Norwegian University
of Life Sciences

Master's Thesis 2018 60 ECTS

Faculty of Environmental Sciences and Natural Resource Management
Ole Christian Lind

Concentration ratios for radionuclides and stable analogues in Rogaland, Norway

Desiree Ong Auren

Master of Science in Radioecology
Faculty of Environmental Sciences and Natural Resource Management

Foreword

This thesis is a joint project between Centre for Environmental Radioactivity (CERAD) and Norwegian Radiation Protection Authority (NRPA) and is a part of the Western Norway Project. The presented work represents the end of my education at the Norwegian University of Life Sciences.

I would like to express my gratitude and appreciation to my main supervisor Ole Christian Lind. Thank you for your patience, support and guidance. I would also want to thank my co-supervisor Håvard Thørring for all information and comments given. Thanks to Karl Andreas Jensen and Solfrid Lohne for guiding me at the laboratory and for answering all of my (hundreds of) questions, as well as Marit N. Pettersen and Yetneberk A. Kassaye for helping me with my samples. I would also like to thank Irene E. Eriksen Dahl and Magdalena Rygalska for all your help, but also good conversations.

To my family and friends, thank you so much! And a special thanks to my “little” family.

Abstract

Norway may be exposed to radioactive deposition from several potential sources due to the country`s geographical location and meteorological conditions (Bartnicki et al., 2016; Klein & Bartnicki, 2018) and it has been demonstrated that a hypothetical accident at the Sellafield reprocessing plant can lead to a significant ^{137}Cs deposition in Rogaland (Ytre-Eide et al., 2009). In cases where events such as this could occur, is it necessary to carry out impact and risk assessments.

Following a deposition will uptake of radionuclides vary depending on the speciation. Radionuclides are present in various physico-chemical forms, where low molecular mass species are considered as mobile and bioavailable, whereas high molecular mass species are regarded as inert. These physico-chemical conditions change over time depending on various mechanisms (as complexation and desorption) in soil (Salbu et al., 2004).

Elemental uptake to plants and animals is often described by factors such as concentration ratios (CR) and transfer coefficients (Harbitz & Skuterud, 1999). Such factors are integrated in impact and risk assessments and as of today are generic values, that are not representative for Rogaland, applied. By application of modeling systems like FDMT (Müller et al., 2003) can these factors be used for calculation of dose estimates to population. Calculated dose is proportional to the transfer factors, hence will the uncertainties associated with such factors be reflected in dose estimates (UNSCEAR, 2015).

In relation to this, was the Western Norway project initiated by CERAD. The main goal of the project is to reduce the uncertainties associated to impact and risk assessments, which includes obtaining site specific data (CERAD, 2016).

The purpose of this study was to provide local concentration ratios for radionuclides (^{137}Cs, Th, U) and stabile analogues of radionuclides (Cs, Co, I, Sr) deriving from agricultural land in Rogaland (referred to as ROGFARM in present work) to minimize uncertainties in impact and risk assessments. This was done by quantifying elements in various sample matrixes (soil, grass, pellets, milk and meat), followed by determination of concentration ratios and transfer coefficients. The results were compared with generic values from both FDMT (Müller et al., 2003) and IAEA (2010). Furthermore, several soil analyses (pH, LOI, grain size, sequential extractions, CEC, XRD) were also carried out to identify any possible influences of soil on concentration ratios.

Comparisons of concentration ratios (CR) for soil-to-plant transfer (geometric mean (GM)) of Cs, ^{137}Cs, Co, Sr, Th and U demonstrated that most of the ROGFARM ratios differed from FDMT and IAEA values with a factor of 2-4, except for U (IAEA) and Th (IAEA) which were 1 and 2 orders of magnitude higher, respectively.
Transfer coefficients (F_m) (GM) for goat`s milk were in general higher (with a factor of 2-4) for data in IAEA and FDMT. Cobalt stood out with IAEA referring to an uptake which was 1 order of magnitude higher than obtained in Rogaland.
Comparisons of uptake in cow`s milk (F_m) (GM) demonstrated that values in both IAEA and FDMT were in general differing from those in present study with a factor of 2-3. Equivalent uptake of ^{137}Cs was observed between ROGFARM, and both IAEA and FDMT.
Transfer coefficients for mutton (GM) between IAEA and ROGFARM differed by a factor of 2. FDMT and ROGFARM had equivalent values for all elements, except for cobalt, where data showed that transfer of Co could be 1 order of magnitude higher in Rogaland than what the FDMT value suggested.
Comparisons of variations in CRs between IAEA and ROGFARM, for both soil-to-plant transfer (GM, GSD) and uptake to animal product (AM, ASD), illustrated that IAEA in general had a relatively greater variation.

The majority of the values compiled in IAEA and applied in FDMT differed from those obtained in present study with at least a factor of 2. It was concluded that by using local data for radionuclides and stable analogues concentration ratios will uncertainties related to impact and risk assessments decrease with a factor of minimum 2, of which corresponding uncertainty will be reduced in dose estimates calculated in FDMT.
Transfer of stable Cs to both grass and milk, correlated with ^{137}Cs transfer, indicating that stable Cs can be used as a proxy for the mobility and bioavailability of ^{137}Cs.
Variation analyses of CR (grass/soil) (Cs, ^{137}Cs, Co, I, Sr, Th, U) and concentrations in mutton (Cs, ^{137}Cs, Co, I, Sr) were performed. There was not identified a significant variation, indicating that the variation within an area is not greater than the variation between the areas.
A significant positive correlation between increased CR (grass/soil) for stable Cs and amount organic matter (OM), increased CR (grass/soil) for ^{137}Cs and OM, and increased uptake of stable Cs in grass and potential bioavailability of Cs, were observed.

Sammendrag

Norge ligger utsatt til for radioaktivt nedfall fra flere potensielle kilder på grunn av landets geografiske plassering og meteorologiske forhold (Bartnicki et al., 2016; Klein & Bartnicki, 2018), og det har blitt demonstrert at en hypotetisk ulykke ved Sellafieldanlegget kan føre til et betydelig ^{137}Cs nedfall i Rogaland (Ytre-Eide et al., 2009). I tilfelle hendelser som dette skulle oppstå er det nødvendig å utføre konsekvens- og risikoanalyser.
Ved nedfall vil opptak av radionuklider variere avhengig av spesiering. Radionuklider finnes i ulike fysisk-kjemiske tilstander, hvorav lav molekylære masser anses som mobile og biotilgjengelige, mens høy molekylære masser beregnes som inerte. Disse fysisk-kjemiske tilstandene endres over tid avhengig av diverse mekanismer (som kompleksdannelse og desorpsjon) i jorda (Salbu et al., 2004).
Opptak til planter og dyr beskrives ofte med faktorer som konsentrasjonsratio (CR) og overføringskoeffisienter (Harbitz & Skuterud, 1999). Slike faktorer er integrert i konsekvens- og risikoanalyser, og per dags dato brukes normalverdier som ikke er representative for Rogaland. Ved bruk av modelleringssystemer som FDMT (Müller et al., 2003), kan disse faktorene brukes til beregning av doseestimat til befolkning. Beregnet dose er proporsjonal med overføringsfaktorene og derfor vil usikkerheter knyttet til slike faktorer reflekteres i doseestimater (UNSCEAR, 2015).
På bakgrunn av dette ble Western Norway prosjektet initiert av CERAD CoE. Hovedmålet med prosjektet er å redusere usikkerheter knyttet til konsekvens- og risikoanalyser, blant annet ved innhenting av lokale data (CERAD, 2016).

Hensikten med dette arbeidet var å fremskaffe lokale konsentrasjonsratioer for radionuklider (^{137}Cs, Th og U) og stabile analoger (Cs, Co, I and Sr) av radionuklider fra landbruk i Rogaland (henvist som ROGFARM i arbeidet) for å redusere usikkerheter i konsekvens- og risikoanalyser. Dette ble gjort ved kvantifisering av grunnstoffer i ulike prøvematrikser (jord, gress, kraftfôr, melk og kjøtt), og deretter ble konsentrasjonsratioer og overføringskoeffisienter bestemt. Resultatene ble sammenlignet med normalverdier fra både FDMT (Müller et al., 2003) og IAEA (2010). Videre ble det også foretatt en rekke jordanalyser (pH, LOI, kornstørrelse, sekvensielle ekstraksjoner, CEC, XRD) for å identifisere mulige påvirkninger av jord på konsentrasjonsfaktorer.

Sammenligninger av konsentrasjonsratioer (CR) for jord til planteoverføring (geometrisk gjennomsnittlig (GM)) av Cs, ^{137}Cs, Co, Sr, Th og U viste at de fleste ROGFARM-ratioene skilte seg fra FDMT- og IAEA-verdier med en faktor på 2-4, med unntak av U (IAEA) og Th (IAEA) som var større med en faktor på henholdsvis 10 og 100.
Overføringskoeffisienter (F_m) (GM) for geitemelk var generelt høyere (med en faktor på 2-4) for data i IAEA og FDMT. Kobolt skilte seg ut, med IAEA som viste til et opptak som var høyere med en faktor på 10 enn utregnet i Rogaland.
Sammenligninger av opptak i kumelk (F_m) (GM) viste at verdier i både IAEA og FDMT generelt skilte seg fra verdier i nåværende studie med en faktor på 2-3. Ekvivalent opptak av ^{137}Cs ble observert mellom ROGFARM, og både IAEA og FDMT.
Overføringskoeffisientene for fårekjøtt (GM) mellom IAEA og ROGFARM varierte med en faktor på 2. FDMT og ROGFARM hadde ekvivalente verdier for alle grunnstoffer, unntatt for kobolt, der dataene viste at overføring av Co kan være en faktor på 10 høyere i Rogaland enn det FDMT-verdien antydet.
Sammenligninger av variasjoner i CRs mellom IAEA og ROGFARM, for både jord til planteoverføring (GM, GSD) og opptak til animalsk produkt (AM, ASD), illustrerte at IAEA generelt hadde en relativt større variasjon.

Mesteparten av verdiene samlet i IAEA og anvendt i FDMT, var forskjellig fra de som ble fremskaffet i nåværende studie med minst en faktor på 2. Det ble konkludert med å bruke lokale data for radionuklider og stabile analoge konsentrasjonsratioer vil usikkerheter knyttet til konsekvens- og risikoanalyser minke med en faktor på minimum 2, hvorav tilsvarende usikkerhet vil reduseres i doseestimater beregnet i FDMT.
Overføring av stabilt Cs til både gress og melk, korrelerte med ^{137}Cs-overføring, som tydet på at stabilt Cs kan brukes som en indikasjon på mobilitet og biotilgjengelighet av ^{137}Cs.
Variasjonsanalyser av CR (gress/jord) (Cs, ^{137}Cs, Co, I, Sr, Th, U), og konsentrasjoner i fårekjøtt (Cs, ^{137}Cs, Co, I, Sr) ble utført. Det ble ikke identifisert signifikant forskjell, som tydet på at variasjonen innad i et område ikke er høyere enn mellom områdene.
En signifikant positiv sammenheng ble observert mellom økt CR (gress/jord) for stabilt Cs ved økt mengde organisk materiale (OM), økt CR (gress/jord) for ^{137}Cs ved økt mengde OM, og økt opptak av stabilt Cs i gress ved økt biotilgjengelighet av Cs.

Content

1. Introduction

1.1. Background

Artificial radioactivity has since it was discovered in 1934 been significant in many scientific disciplines and by the discovery of fission, a new source of energy was available. Induced fission occurs when a fissile atom, most commonly uranium (Lehto & Hou, 2011b), is neutron irradiated creating an unstable nucleus which decays until a stable nuclide is reached. This process originates in fission products, with some being of more radioecological importance than others, depending on half-life, fission yield and biological accumulation (SCOPE 50, 1994).

Norway may be exposed to radionuclide deposition from several nuclear sources, due to geographical and meteorological circumstances. These sources comprise potential accidents from nuclear waste, nuclear power plants as well as nuclear reprocessing sites (Bartnicki et al., 2016; Klein & Bartnicki, 2018). NRPA concluded in a report in 2009 that a possible radioactive fallout from Sellafield could result in considerable greater consequences than the Chernobyl accident, with a ^{137}Cs deposition 0.1-50 times greater in Western Norway compared to the most contaminated sites in Norway (Ytre-Eide et al., 2009).
Impact and risk assessments are developed for environmental protection and are essential in case of radioactive fallout, but as Salbu (2016) explains, such models contain many uncertainties. Comprising factors as sources, transfer to ecosystems and biological effects contribute to the overall uncertainties (Salbu, 2016). Authorities are provided with information regarding countermeasures given by decision support systems (DSS) integrated in the assessments. These systems are dependent on the input parameters, but as of today, these factors are often generic values, thus contributing to additional uncertainties. In relation to this, The Western Norway project, also called Umbrella 8a, was initiated by CERAD CoE. This sub-project simulates a hypothetical radioactive fallout from Sellafield`s waste tanks with a following deposition in Rogaland. The aim is to reduce the overall uncertainties in the assessment (*Figure 1*), thus covering all modules: from sources to consequences.

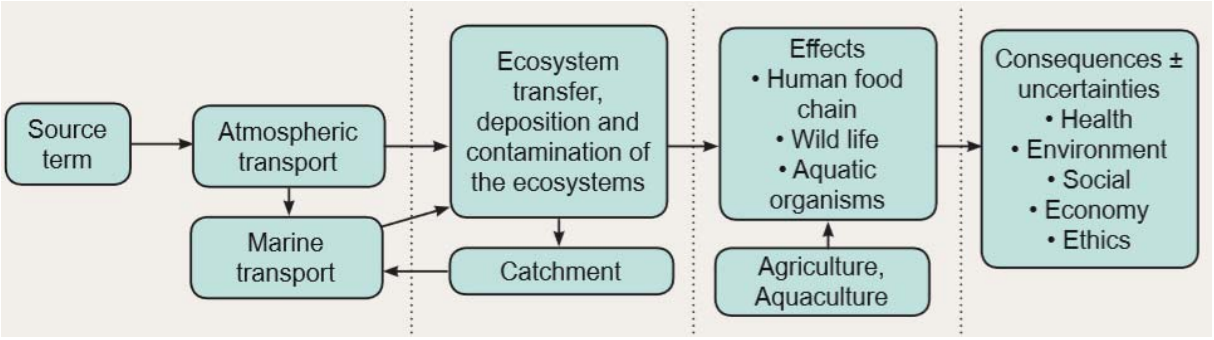


Figure 1. Aspects in impact and risk assessments (CERAD, 2017).

An important assumption in the Western Norway project is that local data will contribute to a decrease in the overall uncertainties (CERAD, 2016). It should be emphasized that Rogaland is an important area concerning both aqua- and agriculture.

The Chernobyl accident raised many questions and a lot of uncertainty in Norway. Experts had different statements, resulting in a lot of concern. This episode underlines how important it is to have a thorough understanding and knowledge about radioactivity, including mobility, bioavailability and transfer of radionuclides in the environment. Elemental transfer is often described by a factor or by a coefficient. The concentration ratio (CR) is frequently used for description of the elemental content between plant and soil, and transfer coefficients (TC) describe the ratio between animal product and daily intake, referred to as F_m (milk) and F_f (flesh) (Harbitz & Skuterud, 1999). Factors representing uptake in animal products are used for quantification of doses to population. Calculation of individual doses can be estimated as follows (*Equation 1*) (UNSCEAR, 2015):

Equation 1. $$D_{ing} = DC_{ing} * AI * Food_{TF} * Deposition\ density$$

*DC_{ing}= dose coefficient (Sv/Bq), AI= average annual intake the foodstuff (kg), $Food_{TF}$= transfer factor for the foodstuff ((Bq a)/kg per (Bq/m^2)), Deposition density (Bq/m^2)

This highlights the practical use of such factors and shows that dose is proportional to the transfer factor. The uncertainties in such factors will therefore be reflected in the quantified dose which is of high relevance in impact and risk assessments.
The transfer model FDMT (Müller et al., 2003), which is incorporated in the impact and risk assessment for Western Norway, applies generic data (Thørring et al., 2016b). Several other generic values are also available, e.g. IAEA (2010) and IAEA (2014). Despite the knowledge

about the uncertainties that follows by using these values, they are often used since obtaining site specific factors is resource demanding (IAEA, 2006).

This thesis comprises calculations of local transfer factors by using stable analogues instead of default values and to try to identify any trends regarding soil characteristics and uptake.

The following chapter includes theory, as well as hypotheses and research questions. Section two provides information about the applied methods and the experimental work. In section three, the results are presented and discussed, ending with the final chapters comprising the conclusions and further work.

1.2. Theory

1.2.1. Speciation

Elemental uptake in plants and animals depend on the mobility and bioavailability of the element which are set by the elemental speciation (Lehto & Hou, 2011c). Elemental speciation is defined as the *"distribution of an element amongst defined chemical species in a system"* (Templeton et al., 2000). Thus, elements can exist in different physical and chemical forms and will behave differently depending on factors as size, charge, oxidation state, complexation, morphology, structure and density. Source term is central in impact and risk assessments and gives information about amount released and the radionuclides physicochemical properties at release point, which is influenced by the release scenario (e.g. pressure and temperature) (Salbu & Skipperud, 2009).

1.2.2. Mobility and bioavailability

Mobility and bioavailability of elements are very specific and are influenced by environmental factors. Elements present as low molecular mass (LMM) species are regarded as mobile and bioavailable, whereas high molecular mass (HMM) species, making up colloids or particles, are considered inert. Elemental speciation is not a permanent physico-chemical form and is controlled by several soil processes. Mechanisms as hydrolysis, complexation and formation of colloids will increase the molecular mass of LMM species, while processes as dissolution and desorption may contribute to weathering of particles. Thus, elements

considered inert may become mobile and bioavailable, whereas LMM species may turn "inert". (Salbu et al., 2004).

Elements have different affinity towards soil components and are differently influenced by soil properties. The degree of importance varies amongst the nuclides, for instance, is soil texture an important factor regarding the mobility and bioavailability of cesium (Cremers et al., 1988). Clay in contrast to sand and silt, represents the reactive part of the soil and has a major role in soil properties due to a high surface area (Lal & Shukla, 2004). It has a permanent negative charge, allowing it to adsorb positively charged ions (Weems, 1904), both by specific adsorption and by ion exchange (Rieuwerts et al., 1998). Cesium (Cs^{+}) gets easily fixed to clayey minerals and binds especially strongly to the interlayers, *frayed edge sites*, of illite, resulting in immobile and non-bioavailable Cs (Cornell, 1993; Cremers et al., 1988). The main factor controlling Sr^{2+} mobility is adsorption by ion exchange; the general trend is an increase in mobility in soils with a high salinity or with a low cation exchange capacity (ATSDR, 2004b). The cation exchange capacity (CEC) is a reversible chemical reaction and a soil`s capacity to retain exchangeable cations depends on the number of negatively charged sites at the surface of clay and organic matter. The cation exchange capacity (CEC) can be quantified (Chapman, 1965; Sumner & Miller, 1996), and in general is a high CEC correlated with fertile soils (McKenzie et al., 2004). Organic matter can also bind metals irreversibly, thus reducing their potential bioavailability (Tessier et al., 1979). In fact are strontium ions readily bound to organic matter and the availability may therefore decrease with an increasing soil organic matter content (Harbitz & Skuterud, 1999).
Finally, metal availability is pH dependent; low pH enhances metal uptake as H^{+} has a higher attraction towards the negative binding sites at colloids (Prasad, 2013).

1.2.3. Stable analogues as indicators of radionuclide behavior

Radionuclides are expected to "over-time" behave like their stable analogues. Elements with a short half-life will not have enough time to follow the same pathways as their stable isotope. With a half-life of 8 days, will ^{131}I not be able to penetrate the soil deep enough for plant uptake, where stable I is taken up by the plant roots (Newman & Unger, 2002). Accordingly, the use of short-lived isotopes as estimators for their long-lived or stable analogues may be restricted as the significance of several long-term processes that might affect their pathways can be excluded. Equilibration of both stable and long-lived radionuclides in environmental

media may primarily be determined by biogeochemical processes (IAEA, 2010), whereas short-lived isotopes are possibly more influenced by physical decay (IAEA, 2010). Deposition of particles is an important factor, and it is essential to not oversee the significance of such particles as they are exposed to weathering. Weathering is a slow process and results in mobilization of LMM species (Salbu et al., 2004). The presence of particles and colloids reflect an inhomogeneous distribution which questions the representativity of samples (Salbu & Skipperud, 2009).

Nevertheless, where study of radioactive elements are not possible, stable isotopes are the best indicators of radioactive elemental behavior. Comparisons of stable analogues and radionuclides showing correlations between both $^{133}Cs/^{137}Cs$ and $^{88}Sr/^{90}Sr$ (Thørring et al., 2016a; Tsukada et al., 2002; Tsukada et al., 2003; Uchida et al., 2007) have been documented.

1.2. Elements

Atmospheric nuclear weapon testing, radioactive discharges, the Chernobyl accident (nrpa, 2013) as well as tropospheric nuclear tests (Wendel et al., 2013) have all contributed to radioactive deposition in Norway. The most significant though are the Soviet weapon testing and the Chernobyl accident which resulted in deposition by primarily the long-lived radionuclides ^{137}Cs and ^{90}Sr (Harbitz & Skuterud, 1999).
In the present work, emphasis is put on the stable analogues of cesium, cobalt, iodine and strontium, and the naturally occurring radioelements, thorium and uranium.

Cesium

Radioactive Cs, especially ^{137}Cs, is one the most significant nuclides within radioecology. It has a long half-life (30.2 y) (Choppin et al., 2013b) and is chemically similar as potassium (SCOPE 50, 1994). Cesium accumulates in soft tissue (Harbitz & Skuterud, 1999) where radiocesium disintegrates by beta decay, followed by gamma emission (Alexander, 2016). Following the Chernobyl accident, intake of radioactive Cs contributed to approximately 95 % of the total effective dose, mostly by consumption of milk and meat. Nursing babies would also be exposed to radioactive Cs as K is present in breast milk (Harbitz & Skuterud, 1999).

Strontium

As cesium, is Sr also of radiological importance. Strontium follows the same pathways as calcium, concentrating primarily in bones. The isotope of most concern is ^{90}Sr because of its long half-life (28.8 years) and the high fission yield (SCOPE 50, 1994) in addition to its release of relatively high energy radiation. It undergoes β decay into its daughter nuclide, Y^{90}, which also emits high β energy (Choppin et al., 2013a). The main source of radioactive Sr is intake of contaminated milk and grain products, thus exposing nursing babies as well (Harbitz & Skuterud, 1999).

Iodine

Even though the fission product ^{131}I (β, γ emitter) is short-lived with a half-life of 8.04 days (Choppin et al., 2013b; Furman, 2017), it may indeed pose a great risk. Like stable iodine, which has an important biological role, it accumulates in the thyroid (Harbitz & Skuterud, 1999). The accumulation in the thyroid is enhanced in populations with a low I status. In fact, an increase in thyroid cancer was seen in the areas around Chernobyl were the daily intake of iodine was relatively low. An important iodine source is through milk, making especially children more vulnerable (Harbitz & Skuterud, 1999).

Cobalt

Radiocobalt is present in large amounts in nuclear power plants and releases of radiocobalt are mainly due to discharges (IRSN, 2012). The most important isotope of Co is the β decaying ^{60}Co with the longest half-life of 5.3 years (ATSDR, 2004a). It emits (high energy) γ rays as well and contributes to relatively high doses at power plants (Lehto & Hou, 2011a).

Thorium and Uranium

Almost 100 % of thorium in the environment exist as ^{232}Th. The radionuclide has a half-life of 140.5 years and disintegrates by alpha decay. Uranium occurs mainly as ^{238}U with a half-life of $2.51*10^9$ years. About 0.7 % (mass weight) exists as the nuclide ^{235}U ($t_{1/2} = 7.04*10^8$ y) (Kathren, 1998). This is the most fissile of the U isotopes and is used as an energy source in nuclear reactors and nuclear weapons. Both elements belong to the group of actinides and all three isotopes disintegrates by alpha decay (Lehto & Hou, 2011b).

Th and U are relatively immobile in soil, but human activities such as mining enhance their mobility (UNSCEAR, 2000) as demonstrated by Popic et al. (2014).

1.3. FDMT- transfer model

Food chain and Dose Module for Terrestrial pathways (FDMT) is a module used within the two decision support systems of ARGOS and JRODOS (Landman et al., 2013). FDMT focuses on radionuclide transfer in food chains in cultivated fields and calculates individual and collective doses to the population arising from ingestion of agricultural products. The transfer of radioactive material is calculated stepwise as shown below (*Figure 2*) (Müller et al., 2003). The model provides information both for short– and long-term, but is most applicable within the first few years after deposition (Thørring, 2018).

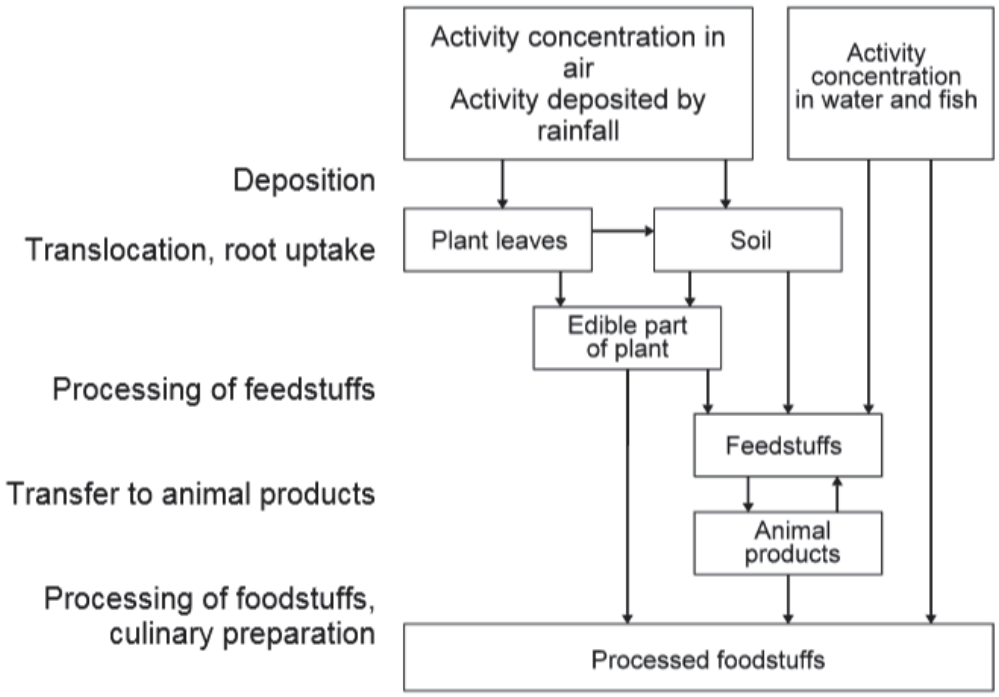


Figure 2. Schematic view of food chain transfer calculations (Müller et al., 2003)

Transfer of radionuclides vary depending on many factors and to increase the applicability of the model, *radioecological regions have* been defined, meaning that regions with similar radioecological conditions may use the same parameters. A country is typically divided in 1 to 5 regions depending on factors as agricultural practice, growing and harvesting time and management of domestic animals. As variations between years would be higher than the variations between more site-specific areas, further divisions are unnecessary (Müller et al., 2003).

1.4. Objectives and hypotheses

The overall goal is to obtain concentration ratios for radionuclides (^{137}Cs, thorium and uranium) and stable analogues (cesium, cobalt, iodine and strontium) of radionuclides in Rogaland to minimize uncertainties in impact and risk assessments by filling gaps in knowledge.

This study addresses the following hypotheses:

- Using locally obtained data for radionuclide and stable analogue concentration ratios will decrease the uncertainties associated with risk assessment modelling
- Stable Cs uptake is correlated with that of radiocesium and can be utilized as a proxy to predict the mobility and bioavailability of ^{137}Cs
- The variation of grass/soil and meat/dry matter concentration ratios can be greater within an area than between areas

The research questions are as follows:

- Will an increase in clay decrease concentration ratios?
- Will an increase in organic matter and/or cation exchange capacity increase concentration ratios?

In order to test the hypotheses and research questions will following be carried out:

- obtaining local concentration ratios for stable isotopes (cesium, cobalt, iodine and strontium) and radionuclides (cesium-137, thorium and uranium)
- quantifying uncertainties in concentration ratios and compare with default values used in models (FDMT and IAEA)
- identifying possible influences of soil on concentration ratios

Elemental concentrations will be quantified in soil, grass, milk, meat and pellets by ICP-MS analyses for calculations of CRs and to identify possible influences of soil properties will organic matter, cation exchange capacity (CEC), grain size, pH, sequential extractions and clay mineral analyses on X-ray diffraction (XRD) be performed.

2. Experimental

2.1. Study area

Sampling was performed as a part of the CERAD funded project ROGFARM at 15 locations on or near five farms (A-E) in Rogaland (*Figure 3*), Norway during an NRPA field work August 25th-27th 2015.

Figure 3. Map of sampling area for ROGFARM.

Samples were collected at 5 farms in Rogaland well separated spatially and representing different types of pastures, soils and land-use. Descriptions of the locations are shown in *Table 1*:

Table 1. Study sites in Rogaland and some characteristics of the farms.

Area	Locality	Type of pasture	Area description	Fertilizer	Animal
A - Nesflaten	A1	Uncultivated*	Steep slope, mainly grass covered. Some forest	Manure from cow, goat and some artificial	Goat
	A2				
	A3				
B - Sandeid	B1	Full grown[1]	Clay rich soil		n.a.[3]
	B2	Cultivated	Humus rich soil (moldjord)		Sheep
	B3			Cow manure and some artificial	Cow
C- Sokna	C1	Cultivated	Grass (high activity area)[2]	Manure from Chicken, sheep/cow and some artificial	Sheep/cow
	C2	Surface cultivated			Sheep/cow
	C3	Full grown[1]			n.a.[3]
D- Lundsneset	D1	Uncultivated	Marshland with organic soil/peat		Wild sheep and some common sheep
	D2	Uncultivated	Humus rich soil (moldjord)		
	D3	Old farmed land		Chicken manure	
E- Brusand	E1	Full grown		Manure from cow, pig and some artificial	Cow
	E2				Cow
	E3	Full grown[1]			n.a.[3]

[1]used for grass production, [2]area limed regularly with sea shell sand to pH 5.5-6, *open lines refers to the locations, thus A1-A3 are all uncultivated, [3] n.a. = not applicable

The sampling sites in area B, C and E were recommended by the farmers as representative locations. For area A the sampling locations were set along a height gradient (A1= 204 masl, A2= 161 masl, A3= 159 masl) and for area D, the chosen representative locations vary in type of pasture.

2.2. Samples and sampling

Soil and grass samples were taken at all locations. Samples of milk, meat and pellets were taken where this was available (*Table 2*).

Table 2. Location and associated samples across all 15 locations. Milk samples represent several individuals as samples were taken from collective tanks.

Area	Locality	Soil samples	Grass samples	Pellets samples*	Milk samples	Meat samples (sheep)
A - Nesflaten	A1	1	1	1 (Formel goat)	1 (goat)	
	A2	1	1			
	A3	1	1			
B - Sandeid	B1	1	1	1 (71010 Formel energy basis 80)	1 (cow)	
	B2	1	1			2
	B3	1	1			
C- Sokna	C1	1	1	1 (71010 Formel energy basis 80)	1 (cow)	5
	C2	1	1			
	C3	1	1			
D- Lundsneset	D1	1	1			
	D2	1	1			
	D3	1	1			
E- Brusand	E1	1	1	1 (71011 Formel energy basis 90)	1 (cow)	
	E2	1	1			
	E3	1	1			
Total		15	15	4	4	7

*pellets are given as a food supplement and are high on energy and/or proteins (Harstad & Vangen, 2015). The pellets are bought at Felleskjøpet.

Milk samples (2 liters) were taken from collecting tanks at areas A, B, C and E. Pellets from the same areas were also sampled. Meat samples (mutton; from neck), were taken from 7 individuals, at area B (n=2) and C (n=5).

Notice that cow in area B in Table 1 is associated to location B3 and in Table 2 to B1. This is because the cows had been in cowshed and been given silages when sampling took place, so for calculation purposes it is most likely better to link the cows to B1 which is used for grass production and giving to the cows prior to milking.

For sampling of soil, a soil probe was used (*Figure 4*):

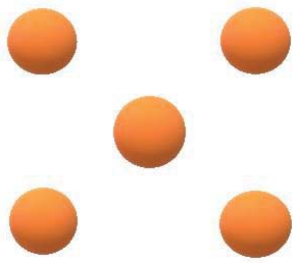


Figure 4. Pattern used for soil sampling.

The distance between the corners were 5x5 meters. The four corners were mixed giving one pooled sample to represent each location, giving 15 samples. The mid samples were divided into separate depths and stored by NRPA for other use. Bulk samples of grass were gathered from the same locations.

2.3. Method

The laboratory work and instrumental analyses described below was been done at the Isotope laboratory and the Soil laboratory of the faculty of Environmental Sciences and Natural Resource Management (MINA), Norwegian University of Life Sciences (NMBU), except for XRD analysis done at the Norwegian National Centre for X-ray diffraction and Scattering, University of Oslo (UiO), as well as some of the sample preparations which were done by Norwegian radiation protection authority (NRPA). All laboratory work regarding ^{137}Cs analysis was also done by NRPA.

2.3.1. Sample preparation

Initial sample preparation was done by NRPA. Soil had been frozen, dried and sieved, while grass was dried and homogenized in a mill. Milk and meat had been through a process of freezing and freeze-drying. Pellets were untreated until transfer to NMBU.
When received at NMBU, the samples were put in a drying cup-board for three days at 40°C degrees, and soil (about 15 g.) and pellets were homogenized. For homogenization of soil, the soil was stirred properly, and an amount was crushed with an automatic mortar for 4 minutes. The soil was then put in small paper envelopes. The concentrates were crushed with a pestle in a sieve, sieved at 2mm and put back in the plastic boxes. The meat samples contain high

concentrations of fat which made homogenization complicated. Meat samples were not subjected to any further treatment. *Figure 5* gives an overview of total sample preparation. All samples had been stored in plastic boxes by NRPA.

