Norwegian University
of Life Sciences

Master's Thesis 2018 60 ECTS
Faculty of Environmental Science and Natural Resource Management

The exposure of terrestrial biota to naturally occurring radiation and stable elements: Case Orrefjell, a risk assessment

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Radioecology

Acknowledgements

This study was part "Case Orrefjell" project supported by the Flagship hazardous substances at the FRAM High North Research Centre for Climate and the Environment, Tromsø, Norway and was performed with collaboration with the Norwegian Radiation Protection Authority (NRPA). This thesis is submitted in partial fulfillment of the requirement for the award of the degree of Master of Science in Radioecology at the Norwegian University of Life Sciences (NMBU), at the Centre for Environmental Radioactivity (CERAD).

I would like to thank my supervisor Prof. Lindis Skipperud (NMBU/CERAD) and my co-supervisors Frøydis Meen Wærsted and Louise Kiel Jensen for allowing me to be part of the "Case Orrefjell" project, valuable suggestions, advice, encouragement and support which has greatly influenced the success of this work. I am greatly thankful to Karl Andreas Jensen, Marit Pettersen and Susanne Birkeland for the assistance and guidance in the laboratory particularly in sample preparation for ICP-MS. Special thanks goes to Louise Kiel Jensen for allowing me to work at the NRPA laboratory (Fram centre, Tromsø) and doing the radiometric analysis, Frøydis for doing the ICP-MS, Håvard Thørring for providing me with the Orrefjell database to use on ERICA tool and the entire team at Isotope lab.

I am greatly thankful to my family for their immeasurable support – to my wife, thank you very much for being part of this journey, your moral, social and spiritual support, not forgetting your constant encouragement has kept me going.

Finally, I greatly thank the almighty God to have enabled me to come this far. Blessing me with good health and helping me to overcome every challenge that I encountered.

Ås, 15/05/2018

Maina, Peter Kariuki

Summary

Naturally occurring radioactive materials (NORM) can be found all over the world. Investigations of possible effects on humans and the environment have mostly focused on exploited sites like mines and their vicinity, while less attention has been given to undisturbed sites despite the potential risk. The Orrefjell area in Northern Norway has one the largest uranium deposits in Norway, and elevated levels of radionuclides in soil and biota can be expected.

This master thesis is part of the project "Case Orrefjell" supported by the flagship hazardous substance at the FRAM High North Research Centre for Climate and Environment, Tromsø, Norway. The aim of this thesis was to examine the risk and possible impact of enhanced levels of NORMs and other selected stable elements in an undisturbed area on non-human biota.

Fieldworks were carried out in September 2016 and September 2017 by a collaboration of scientist from Norwegian radiation protection authority (NRPA), Norwegian University of Life Sciences (NMBU), Norwegian Geological Survey (NGU) and Northern Research Institute (NORUT). A total of 13 sampling station were chosen for the fieldworks carried out in September 2017 and September 2018. The sites were pooled into three groups; Orrefjell high altitude, Orrefjell control and Orrefjell cultivated grassland. The samples collected on this fieldwork included soil, plants (berries and berry leaves, grass and herbs), and earthworms. Sampled were analyzed for radionuclides, ^{238}U, ^{232}Th, ^{226}Ra, ^{210}Pb, ^{210}Po and ^{137}Cs, and selected stable elements As, Cd, Cr, Cu, Ni, Pb, and Zn.

Analysis on soil activity concentration showed elevated levels of radionuclides associated with ^{238}U series. The radionuclides were unevenly distributed among the sites and ^{226}Ra was the most dominant with concentrations ranging from 226 – 6800 Bq/kg dw. The average activity concentration of ^{238}U and ^{226}Ra were above the world average of 33 and 32 Bq/kg, respectively, in both the high-altitude area and the cultivated grassland area, while the average activity concentrations for ^{232}Th was lower than the world average of 45 Bq/kg in all the sites.

The activity concentration of radionuclides in plants were generally lower than in the soils and varied among species and plant parts. However, notable high levels of ^{226}Ra (5770 Bq/kg dw) were measured blueberry (*Vaccinium myrtillus*) leaves at station 10. Soil-to-plants transfer factor were found to be in close agreement with soil to plants transfer factors published by IAEA.

ERICA Tool was used to estimate potential doses from measured radionuclides to non-human biota. The measured activity concentrations of radionuclides in soil together with the tool default transfer parameters were used as input in the initial assessment. A second assessment was run using soil activity concentration and site-specific concentration ratio. Based on the initial assessment on all reference organisms using soil activity concentrations, highest doses were estimated for lichen and bryophytes (813 μGy/h), shrub (325 μGy/h) and grass & herbs (186 μGy/h). Internal exposure to ^{226}Ra-226 was shown to be the major contributor to the total dose rate (83% - 98%). Total dose rate calculated using site-specific activity concentrations from selected vegetation were in agreement with default total dose rates for shrub (364 μGy/h) but lower for grass & herbs (15 μGy/h).

Concentrations of selected stable element, As, Cd, Cr, Cu, Ni, Pb, and Zn in soil showed that the soil at Orrefjell were generally below the Norwegian and European limits for non-polluted soil. However, soil sample from station 11 had elevated Pb concentration and is classified to have moderate soil quality. Maximum concentrations of uranium measured at station 11 (160 mg/kg) and 10 (110 mg/kg) were below 250 mgU/kg predicted no effect concentration (PNEC) for terrestrial plant but slightly above 100 mgU/kg PNEC for soil biota.

Radiation from 238U-related radionuclides has been shown as the main concern for detrimental effects on biota in the Orrefjell. However, a multi-stressor scenario of radiation and chemical toxicity cannot be ruled out in stations where levels were higher than the screening values.

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1. Introduction

Sources of ionizing radiation in the environment can be anthropogenic or natural. Natural radionuclides are the largest contributor to external radiation of the world population (UNSCEAR, 2000). Natural sources are divided into two: cosmic radiation and terrestrial radiation. Cosmic radiation is a result of high energy cosmic rays incident to the earth atmosphere and therefore present everywhere in the environment (UNSCEAR, 2008b).

Natural Occurring Radioactive Materials (NORMs) are defined by EPA (2006), as "Materials which may contain any of the primordial radionuclides or radioactive elements as they occur in nature, such as radium, uranium and thorium and their radioactive products otherwise referred to as daughters, that are undisturbed as a result of human activities." NORM in the environment mainly consists of daughters in the decay series of actinium, uranium, and thorium. The uranium series originates from uranium-238 (^{238}U), the thorium series from thorium-232 (^{232}Th) (Figure 1.1) and actinium series from uranium-235 (^{235}U). The natural percentage abundance of ^{238}U is 99.28% and ^{235}U is 0.71%, therefore the contribution of ^{235}U in the environment is small. There are other singly occurring radionuclides in the environment such as ^{40}K, ^{50}V, ^{87}Rb, ^{115}I, ^{123}Te, ^{138}La, and ^{196}Lu, with ^{40}K being the most abundant.

NORMs have always been present in the earth's crust and are concentrated in some places such as uranium orebodies which may be mined. Investigation of possible effects on human and the environment have mostly focused on exploited sites like mines and their vicinity. Less attention has been given to undisturbed sites despite the potential risk to human and to the environment. One such place is the Orrefjell Mountain located in Salangen valley in Troms County, Northern Norway. The area has one of the largest uranium deposit in Norway and will be subject to this study.

Radionuclides in the environment lead to both external and internal exposure of plants and animals to ionizing radiation. Internal exposure arises following uptake of radionuclide particles via pathways such as ingestion or root uptake and inhalation of radon (such as ^{222}Rn), a member of the ^{238}U series. External exposure arises from gamma irradiation from primordial radionuclides of the earth's crust, such as potassium-40 (^{40}K) and decay chains of uranium (^{238}U, ^{235}U), and thorium (^{232}Th). External exposure depends on various factors including contamination level in the environment, the geometrical

relationship between the radiation source and the organism, organism size, shielding properties of the medium, and the physical properties of the radionuclides present (Balonov et al., 2012).

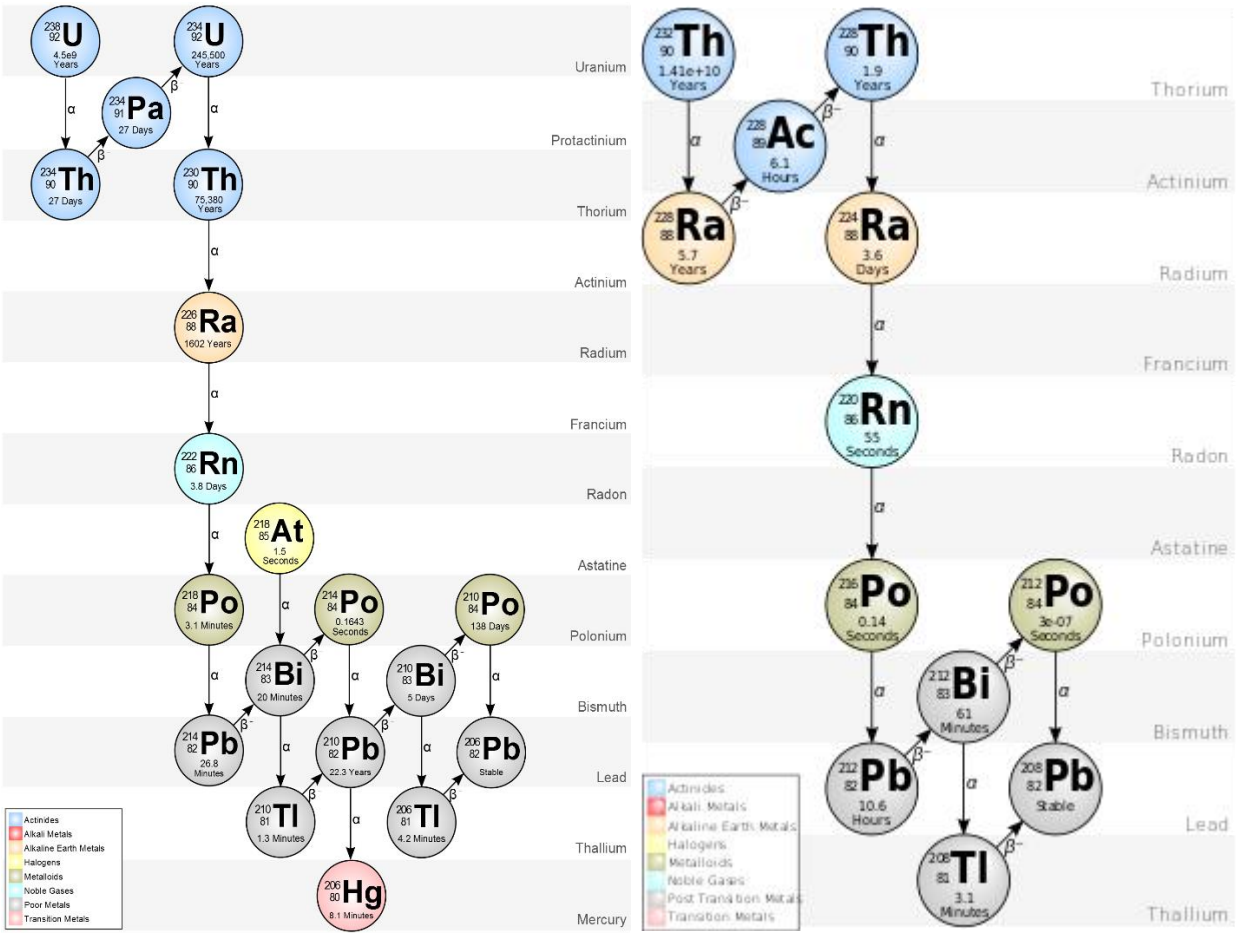

Figure 1.1: ^{238}U and ^{232}Th decay series (Source: Wikipedia).

1.1 Mobility and biological uptake

In terrestrial and aquatic ecosystems, the radionuclides can be transferred from their original site by air emissions for gases such as radon, and particles blown up by wind, leaching, and by running waters such as rivers and streams that pass through such areas, landslides etc. Their mobility and transfer in the ecosystem are controlled by factors such as chemical characteristics of the particular element, the physicochemical properties of soil or water (e.g. pH, organic matter content, competing ions, redox conditions), and to what extent the element is taken up by different types of biota (Chen et al., 2005).

For instance, the transfer of radium (^{226}Ra and ^{228}Ra) to plants is relatively high due to its resemblance to the essential element calcium. Another example includes the radioactive gas ^{222}Rn: the gas is released to the air from soil or bedrock, and as the radon gas decay to metallic elements, the radon daughters (e.g. lead-210 (^{210}Pb) and polonium-210 (^{210}Po)) can be deposited on vegetation surface. Atmospheric deposition have been demonstrated e.g. by the relative high levels of ^{210}Po in lichens. (Skuterud, 2005).

Although both ^{238}U and ^{232}Th are alpha emitters and are characterized as radiotoxic, very low doses are actually received from pure ^{238}U and ^{232}Th due to their long half-life ($t_{1/2}$). Key contribution to dose is associated with the daughter products of ^{238}U and ^{232}Th, especially ^{226}Ra, ^{210}Po and ^{210}Pb produced from ^{222}Rn in ^{238}U series and ^{212}Pb from ^{232}Th series.

The knowledge of the mobility and transfer of radionuclides from source to environmental end-point is vital in any ecological risk assessment. Weathering under different conditions during long periods, together with human activities, leads to mobilization and transport of NORMs through the environment to a variable degree (Popic et al., 2012). Mobilization, ecosystem transfer and impact of radionuclide and trace elements on the surrounding environment are determined mostly by the source which they are released from, by their speciation, binding mechanisms, as well as environmental conditions (Skipperud et al., 2000).

The work looks into the following hypothesis:

Hypothesis 1

H_{01}: We may assume secular equilibrium in the transfer of U and the daughter radionuclide to biota.

H_{a1}: The transfer of U and the daughter radionuclides to biota will differ and not show secular equilibrium.

Hypothesis 2

H_{02}: Since Orrefjell is an undisturbed site, we do not expect doses received from naturally occurring radionuclides to be potentially harmful to biota.

H_{a2}: Even in undisturbed sites, doses received from naturally occurring radionuclides may potentially be harmful to biota.

1.2. Study aims

The main objective of this study was to perform environmental risk assessment from both radionuclides and stable elements at the Orrefjell area. To do this, the following was performed:

1) Analysis of radionuclide concentrations in soil and biota samples collected from an undisturbed NORM rich area (Orrefjell),
2) Estimate of the doses received by non-human biota from selected radionuclides using ERICA Tool.
3) Comparison of the doses and risk quotients estimated by ERICA Tool using default and site-specific parameters.
4) Estimation of the adverse effects on non-human biota from selected stable using Norwegian and European norm values.

2. Environmental impact and risk assessment

In the past, radiation dose limits focused exclusively on human health protection. As a result. assessment frameworks for defining radiation doses to humans and predicting the effects of those doses are much more developed than frameworks dealing with effects caused by non-radioactive contaminants (Hinton et al., 2004). However, since the turn of the millennium, the demand for ecological risk assessment has extended to non-human biota. The former adage that if humans are protected from ionizing radiation, all non-human biota are also protected is no longer accepted (Vives i Batlle et al., 2007) and the need for investigating potential radiation risk to non-human biota and ecosystem is now internationally recognized (IAEA, 2008; ICRP, 2007). In fact current recommendations indicate that environment health and status, including animals and plants should be monitored and preserved from effects of ionizing radiation (Valentin, 2007; Valentin, 2003). In many countries assessment of the risk of ionizing radiation to the environment is now a legislative requirement (Copplestone, 2012).

In contrast to anthropogenic radioactivity which has the distinction of being non-origin in the environment so that there is no question when one should start to investigate it, natural radionuclides are present everywhere in the environment. This necessitates the need to indicate causes of concern and define clear terms for describing the risk scenario at the very beginning of the investigation. In most cases the main concern in the occurrence of NORMs is how severe the radiation exposure is, or when the derived risk is significant from a radiation point of view.

Naturally occurring radioactive materials do not occur alone in the environment and in many cases, they occur together with anthropogenic radionuclides, stable harmful elements or organic pollutants, causing a multiple stressor situation to the biota. Furthermore, some of the heavier long-lived radionuclides, like uranium, poses both a chemical and a radiological risk to living organisms (Ribera et al., 1996; Sheppard et al., 2005).

2.1 The ERICA Integrated Approach and the ERICA Tool

Assessment of risk of radiation contaminants to non-human biota has been made easier by the development of risk assessment tools and models (e.g., RESRAD-BIOTA, ERICA) (Beresford et al., 2008; Brown et al., 2008).

