellet: 0-3 cm(3) | <LOQ | 8.02 | <LOQ | 4.28 | 3.81 | 16.11 |
| Nerlifjellet: 3-7.5 cm(1) | <LOD | 5.65 | <LOQ | 1.55 | 15.22 | 22.42 |
| Nerlifjellet: 3-7.5 cm(2) | <LOD | 1.78 | <LOQ | 1.28 | 2.51 | 5.57 |
| Nerlifjellet: 3-7.5 cm(3) | <LOQ | 8.30 | <LOQ | 2.44 | 15.53 | 26.27 |
| Nerlifjellet: 7.5-13 cm(1) | <LOQ | 17.92 | 4.32 | 9.45 | 141.10 | 172.79 |
| Nerlifjellet: 7.5-13 cm(2) | <LOQ | 9.49 | <LOD | 5.96 | 109.23 | 124.68 |
| Nerlifjellet: 7.5-13 cm(3) | <LOD | 8.95 | 5.23 | 3.02 | 201.46 | 218.65 |
| Groalia: 0-2 cm(1) | <LOD | 10.29 | <LOQ | 7.74 | 203.50 | 221.53 |
| Groalia: 0-2 cm(2) | <LOD | 6.59 | <LOQ | 8.47 | 191.28 | 206.35 |
| Groalia: 0-2 cm(3) | <LOD | 9.02 | <LOD | 9.99 | 281.88 | 300.89 |
| Groalia: 2-3 cm(1) | <LOD | 5.25 | <LOD | 4.29 | 307.59 | 317.13 |
| Groalia: 2-3 cm(2) | <LOD | 7.87 | <LOD | 3.47 | 241.83 | 253.17 |
| Groalia: 2-3 cm(3) | <LOD | 10.77 | <LOD | 3.33 | 252.39 | 266.50 |
| Groalia: 3-8 cm(1) | <LOD | 10.25 | <LOQ | 1.26 | 214.11 | 225.62 |
| Groalia: 3-8 cm(2) | <LOD | 6.79 | <LOD | 0.82 | 165.22 | 172.83 |
| Groalia: 3-8 cm(3) | <LOD | 10.33 | <LOD | 0.89 | 242.23 | 253.45 |

*Table D2: Proportion of $^{133}$Cs in the extracts from each step. The proportion is calculated based on the sum of all $^{133}$Cs measured in the extractions. When one or more of the replicates measured above LOD, values bellow LOD were replaced with ½LOD. When none of the replicates measured above LOD, no value replaced LOD, and the proportion was given 0%.For values between LOD and LOQ, the measured value was used. (1),(2) and (3) gives the core number. Values calculated based on <LOD being estimated to 0% is marked in blue. Values replaced with ½LOD is marked in green. Values bellow LOQ, were the measured value is used is marked in red.*

| Sample | Step 1 | Step 3 | Step 4 | Step 5 | Step 6 |
|---|---|---|---|---|---|
| **Nerlifjellet: 0-3 cm(1)** | 0.5 % | 44.7 % | 22.7 % | 14.3 % | 17.9 % |
| **Nerlifjellet: 0-3 cm(2)** | 1.7 % | 43.8 % | 21.6 % | 17.4 % | 15.5 % |
| **Nerlifjellet: 0-3 cm(3)** | 1.2 % | 40.2 % | 18.0 % | 21.5 % | 19.1 % |
| **Nerlifjellet: 3-7.5 cm(1)** | 0.4 % | 22.2 % | 11.5 % | 6.1 % | 59.8 % |
| **Nerlifjellet: 3-7.5 cm(2)** | 1.4 % | 23.4 % | 25.6 % | 16.7 % | 32.9 % |
| **Nerlifjellet: 3-7.5 cm(3)** | 0.8 % | 27.4 % | 12.4 % | 8.1 % | 51.3 % |
| **Nerlifjellet: 7.5-13 cm(1)** | 0.2 % | 10.4 % | 2.5 % | 5.5 % | 81.5 % |
| **Nerlifjellet: 7.5-13 cm(2)** | 0.2 % | 7.6 % | 0.5 % | 4.7 % | 87.0 % |
| **Nerlifjellet: 7.5-13 cm(3)** | 0.1 % | 4.1 % | 2.4 % | 1.4 % | 92.1 % |
| **Groalia: 0-2 cm(1)** | 0.0 % | 4.6 % | 1.5 % | 3.4 % | 90.5 % |
| **Groalia: 0-2 cm(2)** | 0.0 % | 3.1 % | 1.4 % | 4.0 % | 91.4 % |
| **Groalia: 0-2 cm(3)** | 0.0 % | 3.0 % | 0.2 % | 3.3 % | 93.5 % |
| **Groalia: 2-3 cm(1)** | 0.0 % | 1.7 % | 0.0 % | 1.4 % | 97.0 % |
| **Groalia: 2-3 cm(2)** | 0.0 % | 3.1 % | 0.0 % | 1.4 % | 95.5 % |
| **Groalia: 2-3 cm(3)** | 0.0 % | 4.0 % | 0.0 % | 1.3 % | 94.7 % |
| **Groalia: 3-8 cm(1)** | 0.0 % | 4.5 % | 0.6 % | 0.6 % | 94.3 % |
| **Groalia: 3-8 cm(2)** | 0.0 % | 3.9 % | 0.3 % | 0.5 % | 95.3 % |
| **Groalia: 3-8 cm(3)** | 0.0 % | 4.1 % | 0.2 % | 0.3 % | 95.4 % |

