

Environmental Impact of Radionuclides and Trace Elements
in the Thorium Rich Fen Area in Norway

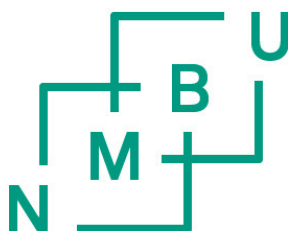
Miljøkonsekvenser knyttet til radionuklider og sporelementer
i det thoriumrike Fensfeltet i Norge

Philosophiae Doctor (PhD) Thesis

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Jelena Mrdaković Popić, Ås, December 2013

Summary

The Fen Complex which is situated in the south of Norway, represents a magmatic bedrock area enriched in thorium (Th), iron (Fe), niobium (Nb) and rare earth elements (REE), and is well known for the elevated levels of natural ionizing radiation. This area has been of public interest from the 17th century when Fe mining started in the central wooded zone of the Fen Complex. Intensive mining of Fe continued until the 20th century, while mining and the production of ferro-niobium were conducted at the site Søve in the western part of the Complex in 1950s. Recently, intensive focus has been directed to the estimated large quantities of Th and REE ores, their value and possibility for future use, but also to environmental issues linked to the legacy enhanced naturally occurring radioactive materials (NORM) in the area. Many studies investigating different aspects of specific bedrock geology, as well as human health risk related to elevated ionizing radiation levels in the area, have been published. Still, no comprehensive investigation of different environmental compartments and radionuclides impact on biota at both legacy enhanced and undisturbed NORM sites in the Fen Complex has been undertaken.

The present work was initiated as an integrated ecological and human impact assessment whose main objectives were to assess the possible radionuclide and trace elements contamination of the Fen Complex environment, impact on biota and radiation doses to humans due to outdoor radiation exposure. The Fen Complex area which is comprised of both legacy NORM and undisturbed ²³²Th-rich sites served as a natural laboratory where environmental compartments and biota could be investigated in the natural state, under realistic conditions. With respect to the specific Fen area and previously published data, the main focus of this work was on radionuclides such as ²³²Th and uranium (²³⁸U) and their progenies, as well on trace elements such as arsenic (As), chromium (Cr), cadmium (Cd) and lead (Pb). The generated data provided information on the current environmental status at both undisturbed and legacy NORM sites in the area, and could be used in planning of eventual remediation activities or for future monitoring.

To assess the impact of radionuclides and trace elements on the ecosystem and humans, information was needed regarding the characterization, mobility and biological uptake of radionuclides and trace elements, as well as their different exposure pathways. These aspects were studied and presented in five scientific papers on which this thesis is based on.

The first paper (Paper I) describes the initial screening of the Fen Complex. It included the analysis of radionuclides and trace elements in samples of soil, rock, water and plants. The high gamma dose rates in outdoor air were recorded. Based on the obtained data, radionuclides in soil were inhomogeneously distributed and "hot spots" with high levels of radionuclides (up to about 7000 Bq/kg ^{232}Th and 150 Bq/kg ^{238}U) and elevated gamma dose rates (up to 10 $\mu\text{Gy/h}$) were identified. "Hot spots" were observed within legacy NORM (former mining) sites, and also in some undisturbed surrounding ^{232}Th -rich sites. The initial ERICA impact assessment demonstrated that dose rates for certain terrestrial organisms were higher than the adopted screening level (10 $\mu\text{Gy/h}$), suggesting the need for more refined analysis. In addition to the previously published literature, the data presented in Paper I assisted in defining future directions of investigation, the choice of sites for more detailed surveys, biota selection and in defining aspects of human exposure that would be investigated.

Binding mechanisms in soil determine the potential mobility and bioavailability of radionuclides and trace elements, and soil fractionation is therefore essential for assessing their behavior in the environment. In the present work, mobility analysis of the investigated elements, based on the results of sequential extractions, was performed (Paper II). Soil fractionation showed that the majority of ^{232}Th and As were irreversibly bound in the soil as they were only leached by concentrated HNO_3 at elevated temperature. With respect to ^{232}Th , the result was in accordance with its chemical nature, established low mobility and tendency to be tightly bound in soil fractions. Uranium and trace elements (Cr, Cd, Pb, Ni, Cu and Zn) were found to be potentially more mobile and associated with pH-sensitive soil phases, redox-sensitive amorphous soil phases and organic soil compounds. Multivariate statistical data analysis provided the link between soil chemical and physical parameters and output data from the sequential extractions. Further mobility investigation was performed by determining the distribution coefficients (K_d). The K_d (^{232}Th) and K_d (^{238}U) suggested elevated dissolution and mobility at legacy NORM sites, especially at decommissioned Nb mining site (346 and 100 L/kg for ^{232}Th and ^{238}U , respectively), while higher sorption of radionuclides was demonstrated at undisturbed ^{232}Th -rich site (10672 and 506 L/kg for ^{232}Th and ^{238}U , respectively).

Earthworms were chosen as biota representative for a detailed analysis of radionuclide and trace element uptake and chronic exposure to radiation (Paper III). Tissues of four different earthworm species, including epigeic and endogeic species, were analyzed and the results were linked to total soil concentrations, bioavailable or extractable soil fractions, and root and litter concentrations in order to predict the

favorable environmental pool for uptake. Variability in individual tissue concentrations of radionuclides was observed to be high as previously demonstrated in scientific literature for heavy metals in earthworms. Differences in uptake between four investigated earthworm species, but also between species collected at legacy NORM and undisturbed ^{232}Th -rich sites were demonstrated. Higher transfer was observed for ^{238}U (TF = 0.09 – 0.25) than for ^{232}Th (TF = 0.03 – 0.08). Radiological dose rates (2.2 – 11.9 $\mu\text{Gy/h}$), obtained by ERICA modeling, were higher than those generally experienced by terrestrial organisms (0.01 – 0.7 $\mu\text{Gy/h}$) in the soil with background radionuclide concentrations. However, no radiation risk could be predicted since the obtained doses were much lower than the internationally (IAEA, 1992; UNSCEAR 2008; US DOE 2002) adopted levels of ionizing radiation below which no measurable population effects would occur (40 and 400 $\mu\text{Gy/h}$).

Further study of biota exposure was performed on nine wild plant species. A wide range of plants was included, with different uptake modes taken into account, and including both roots and aboveground plant parts (Paper IV). Plant tissue concentrations of radionuclides were only slightly enhanced (up to 50 and 5 Bq/kg of ^{232}Th and ^{238}U , respectively), and comparable to the values reported in literature. The levels of trace elements were within the reference range for plants. Roots appeared to be a natural barrier to radionuclide entry into plants. This is illustrated by the finding that the activity concentrations were higher by a factor of 25 in roots than in the aboveground plant parts. Thus, the transfer factors for plants were actually lower ($4 \cdot 10^{-5}$ – $1 \cdot 10^{-2}$ for ^{232}Th and $1 \cdot 10^{-4}$ – $4 \cdot 10^{-2}$ for ^{238}U) than expected from the observed total soil concentrations (about 16000 Bq/kg ^{232}Th and 900 Bq/kg ^{238}U). Based on the ERICA calculation, dose rates up to 23 $\mu\text{Gy/h}$ (in moss and lichen) were predicted.

Previous studies on human health risk in the Fen area have demonstrated that the annual exposure doses were among the highest in Europe (about 14 mSv). The majority of these investigations focused on the doses of indoor gamma radiation and radon (^{222}Rn), as well as on the doses received via ingestion of food and water. In the conclusions presented in several papers, the need for investigating the contribution of outdoor exposure to total exposure doses, as well for measuring the ^{220}Rn concentration in air was highlighted. This information focused our investigation into outdoor ^{220}Rn , ^{222}Rn and terrestrial gamma radiation (Paper V). Compared to the world average, high outdoor gamma dose rates (about 10 $\mu\text{Gy/h}$), high ^{220}Rn (up to 5000 Bq/m³) and moderate ^{222}Rn (up to 200 Bq/m³) concentrations in the air were recorded in the Fen area. Levels of these parameters correlated with the distribution of radionuclides in the bedrock. Due to the high uncertainty when ^{220}Rn is used to calculate the exposure doses, the annual outdoor doses (0.10 – 1.54 mSv) were

obtained by summarizing the doses from terrestrial gamma radiation and ^{222}Rn doses. However, when variations in the exposure times and the ^{220}Rn dose (calculated using the equilibrium factor F from an earlier study in the Fen) are accounted for, an increase in the annual outdoor doses for over 10 mSv for at least some people can be expected.

Based on the overall results, the high concentrations of radionuclides in the soil, the high levels of terrestrial gamma radiation and the high outdoor levels of ^{220}Rn and ^{222}Rn were observed at both legacy NORM and undisturbed ^{232}Th -rich sites in the Fen Complex. However, due to the relatively low mobility of radionuclide ^{232}Th , no significant transport into investigated biota, such as earthworms and plants, was demonstrated so that low radiation doses and no elevated risk were predicted using the ERICA tool. However, the question of synergistic, additive or antagonistic actions of radionuclides and trace elements in biota of the Fen Complex remains. In the analysis of human outdoor exposure, high terrestrial gamma radiation was demonstrated to have a major impact on the magnitude of the received doses. Outdoor exposure doses higher than 10 mSv/y, for at least a certain group of people under specific exposure scenarios, were estimated as possible. Due to the high outdoor ^{220}Rn concentrations, the contribution of ^{220}Rn to the total dose of outdoor exposure could not be excluded, although quantification of such a contribution with the present ^{220}Rn data set is connected to a large uncertainty. With respect to the current Norwegian legislation, an intervention should be considered at the legacy NORM site Søve. Although, there is little that could be done to change the doses at undisturbed high radioactivity sites, it would be reasonable, where possible, to avoid house construction, to restrict the time spent for recreation and to limit the use of the materials with elevated radioactivity.

Sammendrag

Fensfeltet, lokalisert i Sør Norge, representerer et magmatisk berggrunnsområde beriket med thorium (Th), jern (Fe), niob (Nb) og sjeldne jordelementer (REE). Det er velkjent for forhøyet nivå av naturlig ioniserende stråling. Fensfeltet har vært av offentlig interesse siden 1600-tallet da gruvedriften av Fe begynte i sentral skogkledd sone. Det ble drevet intensiv gruvedrift av Fe frem til 1900-tallet, mens gruvedrift og produksjon av ferro-niob ble gjennomført på 1950-tallet i Søve gruver, i den vestlige delen av komplekset. Nylig ble fokus igjen rettet mot Fensfeltet og store estimerte konsentrasjoner av Th og REE, deres verdi og muligheter for fremtidig bruk, samt mot miljøproblematikk knyttet til historiske NORM områder som eksisterer i området. Det har blitt publisert flere studier som forsket på forskjellige aspekter av spesifikk berggrunnsgeologi, samt helserisiko for mennesker relatert til forhøyede nivåer av forhøyet ioniserende stråling i område. Fortsatt har ingen omfattende forskning på forskjellige miljøaspekter og påvirkning av planter i verken tidligere gruveområder eller uforstyrrede NORM områder i Fensfeltet blitt gjennomført.

Dette arbeidet ble initiert som en integrert studie for miljøvurdering. Hovedmålet var å få til risikovurdering av av Fensfeltet fra radioaktivitet og metaller blant annet ved å se på påvirkning av biota og stråledoser for mennesker gjennom utendørs eksponering. Fensfeltet, bestående av både tidligere gruveområder med NORM og uforstyrrede områder rike på ^{232}Th , ble brukt som naturlige laboratorier hvor jord, vann, luft og biota kunne undersøkes under realistiske forhold. Basert på tidligere publiserte data og opplysninger om området, ble hovedfokus rettet mot radionuklider som ^{232}Th , uran (^{238}U) og deres døtre samt sporelementer som arsen (As), krom (Cr), kadmium (Cd) og bly (Pb). Genererte data ga opplysninger om nåværende miljøstatus i både uforstyrrede og historiske NORM områder. Disse dataene kan bli brukt til planlegging av eventuelle aktiviteter for utbedring og fremtidig overvåkning. For å vurdere radionukliderelatert påvirkning av økosystemet og mennesker, var det nødvendig å gjøre karakterisering av forskjellige prøver, undersøke mobilitet og biologisk opptak av radionuklider og sporelementer, samt deres eksponeringsvei og påvirkning av biota og mennesker. Disse aspektene ble studert og presentert i fem vitenskapelige artikler som denne doktoravhandlingen er basert på.

I den første artikkelen (Paper I) ble det gjort en innledende undersøkelse av Fensfeltet. Dette inkluderte analyser av radionuklider og sporelementer i jord, stein, vann og planter. Nivåer av gammastråling i utendørs luft ble registrert. Innhentede data viste at radionuklider i jord var heterogent fordelt og "hotspots" med høye nivåer av radionuklider (opp til 7000 Bq/kg ^{232}Th og 150 Bq/kg ^{238}U) og høye nivåer av

gammastråling (opp til 10 $\mu\text{Gy/h}$) ble identifisert. "Hotspots" med forhøyede verdier av ^{220}Rn ble observert i både tidligere gruveområder med NORM og uforstyrrede ^{232}Th -rike områder. ERICA-modellert vurdering viste at doser for visse organismer var høyere enn vedtatt screening nivå på 10 $\mu\text{Gy/h}$, noe som tyder på behov for mer sofistisert analyse. Sammen med tidligere publiserte data, var dataene presentert i Artikkel I avgjørende for planleggingen av videre forskning, valg av områder for mer detaljert undersøkelse, valg av biota og definisjon av aspekter for menneskelig eksponering som ble undersøkt videre.

Den kjemiske spesieringen forteller noe om mobilitet og biotilgjengelighet av radionuklider og sporelementer, og er derfor avgjørende for å vurdere oppførselen og skjebnen i miljøet. I dette arbeidet ble det gjennomført jord- og vannfraksjonering og mobilitetsanalyse, i tillegg til måling av totale konsentrasjoner (Artikkel II). Jordfraksjonering ved hjelp av sekvensiell ekstraksjon, viste at ^{232}Th og As var irreversibelt bundet i flertallet av jordprøvene. Fordelingen av ^{232}Th var i samsvar med dets kjemiske natur og kjente lave mobilitet, samt tendensen til å være fast bundet i krystalliserte jordfraksjoner. Uran og sporelementer (Cr, Cd, Pb, Ni, Cu og Zn) viste seg å være mer potensielt mobile og assosiert med pH-sensitive jordfaser, redox-sensitive amorfe jordfaser og jord-organisk stoff. Multivariat statistiske analyser av dataene ga oss leddet mellom kjemiske og fysiske parametere i jord og resultater av sekvensiell ekstraksjon. Den videre undersøkelsen av mobilitet ble gjort med analyse og beregning av fordelingskoeffisienten (K_d). De beregnede verdiene viste forhøyet mobilitet og mulighet for transport videre i miljøet ved tidligere gruveområder, spesielt ved gruveområdet for Nb ved Søve (346 and 100 L/kg for henholdsvis ^{232}Th and ^{238}U), mens høy adsorpsjon og dermed lavere mobilitet, ble demonstrert ved det naturlige ^{232}Th -rike området Rullekoll (10672 and 506 L/kg for henholdsvis ^{232}Th and ^{238}U).

Meitemark ble valgt som biota egnet for detaljert analyse av radionuklider og sporelementer, samt opptak og mottak av kroniske stråledoser (Artikkel III). Vev fra forskjellige arter, inkludert epigeiske og endogeiske meitemark fra Fensfeltet, ble analysert for radionuklider og sporelementer. Resultatene ble koblet til totale jordkonsentrasjoner, biotilgjengelige jordkonsentrasjoner og konsentrasjoner i røtter for å kunne forutsi foretrukket medium for opptak. Variasjon i individuelle vevkonsentrasjoner ble observert i likhet med tidligere publiserte data i internasjonal litteratur for tungmetaller i meitemark. Det ble påvist forskjeller i opptak mellom forskjellige arter, men også forskjeller i undersøkte områder. En høyere overføring av ^{238}U (TF = 0,09 – 0,25) enn av ^{232}Th (TF = 0,03 – 0,08) ble demonstrert. ERICA modellering påviste høyere radiologiske doser (2,2 – 3,9 $\mu\text{Gy/h}$) enn gjennomsnittlige

doser (0,01 – 0,7 $\mu\text{Gy/h}$) for jordorganismer som lever i miljøer med lave nivåer av radionuklider i bakgrunn. Likevel kunne man ikke forutsi noen økt strålerisiko, siden beregnede doser fortsatt var mye lavere enn internasjonalt vedtatte nivåer av ioniserende stråling som kan produsere biologiske effekter (40 og 400 $\mu\text{Gy/h}$).

Videre analyse av biotaeksponering ble gjort for et bredt spekter av samlede planter fra Fensfeltet, inkludert både røtter og hele planter av ni forskjellige plantearter (Artikkel IV). Konsentrasjon av radionuklider i plantevev var kun lett forhøyet (opp til 50 Bq/kg ^{232}Th og opp til 5 Bq/kg ^{238}U) og i godt samsvar med verdier sett i litteraturen. Nivåer av sporelementer var innenfor referanseområdet for planter. Røtter virket å representere en naturlig barriere for opptak av radionuklider i planter siden aktivitetskonsentrasjoner i disse var opptil 25 ganger høyere enn i plantedelene over jorda. Generelt sett var overføringsfaktorene for planter ($4 \cdot 10^{-5}$ til $1 \cdot 10^{-2}$ for ^{232}Th and $1 \cdot 10^{-4}$ til $4 \cdot 10^{-2}$ for ^{238}U) lavere enn forventet ut i fra konsentrasjonene i jord (16000 Bq/kg ^{232}Th og 900 Bq/kg ^{238}U). Basert på ERICA-modellering ble det påvist doser opp til 10 $\mu\text{Gy/h}$ (i mose og lav). Tilsvarende som for meitemark, ble konklusjonen at ingen risiko kunne estimeres og det var ikke nødvendig med videre analyser.

Tidligere studier av helserisiko for mennesker i Fensfeltet har vist eksponeringsdoser fra gammastråling som er blant de høyeste i Europa (rundt 14 mSv). Majoriteten av disse studiene fokuserte på innendørs gammastråling og radon (^{222}Rn) doser samt på doser inntatt gjennom mat og vann. I konklusjonen til flere artikler ble det understreket et behov for undersøkelse av bidrag fra utendørs eksponering i beregninger av totale eksponeringsdoser, samt måling av ^{220}Rn og ^{222}Rn konsentrasjoner i luft. Dette rettet vår forskning mot utendørs ^{220}Rn og ^{222}Rn og jordisk gamma stråling (Artikkel V). Sammenlignet med verdensgjennomsnittet ble det i Fensfeltet registrert høye utendørs gammadoser (opp til 10 $\mu\text{Gy/h}$), høye ^{220}Rn - (5000 Bq/m^3) og moderate ^{222}Rn - (200 Bq/m^3) konsentrasjoner i luft. Nivåene av disse parameterne var korrelert med distribusjonen av radionuklider (Th og U) i berggrunn. På grunn av stor usikkerhet ved bruk av ^{220}Rn konsentrasjon i luft for å beregne eksponeringsdoser, ble de totale årlige utendørsdosene (0,10 – 1,54 mSv) gitt som summen av doser fra gamma stråling og doser fra ^{222}Rn . Ved variasjon av eksponeringstid ved beregning av ^{220}Rn -doser (ved hjelp av likevektsfaktor F_i fra tidligere studier i Fensfeltet) ville man få høyere årlige utendørsdoser, over 10 mSv, i hvert fall for noen grupper mennesker.

Basert på de samlede resultatene ble det målt høye konsentrasjoner av radionuklider i jord, samt høy jordisk gammastråling og utendørnivåer av Rn i både tidligere gruveområder og i uforstyrrede ^{232}Th -rike områder i Fensfeltet. På tross av dette ble det, grunnet relativt lav mobilitet av radionukliden ^{232}Th , ikke påvist noen signifikant

transport til undersøkte biota (planter og meitemark). Derfor ble det beregnet lave eksponeringsdoser ved bruk av ERICA-programmet og ingen økt risiko kunne bli estimert. På tross av dette, er spørsmålet om synergistisk, additiv eller antagonistisk virkning av radionuklider og sporelementer i biota fortsatt åpent. I analysen av utendørs eksponering av mennesker, hadde høy gamma stråling hoved-innvirkningen på de årlige strålingsdosene. På grunn av høye ^{220}Rn konsentrasjoner, kan ikke et bidrag fra ^{220}Rn til de totale årlige doser kunne utelukkes, selv om kvantifisering av årlige doser (>10 mSv) er koblet til stor usikkerhet og derfor ikke anbefalt av internasjonale organisasjoner. I følge den nåværende Norske Forurensningsloven, burde man vurdere tiltak i det tidligere gruveområdet Søve. Generelt er det lite som kan gjøres på uforstyrrede områder med naturlige høye nivåer av radioaktivitet, men det kunne vært fornuftig å begrense bygging av hus, bruk av radioaktive materialer fra området og begrense tilgang til områdene med påvist høy radioaktivitet.

Summary of publications

This thesis is based on the following papers which are referred to in the text by their Roman numerals.

- Paper I** Mrdakovic Popic, J., Salbu, B., Strand, T., Skipperud, L. (2011). Assessment of radionuclide and metal contamination in a thorium rich area in Norway. *Journal of Environmental Monitoring* 13, 1730-1738.
- Paper II** Mrdakovic Popic, J., Meland, S., Salbu, B., Skipperud, L. (2014). Mobility of radionuclides and trace elements in soils from legacy NORM and undisturbed ^{232}Th -rich sites. *Environmental Science: Impacts and Processes*, in press, DOI: 10.1039/C3EM00569K.
- Paper III** Mrdakovic Popic, J., Salbu, B., Skipperud, L. (2012). Ecological transfer of radionuclides and metals to free-living earthworm species in natural habitats rich in NORM. *Science of the Total Environment* 414, 167-176.
- Paper IV** Mrdakovic Popic, J., Oughton, D., Salbu, B., Skipperud L. Transfer of radionuclides (^{232}Th , ^{238}U) to wild forest flora species in a thorium rich area, *Manuscript in preparation*.
- Paper V** Mrdakovic Popic, J., Bhatt, C.R., Salbu, B., Skipperud, L. (2012). Outdoor ^{220}Rn , ^{222}Rn and terrestrial gamma radiation levels: investigation study in the thorium rich Fen Complex, Norway. *Journal of Environmental Monitoring* 14, 193-201.

List of Acronyms and Codes

ALARA	“As Low As Reasonable Achievable” radiation safety principle
BSS	Basic Safety Standards
CR	Concentration Ratio
DCC	Dose Conversion Coefficient
EEC	Equilibrium Equivalent Concentration
Eh	Redox Potential
ENRA	Enhanced Natural Radiation Area
ERA	Environmental Risk Assessment
ERICA	Environmental Risk from Ionizing Contaminants: Assessment and Management
FASSET	Framework for Assessment of Environmental Impact
F	Equilibrium Factor
FWHM	Full Width at Half Maximum
GMC	Geiger Müller Counter
GPS	Global Positioning System
HCl	Hydrochloric Acid
HMM	High Molecular Mass
HNO ₃	Nitric Acid
H ₂ O ₂	Hydrogen Peroxide
HPGe	High Purity Germanium Detector
HTGR	High Temperature Gas Cooled Reactor
IAEA	International Atomic Energy Agency
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
ICP-OES	Inductively Coupled Plasma Optical Emission Spectrometry
ICRP	International Commission on Radiological Protection
JSI	Jožef Stefan Institute, Ljubljana, Slovenia
K _d	Soil/Solution Distribution (partition) Coefficient
LMFBR	Liquid Metal cooled Fast Breeder Reactors
LMM	Low Molecular Mass
LNT	Linear No Threshold
LOD	Limit of Detection
LOQ	Limit of Quantification

LWR	Light Water Reactor
MSBR	Molten Salt Breeder Reactor
NH ₄ OAc	Ammonium Acetate
NH ₂ OH · HCl	Hydroxylamine Hydrochloride
NIRS	National Institute of Radiological Science, Chiba, Japan
NOR	Naturally Occurring Radionuclides
NORM	Naturally Occurring Radioactive Material
NRPA	Norwegian Radiation Protection Authority
PCA	Principle Component Analysis
pH	Negative Logarithm of the Activity of the Hydronium Ion
PHWR	Pressurized Heavy Water Reactor
PIPS	Passive Implanted Planar Silicon
PNEDR	Predicted No Effect Dose Rate
REE	Rare Earth Elements
RQ	Risk Quotient
RSD	Relative Standard Deviation
SSNT	Solid State Nuclear Tracks
SV	Screening Value
TENORM	Technologically Enhanced Naturally Occurring Radioactive Material
TF	Transfer Factor
ThO ₂	Thorium Oxide
TLD	Thermo Luminescent Dosimeter
UMB (NMBU)	Norwegian University of Life Sciences
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
UO ₂	Uranium Oxide
UO ₂ ²⁺	Uranyl Ion
USGS	United States Geological Surveys
US EPA	United States Environmental Protection Agency
WHO	World Health Organization

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Paper I

Paper II

Paper III

Paper IV

Paper V

Papers I - V have individual page numbers

1 Introduction

The present study focuses on possible environmental impacts due to increased concentrations of radionuclides and trace elements in the Fen Complex which is situated in the Telemark county, southeastern Norway. Study sites, containing both undisturbed naturally occurring radioactive materials (NORM) and legacy NORM from past mining activities, represented convenient environmental laboratories to study radionuclides and associated elements and their speciation, distribution, impact on biota and humans. Different environmental compartments, i.e., air, soil, water and selected biota were studied. The approaches employed to assess the radionuclides and trace elements contamination and its impact on the environment are presented in the thesis.

An introduction to the Fen Complex area and its naturally occurring radioactive materials, the thesis hypothesis and objectives are given in Chapter 1. The theoretical background, the investigated parameters and approaches used to assess the ecological and human impacts are presented in Chapter 2. Problem formulation, identification of potential hazards, description of study sites, methodologies and measurement techniques are provided in Chapter 3. The obtained results, discussion and evaluation of the impacts based on the applied methodologies are presented in Chapters 4 and 5. The conclusions are in Chapter 6.

1.1 Naturally occurring ionizing radiation

Naturally occurring ionizing radiation, in the form of cosmic and terrestrial radiation, has been ubiquitously present in the environment since the Earth's origin. Exposure to radionuclides from the Earth's crust, together with cosmic rays and cosmogenic radionuclides, accounts for the greatest part of the contribution to the annual human radiation dose of an average individual (Betti *et al.*, 2004; MARINA, 2002; UNSCEAR, 2000, 2008a). Exposure to uranium (^{238}U), thorium (^{232}Th) and their progenies (all of the members of decay series are in Figure 4, section 2.1.1.), ^{235}U and potassium (^{40}K) generate the dose received from terrestrial radiation sources. When present in the body, these radionuclides irradiate an organism internally via alpha and beta particles and gamma rays; externally they irradiate with gamma radiation.

At background concentrations, the naturally occurring radionuclides (NOR) in the ^{238}U , ^{235}U , and ^{232}Th series, including radon (^{222}Rn), contribute to over 80 % of the background radiation to which all humans are continuously exposed (UNSCEAR,

2000, 2008a). The vast majority of the world population receives annual doses around of 2.4 mSv, the average global exposure. More than about 98 % of the population receives annual doses that are lower than 5 mSv, and about 99 % of the population doses lower than 7 mSv. However, there are inhabited areas of the world, so called enhanced natural radiation areas (ENRA), where the average annual exposure doses from natural sources are above 10 mSv (UNSCEAR, 2000, 2008a). Since natural radiation is usually a far more significant contributor to the annual exposure dose than man-made radiation, the dose from naturally occurring radiation represents a baseline value for an annual radiation dose, to which other exposure doses are added under specific circumstances (e.g., due to medical reasons, nuclear accidental situations).

1.2 The thorium rich Fen Complex in Norway

The Fen Complex is located in the Telemark County in southeastern Norway. It is an intrusive complex of alkaline and carbonatite rocks. The central circular intrusive of about 5 kilometers diameter represents a cross-section of the feeder pipe of a Precambrian gneiss volcano. This volcano was active 580 million years ago (Barth and Ramberg, 1966). The area was first described by Brøgger (1921), and since then it has acquired fame in the geological society. Brøgger (1921), who discovered that carbonates in the Fen Complex had a magmatic origin, introduced the term "carbonatite" for carbonate rocks of an apparent magmatic origin and named different specific rocks after localities in the Fen area (Heincke *et al.*, 2008). So far, different aspects of the geology of the Fen, including alkali-metasomatism of the country rock ("fénitisation"), have been studied in great detail by many authors (e.g., Andersen 1984; Bergstøl and Svinndal, 1960; Dahlgren, 1983; Dahlgren, 1987; Sæther, 1957). The fact that the whole Complex was an active volcano implies variable depths for different ores, and the presence of heterogeneous rocks, with mixtures of many minerals and a variety of elements (Eriksen, 2012). Among the most abundant rock types of the Fen Complex area are søvite, rauhaugite, rødbergite, damtjernite and fenite (Figure 1).

Thorium ore resources in the Fen Complex have been estimated to be among the world's largest, with an estimated volume of 170 000 tonnes, which are referred to as the reserve, and 150 000 tonnes, referred to as the reserve base (US GS, 2007). However, knowledge of the geological environments and ²³²Th-enriched minerals in Norway is mainly based on the geological surveys conducted during two periods, after Second World War up to the 1960s, and from 1975 to 1985 (Thorium

Committee, 2008). In these surveys, Th was not a primary exploration target and resources were estimated in relation to U and rare earth element (REE) resources.

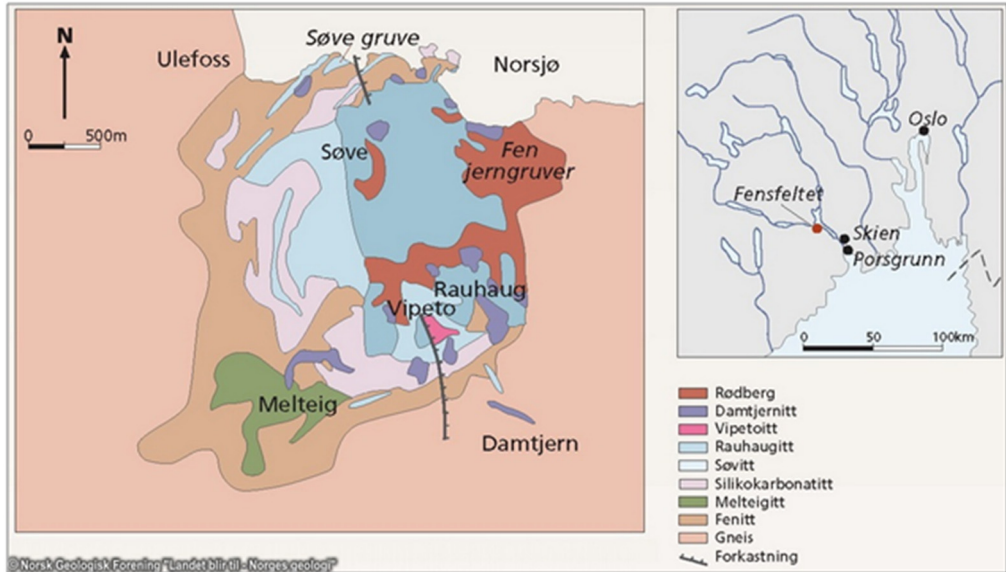


Figure 1. Geological cart of the Fen Complex in Telemark County (Ramberg *et al.*, 2007).

Thus, the uncertainty about the exact quantities of ^{232}Th has been highlighted (Thorium Committee, 2008). According to Berg *et al.* (2012), ^{232}Th resources in the Fen Complex could even reach 675 000 tonnes. As such, these resources could represent potential energy up to 120 times larger than the oil extracted to date by Norway, plus that of the remaining oil reserves (Berg *et al.*, 2012). A need for further investigations to obtain accurate data on the volume size of ^{232}Th resources has been highlighted (Lindahl, 2007).

With respect to potential ^{232}Th extraction, rødbergite is the most interesting rock type, containing the highest concentrations of ^{232}Th (up to 0.4 wt %). However, ^{232}Th has been found to be associated in small mineral grains (Figure 2). Separation of very fine-grained minerals and obtaining satisfactory enrichment are currently the main challenges for industrial exploitation since ore must be milled to fines $<50\ \mu\text{m}$ to extract ^{232}Th -bearing minerals (Berg *et al.*, 2012; Dahlgren, 2012; Eriksen, 2012). Advanced methods for mineral separation should be adapted and utilized for these specific deposits (Thorium Committee, 2008).

Apart from ^{232}Th , the Fen Complex has for a long time been of interest because of other mining possibilities. The iron (Fe) mines in the Fen area ("Fen jerngruver" in Norwegian), located in the eastern parts of the Complex, are rich in rødbergite rock

and have been operating from 1657 to 1927. Reliable data on the mined quantities and produced Fe are not available. Open pits and parts of underground mining tunnels from the Fe production period, as well as the waste area on the slope of the Lake Nordsjø, are still visible.

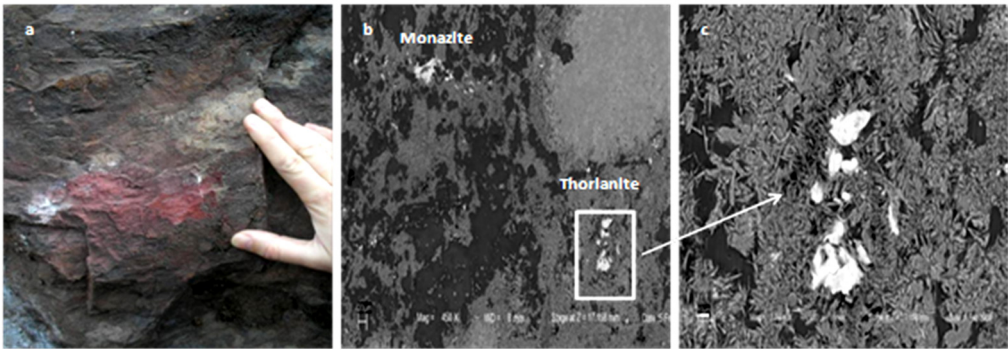


Figure 2. a) Photo image of rødsbergite (J.M.Popic) b) EM - inclusions of monazite and thorianite seen on scale 20 μm (Dahlgren, 2012) c) thorianite better seen on scale 10 μm (Berg *et al.*, 2012).

At the mining site Søvø ("Søvø gruver" in Norwegian) which was in operation from 1953 to 1965, niobium (Nb) was extracted. The main Nb-bearing mineral, pyrochlore, was identified already in 1918 in the søvite rocks. The average grade was about 0.35-0.4 % Nb_2O_5 . During the production period, Nb was extracted from the mineral søvite on a commercial basis. Waste material from the production of ferro-niobium was dumped as an aluminum-rich slag at a small hill nearby. The slag included about 570 tonnes which were covered and sealed with marine clays afterwards. In a remediation action conducted after the mine decommission, site Søvø was covered with sand layer. Recent measurements have shown enhanced gamma radiation dose rates, and elevated Th and U concentrations, due to disturbance of protective sand layer and mixing with contaminated soil (IFE, 2006; NGI-UMB, 2010). Currently, a mechanical engineering firm is located at this former mining site, while the main hazard areas, including a sludge disposal site, wash house and slag heap (NGI-UMB, 2010) are freely accessible.

In addition to Fe and Nb mining, the investigation of REE in the Fen Complex has been in progress in last several decades and has actually revived interest recently. The interest in studying the relationship between geological formations and natural radiation in the area began in 1970-80s (Dahlgren, 1983; Stranden, 1982; Stranden and Strand, 1986; Svinndal, 1973). Dahlgren (1983) undertook the first gamma ray mapping of the terrain and demonstrated high terrestrial dose rates of gamma radiation over the whole area of the Fen Complex. Since health issues associated with

enhanced natural radiation have been of substantial interest for the Fen population, many studies have been directed to risk estimation and radiological protection of humans. The radiological and epidemiological impacts of the Fen mining activities, evaluated by Stranden (1982), showed that miners received an annual dose equivalent of 150 mSv, which is much higher than the occupational dose limit of 20 mSv/y for radiation workers. Furthermore, the indoor ^{222}Rn concentrations in Fen area dwellings, the magnitude of indoor gamma radiation and their dependence on the geological formations of the terrain were also measured and discussed (Smethurst *et al.*, 2006; Solli *et al.*, 1985; Stranden, 1982; Sundal and Strand, 2004). The annual radiation dose to which the population in the Fen area was exposed was estimated to be higher (14 mSv) than the average radiation dose in Norway (2.9 mSv), due to elevated gamma radiation and elevated concentrations of ^{222}Rn daughters (Stranden, 1982; Stranden, 1984; Stranden and Strand, 1986; Sundal and Strand, 2004). The most recent airborne gamma spectrometry mapping that relates indoor ^{222}Rn concentrations to geological parameters (Heincke *et al.*, 2008), has revealed elevated ^{232}Th and moderate ^{238}U concentrations in carbonatite rocks. Positive correlations between local ^{238}U concentrations and indoor ^{222}Rn concentrations were observed.

1.3 Rationale, hypotheses and objectives of the thesis

The rationale behind the current environmental assessment project was based on several points:

- According to the Thorium Committee (2008), ^{232}Th in the Fen Complex in Norway has been considered as a very promising resource, which provides the opportunity for nuclear energy production in the future. However, the available ^{232}Th resources have not been properly characterized; the quoted US GS (2007) weight estimates of the ^{232}Th resources (170 000 tonnes), dating from the 1950s-1960s, are unreliable (as explained in the previous section). Need for further investigation of ^{232}Th in the Fen Complex bedrock has been highlighted. In addition, the Fen carbonatite is one of the very few REE mineral resources in Europe, making it a potentially strategically important deposit (Østergaard, 2011). With respect to both ^{232}Th and REE, the possibilities for mining and exploitation of the Fen Complex bedrock are open for future generations. On the other side, mining of Fe and Nb ores was conducted at several locations in the past, and questions related to possible environmental risk from legacy NORM have already been raised. Thus, considering past and possible future mining and extraction activities, care must be taken to address all environmental issues associated with

radionuclides and trace elements. The present assessment should contribute to understanding of the current radiological exposure situation. Also, it will be valuable in the future if exploitation of Fen resources resumes.

- Investigations in the Fen Complex area have focused on the unique geological formation (Brøgger, 1921; Dahlgren, 1983; Landreth, 1979) and on increased human health risk due to terrestrial gamma radiation and ^{222}Rn inhalation (Smethurst *et al.*, 2006; Solli *et al.*, 1985; Stranden and Strand, 1986; Sundal and Strand, 2004). However, neither investigation of contaminants in different environments compartments nor impact assessment for species other than humans has been performed.
- As of 1 January 2011, a new amendment was enforced whereby radioactive waste and radioactive pollution were integrated in the Norwegian Pollution Control Act from 1981. This means that radioactive waste and radioactive pollution are now regulated under the same legal framework as all other pollutants and hazardous wastes. The amendment established two sets of criteria defining radioactive waste: lower value radioactive waste, and higher value waste - the most common case when waste must be disposed of in a final repository (Liland *et al.*, 2012). According to this, the legacy NORM sites in the Fen Complex need to be investigated in order to determine whether screening levels have been exceeded and intervention is required.

Based on literature data on NORM in the Fen Complex area, the main hypotheses of the current work were as follows:

- H1.** Due to the presence of ^{232}Th -containing minerals in the Fen Complex bedrock, ^{232}Th and associated radionuclides and trace elements are expected to represent an environmental problem in the area.
- H2.** The mobilization of radionuclides and trace elements from surface rock minerals is expected.
- H3.** Transfer of mobile and bioavailable radionuclide and trace element species from soil to biota is expected.
- H4.** The mobility and bioavailability of elements should be enhanced at legacy NORM sites in comparison to undisturbed ^{232}Th -rich sites.
- H5.** Enhanced radiation doses to biota and humans, due to natural radiation in the bedrock, are expected.

The overall objective of this thesis was to assess possible environmental contamination with radionuclides and associated trace elements in the Fen Complex area, and their impacts on biota and humans.

The assessment was organized in tiers common for environmental risk assessments (ERA). The approach was extended to cover the exposure of humans to outdoor radiation (Figure 3). Thus, the final approach represents an integrated radiological impact assessment.

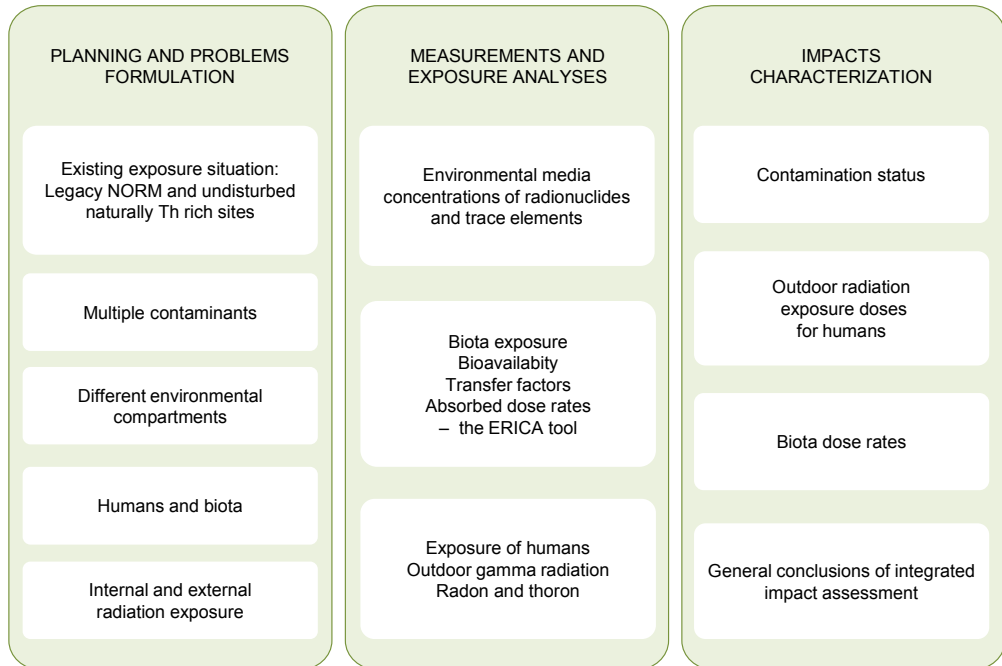


Figure 3. Assessment tiers in the present study.

According to the working tiers, the following sub-objectives were set to meet the main objective:

- 1) Problem identification and characterization of the contaminants in different environmental compartments (Papers I and II)
- 2) Estimation of the transfer and possible bioaccumulation of radionuclides and trace elements in biota (Papers III and IV)
- 3) Assessment of biota exposure, calculation of dose rates (Papers III and IV)
- 4) Identification of the main pathways of humans outdoor radiation exposure and estimation of the radiation doses related to outdoor exposure (Paper V)

2 Thesis background

2.1 Naturally occurring radioactive material (NORM)

Radioactive materials can be divided into two broad categories: naturally occurring radioactive materials (NORM) and man-made materials. Several similar definitions of NORM exist. According to US EPA (2006), NORM is defined as a “material which may contain any of the primordial radionuclides or radioactive elements as they occur in nature, such as radium (^{226}Ra), ^{238}U , ^{232}Th and their radioactive progenies that are undisturbed as a result of human activities”. Human manipulation of NORM for economic ends, such as mining, ore processing, fossil fuel extraction and commercial aviation, leads to what is known as “Technologically Enhanced Naturally Occurring Radioactive Materials” or TENORM (US EPA, 2000; Vearrier *et al.*, 2009). In recent years, the acronym TENORM is often replaced with “industrial NORM” (which refers to enhanced radiation as a consequence of different industrial activities), “legacy NORM” (which refers to enhanced radiation as a consequence of human activities from the past) or simply with “NORM waste”. Still, no clear consensus on the use of these terms exists; similar exposure situations have sometimes been named differently what can even be confusing.

Essentially, exposure to undisturbed NORM is responsible for most of an average person’s annual radiation dose, and therefore, is usually not considered to have any particular health or safety significance. However, as explained above, primordial radionuclides present in the parent materials can become concentrated in wastes during different beneficiation processes. This results in radionuclide concentrations in NORM wastes (e.g., industrial NORM, legacy NORM) that are often orders of magnitude higher than in the parent materials (US EPA 2000). Over time, as potential NORM hazards have been identified, they have increasingly become subject to monitoring and regulation, although legal liability related to NORM waste is loosely defined in most countries (Kosako and Sugiura, 2003). In 1996, the International Agency on Atomic Energy (IAEA) published the Basic Safety Standards (BSS), and although the NORM issue was not considered directly, the exemption levels for each radionuclide, including naturally occurring, were given (IAEA, 1996). Furthermore, during the 1990s and 2000s, international interest in NORM problems increased and several international subgroups and forums were organized (meetings in Australia, Brazil, Malaysia, Vietnam, China, Thailand, etc.). Current recommendations of the International Commission on Radiological Protection (ICRP, 2007) state that control

of the dose of radiation, no matter what the source, is the central aspect of protection of the individual.

Although both ^{232}Th and ^{238}U are alpha emitters and are characterized as radiotoxic, very low doses are actually received from pure ^{232}Th and ^{238}U due to their long half-lives ($T_{1/2}$). Key dose contributors, in the case of NORM waste, are associated with ^{238}U and ^{232}Th progenies, especially ^{226}Ra , polonium (Po) and lead (Pb) isotopes produced from ^{222}Rn . In terrestrial and aquatic ecosystems, these radionuclides can be transferred from the site of origin by air emissions, leaching, and by dissolving in run-off waters, from the soil and into plants, animals and, finally, through the food chain to man (Apps *et al.*, 1988; Bernhard *et al.*, 1996; Stojanovic *et al.*, 2009; Vandenhove *et al.*, 2006; Vandenhove and Van Hees, 2007). It has also been shown that certain fractions of ^{238}U , ^{232}Th and their progenies can be present as radioactive particles (Salbu *et al.*, 2004). Consequently, the transfer to man could occur via inhalation of particles after resuspension and via dietary intake. The exhalation of Rn isotopes from the ground or surface of building materials and subsequent human inhalation is another significant radiation pathway from NORM waste to humans. Thus, improper disposal of NORM waste and/or re-use of NORM contaminated materials can lead to increased exposure of humans, plants and animals to radiation, in a variety of ways. In many situations worldwide, residents, authorities and industries need additional information, assistance and guidance in management and protection from various types of NORM waste.

As described in section 1, the Fen Complex contains large quantities of ^{232}Th in rocks and soil in an undisturbed natural state (NORM). On the other side, decommissioned mining and disposal sites represent legacy NORM waste. Although the acronyms NORM and TENORM were used in Papers I, III and V, in the present thesis a more precise description "undisturbed ^{232}Th -rich sites" has been used for NORM in the Fen area, and "legacy NORM" for waste from the former mining sites. The ICRP (2007) recommendations describe three types of exposure situations: planned, emergency and existing. According to the ICRP (2007) definition ("existing exposure situations are exposure situations that already exist when a decision on control has to be taken"), decommissioned Nb and Fe mining sites in the Fen Complex should be regarded as existing exposure situations. Furthermore, with respect to the amendment of the Norwegian Pollution Control Act (2010), appropriate legislative requirements should be applied in situations where NORM has been enhanced due to human activities as of 1st January 2011. A tiered approach has been developed; a set of activity levels that determine the necessary activities and possible handling has been provided (Liland *et al.*, 2012).

2.1.1 Thorium (Th)

The element thorium (Th) was discovered in 1828 by the Swedish chemist Jons Jakob Berzelius (1779-1849) when he examined rock samples from Norway. He named it after Thor, the Nordic God of thunder. Thorium is a naturally-occurring, silvery white, slightly radioactive metal, produced together with U and other elements heavier than Fe in one or more supernovae over 6 billion years ago (World Nuclear Association, 2006). Naturally occurring Th consists 100 % of the isotope ^{232}Th by weight, a parent nuclide of the Th decay series (Figure 4). The average concentration of ^{232}Th in the Earth's crust is 10 mg/kg, which is about two to four times higher than the concentration of naturally occurring U (UNSCEAR, 2000, 2008a). However, ^{232}Th has a much lower specific activity than ^{238}U due to the difference in their $T_{1/2}$; the activity concentration (Bq/kg) of the two elements is about the same (Kathren, 1998).

As ^{232}Th undergoes radioactive decay, it emits an alpha particle, with accompanying gamma radiation, and forms ^{228}Ra . The decay process continues, radiation (alpha and beta minus particles and gamma rays) releases, and the formation of new radionuclides continues until stable ^{208}Pb is formed (the decay chain consists of 12 different radionuclides) (Figure 4). The half-life of ^{232}Th ($T_{1/2}$) is about 14 billion years. In the ^{232}Th decay chain, ^{228}Ra and ^{228}Th have $T_{1/2} = 5.8$ and $T_{1/2} = 1.9$ years, respectively, while the $T_{1/2}$ of other progenies are short (Choppin *et al.*, 2002).

The major physico-chemical form of ^{232}Th in nature is ion Th^{+4} , i.e., tetravalent (IV) oxidation state with very low solubility (US EPA, 1999). Important factors affecting sorption and dissolution of ^{232}Th are the presence of Fe and manganese (Mn) oxides, organic compounds, ligands such as CO_3^{2-} and humics, Eh and pH (Guo *et al.*, 2008; Rachkova *et al.*, 2010; Syed, 1998; Vandenhove *et al.*, 2009a). In fact, ^{232}Th is considered to be one of the least mobile elements in continental weathering (Braun *et al.*, 1998), and its mobility is expected to increase only in extremely acid environments (Piñeiro *et al.*, 2002).

Thorium occurs in several minerals, such as thorite ((Th, U)SiO₄), thorianite (ThO₂), allanite ((Ce, Ca, La, Y, Th)₂(Al, Fe²⁺, Fe³⁺)₃(SiO₄)₃(OH)), but the most common is the rare earth-thorium-phosphate mineral, monazite ((Ce, La, Nd, Th, Y)PO₄), which contains up to about 1-15 % thorium oxide (ThO₂) (Anthony *et al.*, 2000; Choppin *et al.*, 2002). With respect to ^{232}Th abundance in Norway, three main regions are the Fen Complex in Telemark County, the Permian Oslo Province and the Southeast coast region. A series of ^{232}Th bearing minerals has been identified at these sites. The Fen

Complex is considered to be a very promising resource, with ^{232}Th amounting to about 0.1 – 0.4 wt % in rødbergite and rauhaugite rocks (Thorium Committee, 2008).

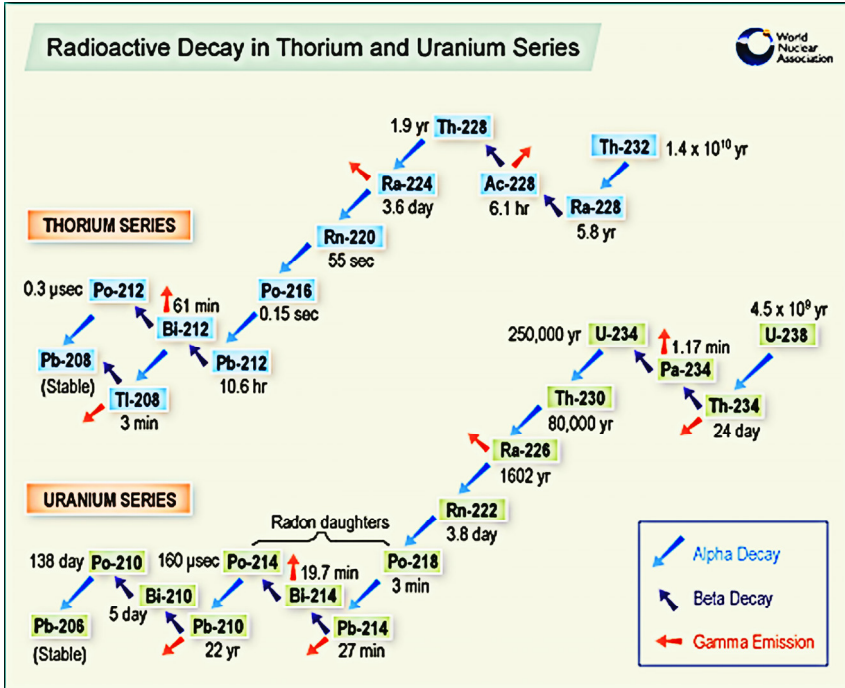


Figure 4. ^{232}Th and ^{238}U radioactive decay chains (<http://world-nuclear.org/>).

Beside the erosion and weathering of the minerals in rocks and soils, windblown dust and volcanic eruptions can also release ^{232}Th in the environment (Andersson *et al.*, 1995; Gill and Condomines 1992; Thomas *et al.*, 2002). A number of worldwide locations have been demonstrated to have elevated natural radiation due to high ^{232}Th contents in soil or bedrock. These include coastal sands of the States of Espiritu Santo and Rio de Janeiro in Brazil and the State of Kerala in India, as well as regions in the Guangdong Province in China (Kathren, 1998). Mining of naturally occurring ^{232}Th , coal burning and commercial processing, if not properly controlled, could release ^{232}Th and its progenies into the environment and could therefore be man-made sources of contamination and enhanced radiation levels (Dowdall *et al.*, 2004; Hutchinson and Toussaint, 1998).

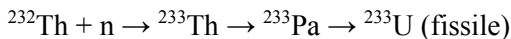
Thorium as nuclear energy source

According to the Thorium Committee (2008), there is a prevailing belief that the current model for the world’s energy policy is not sustainable. Emission of greenhouse gasses and their impact on climate, as well as increasing energy demands

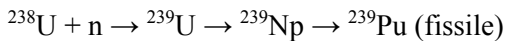
worldwide and the potential for secure energy supplies are among the main reasons for this view. The outlook for nuclear energy power has generally improved worldwide, with a progressive improvement in the operating performance of the existing reactors which ensure economic competitiveness of nuclear electricity in world markets. In June 2009, some of 436 nuclear power plants were in operation worldwide, generating about 16 % of global electricity, while an additional 45 power reactors were under construction (Adamantiades and Kessides, 2009).

Over the last 50 years, there has been increasing interest in the utilization of ^{232}Th as nuclear fuel. Currently, the highest research activity on ^{232}Th as a nuclear energy source is in India, where the utilization of ^{232}Th for large scale energy production is the major goal of the nuclear power program (Thorium Committee, 2008). All of the mined ^{232}Th is potentially useable in a reactor, compared with the 0.7 % of natural U (^{235}U); therefore, about 40 times more energy per unit mass is theoretically available from ^{232}Th . The isotope ^{232}Th is not fissile, which means it cannot undergo fission if bombarded by neutrons and, as such, it is not directly usable in a thermal neutron reactor. On the other hand, it is fertile, which means that in combination with fissile ^{235}U or plutonium (^{239}Pu), upon absorption of a neutron, ^{232}Th will transmute to a new fissile material – ^{233}U . In this respect, it is similar to transmutation of ^{238}U to ^{239}Pu in the common U fuel cycle.

The Thorium-Uranium fuel cycle:



is analogous to the Uranium-Plutonium fuel cycle:



The potential advantages of a Th-based fuel cycle over an U-based fuel cycle are the greater natural abundance of Th, no need for isotope separation (since mined Th consists of a single ^{232}Th isotope), superior thermo-physical and nuclear properties of ThO_2 as compared to UO_2 , a longer fuel cycle and higher burn up, better resistance to the proliferation of nuclear weapons (due to formation of ^{233}U that emits strong gamma radiation) and little Pu or other transuranics products (Hargraves and Moir, 2011; IAEA, 2005). Still, there are several challenges to the use of Th-based fuel. Preparation of Th fuel is somewhat more complex (e.g., high sintering temperature, corrosion in reprocessing plants) and more expensive than the preparation of U fuel, as it requires a fissile material (^{233}U , ^{235}U , ^{239}Pu) so that a chain reaction (and thus supply of surplus neutrons) can be maintained. Further, irradiated Th or Th-based fuel contain significant amounts of ^{233}U ($T_{1/2} = 73.64$ years) associated with strong gamma

emitting daughters ^{212}Bi and ^{208}Tl . As a result, storage of spent Th-based fuel, as well as necessary remote and automated reprocessing and re-fabrication in shielded cells, increase cost in fuel cycle activities (IAEA, 2005). The feasibility of Th utilization in high temperature gas cooled reactors (HTGR), light water reactors (LWR), pressurized heavy water reactors (PHWR), liquid metal cooled fast breeder reactors (LMFBR) and molten salt breeder reactors (MSBR) was demonstrated. However, thus far Th fuels have not been introduced commercially because the estimated U resources are still sufficient (IAEA, 2005).