The ERICA Integrated Approach (Environmental Risks from Ionizing Contaminants: Assessment and Management) was created as result of an EU project "ERICA". This project aimed to develop an integrated approach design to assess the effects of radioactive contaminants on the environment and to ensure that decisions on environmental issues give appropriate weight to the environmental exposure, effects and risk from ionizing radiation with emphasis on ensuring the structure and the function of ecosystems (Beresford et al., 2007). The ERICA Tool can be used to estimate doses to organisms (using selected representative reference organisms), either for screening purposes or identifying the most exposed organisms and to gain insight on the most ecologically relevant radionuclides (Oughton et al., 2013). The main elements in the ERICA Integrated Approach as described by Beresford et al. (2007) and Larsson (2008) are:

- Assessment, where activity concentrations in biota and environmental media are used to estimate radiation doses to the selected reference organisms. The assessment is possible to perform in three defined tiers, depending on the levels of concern and regulatory demand.
- Risk characterization, where results of the assessment are evaluated to estimate probable adverse effects on biota.
- Management, where the pre- and post-assessment decisions are made.

The ERICA Tool is a computerized flexible software system for assessing the radiological risk to biota and supports the ERICA Integrated Approach. It is one of the most comprehensive assessment methods available for evaluation of environmental risks of ionizing radiation, and has been recognized by international organizations as International Commission on Radiological Protection (ICRP), International Atomic Energy Agency (IAEA), and has been tested in international comparison exercises e.g., IAEA EMRAS 1 & 2 programs, (Beresford et al., 2008). ERICA guides the user through the assessment process, keeps records and performs the necessary calculations to estimate exposure dose rate of selected reference organisms (Brown et al., 2008) in either freshwater, marine or terrestrial environments.

The tool is based on a tiered approach and assesses the doses and impact to a series of reference organisms that can be adapted to either generic or site-specific assessments (Brown et al., 2008). As mentioned the tool has three different tiers. Tier 1 is a simple screening assessment where radionuclide

concentrations in environmental media are compared against Environmental Media Concentration Limits (EMCL) with results given as Risk Quotient (RQ):

$$RQ = \sum\left(\frac{M_n}{EMCL_n}\right)$$

Where: RQ = Risk quotient; M_n = measured or predicted maximal activity concentration for radionuclide "n" in the medium in Bq L^{-1} for water, Bq kg^{-1} (dry weight) for soil or sediment or Bq m^{-3} for air; $EMCL_n$= Environmental Media Concentration Limit for radionuclide "n" (same units as media).

The EMCLs are calculated from radionuclide media concentration giving rise to the screening dose which has a default value of 10 μGy h^{-1}. Tier 1 is very conservative, but provides a useful tool for quick assessment to see if further risk assessment (Tier 2) is required. In Tier 2, calculated whole-body doses for individual reference organisms are compared directly with the screening dose rate. The set of reference organisms is intended to represent a range of typical organisms present in freshwater, terrestrial or marine environment. It is also possible to calculate doses and identify the potentially most exposed reference organisms with the users being able to provide their own concentration ratios (CR) and distribution coefficient (K_d) values, where;

$$CR = \frac{Activity\ concentration\ in\ biota\ whole\ body}{Activity\ conentration\ in\ medium}$$

$$K_d = \frac{Activity\ concentration\ in\ sediments}{Activity\ concentration\ in\ water}$$

Where activity concentration is given as Bq/kg dw

The user can also create site-specific organisms and obtain information about expected effects at the calculated dose rates, including an overview of the availability of data for the reference organism of interest in the ERICA library.

Tier 3 allows for the input of site-specific probability distribution functions for the different input data and parameters, thus permits the assessment to be run probabilistically, and provides a quantification of uncertainty in the final dose-rate results.

2.2. Assessment of risk from stable elements

The assessment of ecological risk from stable elements is more developed than for radionuclides. However, there is still a large variation in environmental quality standards (EQS) between countries and assessment context (e.g., screening levels, probable effect levels, serious effect levels, etc.) (Oughton et al., 2013). The starting point of an environmental quality standard is the knowledge of what the human beings and natural environment can withstand. EQS have been derived in many different and range from screening values or predicted no-effect concentrations (PNEC), to maximum permitted concentrations. PNEC represents concentration at which no-effects on environmental biota are to be expected while the maximum permitted represents concentrations associated with significant ecological effects. For the standard of soil quality, these levels are determined by extrapolation from results of toxicity tests for a limited number of species to different chemical substances. The range between the two sets can be large and different countries uses different terminologies and different criteria. In Norway, the Norwegian authorities has set guideline for level of unpolluted soil based on known chemical substances (SFT, 2009) (Table 3.4). In regard to pollution of stable elements, using this guideline it is possible to rank the soil quality based on the concentrations of As, Cd, Cr, Cu, Ni, Pb, and Zn. In addition, the guidelines go further to classify the soils as very good to very bad using traffic light-like color coding for this classification (see Table 3.5).

3. Materials and methods.

3.1 Study area - Orrefjell Area

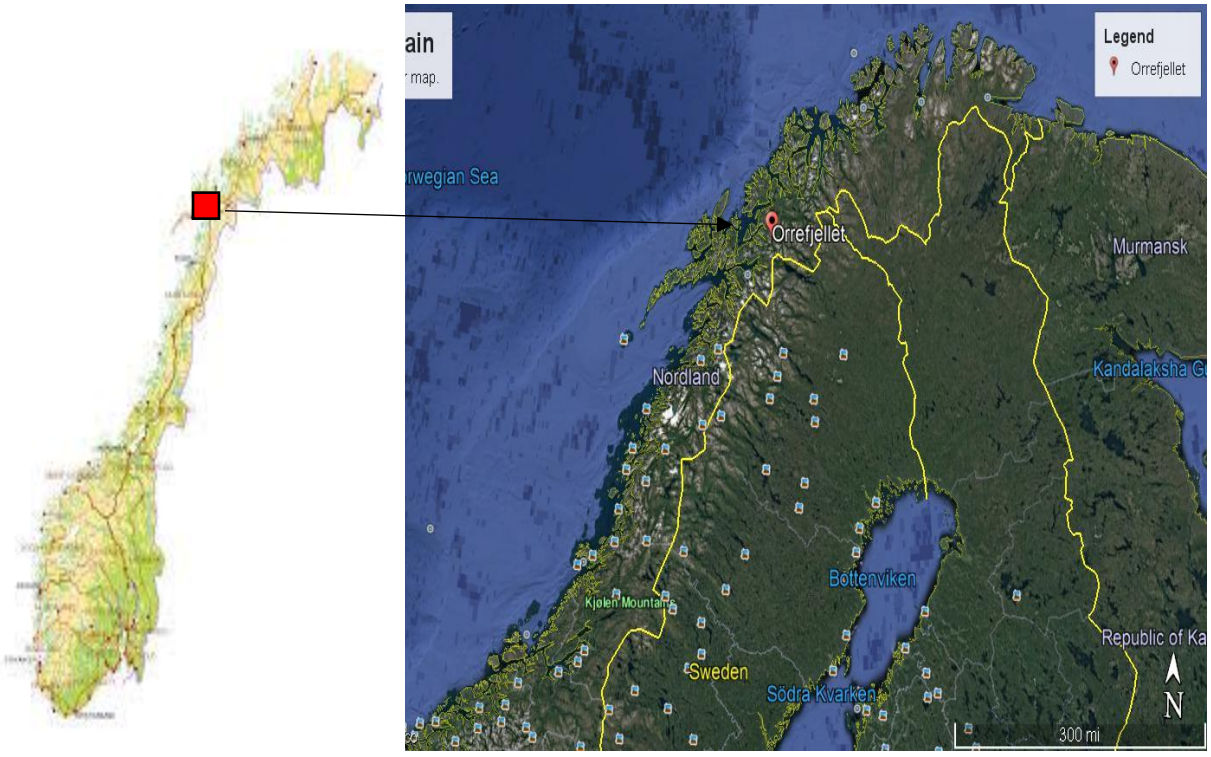

Figure 3.1: Map of Norway to the right indicating the location of Orrefjell mountains (Source: Google map).

Orrefjell is a mountain on the northern side of the Salangen valley (68.89337N, 18.10150E), in Troms County, Northern Norway (Figure 3.1). The area is known to have one of the largest uranium deposits in Norway and due to this, elevated background radiation has been measured in the area. The uranium deposits are related to pegmatite intrusions in Precambrian basement gneisses and were first discovered in the late 50s by Henry Lund from Salangen and documented by Geological Survey Of Norway (NGU) in the 60s, 70s and early 80s (Rindstad, 1981). Scandinavian Highland Holding A/S holds the exploitation rights for Orrefjell. However, it is not known whether there are any plans of exploring the deposits further with the aim of uranium extraction.

While geological properties of the area have been mapped, biotic properties have not been analyzed. Being a NORM rich area with elevated background radiation, the external doses experienced by biota is expected to be substantially higher than the average background radiation in other areas.

The Orrefjell area is used for recreational activities such as hiking and camping and as pastureland for animals with sheep being released for free grazing during summer. One reindeer herd also occasionally visit the area. Human habitation is present within the area with about 15 cabins sited in the mountain area. Agricultural activities are also present in the catchment area south of Orrefjell with developed farms and family houses.

3.2 Sampling Stations

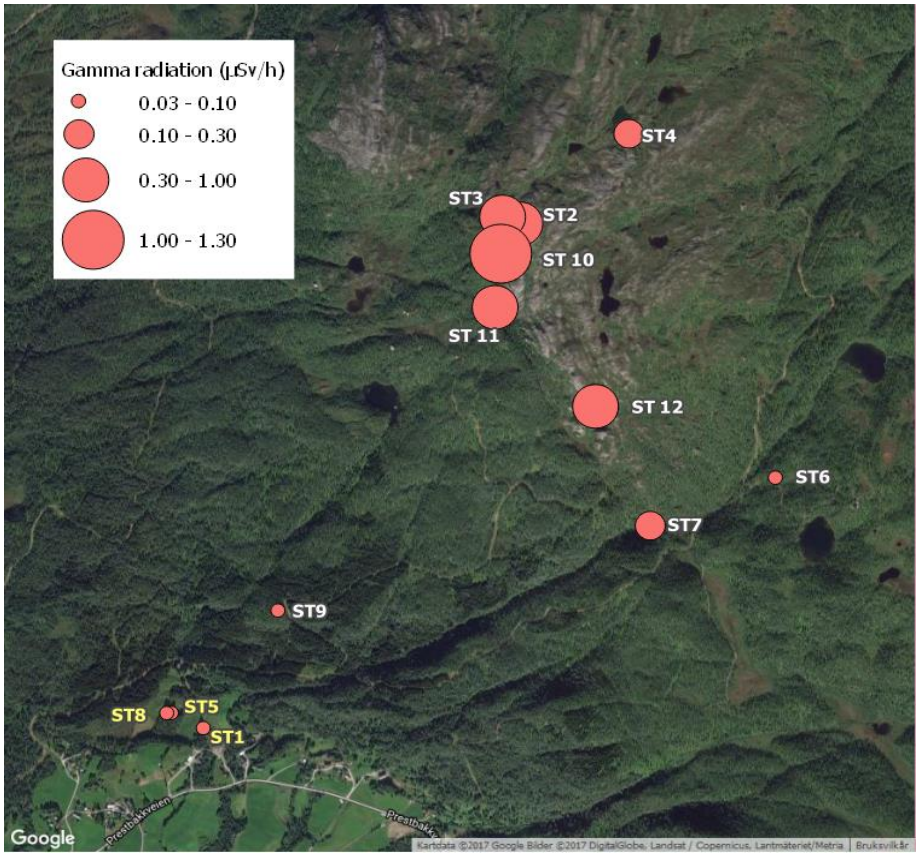

Figure 3.2: Map showing Orrefjell area sampling stations. Stations in yellow represents the cultivated grassland sampled for soil and in white, stations sampled for soil and plants. The size of the circles relates to the background radiation measured at each site (Map by Mari Komperød, NRPA).

The input data for this assessment was the result of analysis of samples collected during fieldwork done in September 2016 and September 2017 in Orrefjell by a collaboration of scientist from Norwegian radiation protection authority (NRPA), Norwegian University of Life Sciences (NMBU), Norwegian Geological Survey (NGU) and Northern Research Institute (NORUT) in the project "Case Orrefjell". The main aim of the project was to study a broad spectrum of the implications of living in an area with elevated levels of naturally occurring radionuclides. The author of this work participated in the fieldwork done in 2017 and prepared all the samples for analysis from this fieldwork. The data on soil and biota activity concentration from both 2016 and 2017 are to be published elsewhere (Thørring et al., in prep), but the data are made available for use in running ERICA Tool.

A total of 10 sampling sites (Stations 1, 2, 3, 5, 5, 6, 7, 7b, 8, and 9) were selected in the field work of 2016 with an additional 3 sites (Stations 10, 11 and 12) and a revisit of stations 2 and 7 selected in 2017. The selection was informed by the geological properties of the Orrefjell area which has been thoroughly mapped by the Norwegian Geological survey (NGU) together with onsite background measurement conducted by the RT-30 Super Ident (Identifier) from Georadis. The instrument was held 1 meter above the ground to measure the ambient dose rate in μSv/h as an average over one minute.

For this study, the sampling stations were divided into three groups as follows:

- Orrefjell mountain area – this covers the high-altitude area with high background and consist stations 2, 3, 4, 7, 7b, 10, 11 and 12
- Orrefjell cultivated grassland area – this covers the low-altitude cultivated area with low background and consist station 1, 5 and 8
- Orrefjell control – this covers the low-altitude area of the mountain with low background and consist stations 6 and 9

The sampling sites coordinates and background measurement are shown in table 3.1.

Table 3.1: Sampling sites coordinates and background radiation in nSv/h

Area	Station	Sampled	Coordinates(DMM)	Background (nSv/h)
Orrefjell mountain area				
	ST2	Sep. 2016	68 53.5849N, 18 05.8442E	580
	ST2*	Sep. 2017	68 53.5849N, 18 05.8442E	576
	ST3	Sep. 2016	68 53.5979N, 18 05.7581E	519
	ST4	Sep. 2016	68 53.7537N, 18 06.3860E	195
	ST7	Sep. 2016	68 53.0193N, 18 06.4920E	124
	ST7*	Sep. 2017	68 53.0193N, 18 06.4920E	287
	ST 10	Sep. 2017	68 53.5290N, 18 05.7494E	1300
	ST 11	Sep. 2017	68 53.4285N, 18 05.7211E	360
	ST 12	Sep. 2017	68 53.2429N, 18 06.220E	974
Orrefjell cultivated area				
	ST1	Sep. 2016	68 52.6399N, 18 04.2712E	60
	ST5	Sep. 2016	68 52.6683N, 18 04.1127E	32
	ST8	Sep. 2016	68 52.6681N, 18 04.0895E	42
Orrefjell Control				
	ST6	Sep. 2016	68 53.1093N, 18 07.1135E	63
	ST9	Sep. 2016	68 52.8604N, 18 04.6420E	51

3.3 Sampling

Figure 3.3: Soil sampling to the left, collected earthworm at the center and blue berry plant (Vaccinium myrtillus).

Soil, plants, and earthworms were identified for sampling (Figure 3.3). The choice of the three types of samples was informed by several reasons: the soil would provide an overview of the activity concentrations of the main naturally occurring radioactive materials in the area and information about site-specific soil characteristics including elemental composition, pH, organic carbon, and dry matter.

The plant that were sampled, which included grass, mushrooms, berries and berry leaves are used as forage for animals and the berries are also consumed by humans. These are included to study transfer from soil to biota and possibly further in the food chain. A detailed list of plant species sampled is shown in Table 3.2. Earthworms are often used as proxy for transfer from soil to soil dwelling organisms. Earthworms are also an important food source for a variety of other animals and thus radionuclides and stable elements accumulated in earthworms may further propagate the ecosystem.

3.3.1 Soil Samples

Soil samples were collected from all the sampling stations by taking the top layer (0 – 3) and transported in zip lock bags to the NRPA lab in Tromsø. A portion of the soil was dried at room temperature and kept aside for determination of soil characteristics including pH, loss of ignition and organic carbon. The pH was measured using 1:2 ratio soil: water method (Kalra, 1995), while organic carbon was estimated from loss on ignition after burning at 550℃ overnight (Krogstad, 1987). The rest of the soil samples were dried at 105℃, homogenized and cleaned from roots and stones by sieving through a 2mm sieve. For radium analysis, the samples were packed in suitable geometries and vacuum packed in aluminum foil. The samples were stored for one month to achieve secular equilibrium between ^{226}Ra and its daughter products. The samples were analyzed on an High-Purity Germanium detector (Canberra) and the activity concentration of ^{226}Ra was calculated as the weighted average of the 295 and the 352keV peak for ^{214}Pb and the 609 keV ^{214}Bi peaks. (Mauring et al., 2014). The activity concentration of 137Cs and 40K was given by the 661.2 and 1460.0 keV peaks, respectively. The analysis was done at NRPA Lab in Tromsø by Louise Kiel Jensen.

