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Natural Radionuclides in Meadow and Pasture land in the Nordic countries

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Abstract

The amount of natural radionuclides in the environment differs between the Nordic countries as shown by previous investigations and also by this study. Agricultural areas of high natural background are predominantly found in Sweden, Southern Finland and Norway while low background areas are typical for Iceland and Denmark. Thus, this study offers possibilities for studying behaviour of natural radionuclides under different conditions such as the influence of different soil types as well as the husbandry. Furthermore the areas also enable studying environmental behaviour of radium and other natural radionuclides under seemingly steady state conditions. However, migration and accumulation of natural radionuclides in cultivated soil is complex involving various processes. Thus, a long term goal of this study was to identify the implications of some of these processes by determining the soil to plant transfer for pasture land under the different conditions that prevail in the Nordic countries. The potential health hazards due to chronic ingestion of low concentrations of naturally occurring radionuclides are fairly unknown but the results of this study may provide valuable background information for assessing these radiation risks. The aim of this project has been to gain knowledge on the status of natural radionuclides in meadow and pasture land and in grassland plants in different Nordic countries and on the transfer of these radionuclides from soil/water to man via the milk/food chain (soil- meadow/pasture grass –cow-milk). Limited data are available on the mobility and the transfer of naturally occurring radionuclides in the ecosystems of the agricultural land. In addition, information concerning the concentrations in meat and dairy products is of interest for assessing exposures of humans to natural radionuclides.

Soil characteristics are known to have significant impact on the mobility and uptake of natural radionuclides. Therefore, the uptake in relation to different soil types such as sandy, organic and/or clay soil was investigated. The use of fertilizers at some of the investigated farms has been considered but no analysis was performed on the content of natural radionuclides in fertilisers.

Key words

Natural Radionuclides & Cesium in soil, grass, milk, water, fodder, 238U, 235U, 232Th, 228Th, 226Ra, 228Ra, 210Pb, 210Po, 40K, and 137Cs

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Final Report from the NKS-B RADPAST (Contract: NKS-AFT/B(11)7)

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Foreword

The amount of natural radionuclides in the environment differs between the Nordic countries as shown by previous investigations and also by this study. Agricultural areas of high natural background are predominantly found in Sweden, Southern Finland and Norway while low background areas are typical for Iceland and Denmark. Thus, this study offers possibilities for studying behaviour of natural radionuclides under different conditions such as the influence of different soil types as well as the husbandry. Furthermore the areas also enable studying environmental behaviour of radium and other natural radionuclides under seemingly steady state conditions. However, migration and accumulation of natural radionuclides in cultivated soil is complex involving various processes. Thus, a long term goal of this study was to identify the implications of some of these processes by determining the soil to plant transfer for pasture land under the different conditions that prevail in the Nordic countries. The potential health hazards due to chronic ingestion of low concentrations of naturally occurring radionuclides are fairly unknown but the results of this study may provide valuable background information for assessing these radiation risks.

The RADPAST 2 and RADPAST 1 projects were coordinated by Klas Rosén (SLU, Sweden) with the assistance of José – Luis Gutiérrez Villanueva (postdoctoral researcher from University of Cantabria, Spain) and (to minor extent) Synnöve Sundell-Bergman.

The other participating organisations have been: STUK—Radiation and Nuclear Safety Authority, Finland (Eila Kostianen, Tuukka Turtiainen and Dina Solatie), The National Laboratory for Sustainable Energy, RISØ-DTU, Denmark (Per Roos), The Icelandic Radiation Safety Authority, IRSA, Iceland (Sigurður Emil Pálsson), The Norwegian Radiation Protection Authority, NRPA, Norway (Lavrans Skuterud and Håvard Thørring), The Norwegian University of Life Sciences, UMB, Norway (Lindis Skipperud and Jelena Mrdakovic Popic).

1. Introduction

Radionuclides from natural decay series are present in the environment in different forms and quantities. The primordial radionuclides uranium and thorium are contained in rocks and soils generally in low concentrations of 2-4 mg/kg (UNSCEAR, 2000) although their concentration may be much higher in e.g. certain felsic rocks, phosphate-rich ores or in uranium/thorium deposits.

Natural radionuclides occur in soil either as dissolved, available for exchanges, as complexes with organic matters or as pure or mixed precipitates as dissolved species in soil water, exchangeable ions in soil matrix, adsorbed species on Fe-Mn oxides, complexes with organic material, pure or mixed precipitates or covalently bound constituents of the mineral lattice. The nuclides might become dispersed in the environment through ground water exchange and/or distributed in drinking water. The radionuclides are to varying degree taken up by plants predominantly via root uptake, a process dependent on the soil characteristics, plant type and the chemical properties of the radionuclides in question (Bettencourt et al., 1988).

The aim of this project has been to gain knowledge on the status of natural radionuclides in meadow and pasture land and in grassland plants in different Nordic countries and on the transfer of these radionuclides from soil/water to man via the milk/food chain (soil-meadow/pasture grass –cow-milk). Limited data are available on the mobility and the transfer of naturally occurring radionuclides in the ecosystems of the agricultural land. In addition, information concerning the concentrations in meat and dairy products is of interest for assessing exposures of humans to natural radionuclides.

Soil characteristics are known to have significant impact on the mobility and uptake of natural radionuclides. Therefore, the uptake in relation to different soil types such as sandy, organic and/or clay soil was investigated. The use of fertilizers at some of the investigated farms has been considered but no analysis was performed on the content of natural radionuclides in fertilisers.

2. Background

Many natural radionuclides are circulating in our environment and the exposure to these radionuclides forms a natural part of our existence. The average annual radiation dose from natural background has been estimated to be about 1 mSv (UNSCEAR, 2000), which is about a quarter of the total dose we usually get per person per year in the Nordic countries (Andersson et al., 2007). Naturally occurring radionuclides in the environment originate mainly from the uranium and thorium decay chains that are contained in rocks, minerals and soil. Uranium (U) as such, is relatively abundant and in the earth's crust. The concentrations may range up to tens of milligrams per kg in rocks (igneous, sedimentary or metamorphic) and from tens to even hundreds of mg per kg in phosphate rich deposits (Qureshi et al., 2001; Langmuir, 1997) or in U ore deposits (Plant et al., 2003). Soluble uranium salts appear also naturally in drinking water (Ek B-M. et al. 2008), sometimes in relatively high concentrations in private wells.

The natural radionuclides in arable soils originate from the bedrock. The bedrock has over millions of years, transformed and eroded, and materials have been moved, and the deposited sediments have formed new rocks (Ivanovich, 1982). These have in turn undergone new conversions before they developed into the shallow, loose soil that after the last ice age in the Nordic countries, following the ongoing land uplift has begun to be used as arable land. Thus the different rock types such as granites, gneisses, diabase, limestone, basalt and shale are the parent material to the moraines and sedimented soils and hence affect their composition and properties (Ivanovich, 1982).

Generally, felsic rocks ($\text{SiO}_2 > 63\%$ by weight), such as granites, tend to have the highest uranium and thorium content which decreases with increasing base-containing (Harmsen & de Haan, 1980). Among sedimentary rocks, shale occupy, formed of clay, the same position with an average of 3-5 mg per kg of uranium and 10 - 20 mg per kg of thorium (Harmsen & de Haan, 1980). Shale with some organic soil may have even higher nuclide contents. Relatively high radionuclide content has the rock phosphate with 30 - 300 mg per kg of uranium or by 5 - 25 mg per kg of thorium. Lowest content has the sandstone with 0.5 to 1.5 mg per kg respectively. Apparently, soils can display a wide variety of natural radionuclide content 1 to 12 mg / kg or ppm.

Arable soil is often of local origin and very heterogeneous with regards to grain size distribution, humus content and chemical properties (Eriksson et al., 2011). Thus it follows that the levels of natural radioactivity and its sequestration in soil should vary in the same proportion. Examples of this might be where the water-based materials have been transported far and had time to be separated before sedimentation. The broad surface of the clay soil and their forestry areas are clear examples. The content of organic matter which depends on the soil progression from peatland to soils during the course of history, contributes to the variation of the surface soil properties. (Eriksson & Rosén, 2000).

The solubility of the radionuclides of the uranium and thorium decay chains in soil is dependent on several factors such as: pH, redox potential, temperature, soil texture, organic and inorganic compounds, moisture and microbial activity (Dupré de Bouloisa et al., 2007). Transport of uranium by ground water usually occurs under oxidizing conditions either as ion or as complex, where maximal sorption occurs on natural material at pH 5 to 8 (Gavrilescu et

al., 2009). Under oxidizing conditions uranium forms very stable and soluble carbonate complexes in soil and water. This is one of the reasons why uranium deficiency is often recorded on the topmost soil layers. However, in presence of iron substrates uranium becomes scarcely mobile. The tendency for sorption to organic matter can result in enrichment in the soil in peat bogs and other organic soils which were subsequently used for farming.

Contrariwise, thorium becomes more soluble at low pH (2-3) and as organic complexes, (Porcelli D, 2008). In natural water systems thorium may be significant but the oxidizing conditions and low pH rarely exist in natural water systems.

Changing oxidation/reduction conditions can influence the mobility of Ra (Chen et al., 2005). Radium is mobile in oxidizing conditions below pH 3 and somewhat mobile in circumneutral pH range provided that iron substrates do not abound. Radium may be sorbed by clay minerals and organic matter in the environment.

According to Vandenhove et al., (2009) a significant relationship between soil K_d (soil vs. water) and pH exists for natural radionuclides due to the abundance of complexing anions in soil. Clay soils seem to show the highest K_d due to higher sorption capacities. There are several possible routes of uptake of ^{210}Po and ^{210}Pb in plants such as direct uptake from soil including the ground water, uptake in the foliar of the plants from the radioactive decay of ^{226}Ra or ^{222}Rn previously adsorbed by the plant and foliar uptake of ^{210}Po and ^{210}Pb deposited on the leaves. The latter processes are expected to be of minor importance except for grass (inedible plant parts).

The transfer of naturally occurring radionuclides from soil to man has not been studied to the same extent as radionuclides of anthropogenic origin although there is an increasing interest from an environmental risk perspective (ICRP 103, 2007). Concerning human exposures the primary pathways for these radionuclides entering the human body are beside inhalation, ingestion of contaminated food and water. To assess the exposures from natural radionuclides it is necessary to determine the rate of transfer from soil to plant, and to the essential food components of the human food chain. The soil to plant transfer factor (TF) as well as the rate of transfer from animal food to milk and meat is important to determine for assessing the doses from known concentrations in the environment. The TF's are highly variable and dependent on e.g. soil characteristics, plant type, the plant part concerned, climate conditions and the physico-chemical form of the elements.

IAEA has recently published a handbook of parameter values for the prediction of radionuclide transfer in terrestrial and freshwater environments (Technical Reports Series 472 IAEA, 2010) to be used worldwide for various risk assessments. However it was concluded that the number of observations concerning the naturally radionuclides U, Ra, Pb, Po and Th was still limited, particularly for clay and organic soils and that large uncertainties were associated with the K_d ¹ estimates (Pröhl 2009; Vandenhove et al., 2009).

For natural nuclides, one should bear in mind that root uptake and transfer can take place from

¹ Solid-liquid partition coefficient, K_d , is the ratio of the concentration of a nuclide on a solid phase (soil or sediment) divided by the equilibrium concentration in the contacting liquid phase (water). Its unit is generally in the form of $l \cdot kg^{-1}$. The equilibrium concentration in the contacting liquid phase (water). Its unit is generally in the form of $l \cdot kg^{-1}$.

all over the soil profile available for plant roots (rhizosphere). The soluble nuclide fraction found in the soil solution is quite different from the insoluble fraction in the soil but the nuclide distribution of both the two fractions affects the availability and concurrently the transfer to the plant. The uptake of radionuclides via the roots depends not only on the availability and the nuclides present in the soil but also on the humidity and the presence of other competing ions. Earth worms may also influence the process by mixing activities. Differences in the mobility of ions, the binding and the transport upwards in the plant are processes that affect the uptake of radionuclides in the plants (Jarvis et.al. 2010).

Farm animals generally form a barrier against the transfer of natural radionuclides from soil via animal feed and animal products into the human diet by low uptake. Concerning radium only one or a few percent will be transferred to the meat section, although this may be about 12 times greater in weight (Technical Reports Series 472 IAEA, 2010). For uranium and thorium the corresponding figures might be 3-15 percent in the meat section (Linsalata et al., 1989, 1991). Szerbin & Popov (1988) found for example in cattle that 9-18% of radium and 3.5 to 12% of thorium used in the feed was recovered in the carcass at slaughter (Eriksson & Rosén, 2000).

Previously Eriksson and Rosén have made an investigation on the concentrations of natural radionuclides in soil and plants in agricultural land (Eriksson & Rosén 2000). The samples were taken from farms situated in areas of average natural background but no detailed information was provided on the geological characteristics of the areas. The average values found in cultivated soils from private farms from three different regions of Sweden were 39 Bq kg⁻¹ (south of Sweden), 51 Bq kg⁻¹ (west of Sweden) and 78 Bq kg⁻¹ (middle of Sweden) for ²²⁶Ra. The values found for ²³²Th were 46 Bq kg⁻¹ (south of Sweden), 56 Bq kg⁻¹ (west of Sweden) and 70 Bq kg⁻¹ (middle of Sweden).

A literature survey on the occurrence of naturally radionuclides in food has been carried out (Myllymaa, 2003) in Finland. This survey was targeted on reindeer and fish but was later expanded to naturally occurring radionuclides and fall-out radionuclides in cereals and grains. Recently another literature survey on the content of natural radionuclides in vegetables and tubers has started in summer 2010.

Measurements of the concentrations of ²¹⁰Po and ²¹⁰Pb in the soil, plants, wild berries and mushrooms of Finnish forest ecosystems have been undertaken by Vaaramaa et al., 2009 and 2010. The mean total inventory in the soil profile, up to 20 cm, of ²¹⁰Pb was 4.0 kBq m⁻² and ²¹⁰Po 5.5 kBq m⁻², the organic soil layer containing 45% of the total inventory of both nuclides. Survey on ²¹⁰Pb and ²¹⁰Po contents in Finnish cereals revealed 2–7 times higher contents than the reference values given by UNSCEAR, (2000) (Turtiainen et al., 2011).

