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TIINA TUOVINEN

TRANSFER OF ELEMENTS RELATED TO THE NUCLEAR FUEL CYCLE – EVALUATION OF LINEARITY IN BOREAL ECOSYSTEMS

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Transfer of elements related to the nuclear fuel cycle – Evaluation of linearity in boreal ecosystems

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ABSTRACT

In radioecological models, the transfer of radionuclides into organisms is commonly described by the concentration ratio (CR), which assumes that the uptake into an organism is linear with respect to the concentration of the element in the medium. This study focused on evaluating this assumption of linearity by examining samples collected from the natural world and experimental meso- and microcosms.

The transfer of essential (Mo, Ni, Zn) and non-essential elements (U, Pb) from soil into three understory species and two tree species was studied at two forest sites. The radionuclides of these elements are potentially important in the risk assessment of radioactive waste disposal. An evaluation of the CRs, as a function of the soil element concentration, revealed that CR was not constant but decreased with increasing soil concentrations, and thus transfer was described better with a non-linear equation than with the linear model.

The transfer of ^{137}Cs from water to fish was studied in two lakes. The water-to-fish CR decreased with increasing ^{137}Cs concentration in water, and the data fitted well with a non-linear equation, similar to that used for describing transfer of elements from soil to plants. There were other findings of potential importance for radioecological modeling; the ^{137}Cs concentration was threefold higher in piscivores than in non-piscivores, but no differences were detected between three non-piscivorous species with different feeding habits.

The transfer of U, Co, Mo, Ni, Pb, Th and Zn into three plant species was studied also in experimental mesocosms. Non-linear transfer was observed in all of the studied elements and plant species, confirming the findings based on samples collected from a natural forest. The mesocosm experiments, and additional microcosm experiments, were also designed to study the transfer of soil elements into snails and earthworms. The data indicated that also transfer from soil or food to animals is non-linear for many of the elements studied. The uptake of U was nearly linear, indicating that different modeling approaches

may be needed for individual elements. However, the results concerning the uptake of Pb did not support the previously proposed simplified hypothesis that transfer would be non-linear only for essential elements.

Finally, the impact of non-linear transfer on radioecological modelling was studied by comparing the linear model, a non-linear model derived from the observations of this study, and a novel model based on the observation that non-linear transfer leads to a practically constant total element concentration in plant tissues. Three models were used to predict transfer of ^{234}U, ^{59}Ni and ^{210}Pb into spruce needles. The predictions of the non-linear model and the novel model were essentially the same, but the linear model underestimated the uptake of radionuclides when the total element concentration in soil was low as typically is the case in a boreal forest. Linear modeling could advantageously be replaced by the new modeling approach since this more realistically reflects the actual processes involved in the uptake of these elements into plants. The proposed new modeling approach could potentially reduce the uncertainty in model predictions. It is also simple to perform and the data needed to estimate the model parameters are readily available in the published literature.

Universal Decimal Classification: 502.21, 504.5, 546.7, 574.4, 574.5, 621.039, 628.4.047

CAB Thesaurus: nuclear energy; radioactive wastes; waste disposal; radionuclides; metallic elements; uranium; lead; molybdenum; nickel; zinc; thorium; caesium; pollution; concentration; ecosystems; models; bioaccumulation; boreal forests; food chains; trophic levels; soil; plants; herbivory; snails; earthworms; fresh water; freshwater lakes; fish

TIIVISTELMÄ

Radioekologisissa malleissa radionuklidien siirtymistä eläviin organismiehin kuvataan yleisesti siirtokertoimella, jonka käyttöön liittyy oletus maaperän tai veden ja eliöiden pitoisuuksien välisestä lineaarisesta riippuvuudesta. Tässä tutkimuksessa arvioidaan edellä mainittua lineaarisuusoletusta luonnosta ja kokeellisista meso- ja mikrokosmoksista kerättyjen aineistojen avulla.