From the radiological protection standpoint, to assess doses associated with Th nuclear fuel, the front end (mining, milling, extraction, fuel fabrication) and the back end (operation, waste storage, reprocessing and waste disposal) of the Th cycle need to be considered. Radiation dose contributors from mining, milling, extraction and fuel fabrication are only naturally occurring radionuclides. Although, the parent radionuclides ^{232}Th and ^{238}U are alpha emitters, they are insignificant contributors due to the very long $T_{1/2}$. The major contributors to increased radiation levels from the ^{232}Th chain are the alpha emitting progenies such as ^{228}Th , ^{224}Ra and short-lived isotopes ^{216}Po , ^{212}Po produced from ^{220}Rn (decay chain in Figure 4). Radium is water soluble, mobile and easily transferred from rocks, soil or sediments into the environment. It is often concentrated in ground water. The inert gas ^{220}Rn does not interact with matter, however, it is easily released from rock, soil, water, and building materials. It disintegrates to particulate, reactive ^{216}Po which can then be further mobilized. In this way, ^{232}Th progenies are displaced from the original deposits and contribute to the exposure dose of humans (Thorium Committee, 2008). However, in contrast to ^{222}Rn and its progenies, ^{220}Rn and its progenies are less significant in terms of dose contribution. Due to a short $T_{1/2}$ of ^{220}Rn ($T_{1/2} = 55.6$ sec), it disintegrates to particulate ^{216}Po before reaching some distance. Thus, it can contribute to the exposure dose only if a person is directly exposed near the source. In contrast, the potential of ^{222}Rn ($T_{1/2} = 3.8$ days) migration before decay is much bigger, and it represents a significant problem at many worldwide locations (UNSCEAR, 2006). Furthermore, the $T_{1/2}$ of the ^{220}Rn progenies (Figure 4) are short (shorter than ^{222}Rn progenies), and consequently their concentrations in the environment and contributions to the exposure dose are low. Due to its short $T_{1/2}$, only traces of high energy gamma emitting thallium (^{208}Tl) will be present per kg of ^{232}Th . Therefore, the impact of ^{232}Th progenies is much lower and easier to handle than the impact of ^{238}U progenies. The doses received at close distance to the source, during Th mining and milling operations, are comparable to the doses received during U mining, while doses at larger distances (associated with crushed rocks and at tailings) are lower than

at U tailings. Radiation protection during the manufacturing of Th-based fuel should be same as for U since they have similar thermodynamic and thermo-physical properties (Thorium Committee, 2008).

With respect to the back-end of the Th cycle, doses to man and the environment are dependent on the composition of the used fuel, the type of reactor system and operations. Thorium-based fuels produce much less Pu and associated minor actinides than U based fuel. In general, the key dose contributions of concern are the generation of protactinium (^{231}Pa), ^{229}Th and ^{233}U (Thorium Committee, 2008).

2.1.2 Uranium (U)

The element uranium (U) was discovered in 1789 by the German chemist Martin Klaproth (1743-1817) in the mineral pitchblende. Klaproth named it after the planet Uranus. Uranium is a naturally-occurring, silvery white, metallic element in the actinide series, produced (as given for Th) in one or more supernovae over 6 billion years ago (World Nuclear Association, 2006). The average concentration of ^{238}U in the Earth's crust is 3-4 mg/kg (UNSCEAR, 2000, 2008a). Under natural conditions, U is found in the environment as a mixture of three isotopes: ^{238}U , ^{235}U and ^{234}U , accounting for 99.3 %, 0.72 % and 0.0057 %, respectively.

The radioactive element U has long half-life ($T_{1/2} = 4.5 \cdot 10^9$ years), which is equal to the age of the Earth. Over time, the ^{238}U decays (Figure 4) into ^{234}U ($T_{1/2} = 248\,000$ years) which decays into ^{230}Th ($T_{1/2} = 80\,000$ years) which further decays into ^{226}Ra ($T_{1/2} = 1620$ years). Under non-disturbed conditions in the soil, ^{226}Ra exhibits the same level of radioactivity as ^{238}U from which it was originally derived due to secular equilibrium. However, when radioactive ^{226}Ra decays, it produces the noble gas ^{222}Rn ($T_{1/2} = 3.8$ days). When produced in the soil close to the air-soil interface, ^{222}Rn can escape to the atmosphere before it decays into the next radioactive element (a non-volatile metal). After several days of residence time in the atmosphere, the ^{222}Rn naturally decays to ^{218}Po ($T_{1/2} = 3$ min), a metallic radionuclide which falls to the earth with dust and rain. A number of subsequent radioactive decays occur over a period of minutes and ^{210}Pb ($T_{1/2} = 22.3$ years) is produced. In two consecutive beta decays, another significant radiation exposure dose contributor ^{210}Po is produced from ^{210}Pb . Both ^{210}Pb and ^{210}Po are particle reactive and tend to end up in the soil and sediments, however, re-mobilization under certain environmental conditions is possible (Persson and Holm, 2011).

Uranium has five valence states (II-VI) (Hirose, 2012). Under most oxidizing conditions, U (VI) is more stable than U (IV) and U (V) and forms the uranyl ion

(UO_2^{2+}) that, depending on the pH and present ligands, forms water-soluble complexes (Barnett *et al.*, 2000; Langmuir, 1978). It was previously reported that ^{238}U is more mobile than ^{232}Th in well-aerated superficial environments (Braun *et al.*, 1998; Kabata-Pendias, 2010). Uranium concentrations are often higher in phosphate-rich soils and in soils where phosphorous fertilizers have been used over a long period. However, concentrations often do not exceed significantly the normal range for uncontaminated soil (Stojanovic *et al.*, 2006). Plants can absorb ^{238}U through their roots and store it in various parts (Raju and Raju, 2000; Unak *et al.*, 2007; Vandenhove *et al.*, 2007a, b).

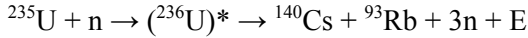
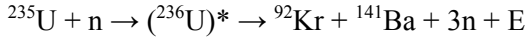
Uranium occurs in a number of different igneous, hydrothermal and sedimentary geological environments. In fact, U deposits worldwide can be grouped into 14 major categories of deposit types, based on the geological setting of the deposits (World Nuclear Association, 2010). Series of U bearing minerals were identified in different rock types (Langmuir, 1978). Sometimes, consideration of the mineral's solubility can be more valuable for the assessment of U in the environment than determination of mobilization parameters, such as the distribution coefficient (K_d) (Elless and Lee, 1998). Common U (IV) minerals are uraninite (UO_2) and coffinite ($\text{U}(\text{SiO}_4)_{1-x}(\text{OH})_{4x}$); common U (VI) minerals are carnotite ($\text{K}_2(\text{UO}_2)_2(\text{V}_2\text{O}_8) \cdot 1-3\text{H}_2\text{O}$), autunite ($\text{Ca}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot 10-12\text{H}_2\text{O}$), U phosphate minerals ($(\text{UO}_2)_3(\text{PO}_4)_2 \cdot 4\text{H}_2\text{O}$), rutherfordine ($\text{U}^{6+}\text{O}_2(\text{CO}_3)$) and uranophane ($\text{Ca}(\text{UO}_2)_2(\text{SiO}_3\text{OH})_2 \cdot 5\text{H}_2\text{O}$) (Anthony *et al.*, 2000; Langmuir, 1978).

Uranium is wide-spread throughout the environment, in rocks, soil and water. In air, it exists as dust that will deposit on surface water, on plants or on soils through settling or rainfall, and then sink to the sediment in water or to the lower soil layers where it can mix with the U that is already present (Jia and Torri 2007; Taboada *et al.*, 2006). Human activities, such as U ore mining, milling and processing, U enrichment and production of nuclear fuel, production of phosphate fertilizers and military use of depleted U, add metal U and its compounds to the environment (US EPA, 2013). The worldwide production of U in 2010 amounted to 53 663 tonnes; the largest producers of U ore are Kazakhstan, Canada, Australia, Namibia, Niger and Russia (IAEA, 2011).

Uranium as a nuclear energy source

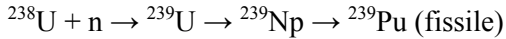
All commercial nuclear power reactors in use today are reliant on the U-Pu fuel cycle, in which ^{235}U is the principle fissile nuclide (present commonly at 3-5 wt %) which provides the fission neutrons needed to maintain criticality and adequate power output.

Examples of fission of ^{235}U atom:



Whereas the ^{235}U nucleus is fissile, that of ^{238}U is fertile (as ^{232}Th). In a nuclear reactor, ^{238}U can capture neutrons and transmute to fissile ^{239}Pu . In fact, about one third of the fuel's energy yield comes from burning ^{239}Pu .

The Uranium-Plutonium fuel cycle:



Processes related to mining, refining, purifying, using, and ultimately disposing of nuclear fuel together make up the nuclear fuel cycle. The front end of the U fuel cycle is comprised of U mining, milling, conversion, enrichment and fuel fabrication. The back end of the U fuel cycle is comprised of reprocessing, reuse and disposal of spent fuel (IAEA, 2011).

Radioactive wastes occur at all stages of the nuclear fuel cycle. Waste produced from the front end of the U nuclear fuel cycle is alpha emitting waste. The parent radionuclide ^{238}U is slightly radioactive (due to a long $T_{1/2}$), and is not of particular concern (as ^{232}Th in the ^{232}Th fuel case). On the other side, the daughters ^{226}Ra , ^{222}Rn and further decay products, such as ^{210}Pb and ^{210}Po , are more significant with respect to exposure doses to man and the environment due to their elevated mobility in the environment (described in sections 2.1.1 and 2.1.3). Radium-226 ($T_{1/2} = 1602$ years) is about one million times more radioactive than ^{238}U , and its inhalation and ingestion are the primary exposure pathways of concern. Unlike the ^{232}Th progeny ^{220}Rn which is of little concern due to its short $T_{1/2}$, ^{222}Rn ($T_{1/2} = 3.8$ days), its progenies (e.g., ^{214}Po , ^{218}Po and ^{210}Po , ^{210}Pb) are internationally recognized as hazards and their occurrence and characteristics have been documented (ICRP, 2007; UNSCEAR 2000, 2006). To prevent risk from inhalation of the listed radionuclides, measures such as dust control, gas dilution through ventilation should be applied at mining and milling sites. The tailings can contain up to 70 % of the initial radioactivity, and measures such as covering with sand layers and re-vegetation, are usually conducted with considerable care to ensure their long term stability (World Nuclear Association, 2013). Processing of the U oxide concentrate into a useable fuel has no effect on the levels of radioactivity and does not produce significant waste.

Wastes from the back end of the fuel cycle contain significant quantities of highly radioactive fission products of ^{235}U . Spent fuel contains U (about 96 %), Pu (about 1 %) and a high level of radioactive waste (about 3 %). Uranium and Pu can be reused

after reprocessing, while the high activity waste is vitrified or converted into a glass to be finally disposed of in a high level waste disposal facility (IAEA, 2011).

2.1.3 Radon (Rn) and Rn daughters

The element radon (Rn) was discovered in 1900 by the German physicist Frederick E. Dorn (1848-1916) who called it "radium emanation", but actually scientists Marie Curie (1867-1934) and Ernest Rutherford (1871-1937) should also share the honor for the discovery. Radon is a colorless, odorless and tasteless noble gas, with three naturally occurring isotopes: ^{219}Rn , ^{220}Rn and ^{222}Rn . The isotope that arises from the ^{232}Th decay chain (^{220}Rn) is historically known as thoron. The isotope that arises from the ^{238}U chain (^{222}Rn) is commonly identified as radon (which sometimes leads to confusion). The isotope ^{222}Rn is the one of most concern regarding the radiation exposure dose in the human population. Thoron and radon (^{220}Rn and ^{222}Rn) are immediate progenies of ^{224}Ra and ^{226}Ra , respectively. They both diffuse from the earth into the atmosphere, undergo decay with different half-lives ($T_{1/2} = 55.6$ sec) and ($T_{1/2} = 3.82$ days), respectively, producing a series of short-lived radionuclides (UNSCEAR, 2000, 2006). Electrically charged progenies attach themselves to small dust particles. These can be inhaled and deposited along the walls of airways of the bronchial tree in the lungs. In fact, the inhalation of progenies of ^{220}Rn (^{212}Pb , $^{212}\text{Bi}/^{212}\text{Po}$) and ^{222}Rn (^{218}Po , ^{214}Pb , $^{210}\text{Bi}/^{210}\text{Po}$) contribute the most to human radiation exposure doses from natural radiation (Porstendörfer, 2001; UNSCEAR, 2000, 2006). The doses are mostly produced by alpha particles emitted by these progenies, although some beta and gamma radiation are emitted too.

According to UNSCEAR (2000, 2006), the outdoor concentrations of $^{220,222}\text{Rn}$ isotopes are on average 10 Bq/m^3 (for each species); however, a wider range, from 1 to more than 100 Bq/m^3 , has been observed worldwide. Air concentrations are not only affected by soil or bedrock concentration of Rn isotopes, but by atmospheric conditions, ground cover, altitude, seasonal and diurnal changes, soil moisture, porosity and grain size, temperature and atmospheric pressure (Kathren, 1998). Data on outdoor ^{220}Rn concentrations are relatively sparse in comparison to ^{222}Rn . In published literature (Doi and Kobayashi, 1994; Nishikawa *et al.*, 1994; Steinhäusler, 1996), the atmospheric ^{220}Rn variability was explained by a significant vertical atmospheric gradient caused by the short ^{220}Rn half live ($T_{1/2} = 55.6$ s). Generally, the outdoor ^{220}Rn concentrations are usually not considered as important and the associated doses are not taken into account (Kávási *et al.*, 2010).

As for outdoor ^{220}Rn , the data on indoor ^{220}Rn are also limited, although it has recently been reported that at some locations, doses from ^{220}Rn and its short-lived

decay products are comparable or even larger than from ^{222}Rn and its decay products (UNSCEAR, 2006). In addition, analyses of compiled data on ^{222}Rn dosimetry have shown possible overlap of ^{220}Rn and ^{222}Rn in indoor measurement studies (Akiba *et al.*, 2010), and ^{220}Rn can thus serve as a source of error in residential studies where two isotopes cannot be clearly distinguished.

In contrast to ^{220}Rn , much literature data has documented the worldwide indoor levels of ^{222}Rn . In the UNSCEAR Report (2000, 2006), values of 40 and 30 Bq/m³ have been reported as arithmetic and geometric means, respectively. The concentration of indoor ^{222}Rn varies according to the geological location and room ventilation of dwellings. In areas with elevated ^{222}Rn levels in the air, the principal entry mechanism into dwellings is through a pressure-driven flow of soil gas through cracks in the floor. Special attention has been devoted to indoor ^{222}Rn levels, due to its important contribution to the dose of exposure in humans (ICRP, 2007). The UNSCEAR reports (2000, 2006) has provided an annual per capita dose, estimated at 1.25 mSv from exposure to $^{220,222}\text{Rn}$, i.e., approximately one half of the estimated exposure dose from all natural radiation sources (2.4 mSv).

The evaluation of the impact of ^{222}Rn inhalation on lung cancer has been in focus worldwide for a long time, primarily in cases with high ^{222}Rn exposure in underground mines, as well as in assessing different environmental ^{222}Rn exposures (UNSCEAR, 2000). The dose-response relationship of ^{222}Rn and lung cancer has been found to be linear and without a threshold (Darby *et al.*, 2005). Cigarette smoking significantly increases lung exposure doses from Pb and Po, and it has been hypothesized that lung cancer risk in smokers is in part attributable to the additional exposure of the bronchial epithelium to alpha emitting Po (Kathren, 1998).

Norway, Sweden and Finland are among the countries with the highest average indoor ^{222}Rn concentrations. Geological conditions and the cool climate pose a considerable challenge with respect to ^{222}Rn levels. Several Norwegian counties, such as Hordaland, Hedmark, Buskerud, Østfold and Hardanger, have elevated ^{222}Rn concentrations due to the geology of the bedrock (Jensen *et al.*, 2006; Smethurst *et al.*, 2008).

Alum shale, the sedimentary rock of the Oslo Rift bedrock, is rich in U and is known to contribute to elevated ^{222}Rn exposure doses (Figure 5). The average indoor ^{222}Rn concentration in Norway is around 90 Bq/m³. It was estimated that around 250-300 people would die annually from lung cancer as a consequence of indoor ^{222}Rn exposure (NRPA, 2013; Strand *et al.*, 2001).

radiation. Gamma rays are indirectly ionizing; about two thirds of the biological damage caused by gamma rays is produced through indirect action via free radicals (Hall and Giaccia, 2006).

The world average outdoor gamma dose rate in the air is 0.059 $\mu\text{Gy/h}$, giving a relatively low annual contribution of 0.07 mSv to the total annual human exposure dose of 2.4 mSv (UNSCEAR, 2000, 2008a). Areas with markedly higher gamma dose rates associated with Th- and U-containing minerals in the soil have been described worldwide (UNSCEAR, 2000, 2008a). In these areas (such as the one investigated in the present work), short-lived gamma emitting radionuclides might be of particular concern as they could considerably contribute to the overall exposure doses in the environment.

2.1.5 *Interaction of ionizing radiation with biological tissue*

The mechanisms of interaction of ionizing radiation with biological matter are the same for humans and biota (although radiosensitivities vary considerably). The biological effects of radiation (e.g., gamma- and X-rays, charged or uncharged particles) result principally from interaction of radiation with the critical targets in cells. During direct action of radiation, ionization or excitation of the cell target is the initial process, followed by a chain of events which in the end produce biological effects (Hall and Giaccia, 2006). Alternatively, during indirect action of radiation, the radiation reacts with other atoms or molecules, particularly with water (H_2O) (since H_2O constitutes about 80 % of the cell), producing the free radicals, i.e., reactive oxygen species (ROS; $\cdot\text{O}_2^-$, $\cdot\text{O}_2^{-2}$, $\cdot\text{OH}$, OH^- , H_2O_2). When cellular production of ROS overwhelms its antioxidant capacity, damage to cellular macromolecules such as DNA ensues (Thannickal and Fanburg, 2000). The DNA molecule is considered to be the major biological target for ionizing radiation. A wide spectrum of possible DNA lesions includes oxidized and methylated bases, DNA adducts and single- and double stranded breaks (Streffer *et al.*, 2004). When DNA damage cannot be repaired by the organisms' natural repair mechanisms, mutations, chromosome rearrangements and eventually cell death (apoptosis, necrosis and mitotic death) occur in somatic, germ and fetal cells. Finally, cancer, heritable genetic, developmental, acute somatic, reproductive and teratogenic effects can develop as the consequence of radiation injury (Little, 2003). The direct action of radiation is the dominant mode for radionuclide alpha emitters, such as ^{232}Th , ^{238}U , ^{226}Ra , while indirect actions predominate in those that emit sparsely ionizing radiation, such as gamma radiation (Hall and Giaccia, 2006).

2.2 Multiple stressors – radionuclides and trace elements

With the current developments of industry and technology, environmental exposure to multiple stressors (combinations of pesticides, fire retardants, polychlorinated biphenyls, endocrine disruptors, heavy metals, trace elements and radionuclides) is common (Muir *et al.*, 2005). Examples of an ecosystem's combined exposure to radiation and other stressor (in most of the cases metal) are observed as releases from nuclear power plants, U mining and milling waste and non-nuclear NORM industry waste (Vanhoudt *et al.*, 2012).

The field of multiple stressors is highly complex. Its main characteristics are several levels of interaction between pollutants, different sensitivities of organisms of concern, multiple pathways and modes of action, multiple target sites and biological endpoints. In addition, there are only single stressor protection tools, sparse information on biological responses to exposures to mixtures, as well as a lack of internationally uniform and recognized risk assessment approaches for multiple stressors. Various contaminants can interact in additive, antagonistic or synergistic manners (Holmstrup *et al.*, 2010; Mothersill and Seymour, 2007; Mothersill and Seymour, 2009; Salbu, 2009). Recent investigations have shown that chemical stressors in the environment can act through similar mechanisms. The outcomes following mixed exposures may not be predictable by consideration of individual stressors (Mothersill and Seymour, 2009). Induced free radicals may result in damage which is reflected in "umbrella" endpoints, such as reproductive and immune system failure, genetic instability and mutation, morbidity and mortality (Salbu, 2009). The lack of knowledge about complex mixtures of pollutants is a major challenge in environmental sciences (Eggen *et al.*, 2004).

The current environmental assessment methodology is to run single-stressor toxicity tests for each of the stressors that make up the mixture, and then to combine the individual effects using a mixture model. Consequently, legislation is largely based on studies that have examined the effects caused by a single contaminant, not mixtures (Glaholt *et al.*, 2012). Radiation protection criteria have also been developed based solely on radiation study (Vanhoudt *et al.*, 2012). Brechignac and Doi (2009) highlighted the need for data on the combined effects from radiation and other stressors in order to determine the extent to which radiation should be included in a multiple pollution context. Furthermore, organisms in natural settings are usually exposed to more than one contaminant. They have to cope with environmental conditions which can interact with or alter the actions of a particular contaminant or mixture of contaminants (Holmstrup *et al.*, 2010). Studies on the cumulative stress

from different combinations of organics, heavy metals, alone and in combination with natural stressors (temperature, oxygen depletion, water pH and occurrence of ligands, weather conditions, etc.) have revealed both synergistic and antagonistic interactions between stressors (Holmstrup *et al.*, 2010; Kortenkamp *et al.*, 2007). Vanhoudt *et al.* (2012) have reviewed multiple stressor studies that included ionizing radiation. The majority of these studies manipulated two or more stressors, most commonly the combination of gamma radiation and one or more heavy metals (Geras'kin *et al.*, 2007; Mothersill *et al.*, 2007; Olsvik *et al.*, 2010; Salbu *et al.*, 2008).

The investigation of the Fen Complex area by NGI-UMB (2010) and initial analyses in the current work (data presented in Paper I) revealed the simultaneous presence of radionuclides (^{232}Th , ^{238}U and progenies) and trace elements (As, Cd, Pb, Cr, Cu, Ni, Zn) in soil and water samples. Concentrations higher than the pollution screening levels (Pollution Control Act, 2010; SFT, 2009) were measured in soil samples at certain sites. Since the absolute soil concentrations of radionuclides and trace elements may not necessarily imply increased risk from multiple stressors, further analytical steps, such as determination of the mobilization potential and biota uptake were conducted in order to characterize the multiple exposure scenarios in the Fen environment.

2.3 Mobilization and ecosystem transfer

Weathering under different environmental conditions during long periods, together with human industrial and mining activities, leads to mobilization and transport of naturally occurring radionuclides through the environment to a variable degree. NORM can reach humans via different pathways, including the food chain, inhalation, ingestion, external exposure (O'Brien and Cooper, 1998). Natural sources of trace elements include geological weathering of parent rock material or derived soil, geothermal sources, volcanoes, forest fires (Landner and Reuther, 2004), while anthropogenic sources include industrial, vehicular and domestic emissions (Adriano, 2001). Depending on the trace element type and source, pathways in the environment include ground deposition, surface run-off, leaching to groundwater, air/soil/water exchange, uptake in plants, and transfer to man.

In general, mobilization, ecosystem transfer and impact of radionuclide and trace elements on the surrounding environment are determined mostly by the source which they are released from, by their speciation (their physical and chemical forms), binding mechanisms, as well as environmental conditions (Salbu, 2000; Vandenhove *et al.*, 2009a). To assess short- and long-term consequences, and to define potential

intervention, information on the source, transport through the environment, biological uptake and biological effects are needed (Figure 6). The investigation of binding mechanisms in soil, speciation analysis of water samples and determination of soil distribution parameter (K_d) were conducted in the present work in order to characterize the potential mobility of radionuclides and trace elements. Uptake in biota was considered by determining transfer factors (TF), while radiation risk was estimated through the biota radiation dose rates ($\mu\text{Gy/h}$).

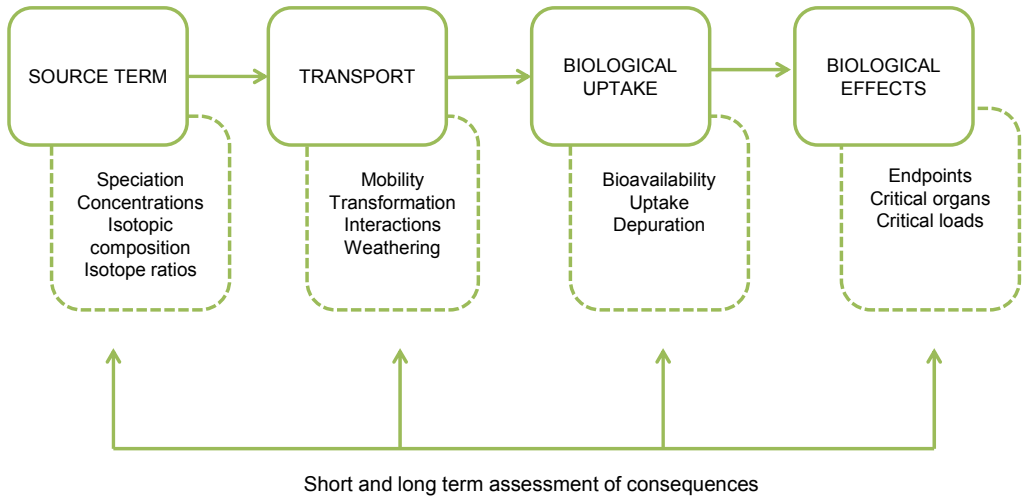


Figure 6. Mobilization of contaminants from source to organism, assessment model and data needed (Salbu, 2000).

2.3.1 Sequential extraction

Total soil concentration of an element of interest is useful for many geochemical applications, but more often the bioavailability is more of an interest in environmental studies or in agriculture (Cottenie *et al.*, 1980). Sequential extraction is a proven tool to identify binding mechanisms of elements with soil/sediment fractions. As such, it allows the estimation of mobility and availability of elements towards biota. Chemical fractionation schemes for the partitioning of elements in soils and sediments have been used extensively since the 1970s, with many of the procedures being slightly modifications of Tessier's schemes (Tessier *et al.*, 1979).

The basic principle for all sequential extraction procedures is consecutive use of reagents with an increasing extraction power, chosen to differentiate between different binding mechanisms (Oughton *et al.*, 1992; Skipperud, 2005; Tessier *et al.*, 1979). By using the sequential extraction procedures which are operationally defined (i.e., results depend on the specific method and reagents used), 3-7 different fractions

(depend on used procedure) of the total element concentration can be reported (Landner and Reuther, 2004). The obtained fractions are referred to in the literature as water soluble, exchangeable, carbonate-bound, Fe- and Mn-oxide-bound, organic compound-bound, residual or lattice material components (Zimmerman and Weindorf, 2010). These fractions reflect binding forces of different nature that operate in soils including their mechanisms.

Fractionation results depend on operational characteristics, such as the used reagents, extraction times, solid-liquid ratios, temperature and separation of phases. Therefore, these factors can influence the analytical results unless standardized (Salbu, 2000). Despite the criticism on the method of sequential extraction such as lack of procedures uniformity, lack of selectivity of reagents used, difficult quality control, data interpretation referring to soil fractions instead of to reagents applied (Kim and Fergusson, 1991; Nirel and Morel, 1990; Salbu, 2000; Tack and Verloo 1995), the extraction schemes are still widely in use (Filgueiras *et al.*, 2002; Haas and Fine, 2010; Skipperud, 2005).

The sequential extraction procedure was applied on soil samples from the Fen Complex area in order to assess the potential radionuclide and trace element mobility based on determined binding mechanisms and soil fractions. Extraction reagents and conditions employed in the current work are detailed described in section 3.2.3.2.

2.3.2 *Distribution coefficients*

The behavior and ultimate impact of substances (radionuclides, trace elements, heavy metals) in soils are largely controlled by their chemical form and different binding processes often classified under the broad term of sorption. The degree of sorption on solid phase in soil is commonly quantified by the distribution coefficient (K_d) (IAEA, 2010). Mathematically, the K_d ($L\ kg^{-1}$) equals the ratio of the concentration in the solid phase (soil), divided by the concentration in the liquid phase (water) under equilibrium conditions (Harvey and Leonard, 2002; IAEA, 2010). A higher value of K_d implies strong sorption and consequently, the possibility for transport through erosion processes; a low value of K_d implies little retention by soil, but the possibility for transport by underground water. Since sorption is element-species- and soil-dependent and is affected by soil mineralogy (the clay content and type, Fe oxides and hydroxides), the organic matter content and soil geochemistry (pH, presence of colloids, presence of counter-ions), the K_d value can vary for the same element by several orders of magnitude, depending on which experimental method is used for its quantification and which conditions are taken into consideration (Vandenhove *et al.*, 2009b). Still, due to its relative simplicity, the K_d model is one of the most used

models in environmental transport assessment studies today. In adopting the K_d concept, though it involves simplification of the reality, it is important to recognize that equilibrium conditions are justifiably assumed (IAEA, 2010). The apparent K_d can change over time due to interactions with soil components, such as clays or organics, weathering of particles (Salbu *et al.*, 2004), and aging effects related to sorption dynamics (Vandenhove *et al.*, 2009b).

Radionuclides and trace element K_d values determined for the Fen Complex soil samples are given in Paper II.

2.3.3 *Transfer factors*

The determination of contaminant transfer from abiotic to biotic systems is well-established and an important step in assessing environmental contamination. Transfer factors (TFs) (given on a dry weight biota basis) or concentration ratios (CRs) (given on a wet weight biota basis) (both terms are widely in use) are defined as the ratio of the concentrations of radionuclide/element in an organism to that in soil (Beresford *et al.*, 2008a; IAEA, 2010; Vandenhove *et al.*, 2009a). By definition, TF describes the amount of an element entering an organism from its relevant environmental compartment (air, soil, water) under equilibrium conditions (IAEA, 2010; Sheppard and Sheppard, 1985). Thus, these proportionality constants provide simple means to estimate radionuclide activities or trace element concentrations in biota, from measured concentrations in abiotic components such as soil or water (Ehlken and Kirchner, 2002; Higley, 2010; IAEA, 2010; Vera Tomé *et al.*, 2003).

However, for many radionuclides and trace elements, transfer will change over time as a result of different parameters and processes such as ecosystem structure, the element's fundamental chemical nature, the physical and chemical characteristics of the soil, temperature regimes and hydrological conditions, migration of elements into the soil, changes in ingestion/uptake quantities, biological half-lives, etc. Therefore, TFs are actually the result of complex interrelationships between an organism, its ecosystem and the fundamental chemical behavior of a particular radionuclide/metal species (Figure 7). The transfer factors were determined in the present thesis (Papers I, III and IV) for radionuclides ^{232}Th , ^{238}U as well as for series of associated trace elements. The concentration ratios were calculated for radionuclides ^{232}Th , ^{228}Ra , ^{238}U , ^{226}Ra and ^{210}Po as they were needed for the ERICA dose rate modeling in biota.

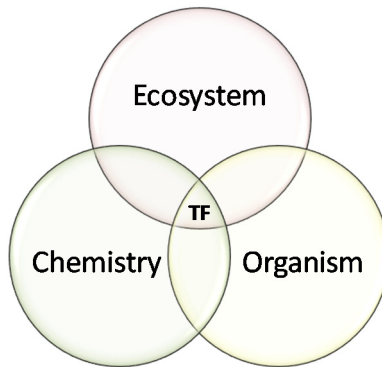


Figure 7. The interrelation and influence of an organism, its ecosystem, and radionuclide chemical behavior on TF (Higley, 2010).

2.4 Impact of legacy and naturally enhanced radiation on humans and the environment

Historically, scientific and regulatory activities have emphasized human health risk and mainly neglected ecological effects and risks to non-human organisms (Suter II, 2007). However, the prevailing anthropocentrism and belief that protection of human health automatically protects non-humans, have been more often replaced with ecological risk assessment (ERA) (ERICA, 2007; Suter II, 2007; USDOE, 2002) or integrated risk or impact assessment for both humans and non-humans (Suter *et al.*, 2003; WHO, 2001). Although approaches are named differently, the practical assessment tiers remain the same, i.e. approaches are rather different in their mandate than in ways of analyzing and managing hazards to the environment. However, the concept of environmental protection and legislative requirements still differ from country to country, and no true consensus has been achieved with regard to environmental protection questions (Suter II, 2007).

The major contribution to the radiation exposure dose of humans is from naturally occurring radionuclides. In ENRAs such as those in Iran, India, Brasil or China (Nair *et al.*, 1999; UNSCEAR, 2000), terrestrial sources of radiation are by far the most predominant contributors to the total radiation dose of the local populations. At legacy NORM sites worldwide, such as former mining sites in Central Asia, the radiological exposure doses in both humans and biota have been of concern for years after a period of active U mining (Salbu *et al.*, 2013; Skipperud *et al.*, 2013a; Stegnar *et al.*, 2013). Parameters obtained in investigations of terrestrial and aquatic biota uptakes of radionuclides in natural conditions are relevant for uptake estimation and

modeling of the consequences of human activities, nuclear pollution or accidental situations. However, the knowledge gap and lack of data for many non-human organisms have been emphasized (Beresford *et al.*, 2008 a, b; Jones *et al.*, 2003).

2.4.1 *Ecological impact assessment*

Sites with enhanced levels of NOR in different parts of the world have been documented in the literature (Geras'kin *et al.*, 2007; Malanca *et al.*, 1993; Quindos *et al.*, 1994; Shiva Prasad *et al.*, 2008; Termizi Ramli *et al.*, 2005; Wang, 2002). Considerations of a contaminated environment in a more integrated manner, analysis of all ecosystem interactions between man, flora and fauna have been initiated during the last decade (Bréchnignac, 2011; Oughton and Strand, 2004). Several approaches have been developed for estimating risk to non-human organisms from radiation in the environment (e.g., Suter II, 2007; ERICA, 2007; ICRP, 2007; USDOE, 2002). In general, different approaches include stages such as planning and problem formulation, analyses and characterization of exposure and effects, risk or impact identification, decision and risk management (Suter II, 2007).

The planning stage is undertaken prior to assessment, where certain issues such as the goals, scope and complexity of the assessment, as well as different management options, are discussed by stakeholder, assessors and managers. Defining the problem includes collecting of all available information concerning sources or contaminants, establishment of screening criteria, definition of assessment endpoints, describing the conceptual model and development of an analysis plan. During the analysis stage, exposure to radiation (contaminants) and possible effects are identified and analyzed. Measurements of radionuclide concentrations, analysis of the distribution, spatial and temporal variations, measurement of effects, dose-response analyses should be included. Impact identification consists of estimation and characterization of the possible impacts. Risk estimation is based on a combination of results, calculations and models, probability analyses, while the associated uncertainties are described and evaluated (Suter II, 2007). The ICRP (2007) acknowledged the difficulty related to and complexity of environmental protection. However, ICRP subscribed to the global needs to and efforts required to maintain biological diversity and to protect the health and status of ecosystems, natural habitats and communities. Accordingly, several frameworks and approaches have introduced the use of "reference organisms" as typical entities that can provide a basis for estimating the radiation dose (ERICA, 2007; FASSET, 2003; ICRP, 2008; UNSCEAR 2008b).

The ERICA integrated approach

As a part of the 6th Euratom-Framework Research and Development Programme, the European project ERICA (Environmental Risk from Ionizing Contaminants: Assessment and Management) was launched in March 2004 and ended in February 2007. This project aimed to develop an integrated approach designed to assess the effects of radioactive contaminants on the environment. At a scientific, decision-making and societal level, this involved focusing on protecting the fauna, flora and ecosystems. The three main elements in the ERICA Integrated Approach, as described by Larsson (2008), are assessment, risk characterization and risk management. The assessment stage comprises estimating and measuring activity concentrations of selected radionuclides in the environmental compartments and organisms, defining the exposure conditions and estimating the radiation exposure dose rates ($\mu\text{Gy/h}$) in selected biota. The assessment is possible to perform in three defined tiers, depending on the level of concern or regulatory demand. Risk characterization is a further stage where the estimation of the probability of adverse effects on selected biota, together with uncertainties, is evaluated. Management is the stage comprising all prior processes, during and after the assessment (Larsson, 2008). To date, the ERICA approach has been widely used for biota risk assessment in different ecosystems.

The ERICA Tool is a computerized flexible software system for assessing the radiological risk to biota. It guides the user through the assessment process, keeps records and performs the necessary calculations to estimate exposure dose rates of selected biota (Brown *et al.*, 2008). Further, the interactions with a number of databases and other functions that help the assessor to estimate environmental media activity concentrations, activity concentrations in biota, and dose rates to biota are included. The Tool allows the user to perform the assessment in three tiers (Figure 8). Tier 1 assessments are media concentration based, with a series of default values for transfer and mobility parameters (e.g., CR, K_d) as well as pre-calculated concentration limits in environment compartments which provide risk estimate. Tier 2 allows the user to edit parameters as CR, K_d , soil water content, dose conversion coefficients, weighting and occupancy factors. Identification of the possible risk is given in a deterministic way by defining a Risk Quotient (RQ) as the ratio of the predicted dose rate to a Predicted No Effect Dose Rate (PNEDR) or screening value. ERICA proposes a screening value of $10 \mu\text{Gy/h}$. Thus, results from tier 2 provide a realistic picture of radiation exposure and risks. Tier 3 assessment offers the same flexibility as tier 2, but allows the option to run the assessment probabilistically (Brown *et al.*, 2008). In the present thesis, the ERICA Tool (Tier 2) was used to calculate the

radiation dose rates of chosen reference organisms and estimate corresponding risks. In contrast to human radiation protection doses (given in unit Sievert (Sv) obtained from absorbed dose unit Gray (Gy) after taking the radiation type and irradiated organism part into the consideration), radiation doses for non-human organisms are given in Gy due to the lack of the information on relationship between absorbed doses and effects in them.

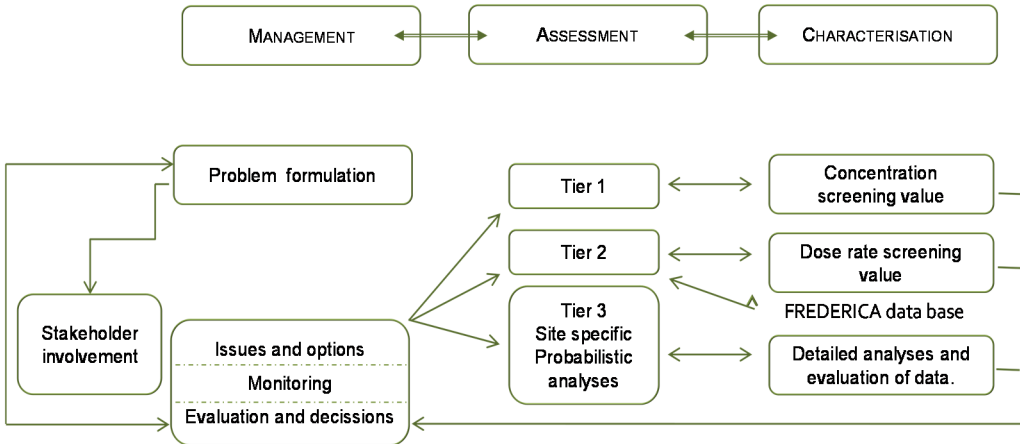


Figure 8. ERICA approach stages (Larsson, 2008).

2.4.2 Human impact assessment

The exposure of human beings to naturally occurring ionizing radiation in the form of cosmic radiation and radionuclides originating in the Earth’s crust, is continuous and an inescapable feature of life on earth (UNSCEAR, 2000, 2008a). The magnitude of natural background radiation exceeds in most of the situations man-made radiation. From a worldwide standpoint, annual exposure to natural radiation would be expected to result in a dose in the range 1-10 mSv, with 2.4 mSv being the present estimate of an average worldwide dose (UNSCEAR, 2000, 2008a). Radiation protection has primarily been concerned on the control and management of exposure to ionizing radiation so that human health is protected (ICRP, 1991). The health effects of exposure to radiation have been divided into two categories: deterministic and stochastic effects. Deterministic effects occur when a person is exposed to radiation levels greater than the threshold levels; the severity of the effects increases at higher dose. These types of effects are not of concern in low radiation exposure scenarios. Stochastic (probabilistic) effects are produced in individuals exposed to low radiation and arise from non-lethal cell modifications. The probability (not the severity),

increases with increasing doses in this case. The ICRP and different national radiation and environmental protection organizations have always accepted a theoretical risk from low dose radiation exposure. The precautionary principle and a linear-no-threshold (LNT) dose-response model (Figure 9) have been widely accepted. According to the LNT model, there is no safe radiation dose. However, many scientific groups have proposed models with curves that deviate from the linear, suggesting possible underestimation of the risk at areas of exposure to low doses and no risks i.e., benefits from low dose radiation.

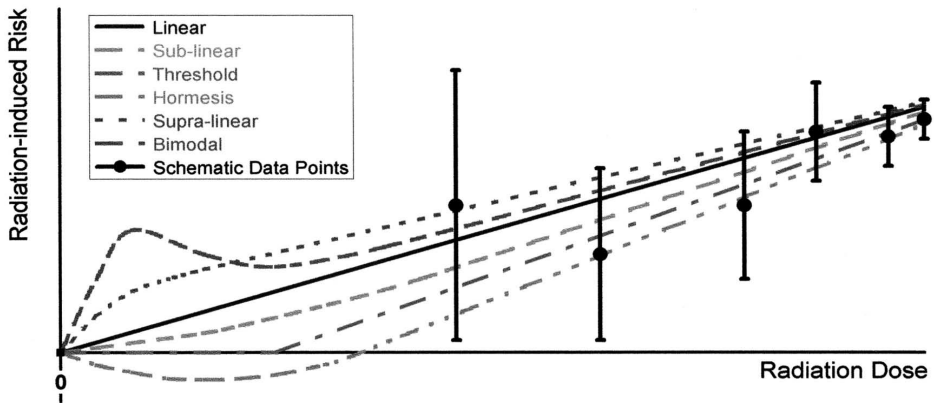


Figure 9. Some possible dose-response curves (Wakeford and Tawn, 2005)

Main physical quantities used in radiological protection of humans are radionuclide activity (Bq) and absorbed dose (Gy). The special radiation protection quantities are equivalent dose (Sv), effective dose (Sv), committed dose and collective dose (man-sievert). According to radiation protection and safety principles (IAEA, 1996; ICRP, 2007), individual doses due to the combination of exposures from all relevant practices should not exceed specified dose limits for occupational (20 mSv/y averaged in 5 years or 50 mSv in a single year) and public (1 mSv/y) exposure. Human radiation exposure dose assessment is to be performed whenever compliance with relevant dose limits needs to be checked or when identification of possible intervention measures aimed at reducing exposure are needed. Internal exposure through ^{222}Rn inhalation, ^{232}Th , ^{238}U and their progenies in the air inhalation, food and water intake and external exposure through gamma radiation are the main exposure routes commonly investigated. According to UNSCEAR (2000, 2008a), ^{222}Rn inhalation makes up at least 50 % of the average annual radiation dose, gamma

radiation (20 %), cosmic radiation (18 %), and radiation exposure from the radionuclides present in water and food (12 %) (UNSCEAR, 2000, 2008a).

Dose assessment can be performed in various ways, depending on many factors. Usually it is a multistage process that requires analyses of the environment and human habits, as well as application of different conversion factors (Figure 10) (ICRP, 2007). The first stage is, as described in the ecological impact assessment, the identification and characterization of the radiation source. The second stage is to collect the data on radionuclide concentrations in environmental compartments, their speciation, distribution, mobilization, etc. Both measuring and modeling can be performed at this stage. The next stage requires an assessment of the combination of radionuclide concentrations and data describing the habits of humans. The dose conversion coefficients and related quantities are used in the fourth stage to calculate internal and external doses. The final stage is summation of the internal and external doses and its expression as a total annual dose. Following the determination of radiation exposure doses, impact and risk assessment and identification of the potential actions should be performed.

Generally, impact of contamination and/or risk assessment is always needed at sites with potentially elevated soil, water, air or food radioactivity levels. Any intervention should be based on the results of assessment and taking the basic principles of radiation protection into account:

- principle of justification
- principle of optimization
- dose constraint

Within the scope of the present work, human radiation exposure doses due to outdoor exposure to high levels of terrestrial gamma radiation and ^{222}Rn were estimated and evaluated. Additionally, a possible dose contribution from outdoor exposure to ^{220}Rn was evaluated and all uncertainties addressed.

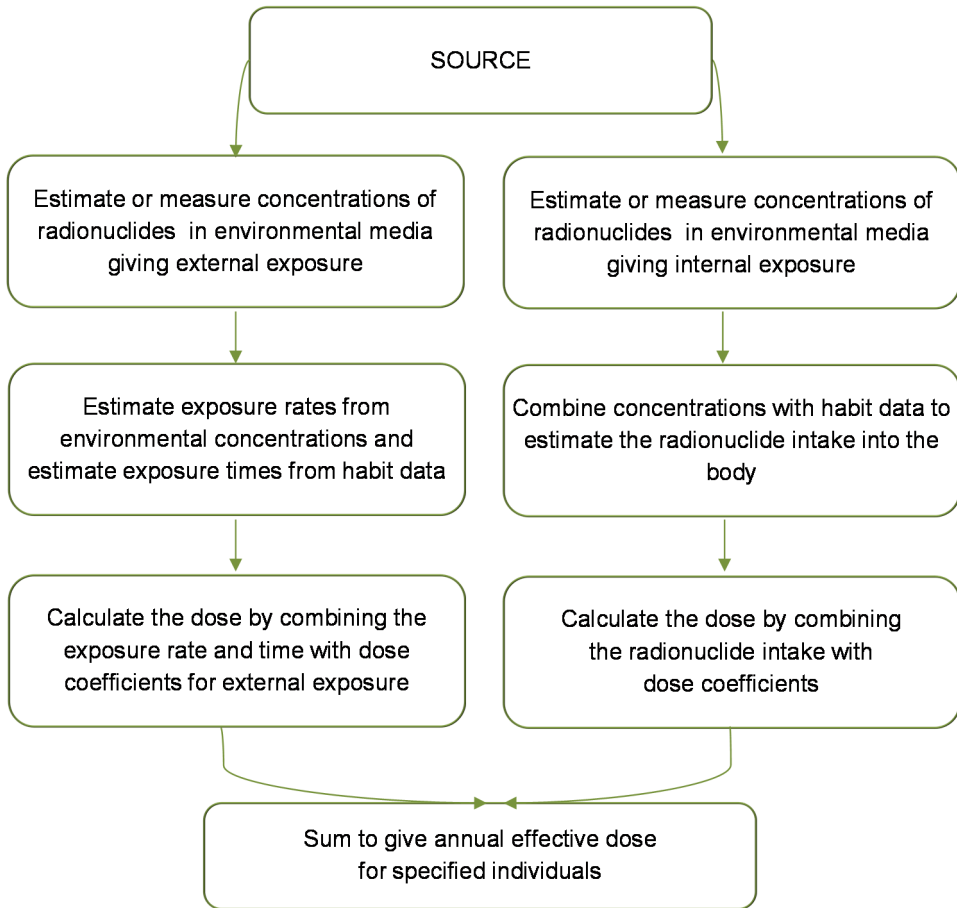


Figure 10. Dose-assessment process (ICRP, 2007)

3 Methodological approach

3.1 Planning and problem formulation

Planning of the current study, its scope and objectives, detailed investigations and their time frame, was undertaken during several meetings in the winter of 2008. The screening criteria were established. Possible exposure scenarios and organisms that might be at risk were identified. Detailed literature search, collecting and analysis of existing data on geology and radiation levels in the Fen Complex area preceded meetings and decision making. This phase can generally include consideration of political and societal issues and involvement of parties, such as stakeholders and politicians. However, the current work was focused "only" on the scientific aspect, i.e., on the assessment of radionuclides and trace elements contamination and their impact characterization. Eventual risk management and decision-making processes in the future could benefit from this study. Possible exposure scenarios included biota and critical groups of people that might be affected, exposure routes and exposure times. The dose assessment for humans took into consideration people working at an engineering firm at the former mining site Søve, people living in the close vicinity of undisturbed wooded sites Bolladalen and Rullekoll, and people regularly spending recreational time in the Fen woods. Investigation of outdoor radiation exposure via two exposure pathways: external irradiation with terrestrial gamma-emitting radionuclides, and internal exposure through $^{220,222}\text{Rn}$ inhalation, was conducted.

3.1.1 *Time frame of the assessment*

The assessment was performed for the "existing exposure scenario" (ICRP, 2007) in the Fen Complex area. The intention was to collect specific data on the current concentrations and behavior of radionuclides and trace elements in the area, and to assess the possible impacts on the whole environment, including both humans and biota. Assessment data, as mentioned in section 1.3., might be of interest in the future for comparison purposes, if new mining and extraction activities begin. The results were obtained from six fieldworks conducted during the period from 2008 to 2011.

3.1.2 *Site description*

The Fen Complex is situated in south eastern Norway, Nome Municipality, Telemark County (N 59° 16.40'; E09° 18.40' N59° 16.90'; E09° 17.16'). It is an early Cambrian intrusive complex of alkaline rocks and carbonatites with a roughly circular outcrop of 9 km². With its specific bedrock geology, the whole area is nationally and

internationally well-known. Today, most of the Fen Complex area is covered with marine clay and silt deposits that are sometimes many meters thick, and is, thus, inaccessible for bedrock examinations (Dahlgren, 2012). On one hand, this means that there is a large uncertainty about the size of the ^{232}Th resources in the area, while on the other, layers most probably contribute to different levels of shielding from radiation from the bedrock under the cover layers. Detailed information on the area is provided in section 1.2.

Two legacy NORM (former mining sites) and three undisturbed ^{232}Th -rich sites were the main subjects of the investigation. In addition, one reference site, outside the main ENRA, was chosen for comparison purposes and was investigated for the same parameters as sites within the ENRA, while three more sampling and measuring points within the surrounding residential area were included for certain measurements and analyses. The positions of the sites are presented in Figure 11. Each of the main sites was divided into a defined number of sampling/investigation points according to the sampling plan. The accurate geographical positions of the sampling and measurement points were recorded using the Global Positioning System (GPS) (Garmin Zumo 10R-023626; GPS-software version 4.00).

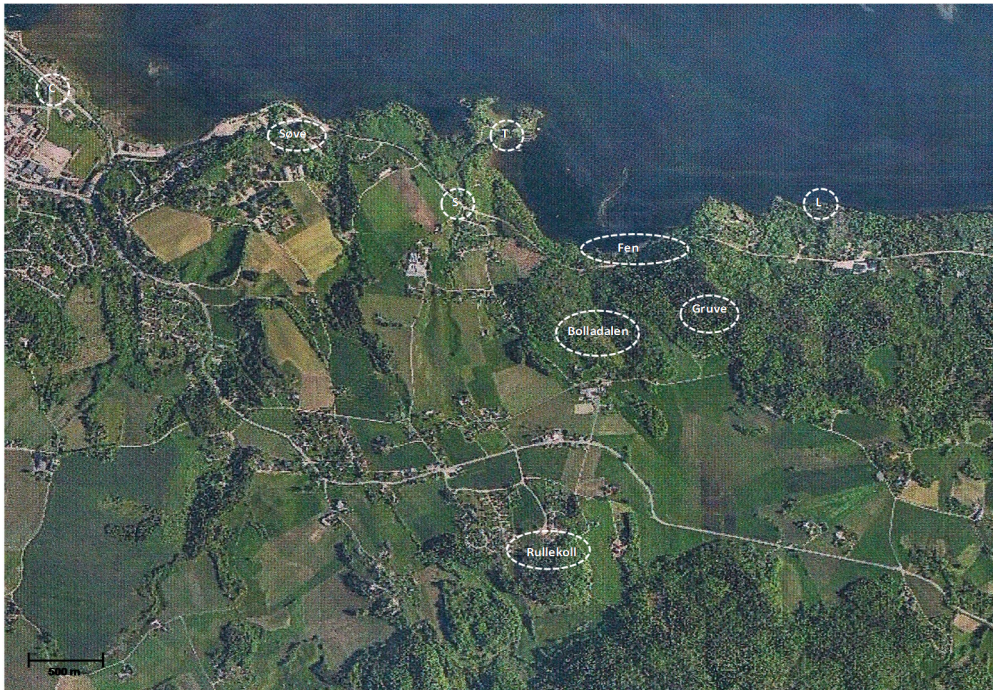


Figure 11. The Fen Complex map with marked study sites (<http://www.gulesider.no/>). Description is given in the text.

3.1.2.1 Legacy NORM (former mining) sites –

The former mining site Fen (N 59°16.625' E 009°18.226') is a part of the larger former Fe mining complex in the area called Gruveåsen. It is located along the shores of Lake Nordsjø (Figure 12). This site consists of the rock type rødbergite (red rock) rich in Fe and ^{232}Th . Mining for Fe was conducted during from 1652 to 1926. Ore exploitation was conducted in both open pits and underground tunnels, many of which are still visible. Data on the exact quantities of the mined ores, produced Fe and waste is not available. The biggest ^{232}Th resources were estimated to be in the Gruveåsen area, where also the highest terrestrial gamma dose rates in the air were measured (Dahlgren, 2012).



Figure 12. **a)** crushed waste rocks at the former mining site Fen; **b)** small stream at the former mining site Fen; **c)** "washing house" part of the former mining site Søve; **d)** material deposited on the slope of the Lake Nordsjø at the former mining site Søve (Photo: J. M. Popic).

The former mining site Søve (N 59° 16.902' E 009°17.162') is a decommissioned facility in the west part of the Fen Complex with rock type søvite predominant in the bedrock. The mining operation was based on this rock which consists mainly (75 – 95

%) of calcium carbonate and some minerals (e.g., pyrochlore, columbite, fersmite) rich in Nb (0.35 % Nb₂O₅) and to a lesser degree radionuclides ²³²Th and ²³⁸U. The ferro-niobium production at this site was conducted during 1953–1965 on a commercial basis. Detailed information about the production volumes, together with waste estimates, is given in Dahlgren (2005). In total, it is estimated that 1.15 million tonnes of søvite were extracted and passed through the subsequent extraction processes. The first, preparation phase of ferro-niobium production, crushing the mineral materials to necessary grain size, separation and centrifuging, is especially important with respect to waste production. In these processes, waste was discharged into the water stream and released into the environment. In next phase, ferro-niobium was produced by thermal exposure of Nb-bearing minerals that were extracted during the first phase. Crucibles from this process were then taken outside, to the rear of the facility where slag was separated from the ferro-niobium (Dahlgren, 2005; Dowdall *et al.*, 2012). Dahlgren (2005) estimated that a total of 302 tonnes of ferro-niobium were produced together with slag in excess of 800 tonnes, with a total volume in excess of 200 m³. After decommissioning of the mining site, it was covered with sand layers as a remediation measure. However, large amounts of waste, including slag and crushed rocks enriched with radioactive materials, were left in the area and only covered with sand. Today, the whole area of the mining and production site is freely accessible and in fact, an engineering firm is in operation in one part of the site. Before the present study was undertaken, several gamma spectrometric measurements (Dahlgren, 1983; Heincke *et al.*, 2008; IFE, 2006; NGI-UMB, 2010) demonstrated elevated gamma radiation levels.