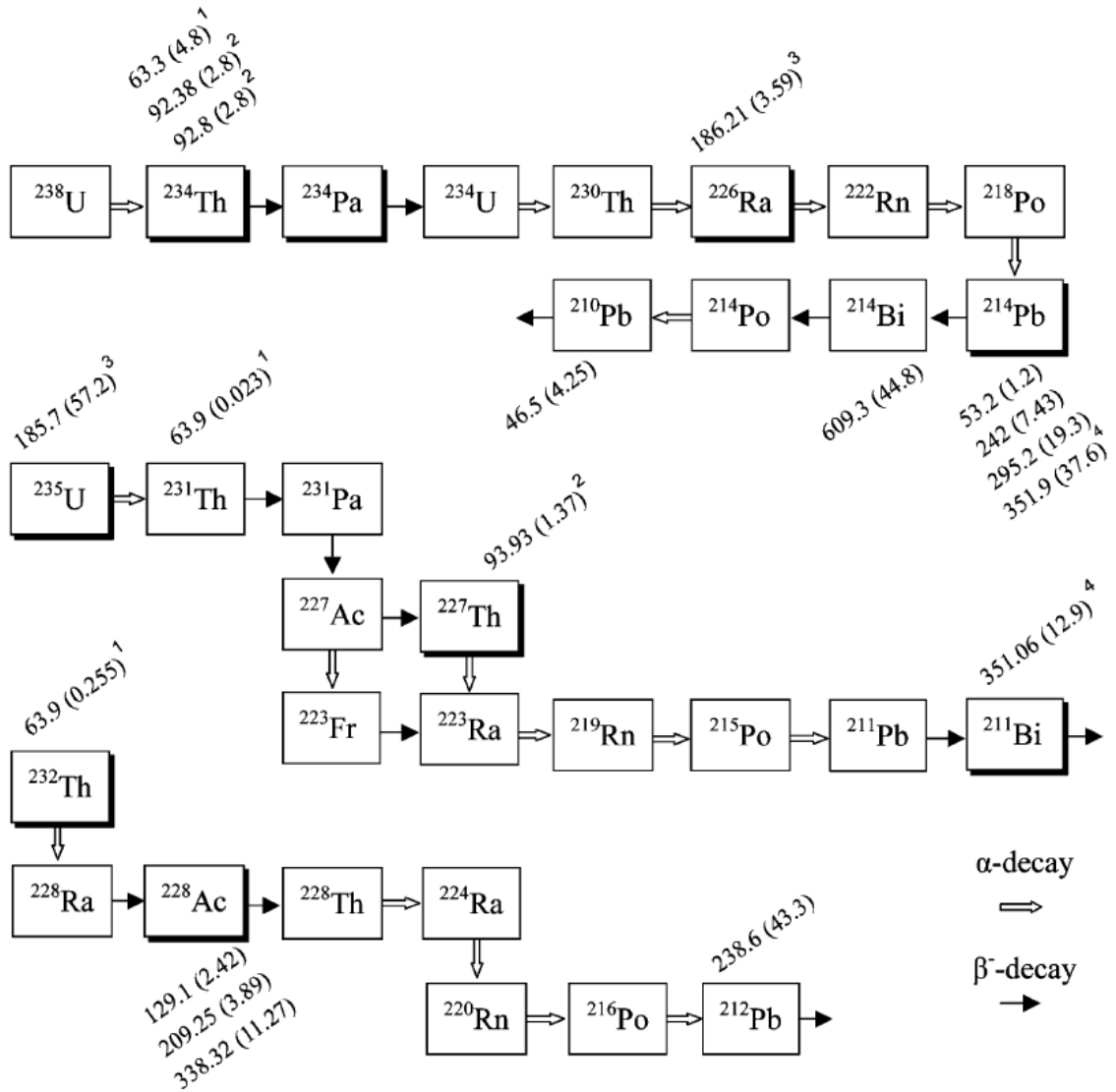


Figure 1: Partial diagram of the uranium, actinium and thorium decay series. The energy (in keV units) and percent emission probability (quoted in parentheses) of the major γ -rays are indicated next to the corresponding nuclide, while emissions recorded in the same spectral peak are marked with superscripts.

3. Material and methods

3.1 Description of sampling work

Sampling was performed in pasture and meadow lands in Sweden, Finland, Denmark, Iceland and Norway during summer/autumn in 2010 and 2011. The number of sampled areas in each country ranged from 2 in Denmark to 4 in Finland. The locations of sampling sites are shown in Figure 2 and Table 1.



Figure 2: Sampling areas in RADPAST project in the Nordic countries.

The grazing season of animals were selected as sampling period. This period is region/country dependent, but is generally between June and October.

In all the countries, soil and grass was sampled in accordance with the scheme shown in Figure 3 slightly modified from that proposed by Rosén et al., (1999).

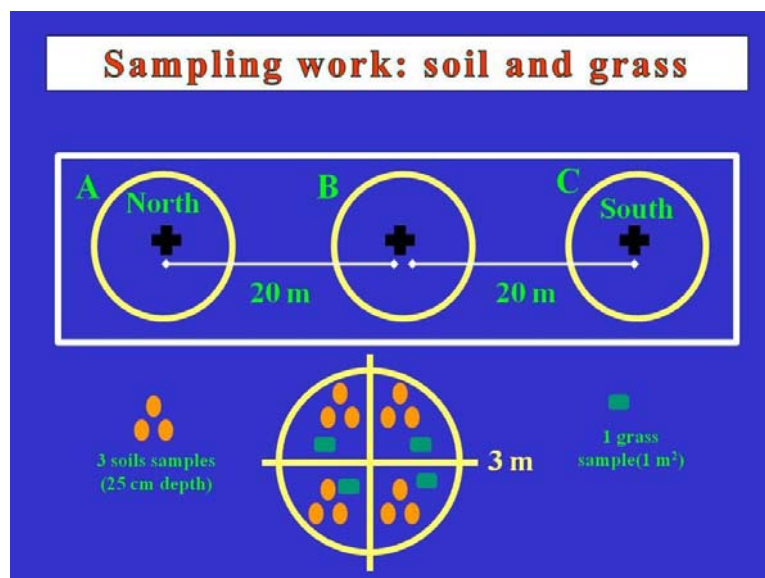


Figure 3: Sampling scheme for soil and grass.

As evident from the scheme, each sampling area consisted of 3 circles with a radius of 3 m. Each circle was subdivided in four sectors, and a composite soil sample based upon 3 plots from each sector was prepared. Soil depth was 25 cm or less; the sampler is shown in figure 4. Moreover, a composite grass sample was made based on 1 m² areas in each sector. All in all, 3 composite soil samples and 3 composite grass samples were taken from each sampling area. The centres of the circles were separated by 20 m, except in the case of Iceland and Norway, where the distance between circles was around 1 km because of the large cow grazing areas (Norway) and separate fields were used at a given farm (Iceland). Bulk samples up to 25 cm depth were collected using similar sampler device in each country (see figure 3). At each sampling site water and/or milk were sampled if possible.



Figure 4: Soil sampler used in RADPAST project. All participants used the same instrument for the sampling work.

Information collected by each country about the sampling sites was included in the study: GPS coordinates; date of sampling; grazing area; number of animals grazing (cows or sheep); type of breed; type of farm; soil characteristics and additional information considered of interest for interpreting the results. In **Table 1** a summary of the information is given for each participant.

Table 1. Description of sampling sites in RADPAST project

Countries	Site	Place	GPS coordinates	Date of sampling	Grazing area (ha)	Grazing period	Number of grazing animals	Type of breed	Milk production (l/cow/year)	Type of farm	Other information
Sweden (SLU)	S1	Hallen	63° 10' 5'' N, 14° 7' 13'' E	10-8-2010	40	1-6-2010/1-9-2010	120	Swedish lowland livestock (slb), cow	9500	intensive ²	ecological farming
	S2	Skogsvallen	60° 10' 1'' N, 17° 11' 19'' E	22-10-2009	2	15-5-2010/15-9-2010	no animals	-	-	extensive	not ploughing since 1986
	S3	Möjsjövik	59° 56' 60'' N, 17° 13' 60'' E	20-10-2009	3	15-5-2010/15-9-2010	10	horses	-	extensive	not ploughing since 1986
	S4	Lövstalöt	59° 57' 10'' N, 17° 35' 0'' E	8-10-2009	50	15-5-2010/15-9-2010	140	Swedish lowland livestock (slb), cow	9000	intensive	ecological farming
Finland (STUK)	F1	Hyvinkää	63° 38' 46'' N, 25° 53' 48'' E	22-6-2010	3	27-5-2009/	16	mainly Ayrshire, cow	9800	intensive	fertilised annually
	F2	Myrskylä	60° 35' 49'' N, 24° 48' 29'' E	10-6-2010	2	25-5-2010/	31	finish Ayrshire and Holstein, cow	8900	intensive	ploughed 2005, nitrogen fertilizer in spring 2010
	F3	Rovaniemi (Jaatila)	66° 13' 37'' N, 25° 20' 10'' E	8-6-2010 and 30-8-2010	3	16-6-2010/	30	Ayrshire, cow	10000	intensive	ploughed in 2008, fertilizer in 2010

² Intensive farms use artificial fertilizers and extensive farm do not use them.

	F4	Sotkamo (Tuhkakylä)	63° 55' 59" N, 28° 6' 50" E	9-6-2010 and 27-8-2010	10	10-6-2010	10	Holstein- Friesian, cow	8000-9000	intensive	ploughing annually, depth 17-21 and fertilized
Denmark (RISØ- DTU)	D1	Hyllinge	55° 42' 52" N, 11° 52' 18" E	30-5-2010	10	-	20	-	-	-	ecological farming
	D2	Mosekær	56° 25' 57" N, 10° 13' 09" E	30-8-2011	35		57	Nordic Red	11140	Ecological	D2
Iceland (IRSA)	I1	Stóra Ármót	63° 59' 19" N, 20° 55' 44" W	10/8/2010	30	1-6- 2010/30-9- 2010	45	Icelandic, cow	6800	extensive	-
	I2	Möðryvellir	65° 45' 56" N, 18° 15' 9" W	19/8/2010	20	1-6- 2010/30-9- 2010	40	Icelandic, cow	6000	extensive	-
Norway	(NRPA)	N1	Haalien	61° 19' 46" N, 8° 47' 3" E	1-9-2010	700	1-7- 2010/1- 9-2010	9	Norwegian red, cow	6000	extensive
		N2	Sanddalen	61° 19' 57" N, 8° 51' 40" E	2-9-2010	100	1-7- 2010/1- 9-2010	10	Norwegian red, cow	7300	extensive
		N3	Langsetra	62° 6' 41" N, 10° 22' 58" E	3-9-2010	-	1-7- 2010/1- 9-2010	13	Norwegian red, cow	5800	extensive

UMB	N4	Skippervold	59° 16' 73'' N 09° 17' 71'' E	28-5- 2011	3	1-7- 2010/1- 9-2010	<10	cow	-	extensive
	N5	Gamlevegen	59° 16' 36'' N 09° 18' 68'' E	29-5- 2011	4	1-7- 2010/1- 9-2010	<10	cow	-	extensive
	N6	Bjørndalen	59° 16' 08'' N 09° 18' 22'' E	29-5- 2011	-	1-7- 2010/1- 9-2010	<20	cow	-	extensive

3.2 Methodology

All participants have used similar methods for the treatment of the samples. The soil and pasture grass samples were dried at 105° C (in some soil samples 30-40 °C was used for drying) until constant weight and then homogenised using a mill or blender. All the soil samples were sieved (Ø 2 mm grain size).

In the case of water and milk samples, the sample preparation was quite similar. Milk samples were freeze dried or evaporated and subsequently ashed in a furnace by slowly increasing temperature to 450 °C (24 hours). This was done in order to reduce total volume of sample, and thereby lowering radionuclide detection limits. The water samples were also evaporated and ashed at 450 °C. In some cases no pre-treatment was applied to liquid samples resulting in higher detection limits with gamma spectrometry.

Table 2. List of radionuclides analysed by each participant

Radionuclide	Sweden (SLU)	Finland (STUK)	Denmark (RISØ-DTU)	Iceland (IRSA)	Norway (NRPA)	Norway (UMB)
²³⁸ U	×	×	×		×	×
²³⁵ U		×		×		
²³² Th						×
²²⁸ Th			×		×	
²²⁶ Ra	×	×	×	×	×	×
²²⁸ Ra	×	×	×	×	×	×
²¹⁰ Pb	×	×	×			
²¹⁰ Po		×				×
⁴⁰ K	×	×	×	×	×	×
¹³⁷ Cs	×	×	×	×	×	

Table 2 shows a summary of the different radionuclides analysed by each participant. Measurement times varied from 1 to 3 days. Activity concentrations of ¹³⁷Cs and ⁴⁰K were determined directly, whereas the other radionuclides were determined using radioactive equilibrium: ^{234m}Pa for determination of ²³⁸U or to use 186 keV doublet from which ²²⁶Ra was subtracted (*STUK*), ²¹⁴Bi and ²¹⁴Pb for ²²⁶Ra and ²²⁸Ac for ²²⁸Ra. In other cases the use of 186 keV doublet from which ²²⁶Ra is subtracted was used. The concentrations of ²³⁸U, ²³⁵U, ²²⁶Ra, ²²⁸Ra, ²²⁸Th, ²¹⁰Pb, ⁴⁰K and ¹³⁷Cs were analysed by different methods and a detailed description of the methods can be found in the Appendix. Table 3 indicates which method has been used by each participant in the project.

For the particular case of the analysis of ²¹⁰Po, the samples were spiked with ²⁰⁹Po tracer and the solid samples digested by using micro wave digestion. Before deposition, hydrazine

monohydrochloride and ascorbic acid were added. Polonium was spontaneously plated on silver discs. The activity of ^{210}Po was measured by alpha spectrometry. All samples were analysed before 6 months had gone from the sampling date. The solution remaining from the first deposition was stored about 6 months to allow the in-growth of ^{210}Po from ^{210}Pb . The second ^{210}Po deposition was then carried out and its ^{210}Po activity measured. The ^{210}Pb content of the samples was calculated from the second deposition and the results of the first deposition were corrected for the radioactive decay between the time of sampling and analysis and the in-growth of ^{210}Pb during sample storage..

Table 3. Summary of methods used by each participant in the project

Participant	Gamma spectrometry	Alpha spectrometry	Mass spectrometry
SLU	<input type="checkbox"/>		
STUK	<input type="checkbox"/>	<input type="checkbox"/>	
RISØ-DTU	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>
IRSA	<input type="checkbox"/>		
NRPA	<input type="checkbox"/>		
UMB	<input type="checkbox"/>		<input type="checkbox"/>

3.2.1 Radon in outdoor air

Three soil samples (each consisting of 2–12 soil cores) were collected at each sampling site F1–F4. The sample volume was recorded for each sample. At the lab, fresh weight, dry weight (oven dried in 105 °C) and weight of the stones were measured for each sample. Samples from each sampling site were partly pooled without the stones and two 100-gramme aliquots from each sampling site were used for analyzing density of radon flux. Density of radon flux at sites I1 and I2 were determined in aliquots from four separate soil samples collected at each site.