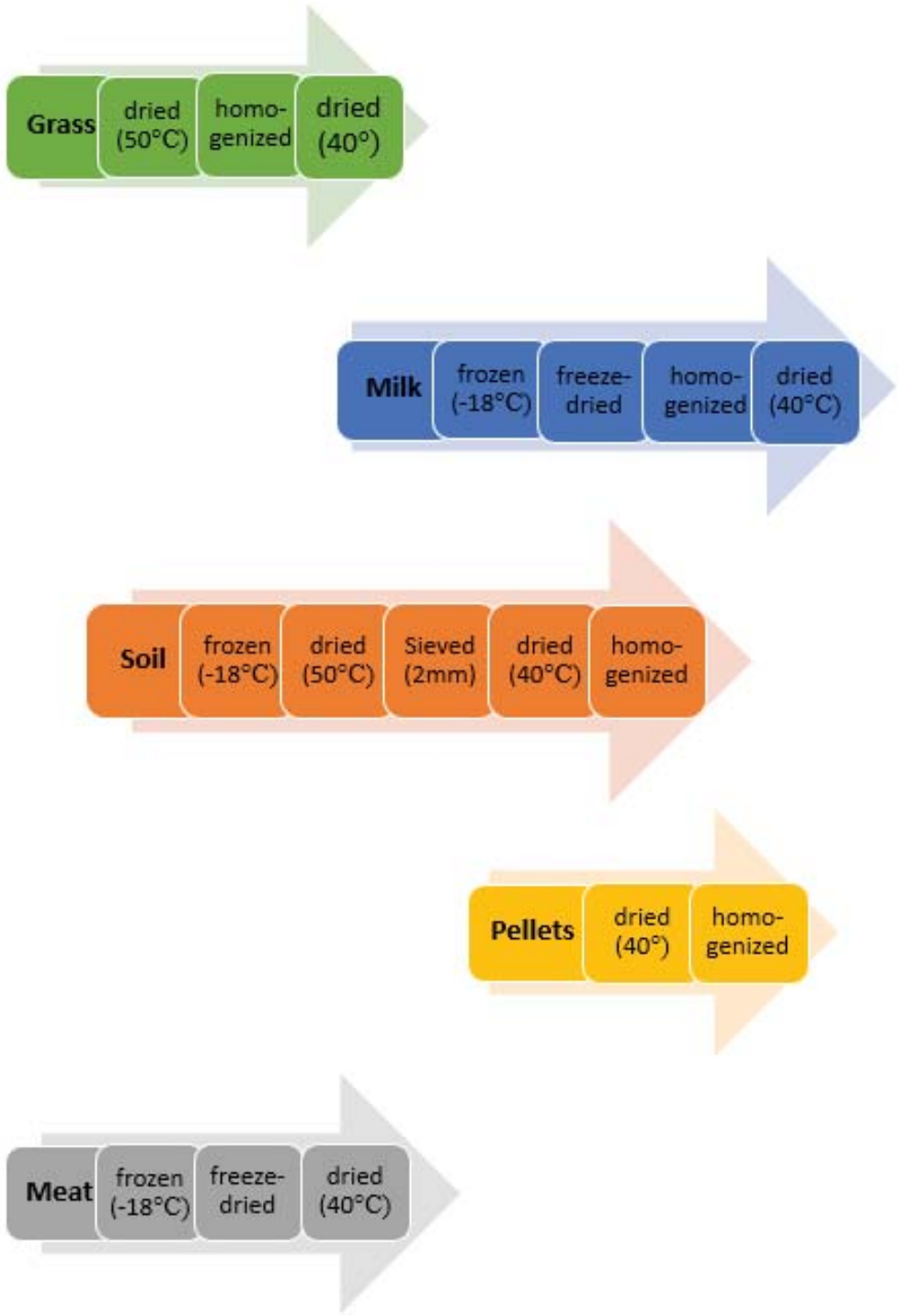


Figure 5. Overview of sample preparation for all matrixes: soil, grass, milk, meat and pellets.

2.3.2. Sample preparation for ICP-MS analysis

Basic dilution (TMAH) of pellets, meat and grass for analysis of primarily I

Pellets, meat and grass (0.2-0.3 g.) were weighed accurately into 15 ml centrifugation tubes, followed by same procedure for certified reference material (CRM), (ERM – BD151, NCS ZC73013 spinage, BCR 129 hay powder). All tubes were added 4 ml of MQ- water (18.1 Ω) and then 1 ml TMAH (Alfa Aesar, electronic grade). Five tubes with blanks containing 4 ml MQ- water and 1 ml with TMAH were also prepared. Samples, CRM`s and blanks were mixed on a vortex and heated at 60°C degrees for 20 hours, followed by re-mixing and heating at 90°C degrees. After two hours, samples were cooled down and diluted with MQ-water, giving a final volume of 10 mL, shaken by hand and centrifuged (4.5 revolutions for 5 min.). The samples were stored for 24 hours allowing the solids to settle, before 2 mL was transferred into a 15 mL centrifuge tube and diluted up to 10 mL with MQ- water (Jensen, 2016).

Decomposition (HNO_3) of pellets, meat and grass for ICP-MS analysis of primarily Cs, Co, Sr, Th and U

Pellets, meat and grass (0.2-0.3 g) were weighed accurately into acid-treated Teflon tubes, along with CRM (NSC ZC73013 spinage, NSC ZC73014 tea, DOLT-5 dogfish liver, 1577b bovine Liver). All tubes were added 2 mL MQ- water (18.1 Ω), 5 mL ultrapure nitric acid (HNO_3) and 250 µL internal standard (IS) containing 2 mg/L Rh, In, Bi and 4 mg/L Ge in 2% (v/v) ultrapure HNO_3. Five blanks containing same amount MQ-water, HNO_3 and IS were prepared. All samples were run in UltraCLAVE (IV Milestone) with a gradually increase in temperature up to 250 ^{O}C degrees and pressure to 160 bar. After decomposition, the samples were transferred into 50 ml centrifuge tubes and diluted with MQ-water up to 50 mL (10% HNO_3) (Jensen, 2016).

Decomposition (HNO_3) of soil for analysis of primarily Th

Soil and CRM (NSC ZC 73007, NSC DC73325) (0.2-0.3 g.) were precisely weighed into acid-treated Teflon tubes, followed by addition of 5 mL ultrapure HNO_3 and 250 µL internal

standard (IS) containing 10 mg/L Rh in 2% (v/v) ultrapure HNO_3. Five blanks containing same amount nitric acid and IS were prepared. All samples were run in UltraCLAVE (IV Milestone) with a temperature at 260 ^{O}C degrees and pressure at 170 bar. The decomposed samples were transferred into 50 mL centrifuge tubes and diluted with MQ-water (18.1 Ω) up to 50 mL (10% HNO_3) (Jensen, 2016).

Basic dilution (TMAH) of soil for analysis of primarily I

Soil and CRM (NSC ZC 73007 and NSC DC 73325) (0.2-0.3 g) were accurately weighed into 15 mL centrifuge tubes. All tubes were added 2 mL of MQ- water (18.1 Ω) and then 1 mL TMAH (Alfa Aesar, electronic grade). Five blanks containing 4 mL of MQ- water and 1 mL TMAH were also prepared. The samples were mixed on a vortex and heated at 60°C degrees for 20 hours, followed by a final procedure of vortex mixing and heating (90°C degrees for two hours). Two hours later, the samples were cooled down, shaken by hand, diluted with MQ- water (up to 10 mL) and centrifuged (4.5 revolutions, 5 min.). The samples were stored for 24 hours, allowing the solids to settle. 2 mL was pipetted into a new centrifuge tube, diluted up to 10 mL with MQ- water, and shaken by hand (Jensen, 2016).

Decomposition (HF) of soil for analysis of primarily Cs, Co, Sr and U

Approximately 0.2-0.3 g of soil and CRM (NSC ZC 73007 and NSC DC 73325) were accurately weighed into 15 mL acid-treated Teflon tubes. 5 mL ultrapure HNO_3 and 250 µL IS (10 mg/L Rh in 2% (V/V) ultrapure HNO_3) and finally 1 mL HF (p.a. quality) were added all tubes. Same amount HNO_3, IS and HF were added five centrifugation tubes for blanks. All samples were run in UltraCLAVE (IV Milestone) with a temperature at 260 ^{O}C degrees and a pressure at 170 bar. After decomposition, the samples were quantitatively moved into 50 mL centrifuge tubes and diluted with MQ-water (18.1 Ω) up to 50 mL (10% HNO_3 and 2% HF). Finally, 1 mL was pipetted into a 10 mL centrifugation tube and diluted up to 10 with MQ-water (1% HNO_3 and 0,2% HF). With HF, further dilution is needed due to its ability to dissolve quartz used in the ICP-MS. Another possibility is to change the vulnerable parts into inert materials (Jensen, 2016).

Basic dilution (BENT) of milk for analysis of primarily I

MQ-water (800 µL, 18.1 Ω) was added 15 mL centrifugation tubes. About 0.2 g of dry milk was weighed accurately into the tubes, followed by addition of 0.100 mL IS (4.00 mg/L Ge, 200 µg/L In, 1.00 mg/L Te, 500 µg/L Tb in 3 % (V/V) NH_4OH) and 0.1 % (w/V) H_4EDTA. Samples were diluted up to 10 mL with BENT, consisting of 4 % (w/V) 1-Butanol, 0,1 % (w/V) H_4EDTA, 5 % (w/V) NH_4OH and 0,1 % (w/V) Triton™ X-100. Five blanks followed the same procedure. For CRM (NCS ZC73013 (spinage), ERM -BD151 (milk)), two centrifugation tubes with 1 mL MQ-water and about 0.1 g. CRM (precisely weighed) were prepared. The tubes were diluted with BENT up to 10 mL (Jensen, 2016).

Decomposition (HNO_3) of milk for analysis of primarily Co, Cs, Sr and U

About 0.4 g. of milk and CRM (1549a (milk), NCS ZC73013 (spinage), ERM - BD151 (milk)) were precisely weighed into acid-treated Teflon tubes. 5 mL MQ-water (18.1 Ω), 5 mL ultrapure HNO_3 and 250 µL IS (2 mg/L Rh, In, Bi and 4 mg/L Ge in 2% (v/v) ultrapure HNO_3) were added all tubes. Identical solutions were also added five acid-treated Teflon tubes for blanks. Finally, all samples were decomposed in UltraCLAVE (IV Milestone) at 260°C degrees and 160 bar and diluted to 50 mL with MQ-water (Jensen, 2016).

ICP-MS analysis

All samples (acid and basic treated), blanks and CRM`s were analyzed at Agilent 8900 QQQ ICP-MS by Head Engineers Marie Vollset and Susanne Birkeland.

2.4. pH measurement

The soil (2mm) was stirred properly to get a homogenized sample as possible. 10 mL soil was transferred to a tube and filled with 25 mL MQ-water. All tubes were properly shaken and left overnight. The following day, all samples were shaken, and the pH-value was measured after roughly 15 minutes. The pH-meter (Orion ROSS combination pH electrode) was calibrated with buffers at pH 4 and pH 7 and controlled by a control solution at pH 6.87 (Krogstad, 1992).

2.5. Loss-on-ignition

As an indicator of total organic carbon (TOC), loss-on-ignition was measured (Krogstad, 1992). The soil (2 mm sieved) was stirred properly to ensure a homogenized sample as possible. About one plastic spoon from each sample was transferred to a previously weighed porcelain crucible. The samples were weighed and dried overnight in 105°C degrees. The next day, all samples were put in desiccators for 30 min. and weighed, giving dry matter. The samples were then put in a calcinating oven at 550 °C degrees for seven hours, cooled down and weighed a final time for determination of loss-on-ignition (*Equation 2* and *Equation 3*).

Equation 2. $$\%\ dry\ matter = \frac{m1}{m2} * 100$$

-where m_1 = sample weight after drying, m_2 = sample weight prior to drying

Equation 3. $$\%\ loss\ on\ ignition = \frac{m1-m3}{m1} * 100$$

-where m_1 = sample weight after drying, m_3 = sample weight after ignition

2.6. Cation exchange capacity (CEC)

The procedure described below is a modified version (Zivanovic, 2011) based on Hesse (1971) and Schollenberger and Simon (1945).

About 3 g. of soil (2 mm sieved) were accurately weighed into 250 mL acid-treated Erlenmeyer flasks. An extraction-solution (1.00 mol/L ammonium acetat, pH 7) was already made according to the reference. 25 mL of the solution was added to all flasks, followed with a thorough turning to ensure fully soaked soil. Six blanks were made. The following day, 250 mL volumetric flasks with filters, were prepared. Filters were saturated with the extraction-solution. The samples were transferred into the newly prepared flasks. This was done by repetitive washing with the extraction-solution until all soil were washed out of the Erlenmeyer flaks and into the filters. Total volume in the end was 250 mL. 20 mL of the extract solution was then titrated with 0.05 M NaOH for calculation of H^+ concentration giving the exchangeable acidity. 15 ml of the extract solution was transferred to 15 mL centrifugation tubes and analyzed at ICP-OES for the quantification of the basic cations, sodium, magnesium, potassium and calcium. The cation exchange capacity was calculated by the use of *Equation 4* and *Equation 5*.

Equation 4. $S = [K^+] + [Mg^{2+}] + [Ca^{2+}] + [Na^+]$

- *where S = total exchangeable bases*

Equation 5. $T = S + [H^+]$

- *where T = cation exchange capacity*

ICP-OES (Agilent MP-4200) analysis was done by Senior Engineer Valentina Zivanovic.

2.7. Grain size

Grain size analysis was done by Engineer Magdalena Rygalska.

The analysis consisted of three parts; pre-treatment, pipetting and sieving, according to Krogstad and Børresen (2015). The first step, pre-treatment, involved primarily oxidizing of organic material and dissolvent of amorphous compounds. In the following step was all samples pipetted giving three fractions of silt (coarse, medium, fine) and clay based on sedimentation time. Finally, the samples were sieved with three sieves (600 µm, 212 µm, and 63µm) for sand fractions. All samples were then dried at 105°C degrees for 24 hours, and then weighed.

2.8. Sequential extraction

Sequential extraction

Sequential extraction was performed by Head Engineer Marit Nandrup Pettersen.

The procedure described is modified (Oughton et al., 1992) and based on Tessier et al. (1979). Sequential extractions involve several steps, dividing the fractions into reversible (step 1-3), irreversibly bound (4-6) and non-soluble fractions. In the present study, the main interest was the reversibly bound fractions which gives an indication of the bioavailability of an element, thus only step 1-3 were done. The first two steps comprise physical sorption, step three electrostatic sorption and the remaining steps (4-6) chemisorped species. The residual fraction represents metals sorped within the crystal lattices of minerals (Salbu, 2006).

Step 1 (water soluble fractions): soil (1-2 g) was weighed into 20 mL scintillation vials and 20 mL Milli-Q water (18.1 Ω) was added, followed by 1 hour mixing at roller-table and centrifugation (10.000 g) for 25 min. Supernatant was transferred by a pipette to a filter. The filtered supernatant was collected in a scintillation vial (20 mL).

Step 2 (exchangeable fractions): 20 mL 1 mol/L NH_4Ac (pH soil in soil) was added to the samples, followed by 2 hours at roller-table and 15 min. centrifugation, prior to filtration.

Step 3 (carbonate associated fractions): 20 mL 1 mol/L NH_4Ac (pH 5) was added to the samples, prior to 2 hours at roller-table, centrifugation for 15 min. and filtration.
Final procedure included washing (10 mL Milli-Q water), shaking and centrifugation for 15 min. and filtration.

ICP-MS preparation

Decomposition and ICP-MS analysis were done by Senior Engineer Karl-Andreas Jensen.
Step 1: solutions were added ultrapure HNO_3 to an acid concentration of 5% (V/V).
Step 2-3: solutions were added 2.5 mL ultrapure HNO_3 and diluted to 50 mL with Milli-Q water (18.1 Ω). Online IS was used. The samples were analyzed on Agilent 8900 QQQ ICP-MS.

2.9. XRD-analysis

Soil was ground with a mortar and pestle and fully packed into a "well-type sample holder" and then pressed flat with a glass slide and leveled with the sample holder surface (Wragg, 2018). The samples were analyzed by Senior Engineer David Wragg on Bruker D8 Discover (step 7) X-ray diffraction.
Bruker-diffrac.suite.EVA V.4.1.1 (database pdf2) was used for phase identification and TOPAS was applied for quantification of the identified minerals.

2.10. Instruments

Several instruments were utilized in this study. UltraCLAVE, ICP-MS, ICP-OES and XRD are presented below.

UltraCLAVE

Prior to elemental detection on ICP-MS the acid diluted samples were put in a UltraCLAVE (IV Milestone) for decomposition of the solid material into liquid form. Based on the principles of microwaves, UltraCLAVE allows digestion of samples requiring high temperatures (Milestone, 2008), resulting in a more efficient decomposition as the reaction time is reduced and samples are more completely digested (Milestone, w.y). Tubes (glass, Teflon or quarts) containing the samples are placed in a rack and manually put in a reaction chamber, starting the process. The chamber is filled with a liquid that absorbs microwaves and heats up the samples rapidly. It is put under high pressure by an inert gas which prevents boiling of samples and cross contamination. After digestion, the chamber cools down and reduces the pressure until ambient pressure is achieved. The described process (*Figure 6*) is performed automatically (Milestone, 2008).

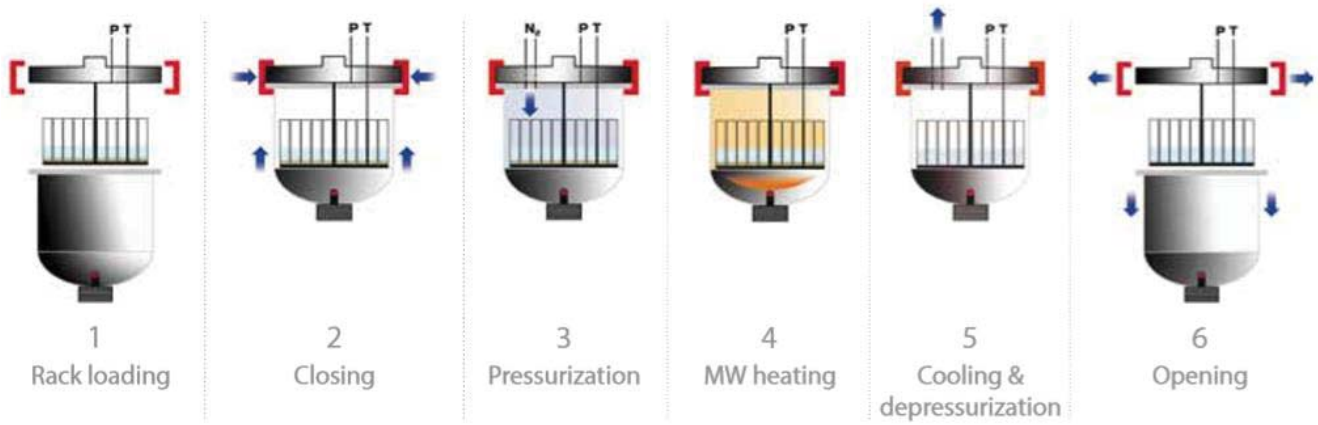


Figure 6. Process of UltraCLAVE digestion (Milestone, 2008).

ICP-MS

Inductively coupled plasma mass spectrometry (ICP-MS) is an analytical tool used for determination of elements and isotopes. It is highly accurate and provides low detection limits compared to other analytical instruments. The ICP source converts elements into ions followed by detection by the mass spectrometer. The samples are first converted into aerosols containing sample matrix and elements, then dried to a solid by an argon plasma (6000 °C)

and transformed into gas. The sample, in gaseous form, absorbs energy in the plasma, resulting in electron loss creating positively charged ions. Before entering the reaction cell, neutral atoms and photons are separated from the analyte ions to prevent instrumental drift and to avoid counting of photons by the detector which would increase background noise and level the detection limits. To remove any possible interferences (ions with identical mass-to-charged ratio) caused by the plasma or sample, the reaction cell contains an inert gas allowing the analyte to pass and a reactive gas that react with the non-analytes, resulting in a highly accurate measurement. The mass spectrometer filters the masses by ejecting the ions that do not have the given mass-to-charge ratio. The analyte ions hit a detector that releases an electron each time it gets struck. The electron signal is amplified until a measurable pulse is created. The pulses are used for calculation of amount ions in the sample.
The ICP-MS is highly effective with a scanning rate at over 5000 atomic mass units/second, allowing rapid analyzation of several atomic masses even though it scans one at time (PerkinElmer, 2011).

The model used in present study, Agilent 8900 Triple Quadrupole ICP-MS, is even more advanced in removal of interferences as it holds a unique tandem MS configuration placed at both sides of the reaction cell (Sakai, 2017).

ICP-OES

Inductively coupled plasma optical emission spectrometry (ICP-OES) determines the elements using their unique spectra created by excitation. ICP-OES consists of the same components as ICP-MS: plasma, spectrometer and a detector (*Figure 7*). As in ICP-MS, the sample is first converted into aerosols and then introduced to the plasma, usually argon gas (10000 °C), where the sample solvent is evaporated, and any remainders are vaporized to prevent chemical interreferences, and molecules are split into atoms. The atoms absorb energy, resulting in a state of excitation. The electrons releases photons when returning to their neutral energy level. These photons are either filtered or measured by the spectrometer depending on the analyte`s wavelengths. (ThermoFischer, w.y). The intensity of the photons is proportional with the elemental concentration. The instrument can quantify about 70 elements at the same time (Egeland, 2018).

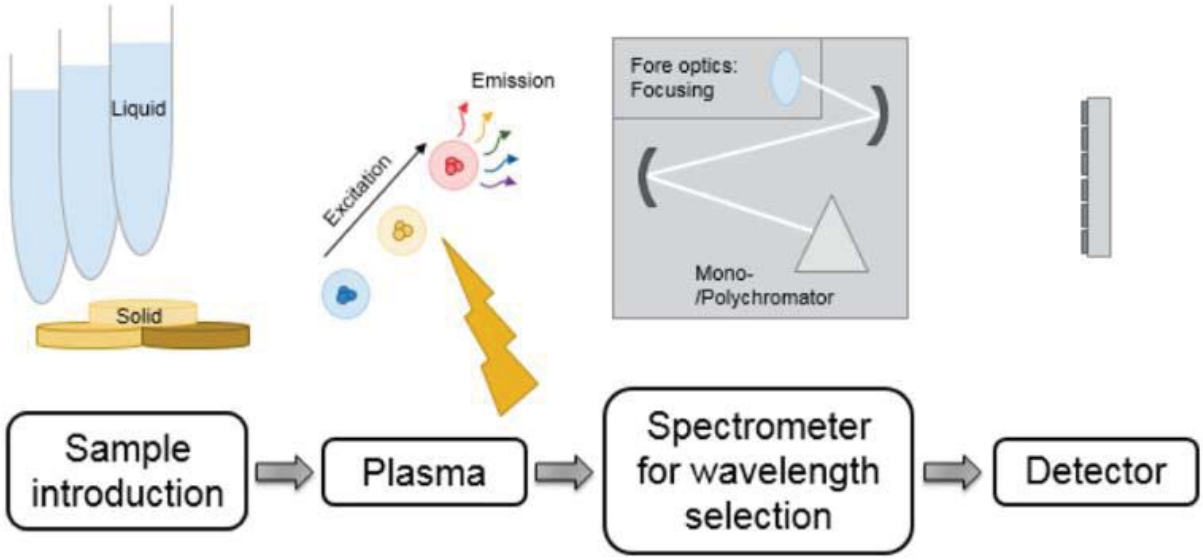


Figure 7. ICP-OES components and process (ThermoFischer, w.y).

XRD

X-ray Powder Diffraction (XRD) is a common method used for identification of phases in crystalline materials. In crystal lattices the atoms are ordered, giving a unique pattern when irradiated by X-rays. The pattern may give information about crystalline structures and quantity, and average grain size. Sample preparation is crucial to get a reliable result. Poor preparation complicates determination of the phases (Bunaciu et al., 2015).

The X-ray tube (*Figure 8*) contains a source which produces the X-rays used for analyzation. The source consists of a cathode emitting electrons and an anode which accelerates these electrons up to ten thousand of volt. As these electrons hit the anode, electrons in the inner shells of the atoms are removed and filled with higher level electrons. When dropping down to a lower energy level, radiation is emitted and passes through the primary optics before irradiating the sample. The primary optics, consisting of soller slits and divergence slit optimizes the output by reducing peak asymmetry and increasing the resolution. The secondary optics have further optimizing effect. It consists of an anti-scatter, soller and receiving slit which reduces both the noise due to air or amorphous scattering, reduces the axial divergence which can cause a lower peak intensity and removes scattered radiation improving instrumental resolution, respectively. Some secondary optics also consist of a monochromator with the purpose of removing disturbing radiation (fluorescent) originating from the sample (ammrf, 2012). The X-rays form a unique diffraction pattern as they scatter

depending on the planes of the atoms in the crystal lattice. These beams are then detected by a "position sensitive detector" which changes the orientation of the focus circle dependent on the beam angle. The powder diffraction is visualized in a diffractogram which is a plot of the intensities of the diffracted beams vs. their diffraction angels. Mineral content can then be determined by qualitative and quantitative analyses of the diffraction pattern (Wragg, w.y).

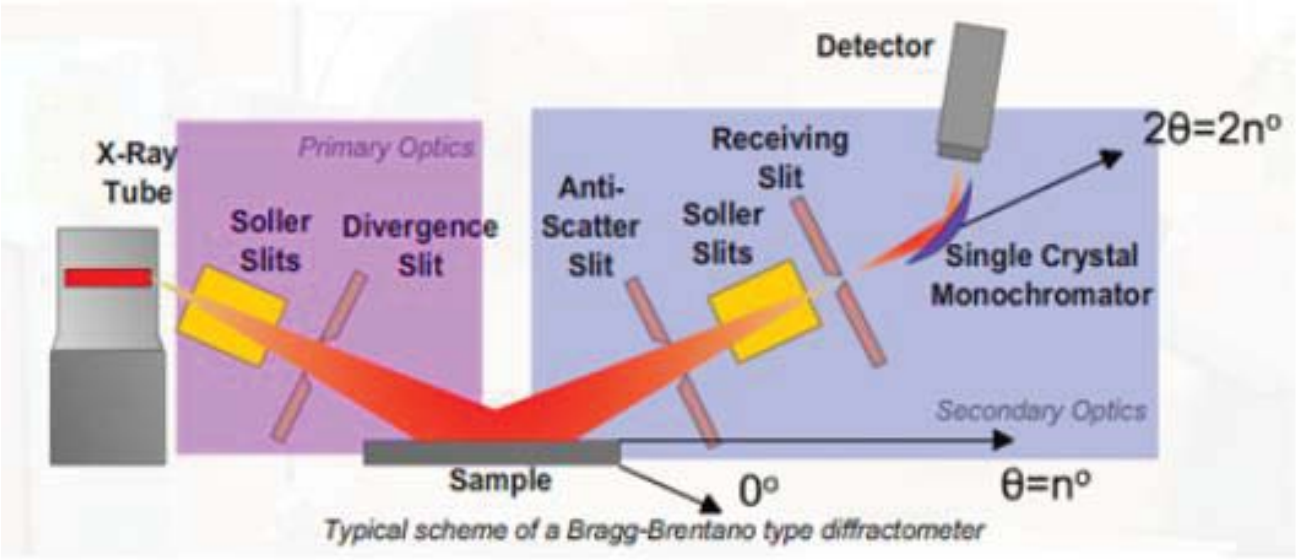


Figure 8. Main components in an XRD (ammrf, 2012)

2.11. Quality assurance and statistical approach

2.11.1. Quality assurance

Experimental work involves several uncertainties, uncertainties from sample preparation to instrumental analysis which is important to consider for assurance of reliable results. These experimental uncertainties can be classified as either random or systematic errors. The latter can be detected and corrected, whereas random errors always are present and cannot be corrected (Harris, 2010).

Moreover, this present study also involves uncertainties regarding concentration ratios which will be presented later.

The quality of the results was assured by considering internal standard, blanks, certified reference material and parallels.

Internal standard

Most of the samples (samples for ICP-MS analyses) were added internal standard (IS), except the samples which were basic diluted with TMAH and the sequential extraction samples. For those, IS was added online to the instrument. Internal standard can correct for loss of analyte during sample preparation and physical interferences, thus improve the analytical precision. A certain concentration of IS is added to all samples, included blanks and CRM. Any drift of the analytes will be detected by a drift in the signal of the IS and is adjusted accordingly (Norris, 2017). The internal standard should differ from the analyte and if not, the added concentration should be negligible. Considerations when deciding which IS to choose is similarities in analyte behavior, hence being close regarding atomic mass and ionization potential (Enger, 2014).

Limit of detection and limit of quantification

Limit of detection (LOD) (*Equation 6*) and limit of quantification (LOQ) (*Equation 7*) were calculated based on the standard deviation of the blanks which had been measured by ICP. The blank samples have been through similar process as the samples, consequently having same content of both chemicals and water.

Equation 6. $$LOD = SDblank * 3$$

Equation 7. $$LOQ = SDblank * 10$$

Standard deviations for Cs, Co, I, Sr, Th and U in soil and biological samples are presented in *Table 3*:

Table 3. Limit of detection (LOD) and limit of quantification (LOQ) for Cs, Co, I, Sr, Th and U in soil, grass, pellets, meat and milk. Th was not determined in milk.

	Soil		Grass, pellets and meat		Milk	
Element	**LOD**	**LOQ**	**LOD**	**LOQ**	**LOD**	**LOQ**
Cs (mg/kg)	0,0004	0,0013	0,0001	0,00033	0,0004	0,0015
Co (mg/kg)	0,004	0,014	0,0001	0,00045	0,00009	0,00029
I (mg/kg)	0,006	0,018	0,003	0,011	0,004	0,013
Sr (mg/kg)	0,03	0,1	0,01	0,032	0,001	0,005
Th (mg/kg)	0,003	0,01	0,00002	0,000082	n.a	n.a.
U (mg/kg)	0,0002	0,00067	0,0002	0,00053	0,00004	0,00012

Most of the elements gave satisfying values, except for uranium in cow`s milk which had 67 % of the samples below LOD and 33 % below LOQ. Uranium had also values (86 %) below LOD in mutton, along with Th which had 14 % of the samples below LOQ (*Table 23*, Appendix E). Finally, $^{137}$Cs had 33 % of total samples in cow`s milk below LOD (*Table 11*, Appendix A). How these data were managed is described in the next subsection.

CRM

Suitable certified reference material (CRM) was used to test the analytical accuracy (*Table 13- Table 15*, Appendix A). Each analyte is certified for a given range and an acceptable accuracy should give analytical results within that range (Harris, 2010).

The results show that most of the elements were within the certified reference areas. Cesium was close to the range for 1 of 2 soil CRMs, while Sr was outside the range for both soil CRMs and close to the range for both biological (meat, pellets, grass) CRMs and near for 1 of 2 milk CRMs. Strontium determination in soil depends on type of soil; Sr is sulphate bound, hence a stronger acid is needed (than HF and HNO_3) for an enhanced decomposition. This underestimation (at least 21 and 55 %) cannot be corrected for as amount bound to sulphate varies depending on the soil (Jensen, 2018). Cobalt was close the certified area for 1 of 4 biological CRMs and finally, I was close for 1 of 3 milk CRMs.

Precision

Precision is a measure of the variation in the results given by repetitive measurements of one sample. It is expressed as the standard deviation (SD) between the parallels, or the relative standard deviation (RSD).

In present study, three or five parallels were taken from one sample for precision determination, except for grain size and CEC analyses.
The relative standard deviation for soil analysis at ICP-MS after decomposition with HNO_3 (Th), HF (Co, Cs, Sr, U) and basic dissolution (I) ranged from respectively 0-52 %, 0-7.8 % and 0-4.4 % (*Table 6*, Appendix A). Cobalt, Sr, I and Th had parallels with identical concentrations, giving RSD of 0 %. The RSD range for Th represents SDs from five sites, with 4 of 5 of these sites having quite low RSDs (0-2.5 %) compared to site 5 (52 %).
RSD varied from 1.8-35 % for acid decomposed grass (Co, Cs, Sr, Th, U). RSD of 35 % represents variation in Th, with the remaining ranging from 1.8-3.9%. For basic diluted grass (I) the RSD was 2.3 %. The high variations of Th in soil and grass might be due to inhomogeneous samples.
Pellets, milk and meat (*Table 7-Table 8*, Appendix A) analyzed by ICP-MS after acid decomposition (Co, Cs, Sr, Th, U) had a RSD ranging from respectively 0.017-53 %, 1.4-17 %, 7.5-48 %. The high RSD (53 %) for pellets is due to variation in Th. Basic diluted (I) RSDs ranged from 22-52 % for pellets, 3.0-5.8 % for milk and was 8.3 % for meat. Homogenization of mutton samples was incomplete due to the high fat content, which may be an explanation for the large variation in meat for both acid (7.5-48 %) and basic treated (8.3 %) samples.

Quality assurances for soil properties

RSD for pH in soil was 0.68 % (n=5) and RSD for loss-on-ignition was 3.2 % (*Table 18*, Appendix B). For sequential extractions three parallels from three locations were selected giving a RSD range of 1.5-25 % (*Table 19*, Appendix B) in addition to a control. CRMs were measured for CEC analysis (*Table 17*, Appendix B) and for grain size, parallels of a control sample was measured, giving RSDs for clay, silt and sand of (8, 4 and 2) %, respectively (*Table 16* in Appendix B).