Kasveille välttämättömien (Mo, Ni, Zn) ja ei-välttämättömien (U, Pb) alkuaineiden siirtymistä maaperästä kolmeen kenttäkerroksen kasvilajiin ja kahteen puulajiin tutkittiin kahdessa eri metsässä. Näiden alkuaineiden radionuklidit ovat potentiaalisesti tärkeitä ydinjätteen loppusijoituksen riskin arvioinnissa. Kun siirtokertoimia tarkasteltiin maaperän alkuainepitoisuuden funktiona, havaittiin, ettei siirtokerroin ollut vakio, vaan se pieneni maaperän alkuainepitoisuuden kasvaessa, ja siksi epälineaarinen yhtälö kuvasi siirtymistä paremmin kuin lineaarinen malli.

Cesium-137 siirtymistä vedestä kalaan tutkittiin kahdessa järvessä. Siirtokerroin vedestä kalaan pieneni veden ^{137}Cs pitoisuuden kasvaessa ja metsäekosysteemissä käytetty epälineaarinen yhtälö kuvasi siirtymistä hyvin myös vesiekosysteemissä. Radioekologisen mallintamisen kannalta tehtiin kaksi muutakin tärkeää havaintoa. Cesium-137 pitoisuudet olivat kolminkertaiset petokaloissa saaliskaloihin verrattuna, mutta saaliskalojen välillä ei havaittua eroa ^{137}Cs pitoisuuksissa, vaikka saaliskalalajien ravinnonkäyttötavat erosivat toisistaan.

Uraanin, Co, Mo, Ni, Pb, Th ja Zn siirtymistä kolmeen kasvilajiin tutkittiin myös kokeellisessa mesokosmoksessa. Kaikkien tutkittujen alkuaineiden siirtyminen oli epälineaarista, mikä vahvisti luonnosta saatuja tuloksia siirtymisen epälineaarisuudesta. Meso- ja mikrokosmoskokeissa tutkittiin myös alkuaineiden siirtymistä lieroihin ja kotiloihin. Tulokset osoittivat, että siirtyminen maasta tai kasvista myös eläimiin oli epälineaarista usean alkuaineen osalta. Uraanin siirtyminen oli

lähes lineaarista ja siksi erilaiset siirtymismallit saattavat olla tarpeellisia eri alkuaineille. Lyijyn siirtyminen ei kuitenkaan tukenut aiempaa yksinkertaista oletusta siitä, että siirtyminen olisi epälineaarista vain välttämättömille alkuaineille.

Lopuksi tutkittiin, miten epälineaarisen siirtymisen huomioiminen vaikuttaa radioekologiseen mallintamiseen vertaamalla lineaarista mallia, epälineaarista mallia, joka kehitettiin tämän tutkimuksen tuloksena, ja uutta mallia, joka perustui havaintoihin siitä, että epälineaarisen siirtymisen johdosta alkuainepitoisuus kasvissa on käytännössä vakio. Näillä kolmella mallilla ennustettiin ^{234}U, ^{59}Ni ja ^{210}Pb siirtymistä kuusen neulasiin. Epälineaarisen mallin ja uuden mallin ennusteet olivat samanlaiset, mutta lineaarinen malli aliarvioi radionuklidien kasviin siirtymistä matalilla maaperän alkuainepitoisuuksilla, jotka ovat tyypillisiä pohjoisen havumetsävyöhykkeen maaperässä. Lineaarisen mallin vaihtaminen uuteen malliin, joka todenmukaisemmin kuvaa tutkittujen alkuaineiden siirtymistä kasviin, voisi tarkentaa mallinnusta. Uusi malli on yksinkertainen ja mallin parametreina tarvittavia tietoja on julkaistu laajasti.