The analytical method in order to determine radon emanation rate per mass (G_m) was based on direct absorption of radon into polar liquid scintillation cocktail which has been described in detail by Turtiainen (2009). The two aliquots representing one sampling site were adjusted to 5 and 25% moist content (w/d.w.) by RO-purified water. The bulk density (ρ_b) of each sample was measured from the oven dried (105 °C) mass of the sample and the original soil core volume (Blake and Hartge, 1986a). Particle density (ρ_s) of the soil sample was determined by volumetric flask method described by Yu et al. (1993). Total porosity (p_t) of the soil sample was calculated according to the following equation:

$$p_t = 1 - \frac{\rho_b}{\rho_s} \quad (1)$$

The fraction of the porosity that is water filled (m) was calculated from the difference of fresh and dry weight (without mass of stones) and total porosity.

Activity concentration of ^{226}Ra (c_{Ra}) was determined by gamma spectroscopy and emanation coefficient (f) was calculated according to the following equation:

$$f = \frac{G_m}{\lambda_{Rn} c_{Ra}}, \quad (2)$$

in which λ_{Rn} is the decay constant of radon. Radon emanation rate per mass (G_m) was temperature corrected by a linear correlation factor of 0.0021/K suggested by Iskandar *et al.* (2004). Radon concentration in soil in the absence of radon transport (c_∞) was assessed as described by Nazaroff *et al.* (1988):

$$c_\infty = G_m \rho_s p_t (1 - p_t)^{-1} (m[k_t - 1] + 1), \quad (3)$$

in which k_t is the partition coefficient (Ostwald coefficient) for radon between water and air. The values of k_t were obtained from Clever (1988). Effective radon diffusion coefficient (D_e) necessary to assess density of radon flux (J_D) was calculated according to Rogers and Nielson (1991):

$$D_e = D_0 p_t e^{-6p_t m - 6m^4 p_t}, \quad (4)$$

in which D_0 denotes radon diffusivity in open air ($1.1 \cdot 10^{-5}$ m²/s). The density of radon flux was calculated according to the following equation:

$$J_D = \sqrt{D_e \lambda_{Rn} \rho_s} \frac{G_m}{\lambda_{Rn}} [1 - p_t (1 + m(k_t - 1))], \quad (5)$$

which has been modified from that presented in UNSCEAR (2000) to account for soil moisture content. This was accomplished by replacing total porosity p_t by $p_a + K_t \times p_w$, in which p_a is the volumetric ratio of air filled pores to total volume and p_w is the ratio of water filled pores to total volume.

3.2.2 External dose

The external exposure was assessed at the pasture lands at sites F1–F4. The representative radionuclide concentrations were averaged over three soil samples collected at each site and converted to represent activity per wet weight at the sampling date. Absorbed dose rate calculations were carried out with WinMat computer programme, which is based on the Markkanen model and Materia programme (Markkanen, 1995). The programme operates in Cartesian geometry and calculates absorbed dose rates from homogenous layers. Two successive layers can be placed in order to calculate attenuation, for example, by air on top of soil. Scattering is taken into account by a build-up factor calculated from Berger–Seltzer expression that has been expanded to cover two successive layers. Interpolation of photon mass-attenuation and energy-absorption coefficients is carried out from the logarithms of the tabled values. The photon mass-attenuation and energy-absorption coefficients for elements, water and air were taken from Hubbell (1981) and those for the soil components (stone, clay) had the elemental composition reported by Turtiainen and Weltner (2007) and Turtiainen *et al.*, (2008). Elemental composition 5% nitrogen, 7% hydrogen and 88% carbon was chosen for the organic component (Poirier *et al.*, 2005). These data were used for calculating photon energy absorption and mass attenuation curves accordingly for each soil (Table 4).

Table 4. Data used for absorbed dose rate assessments

Sampling site	ρ_{wet} [kg/l]	water	stone	clay	organic
F1	1.49	0.22	0.70	0.04	0.04
F2	1.33	0.35	0.43	0.03	0.19
F3	1.46	0.25	0.55	0.16	0.04
F4	1.08	0.19	0.73	0.04	0.04

In order to speed up the integrations, selected X-rays and gamma rays were limited to energies above 30 keV with photon intensities greater than 0.005% amounting to a total of 337 photons for the Th series and 430 photons for the U series. The actinium series was assumed to exhibit 4.66% of the activity of the U series (natural $^{235}\text{U}/^{238}\text{U}$). Radon emanation was not taken into consideration which may lead to about 10% overestimation. Two photo peaks (283 and 662 keV) were used for ^{137}Cs . Information on energies and intensities were retrieved from the Lund/LBNL Nuclear Data Search (Chu *et al.*, 1999). The calculated absorbed dose rates were converted to effective dose rates by 0.7 Sv/Gy recommended by UNSCEAR, (2000). The cosmic radiation component was also taken from UNSCEAR, (2000).

3.2.3 Absorbed dose rate

The estimation of the absorbed dose rate (ADR) from external exposures is done using the dose conversion factors of IAEA (UNSCEAR, 2000). Not all the participating organisations have reported values of the activity concentrations for the same radionuclides with the exception of ^{226}Ra . Thus it is necessary to choose one radionuclide from each of the corresponding natural decay series in order to give an estimation of the absorbed dose rate in air from the activity concentrations in soil.

Not all the radionuclides contribute in the same way to the absorbed dose (Saito, 1995). In the case of the natural series of ^{238}U , one of the most important contributors to the absorbed dose is ^{226}Ra but also the radon daughters ^{214}Pb and ^{214}Bi if only the gamma emitters are considered. The case of the radon daughters with short half lives have not been considered in the evaluation of the absorbed dose rate. The same applies to the natural series of ^{232}Th for the case of ^{224}Ra and the thoron daughters. In this case, ^{232}Th was chosen for the evaluation of the absorbed dose-rates. The radon and thoron short living daughters are of high interest in the case of the calculation of the inhalation dose which is out of the scope of this report.

The equation 1 shows how the absorbed dose has been calculated:

$$ADR = 0.0417 \cdot {}^{40}\text{K} + 0.462 \cdot {}^{238}\text{U}_{\text{series}} + 0.604 \cdot {}^{232}\text{Th}_{\text{series}}, \quad (6)$$

where the numerical factors are the conversion factors from Bq kg⁻¹ to nGy h⁻¹.

3.2.4 Average effective dose rate

In the estimation of the average annual effective dose rate outdoors due to natural radionuclides, the equation 2 has been applied (UNSCEAR, 2000):

$$AEDR = ADR \cdot 8760 \cdot 0.2 \cdot 0.7 \cdot 10^{-6}, \quad (7)$$

where ADR is the absorbed dose rate, 8760 is the numbers of hours in a year, 0.2 is the outdoor occupancy factor, and 0.7 is the conversion coefficient.

4. Results

4.1 Soil characteristics

The reported soil parameters are compiled in *Table 5*. At farms F1–F4, the soil parameter determination was performed on a soil sample pooled from three sampling circles. At farm D1, soil analyses were performed on samples from each sampling circle. The reported nutrient values are not compatible since at farms F1–F4 the nutrients were determined by ammonium acetate leaching method that reveals the exchangeable fraction of the nutrient that is available for root uptake. At farm D1 total dissolution and subsequent MS analysis was performed. The nutrient content of the soil at farms F1 and F2 were generally high or close to the optimum. Only phosphorous content at farm F2 was classified as satisfactory. At farm F3, the nutrient content was generally satisfactory and at F4 passable or poor.

Different standard methods were used to determine the nutrient contents. Therefore the data cannot be easily used for testing hypotheses (for example, K vs. Cs-TF) even if the values are expressed in the same units. The amount of nutrients in the leaching/extraction solution depends on the method used (leaching solution chemical composition, its concentration, pH, soil-to-solution ratio, temperature, agitation, and contact time).

Table 5. Soil parameters at sampling points. Organic content is expressed in % and CEC in meq/100 g. The rest of parameters are in units mg/100 g

	P	K	Ca	Mg	S	organic content	CEC	Soil type
S1	6.9	8	312.2	17.5	2.3	12		loam
S2	2.7	19.1	260.9	21	1.3	10		silty clay
S3	3.9	36.2	959.6	32.4	8.6	82		peat
S4	2.1	8.5	119.2	9.7	2.1	8		clay loam
F1	2.9	36	171	15	1.0	3-5.9	16.2	fine sandy soil
F2	0.6	37	492	34	2.9	20-39.9	50.3	mull
F3	0.8	10	84	25	2.4	3-5.9	11.1	fine sandy soil
F4	0.7	8	32	7.0	3.9	3-5.9	9.65	fine sandy till
D1								
D2								
I1								
I2								
I3								
N1								
N2								
N3								
N4	7.8	5.2	522	19.7	NA	6.4	27.9	
N5	11.9	7	392	60	NA	21.7	31	
N6	8.3	5.5	450	22.5	NA	26.1	31.4	

4.2 Activity concentration in soil, grass, milk, water, fodder and food supplements (natural radionuclides)

4.2.1 Activity concentration in soil

Reported activity concentrations in soils are compiled in Table 6. Generally the measurements from the three sampling circles per farm resulted in similar activity concentrations and the standard deviation of the values was within the uncertainty reported for individual measurements.

Uranium in the environmental samples is generally hard to determine via gamma spectrometry (GS). The normal procedure of using the 1001 keV gamma peak ($I_\gamma = 0.84\%$) of ^{234m}Pa yields in relatively high detection limits. One of the other options is to use the 186 keV gamma doublets of ^{226}Ra and ^{235}U . Uranium can be calculated if the activity concentration of ^{226}Ra is determined from other gamma peaks (such as those of ^{214}Pb and ^{214}Bi). This way lower detection limits can be achieved. It is however best to determine uranium with alpha spectrometry (AS) or mass spectrometry (MS). ^{232}Th has only two weak gamma peaks ($I_\gamma < 0.3\%$) exhibiting low energy ($E_\gamma < 141$ keV) and hence, GS is not a viable method for its determination. However it is possible to measure ^{232}Th using its secular equilibrium with ^{228}Ac and by means of AS or MS.

Table 6. Mean activity concentrations and standard deviation (Sd) in soil samples at the 19 sites. The unit is $\text{Bq} \cdot \text{kg}^{-1}$ (dw). The maximum, minimum and geometric mean (Max, Min, GM) values are given. The values correspond to the radionuclides analysed of the natural decay chains. Values in *italic* mode indicate that the corresponding result is below the MDA

	U-238	Sd	Ra-226	Sd	Pb-210	Sd	Po-210	Sd	Th-232	Sd	Th-228	Sd	Ra-228	Sd	K-40	Sd	U-235	Sd
S1	350	173	72	40	65	7							31	2	681	39		
S2	< 236		65	6	< 121								63	3	1040	26		
S3	672	187	62	29	148								47	10	110	24		
S4	259		39	1	< 34								51	0.3	817	11		
F1	86	11	57	5						42	2	44	3	673	47	4	0.5	
F2	142	23	80	19						58	7	59	7	467	152	7	1	
F3	29	6	28	3						28	3	32	3	628	15	1	0.3	
F4	21	2	17	2	25	1				21	1	23	2	539	34	1	0.1	
D1	19	1	25	4									30	6				
D2	17	1	17	3	28					21			23					
I1			9	8									12	1	99	12	2	0.9
I2			18	6									10	4	123	60	3	0.4
N1			6	1						3	0.4	3	0.3	377	82			
N2			7	3						4	0.5	4	0.3	423	29			
N3	34	17	25	4						24	7	25	6	544	43			
N4	31	5	30	9					269	125			264	158	276	32		
N5	127	5	57	12					647	42			679	4	247	29		
N6	70	0.4	41	41					509	20			527	44	514	65		
Max	672		80		148				647		58		679		1040		7	

Min	19		6		25				269		3		3		99		1	
GM	72		28		51				446		18		38		387		2	

As shown, the geometric mean concentrations of ^{226}Ra and ^{228}Ra were 29 and 17 $\text{Bq} \cdot \text{kg}^{-1}$ respectively; the maximum value were 80 $\text{Bq} \cdot \text{kg}^{-1}$ (F2) and 148 $\text{Bq} \cdot \text{kg}^{-1}$ (S3) respectively and the minimum values were 6 $\text{Bq} \cdot \text{kg}^{-1}$ (N1) and 25 $\text{Bq} \cdot \text{kg}^{-1}$ (F4) respectively.

In most cases, the two isotopes of radium exhibited similar activity concentrations in soils. However, at farms I1 and I2 the ^{228}Ra concentration in soils remains constant, about 10 $\text{Bq} \cdot \text{kg}^{-1}$ whereas the ^{226}Ra concentrations vary considerably 9–18 $\text{Bq} \cdot \text{kg}^{-1}$. At farm S1 the ^{228}Ra concentration was about 31 $\text{Bq} \cdot \text{kg}^{-1}$ and ^{226}Ra about 72 $\text{Bq} \cdot \text{kg}^{-1}$. Nevertheless, soils at farms D1, F1, F3, F4, N3 and S4 can be considered to represent average activity concentration range of 20–40 $\text{Bq} \cdot \text{kg}^{-1}$. The soils at farms I1, I2, N1 and N2 contain notably lower levels of natural radioactivity than the median values, only a few $\text{Bq} \cdot \text{kg}^{-1}$. The soils at farms F1, F2, S1, S2, S3, S4, N4, N5 and N6 contain the highest levels of natural radioactivity. It is interesting to notice that N4, N5 and N6 present an activity concentration of ^{228}Ra (thorium chain) one order of magnitude higher than the rest of the farms.

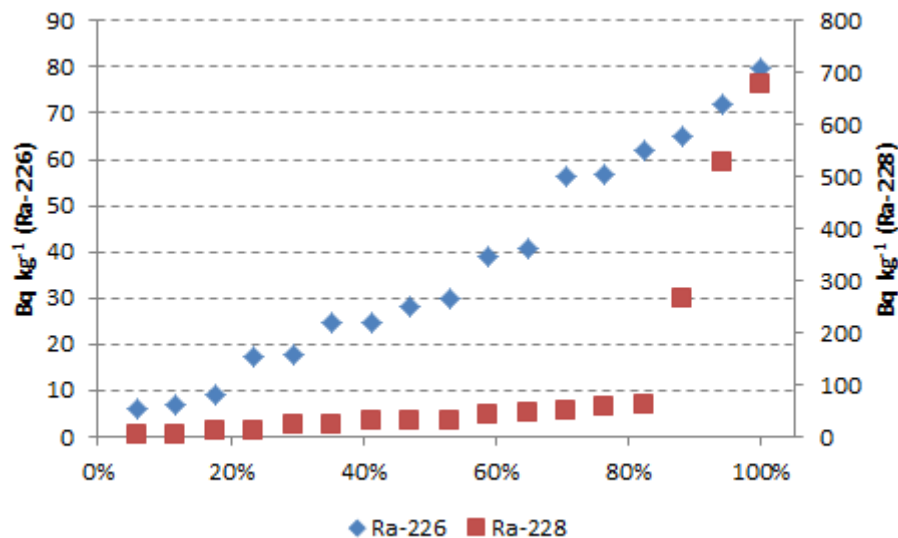


Figure 5. Distribution of activity concentration of radium isotopes in soils. Geometric mean is 29 $\text{Bq} \cdot \text{kg}^{-1}$ for ^{226}Ra and 39 for ^{228}Ra $\text{Bq} \cdot \text{kg}^{-1}$. Percentages represent the activity distribution in all the studied samples.