For stable elements, analysis was conducted by inductively coupled plasma mass spectroscopy (ICP-MS). The samples were prepared by weighing about 0.25 g in two batches of three replicates of each sample into Teflon vials, followed by addition of 0.1 ml 10mg/L Rh solution as internal standard to each sample. The samples were then subjected to 40 min microwave-assisted acid digestion at 260 ℃ in a Milestone Ultra-Clave using 5ml high-grade purity acid HNO_3 on one batch while adding 5 ml

pure HNO_3 and 1ml HF to the other batch. After the digestion, the samples were transferred to 50 ml vials and diluted with Type 1 water to 50ml. NIST 2709a San Joaquin soil, NCS ZC 73007 soil, and NCS DC 73325 soil were used as certified reference materials for elemental concentration in soil. The ICP-MS analysis was carried out by Frøydis Meen Wærsted. The ICP-MS results were given in mg/kg. For uranium and thorium concentration, natural abundance was assumed and the results from ICP-MS analysis were converted to specific activity by multiplying by specific activities of 12.35 Bq ^{238}U/mgU and 4.06 Bq ^{232}Th/mgTh, respectively.

3.3.2 Plant samples

Plant samples were collected from different stations as shown in Table 3.2; they were dried at 105 ℃ and homogenized by milling. Each sample was divided into two batches for radiometric analysis and ICP-MS. The same procedure was used for both radiometric analysis and ICP-MS as used for soil. The only difference being that for digestion before ICP-MS only HNO_3 was used and the Rh internal standard was 10 times diluted compared to the one used for soil. NCS 7C 73014 (tea) and NIST 1575 (pine needles) were used as certified reference materials.

For ^{210}Po, plant samples were analyzed by alpha spectrometry (Canberra Alpha Analyst and Genie-2000) using a slightly modified version of the method described by Flynn (1968). Initially, the samples were dissolved in *aqua regia* and then evaporated at 150 ℃ to almost dryness. Samples were then dissolved in 9 M HCl and a small ascorbic acid and filtered to a deposition cell. Finally, polonium was auto-deposited on to polished silver disk from 5 M HCl at 65 ℃ for 3 hours. Po-209 was used as a yield determinant. After auto-deposition residual polonium was removed by ion exchange. New ^{209}Po tracer was added and the sample stored for 6 months to estimate ^{210}Pb from the ingrowth of its daughter ^{210}Po before it was analyzed again. This analysis was performed by Håvard Thørring at NRPA Lab.

Table 3.2. Plant samples with their sampling stations (sampling done in 2016 and 2017).

Norwegian name	Latin name	English name	Sampling Station
Blokkebær	*Vaccinium uliginosum*	Bog bilberry	
Blokkebærlyng		Bog bilberry leaves	2, 3, 4
Blåbær	*Vaccinium myrtillus*	Blueberry	2, 2*, 3, 4, 6, 7, 9, 10*, 11*, 12*
Blåbærlyng		Blueberry leaves	2, 2*, 3, 4, 6, 7, 9, 10*, 11*, 12*
Krekling	*Empetrum nigrum*	Black crowberry	2
Kreklinglyng		Black crowberry leaves	2, 2*, 4, 7, 9
Skogstorknebb	*Geranium sylvaticum*	Cranesbills.	6, 7, 11*, 12*
Smyle	*Deschampsia flexuosa*	Hair-grass	2, 3, 4, 6, 7, 9
Tyttebær	*Vaccinium vitis-idaea*	Lingonberry	7
Lys reinlav	*Cladonia arbuscula*	Reindeer Lichen	7
Kantarell	*Cantharellus cibarius*	Chanterelle (wild mushroom)	6
Rimsopp	*Cortinarius caperatus*	Gypsy mushroom	3

* -Sampled in 2017.

3.3.2.1 Soil to plants Transfer Factor.

Radionuclides and stable elements in soil follow complex dynamics in which part of its concentration is transported into the soil solution, while another part gradually becomes strongly bound to the particles of the soil. The portion in the soil solution can be incorporated into the plants via the roots. In some cases, this is facilitated by their chemical similarity with other elements essential for plant growth (Manigandan & Manikandan, 2008), as mentioned for ^{226}Ra. To quantify the accumulation of trace elements by plants or transfer of elements from soil to plants through the roots, soil-to-plants transfer factors were used. Transfer factor (TF) is defined as the ratio of specific activity in plant parts and soil and describes the amount of element expected to enter a plant from its substrate under equilibrium conditions (Sheppard & Sheppard, 1985).

From the observed activity concentration of the selected radionuclides in plants and in the corresponding soil, the TF values were calculated according to the equation:

$TF = C_p/C_s$

Where C_p is the concentration of the elements in plants (Bq/kg dw, for radionuclides, mg/kg d.w for stable elements) and C_s is the concentration of elements in soil (Bq/kg dw, for radionuclides, mg/kg d.w for stable elements).

3.3.3 Earthworms

Earthworms were collected from station 1and 5 (2016), 7b (2016, 2017), 8 (2016) and 11 (2017). They were transported to laboratory in plastic boxes with moist soil and perforated lids. However, earthworms sampled in station 1 (2016) and station 7 (2017) escaped while being depurated and therefore their data is not available. The earthworms sampled in 2016 were identified alive by Emmanuel Lapied (NMBU) as *Lumbricus rubellus* while those sampled in 2017 were identified as *Aporrectodea caliginosa* and *Aporrectodea rosea* by visual inspection of their photos as it was not possible to transport them alive from Tromsø to Ås.

The earthworms were depurated on a moist filter paper for 24 hrs to allow them to empty their guts with constant rinsing with distilled water to remove visible soil. They were euthanized by freezing them at -18 ℃. They were later freeze-dried before being prepared for microwave assisted acid digestion. Since their weight was very low, 2.5 ml HNO_3 was used with 0.1ml 4 µg/L of Rh solution used as internal standard and dilution with Type 1 water done to 25ml.

Radium analysis was not performed on the earthworm samples, as the quantity was too small.

3.4 Exposure dose rate

The ERICA tool was used to calculate the radiological dose for biota and to check whether the screening value was exceeded. An initial screening was done on Tier 1 where soil activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra, ^{210}Pb, ^{210}Po (assumed to be in equilibrium with ^{210}Pb in soil) and ^{137}Cs measured were used in Bq/kg dw dry weight. For ^{238}U & ^{232}Th, the ICP-MS results (given in mg/kg) were used after converting them to specific activity assuming natural abundance. This was

done by using specific activities of 12.35 Bq ^{238}U/mgU and 4.06 Bq ^{232}Th/mgTh. A default set of reference organisms given in the ERICA Tool for terrestrial ecosystem was used. The screening included amphibians, annelid, arthropod – detritivorous, bird, flying insects, grasses and herbs, lichen and bryophytes, mammal – large, mammal – small-burrowing, mollusc – gastropod, reptile, shrub, and tree. The ERICA Tool default dose rate screening value of 10 μGy/h, which represent a generic predicted no-effect dose rate for all organisms, was kept.

Since results from Tier 1 for all the stations showed the risk quotient (RQ) for at-least one organism being greater than 1 for all the sites, the decision to proceed to Tier 2 was taken. As mentioned the tool determines the risk quotient (RQ) by comparing the input media concentrations with the most restrictive EMCL for each radionuclide. In Tier 2, assessment was done on the three areas of interest, Orrefjell high-altitude, Orrefjell cultivated grassland and Orrefjell control. In the first run on Tier 2, the input data included activity concentration measured in the soil and default parameters in the ERICA Tool. All the available reference organisms for terrestrial ecosystem were included in the assessment. Due to the heterogeneity of the radionuclide distribution between the sites the assessment was run on both mean and maximum measured soil activity concentration. The model was run with uncertainty factor (UF) of 1 since the aim of the study was to estimate the probable dose to the exposed organism and not to identify the possibility of doses exceeding the pre-set screening value. Default weighting radiation factors i.e. 10 for alpha emitters and 3 for low-level beta emitters, were retained together with default occupancy factors for the organisms. Default parameters of concentration ratios (CRs) for organisms and radionuclides were used. These concentration ratios are based on reviews of available experimental data and are available in the ERICA library. When data is not available, the tool gives a number of options for filling the data gaps such as similar reference organism, from published review, highest animal value, highest plant value, combined methods etc. In the current study, concentration ratio experimental data for Amphibians (for Ra, Th, U), Annelid (Ra), Anthropod – detritivorous (Th), Flying insects (Th), Mollusc – gastropod (Th, U) and reptile (Ra) were missing. Combined method and similar reference organism methods were used when deriving ERICA default concentration ratio values for the missing data as shown in Table 3.3.

Table 3.3. Methods used to derive ERICA Tool default ratio values.

Ref. organism / Radionuclide	Amphibians	Annelid	Arthropod-detritivorous	Flying insects	Mollusc - gastropod	Reptile
Ra	Similar reference organism	Similar reference organism				Similar reference organism
Th	Similar reference organism		Combined method	Combined method	Similar reference organism	
U	Similar reference organism				Similar reference organism	

A second assessment was run on Tier 2 using the measured activity concentration of soil and biota samples collected at the site. The ERICA tool uses fresh weight for biota activity and thus the biota data was converted to fresh weight using the dry weight/fresh weight ratio. The result of this assessment was used to compare the site-specific biota activity concentrations with those modelled within the ERICA Tool and to compare the default concentration ratios (CRs) with the site-specific. The activity concentration measured for berries and berry plants was used to replace shrub, wavy hair-grass (*D. flexuosa*) to replace grass, and earthworm to replace annelid in the reference organism set.

3.5 Soil screening for stable elements

Soil concentrations of selected trace element from the ICP-MS analysis was used to assess the quality of soil in terms of contamination. These trace elements include As. Cd, Cr, Cu, Ni, Pb, and Zn. The Norwegian Authorities (SFT, 2009) for level of unpolluted soil based on known chemical substances guidelines for level of unpolluted soil in Norway (norm values) and European baseline data (De Vos & Tarvainen, 2005) (Table 3.4) were used as screening levels. The soils were further classified as based on the Norwegian Authorities classification criteria (Table 3.5).

Table 3.4. - Soil screening levels for trace elements (mg/kg).

Data/source	As	Cd	Cr	Cu	Ni	Pb	Zn
Norwegian norm values (SFT, 2009)	8	1.5	50	100	60	60	200
European baseline (De Vos and Tarnvainen,2006)	7	0.14	60	13	18	32	52

Table 3.5. - Norwegian classification of soil quality (mg/kg).

Class	**1**	**2**	**3**	**4**	**5**
	Very good	**Good**	**Moderate**	**Bad**	**Very Bad**
Arsenic	<8	8-20	20-50	50-600	600-1000
Lead	<60	60-100	100-300	300-700	700-2500
Cadmium	<1.5	1.5-10	10-15	15-30	30-1000
Copper	<100	100-200	200-1000	1000-8500	8500-25000
Nickel	<60	60-135	135-200	200-1200	1200-2500
Zink	<200	200-500	500-1000	1000-5000	5000-25000
Cr (III)	<50	50-200	200-500	500-2800	2800-25000

4. Results and Discussion

4.1 Soil Characteristics

Basic soil characteristics are presented as average for the three areas studied, Orrefjell high-altitude, Orrefjell cultivated grassland and Orrefjell control, are shown in Table 4.1. Soil characteristics for individual sampling stations are shown on appendix A. The average pH for soil in Orrefjell high-altitude area (pH 4.4) and Orrefjell control (pH 4.2) fall in the range of extremely acidic soil (pH 3.5-4.4) (Soil Survey Staff, 1993). For the Orrefjell cultivated area the average pH (5.5) is in the range of strongly acidic to moderate acidic. This is within the optimum soil pH range for most plants. However, many plants have adopted to thrive at pH value outside this range. Organic carbon content was higher at Orrefjell high-altitude area and Orrefjell cultivated area but lower (10.7±6.2) at the control sites.

Table 4.1. Basic soil characteristics and main element for the three sampling areas in the Orrefjell area. All element concentrations are given in g/kg dw.

	% LOI	Estimated % Org. C*	pH	Fe	Mg	Ca	K	P	S
Orrefjell high-altitude	58.9±34	34±20	4±0.6	19.1±23.3	6.4±7.2	5.9±2.1	5.9±4.5	1.0±0.3	1.4±0.6
Orrefjell cultivated grassland	43.3±25.2	25.1±14.6	5.5±0.8	22.6±13.2	8.6±4.7	22.5±16.3	3.5±1.5	1.3±0.1	3.0±2.3
Orrefjell control	18.4±10.7	10.7±6.2	4.2±0.1	12.4±6.6	4.2±2.4	3.1±1.7	6.2±1.2	0.6±0.1	0.4±0.3

* Org.C = 58% LOI (Krogstad, 1987)

4.2 Activity concentrations of radionuclides of interest in soil and plants

4.2.1 Soil Activity Concentrations

The activity concentration of radionuclides in soils (Bq/kg dw) for the three areas of interest, Orrefjell high-altitude, Orrefjell cultivated grassland and Orrefjell control are shown in Tables 4.2, 4.3 and 4.4, respectively.

Po-210 was not measured in soil, so secular equilibrium with ^{210}Pb was assumed to get an estimate for ^{210}Po activity concentrations. Secular equilibrium is achieved when a short-lived daughter nuclide

reaches the same activity as a long-lived mother nuclide, i.e. when the disintegration rate of the progeny is the same as the rate of production by the disintegration of the mother nuclide. The $^{210}Po/^{210}Pb$ ratio which has been found to be one is frequently used to infer the activity of one based on measurement of the other (Barescut et al., 2005; Parfenov Yu, 1974; Sheppard et al., 2008).

Results from Orrefjell high-altitude sites showed wide ranges for radionuclides activity concentrations, ^{238}U: 10-2000 Bq/kg dw, ^{232}Th: 0.81 - 29 Bq/kg dw, ^{40}K: 41 - 618 Bq/kg dw, ^{210}Pb: 610-1180 Bq/kg dw, ^{210}Po: 610-1180 Bq/kg dw, and ^{226}Ra: 226 – 6840 Bq/kg dw. The wide range shows that the radionuclides are inhomogeneously distributed also within the high-altitude area. The activity concentration of anthropogenic radionuclide ^{137}Cs which may have been as a result of global fallout in the 50's or from Chernobyl, was somehow evenly distributed in all the sampling sites with a narrow range of 20 - 88 Bq/kg dw. The arithmetic mean concentration for uranium in the soils of the Orrefjell high-altitudes sites (n=9) was 642 Bq/kg dw, this exceeds the world average concentration of 33 Bq/kg dw for ^{238}U (UNSCEAR, 2008a). The average value is below the limit of 1 Bq/g for radioactive waste in Norway (Pollution Control Act, 2010) but the soil samples from stations 10 and 11 registered the highest concentrations of ^{238}U (1358 and 1975 Bq/kg dw, respectively) which are above the limit of 1 Bq/g for radioactive waste in Norway (Pollution Control Act, 2010).

The obtained results show that the mean values of ^{40}K concentration soils samples were slightly lower than the world average 370 Bq/kg dw. However, soil samples from station 7 (sampled in 2017), 10, 11, and 12 (610, 418, 591, and 618 Bq/kg dw, respectively) were above the world average. Soil ^{232}Th activity concentrations were considerably low compared to ^{238}U in all the sampling station and were all below the world average activity concentration of 45 Bq/kg dw for ^{232}Th (UNSCEAR, 2008a) and subsequently below the limit (1 Bq/g) for radioactive waste in Norway in regard to ^{232}Th (Pollution Control Act, 2010). The activity concentrations of ^{232}Th in soil at Orrefjell were in the range of 0.8 to 29 Bq/kg dw, this lower than activity concentrations measured at the Fen Central Complex, a thorium rich area in southern Norway. The 232Th activity concentration at the Fen Central Complex were in the range of 69 – 6581 Bq/kg dw (Popic et al., 2011).