*Table D3: The mean proportion of $^{133}$Cs and standard deviation in the extracts from each step of sequential extractions for each of the soil layers for both sites. N=3. SD- standard deviation R SD- relative standard deviation. Values calculated based on <LOD being estimated to 0% is marked in blue.*

| **Layer** | **Step 1** | **SD** | **R SD** | **Step 3** | **SD** | **R SD** | **Step 4** | **SD** | **R SD** | **Step 5** | **SD** | **R SD** | **Step 6** | **SD** | **R SD** |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Nerlifjellet: 0-3 cm** | 1.14 % | 0.62 % | 54.67 % | 42.90 % | 2.37 % | 5.51 % | 20.76 % | 2.44 % | 11.75 % | 17.70 % | 3.61 % | 20.40 % | 17.49 % | 1.82 % | 10.41 % |
| **Nerlifjellet: 3-7.5 cm** | 0.89 % | 0.51 % | 57.52 % | 24.32 % | 2.75 % | 11.29 % | 16.52 % | 7.85 % | 47.54 % | 10.30 % | 5.67 % | 55.02 % | 47.98 % | 13.74 % | 28.63 % |
| **Nerlifjellet: 7.5-13 cm** | 0.15 % | 0.08 % | 57.67 % | 7.33 % | 3.14 % | 42.76 % | 1.78 % | 1.14 % | 64.22 % | 3.86 % | 2.18 % | 56.45 % | 86.88 % | 5.29 % | 6.09 % |
| **Groalia: 0-2 cm** | 0.00 % | | | 3.57 % | 0.87 % | 24.42 % | 1.04 % | 0.73 % | 70.61 % | 3.60 % | 0.39 % | 10.85 % | 91.79 % | 1.55 % | 1.69 % |
| **Groalia: 2-3 cm** | 0.00 % | | | 2.94 % | 1.20 % | 40.96 % | 0.00 % | | | 1.33 % | 0.06 % | 4.89 % | 95.74 % | 1.16 % | 1.21 % |
| **Groalia: 3-8 cm** | 0.00 % | | | 4.17 % | 0.31 % | 7.48 % | 0.38 % | 0.18 % | 47.50 % | 0.46 % | 0.10 % | 22.59 % | 94.99 % | 0.56 % | 0.59 % |

*Table D4: Result of measurement of* $^{137}$*Cs in the fractions from sequential extraction and sum of all measured. Loss of analyte calculated by comparing premeasured activity for each sample and the total measured. (1),(2), and (3) give the core number.*