2.11.2. Data treatment and statistical approach

Microsoft excel (2016) was used for data treatment. Statistical analyses were done in the computer program Rstudio (Rstudio Team 2016). Values which were < limit of concentration and /or limit of quantification were calculated by substitution with ½*LOD or ½*LOQ. There are several other methods to deal with such values, but due to lack of time as well as advanced statistical knowledge, substitution was still used (Barescut et al., 2011; Newman, 1994). Concentration ratios were calculated even though > 50 % of total sample values were below LOD and LOQ. These CRs were only used in statistical analyses and are not listed in present study as associated uncertainties are too high (referring to U in cow`s milk and in mutton).

Multiple linear regression was used for examination of predictors for bioavailability of Cs, Co, I, Sr, Th and U in soil, uptake in grass and in animal. MannWhitney-wilcoxon rank sum test and Mood two-sample test were used for comparison of variation between animals in separate areas and for variation tests of uptake in plants in the five areas.

Calculation of concentration ratios (CRs) and transfer coefficients (F_m, F_f)

For calculation of elemental uptake in grass, *Equation 8* was used:

Equation 8.

$$Concentration\ ratio\ (CR) = \frac{concentration\ in\ grass\ \left(\frac{mg}{kg}\right)(dry\ weight)}{concentration\ in\ soil\ \left(\frac{mg}{kg}\right)(dry\ weight)}$$

CRs were computed for the 15 sites and for calculations of the arithmetic means (AM) and standard deviations (ASD), formulas in Microsoft Excel were applied. For geometric means (GM) and geometric standard deviations (GSD) *Equation 11* and *Equation 12* were used (Thørring et al., 2016a). The CR for ^{137}Cs were calculated likewise, though by the use of activity in grass/soil (Bq/kg) instead.

Several assumptions were made for calculation of concentration ratios and transfer coefficients; 1) the sampled grass is representative for animal consumption, 2) cows and goats

eat 80 % grass and 20 % pellets, representing dry matter (sheep were not given pellets) and 3) a daily intake of 12.5 kg dry matter for cows, 1.5 kg for goats, and 1 kg grass for sheep.

Equation 9 was used for derivation of concentration ratios for both milk and meat/dry matter in wet/dry weight:

Equation 9.

$$CR = \frac{concentration\ in\ animal\ \left(\frac{mg}{kg}\right)(wet\ weight)}{concentration\ in\ dry\ matter\ \left(\frac{mg}{kg}\right)(dry\ weight)}$$

-dry matter calculation: 0.8 * element (mg/kg) in grass + 0.2 * element (mg/kg) in pellets (dry matter SD was corrected for pellets SD (20 %)).

Concentrations derived from ICP-MS analysis reflect the concentration in dry weight for all matrixes. For calculation to wet weight, the ratio between ^{137}Cs (Bq/kg) in wet and dry weight was used (*Table 10, Table 11* in Appendix A). ^{137}Cs (Bq/kg) in milk and meat were computed likewise, but do just include grass as pellets were not subjected to radioactive Cs. Finally, in cases were animals had been grazing in several places, a mean of the dry matter associated to those sites was computed. This applies for calculation of transfer coefficients (TC) (*Equation 10*) as well. TC refers to either F_m (transfer to milk) or F_f (transfer to meat).

Equation 10.

$$TC = \frac{concentration\ in\ animal\ product\ \left(\frac{mg}{kg}\right)(wet\ weight)}{concentration\ in\ dry\ matter\ \left(\frac{mg}{kg}\right) * daily\ intake\ (d/kg)\ (dry\ weight)}$$

Geometric means (GM) (*Equation 11*) and geometric standard deviations (GSD) (*Equation 12*) were computed as followed:

Equation 11. $GM = AM/\sqrt{(\beta^2 + 1)}$

Equation 12. $GSD = exp^{*}\sqrt{(ln(\beta^2 + 1)}$

- *where* $\beta = ASD/AM$

Standard deviations for transfer coefficients and concentration ratios for goat`s milk (n=1) were computed as *a combined standard deviation* as followed in *Equation 13* :

Equation 13. $$ASD = AM_y/AM_x * \sqrt{(\beta_y^2 + \beta_x^2)}$$

Standard deviations for soil/grass (n=15), cow`s milk (n=5) and mutton (n=7) were computed in Excel and represents the variation between their concentration ratios/transfer coefficients.

Equation 13 was also used when computing individual/site specific CRs, F_m and F_f.

3. Results and discussion

The results obtained in present study will be referred to as the project name ROGFARM.
In the present work several elements were determined by ICP-MS in soil, grass, pellets, cow`s and goat`s milk and mutton. Concentrations were used for calculation of concentration ratios and transfer coefficients. The stabile analogues of Co, Cs, I and Sr, and the naturally occurring radionuclides Th and U have been prioritized and are presented and discussed in current chapter. For comparison purposes, values in the technical report series IAEA (2010) and transfer factors based on expert judgement applied in FDMT (Müller et al., 2003) have been added. ^{137}Cesium values from ROGFARM (values given by NRPA) are also presented. The IAEA values are based on worldwide data and are regarded as international reference values.
Concentration ratios for grass/soil are discussed first, followed by soil properties and sequential extractions indicating the potential bioavailability of the elements. Statistics were initiated with principle component analyses to detect any trends followed by multiple regression analyses. Several correlation analyses were performed in order to investigate possible correlations between total soil concentrations, bioavailable elements, irreversibly bound elements, grass concentrations, potential bioavailability and CRs. Variation analyses of concentration ratios between the areas were also done. The next subchapters discuss transfer coefficients and concentration ratios for uptake in milk and mutton, including correlation and variation analyses. Finally, stable Cs and ^{137}Cs were compared.

Concentration ratios and transfer coefficients for the other unprioritized elements (Ag, Al, Ba, Ca, Cd, Ce, Cl, Cu, Eu, Fe, K, La, Mg, Mn, Na, Nb, Pb, S, Sb, Se, Zn, Zr) are listed in Appendix E as well as figures comparing ROGFARM values with those compiled by IAEA and applied in FDMT.

3.1. Transfer from soil to grass

3.1.1. Concentration ratios

IAEAs report includes many transfer values, representing different soil groups and plant groups/compartments. For all values, the factors representing *all* soils groups were chosen, corresponding to soil groups found in ROGFARM (Appendix B). The soil incorporated in

values reported in IAEA (2010), FDMT and ROGFARM were sampled at a depth of 10 cm. Transfer factors by FDMT are reported in wet weight for grass and dry weight for soil and have been calculated to dry/dry weight by the use of IAEA`s standard dry-to-wet weight factor of 0.2 (IAEA, 2010).

ROGFARM concentration ratios for grass/soil (dry/dry weight) for the stable analogues Co, Cs, I, Sr and radioactive Th, U and ^{137}Cs had a range of 0.0060-0.072, 0.0036-1.6, 0.0075-0.13, 0.11-0.55 and 0.00059-0.054, 0.00055-0.024 and 0.0058-1.0, respectively (*Table 24*, Appendix E).These ranges give an insight of the great variation of uptake for an element, but also between the differences amongst them. U had the lowest CR at 0.00055 in contrast to the highest for stable Cs of 1.6. Stable Cs had also the greatest variation in range, described by a magnitude of 3, followed by a factor of 100 for Th, ^{137}Cs and U, and finally a factor of 10 for Co, I and Sr. The big difference for concentration ratios for stable Cs reflects the variation in uptake and the variables controlling CRs.

Concentration ratios calculated for ROGFARM (*Table 25,* Appendix E) and compiled in IAEA (2010), both in geometric mean, dry/dry weight, as well as FDMT values (dry/dry weight) are illustrated below (*Figure 9*).
For Co, Cs and Sr, ROGFARM values were comparable with both IAEA and FDMT; Sr had the highest uptake followed by Cs and Co. In all cases, IAEA soil-to-plant transfer were higher; Co, Cs and Sr with a factor of 2, 3 and 4 respectively, U was one and Th was less than 2 orders of magnitude higher.
FDMT had higher ratios for both iodine (factor of 3) and strontium (factor of 2). But as strontium was underestimated in both CRMs (chapter 2.11.1), the differences between FDMT

(and IAEA) might be even bigger. Soil-to-plant transfer of Co and Cs were higher with a factor of 3 and 2 respectively, for ROGFARM than those for FDMT.
As illustrated (*Figure 9*) are IAEA and ROGFARM values of ^{137}Cs more similar than that for stable Cs, whereas the opposite for FDMT. IAEA and FDMT concentration ratios differ with a factor of less than 2 and 3, respectively, with that for ROGFARM.

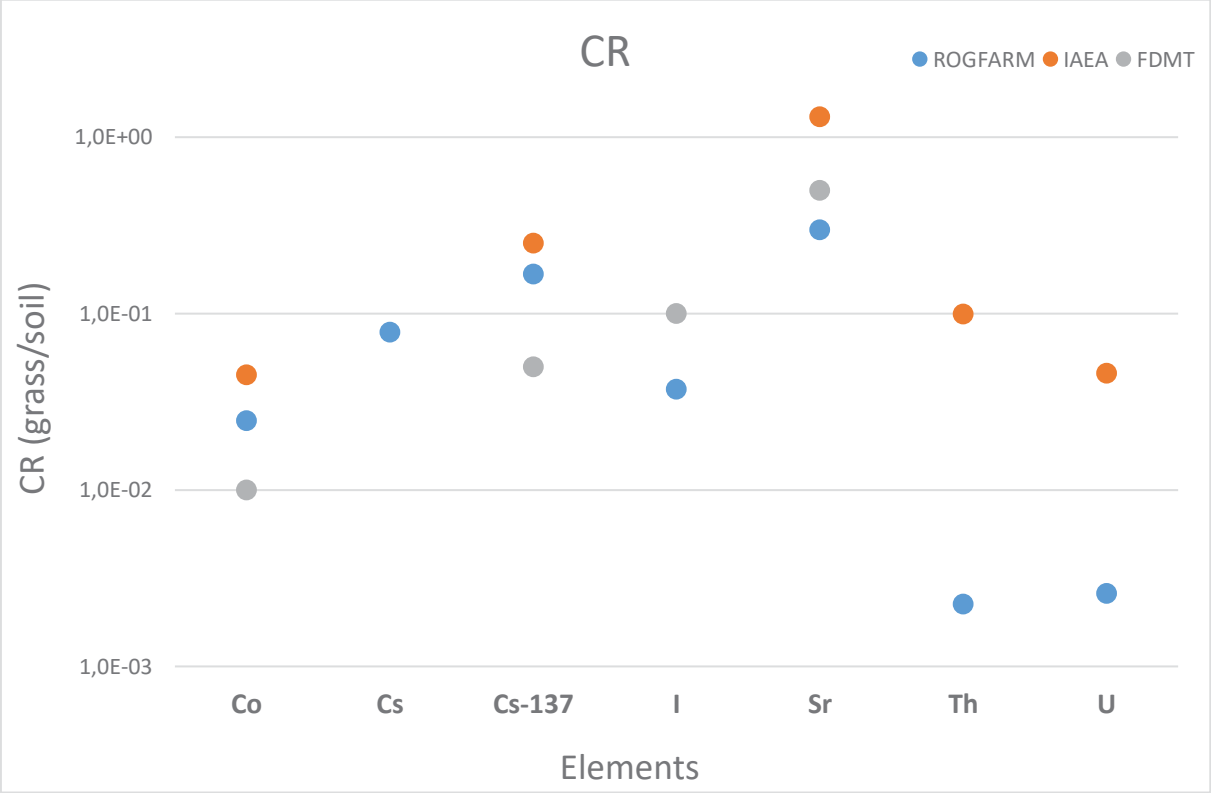


Figure 9. Concentration factors (grass/soil, dry/dry weight) for Co, Cs, ^{137}Cs, I, Sr, Th and U. Factors are given in geometric mean for ROGFARM and IAEA, and FDMT factors are default values. Stable Cs is compared with IAEA and FDMT values of ^{137}Cs. Geometric mean and geometric standard deviations for ROGFARM are in Table 29, Appendix E. Note the logarithmic y-axis.

The geometric means with geometric standard deviations for ROGFARM and IAEA (FDMT values don't have GSD) were compared (*Figure 10*). As shown, IAEA CRs varied more than for those of ROGFARM and all except for Sr overlapped. An explanation may be that 84 % of the Sr values compiled in IAEA represent soils with a maximum organic matter (OM) content of 6.5 %, which is a relatively low OM content compared to the loss-on-ignition results for ROGFARM (presented below). The availability of Sr increases with a decreasing OM soil content (Harbitz & Skuterud, 1999). The differences described above compare the

geometric means and by taking geometric standard deviations into consideration it is clear that the differences may increase due to the great variation for the IAEA values.

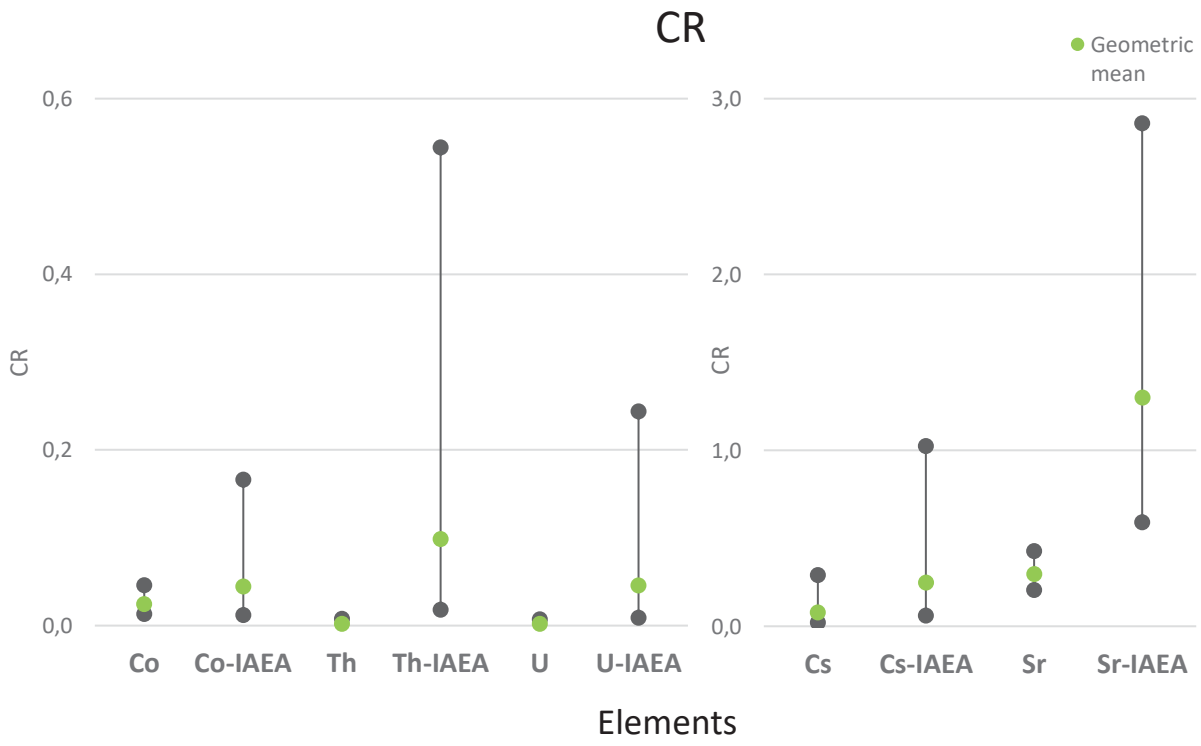


Figure 10. Illustrates the CRs (geometric means with geometric standard deviations) for ROGFARM and IAEA in Figure 9. Note the different scales.

3.1.2. Soil parameters

Organic matter, clay and cation exchange analysis

Sequential extractions and XRD-analysis were done on soil only from following locations: A1-2, B1-2, C1-2, D1 and E1 due to limitation purposes. Grain size analysis was not done for A1, A2, C2 and D1 because of the loss-on-ignition results indicating a high content of organic matter. The method used for grain size analysis applies to an organic material content of max. 20% (Krogstad & Børresen, 2015).

Organic matter content (*Figure 11*) varied from 4.5-62 % (Appendix B, *Table 18*). A1, A2, C2 and D1 had a relatively higher amount of organic matter than the other locations. Compared with the area description (humus rich soils in B2-B3, D2 and peat soil in D1) in *Table 1,* the organic matter content agreed with the peat soil description of D1. Such soils have a high content of organic matter with an organic material content > 30% (dry weight) (Joosten et al., 2015). Ashing of inorganic carbon and evaporation of water in clay may overestimate the OM content. Soils with a low amount of organic matter are more prone for significant errors (Howard & Howard, 1990), thus the results in *Figure 11* have been corrected for water-holding clay based on the clay content in grain size analysis (Krogstad, 1992). See *Table 18* in Appendix B for correction factors.

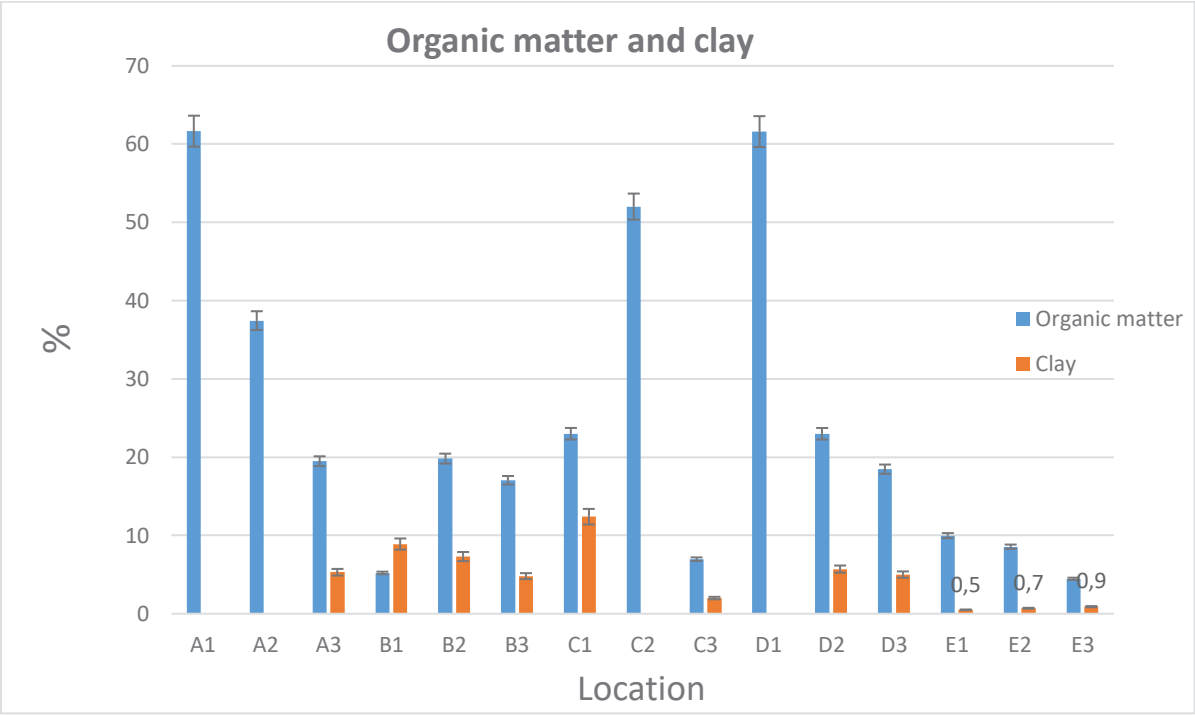


Figure 11. Percentage distribution of organic matter and mineral fractions of clay across all 15 locations, except for A1-2, C2 and D1 for clay due to the high content of OM.

Clay content in mineral fraction varied from 0.5-12 %, with the lowest value in E1 and highest in C1, which was described as a clay rich soil (*Table 1*).
Area E showed a relatively low clay content, but the overall amount clay did not seem to vary significantly. Derived from the grain size analysis, soil in area B was classified as silt loam, area E as sand and locations D2-3 as sandy loam. Site A3, C1 and C3 was categorized respectively as sandy loam, loam and loamy sand (*Table 16*, Appendix B). Finally, sites A1-2, C2 and D1 with their respective high amounts of organic matter may indicate organic soils, but as organic soils refer to an OM content > 40 % in the upper layer of at least 40 cm (Almås, 2018), no further classification will be done.

The cation exchange capacity (CEC) (*Figure 12*) ranged from 14-67 ($cmol^+/kg$) (*Table 17*, Appendix B). Area A along with C1-2 and D1 have high values relative to area B and E, and location C3. pH in soil had a range from 4,8 in D1 to 6,4 in A2 (*Table 18*, Appendix B).

Figure 12. Shows the cation exchange capacity (cmol$^+$/kg) at all locations. CEC ranges from 14-67 cmol+/kg with the lowest in B1 and highest in C2.

Sequential extractions

The soil in present study was put in a freezer after sampling. Sequential extractions should be done on fresh soil as the soil properties may be altered, but an argument for carrying this out anyway was the fact that the soils in this study were naturally subjected to freezing and thawing several times a year, thus the prior was neglected.

As mentioned, sequential extractions were only done at some of the sites, namely the sites were animals had been grazing. The results (*Figure 13*) showed that Th and Cs seemed to be more fixed to soil compared to the other elements, whereas Sr was the most mobile element. Location A1 had the highest bioavailable fractions for all elements, except for Sr. For both Cs and Sr, NH_4Ac (pH~soil) yielded the highest extraction, whereas for Co, Th and U the potential bioavailable fractions were mostly associated with the third extraction step, which is usually interpreted as carbonates.

The fact that cesium is strongly adsorbed to soil (Cornell, 1993; Fuller et al., 2015) is clear in ROGFARM as well, with a potential bioavailable fraction range of 0.22-3.1 % and an arithmetic mean of 0,79 %. Cs was least mobile in site B1 and most in A1, in fact, elemental concentrations in grass (*Table 6*, Appendix A) showed that location A1 (2.6 mg/kg) had a

higher Cs concentration than the other locations. Since clay is the most important factor controlling mobility of cesium it is natural to compare those results, and more significant, the type of clay (Cornell, 1993; Fuller et al., 2015). XRD-analysis suggested the presence of one clay mineral across all sites, the mineral illite located in sites B1-2 (*Table 22*, Appendix D). This correlates with the low mobility of Cs in location B1, but differ from site B2, where Cs is relatively mobile. Possible explanations will be discussed under statistical analyses.
Illite in analyzed soils was quantified to 45 % and 36 % in respectively B1 and B2.

Cobalt had a higher potential bioavailability than Cs with a range of 0.93-13.6 % and an arithmetic mean at 6.0 %. The biggest reversible fractions were fractionated in location D1 followed by A1. Co potential bioavailability was relatively low in site B1, C1 and E1.
Co mobility is controlled by adsorption to different soil constituents (Kim et al., 2006), especially to Fe- and Mn-oxides (Krupka & Serne, 2002), resulting in non-bioavailable fractions (Tessier et al., 1979). In fact, sites with the lowest fractions of reversibly bound Co had the highest Fe concentrations (Mn (mg/kg) was highest in B1) (*Table 33*, Appendix) which may indicate associations to Fe/Mn-OH. Furthermore, the results have an overall agreement with OM/CEC across all sites.

Strontium had an arithmetic mean of 12 % and a range from 3.0-24 % for the potential bioavailability. Sr was more mobile and bioavailable compared to the other elements. The general trend for its availability (an increase as organic matter decreases (Harbitz & Skuterud, 1999)), was not obvious by comparisons with OM (*Figure 11*), in present study. Sites C2 and D1 had the largest quantities of reversible fractions which is in contrast with the high amount of organic matter, however, results from site A1, which also had a high OM content, suggested that Sr was less mobile.

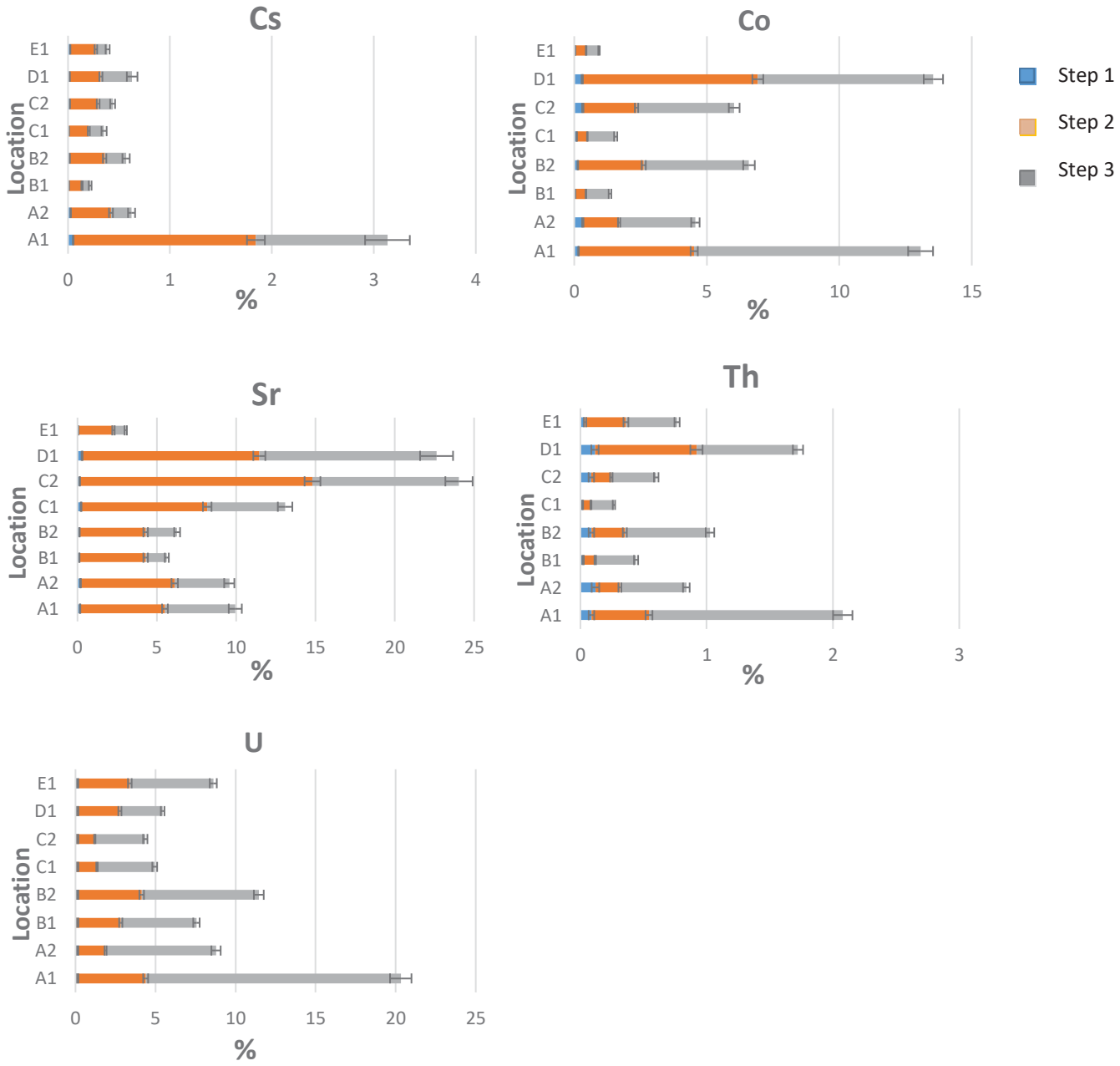


Figure 13. Sequential extractions with standard deviations for Cs, Co, Sr, Th and U in locations A1-2, B1-2, C1-2, D1 and E1. The fractions represent the potential bioavailability in the respective locations. Note the different x-axes.

The potential bioavailability of thorium ranged from 0.44-2.1 %, with an arithmetic mean of 0.97 %. These values reflect that Th is strongly bound to soil (Popic et al., 2014). Location A1 and D1 had higher percentages of potential bioavailable thorium which corresponds to the higher amount of OM. Uranium had a potential bioavailable range of 4.4-20.3 % and an arithmetic mean of 8.9% and was therefore more mobile than Th, as reported in previous

studies (Baeza & Guillén, 2006; Popic et al., 2014). Uranium had a significant higher potential bioavailability in site A1 compared to the other locations.

The general trend showed a higher bioavailability, except of Sr, in site A1, followed by locations D1 and B2 and that all elements were rather more irreversibly associated to soil than reversibly.
Finally, it must be mentioned that step 2 was skipped for D1 due to a soil pH (pH 4.8) lower than extractant pH (pH 5) applied in step 3. It is likely to assume that the fraction in step 3 is representative for step 2, thus step 3 is used as a proxy for the potential bioavailability in step 2. This fraction involves therefore additional uncertainties and might under- or overestimate the fractions associated to NH_4Ac (pH~soil) and might explain the inconsistency with literature for Sr potential bioavailability in site D1.

3.1.3. Statistical analyses

To get an overview of all parameters and to visualize any possible trends, principal component analysis was applied three times, for sequential extractions and soil parameters, for all sites (meaning all parameters excluded sequential extractions as the latter was only done for some sites) and finally for XRD-analysis. In addition to the discussed parameters are some macro- and microelements added as well (*Table 34-Table 35*, Appendix).

Sequential extractions and soil parameters

Principal component analysis (*Figure 14*) of soil parameters (pH, OM, cation exchange capacity, sequential extractions and grain size) and total soil concentration showed that PCA axis 1 and 2 described 58 % of the total variation, respectively 31 % and 27 %. Reversibly bound Cs (Cs1-Cs3) was concentrated in location A1 and B2, moving the opposite direction of clay (mineral fraction). Location A1 had also higher concentrations of reversibly bound cobalt (Co1-Co3), uranium (U1-U3) and thorium (Th1-Th3), with the latter also high in D1, as seen in Figure 13. Sequential extractions showed high potential mobility and bioavailability of Co in D1 as well, in contrast to the PCA-biplot. Both locations in area B and C seemed quite similar, in contrast to A1 and A2.

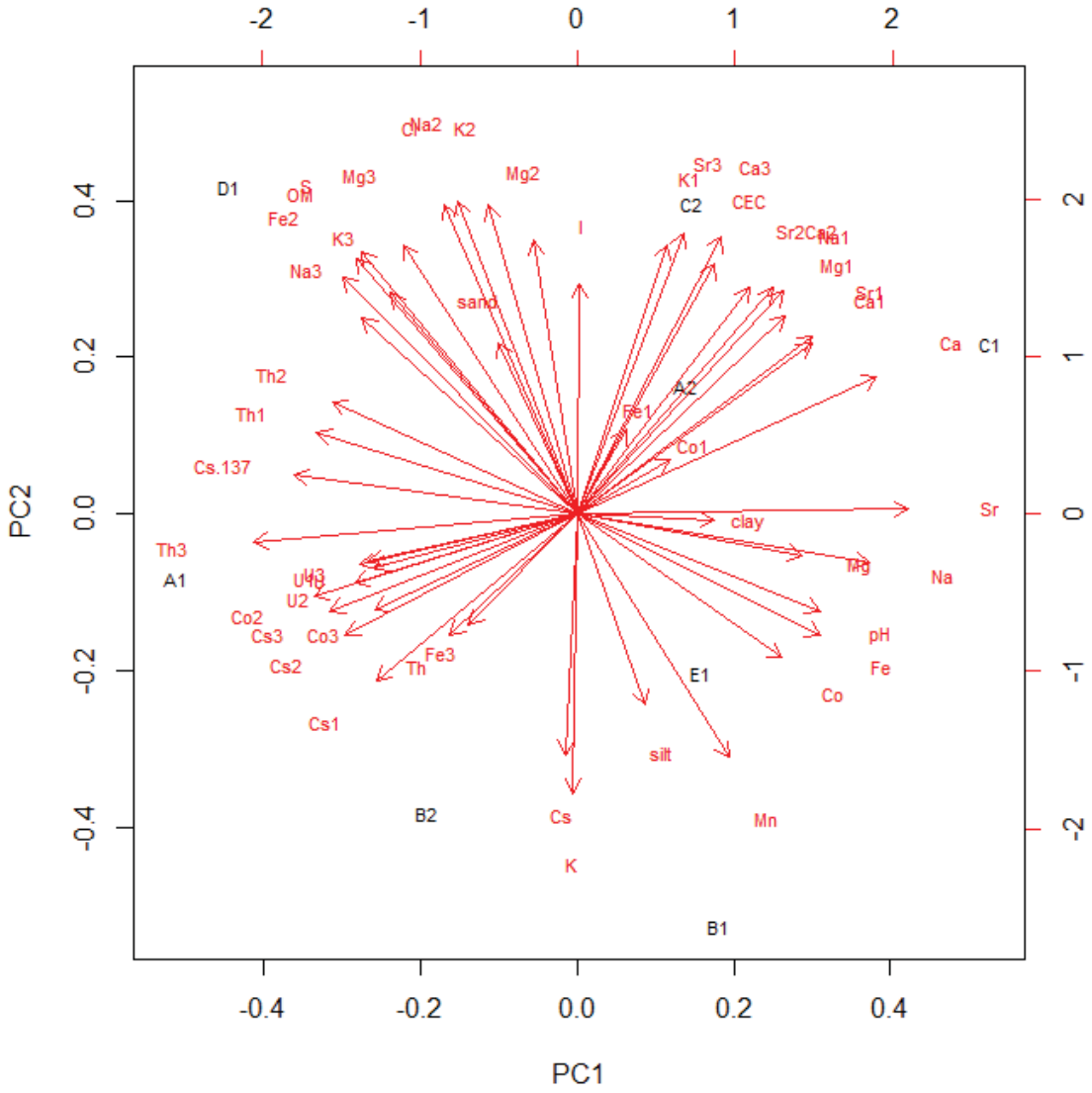


Figure 14. PCA bi-plot of elemental concentration in soil, sequential extractions, pH, OM, CEC and grain size (clay, silt and sand). Note that the PCA just includes data for the 8 locations sequential extractions were done for.