Yleinen suomalainen asiasanasto: ydinenergia; radioaktiiviset aineet; radioaktiiviset jätteet; ydinjätteet; loppusijoitus; ympäristövaikutukset; saastuminen; kulkeutuminen; kertyminen; pitoisuus; alkuaineet; uraani; cesium; koboltti; molybdeeni; nikkeli; sinkki; torium; mallintaminen; ekosysteemit; boreaalinen vyöhyke; ravintoketjut; maaperä; järvet; kasvit; kasvinsyöjät; kalat

“One day I got a horse
and it makes me feel
that everything is possible.
Galloping still makes me feel
that everything is possible,
but the horses in my life
have taught me
that it takes time.”

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Iisalmi, November 2016
Tiina Tuovinen

LIST OF ABBREVIATIONS

AMAP	Arctic Monitoring and Assessment Programme
Bq	Becquerel
Bq g^{-1}	Becquerel per gram
Bq kg^{-1}	Becquerel per kilogram
Bq L^{-1}	Becquerel per litre
Bq mg^{-1}	Becquerel per milligram
CEC	Cation exchange capacity
Ci	Curie
CR	Concentration ratio
GM	Geometric mean
GMS	Geometric standard deviation
HNO_3	Nitric acid
IAEA	International Atomic Energy Agency
ICP-MS	Inductively coupled plasma-mass spectroscopy
ICRP	International Commission on Radiological Protection
K_d	Solid-liquid distribution coefficient
NH_4Ac	Ammonium acetate
NORM	Normally occurring radioactive material
OM	Organic matter
pH	Potential of Hydrogen
SKB	Svensk Kärnbränslehantering Aktiebolag (Swedish Nuclear Fuel and Waste Management Company)
STUK	Säteilyturvakeskus (Radiation and Nuclear Safety Authority)
UF_6	Uranium hexafluoride
UO_2	Uranium dioxide
U_3O_8	Triuranium octoxide

LIST OF ORIGINAL PUBLICATIONS

This thesis is based on data presented in the following articles, referred to in the text by their chapter numbers.

Chapter 2 Tuovinen TS, Roivainen P, Makkonen S, Kolehmainen M, Holopainen T, Juutilainen J. Soil-to-plant transfer of elements is not linear: Results for five elements relevant to radioactive waste in five boreal forest species. *Science of the Total Environment 410-411: 191-197, 2011.*

Chapter 3 Tuovinen TS, Saengkul C, Ylipieti J, Solatie D, Juutilainen J. Transfer of ^{137}Cs from water to fish is not linear in two northern lakes. *Hydrobiologia 700:131-139, 2013.*

Chapter 4 Tuovinen TS, Kasurinen AK, Häikiö E, Tervahauta A, Makkonen S, Holopainen T, Juutilainen J. Transfer of elements relevant to nuclear fuel cycle from soil to boreal plants and animals in experimental meso- and microcosms. *Science of the Total Environment 539:252-261, 2016.*

Chapter 5 Tuovinen TS, Kolehmainen M, Roivainen P, Kumlin T, Makkonen S, Holopainen T, Juutilainen J. Non-linear transfer of elements from soil to plants: impact on radioecological modeling. *Radiation and Environmental Biophysics 55:393-400, 2016.*

AUTHOR'S CONTRIBUTION

Chapter 2 Tiina Tuovinen contributed to the data analysis and writing. Päivi Roivainen contributed to the design of the study, field and laboratory work, data analysis and writing the manuscript. Sari Makkonen and Toini Holopainen contributed to the design of the study and the writing. Mikko Kolehmainen contributed to the writing. Jukka Juutilainen contributed to the design of the study, data analysis and writing.

Chapter 3 Tiina Tuovinen contributed to the design of the study, data analysis and writing the manuscript. Chutarat Saengkul contributed to the data analysis and writing. Jarkko Ylipieti and Dina Solatie contributed to the field and laboratory work and writing. Jukka Juutilainen contributed to the design of the study, data analysis and writing.