As seen in Figure 5 the farms selected for this study represent a wide-range of the levels of natural radioactivity in the Nordic countries

4.2.2 Activity concentration in grass

The reported activity concentrations in grass are compiled in Table 7. The results show significantly higher variance within the three sampling circles per farm than those of the soil samples. This is mostly due to higher uncertainties related to analysing low activity concentrations. The high values

corresponding to the samples S1 – S4 are under investigation at the moment of the production of this report and they will be measured by ICP-MS in order to confirm or modify those values.

Table 7. Mean activity concentrations with standard deviations (Sd) in grass samples from the different farms. The unit is $\text{Bq}\cdot\text{kg}^{-1}$ (dw). Values in *italic* indicate that the corresponding result falls below the MDA

	U-238	Sd	Ra-226	Sd	Pb-210	Sd	Po-210	Sd	Th-232	Sd	Th-228	Sd	Ra-228	Sd	K-40	Sd	U-235	Sd
S1	526	111	< 29		203	16							24	3	856	118		
S2	561	75	220	200	250	25							33	1	543	105		
S3	< 521		319	216	317								32		372	24		
S4	1050		260	163	< 233								69	7	1744	1819		
F1	4	1	3	1	12	1					0.2	0.1	2	0.3	984	112	0.2	0.1
F2			2	0.8							1	1	3	1	1012	320		
F3			4	4							1	0.3	4	1	962	243		
F4			3	1	24	7					0.3	0.2	3	3	1103	179		
D1	0.1	0.04	1	0.3									1	0.3				
D2	0.2		2		22						3							
I1													1	0.2	528	141	0.9	0.4
I2													1	0.3	470	161	1	0.1
N1			3	1											164	45		
N2			4	1											242	90		
N3			6	1							1	0.2	2	0.5	377	213		
N4	5	0.3							23	1								
N5	4	0.7							16	4								
N6	5	2							0.3	0.1								
Max	1050		319		317				23		1		69		1744		1	
Min	0.1		0.7		12				0.3		0.2		1		164		0.2	
GM	11		9		68				5		1		5		595		1	

4.2.3 Activity concentration in milk

The reported activity concentrations in milk samples were even more fragmented than those in grass (Table 8). ^{228}Th , ^{232}Th , and ^{235}U concentrations could not be determined in any of the samples³ but ^{40}K . Fortunately, the ^{226}Ra concentration was reported from eight farms, which thus provides data for assessing the radium transfer to milk. Uranium concentration in milk from farm D1 was analysed with MS and its concentration was $6.4 \cdot 10^{-5} \text{ Bq} \cdot \text{kg}^{-1}$. This shows that MS is a pre-eminent tool for analysing low levels of activity concentrations (uranium). Again, ^{210}Pb concentrations were below detection limits of GS except in one of the samples although with a big uncertainty.

³

Actually, the first milk samples collected at farms F3 and F4 contained 0.11 ± 0.03 and $0.09 \pm 0.03 \text{ Bq} \cdot \text{kg}^{-1}$ (fw) of ^{228}Ra , respectively. These values, however, cannot be utilized as such for grass-to-milk F_m of CR assessment because the cows were not grazing at the time of sample collection.

Table 8. The activity concentrations in milk samples collected at different farms. The unit is Bqkg⁻¹ (fw). Samples F1-F4 are expressed in Bq/l fresh weight for U-238. Those radionuclides with no reported activity concentrations are not shown

	U-238	Sd	Ra-226	Sd	Pb-210	Sd	Po-210	Sd	Ra-228	Sd	K-40	Sd
S1	11		1E-03		3				0.3		32	
S2												
S3												
S4												
F1	7E-04		0.06	0.01	< 0.3				< 0.07			
F2	7E-04		0.02	0.01	< 0.1				< 0.03			
F3	3E-04		0.02	0.01	< 0.1				< 0.1			
F4	2E-04		0.3	0.01	< 0.2				< 0.1			
D1	6E-05		2.8E-03		5.7E-03							
D2	1E-04		4E-03		4E-03		2E-02					
I1									< 0.1			
I2												
N1	< 2		0.3	0.03					< 6E-02			
N2	< 2		0.2	0.04					< 6E-02			
N3	< 2		0.2	0.01					< 5E-02			
N4												
N5												
N6												
Max	11		3E-01		3						32	
Min	6E-05		1E-03		3						32	
GM	1E-03		3E-02		4E-02				0.3		32	

4.2.4 Activity concentration in water, fodder and minerals

The diet of the cows varied greatly between the farms. When intensive farming practices are applied, the cows eat silage, hay, minerals or mineral fodder, protein fodder or supplement, cereal, rape etc. The silage is generally made of grass produced at the farm, but not always. At farms F1–F4 where intensive farming is applied the cows fed on a variety of foods and supplements (Table 9). It is necessary to consider the food with additives and water when assessing the intake of natural radionuclides by the cows. The farms N1–N3 represent extensive farming where the cattle are grazing large pasture areas and drink from natural water ponds and streams. At farm D1, the drinking water contained extremely low amount of uranium.

Table 9. The activity concentrations in cattle's diet. The unit in water is Bqkg⁻¹ (fw), while in food the unit is Bqkg⁻¹ (dw). Data for farms N1, N2 and N3 is taken from Hosseini et al (2010)

	Type	U-238	Sd	Ra-226	Sd	Pb-210	Sd	Th-232	Sd	Th-228	Sd	Ra-228	Sd	K-40	Sd	U-235	Sd
S1	water	< 5		9E-04		3						0.3		0.4			
S1	fodder			136		89						12					
F1	mineral	4	1			2	0.3			1	0.1	1	0.1			0.2	0.03
F1	silage	< 14		2	0.1					< 2		2	1			< 1	
F1	rape fodder	2	0.4	2	1					< 1		1	0.4			< 0.2	
F1	full fodder			1	0.3					1	0.1	1	0.1			0.1	0.02
F1	water	1E-02		5E-03				4E-07		< 2E-03		< 4E-03					
F2	mineral	5	2	0.2						1	0.2	1	0.2			0.1	
F2	fodder	< 39		1	0.2					< 0.4		3	1			0.2	0.1
F2	silage	< 16		1	0.3					< 0.2		< 1				< 2	
F2	amino			1	0.1					9						0.1	0.04
F2	cereal	3	1	1	0.1					1		1	0.3			< 1	
F2	water	8E-02	0.02	0.02				5E-07		< 6E-03		< 8E-03				5E-03	
F3	silage	< 10								< 2		< 3				< 1	
F3	full fodder	< 3								< 0.5		< 1				< 0.1	
F3	protein fodder	< 5		4	0.2					< 2		< 1				< 0.2	
F3	water	1E-03		7E-03				7E-07		< 5E-03		< 7E-03				< 0.1	
F4	silage	< 10		0.2						< 2		6				< 1	
F4	hay	< 9		1	1					< 2		3	1			< 0.4	
F4	full fodder	< 1		4	1					< 1		3	1			< 0.1	
F4	barley	< 5		2	0.2					< 1		< 1				< 0.3	
F4	water	8E-04		0.02		< 5		8E-07		< 0.01		0.03				< 0.1	
D1	water	6E-05															

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N 1	water	0.02	0.02	0.01	0.01	2E-03	1E-03			1E-04	3E-04	5E-03	2E-03			2E-04	7E-05
N 2	water	0.02	0.02	0.01	0.01	2E-03	1E-03			1E-04	3E-04	5E-03	2E-03			2E-04	7E-05
N 3	water	0.02	0.02	0.01	0.01	2E-03	1E-03			1E-04	3E-04	5E-03	2E-03			2E-04	7E-05
N 6	water							0.2	2E-04								

4.3 Activity concentration in soil, grass, milk, water, fodder and food supplements (^{137}Cs)

In Table 10 the activity concentrations of ^{137}Cs in the samples from the studied farms are shown.

Table 10. Results of activity concentrations for ^{137}Cs in soil, grass, milk, water and fodder samples in the studied farms (Bq kg⁻¹ dry weight, milk and water Bq l⁻¹)

	Soil		Grass		Milk		Water		Fodder		
	Act	Sd	Act	Sd	Act	Sd	Act	Sd	Type	Act	Sd
S1	5	1	< 3		3E-02		< 0.03				
S2	75	52	9								
S3	1086	203	168								
S4	73	8	5	0.4							
F1	22	2	0.23	0.06	0.06	8E-03	< 9E-04		mineral	0.3	0.04
F1									rape fodder	0.5	0.1
F1									full fodder	0.8	0.07
F2	236	28	7	6	0.3	0.02			fodder	< 0.3	
F2									silage	0.3	0.07
F2									cereal	1	0.1
F3	4	1			0.09	9E-03	< 2E-03		silage	2	0.3
F3									full fodder	0.3	0.07
F3									protein fodder	0.9	0.2
F4	25	1	2	0.7	0.3	0.02			silage	2	0.4
F4									hay	14	1
F4									full fodder	0.5	0.09
F4									barley	< 0.3	
D1	3	0.2	0.2	0.04	0.01						
D2	6	0.7	< 0.4		0.05						
I1	12	3	14.0	16	0.8	0.1					
I2	6	4	8	7							
N1	202	42	1081	297	207	8	0.3	0.6			
N2	412	269	910	595	173	7	0.3	0.6			
N3	244	106	124	168	12	0.5	0.3	0.6			
N4											
N5											
N6											
Max	1086		1081		207						
Min	3		0.2		0.01						
GM	48		22		1						

The ^{137}Cs concentrations in soil show higher variances between the farms than natural radioactivity levels due to the heterogeneous fall-out from the Chernobyl accident. The geometric mean activity concentration of ^{137}Cs in soils of the farms was $43 \text{ Bq} \cdot \text{kg}^{-1}$. At five farms (F2, N1, N2, N3 and S3) the concentration exceeded $200 \text{ Bq} \cdot \text{kg}^{-1}$. The maximum value, $1086 \text{ Bq} \cdot \text{kg}^{-1}$, was found at farm S3. At farms in which concentrations of ^{137}Cs in soil showed high variance (I2, N2, N3 and S2), even greater variance was detected in grass. Concentrations of ^{137}Cs could be determined from ten milk samples only. For example, at farm S3, where ^{137}Cs concentration in soil was the highest, about $1000 \text{ Bq} \cdot \text{kg}^{-1}$, the concentration in milk was below MDA which may indicate very small transfer coefficient.

4.4 Radionuclide transfer

The values of the transfer factors, transfer coefficients or concentration ratios may range over several orders of magnitude depending on e.g. soil characteristics. Yet another problem as seen in the data presented relates to low values, below detection limits. Therefore, transfer cannot be quantified in many cases. In these cases one option is to use a value of $0.5 \cdot \text{MDA} \pm 0.5 \cdot \text{MDA}$ in the calculations. This, however, will result in distribution functions which may be misleading for our data set.

Table 11. The number of transfer factors determined from the data available

Radionuclide	Number of TF's available	Number of activity concentrations data in grass available	Number of activity concentrations data in soil available
U-238	7	8	12
Ra-226	11	11	17
Pb-210	3	5	3
Po-210	0	0	0
Th-232	3	3	3
Th-228	5	5	7
Ra-228	12	12	17
K-40	13	13	16
U-235	2	3	6
Cs-137	11	12	13

The MDA of gamma spectrometry depends on the measuring time, detector type, background and geometry. Therefore, each measurement has its own unique MDA and those values may vary substantially. If the procedure in one laboratory results in MDA of, let's say, $15 \text{ Bq} \cdot \text{kg}^{-1}$ and the other laboratory is able to report values exceeding $1 \text{ Bq} \cdot \text{kg}^{-1}$, it is not justified to use values of $7.5 \pm 7.5 \text{ Bq} \cdot \text{kg}^{-1}$ for assessment of average values and describing distribution functions. This issue must be underlined when the final analysis of the results is performed. Nevertheless, a preliminary analysis of available data is given in the following section but should be used with caution. The limitations of our data set and the values quantifying radionuclide transfer should be

understood as supplementary to other previously published data if that exists.

4.4.1 Soil-to-plant transfer

The difficulty to determine some radionuclides by gamma spectrometry is demonstrated in Table 11. This table shows the data available for the transfer factor from soil to plant. The MDA values are different from soil to grass in gamma spectrometry and therefore it is not always possible to determine the value of the transfer factor.

4.4.2 Transfer to milk

Two quantities for the transfer of radionuclides to milk are generally used. The more commonly used quantity is the transfer coefficient (F_m) which relates to the equilibrium ratio of a radionuclide concentration in milk to the daily dietary intake of a radionuclide, with the unit $d \cdot L^{-1}$ or $d \cdot kg^{-1}$. The other quantity is the concentration ratio (CR) which is the ratio of a radionuclide concentration in the food product (fresh weight) to a radionuclide concentration in the feed (dry matter). All radionuclide concentrations in milk are hence expressed in activity per mass or volume fresh weight (IAEA, 2010).

Duration of radionuclide intake affects the radionuclide concentrations in milk. In fact, the transfer coefficient represents the equilibrium ratio of radionuclide concentration in milk to that in diet by definition. The diet of cows, however, may vary considerably at different seasons as is the case in Nordic countries. In winter, the cows may eat commercial fodder and supplements. In summer months, the cows are out in the fields grazing on pastures. At farms applying extensive farming practises natural water and pasture grass may constitute the whole diet during summer months. Not all the organisations have reported the daily dietary intake of the cows. Therefore only CRs can be assessed (Table 12).

Table 12. The estimated grass-to-milk concentration factors

Radionuclide	N	Arithmetic mean	Geometric mean	Min	Max
uranium	1	0.0006	0.0006	0.0006	0.0006
radium	7	0.036	0.023	0.005	0.10
caesium	10	0.14	0.12	0.045	0.28

CR for uranium could only be calculated based on the results from farm D1. The concentration ratio, $0.0006 \pm 0.0002 \text{ kg}_{\text{dw}} \cdot \text{kg}_{\text{fw}}^{-1}$ is clearly smaller than that estimated by IAEA, $0.005 \text{ kg}_{\text{dw}} \cdot \text{L}^{-1}$. The reference food-to-milk CR reported by IAEA for caesium is $0.11 \text{ kg}_{\text{dw}} \cdot \text{L}^{-1}$. The value is based on 119 data points the range of which is $0.0036\text{--}0.69 \text{ kg}_{\text{dw}} \cdot \text{L}^{-1}$. Our geometric mean is close to the value. It must, however, be noted that four of our data points are from farms with intensive farming practises where supplementary food is given to the cows (Table 9). The effect of the food has not been included in the assessments at this point.

No values of food-to-milk CR were reported for radium among reference values by IAEA (2010). The reference value (cow's milk) for radium F_m , $3.8 \cdot 10^{-4} \text{ d} \cdot \text{L}^{-1}$, is based on 11 data points only.