3.1.2.2 Undisturbed ²³²Th-rich sites

The sites Bolladalen (N 59°16.424'E 009°18.945') and Gruvehaugen (N 59°16.544'E 009°18.464') are parts of the large wooded zone in the central area of the Fen Complex. Rødbergite is the main rock type in the bedrock at these sites. A more detailed description of the mining area is provided in the technical report on ²³²Th resource estimates by Dahlgren (2012). Since some open pits and parts of tunnels from the Fe mining period were observed in this area, the sites investigated in our work were slightly removed, in an undisturbed wooded environment. Several houses were positioned along the edge of the site Bolladalen, while walking paths were observed at both Bolladalen and Gruvehaugen. Site Rullekoll (N 59°16.865'E 009°17.204') is in an undisturbed wood in the south of the Fen Complex, just above the human settlement area. The predominant rock in the bedrock of this site is rauhaugite. No mining was conducted in the past at this site.

3.1.2.3 Reference sites

The site Torsnes (T) (N59°16.865'E009°17.204') was chosen as the Reference site outside the ENRA, although it was in close vicinity of it. No enhanced gamma radiation dose rates were previously measured at this site.

3.1.2.4 Additional sites

Soil and plant sampling as well as gamma dose rate measurements were performed at the site Skippervoll (S), positioned within the settlement area, in close vicinity to the main NORM sites. Additionally, sampling of water from the Lake Nordsjø and gamma ray measurements were conducted at several sampling sites outside the enhanced radiation area, (Lysnes (L), Ulefoss center (C)).

3.1.3 Radionuclides and trace elements considered

The present assessment focused on the radionuclides ^{232}Th and ^{238}U and trace elements As, Pb, Cd, Cr, Cu, Zn and Ni. These elements were thoroughly analyzed in different samples collected from the Fen Complex. In addition, the radionuclides ^{228}Ra , ^{226}Ra and ^{210}Po were measured in some soil and biota samples. Results were considered together with ^{232}Th and ^{238}U in order to obtain an estimate of the soil contamination status and for the ERICA modeling. The levels of ^{220}Rn and ^{222}Rn in the air were measured to assess the exposure doses to humans after outdoor exposure.

3.1.4 Organisms considered - biota and human assessment

A default set of reference organisms, given in the ERICA Tool for terrestrial and freshwater ecosystems, was used during the initial screening of dose rates for biota (Paper I) since no detailed descriptions with respect to plants and animals inhabiting the Fen Complex area were found in the literature. The screening included amphibians, benthic fish, birds, bivalve molluscs, crustaceans, gastropods, insect larvae, mammals, pelagic fish, phytoplankton, vascular plants and zooplankton for the freshwater ecosystem assessment, and amphibians, birds, bird eggs, detritivorous invertebrates, flying insects, gastropods, grasses and herbs, lichen and bryophytes, mammals (deer, rat), reptiles, shrubs, soil invertebrates (worm) and trees for the terrestrial ecosystem assessment. During the following stages of the study, only terrestrial ecosystem organisms were considered and transfer from soil was studied for nine plants species and four earthworm species (detailed in section 3.2.1.). Site-specific dose rate calculations, using the Fen Complex soil activity concentrations of radionuclides and their CRs, were performed for earthworms, pine trees, lichen and

bryophytes, shrubs, grasses and herbs. Evaluation of outdoor exposure doses for certain groups of people (as defined in the exposure scenarios) was also conducted.

3.1.5 Screening criteria

For the purpose of the assessment of the contamination status and possible impacts on humans and biota, the screening criteria were defined during the initial assessment phase as follows:

- Soil screening levels – New amendments about radioactive waste have been in use in Norway from 1st January 2011 (Pollution Control Act, 2010). A set of activity levels was defined when material is considered to be radioactive waste, when waste is subject to obligatory final disposal and when radioactive discharges require a license. For the present work, a set of values pertaining to the level at which the material is classified as radioactive waste was used. That included screening level of 1 Bq/g for ²³²Th, ²²⁸Ra, ²³⁸U, ²²⁶Ra and ²¹⁰Po.

When more than one radionuclide was to consider in the soil, the decision on radioactive waste was based on the risk quotient (RQ) value:

$$RQ = \sum \frac{Cr}{Csc} \geq 1$$

*Cr is radionuclide activity concentration,
Csc is the proper screening value.*

With respect to soil trace elements, limit concentration values (norm values) for unpolluted soil in Norway (SFT, 2009) and European data provided baseline concentration of chemical elements in natural samples of soil, water and sediment (De Vos and Tarvainen, 2006) was used as screening levels (Table 1).

Table 1. Soil screening levels for trace elements (mg/kg)

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

- Water screening levels – Quality of water with respect to radionuclides and trace elements was estimated using data from KLIF (2012) and De Vos and Tarvainen (2006), which provided the levels of ²³²Th and ²³⁸U in natural waters in Norway and Europe, respectively. In addition, water concentrations were compared to limit

concentrations given by WHO (2011) for drinking waters. Water screening concentrations are given in Table 2.

Table 2. Water screening values ($\mu\text{g/L}$)

Source	As	Pb	Cd	Cu	Zn	Cr	Ni	Th	U
KLIF, 2012	0.15	0.05	0.03	0.3	1.5	0.2	0.5	-	-
De Vos & Tarvainen, 2006	0.63	0.09	0.01	0.9	2.7	0.4	1.9	0.009	0.32
WHO, 2011	10	10	3	2	-	50	70	-	30

- Biota screening levels – Radionuclide and trace element concentrations in selected biota were compared to different literature sources (UNSCEAR, 2008b; individual papers cited in section 5.2.4), while transfer parameters (TFs, CRs) were compared to those given by ERICA (2007), IAEA (1992, 1994, 2010), ICRP (2009) and UNSCEAR (2008b). For calculated biota dose rates, the screening level of 10 $\mu\text{Gy/h}$, as provided by the ERICA Tool, was used. This dose rate is adopted as the Predicted No Effect Dose Rate (PNEDR). In addition, dose rate values of 40 $\mu\text{Gy/h}$ and 400 $\mu\text{Gy/h}$ for terrestrial animals and plants, respectively, provided by international organizations (ICRP, 2008; UNSCEAR, 2008b; US DOE 2002) as dose rates below no effects on population level could be observed, were also considered.
- Radiation dose constraints – Radiation dose limits are intended to serve as a boundary condition that will prevent deterministic effects and limit the probability of stochastic effects in humans (ICRP, 2007). The dose limit, for all radiation sources, for members of the public (in addition to inescapable natural background radiation) for situations having societal benefit, but without individual direct benefit, is set to 1 mSv/y (ICRP, 2007). In addition, the dose of 10 mSv/y, as an existing annual effective dose from all of the summarized radiation sources, was considered as the reference level that served to obtain conclusions with regard to possible interventions (ICRP, 2007).

3.2 Sampling, measurement and analysis

This assessment stage refers to fieldwork, sampling and retrieval of radiation parameters, laboratory handling, analyses and measurements of radionuclide and trace element concentrations in the soil, rock, water and selected organisms.

3.2.1 *In situ measurement and sampling*

3.2.1.1 Measurement of gamma dose rates and Rn concentrations in the air

Terrestrial gamma dose rates and ^{220}Rn and ^{222}Rn activity concentrations in the air were measured at undisturbed ^{232}Th -rich and legacy NORM (former mining) sites in the Fen Complex during fieldworks organized in 2009 and 2010 (Paper V). Portable gamma detector was used for measurement of the external gamma dose rates. Measurements were organized at sub-sites delineated by regular grids 10 x 10 m. Readings were repeated at 1 m above the ground until a constant signal was obtained (Auerbach and Reichle, 1999; Paul *et al.*, 1998). In addition, fifty thermo-luminescent dosimeters (TLD) were used for a long term measurement of gamma radiation. In that way, quality of gamma dose rates data was ensured by both short- and long-term recording and accurate spatial match of measured gamma dose rates with ^{220}Rn and ^{222}Rn levels was obtained (TLDs were put together with ^{220}Rn - ^{222}Rn detectors).

Passive integrating ^{220}Rn - ^{222}Rn discriminative detectors were used for measuring ^{220}Rn and ^{222}Rn concentrations in the outdoor air (Figure 13). Detectors were fixed to the trees, 1 m above the ground during two different seasons, in September-November 2009 and June-August 2010. A total of 86 detectors were placed in the Fen Complex area and 82 were recovered.



Figure 13. Photo of ^{220}Rn - ^{222}Rn detectors placed in the Fen Complex sites in 2009 and 2010 (Photo C.R. Bhatt).

3.2.1.2 Sampling

Soil samples were collected in several fieldworks, in two sampling ways. First, soil for total element analysis and sequential extractions was collected as bulk samples, with the usual sample weight from 0.2 to 1.0 kg. For analysis of the vertical distribution, soil was obtained by digging the pits and then incrementally excavating one wall of the pit to obtain sub-samples from different depths. The typical maximal

depth in both sampling approaches was 20 cm. Rock samples of different type and size were also collected at sites in the Fen Complex. Water sampling was conducted during the fieldwork in 2008. Total water samples were taken from the Lake Nordsjø at several sampling points (within and outside the ENRA) and from small water stream draining through the former mining site Fen. Sampling was conducted with high density polyethylene bottles and tubes. To minimize adsorption losses, the bottles and tubes were rinsed with deionized, lake or stream water before taking samples. Additionally to total water sampling, *in situ* filtration via membrane disc (Milipore, 0.45 μm) and ultrafiltration using hollow fiber tubes (Amicon, cut-off 10 kDa) were performed according to Teien (2005) and Salbu (2000). The aim was to distinguish between radionuclides and trace elements present as low molecular mass (LMM), colloidal or high molecular mass (HMM) species. Cation exchange resins (Chelex 100, BioRad, Na form, 200 mesh size) were applied to distinguish between charged species in the Lake Nordsjø samples. The combined size and charge fractionation system is presented in Figure 14. To preserve samples, ultrapure HNO_3 (1 %, v/v) was added ($\text{pH} < 2$) before transport to laboratory. *In situ* measurements of physical and chemical variables in water, such as conductivity, temperature, pH and ionic strength, were also performed.

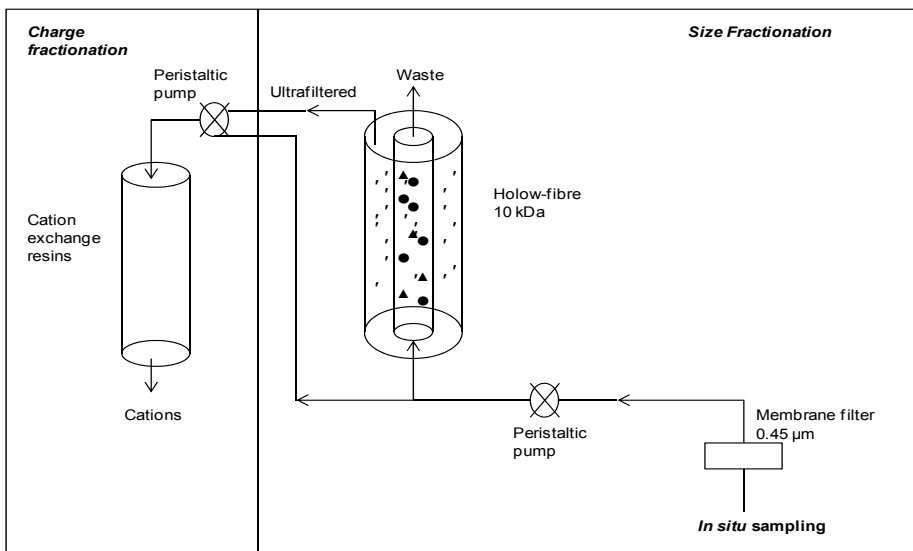


Figure 14. The applied size and charge fractionation system; from Teien (2005).

Plants and earthworms were collected at five to ten sampling points per site whose geographical positions were recorded in GPS. Plants that were found in abundance were regarded as representative, but not all plants were found at each of the sampling

sites. In total, nine different plant and four earthworm species were sampled. Additionally, wild berries and mushrooms were collected during the fieldwork for Msc project in 2011 (Valle, 2012). The chosen plants covered a relatively wide range of wild forest flora, with vascular and non-vascular plants, flowering plants, deciduous and coniferous trees. Leaves and needles were collected from trees, while the other plants were collected as whole plants. Corresponding soil samples were taken during the fieldwork, together with the plants, to ensure accurate determination of TFs. Earthworms were collected by digging the soil and hand sorting of organisms. Earthworms were transported to laboratory in the plastic boxes together with moist soil, roots and leaf litter. Soil, roots and leaf litter were taken at same points as earthworms to allow the evaluation of the earthworms' favorable environmental pools for the radionuclide and trace element uptake.

3.2.2 Sample preparation

Soil samples were dried at 40°C for several weeks, homogenized by sieving through 2 mm sieve and proper sub-samples were selected for analyses. Methods for determining the basic soil physical and chemical parameters (pH, grain size (%), H₂O (%), OM (%), P₂O₅, CaCO₃, CEC, Exch. Ca) are provided in Papers II and III. Decomposition of soil samples prior to ICP-MS is described in section 3.2.3. To release and analyze elements associated with different soil fractions, sequential extraction was conducted (section 3.2.3). Acidified water samples (total and fractionated) were kept in plastic bottles at low temperature (4°C) before the ICP-MS analyses.

Species of collected plants were in the laboratory identified as: *Betula pendula* (birch), *Pinus sylvestris* (pine), *Picea abies* (spruce), *Matteuccia struthiopteris* (fern), *Taraxacum officinale* (dandelion), *Sphagnum sp.* (moss), *Pseudevernia furfuracea* (lichen), *Fragaria vesca* (wild strawberry), *Lolium perenne* (grass), *Rubus idaeus*, *Vaccinium vitis-idaea*, *Vaccinium myrtillus* (wild berries) and *Cantharellus cibarius* (mushrooms). Plants were rinsed from visible soil adherents; roots were divided from the aboveground plant parts. Further, all plant parts were thorough washed and put for drying at 40°C for several weeks. Dry plants were milled and proper sub-samples were selected for the decomposition.

Collected earthworms were identified to belong to four species (*Aporrectodea caliginosa*, *Aporrectodea rosea*, *Dendrodrilus rubidus* and *Lumbricus rubellus*). Only adult organisms were selected for analysis. To obtain depurated earthworm gut contents, all earthworms were starved on moist filter paper for 48 hrs and rinsed several times with distilled water to remove visible soil. They were then separately

frozen in liquid nitrogen, freeze dried and milled into a fine powder. The collected roots and leaf litter were cleaned from visible soil particles, washed with distilled water, dried at 40°C and milled. Decomposition of biota was performed by acid-microwave digestion.

3.2.3 Analytical procedures

3.2.3.1 Acid-microwave digestion

Soil and rock (0.25 – 0.30 g), plant (0.5 g) and earthworm (individual organisms) samples were decomposed by acid digestion with ultrapure concentrated HNO₃ (5 ml) in an Ultraclave (single reaction chamber, MLS-Milestone UltraClave, MLS GmbH). Microwave digestion combines microwave radiation with high pressure and temperature in an autoclave design. This rapid method allows simultaneous digestion of many samples. At the same time, the advantages of this approach are efficient decomposition with minimized risk for sample loss and contamination (Agazzi and Pirola, 2000). The UltraClave program was set to incremental increase of temperature up to 260°C and pressure up to 170 bar. The total decomposition time was 2.5 hrs. After digestion, the solutions were transferred to 50 ml falcon tubes, diluted and measured by ICP-MS and ICP-OES.

3.2.3.2 Sequential extraction of soil

Sequential extraction involves treatment of a sample of soil with a series of reagents in order to partition the trace element/radionuclide content. The consecutively used reagents have different chemical natures, and the steps are performed in order of increasing extracting power. Hence, first isolated species are those already dissolved in soil pore water together with cation exchangeable species of soil. Stepwise attacks on the carbonate, Fe- and Mn-oxide and organic phase follow generally. Finally, more refractory soil components are dissolved in the last phase (Rao *et al.*, 2008; Salbu, 2000). In the present thesis, soil samples (1.00 g) were subjected to the sequential extraction (Salbu, 2000; Tessier *et al.*, 1979), applying the reagents and conditions given in Table 3. The respective extracts were dried by heating on a sand-bath (90°C), the solid was re-dissolved in 3 ml HNO₃ and re-dried for three times. Finally, all extracts were dissolved in 2.5 ml HNO₃ and diluted to 50 ml for analysis on ICP-MS. Results of the sequential extractions of soil samples allowed evaluating the mobility and transferring to biota (Papers II, III and IV).

Table 3. Extracted fractions, reagents and conditions employed in the current work (Salbu, 2000)

Reaction step	Fraction	Extraction reagent	Extraction conditions	
			Contact time, hrs	Temp °C
1	Water soluble	20 ml H ₂ O	1	20
2	Reversibly bound	20 ml 1 M NH ₄ Ac pH soil	2	20
3	Reversibly bound-associated with carbonates	20 ml 1 M NH ₄ Ac pH 5 (pH adjusted with HAc)	2	20
4	Easily reduced compounds (Fe/Mn hydroxides)	20 ml 0.04 M NH ₂ OH HCl in 25 % HAc pH 3	6	80
5	Oxidized compounds soil organic material	15 ml 30 % H ₂ O ₂ (pH 2 adjusted with HNO ₃) 5 ml 3.2 M NH ₄ Ac in 20 % HNO ₃	5.5	80
6	Acid dissolvable	20 ml 7 M HNO ₃	6	80

3.2.3.3 Procedure for ²¹⁰Po determination

The procedure for ²¹⁰Po determination is based on the method given by Chen *et al.* (2001). Though, the method applied here was somewhat modified and adapted in a recent study at the Isotope laboratory, UMB (NMBU), (Skipperud *et al.*, 2013b). A detailed description of processes, reagents and working conditions is given in Table 4. ²⁰⁹Po was used as a tracer. Results for ²¹⁰Po in soil and plant samples were included in the ERICA assessment of dose rates for selected plants (Paper IV).

Table 4. Description of ²¹⁰Po determination method

Process	Reagent	Temperature/Time
Decomposition	7 M HNO ₃ , Microwave (T, P)	0-260°C / 3 hrs
Oxidation	30 % H ₂ O ₂	200°C / 10 min
Digestion	12 M HCl	200°C / 10 min
Dilution	H ₂ O	200°C / 15 min
Complex formation	NH ₂ OH · HCl	200°C / 10 min
Thermal deposition with silver discs for alpha spectrometry counting		90°C / 3 hrs

3.2.4 Measurement techniques

There are two major methods of determining the radionuclide concentrations: radiometric counting methods, which determine the decay rate (counting of the emission of alpha and beta particles and gamma radiation) and measurements of the number of atoms present (using mass spectrometers that include a spectrum of mass spectrometers such as ICP-MS, AMS, TIMS, RIMS, SIMS). Until mid-1980s, radionuclide measurements were done by the traditional counting radiometric methods. Alpha, gamma spectrometry and beta counting still dominate the analysis of most radionuclides. However, since the major break-through with the mass spectrometric technique in 1980s (Chen *et al.*, 1986), there has been a shift in measurements of certain long-lived radionuclides including ^{232}Th . In last 25 years, the performances of mass spectrometric methods have been significantly improved and they have become more competitive regarding radionuclide measurements (Baskaran, 2011; Betti *et al.*, 2006; Goldstein and Stirling, 2003; Hou and Roos, 2008; Santos *et al.*, 2010). This is illustrated by the number of published papers on radionuclide determinations by mass spectrometric methods (Betti, 2000; Becker *et al.*, 2002; Crain, 1996; Mas *et al.*, 2012; Qiao *et al.*, 2009; Shi *et al.*, 2013; Skipperud and Oughton, 2004; Wendel *et al.*, 2013). In general, long-lived radioisotopes are best analyzed by mass spectrometric methods, while short-lived ones are preferably analyzed by radiometric methods (Hou and Roos, 2008). The radionuclide of interest, sample type, the required accuracy and sensitivity, cost of the analysis should be considered before the choice of measurement method.

In the present work, terrestrial gamma dose rates, ^{220}Rn and ^{222}Rn concentrations in the air, as well as radionuclides and trace elements in different samples from the Fen Complex, were measured by following devices and methods:

- Geiger-Müller counter and thermo-luminescent detectors – for measurement of outdoor terrestrial gamma radiation
- Passive discriminative alpha track detectors – for measurement of ^{220}Rn and ^{222}Rn outdoor air concentrations
- Inductively coupled plasma – mass spectrometry (ICP-MS) – for measurement of ^{232}Th , ^{238}U , As, Cd, Cr, Ni, Zn, Cu and Pb in soil, water and biota
- Gamma spectrometry – for measurement of ^{228}Ra and ^{226}Ra in soil and biota
- Alpha spectrometry – for measurement of ^{210}Po in soil and biota
- Digital autoradiography – for soil screening for radioactive heterogeneities

3.2.4.1 Measurement of outdoor terrestrial gamma radiation

Geiger-Müller counters (GMCs) are used to detect ionizing radiation, typically gamma and beta radiation, by detecting the presence and intensity of radiation (particle frequency, rather than energy). A common type of survey-meter for the gamma ray monitoring consists of a portable Geiger-Müller tube, high-voltage supply, and pulse-counting rate meter. The counter tube is the principal organ which is gas filled ion chamber with a hollow cylindrical cathode and a thin central wire anode. When a gas (e.g., argon) is hit by an ionizing radiation through the chamber's window, the ion pairs are produced in the gas and get attracted by the opposite charge electrodes, which generate an electrical signal (Choppin *et al.*, 2002). The anode transfers the pulses of current through a resistor in order to convert them into pulses of voltage. Then, the voltage pulses are recorded by a counting device and eventually, an oscilloscope, LED screen, or other display conveys the particle count to the user. Detailed analysis of the physical processes involved in the Geiger discharge is presented by Wilkinson (1996a, b, c). The Geiger-Müller tube is one of the oldest radiation detectors types in existence due to simplicity, low cost and ease of operations (Knoll, 2010). A portable gamma instrument Automess (Radiacmeter 6150 AD 4 LF), with a response range 0.01 – 9.99 $\mu\text{Gy/h}$, was used in measurements of terrestrial gamma dose rates in the Fen Central Complex.

For a long time period, thermoluminescent dosimeters (TLD) have been the passive detector of primary choice in personal dosimetry, environmental dosimetry and dating of archaeological and geological artefacts. TLD has a sensitive crystalline thermoluminescent material such as lithium fluoride (LiF), calcium fluoride (CaF_2), etc., that absorb ionizing radiation (Attix, 2004). The absorption of ionizing radiation causes some of the electrons in the molecules of crystals to absorb energy and shift to higher energy levels or bands. When the irradiated crystals are heated, they emit visible light. The intensity of this light is proportional to the amount of radiation the crystal has absorbed, which is measured with a photomultiplier tube (Attix, 2004). TLDs can integrate an absorbed dose over a period of time so that an effective or equivalent dose can be calculated (Olko, 2010). The type of TLD system used in the present assessment were MR 200C, with luminescence material CaF_2 doped with Mn [$\text{CaF}_2:\text{Mn}$], developed by the Jožef Stefan Institute (JSI), Ljubljana, Slovenia. It has been used successfully for almost 20 years in personal and environmental dosimetry and has been well suitable for readings of doses of ionizing radiation from 5 μSv to 5 Sv (Zorko *et al.*, 2006). The exposure time was 3 months (June – August 2010) after which the exposed TLDs were mailed to JSI laboratory for dose readings.

3.2.4.2 Passive discriminative measurement of ^{220}Rn and ^{222}Rn in the outdoor air

The environmental $^{220,222}\text{Rn}$ concentration is a function of time and climate conditions. To monitor it, both active and passive techniques have been developed. Active methods, such as grab radon/activated charcoal or grab radon/scintillation cell, are usually used for short-term measurements of Rn and for detailed investigations of individual sites under inspection. Passive methods, such as alpha track detection or electrets ion chamber – long term detection, are more suitable for the assessment of Rn exposure over long time periods and can be used for large-scale surveys at moderate cost.

A newly designed alpha-track detector (Raduet) for discriminatively determining ^{220}Rn and ^{222}Rn concentrations was developed at the National Institute of Radiological Sciences (NIRS), Chiba, Japan (Zhuo *et al.*, 2002). These discriminative monitors have been used throughout the world (Chen *et al.* 2009; Mc Laughlin, 2010; Ramola *et al.*, 2003; Tokonami *et al.*, 2005; Žunić, *et al.*, 2010). Raduet detector is efficient in measuring ^{220}Rn and ^{222}Rn concentrations simultaneously; its lower and higher detection limits are 5-1000 Bq/m³ and 15-1000 Bq/m³, respectively (Tokonami *et al.*, 2005). The detector has two different diffusion chambers from an electro-conductive plastic with a cylindrical inner volume (~30 cm³). The detecting material is CR-39, which is placed at the bottom of the chamber with sticky clays. Radon can penetrate into the chamber via an invisible air gap between the lid and bottom by diffusion. The air gap acts as a high diffusion barrier. Thoron can barely get into the chamber with such a small pathway due to its very short half-life ($T_{1/2} = 55.6$ s) as compared to that of ^{222}Rn ($T_{1/2} = 3.82$ days). To detect ^{220}Rn more effectively, there are six holes of 6 mm in diameter at the side of the other chamber, covered with an electro-conductive sponge. In order to determine the conversion factors of ^{220}Rn and ^{222}Rn concentrations, the detectors were placed into the ^{222}Rn and ^{220}Rn chambers at NIRS respectively (Tokonami *et al.*, 2005). Following the exposure tests, CR-39 plates were taken out of the chamber and chemically etched with a 6.25 M NaOH solution at 90°C in the course of 6 h, and the alpha tracks were counted with a track reading system (Tokonami *et al.*, 2005). Then, using two alpha track densities of low and high air-exchange rate chambers, ^{220}Rn and ^{222}Rn concentrations were obtained with the help of certain mathematical equations (Tokonami, 2010). In the present work, 86 Raduet detectors in total were put at different measuring points in the Fen Complex (as described in section 3.2.1). After the exposure period, detectors were collected and sent to NIRS laboratory for reading.

3.2.4.3 ICP-MS

Inorganic mass spectrometry is a method used for the determination of element concentrations by measuring the number of atoms of the isotopes of the element. As a result of improvements of ICP-MS (comparable detection limits to the radiometric methods and a rapid analytical capacity), it has become a very useful procedure for the determination of long-lived radionuclides (Becker, 2003; Crain, 1996; Crain and Smith, 1995; Hou and Roos, 2008; Skipperud, 2005; Truscott *et al.*, 1999).

In the current work, main measuring method was ICP-MS. The justification of the choice of ICP-MS was based on the possibility for a simultaneous determination of both radionuclides and trace elements in a large number of environmental samples. The time for analysis was much shorter than for alpha spectrometry and quality control were checked in the ways described in section 5.1. Before the measurements, samples were prepared by the acid-microwave digestion procedure described in section 3.2.3. The digested samples were then analyzed for ^{232}Th , ^{238}U and trace elements concentrations by ICP-MS, on a Perkin Elmer ELAN 6000. Thorium easily adheres to sample vessel walls as well as to internal components of ICP, leading to certain sample loss (Bailey, 1993; Holmes and Pilvio, 2000). To avoid this, 5 % HNO_3 is recommended as a matrix solution (Holmes and Pilvio, 2000). Accordingly, rock, soil and biota samples were measured in 5 % acid solutions (although a certain number of soil and rock samples were diluted to 1 % HNO_3). Due to a possible memory effect of Th, a 5 % HNO_3 wash was used, and the wash-out time was varied from 4 min for soil samples, to 2 min for biota samples. The following instrumental parameters were optimized every day before analysis: torch position, nebulizer gas flow, ion lens vs mass calibration, oxide levels and doubly charged ion levels. Typical operating conditions were given by Skipperud (2005). The internal standard (In, Tl, Rh) was added to each sample before microwave – assisted decomposition. The internal standard was added to soil and biota samples before digestion, while to water samples before the instrumental analysis. Calibration was performed using the single element (Spectroscan, Drøbak, Norway) and multi element standard (ICP-MS 68B). Quality check of results was done by measuring standard and blank samples after every 10 samples. Proper standard reference materials were used for all measurements (section 5.1.).

3.2.4.4 Gamma spectrometry

Gamma ray spectrometry is an analytical technique that allows the identification and quantification of gamma emitting isotopes in a variety of matrices without destruction of samples. Many of the alpha and beta emitting radionuclides also emit gamma

radiation and can be determined by gamma spectrometry. Gamma radiation, due to its high frequency and high energy, is indirectly ionizing. The main processes of its absorption in materials that allow detection are photoelectric effect, the Compton process and pair production. Different detectors can be used for measuring gamma rays, such as a scintillation counter using NaI, CsF and ZnS as scintillator, a gas-filled counter, and semiconductor detectors based on Si or high purity Ge. Germanium detector enables measurement of gamma radiation through detection of the charge carriers (electrons and holes), generated by the energy deposited by gamma ray photons. The number of electron-hole pairs is proportional to the energy transmitted by the radiation to the semiconductor. Due to gamma ray characteristic of long penetration in the matter without absorption, gamma emitting samples usually do not need pre-measurement preparation. Thus, risk for contamination in preparation steps is minimized, method is relatively easy and quick (Hou and Roos, 2008). Performance characteristics, advantages and limitations of this method are described elsewhere (Semkov *et al.*, 2002)

A certain number of soil and plant samples were in addition to ICP-MS, analyzed by gamma spectrometry (Paper IV and Paper V). Measurements were performed using a low background high-purity germanium detector (HPGe) at the Norwegian Radiation Protection Authority (NRPA). Soil was prepared in cylindrical geometries, isolated in aluminium foil and kept for a month before measurement to ensure secular equilibrium between ^{222}Rn and its progenies. Measurements were carried out using three different coaxial p-type detectors: Canberra GR2521-7500, with 45 % relative efficiency and FWHM of 1.9 keV; Ortec GEM-40190-S, with 40 % relative efficiency and FWHM of 1.8 keV, and Ortec GEM-33190-S, with 33 % relative efficiency and FWHM of 1.8 keV. Samples were generally counted for one or two days depending on the activity level (a minimum measuring time of 16 h for most active samples was employed). Quantitative analysis of ^{40}K was based on primary photon emissions (1460 keV), while ^{226}Ra was determined via ^{214}Pb (352 keV) and ^{214}Bi (609 keV) and ^{228}Ra via ^{228}Ac (911 keV).

3.2.4.5 Alpha spectrometry

Alpha spectrometry is the technique for measuring the energy spectrum of the alpha radiation emitted from a sample. By measuring the energy of the emitted alpha radiation, alpha emitters can be identified and measured. Because of the alpha particles' short range, sample preparation, chemical separation and sample mounting are necessary steps before counting by alpha spectrometry (Skipperud, 2005). There are several types of detectors that can be used for the measurement of alpha-emitting

radionuclides; examples are Frisch grid ionization chambers, proportional counters, plastic and liquid scintillation detectors and semiconductor detectors (Hou and Roos, 2008; Skipperud, 2005).

In the present work, alpha spectrometry was applied to measure ^{210}Po in soil and plant samples. The used system consisted of 6 Canberra models 7401 α spectrometers linked to a Canberra ADC model 1520. The spectra were acquired using Canberra's Genie PC 2000 software. The passive ion-implanted planar silicon (PIPS) detector (Canberra), a modern edition of silicon charged particle detectors which is commonly used for measurement of alpha spectra from thin sources, was used. Data on ^{210}Po activity concentrations were used in the ERICA calculations of plant exposure dose rates in Paper IV.

3.2.4.6 Digital autoradiography

As described by Lind (2006), digital autoradiography is one of the different imaging techniques that have been used for screening of environmental samples for radioactive heterogeneities. The primary detector is an imaging plate composed of a photostimulable phosphor, sandwiched between a flexible plastic support and a thin protective layer. The sample preparation includes spreading the samples on paper or plastic foils that are in contact with the sensitive film or image plate. The imaging plate (BaFBr:Eu^{2+}) traps the irradiated radioactive energy and stores it until scanning with a laser beam in an Image Plate scanner device, after which it is released as luminescence. The luminescence is then collected into a photomultiplier tube and converted to electrical signals which are stored as digital image information (Lind, 2006). The certain numbers of soil and earthworms from the Fen Complex were screened by digital autoradiography and results are given in Paper II.

3.3 Data analysis

3.3.1 *Used parameters and equations*

To describe the mobility of radionuclides and trace elements and their transfer between abiotic and biotic parts of the environment, parameters distribution coefficient (K_d) and transfer factor (TF) were used.

- Mathematically, K_d was calculated from the equation:

$$K_d = \frac{C_{\text{tot. soil}}}{C_{\text{soil solut.}}}$$

K_d (L/kg) is the distribution coefficient,

$C_{tot.soil}$ is the concentration of the element in the soil,

$C_{soil.solut.}$ is the concentration of the element in the aqueous soil fraction.

- The transfer factor (TF) and concentration ratio (CR) describe transfer of the element from the soil to an organism of concern. Both terms are defined as the ratio of the concentrations of element in an organism to that in soil. TFs are given on the basis of dry biota weight and CRs on the basis of wet biota weight (Beresford *et al.*, 2008a; Vandenhove *et al.*, 2009a). In the present work, the transfer of both radionuclides and trace elements was studied using TF, while CRs were calculated and used only when the ERICA Tool was applied to obtain the radiological dose rates. The equations used were:

$$TF = \frac{C_{biota (d.w)}}{C_{soil (d.w)}} \quad \text{and} \quad CR = \frac{C_{biota (w.w)}}{C_{soil (d.w)}}$$

TF and CR are the transfer factor and concentration ratio (unitless parameters), respectively,

C_{biota} is the concentration of element in biota tissue (based on dry or wet weight as previously explained),

C_{soil} is the concentration of the element in the soil.

To estimate the magnitude of radiation exposure doses which humans could receive, under different exposure scenarios, as a result of outdoor terrestrial gamma radiation and ^{220}Rn and ^{222}Rn outdoor inhalation, the following equations were applied:

- The annual outdoor effective doses from external gamma radiation were calculated according to UNSCEAR (2000, 2006):

$$H_{\text{gamma rad.}} (\text{mSv}) = D (\text{nGy h}^{-1}) \times 8760 \text{ h y}^{-1} \times 0.2 \times 0.7 \times 10^3 \text{ mSv} / 10^9 \text{ nGy}$$

H (mSv) is the annual outdoor effective dose from gamma radiation

D (nGy h⁻¹) is the measured gamma dose rate,

0.2 is the outdoor occupancy factor,

0.7 Sv Gy⁻¹ is the conversion coefficient from the absorbed dose in air to human effective dose equivalent.

- There are two approaches for Rn doses determination and estimation of related risk: dosimetric approach where dose is estimated on the basis of measured deposition of Rn decay products in the air way/lung, and epidemiological approach which takes conversion factors into account from the ^{222}Rn epidemiological studies on underground miners or residential ^{222}Rn (UNSCEAR, 2006). In the latter approach, concentrations of the potential alpha energy of short-

lived progenies of ^{220}Rn and ^{222}Rn are estimated by assuming a state of equilibrium between the parent and progeny nuclides. In practice, an equilibrium factor F is used to characterize the state of equilibrium. F is defined as the ratio of the actual potential alpha energy concentration (PAEC) to the PAEC that would prevail if all the progenies in each series were in equilibrium with the parent ^{220}Rn or ^{222}Rn , as the case may be. A simply way to express the F is as the ratio between the equilibrium equivalent concentration (EEC) and mother ^{220}Rn or ^{222}Rn concentration (UNSCEAR, 2006):

$$F = \frac{\text{ECC Rn}}{C_{\text{Rn}}}$$

F is equilibrium factor,

ECC is Equilibrium Equivalent Concentration (defined as the equivalent concentration of Rn progenies in equilibrium with the parent gas that yield the same potential alpha energy per unit volume as the existing mixture (UNSCEAR, 2006)),

C_{Rn} is measured radon gas air concentration.

In the present assessment, the commonly used epidemiological approach was used to estimate the dose from Rn progenies by measuring air ^{222}Rn gas concentrations and applying conversion factors. However, in case of ^{220}Rn , the dosimetric model is recommended to use since there is paucity of the ^{222}Rn epidemiological studies (UNSCEAR, 2006). The calculation of the lung doses directly from ^{220}Rn measurements is not practical because the F between the gas and short-lived progenies is not well established although much work has been done on it (UNSCEAR, 2006). Thus, it must be emphasized that here applied method should be considered as conservative and indirect approach of ^{220}Rn doses estimation for the Fen Complex area.

The annual effective outdoor doses from ^{220}Rn and ^{222}Rn were calculated by multiplying the measured gas concentration in the air with the proper equilibrium factor, dose conversion factor and the exposure time (UNSCEAR, 2000, 2006):

$$H_i \text{ (mSv)} = C_i \times F_i \times t \times \text{DCF}_i \times 10^{-6} \quad i = (^{220}\text{Rn or } ^{222}\text{Rn})$$

H_i is the annual outdoor effective dose from radon and thoron,

C_i is the ^{220}Rn (^{222}Rn) air concentration (Bq m^{-3}),

F_i is the equilibrium factor 0.003-0.1 for ^{220}Rn , and 0.6 for ^{222}Rn ,

t is the exposure time (h),

DCF is the dose conversion factor 40 nSv (Bq h m^{-3})⁻¹ for ^{220}Rn and 9 nSv (Bq h m^{-3})⁻¹ for ^{222}Rn .

- The total outdoor effective dose was obtained as a sum:

$$H_{\text{tot}} = H_{\text{gamma rad}} + H_{222\text{Rn}} + (H_{220\text{Rn}})$$

H_{tot} is the annual outdoor exposure dose

H_{gamma rad} is the annual outdoor exposure dose from gamma radiation

H_{220Rn} is the annual outdoor exposure dose from thoron inhalation

H_{222Rn} is the annual outdoor exposure dose from radon inhalation.

3.3.2 Statistical analysis

Univariate statistics (arithmetic and geometric mean, median, Kruskal-Wallis test, ANOVA, correlation and regression analyses, student's t-test) was performed using software programs MiniTab (MiniTab 15, 16) and Microsoft Excel (2007). In addition, multivariate statistical analysis (PCA) was conducted using the software CANOCO (Canoco 4.5). The used statistical tests are described in detail in Papers I-V.

4 Summary of scientific papers

4.1 Paper I – Characterization of contaminants in different environmental compartments in the Fen Complex

The Fen Central Complex in southern Norway, a geologically well investigated area of magmatic carbonatite rocks, is assumed to be among the world largest natural reservoirs of thorium (^{232}Th). These rocks, also rich in iron (Fe), niobium (Nb), uranium (^{238}U) and rare earth elements (REE), were mined during past centuries. Waste locations, that give rise to increased levels of both radionuclides and metals, are now situated in the area. Estimation of radionuclide and metal contamination of the environment and radiological risk assessment were done in this study.

The average outdoor gamma dose rate measured in Fen was $2.71 \mu\text{Gy h}^{-1}$. This was significantly higher than the world average dose rate of $0.059 \mu\text{Gy h}^{-1}$. The annual exposure dose from terrestrial gamma radiation, related to outdoor occupancy, ranged from 0.18 to 9.82 mSv. The total activity concentrations of ^{232}Th and ^{238}U in the soil ranged from 69 to 6581 and from 49 to 130 Bq kg^{-1} , respectively. Elevated concentrations were also identified for the metals, arsenic (As), lead (Pb), chromium (Cr) and zinc (Zn), in the vicinity of the former mining sites. Both radionuclide and heavy metal concentrations suggested leaching, mobilization and distribution from rocks into the soil. Correlation analysis revealed different origins for ^{232}Th and ^{238}U , but same or similar origins for ^{232}Th and the metals As, Cr, Zn, nickel (Ni) and cadmium (Cd). The results from *in situ* size fractionation of water demonstrated that radionuclides were predominately present as colloids and low molecular mass (LMM) species, and potentially mobile and available for uptake by aquatic organisms of the Norsjø Lake. Transfer factors, calculated for different plant species, showed the highest radionuclide accumulation in moss and lichens. Uptake in trees was, as expected, lower. Relationship analysis of ^{232}Th and ^{238}U concentrations in moss and soil samples showed a significant positive linear correlation.

4.2 Paper II – Mobility analysis of radionuclides and trace elements in the Fen Complex soil

Investigation of radionuclides (^{232}Th and ^{238}U) and trace elements (Cr, As and Pb) in soil from two legacy NORM (former mining sites) and one undisturbed naturally ^{232}Th -rich site was conducted as a part of the ongoing environmental impact assessment in the Fen Complex area (Norway). The major objectives were to determine the radionuclides and trace elements distribution and mobility in soils as well as to analyze possible differences between legacy NORM and surrounding undisturbed naturally ^{232}Th -rich soils. Inhomogeneous soil distribution of radionuclides and trace elements was observed for each of the investigated sites. Concentration of ^{232}Th was high (up to 1685 mg/kg, i.e., ~7000 Bq/kg) and exceeded the screening value for the radioactive waste material in Norway (1 Bq/g). Based on the sequential extraction results, the majority of ^{232}Th and trace elements were rather inert, irreversibly bound to soil. Uranium was found to be potentially more mobile, as it was associated with pH-sensitive soil phases, redox-sensitive amorphous soil phases and soil organic compounds. Comparison of the sequential extraction data sets from the three investigated sites revealed increased mobility of all analyzed elements at the legacy NORM sites in comparison to the undisturbed ^{232}Th -rich site. Similarly, the distribution coefficients K_d (^{232}Th) and K_d (^{238}U) suggested elevated dissolution, mobility and transportation at the legacy NORM sites, especially at the decommissioned Nb-mining site (346 and 100 L/kg for ^{232}Th and ^{238}U , respectively), while the higher sorption of radionuclides was demonstrated at the undisturbed ^{232}Th -rich site (10672 and 506 L/kg for ^{232}Th and ^{238}U , respectively). In general, although the concentration ranges of radionuclides and trace elements were similarly wide both at the legacy NORM and at the undisturbed ^{232}Th -rich sites, the results of soil sequential extractions together with K_d values supported the expected differences between sites as the consequences of previous mining operations. Hence, mobility and possible elevated bioavailability at the legacy NORM site could be expected and further risk assessment should take this into account when decisions about the possible intervention measures are made.

4.3 Paper III – Biota assessment: Radionuclide and trace element transfer from soil to free-living earthworm species

Transfer of radionuclides (^{232}Th and ^{238}U) and associated metals (As, Cd, Pb and Cr) from soil to free-living earthworm species was investigated in a thorium (^{232}Th) rich area in Norway. Sampling took place within former mining sites representing the technologically enhanced naturally occurring radioactive materials (TENORM), at an undisturbed site with a unique ^{232}Th -rich bedrock which was assumed to represent naturally occurring radioactive materials (NORM), and at site outside the ^{232}Th -rich area that was taken as the Reference background site. Soil analyses revealed increased levels of the investigated elements at NORM and TENORM sites. Based on sequential extraction, uranium (^{238}U) and cadmium (Cd) were quite mobile, while the other elements were strongly associated with mineral components of the soil.

Four investigated earthworm species (*Aporrectodea caliginosa*, *Aporrectodea rosea*, *Dendrodrilus rubidus* and *Lumbricus rubellus*) exhibited large individual variability in the accumulation of radionuclides and metals. The differences in uptake by epigeic and endogeic species, as well as differences within same species from NORM, TENORM and Reference sites were also observed. Based on the total concentrations in soil, the transfer factors (TF) ranged from 0.03 to 0.08 and from 0.09 to 0.25, for ^{232}Th and ^{238}U , respectively. The TFs for lead (Pb), chromium (Cr) and arsenic (As) were low (less than 0.5), while the TFs for Cd were higher (about 10).

Using the ERICA tool, the estimated radiation exposure dose rate of the earthworms ranged from 2.2 to 3.9 $\mu\text{Gy/h}$. The radiological risk for the investigated earthworms was low (0.28). The obtained results demonstrated that free-living earthworm species can survive in soil containing elevated ^{232}Th and ^{238}U , as well As, Cd, Pb and Cr levels, although certain amounts of radionuclides were accumulated in their bodies. The present investigation contributes to a general better understanding of complex soil-to-biota transfer processes of radionuclides and metals, and to the assessment of risk for non-human species in the ecosystem with multiple contaminants.

4.4 Paper IV – Biota assessment: Radionuclide transfer from soil to wild plant species

A study of transfer of the naturally occurring radionuclides thorium (^{232}Th) and uranium (^{238}U) from soil to wild plant species was performed as a part of an environmental impact assessment in an area with legacy and naturally enhanced radioactivity in Norway. To investigate possible differences in uptake, three sites were chosen for the study: a former iron (Fe) mining site as a legacy NORM site, undisturbed ^{232}Th -rich as a NORM site, and a site outside the enhanced radiation area that served as a Reference site. Tissue concentrations, transfer factors and radiological exposure dose rates were determined in nine wild plant species, including vascular and non-vascular plants, deciduous and coniferous trees. High soil activity concentrations of ^{232}Th , up to 5069 Bq/kg and 16516 Bq/kg, were measured in samples from the legacy NORM and undisturbed ^{232}Th -rich sites, respectively, while soil from the Reference site exhibited a statistically lower activity concentration of ^{232}Th (up to 2270 Bq/kg). Soil activity concentrations of ^{238}U were lower than ^{232}Th and in terms of the investigated sites, they were statistically indistinguishable, with maximal concentrations 156, 859 and 109 Bq/kg at the legacy NORM, undisturbed ^{232}Th -rich and Reference sites, respectively. Despite the increased total soil radionuclide levels, the mobile soil fractions were low (up to 157 Bq/kg of ^{232}Th and 27 Bq/kg of ^{238}U). Aboveground plant tissue activity concentrations of ^{232}Th ranged from non-detectable (tree needles and leaves) to 49 Bq/kg (grass); ^{238}U ranged from 0.03 (tree needles and leaves) to 5 Bq/kg (moss). No differences in the uptake of the radionuclide ^{232}Th by plants at the three investigated sites were demonstrated; uptake of the radionuclide ^{238}U was significantly lower at the Reference site. As expected, plants roots served as a natural translocation barrier, as the concentration of radionuclides in the analyzed samples were up to 25-fold higher than in the corresponding aboveground plants. The transfer factors for ^{232}Th and ^{238}U in aboveground plant parts ranged from $4 \cdot 10^{-5}$ to $1 \cdot 10^{-2}$ and $1 \cdot 10^{-4}$ to $4 \cdot 10^{-2}$, respectively. Radiological exposure dose rates, calculated using the ERICA software programme, were from 5.8 to 22.6 $\mu\text{Gy/h}$. Although the dose rate calculated for lichen and bryophytes (22.6 $\mu\text{Gy/h}$) was above the adopted screening level of 10 $\mu\text{Gy/h}$, this value is still below the dose rates expected to cause impacts to plants. Based on obtained results, no significant uptake of ^{232}Th and ^{238}U was demonstrated in plants, as initially expected due to the high soil radionuclide levels.

4.5 Paper V: Estimation of possible human outdoor exposure doses in the Fen Complex area

The present study was performed in the Fen Complex, a Norwegian area rich in naturally occurring radionuclides, especially thorium (^{232}Th). Measurement of radioactivity levels was conducted at the decommissioned iron (Fe) and niobium (Nb) mining sites (TENORM), as well as at the undisturbed wooded sites (NORM), all open for free public access. The soil activity concentrations of ^{232}Th (3280 – 8395 Bq kg^{-1}) were significantly higher than the world and the Norwegian average values and exceeded the Norwegian screening level (1 Bq g^{-1}) for radioactive waste; radium (^{226}Ra) was present at slightly elevated levels (89 – 171 Bq kg^{-1}). Terrestrial gamma dose rates were also elevated, ranging from 2.6 to 4.4 $\mu\text{Gy h}^{-1}$. Based on long-term surveys, the air concentrations of thoron (^{220}Rn) and radon (^{222}Rn) were 1786 and 82 Bq m^{-3} , respectively. Seasonal variations in the outdoor gamma dose rates and Rn concentrations were confirmed. Correlation analyses showed a linear relationship between air radiation levels and the abundance of ^{232}Th in soil. The annual outdoor effective radiation doses for humans (occupancy 5 h day^{-1}) were estimated to be in the range of 3.0 – 7.7 mSv; this is comparable to or higher than the total average (summarized indoor and outdoor) exposure dose for the Norwegian population (2.9 mSv year^{-1}). On the basis of all of the obtained results, it was concluded that this area in Norway should be considered as an enhanced natural radiation area (ENRA).

5 Results and discussion

The overall results are discussed with respect to data quality, the Fen Complex area contamination with radionuclides and trace elements, biota exposure to multiple stressors and human outdoor radiation exposure.

5.1 Quality of the data

To ensure good data quality, the prescribed protocols were followed and necessary care was made during sampling, storage, preparation and measurements. The quality of the sample preparations and measurements were controlled as follows:

- The acceptable accuracy and precision were established by repeatedly measuring standard reference materials throughout whole current work. Certified standard reference materials NCS DC 73324, NCS DC 73325, GBW 07401 for soil, NCS DC 73348 for biota were used. The percentage of error lower than 10 % confirmed good data quality. Examples of accuracy control are presented in Table 5. High standard deviations in soil samples reflect the significant inhomogeneous spatial distribution of radionuclides in the Fen area soil rather than variations in the analysis. A precision test, performed on 6 parallel soil samples that were digested and analyzed by ICP-MS in the same way as other soil samples, had a relative standard deviation (% RSD) for all of the investigated elements below 8 %. According to literature (Brown, 1998), RSD values below 30 % can be considered as an indication of good to fairly good precision in a data set.
- Nine soil samples from the former mining site Søve were analyzed by ICP-MS in an outside commercial chemical laboratory. All reported values for radionuclides and trace elements were within a 15 % difference as compared to the results from this thesis for the same soil samples.
- For the Msc project at the Isotope Laboratory (Valle, 2012), soil and plant samples were collected in the Fen and analyzed with respect to ^{232}Th , ^{238}U and trace elements by another operator and on another instrument ICP-MS (8800 Triple Quadrupole ICP-MS Agilent Technologies). Although samples were taken at same sites, but not exactly at the same sampling points, the obtained ranges for the analyzed elements were comparable, and student's t-test for the two data sets did not show statistically significant differences.

Table 5. Concentrations of radionuclides and trace elements (mg/kg) in standard reference materials

Reference material	Element	²³² Th	²³⁸ U	As	Cr	Cd	Pb	Cu	Zn
NCS DC 73324 soil	Measured	25.1 ± 0.6	7.1 ± 0.3	228 ± 27	69 ± 8	0.11 ± 0.02	344 ± 27	382 ± 22	102 ± 4
	Expected	23 ± 2	6.7 ± 0.7	220 ± 14	75 ± 6	0.13 ± 0.03	314 ± 13	390 ± 14	96 ± 6
NCS DC 73325 soil	Measured	7.9 ± 0.3	2.0 ± 0.5	4.9 ± 0.6	397 ± 16	0.09 ± 0.02	13 ± 1	88 ± 2	119 ± 25
	Expected	9.1 ± 0.7	2.2 ± 0.4	4.8 ± 1.3	410 ± 23	0.08 ± 0.02	14 ± 3	97 ± 6	142 ± 11
NCS DC 73348 plant	Measured	0.37 ± 0.04	0.10 ± 0.02	1.03 ± 0.08	2.6 ± 0.5	0.16 ± 0.05	7.0 ± 0.4	4.9 ± 0.8	21.3 ± 1.9
	Expected	0.37 ± 0.02	0.11*	0.95 ± 0.12	2.3 ± 0.3	0.14 ± 0.06	7.1 ± 1.1	5.2 ± 0.5	20.6 ± 2.2

* Value for standard reference material given as single value without standard deviation in proper reference sheet.

- Parallel measurements of ten soil samples from the site Bolladalen were done by ICP-MS and gamma spectrometry. The systematically higher values obtained by gamma spectrometry were expected since non-destructive gamma spectrometry measures total radionuclide concentrations in the soil, while the ICP-MS measures radionuclide in the digested soil fractions. Correlations between results obtained by two different methods are given in Figure 15. Differences in measured concentrations between the two data sets ranged between 6.4 – 21.3 % and 11.2 – 23.7 %, for ^{232}Th and ^{238}U , respectively. In ^{232}Th parallel measurements, the demonstrated differences were between 10.0 and 20.0 % for 7 of 10 measured samples, <10 % for 2 samples and 21 % for 1 sample. In ^{238}U parallel measurements, differences were between 10 – 20 % in 9 of 10 measured samples. However, the obtained results demonstrated a high degree of overlap of ranges, showing that in some cases lower difference existed in values obtained by two methods for the same soil sample than in values obtained by same method but for the two different samples from the same site.

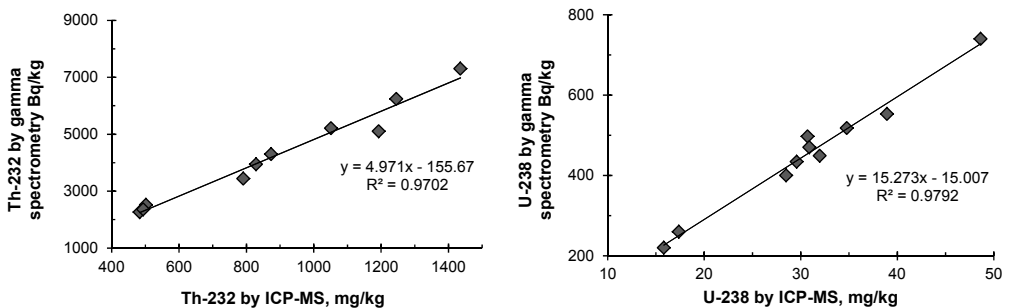


Figure 15. Comparison of ^{238}U and ^{232}Th concentrations measured by gamma spectrometry and ICP-MS.

5.2 Contamination status of the Fen Complex environment

Based on the study results, according to the ICRP (2007) classification, the Fen Complex area should be characterized as an "existing exposure situation", with both ^{232}Th naturally enriched bedrock and legacy NORM waste giving rise to elevated radiation in the area.

5.2.1 Outdoor terrestrial gamma dose rates and ^{220}Rn , ^{222}Rn concentrations

Recorded terrestrial gamma dose rates in the air ranged from 0.07 to 9.24 $\mu\text{Gy/h}$ in residential and wooded (former mining) areas, respectively. The data were in good agreement with data already published for the same area (Dahlgren, 1983; Heincke *et*

al., 2008; IFE, 2006; NGI-UMB, 2010). Summary of the results from Papers I and V showed that elevated gamma dose rates at both legacy NORM (former mining sites) and undisturbed ^{232}Th -rich sites were up to 120-fold higher than the average Norwegian value of $0.073 \mu\text{Gy/h}$ (UNSCEAR, 2000, 2008a). Although a statistically significant difference ($p = 0.005$) was observed between certain sites (Paper V), maximal recordings were seen at both the former Nb mining site Søve, and the undisturbed ^{232}Th -rich sites Bolladalen and Rullekoll, suggesting that both the presence of radioactive mining waste and radionuclide-rich minerals were responsible for the high gamma radiation dose rates in the air. Seasonal variations did not exhibit any common patterns for the sites, although maximal readings were obtained in early September (5th and 6th September 2009) when the weather conditions were stable, while minimal readings were measured in late November (29th November 2009) in weather conditions with rain, wind and snow.