Chapter 4 Tiina Tuovinen contributed to the design of the study, field, garden and laboratory work, data analysis and writing of the manuscript. Anne Kasurinen and Elina Häikiö contributed to the design of the experimental systems, garden work and writing. Arja Tervahauta contributed to establishing the experimental systems and to the writing. Sari Makkonen and Toini Holopainen contributed to the design of the study and writing. Jukka Juutilainen contributed to the design of the study, data analysis and writing.

Chapter 5 Tiina Tuovinen contributed to the design of the study, data analysis, modeling and writing the paper. Mikko Kolehmainen contributed to the

modelling and writing. Päivi Roivainen and Timo Kumlin contributed to the data analysis and writing. Sari Makkonen and Toini Holopainen contributed to the writing. Jukka Juutilainen contributed to the design of the study and writing.

Contents

1 General introduction

1.1 BACKGROUND

Radioecology is the discipline that links the sources of radionuclides to their impacts and risks of exposure in ecosystems. This radioecological study focused on the transfer of radionuclides from soil and water to biota. This form of transfer is an important step for evaluating how these agents move further up the food web. Traditionally, radioecological modeling has been performed to assess possible adverse effects of radiation on humans, and non-human biota has been assumed to be unaffected as long as humans are protected (ICRP 1977, 1991). More recently, the focus of radioecology has expanded to cover the protection of non-human biota (IAEA 2002a, ICRP 2007). As previous data was limited to organisms important in food chains terminating in humans, this means that an extensive amount of new information is needed for assessing the exposure of other key species in different ecosystems.

In radioecology, an exposure pathway describes the transfer of radionuclides in a foodweb. Radioecological knowledge has expanded due to nuclear incidents. After the Chernobyl nuclear power plant accident, the grass – cow – milk – human pathway was the main exposure pathway, as the radionuclides were transported in the atmosphere and deposited on the Earth's surface during the pasturing season (AMAP 1998). Another vulnerable pathway, especially related to boreal forests, was the lichen – reindeer – human pathway (AMAP 1998), which had been studied already after atmospheric contamination resulting from nuclear weapons tests. In addition to terrestrial sources, also marine and freshwater systems were contaminated by radionuclides and increased levels of radioactivity were

detected in fish species in Scandinavian countries as a consequence of the fallout from Chernobyl (AMAP, 1998).

The context of the present study is to assess possible risks of the normal use of nuclear power. In contrast to the atmospheric deposition resulting from nuclear weapons tests and major nuclear accidents, the normal nuclear fuel cycle can lead to soil contamination by radionuclides from below-ground sources such as final disposal or other radioactive waste repositories. In this context, uptake by plants is the key process for the entry of radionuclides into the food web.

The boreal vegetation zone, covering 15% of our planet's land surface, is of particular interest for assessing radioecological risks, especially in Finland. The boreal forest is a mixture of coniferous and deciduous tree species growing at northern latitudes, where there is a large seasonal variation between summer and winter (Bonan and Shugart 1989). Because of the short summers, cold winters, snow cover, and higher precipitation compared to evapotranspiration, soil in boreal forest is generally moist and cold, which decreases the decomposition of organic matter and slows down nutrient cycling (Bonan and Shugart 1989). Boreal soil is usually layered (Lundström et al. 2000a,b) and its organic matter consists mainly of pine needles which excrete acids, creating "acidic" conditions in the soil (Bonan and Shugart 1989). This layered and acidic soil type is podzol and it is common in boreal forest (Lundström et al. 2000a,b).

Low temperature, seasonality in light intensity and ice cover are environmental characteristics, which increase the vulnerability of an ecosystem to the effects of radiation (AMAP, 1998), by affecting, for example, mobility and persistence of radionuclides in the environment. The radiosensitivity of coniferous forests and boreal plant species was among the highest observed in a Northern American study (Woodwell and Sparrow 1965). As soil conditions, frequency of lakes, seasonality and species, in particular the possible elevated radiosensitivity of boreal species, differ from those of other

vegetation zones, it is important to examine the transfer of radionuclides in boreal terrestrial and aquatic ecosystems.