The activity concentration of $^{210}Pb/^{210}Po$ ranged from 75 – 1180 Bq/kg dw and was higher than the activity concentration of ^{226}Ra in all the station it was measured. This would be because deposition of $^{210}Pb/^{210}Po$ produced in the atmosphere from the decay of ^{222}Rn is thought to occur once it becomes

attached to atmospheric particles that deposit through wet and dry deposition. Another reason could be the washing out of ^{226}Ra by rain water since Ra is more soluble in water. Therefore, surface soil can become enriched in ^{210}Pb/^{210}Po relative to the parent ^{226}Ra present in the soil. Lead-210 decays to produce ^{210}Po, and in most cases, these are in secular equilibrium in the soil (Sheppard et al., 2008).

Although Orrefjell area is an undisturbed NORM site, activity concentration of ^{238}U and ^{226}Ra in soil samples of some stations are comparable with activity concentrations of the same radionuclides measured in areas where the activity concentrations have been technologically enhanced by human activities, "technologically enhanced natural occurring radioactive materials (TENORM)" sites. Concentration of ^{238}U in soil samples from station 2 (2017), 4, 7b (2017), 10, 11 and 12 at Orrefjell high-altitude area ranging from 246 – 1975 Bq/kg dw, can be compared with those observed in mining sites in Central Asia. Uranium concentrations in Kazakhstan, Kyrgyzstan and Tajikistan mining sites in Central Asia were in the range of 71 – 1455 Bq/kg dw, 1082 – 5858 Bq/kg dw and 296 – 590 Bq/kg dw, respectively (Skipperud & Salbu, 2011). Concentration of ^{226}Ra in soils samples from station 2, 3, 4, 7, 10, 11 and 12 ranging from 225 – 6840 Bq/kg dw are in comparison with ^{226}Ra concentration in soils at Kazakhstan (114 – 2188 Bq/kg dw) and Kyrgyzstan (1285 – 4990 Bq/kg dw) mining sites in Central Asia (Skipperud & Salbu, 2011). It is worth mentioning that station 10 stand out as the ^{226}Ra activity concentration measured here was almost five times higher than second highest ^{226}Ra concentration site i.e. 6840 Bq/kg dw compared to 1465 Bq/kg dw. A future analysis at the site the stations, (10, 11, and 12) may be important as the soils are above the 1 Bq/g level for radioactive waste in Norway. Such assessment of the stations should include sequential extraction to give more information the mobility of the radionuclides. Today there is no information on bioavailability of the radionuclides and thus no information on to what degree the elevated concentrations of radionuclides in soil are accessible to plants.

Table 4.2. Activity concentration of radionuclides (Bq/kg dw dry weight) in soil at the Orrefjell high-altitude area.

Radio-nuclide	St.2*	St.3	St.4	St.7	St.7b*	St.10**	St.11**	St.12**	Mean	StDev	Min.	Med.	Max.
Cs-137	58	60	43	37	21	88	43	45	50	20	21	44	88
K-40	76	234	43	219	567	418	591	618	346	233	43	326	618
Pb-210	1180	610	1164	1104	nm	nm	nm	nm	1015	272	610	1134	1180
Po-210* **	1180	610	1164	1104	nm	nm	nm	nm	1015	272	610	1134	1180
Ra-226	656	226	490	301	579	6840	1106	1465	1458	2214	226	618	6840
Th-232	1.7	4.5	0.8	2.2	23	29	26	23	14	12	0.8	14	29
U-238	73	30	247	10	802	1358	1,975	642	642	714	10	444	1975

** -Mean values based on 2016 and 2017 fieldwork.*
*** -Sampled in 2017.*
**** Po-210 assumed to be in secular equilibrium with Pb-210(Barescut et al., 2005; Parfenov Yu, 1974; Sheppard et al., 2008).*
nm - Not measured.

In the Orrefjell grassland cultivated area, activity concentrations were 57, 111, and 235 Bq/kg dw of ^{238}U and 22, 20, and 7.7 Bq/kg dw ^{232}Th for stations 1, 5, and 8, respectively, (Table 4.3). The mean activity concentration for 238U was 134 Bq/kg dw, which exceeds the world average activity concentration (33 Bq/kg dw) while the mean activity concentration of ^{232}Th was 17 Bq/kg dw which is below the world average (45 Bq/kg dw) for ^{232}Th. Activity concentrations of ^{137}Cs, ^{210}Pb/^{210}Po, and ^{226}Ra were generally low compared to the high-altitude sites, while high activity concentrations were measured for ^{40}K in some stations in the grassland cultivated area compared to the high-altitude area.

The activity concentration of all the radionuclides measured were similar for the two stations sampled at the Orrefjell control sites despite the stations being on opposite sides of the mountain (figure 3.2). The mean activity concentrations of ^{137}Cs (49 Bq/kg dw), ^{40}K (596 Bq/kg dw), and ^{210}Pb/^{210}Po (181 Bq/kg dw) were however higher than the mean activity concentration in the cultivated grassland sites, while the mean activity concentration of ^{226}Ra (36 Bq/kg dw), ^{232}Th (8.7 Bq/kg dw), and ^{238}U (14 Bq/kg dw) were lower than mean activity concentration for the same radionuclides at Orrefjell cultivated grassland sites. While the mean activity concentration of ^{40}K (596 Bq/kg dw) was above the

world average of 370 Bq/kg dw for ^{40}K and ^{226}Ra (36 Bq/kg dw) slightly higher than the world average of 32 Bq/kg dw for ^{226}Ra, the activity concentration of ^{232}Th (8.7 Bq/kg dw) and ^{238}U (14 Bq/kg dw) were both below the world average of 45 Bq/kg dw and 33 Bq/kg dw, respectively

Table 4.3. Activity concentration of radionuclides (Bq/kg dw dry weight) in soil at the Orrefjell cultivated grassland sites.

Radio-nuclide	St.1	St.5	St.8	Mean	StDev	Minimum	Median	Maximum
Cs-137	23	26	32	27	4.6	23	26	32
K-40	550	351	121	341	214	121	351	550
Pb-210	75	85	165	109	49	75	85	165
Po-210*	75	85	165	109	49	75	85	165
Ra-226	49	37	46	44	6.2	37	46	49
Th-232	22	20	7.7	17	7.7	7.7	20	22
U-238	57	111	235	134	91	57	111	235

* *Po-210 assumed to be at secular equilibrium with Pb-210(Barescut et al., 2005; Parfenov Yu, 1974; Sheppard et al., 2008).*

Table 4.4. Activity concentration of radionuclides (Bq/kg dw dry weight) in soil at the Orrefjell control sites.

Radio-nuclide	St.6	St.9	Mean	StDev	Minimum	Median	Maximum
Cs-137	49	48	49	0.3	48	49	49
K-40	630	561	596	49	561	596	630
Pb-210	117	244	181	90	117	181	244
Po-210*	117	244	181	90	117	181	244
Ra-226	33	38	36	3.1	33	36	38
Th-232	7.3	10	8.7	2.0	7.3	8.7	10.2
U-238	16	12	14	2.6	12	14	16

**Po-210 assumed to be at secular equilibrium with Pb-210(Barescut et al., 2005; Parfenov Yu, 1974; Sheppard et al., 2008)*

The soil activity concentrations presented for all the sites indicates that the assumption that uranium at its natural state is at secular equilibrium with its daughter radionuclides is not true. Activity

concentration of ^{210}Pb at station 7 was 110 times higher than activity concentration of ^{238}U, while activity concentrations of ^{226}Ra was 30 times higher than ^{238}U at the same station. At station 2, activity concentration of ^{210}Pb and ^{226}Ra were 16 and 8 times, respectively higher than activity concentration of ^{238}U.

4.2.2 Plant Activity Concentrations

Uranium concentration in plants are generally several orders of magnitude lower than in soil (Mitchell et al., 2013) and varies between plant species depending on accumulation ability of different plant species (Salbu et al., 2011). At Orrefjell the activity concentration of ^{238}U analyzed in plants species was low, ranging from 0.0037 to 31 Bq/kg dw (dw) (Table 4.5). The maximum activity concentration was measured in cranesbill leaves (*G. sylvaticum*) found in station 11, which was also the station with the highest activity concentration of ^{238}U in the soil. Although station 11 had the highest concentration of ^{238}U, the high concentration of ^{238}U in *G. sylvaticum* does not qualify to conclude that activity concentration uranium increases with activity concentration of soil. Contrasting findings have been reported regarding uptake of uranium from soil. Sheppard et al. (2004) showed that concentration ratios for uranium are not constant with soil concentration, declining as concentration increases. However, this was contrasted by Tracy et al. (1983) who reported a linear relationship between plants and soil concentration in the same geographic area.

There was a strong positive correlation between activity concentrations of ^{238}U in soil and in berry leaves ($r = 0.725$, $p = 0.002$) but a negligible relation between concentration of ^{238}U in soil and the berries ($r = 0.05$) (Figure 4.1). The strong positive correlation agrees with the findings of Tracy et al. (1983) who reported a linear relationship between plants and soil concentration in the same geographic area.

For blueberry, activity concentration of ^{238}U was up to one order of magnitude higher in the leaves than in the berries. Activity concentrations in roots was not analyzed in this study, but several studies have shown that uranium content is high in roots than in other plants parts. As reported on Mitchell et al. (2013), a study using tomato plants growing in a contaminated site found the greatest transfer factors were for roots, then shoots, then for fruits (Kohler et al., 2000). Pereira et al. (2009) also reported low or to no accumulation of uranium in the above-ground parts of some plant species.

Table 4.5. Activity concentration of ^{238}U (Bq/kg dw) in selected vegetation species.

Species (Plants)	**St.2**	**St.2***	**St.3**	**St.4**	**St.6**	**St.7**	**St.9**	**St.10***	**St.11***	**St.12***
Vaccinium myrtillus	1.5	2.7	0.4	3.5	0.01	0.02	0.1	2.6	8.4	0.9
Empetrum nigrum	5.9	0.8		1.5		0.1	0.1			
Vaccinium uliginosum	0.1		0.1	0.6						
Average	**2.5**	**1.7**	**0.2**	**1.8**	**0.01**	**0.1**	**0.1**	**2.6**	**8.4**	**0.9**
Species (Berries)										
Vaccinium myrtillus	0.1	0.1	0.6	0.05	0.1	0.02	0.004	0.5		0.1
Empetrum nigrum		0.1								
Vaccinium vitis-idaea						0.0				
Average	**0.1**	**0.1**	**0.6**	**0.05**	**0.1**	**0.01**	**0.004**	**0.5**		**0.1**
Grass and herbs										
Deschampsia flexuosa	0.2		0.1	0.6	0.1	0.02	0.1			
Geranium sylvaticum					0.1	0.6			31	0.9

** Sampled in 2017*

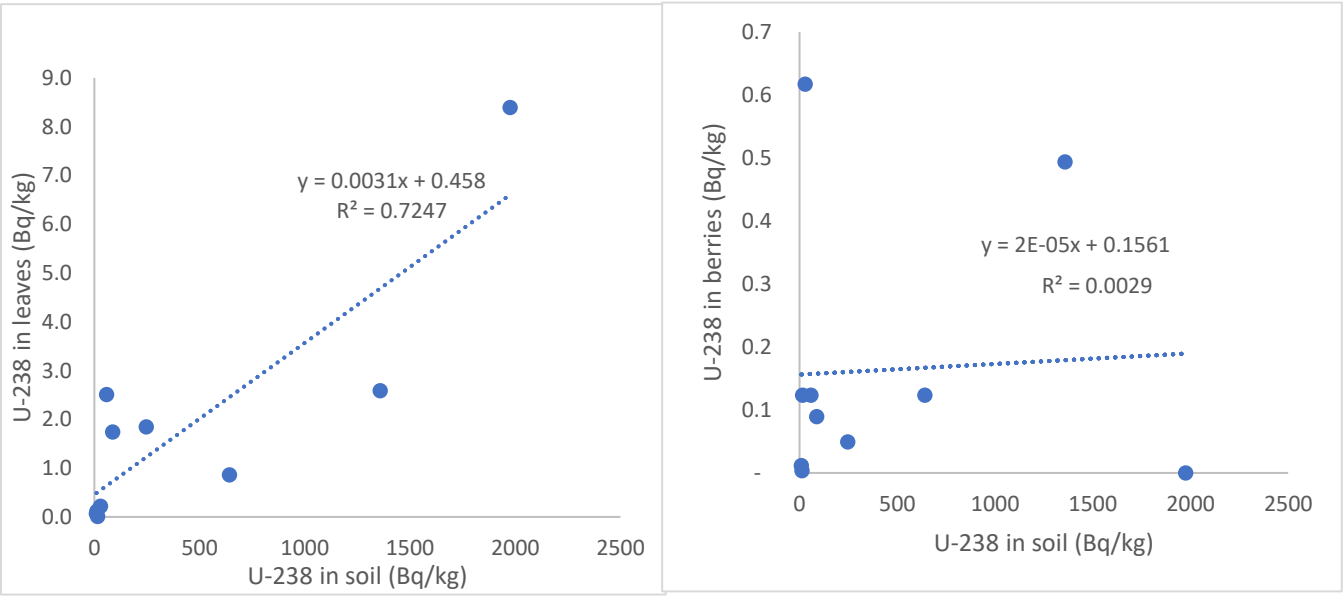

Figure 4.1. *Scatter plot showing relation between ^{238}U in soil and ^{238}U in plants and berries.*

The activity concentration of ^{226}Ra in the plant species sampled ranged from non-detectable (<LOD) to 5770 Bq/kg dw (table 4.6). Maximum levels were observed in blueberry (*V. myrtillus*) leaves (5770 Bq/kg dw). As for uranium, activity concentration of ^{226}Ra was higher in the leaves than in the berries

(figure 4.3). Based on Pearson correlation a strong positive correlation was observed between ^{226}Ra activity concentration in soil and activity concentration of ^{226}Ra in plant (r = 0.973, p = 0.001) and berries (r = 0.957, p = 0.001), as also shown on figure 4.2. This strong positive correlation can be explained by the fact that radium being an alkali earth element can mimic other essential earth elements like calcium and magnesium and thus taken up by plants and distributed evenly but not equally in all parts of the plants.

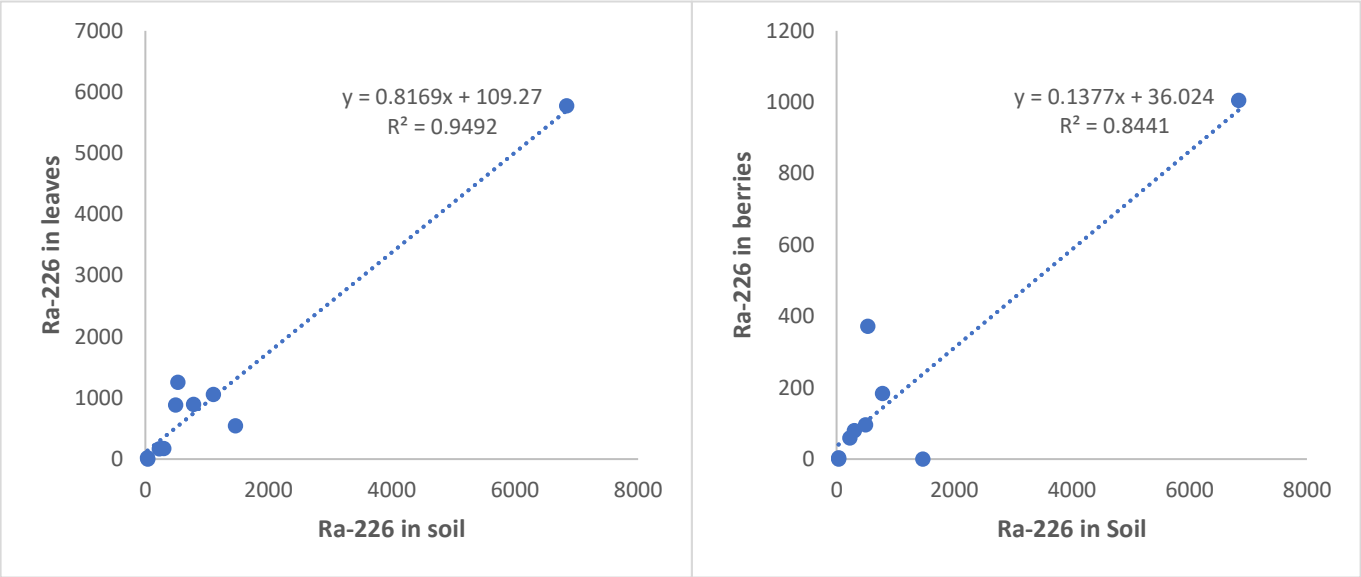

Figure 4.2. *Scatterplot showing a strong relation between ^{226}Ra in soil and ^{226}Ra in plants and berries.*

Table 4.6. Concentration of ^{226}Ra (Bq/kg dw) in selected vegetation species.