The UNSCEAR (2006) Report suggests that typical outdoor levels of ^{220}Rn and ^{222}Rn were each 10 Bq/m^3 . However, long term surveys provided ^{222}Rn concentrations as high as 100 Bq/m^3 at certain sites with high ^{222}Rn exhalation over large areas. Because of the short half-life of ^{220}Rn , considerable variability has been observed at close measuring points, indicating large differences in exhalation rate; however, usually with the outdoor values below 15 Bq/m^3 (UNSCEAR, 2006). Based on long-term surveys presented in detail in Paper V, a wide range of ^{220}Rn levels in the air was measured in the Fen Complex area, ranging from 7 Bq/m^3 (recorded at former mining site Søve) to about 5000 Bq/m^3 (recorded at former mining site Fen). The few publications examining outdoor ^{220}Rn levels, although conducted in the areas with no elevated primordial radionuclides in the bedrock/soil, present much lower outdoor values (Doi and Kobayashi, 1994; Porstendörfer et al., 1991). The concentrations of ^{222}Rn in the air were substantially lower than concentrations of ^{220}Rn , ranging from 4 to 210 Bq/m^3 .

The statistically lower concentrations ($p = 0.0005$) of ^{220}Rn at the former mining site Søve (though only summer survey data allowed statistical analysis) were attributed to the presence of a protective cover layer at the measuring points. It corresponded to results from soil analysis, showing significantly lower ^{232}Th at the sampling points where the remediation cover layers were intact. However, it must be highlighted that higher values of ^{220}Rn gas were expected to be found at least at some detection points since a higher (not statistically different from other sites) ^{232}Th concentration in the soil was demonstrated at points where mixing of ^{232}Th -enriched mining waste occurred (Paper II). The discrepancy between the expected and measured values is attributed to a certain degree to the positions of the detectors, but also to the short

^{220}Rn half-life and different soil parameters that could affect the exhalation rate. At other investigated sites, including both legacy NORM and undisturbed ^{232}Th -rich sites, comparable ranges, displaying high variations within sites, were detected. High measured ^{220}Rn levels were there assigned to increased, but inhomogeneous ^{232}Th activity concentrations in the bedrock. The statistically lower values for ^{220}Rn ($p = 0.0039$) and ^{222}Rn ($p = 0.0054$) air concentrations obtained in the autumn compared to those in summer, contrasted with the typical behavior of these radionuclides (Chan *et al.*, 2010; Oikawa *et al.*, 2003). However, results similar to here presented data have been seen at some sites worldwide previously (Arnold *et al.*, 2009; Baciú, 2005). For a better understanding of seasonal differences, a detailed knowledge of the precipitation levels, movements of air masses, temperature and atmospheric pressure fluctuations, the presence of a cloud cover, air humidity and soil moisture is needed. The prevalence of ^{220}Rn in comparison to ^{222}Rn in the air, confirmed statistically ($p = 0.0005$), was expected since higher soil/bedrock concentrations of mother ^{232}Th were also measured in comparison to ^{238}U .

5.2.2 Radionuclides and trace elements in soil

Results of total soil radionuclides and trace element measurements, as well as analysis of the distribution and potential mobility are given in detail in Papers I – IV. Concentrations of ^{232}Th and ^{238}U in the Fen Complex area were ranged from 60 to 16516 and from 36 to 2161 Bq/kg, respectively. With respect to ^{232}Th and ^{238}U , the reference site Torsnes, outside ENRA, had a statistically lower (for 1 to 2 orders of magnitude) soil radionuclide concentration ($p = 0.026$). The obtained results are in good agreement with the results presented in the NGI-UMB (2010) report for the former mining site Søve and the area around it. Screening of soil samples from the Fen Complex area against the set of values for classifying the material as radioactive waste in Norway (Pollution Control Act, 2010) showed contamination with regard to ^{232}Th and ^{228}Ra . Uranium activity concentrations were generally below the screening level of 1 Bq/g, with the exception at several sampling points at the former mining site Søve. To estimate the contamination status of the soil, additional radionuclides, progenies ^{228}Ra , ^{226}Ra , and ^{210}Po , were analyzed and found to be in ranges 134 – 7300, 77 – 250, and 38 – 76 Bq/kg, respectively. Taking all of the analyzed radionuclides into the consideration, and their maximal values, the risk quotient (RQ) could reach 27, clearly above the screening value of 1 (section 3.1.5). Trace elements (As, Cd, Cr, Pb) were only slightly enhanced at certain sampling points, and were just above the norm values for soil in Norway (SFT, 2009).

Considerable variation in the concentrations of ^{232}Th and ^{238}U was observed both between and within sites, indicating an intensive spatial inhomogeneous distribution. Heterogeneities on a macro scale were demonstrated by autoradiography, even within the same soil samples. Further characterization of isolated radioactive particles is currently ongoing (Figure 16). No differences were demonstrated in total radionuclide ^{232}Th and ^{238}U soil concentrations between legacy NORM and undisturbed ^{232}Th -rich sites (the exception are the concentrations measured at the former mining site Søve, presented in Paper I, which were considerably lower than at the other sites, due to the fact that only the cover sand layer poor in radionuclides was sampled in the first fieldwork and included in the soil analysis). However, for a better understanding of the identified, high and equally variable radionuclide levels at the investigated sites, some sites characteristics should be taken into the consideration. Bedrock of the former mining site Fen (legacy NORM) and sites Bolladalen, Gruvehaugen and Rullekoll (undisturbed ^{232}Th -rich sites) consists of rødbergite and rauhaugite rock type which were shown to be rich in ^{232}Th (Paper I; Sundal and Strand, (2004)). Consequently, high concentrations of ^{232}Th in the soil were expected and demonstrated at all of the sites. However, based on previous experiences with radionuclide enrichment of the environment at mining and tailing sites (Lind *et al.*, 2013; Salbu *et al.*, 2013; Skipperud *et al.*, 2013a, b), it was further expected that the former mining site Fen would exhibit elevated soil radionuclide concentrations in comparison to undisturbed sites, due to soil and rocks disturbances caused by human mining activities. Nevertheless, with respect to total soil concentrations, the enrichment of soil at this legacy NORM site was not observed since all of the above mentioned sites displayed similar ranges of soil ^{232}Th and ^{238}U concentrations. A similar consideration of the former mining site Søve resulted in an opposite conclusion. The bedrock at the site Søve consists mainly of rock søvite that was demonstrated to have lower ^{232}Th activity concentrations (Paper I; Sundal and Strand, (2004)). Taking again rock concentrations as a starting point, statistically lower ^{232}Th concentrations could be expected in soil if only natural weathering of rock søvite occurred at this site. As in case of former mining site Fen, the question of soil radionuclide enrichment due to different mining operations in the past was raised. In that sense, higher than expected and comparable to other sites soil ^{232}Th concentrations at Søve (Papers II, V) might be the results of enrichment from mining in the past.

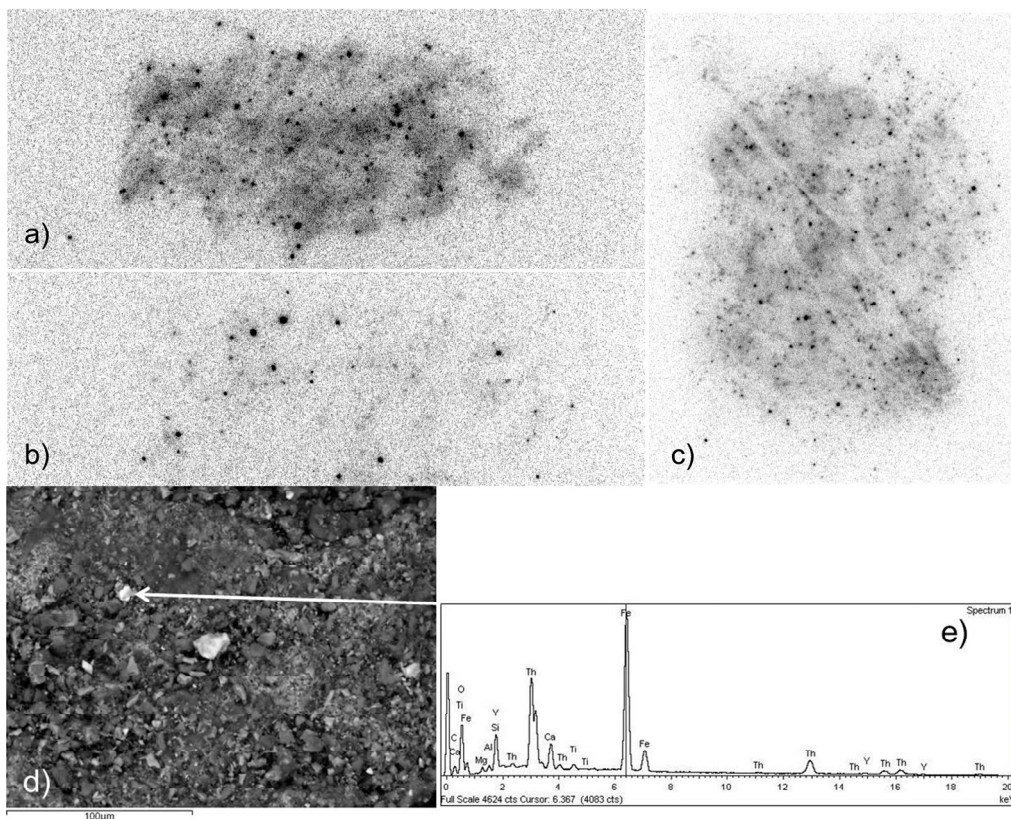


Figure 16. Heterogeneous distribution of radionuclides demonstrated by digital autoradiograms of soil samples from
a) former mining site Fen **b)** former mining site Søve **c)** undisturbed ^{232}Th rich site Rullekoll
d) ESEM image of rock rødbergite **e)** EDX showing Th and Y (Lind, pers. comm., 2014)

The concept of a total concentration of radionuclides and trace elements in soil provides limited insight into the actual behavior of elements in the soil, and is thus of limited use. Both radionuclides and trace elements can be associated with a number of different soil phases that determine their mobility, behavior, bioavailability and toxicity to biota (Alloway, 1995; Anju and Banerjee, 2011; Garcia *et al.*, 2005; Guo *et al.*, 2007; 2008; Lambrechts *et al.*, 2011). Sequential extractions of soil were conducted to obtain the information on radionuclides and trace elements binding mechanisms and their distribution in the soil (Paper II). Based on the obtained results, the majority of ^{232}Th (77 – 94 %) and As (70 – 89 %) was rather inert, irreversibly bound in soil. Uranium, Cr and Pb were found to be potentially more mobile as up to 62, 45 and 49 %, respectively, were associated with pH-sensitive soil phases, redox-sensitive amorphous soil phases and soil organic compounds. Principal component

analysis (PCA), performed on the complete sequential extraction data set indicated a strong gradient, i.e., the concentrations of elements generally increased with every sequential extraction step (Figure 17A and B). Similar behaviors of ^{232}Th and Cr and of ^{238}U and Pb and As towards soil fractions was suggested by their positive correlation along the first canonical axis. By comparing sequential extraction results between sites, stronger and irreversible binding of all elements was observed in soils from the undisturbed ^{232}Th -rich site Rullekoll than in soils from the legacy NORM sites.

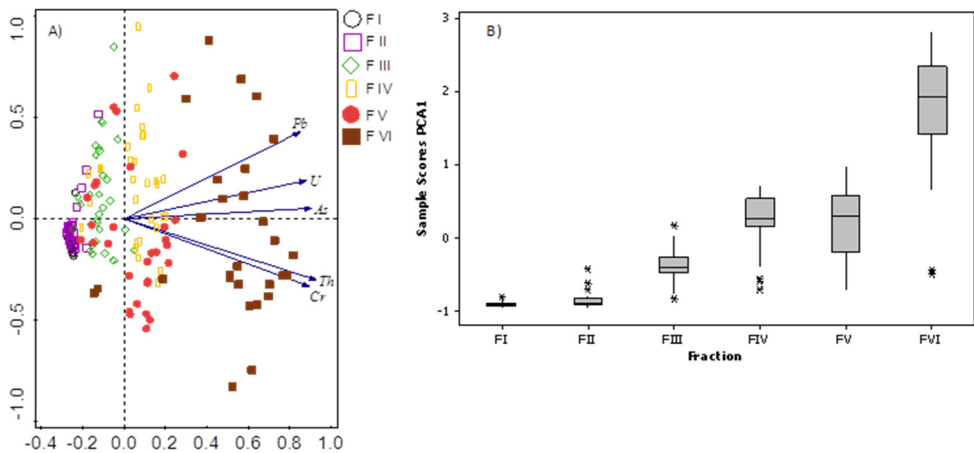


Figure 17. A) Bi-plot displaying the results obtained from PCA using all data from sequential extraction FI-FVI. **B)** Sample scores grouped according to their respective sequential extraction step obtained from PCA axis 1. Increasing sample score indicates increasing element concentrations.

Distribution coefficients were determined for legacy NORM and undisturbed ^{232}Th -rich soils in order to study the distribution of elements between the soil solid and aqueous phase, and hence, to estimate their potential mobility in soil (Paper II). Geometric mean values for distribution coefficients K_d (^{232}Th) and K_d (^{238}U) ranged within 346 – 10672 and 100 – 506 L/kg, respectively. According to Vandenhove *et al.*, (2009b), pH, soil grain size distribution and organic content could have especially important roles in various processes determining sorption and dissolution in soil. Thus, the current study K_d s were grouped, compared and subsequently found to be in accordance with the existing literature with respect to basic physical and chemical characteristics (IAEA, 1994; 2010; Sheppard *et al.*, 2006; Vandenhove *et al.*, 2009b). Trace elements exhibited lower K_d s than radionuclides in a pooled range 6 – 762 L/kg, with the following order of decreasing: K_d (Pb) > K_d (As) > K_d (Cr). Analysis of

K_{ds} with respect to different sites suggested elevated dissolution, transport and mobility at legacy NORM sites, especially at the decommissioned Nb mining site Søve, while higher sorption of radionuclides was demonstrated at the undisturbed ^{232}Th -rich site Rullekoll.

5.2.3 Radionuclides and trace elements in water

The largest water source in the Fen Complex area is the Lake Nordsjø. Three sites, included in the water investigation (the former mining sites Fen and Søve and the Reference site outside the area Torsnes), were directly positioned or in close vicinity of this lake. Since the large quantities (assumed, not precise data) of waste materials from different mining operations at both Nb and Fe mining sites were deposited near water, the mobilization and leaching of radionuclides and trace elements into water were hypothesized. Two additional sampling points at the Lake Nordsjø within the residential Fen area and one sampling point at a small stream which flows through the former mining site Fen were included in the investigation.

Results of total and fractionated water analysis (Paper I) showed very low concentrations of both radionuclides and trace elements as compared to the results from similar assessment studies at abandoned mining sites in Central Asia (Lind *et al.*, 2013; Salbu *et al.*, 2013; Skipperud *et al.*, 2013). Based on all analyzed samples, no contamination of the lake nor of the small stream was observed. Measurements of *in-situ* size fractionated water demonstrated that the ^{232}Th was present as particulate (HMM species) and colloidal matter, while the ^{238}U was mainly found as a colloidal species (Figure 18). Therefore, although they were present at low concentrations, both radionuclides could be subjected to colloidal transport.

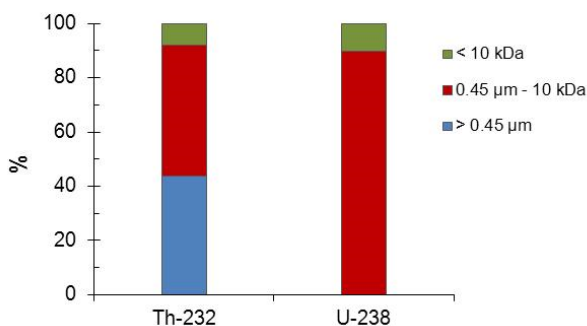


Figure 18. Results of size fractionation of ^{232}Th and ^{238}U in water samples from the Lake Nordsjø.

5.2.4 Radionuclides and trace elements in biota

Different earthworms and plant species were chosen for the biota analyses (Paper III and IV, respectively). Radionuclide and trace element concentrations in the aboveground plant parts and roots are given in Figure 19. The activity concentrations of ^{232}Th in the analyzed aboveground plant parts ranged from undetectable ($< \text{LOD}$) to 49 Bq/kg and reflected the variety of collected biota (vascular, non-vascular plants, flowers, deciduous and coniferous trees, etc.) rather than the difference in radionuclide concentrations in the soil at different sampling sites. Maximal levels were detected in dandelion (32 Bq/kg), grass (49 Bq/kg) and moss (22 Bq/kg). The concentrations of ^{232}Th in plants, although higher, were still comparable to those from other similar studies (Belivermiş and Çotuk, 2010; Dowdall *et al.*, 2005; Dragovic *et al.*, 2010; Martinez-Aguirre *et al.*, 1997). The activity concentrations of ^{238}U were several times lower than of ^{232}Th , ranging from 0.031 to 5 Bq/kg. At all investigated sites, moss had a statistically higher ^{238}U concentration than other plant species, while trees (leaves and needles) had minimal ^{238}U concentrations ($p = 0.002$). As observed for ^{232}Th , the concentrations of ^{238}U in plants were also in good agreement with the previously published ranges measured in the same or similar species in different regions worldwide (Belivermiş and Çotuk, 2010; Dowdall *et al.*, 2005; Dragovic *et al.*, 2010). In addition, berries, leaves and stem samples of raspberries, lingonberries, and blueberries were analyzed (Valle, 2012). Although slightly higher concentrations ^{232}Th , ^{238}U and ^{210}Po were demonstrated for plants and stems in comparison to berries, those values were comparable to values obtained herein for trees leaves. It should be highlighted that these concentrations are low and comparable to the levels observed in areas with no enhanced NOR. The concentration of ^{210}Po in mushroom samples reached 5 Bq/kg, while ^{232}Th and ^{238}U were below 1 Bq/kg.

Trace element concentrations in plants varied also with plant species, but were found to be generally low, suggesting no or insignificant uptake from the soil (Figure 19). Overall, the trend of highest trace element concentrations was seen in moss. On the contrary, the lowest levels of investigated elements were detected in spruce needles. Although the highest concentrations of Cr and Pb detected in moss were above the norm plant values reported by Kabata-Pendias (2010), results similar to current study results have been published previously (Bublinec, 1994; Maňkowska, 1996; Markert, 1992). The concentrations of As and Cd in plant tissues were comparable to normal background levels (Allen, 1989; Bowen, 1979; Kabata-Pendias, 2010; Zu *et al.*, 2004).

The analysis of roots in selected plants was conducted in order to assess the translocation of radionuclides and trace elements from the soil. As expected, the

concentrations in roots were higher, up to 25-fold higher than in aboveground plant parts. This result is in agreement with previously published results showing that roots act as a natural barrier for radionuclide and trace metal translocation to other plant parts (Chang *et al.*, 2005; Shahandeh and Hossner, 2002; Shtangeeva and Ayrault, 2004; Shtangeeva, 2010).

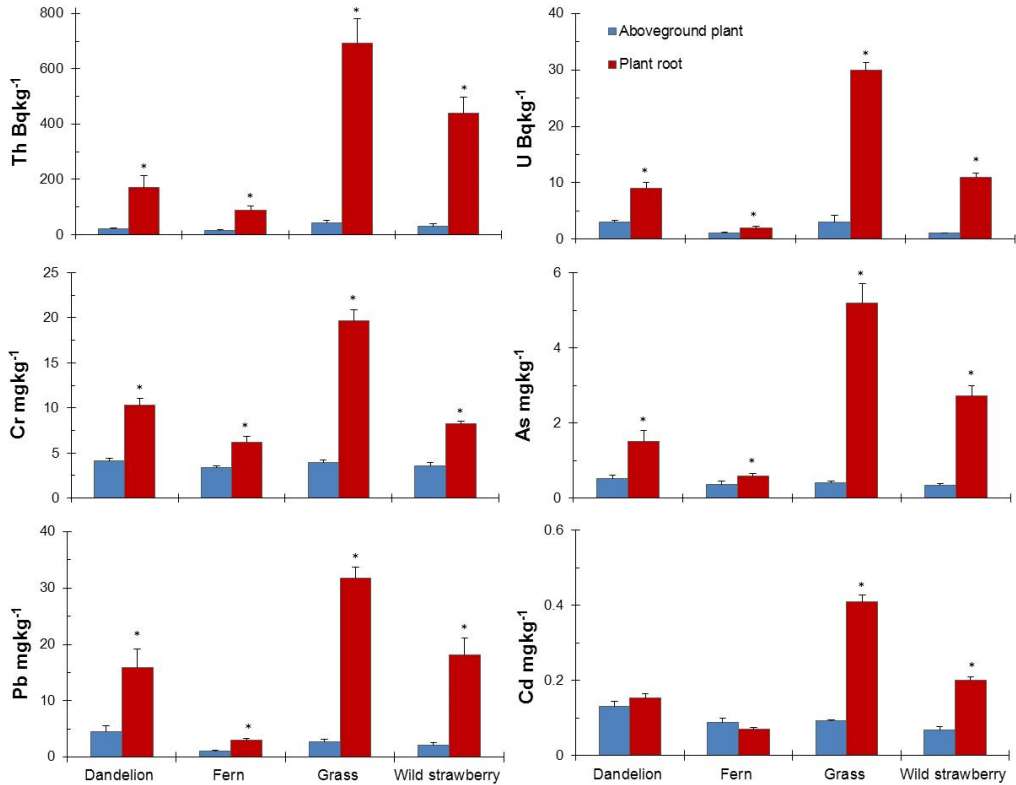


Figure 19. Concentrations of radionuclides and trace elements in aboveground plant parts and roots. Significant differences in concentrations in roots and aboveground parts are marked with *. Note the different units for radionuclides and trace elements as well as different axis scales.

Free-living epigeic and endogeic earthworm species were sampled and their tissues were analyzed for the presence of ^{232}Th , ^{238}U and trace elements. The description of the analytical procedures and detailed results of species-specific accumulation, environmental pools favoring uptake, differences between endogeic and epigeic species, as well between sites, are presented in Paper III. The concentration of ^{232}Th in earthworms varied from 45 to 231 Bq/kg. Species-specific accumulation was observed, but no clear patterns with regard to epigeic and endogeic species were

identified. Still, with regard to endogeic species, the highest concentrations of ^{232}Th were demonstrated in earthworms collected at legacy NORM sites. This was expected considering the manner of feeding of endogeic earthworms which involves the processing of large amounts of soil. As a result of previous mining activities, the disturbances in soil profiles could also result in increased availability of radionuclide-rich particles. Autoradiography of earthworms showed the radioactive soil inside the earthworms (Figure 20). The ^{232}Th concentrations in earthworms were up to 80-fold higher than the ^{232}Th concentration in those living in the average background soil, according to a study of natural background radioactivity dose rates of wildlife in the UK (Beresford *et al.*, 2008b). The concentrations of ^{238}U in earthworms were considerably lower than ^{232}Th , ranging between 7 and 12 Bq/kg.

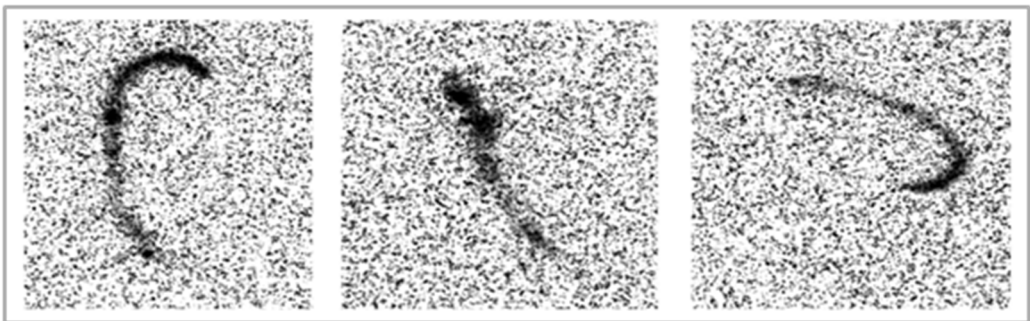


Figure 20. Autoradiography of earthworms collected in the Fen Complex area.

5.3 Exposure of biota

To characterize the impact of terrestrial radionuclides and associated trace elements in the Fen Complex area, the following parameters were investigated and discussed:

- Biota tissue concentrations (given in the previous section)
- Transfer factors
- Radiological dose rates

From the radiological protection view, the most significant processes regarding terrestrial decay chains (^{232}Th and ^{238}U) are mobilization in the environment of alpha emitters, ^{232}Th , ^{238}U , and in particular the progenies ^{226}Ra and ^{228}Ra , ^{210}Po , ^{210}Pb , exhalation of ^{220}Rn and ^{222}Rn , and gamma emissions from various progenies throughout the decay chains.

5.3.1 Transfer factors

Within the scope of the present thesis, transfer factors were calculated only for radionuclides ^{232}Th and ^{238}U , since very low trace element concentrations were demonstrated in plant tissue (Paper IV). Pooled TFs were log-normally distributed, suggesting differences in uptake in different plant species. The geometric means of TFs (^{232}Th) and TFs (^{238}U) in plants ranged from $4 \cdot 10^{-5}$ to $1 \cdot 10^{-2}$ and $1 \cdot 10^{-4}$ to $4 \cdot 10^{-2}$, respectively. Obtained values were in good agreement with the TF values published in TRS-364 (IAEA, 1994), TRS-472 (IAEA, 2010) and with those given by Vandenhove *et al.*, (2009a), although it should be mentioned that wide TF ranges were found both in the current work and in cited literature. No differences in uptake at former mining sites and undisturbed ^{232}Th -rich sites were demonstrated. Transfer to different plant parts differed significantly, with transfer to the roots being one order of magnitude higher than transfer to aboveground parts, on average. This was in accordance with findings obtained in similar studies (Roivainen *et al.*, 2011; Shahandeh and Hossner, 2002). More detailed analysis for plant species and investigated sites are given in Paper IV.

The earthworms TFs for ^{232}Th were comparable among different species (0.03 – 0.08) and the averaged TF was 0.06. No data on ^{232}Th transfer from soil to earthworms in natural conditions was found in the literature. The average TF for ^{238}U in all earthworms was 0.20 (ranging from 0.09 – 0.25, depending on the species). Similar levels of naturally occurring U uptake in earthworms have been published recently (Beresford *et al.*, 2008b; Giovanetti *et al.*, 2010; Yoshida *et al.*, 2005). The order of TFs for trace elements decreased as follows: $\text{Cd} > \text{As} > \text{Pb} > \text{Cr}$. Similar differences in TF orders of magnitude were found for earthworms in Norway (Holmstrup *et al.*, 2011). The present study showed, however, that TF values were generally one order of magnitude lower than those obtained by Ernst *et al.* (2008), suggesting low bioaccumulation.

5.3.2 Exposure dose rates for terrestrial organisms

In the initial stage of the present work, the ERICA calculations of the radiological dose rates were performed both for freshwater and terrestrial organisms in order to establish the possible magnitude of doses and to check whether the screening level was exceeded. The maximal measured soil activity concentrations of ^{232}Th , ^{228}Ra , ^{238}U , ^{226}Ra and ^{210}Po and water activity concentrations of ^{232}Th and ^{238}U were used together with default values for K_d and CR provided in the ERICA Tool. Default reference organisms were selected. The obtained dose rates per organism group

determined in the initial screening of freshwater and terrestrial ecosystem in the Fen Complex are shown in Figure 21.

For freshwater organisms, the dose rates were in the range from 0.04 to 3.69 $\mu\text{Gy/h}$, with maximal values estimated for vascular plants. No organisms with dose rates above the 10 $\mu\text{Gy/h}$ default screening level were observed. Thus, based on the findings from the present work and previously published data (Stranden, 1982), no risk for freshwater organisms in the Lake Norsjø was established so that no further investigation of the impact on freshwater organisms was conducted. However, several things that could influence the magnitude of the doses should be considered. Apart from ^{232}Th and ^{238}U , no other potential radionuclide-dose contributors were included in the dose calculation. Based solely on that fact, the dose underestimation is likely. However, due to the very low mother radionuclide concentrations, and considering the results of a previously published water survey showing insignificant ^{226}Ra and ^{228}Ra radionuclide concentrations in lake water (Stranden, 1982), the potential underestimate should not be important in terms of the environmental impact. On the other side, dose rates for the freshwater organisms, calculated with the site-specific transfer parameters (K_d , CR), would probably be lower than here estimated with the default K_d and CR used in the calculations (like demonstrated for terrestrial organisms in the Fen Complex).

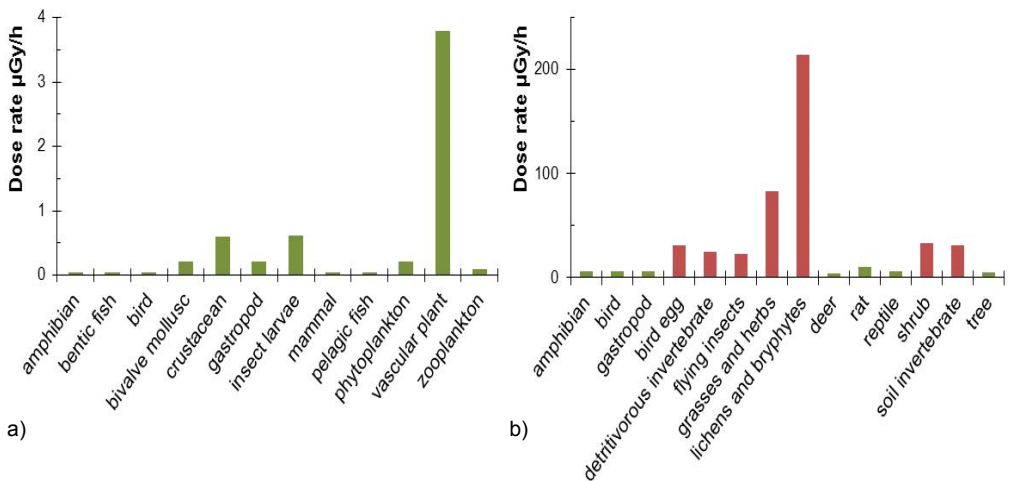


Figure 21. Dose rates per organism group for a) freshwater b) terrestrial ecosystem. Doses are calculated using the maximal sediment and soil concentrations and default CRs.

The dose rates for terrestrial organisms were from 3.7 to 214.0 $\mu\text{Gy/h}$, with the values for bird egg, detritivorous invertebrates, flying insects, grasses and herbs, lichen and

bryophytes, shrubs and soil invertebrates clearly above the screening value of 10 $\mu\text{Gy/h}$. With respect to the calculated dose rates, more detailed site-specific assessment continued on plants (nine different plant species including grasses and herbs, lichen and moss, berries, trees leaves and needles), earthworms and mushrooms. The dose rates (1.3 – 23.1 $\mu\text{Gy/h}$) calculated by the ERICA Tool using the site-specific data (soil and biota median radionuclide activity concentrations as well as calculated CRs), are presented in Figure 22. By comparing the doses obtained in the initial calculation (based only on soil activity concentrations) and doses obtained in the second site-specific calculation, changes in dose magnitude were seen in grasses and herbs (6 $\mu\text{Gy/h}$), lichen and bryophytes (23 $\mu\text{Gy/h}$), earthworms (13.1 $\mu\text{Gy/h}$), shrubs (1.3 $\mu\text{Gy/h}$) and trees (1.9 $\mu\text{Gy/h}$). Decrease in calculated dose rates were attributed to the lower soil activity concentrations of radionuclides and lower site-specific CRs used in the second calculation in comparison to those used in the initial screening. It should be mentioned that somewhat higher doses for earthworms were obtained when the actual measured concentrations of ^{226}Ra and ^{238}Ra were used in the site-specific calculation, rather than when secular equilibrium between mother-daughters was assumed (Paper III). More detailed discussion on radionuclides-dose contributors is provided in paper IV. Although dose rates for selected Fen Complex biota were higher than the background range for terrestrial plants and animals (0.01 – 0.7 $\mu\text{Gy/h}$) (UNSCEAR, 2008b), they do not imply elevated risk. In fact, the obtained doses lie below the screening levels of 40 $\mu\text{Gy/h}$ and 400 $\mu\text{Gy/h}$ for terrestrial animal and plants, respectively, adopted by international organizations as values below which no effects on population levels should be expected (IAEA, 1992; US DOE, 2002; UNSCEAR, 2008).

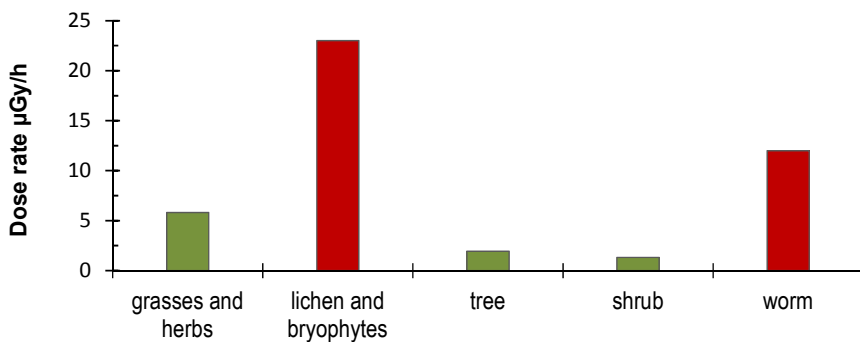


Figure 22. Dose rates for the selected terrestrial organisms from the Fen Complex area. Doses were calculated using site-specific data to represent a more realistic exposure scenario.

5.4 Human outdoor radiation exposure

The presence of NORM can lead to radiation doses that are not insignificant from the radiation protection point of view. In the last decade, international organizations (IAEA, ICRP, UNSCEAR) have addressed NORM problems as part of occupational exposure, and a number of international meetings with NORM as the main subject were organized, raising awareness of this problem. Nevertheless, there is still a backlog in the knowledge on NORM radiation problems, especially with practical issues related to legacy and industry NORM and legislation (Van der Steen and Van Weers, 2004). Individuals are exposed to radiation in different ways, referred to as exposure pathways. According to UNSCEAR (2008a), the most significant NORM exposure pathways are:

- External exposure to terrestrial gamma radiation
- Internal exposure through inhalation of ^{222}Rn (^{220}Rn)
- Internal exposure through ingestion (food and water)

Within the scope of the present work, outdoor exposure to terrestrial gamma radiation and exposure to outdoor ^{220}Rn and ^{222}Rn were investigated and presented in Paper V. A somewhat altered exposure scenario (than that given in Paper V) is provided in the thesis in order to obtain more realistic information on the possible magnitude of outdoor exposure doses. Doses from internal exposure resulting from food and water intake were not considered, since initial investigations of pasture and meadow land (soil, plants and water investigations) did not reveal higher radionuclide transfer from soil to plants and water. Similarly, a NRPA rapport (Stranden, 1982) showed insignificant doses from ingestion of food in the Fen Complex area.

External exposure to outdoor terrestrial gamma radiation in the Fen Complex was estimated as significant. Tabulated absorbed gamma dose rates and corresponding annual doses in some of the possible exposure scenarios are presented (Table 6). To predict outdoor radiation doses from terrestrial gamma radiation in a realistic way, several exposure scenarios with respect to specific sites were considered. For the former mining site Søve where an engineering firm with several employees is currently operating, an exposure time of 1 hour per day (250 days in a year) was assumed; the exposure time for people's recreational walking in the Fen Complex in the undisturbed woods (Bolladalen, Gruvehaugen and Rullekoll) and former mining site Fen (unrestricted area) was set to 1 h per week or 50 h per year. These exposure scenarios should be considered only as one option rather than a real situation. Consequently, doses should be considered just as estimates, since both under- and

overestimation is possible. For the residential Fen Complex area, the commonly accepted occupancy of 1752 h per year was used in the calculations (UNSCEAR, 2000, 2008a). Variable gamma dose rates were recorded at different sites in the Fen Complex. Consequently, annual outdoor gamma doses could vary and easily reach maximal values (0.16 – 1.49 mSv) (Table 6). Based on the presented data for maximal annual doses from outdoor exposure solely, concern should be directed to the former mining site Søve, where the annual dose could reach up to 1.49 mSv. At this site, the dose constraint for the general public of 1 mSv is received in 0.7 years (8.5 months) by exclusively outdoor exposure to terrestrial gamma radiation. People working at the site Søve are not radiation workers and doses should not be considered in terms of occupational exposure. For people walking in the undisturbed Fen woods, annual exposure doses are considerably lower (0.16 – 0.32 mSv) due to lower predicted occupancy. For the accurate estimation, habit data on Fen village people is required. However, this was outside the scope of the present thesis. In a very conservative exposure scenario with a higher exposure time of 700 h per year (in backyards and gardens of houses) for people living along the edge of the undisturbed ²³²Th-rich sites Bolladalen and Rulekoll, the annual doses due to outdoor exposure to terrestrial gamma radiation would increase to 2.33 and 2.72 mSv, respectively.

Table 6. Possible annual outdoor exposure doses for different exposure scenarios in the Fen Complex

Site	Exp. time hrs	Mean gamma μ Gy/h	Max gamma μ Gy/h	Mean gamma dose mSv	Max gamma dose mSv	Time to receive 1 mSv*
Legacy Søve	250	2.51	8.51	0.44	1.49	0.7 y
NORM Fen	50	2.55	4.46	0.09	0.16	6 y
Gruvehaugen	50	5.59	9.24	0.19	0.32	3 y
NORM Bolladalen	50	3.21	4.76	0.11	0.17	6 y
Rullekoll	50	2.66	5.56	0.09	0.19	5 y
Residential Fen village	1750	0.17	0.37	0.20	0.45	2 y
World average	1750	0.059	-	0.07	-	14 y

*Times after which a dose constraint of 1 mSv will be received, calculated with respect to maximal annual outdoor exposure doses.

The gamma radiation dose rates in the residential zone of the Fen Complex were lower than those in wooded and former mining zones and comparable to the rest of Norway. The annual exposure doses were up to 0.45 mSv when calculated with exposure time of 1750 h (UNSCEAR, 2008a). Still, these doses are 6-fold higher than

the average annual world doses from outdoor gamma radiation (0.07 mSv) (UNSCEAR, 2008a).

Inhalation of short-lived progenies of ^{222}Rn , and to a lesser degree of ^{220}Rn , and their deposition on the walls of the bronchial tree are major modes of lung exposure. Evaluation of the exposure doses must thus take into the account the actual activity concentrations of different alpha-emitting radionuclides in the air originating from both chains (UNSCEAR, 2000, 2006). However, in most cases, instead of individual progenies activities, ^{220}Rn and ^{222}Rn gas concentrations in the air were measured. This further implies indirect determination of the exposure dose, assuming certain values for the equilibrium factor (F) and the dose conversion coefficients (DCC) are correct. Although different values for F for outdoor ^{222}Rn ranging from 0.2 to 1.0 were determined, the UNSCEAR (2000, 2006) has adopted 0.6 as the appropriate value for the outdoor environment. Similarly, a range from 6 to 15 nSv (Bq h m⁻³)⁻¹ was reported for DCC, and a value of 9 nSv (Bq h m⁻³)⁻¹ has been recommended as appropriate for dose calculations (UNSCEAR, 2006). Still, uncertainties in exposure doses calculated using such parameters must be borne in mind. When it comes to estimating the doses from ^{220}Rn inhalation, the situation is even more complicated because of the very short half-life of ^{220}Rn ($T_{1/2} = 55.6$ sec) and a large space-time variation. Using the equilibrium factor F to calculate the exposure dose is thus not recommended, since large variations have been observed (reported worldwide F values from 0.003 to 0.1) (UNSCEAR, 2000, 2006). Instead, the Equilibrium Equivalent Concentrations (EEC) should be measured and included in the calculation. With respect to this, possible radiation doses for several outdoor exposure scenarios in the Fen Complex are given herein as sums of the exposure dose from terrestrial gamma and exposure dose from ^{222}Rn in the air (Table 7). The somewhat different (more conservative) exposure scenarios and possible contribution of ^{220}Rn (explained in details in Paper V) was also considered, although a large uncertainty is associated with it.

The total annual outdoor exposure doses at the Fen Complex sites ranged from 0.10 to 0.47 mSv for defined exposure times (50 and 250 hrs). Values at undisturbed ^{232}Th -rich sites (Bolladalen, Gruvehaugen, Rullekoll) (0.10 – 0.21 mSv/y) are comparable to the annual exposure dose (0.16 mSv) obtained by summarizing the outdoor gamma radiation (0.07 mSv) and outdoor ^{222}Rn dose (0.095 mSv), given as world average values for typical outdoor exposure time of 1752 h (UNSCEAR, 2000), while the dose at the former mining site Søve (0.47 mSv) was higher. The difference in considered exposure times should be noted.

Table 7. Possible annual outdoor exposure doses at legacy and undisturbed NORM sites in the Fen Complex (geometric mean values of the measured parameters used in the equations)

Site	Exposure time hrs	Gamma dose rate $\mu\text{Gy/h}$	^{222}Rn conc. Bq/m^3	Gamma dose mSv	^{222}Rn dose mSv	Total dose mSv	^{220}Rn conc. Bq/m^3	^{220}Rn dose mSv*
Søve	250	2.51	21	0.44	0.03	0.47	37	0.04
Fen	50	2.55	32	0.09	0.009	0.10	818	0.16
Bolladalen	50	3.21	34	0.11	0.009	0.12	868	0.17
Gruvehaugen	50	5.59	29	0.20	0.008	0.21	1123	0.22
Rullekoll	50	2.66	25	0.09	0.007	0.10	1091	0.22
World average	1760	0.059	10	0.07	0.095	0.17	10	0.007

*F = 0.1 (Stranden, 1982; UNSCEAR, 2000; Kant *et al.*, 2009; Ramachandran *et al.*, 2010).

Overall, the most significant source of radiation is terrestrial gamma radiation, contributing from 81.8 to 95.2 % to the total outdoor exposure dose. Taking doses from ^{220}Rn into the consideration, with all of its limitations and uncertainties, would not give much higher total doses, and they would then be in the range from 0.26 to 0.51 mSv. The contribution of ^{220}Rn in this case ranged 7.8 – 68.7 %. However, an increase of here estimated exposure time would significantly influence the outdoor exposure doses.

An estimation of the possible annual outdoor exposure doses in a conservative scenario, with considered maximal values of measured radiation parameters, is given in Table 8. The annual outdoor exposure doses in such scenario would range from 0.21 to 1.54 mSv. Annual dose calculated for the people working at engineering firm at former mining site Søve was calculated to be 1.54 mSv, above the dose constraint of 1 mSv for public exposure. The very significant contribution of gamma radiation to the total dose remained (72.7 – 96.7 %). An increase in annual exposure doses would be acquired by people living at the edges of the ^{232}Th -rich Bolladalen and Rullekoll woods, up to 3 mSv, assuming an exposure time of 700 h per year. Such a scenario is actually very likely. The high values of ^{220}Rn (from 362 to 4996 Bq/m^3) in the air could contribute to the total effective dose. Possible magnitudes of annual doses due to ^{220}Rn inhalation (0.40 – 1.0 mSv) are given in Table 8. Total doses above the dose constraint of 1 mSv would be observed in such a case at the former mining sites Søve and Fen (1.94 and 1.22 mSv, respectively). Annual doses at the undisturbed ^{232}Th -rich site Gruvehaugen would be also elevated (1.05 mSv).

Table 8. Possible annual exposure doses at legacy and undisturbed NORM sites in a hyper conservative exposure scenario (maximal doses of measured parameters used in equations)

Site	Exposure time hrs	Gamma dose rate $\mu\text{Gy/h}$	^{222}Rn conc. Bq/m^3	Gamma dose mSv	^{222}Rn dose mSv	Total dose mSv	^{220}Rn conc. Bq/m^3	^{220}Rn dose mSv^*
Søve	250	8.51	37	1.49	0.05	1.54	362	0.40
Fen	50	4.46	210	0.16	0.06	0.22	4996	1.00
Bolladalen	50	4.76	157	0.17	0.04	0.21	2801	0.56
Gruvehaugen	50	9.24	120	0.32	0.03	0.35	3495	0.70
Rullekoll	50	5.56	72	0.19	0.02	0.21	2047	0.41

*F = 0.1 (Stranden, 1982; UNSCEAR, 2000; Kant *et al.*, 2009; Ramachandran *et al.*, 2010).

To summarize applying all available data and all constraints, the annual outdoor doses at the selected locations in the Fen area would be most likely within the range 0.10 – 1.54 mSv, based on terrestrial gamma radiation and ^{222}Rn . By far, the most significant contribution is from terrestrial gamma radiation. However, varying several factors (exposure time, use of ^{220}Rn concentrations) would lead to elevated annual outdoor doses, up to 10 mSv.

Additionally, variable indoor gamma radiation dose rates (0.035 – 0.62 $\mu\text{Gy/h}$), as well as elevated ^{222}Rn levels in a certain number of the Fen dwellings (10 – 1250 Bq/m^3), as measured by Sundal and Strand (2004), would give annual exposure doses of 0.2 – 3 mSv and 0.2 – 27 mSv for indoor exposure, respectively. Measurements of ^{220}Rn equilibrium equivalent concentrations, as well as a detailed survey of people’s habits in order to obtain accurate exposure times, are required for a calculation of the exposure doses in the Fen Complex. Nevertheless, the magnitude of the total annual dose for at least critical groups of people in the worst-case scenario (all maximal doses summarized) could be above 40 mSv, implying the need for a detailed and focused survey, and possible interventions (Figure 23). This is in good agreement with the previous estimate that 50 % of Fen village residents receive >10 – 13 mSv/y, 30 % receive >15 – 20 mSv/y, and 1 % might receive a dose from 52 to 68 mSv/y (Stranden, 1982).

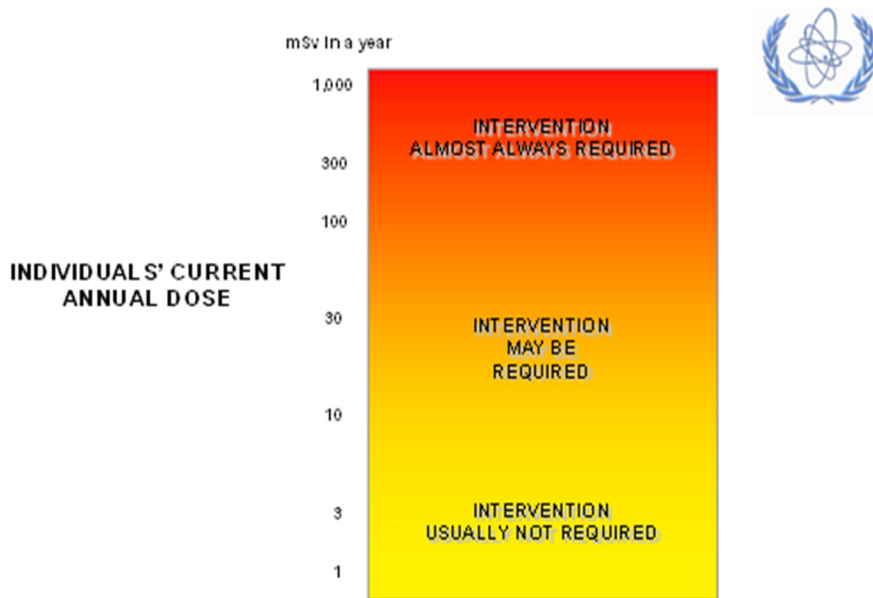


Figure 23. Intervention criteria (<http://www.iaea.org/> 2013).

5.5 Uncertainties in the assessments

The impact assessment performed in the present thesis required the analysis of a large number of different samples, sequential extractions and use of parameters (K_d , TF, CR) to determine the mobility and transfer through the environment and uptake in biota. Modeling to predict the biota dose rates, consideration of different exposure scenarios and the use of constants and coefficients in dose calculations followed in the later stages of the assessment. These activities and factors are commonly addressed as sources of uncertainty, rendering the assessment complicated (Oughton *et al.*, 2008; Salbu *et al.*, 2004).

To help in the evaluation of the presented data, an overview of uncertainties is given as follows:

- Representative sampling – an inhomogeneous distribution of radionuclides in soil and rocks was demonstrated; a wide range of soil activity concentrations, and consequently a wide range of gamma dose rates and $^{220,222}\text{Rn}$ in the air were obtained within the same sites. Variations in soil activity concentrations of ^{232}Th and ^{238}U were observed in vertical soil profiles also. Thus, under the environmental conditions that prevail in the Fen Complex area, the question of representative sampling remains opened and should be highlighted.

- Measurement uncertainties – a certain degree of uncertainty is typical for all laboratory data (Ellison and Williams, 2012). The present investigation of samples included sample storage, preparation and measurements. Different activities influenced the precision, accuracy and quality of the results. Section 5.1. shows how the quality of the present work was ensured.
- Uncertainty in sequential extraction – soil fractionation was performed by applying the sequential extraction model according to Salbu (2000), in order to obtain the information on reversibly- and irreversibly-bound radionuclides and trace elements in the soil. Although the sequential extraction approach is generally accepted and variations of the procedure are in use worldwide, the question of uncertainty due to the choice of reagents and conditions of extractions has been raised in the literature (Courchesne *et al.*, 2008; Sauv e, 2002).
- Uncertainty in mobility analysis – the concept of the distribution coefficients (K_d) is based on the assumption of equilibrium, and as such it is applied to radionuclides/trace elements in Fen Complex soil. However, the K_d change over time due to interactions of element species with soil and clay can cause the uncertainty in K_d to reach a factor 10^2 - 10^3 if speciation is not included in the analysis (Salbu *et al.*, 2004). Further, several experimental methods (e.g., laboratory batch method, laboratory column method, field batch method) for K_d determination have been developed, some with contrasting experimental conditions and specific uncertainties (e.g., due to aging effects and sorption dynamics) have been attributed to all procedures. Hence, wide ranges of K_d values for both radionuclides and trace elements are reported in the literature (Vandenhove *et al.*, 2009b), making comparisons and consequent conclusions unreliable.
- Uncertainty in applicability of the transfer factor (TF) concept in the case of earthworms – significant heterogeneity of radionuclides and trace element distributions were demonstrated in the Fen Complex area. Hence, the question to what extent the soil where the earthworms were found is representative of the soil in which they spent most of their time was raised. Most earthworm species can and do traverse considerable distances in the soil. However, the sampling points were chosen in areas where high radionuclide levels at all of the investigation points were previously demonstrated. The length of time that earthworms, despite their movement, spend in these soils is, in our opinion, long enough to allow earthworms to be representative organisms in the uptake study.

- Uncertainties in The ERICA calculations – in the current work, the ERICA Tool modeling was used to calculate the dose rates of biota. Models are always simplified versions and simulations of real systems, and as such they are associated with uncertainties, such as numerical uncertainties (e.g., physical quantities used as input data, parameters within the models like K_d , CR, occupancy time), model and scenario uncertainties (e.g., model structural errors, computational and mechanistic, time and spatial variations), conceptual (e.g., compartment models that represent reality, the assumption that dose rate is proportional to harm) (Oughton *et al.*, 2008). Initial screening of dose rates in the Fen area, using default parameters for CR and default occupancy for reference organisms, resulted in doses that were later shown to be overestimated when the model was run with a site-specific data set. It has been acknowledged that data on CR for a number of radionuclides are scarce or even unavailable, and several methods were used to obtain the best estimated values (Beresford *et al.*, 2008a; Oughton *et al.*, 2008). The degree of uncertainty in the second assessment was lowered; however, many of the above mentioned uncertainties are linked to the values of dose rates.
- Uncertainty in prediction of biota risk/effects – in order to study the possible effects on biota, the concentrations of radionuclides and trace elements in biota tissues, the TFs and biota dose rates were measured and calculated. Based on the obtained results (low biota tissue concentrations, low TFs and dose rates), no risk could be estimated for the Fen Complex biota, and no changes at the population level in biota were predicted. However, the simultaneous presence of both radionuclides and trace elements was demonstrated in plants and earthworms. Research on non-radioactive contaminants has shown that certain compounds can induce adverse effects as components of a mixture despite being present at concentrations at which as single contaminants they do not produce any effects (Baas *et al.*, 2010; Kortenkamp *et al.*, 2007). Thus, although concentrations of investigated radionuclides and trace elements were not demonstrated to be at high levels, the diverse effects (though not likely significant at the population level), could not be excluded and may needed further examination.
- Uncertainty in estimating human outdoor exposure – The largest uncertainty in the present thesis is linked to the difficulty in including high ^{220}Rn levels in the evaluation of doses in the Fen critical groups. It is known that the short half-life ($T_{1/2} = 55.6$ s) of ^{220}Rn contributes to spatial heterogeneity, and that a change in distance of 1 m could result in factor of 10 in the difference of recorded ^{220}Rn air concentrations (UNSCEAR, 2006). Detectors were placed according to prescribed

procedures, but some variation in readings due to inconsistent distances from the soil could not be excluded. However, in the case of the Fen Complex, such variations (and uncertainties) were most likely to be lower than variations in the air concentration of the gas due to the very inhomogeneous distribution of ^{232}Th in the soil, and thus were of no concern. To predict the radiation exposure dose based on ^{220}Rn is difficult and linked to a considerable uncertainty and not recommended (UNSCEAR, 2006; Yamada *et al.*, 2006). In the present thesis, such a recommendation was followed, and the major total doses were estimated as a sum of the dose of gamma radiation and ^{222}Rn dose. The magnitude of the possible contribution of ^{220}Rn was discussed, and all of its limitations were acknowledged. Additionally, the conversion coefficients and factors, although internationally accepted, can vary (Kranrod *et al.*, 2010; UNSCEAR, 2006), leading to over- or underestimation of doses. In the same way, adopted exposure scenarios are crucial for the dose magnitude; this can be observed by comparing the results from Paper V, where the usual outdoor occupancy of 1760 h was considered, and the results presented in section 5.4 of the thesis, where more realistic and specific exposure times for each of the study sites were provided, and the doses were discussed taking these factors into consideration.

6 Conclusions

The Fen Complex in Norway is an area well-known for its specific magmatic bedrock, rich in thorium (Th), iron (Fe), niobium (Nb) and rare earth elements (REE). In the past, intensive mining was conducted at several sites in the area, giving rise to enhanced radiation levels. Previous human health studies demonstrated that the annual exposure doses were among the highest in Europe. In the present work, the contamination status with respect to radionuclides and trace elements, their possible impact on humans and biota were investigated at legacy NORM and undisturbed surrounding ^{232}Th -rich sites.

Significant heterogeneous radionuclide (^{232}Th , ^{238}U and progenies) distribution in soil was demonstrated both at legacy NORM sites and undisturbed ^{232}Th -rich sites. Thorium activity concentration levels of soil exceeded the screening levels for radioactive waste material given by the Norwegian Pollution Control Act (2010). Based on the sequential extraction results, it was observed that the mobility of ^{232}Th and trace elements were low, although higher at the legacy NORM than at undisturbed ^{232}Th -rich sites. Uranium was present at considerable levels (>50 %) in pH- and redox-sensitive soil fraction, and associated with soil organic compounds. However, no significant transport to the largest water source, Lake Nordsjø, was observed. Short and long-term surveys of the outdoor terrestrial gamma dose rates and the concentrations of thoron (^{220}Rn) and radon (^{222}Rn) in the air demonstrated enhanced levels (up to 9.24 $\mu\text{Gy/h}$, 5000 Bq/m^3 and 200 Bq/m^3 , respectively), with seasonal variations.

To assess the impact on biota, wild plant species, free-living earthworm species, wild berries and mushrooms were collected from both former mining sites (legacy NORM) and undisturbed ^{232}Th -rich sites. Radionuclide (^{232}Th and ^{238}U as well progenies ^{228}Ra , ^{226}Ra , ^{210}Po) levels in biota were demonstrated to be in accordance with worldwide published literature, with calculated transfer factors actually lower than expected. The radiation dose rates obtained by the ERICA Tool using site-specific data, ranged from 1.3 – 23.1 $\mu\text{Gy/h}$, with maximal values estimated for earthworms, lichens and bryophytes. Although the calculated dose range for terrestrial organisms was higher than the typical world background doses (0.01 – 0.44 $\mu\text{Gy/h}$), the biota exposure doses were still very low compared to values below which no effects could be seen (40 and 400 $\mu\text{Gy/h}$ for terrestrial plants and animals, respectively). Thus, based on the here presented ERICA dose rates no risk for the Fen biota could be estimated.