1.2 ENVIRONMENTAL RADIOACTIVITY

Natural background radiation includes cosmic radiation from sun and space and primordial radiation from the Earth's crust. Cosmic radiation is constantly regenerating some relatively short living radionuclides (^{14}C, ^{3}H) in the air, which are subsequently deposited on soil, water bodies and biota (AMAP 1998). Primordial radionuclides have long half-lives comparable to the age of Earth (4.6×10^{9} years) and they are believed to have existed long before the Earth was formed (AMAP 1998). The decay chains of U, Th and ^{235}U (actinium) have a primordial origin. Under natural conditions, primordial radionuclides are slowly weathered from the Earth's crust into the soil and the concentration of these radionuclides varies with local geology (AMAP 1998). Naturally occurring radioactive material (NORM) consists of primordial radionuclides and their daughters. Continuous anthropogenic activities, like mining, fertilizer manufacture, gas and oil production and coal combustion, typically increase the impact of NORM by increasing the mobility and availability of NORM (AMAP 1998). For example, the mining of phosphate for fertilizers elevates the amounts of naturally occurring ^{238}U, ^{232}Th and ^{226}Ra in the environment (Sahu et al. 2014).

Environmental radioactivity outside the NORM definition includes artificially produced radionuclides from nuclear weapons tests and nuclear plant accidents. Nuclear weapons tests, which occurred mostly in the 1950s, are responsible for the largest radioactive contamination on Earth and long-lived radionuclides from these tests can still be detected all around the world (AMAP 1998). Considerable radioactive contamination was caused also by the two recent nuclear power plant accidents, Chernobyl in 1986 and Fukushima in 2011. In Chernobyl, radionuclides were uncontrollably released into the

atmosphere for one whole week; they were transported widely by winds and deposited on the Earth's surface, exposing mainly boreal plant and animal species and European people (AMAP 1998). The explosion zone around the Chernobyl nuclear power plant was very radioactive and the effects on the local ecosystem have been monitored since the accident. In Fukushima, radioactive contamination involved minor and short-term air releases that became deposited on Japanese territory and in the Pacific Ocean (Morino et al. 2011), whereas the majority of the emissions were controlled releases into the sea, where the contamination spread with the ocean currents (Buesseler 2014). After the accident, transfer of radionuclides into non-human organisms has been observed, but transfer into humans was minor compared to the situation in Chernobyl. If one considers the radionuclides from nuclear weapons tests and nuclear plant accidents, then ^{137}Cs, ^{129}I, ^{90}Sr as well as isotopes of Pu and radioisotopes of Am are the radionuclides that cause the greatest long-term radiological concern (AMAP 1998, Livingstone and Povinec 2000, Buesseler 2014).

In the environment, a chemical element can exist as several stable and radioisotopes, differing in their mass number (Choppin et al. 2002). Both natural and man-made radionuclides have excess energy in their nucleus and, due to this energy, they are able to spontaneously undergo radioactive decay towards more stable forms until they are no longer radioactive. The decay of a radionuclide results in the emission of particles, such as alpha or beta particles, or gamma rays. An alpha particle consists of two neutrons and two protons. Natural heavy elements with a high atomic number, for example U and Ra, are typically alpha emitters. In beta decay, a neutron is converted into a proton, and an electron (beta particle) is emitted. This is typical for many kinds of radionuclides. Alpha and beta particle emissions lead to changes in the atomic or mass number of the original isotope and naturally existing decay chains of U, Th and ^{235}U are formed as mother radionuclides transmute into daughter radionuclides by alpha and beta decays. The decay chain of uranium includes 13 alpha and beta emitting

radionuclides between ^{238}U down to the stable element, lead. Gamma rays are photons (energy involving no particles); they are emitted when a nucleus in an excited state decays into a lower energy state. This kind of gamma decay often occurs after other forms of decay. (Choppin et al. 2002)

Alpha, beta and gamma emissions are ionizing radiation, which has enough energy to move electrons from other atoms. Alpha particles cannot penetrate into plant tissue or skin, and therefore they are harmful only as internal exposure i.e. if inhaled or they otherwise gain internal access to the organism (Harten 1998). In contrast to alpha particles, small beta particles and gamma rays are penetrating and these can be harmful also outside the organism i.e. as external exposure (Harten 1998).