Species (Plants)	St.2	St.2*	St.3	St.4	St.6	St.7	St.9	St.10*	St.11*	St.12*
Vaccinium myrtillus	1692	857	293	464	18	208	<LOD	5770	1053	540
Empetrum nigrum	1235	932		1245		134	<LOD			
Vaccinium uliginosum	826		33	932						
Average	**1251**	**895**	**163**	**880**	**18**	**171**	**<LOD**	**5,770**	**1,053**	**540**
Species (Berries)										
Vaccinium myrtillus	372	352	59	96	3.5	90	<LOD	1005		
Empetrum nigrum		15								
Vaccinium vitis-idaea						69				
Average	**372**	**184**	**59**	**96**	**3.5**	**79**	**<LOD**	**1005**		**<LOD**
Grass and herbs										
Deschampsia flexuosa	71		19	49	<LOD	39	<LOD			
Geranium sylvaticum					6.0					85

<LOD -below detectable limit.

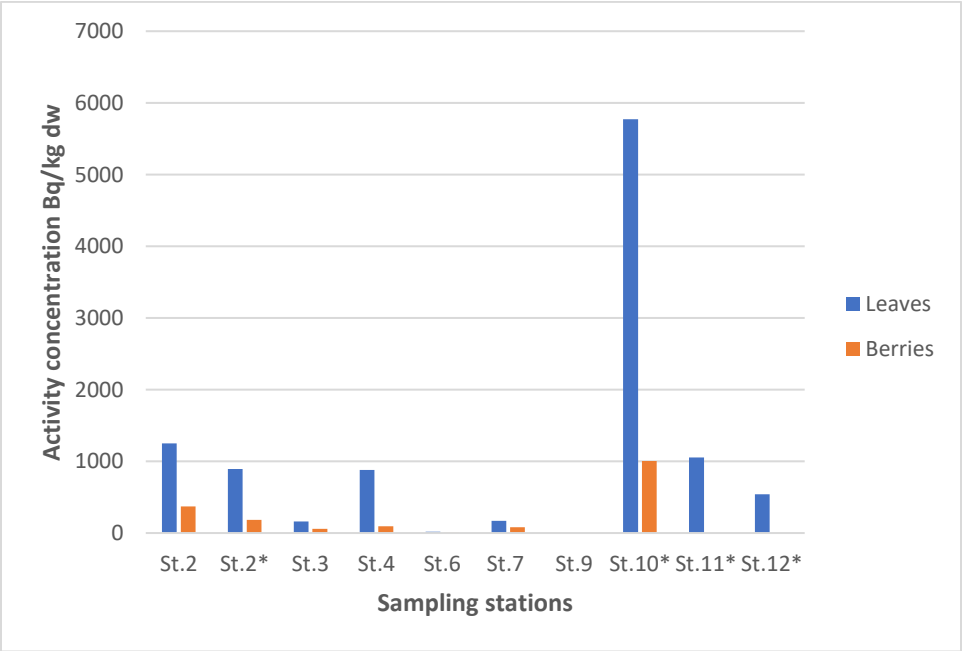

Figure 4.3. *Comparison of average activity concentration of ^{226}Ra in leaves and berries.*

4.2.2.1 Soil-to-plants transfer factor.

Soil to plants transfer factors for selected radionuclides calculated from the pooled data of the species are presented in table 4.7, for plants, berries and grass. Transfer factors for leaves were found to be in the range 0.001 – 0.04 (^{238}U), 0.37 – 2.36 (^{226}Ra), 0.23 – 0.6 (^{137}Cs), and 0.01 – 2.03 (^{40}K) while for berries the range was <0.0001 – 0.02 (^{238}U), 0.11 – 0.7 (^{226}Ra), 0.15 – 0.75 (^{137}Cs), and 0.32 – 4.55 (^{40}K). TF for grass were in the range of <0.0001 – 0.01 (^{238}U), 0.08 – 0.13 (^{226}Ra), 0.11 – 0.29 (^{137}Cs) and 0.79 – 9.05 (^{40}K). Transfer factors for ^{232}Th are not presented here as activity concentration in most plants were below detectable limits. This can also be explained by the low mobility of ^{232}Th due to its low solubility as reported by Martínez-Aguirre et al. (1995). Even though there was not much data for activity concentration of ^{210}Pb, the few samples available showed a decrease in ^{210}Pb transfer factor in blue berry *(V. myrtillus)* leaves with increasing ^{210}Pb concentration in soil (figure 4.4). Transfer Factors were found to be 0.04 for both station 2 and 7 with ^{210}Pb activity concentration in soil of 1180 and 1104 Bq/kg dw, respectively compared to TFs of 0.17 (station 6) and 0.11 (station 9) with ^{210}Pb activity concentration in soil of 117 and 244 Bq/kg dw, respectively. This observation has been made elsewhere by Sheppard et al. (2008) and Martinez-Aguirre et al. (1997).

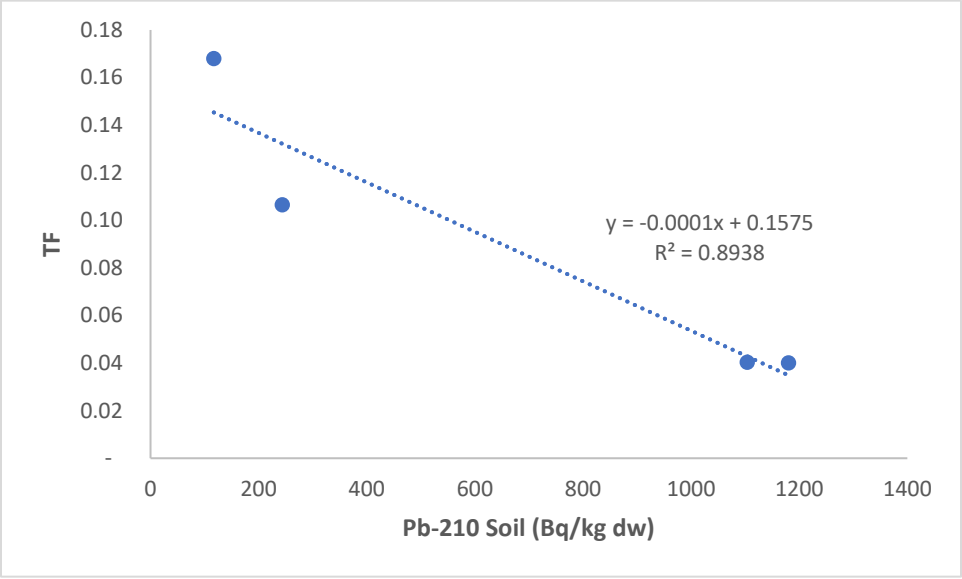

Figure 4.4: *Pb-210 soil-to-plant transfer factor against Pb-210 activity concentration in soil for blue berry (Vaccinium myrtillus) leaves.*

Table 4.7. Soil to plants transfer factors for selected radionuclides calculated from the mean of pooled data for berries, and berry leaves and grass *(Deschampsia flexuosa).*

	U-238			Ra-226			Cs-137			K-40		
Station	**Leaves**	**Berries**	**Grass**	**Leaves**	**Berries**	**Grass**	**Leaves**	**Berries**	**Grass**	**Leaves**	**Berries**	**Grass**
St.2	0.04	0.002	0.003	2.36	0.70	0.13	0.43	0.47	0.26	1.39	4.11	9.05
St.2*	0.02	0.001	0.000	1.14	0.23	-	0.36	0.40	-	0.72	1.38	-
St.3	0.01	0.02	0.004	0.72	0.26	0.08	0.36	0.75	0.29	0.18	0.93	1.67
St.4	0.01	0.0002	0.003	1.80	0.20	0.10	0.32	0.38	0.20	2.03	4.55	6.23
St.6	0.001	0.01	0.01	0.54	0.11		0.17	0.15	0.11	0.01	0.32	0.70
St.7	0.01	0.001	0.002	0.57	0.26	0.13	0.23	0.18	0.16	0.25	0.84	1.78
St.9	0.01	0.0003	0.01				0.17	0.20	0.14	0.14	0.42	0.79
St.10*	0.002	0.0004	0.00	0.84	0.15	-	0.60	0.56	-	0.13	0.46	-
St.11*	0.004	0.0000	0.00	0.95	-	-	0.25	-	-	0.02	-	-
St.12*	0.001	0.0002	0.00	0.37		-	0.18		-	0.01	< LOD	-

** Sampled in 2017*

In general, the transfer factors were found to be in close agreement with soil to plants transfer factors published by IAEA. The IAEA soil to plant transfer factors are in the range of 0.01 – 1.0 (^{226}Ra), 0.49 – 5.6 (^{40}K), 0.02 – 3.2 (^{137}Cs), and 0.02 (^{238}U) (IAEA, 2010). Transfer factors were found to be negatively correlated with pH, being high on sampling stations that had low pH for mostly all the radionuclides.

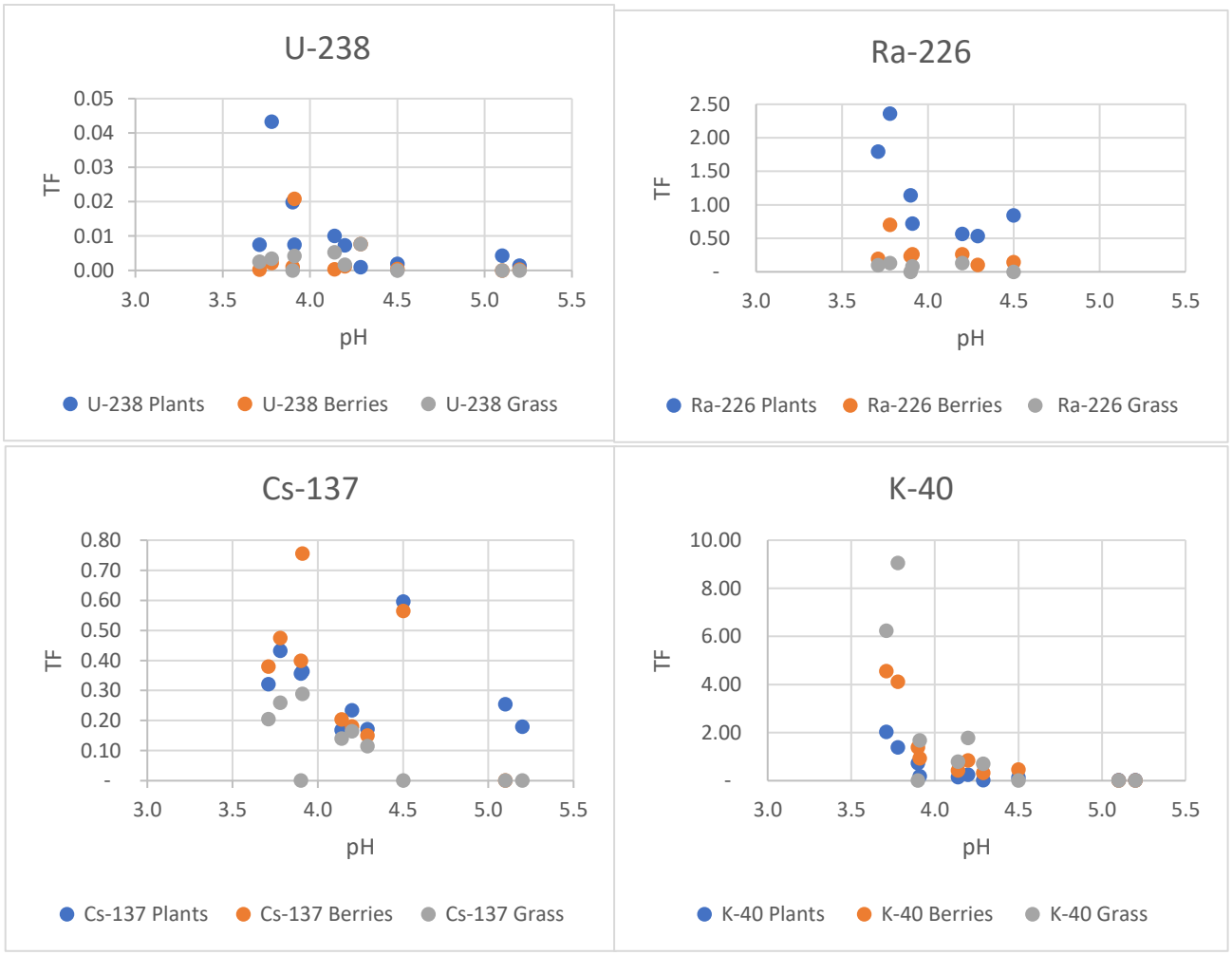

Figure 4.5: *Soil to plant transfer factor (TF) against soil pH (decrease in transfer factor with increase in pH).*

4.3 Concentration of main elements contaminants in soil

The concentrations of stable elements in soil and biota was included to check the possibility for a multiple stressor scenario, with contamination of both radionuclides and stable elements in the area. The elements analyzed included; As, Cd, Cr, Cu, Ni, Pb and Zn. A summary of obtained concentrations on the metals of interest in soil is shown in table 4.8 together with the concentrations of ^{238}U and ^{232}Th in mg/kg.

The obtained concentrations were compared with the default upper limit values for soils that can be considered as non-polluted in Norway given by Norwegian Authorities - SFT (2009) and European data as given in De Vos and Tarvainen (2005) (Table 3.4 and 4.8).

Table 4.8. - Soil concentration of elements of interest (mg/kg) for all the stations sampled compared with Norwegian norm values and European baseline values.

	As	Cd	Cr	Cu	Ni	Pb	Zn	Th	U
St. 1	3.8	0.1	37	19	19	18	69	5.3	4.6
St. 2	0.4	0.6	1.6	9.4	3.5	18	46	0.2	4.7
St. 2*	1	0.6	5.5	10	3.8	23	48	0.6	7.1
St. 3	1.6	1.3	8.2	11	5.5	19	43	1.1	2.4
St. 4	0.4	0.4	2.3	18	9.7	15	50	0.2	20
St. 5	2	0.4	35	56	29	15	77	5	9
St. 6	0.9	0.1	18	7.5	2.5	16	29	1.8	1.3
St. 7	0.7	0.5	7.7	6.6	4.3	15	89	0.5	0.8
St. 7b	1.3	0.2	45	26	16	20	54	5.2	66
St. 7b*	2.2	0.2	64	25	20	20	76	6	64
St. 8	0.7	0.3	15	40	21	10	27	1.9	19
St. 9	0.3	0.2	32	26	14	21	41	2.5	1
St. 10	1.6	0.4	59	28	18	45	23	7.1	110
St. 11	3.3	0.5	66	28	24	190	74	6.5	160
St. 12	12	0.4	62	20	24	81	170	5.7	52
SFT	8	1.5	50	100	60	60	200		
De Vos	7	0.14	60	13	18	32	52		

**- Sampled in 2017.*
SFT - Norwegian norm value by SFT (2009).
De Vos – European baseline by De Vos Tarvainen (2005).

The soil concentrations of Cd, Cu and Zn in all the sampling stations were below than the value of 1.5 mg/kg, 100 and 20 mg/kg for non-polluted Norwegian soil. The concentrations of As were also below the Norwegian value of 8 mg/kg and European value of 7 mg/kg in all the sampled station apart from soils from station 12 where concentrations of up to 12 mg/kg were measured. For Cr, concentrations were slightly higher than both Norwegian and European limits for stations 7, 10, 11, and 12 all sampled in 2017, while Pb concentrations were found to be high on station 11 and 12 with 190 mg/kg and 81 mg/kg, respectively.

Based on the Norwegian norm values, soil from the Orrefjell area can generally be described in as good. In all the stations sampled, the soil ranges from very good to moderate with only soil from station 11 having Pb concentration as 190 mg/kg, although its 3 times more than the Norwegian norm value and 6 times more than European baseline value, it can still be classified as moderate.

The soil concentrations of U at station 10 and 11 were high; 110 mg/kg (1358 Bq/kg) and 160 mg/kg (1975 Bq/kg), respectively. These concentrations exceed the screening value for the radioactive waste material in Norway (1 Bq/g). This reflects the abundance of uranium in the rocks at this sites that are potential for mining.