The calculated annual exposure doses for critical groups of people due to outdoor exposure, ranged from 0.10 to 1.54 mSv, based on the doses of terrestrial gamma radiation and inhalation of the outdoor ^{222}Rn . The terrestrial gamma radiation was identified to be the most significant dose contributor. Varying the exposure times and taking into account ^{220}Rn doses could increase the doses up to over 10 mSv, although with a very large uncertainty. A rough summary with previously published data on indoor doses for the Fen village population suggests that the total annual exposure dose could exceed 40 mSv in certain groups of people in ‘the worst-case scenario’. Together, these findings identify several ‘hot-spots’ in the Fen Complex, including both legacy NORM and undisturbed ^{232}Th -rich sites. With respect to the current Norwegian legislation, an intervention action should be considered at the former mining site Søve.

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Paper I

Assessment of radionuclide and metal contamination in a thorium rich area in Norway

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The Fen Central Complex in southern Norway, a geologically well investigated area of magmatic carbonatite rocks, is assumed to be among the world largest natural reservoirs of thorium (^{232}Th). These rocks, also rich in iron (Fe), niobium (Nb), uranium (^{238}U) and rare earth elements (REE), were mined in several past centuries. Waste locations, giving rise to enhanced levels of both radionuclides and metals, are now situated in the area. Estimation of radionuclide and metal contamination of the environment and radiological risk assessment were done in this study. The average outdoor gamma dose rate measured in Fen, $2.71 \mu\text{Gy h}^{-1}$, was significantly higher than the world average dose rate of $0.059 \mu\text{Gy h}^{-1}$. The annual exposure dose from terrestrial gamma radiation, related to outdoor occupancy, was in the range $0.18\text{--}9.82 \text{ mSv}$. The total activity concentrations of ^{232}Th and ^{238}U in soil ranged from 69 to 6581 and from 49 to 130 Bq kg^{-1} , respectively. Enhanced concentrations were also identified for metals, arsenic (As), lead (Pb), chromium (Cr) and zinc (Zn), in the vicinity of former mining sites. Both radionuclide and heavy metal concentrations suggested leaching, mobilization and distribution from rocks into the soil. Correlation analysis indicated different origins for ^{232}Th and ^{238}U , but same or similar for ^{232}Th and metals As, Cr, Zn, nickel (Ni) and cadmium (Cd). The results from *in situ* size fractionation of water demonstrated radionuclides predominately present as colloids and low molecular mass (LMM) species, being potentially mobile and available for uptake in aquatic organisms of Norsjø Lake. Transfer factors, calculated for different plant species, showed the highest radionuclide accumulation in mosses and lichens. Uptake in trees was, as expected, lower. Relationship analysis of ^{232}Th and ^{238}U concentrations in moss and soil samples showed a significant positive linear correlation.

Introduction

Naturally occurring radioactive materials (NORM) are responsible for the major contribution to the total effective dose of

ionizing radiation of the world population.^{1,2} The radiation dose from natural sources is primarily generated by external and internal exposures to ^{238}U , ^{232}Th and their decay products. Mining and tailing are considered to be hazardous steps with respect to technologically enhanced naturally occurring radioactive material (TENORM) and metal contamination of the environment, and also in terms of radiation doses to man.³ Key problems are associated with transport of ^{238}U and ^{232}Th and their daughters, especially radium (Ra)-, polonium (Po)- and lead (Pb)-isotopes. In terrestrial and aquatic ecosystems, these

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Environmental impact

The Fen area in Norway, subject of this study, is assumed to be among the world largest deposits of thorium (^{232}Th) ore. Naturally occurring radioactive materials (NORM) are responsible for the major contribution to the total effective dose from ionizing radiation. The annual radiation dose, received by the population of this area, is estimated to be among the highest in Europe. The presence of metals along with ^{232}Th and uranium (^{238}U) gives the multiple stressors scenario and makes the whole exposure even more complicated. The information on radionuclide and metal concentrations, distribution and mobility in environmental compartments, obtained in this study, is highly relevant for estimation of pollution level, risk and possible biological effects. Further, data on transfer of ^{232}Th and ^{238}U from soil to vegetation in natural (and not experimental, designed) conditions are significant in radiological risk estimation of non-human biota and have been emphasized as needed by many working groups worldwide.

radionuclides can be transferred from the site of origin by air emissions, leaching and by run-off water, from soil into plants, animals and finally to man.

Substantial work associated with the evaluation of the radiological situation in ^{238}U and ^{232}Th deposit regions and in vicinity of mines has been performed in several regions such as Brazil, Australia, Europe, USA and Canada.⁴⁻⁸ Some studies have shown various pathways of radionuclide and metal leaching from rocks and soil, as well as their transportation and concentration of hazardous decay products into the water, plants and animals in different degrees.^{7,9-12} It has also been shown that radionuclides could be present as radioactive particles. Consequently, the transfer to man could occur *via* inhalation of particles from resuspension, as well as, *via* dietary intake.¹³

Radionuclides released from contaminated sites rarely occur alone, but more often along with metals and persistent organic pollutants, giving multiple stressors acting in nature. In mixtures of different stressors, multiple types of their interactions, as well as interactions with multiple target sites, may occur, inducing various biological effects. Thus, these stressors need to be investigated together in order to obtain more detailed assessment of possible risks related to environmental pollution.¹⁴⁻¹⁶

The Fen area (county of Telemark, southern Norway) is an area rich in NORM.¹⁷ It contains significant levels of ^{232}Th , ^{238}U and progenies, as well as, high levels of metals, As, Cr, Pb and Zn.¹⁸ Extensive mining activities have been conducted on several places in the area during previous centuries, ending in 1960s. Thus, beside NORM, several TENORM sites also occur there. These sites should be relevant for the national assessment of hazard, impact and risks from radionuclides and metal exposures, as the factual database is relatively poor. In fact, NORM and mostly TENORM (except waste from oil industry) have not been regulated in Norway until recently, but the new Norwegian Pollution Act considers and regulates radionuclides as pollutants, together with other kinds of man-made pollutants. With respect to that, a proper factual base from Fen is highly significant. Further, need for regulation and radiation protection and necessity to raise awareness about TENORM have been stated previously.^{19,20} On the other side, data on concentrations of NORM and TENORM in the environment are important for assessment of radiation impact on non-human species, as emphasized by Brown *et al.*,²¹ ICRP²² and Beresford *et al.*^{23,24}

The annual total effective radiation dose to population in the Fen area is up to a factor of 4 higher than the average radiation dose (2.9 mSv per annum) estimated for the Norwegian people.²⁵⁻²⁷ Focus of many investigations, previously done in this area, has been on Fen complex unique geology formation and its relationship with the enhanced content of ^{232}Th , ^{238}U and REE,²⁸⁻³⁰ increased gamma radiation dose rates and human radon risk estimation.^{26,27,31,32} Measurements of Dahlgren³⁰ have shown very high outdoor gamma radiation dose rates of $\sim 4 \mu\text{Gy h}^{-1}$ at some TENORM points. The arithmetic mean of indoor gamma radiation dose rate in Fen has been found to be $0.2 \mu\text{Gy h}^{-1}$, which is 3 times higher than the national average of $0.079 \mu\text{Gy h}^{-1}$ and comparable to the highest dose rates recorded in Europe.²⁷ The radiological and epidemiological impact of former mining activities, evaluated by Strandén,²⁵ has shown that workers received an annual dose equivalent of 150 mSv, obviously much higher than the occupational dose limit of

20 mSv per annum for radiation workers and even 3 times higher than the allowed maximum dose of 50 mSv in one year in special situations.

However, beside investigations considering human radiation doses and risk levels, neither investigations of contaminants in different environmental compartments nor the ecological risk assessments for species other than humans have been done at these locations.

The main objective of this work was to determine the concentrations of radionuclides ^{232}Th , ^{238}U and metals As, Cr, Pb, Cd, Ni, Cu and Zn in samples of soil, rocks, water and vegetation taken in the Fen Central Complex area. On the basis of results, an overall picture about radionuclide and metal dispersion to different environmental compartments was obtained. Relationship analysis of identified radionuclide concentrations and outdoor radiation dose rates, preliminary estimation of contamination degree due to former mining activities and general radiation risk assessment were also performed.

Materials and methods

Study area

The Fen region (Fig. 1) is located in Nome municipality, Telemark County in south-eastern Norway. Typical Fen Central Complex rock types, first described by Brøgger,²⁸ are carbonatite rocks of magmatic origin. The most abundant rock types of the area are: sövite (calcite carbonatite), rauhaugite (dolomite carbonatite), rödbergite (hematite-calcite carbonatite) and fenite (alkali-metasomatised granitic gneiss).³³ After ending of mining activities in the past century, several recordings of elevated natural gamma radiation doses in this complex have been done, either by portable hand detectors^{25,30} (Fig. 1) or by helicopter,³⁴ and all gave consistent high values.

Fieldwork and preparation of samples

Rock samples of different sizes were randomly taken at six locations in Fen (Table 1), and later identified as rock types sövite and rödbergite, typical for Fen geology complex. At least

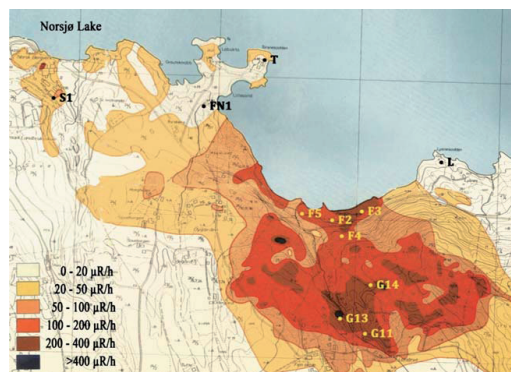


Fig. 1 Radiation map of the study area,³⁰ with marked sampling sites (fieldwork 2008) in Fen, Norway.

ten separate rocks were taken at each sampling location. After identifying, rocks were milled and stored until analysis.

Soil samples were collected at seven locations (NORM and TENORM) with, previously measured, different gamma radiation levels (Table 1). One sampling site (T) was chosen outside the mining area to represent the background level. At each sampling site soil was collected at minimum five separate points, at depth 0–20 cm. Samples were air dried at ambient temperature for several weeks, ground to fine powder and homogenized by sieving through 2 mm sieve. Analyses of pH, water and organic matter content were done for all soil samples.

Vegetation species found to be characteristic for the study area, *i.e.*, moss, lichen, birch leaves, fern, spruce and pine needles, were collected in paper bags at four field sites with variation in the outdoor gamma dose rate measured (Table 1). Plants were cleaned in the laboratory from adhered, visible soil, washed twice with distilled water, air dried for two weeks and homogenized by milling. Further, samples of rocks, soil and plants were oven dried (105 °C) to constant mass before acid microwave decomposition.

Water samples were taken at five locations as shown in Table 1. Water of the Norsjø Lake was sampled at three sites within the Fen public area (with different distance to the area with high background radiation) and at one site in the Fengruve area rich in ²³²Th bearing rocks. Additionally, water from a small river below Fengruve, that flows from the Fen bog area, an intensive Fe mining area in the past, was taken for analysis. Temperature, pH values and conductivity of water samples were measured *in situ* with a portable pH-meter, WTW Multi 340i. In order to obtain the information regarding the size distribution of metals and radionuclides of Norsjø Lake water, *in situ* filtration with a 0.45 µm membrane filter and ultrafiltration with a 10 kDa hollow-fibre were performed according to Salbu.³⁵ All water samples were conserved by acidifying with concentrated HNO₃ to obtain pH approximately below 2 (0.5% acid concentration).

Analytical method

Concentrations of radionuclides ²³²Th, ²³⁸U and metals As, Cr, Pb, Cd, Ni, Cu and Zn in samples were measured with ICP-MS analysis. Extracts of rocks, soil and plants were obtained after microwave decomposition (Milestone Inc., Ultraclave High performance reactor, Shelton, CT), employing high grade purity acid HNO₃ for soil and rocks and HNO₃–H₂O for plants. An internal standard containing 4 mg L⁻¹ of In, Tl and Rh in 2%

HNO₃ was added systematically to each sample before decomposition. ICP-MS analysis was done using a Perkin Elmer Sciex Elan 6000 (Norwalk CT, USA). Rock and soil samples were measured in 1% acid solutions and plant samples in 3.5% acid solutions. Each sample was determined by 3 consecutive injections. Detection limits were calculated as three times the standard deviation of blank samples measurements. The accuracy of the analysis was assured using certified reference materials NCS DC 73325 for soil and NCS DC 73348 for plants. Calibration standards, reagent and method blanks were also systematically used throughout ICP-MS measurements.

Statistical analyses

Statistical analyses were performed using Minitab 15 (Minitab Inc.). Data are presented as mean ± standard deviation (SD). Median values are presented together with the mean and range in the case of gamma exposure dose rates since values were not normally distributed on some sites. The normality was tested using the Anderson–Darling test. Difference analyses were done by ANOVA or Kruskal–Wallis test. Correlation analyses were performed using the Pearson correlation coefficient with *p* < 0.05 as a criterion for significance.

Results and discussion

Terrestrial gamma radiation measurements

Measurements of gamma radiation dose rates (Table 1) showed values in the range 0.07–9.24 µGy h⁻¹ for different Fen locations. These values are significantly higher than the world average terrestrial dose rate of 0.059 µGy h⁻¹,³⁶ but comparable with dose rates found in similar investigations of worldwide areas rich in NORM.^{6,8,37}

The maximum gamma dose rate was measured at site G13, in a past mining area called Gruvehaugen, where similarly, significant concentrations of ²³²Th and ²³⁸U were found in r dbergite rocks, soil and vegetation samples. High readings were also obtained at sampling locations F2, F3 and F4 in Fengruve area, in the vicinity of the waste location of abandoned Fe mines. However, at the location of the former Nb mining site, S vegruve (S1), the wide variation in readings was noticed. High gamma dose rates, up to 8.51 µGy h⁻¹, were recorded at some small, limited points, while majority of the terrain showed much lower gamma dose rates what resulted in a median of 0.63 µGy h⁻¹. This inconsistent gamma activity of the area could be explained with existing of intact protective sand covers at this site. Public

Table 1 Gamma radiation dose rates (µGy h⁻¹) in the Fen area

Sites, samples	Mean	Median	Range
S1, soil, rocks	2.22	0.63	0.24–8.51
F2, water	3.07	2.95	1.95–4.46
F3, soil, rocks, plants	3.42	3.40	2.94–3.77
F4, soil, rocks, plants	2.83	2.69	0.81–3.89
T, soil, plants, water	0.19	0.17	0.09–0.35
F5, water	0.98	0.92	0.66–1.46
L, water	0.24	0.22	0.14–0.37
FN1, water	0.15	0.14	0.07–0.24
G11, soil, rocks	4.85	4.87	4.11–5.57
G13, soil, rocks, plants	7.97	8.07	6.50–9.24
G14, soil, rocks	3.94	3.98	3.32–4.57

Table 2 Annual effective doses (mSv) related to outdoor occupancy, based on terrestrial gamma dose rates (µGy h⁻¹)

Site	Dose rate	Effective dose ^a	Effective dose ^b
FN1 (public zone)	0.15	0.92	0.18
S1 (public zone, former Nb mine)	2.22	13.61	2.72
G14 (wooden zone, former Fe mine)	7.97	48.87	9.82
World average	0.059	0.36	0.07

^a Exposure time 8760 h. ^b Exposure time 1760 h.

locations in Fen, that were chosen as examples of Fen background sites, T, L and FN1 had, as expected, the lowest gamma radiation dose rates measured, *i.e.*, 0.07–0.37 $\mu\text{Gy h}^{-1}$. Still, these values are several times higher than the world average level 0.059 $\mu\text{Gy h}^{-1}$.³⁶ Results obtained in this study are comparable with previous measurements (Fig. 1) done in this Norwegian area.³⁰ Likewise, the presence of enriched ^{232}Th and ^{238}U rocks has shown to cause similar high outdoor radiation dose worldwide.^{38,39}

The average annual radiation effective doses due to external exposure to natural terrestrial sources of radiation, for certain sites within public and wooden area (FN1, S1 and G14), are given in Table 2. Shown effective radiation doses are related to the outdoor occupancy, and calculated using measured absorbed dose rates in the air and 0.7 Sv Gy^{-1} as the conversion coefficient from the absorbed dose rate in the air to the effective dose received by adults.³⁶ Two exposure periods were taken into the consideration, *i.e.*, 8760 and 1760 hours. As it is shown in Table 2, the annual exposure dose due to terrestrial radiation within public place is approximately 3 times higher than the world average value. However, extremely high gamma radiation dose rates in the vicinity and around the former mining sites in the wooden zone of Fen gave significantly high exposure doses, up to 140 times higher than the world average. Calculated outdoor exposure doses for NORM and TENORM area are even higher than the world average total exposure dose that is in the range 0.3–1 mSv per annum³⁶ for most of the countries. On the basis of our data, it was concluded that radiation protection intervention actions, according to ICRP,⁴⁰ should be applied on these open and free accessible sites.

Radionuclide and metal concentrations in soil

Results of soil analysis (Table 3) showed wide ranges for radionuclides activity concentrations, 69–6581 Bq kg^{-1} and 49–130 Bq kg^{-1} , for ^{232}Th and ^{238}U , respectively. These values clearly exceed the world average values, 45 Bq kg^{-1} for ^{232}Th and 33 Bq kg^{-1} for ^{238}U for soil, given by UNSCEAR.³⁶

The highest ^{232}Th concentration was found in soil at site G11, area Gruvehaugen, and the lowest at site S1, area Søvgruve (Table 3). The pattern of ^{232}Th distribution in soil was in connection with the abundance of the rock type rödbergite (with high ^{232}Th level) and with the abundance of the rock type sövite (considerable lower ^{232}Th level). Also, as mentioned above, the former Nb mining site Søvgruve was covered with a protective layer of clay and sand material which contributed to lower radionuclide results for soil. Comparison of the ^{232}Th concentration median in TENORM rich and background soil samples showed a significant difference (non-parametric Kruskal–Wallis test, $H = 31.50$, $p < 0.05$). The maximum ^{238}U concentration, 130 Bq kg^{-1} , was measured on site T, which is an unexpected high value considering that this sampling site was chosen to represent the background area. It indicated that high radionuclide concentrations could be measured not only on TENORM but on other non-disturbed sites in the area.

Correlation analysis of ^{232}Th and ^{238}U in soil did not show any significant relationship. The most likely explanation is that different and not correlated minerals were origins for these two

Table 3 Radionuclides activity concentrations (Bq kg^{-1}) in soil (mean \pm SD)

Site	$^{232}\text{Th}^a$	$^{238}\text{U}^a$
S1	69 \pm 8	49 \pm 12
F3	4011 \pm 1108	111 \pm 37
F4	4563 \pm 93	111 \pm 37
T	1059 \pm 28	130 \pm 5
G11	6581 \pm 364	86 \pm 62
G13	5781 \pm 848	68 \pm 6
G14	4839 \pm 126	85 \pm 5

^a Activity concentrations (Bq kg^{-1}) obtained by multiplying ICP-MS concentrations with the factor 4.06 and 12.35 for ^{232}Th and ^{238}U , respectively.

radionuclides in rocks. The presence of rocks particularly rich in some minerals highly affected soil concentration.

Generally, soil concentration results suggested that erosion and weathering of rocks led to formation of soil layers enriched in radionuclides. Intensive mining activities in the past gave, additionally, an increase of radiation locally. Similar results for soil ^{232}Th and ^{238}U distribution were obtained in the study of soil from a region in Malaysia, with an abundance of basic volcanic rocks rich in monazite.⁶ Investigation of carbonatite rocks, type sövite, in Uganda³⁹ showed higher, but comparable to Fen values for ^{232}Th and similar for ^{238}U .

The dependence of air measured gamma dose rates on ^{232}Th activity concentration in soil was confirmed by a positive linear correlation found (Fig. 2) ($r = 0.78$, $p < 0.001$, $n = 30$). However, it is possible that some mother–daughter equilibriums in ^{232}Th and ^{238}U radionuclides decay chains were disturbed (*e.g.*, due to radium (^{226}Ra) mobility or radon (^{222}Rn) emission), giving the gamma daughters that contributed to some extremely high measured dose rates. Thus, this relationship showed the need for more detailed investigation of the actual concentration of ^{232}Th and ^{238}U daughter products.

To check the possibility for a multiple radiological and metal contamination in this area, soil samples were simultaneously analyzed for metals such as As, Cr, Ni, Cd, Pb, Cu and Zn. Obtained concentrations of metals are given in Table 4. Default upper limit values for soil that can be considered as non-polluted in Norway, given by SFT,⁴¹ are also presented in Table 4, for comparison purpose.

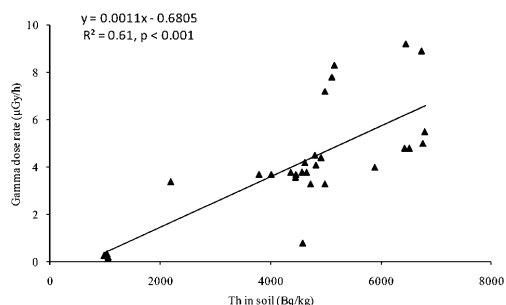


Fig. 2 Relationship between terrestrial gamma dose rates and ^{232}Th activity concentration in soil, measured in Fen wood.

Concentrations of As in soil samples (with an exception of site S1 with protective sand layer) were higher than the value of 8 mg kg^{-1} for non-polluted Norwegian soil. Earlier investigation of soil samples from Fen area¹⁸ has shown As values similar to the values we obtained. Analysis of the relationship between As and ^{232}Th concentrations showed a strong positive linear correlation ($r = 0.86$, $p < 0.05$, $n = 35$) (Fig. 3). However, no correlation between As and ^{238}U was found. This result could indicate that As was present in same minerals as ^{232}Th . The concentration range for Pb in samples of soil taken in Fen area was $47\text{--}287 \text{ mg kg}^{-1}$, with a median value of 63 mg kg^{-1} . Obtained results for 4 of 7 investigated sites are higher than the SFT⁴¹ value, but in accordance with results previously obtained for southern Norway.^{42,43} A moderate positive correlation was found between Pb concentrations and ^{238}U concentrations in soil ($r = 0.61$, $p < 0.05$, $n = 35$), but a weak negative correlation was found with ^{232}Th ($r = -0.37$, $p = 0.028$, $n = 35$). Opposite to As, it is likely that Pb is connected with ^{238}U in the same mineral types. Cd concentrations in soil were in ranges often given for soil in the vicinity of sedimentary or Fe rich rocks and soil in different parts of Norway.^{41,42,44-46} Zn was found to be present in different levels in soil, from $47\text{--}1021 \text{ mg kg}^{-1}$. These values are 3–10 times higher than those found in Norwegian surface soil, by Steinnes and Ruhling.⁴² Cr and Ni exceeded SFT⁴¹ values in samples taken at sites G11, G13 and G14 in the wooden area of Gruvehaugen, where high radionuclides levels were also found. Results of Cu levels in soil were in agreement with the different abundance of Cu, $2\text{--}250 \text{ mg kg}^{-1}$, that has been recorded in different soil types.^{42,45} Cd, Zn and Cr showed a moderate positive linear correlation with ^{232}Th concentrations ($r = 0.63$, $r = 0.58$ and $r = 0.44$, respectively, all $p < 0.05$, $n = 35$). The relationship of soil Ni and ^{232}Th concentrations was, as in the case of As, positive ($r = 0.74$, $p < 0.05$, $n = 35$) (Fig. 4). Cu levels were negatively correlated to ^{232}Th in soil.

In general, distribution of all metals was found to be very inhomogeneous, even on close sampling points. High levels of metals on some sampling sites could be the result of local loading due to weathering of bedrocks and leaching upon different environmental conditions. However, vicinity of contaminated hot spots to past mining sites indicated correlation with human activities in past centuries. Positive correlations of mentioned metals with ^{232}Th should mean that they probably had the same origin. Therefore, not only extremely high radionuclide contamination but mixture of radiological and chemical contamination could affect populations living in Fen. These mixtures could cause diverse biological effects—consequences of

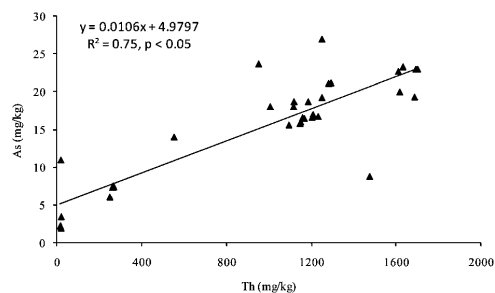


Fig. 3 Relationship between As and ^{232}Th concentrations in soil.

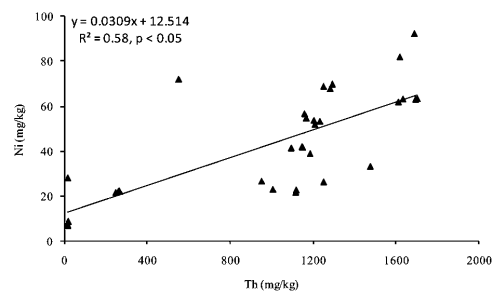


Fig. 4 Relationship between Ni and ^{232}Th concentrations in soil.

additive or synergistic acting.¹⁵ Thus, future work will be focused on radionuclide and metal mobility and transfer factors into vegetation and animal species of the region, as well as on possible, induced, biological effects.

Fen Central Complex rocks

The mean concentration of ^{232}Th in r dbergite rocks collected at Gruvehaugen and Fenguve locations (Table 5) was 1552 mg kg^{-1} . The mean concentration for ^{232}Th in s vite rock samples was found to be considerably lower, 64 mg kg^{-1} . Results of rock samples analysis (Table 5), previously reported,^{27,30,34} showed comparable concentrations for ^{232}Th in these rock types. Values for r dbergite were significantly higher than typical ^{232}Th values of $0.1\text{--}87.5 \text{ mg kg}^{-1}$ for Norwegian bedrock,⁴⁷ placing it in a position of rock with highest ^{232}Th level in Norwegian

Table 4 Metal concentrations (mg kg^{-1}) in soil (mean \pm SD)

Site	As	Cd	Pb	Cr	Ni	Cu	Zn
S1	4 \pm 4	0.26 \pm 0.01	47 \pm 5	26 \pm 23	14 \pm 15	45 \pm 24	210 \pm 52
F3	20 \pm 5	0.60 \pm 0.14	134 \pm 95	33 \pm 19	37 \pm 20	9 \pm 12	47 \pm 5
F4	17 \pm 1	0.73 \pm 0.07	121 \pm 23	40 \pm 8	34 \pm 11	9 \pm 7	891 \pm 334
T	7 \pm 1	0.48 \pm 0.02	287 \pm 10	30 \pm 0	22 \pm 1	12 \pm 0	242 \pm 3
G11	20 \pm 6	1.88 \pm 0.59	55 \pm 7	54 \pm 14	57 \pm 13	5 \pm 2	1021 \pm 300
G13	20 \pm 1	0.68 \pm 0.07	65 \pm 10	66 \pm 4	76 \pm 11	6 \pm 1	258 \pm 28
G14	16 \pm 1	0.54 \pm 0.07	59 \pm 2	89 \pm 3	54 \pm 2	6 \pm 1	268 \pm 27
SFT ^a	8	1.5	60	50	60	100	200

^a SFT (Norwegian Pollution Control Authority) standard values—criteria for non-polluted soil in Norway.

Table 5 Results of rock radionuclide analysis (mg kg⁻¹), comparison with earlier investigations

Rock type	²³² Th mean (range)	²³⁸ U mean (range)	²³² Th/ ²³⁸ U ratio
Sövite ^a	64 (51–77)	5 (4–6)	11
Sövite ^b	(1–50) ^c	(1–60) ^c	—
Sövite ^c	20 (5–48)	1.6 (0.8–4.9)	—
Sövite ^d	(10–150) ^e	(2–17) ^e	—
Rödbergite ^a	1552 (897–1969)	6 (3–12)	342
Rödbergite ^b	(560–3000) ^c	(5–40) ^c	—
Rödbergite ^c	775 (97–1475)	5.7 (1.6–8.9)	—
Rödbergite ^d	(183–1060) ^e	(1.8–25.1) ^e	—

^a This study results. ^b Dahlgren.³⁰ ^c Sundal and Strand.²⁷ ^d NGI.³⁴
^e Mean values not provided.

bedrock.²⁷ ²³⁸U in rocks was present in significantly lower concentrations, 6 mg kg⁻¹ and 5 mg kg⁻¹, in rödbergite and sövite, respectively. Obtained results are consistent with those reported by Dahlgren,³⁰ Sundal and Strand²⁷ and NGI.³⁴ As presented in Table 5, the ²³²Th/²³⁸U ratio for sövite rock type was 11 and for rödbergite rock type was 342. These distinguished values reflected a high difference in the ²³²Th concentration level compared with the ²³⁸U concentration level in the two different rock types. The ²³²Th/²³⁸U ratio value obtained for sövite was similar to the ratio of 7.9 previously published by Heincke *et al.*⁴⁸ However, the value for rödbergite was higher than 54.7 obtained in the same study.⁴⁸

²³²Th rich rocks in Fen are estimated as valuable energy sources for future.¹⁷ Thus, the investigation of rocks radionuclide levels and possible mobilization of radionuclides toward other environmental compartments is of importance in light of possible future excavation and mining processes.

Water analysis

Radionuclide concentrations in water ranged from 0.038 to 0.187 µg L⁻¹ and from 0.136 to 4.414 µg L⁻¹, for ²³²Th and ²³⁸U, respectively, (Table 6). These numbers exceed values given by UNSCEAR³ for the world average concentration in water that contribute to exposure from natural sources, 0.012 and 0.083 µg L⁻¹ for ²³²Th and ²³⁸U, respectively. However, values for both radionuclides were still far below the significant contamination levels considering guideline for drinking water.⁴⁹ Previously, Stranden²⁵ has also reported no radiation dose contribution for Fen human population, regarding Norsjø Lake water. The highest ²³²Th concentration, 0.187 µg L⁻¹, was obtained as a mean value for stream water samples from the

Table 6 Radionuclides concentration in water samples (µg L⁻¹) (mean ± SD)

Site	²³² Th	²³⁸ U
F2	0.097 ± 0.031	0.172 ± 0.006
F5	0.187 ± 0.015	4.414 ± 0.105
FN1	0.038 ± 0.014	0.206 ± 0.007
T	0.041 ± 0.014	0.149 ± 0.003
L	0.041 ± 0.012	0.136 ± 0.003

Fengruve wood. This higher concentration could be explained with ²³²Th leaching and mobilization from the surrounding area, significantly rich in ²³²Th. Comparison of ²³²Th and ²³⁸U levels showed lower levels of dissolved ²³²Th in all water samples. This is in agreement with generally low chemical solubility of ²³²Th due to high affinity for sedimentation and particle formation. The highest ²³⁸U concentration, 4.14 µg L⁻¹ was also found in a river stream in the Fengruve area.

Concentrations of Pb, Cd, As, Ni, Cu and Zn were similar to those reported for Norwegian rivers and lakes from different parts of country.^{46,50} Low metals concentrations, in water samples taken on sites with soil rich in these metals, indicated no leaching and mobilization from soil and rocks into the water.

However, the analysis of size distribution of both radionuclides and metals, important for estimation of possibility for water living organisms uptake, showed interesting results (Fig. 5). ²³²Th size distribution was as follows: 43.75% was present as particulate, 48.44% as colloid and 7.81% as low molecular mass (LMM) species. Regarding typical chemical behaviour of ²³²Th, *i.e.*, extremely low solubility and tendency to settle down in the form of particles, we expected a considerable higher percentage in phase >0.45 µm than what was found, but results were comparable to those found in other investigations.^{51,52} Still, more than 50% of ²³²Th was present in mobilizable colloidal and LMM form. Thus, the colloidal transportation is expected and might induce further distribution and bio-concentration in living species of this water system. Analysis of ²³⁸U showed that it was present mainly as colloids (90%) (Fig. 5). Consequently, it is expected that mobile, soluble ²³⁸U could be a subject of active uptake by aquatic biota. The most abundant fraction of all investigated metals was LMM, below 10 kDa. This fraction is considering active biological uptake, environmental fate and influence, the most important one.⁵³ Thus, accumulation of these metals could also be possible in living organisms of the Norsjø Lake and should be investigated.

Radionuclides and metals in vegetation

The absorbed dose rates and preliminary risk assessment for non-human species living in the area were calculated with the ERICA software tool⁵⁴ (Fig. 6). The calculation of total exposure doses is based on soil concentrations of radionuclides and default

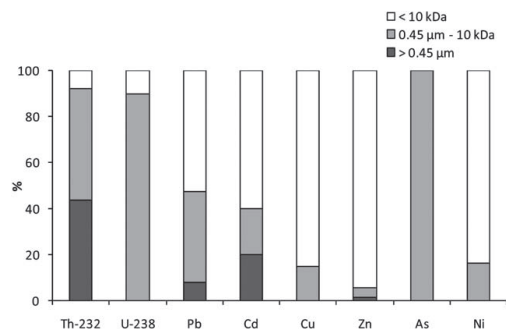


Fig. 5 Size fractionation of radionuclides and metals in water from Norsjø Lake.

values for concentration ratios for organisms of interest. The screening value for risk estimation is set on $10 \mu\text{Gy h}^{-1}$. No relevant biological effects, consequences of radiation exposure, have been seen under this limit.⁵⁴ As it is shown in Fig. 5, grasses, herbs, lichens and bryophytes are groups of organisms that could potentially be considered as under the risk in Fen conditions.

Plants uptake of radionuclides was investigated in order to estimate the possibility for bioconcentration and further bioaccumulation in food chain. Certain studies have earlier shown that accumulation in mosses and lichens is in agreement with radionuclide and metal contamination of soil.^{55,56}

In our study, variable sensitivity and capacity for uptake was obtained. The highest ^{232}Th concentrations were observed in moss and lichen plants, 4.70 and 4.46 mg kg^{-1} , respectively. The lowest ^{232}Th concentrations were observed in fern, spruce and birch, in the range $0.02\text{--}0.09 \text{ mg kg}^{-1}$. Lower but comparable results for moss and grass samples were obtained in similar investigations of Ramli *et al.*⁶ and Vandenhove *et al.*⁷ Concentrations of ^{238}U were, also, the highest in moss and lichen, 0.53 and 0.21 mg kg^{-1} , respectively. Summarized, ^{232}Th and ^{238}U in plant samples, except the fern sample, were higher than reference values for leafy plants (0.004 mg kg^{-1} and 0.005 mg kg^{-1} for ^{232}Th and ^{238}U respectively), given by UNSCEAR.³

Significant positive linear correlations ($r = 0.88$, $p = 0.001$, $n = 10$ and $r = 0.95$, $p = 0.000$, $n = 10$) were obtained for ^{232}Th and ^{238}U , respectively, in moss and corresponding soil samples (Fig. 7).

However, no positive correlation was found for ^{232}Th and ^{238}U concentrations in lichen and soil samples taken under the plants. A possible different mechanism of uptake could be explanation for the correlation absence.

In addition to radionuclides, metal concentrations were analyzed in all plants, but obtained results showed no significant levels of uptake.

Transfer factors were calculated as ratio between dry weight plant concentration and dry weight soil concentration. Values for transfer factors for ^{232}Th (Table 7) in plants were 1–2 orders of magnitude lower than previously published.^{6,24,57,58} ^{238}U transfer factors were lower than those obtained by Dragovic *et al.*⁵⁸ and Ramli *et al.*⁶

^{238}U was more than ^{232}Th taken up by analyzed plants. This was expected since ^{238}U is a more soluble and mobile radionuclide, and that also may mimic essential elements.⁵⁹

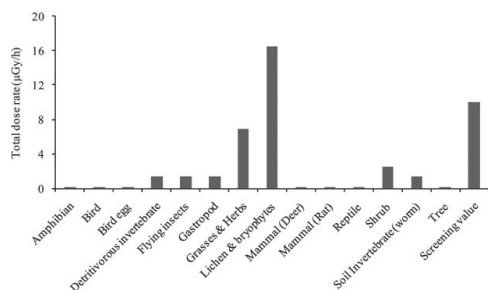


Fig. 6 Radiation exposure dose rates per organism group, by the ERICA software tool (Brown *et al.*⁵⁴).

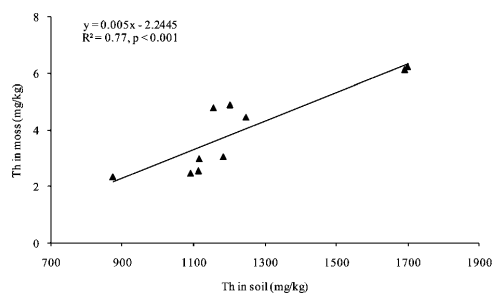


Fig. 7 Relationship of ^{232}Th concentration in moss and corresponding soil samples.

Table 7 Vegetation transfer factors (TF)

Vegetation	^{232}Th	^{238}U
Moss	3.33×10^{-3}	6.25×10^{-2}
Lichen	4.90×10^{-3}	2.59×10^{-2}
Spruce	4.55×10^{-5}	4.10×10^{-4}
Pine	1.80×10^{-4}	1.90×10^{-3}

Generally, low transfer factors were obtained despite high activity concentrations of both radionuclides in plants. Possible explanations could be limited plants uptake capacity and, in the same time, extremely high total soil radionuclides concentration used in calculations. To obtain more accurate transfer factors, the study should be expanded on investigation of different soil fractions, calculation of available transfer factors (ATF), differences between species or age of the plants and possible different mechanisms of uptake.

Conclusion

Environmental risk assessment was done in a thorium rich area in Norway. Terrestrial gamma dose rates in Fen Central Complex were in the range $0.15\text{--}7.97 \mu\text{Gy h}^{-1}$, significantly higher than the world average of $0.059 \mu\text{Gy h}^{-1}$.³⁶ These gamma dose rates highly reflected the presence of radionuclides bearing minerals in volcanic rocks of the terrain. Consequently, annual effective doses due to outdoor external exposure to natural terrestrial radiation were in the range $0.18\text{--}9.82 \text{ mSv}$. The majority of investigated, free accessible sites in Fen had the exposure dose considerably higher than the world average of 0.07 mSv per annum for outdoor exposure.³⁶ These numbers put Fen area in a position of highly enhanced radiation area not only in Norway but in Europe also. Further, the risk could be even higher if thoron (^{220}Rn) and radon (^{222}Rn) air levels were taken into the consideration and assessment. Radiation protection intervention actions are recommended.

Soil ^{232}Th concentrations were found to be higher than world and Norwegian average soil values, and positively correlated with the presence of certain ^{232}Th rock types. No positive correlation was found between ^{232}Th and ^{238}U , suggesting different and separate types of minerals, carriers of radionuclides. Inhomogeneous distribution of radiation was found in the

whole area. Radionuclide and metal concentrations were particularly high in the vicinity of former Fe mining sites. It is most likely that intensive mining activities from past, coupled with rocks weathering upon different environmental conditions in long period of time, have given an increase to both, concentration of ^{232}Th , ^{238}U and to the concentration of some metals. A strong positive correlation of As and Ni, as well as moderate of Cd, Zn and Cr, with ^{232}Th could mean that these metals are present in the same minerals as ^{232}Th .

Analyzed water samples showed elevated concentrations of radionuclides according to UNSCEAR,³ but still in compliance with requirements for clean water.⁴⁹ However, results of size fractionation, *i.e.*, presence of all metals and high percentage of both radionuclides in the biologically most important colloidal and low molecular mass fractions, could lead to accumulation in living organisms of Nørsjø Lake.

The assessment of plants uptake showed lower transfer factors of both radionuclides than worldwide published values despite the high concentrations in plants. ^{238}U was more readily taken up than ^{232}Th by all plant species. Mosses and lichens showed 1–2 orders of magnitude higher accumulation than trees. Assumption of linearity of plant and soil concentrations was confirmed in the case of moss for both ^{232}Th and ^{238}U .

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Paper II

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ARTICLE TYPE

Mobility of radionuclides and trace elements in soil from legacy NORM and undisturbed naturally ²³²Th-rich sites

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10 Investigation of radionuclides (²³²Th and ²³⁸U) and trace elements (Cr, As and Pb) in soil from two legacy NORM (former mining sites) and one undisturbed naturally ²³²Th-rich site was conducted as a part of the ongoing environmental impact assessment in the Fen Complex area (Norway). The major objectives were to determine the radionuclides and trace elements distribution and mobility in soils as well as to analyze possible differences between legacy NORM and surrounding undisturbed naturally ²³²Th-rich soils.

15 Inhomogeneous soil distribution of radionuclides and trace elements was observed for each of the investigated sites. Concentration of ²³²Th was high (up to 1685 mg/kg, i.e., ~7000 Bq/kg) and exceeded the screening value for the radioactive waste material in Norway (1 Bq/g). Based on the sequential extraction results, the majority of ²³²Th and trace elements were rather inert, irreversibly bound to soil. Uranium was found to be potentially more mobile, as it was associated with pH-sensitive soil phases,

20 redox-sensitive amorphous soil phases and soil organic compounds. Comparison of the sequential extraction data sets from the three investigated sites revealed increased mobility of all analyzed elements at the legacy NORM sites in comparison to the undisturbed ²³²Th-rich site. Similarly, the distribution coefficients K_d (²³²Th) and K_d (²³⁸U) suggested elevated dissolution, mobility and transportation at the legacy NORM sites, especially at the decommissioned Nb-mining site (346 and 100 L/kg for ²³²Th and

25 ²³⁸U, respectively), while the higher sorption of radionuclides was demonstrated at the undisturbed ²³²Th-rich site (10672 and 506 L/kg for ²³²Th and ²³⁸U, respectively). In general, although the concentration ranges of radionuclides and trace elements were similarly wide both at the legacy NORM and at the undisturbed ²³²Th-rich sites, the results of soil sequential extractions together with K_d values supported the expected differences between sites as the consequences of previous mining operations. Hence,

30 mobility and possible elevated bioavailability at the legacy NORM site could be expected and further risk assessment should take this into account when decisions about the possible intervention measures are made.

Introduction

Most of the naturally occurring radionuclides (NOR) exist in soils and rocks at levels that are of no concern to human health or the environment.¹ However, due to their specific geological composition, some areas are particularly rich in primordial radionuclides uranium (²³⁸U) and thorium (²³²Th), often accompanied with different heavy metals. Mining activities in

40 such areas usually generate various amounts of radionuclide and metal contaminated waste in the form of disposal dumps on the soil surface. Increasing attention has recently been paid to such naturally occurring radioactive materials (NORM) rich sites due to the common problem of large amounts of waste, often

45 containing some long-lived, highly radiotoxic radionuclides.² In most cases, defined legislative requirements are applied to the exploitation sites in order to protect people from radioactive contamination. Nevertheless, there still is a backlog in the knowledge on the radiation protection problems with NORM, in

50 comparison to artificial radionuclides. Consequently, there is a general lack of awareness and assessment of radiological hazards and exposure levels by legislators, regulators and operators.² Additionally, recent shift from an anthropocentric to an ecocentric view in radiation protection³ makes risk assessment

55 and decision making at both legacy NORM and undisturbed NORM rich sites more demanding.

The Fen Complex, located in Precambrian gneisses of Telemark (south-eastern Norway), is an area well known and

fully described with respect to its specific volcanic bedrock geology.^{4,5} Several types of magmatic rocks in the Fen Complex are particularly rich in the radionuclide ²³²Th, iron (Fe), niobium (Nb) and rare earth elements (REE). Mining for Fe and Nb was conducted in the area during past centuries.

Elevated terrestrial gamma radiation in the Fen area and possible human health effects due to former mining activities and naturally high radiation, were in focus of investigation in the last 30 years.⁶⁻¹⁰ The high radiation exposure doses from terrestrial gamma radiation and indoor radon (²²²Rn) were reported.^{9,10}

As soil radionuclide concentrations are determined by those in the parent material and by physical and chemical phenomena associated with weathering,¹¹ the Fen Complex soils were shown to have significantly elevated ²³²Th and moderate ²³⁸U concentrations.^{7,12} However, the concept of total concentration analysis of radionuclides and trace elements in soil provides the limited insight into the actual behaviour of elements in soil and, thus, is of partial value for the assessment of environmental impacts and risks. Both radionuclides and trace elements are associated with a number of different phases in soils and knowledge about these forms is required for understanding and predicting their future behaviour, mobility, bioavailability and toxicity to biota.¹³⁻¹⁷ Furthermore, the issue of potentially different speciation and mobility of investigated elements in soils from the legacy NORM and the undisturbed ²³²Th-rich sites was raised.

The objectives of the present study were to determine distribution and mobility of radionuclides (²³²Th, ²³⁸U) and trace elements (chromium (Cr), arsenic (As) and lead (Pb)) in soil of the Fen Complex area. Vertical soil concentration profiles and possible differences between legacy NORM sites and undisturbed ²³²Th-rich site were analyzed and discussed. The current study provides valuable results for the Fen Complex area which could be considered in the context of the new amendments of Norwegian Pollution Control Act regarding radioactive pollution and waste in Norway (Pollution Control Act; FOR 2010-11-01 nr.1394).¹⁸

Materials and methods

Study sites

The results discussed in this paper were obtained from samples taken in several field campaigns during the period 2008-2011. Three sampling sites (former mining sites Fen and Søve as legacy NORM sites and Rullekoll as undisturbed ²³²Th rich site) were chosen to meet the main objectives of the present study. The detailed description of the geology of the Fen Complex and map with positions of the study sites is given by Mrdakovic Popic et al.¹⁹

The former mining site Søve is located in the western part of the Fen Complex (N 59° 16.902' E 009° 17.162'), occupied by the rock Søvite (calcite-carbonatite). It is a decommissioned Nb-mining complex, which was active in the period 1953-1965. In order to reduce the risk from contamination, after the mine decommission, the site was covered by sand layers in a remediation action. However, recent investigations demonstrated the high levels of soil radioactivity together with elevated air radon (^{220,222}Rn) concentrations and high gamma dose rates in the

air,^{7,19,20} suggesting cover disturbance and mixing with underlying waste during the time.

The former mining site Fen comprises of wooded zone on the shores of the Lake Nordsjø in the north-central part of the Fen Complex (N 59°16.625' E 009°18.226'). Bedrock of the area consists mainly of the rock Rødbergite (hematite-calcite-carbonatite). Mining of Fe was conducted during several centuries in the past (17th-20th century). Nowadays, large quantities of crushed waste rocks are deposited at this freely-accessible place.

The Rullekoll site is a small wood situated in the south of the Fen Complex zone (N 59°16.002' E 009°18.110'), outside the mining areas. This site was chosen as undisturbed site with bedrock consisting of the rock Rauhaugite (dolomite-carbonatite) demonstrated to be rich in ²³²Th.

Soil analyses

Soil was sampled in two different ways. Samples for total soil analysis and sequential extraction were taken in the first fieldwork as bulk samples, using a soil corer device of 10 cm diameter, to the depth of approximately 5-20 cm. Typical soil sample weighed 0.2-1.0 kg. In the subsequent fieldwork, samples were obtained by digging pits, and then incrementally excavating one wall of the pit to obtain sub-samples from different depths. In most cases, the total depth was limited to 20 cm where the parent rock material was already present in abundance. Soil was collected from the vertical profiles at approximately 0-5, 5-10, 10-15 and 15-20 cm. The samples were transported to the laboratory in polyethylene bags, dried for several weeks at 40°C and then sieved to <2 mm particle size. Measurement of total soil concentrations, analysis of soil vertical profiles, sequential extraction and autoradiography were conducted right after soil drying in the laboratory. In such way, it was ensured that results of undertaken chemical analyses reflected distribution and mobility as they were in the environment. Samples taken for the determination of K_d were transported to the laboratory in polyethylene bags and analyzed during the same day. Organic matter (OM), grain size, pH, cation exchange capacity (CEC) and stable elements (Fe, Mn) were determined as part of basic soil characterization. Loss on ignition (550°C, 12h) was used to determine OM, while grain size was analyzed by an in-house pipette method. After suspending soil samples in ultrapure water (prepared by Millipore) in the ratio 1:5 (w:v) and allowing to settle for 24h, pH was measured using a glass electrode (Mettler Toledo, SenTix 21). Exchangeable bases were determined with ICP-OES in NH₄OAc extracts of soil according to Reeuwijk.²¹ Stable elements (Fe, Mn) were measured by ICP-OES after microwave assisted acid digestion of soil.

In order to decompose soil, dry sub-samples were weighed (0.3-0.5 g) in Teflon containers and 5 ml of ultrapure nitric acid (HNO₃) was added together with the internal standard (Indium (In), Rhodium (Rh) and Thallium (Tl)). Samples were digested in a microwave oven (MLS-Milestone UltraClave, MLS GmbH) with an increasing temperature and pressure regime, quantitatively transferred into 50 ml volumetric flasks and diluted to volume with deionized water.

In addition, ten soil samples from each site were treated by sequential extraction²²⁻²⁴ to satisfy the scientifically recognized need for information on binding mechanisms in soil in order to

understand their environmental fate.^{14,25-27} Assessment of element mobility and bioavailability in soils, using the sequential extraction, assumes that mobility and bioavailability decrease in the order of extraction, implying that elements in exchangeable fractions are most mobile and bioavailable, whereas those in residual fractions are tightly bound and are least mobile under natural conditions.¹⁴ The reagents, used in the current work, generally followed the sequence – FI: water; FII: inert electrolyte (NH₄OAc, pH of soil); FIII: weak acid (NH₄OAc, pH = 5); FIV: reducing agent (NH₂OH·HCl in HOAc); FV: oxidizing agent (H₂O₂); FVI: strong acid (HNO₃). Fractions FI-FV were referred to as non-residual reversibly bound fractions, labile or potentially available under certain environmental conditions. Fraction FVI was regarded as residual and poorly transferred during a short-time scale under natural conditions. Detailed description of the procedure, reactants and experimental conditions is given by Salbu.²³ Differences in fractionation results between soil samples from the legacy NORM and the undisturbed naturally ²³²Th-rich sites were evaluated.

Measurement of radionuclide and trace element concentrations in extracts was done by ICP-MS (Perkin Elmer Sciex, ELAN 6000). Blanks, calibration standards and standard reference materials were used throughout the analysis to ensure good quality of the data. Detection limits were calculated as three times the value obtained for blank samples. In addition, the quality control was done by measuring a certain number of same soil samples by gamma spectrometry. Sample preparation and method description is given by Mrdakovic Popic et al.¹⁹

In order to study the element transfers to soil pore water and hence the possibility of the further mobilization, the distribution coefficients (K_d , L/kg) were determined using the equation:

$$K_d = \text{concentration of element in soil solid phase} / \text{concentration of element in soil solution}$$

The laboratory batch method for K_d determination was used.²⁸ The concentrations of investigated elements in aqueous and solid soil fractions were measured by ICP-MS. Furthermore, dry soil samples from the study sites were screened for heterogeneities using the digital phosphor imaging (Typhoon 8600, Molecular Dynamics).

40 Data analyses

Statistical analysis of data was performed by MiniTab 16 (Minitab Inc.). Detection of outliers was performed using the box-whisker plots. The Anderson–Darling test was applied to test the normality of radionuclide and trace element concentrations in soil samples. Differences among soil samples were evaluated by ANOVA (with Tukey's post-hoc analyses) of log-transformed data. The criterion of significance was $p < 0.05$.

50 Multivariate statistics

Principal component analysis (PCA) was conducted using the CANOCO 5 software package. PCA was performed to evaluate the overall variation and patterns in the data related to the element concentrations obtained from the various sequential extraction steps. In addition, the PCA was performed on the sequential extraction steps FIII – FVI and the soil parameters.

The data set consisted of many samples, especially in sequential extraction step I and II, with element concentrations below LOD. To evaluate such data, a commonly applied method is to substitute samples with concentrations below LOD with LOD·0.5. However, if a chemical compound/element appears with concentrations below LOD in too many of the samples, it should be excluded from the analysis. Hence, in the overall evaluation of the various extraction steps all samples were included, but in the detailed PCAs on samples from each extraction step, only the steps FIII–FVI were included. The data was $\log(x+1)$ transformed to reduce the effects of extreme values, and in addition, centred and standardized (i.e., bringing their means to zero and their variance to one).

70 Results and discussion

Data quality

The results obtained for the standard reference material (NCS DC 73325-soil) were within 10% of the certified values for all investigated elements. Good reproducibility of results was evident from the relative standard deviations (RSD) lower than 5%. The efficiency of radionuclide and trace element recovery attained by sequential extraction was evaluated by comparing the cumulative concentrations from all extraction steps with values obtained by total digestion of soil from the same sampling points. Satisfactory recovery below 15% was obtained.

The quality control was additionally performed by measuring samples with ICP-MS and gamma spectrometry in parallel. The obtained results were comparable, i.e., differences ranged between 7.6 – 18 % and 5.2 – 10.0 %, for ²³²Th and ²³⁸U, respectively. Higher values obtained by gamma spectrometry should be assigned to differences in methods (non-destructive versus destructive) and heterogeneity inside the samples rather than to lack of quality procedures. Gamma spectrometry as non-destructive method measures total soil activity concentrations of radionuclides (in all soil fractions), while ICP-MS measures concentrations of those radionuclides present in decomposed part of material, i.e., in digested soil solution. Since HF was not used in the current work, soil silicates remained non-dissolved and some underestimation in ICP-MS results (and hence discrepancy between results from two methods) must be considered.

Table 1 Basic soil parameters (n = 10)

Site	pH	OM (%)	Sand (%)	Silt (%)	Clay (%)	CEC (cmol/kg)	Fe (g/kg)	Mn (g/kg)
Søve, former mining site	8.0	7.0	87.5	11.7	0.8	22.8	5	2
Fen, former mining site	6.9	19.7	33.1	52.7	14.2	48.8	12	1
Rullekoll, undisturbed ²³² Th-rich site	6.0	19.1	40.7	46.9	12.4	40.0	13	3

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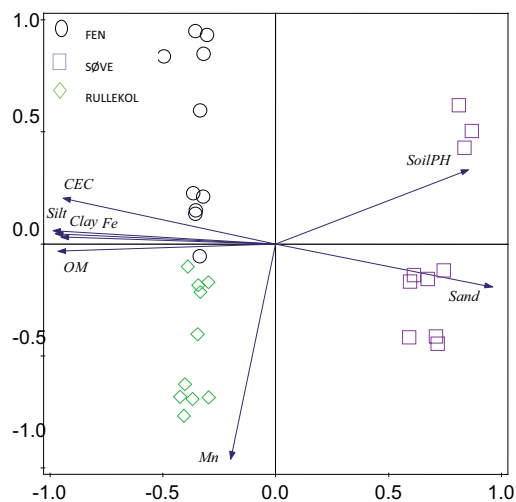


Fig.1 PCA bi-plot of soil characteristics at the former mining sites Fen and Søve, and at the undisturbed ^{232}Th -rich site Rullekoll.

Radionuclides and trace elements in soil

Basic soil parameters (pH, OM, CEC, Fe, Mn and soil texture) are given in Table 1. The similarities between the former mining site Fen and the undisturbed ^{232}Th -rich site Rullekoll, as well as their difference with the former mining site Søve, were demonstrated by principal components analysis (PCA) (Fig.1).

Analysis showed that PCA axis 1 described 79% of the total variation in the soil data. Soil pH and percentage of sand fraction were positively correlated with PCA axis 1 and were higher in samples from the former mining site Søve. Other parameters CEC, OM, silt and clay fraction and Fe content were negatively correlated with PCA axis 1 and appeared dominant for the samples from the former mining site Fen and to less extent for samples from the undisturbed ^{232}Th -rich site Rullekoll.

Soil concentrations of radionuclides and trace elements are shown in Table 2. The median values of ^{232}Th concentration in soils (329 – 1401 mg/kg) were elevated in comparison with the world average soil concentration (11 mg/kg)^{29,30} and exceeded the screening value for radioactive waste materials in Norway (1 Bq/

g, approx. 245 mg/kg).¹⁸ The wide ranges of ^{232}Th concentration were demonstrated for each of the investigated sites. No significant differences in ^{232}Th concentrations between the sites were demonstrated by ANOVA.