The unit of radioactivity described in the International System of Units (SI) is the Becquerel (Bq), which is defined as radioactive decays per second. An alternative way to describe activity is the Curie ($1\ Ci = 3.7 \times 10^{10}\ Bq$). In environmental measurements, the activity concentration of media ($Bq\ L^{-1}$, $Bq\ kg^{-1}$) is often used. The specific activity is the activity per quantity of atoms of a specific radionuclide and it is a property of that radionuclide.

Radioactivity decreases via its decay. The physical half-life of a radionuclide is the time in which its activity has decreased to half of the original value. Radioecologically relevant radionuclides are usually relatively long-lived and because of their persistence in the environment, they display a wide dispersion and transfer in food webs and therefore there can be extensive exposure of organisms and ecosystems. Biological half-life is the time in which the radioactivity in an organism has declined to half of its original value. Biological half-life is typically shorter than the physical half-life, because radionuclides are excreted out of the organism (AMAP 1998). The biological half-life is relevant if an organism is transported to a clean environment after contamination, and fed with uncontaminated food and water. However, if an animal or plant stays in the contaminated environment, the decreasing activity concentration in the organism is more properly described by the

ecological half-life, which includes environmental processes influencing the transfer of the radionuclide (IAEA 2010). Root uptake (IAEA 2010) and leaching of radionuclides from the rooting zone (Pröhl et al. 2006) are important processes affecting the ecological half-life. Because of continuing intake of radionuclides, the ecological half-life is longer than the biological half-life (IAEA 2010).

1.3 NUCLEAR FUEL CYCLE

According to the World Nuclear Association, a total of 14 % of the world's energy needs are produced by nuclear reactors. The nuclear fuel cycle includes mining, milling, conversion and enrichment of uranium, nuclear fuel fabrication, power generation in a reactor, spent nuclear fuel storage, and the final disposal of spent nuclear fuel. A nuclear fuel cycle can be open or closed; the cycle is referred to as being closed if spent nuclear fuel is reprocessed and recycled (IAEA 2011).

Uranium is a common metal in the Earth's crust and it can be mined in open or underground mines or it can leach *in situ* from the ore (IAEA 2011). The largest producers of uranium are Kazakhstan, Canada and Australia (IAEA 2011).

During milling, uranium is chemically extracted from the mined uranium ore, resulting in the formation of uranium oxide (U_3O_8; "yellowcake") and waste rock (IAEA 2011). In the yellowcake, uranium is concentrated to as much as 80 %, while uranium ore typically contains 0.03 - 20 % of uranium (IAEA 2011). Solid yellowcake is transported from a mill to further processing units, whereas waste rock is dumped near the mine. Not only is waste rock radioactive it may also include toxic materials such as heavy metals, and therefore mine dumps need to be isolated from the surrounding environment in order to avoid releases (IAEA 2002b).

The next step in nuclear fuel cycle is conversion, in which solid uranium oxide is converted into uranium hexafluoride (UF_6) which can be used in enrichment plants (IAEA 2011). In an

enrichment plant, the isotope composition of the uranium is changed to make it suitable for installation in nuclear reactors (IAEA 2011). Since the majority of natural uranium is non-fissile ^{238}U and only 0.7 % is fissile ^{235}U, isotope separation is needed to increase the level of ^{235}U to about 4 % (IAEA 2002b).