A positive significant correlation ($p<0.01$) was also seen between the potential bioavailability of Co and organic matter ($r^2=0.76$), confirming the pattern similarities between sequential extractions results and organic matter, and between the potentially bioavailable Sr and OM/pH ($r^2=0.92$). A positive correlation suggests an increase of strontium availability as OM/pH increases, which is contradicting to the general trend of Sr potential bioavailability (Harbitz & Skuterud, 1999). But regression analysis did though identify a negative significant relation ($p<0.05$) between Sr content in grass and organic matter ($r^2=0.60$), giving weight to the fact that an increase in OM decreases its availability. Van Bergeijk et al. (1992) found

similar results though for soil-to-plant uptake and OM/pH and Nisbet and Woodman (2000) did also identify correlation between uptake in crops and OM/pH (r^2=0.30).

Further multiple regression analyses were also done for grass concentrations and soil parameters as well as for concentration ratios and soil parameters. A positive significant relation ($p<0.05$) was identified between the Cs concentration in grass and its potential bioavailability, describing 64% of the variability. For concentration ratios, an even stronger significant positive correlation ($p<0.001$), accounting for 92% of the variability, was found between Cs CR (grass/soil) and the potential bioavailability of Cs, indicating that the potential availability of Cs in soil indeed is an important factor for its soil-to-plant transfer. There was also seen a significant correlation ($p<0.01$) between soil-to-plant transfer of stable Cs and organic matter (r^2=0.45) and between ($p<0.05$) ^{137}Cs CR (grass/soil) and OM (r^2=0.28).
Cs adsorption is primarily controlled by clay minerals (chapter 1.2.2.) and the role of soil organic matter seems to be unclear, at least concerning radiocesium; Rigol et al. (2002) reported that in soils with a high organic matter content (>95 %) is adsorption to non-specific sites predominant, whereas others have documented none effects of OM (Lofts et al., 2002), though in soils with an organic matter content of 85 %, while a another study did demonstrate an increase of uptake in soils with OM content < 5 % up to > 50 % (Van Bergeijk et al., 1992). Either way, did Thørring et al. (2016a) also find a positive correlation between stable Cs and soil organic matter (*Figure 15*). For ^{137}Cs no correlation was seen, but the uptake is quite similar in soils with a high OM content.

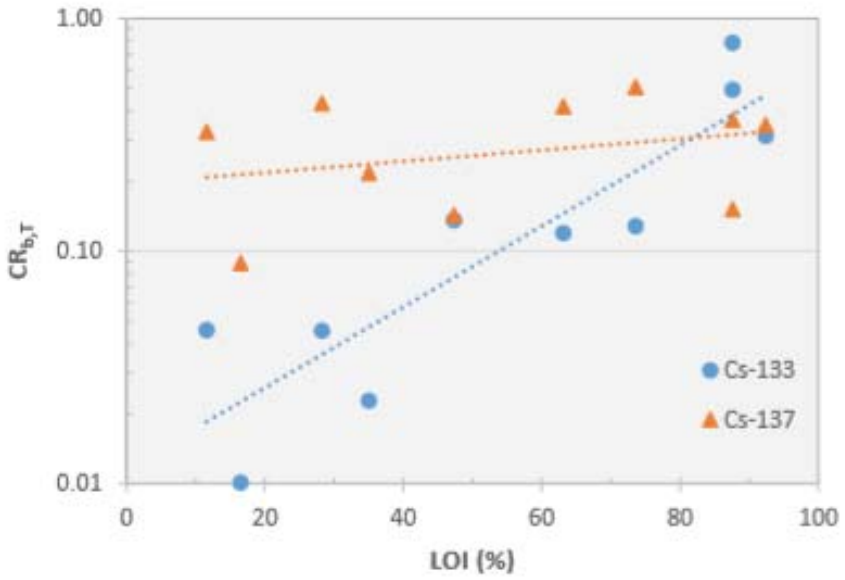


Figure 15. Stable and radioactive Cs as a function of LOI (%) in Tjøtta, Norway (Thørring et al., 2016a).

Results from ROGFARM show parallels in uptake in soils with OM content around 60 % (*Figure 16*). The similarities in uptake in soils with a higher organic matter content, in both figures, indicate that reversible sorption is more significant than fixation by clay in such soils, as reported in Thørring et al. (2016a).

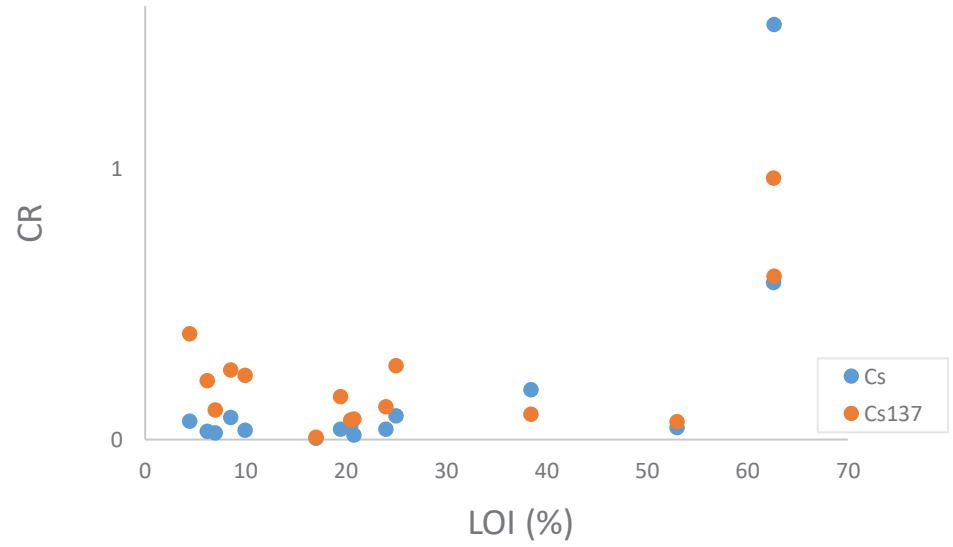


Figure 16. CR of stable and radioactive Cs as a function of LOI (%) based on data from sites A1-E3 in the present work.

Finally, for ^{137}Cs, there was identified a negative significant correlation ($p<0.05$) between its concentration in grass and pH ($r^2=0.70$).
No further correlations between concentration ratios, potential bioavailability, concentration in grass and soil parameters were identified.

Variance analysis

To see if the CRs (grass/soil) varied more between the areas than within an area, variation analyses of Co, Cs, ^{137}Cs, I, Sr, Th and U were carried out by using nonparametric tests in Rstudio. MannWhitney-Wilcoxon/Mood two-sample test in Rstudio were applied depending on if the variances were equal or not. F-test done in word Excel showed that Sr had unequal variances, thus Mood-two sample test was utilized, while the others were run in MannWhitney-Wilcoxon test. There was not enough evidence to conclude that CR for any of the elements above varied more between area A-E than within ($p>0.05$). This may give weight to the statement mentioned above regarding that countries are divided into 1-5 regions in accordance to similarities (agricultural practices, etc) and that further subdivisions would be unnecessary as variations from year to year are expected to be more pronounced (Müller et al., 2003). However, it should be emphasized that sample size was quite low and that this could affect the result.

PCA of soil parameters in all locations

Bi-plot of principal component analysis of total soil concentrations, pH, organic matter, cation exchange capacity (CEC) and mineral fractions; silt, sand and clay for all 15 locations are illustrated below (*Figure 17*). PCA axis 1 and 2 described respectively, 31% and 26 % of the total variation. Clay and Cs were positively correlated, along with Sr and Ca, and CEC and organic matter. CEC was positively correlated with organic matter ($p<0.001$), as expected, with 89 % of the variation in OM describing the variation in CEC. The bi-plot showed that area C varied more regarding total soil concentrations and soil parameters compared to other areas.

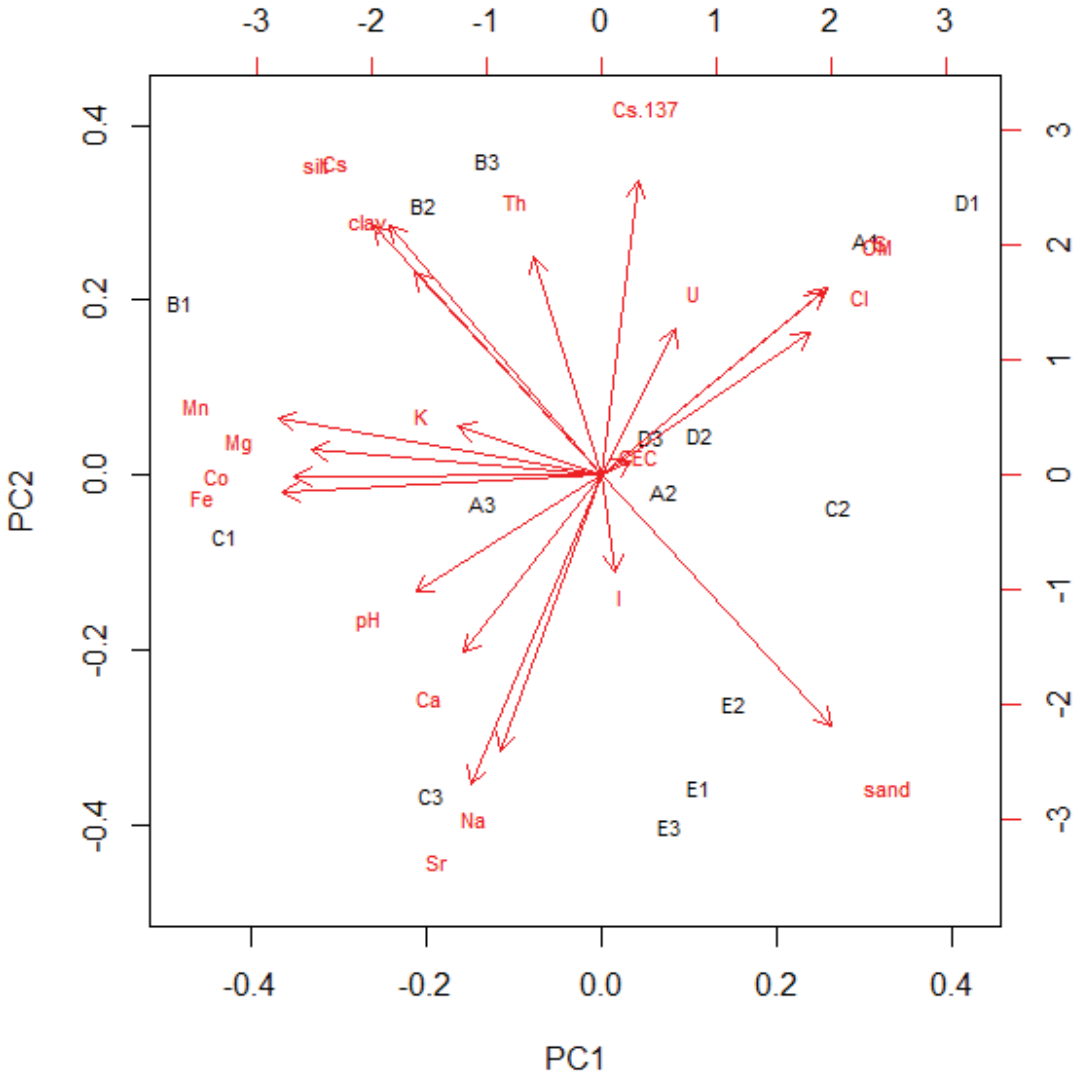


Figure 17. PCA bi-plot of elemental concentration in soil, pH, OM, CEC and grain size. All 15 locations are included.

PCA of XRD-results

Principal component analyses were performed on data for illite, determined by XRD-analysis, mineral fraction of clay, total Cs and Cs-137 and potential bioavailable Cs (Cs1-3) (*Figure 18*). The PCA axis 1 and 2 described 82% of the total variation, respectively 55% and 27%. Clay, illite and total Cs were positively correlated and were predominant in sites B1-2. Potential bioavailable Cs (C1-3) and total ^{137}Cs were more concentrated in site A1. Illite was found in sites B1-2 and was the only clay mineral detected by XRD-analysis (Appendix D, *Figure 22*) as described above, (3.1.2 Sequential extractions) and as stated is the presence of illite in agreement with the high amount of Cs that is irreversibly bound in site B1, in contrast to site B2. Sample homogeneity is very important in analyses and the presence of a particle

could give elevated, hence not representative results, which might be an explanation for the contrasting results. Anyways did multiple regression analyses not reveal any correlations between the reversibly bound fractions of Cs and soil parameters, suggesting that other factors are affecting the bioavailability. In fact, is the radiocesium interception potential (RIP) expected to be analyzed and a comparison with results obtained in present work may contribute to useful information.

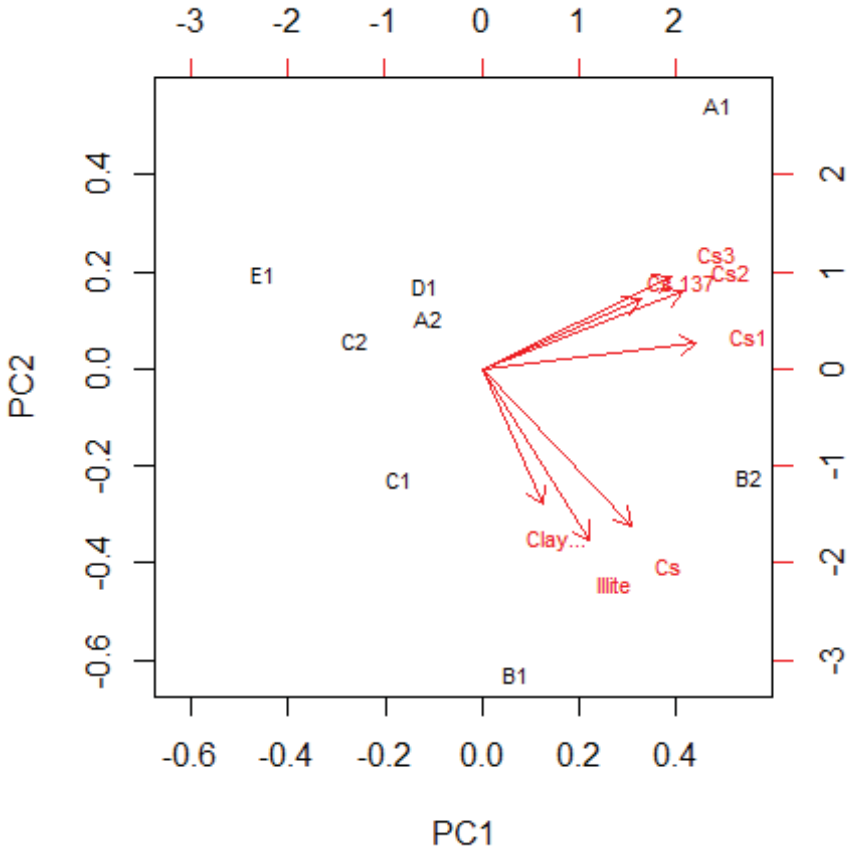


Figure 18. PCA bi-plot of illite, derived from XRD-analysis, mineral fraction of clay, total Cs and Cs-137 and potential bioavailable Cs (Cs1-3) in location A1-2, B1-2, C1-2, D1 and E1.

Based on XRD, about 5 % zeolite was identified in soil at location C1 (*Figure 25*, Appwndix D). Zeolite is a mineral that readily adsorbs radionuclides and metals, particularly monovalent ions such as Cs^+ (Misaelides, 2011). The mineral has a high CEC (Ming & Dixon, 1987), probably contributing to the relatively high CEC observed in location C1 (*Figure 12*).

3.2. Transfer from feed to animal product

3.2.1. Transfer coefficients (F_m) - transfer to goat`s milk

Milk samples (n=1) were collected from goats grazing in locations A1-3 (*Table 2*). The standard deviations for transfer coefficients and concentration ratios represent the variation between milk parallels (n=5) and the variation between dry matter for A1-3. For ^{137}Cs, the GSD expresses the analytical uncertainty for milk (n=1) and variation between grass samples A1-3. Geometric mean is used for all transfer coefficients, except for FDMT factors which are default values. Concentration ratios are in arithmetic mean.

ROGFARM transfer coefficients of Co, Cs, ^{137}Cs, I, Sr and U had ranges of respectively, (0.00026-0.00080), (<LOD-0.25), (<LOD-0.65), (0.12-0.25), (0.0084-0.0096) and (0.00034-0.00037) d/L, representing the variation of elemental uptake in goat`s milk (*Table 26*, Appendix E). Cobalt, I, Sr and U had narrower ranges than Cs and ^{137}Cs, reflecting a greater variation for the latter two. The ranges for both Cs and ^{137}Cs can be explained by their respective variation in dry matter (dry matter and grass), which both had relatively high RSDs (~130 %) (*Table 6*, *Table 9*, Appendix A) in contrast to milk RSDs of < 5 %. As mentioned above had grass in A1 a significant higher concentration compared to the other sites. This applies for ^{137}Cs as well. The low RSDs are not surprising as goat`s milk (n=1) for Cs and ^{137}Cs demonstrate respectively, parallel variation and analytical uncertainty.

ROGFARM, IAEA and FDMT transfer coefficients (wet/dry weight) for uptake in goat`s milk (*Figure 19*) showed that all IAEA F_m, except for ^{137}Cs, were higher than for those of ROGFARM. Similar observations were seen for FDMT, except for Cs and ^{137}Cs.
The biggest difference of uptake was for Co, with a factor of 10, between IAEA/ROGFARM. IAEA transfer of Cs, Sr and U were higher with a factor of respectively 2,3 and 4. FDMT uptake of Sr, I and Co were higher 2, 3 and 4 times higher respectively. IAEA F_m for I was only a bit higher and can be considered as equivalent to that for ROGFARM, as well as for ROGFARM/FDMT of Cs. Uptake of ^{137}Cs in Rogaland was quantified as higher than both IAEA and FDMT with a factor of less than 2 and 3, respectively.
A trend in uptake is illustrated below, where I had the highest uptake in goat`s milk, followed by Cs, Sr and Co. U had the lowest transfer coefficient for both ROGFARM and IAEA.

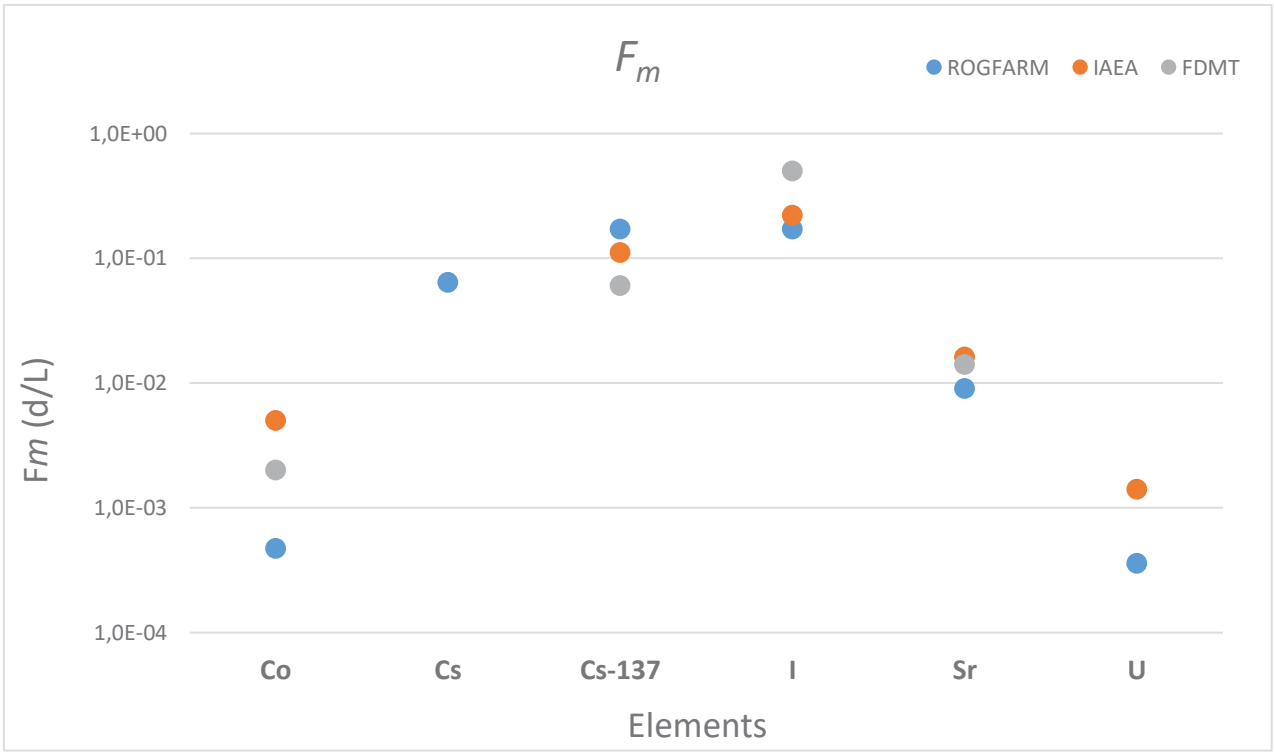


Figure 19. Transfer coefficients (F_m) (wet/dry weight) of uptake in goat`s milk of Co, Cs, ^{137}Cs, I, Sr and U for ROGFARM, IAEA and FDMT. Stable Cs is compared with IAEA and FDMT values of ^{137}Cs. F_m for ROGFARM and IAEA are given as geometric mean, FDMT have been derived by expert judgement. Note the logarithmic y-axis.

As transfer coefficients consider the daily consume, the assumed intake is important to compare. For ROGFARM the daily intake was estimated to 1.5 kg dry matter (80 % grass and 20 % pellets), and for IAEA and FDMT the daily intake was set to respectively, 1.5 and 2.6 kg. The latter do not consider pellets which might underestimate nutrient intake, thus overestimating F_m and with an adjustment of FDMT daily intake to 1.5 kg the coefficients would be higher. Variation of dry matter intake is not expected to influence transfer coefficients more than a factor of 2-3 according to IAEA (2010).

IAEA and ROGFARM concentration ratios (goat`s milk/dry matter) (*Table 27*, Appendix E) with standard deviations for ^{137}Cs, I and Sr (*Figure 20*) showed that IAEA had higher CRs for iodine and Sr. Strontium and I for ROGFARM were within the ranges of IAEA, whereas radiocesium of IAEA was within the range of uptake in Rogaland. CRs of ^{137}Cs and I were differing with a factor of 2 and Sr with a factor of 3. ROGFARM had a broader range for ^{137}Cs, whereas ranges for I and Sr were more extensive for IAEA CRs.

IAEA Cs, I and Sr ratios were computed from n= (12, 3 and 5), respectively, whereas in the present work concentration ratios have been derived from one sample. Individuals might have a selectivity regarding food intake and the ranges for IAEA CRs would illustrate the individual differences between the animals in a greater extent in contrast to ROGFARM, thus a lower variation in Rogaland values can be expected. Another aspect is that IAEA values reflect a collection of CRs from different countries, thus taking a lot of uncertainties and variation into account.

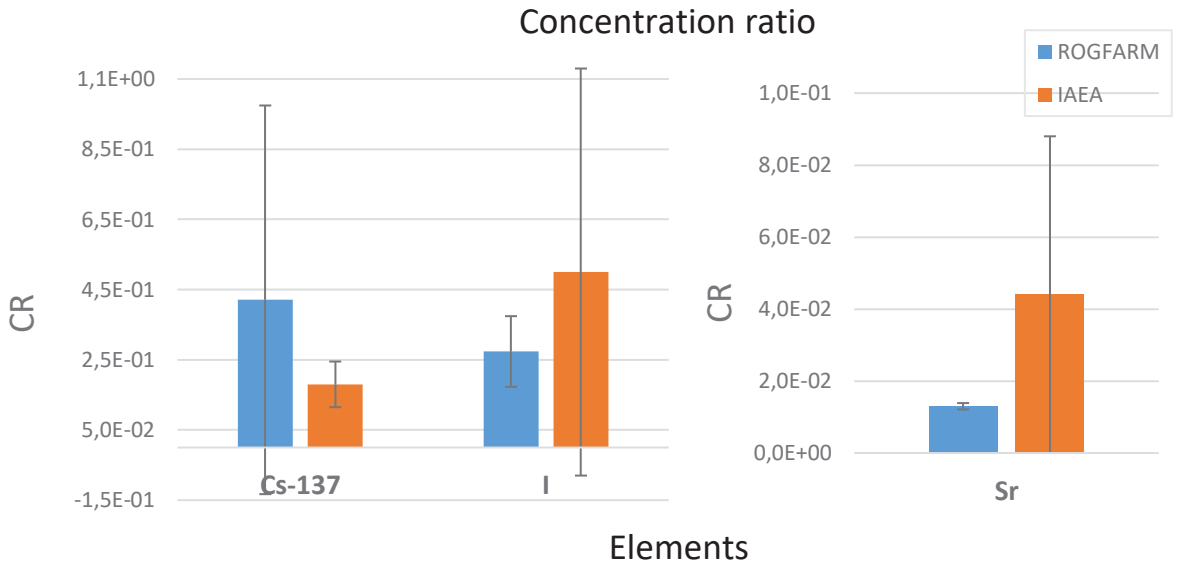


Figure 20. Illustrates the concentration ratios (wet/dry weight) for ^{137}Cs, I and Sr in goat`s milk (arithmetic mean with standard deviation) for ROGFARM and IAEA. In the case of IAEA, the respective elements were the only ones where associated standard deviations were provided. Note the different y-axes.

3.2.2. Transfer coefficients (F_m) – transfer to cow`s milk

Transfer coefficients *(F_m)* of Co, Cs, ^{137}Cs, I and Sr for cow`s milk in Rogaland had respective ranges of ((0.000058-0.00016), (0.00086-0.015), (<LOD-0.011), (0.0022-0.017) and (0.00050-0.00088)) d/L (*Table 28,Table 29* in Appendix E). Like goat`s milk, transfer coefficients for Cs and ^{137}Cs had broader ranges for cow`s milk as well. In contrast to goat`s milk the ranges for cow`s milk reflect the variation between F_m in 3 areas (A, B and E). For both Cs and ^{137}Cs the F_m were in the increasing order of C<E<B (*Table 6, Table 7, Table 9* and *Table 11* in Appendix A). Radiocesium had values (33 %) below LOD, which may contribute to a higher variation.

All values for uranium was below LOD/LOQ, thus transfer coefficients and concentration ratios were only used in statistical analyses and will not be discussed any further here due to unacceptable uncertainties.

Cow`s milk (F_m) for ROGFARM, IAEA and FDMT are illustrated below (*Figure 21*). IAEA had higher transfer coefficient for Sr (2 times) and Cs (less than 2 times), whereas Co and I uptake were similar. FDMT F_m were higher for Co and Sr, described by a factor of 2 and 3 respectively, whereas ROGFARM had almost 2 times higher values for Cs and I.
Transfer coefficients for ^{137}Cs were equivalent in both cases. Others have also documented similar results for radiocesium (Karunakara et al., 2013; Tsukada et al., 2003).
As goat`s milk, the overall uptake shows a pattern; I and Cs had quite similar and also the highest $F_m$, followed by Sr and Co. All $F_m$ for goat`s milk were higher than for those of cow`s milk, with the former also having relatively broad ranges for both isotopes of Cs which can possibly be explained by the great variation of Cs content in area A. Transfer coefficients for Co, Cs, I and Sr were 10 times higher in goat and ^{137}Cs was higher with a magnitude of 2.

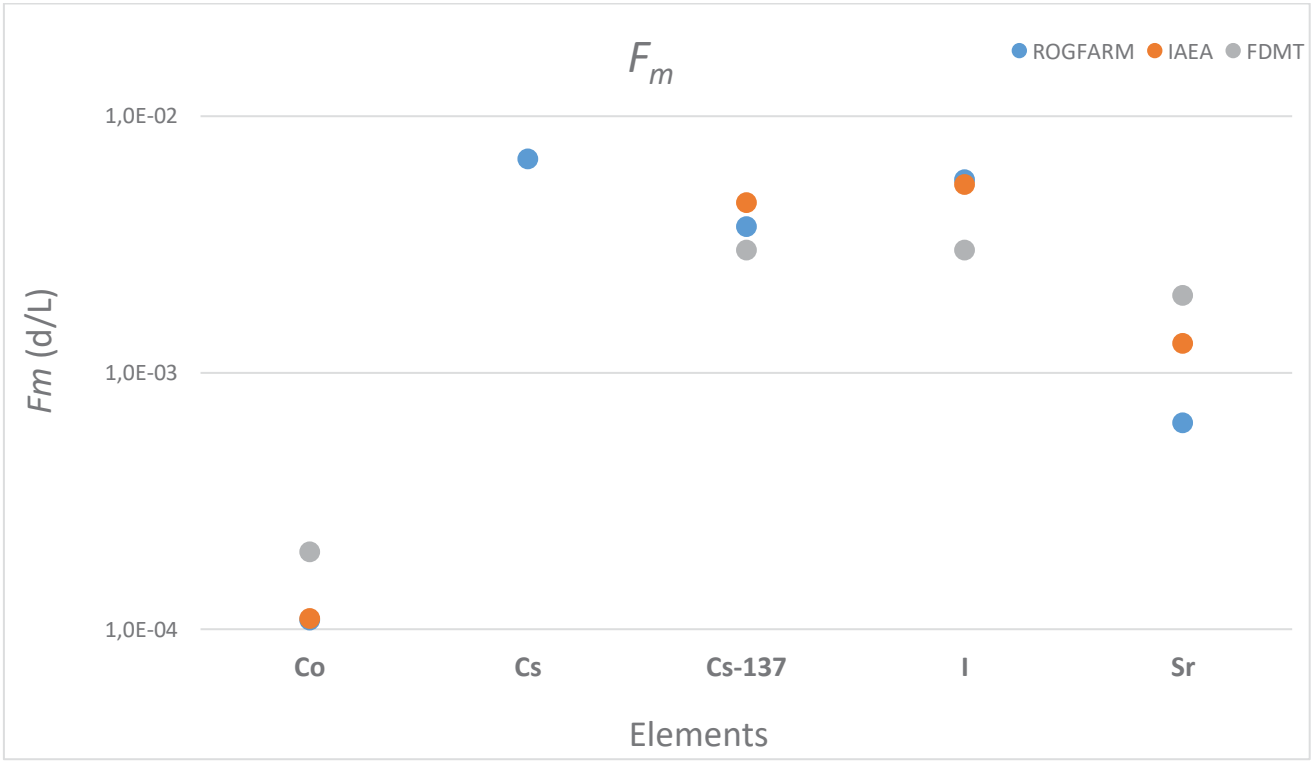


Figure 21. Transfer coefficients (F_m) (wet/dry weight) of uptake in cow`s milk of Co, Cs, ^{137}Cs, I and Sr for ROGFARM, IAEA and FDMT in geometric mean, except for ARGOS which have been derived by expert judgement. Stable Cs is compared with IAEA and FDMT values of ^{137}Cs. Note the logarithmic y-axis.

The assumed daily intake for cows were set to 16, 14 and 12.5 kg by respectively, IAEA, FDMT and ROGFARM, with the latter correcting for pellets (20 %) as well. The higher assumed intake by IAEA and FDMT and non-inclusion of pellets affect their respective transfer coefficients regarding both under- and overestimations in relative to ROGFARM values.

ROGFARM concentration ratios (*Figure 22*) (*Table 30*, Appendix E) were compared with IAEA values where standard deviations were available. IAEA had higher values for all three; ^{137}Cs was higher with a factor of 2, and I and Sr, with a factor of 3. All IAEA CRs had wider ranges, with the biggest difference for Sr. Both Cs and Sr (ROGFARM) were within the range of IAEA and I had a bit lower minimum value for ROGAFARM. Cs, Sr and I concentration ratios derived by IAEA were computed from respectively, n= (119, 44 and 43), whereas ROGFARM CRs derived from 3 samples. IAEA values have been gathered from an extensive database covering huge differences in climate and soil properties in contrast to ROGFARM, which represent local data. In this manner a greater variation for IAEA data is not surprising. Another aspect is that ROGFARM values represent fewer samples, which might affect the variations, in both directions, of the obtained CR values.

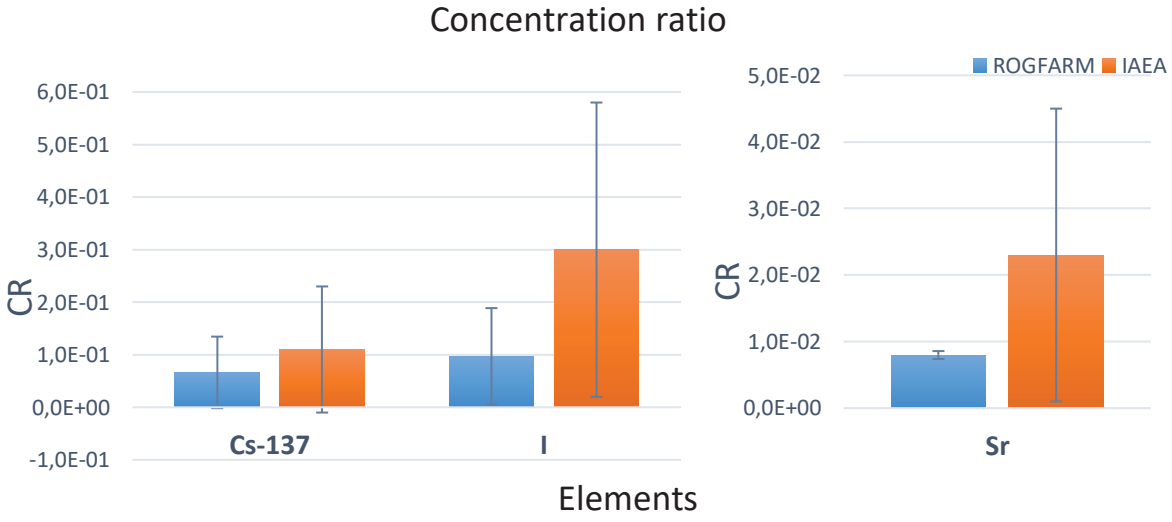


Figure 22. Shows concentration ratios for cow`s milk in arithmetic mean and standard deviation (wet/dry weight) for ^{137}Cs, I and Sr for ROGFARM and IAEA. In the case of IAEA, the respective elements were the only ones where associated standard deviations were provided. Note the different y-axes.