4.5 Results of radon in outdoor air

The results from the measurements of the physical parameters of the soil samples are presented in Table 13. The total porosity of the samples was high, 0.5–0.7. It must be noted that total porosity (including micro and macro pores) in soils does not remain constant, particularly in clayey soil, and these results represent the situation at the time of sampling.

Soils at sites F1–F3 were very moist as 70–80% of the total pore volume was water saturated and can be assumed to be close to their field capacities. The particle density at sites F2, I1 and I2 were the lowest indicating greater proportion of organic matter in the soil. Soils at sites F1, F3 and F4 represented fine sand soil or fine sand moraine and their particle densities were close to that of stone (2.65 kg/l).

Table 13. Soil physical data used for radon in outdoor air determinations

Sampling site	Bulk density [kg/L]	Particle density [kg/L]	Total porosity	Volumetric water content [m ³ /m ³]	H ₂ O-filled fraction of pores	Ra-226 [Bq/kg]
F1	1.1 ±0.3	2.5 ±0.1	0.53 ±0.12	0.34 ±0.10	0.73 ±0.39	57 ±6
F2	0.9 ±0.2	2.0 ±0.1	0.58 ±0.09	0.47 ±0.12	0.80 ±0.11	80 ±19
F3	1.1 ±0.2	2.5 ±0.1	0.57 ±0.10	0.37 ±0.25	0.67 ±0.19	28 ±3
F4	0.8 ±0.1	2.5 ±0.1	0.68 ±0.05	0.25 ±0.07	0.36 ±0.09	17 ±2
I1	n/a	1.8 ±0.1	n/a	n/a	n/a	7.5 ±0.5
I2	n/a	2.3 ±0.1	n/a	n/a	n/a	14 ±1

The results of the radon emanation rate measurements are presented on Table 14. As can be seen, increasing moisture content results in higher emanation rates most notably in soils where particle densities were the lowest (and thus the organic content was the highest). Radon emanation rates increased with increasing radium concentration in the soil, which was expected. Soils containing more organic component (as opposed to mineral component) have generally higher emanation coefficients which was also evident in this study (Figure 6).

When assessing densities of radon flux, climate dependent parameters must also be considered. Radon emanation coefficient is affected by both soil moisture content and temperature, and Ostwald coefficient (K_t) by temperature. The effective radon diffusivity depends on total porosity (which is not a constant quantity) as well as on moisture which most notably on top-soil depends on precipitation and ground water diffusion.

Table 14. The radon emanation rates and emanation coefficients in 22 °C at two different moisture contents

Sampling site	Moisture [kg/kg ⁻¹ d.w.]	$G_m \times 10^{-6}$ [Bq/(kg s)]	f
F1	0.05	52 ± 3	0.44 ± 0.05
	0.25	55 ± 3	0.46 ± 0.05
F2	0.05	35 ± 2	0.21 ± 0.05
	0.25	140 ± 7	0.82 ± 0.20
F3	0.05	16 ± 1	0.27 ± 0.03
	0.25	26 ± 1	0.44 ± 0.05
F4	0.05	8.1 ± 0.6	0.22 ± 0.03
	0.25	14 ± 1	0.38 ± 0.06
I1	0.05	7.2 ± 0.4	0.46 ± 0.05
	0.25	9.3 ± 0.5	0.59 ± 0.06
I2	0.05	9.5 ± 0.4	0.31 ± 0.03
	0.25	14 ± 6	0.45 ± 0.04

We have made two radon flux density assessments (Table 15), one for spring/autumn months (temp=6 °C) and one for summer (temp=13 °C). Temperature data was retrieved from Yli-Halla and Mokma (1998) and volumetric soil moisture content (m³/m³) of 0.3 was used for assessing the fraction of the porosity that is water filled (m) during both seasons (to 1-meter depth) as assessed by Hakojärvi et al. (2010).

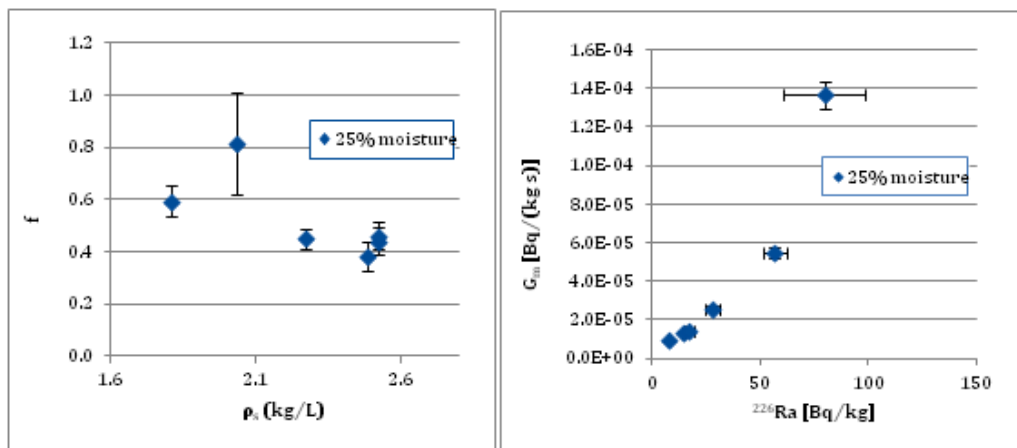


Figure 6: The emanation coefficient as a function of particle density (left graph) shows that soils containing more organic component lead to higher emanation coefficient. Radon emanation rate depends on the ²²⁶Ra concentration of the soil (right graph).

Table 15. The fraction of the porosity that is water filled (m), effective radon diffusivity (D_e), temperature adjusted radon emanation rate (G_m) and density of radon flux (J_D) during spring and autumn seasons with soil temperature of 6 °C ($K_t=0.42$) and 13 °C ($K_t=0.3$)

Sampling site	m	$D_e \times 10^{-6} \text{ m}^2/\text{s}$	$G_{m,6^\circ\text{C}} \times 10^{-6} \text{ Bq}/(\text{kg s})$	$G_{m,13^\circ\text{C}} \times 10^{-6} \text{ Bq}/(\text{kg s})$	$c_{\infty, 6^\circ\text{C}} \text{ kBq}/\text{m}^3$	$c_{\infty, 13^\circ\text{C}} \text{ kBq}/\text{m}^3$	$J_{D,6^\circ\text{C}} \text{ Bq}/(\text{m}^2 \text{ s})$	$J_{D,13^\circ\text{C}} \text{ Bq}/(\text{m}^2 \text{ s})$
F1	0.57	0.87	57	57	93	100	0.060	0.063
F2	0.52	1.0	140	140	140	150	0.12	0.13
F3	0.53	1.0	27	26	35	37	0.028	0.029
F4	0.44	1.2	15	15	11	11	0.014	0.014

The assessed effective radon diffusion coefficient (D_e) varied 0.9×10^{-6} – 1.2×10^{-6} , which is about half of the generic value suggested for soils, 2.0×10^{-6} (Yu *et al.*, 1993). As the effective radon diffusion coefficient decreases with increasing moisture content, it is natural that agricultural soils exhibiting good field capacities have lower than average D_e values. In practise, our values translate to effective diffusion lengths of 0.6–0.8 metres which indicates that >70% of the radon released from the soils is attributable to the first metre. Radon concentration in air in soil pores was 11–150 kBq/m³. If we had used the moisture contents measured at the sampling date, the range would have been 10–190 kBq/m³. These values compare favourably to the average radon concentration in air of soil pores, 50 kBq/m³, assessed by Weltner *et al.*, (2003). In this assessment, the small difference between radon flux densities (J_D) in spring/autumn and summer is explained by the temperature dependent Ostwald coefficient since the moisture content in the assessment had been assigned a constant value of 0.3 m³/m³. If we had used the moisture contents measured at the date of sampling ($t=13$ °C), the radon flux densities would have been 0.040, 0.053, 0.022 and 0.015 Bq/(m² s) at sampling sites F1, F2, F3 and F4, respectively. In the model used in this study, radon flux density is clearly more sensitive to soil moisture than to temperature and had the largest effect on soil at F2. In any case, all assessed values are similar to the global average value of 0.016 Bq/(m² s) cited by UNSCEAR, (2000).

Radon flux density values assessed in “European ²²²Rn flux map for atmospheric tracer applications” –project form another source for reviewing the assessed values (Szegvary, 2009). The mean radon flux in Finland (with spatial standard deviation) was assessed as 0.30 (0.25) atom cm⁻² s⁻¹. This translates to 0.060 ± 0.052 Bq/(m² s). Radon flux density at site F2 was assessed higher than the generic value for Finland. However, if we had used the moisture content determined on the sampling date, this value would also have been within the range. In Finland, annual radon concentration in outdoor air has only been investigated at 33 sampling sites, in six municipalities. In these municipalities, higher than average radon concentration in indoor air had been found (Turtiainen, 2000). The concentration was recorded with integrating radon dose meters which were hung at 0.6–1.0 meter height from the ground and protected against rain and wind by a plastic capsule. One of the municipalities was that of site F2 where the highest radon flux density was also measured in this study. The results of these two measurement points are presented in Figure 7.

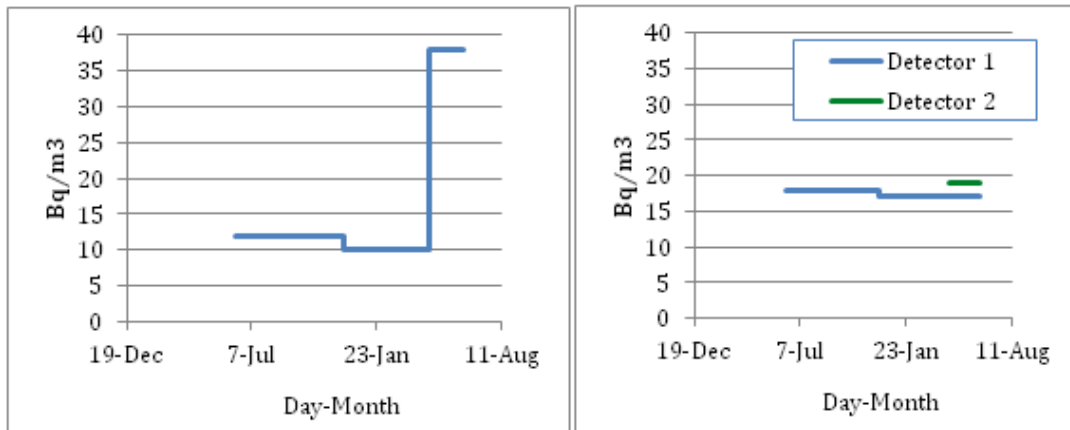


Figure 7: Radon measurements at two sites in the municipality of sampling site F2. The annual mean radon concentration in outdoor air was 15 and 17 Bq/m³ at site #1 (left) and at site #2 (right). A short measurement period (56 d) was performed 16.4–11.6. at site.

Radon concentrations in outdoor air depend greatly on climatic conditions such as wind speed and direction, closeness of open sea, precipitation, duration of snow cover etc. The average life time of a radon atom is 5.5 days during which time it can move several hundreds of kilometres before decaying into radon progeny. Vegetation can actively transport radon into outdoor air (Jayaratne et al., 2011). Therefore, radon flux densities cannot be converted to outdoor air radon concentrations without complex atmospheric modelling. In any case, we have shown that even with the highest assessed radon flux densities 0.13 Bq/(m² s), radon concentration in outdoor air remain <40 Bq/m³ but more typically <20 Bq/m³. Turtiainen (2000) suggested a generic annual mean value of 15 Bq/m³ for radon concentration in outdoor air in Finland which is the same as that used by US-EPA. For our dose assessment, this value will be used for assessments at sites F1–F4.

4.6 External exposure

4.6.1 The external dose of ¹³⁷Cs

Two of the calculated photon mass-attenuation and energy-absorption curves are presented in Figure 8. Lower values were obtained for F2, in which water and organic component formed a substantive proportion of the total mass. The terrestrial components of absorbed dose rates varied 46–97 nGy/h being highest at site F2.

All sites F1–F4 had undergone ploughing several times to 20–25 cm depth after the 1986 fall-out. Therefore, ¹³⁷Cs was thought to be evenly distributed in this ploughing layer. In addition, ¹³⁷Cs was restricted to the cultivated layer, since diffusion by water or soil fauna is limited (VandenBygaart *et al.*, 1999). The contribution of ¹³⁷Cs to the terrestrial component was highest at site F2, where its proportion was 35%. The proportions of ¹³⁷Cs component at sites F1, F3 and F4 were 5.1, 1.3 and 9.5% respectively. The effective external dose rates including the cosmic ray doses varied 0.07–0.11 μSv/h (Table 16).

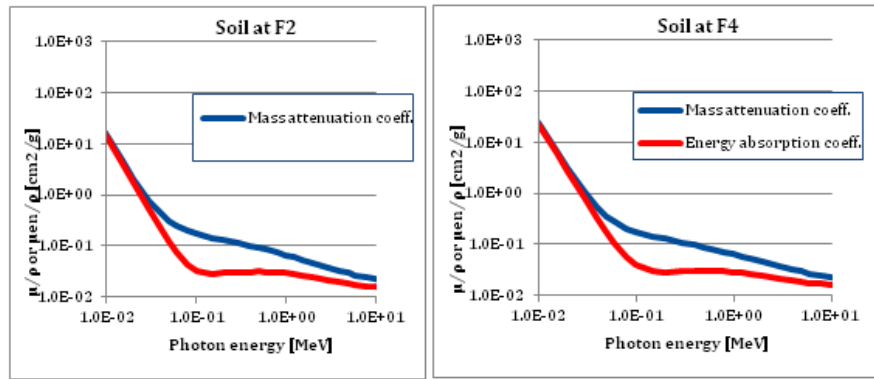


Figure 8: Photon mass-attenuation and energy-absorption curves calculated for sites F2 and F4.

Table 16. The external dose rate assessment at sites F1–F4

Sampling site	Terrestrial component [nGy/h]	Terrestrial component [μ Sv/h]	Cosmic component [μ Sv/h]	Total dose rate [μ Sv/h]
F1	65	0.046	0.043	0.089
F2	97	0.068	0.043	0.11
F3	46	0.032	0.043	0.075
F4	38	0.026	0.043	0.069

4.6.2 The absorbed dose rate from natural radionuclides (external)

The value of the absorbed dose rate was calculated using equation 6. A representation of the data from the participants can be seen in Figure 9.