In fuel fabrication, ceramic pellets of nuclear fuel are formed from pressed uranium oxide (UO_2) which is baked at a very high temperature (1400°C) so that the pellets have a high density and good stability (IAEA 2011). Then, the nuclear fuel pellets are encased in metal tubes (IAEA 2011). The metal tubes (fuel rods) are arranged in fuel assemblies which are ready for use in a nuclear reactor (IAEA 2011). Several hundred fuel assemblies in the reactor form the nuclear core, where a controlled fission of ^{235}U occurs (IAEA 2002b). Some of the ^{238}U in the reactor core is converted into plutonium, part of which also experiences fission (IAEA 2002b). The heat produced in fission is converted into electricity through the heating of water to generate steam to turn the turbines (IAEA 2011). Approximately one third of the reactor's nuclear fuel is replaced with new fuel every year to ensure that fission continues at the optimum level (IAEA 2011).

Spent nuclear fuel removed from the reactor is very hot and radioactive and therefore fuel assemblies are stored near the reactor in a water pool which cools down the fuel and shields the surroundings from radiation (IAEA 2011). Spent nuclear fuel consists mainly of uranium (96%), but it also contains small amounts of plutonium and other high level waste products (IAEA 2002b).

After several years, spent nuclear fuel can be moved from the nuclear plant to an interim wet or dry storage site or it can be recycled back to nuclear fuel by separating uranium and plutonium from the waste products (IAEA 2011). During the interim storage of 40 years, the radioactivity of spent nuclear fuel decreases to one thousandth of its initial levels, which permits the final disposal of spent nuclear fuel (IAEA 2011).

Liquid high-level waste is prepared for final disposal by heating it to produce a dry powder which is immobilized in a ceramic form (2002b). The spent nuclear fuel is categorized as

high-level waste due to its high amount of radioactivity (IAEA 2011). At the moment, there are no functioning final disposal facilities (IAEA 2011), but a deep geological repository is being constructed in Finland (see 1.3.1), and studies into final disposal practices and locations are being widely conducted.

1.3.1 Nuclear facilities in Finland

In Finland, about 30 percent of the country's electricity is produced by four working nuclear power plants (two in Loviisa and two in Olkiluoto). One new reactor is under construction in Olkiluoto, and one more is being planned. It is stated in the Finnish Nuclear Energy Act (990/1987) that all nuclear waste produced in Finland must also be disposed of in Finland. To make this possible, a deep geological repository, called "Onkalo", is being built in Olkiluoto in granite bedrock at a depth of 500 meters. Onkalo has been being constructed since 2004 and it is expected to be ready in 2020. In the 2020s, spent nuclear fuel from the first reactors of Loviisa and Olkiluoto will have been cooling down for 40 years and the final disposal can be started in Onkalo. It is estimated that Onkalo has the capacity to store all of Finland's nuclear waste of the next hundred years and therefore the final encapsulation and burial will take place and the access tunnel to Onkalo will be backfilled and sealed in 2120.

Nuclear waste burial in Onkalo is based on the KBS-3 method developed by the Swedish nuclear fuel and waste management Co (Posiva 2007-10). In this method, ceramic state spent nuclear fuel is arranged in assemblies that are placed into a boron steel canister. The canister is enclosed in a copper capsule, and the copper capsule is placed in its own cave in the bedrock and then buried in bentonite clay. This KBS-3 method follows the multiple barrier principle; the ceramic form of spent nuclear fuel, the copper capsule, bentonite clay and bedrock are the barriers that prevent release of radionuclides from its repository into the environment. (Posiva 2007-10)

As Onkalo will be one of the world's first long-term underground repository, it has also been used for research

purposes during its construction. The company responsible for the construction of Onkalo and final disposal of spent nuclear fuel in Finland (Posiva Oy) has published many research reports, taking into account the local environmental conditions of the Olkiluoto Island (Posiva 2012-11).