It has been suggested that concentration ratios have a lower variability than transfer coefficients across species (Beresford et al., 2007). IAEA CRs for Cs, I and Sr were 2 times higher in goat`s milk (*Figure 20*) relative to cow`s milk and maximum 3 times higher in goat`s milk in Rogaland, whereas the differences between transfer coefficients were ranging from a factor of 10-40, thus a higher variation for F_m across species is seen here as well.

For both Cs and U, a significant relationship ($p<0.01$) was identified between milk concentration (multiple regression analyses done at n=4, including both goat`s and cow`s milk) and their concentration in dry matter, with dry matter variability describing the variation in Cs and U milk with 99 % and 100 %, respectively. As uranium in cow`s milk had values > 50 % below LOD it must be emphasized that the identified correlation involves unknown uncertainties.
A positive significant correlation ($p<0.05$) was also found between ^{137}Cs in milk and grass ($r^2=0.98$).

3.2.3. Transfer coefficients (F_f) - transfer to mutton

ROGFARM ranges of transfer coefficients (wet/dry weight, d/kg) for Co, Cs, ^{137}Cs, I, Sr and Th were ((0.0057-0.076), (0.058-0.77), (0.15-0.37), (0.0071-0.029), (0.00089-0.020) and (<LOD-0.094)) d/kg, respectively (*Table 31, Table 32* in Appendix E). Cs and ^{137}Cs had higher F_f and in contrast to goat`s and cow`s milk, the isotopes had narrower ranges for mutton F_f, reflecting a more balanced uptake in sheep. The relatively high F_f of Cs in mutton is expected as Cs accumulates in muscles (Harbitz & Skuterud, 1999). Thorium had a quite broad range which may be explained by values (14 %) below LOQ (*Table 23*, Appendix E).

Transfer coefficients (wet/dry weight) for mutton in ROGFARM and derived by IAEA and FDMT are illustrated below (*Figure 23*). ROGFARM values were almost two times higher for Co, Cs and Sr than those for IAEA and three times higher for ^{137}Cs, whereas reported value of iodine by IAEA was higher with a factor of 2. FDMT and ROGFARM had equivalent values for transfer coefficients of Cs, I and Sr as well as for ^{137}Cs.
The biggest difference between uptake in mutton was for Co which was 10 times higher in Rogaland than the default value in FDMT.

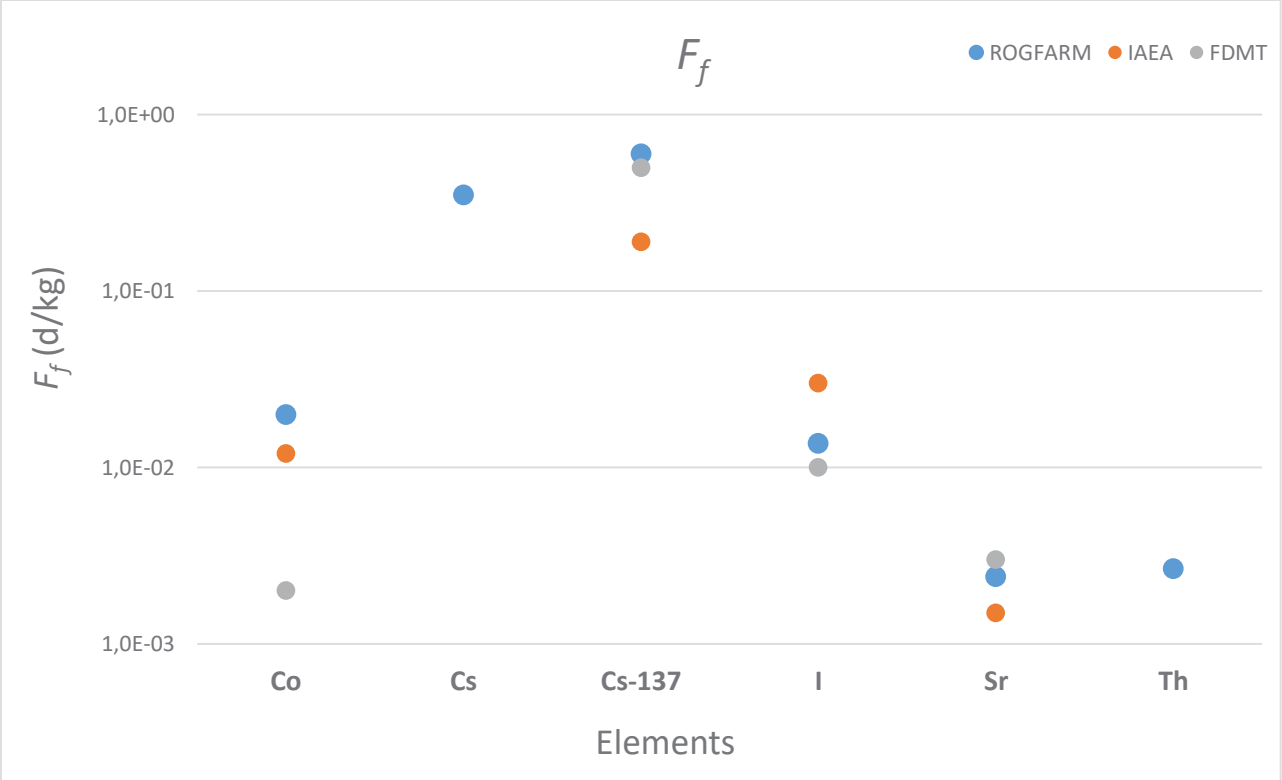


Figure 23. Transfer coefficients (F_f) (wet/dry) of Co, Cs, ^{137}Cs, I, Sr and Th for mutton. Stable Cs is compared with IAEA and FDMT values of ^{137}Cs. ROGFARM and IAEA values are in geometric mean, while FDMT are default values. Note the logarithmic scale for y-axis.

The assumed daily intake was 1 kg for both ROGFARM and FDMT and 1.5 kg for IAEA. In contrast to F_m (*Figure 19, Figure 21*) are the F_f more similar in terms of amount consumed along with that pellets are not included in neither. Despite the variability of intake between the reported transfer coefficients, the differences between ROGFARM, IAEA and FDMT *Fm* for cow`s milk were only greater for Sr than for those of mutton. In fact, the differences between Co and I were less and similar for Cs in mutton which might reflect the fact that the variation of transfer coefficients is expected not to vary more than a factor of 2-3 because of an unbalanced consume (IAEA, 2010).

Concentration ratios (*Figure 24*) for ^{137}Cs (*Table 32*, Appendix E) derived by IAEA (n=51) were higher (less than a factor of 3) and the variation was larger than for the ones obtained for Rogaland (n=7). However, the ROGFARM CR values fall within the range of the IAEA values. In contrast to ROGFARM, IAEA had a large sample size in addition to a greater variability reflecting the huge differences of the uptake in mutton.

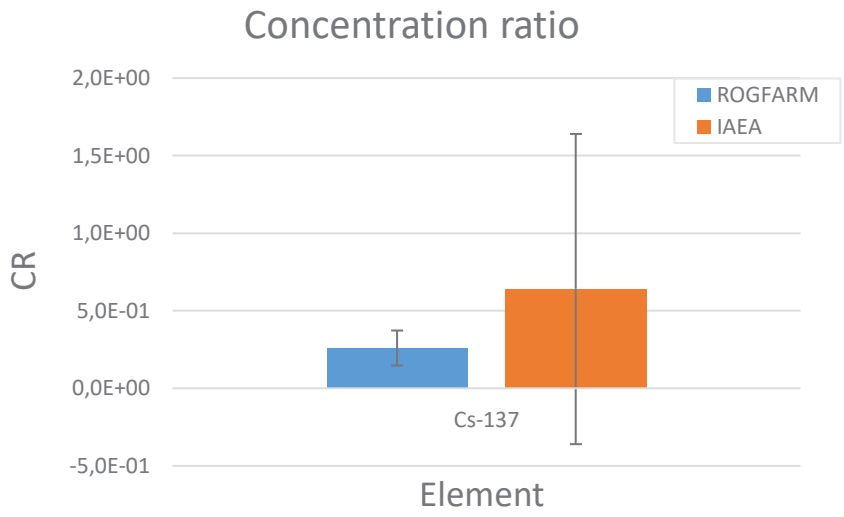


Figure 24. Concentration ratio (arithmetic mean and ASD) in wet/dry weight in mutton for ^{137}Cs in ROGFARM and by IAEA. In the case of IAEA, Cs was the only element for which associated standard deviation was provided.

Variance analyses of elemental concentration in mutton

To test whether the concentration of Cs, ^{137}Cs, Co, Th, I and Sr in mutton were different in area B and C or not, data were tested in Rstudio using either MannWhitney-Wilcoxon rank sum test (assumes equal variance) or Mood two-sample test (assumes unequal variance), depending on the variance were equal or not. Data were first tested by F-test in Excel, to check for equal variance.

For Cs, Cs-137, Co, Th, I and Sr, the difference between the median weights were not significant (p-value > 0,05), meaning that the concentration medians of mentioned elements are equal in location B and C. Thus, there are not enough evidence to state that the variation within an area is greater than between the two areas. An emphasize on sample size should be mentioned as area B, with n=2, is very small and questions the reliability of the results.

3.3. Comparisons of Cs and ^{137}Cs

A significant correlation ($p<0.05$) was identified between the soil-to-plant uptake of Cs and ^{137}Cs ($r^2=0.44$). Concentration ratio (geometric mean) of ^{137}Cs was a factor of 2 higher than that of stable Cs and the variability was greater for the latter (*Table 25,* Appendix E). Equivalent difference in GMs with a greater variability for stable Cs have been reported by Thørring et al. (2016a) as well. Others have also documented a higher uptake, varying from 3

– 11 times, for radioactive Cs (Tsukada et al., 2002; Tsukada et al., 2003; Uchida & Tagami, 2007) in grass and crops. All reported correlations between Cs/^{137}Cs, suggesting that Cs can be used as a predictor for soil-to-plant uptake of ^{137}Cs. However, the elevated uptake of radioactive Cs may reflect a disequilibrium for the isotopes; stable Cs originates from soil and would therefore be more fixed to soil compounds as mineral fixation is a slow process (Comans et al., 1991). As documented above (*Figure 15* and *Figure 16*), was the uptake quite similar in organic soils were clay fixation is negligible, hence giving emphasis to this statement.

The transfer coefficient (GM) to goat`s milk (*Table 26*) was higher for $^{137}$Cs (less than 3 times) than that for Cs and the variability was equivalent. However, in cow`s milk (GM) (*Table 29*), was Cs less than 2 times higher than that for ^{137}Cs and the variability was greater for the latter. Regression analyses including both goat`s and cow`s milk identified a positive significant correlation ($p<0.01$) between stable Cs and ^{137}Cs in milk ($r^2=0.99$ %), with identical results for the isotopes transfer coefficients. Similar results of Cs/^{137}Cs F_m have been reported previous (Karunakara et al., 2013; Tsukada et al., 2003).
A link between Cs concentrations in soils up to milk have been identified; there was found a significant relation between Cs concentration in grass and potential bioavailable Cs, and a correlation between milk/dry matter. The same regression analysis was done for just grass, giving identical outcome, which might indicate that Cs introduced via pellets do not contribute significantly to accumulation in milk.

Comparisons of F_f (GM*) (Table 32)* in mutton showed that ^{137}Cs was less than 2 times higher than that for Cs and the standard deviations were equal. No correlations were identified between the transfer coefficients of Cs and ^{137}Cs in mutton, nor between their accumulation is meat.

3.4. Summary of uncertainties and uncertainties in general

Several factors contribute to overall uncertainties. An overview is presented below (*Table 4*) with the use of ^{137}Cs uptake in milk and mutton (*Figure 20*, *Figure 22*, *Figure 24*) and Sr in milk as examples.

The analytical RSD (%) for ^{137}Cs uptake in goat`s milk in ROGFARM varied from 3-8 % and the variation within sites, thus overall RSD was 132 %. As the grazing sites were A1-3, the "within and overall RSDs (%)" reflect the same variation. Analytical variation for cow`s milk ranged from 5-26 % and the variation within sites from 26 to 100 %, with the highest RSD almost equal to the overall RSD (%) of 102 %. Mutton had a range of 5-15 % for analytical variation. The RSDs (%) within sites and overall were, 3-48 % and 43 %, respectively. In comparison with IAEA, is overall variation for cow`s milk a bit higher for IAEA, while goat`s milk and mutton vary significantly. The biggest difference between the concentrations ratios was for mutton (2.5), then goat`s milk (2.3) and finally cow`s milk (1.7). The use of IAEA concentration ratios could possibly underestimate uptake in goat`s milk and overestimate uptake in both cow`s milk and mutton, by the respective factors.

The analytical RSD for Sr (ROGFARM) varied from 0.41-5.0 % in goat`s milk and the replicate RSD had a range of 1.8-2.7 %. The variation within sites, which also represents the overall variation, was 6.9 %. IAEA had an overall RSD of 100 % and the CR was higher with a factor of 3.4. For cow`s milk did ROGFARM analytical RSD vary from 0.38-3.5 %, while the variation between the replicates was 1.4-1.8 %. The variation within and overall sites were, respectively 3-28 % and 7.5 %. IAEA had an overall RSD of 96 % and had a CR higher with a factor of 2.8. Thus, by using IAEA values for goat`s and cow`s milk the uptake could possibly be overestimated by the respective factors. In addition, did IAEA values in both cow`s and goat`s milk vary significantly more, contributing to even more uncertainties.
The differences between the variations for strontium are relatively greater compared to ^{137}Cs between ROGFARM and IAEA, but there are some similarities. Both analytical and replicate RSDs were in all cases relatively small compared to the other uncertainties. Another similarity is that within sites RSDs had ranges above the overall RSDs, thus contributing significantly to the overall variation.

Table 4. Overview of all relative standard deviations (RSD) contributing to uncertainties of ^{137}Cs and Sr concentration ratio (arithmetic mean). Analytical RSD (%) represents the minimum and maximum value for instrumental variation. Replicate RSD (%) shows the minimum and maximum value for the variation between the replicates taken (replicates were only taken for Sr). Within sites RSD (%) are the min.-max. value of variation within the respective sites. Overall RSD (%) represents the variation between all sites for ROGFARM and for IAEA it describes the variation of all input data. Ratio (CR/CR) is the ratio between the CRs, which was CR-IAEA/CR-ROGFARM for all, except for ^{137}Cs in goat`s milk. Uptake in soil was not included since IAEA values were given as geometric mean, thus GSD. See Table 36,Table 37, Table 38 in Appendix for calculations of cow`s milk and mutton (^{137}Cs) and Table 27 for goat`s milk.

	Element	Matrix	CR	Replicate RSD (%)	Analytical RSD (%)	Within sites RSD (%)	Overall RSD (%)	Ratio (CR/CR)
ROGFARM	^{137}Cs	Milk[1]/dry matter	0.42	-	3-8	132	132*	2.3
IAEA	^{137}Cs	Milk[1]/ dry matter	0.18	-	-	-	36	-
ROGFARM	^{137}Cs	Milk[2]/ dry matter	0.066	-	5-26	26-100	102	-
IAEA	^{137}Cs	Milk[2]/ dry matter	0.11	-	-	-	109	1.7
ROGFARM	^{137}Cs	Mutton/ dry matter	0.26	-	5-15	3-48	43	-
IAEA	^{137}Cs	Mutton/ dry matter	0.64	-	-	-	156	2.5
ROGFARM	Sr	Milk[1]/ dry matter	0.013	1.8-2.7	0.41-5.0	6.9	6.9*	
IAEA	Sr	Milk[1]/ dry matter	0.044		-	-	100	3.4
ROGFARM	Sr	Milk[2]/ dry matter	0.0080	1.4-1.8	0.38-3.5	3-28	7.5	
IAEA	Sr	Milk[2]/ dry matter	0.023		-	-	96	2.8

[1]goat`s milk, [2]cow`s milk, [*]the goat`s had been grazing in area A, thus Overall RSD (%) and Within sites RSD (%) are representing the same.

Uncertainties regarding the *assumed* intake concerning both amount for calculations of transfer coefficients and the representativity of grass/pellets are also an important factor. Another aspect is the individual selectivity for plant species (Cingolani et al., 2005; Rook et al., 2004), hence the accumulation of elements in animals might correlate to plant species rather than bulked samples. But in fact, a plant`s ability to release colloidal elements varies depending on genotype, thus there are variations within a specie as well (Prasad, 2013) and will therefore always be a faction that cannot be noted. In addition, there have been reported circumstances where the preference for certain plants have been copied to offspring, hence a family line might have a different concentration ratio than another (Thørring, 2018). Whether or not these uncertainties are comprised within the calculated standard deviations are

depending on the representativity of the samples and the probability for this increases with an increasing sample size.

To emphasize differences between soil properties, are typical soil values for factors compiled in IAEA (2010) presented along with those for Rogaland (*Table 5*):

Table 5. Shows the typical ranges of soil parameters for the soil groups comparable with those for ROGFARM. Blue = ROGFARM values, orange = IAEA (2010) values. pH, organic matter content (%), cation exchange capacity (cmol$_+$/kg), sand content in the mineral matter fraction (%) and clay content in the mineral matter fraction (%). ROGFARM soil groups have been classified as followed in the IAEA report: sand (≥65, <18) % content in the mineral matter fraction, Clay (≥35) % content in the mineral matter fraction, organic matter content (≥20) % and loam for the rest.

Soil group	pH	pH	OM (%)	OM (%)	CEC (cmol$_+$/kg)	CEC (cmol$_+$/kg)	Sand (%)	Sand (%)	Clay (%)	Clay (%)
Sand	5.6-6.1	3.5-6.5	4.5-19.5	0.5-3.0	15-35	3.0-15.0	≥69.1	≥65	<5.3	<18
Loam	5.4-5.9	4.0-6-0	5.2-19.8	2.0-6.5	14-30	5.0-25.0	14-55.7	65-82	4.8-8.9	18-35
Organic	4.8-6.4	3.0-5.0	≥23	≥20	30-67	20.0-200.0	-	-	-	-
Clay	-	5.0-8.0	-	3.5-10.0	-	20.0-70.0	-		-	≥35

pH for all soil groups were more acidic for IAEA, sand and loam soils had a greater organic matter content in Rogaland, CEC- values were varying greatly and clay content was lower for ROGFARM. The overall differences in the soil parameters might give a better understanding of the differences between the transfer factors.

4. Conclusions

Comparisons of concentration ratios for soil-to-plant transfer (GM) and transfer coefficients (F_m, F_f) (GM) of Cs,^{137}Cs, Co, I, Sr, Th and U show that the majority of the values obtained in present study were differing from the factors compiled in IAEA (2010) and the default values in FDMT (Müller et al., 2003) with at least a factor of 2. As FDMT values are used in dose estimations, will the uncertainties by using the default values be reflected in calculations of doses to population. In addition, the comparisons of overall variations in CRs (AM) between IAEA and ROGFARM illustrated a trend: a relatively greater variation for IAEA values which emphasizes how such factors may vary in terms of uncertainties.
Thus, it is likely to conclude that by using locally obtained data for radionuclides and stable analogues concentration ratios, the uncertainties associated with impact and risk assessment modelling will decrease with at least a factor of 2. Moreover, will uncertainties regarding calculations of possible doses to population decrease similarly.

There was identified correlations between stable Cs and ^{137}Cs in transfer to grass and to animal (milk), indicating that stable Cs can be used as a proxy for prediction of ^{137}Cs mobility and bioavailability.
Variation analyses of CRs (grass/soil) for Cs, ^{137}Cs, Co, I, Sr, Th and U and elemental concentrations (Cs, ^{137}Cs, Co, I and Sr) in mutton for were performed, and no significant differences were identified. Thus, there are not enough evidence to state that the variation within an area is greater than between the respective areas. Though it should be mentioned that sample sizes were relatively small and questions the reliability of the analyses.
Positive significant correlations were identified between CR (grass/soil) of stable Cs and organic matter (OM), as well as between ^{137}Cs and OM, indicating that an increase of soil organic matter is followed by a higher soil-to-plant uptake of both Cs and ^{137}Cs. There was also identified a positive significant correlation between CR (grass/soil) of stable Cs and the potential bioavailability of Cs.
Finally, no further correlations between concentration ratios of Cs, ^{137}Cs, Co, I, Sr, Th and U, and soil parameters were found which possibly can be attributed to the relatively small sample sizes.

For statistical purposes it would be preferable with more samples, both for variation and correlation analyses. This would increase the reliability of the results and could possibly give a better understanding of variations and factors influencing uptake in Rogaland. It would also be interesting with a second XRD analysis of soil due to inconsistency with clay mineral fraction and Cs.

5. References

Alexander, G. A. (2016). Radiation Decontamination. In Ciottone, G. R. (ed.) *Ciottone's Disaster Medicine (Second Edition)*, pp. 519-523. Philadelphia: Elsevier.

Almås, Å. R. (2018). jord. In *Store norske leksikon*. Available at: https://snl.no/jord (accessed: 11.07.2018).

ammrf. (2012). *Parts of the machine*. Available at: http://www.ammrf.org.au/myscope/xrd/background/whatisxrd/ (accessed: 10.08.2018).

ATSDR. (2004a). *Toxicological profile for cobalt*. In U.S. Department of Health and Human Services, P. H. S., Agency for Toxic Substances and Disease Registry (ed.). Available at: https://www.atsdr.cdc.gov/ToxProfiles/tp33.pdf (accessed: 05.09.2018).

ATSDR. (2004b). *Toxicological profile for strontium*. In U.S. Department of Health and Human Services, P. H. S., Agency for Toxic Substances and Disease Registry (ed.). Available at: https://www.atsdr.cdc.gov/toxprofiles/tp159.pdf (accessed: 08.09.2018).

Baeza, A. & Guillén, J. (2006). Influence of the soil bioavailability of radionuclides on the transfer of uranium and thorium to mushrooms. *Applied Radiation and Isotopes*, 64 (9): 1020-1026. doi: https://doi.org/10.1016/j.apradiso.2006.04.003.

Barescut, J., Lariviere, D., Stocki, T., Wood, M., Beresford, N. & Copplestone, D. (2011). Limit of detection values in data analysis: Do they matter? *Radioprotection*, 46 (6): S85-S90.

Bartnicki, J., Amundsen, I., Brown, J., Hosseini, A., Hov, Ø., Haakenstad, H., Klein, H., Lind, O. C., Salbu, B., Szacinski Wendel, C. C., et al. (2016). Atmospheric transport of radioactive debris to Norway in case of a hypothetical accident related to the recovery of the Russian submarine K-27. *Journal of Environmental Radioactivity*, 151: 404-416. doi: https://doi.org/10.1016/j.jenvrad.2015.02.025.

Beresford, N. A., Mayes, R. W., Barnett, C. L. & Howard, B. J. (2007). The transfer of radiocaesium to ewes through a breeding cycle – an illustration of the pitfalls of the transfer coefficient. *Journal of Environmental Radioactivity*, 98 (1): 24-35. doi: https://doi.org/10.1016/j.jenvrad.2006.10.004.

Bunaciu, A. A., Udristioiu, E. G. & Aboul-Enein, H. Y. (2015). X-ray diffraction: instrumentation and applications. *Crit Rev Anal Chem*, 45 (4): 289-99. doi: 10.1080/10408347.2014.949616.

CERAD. (2016). *ANNUAL REPORT 2016*. CERAD Annual Reports. Available at: https://www.nmbu.no/download/file/fid/24315 (accessed: 10.07.2018).

CERAD. (2017). *ANNUAL REPORT 2017*. CERAD Annual Reports. Available at: www.nmbu.no/download/file/fid/31456 (accessed: 22.08.2018).

Chapman, H. (1965). Cation-Exchange capacity. In *Methods of soil analysis. Part 2. Chemical and microbiological properties*, pp. 891-901: American Society of Agronomy, Soil Science Society of America.

Choppin, G., Liljenzin, J.-O., Rydberg, J. & Ekberg, C. (2013a). Chapter 5 - Unstable Nuclei and Radioactive Decay. In Choppin, G., Liljenzin, J.-O., Rydberg, J. & Ekberg, C. (eds) *Radiochemistry and Nuclear Chemistry*, pp. 85-123. Oxford: Academic Press.

Choppin, G., Liljenzin, J.-O., Rydberg, J. & Ekberg, C. (2013b). Chapter 22 - Behavior of Radionuclides in the Environment. In Choppin, G., Liljenzin, J.-O., Rydberg, J. & Ekberg, C. (eds) *Radiochemistry and Nuclear Chemistry*, pp. 753-788. Oxford: Academic Press.

Cingolani, A. M., Posse, G. & Collantes, M. B. (2005). Plant functional traits, herbivore selectivity and response to sheep grazing in Patagonian steppe grasslands. *Journal of Applied Ecology*, 42 (1): 50-59.

Comans, R. N. J., Haller, M. & De Preter, P. (1991). Sorption of cesium on illite: Non-equilibrium behaviour and reversibility. *Geochimica et Cosmochimica Acta*, 55 (2): 433-440. doi: https://doi.org/10.1016/0016-7037(91)90002-M.

Cornell, R. M. (1993). Adsorption of cesium on minerals: A review. *Journal of Radioanalytical and Nuclear Chemistry*, 171 (2): 483-500. doi: 10.1007/bf02219872.

Cremers, A., Elsen, A., Preter, P. D. & Maes, A. (1988). Quantitative analysis of radiocaesium retention in soils. *Nature*, 335: 247-249. doi: 10.1038/335247a0.
Egeland, E. S. (2018). ICP. In: Store norske leksikon. Available at: https://snl.no/ICP (accessed: 10.08.2018).
Enger, Ø. (2014). *Development of method for determination of 12 elements in breast milk using Inductively coupled plasma mass spectrometry*. Master thesis. Ås: Norges miljø- og biovitenskapelige universitet. Available at: https://brage.bibsys.no/xmlui/bitstream/handle/11250/221405/MSc%20Thesis%20Øyvind%20Enger.pdf?sequence=1.
Fuller, A. J., Shaw, S., Ward, M. B., Haigh, S. J., Mosselmans, J. F. W., Peacock, C. L., Stackhouse, S., Dent, A. J., Trivedi, D. & Burke, I. T. (2015). Caesium incorporation and retention in illite interlayers. *Applied Clay Science*, 108: 128-134. doi: https://doi.org/10.1016/j.clay.2015.02.008.
Furman, B. L. (2017). Radioactive Iodine. In *Reference Module in Biomedical Sciences*: Elsevier.
Harbitz, O. & Skuterud, L. (1999). *Radioaktiv forurensning - betydning for landbruk, miljø og befolkning*: Landbruksforlaget.
Harris, D. C. (2010). *Quantitative Chemical Analysis*: W. H. Freeman.
Harstad, O. M. & Vangen, O. (2015). Kraftfôr. In *Store norske leksikon*. Available at: https://snl.no/kraftfôr (accessed: 02.07.2018).
Hesse, P. R. (1971). *A textbook of soil chemical analysis*: J. Murray.
Howard, P. J. A. & Howard, D. M. (1990). Use of organic carbon and loss-on-ignition to estimate soil organic matter in different soil types and horizons. *Biology and Fertility of Soils*, 9 (4): 306-310. doi: 10.1007/BF00634106.
IAEA. (2006). *The classification of soil systems on the basis of transfer factors of radionuclides from soil to reference plants*. IAEA-TECDOC-1497. Vienna: IAEA.
IAEA. (2010). *Handbook of parameter values for the prediction of radionuclide transfer in terrestrial and freshwater environment*. TECHNICAL REPORTS SERIES No. 472, 9201130090. Vienna: IAEA.
IAEA. (2014). *Handbook of Parameter Values for the Prediction of Radionuclide Transfer to Wildlife*. TECHNICAL REPORTS SERIES No. 479. Vienna: IAEA.
IRSN. (2012). Cobalt-60 and the environment.
Jahn, R., Blume, H.-P., Asio, V. B., Spaargaren, O., Schad, P., Langohr, R., Brinkman, R., Nachtergaele, F. O. & Krasilnikov, R. (2006). *Guidelines for soil description*: FOOD AND AGRICULTURE ORGANIZATION OF THE UNITED NATIONS.
Jensen, K. A. (2016). *Sample preparation for ICP-MS (Personal Communication)*.
Jensen, K. A. (2018). *Discussion of results*.
Joosten, H., Barthelmnes, A., Couwenberg, J., Hassel, K., Moen, A., Tegetmeyer, C. & Lyngstad, A. (2015). *Metoder for å beregne endring i klimagassutslipp ved restaurering av myr*. NTNU Vitenskapsmuseet naturhistorisk rapport. Available at: https://www.ntnu.no/documents/10476/1262347829/2015-10+Rapport+Klimagassutslipp.pdf/c83fab53-b626-4c6e-988e-efcc675e8ff2 (accessed: 22.08.2018).
Karunakara, N., Ujwal, P., Yashodhara, I., Rao, C., Sudeep Kumara, K., Dileep, B. N. & Ravi, P. M. (2013). Studies on soil to grass transfer factor (Fv) and grass to milk transfer coefficient (Fm) for cesium in Kaiga region. *J Environ Radioact*, 124: 101-12. doi: 10.1016/j.jenvrad.2013.03.008.
Kathren, R. L. (1998). NORM sources and their origins. *Applied Radiation and Isotopes*, 49 (3): 149-168. doi: https://doi.org/10.1016/S0969-8043(97)00237-6.
Kim, J. H., Gibb, H. J. & Howe, P. D. (2006). *Cobalt and inorganic cobalt compounds*. Concise International Chemical Assessment Document 69. Available at: http://www.who.int/ipcs/publications/cicad/cicad69%20.pdf (accessed: 05.09.2018).

Klein, H. & Bartnicki, J. (2018). *Worst Case Meteorological Scenario for Norway in Case of an Accident in Sellafield Nuclear Site*, Cham: Springer International Publishing.
Krogstad, T. (1992). *Metoder for jordanalyser*, 6/92: Institutt for jordagfag (Ås-NLH).
Krogstad, T. & Børresen, T. (2015). *JORD200: FIELD AND LABORATORY METHODS*: Department of environmental sciences (Norwegian University of Life Sciences).
Krupka, K. M. & Serne, R. J. (2002). *Geochemical Factors Affecting the Behavior of Antimony, Cobalt, Europium, Technetium, and Uranium in Vadose Zone Sediments*. PNNL-14126: Pacific Northwest National Lab.(PNNL), Richland, WA (United States). Available at: https://www.pnnl.gov/main/publications/external/technical_reports/PNNL-14126.pdf (accessed: 06.09.2018).
Lal, R. & Shukla, M. (2004). *Principles of Soil Physics*.
Landman, C., Päsler-Sauer, J., Raskob, W. & Trybushnyi, D. (2013). *A proposed countermeasure simulation model for the new ICRP recommendations*, vol. 48.
Lehto, J. & Hou, X. (2011a). Radiochemistry of the 3d-Transition Metals. In *Chemistry and analysis of radionuclides*, pp. 123-137. Germany: Wiley-VCH.
Lehto, J. & Hou, X. (2011b). Radiochemistry of the Actinides. In *Chemistry and Analysis of Radionuclides*, pp. 239-275. Germany: Wiley-VCH.
Lehto, J. & Hou, X. (2011c). Speciation Analysis. In *Chemistry and analysis of radionuclides*, pp. 311-355. Germany: Wiley-VCH.
Lofts, S., Tipping, E. W., Sanchez, A. L. & Dodd, B. A. (2002). Modelling the role of humic acid in radiocaesium distribution in a British upland peat soil. *Journal of Environmental Radioactivity*, 61 (2): 133-147. doi: https://doi.org/10.1016/S0265-931X(01)00118-7.
McKenzie, N., Jacquier, D., Isbell, R. & Brown, K. (2004). *Australian Soils and Landscapes: An Illustrated Compendium*: CSIRO PUBLISHING.
Milestone. (w.y). *Productivity benefits of single reaction chamber (SRC) microwave digestion- the ultraclave advantage*. Available at: http://ats-scientific.com/uploads/products/docs/Milestone-UltraWave-Single-Reaction-Chamber-Technical-Note.pdf (accessed: 09.08.2018).
Milestone, S. r. l. (2008). *UltraCLAVE Microwave Digestion System, Brochure*. Available at: http://milestonesci.com/wp-content/uploads/2017/08/UltraCLAVE_Brochure.pdf (accessed: 04.07.2018).
Ming, D. W. & Dixon, J. B. (1987). Quantitative determination of clinoptilolite in soils by a cation-exchange capacity method. *Clays and Clay Minerals*, 35 (6): 463-468.
Misaelides, P. (2011). Application of natural zeolites in environmental remediation: A short review. *Microporous and Mesoporous Materials*, 144 (1-3): 15-18.
Müller, H., Gering, F. & Pröhl, G. (2003). *Model Description of the Terrestrial Food Chain and Dose Module FDMT in RODOS PV6.0*.
Newman, M. C. (1994). *Quantitative methods in aquatic ecotoxicology*: CRC press.
Newman, M. C. & Unger, M. A. (2002). *Fundamentals of Ecotoxicology, Second Edition*: Taylor & Francis.
Nisbet, A. F. & Woodman, R. F. (2000). Soil-to-plant transfer factors for radiocesium and radiostrontium in agricultural systems. *Health Phys*, 78 (3): 279-88.
Norris, S. J. E. (2017). *Improve your ICP-OES Performance by using an Internal Standard*. Available at: https://www.armi.com/blog/how-an-internal-standard-can-improve-your-icp-oes-performance (accessed: 02.07.2018).
nrpa. (2013). *Kilder til radioaktiv forurensning*: Norwegian Radiation Protection Authority. Available at: Kilder til radioaktiv forurensning (accessed: 12.09.18).
Oughton, D. H., Salbu, B., Riise, G., Lien, H., Østby, G. & Nøren, A. (1992). Radionuclide mobility and bioavailability in Norwegian and Soviet soils. *Analyst*, 117 (3): 481-486.
PerkinElmer, I. (2011). *The 30-Minute Guide to ICP-MS*. Available at: https://www.perkinelmer.com/CMSResources/Images/44-74849tch_icpmsthirtyminuteguide.pdf (accessed: 10.08.2018).