| Sample | Step 1 | Step 3 | % RU | Step 4 | Step 5 | % RU | Step 6 | % RU | Residue | % RU | Total Measured | Total premeasured | Loss of analyte % |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Nerlifjellet: 0-3 cm(1) | <LOD | <LOD | | <LOD | <LOD | | 0.33300 | 11.7 % | 0.585 | 0.06 | 0.918 | 2.04 | 55 % |
| Nerlifjellet: 0-3 cm(2) | <LOD | <LOD | | <LOD | <LOD | | 0.363 | 12.1 % | 0.475 | 0.03 | 0.838 | 1.691 | 50 % |
| Nerlifjellet: 0-3 cm(3) | <LOD | <LOD | | <LOD | <LOD | | 0.498 | 12.1 % | 1.928 | 0.03 | 2.426 | 2.827 | 14 % |
| Nerlifjellet: 3-7.5 cm(1) | <LOD | <LOD | | <LOD | <LOD | | 0.313 | 8.1 % | 0.014 | 0.05 | 0.313 | 0.935 | 67 % |
| Nerlifjellet: 3-7.5 cm(2) | <LOD | <LOD | | <LOD | <LOD | | <LOD | | 0.515 | 0.08 | 0.515 | 0.952 | 46 % |
| Nerlifjellet: 3-7.5 cm(3) | <LOD | <LOD | | <LOD | <LOD | | 0.312 | 7.7 % | 0.382 | 0.08 | 0.694 | 1.502 | 54 % |
| Nerlifjellet: 7.5-13 cm(1) | <LOD | <LOD | | <LOD | <LOD | | 0.952 | 9.3 % | 0.623 | 0.06 | 1.575 | 1.55 | -2 % |
| Nerlifjellet: 7.5-13 cm(2) | <LOD | <LOD | | <LOD | <LOD | | 1.07 | 5.2 % | 0.551 | 0.04 | 1.621 | 1.639 | 1 % |
| Nerlifjellet: 7.5-13 cm(3) | <LOD | <LOD | | <LOD | <LOD | | 0.573 | 6.1 % | <LOD | | 0.573 | 0.68 | 16 % |
| Groalia: 0-2 cm(1) | <LOD | <LOD | | <LOD | <LOD | | 5.101 | 9.3 % | 7.416 | 0.05 | 12.834 | 12.85 | 0 % |
| Groalia: 0-2 cm(2) | <LOD | 0.1766 | 20.0 % | <LOD | 0.883 | 11.1 % | 15.013 | 3.7 % | 25.313 | 0.03 | 41.3856 | 42.2 | 2 % |
| Groalia: 0-2 cm(3) | <LOD | <LOD | | <LOD | <LOD | | 1.616 | 8.5 % | 2.664 | 0.09 | 4.28 | 4.43 | 3 % |
| Groalia: 2-3 cm(1) | <LOD | 0.418 | 9.8 % | <LOD | <LOD | | 5.167 | 3.4 % | 8.069 | 0.05 | 13.78 | 14.134 | 3 % |
| Groalia: 2-3 cm(2) | <LOD | <LOD | | <LOD | <LOD | | 3.11 | 6.8 % | 10.583 | 0.04 | 13.693 | 13.4 | -2 % |
| Groalia: 2-3 cm(3) | <LOD | 0.424 | 5.5 % | <LOD | <LOD | | 3.022 | 6.9 % | 7.451 | 0.04 | 10.897 | 10.945 | 0 % |
| Groalia: 3-8 cm(1) | <LOD | <LOD | | <LOD | <LOD | | 0.366 | 6.3 % | 1.412 | 0.03 | 1.778 | 1.646 | -8 % |
| Groalia: 3-8 cm(2) | <LOD | <LOD | | <LOD | <LOD | | 0.133 | 14.2 % | 0.623 | 0.04 | 0.7557 | 0.959 | 21 % |
| Groalia: 3-8 cm(3) | <LOD | <LOD | | <LOD | <LOD | | 0.109 | 8.9 % | <LOD | | 0.109 | 0.160 | 32 % |

*Table D5: Proportion of* $^{137}$*Cs in the extracts from each step. The proportion is calculated based on the sum of all* $^{173}$*Cs measured in the extractions. For values below LOQ were another replicate had a value over LOQ, the values were replaced with ½LOQ. When non of the replicates had higher values than LOQ, value was replaced with 0% (marked blue), these values is marked in blue. Vaues being replaced by ½LOQ is marked in green (1), (2), and (3) give the core number.*