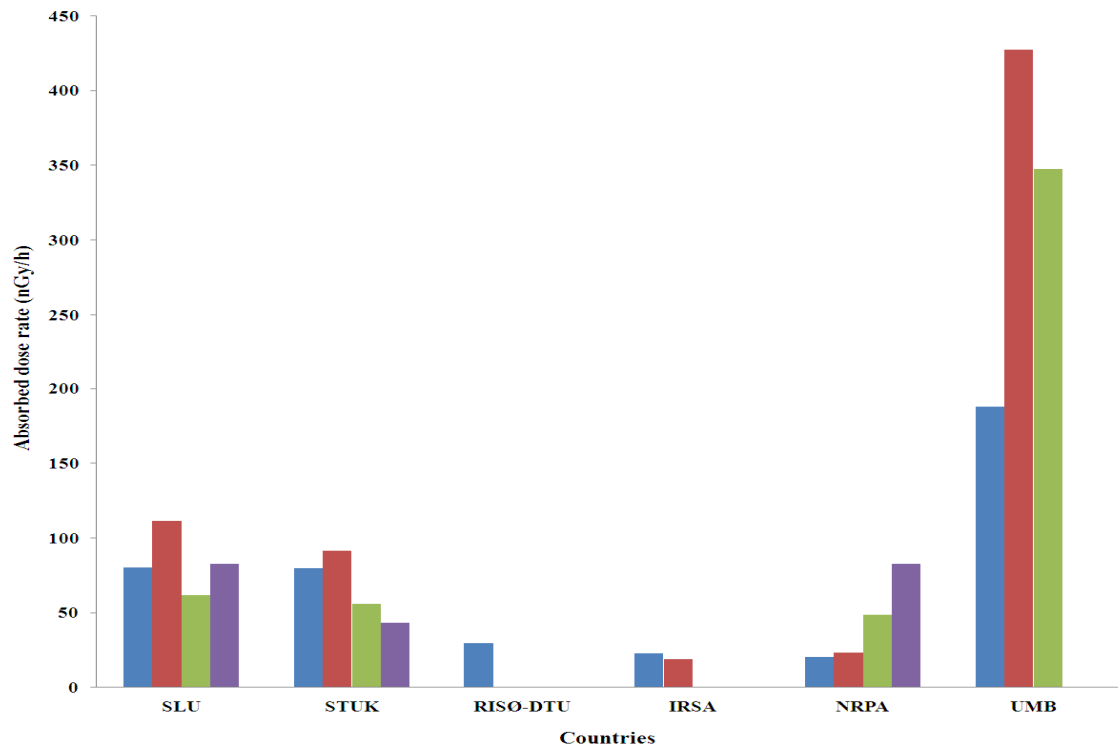


Figure 9: Absorbed dose rate in the different points analysed by the participant countries.

5. Discussion

5.1 The activity concentrations in soil samples

The variation in the concentration of naturally occurring radionuclides is less compared to ^{137}Cs at the analysed farms. The variation at the farms I2, N2, N3 and S2 is bigger than at other places. This can be explained by uneven fall-out, distance between sampling circles and different caesium retention characteristics of the soil in the sampling circles. At farm S3, however, the two ^{226}Ra concentrations measured were 82.2 ± 13.2 and $41.5 \pm 6.7 \text{ Bq} \cdot \text{kg}^{-1}$. At farms I1 and I2 one of the three soil samples contained significantly more ^{226}Ra than the other samples. Table 18 shows the activity ratios between $^{226}\text{Ra}/^{238}\text{U}$ and $^{228}\text{Ra}/^{238}\text{U}$.

We can observe that there is no equilibrium between ^{226}Ra and ^{238}U at most of the farms except in the case of F3 and N4. The rest of the farms present much more content of ^{238}U . In some cases it was not possible to determine the ratios because one of the two elements (^{238}U) was not detected, indicating the difficulty of measuring ^{238}U by means of gamma spectrometry. Interestingly the behaviour of farms N4, N5 and N6 is clearly different from the others in terms of the ratio $^{228}\text{Ra}/^{238}\text{U}$. These farms have a high content due to the presence of ^{232}Th , which also relate to the farms F3, F4 and D1, although less evident. Some studies have showed that the breaking of the radioactive equilibrium between members of the same radioactive decay chain gives an indication of the increased mobility of the single radioisotope (P. Blanco Rodríguez et al., 2008). Higher ^{226}Ra activities at some farms could be due to high clay content and therefore enhanced interaction between the nuclide and the soil particles due to a weak nutrient content (the lowest registered in both provinces). According to previous studies (El-Arabi et al., 2006) the reason for high radionuclide activity in clays is due to the fact that clay minerals are mainly composed by aluminum silicates and they are characterized by small sized grain and negatively charged surface. In this way clay particles easily absorb cations on their surface. The highest ^{226}Ra activity concentration in the soil was found at the farm F2 which also has one of the highest content on Ca (492 mg/100 g). Farm S3 with a Ca content of 959.6 mg/100 g has a high ^{226}Ra activity concentration too.

Table 17. The activity ratios of the two natural decay chains obtained at the studied sites

	Ra-226/U-238	Ra-228/U-238
S1	0.21	0.09
S2		
S3	0.09	0.07
S4	0.15	0.20
F1	0.66	0.51
F2	0.56	0.42
F3	0.98	1.12
F4	0.82	1.08
D1	1.33	1.61
D2	1.0	1.33
I1		
I2		
N1		
N2		
N3	0.73	0.72
N4	0.97	8.52
N5	0.44	5.35
N6	0.59	7.55

Finally the activity concentrations from natural and artificial sources in terms of ^{226}Ra and ^{137}Cs respectively are compared. Figure 10 shows the correlation between these parameters and we can observe that there is no significant correlation. This is the expected behaviour since the presence of the two radionuclides in the soil is due to different origins: ^{137}Cs is mainly due to the Chernobyl fallout and ^{226}Ra is a natural radionuclide which is presented in the soil since its origin. The correlation between ^{226}Ra and ^{137}Cs as seen in Figure 10 is poor which shows that ^{137}Cs is not suitable as nuclide vector for ^{226}Ra when performing risk assessments.

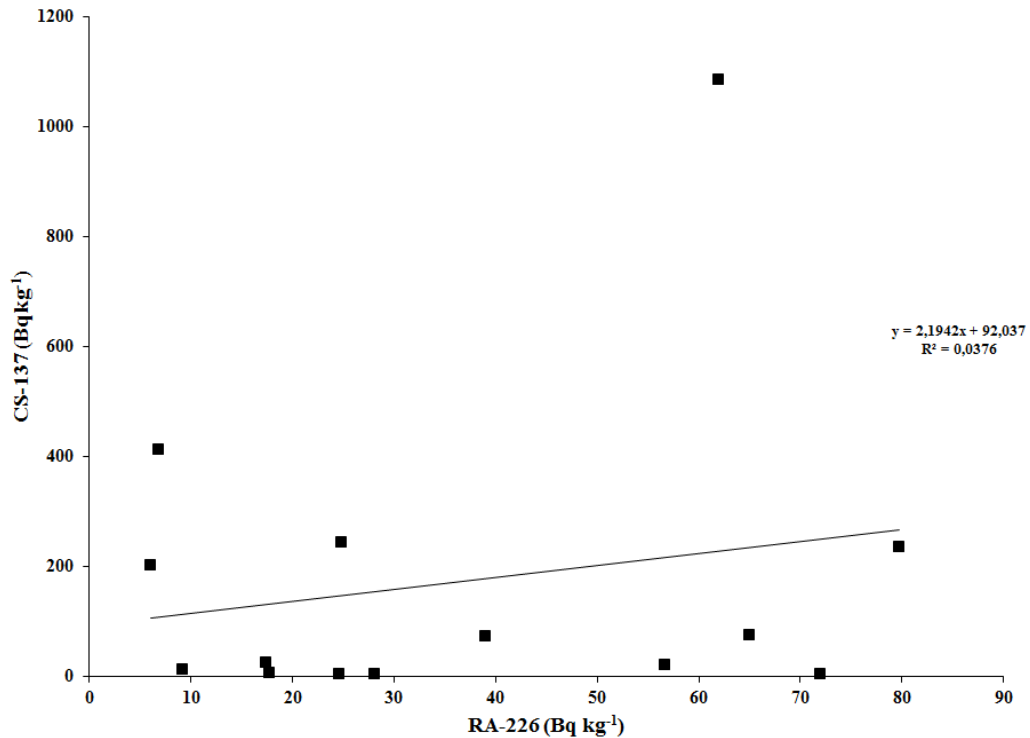


Figure 10: Comparison between ^{226}Ra and ^{137}Cs activity concentrations in soils.

5.2 The activity concentration in grass samples

Table 6 also shows the difficulty in determining low activity levels of radium isotopes by gamma spectrometry (GS). While the laboratories were able to determine ^{226}Ra and ^{228}Ra concentration in most of the soil samples, the concentrations were determined in a significant less number for the grass samples. It is worth mentioning that grass and milk sampling was carried out twice at farms F3 and F4 because during the first sampling the cows were not grazing because of the cold weather. ^{228}Th turned out to be an even more demanding radionuclide to measure. When subtracting the activity of ^{228}Th produced by ^{228}Ra during the period from sampling to measurement, the uncertainties increase and the results are generally below detection limits. Uranium (either ^{238}U or ^{235}U) is also more demanding to determine by gamma spectrometry. Only seven grass samples could be measured by gamma spectrometry whereas mass spectrometry performed on the three samples from farm D1 was able to determine uranium from each sample.

Difficulties were also encountered with the measurements of ^{137}Cs . At three farms, the activity concentrations in grass were below detection limits. At farm F3, the ^{137}Cs concentration in soil was about $4 \text{ Bq} \cdot \text{kg}^{-1}$ and all four grass samples contained less than $0.47\text{--}0.56 \text{ Bq} \cdot \text{kg}^{-1}$ of ^{137}Cs . At farms S1 and S4, the ^{137}Cs concentrations in soils were 5 and $73 \text{ Bq} \cdot \text{kg}^{-1}$, respectively, and the corresponding grass samples had less ^{137}Cs than the MDA.

5.3 Transfer factors

Radium isotopes (^{226}Ra and ^{228}Ra) are relatively easy to measure by gamma spectrometry in (constant or monitored) low radon background facility. Therefore, most partners were able to report radium in both soil and plant from most of the sampling points. Soil characteristics affect greatly

the soil-to-plant TFs, which may vary even in the range of several orders of magnitude. Chemical analogue pairs such as Cs-K and Sr-Ca are most notable but also cation exchange capacity, pH value, sulphate anions etc. have been reported to affect the transfer (IAEA, 2010). Standard soil analyses carried out by agricultural laboratories usually apply leaching of available nutrients by ammonium acetate solution. In this way, exchangeable nutrients available for root uptake are measured. For example, potassium residing in the mineral lattice of the soil would have little effect on the uptake of caesium and hence its concentration offers little explanatory value. Of course, the same applies to naturally occurring radionuclides: uranium or thorium in monazite or xenotime crystal lattice cannot transfer to plants whereas leached secondary species found in soil exchange sites can. It has however become a standard to analyse total radionuclide content of the soil rather than the exchangeable species although radionuclide solid/liquid partition coefficient has been used in some studies (Vandenhove and Van Hees, 2007).

Fall-out radionuclides such as ^{90}Sr and ^{137}Cs have been deposited on soils and hence there is no controversy in measuring their total amount in soils when assessing transfer factors. The thorium isotope ^{228}Th , however, is relatively easy to determine by gamma spectroscopy by utilizing the gamma peaks of ^{212}Pb and ^{208}Tl , which are assumed to be in secular equilibrium in the samples. The problem with ^{228}Th is that it is constantly produced by decay of ^{228}Ra which is also transferred into vegetation. The ^{228}Th concentrations therefore increase with time and hence this isotope suits poorly to assessing soil-to-plant transfer factors (but is naturally considered in intake assessments).

Unfortunately, the same technique for soil analysis was not used for all the farms and therefore, the concentrations are not comparable. Electrical conductance, cation exchange capacity (CEC), pH and sulphur were reported only at farms F1–F4. Soil organic content data was available from farms F1–F4 and S1–S4. Our data will thus be far too fragmental to clearly demonstrate any correlations between TFs and soil characteristics.

5.3.1 Soil-to-plant TF of uranium

Soil-to-plant transfer factors for uranium could be determined from samples representing ten farms. Uranium concentrations in samples from farm D1 were determined by MS and hence constitute the most reliable source of information for the study. There, the range of the TF values is close to IAEA reference values⁴ of 0.017 and 0.046 $\text{kg} \cdot \text{kg}^{-1}$ for temperate environment grasses and natural pasture, respectively. The TF from farm F1 ($0.045 \pm 0.016 \text{ kg} \cdot \text{kg}^{-1}$) is also close to the reference value suggested by IAEA. The highest value found corresponds to farm S4 (4.05) which is out of the range. This TF corresponds with a grass sample with a very high ^{238}U content which is unusual and shows a possible soil contamination of the grass sample.

5.3.2 Soil-to-plant TF of radium

Soil-to-grass transfer factors calculated from the two isotopes of radium (^{226}Ra and ^{228}Ra) were first pooled assuming similar transfer characteristics for the both. The TF was determined from five (^{226}Ra) and six (^{228}Ra) farms employing intensive farming practices and from six farms applying extensive farming practices. In the sampling circles representing extensive practices, the arithmetic mean and geometric mean values of TF (0.95 and $0.29 \text{ kg} \cdot \text{kg}^{-1}$, respectively) were significantly

The IAEA reference values are geometric mean values from all available data.

higher than those representing intensive farming (0.86 and $0.18 \text{ kg} \cdot \text{kg}^{-1}$, respectively). The TF values form a nearly lognormal distribution (Figure 11).

Farming type was unfortunately not reported at one farm. The arithmetic and geometric mean values of TFs (\pm SD) at this farm were very small, only 0.031 and $0.030 (\pm 0.010) \text{ kg} \cdot \text{kg}^{-1}$, respectively.

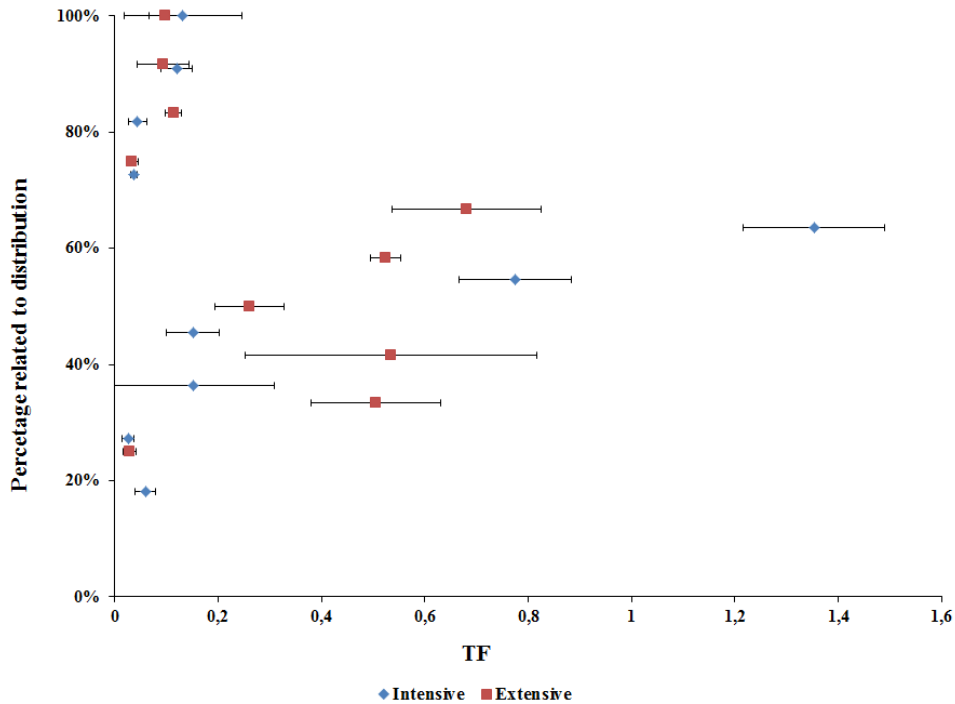


Figure 11: Soil-to grass transfer factor for radium at farms applying intensive and extensive farming practises with percentages relating to distribution. TF clearly out of range are not represented.