Popic, J. M., Meland, S., Salbu, B. & Skipperud, L. (2014). Mobility of radionuclides and trace elements in soil from legacy NORM and undisturbed naturally 232 Th-rich sites. *Environmental Science: Processes & Impacts*, 16 (5): 1124-1134.

Prasad, M. N. V. (2013). *Heavy metal stress in plants: from biomolecules to ecosystems*: Springer Science & Business Media.

Rieuwerts, J. S., Thornton, I., Farago, M. E. & Ashmore, M. R. (1998). Factors influencing metal bioavailability in soils: preliminary investigations for the development of a critical loads approach for metals. *Chemical Speciation & Bioavailability*, 10 (2): 61-75. doi: 10.3184/095422998782775835.

Rigol, A., Vidal, M. & Rauret, G. (2002). An overview of the effect of organic matter on soil–radiocaesium interaction: implications in root uptake. *Journal of Environmental Radioactivity*, 58 (2-3): 191-216.

Rook, A. J., Dumont, B., Isselstein, J., Osoro, K., WallisDeVries, M. F., Parente, G. & Mills, J. (2004). Matching type of livestock to desired biodiversity outcomes in pastures – a review. *Biological Conservation*, 119 (2): 137-150. doi: https://doi.org/10.1016/j.biocon.2003.11.010.

Sakai, K. (2017). *Benefits of the Agilent 8900 ICP-QQQ with MS/MS operation for routine food analysis*. Available at: https://www.agilent.com/cs/library/applications/5991-6943EN.pdf (accessed: 10.08.2018).

Salbu, B., Lind, O. C. & Skipperud, L. (2004). Radionuclide speciation and its relevance in environmental impact assessments. *Journal of Environmental Radioactivity*, 74 (1): 233-242. doi: https://doi.org/10.1016/j.jenvrad.2004.01.008.

Salbu, B. (2006). Speciation of Radionuclides in the Environment. In Meyers, R. A. (ed.) *Encyclopedia of Analytical Chemistry*, pp. 12993–13016. Chichester: John Wiley & Sons Lt.

Salbu, B. & Skipperud, L. (2009). Speciation of radionuclides in the environment. In vol. 100, pp. 281-2.

Salbu, B. (2016). Environmental impact and risk assessments and key factors contributing to the overall uncertainties. *Journal of Environmental Radioactivity*, 151: 352-360. doi: https://doi.org/10.1016/j.jenvrad.2015.09.001.

Schollenberger, C. J. & Simon, R. H. (1945). DETERMINATION OF EXCHANGE CAPACITY AND EXCHANGEABLE BASES IN SOIL—AMMONIUM ACETATE METHOD. *Soil Science*, 59 (1): 13-24.

SCOPE 50. (1994). Radioecology after Chernobyl: Biogeochemical Pathways Artificial Radionuclides *Journal of Chemical Education*, 71 (1): A23. doi: 10.1021/ed071pA23.5.

Sumner, M. & Miller, W. (1996). Chapter 40: Cation exchange capacity and exchange coefficients. In *Methods of Soil Analysis Part 3—Chemical Methods*, pp. 1201-1229: Soil Science Society of America and American Society of Argonomy.

Templeton, D. M., Ariese, F., Cornelis, R., Danielsson, L.-G., Muntau, H., van Leeuwen, H. P. & Lobinski, R. (2000). Guidelines for terms related to chemical speciation and fractionation of elements. Definitions, structural aspects, and methodological approaches (IUPAC Recommendations 2000). *Pure and applied chemistry*, 72 (8): 1453-1470.

Tessier, A., Campbell, P. G. & Bisson, M. (1979). Sequential extraction procedure for the speciation of particulate trace metals. *Analytical chemistry*, 51 (7): 844-851.

ThermoFischer. (w.y). *ICP-OES Systems and Technologies*. Available at: https://www.thermofisher.com/no/en/home/industrial/spectroscopy-elemental-isotope-analysis/spectroscopy-elemental-isotope-analysis-learning-center/trace-elemental-analysis-tea-information/icp-oes-information/icp-oes-system-technologies.html (accessed: 10.08.2018).

Thørring, H., Brown, J., Aanensen, L. & A., H. (2016a). *Tjøtta- ICRP reference site in Norway*. StrålevernRapport: Østerås, Statens strålevern, 2016.

Thørring, H., Dyve, J., Hevrøy, T., Lahtinen, J., Liland, A., Montero, M., Real, A., Simon-Cornu, M. & Trueba, C. (2016b). *Set of improved parameter values for Nordic and Mediterranean ecosystems for Cs-134/137, Sr-90, I-131 with justification text*. COMET project deliverable COMET IRA-Human-D3. Available at: https://www.radioecology-

exchange.org/sites/default/files/files/COMET%20IRA-Human%20Food%20Chain-D3.pdf (accessed: 05.09.2018).
Thørring, H. (2018). *personal communication* (28.06.2018).
Tsukada, H., Hasegawa, H., Hisamatsu, S. i. & Yamasaki, S. i. (2002). Transfer of 137Cs and stable Cs from paddy soil to polished rice in Aomori, Japan. *Journal of Environmental Radioactivity*, 59 (3): 351-363. doi: https://doi.org/10.1016/S0265-931X(01)00083-2.
Tsukada, H., Hisamatsu, S. & Inaba, J. (2003). Transfer of 137Cs and stable Cs in soil-grass-milk pathway in Aomori, Japan. *Journal of Radioanalytical and Nuclear Chemistry*, 255 (3): 455-458. doi: 10.1023/a:1022507710447.
Uchida, S. & Tagami, K. (2007). Soil-to-plant transfer factors of fallout 137Cs and native 133Cs in various crops collected in Japan. 273: 205-210. doi: 10.1007/s10967-007-0737-5.
Uchida, S., Tagami, K. & Hirai, I. (2007). Soil-to-plant transfer factors of stable elements and naturally occurring radionuclides (1) upland field crops collected in Japan. *Journal of Nuclear Science and Technology*, 44 (4): 628-640.
UNSCEAR. (2000). *Sources and Effects of Ionizing Radiation, Report to the General Assembly, with Scientific Annexes*. New York: United Nations. Available at: http://inis.iaea.org/search/search.aspx?orig_q=RN:32002971.
UNSCEAR. (2015). *ATTACHMENT C-13: METHODOLOGY FOR THE ASSESSMENT OF DOSE FROM INGESTION OF RADIOACTIVE MATERIAL*. In Nations, U. (ed.). UNSCEAR 2013 Report, Annex A, Levels and effects of radiation exposure due to the nuclear accident after the 2011 great east-Japan earthquake and tsunami, Appendix C (Assessment of doses to the public). Available at: http://www.unscear.org/docs/reports/2013/UNSCEAR_2013A_C-13_Methodology_ingestion_2015-01.pdf (accessed: 15.09.2018).
Van Bergeijk, K., Noordijk, v. H., Lembrechts, J. & Frissel, M. (1992). Influence of pH, soil type and soil organic matter content on soil-to-plant transfer of radiocesium and-strontium as analyzed by a nonparametric method. *Journal of Environmental Radioactivity*, 15 (3): 265-276.
Weems, J. (1904). Chemistry of clays. *Iowa Geological Survey Annual Report*, 14 (1): 319-346.
Wendel, C. C., Fifield, L. K., Oughton, D. H., Lind, O. C., Skipperud, L., Bartnicki, J., Tims, S. G., Høibråten, S. & Salbu, B. (2013). Long-range tropospheric transport of uranium and plutonium weapons fallout from Semipalatinsk nuclear test site to Norway. *Environment International*, 59: 92-102. doi: https://doi.org/10.1016/j.envint.2013.05.017.
Wragg, D. (2018). *Sample preparation for XRD-analysis*.
Wragg, D. (w.y). *Basic training for the X-ray Laboratories*: University of Oslo. Unpublished manuscript.
Ytre-Eide, M. A., Standring, W. J., Amundsen, I., Sickel, M., Liland, A., Saltbones, J., Bartnicki, J., Haakenstad, H. & Salbu, B. (2009). *Konsekvenser for Norge ved en tenkt ulykke ved Sellafield-anlegget: potensielt utslipp–transport og nedfall*, 0804-4910: StrålevernRapport 2009: Nr.6 Østerås: Statens strålevern, 2009.
Zivanovic, V. (2011). *CEC- bestemmelse av kationbyttekapasitet og basemetningsgrad*. Unpublished manuscript.

Appendix A
Elemental concentrations and certified reference material

A.1. Elemental concentrations

Table 6 shows concentrations (dry weight) and relative standard deviation (RSD) for Co, Cs, I, Sr, Th and U in soil and grass for all sites in area A-E. As a measure of precision, three parallels were taken from soil sample B1, B2, C1, C2 and E1. The highest RSD were used as a proxy for the others. For grass, five parallels from site C1 were taken.

Table 6. Table of soil (dry weight) and grass (dry weight) concentration and RSD for Co, Cs, I, Sr, Th and U in all locations. Red squares represent samples taken parallels of.

	Co (mg/kg)					Cs (mg/kg)			
Location	**Soil**	**RSD**	**Grass**	**RSD**	**Location**	**Soil**	**RSD**	**Grass**	**RSD**
A1	5,1	7,8	0,10	3,7	**A1**	1,7	6,6	2,6	3,1
A2	4,1	7,8	0,29	3,7	**A2**	1,7	6,6	0,31	3,1
A3	6,8	7,8	0,053	3,7	**A3**	3,3	6,6	0,15	3,1
B1	11	0,0	0,10	3,7	**B1**	4,9	3,1	0,14	3,1
B2	8,3	0,0	0,40	3,7	**B2**	5,8	6,6	0,092	3,1
B3	6,3	7,8	0,45	3,7	**B3**	5,7	6,6	0,021	3,1
C1	16	0,0	0,27	3,7	**C1**	2,1	2,8	0,18	3,1
C2	1,5	0,0	0,052	3,7	**C2**	1,2	4,8	0,049	3,1
C3	7,2	7,8	0,046	3,7	**C3**	1,7	6,6	0,038	3,1
D1	1,6	7,8	0,047	3,7	**D1**	1,4	6,6	0,81	3,1
D2	2	7,8	0,063	3,7	**D2**	2,3	6,6	0,086	3,1
D3	2,2	7,8	0,079	3,7	**D3**	2,1	6,6	0,077	3,1
E1	4,9	7,8	0,087	3,7	**E1**	0,3	3,3	0,010	3,1
E2	4,7	7,8	0,13	3,7	**E2**	0,3	6,6	0,023	3,1
E3	5,1	7,8	0,11	3,7	**E3**	0,4	6,6	0,023	3,1
	I (mg/kg)					**Sr (mg/kg)**			
Location	**Grass**	**RSD**	**Grass**	**RSD**	**Location**	**Soil**	**RSD**	**Grass**	**RSD**
A1	17	4,4	1,0	2,3	**A1**	9,1E+01	0,4	26	1,8
A2	6,5	4,4	0,4	2,3	**A2**	1,6E+02	0,9	27	1,8
A3	4,6	4,4	0,1	2,3	**A3**	2,1E+02	0,2	23	1,8
B1	5,5	1,8	0,2	2,3	**B1**	1,8E+02	0,0	78	1,8
B2	13	0,0	1,6	2,3	**B2**	1,2E+02	4,7	56	1,8
B3	5,3	4,4	0,3	2,3	**B3**	8,8E+01	1,6	47	1,8
C1	27	4,4	0,5	2,3	**C1**	2,6E+02	2,3	83	1,8
C2	20	2,9	0,4	2,3	**C2**	2,1E+02	2,7	50	1,8
C3	34	4,4	0,3	2,3	**C3**	3,1E+02	0,8	86	1,8
D1	20	4,4	1,1	2,3	**D1**	8,1E+01	0,9	28	1,8
D2	14	4,4	0,6	2,3	**D2**	1,4E+02	1,4	66	1,8
D3	11	4,4	0,4	2,3	**D3**	1,7E+02	2,1	63	1,8
E1	6,1	3,4	0,3	2,3	**E1**	2,4E+02	0,0	59	1,8
E2	18	4,4	0,3	2,3	**E2**	1,8E+02	1,1	42	1,8
E3	13	4,4	0,6	2,3	**E3**	2,6E+02	0,6	71	1,8
	Th (mg/kg)					**U (mg/kg)**			

Location	Soil	RSD	Grass	RSD	Location	Soil	RSD	Grass	RSD
A1	3,3	52	0,0052	35	**A1**	15	1,4	0,0095	3,9
A2	2,3	52	0,0057	35	**A2**	2,9	0,8	0,0058	3,9
A3	5,7	52	0,0034	35	**A3**	3,6	0,6	0,0024	3,9
B1	8,2	1,2	0,0084	35	**B1**	2,9	2,0	0,0036	3,9
B2	6,9	2,5	0,050	35	**B2**	2,6	2,2	0,017	3,9
B3	8,0	52	0,0053	35	**B3**	2,2	1,2	0,0022	3,9
C1	3,2	1,8	0,13	35	**C1**	2,1	2,8	0,046	3,9
C2	2,7	0,0	0,0058	35	**C2**	1,4	4,1	0,0026	3,9
C3	5,0	52	0,011	35	**C3**	2,0	0,7	0,0033	3,9
D1	7,9	52	0,011	35	**D1**	1,7	2,6	0,0044	3,9
D2	8,6	52	0,021	35	**D2**	2,7	1,4	0,0083	3,9
D3	10	52	0,013	35	**D3**	3,1	1,5	0,011	3,9
E1	2,1	52	0,014	35	**E1**	0,7	0,8	0,0017	3,9
E2	2,8	52	0,0065	35	**E2**	0,7	1,7	0,0017	3,9
E3	1,8	52	0,0055	35	**E3**	0,9	2,6	0,012	3,9

Table 7 and *Table 8* presents the concentrations in pellets and milk, and in meat.
It was taken 5 parallels of pellets (area A, C, D), milk (A and C) and meat (B). The highest RSDs were used as proxies.

Table 7. Concentrations (dry weight) and RSDs for Cs, Co, I, Sr, Th and U in pellets and milk. Red squares represent samples taken parallels of. All parallels for U in milk (C1-2) were < LOD. For milk values in wet weight, use correction factors in Table 11. Location A1-3 represents goat, while the others are related to cow.

	Cs (mg/kg)					Co (mg/kg)			
Location	**Pellets**	**RSD (%)**	**Milk**	**RSD (%)**	**Location**	**Pellets**	**RSD (%)**	**Milk**	**RSD (%)**
A1-3	0,023	2,5	1,1	4,1	**A1-3**	0,45	0,035	0,0014	17
B1	0,025	2,5	0,16	4,1	**B1**	0,53	0,079	0,0019	17
C1-2	0,026	2,2	0,021	2,1	**C1-2**	0,43	0,017	0,0024	3,5
E1-2	0,029	2,0	0,015	4,1	**E1-2**	0,78	0,079	0,0024	17
	I (mg/kg)					**Sr (mg/kg)**			
Location	**Pellets**	**RSD (%)**	**Milk**	**RSD (%)**	**Location**	**Pellets**	**RSD (%)**	**Milk**	**RSD (%)**
A1-3	2,7	22	2,1	5,8	**A1-3**	37	4,7	3,08	2,7
B1	8,7	52	0,43	5,8	**B1**	16	6,3	3,6	2,7
C1-2	4,0	42	0,55	3,0	**C1-2**	16	6,3	3,94	1,4
E1-2	3,3	52	1,4	5,8	**E1-2**	16	6,3	2,6	2,7
	U (mg/kg)					**Th (mg/kg)**			
Location	**Pellets**	**RSD**	**Milk**	**RSD**	**Location**	**Pellets**	**RSD**		
A1-3	2,1	4,8	0,0019	4,5	**A1-3**	0,033	9,1		
B1	0,36	9,2	<LOD	-	**B1**	0,011	9,1		
C1-2	0,23	9,2	<LOD	0	**C1-2**	0,014	53		
E1-2	0,50	3,5	<LOQ	-	**E1-2**	0,020	2,9		

*Table 8. Concentrations (dry weight) of Cs, Co, I, Sr, Th and U in meat (mutton). The red squares represent RSDs. All parallels for U (B2), were < LOD. For Th, 2/5 were <LOD, so ½ *LOD was used. For meat values in wet weight, use correction factors in Table 10.*

Cs (mg/kg)			Co (mg/kg)		
Location	**Meat**	**RSD**	**Location**	**Meat**	**RSD**
B2	0,11	7,5	**B2**	0,0060	8,9
B2	0,061	7,5	**B2**	0,016	8,9
C1-2	0,075	7,5	**C1-2**	0,017	8,9
C1-2	0,073	7,5	**C1-2**	0,0085	8,9
C1-2	0,094	7,5	**C1-2**	0,0073	8,9
C1-2	0,093	7,5	**C1-2**	0,028	8,9
C1-2	0,063	7,5	**C1-2**	0,019	8,9
I (mg/kg)			**Sr (mg/kg)**		
Location	**Meat**	**RSD**	**Location**	**Meat**	**RSD**
B2	0,047	8,3	**B2**	0,17	7,7
B2	0,16	8,3	**B2**	0,13	7,7
C1-2	0,043	8,3	**C1-2**	2,2	7,7
C1-2	0,030	8,3	**C1-2**	0,25	7,7
C1-2	0,033	8,3	**C1-2**	0,19	7,7
C1-2	0,042	8,3	**C1-2**	0,22	7,7
C1-2	0,080	8,3	**C1-2**	0,75	7,7
Th (mg/kg)			**U (mg/kg)**		
Location	**Meat**	**RSD**	**Location**	**Meat**	**RSD**
B2	0,000084	48	**B2**	<LOD	-
B2	0,00023	48	**B2**	<LOD	-
C1-2	<LOQ	-	**C1-2**	<LOD	-
C1-2	0,00011	48	**C1-2**	<LOD	-
C1-2	0,00010	48	**C1-2**	<LOD	-
C1-2	0,00011	48	**C1-2**	<LOD	-
C1-2	0,0060	48	**C1-2**	0.0022	-

Table 9, *Table 10*, *Table 11* show ^{137}Cs concentration in soil, grass, meat and milk. RSDs (%) represent instrumental variation (High purity germanium detector).

Table 9. ^{137}Cs (Bq/kg) in soil and grass with respective relative standard deviations (RSD), and ^{137}Cs (Bq/m^2) in soil with RSDs.

	Cs-137					
Location	**Soil (Bq/kg)**	**RSD%**	**Grass (Bq/kg)**	**RSD%**	**Soil (Bq/m^2)**	**RSD%**
A1	56	6	34	3	1189	6
A2	37	3	3,4	8	1133	3
A3	43	3	3,0	8	2447	3
B1	16	3	3,5	9	1807	3
B2	69	3	5,1	4	4105	3
B3	84	6	0,65	25	4694	6
C1	23	3	6,3	5	1849	3
C2	26	3	1,7	15	1244	3
C3	6,8	3	0,73	28	879	3
D1	62	3	60	3	1808	3
D2	20	3	2,3	16	1332	3
D3	15	3	2,4	9	1173	3
E1	10	3	2,3	19	991	3
E2	9,0	3	2,3	9	849	3
E3	8,5	3	3,3	7	1173	3

Table 10. Activity (Bq/kg) of ^{137}Cs in meat in fresh and dry weight with relative standard deviation (%) and correction factor (dry to wet weight). Correction factor obtained by dividing wet with dry weight.

	Cs-137 (Bq/kg)			
Location (ID)	**Meat (wet weight)**	**Meat (dry weight)**	**RSD (%)**	**Correction factor (dry to wet)**
B2	1,83	4,4	6	0,41
B2	1,91	3,8	4	0,50
C1-2	0,45	1,2	14	0,38
C1-2	0,63	1,6	8	0,40
C1-2	1,41	3,5	5	0,40
C1-2	1,19	3,2	4	0,37
C1-2	0,62	1,6	10	0,38

*Table 11. Activity (Bq/kg) of ^{137}Cs in milk in fresh and dry weight with RSD (%) and correction factor (dry to wet weight). Correction factor obtained by dividing wet with dry weight. *below limit of detection. LOD for milk in area C was 0.6 (wet weight).*

	Cs-137 (Bq/kg)			
Area (animal)	**Milk (wet weight)**	**Milk (dry weight)**	**RSD**	**Correction factor (dry to wet)**
A (goat)	5.6	46.3	3	0.12
B (cow)	0.50	3.67	6	0.14
C (cow)	0.074*	0.3*	0	0.12
E (cow)	0.11	0.84	26	0.13

A.2. Certified reference material

Certified reference material (CRM) is presented in *Table 12- Table 15*:

Table 12. Certified reference material (CRM) used for decomposition with HNO_3, HF and basic dilution with TMAH and BENT.

Material	Type material	Supplier
NSC ZC73013 Spinage	Certified reference material	China National Accreditation Council for Registrars
NSC ZC73014 Tea	Certified reference material	China National Accreditation Council for Registrars
DOLT-5 Dogfish Liver	Certified reference material	National Research Council Canada
Standard Reference Material 1577b Bovine Liver	Certified reference material	National Institute of Standards & Technology
NSC ZC 73007 Soil	Certified reference material	China National Accreditation Council for Registrars
NSC DC 73325 Soil	Certified reference material	China National Accreditation Council for Registrars
Standard Reference Material® 1549a Whole Milk Powder	Certified reference material	National Institute of Standards & Technology
ERM®-BD151 Skimmed Milk Powder	Certified reference material	Institute for Reference Materials and Measurements

Table 13. Results of the certified reference materials (CRM) NSC Z73007 and NSC DC73325 analyzed by ICP-MS for soil. I was basic diluted, Th was decomposed with HNO_3 and Cs, Co, Sr and U were decomposed with HF. Green = value within certified range, red = outside certified range, orange = close to certified range. [1]= CRM range, [2]= quantified amount by ICP-MS.

Element	NSC ZC73007 (mg/kg)	NSC DC73325 (mg/kg)
Cs	13.02-14.06 mg/kg[1] 11 mg/kg[2]	
Co		
I		
Sr	62-72 mg/kg[1] 49 mg/kg[2]	22-30 mg/kg[1] 12 mg/kg[2]
Th		
U		

Table 14. CRM used for acid decomposition and basic dilution of grass, pellets and meat. Green = value within certified range, red = outside certified range, orange = close to certified range. NSC ZC73014 is certified for I, but was not basic diluted, hence the white square. [1]= CRM range, [2]= quantified amount by ICP-MS.

Element	ERM® BD151 (mg/kg)	BCR®- 129 (mg/kg)	NSC ZC73014 (mg/kg)	NSC ZC73013 (mg/kg)	DOLT – 5 (mg/kg)	Bovine Liver (mg/kg)
Cs						
Co						0.282-0.318 mg/kg[1] 0.23 mg/kg[2]
I						
Sr			8.9-9.3 mg/kg[1] 7.7 mg/kg[2]	82-92 mg/kg[1] 81 mg/kg[2]		91.1-99.5 µg/kg[1] 0.13 mg/kg[2]
Th						
U						

Table 15. CRM used for acid decomposition and basic dilution of milk. Green = value within certified range, red = outside certified range, orange = close to certified range. [1]= CRM range, [2]= quantified amount by ICP-MS.

Element	1549a (mg/kg)	NSC ZC73013 (mg/kg)	ERM BD151 (mg/kg)
Cs			
Co			
I			1.61-1.95 mg/kg[1] 1.5 mg/kg[2]
Sr		82-92 mg/kg[1] 74 mg/kg[2]	
U			

Appendix B

Soil properties: grain size, CEC, pH, loss-on-ignition and sequential extractions

B.1. Grain size

Grain size analysis (*Table 16*) on sample A1-2, C2 and D1 was not done because of the loss-on-ignition results, indicating a TOC content of respectively (62.7, 38.5, 53.0, 62.6) %. The method used for grain size analysis applies to an organic material content of max. 20% (Krogstad & Børresen, 2015). Method accuracy was measured by a homemade control which was run in parallel. The relative standard deviations for the control`s fractions of clay, silt and sand was respectively (8, 4, 2) %.

Table 16. Amount clay, silt and sand and soil classification (Jahn et al., 2006). The remaining locations are categorized as organic soils.

Sample	Clay (%) (<2 µm)	Silt (%) (20-63 µm)	Sand (%) (63-2000 µm)	Soil type
A3	5,30	25,6	69,1	Sandy loam
B1	8,90	77,1	14,0	Silt loam
B2	7,30	53,7	39,0	Silt loam
B3	4,80	50,7	44,5	Silt loam
C1	12,4	44,2	43,4	Loam
C3	2,00	19,5	78,5	Loamy sand
D2	5,70	28,1	66,2	Sandy loam
D3	5,00	39,3	55,7	Sandy loam
E1	0,50	4,80	94,7	Sand
E2	0,70	3,70	95,6	Sand
E3	0,90	4,80	94,3	Sand

B.2. Cation exchange capacity (CEC)

Table 17 gives an overview of CEC data. Two CRMs were measured for analytical accuracy. The majority were within the certified range (green). Ca and K were close (yellow) for 1 of 2 CRMs.

Table 17. Shows concentrations of (Ca^{2+}, K^{+}, Mg^{2+}, Na^{+} and H^{+}) mol^{+}/kg, V NaOH used for titration, limit of detection (LOD), limit of quantification (LOQ) and certified reference values. [1]= CRM range, [2]= quantified amount by ICP.. Green = value within certified range, orange = close to certified range.

	Ca^{2+} (cmol+/kg)	K^{+} (cmol+/kg)	Mg^{2+} (cmol+/kg)	Na^{+} (cmol+/kg)	H^{+} (cmol+/kg)	CEC (cmol+/kg)	NaOH (ml) used for titration
LOD	0,0183	0,0302	0,0109	0,0244	1,47		
LOQ	0,0611	0,1008	0,0362	0,0815	4,89		
A1	12	1,3	4,2	0,53	44	62	2,45
A2	27	1,8	7,5	0,45	12	49	0,9
A3	17	0,89	5,8	0,22	11	35	0,87
B1	5,0	0,21	0,68	0,15	7,9	14	0,7
B2	2,8	0,76	1,8	0,27	16	22	1,1
B3	1,9	0,82	1,4	0,21	15	19	1,02
C1	26	1,7	3,0	0,54	16	47	1,09
C2	28	1,1	5,1	0,84	32	67	1,84
C3	7,2	0,14	0,82	0,2	9,2	18	0,76
D1	5,0	1,8	4,3	0,77	50	62	2,7
D2	11	1,0	1,4	0,35	16	30	1,1
D3	8,1	0,92	1,5	0,35	19	30	1,22
E1	3,1	0,61	1,5	0,28	9,6	15	0,78
E2	2,7	0,47	1,3	0,22	14	19	0,97
E3	2,1	0,18	0,69	0,17	13	16	0,93
CRM85113	22,5±1[1] 20[2]	0,94±0,06[1] 0,86[2]	5,6	0,27			
CRM85101	17,8±1,0[1] 17[2]	0,99±0,06[1] 1,0[2]	4,5	0,36			

B.3. pH and loss-on-ignition

Five parallels were taken from soil C1 for pH and loss-on-ignition, giving relative standard deviations of respectively, 0.68 % and 3.2 % (*Table 18*). Loss-on-ignition, *corrected* is adjusted for water-holding clay based on the clay content in grain size analysis. By subtracting loss-on-ignition (%) with a correction factor, loss-on-ignition (%), corrected is provided. A factor of 1 for clay content of 5-9 % and 2 for above 10 % was used in present study (Krogstad, 1992).

Table 18. Overview of pH, dry weight (%), loss-on-ignition (%) and loss-on-ignition (%) corrected at the respective locations.

Location	pH	Dry weight (%)	Loss-on-ignition (%)	Loss-on-ignition (%) corrected
A1	5,7	93,1	62,7	61,7
A2	6,4	95,2	38,5	37,5
A3	6,1	97,0	20,5	19,5
B1	5,9	98,9	6,2	5,2
B2	5,8	97,3	20,8	19,8
B3	5,4	97,9	17,1	17,1
C1	6,2	95,8	25,0	23,0
C2	5,7	94,1	53,0	52,0
C3	5,8	98,6	7,0	7,0
D1	4,8	93,3	62,6	61,6
D2	5,7	96,9	24,0	23,0
D3	5,5	97,7	19,5	18,5
E1	5,8	98,7	10,0	10,0
E2	5,8	98,8	8,6	8,6
E3	5,6	99,3	4,5	4,5

B.4. Sequential extractions

Table 19 shows sequential extractions for step 1 (water soluble fraction), step 2 (exchangeable fraction) and step 3 (carbonate associated fractions), extracted with respectively Milli-Q water, NH_4Ac (pH soil) and NH_4Ac (pH 5). Three parallels were taken from sample A1, B1 and B2. Step 2 was not done for D1, due to a soil pH at 4.8. For calculation of the potential bioavailability, values from step 3 were used as a proxy. LOD and LOQ value are shown in *Table 20*. A control standard which was used for analytical accuracy gave satisfying results.

Table 19. Sequential extractions (mg/kg, dry weight) and standard deviations (%) (RSD) for Cs, Co, Sr, Th and U in locations A1-2, B1-2, C1-2, D1 and E1.

Cs (mg/kg)

Location	Step 1	RSD (%)	Step 2	RSD (%	Step 3	RSD (%
A1	8,6E-04	5,9	3,0E-02	4,9	2,2E-02	2,3
A2	4,5E-04		6,7E-03		3,4E-03	
B1	4,1E-04	2,0	6,3E-03	3,7	3,9E-03	4,7
B2	9,4E-04	6,6	2,0E-02	1,8	1,2E-02	17
C1	1,7E-04		4,1E-03		3,1E-03	
C2	2,1E-04		3,3E-03		1,7E-03	
D1	2,2E-04		N.A.		4,3E-03	

E1	6,5E-05		7,6E-04		3,5E-04	
			Co (mg/kg)			
Location	**Step 1**	**RSD (%**	**Step 2**	**RSD (%**	**Step 3**	**RSD**
A1	8,2E-03	7,7	2,2E-01	3,0	4,4E-01	4,5
A2	1,3E-02		5,6E-02		1,2E-01	
B1	4,2E-03	7,1	4,4E-02	1,6	1,0E-01	4,8
B2	1,1E-02	4,1	2,1E-01	3,1	3,3E-01	5,4
C1	1,5E-02		6,4E-02		1,7E-01	
C2	4,8E-03		3,0E-02		5,5E-02	
D1	4,8E-03		N.A.		1,1E-01	
E1	2,0E-03		2,0E-02		2,4E-02	
			Sr (mg/kg)			
Location	**Step 1**	**RSD (%**	**Step 2**	**RSD (%**	**Step 3**	**RSD (%**
A1	0,15	10	4,9	3,4	4,0	1,7
A2	0,31		9,5		5,5	
B1	0,21	6,2	7,5	1,1	2,4	9,3
B2	0,16	9,9	5,1	3,2	2,5	7,8
C1	0,59		20		13	
C2	0,30		31		20	
D1	0,23		N.A.		9,1	
E1	0,12		5,3		1,9	
			Th (mg/kg)			
Location	**Step 1**	**RSD (%**	**Step 2**	**RSD (%**	**Step 3**	**RSD (%**
A1	0,0029	25	0,015	6,0	0,051	
A2	0,0027		0,0045		0,012	
B1	0,0018	6,4	0,0079	3,7	0,027	
B2	0,0060	6,5	0,018	1,1	0,047	
C1	0,00055		0,0021		0,0059	
C2	0,0023		0,0043		0,010	
D1	0,0091		N.A.		0,064	
E1	0,00079		0,0069		0,0086	
			U (mg/kg)			
Location	**Step 1**	**RSD (%**	**Step 2**	**RSD (%**	**Step 3**	**RSD (%**
A1	0,017	4,8	0,64	3,6	2,4	1,5
A2	0,0054		0,049		0,20	
B1	0,0017	2,8	0,081	3,0	0,14	4,2
B2	0,0048	2,2	0,10	2,5	0,19	3,9
C1	0,0018		0,026		0,075	
C2	0,0013		0,016		0,045	
D1	0,0018		N.A.		0,045	
E1	0,0014		0,023		0,038	

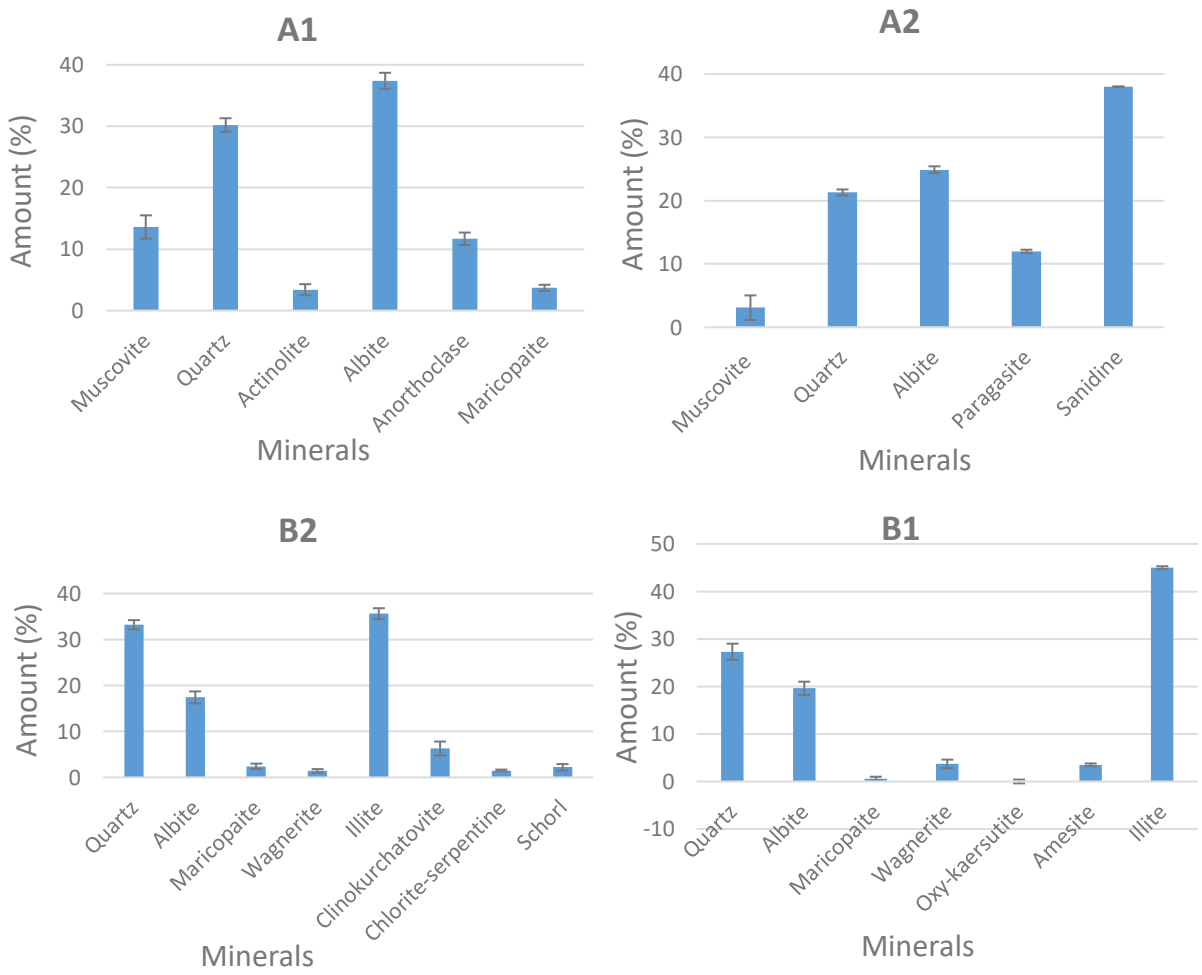
Table 20. Limit of detection (LOD) and limit of quantification (LOQ) for the sequential extractions.