On uranium chemical toxicity, a summary on predicted no effect concentrations (PNEC) for uranium by Sheppard et al. (2005) shows no effects are expected to occur on terrestrial plants and soil biota under 250 and 100 mg U/kg dry soil, respectively. At Orrefjell, maximum concentrations for U were found at stations 10 and 11 with 110 and 160 mg U/kg, respectively, which are below 250 mg U/kg PNEC for terrestrial plant but slightly above 100 mg U/kg PNEC for soil biota. However, recent studies have shown concentration of 5 – 15 mg U/kg causing DNA damage and adverse effects on earthworms *Eisenia fetida* (Giovanetti et al., 2010). This implies that its possible to have DNA damage

Uranium has a strong positive correlation with Cr ($r = 0.763$, $p = 0.002$), Pb ($r = 0.842$, $p = 0.004$) and Th ($r = 0.716$, $p = 0.004$), but a weak correlation with As ($r = 0.281$, $p = 0.330$), Co ($r = 0.395$, $p = 0.162$), Cu ($r = 0.250$, $p = 0.388$). A negligible correlation was found for U and As ($r = 0.281$ $p = 0.330$) and Cd ($r = -0.033$, $p = 0.911$). Thorium was found to have a strong positive correlation with cobalt ($r = 0.735$, $p = 0.003$), chromium ($r = 0.961$, $p = 0.000$) and nickel ($r = 0.808$, $p = 0.000$). A positive correlation between U, Cr and Pb could mean that they share the same bedrock and an increase in uranium concentrations due to human activity would then mean a concomitant increase in chromium and lead concentrations. This can cause a multiple stressor scenario.

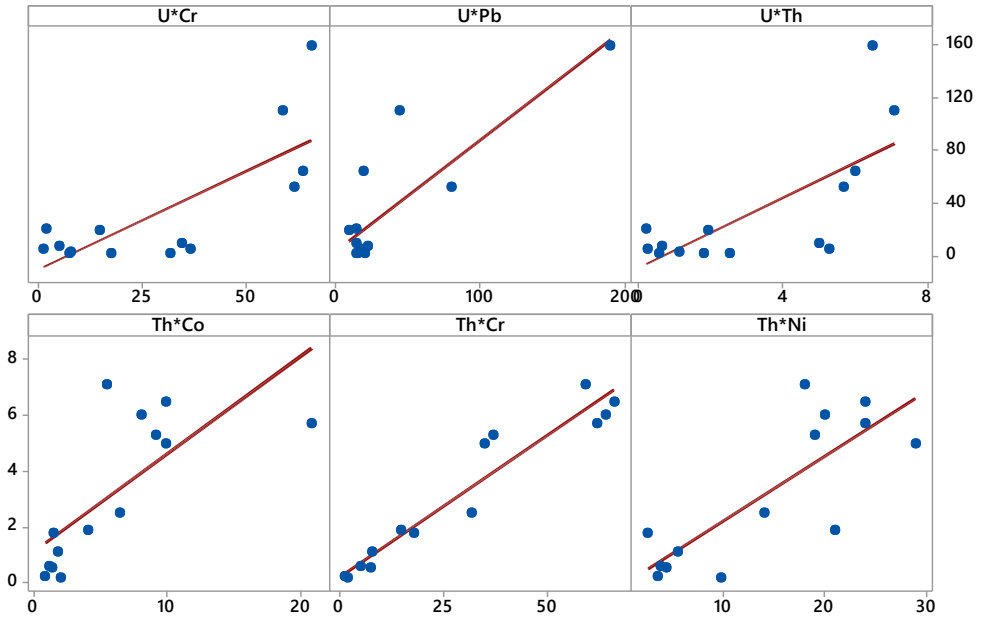

***Figure 4.6**. Scatter plot showing the relation of uranium and thorium with selected stable elements.*

4.4 Activity concentration of specific radionuclides and accumulation of stable elements in earthworms

The concentrations of the selected elements in earthworms are shown in table 4.9. Since the study was not about the uptake of specific earthworm species but rather the population of earthworms, the average concentration of all the earthworms in each site was used. Concentrations of selected elements for individual earthworms are given in appendix B. A large variability between individual earthworms was found for all the selected stable elements and radionuclides which is typical to most studies. This variability can be explained by the fact that the uptake of radionuclides and stable elements is a dynamic process where the individual earthworm organism reach a maximum load, after which excretion starts. Meaning the earthworm body concentration could be dependent on prevailing process of either active uptake or excretion (van Vliet et al., 2005).

Based on soil concentration of the sites where the earthworms were sampled, the transfer factor for ^{238}U was in the range of 0.04 – 0.32 (average 0.14), this is close to the transfer factors reported by Popic et al. (2012) on earthworms at the Fen Complex in Norway, where they found the average TF for ^{238}U to be 0.20 (range 0.09 – 0.25). The transfer factors for ^{232}Th were found to be almost similar

in all the stations sampled with a range of 0.01 – 0.04 (average 0.02). This also compares to the results reported by Popic et al. (2012) where the found the average transfer factors for earthworms at Fen complex to be 0.06.

For metals low transfer factors were observed for As, Cr, Cu, Ni and Pb (less than 0.5), with exception of earthworms in station 5 and 8 with TF of 1.1 and 2.8, respectively. The transfer factor for Cd and Zn were high (7 – 53 for Cd, and 3 – 8 for Zn). The accumulation of Cd can be explained by the fact that Cd, as a nonessential element is hardly eliminated from the body and therefore accumulates for a long period of time in worms (Spurgeon & Hopkin, 1999).

Table 4.9. Earthworm concentration of stable elements (mg/kg d.w) averaged for each site sampled (mean±standard deviation).

Station	U*	Th*	As	Cd	Cr	Cu	Ni	Pb	Zn
St.5 (n=7)	0.9±0.4	0.1±0.1	2.3±1.4	4.8±3.5	1.0±0.6	21±4	2.2±0.7	0.4±0.2	279±51
St.7(n=4)	5.4±2.6	0.2±0.1	0.3±0.1	8.0±2.3	2.4±2.1	11±2.6	1.0±0.7	0.9±0.6	393±105
St.8(n=8)	0.8±0.4	0.02±0.017	1.9±0.9	2.4±1.4	0.3±0.2	9.4±1.3	1.0±0.4	0.3±0.2	214±57
St.11(n=18)	52±29	0.2±0.3	1.4±0.5	10±6.2	2.5±3.2	9.1±2.8	2.3±2.4	380±561	298±106

** Activity concentration (Bq/kg dw) of radionuclides ^{238}U and ^{232}Th can be calculated by multiplying concentration in mg/kg by 12.35 (U) and 4.06 (Th).*

4.5 Total doses to biota – based on soil activity concentrations

An initial screening of all the sites was performed using Tier 1 of the ERICA Tool. Here only soil activity concentration of the radionuclides of interest (^{137}Cs, ^{210}Pb, ^{226}Ra, ^{232}Th, and ^{238}U) were used as input, with all the reference organisms in the tool selected. The results showed that in all the sites sampled the total risk quotient (RQ) for all the sites was greater than 1, meaning that in all the station the default screening dose of 10 μGy/h was exceeded. Risk Quotient is the result of comparison of radionuclide concentration in the media against Environmental Media Concentration Limits (EMCL). From the results the limiting reference organism was found to be lichens and bryophytes for all the sites with ^{226}Ra and ^{210}Po presenting a risk worthy of further investigation.

Further assessment of doses to terrestrial organisms in the three areas, Orrefjell high-altitude, Orrefjell cultivated grassland and Orrefjell control, was carried out using Tier 2 of the ERICA Tool. Due to inhomogeneous distribution of radionuclides in the area, both maximum and mean soil activity

concentrations of radionuclides were used. The results and comparison of total dose received by reference organisms in the three studied areas are shown in table 4.10 below.

Table 4.10. Total dose received by ERICA Tool reference organisms in µGy/h, using maximum and mean soil activity concentration

	High Altitude (n=9)		Cultivated (n=3)		Control (n=2)	
Organism	**Mean**	**Max.**	**Mean**	**Max.**	**Mean**	**Max.**
Amphibian	13	51	0.7	0.9	0.6	1.1
Annelid	10	49	0.5	0.6	0.3	0.4
Arthropod - detritivorous	9.8	48	0.4	0.5	0.3	0.4
Bird	7.9	38	0.3	0.3	0.2	0.3
Flying insects	9.0	43	0.4	0.4	0.3	0.3
Grasses & Herbs	44	186	2.5	3.5	1.9	3.2
Lichen & Bryophytes	228	813	16	23	13	24
Mammal - large	12	47	0.6	0.8	0.6	1.0
Mammal - small-burrowing	12	50	0.6	0.9	0.6	1.0
Mollusc - gastropod	10	49	0.5	0.6	0.3	0.4
Reptile	13	52	0.8	1.0	0.7	1.2
Shrub	73	325	3.3	4.3	2.8	4.3
Tree	5.0	16	0.4	0.5	0.3	0.6

The maximum calculated doses to all reference organisms were as expected, highest at the Orrefjell high altitude area, with the highest dose calculated for lichens and bryophytes (813 µGy/h) followed by shrub (325 µGy/h) and grasses & herbs (186 µGy/h). Calculated dose to all reference organism exceed the screening dose value of 10 µGy/h with trees having the least dose of 16 µGy/h with the rest of the reference organism receiving doses from 38 µGy/h to 51 µGy/h. Total dose rates to reference organisms calculated using the mean soil activity concentration of radionuclides were up to 5 times less than the total dose calculated using the maximum soil activity concentration. Here, the highest dose rates were calculated for lichen and bryophytes at 228 µGy/h followed by shrub, and grasses and herbs at 73 and 44 µGy/h, respectively, at the high-altitude stations. When using the mean soil activity concentration, doses for trees (5.0 µGy/h), birds (7.9 µGy/h), flying insects (9.0 µGy/h) and Arthropod – detritivorous (9.8 µGy/h) were all below the screening value of 10 µGy/h at the high-altitude stations. This suggests that the use of mean activity concentrations only to quantify risk to organisms in an area with inhomogeneous distribution of radionuclides can lead to underestimation of doses to organism in such areas. Comparison for total dose received by reference organisms using maximum and mean soil activity concentration for radionuclides are shown in figure 4.5.

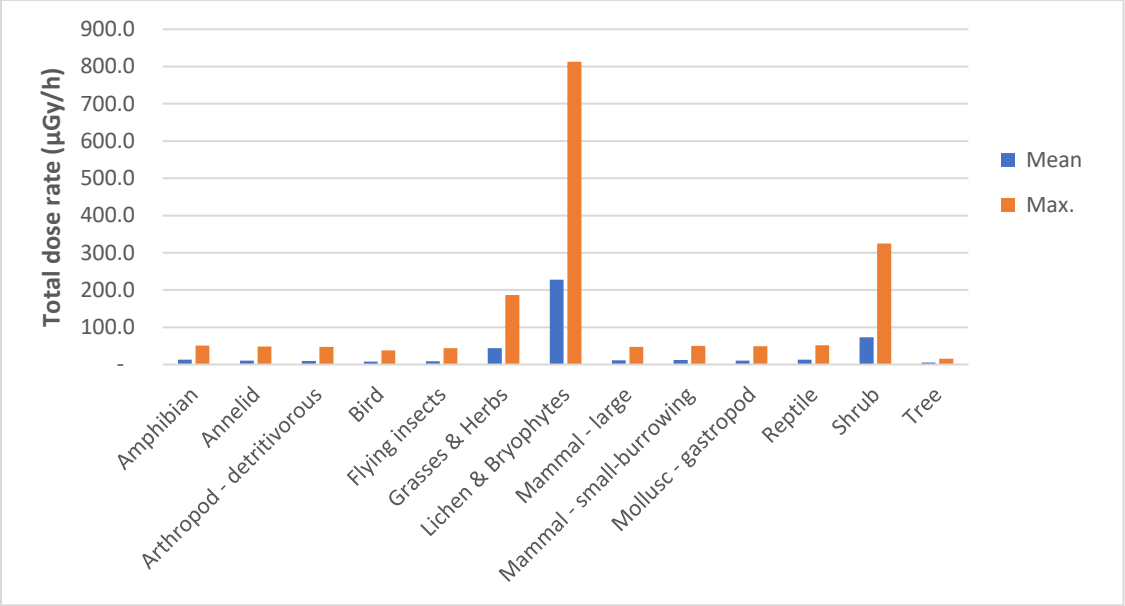

Figure 4.7. *Total dose received by reference organism at Orrefjell high altitude sites, calculated using mean and maximum soil activity concentration.*

In all the reference organisms, internal alpha exposure to ^{226}Ra was the major contributor to the total dose, contributing 70% - 99% of the total dose. The only other radionuclides that alone contributed to a dose rate above the adopted screening value of 10 µGy/h were ^{210}Po (94 µGy/h and 10 µGy/h to lichen & bryophyte and grass & herb, respectively), and ^{238}U (43 µGy/h) to lichens and bryophytes. External exposure contributed little to the total dose received by the organisms.

Assessment done on individual stations (Appendix C), estimates the highest dose rates for all reference organisms in station 10, with the maximum dose calculated for lichen and bryophyte (705 µGy/h) (figure 4.6). This is due to the high activity concentration of ^{226}Ra (6840 Bq/kg dw) in the soil. However, the real dose rate might be even higher, as ^{210}Pb and ^{210}Po were not measured in this station and are likely present in concentration giving a considerable contribution to the dose rate. It is worth noting that doses to lichen and bryophytes at station 10 (705 µGy/h) was above the level at which reproductive effects have been observed in a laboratory (about 400 µGy/h) (Andersson et al., 2009). International organisations have adopted levels of 40 and 400 µGy/h for terrestrial animals and plants, respectively, as values below which no effect on population levels should be expected (UNSCEAR, 2008). Other notable highly exposed stations are station 11 and 12 where doses of 153 µGy/h and 159

μGy/h were calculated for lichen and bryophytes, respectively. This suggest that a further analysis would be important especially for this station.

Orrefjell as an undisturbed NORM sites, doses from naturally occurring radionuclides are not expected to be potentially harmful to the plants. However, assessment on station 10 shows considerably high doses estimated for lichen and bryophytes above the adopted level of 400 μGy/h above which biological effects can be observed in plant population.

For both the cultivated grassland area and the control area, doses received by reference organisms were up to 3 orders of magnitude lower than doses in the high-altitude area. Total dose rates received by reference organisms in these two areas were close to identical. Maximum doses for lichen and bryophytes were 23 μGy/h for the cultivated area and 24 μGy/h for the control area. The rest of the reference organisms were estimated to receive less than the screening value of 10 μGy/h. This dose rates however, were higher than those generally experienced by terrestrial organism (0.01 – 0.7 μGy/h). The main contributors to the total dose in both the cultivated and control areas were internal exposure to ^{226}Ra, ^{210}Po and ^{238}U. This is different from Orrefjell high-altitude area where ^{226}Ra was the only major contributor to total dose. Internal exposure to ^{226}Ra to lichen & bryophytes in the cultivated grassland stations constituted 16-75% of the total dose rate, while ^{210}Po contributed 8-73%, while in the control area 226Ra constituted 12 – 85% of the total dose.