The pooled concentration range for ^{238}U (3 – 175 mg/kg) in soil was much narrower than the range for ^{232}Th (15 – 1685 mg/kg). Although ^{238}U concentrations in most samples were above the world average (3 mg/kg),^{29,30} the screening value (1 Bq/g, approx. 81 mg/kg)¹⁸ for radioactive waste was exceeded only in a few samples taken at the former mining site Søve. The median concentrations of Cr (14 – 97 mg/kg), As (8 – 18 mg/kg) and Pb (80 – 156 mg/kg) were found to be elevated (with the exception of Cr in soil at site Søve) with respect to the screening values for polluted soil in Norway (50, 8 and 60 mg/kg for Cr, As and Pb, respectively).³¹ No significant differences in ^{238}U and trace element concentrations between investigated sites were observed.

Digital autoradiography

Significant variation of the total soil concentrations in samples within sites suggested inhomogeneous spatial distribution of the radionuclides both at legacy NORM and undisturbed ^{232}Th -rich sites. Digital autoradiograms confirmed heterogeneity at macro-level and demonstrated the presence of radioactive particles both at former mining sites and at undisturbed natural site (Fig. 2).

Both macro-level heterogeneity of samples and spatial inhomogeneous distribution of radionuclides seen at each of the sites should be contributed to the mineralogical composition of the bedrock. Søvite, Rødbergite and Rauhaugite are the dominant rocks in the bedrock of the former mining site Søve, the former mining site Fen and the undisturbed ^{232}Th -rich site Rullekoll, respectively. The lowest degree of radioactive heterogeneities seen in samples from the site Søve was in accordance with previously demonstrated lower radionuclide levels in the rock Søvite than radionuclide levels in the rocks Rødbergite and Rauhaugite.¹² Small (μm size) mineral grains and inclusions of monazite and thorianite have previously been identified in the Fen Complex rocks by Berg et al.³² and Dahlgren.³³ Weathering of surface parts of the rocks containing such minerals led to the inhomogeneous environmental radionuclide distribution.¹² Further isolation and characterization of the radioactive particles from the former mining sites in the Fen Complex on micrometer scale is in progress.

Table 2 Radionuclide and trace element concentrations in soil (mg/kg d.w.) (median values accompanied with ranges in parentheses; n = 12 – 23)

Site	^{232}Th	^{238}U	Cr	As	Pb
Søve, former mining site	329	87	14	8	80
	(15 – 720)	(3 – 175)	(4 – 78)	(3 – 11)	(31 – 120)
Fen, former mining site	507	8	66	16	156
	(391 – 846)	(5 – 12)	(20 – 91)	(14 – 45)	(120 – 226)
Rullekoll, undisturbed	1401	23	97	18	143
^{232}Th -rich site	(1105 – 1685)	(7 – 42)	(18 – 104)	(13 – 32)	(74 – 358)

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Vertical distribution of radionuclides and trace elements in soil

The variation of radionuclide and trace element concentrations in soil with sampling depth is given in Fig. 3. The pattern of vertical distribution for ^{232}Th and ^{238}U was similar at each of the investigated sites. The surface soil layer (0-5 cm) at the former mining site Søve had, as expected, significantly lower concentrations ($p < 0.05$) of both ^{232}Th (16 mg/kg) and ^{238}U (3 mg/kg) in comparison to deeper soil layers (256 – 655 mg/kg of ^{232}Th and 83 – 153 mg/kg of ^{238}U), suggesting that remedial cover layers exist intact at sampling points in the current study. Deeper soil layers showed variable radionuclide concentrations likely due to the mixing of waste materials from previous mining activities, which are enriched in both ^{232}Th and ^{238}U . The presence of different minerals with variable radionuclide contents might also contribute to variation of radionuclide concentrations in soil depth profile at the former mining site Søve.

The vertical distribution of ^{232}Th and ^{238}U at the former mining site Fen (404 – 714 mg/kg and 6 – 11 mg/kg, respectively) and at the undisturbed ^{232}Th -rich site Rullekoll (1128 – 1322 mg/kg and 23 – 28 mg/kg, respectively) was relatively even. The highest concentrations of both radionuclides were determined at the deepest investigated soil layers where fragments of undisturbed parental Rødbergite and Rauhaugite rock, respectively, were dominant.

The analysis of trace elements vertical distribution showed enhanced Cr (68 mg/kg) and As (30 mg/kg) concentration in surface soil layers at the former mining sites Søve and Fen, respectively, suggesting possible surface enrichment through past mining activities. At deeper soil layers, as well as in all soil layers at the undisturbed ^{232}Th -rich site Rullekoll, no significant difference in trace elements concentrations was seen.

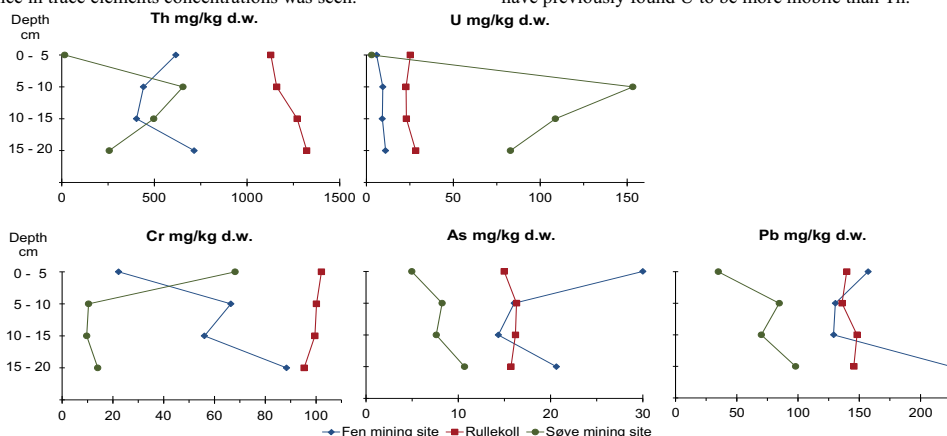


Fig. 3 Variation of radionuclide and trace element concentrations with sampling depth at former mining sites Fen and Søve, and at undisturbed ^{232}Th rich site Rullekoll.

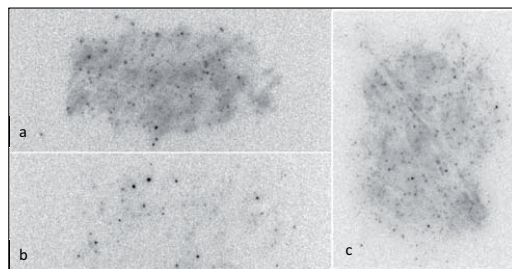


Fig. 2 Heterogeneous distribution of radionuclides demonstrated by digital autoradiograms of soil samples from a) former mining site Fen b) former mining site Søve c) undisturbed ^{232}Th -rich site Rullekoll.

Results of sequential extractions

The sequential extraction procedure was applied for the Fen Complex soils with the results presented in Fig. 4. Thorium in soil samples from three sites was predominantly strong associated with soil mineral fraction; between 77 and 94% were obtained in the irreversible soil fraction, after attack with concentrated HNO_3 at high temperature. This accorded to ^{232}Th chemical nature, as it is considered to be one of the least mobile elements in continental weathering.³⁴ Thorium has high sorption tendency, it is tightly bound to the soil or sediment fractions and increased removal is anticipated only in extremely acid environments.³⁵ Nevertheless, considering the high total ^{232}Th concentration in soil samples, potentially mobile ^{232}Th should not be neglected at the former mining site Fen.

Rather different binding of ^{238}U in comparison to ^{232}Th was demonstrated in soil samples, showing the lower percentages (38 – 69%) of ^{238}U irreversibly bound in soil. Similarly, other studies have previously found U to be more mobile than Th.^{34,36,37}

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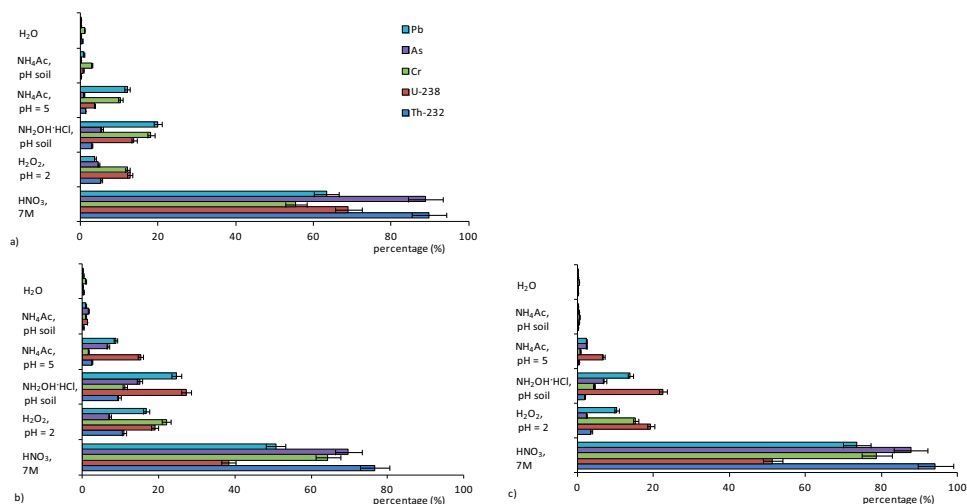


Fig. 4 Sequential extraction of soil from a) former mining site Søve b) former mining site Fen c) undisturbed ^{232}Th rich site Rullekoll ($n = 10$).

Under oxidizing conditions, ^{238}U is present as uranyl ion (UO_2^{2+}) that can form water-soluble complexes depending on pH and other present ligands.^{38,39} It has been explained that complex sorption and dissolution mechanisms determine the environmental behaviour of ^{238}U .^{38,40,41} These mechanisms depend on various factors such as soil pH, Eh, Fe- and Mn-content, organic compounds, clays, different ions, complex ligands.^{38,40-43} In soil with $\text{pH} \geq 6$ and in presence of CO_3^{2-} , ^{238}U forms uranyl-carbonates that are readily dissolved in water.⁴¹ It was previously demonstrated that these parameters could have an important role in the ^{238}U behaviour in the Fen Complex soils.⁴⁴ Marked difference in ^{238}U soil distribution ($p = 0.026$) was observed at the former mining site Fen, where up to 16% of the total ^{238}U was readily available in the environment (extracted in FI-FIII). Mobilization and further transport of ^{238}U could be expected at this legacy NORM site.

Regarding the trace elements, As was mainly irreversibly bound in soil, while Cr and Pb were associated with redox-sensitive amorphous fractions and organic compounds in soil. Having compared sequential extraction results between sites, stronger association of investigated radionuclides and trace elements in soil was observed at the undisturbed ^{232}Th -rich site Rullekoll than in soils from the former mining sites Fen and Søve.

More detailed description of the identified associations of radionuclides and trace elements with soil fractions, relationships among found associations and soil basic characteristics, as well as the description of differences among the sites were provided by performing principal component analysis (PCA). Analysis

performed on the complete data set (sequential extraction step FI-FVI) is presented in Fig. 5. The data were well described by the PCA, and 80% of the total variation was assigned to the first PCA axis. This indicated a strong gradient in the data (Table 3), i.e., the element concentrations generally increased with the increase of sequential extraction step (Fig. 5 A and B). Similar behaviour of ^{232}Th and Cr towards soil fractions was indicated by their positive correlation along PCA axis 1.

Furthermore, the PCA was done separately for each of the data sets obtained for element concentrations in FIII-FVI fractions of the sequential extractions. Correlations of elements and sampling sites along the PCA axis are given in bi-plots (Fig. 6). The PCA on data from sequential extraction step III showed that PCA axis 1 and 2 described 48 and 23% of the total variation, respectively (Table 3). Thorium, ^{238}U and As were positively correlated with PCA axis 1 indicating a slightly higher concentrations of these elements in samples from the undisturbed ^{232}Th -rich site Rullekoll and the former mining site Fen than in samples from the former mining site Søve. In contrast, Pb was negatively correlated with PCA axis 1 and appeared at higher levels in samples from the former mining site Søve. The ion exchangeable fraction and carbonates (pH sensitive NH_4Ac soil fraction) are expected to be released in step FIII, and enhanced concentrations of ^{232}Th , ^{238}U and As in samples from the former mining site Fen may be explained by soil properties to some degree. For example, CEC appeared higher in samples from the former mining site Fen and the undisturbed ^{232}Th -rich site Rullekoll compared to the samples from the former mining site Søve (Fig. 1), suggesting the possibility for higher concentrations of exchangeable

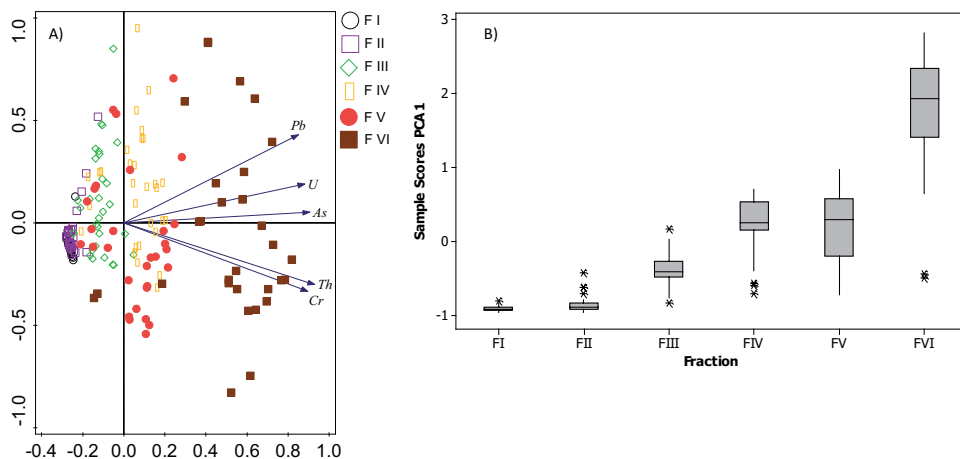


Fig. 5 A) Bi-plot displaying the results obtained from the PCA using all data from the sequential extraction FI-FVI. B) Sample scores are grouped according to their respective sequential extraction step obtained from PCA axis 1. Increasing sample score indicates increasing element concentrations.

radionuclides and trace elements to be found at these sites in FIII fraction. Furthermore, as presented in Fig. 6., Cr concentrations appeared to be higher at the former mining site Søvle than at the other two sites. Based on alkaline soil pH and sand texture seen at the former mining site Søvle (Fig. 1), we could assume dominant adsorption of Cr to soil particles and hydrolysed alkaline species at this site, as it has been described previously for other locations.^{45,46} This kind of sorption is highly dependent on pH.⁴⁶ Thus, the change in pH (that happens by adding acid in FIII phase of sequential extraction) could then lead to increase of Cr solubility and mobilization at the former mining site Søvle.

Principal component analysis of data from FIV showed that PCA axis 1 and 2 described 43 and 27% of the total variation, respectively (Table 3). Analyzed elements were negatively correlated with PCA axis 1 (Fig. 6). High concentrations of ²³⁸U and Pb in samples from undisturbed ²³²Th-rich site Rullekoll and ²³²Th, As and Cr in samples from former mining site Fen were indicated. High Fe soil contents at Fen and Rullekoll sites, seen in Fig. 1., could mean the elevated presence of Fe amorphous oxides and hence higher degree of radionuclides and trace elements association with it at these two sites in comparison to former mining site Søvle with lower Fe content in soil.

Table 3 Explained variation in % for PCA axes 1, 2 and cumulative explained variation

PCA Model	Explained variation (%)		Total
	PCA axis 1	PCA axis 2	
F I-VI	80	9	89
F III	48	23	71
F IV	43	27	70
F V	46	26	72
F VI	56	25	81
Soil Data	79	14	93

Decomposition of organic matter in soil samples released the radionuclides and trace elements associated with it in step FV of the sequential extraction. PCA axis 1 and 2 explained 46 and 26% of the variation in the data, respectively. Positive correlation of ²³⁸U, As and Pb and correlation of ²³²Th and Cr is also clear from the PCA bi-plot (Fig.6). The higher concentrations ²³⁸U, As and Pb and ²³²Th and Cr at former mining site Fen and undisturbed ²³²Th-rich site Rullekoll were indicated. Similarly high organic matter content at these sites most likely (Fig.1) contributed to the result.

Analysis of extraction data from residual soil fraction (FVI) showed that PCA axis 1 and PCA axis 2 described 56 and 25% of the total variability in data, respectively (Fig.6). All analyzed elements were negatively correlated with PCA axis 1. As in previous phase, ²³²Th and Cr were correlated and appeared to be at high percentages in soils from the undisturbed ²³²Th-rich site Rullekoll.

Distribution coefficients

Soil sorption of an element determines its fate in the environment, from mobilization and transport through underground water to uptake in plants. According to previous studies,^{47,48} the sorption is a combination of processes that are element- and soil-type dependent and affected by various factors, such as soil texture, soil pH, mineral composition, Fe oxide and hydroxide content, content of OM, phosphates, carbonates, CEC and their combination. The K_d is one of the parameters (e.g., TFs, CRs, Fm/f, BCF) commonly used to describe element partitioning between environmental compartments. It is based on the equilibrium concept assuming that an element in solid soil phase is in equilibrium with its content in liquid soil phase.⁴⁹ However, the uncertainty about the achieved equilibrium should be acknowledged since concept takes no explicit account of sorption mechanisms and aging effect which are known to affect the

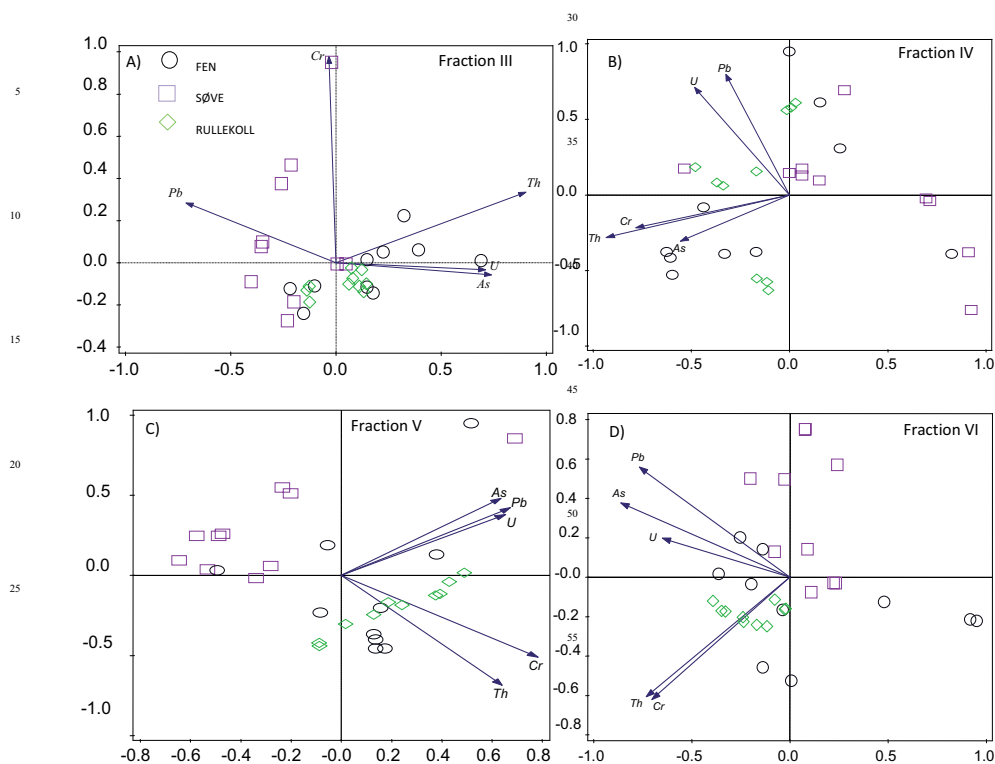


Fig. 6 PCA bi-plots showing radionuclides and trace elements in different sequential extraction steps (FIII-FVI) together with sampling sites.

magnitude of K_d . The samples included in the current analysis were taken from the Fen Complex sites either under undisturbed environmental conditions (Rullekoll site) or at sites where mining activities were finished before the long time period (i.e., 40 years) (former mining sites Fen and Søve) and analyzed by the commonly used batch equilibrium method.²⁸ As given by Strandberg and Fortkamp,⁵⁰ this method has both advantages (e.g., widely spread method that allows comparing to already published data, fast and cheap method, no chemicals are involved that are not already in the soil) and disadvantages (e.g., solid-solution ratios could influence the results, the questionable equilibrium time, difficulty of extracting of all the supernatant from soil rest after the centrifugation). In the current study, the solid : solution ratio 1:2 and contact time of 72 h was applied to ensure the equilibrium between radionuclides (trace elements) in the aqueous and solid phase. Although these conditions are commonly applied in many studies worldwide,²⁸ certain uncertainty related to the K_d concept and here applied method exist. According to IAEA,⁴⁹ the equilibrium concept could be especially justified in field samples with high radionuclide concentrations. However, the authors emphasize that K_d quantified by the approach used in this study should be

considered as the apparent K_{ds} . The presented K_{ds} could be higher than those resulted from a laboratory sorption test with additionally added radionuclide (trace element), because the radionuclide (trace element) quantified in the soil in the present work included also those tightly bound in soil fractions and not available for exchange with the soil solution due to the fact that the primary source of radionuclides (trace elements) is from bedrock minerals weathering.

Distribution coefficients (K_d) of radionuclides and trace elements in soils from the Fen Complex are shown in Table 4. The major natural form of ^{232}Th is the ion $^{232}\text{Th}^{4+}$ which is very poorly soluble in natural water.²⁸ Important factors affecting the sorption and dissolution of ^{232}Th are the presence of Fe- and Mn-oxides, organic compounds and pH.^{47,48,51} The K_d (^{232}Th) values were statistically different ($p = 0.03$) regarding different investigated sites. Minimal sorption was determined for the former mining site Søve (346 L/kg) while maximal sorption was observed at undisturbed ^{232}Th -rich site Rullekoll (10672 L/kg). As stated earlier,^{40,48,52} the available soil characteristics should be considered for better understanding of K_d values. Current study K_d (^{232}Th) values suggested lower sorption at the former mining site Søve with a dominant sand structure (Table 1) (87.5%) of

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Table 4 Distribution coefficients (K_d , L/kg) as arithmetic and geometric means with standard deviations (n = 6)

Site	Parameter	^{232}Th	^{238}U	Cr	As	Pb
Søve, former mining site	AM \pm SD	372 \pm 424	155 \pm 138	27 \pm 22	75 \pm 53	279 \pm 91
	GM \pm GSD	346 \pm 2 (a)	100 \pm 3 (a)	23 \pm 2 (a)	58 \pm 2 (a)	182 \pm 3 (a)
Fen, former mining site	AM \pm SD	5620 \pm 4668	277 \pm 316	8 \pm 7	25 \pm 11	427 \pm 515
	GM \pm GSD	4173 \pm 2 (b)	177 \pm 3 (a)	6 \pm 2 (a)	22 \pm 2 (a)	239 \pm 4 (a)
Rullekoll, undisturbed ^{232}Th -rich	AM \pm SD	10753 \pm 1436	1267 \pm 2153	16 \pm 12	108 \pm 55	969 \pm 689
	GM \pm GSD	10672 \pm 1 (c)	506 \pm 4 (b)	13 \pm 2 (a)	94 \pm 2 (b)	762 \pm 2 (b)

^a Geometrical means marked with different letters are significantly different (ANOVA, $p < 0.05$).

the soil, while K_d (^{232}Th) was one order of magnitude higher at the former mining site Fen and at the undisturbed ^{232}Th -rich site Rullekoll with more loam in their soil texture (52.7 and 46.9%, respectively). Furthermore, higher OM, Fe- and Mn-content in soil at the former mining site Fen and the undisturbed ^{232}Th -rich site Rullekoll than in soil at the former mining site Søve might be considered as important binding factors leading to higher ^{232}Th sorption. Comparison of K_d values grouped according to soil pH values demonstrated lower K_d (^{232}Th) for the former mining site Søve soil samples (pH = 8) than for the former mining site Fen and the undisturbed ^{232}Th -rich site samples (pH = 6-7). These results accorded to the previously published dependence and relationship between K_d (^{232}Th) and soil pH.^{28,48} The magnitude of K_d (^{232}Th) geometric means was also comparable to wide ranges of K_{ds} published in compilation given for natural non-contaminated soils by Vandenhove et al.⁴⁸ and in reports of EPA²⁸ and IAEA.⁴⁹ However, it has been acknowledged that reliable data on K_d (^{232}Th) is still scarce and majority of published values come from reports and papers from different field and laboratory experiments, from 1990 onwards, sometimes with contrasting experimental conditions applied, without soil characterization or with limited sample number.⁴⁸ According to K_d (^{232}Th) values for the Fen Complex sites, minimal transportation and mobility towards biota should be expected at the undisturbed naturally ^{232}Th -rich site Rullekoll. Similarly as the sequential extraction results, K_d (^{232}Th) values indicated low mobility and bioavailability at this site, despite the high total soil concentration. This is important in terms of the environmental risk. On the contrary, based on K_d values, ^{232}Th availability was enhanced at the former mining sites, especially at site Søve. This again was in accordance with the soil ^{232}Th distribution demonstrated by the sequential extraction and most probably is the consequence of mining activities from the past.

As stated earlier, the behaviour of ^{238}U in the environment is affected by specific combinations of soil physical and chemical parameters.^{38,40,41,43,53} Although soil texture, OM and pH were traditionally considered as determining for soil sorption and K_d (^{238}U) magnitude,^{41,44,48} some recent studies and compilation analysis showed no simple relationship between soil texture or organics and K_d (^{238}U) values.^{28,52,54} In the current study, comparably lower K_d (^{238}U) values were found for soil from the two former mining sites (100 and 177 L/kg at site Søve and Fen,

respectively), while the significantly higher K_d (^{238}U) values for soil from the undisturbed ^{232}Th -rich site Rullekoll (506 L/kg) ($p = 0.003$). Such difference suggested greater sorption and lower bioavailability at the undisturbed natural site. Previously proposed K_d (^{238}U) values, depending on soil parameters, were 40, 200, 200 and 2000 L/kg for sand, loam, clay and organics, respectively, according to Sheppard et al.⁵² For the same soil classification, values given by Vandenhove et al.⁴⁸ were 110, 310, 28 and 1200 L/kg, respectively. The current study K_{ds} (^{238}U) for sandy (site Søve) and sandy-loam (sites Fen and Rullekoll) soil were in accordance with the above listed results. When K_{ds} (^{238}U) were grouped with respect to soil OM (low OM at site Søve and high at sites Fen and Rullekoll), lower K_d (^{238}U) values were determined in the present study in comparison to literature data.^{48,52,54} Values of K_d (^{238}U) in natural soils, given by Vandenhove et al.,⁴⁸ were one order of magnitude higher in soil with $5 < \text{pH} < 7$ than in soil of $\text{pH} > 7$. Similarly, higher K_{ds} (^{238}U) were demonstrated for soil from the former mining site Fen with pH 6.9 and from the undisturbed ^{232}Th -rich site Rullekoll with soil pH 6.0. than in soil from the former mining site Søve with pH 8. Furthermore, results from the present work were in accordance with K_{ds} (^{238}U) given in the TRS-472 by IAEA.⁴⁹

The distribution coefficients of Cr and As were one to three orders of magnitude lower than those for radionuclides ^{232}Th and ^{238}U , whereas K_d values for Pb were comparable to those found for ^{238}U . Based only on K_d results, higher mobilization of trace elements Cr and As than the mobilization of the radionuclides from soil towards other environmental compartments could be expected. No statistically significant difference in K_{ds} (Cr, Pb) was observed between the investigated sites, while K_d (As) at the undisturbed ^{232}Th -rich site Rullekoll was significantly higher ($p = 0.037$) than at the two former mining sites.

Conclusion

Radionuclides (^{232}Th and ^{238}U) and trace elements (Cr, As and Pb) in soils from two legacy NORM and one undisturbed naturally ^{232}Th -rich sites in the Fen Complex area were analyzed to obtain distribution and mobility data within the ongoing environmental impact assessment study. Inhomogeneous soil distribution of radionuclides with significantly elevated ^{232}Th (up to 1685 mg/kg) and moderate ^{238}U (up to 175 mg/kg)

concentrations was observed. Soil heterogeneity at macro-level was demonstrated by digital autoradiography imaging. Trace element concentration ranges in soil were wide (i.e., from 4 to 104; 3 to 45 and 31 to 358 mg/kg for Cr, As and Pb, respectively). Variation of concentrations in vertical soil profiles were attributed to both different minerals present in bedrock layers (Fen and Rullekoll sites) and to waste from mining activities in the past (Søve site). Results from the sequential extractions showed the majority of the ^{232}Th and trace elements in the residual soil fraction. Uranium was found to be more mobile, especially at the former mining site Fen where 15% of ^{238}U was present in soil as easily available. Although no differences in total soil concentrations of radionuclides and trace elements between the sites were observed, the elevated availability was confirmed at the legacy NORM sites in comparison to the undisturbed ^{232}Th -rich site. Similarly, the distribution coefficients K_d (^{232}Th) and K_d (^{238}U) suggested enhanced mobility at the former mining sites, especially at site Søve, while higher sorption of radionuclides at the undisturbed ^{232}Th -rich site Rullekoll. Hence, mobility and possible elevated bioavailability at the legacy NORM sites could be expected and further risk assessment should take this into account when decisions on intervention are made.

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Paper III

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Ecological transfer of radionuclides and metals to free-living earthworm species in natural habitats rich in NORM

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ABSTRACT

Transfer of radionuclides (^{232}Th and ^{238}U) and associated metals (As, Cd, Pb and Cr) from soil to free-living earthworm species was investigated in a thorium (^{232}Th) rich area in Norway. Sampling took place within former mining sites representing the technologically enhanced naturally occurring radioactive materials (TENORM), at undisturbed site with unique bedrock geology representing the naturally occurring radioactive materials (NORM) and at site outside the ^{232}Th rich area taken as reference Background site. Soil analysis revealed the elevated levels of investigated elements at NORM and TENORM sites. Based on sequential extraction, uranium (^{238}U) and cadmium (Cd) were quite mobile, while the other elements were strongly associated with mineral components of soil. Four investigated earthworm species (*Aporrectodea caliginosa*, *Aporrectodea rosea*, *Dendrodrilus rubidus* and *Lumbricus rubellus*) showed large individual variability in the accumulation of radionuclides and metals. Differences in uptake by epigeic and endogeic species, as well as differences within same species from the NORM, TENORM and Background sites were also seen. Based on total concentrations in soil, the transfer factors (TF) were in ranges 0.03–0.08 and 0.09–0.25, for ^{232}Th and ^{238}U , respectively. TFs for lead (Pb), chromium (Cr) and arsenic (As) were low (less than 0.5), while TFs for Cd were higher (about 10).

Using the ERICA tool, the estimated radiation exposure dose rate of the earthworms ranged from 2.2 to 3.9 $\mu\text{Gy/h}$. The radiological risk for investigated earthworms was low (0.28).

The obtained results demonstrated that free-living earthworm species can survive in soil containing elevated ^{232}Th and ^{238}U , as well as As, Cd, Pb and Cr levels, although certain amount of radionuclides was accumulated within their bodies.

The present investigation contributes to general better understanding of complex soil-to-biota transfer processes of radionuclides and metals and to assessment of risk for non-human species in the ecosystem with multiple contaminants.

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1. Introduction

The knowledge gap and the need for comprehensive data on naturally occurring radionuclides in terrestrial ecosystems have been reported and emphasized (Beresford et al., 2008a; Gomez-Ros et al., 2004; Jones et al., 2003; Pentreath, 1999, 2002). It has been suggested that, when assessing radiation impacts on non-human biota, estimated dose rates due to anthropogenically released radionuclides should be compared to dose rates from natural background radiation normally experienced by animals and plants (Beresford et al., 2008a; Pentreath, 1999, 2002). However, radionuclides in polluted environmental conditions rarely occur alone, but usually in a mixture with a variety of other chemicals (e.g., heavy metals, persistent organic pollutants, solvents, pesticides), making the exposure situation

complex. As multiple stressors can affect living organisms in mixed contaminated environments, causing different combinations of interactions and, consequently, diverse combined biological effects (Eggen et al., 2004; Salbu et al., 2005), the analytical strategy should not be restricted to radionuclides only.

A recent investigation of radionuclide and metal concentrations in a thorium rich Fen Complex area in Norway demonstrated a presence of significantly elevated soil levels of ^{232}Th (up to 6581 Bq/kg) and slightly elevated levels of ^{238}U (up to 130 Bq/kg) and metals As, Cr, Pb and Cd (20, 89, 287 and 1.88 mg/kg, respectively) (Mrdakovic Popic et al., 2011). Based on soil analysis, the possibility for biota simultaneous uptake of multiple contaminants (radionuclides ^{232}Th , ^{238}U and their daughters and series of metals) from Fen Complex soil could not be excluded. With respect to that, the aim of the current study was to investigate the transfer and potential bioaccumulation of ^{232}Th , ^{238}U , Cr, As, Cd and Pb from soil into the earthworms.

Depending on characteristics of ecosystem, up to 80% of the total biomass of soil fauna could be the earthworms (Ernst et al., 2008;

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Ireland, 1983; Lee, 1985). As key organisms responsible for mixing of soil constituents and maintaining the structure of soil, they are regarded as one of the most suitable animals for testing the toxicity of chemicals and have been adopted as standard organisms in ecotoxicological tests (Lee et al., 2008). Acute and chronic tests with contaminants have shown changes in different endpoints of earthworms, such as mortality, changes in biomass, reproduction rates and behavioral responses. Currently, the extensive database on the effects of different chemicals on earthworms exists (Lee et al., 2008).

Efficient accumulation of heavy metals from soil to different earthworm species has been demonstrated (Dai et al., 2004; Ernst et al., 2008; Langdon et al., 2001; Lee, 1985; Lukkari et al., 2006; Morgan and Morgan, 1999; Nahmani et al., 2007; Spurgeon et al., 2003). Since animals from many taxa including insects, amphibians, reptiles, birds and mammals prey on earthworms (Eijsackers, 2004; Gish and Christensen, 1973; Maleri et al., 2008), the contaminants do not necessarily affect only earthworms, but organisms at higher trophic levels of the food chain through biomagnifications (Reinecke, 1992). However, the uncertainty about the interpretation of the results and conclusions from studies commonly conducted in laboratory conditions or highly contaminated areas and about their use in the areas with low or moderate pollution levels has been emphasized (Vliet et al., 2005). Furthermore, few data on uptake of natural radionuclides or mixtures of radionuclides and metals by earthworms is available in literature.

In the present work, the ecological transfer of the radionuclides (^{232}Th , ^{238}U) and metals (As, Cr, Pb and Cd) into different free-living earthworm species has been investigated and the associated radiation exposure dose rates and radiological risk estimated. The study complies with increased emphasis put forward by the International Commission on Radiological Protection (ICRP) to monitor and demonstrate the protection of non-human species from ionizing radiation. Moreover, the most information on transfer to earthworms is based on controlled laboratory designed experiments with designated earthworms feed with artificial soil spiked with limited number of commercial radionuclides or metals. No information on transfer to free-living species under natural conditions in areas characterized by a heterogeneous mixture of NORM, TENORM and metals is available.

2. Materials and methods

2.1. Study area

The Fen Complex (FC), situated in south eastern Norway (from N 59° 16.40'; E09° 18.40' to N59° 16.90'; E09° 17.16') consists of carbonatite rocks of magmatic origin (Rödbergite, Rauhaugite, Sövite and Fenite). The area is rich in NORM, particularly in ^{232}Th (USGS, 2007). The high levels of ^{232}Th , ^{238}U , rare earth elements (REE) and metals, especially iron (Fe) and niobium (Nb) are due to the specific bedrock geology (Brøgger, 1921). Following intensive mining activities during the last few centuries, TENORM are also present in this area.

Previous studies in FC area have shown the elevated radiation exposure for humans due to radon inhalation (Stranden and Strand, 1986; Sundal and Strand, 2004). Recent investigations (Mrdakovic Popic et al., 2011; NGI, 2010) have demonstrated that soils from mines and forest within FC area had significantly elevated ^{232}Th and moderately to slightly elevated ^{238}U and metals Pb, Cd, As and Cr concentrations in comparison with soil outside the FC area.

For the investigation purposes of the current study, three sites within the circular FC that has an approximate diameter of three kilometers, and one reference Background site outside the FC, were chosen. NORM site was situated at the south end of FC, 2 km outside the mining areas, and represented the undisturbed natural conditions. Two TENORM sites (TENORM 1 and 2) were positioned in the north and central FC zones, respectively, with the distance of 500 m

between them. Site TENORM 1 comprised the disused Fe mining site along the shores of the Lake Nordsjø with abundance of waste rocks from Fe mining activities in the past. Site TENORM 2 comprised the wooden zone above the Fe mining site with bedrock rich in ródbergite and many visible disused mining holes and open tunnels. The Background site, chosen as reference site outside the naturally enhanced radiation area, was situated 5 km far from the FC. The radiation and geological maps (Barth and Ramberg, 1966; Dahlgren, 1983; Sundal and Strand, 2004), showing that Background site is positioned outside the area with specific bedrock geology of elevated radiation, as well as in-situ gamma dose rate measurements showing values comparable to the average Norwegian values (0.07–0.1 $\mu\text{Gy/h}$), were basis for Background site choice.

2.2. Sample collection and analytical procedures

Previous studies have suggested that polluted soil particles, soluble soil fractions, leaf and root litter are the main environmental pools of importance for uptake by earthworms (Ernst et al., 2008; Hobbelen et al., 2006; Spurgeon and Hopkin, 1996; Vliet et al., 2005). According to that, two field expeditions (May 2008 and September 2009) were organized to collect earthworms and corresponding soil, root and leaf litter samples. At each site, at least five sampling points were laid out, with an average distance of 5 m between them. Soil pits, 20×20×20 cm in size, for collection of earthworms and roots, were excavated at each sampling point. Earthworms were collected by hand sorting and transported in plastic bearers with moist soil to the laboratory for identification and analysis. The number of collected earthworms was 85 in total (adults and immature). Roots and leaf litter were separated into paper bags and taken to the laboratory for cleaning and analysis. Additionally, soil samples were taken at sampling points of each site using split corer (10 cm diameter).

Earthworm species were identified in the laboratory as *Aporrectodea caliginosa*, *Aporrectodea rosea*, *Lumbricus rubellus* and *Dendrodrilus rubidus*. *D. rubidus* and *L. rubellus* are epigeic earthworms, living in the upper soil layers and feeding mainly on leaf litter, surface soil and roots. In contrast, *A. caliginosa* and *A. rosea* are endogeic worms from deeper soil layers, feeding mainly on soil particles. The presence of different earthworm species (Sims and Gerard, 1985) allowed us to compare and investigate possible differences between species and correlations with different soil fractions, roots and leaf litter. All mature earthworms were starved on moisture filter paper for 48 h to deplete their gut contents and rinsed with distilled water to remove visible soil particles. They were then weighed for fresh earthworm masses, separately frozen in liquid nitrogen, freeze dried and milled into fine powder. Soil samples were dried at 40 °C for several weeks, crushed and homogenized by sieving through a 2 mm sieve. The physico-chemical characteristics were measured according to standard soil test methods. Samples of collected roots and leaf litter were cleaned from visible soil particles, washed with distilled water, dried at 40 °C and milled. Before the digestion procedure, samples of soil and root and leaf litter were oven dried (105 °C) to constant mass.

Earthworms, soil, root and leaf litter samples were acid microwave decomposed (MLS-Milestone UltraClave, MLS GmbH) with high purity grade concentrated nitric acid. The radionuclides and metals were measured in obtained solutions by ICP-MS (Perkin Elmer Elan 6000). An internal standard was systematically added to each sample before decomposition. Furthermore, standard reference material (NCS DC 73325), calibration standard, reagent and method blanks were used to ensure high quality performance. The measurements of the standard reference material were all within 10% of the certified value. The detection limits (LOD) were calculated as three times the standard deviation of blank sample measurements and they were: 0.015 (^{232}Th), 0.001 (^{238}U), 0.135 (As), 0.003 (Cd), 0.005 (Pb) and 0.380 (Cr) mg/kg d.w.

In addition to total analysis, the soil samples were subjected to sequential extraction according to Salbu (2000) (Table 1). The aim was to estimate the environmental availability of radionuclides and metals in soil. Water soluble and reversibly bound (NH_4OAc , soil pH + NH_4OAc , pH=5) soil fractions of radionuclides and metals were considered to reflect the environmental availability.

2.3. Data analyses

Statistical analyses were performed using Minitab 16 (Minitab Inc.). Normality of data was assessed with the Anderson–Darling test and not normalized data were \log_{10} transformed prior to statistical tests. The differences in radionuclide and metal concentrations between soil samples, as well as between earthworms, collected at different sites, were tested with ANOVA followed by Tukey's post hoc analyses. Correlation analyses were performed using the Pearson correlation coefficient. Regression analysis was used to express the accumulated ^{232}Th and ^{238}U in earthworms as a function of the following parameters: total soil concentration, water soluble and reversible bound soil concentrations, and root and leaf litter concentrations. For all the analyses, the p-value lower than 0.05 is considered statistically significant.

In order to quantify the transfer of investigated elements from soil to earthworms, the transfer factors (TF) and available transfer factors (ATF) were calculated according to:

$$\text{TF} = \text{earthworm concentration (mg/kg d.w.)} / \text{soil concentration (mg/kg d.w.)}$$

$$\text{ATF} = \text{earthworm concentration (mg/kg d.w.)} / \text{soil environmentally available fraction (mg/kg d.w.)}$$

Available transfer factors (ATF) (Baeza and Guillen, 2006) referred to the first three steps of the sequential extraction procedure. Although the TF concept based on total soil concentrations is widely used within ecology and radioecology, the ATF concept takes into consideration the speciation and actually available, mobile species and to certain degree better explains the capacity of an organism to bioaccumulate the contaminant (Salbu, 2000).

The ERICA tool (Environmental Risk from Ionizing Contaminants: Assessment and Management), tier 2, was used for calculation of radiation exposure dose rate in earthworms and radiological risk assessment (Brown et al., 2008; Larsson, 2008). Soil and earthworm radionuclide activity concentrations (Bq/kg d.w. and Bq/kg f.w., respectively), calculated concentration ratios (CR) and $10 \mu\text{Gy/h}$ as the screening dose rate confidence level that environmental risks are negligible (Andersson et al., 2009), were used in this approach.

3. Results and discussion

3.1. Soil analysis

Some of the soil characteristics are presented in Table 2. Values of pH showed no large differences between studied sites and ranged from 6.3 to 6.9. The organic matter content was high at all sites

(>20%), while moisture content was below 5%. Cation exchange capacity varied from 22.3 to 45.7 cmol/kg, while exchangeable Ca varied from 15.3 to 27.2 cmol/kg. The content of Fe and Mn ranged from 2 to 14 and from 0.3 to 3 g/kg, respectively. The carbonates and phosphates were obtained in the ranges of 0.9–10.9 and 0.08–0.46%, respectively.

Radionuclide and metal concentrations in soil are presented in Table 3. Total radionuclide concentrations in soil ranged from 101 to 954 mg/kg and from 3 to 7 mg/kg, for ^{232}Th and ^{238}U , respectively. The ^{232}Th concentrations were significantly higher than the world soil average of 8–12 mg/kg (UNSCEAR, 2000, 2008). Soil samples collected at NORM and TENORM sites exceeded the screening value for radioactive waste (1000 Bq/kg for ^{232}Th) given by the Norwegian Pollution Act (2011). The statistical analysis demonstrated that only ^{232}Th levels in soil at Background site were significantly lower ($p=0.026$), although even this reference site soil had elevated concentrations of ^{232}Th (up to 146 mg/kg) in comparison to normal background levels in areas with no enhanced natural radiation (12 mg/kg) (UNSCEAR, 2000, 2008). Although the ^{232}Th median values of soil samples from TENORM sites were lower than median value of samples from the NORM site, the statistically significant difference in soil concentrations between these sites was not observed, suggesting that radionuclide levels are reflection of the abundance of ^{232}Th in rocks and not of the expected enhancement due to mining activities in the past.

Values for ^{238}U were higher than the world average of 4 mg/kg, but still far below the screening value (1000 Bq/kg) for radioactive waste in Norway (Norwegian Pollution Act, 2011). No significant difference in soil ^{238}U concentrations between NORM, TENORM and Background sites was observed.

Generally, there were inhomogeneous distributions of both ^{232}Th and ^{238}U in soil. Wide concentration ranges of both radionuclides (Table 3) indicated the differences even between samples taken at the same site. This could be explained by variation in the abundance of ^{232}Th and ^{238}U rich soil particles originating from different rock types. Ranges of radionuclides comparable to current study results have been published in NORM and TENORM investigations worldwide (Malanca et al., 1993; Quindos et al., 1994; Termizi Ramli et al., 2005).

Total concentrations of As in soils ranged within 10–15 mg/kg on NORM and TENORM sites, which slightly exceeded the screening value for polluted soil in Norway (8 mg/kg) given by the Norwegian Pollution Control Authority (2009) as level that implies the need for more refined study analysis and risk assessment. Ranges of As comparable to this study ranges have been reported previously by NGI (2010) for site located in the Fen Complex area, in the vicinity to sites investigated in the present study. Differences in total soil concentration of Cr and Cd among individual soil samples taken at the same site were found to be large. That resulted in wide Cr and Cd ranges, but median values (9–57 and 0.70–0.97 mg/kg for Cr and Cd, respectively) were still below the screening limits for polluted soil (50 mg/kg for Cr and 1.5 mg/kg for Cd) (Norwegian Pollution Control Authority, 2009). Both Cr and Cd concentrations in soil were

Table 1
Sequential extraction procedure; description (Salbu, 2000).

Process	Model	Agents	Reagents
Physical sorption	Consecutive layers, reversible reaction	Indifferent electrolyte	H_2O HN_4OAc , pH of soil
Reversible chemisorption Electrostatic sorption	Reversible reaction	Ion-exchangeable species, increased pH	HN_4OAc , pH = 5
Irreversible chemisorption	Irreversible reaction	Red/Ox. agents, temperature increase	Weak reducing: $\text{NH}_2\text{OH} \cdot \text{HCl}$ Weak oxidizing: H_2O_2 , pH=2 Strong oxidizing: 7M HNO_3 , 80 °C

Table 2
Physico-chemical characteristics of soil.

Site	pH	H ₂ O (%)	OM (%)	CEC (cmol _c /kg)	Exch. Ca (cmol _c /kg)	P ₂ O ₅ (%)	CaCO ₃ (%)	Content of Fe (g/kg)	Content of Mn (g/kg)
NORM	6.6	4.3	20.1	35.7	27.2	0.24	3.5	12	2
TENORM 1	6.9	2.7	22.8	45.7	36.2	0.10	10.9	11	1
TENORM 2	6.3	4.8	24.5	34.5	26.7	0.08	2.4	14	3
Background	6.6	4.9	23.1	22.3	15.3	0.46	0.9	2	0.3

similar to ranges published earlier for Norwegian soil (Steinnes and Ruhling, 2002). In NORM and TENORM soil samples, Pb ranged from 70 to 159 mg/kg, exceeding the criterion for polluted soil concentration of 60 mg Pb/kg dry soil (Norwegian Pollution Control Authority, 2009), and indicating enrichment. This range is in accordance with results obtained for southern Norway (AMAP, 1998; Steinnes and Ruhling, 2002) and comparable to values reported for natural forest soils in Switzerland, Finland and Germany (Ernst et al., 2008; Lukkari et al., 2006; Rahtkens and von der Trenck, 2006).

In general, metal concentrations in soil were higher than the Norwegian screening levels above which soil should be considered as polluted. However, according to the Norwegian Pollution Control Authority further risk estimation with respect to exposure scenario should be performed to give the site specific acceptance criteria and to allow the final conclusions and decision making.

The environmental availability varied according to investigated radionuclides and metals (Fig. 1). The complex combination of physico-chemical conditions in soil controls the equilibrium between the soluble and adsorbed forms of radionuclides and metals and consequently their availability (Echevarria et al., 2001; Guo et al., 2008; Stojanovic et al., 2011; Vandenhove and Van Hees, 2007). Thorium was rather immobile with a large part (86.5–95.1%), associated with the crystalline soil fraction. Similarly, As, Cr and Pb, were mostly present in the irreversibly bound fraction (81.5–90.5%, 68.2–81.3% and 62.5–71.6%, respectively). These results suggested low bioavailability and transfer to biota under natural conditions. It seemed that the adsorption on organic matter and Fe and Mn oxides, all present at high levels, was dominant process in these soils. Almost neutral soil pH and precipitation on soil phosphates also contributed to lower solubility. Similar effects of pH, Fe and Mn oxides and organics have previously been demonstrated (Guo et al., 2008; Martínez and Motto, 2000; Syed, 1998; Vandenhove et al., 2009a). In contrast, the percentage of ²³⁸U in crystalline soil fraction was lower (39.2–52.5%), but considerable amount of ²³⁸U was released from the more available soil fractions by ion exchange reactions (7.5–13.5%), weak reducing (18.9–25.1) and oxidizing (17.6–22.0%) reagents. The variety and complexity of sorption/desorption mechanisms involved in the dynamics of ²³⁸U in soil have been described by many authors (Echevarria et al., 2001; Koch-Steindl and Pröhl, 2001; Langmuir, 1978; Stojanovic et al., 2011; Vandenhove et al., 2007; Zheng et al., 2003). Based on present soil physico-chemical parameters, the higher ²³⁸U solubility is, most likely, best explained by neutral pH and carbonates in soil. Koch-Steindl and Pröhl (2001) showed that the presence of dissolved carbonates and pH above 6 lead to formation of uranyl-carbonate complexes that are soluble and readily transported in soil. It seems that these parameters were

prevailing since other factors (organic matter, phosphate content, Fe and Mn content), that actually should contribute to lower ²³⁸U solubility, were relatively diminished. Similarly to ²³⁸U, Cd was also to a large extent potentially available (up to 84.2%). The higher mobility of Cd in comparison to many other heavy metals has been explained in literature by its weak affinity for soil colloids (Alloway, 1995). The additional solubility contributing factor for Fe soil might be exchangeable Ca²⁺. Research has shown that Cd²⁺ ion, with same charge and similar size, is in competition with Ca²⁺ for exchange sites in soil. However, when soil is saturated with Ca, consequently, mobilization and higher soil solution Cd concentrations are expected (Kookana and Naidu, 1998; Naidu et al., 1994).

3.2. Species specific radionuclide and metal accumulation in earthworms

The concentrations of investigated elements in four earthworm species pooled for all sites are given in Table 4. The concentration levels of elements in individual species collected at different sampling sites are shown in Fig. 2. Large variability between individual earthworms was found for all investigated radionuclides and metals (Table 4), which accords with previously published data (Ernst et al., 2008; Maleri et al., 2008; Morgan and Morgan, 1999; Nahmani et al., 2007; Vliet et al., 2005). The uptake of radionuclides and metals is a dynamic process where the individual earthworm organisms reach a maximum load, after which elimination starts. With respect to that, earthworm body concentration could be dependent on prevailing process of either active uptake or excretion (Vliet et al., 2005). It has also been reported that a kind of regulatory mechanism occurs in earthworms, leading to elimination of any higher body content of metal via excretion, and allowing that earthworms can survive in highly polluted environments (Kennette et al., 2002; Maleri et al., 2008). Moreover, different age and physiological status of the earthworms could also affect individual uptake to a certain level (Maleri et al., 2008).

The concentrations in individual species collected at different sites (Fig. 2) were compared in order to reveal differences in uptake within each species depending on the site characteristics.

The ²³²Th concentrations in earthworms collected within the whole FC area varied from 11 to 57 mg/kg. Species specific accumulation was observed (p=0.041). The found concentrations are significantly higher (up to 80 times) than earthworm ²³²Th concentration given in a study of natural background dose rates of wildlife in the UK (Beresford et al., 2008a). It was expected that endogeic species, which live in the mineral layers of soil on a diet consisting mainly of soil organic matter, would accumulate larger amounts of radionuclides and metals than epigeic, surface living earthworms

Table 3
Radionuclide and metal concentrations (mg/kg d.w.) in soil (median values and range (in parenthesis) are shown; n≥5).

Site	²³² Th ^a	²³⁸ U ^a	As	Cr	Cd	Pb
NORM	954 (566–964)	7 (4–10)	15 (11–20)	57 (15–76)	0.9 (0.8–1.1)	142 (46–168)
TENORM 1	557 (288–984)	4 (3–6)	10 (7–22)	45 (13–72)	1.1 (0.3–1.3)	70 (37–103)
TENORM 2	435 (411–2773)	6 (3–32)	14 (10–62)	17 (10–78)	0.9 (0.7–1.9)	159 (51–397)
Background	101 (39–126)	3 (1–3)	2 (1–3)	9 (9–17)	0.7 (0.2–0.8)	16 (12–26)

^a Activity concentrations (Bq/kg) can be calculated by multiplying given concentrations (mg/kg) with factors 4.06 and 12.35 for ²³²Th and ²³⁸U, respectively.

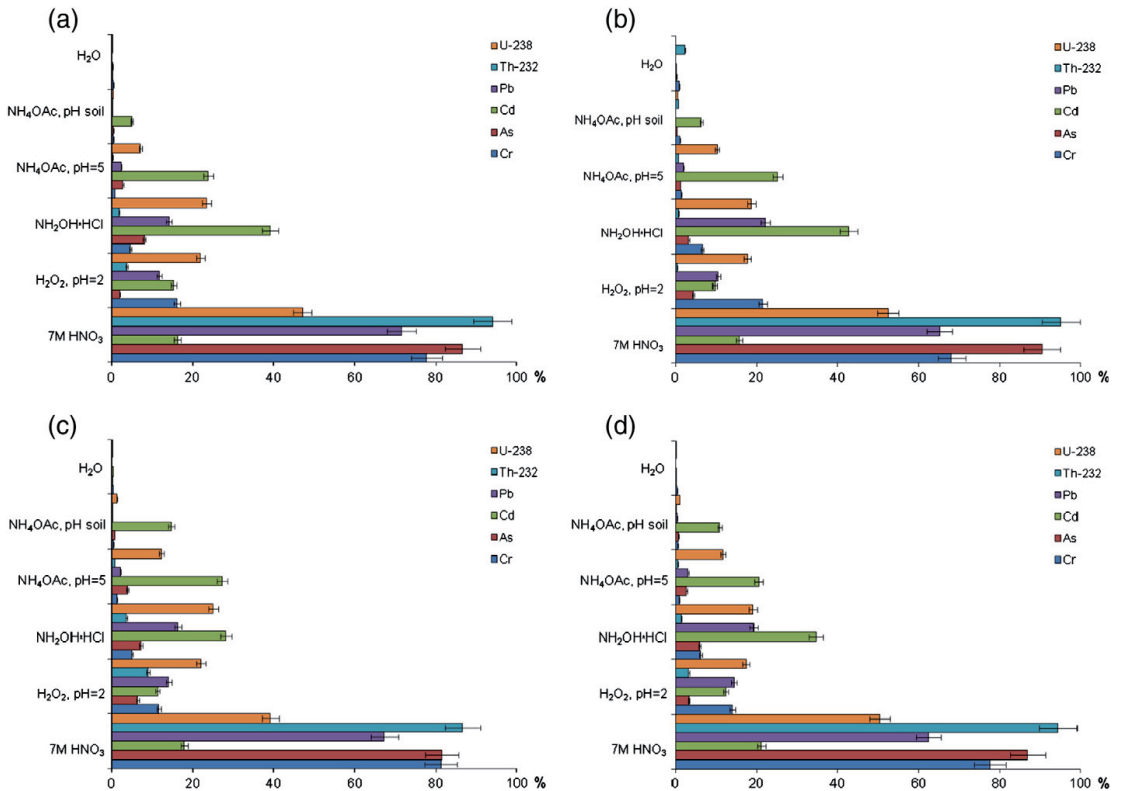


Fig. 1. Radionuclide and metal fractions in soil, obtained by sequential extraction of a) NORM soil, b) Background soil, c) TENORM 1 soil, d) TENORM 2 soil. Percentages accompanied with standard errors are shown; n ≥ 5.

that feed mainly on surface litter material. However, the maximal ²³²Th concentration was obtained for epigeic *L. rubellus*, and the lowest for endogeic *A. rosea*. High concentration of ²³²Th in root and leaf litter (216–416 mg/kg) could provide the explanation for high body concentrations in epigeic *L. rubellus*, although the lower concentration in endogeic *A. rosea* remained unclear. Similarly, the inverse findings with respect to endogeic and epigeic earthworm uptake were reported for heavy metals by Vliet et al. (2005). The more detailed knowledge on species specific characteristics, as well as knowledge on element accumulation and elimination in relation to earthworm physiological dynamic, might provide a better understanding of complex field data (Vliet et al., 2005).