| | **Step 1** | **Step 3** | **Step 4** | **Step 5** | **Step 6** | **Residue** |
|---|---|---|---|---|---|---|
| **Nerlifjellet: 0-3 cm(1)** | 0.0 % | 0.00 % | 0.00 % | 0.00 % | 36.27 % | 63.73 % |
| **Nerlifjellet: 0-3 cm(2)** | 0.0 % | 0.00 % | 0.00 % | 0.00 % | 43.32 % | 56.68 % |
| **Nerlifjellet: 0-3 cm(3)** | 0.0 % | 0.00 % | 0.00 % | 0.00 % | 20.53 % | 79.47 % |
| **Nerlifjellet: 3-7.5 cm(1)** | 0.0 % | 0.00 % | 0.00 % | 0.00 % | 95.72 % | 4.28 % |
| **Nerlifjellet: 3-7.5 cm(2)** | 0.0 % | 0.00 % | 0.00 % | 0.00 % | 2.65 % | 97.35 % |
| **Nerlifjellet: 3-7.5 cm(3)** | 0.0 % | 0.00 % | 0.00 % | 0.00 % | 44.96 % | 55.04 % |
| **Nerlifjellet: 7.5-13 cm(1)** | 0.0 % | 0.00 % | 0.00 % | 0.00 % | 60.44 % | 39.56 % |
| **Nerlifjellet: 7.5-13 cm(2)** | 0.0 % | 0.00 % | 0.00 % | 0.00 % | 66.01 % | 33.99 % |
| **Nerlifjellet: 7.5-13 cm(3)** | 0.0 % | 0.00 % | 0.00 % | 0.00 % | 97.61 % | 2.39 % |
| **Groalia: 0-2 cm(1)** | 0.0 % | 0.36 % | 0.00 % | 0.36 % | 40.46 % | 58.82 % |
| **Groalia: 0-2 cm(2)** | 0.0 % | 0.43 % | 0.00 % | 2.13 % | 36.28 % | 61.16 % |
| **Groalia: 0-2 cm(3)** | 0.0 % | 1.03 % | 0.00 % | 1.03 % | 36.98 % | 60.96 % |
| **Groalia: 2-3 cm(1)** | 0.0 % | 3.06 % | 0.00 % | 0.00 % | 37.84 % | 59.10 % |
| **Groalia: 2-3 cm(2)** | 0.0 % | 0.33 % | 0.00 % | 0.00 % | 22.64 % | 77.03 % |
| **Groalia: 2-3 cm(3)** | 0.0 % | 3.89 % | 0.00 % | 0.00 % | 27.73 % | 68.38 % |
| **Groalia: 3-8 cm(1)** | 0.0 % | 0.00 % | 0.00 % | 0.00 % | 20.58 % | 79.42 % |
| **Groalia: 3-8 cm(2)** | 0.0 % | 0.00 % | 0.00 % | 0.00 % | 17.56 % | 82.44 % |
| **Groalia: 3-8 cm(3)** | 0.0 % | 0.00 % | 0.00 % | 0.00 % | 88.62 % | 11.38 % |

*Table D6: Proportion of $^{137}$Cs in the extracts from each step. The proportion is calculated based on the premeasured activity of the sample. (1), (2), and (3) give the core number.*

| Layer | Step 1 | Step 3 | Step 4 | Step 5 | Step 6 | Residue |
|---|---|---|---|---|---|---|
| Nerlifjellet: 0-3 cm(1) | <LOQ | <LOQ | <LOQ | <LOQ | 36.27 % | 63.73 % |
| Nerlifjellet: 0-3 cm(2) | <LOQ | <LOQ | <LOQ | <LOQ | 43.32 % | 56.68 % |
| Nerlifjellet: 0-3 cm(3) | <LOQ | <LOQ | <LOQ | <LOQ | 20.53 % | 79.47 % |
| Nerlifjellet: 3-7.5 cm(1) | <LOQ | <LOQ | <LOQ | <LOQ | 95.72 % | 4.28 % |
| Nerlifjellet: 3-7.5 cm(2) | <LOQ | <LOQ | <LOQ | <LOQ | <LOQ | 97.35 % |
| Nerlifjellet: 3-7.5 cm(3) | <LOQ | <LOQ | <LOQ | <LOQ | 44.96 % | 55.04 % |
| Nerlifjellet: 7.5-13 cm(1) | <LOQ | <LOQ | <LOQ | <LOQ | 60.44 % | 39.56 % |
| Nerlifjellet: 7.5-13 cm(2) | <LOQ | <LOQ | <LOQ | <LOQ | 66.01 % | 33.99 % |
| Nerlifjellet: 7.5-13 cm(3) | <LOQ | <LOQ | <LOQ | <LOQ | 97.61 % | <LOQ |
| Groalia: 0-2 cm(1) | <LOQ | <LOQ | <LOQ | <LOQ | 40.46 % | 58.82 % |
| Groalia: 0-2 cm(2) | <LOQ | 0.43 % | <LOQ | 2.13 % | 36.28 % | 61.16 % |
| Groalia: 0-2 cm(3) | <LOQ | <LOQ | <LOQ | <LOQ | 36.98 % | 60.96 % |
| Groalia: 2-3 cm(1) | <LOQ | 3.06 % | <LOQ | <LOQ | 37.84 % | 59.10 % |
| Groalia: 2-3 cm(2) | <LOQ | <LOQ | <LOQ | <LOQ | 22.64 % | 77.03 % |
| Groalia: 2-3 cm(3) | <LOQ | 3.89 % | <LOQ | <LOQ | 27.73 % | 68.38 % |
| Groalia: 3-8 cm(1) | <LOQ | <LOQ | <LOQ | <LOQ | 20.58 % | 79.42 % |
| Groalia: 3-8 cm(2) | <LOQ | <LOQ | <LOQ | <LOQ | 17.56 % | 82.44 % |
| Groalia: 3-8 cm(3) | <LOQ | <LOQ | <LOQ | <LOQ | 88.62 % | <LOQ |