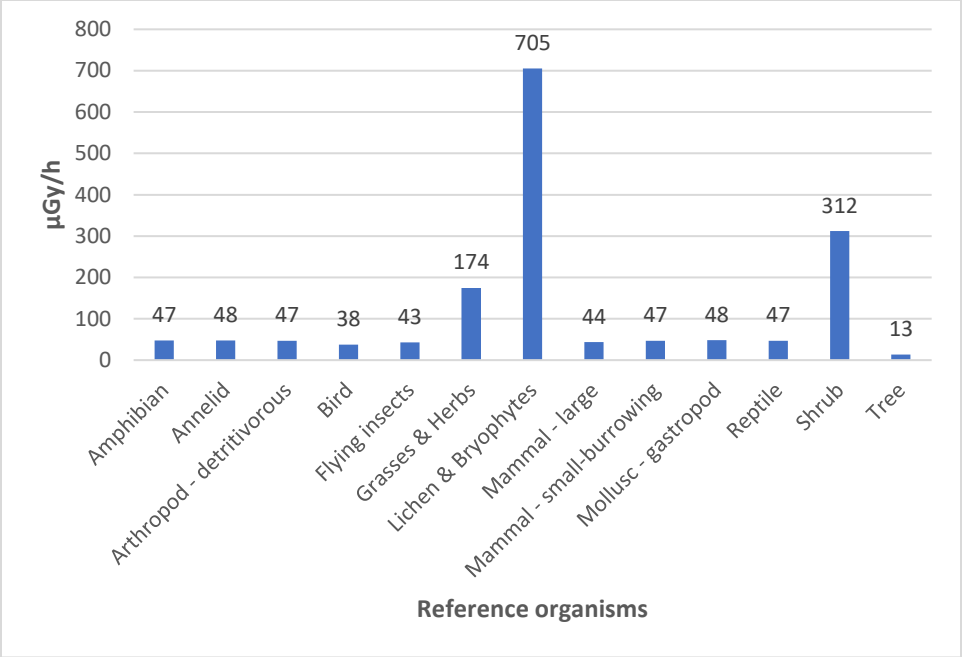

Figure 4.8: *Calculated total doses to reference organisms at station 10 using soil activity concentrations only.*

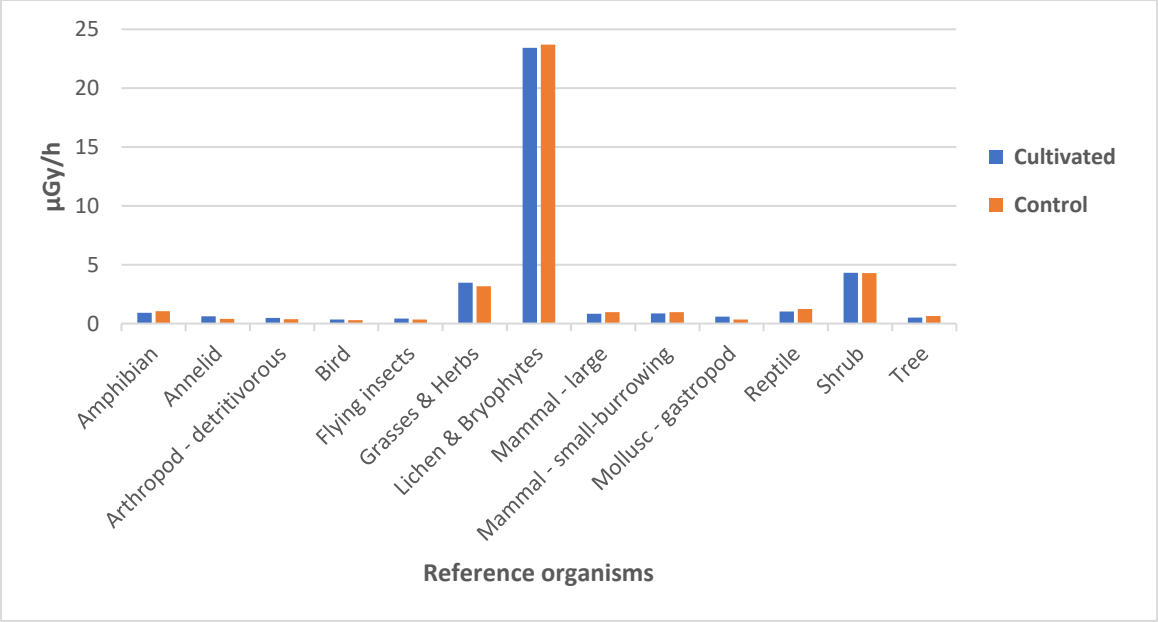

Figure 4.9. *Total dose received by reference organism at Orrefjell cultivated grassland site and control sites.*

4.6 Total doses to biota based on soil and biota activity.

The second ERICA assessment was done using site-specific activity concentrations of soil and selected biota for the radionuclides ^{137}Cs, ^{210}Pb, ^{210}Po, ^{226}Ra, ^{232}Th and ^{238}U. This was to provide a comparison with the dose rates calculated using the tool default transfer parameters. The most numerous plant samples collected were used as sample representative to the reference organisms in the ERICA Tool: Blueberry leaves (*V. myrtillus*) concentrations were used as input for shrub, hair-grass (*D. flexuosa*) concentrations as input for grass and cranesbills (*G. sylvaticum*) as input data reference herbs. Hair-grass (*D. flexuosa*) and cranesbills (*G. sylvaticum*) activity concentrations were pooled and arithmetic mean and maximum used in ERICA Tool as input for reference grass and herbs. Though there was no data for lichen and bryophytes and tree, the two were kept in the second assessment as they received maximum and minimum doses, respectively in the initial assessment.

The recalculated total dose rate at Orrefjell high-altitude area using maximum activity concentrations of radionuclides in soil and selected vegetation, was lower for grass & herbs (15 µGy/h compared to 187 µGy/h) and a bit higher for Shrub (365 µGy/h compared to 325 µGy/h) (Figure 4.8). The decrease in dose rate for grass and herbs is due to a lower site-specific CRs for grass & herbs compared to the default CRs. This shows an overestimation by of dose received by grass & herbs using default concentration ratio in ERICA Tool in this particular case. However, for the shrub the ERICA tool performs very well in this case. It is worth noting that the reference organism "Grass & herbs" and indeed all the reference organisms in ERICA tool are considered to cover a range of different species with different life strategies and ecology and thereby possibly different uptake. This can cause uncertainty when estimating doses to organism when only one species is considered.

Site specific concentration ratios (CRs) were one to two orders of magnitude lower than the default ERICA Tool for grass & herbs for all the selected radionuclides (Table 4.11). For blueberry leaves the site-specific concentration ratio for ^{226}Ra was in good agreement with the default concentration ratio for shrub in ERICA Tool while for the other radionuclides, sites specific CRs were one to two orders of magnitude lower than the default ERICA CR (Table 4.12). The same ^{226}Ra concentration ratio in shrub explains why total dose received by shrub does not change when site-specific biota activity concentrations are used since ^{226}Ra is the main contributor to total dose. It also explains why there was

a decrease in dose received by grass and herbs. This confirmed the uncertainty related to use of transfer parameters in the assessment studies (Oughton et al., 2008).

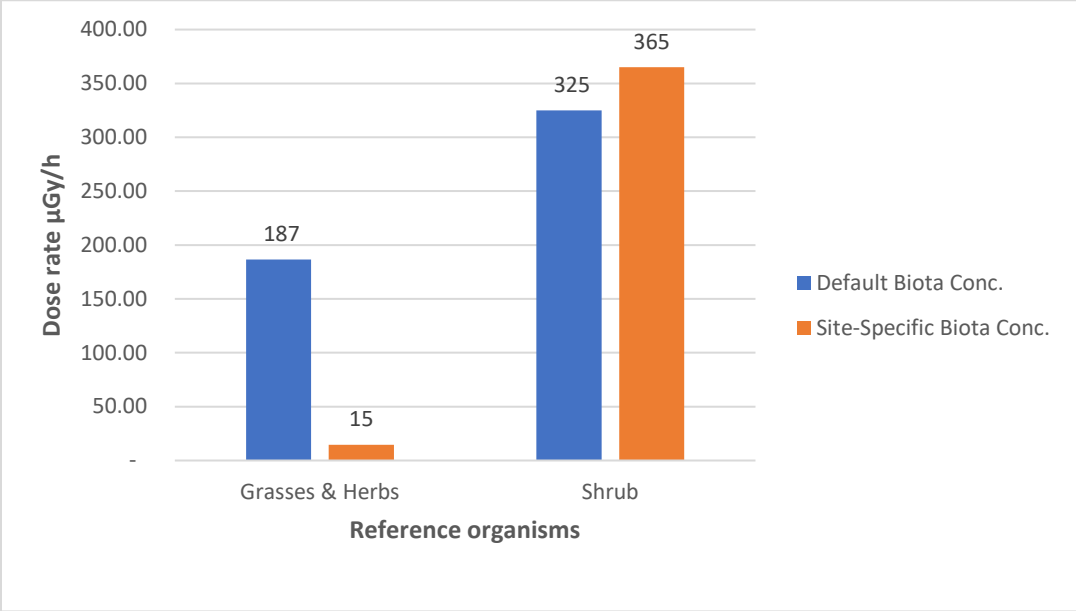

***Figure 4.10**: Total dose rate (µGy/h) received by reference organisms at Orrefjell high-altitude area using default ERICA concentration ratios and site-specific activity concentrations (only for grass & herbs and shrub). Maximum activity concentrations were used.*

Internal exposure to ^{226}Ra remained the biggest contributor of total dose to the reference organisms. At Orrefjell high-altitude sites, 79% (12 µGy/h) of the total dose received by grasses and herbs came from internal exposure to ^{226}Ra which was a decrease from 90% when using default parameters (Figure 4.9). External exposure to ^{226}Ra constituted about 15% (2.3 µGy/h) of the total dose rate. For shrubs, there was only a slight difference in percentage contribution, with internal exposure to ^{226}Ra contributing 99% (362 µGy/h) of the total dose. Contribution by other radionuclides to both grasses & herbs, and shrub were below 5%.

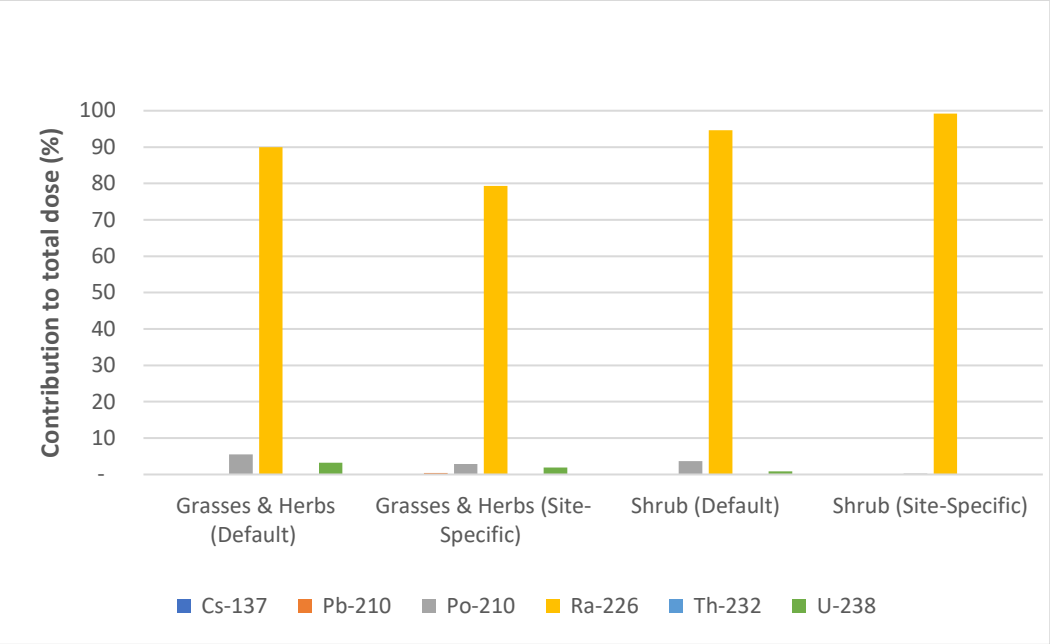

Figure 4.11: *Radionuclide contribution (%) on the total dose received by grasses & herbs, and shrub at Orrefjell high-altitude sites using default and site-specific parameters.*

In the control sites the main contributors to total dose received by grasses and herbs, and shrubs were ^{226}Ra and ^{210}Po. For grasses and herbs, 85% of the total dose was from ^{226}Ra, while 13% came from ^{210}Po. (Figure 4.10). Contribution from the other radionuclides was below 2% and less than 1% from external exposure. For shrubs, 88% was from ^{226}Ra, while 11% came from ^{210}Po. The other radionuclides only contributed less than 1% of the total dose.

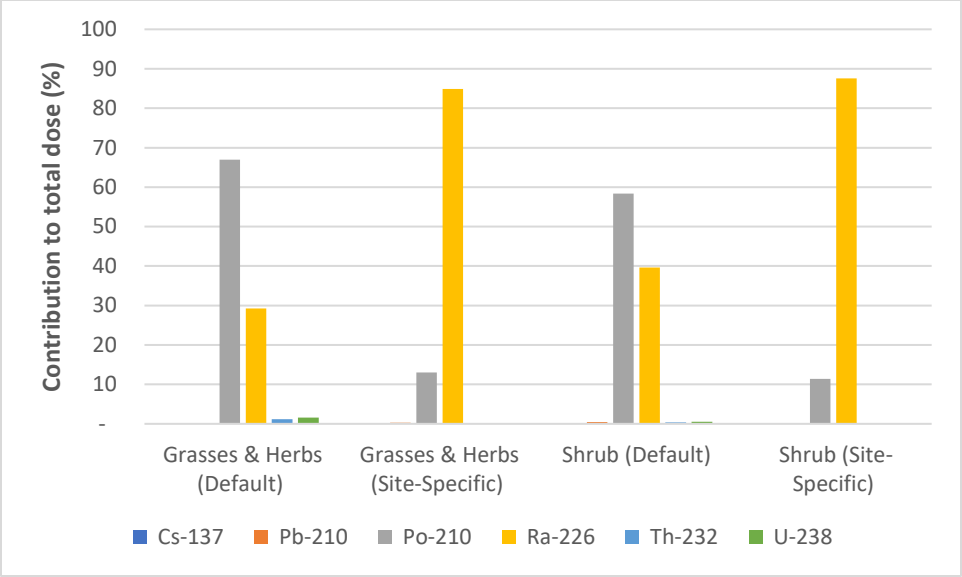

Figure 4.12: *Radionuclide contribution (%) on the total dose received by grasses and herbs, and shrub at Orrefjell control sites using default and site-specific parameters.*

ERICA Tool predicted activity concentrations for the selected reference organisms was also compared to the measured site-specific activity concentration for sampled vegetation (Table 4.13). Predicted ^{226}Ra activity concentrations in shrub using soil activity concentration were in good agreement with the measured site-specific activity concentrations of ^{226}Ra in blueberry (*V. myrtillus*) leaves further explaining why there is no significant difference between dose rate calculated using default values and site-specific values for shrubs. Predicted activity concentration of other radionuclides tended to be higher than measured activity concentration for the same species. Predicted activity concentration for ^{210}Pb were similar to those measured in hair grass (*D. flexuosa*). For the other radionuclides the predicted activity concentration tended to be lower than those measured in the same species.

Table 4.11. Comparison of reference organism doses with site-specific doses (µGy/h) and Concentration Ratios in hair grass (*Deschampsia flexuosa*) at Orrefjell high-altitude sites.

	Total Dose (µGy/h)		**Concentration ratio (CR)**	
Radionuclide	Reference grass&herb	Site-specific (Hair grass)	ERICA	Site-specific*
Cs-137	2.35E-02	1.07E-02	1.12E+00	9.40E-02
Pb-210	3.29E-02	6.00E-02	1.20E-01	6.26E-03
Po-210	1.02E+01	4.25E-01	2.80E-01	5.76E-03
Ra-226	1.70E+02	1.39E+01	1.80E-01	4.75E-02
Th-232	1.06E-01	9.23E-04	1.60E-01	2.03E-03
U-238	6.06E+00	2.82E-01	1.28E-01	1.13E-03

** Geometric mean (n=8) of pooled data at Orrefjell high-altitude sites.*

Table 4.12. Comparison of reference organism doses with site-specific doses (µGy/h) and Concentration Ratios in blueberry (*Vaccinium myrtillus*) leaves at Orrefjell high-altitude sites.

	Total Dose (µGy/h)		**Concentration ratio (CR)**	
Radionuclide	Reference Shrub	Site-specific (Blueberry leaves)	ERICA	Site-specific*
Cs-137	3.39E-02	1.31E-02	1.96E+00	1.63E-01
Pb-210	8.66E-02	6.92E-03	3.20E-01	2.48E-02
Po-210	1.21E+01	7.35E-01	3.30E-01	2.01E-02
Ra-226	3.10E+02	3.64E+02	3.30E-01	3.77E-01
Th-232	4.05E-02	4.61E-04	6.10E-02	7.94E-04
U-238	2.89E+00	1.03E-01	6.10E-02	1.91E-03

** Geometric mean (n=7) of pooled data at Orrefjell high-altitude sites*

Table 4.13. Comparison of site-specific radionuclides activity concentration (Bq/kg dw) in selected plants (*Vaccinium myrtillus* and *Deschamsia flexuosa*) with ERICA Predicted activity concentration to reference organisms.

	Radionuclide concentration (Bq/kg dw)					Radionuclide concentration (Bq/kg dw)				
	Shrub					**Grasses & Herbs**				
	Site-specific			**ERICA Predicted**		**Site-specific**			**ERICA Predicted**	
	n	Mean	Max.	**Mean**	**Max**	n	Mean	Max.	**Mean**	**Max**
Cs-137	8	12	24	99	173	6	10	17	56	98
K-40	8	102	171	-	-	6	362	502	-	-
Pb-210	2	23	29	325	378	3	131	349	122	142
Po-210	2	15	24	335	389	2	23	33	284	330
Ra-226	8	690	2654	447	2257	5	97	305	244	1231
Th-232	8	0.01	0.02	0.8	1.8	7	0.04	0.1	2.0	4.6
U-238	8	1.4	4.3	35	120	7	4.8	31	74	253

4.7 Exposure dose rate to Earthworms

Radiation exposure dose rates of the earthworms were estimated in station 11 which was the most contaminated station sampled for earthworms. Based on the soil activity concentration and the default transfer parameter in ERICA tool, total dose rate calculated using tier 2 was 9 μGy/h with ^{226}Ra being the major contributor to the dose with 71% contribution to the total dose rate. The only other radionuclide with significant contribution to total dose rate was ^{238}U with 17% contribution to the total dose rate. A second assessment using site-specific activity concentration of ^{238}U and ^{232}Th was carried out. The result showed an increase in total dose rate to 23 μGy/h with internal exposure to ^{238}U being the main contributor to total dose rate with 67%. This consequently reduced the contribution of ^{226}Ra to 28%. This can be explained by the use of site-specific concentration ratio (0.32) which was more than 9 times higher than the ERICA default concentration ratio for U (0.03). This consequently explains the increase in total dose rate from 9 μGy/h to 23 μGy/h. This shows an underestimation of total dose by ERICA tool using default transfer parameters.