The IAEA reference values of soil-to-plant TF of radium are 0.13 and $0.071 \text{ kg} \cdot \text{kg}^{-1}$ for temperate environment grasses and natural pasture, respectively. The geometric mean of TF obtained from samples representing intensive farming practices ($0.18 \text{ kg} \cdot \text{kg}^{-1}$) is close to the reference value suggested by IAEA for grasses. Our data set, however, suggests a higher TF value for natural pasture ($0.26 \text{ kg} \cdot \text{kg}^{-1}$) than the IAEA value.

It has been suggested that increased exchangeable calcium, increasing pH and high sulphate concentration decrease radium transfer to plants (IAEA, 2010). It has, however, also been debated whether divalent calcium can compete with divalent radium during the ion exchange related to root uptake due to significantly different ion radii (Vandenhove and Van Hees, 2007). In addition, Mg also forms a divalent cation and should therefore be investigated in more detail.

The exchangeable Ca and Mg data were obtained from four farms (F1–F4). At each farm, the soil parameters were determined from one pooled sample representing three sampling circles. Therefore, the radium concentrations in soil and grass were also calculated for pooled samples and the TF calculated accordingly. At farm D1, total Ca and Mg data was available from all three sampling circles. It is clear that our data has little statistical power. The ostensible correlations between TF

and CEC as well as TF and exchangeable Ca, however, follow the trends reported earlier (IAEA 2010).

Naturally occurring radionuclides originate from the earth's lithosphere. During aeons of geological processing these radionuclides have leached from the rock minerals and entered the organic portion of the soil. Organic content of agricultural soils, however, varies from organically poor sandy soils to mostly organic peat soils. Significantly different transfer factors should hence be expected even though the activity concentration and available nutrient levels in soil would be the same.

5.3.3 Soil-to-plant TF of caesium

The soil-to-plant TF of caesium was determined from 3 farms employing intensive farming practices and from seven farms applying extensive farming practices. In the sampling circles representing extensive practices, the arithmetic mean and geometric mean values of TF (1.5 and 0.7, respectively) were significantly higher than those representing intensive farming (0.053 and 0.050, respectively). Again, the TF values form a nearly lognormal distribution which is presented in Figure 12. Farming type was not reported at one farm. When combining all data (N=31), the arithmetic mean and geometric mean values of TF are 0.99 and 0.29 $\text{kg} \cdot \text{kg}^{-1}$, respectively.

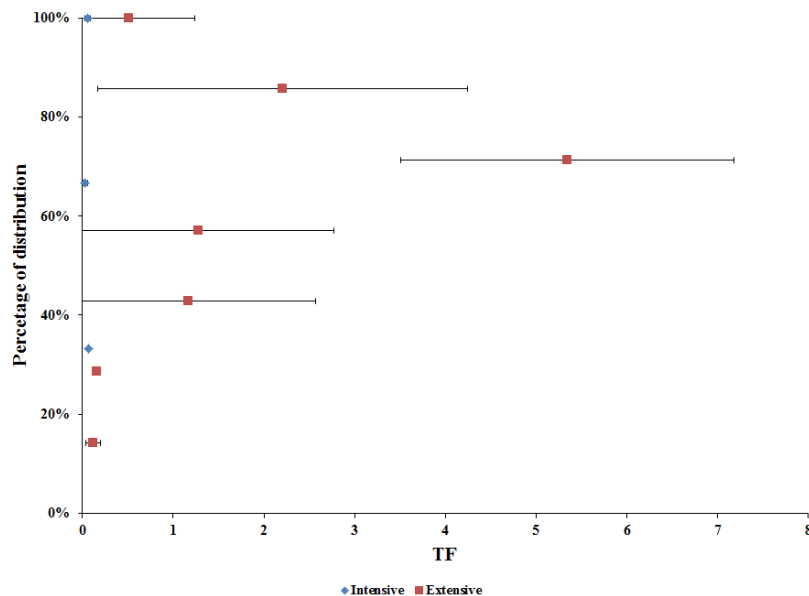


Figure 12: Soil-to grass transfer factor of caesium at farms applying intensive and extensive farming practises. Percentages are related to distribution.

The IAEA reference values for soil-to-plant TF of caesium are 0.063 and 0.25 $\text{kg} \cdot \text{kg}^{-1}$ for temperate environment grasses and natural pasture, respectively. The geometric mean of TF obtained from samples representing intensive farming practices (0.05 $\text{kg} \cdot \text{kg}^{-1}$) is somewhat lower than the reference value for grasses suggested by IAEA (2010). Our values, however, fit amply within the reported range which is 0.0002–5.5 $\text{kg} \cdot \text{kg}^{-1}$. In case of natural pasture, our geometric mean 0.77 $\text{kg} \cdot \text{kg}^{-1}$ is higher than that suggested by IAEA. The ranges of previously reported TFs in natural pastures are 0.01–5.0 $\text{kg} \cdot \text{kg}^{-1}$ and in Chernobyl affected meadow grasses 0.5–33 $\text{kg} \cdot \text{kg}^{-1}$ (IAEA, 2010). Thus, all the detected TFs fall within the reported ranges.

Potassium and caesium are chemical analogues forming aqueous monovalent cation species in soil. Only weak aqueous complex with SO_4^{2-} , Cl^- and NO_3^- have been reported. Therefore, as exchangeable potassium in soil increases, the transfer factor of caesium decreases (IAEA, 2010). Data on exchangeable potassium was available from four farms (F1–F4) but the TF could only be determined at three farms due to negligible caesium fall-out at one farm (F3). At each farm, the soil parameters were determined from one pooled sample representing three sampling circles. Therefore, the caesium concentrations in soil and grass were also calculated for pooled samples and the TF calculated accordingly. At farm D1, total K data was available from all three sampling circles. The correlations are presented in Figure 12.

The potassium concentration in soil (exchangeable and total) and soil-to-plant TF of caesium presented in Figure 13 have little statistical power due to the extremely limited number of data points (N=3). The ostensible correlations between TF and exchangeable K, however, follow the trends reported elsewhere. The concentration of exchangeable potassium in fertilized soils (intensive farming practise) is generally higher than those where fertilizing is not allied. This offers a straightforward explanation for the difference of TF values between samples representing intensive and extensive farming practises (Figure 13).

5.4 Estimation of doses

The main contributors to the external radiation doses from natural sources are ^{238}U , ^{226}Ra and ^{232}Th or ^{228}Ra (^{232}Th was only considered in the case of the farms N4, N5 and N6). ^{210}Po is also another important contributor to the dose but primarily from intake. The estimation of the internal dose was not considered in this report partly because of the problem with the measurements of ^{210}Po by part of the organisations.

As the artificial source the ^{137}Cs deposition was considered. Only farms from Sweden, Finland and partly Norway reported values for ^{137}Cs . All the Norwegian farms which reported the ^{137}Cs deposition stated that ^{137}Cs is the main contributor to the annual effective dose at those sites. However, four of the farms show clearly that the natural component is of importance, most remarkable in the case of S1, located in the north of Sweden an area with very high levels of uranium (alum shale). At this farm almost 99 % of the annual effective dose is due to natural sources. The importance of natural radiation can also be seen when calculating the absorbed dose rate. Most of the farms show similar values as we can see in Figure 6. Nevertheless the farms N4, N5 and N6 have the highest values which are mainly due to the high content on ^{232}Th .

6. Conclusions

- Fourteen farms were selected as test sites for this study. Seven of the farms represented extensive farming practises, six intensive farming practises and one farm was not categorized.
- In total, 42 soil samples were collected at the farms. ^{226}Ra and ^{228}Ra could be determined by gamma spectrometry in 35 and 41 samples, respectively. Uranium could be determined in 15 soil samples only. ^{137}Cs could be determined in all samples. Analyses of ^{210}Pb are carried out by radiochemical methods and the results are not available at present.
- ^{226}Ra and ^{228}Ra were employed to represent uranium and thorium series, respectively. The levels for both isotopes varied from a few becquerels per kilogram to nearly one hundred. The test sites form thus a representative array of natural radioactivity levels in the different Nordic Countries.
- ^{137}Cs concentrations (from anthropogenic origin) in the soils at the selected farms range from a few becquerels per kilogram to over one thousand.
- Soil parameter analyses were not reported by all partners. Therefore, the main variable when investigating radionuclide transfer was confined to the type of farming practises (intensive/extensive).
- In total, 44 grass/pasture samples were collected at 14 farms. ^{226}Ra and ^{228}Ra could be determined by gamma spectrometry in 26 and 21 samples, respectively. Uranium could be determined in 7 grass samples only. ^{137}Cs could be determined in 30 samples. Analyses of ^{210}Pb are carried out by radiochemical methods and the results are not available at the time of writing.
- In total, 16 milk samples were collected at 14 farms. ^{226}Ra and ^{228}Ra could be determined by gamma spectrometry in 8 and 2 samples, respectively. Uranium could be determined in one milk sample only. ^{137}Cs could be determined in 12 samples. Analyses of ^{210}Pb are carried out by radiochemical methods and the results are not available at present.
- Radionuclide concentrations in the drinking water were reported at six farms. Sixteen samples of additional foods, supplements and minerals used commonly at farms applying intensive farming were measured and reported at four farms.
- Soil-to-plant transfer factors could be calculated for radium and caesium.
- Also, concentration ratio (milk/food) could be assessed for radium and caesium although some lack of data.
- Table 19 summarises our findings and provides a comparison to the reference values reported by IAEA. As can be seen, this project has been able to supplement available data, in some cases considerably.

Table 18. Summary of radionuclide transfer. TF from RadPast project and IAEA reference values are geometric mean values of the data

Source	transfer quantity	Unit	U	N	Ra	N	Cs	N
RadPast	soil-to-grass/pasture TF	$\text{kg} \cdot \text{kg}^{-1}$	0.010	4	0.11	47	0.27	31
IAEA	soil-to-grass TF	$\text{kg} \cdot \text{kg}^{-1}$	0.017	147	0.13	62	0.063	64
IAEA	soil-to-pasture TF	$\text{kg} \cdot \text{kg}^{-1}$	0.046	53	0.071	42	0.25	401
RadPast	feed-to-milk CR	$\text{kg}_{\text{dw}} \cdot \text{kg}_{\text{fw}}^{-1}$	0.0006	1	0.023	10	0.12	10
IAEA	feed-to-milk CR	$\text{kg}_{\text{dw}} \cdot \text{L}^{-1}$	0.005	n/a	n/a	n/a	0.11	119

- Transfer of ^{137}Cs deposition to milk was not quantified at this point.
- Assessment of exposure to radon in outdoor air was based on measuring radon emanation rates and soil properties which were used for modelling radon flux densities from the soil. The difficulty in interpreting the results is evident due to several climatic factors discussed in the previous chapter. In modelling, soil is also considered a homogenous layer exhibiting the same activity concentrations and soil properties throughout, which is naturally not the case in real situations. More accurate exposure assessment would have been obtained by giving personal radon dose meters to the farmers, monitoring radon concentrations inside the houses and by recording times spent indoors and outdoors. Nevertheless, our results suggest that no significant exposure to radon gas takes place at work on the fields. Farmer's work also includes a lot of work indoors, which was not investigated here and is most probably the most significant exposure route to the farmers.
- External gamma doses to the farmers at sites F1-F4 was also assessed from the measurements at lab: activity concentration and soil composition. The assessed doses were based on computing available data and were not validated by direct measurements. Nevertheless, the results suggest that no significant excessive external exposure takes place at work.
- Exposure to radioactive aerosols was not considered here. When ploughing dry soil, airborne soil dust is created and can cause doses via inhalation. Handling of hay stacks may also cause dust emissions which was not investigated.

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Appendix A: General description on methods to analyse natural radionuclides

There are several possible analytical methods available when determining the radioisotopes in the naturally occurring decay chains. These are mainly based on radioanalytical methods including alpha, beta and gamma spectrometric techniques. Due to the low specific activity of several of the decay chain isotopes and the recent development of mass spectrometric methods this technique has gained increasing importance for those radioisotopes. Apart from these mass spectrometric techniques there is also several atomic emissions (eg fluorometry for uranium) techniques of which some are more sensitive than radiometric techniques but which are not isotope selective. In recent years however improved atomic emission techniques (based on laser or ICP) have emerged which enable isotope selective determination and which in the future will play a more pronounced importance than they do today. In this review these recent techniques have been omitted due to their today limited use. This review is therefore mainly meant to give a summary of the radiometric and mass spectrometric techniques used for determination of the natural decay chain radioisotopes. Furthermore, the review will only cover those radioisotopes which commonly are dealt with in the decay chains and which may have some dosimetric implications. These are the uranium, thorium and radium isotopes as well as the ^{210}Pb - ^{210}Bi - ^{210}Po isotopes.

Gamma spectrometry

Although gamma spectrometry of the natural decay chain radioisotopes were useful already using NaI-detectors the introduction of high resolution gamma spectrometry using Ge-detectors significantly improved the analytical capabilities. The main advantage of using gamma spectrometry over other radiometric methods is the ease of with it can be performed and that the method is non destructive. Another important advantage is that several gamma emitting radioisotopes can be determined simultaneously. The main disadvantage is that the sensitivity often required to analyse decay chain radioisotopes in biological samples is not sufficient. Therefore gamma spectrometry is normally used to determine concentrations of these radioisotopes in soils and sediments where levels are high enough. Apart from a few radioisotopes such as ^{210}Pb and ^{235}U the determination of most other members of the decay chain isotopes are determined via some or several of their daughter radioisotopes. The ^{238}U isotopes is for instance determined via its short lived (24d) daughter ^{234}Th , the ^{232}Th isotope (in aged samples) via the ^{228}Ac isotope, ^{226}Ra via its ^{222}Rn daughters ^{214}Bi and ^{214}Pb , ^{228}Ra via ^{228}Ac and ^{228}Th via ^{208}Tl . The reason for choosing the short-lived daughter radioisotopes is simply because of their abundant number of suitable gamma lines (high branching ratio and suitable emission energies in the 100-2000 keV range) in combination with poor abundance and/or low energy emission of gamma lines from the mother isotopes. Unfortunately there are several decay chain radioisotopes which lacks gamma emission to the extent that they can be analysed in most normal types of samples. These isotopes include ^{234}U , ^{230}Th and ^{210}Po .