	Step 1		Step 2		Step 3	
Element	**LOD**	**LOQ**	**LOD**	**LOQ**	**LOD**	**LOQ**
Cs (mg/kg)	0,00001	0,000038	0,00004	0,0001	0,00008	0,0003
Co (mg/kg)	0,00001	0,000046	0,0001	0,0004	0,0008	0,003
Sr (mg/kg)	0,01	0,047	0,09	0,3	0,07	0,2
Th (mg/kg)	0,000004	0,000012	0,0001	0,0004	0,00008	0,0003
U (mg/kg)	0,00002	0,000067	0,0003	0,0009	0,0004	0,001

Appendix D

X-ray diffraction (XRD) analysis

Results from X-ray diffraction analysis done at sites A1-2, B1-2, C1-2, D1 and E1 are shown below (*Figure 25* and *Table 21):*



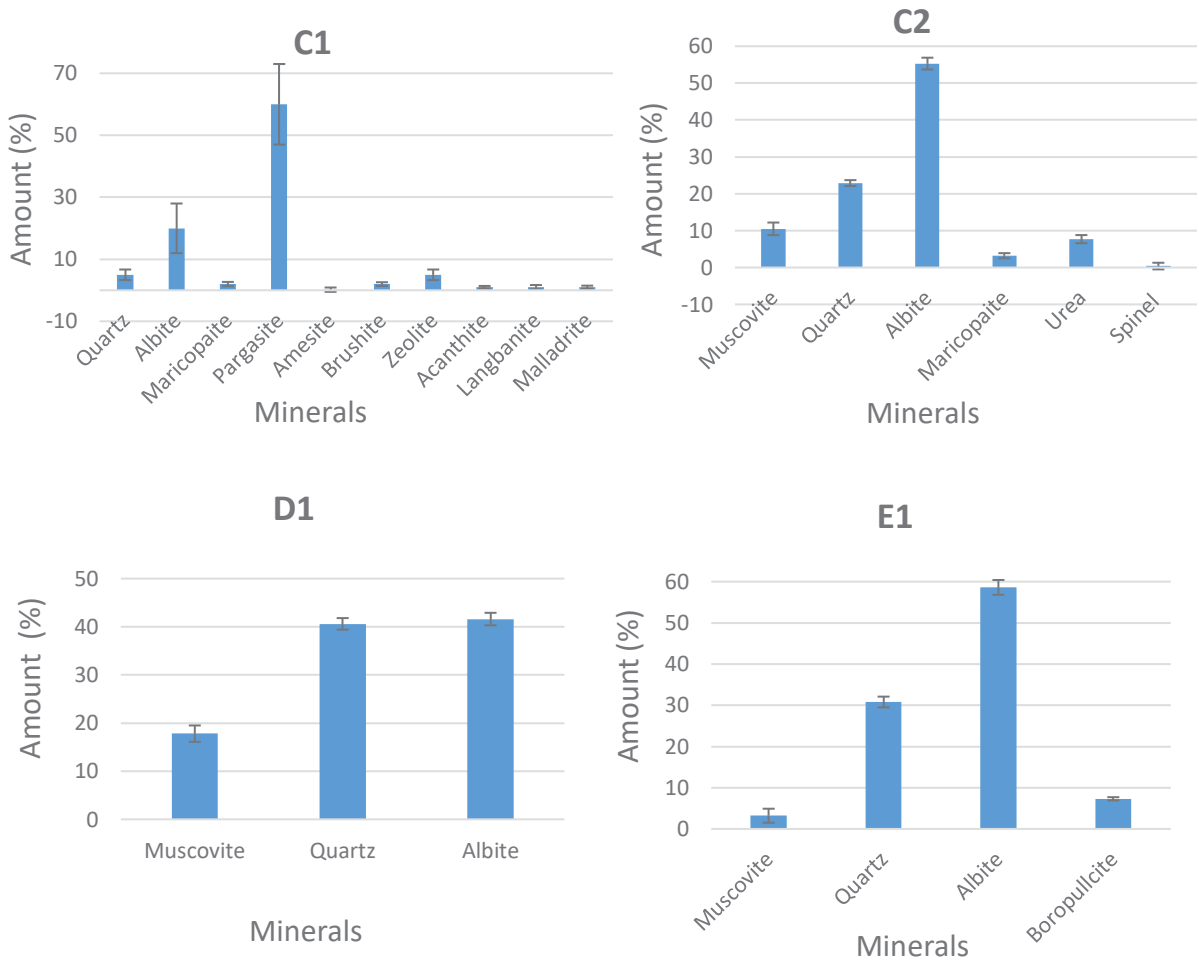


Figure 25. Shows minerals in location A1-2, B1-2, C1-2, D1 and E1 obtained by XRD-analysis.

Table 21. Minerals detected by XRD-analysis, elemental composition and qualitative PDF (reference) (TOPAS).

	Elemental composition	**PDF (reference)**
Muscovite	(Ba,K)Al2(Si3Al)O10(OH)2	00-010-0490
Quartz	SiO2	01-070-7344
Actinolite	Ca2 (Mg,Fe $^{2+}$)5 Si8O22 (OH)	01-089-5367
Albite	NaAlSi3O8	00-009-0466
Anorthoclase	(K,Na)AlSi3O8	00-009-0478
Maricopaite	Pb7Ca2(Si,Al)48O100·32H2O	01-073-9748
Pargasite	NaCa2(Mg,Fe++)4Al(Si6Al2)O22(OH)2	01-085-2159
Sanidine	KalSi3O8	00-010-0357
Wagnerite	(MgFe)2PO4F	01-071-5310
Oxy-kaersutite	NaCa2Mg4Ti(Si6Al2O23)(OH)2	01-086-1716
Amesite	$Mg_2Al_2SiO_5(OH)_4$	01-082-9425
Illite	$(K,H_3O)(Al,Mg,Fe)_2(Si,Al)_4O_{10}[(OH)_2,(H_2O)]$	00-058-2016
Clinokurchatovite	Ca(Mg,Fe++,Mn)B2O5	01-084-0035
Chlorite-serpentine	$(Mg_5Al)(AlSi_3)O_{10}(OH)_8$	00-052-1044

Schorl	$NaFe^{2+}{}_3Al_6(BO_3)_3Si_6O_{18}(OH)_4$	01-084-6120
Brushite	Ca(HPO4)(H2O)2	01-072-0713
Zeolite	(Na,Ca)Al3Si3O12	00-019-1183
Acanthite	Ag2S	01-080-8434
Langbanite	$(Mn^{2+}$,Ca)4(Mn3+,Fe3+)9Sb5+Si2O24	00-014-0195
Malladrite	$(Na,Cs)_2SiF6$	01-071-6476
Urea	$CO(NH2)_2$	01-072-0118
Spinel	(Cu,Al)Cr2Se4	01-078-3257
Boropollucite	CsBSi5O12	01-083-8309

Output of results from qualitative and quantitative analyses of XRD-results (site B1) are shown in *Figure 26 and Figure 27*:

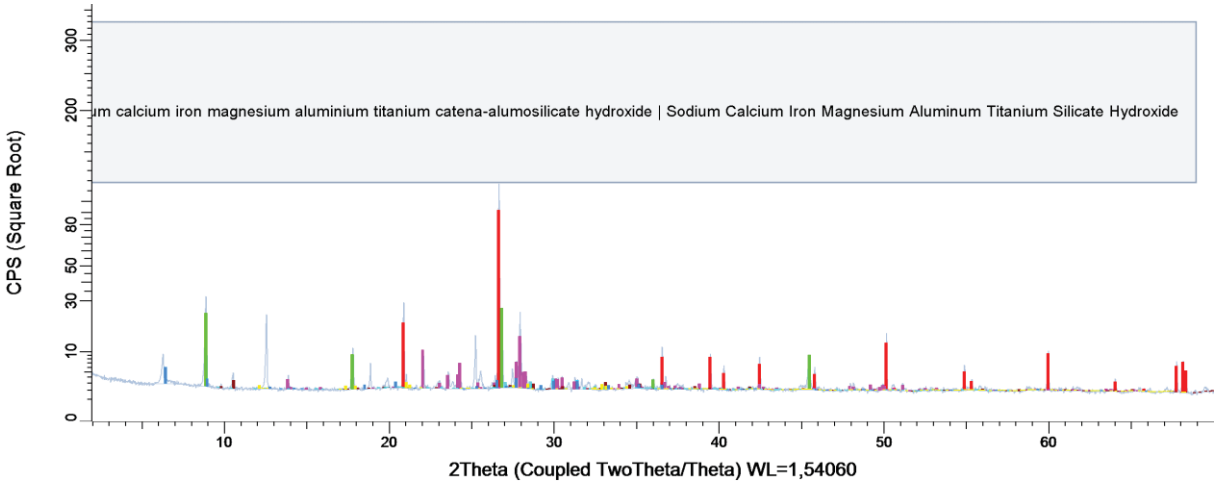


Figure 26. Phase ID of minerals originating from location B1.

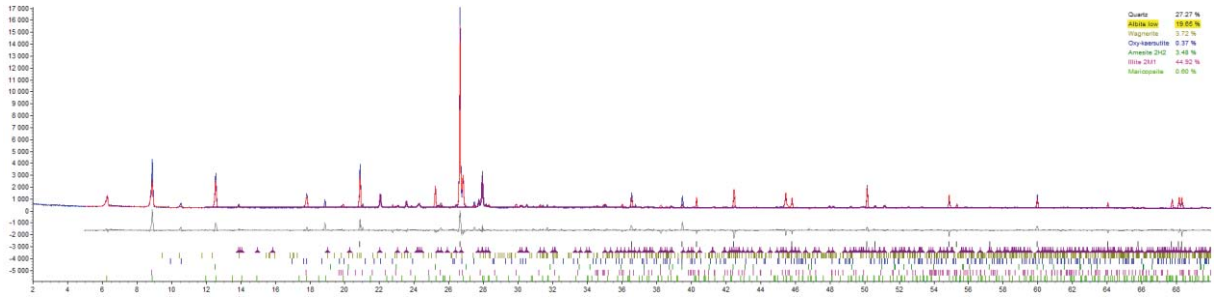


Figure 27. Quantitative determination of minerals from location B1.

For principal component analysis purposes, locations where the clay minerals (illite) were not detected (N.A.), ½*LOD were used (*Table 22*). For Rietveld quantitative analysis it is common to use 4% as

LOD, but for phases were peaks are clear, percentages less than that are acceptable (Wragg, 2018) However, 0.5*4% was used as LOD in present study.

Table 22. Clay particles detected by XRD.

Location	Illite (%)
A1	N.A.
A2	N.A.
B1	45
B2	36
C1	N.A.
C2	N.A.
D1	N.A.
E1	N.A.

Appendix E

Concentration ratios and transfer coefficients

Table 23 shows elements which had values below the detection of limit and/or the quantification limit.

Table 23. Total sample values < detection limit (LOD) and quantification limit (LOD).

Matrix	Element	<LOD (%)	<LOQ (%)
Milk – goat	Cd	100	
	Sb	80	20
Milk – cow	Ce	0	43
	Cd	100	
	Eu	86	14
	Nb		100
	La	43	29
	Sb	86	14
	U	67	33
	Zr	0	43
Meat	Ba	0	64
	Ce	0	64
	Cd	100	
	Eu	27	18
	La	0	64
	Mn	0	82
	Pb	91	0
	Sb	18	82
	Th	0	14
	U	86	0
	Zr	91	0

E.1. Concentration ratios (grass/soil)

Site specific values

Values used for calculation of concentration ratios in *Table 24* are in *Table 6* Appendix A. Ranges ($min_{(CR-SD)}$ – $max_{(CR+SD)}$) for Cs, ^{137}Cs, Co, I, Sr, Th and U were 0.0036-1.6, 0.0058-1.0, 0.0060-0.072, 0.0075-0.13, 0.11-0.55, 0.00059-0.054, 0.00055-0.024, respectively, representing following sites B3-A1, B3-D1, C3-B3, C3-B2, A3-B3, A3-C1 and A3-C1.

*Table 24. Concentration ratios (CR)(mg*kg^{-1}/mg*kg^{-1}), arithmetic standard deviation (ASD) and minimum and maximum values for Cs, ^{137}Cs, Co, I, Sr, Th and U for all sites A1-E3. Transfer coefficients (TC) (m^2/kg) for ^{137}Cs with ASD are also listed. n=1 for all values.*

Cs

Site	CR	SD	Min.	Max.
A1	1,5E+00	2,1E-02	1,5E+00	1,6E+00
A2	1,8E-01	1,4E-03	1,8E-01	1,9E-01
A3	4,5E-02	2,5E-04	4,5E-02	4,6E-02
B1	2,8E-02	8,8E-04	2,7E-02	2,9E-02
B2	1,6E-02	1,0E-03	1,5E-02	1,7E-02
B3	3,7E-03	1,4E-05	3,6E-03	3,7E-03
C1	8,5E-02	2,4E-03	8,2E-02	8,9E-02
C2	4,2E-02	2,1E-03	4,0E-02	4,4E-02
C3	2,2E-02	1,5E-04	2,2E-02	2,3E-02
D1	5,8E-01	1,5E-02	5,7E-01	5,9E-01
D2	3,7E-02	5,5E-04	3,7E-02	3,8E-02
D3	3,7E-02	5,3E-04	3,6E-02	3,7E-02
E1	3,3E-02	1,1E-03	3,2E-02	3,4E-02
E2	7,9E-02	1,3E-03	7,7E-02	8,2E-02
E3	6,6E-02	1,7E-03	6,4E-02	6,8E-02

Co

Site	CR	SD	Min.	Max.
A1	1,9E-02	2,6E-04	1,8E-02	1,9E-02
A2	7,1E-02	5,3E-04	7,0E-02	7,1E-02
A3	7,8E-03	4,3E-05	7,3E-03	8,3E-03
B1	9,0E-03	0,0E+00	8,9E-03	9,1E-03
B2	4,8E-02	0,0E+00	4,8E-02	4,9E-02
B3	7,1E-02	2,9E-04	7,1E-02	7,2E-02
C1	1,7E-02	0,0E+00	1,6E-02	1,8E-02
C2	3,5E-02	0,0E+00	3,4E-02	3,6E-02
C3	6,4E-03	4,3E-05	6,0E-03	6,8E-03
D1	2,9E-02	7,7E-04	2,7E-02	3,2E-02
D2	3,2E-02	4,6E-04	3,0E-02	3,3E-02
D3	3,6E-02	5,2E-04	3,5E-02	3,7E-02
E1	1,8E-02	1,4E-03	1,6E-02	1,9E-02
E2	2,8E-02	4,6E-04	2,7E-02	2,8E-02
E3	2,2E-02	5,6E-04	2,0E-02	2,3E-02

I

Site	CR	SD	Min.	Max.
A1	5,6E-02	7,8E-04	5,5E-02	5,8E-02
A2	5,8E-02	4,4E-04	5,8E-02	5,9E-02
A3	3,0E-02	1,7E-04	3,0E-02	3,1E-02
B1	3,3E-02	6,0E-04	3,2E-02	3,4E-02
B2	1,2E-01	0,0E+00	1,2E-01	1,3E-01
B3	6,4E-02	3,9E-03	6,1E-02	6,7E-02
C1	1,8E-02	7,5E-04	1,7E-02	1,9E-02
C2	2,1E-02	5,9E-04	2,0E-02	2,1E-02
C3	7,6E-03	5,2E-05	7,5E-03	7,8E-03
D1	5,5E-02	1,4E-03	5,4E-02	5,6E-02
D2	4,3E-02	6,3E-04	4,2E-02	4,4E-02
D3	3,5E-02	5,1E-04	3,4E-02	3,7E-02
E1	5,1E-02	1,8E-03	4,9E-02	5,3E-02
E2	1,9E-02	3,2E-04	1,8E-02	2,0E-02
E3	4,5E-02	1,2E-03	4,2E-02	4,7E-02

Sr

Site	CR	SD	Min.	Max.
A1	2,9E-01	4,0E-03	2,8E-01	2,9E-01
A2	1,7E-01	1,3E-03	1,7E-01	1,7E-01
A3	1,1E-01	6,0E-04	1,1E-01	1,1E-01
B1	4,3E-01	0,0E+00	4,3E-01	4,4E-01
B2	4,5E-01	2,1E-02	4,3E-01	4,8E-01
B3	5,3E-01	7,9E-03	5,2E-01	5,5E-01
C1	3,2E-01	7,3E-03	3,2E-01	3,3E-01
C2	2,3E-01	6,3E-03	2,3E-01	2,4E-01
C3	2,8E-01	1,9E-03	2,7E-01	2,8E-01
D1	3,5E-01	9,0E-03	3,4E-01	3,5E-01
D2	4,7E-01	6,9E-03	4,6E-01	4,8E-01
D3	3,7E-01	5,3E-03	3,6E-01	3,8E-01
E1	2,5E-01	0,0E+00	2,4E-01	2,5E-01
E2	2,3E-01	3,9E-03	2,3E-01	2,4E-01
E3	2,7E-01	7,1E-03	2,6E-01	2,8E-01

Th

U

Site	CR	SD	Min.	Max.	Site	CR	SD	Min.	Max.
A1	1,6E-03	2,2E-05	1,5E-03	1,6E-03	A1	6,3E-04	3,7E-05	6,0E-04	6,7E-04
A2	2,5E-03	1,9E-05	2,4E-03	2,5E-03	A2	2,0E-03	4,1E-05	2,0E-03	2,0E-03
A3	6,0E-04	3,3E-06	5,9E-04	6,1E-04	A3	6,7E-04	1,2E-04	5,5E-04	7,8E-04
B1	1,0E-03	1,2E-05	9,9E-04	1,1E-03	B1	1,2E-03	2,8E-05	1,2E-03	1,3E-03
B2	7,2E-03	1,8E-04	7,1E-03	7,4E-03	B2	6,5E-03	1,5E-04	6,3E-03	6,6E-03
B3	6,6E-04	1,0E-05	6,5E-04	6,8E-04	B3	1,0E-03	4,6E-05	9,5E-04	1,0E-03
C1	4,0E-02	7,1E-04	2,6E-02	5,4E-02	C1	2,2E-02	1,1E-03	2,1E-02	2,4E-02
C2	2,1E-03	4,3E-19	2,1E-03	2,2E-03	C2	1,8E-03	1,7E-04	1,6E-03	2,0E-03
C3	2,2E-03	1,5E-05	2,1E-03	2,3E-03	C3	1,7E-03	1,5E-04	1,5E-03	1,8E-03
D1	1,4E-03	3,6E-05	1,3E-03	1,4E-03	D1	2,6E-03	1,3E-04	2,5E-03	2,7E-03
D2	2,4E-03	3,6E-05	2,4E-03	2,5E-03	D2	3,1E-03	8,5E-05	3,0E-03	3,2E-03
D3	1,3E-03	1,9E-05	1,3E-03	1,3E-03	D3	3,5E-03	1,4E-04	3,4E-03	3,7E-03
E1	6,6E-03	3,3E-03	3,3E-03	9,9E-03	E1	2,4E-03	9,6E-05	2,3E-03	2,4E-03
E2	2,3E-03	3,9E-05	2,2E-03	2,5E-03	E2	2,5E-03	1,1E-04	2,4E-03	2,6E-03
E3	3,1E-03	7,9E-05	2,9E-03	3,2E-03	E3	1,4E-02	5,3E-04	1,3E-02	1,4E-02
^{137}Cs (CR)					^{137}Cs (TC) (m^2/kg)				
Site	CR	SD	Min.	Max.	Site	TF	SD	Min.	Max.
A1	6,0E-01	4,0E-02	5,6E-01	6,4E-01	A1	2,8E-02	1,9E-03	2,6E-02	3,0E-02
A2	9,1E-02	7,8E-03	8,3E-02	9,9E-02	A2	3,0E-03	2,5E-04	2,7E-03	3,2E-03
A3	7,0E-02	6,0E-03	6,4E-02	7,6E-02	A3	1,2E-03	1,1E-04	1,1E-03	1,3E-03
B1	2,2E-01	2,0E-02	2,0E-01	2,4E-01	B1	1,9E-03	1,8E-04	1,8E-03	2,1E-03
B2	7,4E-02	3,7E-03	7,0E-02	7,8E-02	B2	1,2E-03	6,2E-05	1,2E-03	1,3E-03
B3	7,7E-03	2,0E-03	5,8E-03	9,7E-03	B3	1,4E-04	3,6E-05	1,0E-04	1,7E-04
C1	2,7E-01	1,6E-02	2,6E-01	2,9E-01	C1	3,4E-03	2,0E-04	3,2E-03	3,6E-03
C2	6,4E-02	9,8E-03	5,4E-02	7,4E-02	C2	1,4E-03	2,1E-04	1,2E-03	1,6E-03
C3	1,1E-01	3,0E-02	7,7E-02	1,4E-01	C3	8,3E-04	2,3E-04	5,9E-04	1,1E-03
D1	9,6E-01	4,1E-02	9,2E-01	1,0E+00	D1	3,3E-02	1,4E-03	3,2E-02	3,4E-02
D2	1,2E-01	1,9E-02	1,0E-01	1,4E-01	D2	1,7E-03	2,8E-04	1,5E-03	2,0E-03
D3	1,6E-01	1,5E-02	1,4E-01	1,7E-01	D3	2,0E-03	1,9E-04	1,8E-03	2,2E-03
E1	2,4E-01	4,5E-02	1,9E-01	2,8E-01	E1	2,3E-03	4,4E-04	1,9E-03	2,8E-03
E2	2,6E-01	2,4E-02	2,3E-01	2,8E-01	E2	2,7E-03	2,6E-04	2,5E-03	3,0E-03
E3	3,9E-01	3,0E-02	3,6E-01	4,2E-01	E3	2,8E-03	2,1E-04	2,6E-03	3,0E-03

Soil-to-plant transfer for ROGFARM

Uptake in grass for entire ROGFARM is listed (*Table 25*) and illustrated (*Figure 28*) below.

Table 25. Arithmetic mean (AM), arithmetic standard deviation (ASD), geometric mean (GM), geometric standard deviation (GSD) and minimum and maximum values (AM ± ASD) of concentration ratios (dry/dry weight). The same parameters are given for ^{137}Cs, transfer coefficient (m^2/kg). n=15. Unit is ($mg*kg^{-1}/mg*kg^{-1}$), except for Ca, Cl, Fe, K, Mg, Mn, Na and Zn ($g*kg^{-1}/g*kg^{-1}$).

Element	AM	ASD	GM	GSD	Min.	Max.
Ag	5,0E-02	2,7E-02	4,4E-02	1,7E+00	1,7E-02	1,1E-01
Ba	7,5E-02	4,4E-02	6,5E-02	1,7E+00	2,6E-02	1,8E-01
Ca	8,0E-01	5,1E-01	6,7E-01	1,8E+00	1,1E-01	2,3E+00
Cd	1,6E-01	8,5E-02	1,4E-01	1,7E+00	6,3E-02	3,5E-01
Ce	5,1E-03	7,1E-03	2,9E-03	2,8E+00	5,8E-04	3,0E-02
Cl	1,4E+02	1,1E+02	1,1E+02	2,0E+00	1,2E+01	4,2E+02
Co	3,0E-02	2,0E-02	2,5E-02	1,9E+00	6,0E-03	7,2E-02
Cs	1,9E-01	4,0E-01	7,9E-02	3,7E+00	3,6E-03	1,6E+00
Cs-137	2,4E-01	2,5E-01	1,7E-01	2,4E+00	5,8E-03	1,0E+00
Cu	9,3E-01	5,6E-01	7,9E-01	1,8E+00	3,8E-01	2,3E+00
Eu	7,0E-03	4,1E-03	6,1E-03	1,7E+00	1,7E-03	1,7E-02
Fe	1,0E-02	5,1E-03	9,1E-03	1,6E+00	3,1E-03	2,0E-02
I	4,4E-02	2,8E-02	3,7E-02	1,8E+00	7,5E-03	1,3E-01
K	3,9E+00	4,9E+00	2,4E+00	2,7E+00	1,5E+00	2,2E+01
La	6,4E-03	7,8E-03	4,0E-03	2,6E+00	6,3E-04	3,3E-02
Mg	1,4E+00	8,9E-01	1,2E+00	1,8E+00	2,4E-01	3,1E+00
Mn	4,0E-01	2,9E-01	3,3E-01	1,9E+00	5,5E-02	1,2E+00
Na	1,3E-01	1,2E-01	9,2E-02	2,2E+00	2,2E-02	4,4E-01
Nb	2,9E-03	4,4E-03	1,6E-03	3,0E+00	2,9E-04	1,9E-02
Pb	7,2E-03	6,0E-03	5,5E-03	2,1E+00	2,8E-03	2,8E-02
S	4,6E+00	4,0E+00	3,4E+00	2,1E+00	9,7E-01	1,6E+01
Sb	2,2E-01	2,5E-01	1,4E-01	2,5E+00	3,2E-02	7,9E-01
Se	5,6E-02	3,8E-02	4,7E-02	1,8E+00	1,0E-02	1,1E-01
Sr	3,2E-01	1,2E-01	3,0E-01	1,4E+00	1,1E-01	5,5E-01
Th	5,0E-03	9,9E-03	2,3E-03	3,5E+00	5,9E-04	5,4E-02
U	4,4E-03	6,0E-03	2,6E-03	2,8E+00	5,5E-04	2,4E-02
Zn	7,7E-01	3,1E-01	7,2E-01	1,5E+00	4,1E-01	1,4E+00
Zr	2,6E-03	5,8E-03	1,0E-03	3,9E+00	2,9E-04	2,4E-02
			TC (m^2/kg)			
^{137}Cs	5,7E-03	1,0E-02	2,8E-03	3,3E+00	1,0E-04	3,4E-02

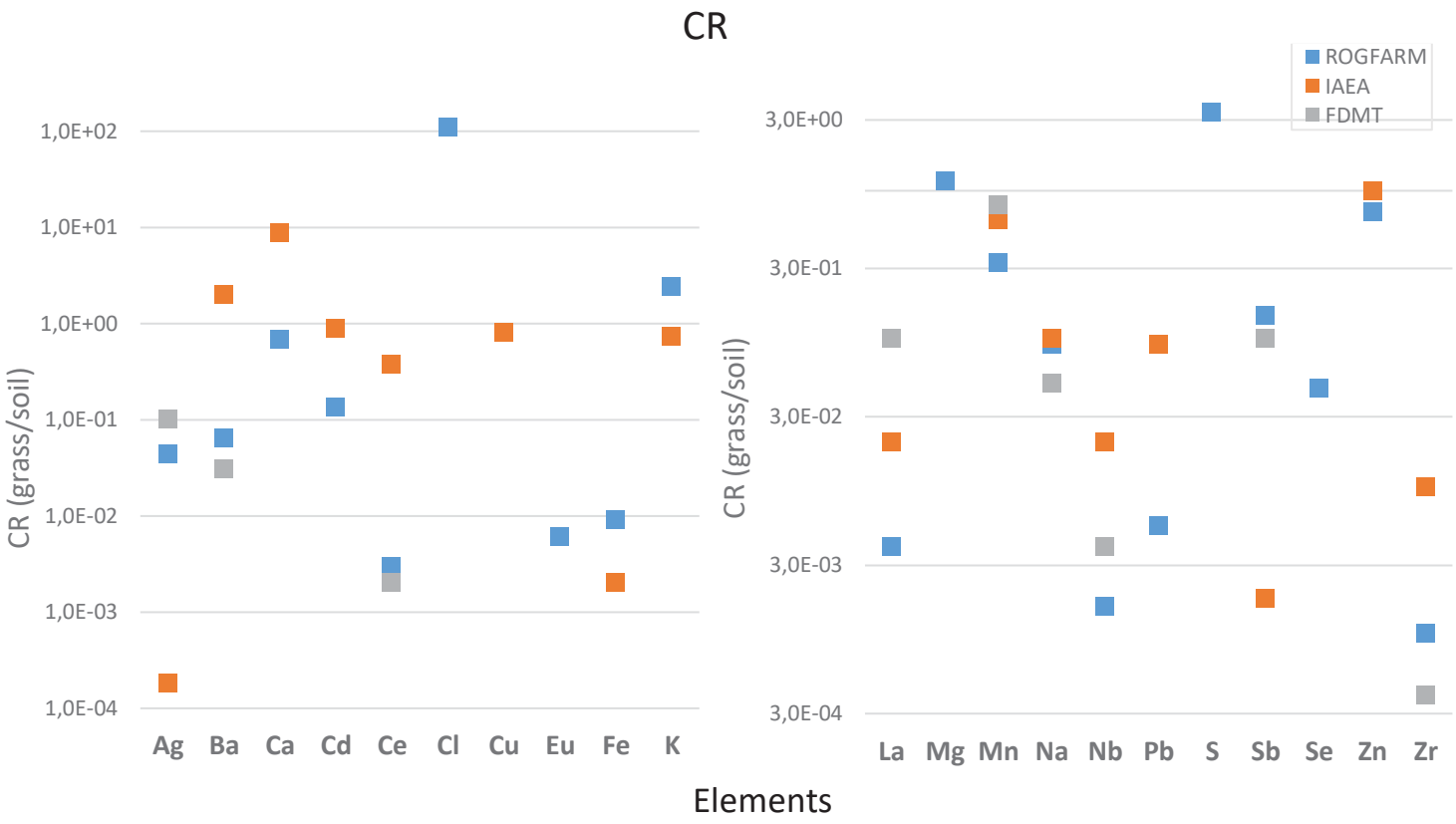


Figure 28. Shows concentration ratios (dry/dry weight) in geometric mean for ROGFARM and IAEA and "expert judgement" values for FDMT. Note the logarithmic y-axis and the different scales. Cesium, ^{137}Cs, Co, I, Sr, Th, U are not added.

E.2. Uptake in goat`s milk

Uptake in goat`s milk are listed as transfer coefficients (F_m) (*Table 26*) and illustrated (*Figure 29*) below. Concentration ratios (CRs) are also presented (*Table 27*). Values are given in arithmetic mean (AM), arithmetic standard deviation (ASD), geometric mean (GM), geometric standard deviation (GSD) and minimum and maximum values, and are given in wet/dry weight.

Table 26. F_m (d/L) of uptake in goat`s milk, n=1.