Regarding endogeic species, *A. caliginosa* and *A. rosea*, the highest ²³²Th concentrations were found in earthworms collected at TENORM sites (Fig. 2). It was expected considering the way endogeic

earthworms feed, i.e., regarding large amounts of soil processed. Due to previous mining activities, the disturbances of the soil profiles could also result in more available radionuclide rich particles. In contrast, in the epigeic species, *D. rubidus* and *L. rubellus*, the maximal ²³²Th levels were seen in organisms from the NORM site where the highest total ²³²Th concentration was found in surface soil, but also considerable ²³²Th concentration was found in root and leaf litter (217 mg/kg).

As expected the lowest levels of ²³²Th were found in earthworms from the Background site. The background earthworm concentrations were significantly lower than those in earthworms collected at the NORM and TENORM sites for *A. caliginosa* (p=0.036), *A. rosea* (p=0.018) and *D. rubidus* (p=0.033).

The concentrations of ²³⁸U in earthworms were considerably lower than those of ²³²Th, ranging between 0.6 and 1.0 mg/kg. Although, the highest ²³⁸U concentration in pooled data was observed in endogeic *A. caliginosa* and the lowest in epigeic *L. rubellus*, no significant differences in accumulation between species were found. Comparable body loads of ²³⁸U in earthworms have been obtained previously in study of natural exposure doses of biota in the UK (Beresford et al., 2008a). Based on work of Sheppard et al. (1992), the concentrations of ²³⁸U in earthworms from the present study would not be expected to affect survival and reproduction in biota. Statistical analysis of differences showed that *A. caliginosa* earthworms collected at Background site accumulated significantly lower amounts of ²³⁸U than the earthworms collected at NORM and TENORM sites (p=0.033) (Fig. 2). However, no differences

Table 4

Earthworm body concentrations of radionuclides and metals (mg/kg d.w.) averaged for all sites (mean ± standard deviation, n = 10–16).

Earthworm species	²³² Th ^a	²³⁸ U ^a	As	Cr	Cd	Pb
<i>A. caliginosa</i>	47 ± 38	1.0 ± 0.6	4.9 ± 2.6	4.2 ± 3.5	7.0 ± 3.2	8.9 ± 7.9
<i>A. rosea</i>	11 ± 9	0.8 ± 0.4	2.8 ± 0.5	3.2 ± 1.2	6.3 ± 1.0	3.2 ± 2.5
<i>D. rubidus</i>	38 ± 22	0.6 ± 0.2	2.9 ± 1.8	3.5 ± 0.9	8.8 ± 6.8	5.2 ± 3.6
<i>L. rubellus</i>	57 ± 27	0.6 ± 0.3	3.5 ± 1.9	4.2 ± 2.5	7.2 ± 2.6	3.2 ± 1.8

^a Activity concentrations of radionuclides can be calculated by multiplying concentrations with factors 4.06 and 12.35, for ²³²Th and ²³⁸U, respectively.

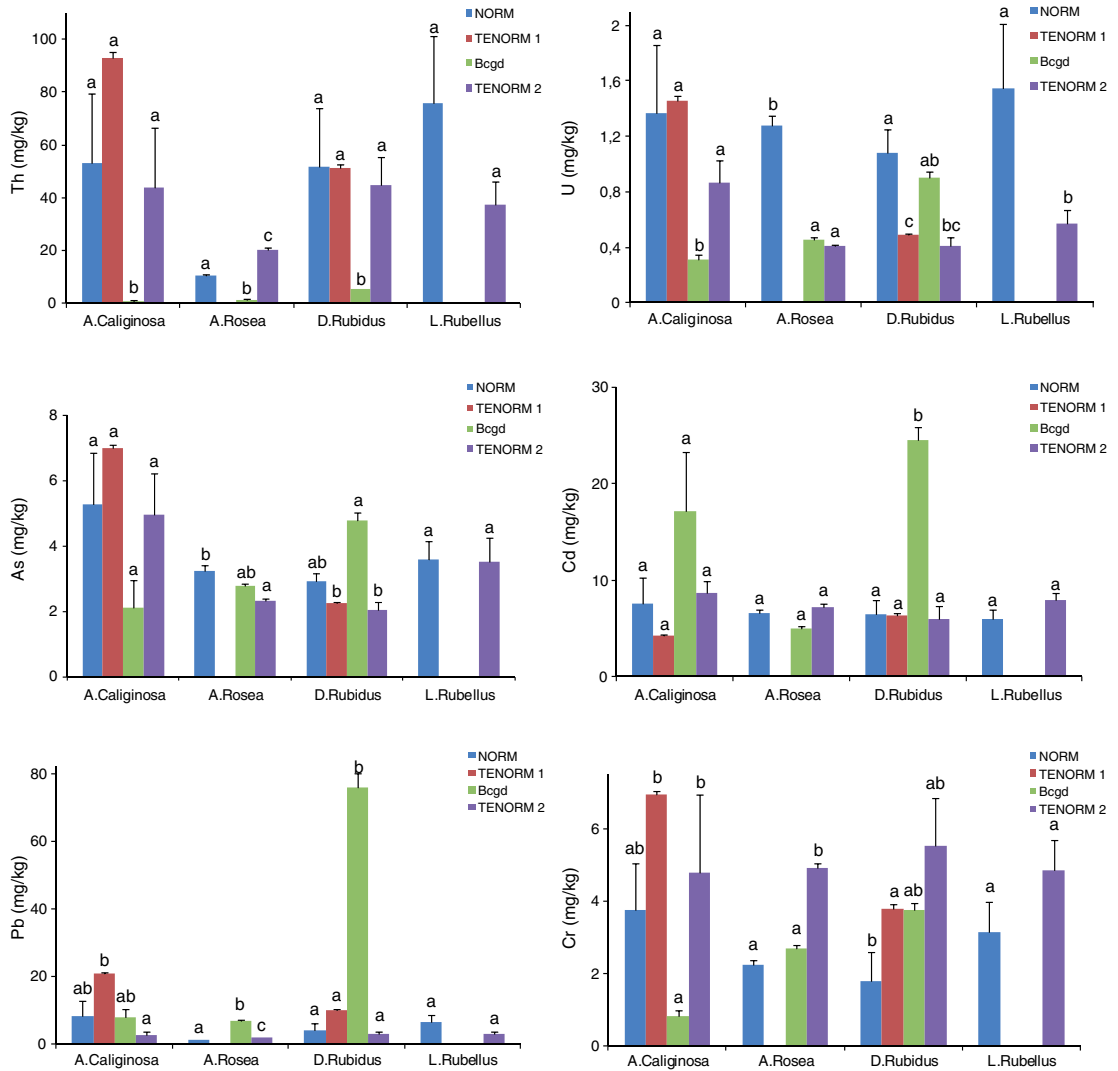


Fig. 2. Mean concentrations ± standard error (d.w.) of radionuclides and metals in earthworm species from different sites (n=14–16). The species *A. rosea* was not found on TENORM 1; and *L. rubellus* not on TENORM 1 and Background sites. Significant differences between earthworms from NORM, TENORM 1, 2 and the Background sites are indicated with different letters (ANOVA, Tukey's post hoc, p < 0.05).

between ²³⁸U concentrations in *A. rosea* and *D. rubidus* from the Background site and those from NORM and TENORM sites were found. Instead, there were significant differences in ²³⁸U uptake between the earthworms collected at NORM and those collected at TENORM sites (p = 0.024, p = 0.008 and p = 0.012, for *A. rosea*, *D. rubidus* and *L. rubellus*, respectively).

The concentration ranges of As and Cr in earthworms were 2.8–4.9 mg/kg and 3.2–4.2 mg/kg, respectively (Table 4). The values for As concentrations in earthworms are similar to those previously demonstrated for earthworms collected in uncontaminated soil (Holmstrup et al., 2011; Langdon et al., 2003). Differences between species in accumulation of As were confirmed (p = 0.019). The highest individual As concentration was obtained for *A. caliginosa*.

Concentrations of As within the same earthworm species collected at the NORM and TENORM sites were significantly different only for species *A. rosea*. Similar to the findings of Langdon et al. (2003), *L. rubellus* accumulated more As than *D. rubidus* within each site.

The maximal concentration of Cr was obtained for earthworms from the TENORM 1 site and the lowest for earthworm from the Background site. No species specific accumulation of Cr for pooled data was observed. Elevated and significantly different Cr concentrations were found in earthworms *A. caliginosa* (p = 0.011), *A. rosea* (p = 0.0005) and *D. rubidus* (p = 0.044) from the TENORM sites in comparison to those from NORM and Background sites.

Considering both species and sites, Cd was found to be relatively evenly distributed (6.3–8.8 mg/kg). Earthworm species showed no

differences in accumulation. The highest mean concentrations were found in earthworms from the Background site in *A. caliginosa* and *D. rubidus*, two ecophysiologicaly different earthworm species (Fig. 2). These higher earthworm concentrations were reflections of large availability of Cd at Background site, i.e., only 15.8% of soil Cd from this site was irreversibly associated with the crystalline soil fraction. The lower Background soil exchange capacity for cations (Table 2) resulted in higher available Cd concentration in soil. In addition, lower concentrations of exchangeable Ca^{2+} ion at Background site led to weaker competition between Ca^{2+} and Cd^{2+} (Table 2). With similar size and same charge, Cd^{2+} was readily taken up by earthworms instead of Ca^{2+} during feeding.

The concentration of Pb in pooled earthworm data ranged from 3.2 to 8.9 mg/kg, depending on earthworm species. The species specific accumulation was demonstrated ($p=0.032$). *A. caliginosa* showed the highest mean Pb value. According to Spurgeon and Hopkin (1996), *A. caliginosa* has a slower Ca-metabolism, which results in poor ability to detoxify Pb and consequently in its accumulation inside the earthworms' body. As for Cd, the analysis of the species individually for each site showed the maximal mean Pb uptake in earthworms from the Background site. Values for earthworm Cd and Pb concentrations in this study were in accordance with those obtained for earthworms collected in uncontaminated natural soil conditions (Ernst et al., 2008), but one order of magnitude lower than those obtained for earthworms in polluted soils (Hobbelen et al., 2006).

3.3. Relationship analysis of radionuclides in earthworms and possible environmental pools for uptake

The total concentrations of radionuclides in soil, concentrations in water soluble and environmentally available soil fractions, as well as in leaf litter and roots were statistically analyzed, simultaneously with earthworm concentrations, in order to identify the most important environmental pool for enhanced uptake of radionuclides. Concentrations in these media were tested for the existence of a positive relationship with total body concentrations in earthworms. The linear regression models obtained are shown in Table 5. The value of the regression formula was judged by $R^2_{adjusted}$, which was the indicator of magnitude of variation in the data that was explained by the model.

When all earthworm species were considered as pooled data, the variation in earthworm body concentrations of ^{232}Th was best explained by the variation in the total soil and available soil concentrations. In the case of ^{238}U , body load in earthworms was best

correlated with total soil concentration and concentration in roots and leaf litter.

The concentration of ^{232}Th in *D. rubidus*, an epigeic earthworm, was best described by the variation in ^{232}Th total concentration of soil and by variation in ^{232}Th concentration in roots and leaf litter. The observed relationship was in accordance with expectations considering habitat and feeding manner. Concentration of ^{238}U in the same earthworm was dependent only on the ^{238}U concentrations in roots and leaf litter. However, the significant correlation neither for ^{232}Th nor for ^{238}U concentration in earthworm body and roots and leaf litter was obtained for the ecophysiologicaly similar species, *L. rubellus*.

Regression models obtained for ^{232}Th in the endogeic species, *A. caliginosa* and *A. rosea*, demonstrated soil as primary source of radionuclides and metals for earthworm uptake. It suggested that these earthworms, living in deeper soil layers, accumulated ^{232}Th within their bodies mainly via radioactive particles in soil. According to the obtained models, ^{238}U concentrations in endogeic earthworms were also dependent on the total concentrations of ^{238}U in soil, as well as on the available soil (*A. caliginosa*), roots and leaf litter ^{238}U concentrations (*A. rosea*). The suggested uptake mechanism via roots and leaf litter for these earthworms was in accordance with the higher ^{238}U concentration in environmentally available soil fractions. The absence of a positive correlation between earthworm concentrations and water soluble soil concentrations remains unclear.

The obtained regression models suggested total soil as the primary environmental pool for the earthworms' uptake of radionuclides.

3.4. Transfer from soil to earthworms

Transfer factors (TF) are commonly used to evaluate radionuclide and metal bioavailability and ecological transfer from soil to biota (Neuhauser et al., 1995; Suthar and Singh, 2009; Vandenhove et al., 2009b). Soil sequential extraction in current study showed, however, that the mobility and environmental availability of the most of the elements investigated was low. To obtain more realistic information about the ability of an organism to accumulate radionuclides and metals, the available transfer factors (ATF) have proved to be the most useful, taking the mobile fractions in soil into account (Baeza and Guillen, 2006). To determine the transfer from soil to earthworms, TF and ATF were calculated for the investigated elements (Table 6). However, the uncertainty of TF concept for earthworms living in this area has to be emphasized. The observed inhomogeneous distribution

Table 5

Linear regression models (concentrations of radionuclides in earthworms taken as dependent variables; total soil concentrations, environmentally available soil concentrations and root and leaf litter concentrations taken as predictor variables). Only models containing significant predictors are shown ($p<0.05$).

Model	F	p	R ²	R ² _{adjusted}
<i>D. rubidus</i>				
$\log (Th_{worm}) = -0.48 + 0.58 \log (Th_{tot,soil}) + 0.16 \log (Th_{root-leaf})$	17.21	0.005	0.758	0.714
$\log (U_{worm}) = -0.68 + 0.59 \log (U_{root-leaf})$	9.52	0.010	0.464	0.415
<i>L. rubellus</i>				
$\log (Th_{worm}) = -2.20 + 1.32 \log (Th_{tot,soil})$	11.47	0.005	0.469	0.428
<i>A. caliginosa</i>				
$\log (Th_{worm}) = -4.40 + 2.11 \log (Th_{tot,soil})$	61.74	0.005	0.849	0.835
$\log (U_{worm}) = -0.44 + 0.60 \log (U_{tot,soil})$	6.61	0.026	0.375	0.319
$\log (U_{worm}) = 0.13 + 0.57 \log (U_{av,soil})$	6.33	0.029	0.365	0.308
<i>A. rosea</i>				
$\log (Th_{worm}) = -0.84 + 0.69 \log (Th_{tot,soil})$	24.85	0.001	0.756	0.726
$\log (U_{worm}) = -0.87 + 0.76 \log (U_{root-leaf})$	28.71	0.001	0.782	0.755
<i>All species combined</i>				
$\log (Th_{worm}) = -2.13 + 1.30 \log (Th_{tot,soil}) - 0.09 \log (Th_{av,soil})$	31.99	0.005	0.598	0.579
$\log (U_{worm}) = -0.58 + 0.21 \log (U_{tot,soil}) + 0.33 \log (U_{root-leaf})$	6.03	0.005	0.219	0.183

Table 6
Transfer factors (TF) and available transfer factors (ATF) for radionuclides and metals in earthworms (mean \pm standard deviation, n = 10–16).

Worms	^{232}Th		^{238}U		As		Cd		Pb		Cr	
	TF	ATF	TF	ATF	TF	ATF	TF	ATF	TF	ATF	TF	ATF
<i>A. caliginosa</i>	0.05 \pm 0.05	11 \pm 10	0.25 \pm 0.16	2.0 \pm 0.8	0.47 \pm 0.30	22 \pm 16	9 \pm 5	32 \pm 21	0.27 \pm 0.31	14 \pm 16	0.12 \pm 0.10	8 \pm 5
<i>A. rosea</i>	0.03 \pm 0.02	4 \pm 5	0.23 \pm 0.12	3 \pm 1	0.39 \pm 0.33	22 \pm 19	12 \pm 6	37 \pm 18	0.18 \pm 0.27	10 \pm 12	0.19 \pm 0.16	8 \pm 5
<i>D. rubidus</i>	0.08 \pm 0.04	9 \pm 7	0.25 \pm 0.23	2 \pm 2	0.19 \pm 0.07	19 \pm 26	8 \pm 5	26 \pm 14	0.08 \pm 0.08	8 \pm 8	0.25 \pm 0.17	8 \pm 3
<i>L. rubellus</i>	0.06 \pm 0.04	24 \pm 17	0.09 \pm 0.04	0.9 \pm 0.3	0.24 \pm 0.11	11 \pm 6	9 \pm 3	28 \pm 14	0.02 \pm 0.03	3 \pm 5	0.12 \pm 0.07	10 \pm 5
All worms	0.06 \pm 0.04	12 \pm 13	0.20 \pm 0.16	1.9 \pm 1.5	0.33 \pm 0.25	18 \pm 18	10 \pm 5	31 \pm 17	0.14 \pm 0.22	9 \pm 11	0.17 \pm 0.14	9 \pm 5

of investigated elements in soil leads to a question of applicability of TF concept for organisms that might not have spent enough time in the soil that was actually taken into the calculation of TF as representative. High standard deviations provided with mean values of TF and ATF are the reflections of large differences between soil concentrations rather than differences in earthworm body concentrations.

The TF for ^{232}Th were comparable among different species and averaged TF were 0.06. No data on TFs for ^{232}Th from soil to earthworms could be found in the literature. It has been previously stated that the key source of uncertainty in radiological risk prediction for non-human biota is transfer parameters used (Oughton et al., 2008). According to Wood et al. (2009) the development of transfer variable databases should be a focus of future research efforts. In the description of the methodology used to derive the default concentration ratio database within the ERICA tool, Beresford et al. (2008b) provided explanation that available concentration ratio values for biogeochemically similar element (i.e., ^{238}U) are in use for ^{232}Th in case of terrestrial invertebrates due to the absence of actual ^{232}Th transfer data.

The average TF for ^{238}U in all earthworms were 0.20 (range 0.09–0.25, depending on the species). Similar levels of natural uranium uptake were found recently (Beresford et al., 2008a; Giovanetti et al., 2010; Yoshida et al., 2005). The TF for ^{238}U were approximately three times higher than for ^{232}Th . However, when environmentally available concentrations in soil were considered to reflect the capacity of organisms to bioaccumulate the investigated elements in the more realistic way (Salbu, 2000), obtained ATF were reversed. The magnitude of both ATF for ^{232}Th and ATF for ^{238}U increased significantly, indicating that ^{232}Th was actually bioaccumulated to a higher degree than ^{238}U , although ^{238}U was much more bioavailable in the soil.

For metals in earthworms, the order of TF and ATF decreased according to Cd > As > Pb > Cr. Similar TF order and magnitude have been found for earthworms collected at sampling sites (polluted and reference) in Øvre Ardal in Norway (Holmstrup et al., 2011). The present study showed, however, that TF values were one order of magnitude lower than those obtained by Ernst et al. (2008), suggesting low bioaccumulation. When TF for individual earthworm

species were compared separately, they decreased according to: *A. caliginosa* > *A. rosea* > *L. rubellus* > *D. rubidus*, for As and Pb; *A. rosea* > *A. caliginosa* > *L. rubellus* > *D. rubidus* for Cd and *D. rubidus* > *A. rosea* > *A. caliginosa* > *L. rubellus* for Cr. As expected, the TF for the non-essential elements Cd, Pb and As from soil were higher in endogeic species. TF for Cr were more equally distributed with no difference between endogeic and epigeic species.

3.5. Radiation exposure dose rates and risk estimation

The exposure dose rates ($\mu\text{Gy/h}$) from ^{232}Th and ^{238}U and radiological risk quotient (RQ) for earthworms were calculated using the ERICA tool, tier 2. Whole-body weighted absorbed dose rates were summarized for internal and external dose rates. Obtained values were in the range 0.05–2.6 $\mu\text{Gy/h}$ (Fig. 3), comparable to absorbed dose rates for earthworms in the study of background exposure rates of terrestrial wildlife in England and Wales (Beresford et al., 2008a).

No statistically significant differences were found in absorbed dose rates of different earthworm species. In all species, the major contribution to the total dose was from internal exposure to ^{232}Th (81–96%). Furthermore, when dose rate calculation was expanded to include the radium isotopes (^{226}Ra and ^{228}Ra), assuming the secular equilibrium between mother and daughter in soils, higher dose rates (2.2–3.9 $\mu\text{Gy/h}$) were obtained. Still, all dose rates were lower than 10 $\mu\text{Gy/h}$, a value below which no evidence for ecologically relevant biological effects has been found (Andersson et al., 2009).

The expected radiation risk for earthworms in this area was low (RQ = 0.28). However, the additional dose contribution from high soil levels of thoron (^{220}Rn) and radon (^{222}Rn) (Mrdakovic Popic et al., unpublished data), as well as the contribution from other daughter radionuclides (e.g., ^{210}Po , ^{210}Pb), not considered in the present study, will actually enhance the doses received by earthworms. In that sense, the present evaluation could somewhat underestimate the presented exposure dose rates and risk quotient.

4. Conclusion

Transfer from soil to free-living earthworm species in Fen Complex area was investigated in the present study. Soil analysis showed significantly elevated levels of ^{232}Th and slightly elevated levels of ^{238}U and metals As, Pb, Cd and Cr at the NORM and TENORM sites in comparison to natural uncontaminated Norwegian soil. The inhomogeneous distribution of radionuclides and metals was observed at all sites. Based on sequential extraction results, the mobility and bioavailability of ^{232}Th , As, Pb and Cr were low, while ^{238}U and Cd concentrations in available soil fractions were elevated. The species specific earthworm uptake was demonstrated for ^{232}Th , As and Pb. The observed variability between individual organisms was high for all investigated elements. Transfer factors (TF) of ^{238}U were found to be higher than those of ^{232}Th and in accordance with the chemical nature of both elements and their bioavailability in soil of FC area. However, based on available transfer factors (ATF), the higher uptake of mobile and bioavailable ^{232}Th than ^{238}U species was actually seen in different earthworm species. No clearly distinguished

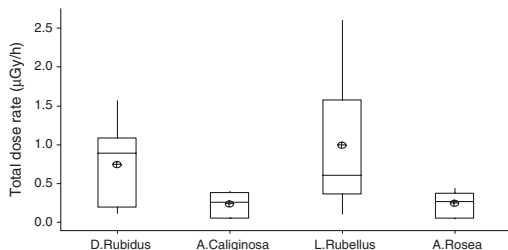


Fig. 3. Total radiation dose rate per earthworm species (n = 10–16) calculated by the ERICA tool. Box plot: the horizontal lines show the median; crossed circles average values; the top and the bottom of the box give the 75th and 25th percentiles. The top and the bottom of the whiskers show the maximum and minimum values.

differences in uptake of ^{232}Th and ^{238}U were found regarding epigeic and endogeic species. However, in the case of Cd, Pb and As, as expected, the endogeic species showed higher uptake. According to the regression models, primary sources for earthworm uptake of ^{232}Th were total soil and environmentally available soil fractions. For ^{238}U in earthworms, positive correlations were also found for total and available soil fraction and for roots and leaf litter. The calculated exposure dose rates from ^{232}Th and ^{238}U ranged from 0.05 to 2.60 $\mu\text{Gy/h}$, suggesting low radiological risk for earthworms.

In general, present study confirmed, as reported previously for metals, that earthworms can survive within contaminated media. Nevertheless, more attention should be given to additional investigating of daughter radionuclides (e.g., ^{210}Pb , ^{210}Po , ^{220}Rn , ^{222}Rn) that could affect the earthworms. As the authors are aware, this study is one of very few that have shown transfer from soil and bioaccumulation of both radionuclides and metals in different earthworm groups in natural forest conditions.

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Paper IV

Transfer of radionuclides (^{232}Th , ^{238}U) to wild forest flora species in an area with enhanced legacy and natural radioactivity in Norway

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Abstract:

A study of transfer of the naturally occurring radionuclides thorium (^{232}Th) and uranium (^{238}U) from soil to wild plant species was performed as a part of an environmental impact assessment in an area with legacy and naturally enhanced radioactivity in Norway. To investigate possible differences in uptake, three sites were chosen for the study: a former iron (Fe) mining site as a legacy NORM site, undisturbed ^{232}Th -rich as a NORM site, and a site outside the enhanced radiation area that served as a Reference site. Tissue concentrations, transfer factors and radiological exposure dose rates were determined in nine wild plant species, including vascular and non-vascular plants, deciduous and coniferous trees. High soil activity concentrations of ^{232}Th , up to 5069 Bq/kg and 16516 Bq/kg, were measured in samples from the legacy NORM and undisturbed ^{232}Th -rich sites, respectively, while soil from the Reference site exhibited a statistically lower activity concentration of ^{232}Th (up to 2270 Bq/kg). Soil activity concentrations of ^{238}U were lower than ^{232}Th and in terms of the investigated sites, they were statistically indistinguishable, with maximal concentrations 156, 859 and 109 Bq/kg at the legacy NORM, undisturbed ^{232}Th -rich and Reference sites, respectively. Despite the increased total soil radionuclide levels, the mobile soil fractions were low (up to 157 Bq/kg of ^{232}Th and 27 Bq/kg of ^{238}U). Aboveground plant tissue activity concentrations of ^{232}Th ranged from non-detectable (tree needles and leaves) to 49 Bq/kg (grass); ^{238}U ranged from 0.03 (tree needles and leaves) to 5 Bq/kg (moss). No differences in the uptake of the radionuclide ^{232}Th by plants at the three investigated sites were demonstrated; uptake of the radionuclide ^{238}U was significantly lower at the Reference site. As expected, plants roots served as a natural translocation barrier, as the concentration of radionuclides in the analyzed samples were up to 25-fold higher than in the corresponding aboveground plants. The transfer factors for ^{232}Th and ^{238}U in aboveground plant parts ranged from $4 \cdot 10^{-5}$ to $1 \cdot 10^{-2}$ and $1 \cdot 10^{-4}$ to $4 \cdot 10^{-2}$, respectively. Radiological exposure dose rates, calculated using the ERICA software programme, were 5.8 $\mu\text{Gy/h}$ for grasses and herbs, 22.6 $\mu\text{Gy/h}$ for lichen and bryophytes, 1.3 $\mu\text{Gy/h}$ for shrubs and 1.9 $\mu\text{Gy/h}$ for trees. Although the dose rate calculated for lichen and bryophytes was above the adopted screening level of 10 $\mu\text{Gy/h}$, this value is still below the dose rates expected to cause impacts to plants. Based on plant tissue activity concentrations and calculated transfer factors, no significant uptake of ^{232}Th and ^{238}U was demonstrated in plants, as initially expected due to the high soil radionuclide levels.

Keywords: thorium, uranium, plants, transfer factors, radiation dose

1. Introduction

The recommendations of the International Commission on Radiological Protection (ICRP, 1991) and the research community have been exclusively focused on the protection of humans from ionizing radiation. Up to the last decade, it was assumed that if humans are protected, the quality of the living environment is not impaired. However, current recommendations (ICRP, 2003, 2007, 2008) indicate that environment health and status, including animals and plants should be monitored and preserved from the effects of ionizing radiation. Numerous monitoring studies on different vegetation species (mainly domestic, cultivated plants) have been conducted worldwide to investigate the transfer of radionuclides (e.g., ^{137}Cs , ^{238}U , ^{226}Ra , ^{210}Po) through the food web and to humans (Al-Masri *et al.*,

2008; Bunzl and Trautmannsheimer, 1999; Carvalho *et al.*, 2009; Schimmack *et al.*, 2004; Skutterud *et al.*, 2012). On the contrary, the impact assessments for species other than humans and data on the transfer of naturally occurring radionuclides in natural, non-disturbed conditions are less frequent, although interest in this has been increasing in the last years (Dragovic *et al.*, 2010; Chen *et al.*, 2005; Shtangeeva, 2010; Vandenhove *et al.*, 2009a,b) and the need for this kind of data has been emphasized (Beresford *et al.*, 2008). With respect to the radiological protection of non-human species, a set of reference organisms (animals and plants) is currently used in radioecological studies in order to establish an integrated approach (Beresford *et al.*, 2008; ICRP, 2008).

Radionuclides ^{232}Th and ^{238}U are present in the Earth's crust from its origin. These naturally occurring radionuclides are part of the biosphere and exist in soil and sediments, water and biota. Although many reports can be found in the literature of today on the different aspects of geochemical behavior of ^{232}Th and ^{238}U (Galindo *et al.*, 2007; Sheppard and Evenden, 1988; Thiry *et al.*, 2005; Vera Tomé *et al.*, 2002, 2003), most of them are the results of work performed under laboratory conditions or from studies at former mining sites with highly contaminated soil, water or air. With this in mind, the investigation of undisturbed naturally occurring radionuclides and their transfer to biota is of significance (Shtangeeva, 2010), especially since data for many radionuclides and biota species are still missing (Beresford *et al.*, 2008).

The Fen Complex in Norway is an area rich in naturally occurring radionuclide thorium (^{232}Th) and, to lesser degree, uranium (^{238}U). In the past, several locations in this area were mined for iron (Fe), niobium (Nb) and some rare earth elements (REE). Nowadays, both disturbed and undisturbed environmental conditions can be seen in the area. Thus, these sites could be regarded as very appropriate natural laboratories to investigate the mobilization of radionuclides and associated trace elements. Previous studies in the Fen Complex area focused on human risk estimations and demonstrated the elevated annual exposure dose due to gamma radiation, radon (^{222}Rn) and thoron (^{220}Rn) exposure (Mrdakovic Popic *et al.*, 2012; NGI-UMB, 2010; Stranden and Strand, 1896; Sundal and Strand, 2004). Recent investigation into radionuclide and trace element levels in the Fen Complex soils demonstrated a significantly inhomogeneous distribution and elevated soil concentrations of ^{232}Th in comparison to soil from other areas in Norway. In addition, concentrations of ^{238}U and trace elements such as arsenic (As), chromium (Cr), cadmium (Cd) and lead (Pb) were moderately increased at some of the Fen Complex locations (Mrdakovic Popic *et al.*, 2011).

The current work is part of an environmental impact assessment project in the Fen Complex in Norway. The transfer of radionuclides from soil to plant is a key process in mediating the effects on plants as well on other biota (Kabata-Pendias, 2010). In this regard, the aim of present study was to investigate the uptake, soil-to-root and soil-to-plant transfer of radionuclides ^{232}Th and ^{238}U in the wild flora of an enhanced ionizing radiation area. The dose rates for selected wild plant species were calculated and major dose contributors discussed.

2. Materials and methods

2.1. Study area

The assessment of soil-to-plant radionuclide transfer presented in the present paper was based on input data from several field expeditions (2008 and 2009) in the Fen Complex, Norway. This area has unique bedrock geology and is well-known as an enhanced natural radiation area. Based on recent investigations in the Fen Complex (Mrdakovic Popic *et al.*, 2011; NGI-UMB, 2010), three different sites (Fen, Bolladalen and Torsnes) were chosen for the current study. The former iron (Fe) mining

site, Fen, and an undisturbed ^{232}Th -rich site, Bolladalen, were considered representative for legacy NORM and non-altered NORM sites, respectively. The site Torsnes was chosen as the Reference site outside the main enhanced radiation area. Detailed descriptions of the area and study sites are given in Mrdakovic Popic *et al.* (2011, 2012).

2.2. Sampling

The plant species which were abundant at the sampling sites were collected for analysis. In total, nine different plants were sampled. The chosen plants covered a relatively wide range of wild forest flora, with vascular and non-vascular plants, deciduous and coniferous trees. Each study site was divided into 5 sampling points whose geographical positions were recorded in by Global Positioning System (GPS Germin, USA). Not all plant species were found at each study site. Leaves and needles were collected from trees, while the other plants were collected as whole plants after removing adherent soil from the roots. Samples were transported in paper bags. The plant taxonomy was determined in the laboratory. Visible soil particles were removed; samples were washed and divided into root and aboveground plant parts. All samples were then dried at 40°C , milled and homogenized prior to analysis. Samples of underlying soil in the A horizons (about 500 g f.w.) were taken at the same points as plants, allowing the calculation of transfer factors (TF) and uptake estimation. In the laboratory, vegetation and other debris were removed from the soil samples, which were then dried and homogenized by passing through a 2-mm mesh sieve.

2.3. Sample preparation and analyses

Plant samples were decomposed by microwave-assisted acid digestion. Plants were weighed (0.5 g) into Teflon vessels and 3.5 ml of ultrapure concentrated nitric acid, 1.5 ml of distilled water and the internal standard were added to each vessel. Microwave digestion (MLS-Milestone UltraClave, MLS GmbH), combining high pressure and high temperature with concentrated acid, efficiently decomposed the samples. After digestion, the solutions were transferred to 50-ml plastic tubes and diluted (7% HNO_3) with distilled water.

Soil samples were divided into subsamples for soil characterization, measurement of total radionuclide concentration and sequential extraction. Soil pH was measured in distilled water and 1 M KCl solution, at the solid-liquid ratio 1:2.5 (Reeuwijk, 2002). Particle size analysis was performed by the standard pipette method and organic matter determination by loss on ignition (LOI). The total cation exchange capacity (CEC) of the sorptive complex was calculated as the sum of the hydrolytic acidity and total exchangeable basis, both measured according to a modified version of Reeuwijk (2002). The decomposition of soil (0.3-0.5 g) was performed by ultrapure concentrated HNO_3 (5 ml) digestion in a microwave oven, in the same way as for plants, giving 10 % acid solutions for radionuclide measurements. In addition, the sequential extraction procedure (Oughton *et al.*, 1992; Tessier *et al.*, 1979) was carried out on a certain number of soil samples to obtain information on potentially mobile soil fractions of radionuclides. Solutions obtained after extraction with distilled water, NH_4Ac (buffered at soil pH) and NH_4Ac (buffered at pH 5) were analyzed and summarized results considered as potentially mobile soil ^{232}Th and ^{238}U fractions.

Concentrations of ^{232}Th and ^{238}U were measured in all plant and soil solutions by ICP-MS (Perkin Elmer Sciex ELAN 6000). Prior to ICP-MS, soil solutions were diluted to 5 % acid. All samples were analyzed according to current laboratory quality-assurance procedures involving standard reference materials (NSC DC 73325 and NCS DC 73348), blanks, internal standard and in-house calibration standards. Detection and quantification limits were calculated as three and ten times the standard

deviation of blank sample measurements, respectively. Plant Sc and Ti concentrations, used as indicators of soil contamination, were generally below 1 % in the plants relative to the levels in the corresponding soils. In addition to ICP-MS, daughter radionuclides ^{226}Ra and ^{228}Ra were measured by gamma- and ^{210}Po by alpha spectrometry. Measurement characteristics are provided in Mrdakovic Popic *et al.*, (2012) and Skipperud *et al.*, (2013). The obtained data were used for the ERICA Tool calculation of dose rates per reference organism group.

2.4. Data analysis

Radionuclide soil-to-root and soil-to-plant ratios were quantified by commonly used transfer factors (TF) (IAEA, 1994, 2010). The TF is defined as the ratio of the concentration in biota and concentration in soil:

$$\text{TF} = \frac{\text{C biota (d.w)}}{\text{C soil (d.w)}}$$

It describes the amount of element expected to enter a plant from its substrate under equilibrium conditions (Sheppard and Sheppard, 1985; Vandenhove *et al.*, 2009b; Vera Tome *et al.*, 2003).

The assessment of the radiation dose rates was done using the ERICA software programme developed within an EC EURATOM-funded ERICA project to assess radiological risk to biota (Brown *et al.*, 2008). Tool is based on a tiered approach that can be adapted to either generic or site-specific assessments and run for a set of reference organisms. For our work, the assessment was run at Tier 2 in two ways. Initial dose rates calculation (using the Fen soil radionuclide activity concentrations and the default ERICA CRs values) provided insight into which referent plants are likely to have the highest doses and which radionuclides would contribute the most. The second dose rates calculation was done using site-specific data, including both soil and plant radionuclide activity concentrations and calculated CR values. The resulting dose rates, radionuclide contribution and exposure routes were discussed with respect to those obtained when the model was run with generic values.

Statistical data analyses were performed using Minitab 16 (Minitab Inc.). Specific activities of $4.06 \text{ Bq } ^{232}\text{Th}\cdot\text{mg}^{-1}\text{Th}$ and $12.35 \text{ Bq } ^{238}\text{U}\cdot\text{mg}^{-1}\text{U}$ were used to obtain radionuclide activity concentrations from total soil Th and U determinations. Normality was tested using the Anderson-Darling test. Soil and plant concentrations were log-normally distributed and are presented with median and ranges. TFs are presented as geometrical mean accompanied by geometrical standard deviation. All data were log10 transformed prior to statistical tests. One-way ANOVA followed by Tukey Simultaneous Tests were applied to test for any differences in concentration and in TFs between analyzed plant species and among investigated sites. The difference between the set of TFs obtained on the basis of total soil radionuclide concentrations and that obtained on the basis of mobile soil radionuclide concentrations was tested with a two-sample t-test. Results with $p \leq 0.05$ were considered statistically significant.

3. Results and discussion

3.1. Soil analyses

Basic soil characteristics are presented in Table 1. Soil pH was neutral at the former mining site Fen and slightly acid at the sites Bolladalen and Torsnes, while organic matter content was high (above 15 %) at each of the investigated sites. Soil texture analysis showed silt and sand as main fractions at both Fen and Bolladalen, while in soil samples from the Reference site Torsnes silt was dominant. Total and mobile soil activity concentrations of ^{232}Th and ^{238}U are given in Table 2. Thorium and ^{238}U are

primordial, long-lived radionuclides, ubiquitous in the Earth's crust. Their levels are found to be in close connection with the geological composition and current conditions of the underlying formation (El-Dine, 2008), as demonstrated in previous studies in the Fen Complex area (Mrdakovic Popic *et al.*, 2011; NGI-UMB, 2010; Sundal and Strand, 2004). Similar to our previous results (Mrdakovic Popic *et al.*, 2011; Mrdakovic Popic *et al.*, 2012), large variations in ^{232}Th activity concentrations within the sites were observed (1589-5069, 2146-16516 and 55-2270 Bq kg⁻¹ for soil samples collected at sites Fen, Bolladalen and Torsnes, respectively), suggesting an inhomogeneous radionuclide distribution, even at close sampling points. Median soil ^{232}Th activity concentrations (2727, 6835 and 664 Bq/kg for Fen, Bolladalen and Torsnes) were about 2 orders of magnitude higher than the world and Norwegian soil concentrations (45 Bq kg⁻¹) given by UNSCEAR (2000, 2008). In addition, the activity concentrations of ^{232}Th at the Fen and Bolladalen sites exceeded the screening value (1 Bq/g) for radioactive waste in Norway (Pollution Control Act, 2010). Significantly lower activity concentrations of ^{232}Th were observed for soil at the Reference site Torsnes ($p = 0.003$). Accordingly with our previous results (Mrdakovic Popic *et al.*, 2011), no statistically significant difference in activity concentration was observed between soil samples from the former mining site Fen (legacy NORM) and the undisturbed ^{232}Th -rich site Bolladalen (undisturbed NORM).

The soil ^{238}U activity concentrations (61 – 156, 96 – 859 and 8 – 109 Bq/kg at sites Fen, Bolladalen and Torsnes, respectively) were considerably lower than ^{232}Th values, but at both Fen and Bolladalen median values were still higher than the reference worldwide value (30 Bq kg⁻¹) and Norwegian average value (32 Bq kg⁻¹) (UNSCEAR, 2000, 2008). The screening value for radioactive waste materials with respect to ^{238}U (1.0 Bq/g) (Pollution Control Act, 2010) was not exceeded in the analyzed samples. These concentration levels corresponded to lower ^{238}U abundance in the bedrock of the study area, already seen in previous studies (Mrdakovic Popic *et al.*, 2011; NGI-UMB, 2010). No statistically significant differences in soil ^{238}U concentration were found between sites.

Mobile soil fractions were considerably lower than the total soil concentrations ($p = 0.004$) for both ^{232}Th and ^{238}U . No significant differences in mobile ^{232}Th or mobile ^{238}U activity concentrations between sites were observed. The low radionuclide percentages in the environmentally mobile soil fractions (0.03 – 0.69 and 2.35 – 4.92 % for ^{232}Th and ^{238}U , respectively) suggested limited transport and low bioavailability. Correlation analysis of radionuclide concentrations and soil parameters revealed some weak linear (positive and negative) relationships for mobile soil ^{232}Th concentration and total soil ^{232}Th concentration ($r = 0.57$, $p = 0.035$), pH of soil ($r = -0.61$; $p = 0.022$), and OM content ($r = -0.59$; $p = 0.027$). Similarly, negative linear relationships were found for mobile ^{238}U soil concentrations and pH ($r = -0.75$; $p = 0.002$), as well as for mobile ^{238}U soil concentrations and the OM content ($r = -0.68$; $p = 0.007$). Considering these relationships, soil pH and organic matter play an important role in radionuclide mobility and transport in Fen soils. According to Syed (1998), large Th^{4+} is hydrolyzed and significantly adsorbed on organics, clay and oxides in the soil in natural conditions, i.e., rather immobile in the environment. The significant positive correlation in the study of Vandenhove *et al.* (2009a) was seen between the soil distribution coefficient (K_d) and organic matter content, suggesting increased sorption of positively valent U-forms on the exchange sites of organic matter. Guo *et al.* (2008) showed that pH and organic matter are main contributors to the dynamic processes of ^{232}Th adsorption and mobilization in soil.

The pooled data on activity concentrations of ^{228}Ra , ^{226}Ra and ^{210}Po for the Fen Complex soil (obtained by gamma and alpha spectrometry) were in following ranges: 134 – 7300, 77 – 251 and 38 – 76 Bq/kg, respectively. As for the parent radionuclides ^{232}Th and ^{238}U , maximal activity concentrations of ^{228}Ra , ^{226}Ra and ^{210}Po were measured in soil from the site Bolladalen, while minimal in soil from the site Torsnes.

3.2. Plant concentrations

The activity concentration of ^{232}Th in the analyzed plant species was in the range from non detectable (< LOD) to 49 Bq kg^{-1} (Fig. 1). Maximal levels were observed in dandelion (32 Bq kg^{-1}), grass (49 Bq kg^{-1}) and moss (22 Bq kg^{-1}). Although a different mode of uptake is known for moss and lichen without wax cuticles and root systems in comparison to vascular plants with developed root systems (Bell *et al.*, 1988; Ugur *et al.*, 2003), the levels of ^{232}Th did not differ statistically in dandelion, grass and moss samples. However, the concentrations in birch leaves, spruce and pine needles did differ statistically from other analyzed plant species ($p = 0.001$) since they were below the limit of detection (LOD) and quantification (LOQ). Although fern, grass and moss collected at Torsnes had the lowest demonstrated activity concentration of ^{232}Th , no statistically significant difference was observed in these plants with respect to the different sites. Somewhat wider ^{232}Th ranges were seen at the former mining site Fen for all analyzed plants. Generally, activity concentrations of ^{232}Th in plants obtained in the current study were higher, but still comparable with results from similar studies, either in regions with no elevated primordial radionuclide levels or in areas with legacy NORM (Belivermiş and Çotuk, 2010; Dowdall *et al.*, 2005; Dragovic *et al.*, 2010; Martinez-Aguirre *et al.*, 1997).

The activity concentrations of ^{238}U were several times lower than those of ^{232}Th , and were in the range from 0.031 to 5 Bq kg^{-1} (Fig. 2). At all sites, moss exhibited a statistically higher ^{238}U concentration than other plant species ($p = 0.002$), while trees (leaves and needles) had the lowest concentrations ($p = 0.004$). By comparing concentrations in the same plant species from the three different sites, equally high and statistically not different plant activity concentrations of ^{238}U were found at the Fen and Bolladalen sites, while significantly lower values were obtained for all analyzed plant species from the reference site Torsnes. Good agreement of ^{238}U concentrations in the plants from the current study with previously published ranges for comparable vegetation in various worldwide regions was obtained (Dowdall *et al.*, 2005; Dragovic *et al.*, 2010; Belivermiş and Çotuk, 2010).

Activity concentrations of ^{228}Ra , ^{226}Ra and ^{210}Po were in plants in following ranges $35 - 290 \text{ Bq/kg}$, $12 - 67 \text{ Bq/kg}$ and $16 - 620 \text{ Bq/kg}$, respectively. The highest activity concentrations of ^{226}Ra and ^{228}Ra were measured in grass, while the highest concentrations of ^{210}Po were measured in samples of lichen. It has been previously shown that lichens contain significantly higher ^{210}Po and ^{210}Pb concentrations than vascular plants or funghi (Jaworovski, 1969; Kauranen and Miettinen, 1967, 1969; Skwarzec and Jakusik, 2003). Polonium-210 activity concentrations in lichen from the Fen Complex were higher than concentrations ($70 - 212 \text{ Bq/kg}$) in study of Skuterud *et al.* (2005) where lichens from two areas in central and southern Norway were analyzed. Obtained higher plants activity concentrations of ^{232}Th and ^{238}U progenies indicated enhanced mobility, bioavailability and uptake of these progenies and need for their further analyses. Radionuclide bioavailability and accumulation in plants is associated with numerous factors – speciation, soil and climatic conditions, plant genotype, active or passive transport processes, redox state of substrate, plant part and age, the type of plant root system and competition with essential elements (Dragovic *et al.*, 2010, Kabata-Pendias 2010; Vandenhove *et al.*, 2007). As discussed, soil pH and organic matter in particular might have had a crucial role in the mobility and bioavailability of the here investigated radionuclides.

The incorporation of radionuclides in plants, depending on plant species and bioavailable radionuclides, takes place through activity interception by the external plant surface or through uptake via the root system. In this paper, two different modes of uptake were studied by moss/lichen and vascular plants, respectively. Translocation of the radionuclides ^{232}Th and ^{238}U from the soil to vascular plants is evaluated by root-to-aboveground parts (Fig. 3). The concentration of ^{232}Th was elevated in roots than in aboveground plant parts for all analyzed plants (higher from a factor of 3 in

dandelion to a factor of 25 in wild strawberries). Regarding ^{238}U , the highest root-to-aboveground plant ratio was obtained for grass and fern, while lowest was for dandelion and wild strawberry.

3.3. Transfer factors

Transfer factors (TFs) were included in the present work since the determination of transfer of contaminants from abiotic to biotic systems is a well-established and important step in the assessment of environmental contamination. TF values (^{232}Th , ^{238}U) for both soil-to-root and soil-to-plant are shown in Tables 3 and 4. In general, for each of the analyzed plant species the TF values varied significantly for different plant parts, i.e., for roots and aboveground parts ($p = 0.037$). This was expected and is in accordance with findings in similar studies (Roivainen *et al.*, 2011; Shahandeh and Hossner, 2002). In the pooled data from investigated sites, geometric means for TFs (^{232}Th) in vascular plants, moss and lichen, and trees were in the ranges $2.26 \cdot 10^{-3} - 4.46 \cdot 10^{-2}$, $4.81 \cdot 10^{-3} - 8.97 \cdot 10^{-3}$ and $4.50 \cdot 10^{-5} - 8.63 \cdot 10^{-4}$, respectively. The determined values were generally in accordance with the TFs published in TRS-364 and TRS-472 by IAEA (1994, 2010) and by Vandenhove *et al.* (2009b), although it should be highlighted that wide ranges were observed either after considering pooled data in the current study, or data in the cited literature.

Uranium TFs (pooled data) for vascular plants ranged from $3.11 \cdot 10^{-3}$ to $1.22 \cdot 10^{-1}$, for moss and lichen from $1.10 \cdot 10^{-2}$ to $4.12 \cdot 10^{-2}$ and for trees from $1.43 \cdot 10^{-4}$ to $5.33 \cdot 10^{-3}$. Values for vascular plants were in accordance with the ranges reported for fodder, pastures/grasses, and herbs from a compiled study on radionuclide TFs by Vandenhove *et al.* (2009b), and were comparable to the generic TF (^{238}U) value given in the same study. Furthermore, grasses TFs from the present study were comparable to those given in TRS 364 and TRS 472 by IAEA (1994, 2010) and Sheppard *et al.* (2006). Uptake of ^{238}U in plants was on average 10-fold higher than the uptake of ^{232}Th , most likely due to the higher solubility of ^{238}U in comparison to ^{232}Th and the higher mobility of ^{238}U in the Fen soils.

The ERICA software programme includes default concentration ratios (CR) for ^{238}U and ^{232}Th (Beresford *et al.*, 2008). These values are based on fresh weight biota, and to allow the comparison, data from the current study were converted to fresh weights. For ^{232}Th and ^{238}U in grasses and herbs, CR values in the ERICA ($4.37 \cdot 10^{-2}$ and $1.46 \cdot 10^{-2}$, respectively) were based on the work of Lapham and Millard (1989) and Mahon and Mathewes (1983). The converted CR values for grasses from the Fen area ($1.61 \cdot 10^{-3}$ and $2.81 \cdot 10^{-3}$, for ^{232}Th and ^{238}U , respectively), were lower than the ERICA adopted values. Further, the values for CR of ^{232}Th ($1.63 \cdot 10^{-3}$) and ^{238}U ($4.82 \cdot 10^{-3}$) in lichen were two and one order of magnitude lower respectively than the ERICA default values for ^{232}Th ($1.03 \cdot 10^{-1}$) and ^{238}U ($7.09 \cdot 10^{-2}$) (Litver *et al.*, 1976; Verhovskaya 1972). Similarly, obtained CR values for ^{232}Th and ^{238}U in trees, $1.15 \cdot 10^{-5}$ and $4.55 \cdot 10^{-4}$, respectively, were two and one order of magnitude lower than the ERICA default CR values for ^{232}Th ($1.08 \cdot 10^{-3}$) and ^{238}U ($6.79 \cdot 10^{-3}$) (Hinton *et al.*, 2005; Mahon and Mathewes, 1983; Sheppard and Evenden, 1988).

Student's t-test, applied on sets of TF data calculated with total soil concentration and with mobile soil radionuclide fraction, demonstrated significant difference ($p < 0.0005$) in all plant species, for roots and aboveground parts, for both ^{232}Th and ^{238}U . Values for TFs based on mobile soil fractions were factor 5-50 higher than TFs based on total soil concentrations. Although higher TF values calculated with mobile soil radionuclide fractions are a direct reflection of lower denominator, and the approach which considers TFs with bioavailable instead of total soil concentrations has not been usually accepted nor justified, these values provide better insight into the real mobility and uptake of radionuclides. Many studies conducted in the last decades presented TFs for long-lived radionuclides in wide ranges and variation, which may exceed three orders of magnitude (Frissel, 1992). According

to Ehklen and Kirchner (2002), such variability in the whole TF concept could be reduced by using the bioavailable soil radionuclide fraction in derivation of TFs. However, the available TFs concept might have other sources of uncertainty, such as choice of leaching method that is optimal for obtaining the bioavailable fractions.

3.4. Assessment of radiation doses to biota – application of the ERICA Tool

The ERICA Tool was used to calculate the exposure dose rates of terrestrial plants in the Fen Complex. The detailed description of the ERICA assessment tool and examples of assessment are given elsewhere (Brown *et al.*, 2008; Hosseini *et al.*, 2011; Larsson, 2008; Oughton *et al.*, 2013). Whole body plant doses were calculated using pooled data for the Fen former mining site and undisturbed ^{232}Th -rich Bolladalen site, since significant differences were seen neither in soil nor in plant radionuclide concentrations at these sites. Two separate dose rate calculations, initial and site-specific, were done (Fig. 4). In the initial screening, the maximal measured Fen soil radionuclide activity concentrations (16516, 859, 7300, 251 and 76 Bq/kg of ^{232}Th , ^{238}U , ^{228}Ra , ^{226}Ra and ^{210}Po , respectively) and concentrations of ^{228}Th and ^{234}U assumed to be in secular equilibrium with the parent radionuclide in soil were used as input data. The default CR values from the ERICA programme were selected. Doses were calculated for grasses and herbs, lichens and bryophytes, shrubs and trees. The screening level for radiation dose rates was 10 $\mu\text{Gy/h}$. The predicted dose rates ranged from 5 to 206 $\mu\text{Gy/h}$. The lowest dose was estimated for trees (5 $\mu\text{Gy/h}$), the highest for lichen and bryophytes (206 $\mu\text{Gy/h}$), while the doses higher than screening level, 82 and 32 $\mu\text{Gy/h}$, were calculated for grasses and herbs and for shrubs, respectively. Internal exposure to ^{228}Th and ^{232}Th gave the major contribution to the doses for grasses, herbs and shrubs. For lichen and bryophytes, beside the main contribution from ^{228}Th and ^{232}Th , the additional dose contribution was estimated to be from the internal exposure to ^{210}Po and ^{226}Ra . On the contrary, the major exposure route estimated for trees was via external exposure to ^{228}Th and ^{228}Ra . Dose rate values above the adopted screening value of 10 $\mu\text{Gy/h}$ suggested a need for more refined analysis.

The site-specific data used in the second ERICA assessment were soil and plants activity concentrations of radionuclides ^{232}Th , ^{228}Ra , ^{238}U , ^{226}Ra and ^{210}Po (sections 3.1 and 3.2), soil activity concentrations of ^{228}Th and ^{234}U assumed to be in secular equilibrium with the parent radionuclides, plant activity concentration of ^{234}U assumed to be in secular equilibrium with the ^{238}U , site-specific CRs recalculated from TFs (section 3.3). The range of recalculated dose rates (1 – 23 $\mu\text{Gy/h}$) was different and much lower than the one obtained by the initial screening. Somewhat different contribution of radionuclides and exposure routes were also predicted. For grasses and herbs, main contribution to the dose rate (6 $\mu\text{Gy/h}$) was from internal exposure to ^{228}Th and ^{226}Ra and external exposure to ^{228}Ra . About 75 % of the lichen dose (23 $\mu\text{Gy/h}$) was estimated to be from the internal exposure to ^{210}Po , while the rest of 25 % due to internal exposure to ^{226}Ra and ^{228}Th . In cases of shrubs (1 $\mu\text{Gy/h}$) and trees (2 $\mu\text{Gy/h}$), the external exposures to ^{228}Th and ^{228}Ra were the most important regarding the doses contribution. The decrease in the dose rates in the site-specific assessment of the Fen Complex plants in comparison to the initial one was contributed mostly to two things. First, more realistic exposure scenario using the median (and not maximal measured) soil activity concentrations was used in the second calculation. Second, the site-specific CRs of Th and U were lower than the default CRs in the ERICA Tool what consequently diminished the internal doses from ^{228}Th and ^{232}Th which were shown to be the major doses contributors in the initial screening. In lichen and bryophytes, the dose from the internal exposure to ^{210}Po remained about the same due to the comparable site-specific and the default CRs. However, the contribution from ^{228}Th which was the main contribution in the initial assessment decreased significantly due to the much lower site-specific CR used for ^{228}Th in

the site-specific calculations. The obtained lower results in the site-specific assessment confirmed the uncertainty related to use of mobility and transfer parameters in the assessment studies (Oughton *et al.*, 2008; Salbu, 2004). The CR concept takes into the consideration the total soil activity concentrations and as such is commonly accepted. However, without the knowledge on the actual mobility of radionuclides in the soil, both under and over estimation of the soil-to-plant transfer could happen. Although the dose rates for the selected Fen Complex wild plants (1 – 23 $\mu\text{Gy/h}$) were higher than the background range for terrestrial plants (0.02 – 0.7 $\mu\text{Gy/h}$) and animals (0.01 – 0.44 $\mu\text{Gy/h}$) (UNSCEAR, 2008), they did not imply an elevated radiation risk. In fact, the calculated dose rates lie below the levels of 40 and 400 $\mu\text{Gy/h}$ for terrestrial animal and plants, respectively, adopted by international organizations as values below which no effects on population levels should be expected (IAEA, 1992; UNSCEAR, 2008).