Most of the gamma lines used when determine the decay chain radioisotopes are in the range 100-2000 keV. Exceptions are lines from ^{210}Pb (46 keV) and ^{238}U - ^{234}Th (63 & 93 keV). The low energy lines pose problems related to self absorption in the sample as well as low detection efficiency unless N-type HPGe detectors are used (with thin entrance window). The self absorption in the sample furthermore sets a limit as to what amount of sample that is useful to measure. The self absorption is both density and atomic number dependent and especially the latter may become important when using these low energies in samples rich in minerals having high atomic number (eg pitchblende). Corrections for self absorption which only relies on density will then underestimate the true content of the sample. This is a problem for low energy gammas, for higher energies the absorption in practice depends mainly on the density and to a limited degree on the

chemical (atomic number). For more correct determinations, transmission based techniques may be used. In this case the sample self absorption is measured using an external source of ^{210}Pb and ^{238}U (^{234}Th).

A factor limiting the sensitivity in determining the natural decay chain isotopes using gamma spectrometry is the presence of these isotopes in nearly all the material surrounding the detector and even inside the detector cryostat and electrical components. Effective shielding and choice of materials is an effective way of decreasing this contribution but for some of the radioisotopes (eg ^{226}Ra determination via Rn-daughters as well as ^{210}Pb) a considerable pathway is the air around the detectors. Constant flushing of the interior using N₂-gas can significantly reduce this background. Further background reduction in order to reduce first the neutron component (prompt gammas and activation products) as well as the muon component requires underground laboratories. Although connected to considerable cost such underground laboratories show detection capabilities which even match alpha spectrometry and in some cases mass spectrometry.

It should also be mentioned that gamma spectrometry may be used in connection with radiochemical methods where the element in question is isolated from the sample to improve counting efficiency. An example of this technique is the efficient extraction of ^{234}Th from hundreds or even thousands of liter of seawater using MnO₂ impregnated filters which then are ashed and packed into small canisters for counting. Direct counting of the ^{234}Th in seawater is not possible. The concentrations are in the range 0.01-0.03 Bq per litre.

Beta counting and beta spectrometry

The nature of the continuous beta spectrum from beta emitters and the energy degradation during electron interaction with matter makes it necessary to perform element separation by chemical methods in order to analyse decay chain radioisotopes using beta counting. Spectrometric methods (eg proportional counter or liquid scintillation spectrometers) provide spectrometric information but spectra usually require unfolding techniques to be able to separate contributions from individual emitters. In practice the GM-counters following proper radiochemical separation is commonly used to determine isotopes like ^{234}Th (^{234}Pa) and ^{210}Pb - ^{210}Bi . Purity of the isolated sample is usually determined from repeated counting and fitting the decay (^{234}Th) or ingrowth (^{210}Bi) to the known half-life. Both ^{234}Th and ^{210}Pb may be determined using gamma spectrometry but as mentioned above they both emit low energy lines making the reliability and sensitivity of gamma spectrometry low. The extensive use of both ^{234}Th and ^{210}Pb in marine sciences (mainly in connection with removal of particulate carbon from the euphotic zone as well as sediment dating and mixing determinations) has made beta counting of these two radioisotopes an important topic. When using radiochemical methods to separate these elements suitable yield tracer isotopes must be used. For the ^{234}Th isotope usually ^{229}Th is used. The emission of electrons from other isotopes in the purified Th-sample (conversion-, auger electrons and beta particles) will increase the background and make it time dependent due to ingrowth of daughter isotopes so it is important to also quantify the content of other Th-isotopes in the sample (notably ^{228}Th) which normally is done using alpha spectrometry. Use of beta counting for other decay chain radioisotopes is rare.

Alpha spectrometry

Due to the alpha particle emission from several of the natural decay chain radioisotopes, alpha spectrometry is a suitable detection method to determine concentrations of these elements. The main radioisotopes determined by alpha spectrometry are all the uranium isotopes, the ^{238}U , ^{235}U and ^{234}U , nearly all the Th-isotopes ^{232}Th , ^{230}Th , ^{228}Th (but not ^{234}Th and ^{231}Th). Also the dosimetric important ^{210}Po is most favourable determined using alpha spectrometry. Although the alpha particle energy for each isotope has an inherent energy spread which is negligible the rapid energy

degradation of alpha particles in matter quickly produces tailing towards lower energy. Alpha particle spectrometry is to a large extent about spectrum resolution. Even with the best available spectrometric equipment of any kind this energy degradation necessitates radiochemical separation methods in order to both separate different alpha emitting elements from each other and to isolate the element in question from all matrix elements so that an infinitesimal thin source can be prepared from which the loss of alpha particle energy is negligible. Such sources may in practice best be prepared using electrolytic deposition onto some smooth material such as mirror blank stainless steel discs. The inherent loss of energy of 5 MeV alpha particles from such a source may be reduced to a few keV. Main sources of loss of resolution in semiconductor detectors is through the entrance window and through detector edge effects (where charge collection is less effective) and the capacitive coupling (the detector acts as capacitor and the corresponding leakage currents limits the resolution). This leakage current increase, and thus makes resolution worse, the thinner the detector depletion depth and the larger the surface area. As with all semiconductor detectors leakage currents are also proportional to the temperature but silicon based alpha particle detectors have little gain in being used cooled and are thus operated at room temperature. If using a proportional counter or a LSC the energy required to produce ion pairs in the medium will be large (30 eV to 1 keV) which therefore will be the main source of poor energy resolution and these two detection techniques cannot compare with semiconductor based alpha spectrometry where ion pairs are created using only a few eV. The dominating detector type used for alpha spectrometry is semiconductor based detectors. The advantages of these detectors are several. They have a good energy resolution and can be placed very close (~mm) to the source to achieve high counting efficiency (~30% readily achievable), detector background is excellent with count rates less than 1 count per day in a chosen peak energy window. The alpha particle part of the energy spectrum (4-9 MeV) is furthermore in principle completely free from beta particle and gamma photon interferences. Detectors are cheap and last for many years if taken proper care of. The main disadvantage of using such detectors is the need for extensive chemical cleanup to produce the thin source needed. When large soil or sediment samples in some occasions are required to analyse the more rare U-isotopes (eg ^{236}U from nuclear installations) the amount of uranium itself on the steel disc will limit the resolution. The specific activity of ^{238}U is 12.3 Bq/mg and for ^{232}Th 4 Bq/mg. Already at depositions of around some tens of micrograms of uranium and thorium on the disc resolution starts to become worse but due to large energy difference between peaks the tolerance is usually some hundred micrograms of each element on the disc. If larger amounts exist mathematical methods may be used to unfold the spectra.

For both uranium and the thorium isotopes excellent yield tracers exist as ^{232}U and ^{229}Th . Although ^{233}U and ^{236}U also may be used as yield tracers for uranium their alpha peaks partly interfere with ^{234}U and ^{235}U respectively. The main disadvantage with the ^{232}U tracer is the ingrowth of the ^{228}Th daughter which requires frequent removal in case the sample is analysed for thorium isotopes in a sequential way. For ^{210}Po the common tracers are ^{208}Po and ^{209}Po . Both are commonly used. The energy separation between the ^{209}Po and ^{210}Po is larger (400 keV) than the ^{208}Po - ^{210}Po (200 keV) which imply that a worse energy resolution may be accepted when using ^{209}Po as tracer as when ^{208}Po is used. The relative short half life of the ^{209}Po isotope (2.8y) is another disadvantage.

For the radium isotopes (^{226}Ra , ^{228}Ra , ^{224}Ra & ^{223}Ra) measurement are usually focused on the two first isotopes, ^{226}Ra and ^{228}Ra due to their longer half lives. Since source preparation of radium for solid state alpha spectrometry is more difficult than for the actinides and polonium isotopes other analytical techniques are usually employed. For ^{226}Ra the absolutely most efficient and easiest method is by using radon emanation. Radium may be isolated chemically but the cleanup of the sample is not so critical as when working with solid state detectors. The purified radium sample is usually taken up in a small (10ml) water sample and placed in a LSC cell to monitor ^{222}Rn ingrowth or similarly placed in a glass tube, evacuated and bubbled to release radon into an Lucas ZnS cell

for scintillation counting following 3 weeks of ^{222}Rn ingrowth to reach equilibrium with ^{226}Ra . The main disadvantage with these methods (as compared with determination using gamma spectrometry) is the lack of suitable tracers for the chemical work of radium. The only isotopic tracer is the 10d half live ^{225}Ra produced in the ^{229}Th decay chain. Its presence however requires analysis using a separate detector system, usually semiconductor detectors which therefore make its use limited. Instead ^{133}Ba has been used as a chemical analogue in the chemical work. Barium is chemically very similar to radium but separation between them may occur during the processing of samples. Such separation must then be identified. The determination of the weak beta emitter ^{228}Ra isotope is not straight forward. If not determined through gamma spectrometry using the short lived (6h) ^{228}Ac it may be determined by its ^{228}Th daughter following a suitable ingrowth period, usually some months depending on the expected ^{228}Ra in the sample.

It is difficult to compare alpha and gamma spectrometry but in general alpha spectrometry can be said to be in the order of 10 to 1000 times more sensitive than gamma spectrometry dependent on sample size, isotope and counting geometry, time etc.

Mass-spectrometry

With development of mass spectrometric techniques during the last 20y this technique has gained a growing interest among users in the field of determining natural decay chain radioisotopes. Since mass spectrometry is about counting the atoms rather than the emission from the isotopes mass spectrometry has preferably been applied to long lived radioisotopes (low specific activity) and thus been applied to all the U-isotopes, the ^{232}Th and ^{230}Th (but not ^{228}Th which is far too short lived) and ^{226}Ra . Three techniques have dominated the field, Thermal ionization mass spectrometry (TIMS), inductively coupled plasma mass spectrometry (ICP-MS) and accelerator mass spectrometry (AMS). Of the techniques the TIMS method has been employed for decades and has been the ultimate tool to perform high precision isotope ratio measurements with. For isotopes having half lives larger than about 1ky this technique shows higher sensitivity than any radiometric technique. Chemical separation of the elements to be analysed is of similar magnitude as with alpha spectrometry. Unfortunately TIMS instruments are expensive and require relatively high skills to operate. Sample throughput rate is relatively low.

In contrast to TIMS the ICP-MS instruments have developed radically during the last 10-15 years. Today multi collector ICP-MS instruments have surpassed commercial TIMS instruments with respect to sensitivity and isotope ratio precision. The relatively low cost of standard type ICP-MS instruments means that nearly all U & Th measurements have been transferred from alpha spectrometry to ICP-MS. This however only holds if the ^{228}Th isotope is not to be analysed. Due to its short half life alpha spectrometry is the only possibility to determine it at normal environmental concentrations. Similarly the ^{230}Th may pose problems when using mass spectrometry. The activity ratio ^{230}Th to ^{232}Th is usually in the order of 0.5 – 2 in most environmental samples but the corresponding mass ratio is in the order of 10^{-7} . This sets high requirements on the analyst to perform all necessary corrections such as dead time and detector linearity corrections.

Finally, AMS, has emerged as a technique where extreme isotope ratios may be determined. Also its sensitivity will continue to develop and reach to levels where the other techniques cannot follow. Already now mass spectrometric techniques routinely analyse ^{238}U and ^{232}Th in the μBq range in a few minutes counting time. Analysis of nano Bq is possible using most type of mass spectrometers and even the next step (pico Bq) is within reach. The absolutely dominating problem when approaching these levels is not the instrument sensitivity as such but the blank problems associated with handling the samples.

Although mass spectrometric techniques are very useful for both high precision ratio instruments and to perform ultrasensitive analysis of selected radioisotopes the main disadvantage is that they

don't measure dosimetric important radioisotopes – those having a relatively high specific activity (^{226}Ra , ^{210}Pb , ^{210}Po). Thus a combination of radiometric and mass spectrometric techniques is to be preferred.

Neutron activation and fission track techniques

Finally, there are some techniques which are very useful when analyzing radioisotopes having a high cross section for neutron induced fission and/or neutron activation. In the case of fission the particle tracks are detected using a plastic etchable film in a similar way as when using radon track etch film. The ^{235}U isotope have a high cross section for fission and this combined with a more or less fixed $^{235}\text{U}/^{238}\text{U}$ ratio in environmental samples makes this technique useful for uranium determination. Both ^{238}U and ^{232}Th may be neutron activated to the short lived ^{239}U and ^{233}Th isotopes which are easily determined by gamma spectrometry several order of magnitude easier than the ^{238}U and ^{232}Th themselves. Advantages of neutron activation techniques are that they can be done simultaneously on several (hundreds) samples. The obvious disadvantage is the need of a strong neutron source such as a nuclear reactor. Although several of the other natural decay chain radioisotopes show high neutron capture cross sections their atomic abundance is far too low to be useful for this technique.

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Abstract	<p>The amount of natural radionuclides in the environment differs between the Nordic countries as shown by previous investigations and also by this study. Agricultural areas of high natural background are predominantly found in Sweden, Southern Finland and Norway while low background areas are typical for Iceland and Denmark. Thus, this study offers possibilities for studying behaviour of natural radionuclides under different conditions such as the influence of different soil types as well as the husbandry. Furthermore the areas also enable studying environmental behaviour of radium and other natural radionuclides under seemingly steady state conditions. However, migration and accumulation of natural radionuclides in cultivated soil is complex involving various processes. Thus, a long term goal of this study was to identify the implications of some of these processes by determining the soil to plant transfer for pasture land under the different conditions that prevail in the Nordic countries. The potential health hazards due to chronic ingestion of low concentrations of naturally occurring radionuclides are fairly unknown but the results of this study may provide valuable background information for assessing these radiation risks.</p> <p>The aim of this project has been to gain knowledge on the status of natural radionuclides in meadow and pasture land and in grassland plants in different Nordic countries and on the transfer of these radionuclides from soil/water to man via the milk/food chain (soil- meadow/pasture grass –cow-milk). Limited data are available on the mobility and the transfer of naturally occurring radionuclides in the ecosystems of the agricultural land. In addition, information concerning the concentrations in meat and dairy products is of interest for assessing exposures of humans to natural radionuclides.</p> <p>Soil characteristics are known to have significant impact on the mobility and uptake of natural radionuclides. Therefore, the uptake in relation to different soil types such as sandy, organic and/or clay soil was investigated. The use of fertilizers at some of the investigated farms has been considered but no analysis was performed on the content of natural radionuclides in fertilisers.</p>
Key words	Natural Radionuclides & Cesium in soil, grass, milk, water, fodder, 238U, 235U, 232Th, 228Th, 226Ra, 228Ra, 210Pb, 210Po, 40K, and 137Cs