*Table D7: Mean proportion and standard deviation of $^{137}$Cs in the extracts from each step for each layer for each site. The proportion is calculated based on the sum of all $^{137}$Cs measured in the extractions. SD- standard deviation R SD- relative standard deviation. Values calculated based on <LOQ being estimated to 0% is marked in blue.*

| **Layer** | **Step 1** | **Step 3** | **SD** | **R SD** | **Step 4** | **Step 5** | **SD** | **R SD** | **Step 6** | **SD** | **R SD** | **Residuals** | **SD** | **R SD** |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Nerlifjellet: 0-3cm** | 0.0 % | 0.00 % | | | 0.00 % | 0.00 % | | | 42.25 % | 32.81 % | 77.64 % | 57.75 % | 32.81 % | 56.81 % |
| **Nerlifjellet: 3-7.5 cm** | 0.0 % | 2.35 % | 1.63 % | 69.2 % | 0.00 % | 0.00 % | | | 29.42 % | 6.30 % | 21.41 % | 68.23 % | 7.40 % | 10.84 % |
| **Nerlifjellet: 7.5-13 cm** | 0.0 % | 0.60 % | 0.30 % | 50.0 % | 0.00 % | 1.17 % | 0.73 % | 62.4 % | 38.15 % | 1.85 % | 4.84 % | 60.71 % | 1.16 % | 1.91 % |
| **Groalia: 0-2 cm** | 0.0 % | 0.00 % | | | 0.00 % | 0.00 % | | | 74.69 % | 16.37 % | 21.92 % | 25.31 % | 16.37 % | 64.67 % |
| **Groalia: 2-3 cm** | 0.0 % | 0.00 % | | | 0.00 % | 0.00 % | | | 47.77 % | 38.05 % | 79.64 % | 52.23 % | 38.05 % | 72.85 % |
| **Groalia: 3-8cm** | 0.0 % | 0.00 % | | | 0.00 % | 0.00 % | | | 33.37 % | 9.53 % | 28.55 % | 66.63 % | 9.53 % | 14.30 % |

*Table D8: Mean proportion of reversibly bound (steps 1 and 3). irreversibly bound (step 4. step 5 and step 6) and inert (residue) $^{137}$Cs and standard deviation for each layer in each site. Values calculated based on <LOQ being estimated to 0% is marked in blue.*