Element	AM	ASD	GM	GSD	Min.	Max.
Al	1,4E+00	1,6E+00	9,3E-01	2,5	<LOD	3,0E+00
Ba	2,9E-03	9,0E-04	2,8E-03	1,4	2,0E-03	3,8E-03
Ca	7,5E-02	3,7E-03	7,5E-02	1,1	7,1E-02	7,8E-02
Cd*	-	-	-	-	-	-
Ce	1,3E-04	1,2E-04	9,7E-05	2,2	1,4E-05	2,5E-04
Cl	1,3E-01	3,1E-02	1,3E-01	1,3	9,8E-02	1,6E-01
Co	5,3E-04	2,7E-04	4,7E-04	1,6	2,6E-04	8,0E-04
Cs	1,1E-01	1,4E-01	6,4E-02	2,8	<LOD	2,5E-01
Cs-137	2,8E-01	3,7E-01	1,7E-01	2,7	<LOD	6,5E-01
Cu	3,2E-03	2,8E-04	3,1E-03	1,1	2,9E-03	3,4E-03
Eu	2,0E-03	6,7E-04	1,9E-03	1,4	1,3E-03	2,7E-03
Fe	1,3E-03	2,5E-04	1,2E-03	1,2	1,0E-03	1,5E-03
I	1,8E-01	6,7E-02	1,7E-01	1,4	1,2E-01	2,5E-01
K	5,9E-02	1,7E-02	5,7E-02	1,3	4,2E-02	7,6E-02

La	9,0E-05	6,4E-05	7,3E-05	1,9	2,6E-05	1,5E-04
Mg	3,0E-02	1,3E-03	3,0E-02	1,1	2,8E-02	3,1E-02
Mn	2,7E-04	8,9E-05	2,5E-04	1,4	1,8E-04	3,6E-04
Na	1,6E-01	2,6E-02	1,6E-01	1,2	1,4E-01	1,9E-01
Nb	2,2E-03	5,2E-04	2,2E-03	1,3	1,7E-03	2,7E-03
Pb	7,9E-04	1,4E-04	7,8E-04	1,2	6,5E-04	9,4E-04
Sb*	-	-	-	-	-	-
Se	6,3E-02	8,0E-03	6,2E-02	1,1	5,5E-02	7,1E-02
Sr	9,0E-03	5,9E-04	9,0E-03	1,1	8,4E-03	9,6E-03
U	3,6E-04	1,6E-05	3,6E-04	1,1	3,4E-04	3,7E-04
Zn	3,9E-02	5,6E-03	3,8E-02	1,2	3,3E-02	4,4E-02
Zr	3,1E-04	1,4E-04	2,8E-04	1,6	1,6E-04	4,5E-04

*total samples > 50% below LOD

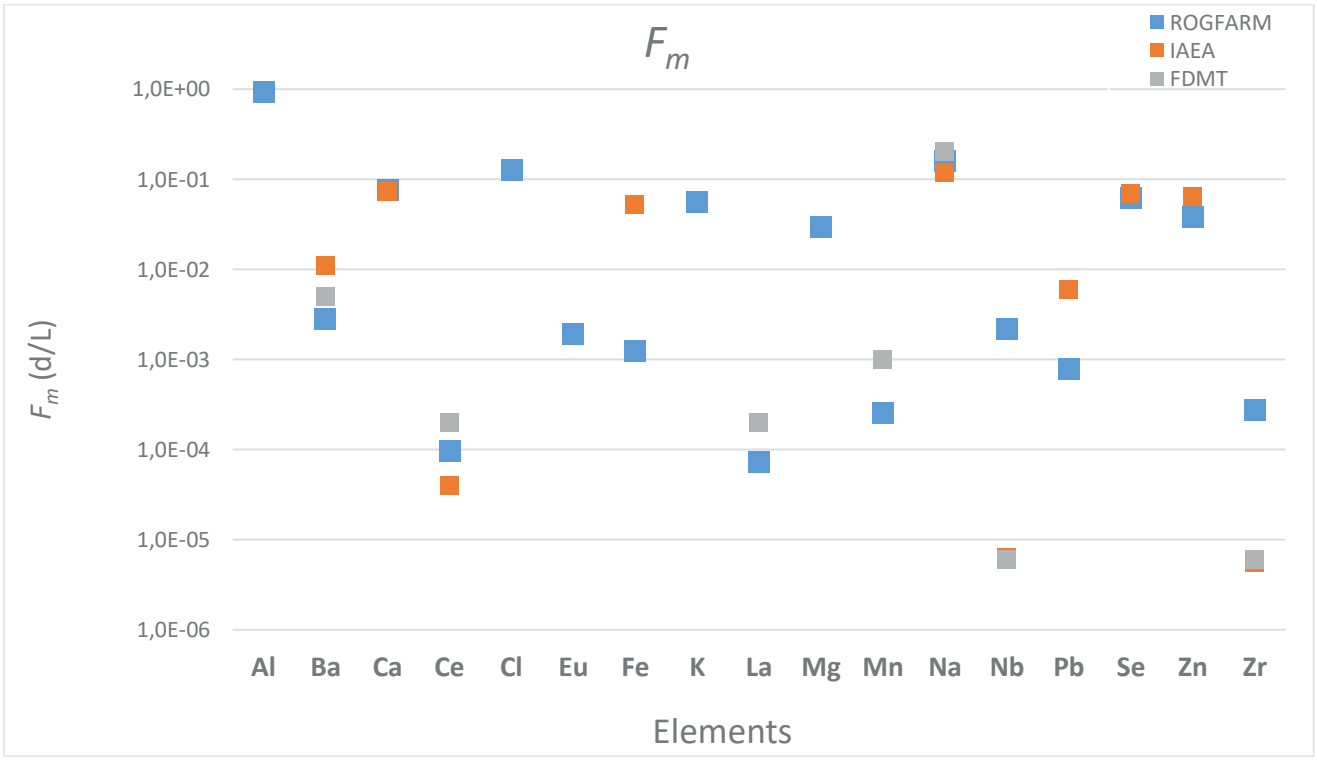


Figure 29. Transfer coefficients (GM) for ROGFARM and IAEA (2010) and "expert judgement" values for FDMT (except for those presented in the results and discussion part (Cs, ^{137}Cs, Co, I, Sr, Th, U). Note the logarithmic y-axis.

Table 27. Concentration ratios of goat`s milk, n=1. Unit is ($mg*kg^{-1}/mg*kg^{-1}$), except for Al, Ca, Cl, Fe, K, Mg, Mn, Na and Zn ($g*kg^{-1}/g*kg^{-1}$).

Element	AM	SD	GM	GSD	Min.	Max.
Al	2,1E+00	2,4E+00	1,4E+00	2,5	<LOD	4,6E+00
Ba	4,4E-03	1,3E-03	4,2E-03	1,35	3,1E-03	5,8E-03
Ca	1,1E-01	5,5E-03	1,1E-01	1,1	1,1E-01	1,2E-01
Cd*	-	-	-	-	-	-
Ce	1,9E-04	1,7E-04	1,5E-04	2,2	2,1E-05	3,7E-04
Cl	1,9E-01	4,7E-02	1,9E-01	1,3	1,5E-01	2,4E-01
Co	7,9E-04	4,1E-04	7,0E-04	1,6	3,8E-04	1,2E-03
Cs	1,6E-01	2,1E-01	9,5E-02	2,8	<LOD	3,7E-01
Cs-137	4,2E-01	5,5E-01	7,0E-01	2,7	<LOD	9,7E-01
Cu	4,7E-03	4,2E-04	4,7E-03	1,1	4,3E-03	5,2E-03
Eu	3,0E-03	1,0E-03	2,9E-03	1,4	2,0E-03	4,0E-03
Fe	1,9E-03	3,8E-04	1,9E-03	1,2	1,5E-03	2,3E-03
I	2,7E-01	1,0E-01	2,6E-01	1,4	1,7E-01	3,7E-01
K	8,8E-02	2,6E-02	8,5E-02	1,3	6,2E-02	1,1E-01
La	1,3E-04	9,6E-05	1,1E-04	1,9	3,9E-05	2,3E-04
Mg	4,4E-02	2,0E-03	4,4E-02	1,1	4,2E-02	4,7E-02
Mn	4,0E-04	1,3E-04	3,8E-04	1,4	2,7E-04	5,3E-04
Na	2,5E-01	3,9E-02	2,4E-01	1,2	2,1E-01	2,8E-01
Nb	3,3E-03	7,7E-04	3,3E-03	1,3	2,6E-03	4,1E-03
Pb	1,2E-03	2,1E-04	1,2E-03	1,2	9,8E-04	1,4E-03
Sb*	-	-	-	-	-	-
Se	9,4E-02	1,2E-02	9,4E-02	1,1	8,2E-02	1,1E-01
Sr	1,3E-02	8,9E-04	1,3E-02	1,1	1,3E-02	1,4E-02
U	5,4E-04	2,4E-05	5,3E-04	1,1	5,1E-04	5,6E-04
Zn	5,8E-02	8,4E-03	5,7E-02	1,2	5,0E-02	6,6E-02
Zr	4,6E-04	2,1E-04	4,2E-04	1,6	2,5E-04	6,7E-04

*total samples > 50% below LOD

E.3. Uptake in cow`s milk

Site specific transfer coefficients (F_m) (*Table 28*), as well as F_m (*Table 29*) and concentrations ratios (CR) (*Table 30*) for entire ROGFARM are presented below. F_m are also illustrated (*Figure 30*). Values are given in arithmetic mean (AM), arithmetic standard deviation (ASD), geometric mean (GM), geometric standard deviation (GSD) and minimum and maximum values. All values are in wet/dry weight.

Table 28. Site specific transfer coefficients (d/L) in AM of cow`s milk in Rogaland, n=1. Uranium are excluded since over 50% of the total sample values were below the limit of detection.

Cs					Co				
Site	**F_m**	**SD**	**Min.**	**Max.**	**Site**	**F_m**	**SD**	**Min.**	**Max.**
B1	1,5E-02	5,4E-04	1,4E-02	1,5E-02	**B1**	1,1E-04	5,1E-06	1,1E-04	1,2E-04
C1-2	2,2E-03	1,3E-03	8,6E-04	3,5E-03	**C1-2**	1,1E-04	5,1E-05	5,8E-05	1,6E-04
E1-2	8,4E-03	2,7E-03	5,7E-03	1,1E-02	**E1-2**	1,0E-04	9,1E-06	9,5E-05	1,1E-04
I					Sr				
Site	**F_m**	**SD**	**Min.**	**Max.**	**Site**	**F_m**	**SD**	**Min.**	**Max.**
B1	2,5E-03	2,6E-04	2,2E-03	2,7E-03	**B1**	6,0E-04	1,3E-05	5,8E-04	6,1E-04
C1-2	4,7E-03	1,9E-04	4,5E-03	4,9E-03	**C1-2**	6,9E-04	1,9E-04	5,0E-04	8,8E-04
E1-2	1,6E-02	5,6E-04	1,6E-02	1,7E-02	**E1-2**	6,3E-04	1,1E-04	5,2E-04	7,4E-04

^{137}Cs				
Site	**F_m**	**SD**	**Min.**	**Max.**
B1	1,1E-02	1,2E-03	1,0E-02	1,3E-02
C1-2	7,5E-04	6,4E-04	1,1E-04	1,4E-03
E1-2	3,9E-03	1,0E-03	2,9E-03	4,9E-03

Table 29. Transfer coefficients (d/L) for cow`s milk in ROGFARM, n=3.

Element	AM	SD	GM	GSD
Al	4,6E-02	5,7E-02	2,9E-02	2,6
Ba	2,3E-04	4,9E-05	2,2E-04	1,2
Ca	1,5E-02	4,7E-03	1,4E-02	1,4
Cd*	-	-	-	-
Ce	2,1E-05	2,2E-05	1,5E-05	2,3
Cl	4,5E-03	2,2E-04	4,5E-03	1,1
Co	1,1E-04	3,9E-06	1,1E-04	1,0
Cs	8,5E-03	6,3E-03	6,8E-03	1,9
Cs-137	5,3E-03	5,4E-03	3,7E-03	2,3
Cu	3,1E-04	1,0E-04	2,9E-04	1,4
Eu*	-	-	-	-
Fe	7,8E-05	4,2E-05	6,9E-05	1,7
I	7,8E-03	7,3E-03	5,6E-03	2,2
K	3,8E-03	4,8E-04	3,7E-03	1,1

La	5,3E-06	5,2E-06	3,7E-06	2,3
Mg	2,6E-03	1,6E-04	2,6E-03	1,1
Mn	9,0E-06	2,6E-06	8,6E-06	1,3
Na	1,3E-02	3,3E-03	1,2E-02	1,3
Nb**	-	-	-	-
Pb	1,3E-04	8,9E-05	1,1E-04	1,9
Sb*	-	-	-	-
Se	1,0E-02	1,4E-03	1,0E-02	1,1
Sr	6,4E-04	4,8E-05	6,4E-04	1,1
U*	-	-	-	-
Zn	5,9E-03	4,7E-04	5,8E-03	1,1
Zr	4,8E-05	4,6E-05	3,5E-05	2,2

*total samples > 50% below LOD, ** total samples > 50% below LOQ

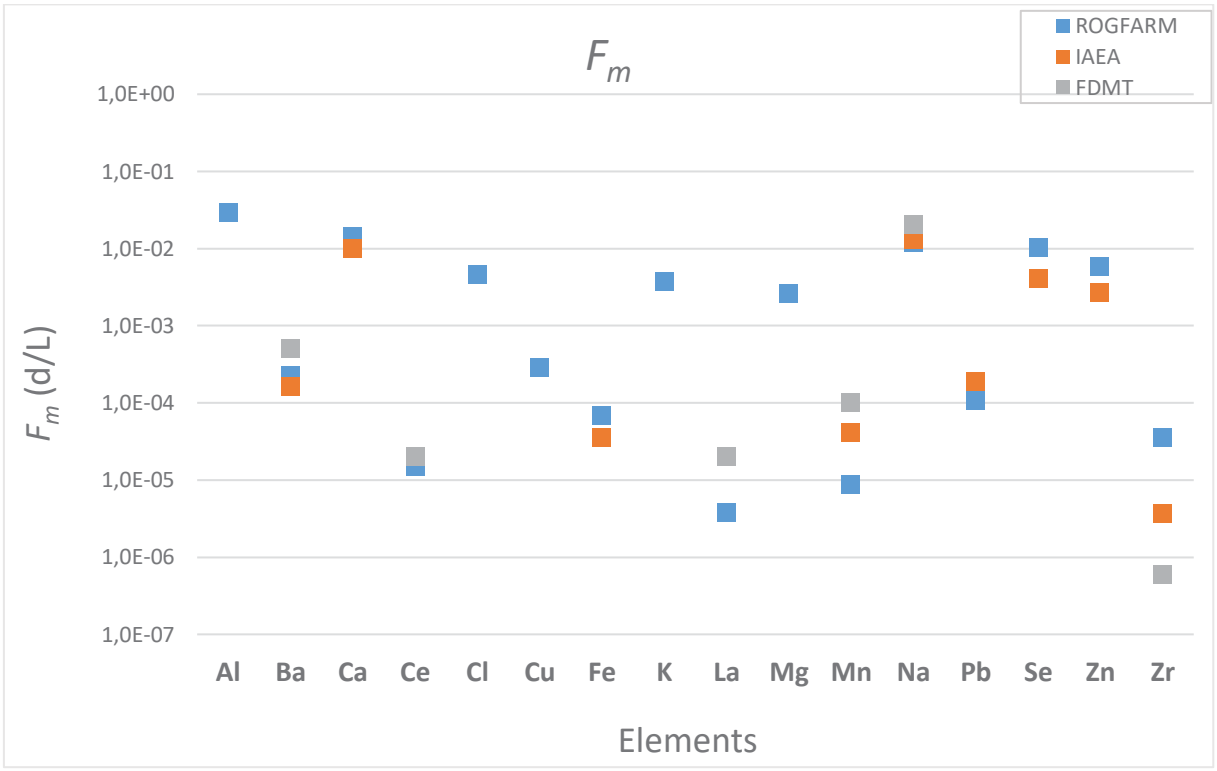


Figure 30. Transfer coefficients for (milk/dry matter, wet/dry weight) in geometric mean for ROGFARM and IAEA and "expert judgement" values for FDMT. Note the logarithmic y-axis.

*Table 30. Concentration ratios for cow`s milk in Rogaland, n=15. Unit is (mg*kg^{-1}/mg*kg^{-1}), except for Al, Ca, Cl, Fe, K, Mg, Mn, Na and Zn (g*kg^{-1}/g*kg^{-1}).*

Element	AM	SD	GM	GSD
Al	5,7E-01	7,1E-01	3,6E-01	2,6
Ba	2,9E-03	6,1E-04	2,8E-03	1,2
Ca	1,9E-01	5,9E-02	1,8E-01	1,4
Cd*	-	-	-	-
Ce	2,7E-04	2,7E-04	1,9E-04	2,3
Cl	5,7E-02	2,8E-03	5,7E-02	1,1
Co	1,4E-03	4,9E-05	1,4E-03	1,0
Cs	1,1E-01	7,9E-02	8,5E-02	1,9
Cs-137	6,6E-02	6,8E-02	4,6E-02	2,3
Cu	3,9E-03	1,2E-03	3,7E-03	1,4
Eu*	-	-	-	-
Fe	9,8E-04	5,3E-04	8,6E-04	1,7
I	9,7E-02	9,2E-02	7,1E-02	2,2
K	4,7E-02	6,0E-03	4,7E-02	1,1
La	6,6E-05	6,6E-05	4,7E-05	2,3
Mg	3,2E-02	2,0E-03	3,2E-02	1,1
Mn	1,1E-04	3,2E-05	1,1E-04	1,3
Na	1,6E-01	4,1E-02	1,5E-01	1,3
Nb**	-	-	-	-
Pb	1,6E-03	1,1E-03	1,3E-03	1,9
Sb*	-	-	-	-
Se	1,3E-01	1,7E-02	1,3E-01	1,1
Sr	8,0E-03	6,0E-04	8,0E-03	1,1
U*	-	-	-	-
Zn	7,3E-02	5,9E-03	7,3E-02	1,1
Zr	6,0E-04	5,8E-04	4,4E-04	2,2

*total samples > 50% below LOD, ** total samples > 50% below LOQ

E.4. Uptake in mutton

Site specific transfer coefficients (F_f) (*Table 31*) as well as F_f and concentrations ratios (CR) for entire ROGFARM are presents below (*Table 32*). F_f are also illustrated in *Figure 31*. Values are given in arithmetic mean (AM), arithmetic standard deviation (ASD), geometric mean (GM), geometric standard deviation (GSD) and minimum and maximum values. All values are in wet/dry weight.

Table 31. Individual transfer coefficients (d/L) in AM for Cs, Co, I, Sr and Th in mutton in Rogaland. In fact, the transfer coefficients below are similar as concentration ratios since daily intake was set to 1 kg, n=1.

Cs

Site	F_f	SD	Min.	Max.
B2	3,2E-01	5,7E-02	2,6E-01	3,8E-01
B2	7,0E-01	7,2E-02	6,3E-01	7,7E-01
C1-2	3,0E-01	2,3E-01	6,8E-02	5,2E-01
C1-2	3,0E-01	2,3E-01	7,1E-02	5,4E-01
C1-2	4,0E-01	3,0E-01	9,5E-02	7,0E-01
C1-2	3,6E-01	2,8E-01	8,6E-02	6,4E-01
C1-2	2,5E-01	2,0E-01	5,8E-02	4,5E-01

Co

Site	F_f	SD	Min.	Max.
B2	1,6E-02	1,5E-03	1,4E-02	1,7E-02
B2	7,1E-03	6,9E-04	6,4E-03	7,8E-03
C1-2	3,0E-02	1,7E-02	1,2E-02	4,7E-02
C1-2	1,6E-02	9,1E-03	6,6E-03	2,5E-02
C1-2	1,4E-02	7,9E-03	5,7E-03	2,2E-02
C1-2	4,8E-02	2,8E-02	2,0E-02	7,6E-02
C1-2	3,4E-02	2,0E-02	1,4E-02	5,4E-02

I

Site	F_f	SD	Min.	Max.
B2	2,2E-02	1,8E-03	2,0E-02	2,4E-02
B2	7,8E-03	6,5E-04	7,1E-03	8,4E-03
C1-2	1,4E-02	1,2E-03	1,3E-02	1,5E-02
C1-2	1,0E-02	9,0E-04	9,3E-03	1,1E-02
C1-2	1,1E-02	1,0E-03	1,0E-02	1,2E-02
C1-2	1,3E-02	1,2E-03	1,2E-02	1,5E-02
C1-2	2,6E-02	2,3E-03	2,4E-02	2,9E-02

Sr

Site	F_f	SD	Min.	Max.
B2	1,1E-03	8,9E-05	1,0E-03	1,2E-03
B2	1,8E-03	1,5E-04	1,7E-03	2,0E-03
C1-2	1,5E-02	5,0E-03	9,6E-03	2,0E-02
C1-2	1,8E-03	6,0E-04	1,2E-03	2,4E-03
C1-2	1,4E-03	4,6E-04	8,9E-04	1,8E-03
C1-2	1,4E-03	4,9E-04	9,5E-04	1,9E-03
C1-2	5,1E-03	1,7E-03	3,4E-03	6,8E-03

Th

Site	F_f	SD	Min.	Max.
B2	2,3E-03	1,4E-03	9,1E-04	3,6E-03
B2	1,0E-03	6,0E-04	4,0E-04	1,6E-03
C1-2	2,7E-04	3,6E-04	-8,6E-05	6,3E-04
C1-2	7,7E-04	1,0E-03	-2,4E-04	1,8E-03
C1-2	7,1E-04	9,3E-04	-2,3E-04	1,6E-03
C1-2	7,2E-04	9,5E-04	-2,3E-04	1,7E-03
C1-2	4,1E-02	5,3E-02	-1,3E-02	9,4E-02

Table 32. Transfer coefficients/concentration ratios for mutton in Rogaland (daily intake was set to 1 kg, thus F_f (d/L) is equal to CR ($mg*kg^{-1}/mg*kg^{-1}$). Al, Ca, Cl, Fe, K, Mg, Mn, Na and Zn is in ($g*kg^{-1}/g*kg^{-1}$), n=7.

Element	AM	SD	GM	GSD
Ag	5,4E-02	4,1E-02	4,3E-02	2,0
Al	1,5E-02	3,7E-02	5,8E-03	4,0
Ba**	-	-	-	-
Ca	1,8E-02	2,6E-02	9,9E-03	2,9
Cd*	-	-	-	-
Ce**	-	-	-	-
Cl	4,4E-02	9,2E-03	4,3E-02	1,2
Co	2,3E-02	1,4E-02	2,0E-02	1,8
Cs	3,8E-01	1,5E-01	3,5E-01	1,5
Cs-137	2,6E-01	1,1E-01	6,0E-01	1,5
Cu	3,9E-02	1,2E-02	3,8E-02	1,3
Eu	1,4E-02	2,9E-02	6,3E-03	3,6
Fe	3,3E-02	2,3E-02	2,7E-02	1,9
I	1,5E-02	6,7E-03	1,4E-02	1,5
K	8,2E-02	3,2E-03	8,2E-02	1,0
La**	-	-	-	-
Mg	4,4E-02	3,5E-03	4,4E-02	1,1
Mn**	-	-	-	-
Na	4,1E-01	3,8E-02	4,1E-01	1,1
Nb	1,4E-02	2,3E-02	7,5E-03	3,1
Pb*	-	-	-	-
S	4,4E-01	1,7E-02	4,4E-01	1,0
Sb**	-	-	-	-
Se	2,7E-01	1,2E-01	2,4E-01	1,6
Sr	3,9E-03	4,9E-03	2,4E-03	2,7
Th	6,6E-03	1,5E-02	2,7E-03	3,8
U*	-	-	-	-
Zn	5,4E-01	2,8E-02	5,4E-01	1,1
Zr*	-	-	-	-

*total samples > 50% below LOD, ** total samples > 50% below LOQ

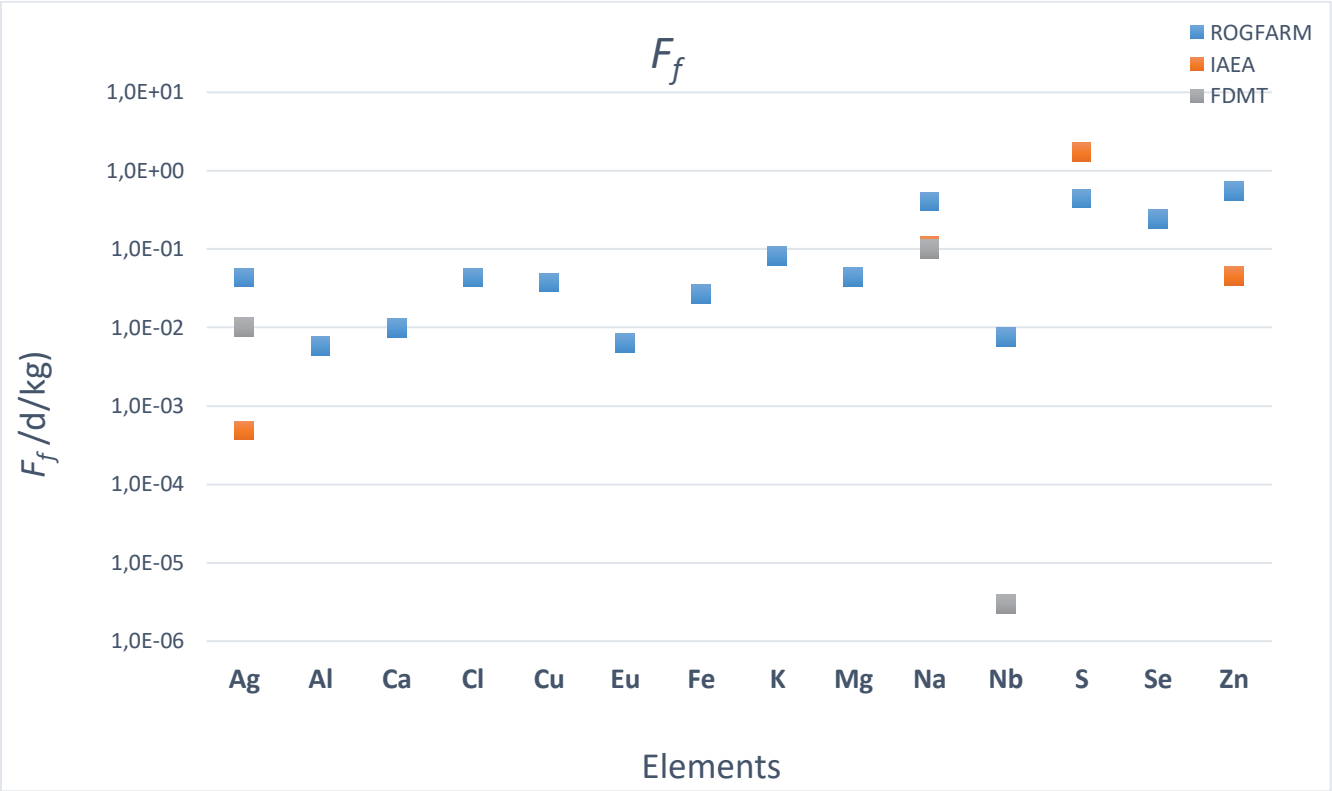


Figure 31. Transfer coefficients (mutton/grass, wet/dry weight) in geometric mean for ROGFARM and IAEA and "expert judgement" values for FDMT. Note the logarithmic y-axis

Appendix F
Additional data

F.1. Additional data of results from sequential extractions

Iron (Fe) and manganese (Mn) soil concentrations at sites it was taken sequential extractions of are listed below (*Table 33*):

Table 33. Concentrations (mg/kg) of Fe and Mn from sites subjected to sequential extractions.

Site	Fe (mg/kg)	Mn (mg/kg)
A1	13	0,30
A2	15	0,41
B1	36	1,2
B2	27	0,83
C1	49	0,81
C2	14	0,26
D1	6,3	0,25
E1	15	0,35

Additional data applied in the thesis are presented below (*Table 34* and *Table 35*):

Table 34. Sequential extractions result of Ca, Fe, Na, K and Mg (mg/kg, dry weight) in sites A1-2, B1-2, C1-2, D1 and E1

	Ca (mg/kg)			Fe (mg/kg)		
Location	**Step 1**	**Step 2**	**Step 3**	**Step 1**	**Step 2**	**Step 3**
A1	1,7E-02	1,1E+00	7,5E-01	1,9E+00	7,6E+00	4,0E+01
A2	4,7E-02	2,9E+00	1,5E+00	4,7E+00	5,0E+00	1,7E+01
B1	2,2E-02	7,3E-01	2,0E-01	3,4E+00	1,4E+00	1,4E+01
B2	1,6E-02	3,8E-01	1,6E-01	5,7E+00	6,7E+00	6,9E+01
C1	8,2E-02	2,9E+00	1,7E+00	4,0E+00	5,4E+00	2,2E+01
C2	3,4E-02	3,0E+00	1,8E+00	4,8E+00	7,1E+00	3,5E+01
D1	2,4E-02	6,9E-01	6,9E-01	4,3E+00	1,2E+01	1,2E+01
E1	1,1E-02	4,3E-01	1,4E-01	2,2E+00	3,3E+00	2,0E+01
	Na (mg/kg)			**K (mg/kg)**		
Location	**Step 1**	**Step 2**	**Step 3**	**Step 1**	**Step 2**	**Step 3**
A1	5,2E-02	7,4E-02	3,9E-02	4,2E-02	2,3E-01	1,2E-01
A2	7,7E-02	5,6E-02	2,0E-02	1,1E-01	3,0E-01	9,7E-02
B1	4,6E-02	2,0E-02	2,0E-02	8,7E-03	4,6E-02	1,4E-02
B2	7,5E-02	2,6E-02	2,0E-02	4,5E-02	1,6E-01	4,6E-02
C1	1,2E-01	5,5E-02	2,0E-02	1,2E-01	3,2E-01	1,1E-01
C2	9,9E-02	1,2E-01	3,7E-02	5,9E-02	2,4E-01	7,2E-02
D1	7,8E-02	1,2E-01	1,2E-01	9,3E-02	4,6E-01	4,6E-01
E1	8,2E-02	3,7E-02	2,0E-02	4,4E-02	1,2E-01	2,8E-02

	Mg (mg/kg)		
Location	**Step 1**	**Step 2**	**Step 3**
A1	5,0E-03	2,8E-01	1,5E-01
A2	1,4E-02	5,9E-01	2,3E-01
B1	6,0E-03	7,2E-02	1,5E-02
B2	5,0E-03	1,7E-01	4,9E-02
C1	1,7E-02	2,4E-01	9,2E-02
C2	8,7E-03	4,1E-01	1,5E-01
D1	7,9E-03	4,0E-01	4,0E-01
E1	3,7E-03	1,4E-01	3,0E-02

*Table 35. Soil concentrations of Ca, Fe, Na, K, Mg, Mn, S and Cl (mg/kg, dry weight) in all locations. *represents sites subjected for sequential extractions.*

Location	Ca (mg/kg)	Fe (mg/kg)	Na (mg/kg)	K (mg/kg)	Mg (mg/kg)
A1*	6,0	13	6,5	6,1	2,4
A2*	13	15	14	15	3,3
A3	13	27	17	17	5,7
B1*	9,3	36	14	19	5,9
B2*	4,3	27	11	19	4,7
B3	2,8	24	8,3	20	4,6
C1*	26	49	17	4,4	13
C2*	17	14	14	2,3	1,2
C3	16	29	25	15	5,0
D1*	3,7	6,3	7,9	8,1	1,0
D2	6,5	8,4	18	18	1,5
D3	7,1	7,3	18	19	1,5
E1*	9,2	15	22	13	1,5

E2	9,4	16	9,0	11	1,5
E3	9,7	18	23	14	1,5

Location	Mn (mg/kg)	S (mg/kg)	Cl (mg/kg)
A1*	0,30	2,5	0,29
A2*	0,41	1,7	0,18
A3	0,59	0,88	0,10
B1*	1,2	0,4	0,041
B2*	0,83	1,2	0,15
B3	0,80	1	0,11
C1	0,81	1,3	0,26
C2	0,26	2,2	0,45
C3	1,0	0,49	0,049
D1*	0,25	2,5	0,51
D2	0,37	1,6	0,18
D3	0,39	0,98	0,16
E1*	0,35	0,73	0,11
E2	0,35	0,67	0,091
E3	0,32	0,43	0,053

F.2. Calculations of individual concentration ratios of ^{137}Cs and Sr

Calculation of individual concentration ratios (CR) (wet/dry weight) and standard deviations (SD) of ^{137}Cesium (cow`s milk and mutton) and strontium are shown below (*Table 36-Table 38*). Values are in arithmetic mean.

Table 36. Values used for calculation of individual ^{137}cesium CRs (cow`s milk). For calculation of SD was Equation 13 (chapter 2.11.2) used.

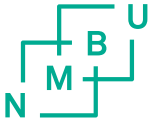
Site	Grass	SD	Milk	SD	CR	SD	RSD (%)
B1	3,5	3,5E+00	5,0E-01	3,0E-02	1,4E-01	1,4E-01	100
C1-2	4,0	3,2E+00	3,7E-02	9,7E-03	9,4E-03	8,0E-03	85
E1-2	2,3	7,1E-03	1,1E-01	2,9E-02	4,8E-02	1,3E-02	26

Table 37. Values used for calculation of individual ^{137}Cs CRs (mutton). Values used for CRs is in Appendix A Table 9 and Table 10.

Site	CR	SD	RSD (%)
B2	0,36		
B2	0,38	0,012	3,3
C1-2	0,11		
C1-2	0,16		
C1-2	0,36		
C1-2	0,30		
C1-2	0,16	0,10	48

Table 38. Values used for calculation of individual strontium CRs (cow`s milk). For calculation of SD was Equation 13 (chapter 2.11.2) used.

Site	Fodder	SD	Milk	SD	CR	SD	RSD (%)
B1	6,6E+01	1,8	4,9E-01	6,8E-03	7,5E-03	2,3E-04	3,0
C1-2	5,7E+01	16	4,9E-01	6,8E-03	8,6E-03	2,4E-03	28
E1-2	4,4E+01	8,2	3,4E-01	4,8E-03	7,8E-03	1,5E-03	19



Norges miljø- og biovitenskapelige universitet
Noregs miljø- og biovitskapelege universitet
Norwegian University of Life Sciences

Postboks 5003
NO-1432 Ås
Norway