5. Hypothesis evaluation and conclusion

Hypothesis 1

Based on the big difference in activity concentration among radionuclides in the U-series, this study has shown that there is no secular equilibrium between U and its daughter radionuclides. For example, in stations where activity concentration of ^{210}Pb was measured, it was 4 – 110 times higher than ^{238}U. Activity concentrations of ^{226}Ra was 30 times higher than ^{238}U at the same station. Therefore, we reject the null hypothesis that U is in equilibrium with the daughter radionuclides.

Hypothesis 2

Based on the dose estimates from ERICA tool, the estimated doses e.g. for station 10 are higher enough to give detrimental effect on biota. Thus, if the dose estimates are correct, even undisturbed NORM sites can give harmful doses to biota. Therefore, the null hypothesis in this case is false.

Soil activity concentrations at Orrefjell showed elevated levels of radionuclides associated with the uranium decay series. The radionuclides were unevenly distributed among the sites and the most dominant radionuclide was ^{226}Ra with activity concentration ranging from 226 – 6800 Bq/kg dw in the high-altitude area. The activity concentrations for ^{238}U and ^{210}Pb/^{210}Po ranged from 10 – 2000 Bq/kg dw and 610 – 1180 Bq/kg dw, respectively. While the mean concentration of ^{238}U (538 Bq/kg dw) and was above the world average value of 33 Bq/kg dw, it was below the limit of 1 Bq/g for radioactive waste in Norway. However, soil samples from stations 10 and 11 had ^{238}U activity concentration of above the limit of 1 Bq/kg dw (1358 and 1975 Bq/kg dw, respectively). Further studies of these sites with high activity levels might be needed to improve the environmental risk analysis.

Secular equilibrium was assumed between ^{210}Pb and ^{210}Po. However, based on the big difference in among radionuclides in the ^{238}U series that has been demonstrated here, this assumption might be false, but the high radiotoxicity of ^{210}Po made us chose to include it as it very important for dose calculation.

This is supported by our results showing ^{210}Po gives a great part of dose. The assumption of secular equilibrium is the best estimate from the available data.

Activity concentration of radionuclides in plant species sampled were generally low and varied among species and plant part. However, notable high levels of ^{226}Ra (5770 Bq/kg dw) were measured blueberry (*V. myrtillus*) leaves at station 10. Calculated soil-to-plant transfer factor showed a close agreement with transfer factor published by IAEA.

Obtained concentration of inorganic elements As, Cd, Cr, Cu, Ni, Pb, and Zn in soil were compared with the default upper limits for soils that can be considered non-polluted in Norway and Europe. The results showed that the soil at Orrefjell were generally below the Norwegian and European limits for non-polluted soil. However, soil sample from station 11 had elevated Pb concentration and is classified to have moderate soil quality. Maximum concentrations of uranium measured at station 11 (160 mg/kg) and 10 (110 mg/kg) were below 250 mgU/kg PNEC for terrestrial plant but slightly above 100 mgU/kg PNEC for soil biota.

At all sites sampled, the maximum estimated dose rates were for lichen & bryophytes, followed by shrubs, and grass & herbs. Internal alpha dose from ^{226}Ra was the biggest contributor to total dose with ^{210}Po being second in the sites where it was measured, with the external dose and radiation from other radionuclides analyzed having very low contribution to the total dose. As expected from activity concentration in soil, highest doses to terrestrial biota were calculated in the high-altitude sites, with organisms from station 10, 11 and 12, being the most exposed with doses reaching 705, 153, and 158 µGy/h for lichen and bryophytes in station 10, 11, and 12, respectively. At these same sites, doses calculated for shrub were 312, 53, and 67 µGy/h, respectively, while the dose for grass and herbs were 174, 34, and 38 µGy/h, respectively. Most of the sites, particularly in the Orrefjell high-altitude area, showed doses being higher than the ERICA Tool default screening value of 10 µGy/h for most organism. This default value is a conservative estimate representing the generic dose below which no adverse effect would be expected on populations of any species.

Radiation doses obtained using site-specific biota concentration showed lower total dose rate for grass & herbs (15µGy/h) compared to dose rate obtained using default ERICA concentration ratios (CRs) (187µGy/h), while they were in agreement for shrub. It should be noted that the number of species

sampled at the sites was limited and very few reference organisms listed on the ERICA Tool were sampled at the site. The few samples collected may not necessarily be representative of the most exposed organisms but the assessment of the most exposed organism as well as the uncertainty of the bioavailability of the radionuclides can offer important information for planning future field study sampling.

The main concern for detrimental effects on biota in the Orrefjell is radiation from ^{238}U-related radionuclides, while other measured natural and anthropogenic radionuclides (^{232}Th, ^{40}K, and ^{137}Cs) and stable elements were of little concern. This work shows that also undisturbed NORM sites can give elevated doses to biota with potential harmful effects.

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7. Appendixes

Appendix A: Orrefjell soil characteristics and concentration of selected stable elements

Sample	% LOI	Estimated % Org. C**	pH	Fe***	Mg***	Ca***	K***	P***	S***
St.1	21,7	12,6	4,6	3,50E+04	1,30E+04	6,40E+03	4,70E+03	1,3E+03	8,2E+02
St.2	95.6	55.4	3,8	8,50E+02	1,40E+03	5,30E+03	1,00E+03	9,7E+02	1,9E+03
St.2*	90,1	52,3	3,9	2,10E+03	1,30E+03	4,70E+03	2,90E+03	6,2E+02	1,6E+03
St.3	74,8	43,4	3,9	5,30E+03	1,90E+03	6,60E+03	3,00E+03	1,1E+03	1,8E+03
St.4	95,0	55,1	3,7	1,20E+03	1,40E+03	4,60E+03	1,20E+03	1,1E+03	1,9E+03
St.5	37,1	21,5	6,0	2,40E+04	9,30E+03	2,20E+04	3,90E+03	1,3E+03	2,9E+03
St.6	10,8	6,3	4,3	7,70E+03	2,50E+03	1,90E+03	7,00E+03	5,4E+02	2,4E+02
St.7	71,9	41,7	4,2	3,10E+03	2,00E+03	6,00E+03	3,40E+03	1,5E+03	1,7E+03
St.7*	12,1	7,0	5,2	3,80E+04	1,10E+04	4,90E+03	1,10E+04	1,2E+03	6,1E+02
St.8	71,0	41,2	6,0	8,80E+03	3,60E+03	3,90E+04	1,80E+03	1,4E+03	5,4E+03
St.9	26,0	15,1	4,1	1,70E+04	5,90E+03	4,30E+03	5,30E+03	6,6E+02	6,0E+02
St.10*	48,7	28,2	4,5	1,80E+04	2,80E+03	4,20E+03	8,00E+03	1,1E+03	1,5E+03
St.11*	18,1	10,5	5,1	3,60E+04	1,80E+04	5,40E+03	1,20E+04	8,0E+02	5,9E+02
St.12*	23,9	13,9	5,2	6,70E+04	1,80E+04	1,10E+04	1,10E+04	5,0E+02	5,6E+02

** Sampled in 2017*

*** 58% of LOI is Org.C (Øien og Krogstad, 1987)*

**** Concentration in mg/kg dry weight*

Appendix B. Concentration of selected elements in individual earthworms in mg/kg

	As	Cd	Cr	Cu	Ni	Pb	Th	U	Zn
st 11 Earthworm 1	2.1	8.8	0.9	7.8	0.9	83	0.1	23	240
st 11 Earthworm 2	0.9	4.1	0.8	6.3	0.7	4.9	0.1	32	240
st 11 Earthworm 3	1.7	15	1.4	11	2.4	370	0.1	40	470
st 11 Earthworm 4	1.5	7.0	0.2	6.6	0.6	1300	0.1	66	330
st 11 Earthworm 5	1.4	10	1.2	9	1.5	280	0.1	36	260
st 11 Earthworm 6	0.5	6.3	0.2	11	1.8	1.6	0.01	75	240
st 11 Earthworm 7	1.4	7.6	11	15	11	150	0.8	40	280
st 11 Earthworm 8	0.9	6.0	0.5	8.6	1.4	130	0.03	65	310
st 11 Earthworm 9	0.9	3.7	0.3	4.2	0.9	370	0.03	21	210
st 11 Earthworm 10	1.0	2.3	7.0	6.7	3.9	220	0.6	27	220
st 11 Earthworm 11	1.8	12	8.6	15	3.6	100	0.6	87	200
st 11 Earthworm 12	2.0	18	4.8	10	3.3	810	0.6	74	470
st 11 Earthworm 13	1.7	5.1	3.8	9.4	3.0	23	0.6	57	270
st 11 Earthworm 14	2.6	25	0.3	11	1.1	2300	0.1	140	510
st 11 Earthworm 15	2.0	23	1.4	10	1.5	280	0.2	60	130
st 11 Earthworm 16	1.1	11	0.2	8.8	0.7	77	0.1	27	250
st 11 Earthworm 17	1.1	10	0.2	7.0	1.0	66	0.04	34	260
st 11 Earthworm 18	1.5	8	1.6	6.3	1.4	280	0.2	29	470
AM (n=18)	**1.4**	**10**	**2.5**	**9.1**	**2.3**	**380**	**0.2**	**52**	**298**
StDev	**0.5**	**6**	**3.2**	**2.8**	**2.4**	**561**	**0.3**	**29**	**106**
max.	**2.6**	**25**	**11**	**15**	**11**	**2300**	**0.8**	**140**	**510**
median	**1.5**	**8.4**	**1.0**	**8.7**	**1.5**	**185**	**0.1**	**40**	**260**
min.	**0.5**	**2.3**	**0.2**	**4.2**	**0.6**	**1.6**	**0.0**	**21**	**130**
St 5 Earthworm 1	2.9	4.1	0.3	22	1.3	0.1	0.02	0.5	250
St 5 Earthworm 2	0.4	3.2	1.3	19	2.4	0.4	0.2	0.9	280
St 5 Earthworm 3	4.8	11	1.5	23	3.4	0.7	0.2	1.6	400
St 5 Earthworm 4	0.4	2.0	0.8	22	2.0	0.3	0.1	0.8	260
St 5 Earthworm 5	2.5	1.0	1.9	13	2.8	0.7	0.2	1.3	260
St 5 Earthworm 6	2.0	3.4	0.3	20	1.1	0.1	0.01	0.3	250
St 5 Earthworm 7	2.7	9.2	0.7	27	2.3	0.3	0.1	0.9	250
AM (n=7)	**2.3**	**4.8**	**1.0**	**21**	**2.2**	**0.4**	**0.1**	**0.9**	**279**
StDev	**1.4**	**3.5**	**0.6**	**4**	**0.7**	**0.2**	**0.1**	**0.4**	**51**
max	**4.8**	**11**	**1.9**	**27**	**3.4**	**0.7**	**0.2**	**1.6**	**400**
median	**4.8**	**11**	**1.9**	**27**	**3.4**	**0.7**	**0.2**	**1.6**	**400**
min.	**0.4**	**1.0**	**0.3**	**13**	**1.1**	**0.1**	**0.01**	**0.3**	**250**
St 7 Earthworm 1	0.2	4.5	0.2	7	0.2	0.1	0.01	1.6	490
St 7 Earthworm 2	0.2	8.7	1.3	13	0.9	0.6	0.1	5.3	230
St 7 Earthworm 3	0.4	7.8	5.7	13	2.0	1.6	0.4	6.0	370
St 7 Earthworm 4	0.3	11	2.5	13	1.0	1.4	0.2	8.8	480
AM (n=4)	**0.3**	**8.0**	**2.4**	**11**	**1.0**	**0.9**	**0.2**	**5.4**	**393**
StDev	**0.1**	**2.3**	**2.1**	**2.6**	**0.7**	**0.6**	**0.1**	**2.6**	**105**
max	**0.4**	**11**	**5.7**	**13**	**2.0**	**1.6**	**0.4**	**8.8**	**490**
median	**0.2**	**8.3**	**1.9**	**13**	**0.9**	**1.0**	**0.2**	**5.7**	**425**
min.	**0.2**	**4.5**	**0.2**	**7**	**0.2**	**0.1**	**0.01**	**1.6**	**230**

St 8 Earthworm 1	1.6	1.5	0.3	10	1.1	0.4	0.04	0.7	150
St 8 Earthworm 2	1.5	2.0	0.1	9	0.6	0.1	0.001	0.4	220
St 8 Earthworm 3	2.4	4.9	0.4	10	1.3	0.5	0.05	1.2	340
St 8 Earthworm 4	1.7	1.5	0.1	10	0.4	0.1	0.001	0.3	210
St 8 Earthworm 5	3.9	4.6	0.4	10	1.5	0.9	0.04	1.6	140
St 8 Earthworm 6	0.8	1.9	0.4	11	0.9	0.4	0.03	0.9	210
St 8 Earthworm 7	2.2	1.7	0.2	6	0.7	0.1	0.005	0.5	210
St 8 Earthworm 8	1.2	1.4	0.5	10	1.2	0.2	0.02	0.6	230
AM (n=8)	**1.9**	**2.4**	**0.3**	**9**	**1.0**	**0.3**	**0.02**	**0.8**	**214**
StDev	**0.9**	**1.4**	**0.2**	**1**	**0.4**	**0.2**	**0.02**	**0.4**	**57**
max	**3.9**	**4.9**	**0.5**	**11**	**1.5**	**0.9**	**0.05**	**1.6**	**340**
median	**1.7**	**1.8**	**0.4**	**10**	**1.0**	**0.3**	**0.03**	**0.7**	**210**
min.	**0.8**	**1.4**	**0.1**	**6**	**0.4**	**0.1**	**0.00**	**0.3**	**140**

Appendix C: Total dose rate (μGy/h) received by reference organisms at individual stations.

Organism	St.1	St.2	St.2*	St.3	St.4	St.5	St.6	St.7	St.7*	St.8	St.9	St.10*	St.11*	St.12*
Amphibian	0.6	7.4	5.4	3.5	7.1	0.6	0.6	5.6	3.3	0.9	1.1	47	7.9	10
Annelid	0.4	4.2	5.4	1.8	4.0	0.4	0.3	2.5	3.8	0.6	0.4	48	9.1	11
Arthropod - detritivorous	0.4	4.1	5.4	1.8	3.9	0.3	0.3	2.5	3.4	0.4	0.4	47	8.0	10
Bird	0.3	3.3	4.3	1.5	3.1	0.2	0.2	2.0	2.6	0.3	0.3	38	6.2	8.1
Flying insects	0.4	3.8	4.9	1.7	3.6	0.3	0.3	2.3	3.1	0.4	0.3	43	7.4	9.3
Grasses & Herbs	2.1	24	20	11	23	2.1	1.9	17	14	3.3	3.2	174	34	39
Lichen & Bryophytes	12	148	79	72	147	13	13	118	64	23	24	705	153	159
Mammal - large	0.6	6.7	5.1	3.2	6.4	0.5	0.6	5.0	3.1	0.8	1.0	44	7.3	9.4
Mammal - small-burrowing	0.6	6.9	5.4	3.3	6.6	0.5	0.6	5.1	3.3	0.8	1.0	47	7.9	10
Mollusc - gastropod	0.4	4.1	5.4	1.8	3.9	0.4	0.3	2.4	3.8	0.6	0.4	48	9.2	11
Reptile	0.7	8.3	5.4	4.0	8.0	0.6	0.7	6.5	3.3	1.0	1.2	47	7.8	10
Shrub	3.1	36	36	17	35	2.8	2.8	25	22	4.1	4.3	312	53	67
Tree	0.3	3.7	1.5	1.8	3.6	0.3	0.3	3.1	1.0	0.5	0.6	13	2.4	2.9

**Based on 2017 field work, does not include ^{210}Pb and ^{210}Po data*