| | **Reversably bound** | **SD** | **R SD** | **Irreversibly bound** | **SD** | **R SD** | **Inert** | **SD** | **R SD** |
|---|---|---|---|---|---|---|---|---|---|
| **Nerlifjellet: 0-3cm** | 0.0 % | 0.0 % | 0.0 % | 33.4 % | 9.5 % | 28.5 % | 66.6 % | 9.5 % | 14.3 % |
| **Nerlifjellet: 3-7.5 cm** | 0.0 % | 0.0 % | 0.0 % | 47.8 % | 38.0 % | 79.6 % | 52.2 % | 38.0 % | 72.9 % |
| **Nerlifjellet: 7.5-13 cm** | 0.0 % | 0.0 % | 0.0 % | 74.7 % | 16.4 % | 21.9 % | 25.3 % | 16.4 % | 64.7 % |
| **Groalia: 0-2 cm** | 0.3 % | 0.1 % | 45.6 % | 39.0 % | 1.3 % | 3.3 % | 60.7 % | 1.2 % | 1.9 % |
| **Groalia: 2-3 cm** | 2.4 % | 1.6 % | 69.2 % | 29.4 % | 6.3 % | 21.4 % | 68.2 % | 7.4 % | 10.8 % |
| **Groalia: 3-8cm** | 0.0 % | 0.0 % | 0.0 % | 42.3 % | 32.8 % | 77.6 % | 57.7 % | 32.8 % | 56.8 % |

*Table D9: Test of significant differences in proportions of $^{137}$Cs and of $^{133}$Cs. Marked p values- significant*

| Test: | Isotope | Matrix 1 | Matrix 2 | p-value |
|---|---|---|---|---|
| Dependent t-test | $^{137}$Cs | Step 6% | Residuals % | 0.295 |
| | | Irreversibly bound (%) | Inert (%) | 0.419 |
| Independent t-test | | Step 6 (%) Nerlifjellet | Step 6(%) Groalia | 0.243 |
| | | Residuals(%) Nerlifjellet | Residuals (%) Groalia | 0.286 |
| Welsh's t-test | | Loss (%$^{137}$Cs) in >40% | Loss (%$^{137}$Cs) <40% | <0.001 |
| | | Step 6 (%) >40% organic matter | Step 6 (%) <40% organic matter | 0.72 |
| | | Residuals (%) >40% organic matter | Residuals (%) >40% organic matter | 0.66 |
| Dependent t-test | $^{133}$Cs | Step 6 (%) | Step 3(%) | <0.001 |
| | | Step 6 (%) | Step 4(%) | <0.001 |
| | | Step 6 (%) | Step 5 (%) | <0.001 |
| Welsh's t-test | | Step 6(%) >40% organic matter | Step 6(%) <40% organic matter | <0.001 |
| | | Step 5 (%) >40% organic matter | Step 5 (%) <40% organic matter | 0.005 |
| | | Step 4 (%) >40% organic matter | Step 4 (%) <40% organic matter | <0.001 |
| | | Step 3 (%) >40% organic matter | Step 3 (%) <40% organic matter | 0.001 |

*Table D10: Correlation tests between activity proportion of $^{133}$Cs and $^{137}$Cs in different extracts and organic matter and between activity proportion of $^{133}$Cs and $^{137}$Cs in different extracts and pH. Marked p values- significant.*

| Test: | Isotope | Matrix 1 | Matrix 2 | R-value | p-value |
|---|---|---|---|---|---|
| Pearson correlation test | $^{137}$Cs | Step 6 (%) | Organic matter content | -0.16 | 0.527 |
| | | Step 6 (%) | pH | -0.36 | 0.17 |
| | | Residuals (%) | Organic matter content | 0.17 | 0.488 |
| | | Residuals (%) | pH | 0.32 | 0.189 |
| | $^{133}$Cs | Step 3 (%) | Organic matter content | 0.89 | <0.001 |
| | | Step 3 (%) | pH | -0.45 | 0.063 |
| | | Step 4 (%) | Organic matter content | 0.92 | <0.001 |
| | | Step 4 (%) | pH | -0.44 | 0.068 |
| | | Step 5 (%) | Organic matter content | 0.89 | <0.001 |
| | | Step 5 (%) | pH | -0.38 | 0.12 |
| | | Step 6 (%) | Organic matter content | -0.93 | <0.001 |
| | | Step 6 (%) | pH | 0.44 | 0.066 |