4. Conclusions

Based on the high ^{232}Th and moderate ^{238}U activity concentrations in the soil of the Fen Complex, the mobilization of bioavailable radionuclide species and their accumulation in terrestrial plants was expected. To assess the potential impact, nine wild plant species were collected, and tissue concentrations, transfers from soil-to-plant and radiation exposure dose rates were studied. Although significantly high ^{232}Th activity concentrations in soil were demonstrated at both legacy NORM and the undisturbed surrounding NORM site, maximal plant tissue concentrations measured in grass, dandelion and moss were comparable to previously published values in plants worldwide. Uranium concentrations in plants were several times lower than ^{232}Th concentrations, reflecting more the lower abundance of ^{238}U in the soil than the higher ^{232}Th bioavailability. It was demonstrated that the roots of vascular plants served as a kind of transport barrier, as concentrations of both radionuclides were higher up to a factor of 25 in roots than in aboveground plant parts. With respect to all analyzed plant species, the geometric means of TF (^{232}Th) and TF (^{238}U) were in ranges comparable to those given in reports TRS-364 and TRS-472 by the IAEA (1994, 2010) for terrestrial vegetation. Soil-to-roots transfer was, on average, one order of magnitude higher with regard to both analyzed radionuclides. The exposure doses for the selected terrestrial plants ranged from 1 to 23 $\mu\text{Gy/h}$ after calculation with site-specific data, including the primordial radionuclides ^{232}Th and ^{238}U , and their progenies as well. Maximal doses were obtained for lichen and bryophytes, with internal exposure to ^{210}Po and ^{228}Th dominating the assessment. In general, the demonstrated doses were below the values that could cause harm to plants.

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Table 1. Soil characteristics of investigated soil (n = 10)

	pH	OM (%)	Sand (%)	Silt (%)	Clay (%)
Fen	6.7	20.1	39.6	48.0	12.4
Bolladalen	5.9	20.6	31.7	54.5	13.8
Torsnes	6.1	15.5	16.0	65.7	18.3

Table 2. Radionuclide activity concentrations in soil (Bq kg⁻¹ d.w; median and ranges shown; n = 7 – 20). Percentages of mobile soil radionuclide fraction in are given (% are in parenthesis).

Site (description)	Total soil concentration		Mobile soil concentration	
	²³² Th	²³⁸ U	²³² Th	²³⁸ U
Fen	2737 (1589-5069)	115 (61-156)	14 (0.69) (4-55)	5 (4.70) (2-9)
Bolladalen	6835 (2146-16516)	228 (96-859)	13 (0.17) (3-157)	12 (4.92) (4-27)
Torsnes	664 (55-2270)	34 (8-109)	0.2 (0.03) (0.05-0.6)	0.8 (2.35) (0.7-1.1)

Table 3. Transfer factors of ^{232}Th for different plant species (geometric mean with geometric standard deviation in parenthesis)

Plant	Type of TF	TF (^{232}Th)		
		Fen (n = 5 - 12)	Bolladalen (n = 5 - 14)	Torsnes (n = 3)
Fern	soil-to-root	3.15E-02 (1.46) (b)	5.91E-03 (1.39) (a)	-
	soil-to-plant	6.10E-03 (2.56) (abc)	2.38E-03 (6.87) (a)	4.588E-03 (1.79) (bc)
Grass	soil-to-root	9.40E-02 (1.41) (a)	1.81E-02 (1.34) (a)	1.35E+00 (1.38)
	soil-to-plant	9.41E-03 (2.43) (a)	42E-03 (3.75) (a)	4.46E-02 (1.05) (a)
Dandelion	soil-to-root	1.15E-01 (2.11) (a)	2.41E-03 (1.76) (b)	-
	soil-to-plant	7.18E-03 (3.07) (abc)	7.90E-03 (1.70) (a)	-
Wild strawberry	soil-to-root	1.43E-01 (1.26) (a)	1.58E-02 (1.48) (a)	-
	soil-to-plant	1.02E-02 (1.92) (abc)	2.26E-03 (1.53) (a)	-
Moss	soil-to-plant	8.97E-03 (1.56) (abc)	6.04E-03 (2.46) (a)	6.03E-03 (1.61) (b)
Lichen	soil-to-plant	8.89E-03 (2.42) (ab)	4.80E-03 (1.49) (a)	-
Spruce*	soil-to-needles	6.51E-05 (1.48) (bc)	4.55E-05 (1.52) (b)	7.45E-04 (1.50) (c)
Birch*	soil-to-leaves	4.99E-05 (1.31) (c)	5.57E-05 (1.48) (b)	3.07E-04 (1.57) (c)
Pine*	soil-to-needles	3.33E-04 (3.19) (abc)	8.62E-04 (1.76) (a)	-

*The calculation of TFs for ^{232}Th in plants where measured concentrations were lower than detection limit was done by using value $0.5 \cdot \text{LOD}$; soil-to-root or soil-to-plant transfer factors of analyzed plants sharing same letters are not statistically different (ANOVA, Tukey's post-hoc, $p \leq 0.05$).

Table 4. Transfer factors of ^{238}U for different plant species (geometric mean with geometric standard deviation in parenthesis)

Plant	Type of TF	TF (^{232}Th)		
		Fen (n = 5 - 12)	Bolladalen (n = 5 - 14)	Torsnes (n = 3)
Fern	soil-to-root	6.80E-02 (1.61) (b)	1.76E-02 (1.34) (b)	-
	soil-to-plant	9.18E-03 (1.94) (bcd)	3.81E-03(3.29) (ab)	5.74E-03 (1.30) (b)
Grass	soil-to-root	2.66E-01 (1.45) (a)	1.49E-01 (1.48) (a)	9.87E-01 (1.41)
	soil-to-plant	2.58E-02 (1.67) (ab)	3.77E-03 (4.71) (ab)	1.22E-01 (1.25) (a)
Dandelion	soil-to-root	1.24E-01 (1.49) (b)	2.33E-02 (1.35) (b)	-
	soil-to-plant	2.00E-02 (1.86) (bc)	6.67E-03 (2.59) (ab)	-
Wild strawberry	soil-to-root	1.13E-01 (1.63) (b)	4.67E-02 (1.34) (b)	-
	soil-to-plant	1.12E-02 (1.60) (bcd)	2.71E-03 (1.66) (ab)	-
Moss	soil-to-plant	4.07E-02 (1.87) (a)	1.33E-02 (2.97) (ab)	1.11E-02 (1.13) (b)
Lichen	soil-to-plant	2.20E-02 (1.49) (bc)	2.65E-02 (1.52) (a)	-
Spruce*	soil-to-needles	4.31E-04 (1.60) (d)	1.40E-04 (2.10) (b)	9.73E-04 (1.28) (b)
Birch*	soil-to-leaves	1.63E-03 (1.35) (d)	4.16E-04 (3.04) (ab)	8.67E-04 (1.93) (b)
Pine*	soil-to-needles	5.33E-03 (1.85) (cd)	4.72E-03 (1.38) (ab)	-

*The calculation of TFs for ^{238}U in plants where the measured concentrations were below the detection limit was performed using value $0.5 \cdot \text{LOD}$; soil-to-root or soil-to-plant transfer factors of analyzed plants sharing the same letters are not statistically different (ANOVA, Tukey's post-hoc, $p \leq 0.05$).

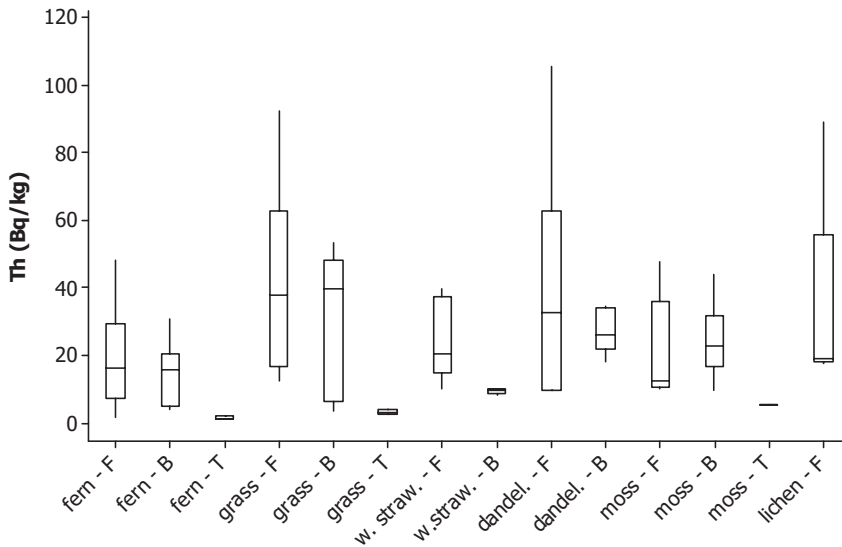


Figure 1. Activity concentrations of ^{232}Th (Bq/kg) in plants. Plant name is accompanied with letters F, B, T for Fen, Bolladalen and Torsnes, respectively. Boxes represent 25th percentile, median and 75th percentile. Error bars represent minimum and maximum values. Concentrations in birch, spruce and pine leaves were below the detection limit and were excluded from Fig.1. Dandelion and wild strawberries were not found at site Torsnes, whereas lichen was not collected from Bolladalen or Torsnes.

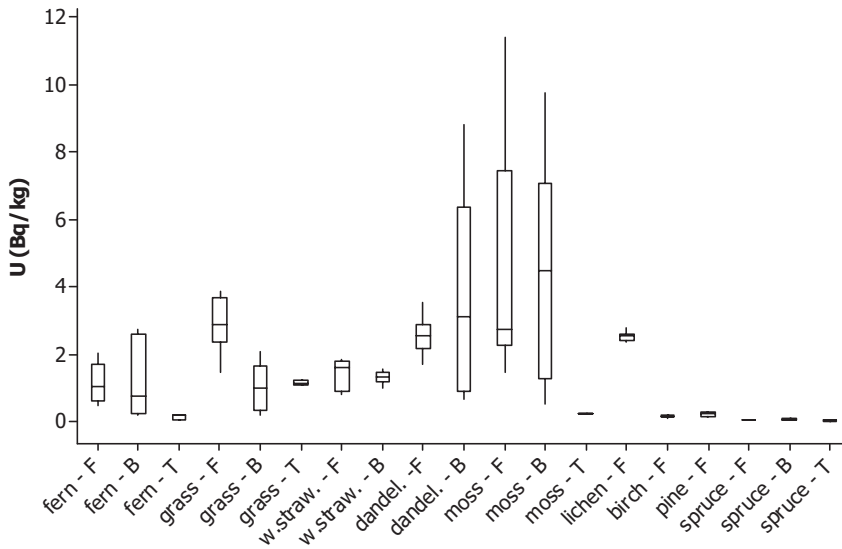


Figure 2. Activity concentrations of ^{238}U (Bq/kg) in plants. Plant name is accompanied with letters F, B, T for Fen, Bolladalen and Torsnes, respectively. Boxes represent 25th percentile, median and 75th percentile. Error bars represent minimum and maximum values. Dandelion and wild strawberries were not found at Torsnes, whereas lichen was not collected from Bolladalen or Torsnes.

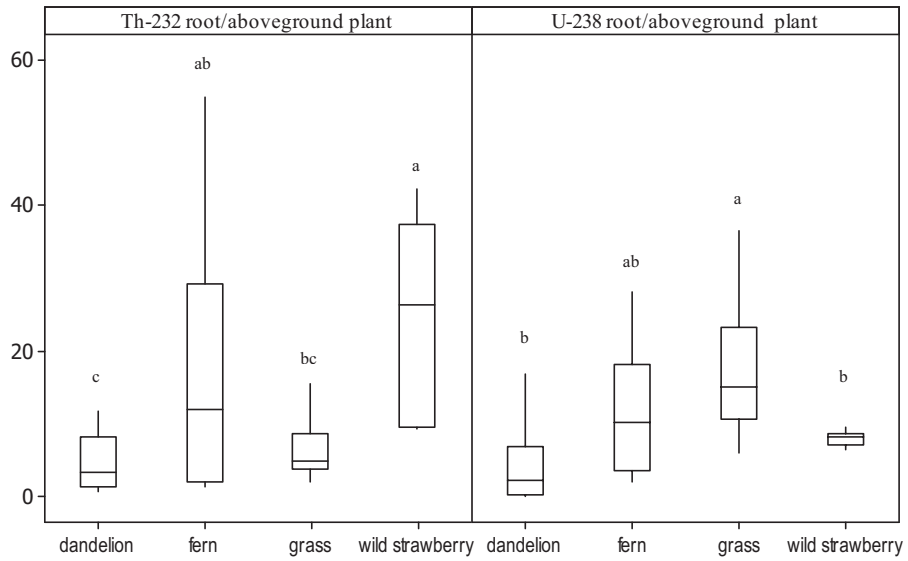


Figure 3. ^{232}Th and ^{238}U root/aboveground plant ratio in different plant species; plants sharing the same letter within each panel demonstrated no significant difference in root/aboveground plant ratio (ANOVA, Tukey's post hoc analysis, $p \leq 0.05$).

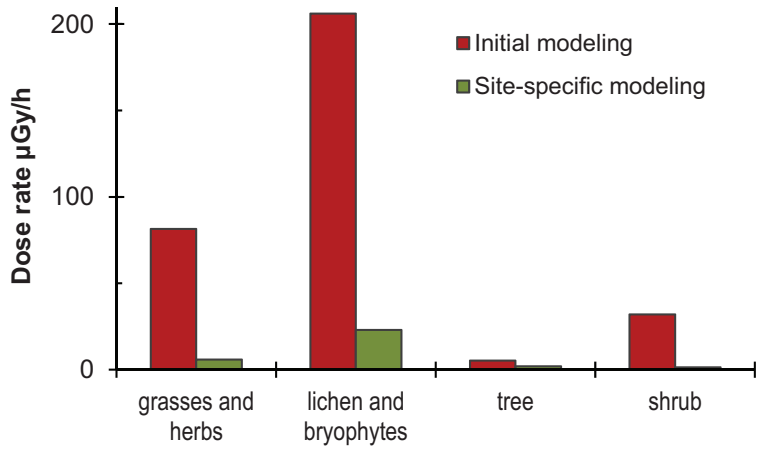


Figure 4. Calculated dose rates for selected terrestrial plants in the Fen Complex area. Results from two different calculations are shown.

Paper V

Outdoor ^{220}Rn , ^{222}Rn and terrestrial gamma radiation levels: investigation study in the thorium rich Fen Complex, Norway

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The present study was done in the Fen Complex, a Norwegian area rich in naturally occurring radionuclides, especially in thorium (^{232}Th). Measurement of radioactivity levels was conducted at the decommissioned iron (Fe) and niobium (Nb) mining sites (TENORM) as well as at the undisturbed wooded sites (NORM), all open for free public access. The soil activity concentrations of ^{232}Th (3280–8395 Bq kg^{-1}) were significantly higher than the world and the Norwegian average values and exceeded the Norwegian screening level (1000 Bq kg^{-1}) for radioactive waste, while radium (^{226}Ra) was present at slightly elevated levels (89–171 Bq kg^{-1}). Terrestrial gamma dose rates were also elevated, ranging 2.6–4.4 $\mu\text{Gy h}^{-1}$. Based on long-term surveys, the air concentrations of thoron (^{220}Rn) and radon (^{222}Rn) reached 1786 and 82 Bq m^{-3} , respectively. Seasonal variation in the outdoor gamma dose rates and Rn concentrations was confirmed. Correlation analyses showed a linear relationship between air radiation levels and the abundance of ^{232}Th in soil. The annual outdoor effective radiation doses for humans (occupancy 5 h day^{-1}) were estimated to be in the range of 3.0–7.7 mSv, comparable or higher than the total average (summarized indoor and outdoor) exposure dose for the Norwegian population (2.9 mSv year^{-1}). On the basis of all obtained results, this Norwegian area should be considered as enhanced natural radiation area (ENRA).

1. Introduction

Natural radiation, comprising cosmic and radiation from terrestrial radionuclides with half-lives comparable to the age of the earth, is ubiquitous. Specific radiation levels due to terrestrial background radiation are positively correlated with geological structure of the terrain and current geological conditions.¹ According to UNSCEAR,² exposure from naturally occurring radioactive materials gives the major contribution to the total

effective radiation dose of the population. The exposure pathways include internal exposure, mainly due to the inhalation of radon (^{222}Rn , half-life 3.8 d) and its progeny, and external exposure due to the gamma irradiation with radionuclides originated in primordial radionuclides of earth's crust (decay chains of thorium (^{232}Th) and uranium (^{238}U , ^{235}U), as well as radionuclide potassium (^{40}K)).

The world average annual effective dose per capita is estimated to be 2.4 mSv of which 1.1 mSv results from inhalation of indoor ^{222}Rn and its progeny.³ The indoor ^{222}Rn levels, emanation processes, measurement methods and associated health risks have been heavily investigated.^{4–7} In contrast, information on thoron (^{220}Rn , half-life 56 s) is scarce, probably due to the short

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Environmental impact

Natural radiation, comprising cosmic and radiation from terrestrial radionuclides with half-lives comparable to the age of the earth, is ubiquitous. In enhanced natural radiation areas (ENRA), like the one investigated in the current study, the contribution of exposure to terrestrial gamma radiation and radon ($^{220,222}\text{Rn}$) levels could overcome other radiation exposures and pose a serious risk for people living there. The Fen Complex, in southern Norway, is an area significantly rich in thorium (Th) ore. Several assessment studies have previously shown the increased risk for humans correlated mainly with indoor exposure to radon (^{222}Rn). However, the current study is the first one showing the outdoor levels of gamma radiation, ^{222}Rn and thoron (^{220}Rn), and their possible significant contribution to the total radiation exposure dose. The analyses of the magnitude of radiation parameters, their seasonal variation, and correlation with mother radionuclides in the soil (^{232}Th and ^{238}U) put additional light on the complex radiation exposure scenario in this area.

half-life, difficulties in measurement techniques and limited abundance in nature. Still, ^{230}Rn has recently been recognized as a potential health hazard in worldwide areas rich in ^{232}Th , and therefore, its determination and risk estimation are emphasized as being important.^{3,8–10}

The Fen Complex area, situated in the Norwegian county of Telemark, is well known for its specific bedrock of volcanic magmatic origin,^{11,12} containing ^{232}Th and ^{238}U rich rock types (e.g., rödbergite, rauhaugite, sövite, fenite). Due to the elevated levels of a series of metals, mining of iron (Fe) ores and rare earth elements (e.g., niobium (Nb)) has been performed during the past centuries. Previous studies in the Fen Complex have demonstrated elevated levels of radionuclides within the mining areas, as well as elevated indoor radiation exposure and associated human risk.^{13–16} The major health issue in terms of radiation exposure has been associated with the inhalation of ^{222}Rn progeny.^{14,17} The estimated annual doses in the Fen Complex area are reported up to four times higher than the Norwegian average effective dose of 2.9 mSv.¹³ However, the contribution of the outdoor exposure to the total radiation dose has not been previously determined.

Even though data on radiation doses due to outdoor occupancy are generally much less available compared to those due to indoor occupancy, such data are important for estimating human doses, especially in the areas of high natural radiation.³ With respect to that, the results from the present study could give a valuable insight into outdoor radiation exposure issues.

The objective of this study was to investigate the air concentration levels of ^{220}Rn and ^{222}Rn , as well as terrestrial gamma dose rates, at sites rich in naturally occurring radioactive materials (NORM) and technologically enhanced naturally occurring radioactive materials (TENORM). The sites are freely accessible to the public and significantly rich in ^{232}Th and moderately in ^{238}U . The seasonal variations and correlations between radiation variables were also evaluated. Based on the attained results, the possible annual effective radiation dose received by humans outdoor was estimated.

2. Materials and methods

2.1. Study area

The study area is located in a small village Fen, in south-eastern Norway, about 009° 18' E and 59° 16' N. The whole area is known as the Fen Complex and has unique bedrock geology of volcanic origin. Based on our previous work,¹⁸ two TENORM sites (Fen and Söve mining sites) and three NORM sites (Bolladalen, Gruvehaugen and Rullekoll) that had high terrestrial gamma dose rates and elevated rocks and soil ^{232}Th and ^{238}U activity concentrations were selected for the investigation of outdoor radiation levels (Fig. 1).

The Söve site (TENORM) is an abandoned mining facility on the west part of the Fen Complex. The mining operation was based on limestone mineral sövite, present in abundance at this site. Sövite consists mainly (75–95%) of calcium carbonate and some minerals (e.g., pyrochlore, columbite, fersmite) rich in Nb (0.35% Nb₂O₅) and to a lesser degree radionuclides ^{232}Th and ^{238}U . The mining activities, related to ferro-niobium production, were conducted during 1953–1965. Large amounts of waste in

the form of crushed rocks and slag, enriched with radioactive elements (^{232}Th and ^{238}U and their daughters), were left out in the area following the mining activities.^{16,19} The site, in fact, was covered with sand layers in a remediation action conducted after the decommission. However, several points in this area have recently been investigated^{16,18,20} revealing significantly inhomogeneous and elevated radionuclide concentrations in soil, as well as elevated terrestrial gamma radiation dose rates. Currently, a mechanical workshop is in operation at one part of this site.

Mining of Fe was conducted in the Fen Complex at several locations during 1650–1929. As a consequence, certain wooded zones are found to be with elevated radiation levels.¹⁸ Major Fen rock types, rauhaugite (magnesium–calcium–carbonate) and rödbergite (ferro–carbonate), contain little of Nb, but are enriched in ^{232}Th , rare earth elements, and Fe. Rödbergite, in particular, is considered as a possible source for future Th mining and exploitation.¹⁶ The Fen site (TENORM) is situated in the north part of the Fen Complex, along the shores of Lake Norsjø where Fe mining waste rocks are still deposited.

Bolladalen and Gruvehaugen sites comprise the area in the central Fen Complex wooded zone. The investigated subsites within these sites were considered as NORM since no waste from the previous Fe mining was found there, although some mining holes and open tunnels were observed (limited and not accessible).

The Rullekoll site (NORM) is an undisturbed site located in the south of the Fen Complex, consisting of a small forest just above the human settlement area. Radionuclide ^{232}Th and its progeny, significantly elevated in rödbergite rock found at this site, result in higher radiation levels in the soil and air.¹⁸

2.2. Study design

Five field expeditions (2008–2010) were organized in different seasons, allowing the investigation of the radiation parameters not only in stable spring–summer conditions (May–August 2008, 2009 and 2010), but also in rainy and snowy conditions (October and November 2009). Since four of the five investigated sites (F, B, G, R) (Fig. 1) were large and comprised of wooden areas, a certain number of subsites, within each of the main sites, were investigated in order to obtain more reliable data. The geographical positions of all sampling and measurement points within sites were recorded with the Global Positioning System (GPS, Garmin, USA). Additionally, the sites were photographed, enabling easy return in the following field expeditions.

To obtain all necessary data for this study, the following activities were conducted:

- Soil sampling for *ex situ* gamma ray spectrometry,
- *In situ* measurements of gamma dose rates,
- Continuous measurements of ^{220}Rn and ^{222}Rn air concentrations (two seasonal, three months long surveys).

2.2.1. Soil sampling and *ex situ* radiometric analysis. Soil sampling was conducted simultaneously with the gamma radiation measurements and detector placement at GPS recorded sites. Samples were collected using the soil corer with a size diameter of 10 cm. At each site, soil corers were taken from at least 5 sampling points with a distance of 5 m between them, at depth up to a maximum of 20 cm. The abundance of



Fig. 1 Map showing the studied sites in the Fen Complex, Norway: Søve mining site (S), Fen mining site (F), Bolladalen (B), Gruvehaugen (G) and Rullekoll (R) (source of original map: Norwegian Mapping Authority).

rocks in soil layers was a limiting factor regarding the sampling depth. The soil was packed into the polyethylene bags and properly marked. In the laboratory, the collected soil was dried at 110 °C, crushed and sieved to less than 2 mm. The gamma spectrometry measurement of samples was done at the Norwegian Radiation Protection Authority (NRPA). The soil was prepared in cylindrical geometries, isolated in aluminium foil and kept for a month before measurement to ensure secular equilibrium. Measurements were carried out on three different coaxial p-type detectors: a Canberra GR2521-7500 with 45% relative efficiency and a FWHM of 1.9 keV; an Ortec GEM-40190-S with 40% relative efficiency and a FWHM of 1.8 keV and an Ortec GEM-33190-S with 33% relative efficiency and a FWHM of 1.8 keV. Samples were generally counted for one or two days depending on the activity level (minimum measuring time of 16 hours for the most active samples). Quantitative analysis of ^{40}K was based on primary photon emissions (1460 keV), while ^{226}Ra was determined via ^{214}Pb (352 keV) and ^{214}Bi (609 keV) and ^{232}Th via ^{228}Ac (911 keV). Spectra were obtained through an Ortec Maestro v6 and spectrum analysis was carried out using self-written Sampo-based software. The measurement uncertainties at the 1σ level were in the range 4–8, 1–6 and 5–9% for ^{226}Ra , ^{232}Th and ^{40}K , respectively. To ensure the quality of analysis, the control of detector efficiency calibration and background levels were in place, as well monitoring of energy calibration and laboratory climate.

2.2.2. Gamma dose rate measurements. *In situ* spatial surveys of gamma dose rates ($\mu\text{Gy h}^{-1}$) were carried out by a portable gamma detector (Automess, Radiacmeter 6150 AD 4 LF) calibrated with ^{137}Cs source. The response range of the instrument was 0.01 $\mu\text{Gy h}^{-1}$ to 9.99 mGy h^{-1} . Measurements were

conducted at 1 m above the ground surface. A regular grid 10×10 m was used at the subsites, and readings were repeated until constant signal. The dose rate in the air at each of the sites was obtained as the arithmetic mean of all measurements at the subsites. The data collection was performed for different months (May, September, November and June), allowing us to identify the seasonal variation. Besides the studied sites, several readings at public places (road, vicinity of the houses, schoolyard) were also recorded, providing the background dataset of dose rates for comparison purposes.

2.2.3. Measurements of ^{220}Rn and ^{222}Rn air concentrations. Continuous measurements of ^{220}Rn and ^{222}Rn concentrations (Bq m^{-3}) in the air were performed in two separate periods, during September–November 2009 and June–August 2010. Passive integrating ^{222}Rn – ^{220}Rn discriminative detectors, type RADUET (RadoSys Co., Ltd., Budapest, Hungary), were used.²¹ The detectors, consisting of two different diffusion chambers, were developed and evaluated at the National Institute of Radiological Studies (NIRS), Chiba, Japan.²² These detectors have been widely used in surveys throughout the world.^{9,23}

In the current study, the detectors were fixed to the trees, lying 1 m above the ground surface. A total of 86 detectors were placed at five chosen sites, and 82 were recovered. After collecting the detectors, they were sent to the Japan Chemical Analysis Center, Chiba, for counting. The detailed performance has been described by Zhuo *et al.*²¹ and Tokonami *et al.*²⁴

2.3. Data analyses

Statistical analyses were performed using a Minitab 16 (Minitab Inc.). Normality was assessed with the Anderson–Darling test. The ^{220}Rn and ^{222}Rn concentrations were logarithmic

transformed prior to statistical tests. Difference analyses of two measurement sets (^{220}Rn and ^{222}Rn) from different seasons (summer and fall) were done with the Student's *t*-test. One-way ANOVA, followed by Tukey's *post hoc* description, was used in difference analysis of ^{220}Rn , ^{222}Rn and gamma dose rates measured at the investigated sites, as well as in comparison of gamma dose rates measured in four different months. Correlation analyses were performed using the Pearson correlation coefficient. For all the analyses, the *p*-value lower than 0.05 is considered statistically significant.

In addition to the measurements, the gamma dose rates in the air were calculated on the basis of soil gamma spectrometry results, following the guidelines of UNSCEAR.²

$$D(\text{nGy h}^{-1}) = 0.042C_{\text{K}} + 0.604C_{\text{Th}} + 0.462C_{\text{Ra}} \quad (1)$$

where *D* is the gamma dose rate; 0.042, 0.604 and 0.462 are conversion factors expressed in nSv h⁻¹ per activity unit and *C*_K, *C*_{Th} and *C*_{Ra} are soil activity concentrations (Bq kg⁻¹) of ^{40}K , ^{232}Th and ^{226}Ra , respectively. Contributions of other soil radionuclides, e.g., ^{90}Sr , ^{137}Cs and ^{235}U , were considered as insignificant and were not taken into the calculation of the dose rates.²⁵ Calculated gamma dose rates were compared to the measurements obtained with the portable detectors using the Student's *t*-test.

The annual outdoor effective doses from external gamma radiation were determined using the following equation:²

$$H_{\text{gamma rad.}} (\text{mSv}) = D (\text{nGy h}^{-1}) \times 8760 \text{ h} \times 0.2 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-6} \quad (2)$$

where *H* is the effective dose, *D* is the measured gamma dose rate, 0.2 is the outdoor occupancy factor and 0.7 Sv Gy⁻¹ is the conversion coefficient from absorbed dose in air to human effective dose equivalent.

Effective doses from ^{220}Rn and ^{222}Rn outdoor were estimated as:²

$$H_i (\text{mSv}) = C_i \times F_i \times t \times \text{DCF}_i \times 10^{-6} \quad (3)$$

i = (^{220}Rn or ^{222}Rn)

where *H*_{*i*} is the effective dose, *C*_{*i*} is the ^{220}Rn (^{222}Rn) air concentration (Bq m⁻³), *F*_{*i*} is the equilibrium factor 0.003 for ^{220}Rn (ref. 2 and 3) and 0.6 for ^{222}Rn (ref. 2); *t* is the exposure time (1752 hours) and DCF is the dose conversion factor 40 nSv Bq⁻¹ equivalent equilibrium concentration (EEC) h m⁻³ for ^{220}Rn and 9 nSv Bq⁻¹ EEC h m⁻³ for ^{222}Rn .

Finally, total outdoor effective dose was obtained as sum:

$$H_{\text{tot}} = H_{\text{gamma rad}} + H_{220\text{Rn}} + H_{222\text{Rn}} \quad (4)$$

3. Results and discussion

3.1. Soil analysis

The activity concentrations of the terrestrial radionuclides in soil samples from different Fen Complex sites are given in Table 1. The world average soil activity concentrations of ^{226}Ra , ^{232}Th and

Table 1 Radionuclide activity concentration of the Fen Complex soil (median and range (in parenthesis) are given)

Site	N	$^{226}\text{Ra}/$	$^{232}\text{Th}/$	$^{40}\text{K}/$
		Bq kg ⁻¹	Bq kg ⁻¹	Bq kg ⁻¹
Bolladalen	5	127 (66–234)	8395 (1740–15 500)	654 (496–921)
Fen mining site	5	122 (88–160)	3280 (2360–3940)	404 (366–429)
Gruvehaugen	8	110 (65–272)	8020 (5100–11 500)	500 (304–637)
Rullekoll	5	171 (38–376)	6655 (5560–9270)	439 (343–640)
Søve mining site	5	89 (77–101)	5650 (5130–6170)	551 (489–614)

^{40}K are estimated to be 32, 45 and 420 Bq kg⁻¹, respectively.^{2,26} The average Norwegian soil values are somewhat higher, i.e., 50, 45 and 850 Bq kg⁻¹ for ^{226}Ra , ^{232}Th and ^{40}K , respectively.^{2,26} The median values of ^{232}Th soil concentrations in the present work (3280–8395 Bq kg⁻¹) were two orders of magnitude higher, exceeding the screening value (1000 Bq kg⁻¹) for radioactive materials given by the Norwegian Pollution Act.²⁷ Wide ranges, showing the difference in ^{232}Th concentration between close sampling spots, suggested significantly inhomogeneous distribution at each of the investigated sites. This distribution could be considered as a consequence of bedrock weathering, formation of space localized radionuclides rich soil and existence of ^{232}Th rich soil particles. No statistically significant difference in ^{232}Th concentration was found regarding NORM and TENORM sites. The relationship analysis has previously shown the positive correlation (*r* = 0.78, *p* < 0.001) between gamma dose rates in the air and ^{232}Th concentration in the soil.¹⁸

Soil activity concentrations of ^{226}Ra were only slightly enhanced (89–171 Bq kg⁻¹), which was expected since the mother radionuclide (^{238}U) was present in rocks and soil to a significantly less degree than ^{232}Th .^{16,18} The lowest level was obtained at site Søve, but no significant differences between sites were confirmed. Similar results, regarding both ^{232}Th and ^{226}Ra , have been published for a high natural background radiation area (monazite and zircon rich) in India.²⁸

The concentration of ^{40}K (404–654 Bq kg⁻¹) was in the normal variation range for Norway given by UNSCEAR.^{2,26}

3.2. Gamma dose rates

Gamma dose rates, recorded with a portable detector at NORM and TENORM sites, were in the range of 2.6–4.4 μGy h⁻¹ (Table 2). Generally, all gamma dose rates were 30–80 times higher than the Norwegian average of 0.073 μGy h⁻¹ and the

Table 2 Gamma dose rates directly measured and calculated from soil activity concentrations (mean ± standard deviation); percentage contribution of ^{226}Ra , ^{232}Th and ^{40}K to calculated gamma dose rates

Site	Measured gamma dose rate/	Calculated gamma dose rate/			^{40}K (%)
	μGy h ⁻¹	μGy h ⁻¹	^{226}Ra (%)	^{232}Th (%)	
Bolladalen	3.2 ± 0.8	5.2 ± 0.4	1.1	98.3	0.5
Fen mining site	2.8 ± 0.3	2.1 ± 0.4	2.8	96.1	1.0
Gruvehaugen	4.3 ± 1.8	4.9 ± 1.6	1.0	98.5	0.4
Rullekoll	2.6 ± 0.4	4.1 ± 1.1	1.9	97.6	0.4
Søve mining site	4.4 ± 2.8	3.5 ± 0.4	1.2	98.1	0.7

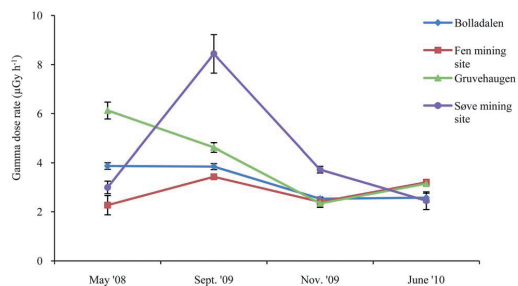


Fig. 2 Monthly variation of gamma dose rates in the Fen Complex (mean value \pm standard error); site Rullekoll is not shown because of high measurement uncertainty.

world average of $0.058 \mu\text{Gy h}^{-1}$ given by UNSCEAR,^{2,26} as well as higher than those obtained for different reference sites worldwide.^{29–31} However, results were comparable and confirmed the previous Fen Complex measurements obtained by Dahlgren,³² Heincke *et al.*,¹⁵ and IFE.²⁰ Significant differences ($p = 0.0005$) were found between some of the investigated sites. Although it indicated the site specific radioactivity enrichment, the comparison of soil activity concentrations at TENORM and NORM sites did not demonstrate any significant correlation between the enrichment and former mining activities. The highest mean gamma dose rate ($4.4 \mu\text{Gy h}^{-1}$) was obtained at a TENORM site, *i.e.*, at decommissioned Nb mining site Søve. However, the gamma dose rate obtained for NORM site Gruvehaugen had almost the same mean value ($4.3 \mu\text{Gy h}^{-1}$), while another NORM site (Rullekoll) showed the lowest gamma dose rate ($2.6 \mu\text{Gy h}^{-1}$). Furthermore, Fen, the former mining site (TENORM), showed similar dose rates ($2.8 \mu\text{Gy h}^{-1}$) to those of the Rullekoll site (NORM). Based on these results, the variation of the gamma dose rates is more likely related to the variation in the abundance of the certain rock types at chosen sites than to the presence or absence of former mining activities at the investigated sites.

The gamma dose rates, recorded outside the forest area (within the Fen living areas), with a mean value of $0.18 \mu\text{Gy h}^{-1}$, were statistically lower ($p = 0.0005$) than those recorded at NORM and TENORM sites. Still, these public sites had also the slightly enhanced gamma radiation dose rates in comparison to the world average.^{2,26}

In addition to measurements, a theoretical approach of gamma dose rate estimation, based on soil activity

concentrations of radionuclides (eqn (1)), was used and obtained results are shown in Table 2.

Computed gamma dose rates in the air due to naturally occurring radionuclides varied in the range of 2.1 – $5.2 \mu\text{Gy h}^{-1}$. The highest radionuclide contribution (>96%) to the outdoor gamma dose rates was from radionuclide ^{232}Th and its short lived gamma progeny, at all investigated sites. The comparison of gamma dose rates estimated from soil radionuclide activity concentrations and gamma dose rates directly measured showed no significant differences ($p = 0.4$).

Observed seasonal change of measured gamma dose rates is shown in Fig. 2. Statistically significant differences ($p < 0.001$, for each of the investigated sites) between gamma dose rate values in different months confirmed the seasonal variation at each of the investigated sites. The maximal mean value was obtained in early September (2009) at TENORM site Søve, showing the considerable difference ($p < 0.05$) from values obtained at other locations (Fig. 2). High readings at this site would give the annual absorbed gamma dose in the air up to 38.5 mGy (exposure time 8760 h). These considerably high but spatially limited gamma radiation measurements at Søve corresponded to inhomogeneous distribution of soil radioactivity, previously reported at the same location.¹⁶ Gamma dose rates, similar to present findings, have been previously reported for the Søve site by IFE²⁰ and NGI.¹⁶ For three of the four investigated sites, the mean value of gamma dose rate was highest in September, while lowest in November. The dry weather with no wind, recorded during the expedition in early September, and in contrast, strong wind, rain and snow cover of approximately 10 cm at the end of November, could provide the explanation for obtained readings. However, it is emphasized that no common seasonal variation pattern applicable to all sites was observed. The detailed recording of all atmospheric and weather conditions and other factors (*e.g.*, soil humidity and permeability) is essential to obtain a much accurate explanation of variation in gamma dose rates.

3.3. Continuous measurements of ^{220}Rn and ^{222}Rn air concentrations

The values for ^{220}Rn and ^{222}Rn concentrations in the air, obtained in summer and fall surveys, are given in Tables 3 and 4 respectively.

UNSCEAR^{2,3} suggests the typical outdoor concentration of ^{220}Rn is of the order 10 Bq m^{-3} , with the range from 1 to 100 Bq m^{-3} . The air ^{220}Rn concentrations from the summer continuous

Table 3 Air ^{220}Rn concentrations in the Fen Complex

Site	Summer survey				Fall survey			
	Arithmetic mean/ Bq m^{-3}	Standard deviation/ Bq m^{-3}	Geometric mean/ Bq m^{-3}	Range/ Bq m^{-3}	Arithmetic mean/ Bq m^{-3}	Standard deviation/ Bq m^{-3}	Geometric mean/ Bq m^{-3}	Range/ Bq m^{-3}
Bolladalen	1294	863	1029	284–2801	743	320	707	516–969
Fen mining site	1442	1155	1230	765–4996	445	259	406	262–628
Gruvehaugen	1786	860	1579	495–3495	741	359	668	339–1029
Rullekoll	1231	339	1189	709–2047	1000	138	994	896–1156
Søve mining site	91	90	67	24–362	7 ^a	—	7 ^a	—

^a One detector recovered at site Søve.

Table 4 Air ^{222}Rn concentrations in the Fen Complex

Site	Summer survey				Fall survey			
	Arithmetic mean/ Bq m^{-3}	Standard deviation/ Bq m^{-3}	Geometric mean/ Bq m^{-3}	Range/ Bq m^{-3}	Arithmetic mean/ Bq m^{-3}	Standard deviation/ Bq m^{-3}	Geometric mean/ Bq m^{-3}	Range/ Bq m^{-3}
Bolladalen	73	40	59	8–157	9 ^a	—	—	—
Fen mining site	82	56	64	9–210	<LOD	—	—	—
Gruvehaugen	61	31	53	21–120	5 ^a	—	—	—
Rullekoll	47	21	41	9–72	8	3	7	4–11
Søve mining site	29	5	28	24–37	13 ^b	—	—	—

^a Only one detector showed value > LOD. ^b One detector recovered at site Søve.

survey were found to be enhanced in comparison to the average world value, with the arithmetic and geometric mean in the range of 91–1786 and 67–1579 Bq m^{-3} , respectively. The fall survey demonstrated values with the arithmetic and geometric mean in the range of 7–1000 and 7–994 Bq m^{-3} , respectively.

Summer survey ranges were wider at each of the investigated sites than those recorded in fall, indicating the greater differences between subsites. However, these differences could be due to a higher number of detectors placed in the summer survey, giving more representative information, than due to the actual decrease in variation between subsites in the fall period. The demonstrated variation corresponded also to the significant inhomogeneous distribution of ^{232}Th in the soil of same subsites and suggested strong dependence of air levels on the soil concentration.

The decrease in the values of the fall ^{220}Rn concentration was noticed at all sites in comparison with summer values. However, except for the Søve site, the concentration values were still higher than the world average value. Sparse publications on outdoor ^{220}Rn have shown much lower air concentration in comparison to this study.^{33,34}

Difference analysis showed that the ^{220}Rn air concentration at mining site Søve was significantly lower ($p = 0.0005$) than ^{220}Rn air concentrations at other investigated sites in the summer survey. The lack of detectors at Søve in the fall survey did not allow the statistical analysis of difference, but the lowest value in the fall survey was also recorded for the Søve site. The possible explanation is the sand covering placed on the soil surface of the site to reduce the public exposure. No other significant

differences in ^{220}Rn air concentrations between NORM and TENORM sites were obtained, suggesting no ^{220}Rn air enrichment in terms of former mining.

The presence of much less ^{222}Rn than ^{220}Rn in the air in both seasons was demonstrated, reflecting again the bedrock and soil abundance of ^{232}Th and moderate levels of ^{238}U . The values of outdoor ^{222}Rn in the summer period (Table 4) were in the range of 8–210 Bq m^{-3} , with the arithmetic and geometric mean in the range 29–82 Bq m^{-3} and 28–64 Bq m^{-3} , respectively. These measurements were in agreement with the wide outdoor background range (1–100 Bq m^{-3}) given by UNSCEAR,^{2,3} but still higher than the world average^{2,3} of 10 Bq m^{-3} and higher than results published in the similar studies worldwide.^{35–37} A significant decrease of ^{222}Rn in the air was observed in the fall survey at all sites, giving non-measurable values at the majority of exposed detectors. No statistical differences were found in the ^{222}Rn air concentration between sites in the summer survey.

Seasonal (summer and fall) variation in ^{220}Rn and ^{222}Rn air concentrations is presented in Fig. 3. Student's *t*-test showed significant seasonal difference for both ^{220}Rn ($p = 0.0039$) and ^{222}Rn ($p = 0.0054$). In the present study, the summer values of both ^{220}Rn and ^{222}Rn were higher than those obtained in fall, in contrast to their characteristic 'high in winter and low in summer' behaviour.^{36,37} It seems that the Scandinavian weather conditions could be the reasons for these results. Decreased values of investigated variables in the fall could be explained with the low emanation and exhalation processes due to the significantly increased moisture content, snow coverage and frozen soil in fall

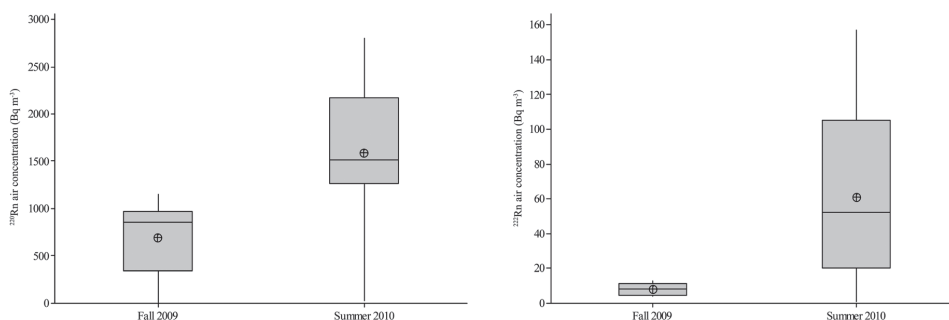


Fig. 3 Seasonal variation of ^{220}Rn (left) and ^{222}Rn (right) air concentrations. Box plot: the horizontal lines show the median; crossed circles average values; the top and bottom of the box show the 75th and 25th percentiles. The top and bottom of the whiskers show the maximum and minimum values.

months of 2009 (late October and November). The influence of soil humidity and snow coverage, on air ^{220}Rn and ^{222}Rn levels, has been previously studied and they have been suggested as the diminishing factors for ^{220}Rn and ^{222}Rn emanation.^{38,39} Additionally, more wind, rain and snow conditions in the air might act as the removal processes that contributed to lower diffusion and concentration of radon gases.

Seasonal behaviour similar to behaviour obtained in the current study has previously been published.^{6,40} For a better understanding of these seasonal differences, a detailed knowledge on precipitation levels, moving of air masses, temperature and atmospheric pressure, presence of clouds, humidity of air and soil moisture is needed.

3.4. Correlation analyses

Based on the present results, a scatter diagram, examining the correlation between ^{220}Rn and ^{222}Rn concentrations in the air, is presented in Fig. 4. The regression equation with a coefficient of $R^2 = 0.49$, an intercept of 462 and a slope of 16 was obtained ($p = 0.0005$). The correlation coefficient found in this study (0.7) is higher than previously published (0.009–0.42).^{7,9,41} The obtained y -axis intercept (462) and ratio (16) between ^{220}Rn and ^{222}Rn air concentrations suggested highly relevant prevalence of ^{220}Rn . It was expected since a significant soil enrichment of ^{232}Th has been previously reported¹⁸ and confirmed in the current study.

Correlation analysis demonstrated a moderate positive relationship between gamma dose rates and ^{220}Rn air concentrations ($r = 0.56$, $p = 0.001$), as well between gamma dose rates and ^{222}Rn air concentrations ($r = 0.64$, $p = 0.001$). The correlations, similar to those we obtained, have been published elsewhere.^{14,36}

Furthermore, a positive linear correlation between air ^{220}Rn concentrations and soil ^{232}Th activity concentrations was observed (Fig. 5).

The Pearson correlation coefficient of 0.66 ($p = 0.001$) suggested the dependence of air ^{220}Rn on geological composition of terrain. However, the value of slope (0.14) implicated limited ^{220}Rn in the air in comparison to what might be expected from ^{232}Th concentrations in soil. The spatial variation and possible significant decrease of air ^{220}Rn with distance from the soil are the most reasonable explanation. In addition, the emanation of ^{220}Rn and its diffusion through soil, which is highly dependent on soil conditions, might also be limited and hence affect the results.

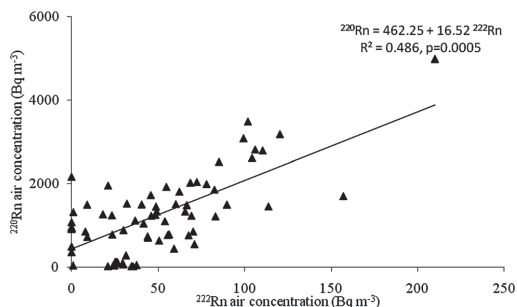


Fig. 4 Correlation between ^{220}Rn and ^{222}Rn air concentrations.

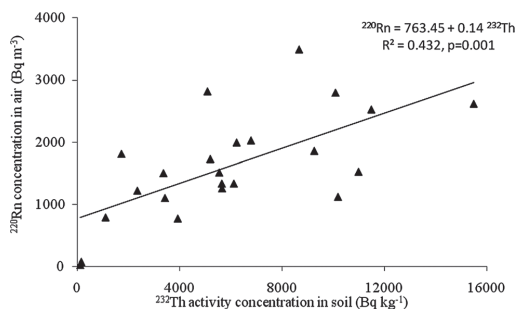


Fig. 5 Correlation between ^{220}Rn concentrations in air and the corresponding soil ^{232}Th activity concentrations.

3.5. Effective doses related to outdoor exposure to radiation

The inhalation of ^{220}Rn , ^{222}Rn and consequently the deposition of their stable progenies inside the human bodies together with external irradiation with gamma emitting radionuclides contribute significantly to the exposure dose of humans.

The total annual effective doses were calculated in the present study using the following relatively rough assumptions:

- Summer radiation dataset (^{220}Rn , ^{222}Rn and gamma dose rates) was considered as constant for the period of six months with relatively stable atmospheric conditions. Therefore, they were used in calculation of effective doses for a half-year exposure period (eqn (2) and (3)).

- Fall radiation dataset (^{220}Rn , ^{222}Rn and gamma dose rates) was considered as constant for the period of six months with unstable conditions (snow, rain, wind). Effective doses were derived from it for the other half-year period (eqn (2) and (3)).

Total annual effective doses were then obtained by summarizing the effective doses from two periods (eqn (4)). Although the calculations with these assumptions do not give completely accurate values of doses, they are useful as they provide the upper limits of possible doses and allow preliminary risk estimation.

Total exposure doses to the population due to outdoor exposures and comparison with the ICRP⁴² annual constraint value of 1 mSv are given in Fig. 6. The estimated total annual outdoor

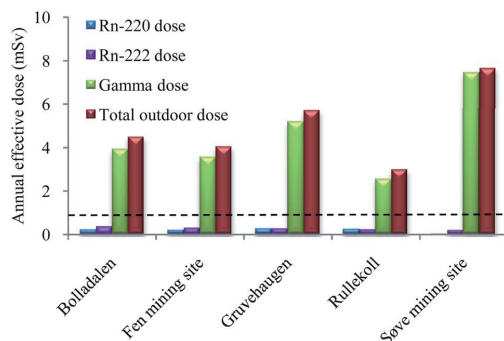


Fig. 6 Total annual outdoor effective dose for Fen Complex sites; the dashed line presents maximum constraint of radiation for the general public (ICRP).⁴²

effective doses were in the range 3.0–7.7 mSv. The lowest dose was obtained at NORM site (Rullekoll), while the highest at TENORM site (Søve). The major contributor to the total outdoor effective doses at all investigated sites was the dose from terrestrial gamma radiation (82.4–97.3%), both in stable and unstable weather conditions. The contribution of ^{220}Rn and ^{222}Rn was significantly lower, 0.1–7.8% and 2.5–8.0%, respectively. Estimated summer month doses were higher (up to 3 times) than fall at all sites. In all the cases, the outdoor doses were higher than the constraint value of 1 mSv given by ICRP⁴² for total (summarized outdoor and indoor) exposure of humans. Values of indoor radiation doses, previously published, could provide an additional insight into total annual exposure doses for the Fen population. The indoor concentrations of radon exceeding 200 and 400 Bq m⁻³ have been found in 37 and 11% of investigated Fen Complex dwellings.¹⁴ The effective dose from indoor gamma radiation has been found in the range of 0.2–3 mSv year⁻¹. If all available data for the Fen Complex were roughly considered together, total exposure dose could exceed 10 mSv year⁻¹ for certain limited parts of the Fen Complex population. Similar results have previously been published.¹³

However, some uncertainties must be highlighted:

- An exposure period of 20% of the year (1752 hours) might not be the most realistic exposure scenario since the majority of measuring points were in a wooded area. Instead, an exposure period of 350 hours per year would give approximately 5 times lower outdoor doses, in the range of 0.6–1.5 mSv.

- The assumption of uniform distribution of ^{220}Rn was probably not justified because of its very short half-life. Spatial changes^{2,43} which were not taken into account in this paper, could lead to actually much different (both lower and higher) ^{220}Rn doses, especially under different atmospheric conditions.

- Numerous studies worldwide^{3,8,44–48} have published the ^{220}Rn equilibrium factor values in the range of 0.003–0.1.

With respect to the differences in the equilibrium factor for ^{220}Rn , the additional dose estimation was done applying a significantly higher equilibrium factor of 0.1. Total annual effective doses were then found to be in the range of 8.0–14.3 mSv.

The contribution of ^{220}Rn to total dose increased in this case to 74%, ten times higher than the values we derived in the first calculation (with equilibrium factor 0.003). Stranden¹³ reported inhaled ^{220}Rn progenies as the main contributors to the effective dose at mining sites in the Fen Complex area. Considering that and our recalculations, the estimated total effective doses for Fen population could easily exceed 17 mSv year⁻¹.

Based on all given facts, an accurate estimation of outdoor radiation dose should comprise the direct measurement of ^{220}Rn decay products and different exposure scenarios reflecting the realistic behaviour of people living in the area.

UNSCEAR²⁶ provided a list of worldwide ‘enhanced natural radiation areas’ (ENRA), although the specific criteria to characterize an area as ENRA are still needed. Review of the current list showed that these areas mainly have high absorbed gamma dose rates in the air (>300 nGy h⁻¹), enhanced soil radionuclides activity concentrations and ^{222}Rn concentrations in the air (indoor ^{222}Rn > 150 Bq m⁻³). According to results of the present study, the Fen Complex in Norway should be considered as an ENRA. These locations are of interest to illustrate chronic

human exposure to elevated natural radiation levels, as well as in studies of possible effects due to low dose exposure.²⁶

4. Conclusion

The survey of outdoor radiation levels in a ^{232}Th rich Norwegian area was done in the present study. In addition, the seasonal variation and exposure doses due to outdoor radiation were evaluated. Significantly high air concentrations of ^{220}Rn and high terrestrial gamma dose rates in the air were observed for all investigated sites. These high values were attributed to the primordial radiation in the bedrock of the area. Based on long-term surveys, the air concentrations of thoron (^{220}Rn) and radon (^{222}Rn) reached 1786 and 82 Bq m⁻³, respectively. Regression analysis results suggested highly relevant prevalence of ^{220}Rn in the air. It corresponded to the significant soil enrichment with ^{232}Th , found by gamma spectrometric analysis. Statistically significant seasonal variation was obtained regarding ^{220}Rn , ^{222}Rn and gamma dose rates in the air. The effective radiation doses from outdoor exposure (5 h day⁻¹) were estimated to be in the range of 3.0–7.7 mSv year⁻¹. More conservative exposure of spending one hour daily at these locations would give lower doses in the range 0.6–1.5 mSv. However, calculations using a higher equilibrium factor for ^{220}Rn ($F = 0.1$) would give doses up to 14.3 mSv, although with high associated uncertainty.

In general, outdoor doses exceeded the value of the average dose constraint of 1 mSv for public exposure (both outdoor and indoor).⁴² Based on the results presented in this study, the Fen Complex area should be considered as an ENRA.

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1. Mrdakovic Popic, J., Salbu B. and Skipperud, L. (2011), Ecological transfer of radionuclides and heavy metals at NORM and TENORM sites in Norway. International Conference on Radioecology and Environmental Radioactivity, Hamilton, Canada, 19–24 June 2011.
2. Mrdakovic Popic, J. (2011). Environmental risk assessment at NORM and TENORM sites in Norway. International Workshop on Demonstrating Compliance with Environmental Protection Objectives for Radioactive Waste Disposal Facilities, Herrankukkaro, Finland, 30-31 March 2011.
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