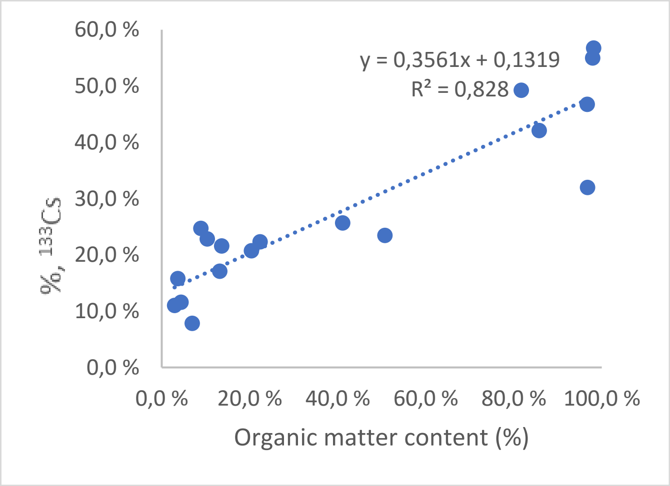

*Figure D1: Linear regression analysis of the proportion of $^{133}$Cs in step 3 compared to organic matter content.*

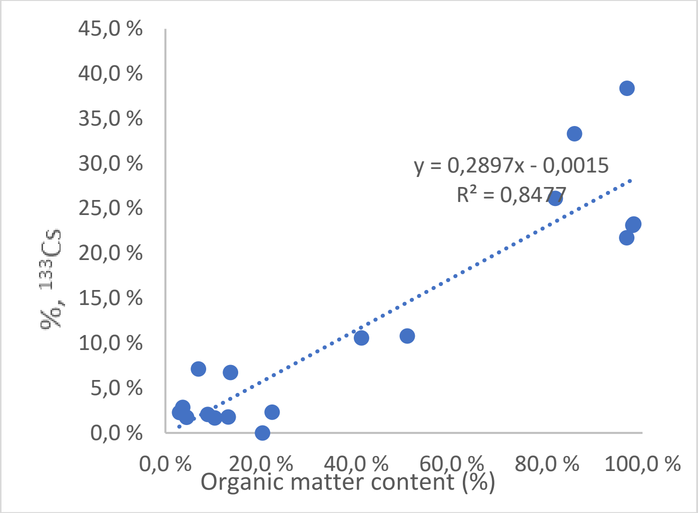

*Figure D2: Linear regression analysis of the proportion of* $^{133}$*Cs in step 4 compared to organic matter content.*

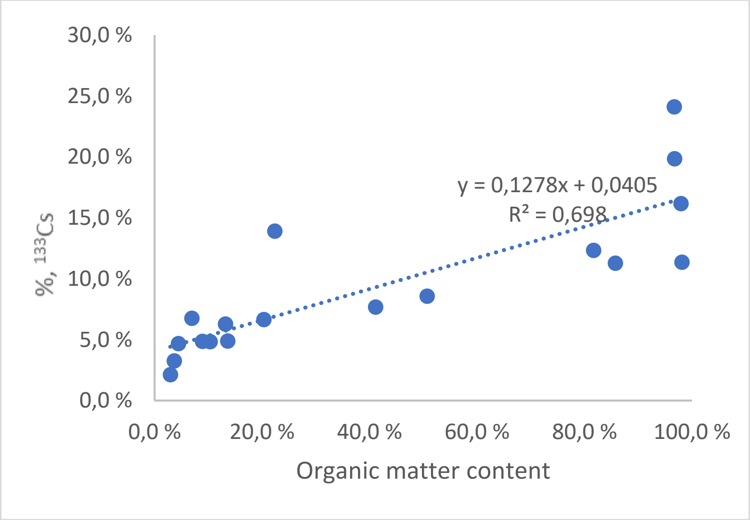

*Figure D3: Linear regression analysis of the proportion of* $^{133}$*Cs in step 5 compared to organic matter content.*

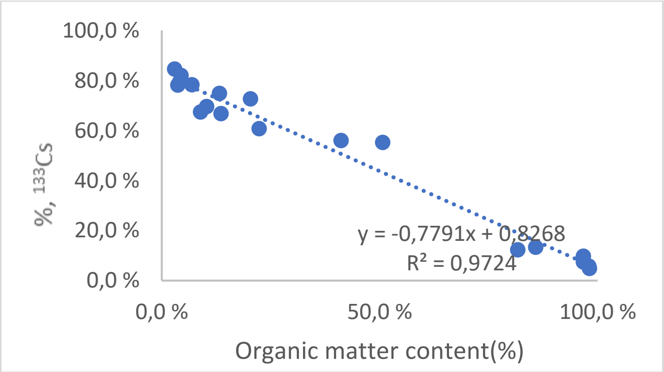

*Figure D4: Linear regression analysis of the proportion of* $^{133}$*Cs in step 6 compared to organic matter content.*

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