1.3.2 Elements related to nuclear fuel cycle

Uranium occurs naturally as a mixture of three isotopes ^{238}U, ^{235}U and ^{234}U, of which ^{238}U is the most abundant accounting for 99,275 % of natural uranium. All isotopes of U are radioactive and the World Nuclear Association has reported that radioactivity in uranium ore is mainly from ^{238}U decay series, which includes among others ^{234}Th, ^{234}U, ^{230}Th, ^{226}Ra, ^{222}Rn, ^{210}Pb, ^{210}Bi and ^{210}Po. After U is processed and removed from the ore and short-lived ^{234}Th and ^{234}Pa have disappeared, then ^{230}Th, ^{226}Ra and ^{222}Rn are the major contributors to radioactivity in the mine.

Fission in a nuclear reactor core produces large quantities of fission and activation products. Fission products include isotopes with atomic masses between 70 and 170 such as ^{89}Sr, ^{90}Sr, ^{91}Y, ^{95}Zr, ^{95}Nb, ^{99}Mo, ^{103}Ru, ^{131}I, ^{133}Xe, ^{137}Cs, ^{140}Ba, ^{140}La, ^{141}Ce, ^{144}Ce, ^{143}Pr and ^{147}Nd, of which the radioecologically most important in the short term are ^{89}Sr, ^{90}Sr, ^{131}I and ^{137}Cs and in the long term, ^{90}Sr and ^{137}Cs, when possible doses to humans in arctic areas are considered (AMAP 1998). The activation products formed in nuclear reactors include ^{51}Cr, ^{54}Mn, ^{55}Fe, ^{59}Fe, ^{60}Co, ^{63}Ni, ^{65}Ni, ^{64}Cu, ^{65}Zn, ^{69}Zn, ^{110}Ag, ^{109}Cd, ^{134}Cs, ^{236}U and ^{239}U. Transuranic nuclides which have an atomic number higher than U, such as ^{239}Np and ^{239}Pu, are short-lived activation products of ^{239}U (AMAP 1998). Spent nuclear fuel contains large amounts of fission and activation products (AMAP 1998).

Peak soil concentrations from the interim storage of intermediate and low radioactive waste are related to radionuclides of ^{14}C, ^{36}Cl, ^{59}Ni, ^{79}Se, ^{99}Tc, ^{129}I, ^{135}Cs, ^{210}Po, ^{210}Pb, ^{226}Ra, ^{227}Ac, ^{230}Th, ^{231}Pa, ^{234}U, ^{235}U, ^{238}U, and ^{239}Np, of which ^{226}Ra, ^{210}Po, ^{234}U, ^{230}Th, and ^{238}U have the highest potential for impact on human beings (Jones et al. 2003). In addition, ^{3}H, ^{90}Sr and

^{239}Pu are also listed, but soil concentration values for these radionuclides are not given (Jones et al. 2003).

According to Posiva, there are 11 radionuclides that are the most important in the long-term safety assessment of the final disposal of spent nuclear fuel. The top priority radionuclides are ^{14}C, ^{36}Cl and ^{129}I, and high priority radionuclides are ^{93}Mo, ^{94}Nb, ^{135}Cs, ^{59}Ni, ^{79}Se, ^{90}Sr, ^{107}Pd, ^{126}Sn (Hjerpe et al. 2010). Due to updated information related to spent nuclear fuel canisters, ^{90}Sr will most probably be removed from the high priority list, but ^{108}Ag will be incorporated (Posiva 2012-06). Top priority radionuclides or their progeny radionuclides are expected to dominate the dose in most biosphere prediction cases, especially the most realistic ones, whereas high priority radionuclides or their progeny radionuclides are expected to give a significant contribution to the doses in some biosphere prediction cases, or even dominate in one or more biosphere prediction cases (Hjerpe et al. 2010). These calculations exclude all radionuclides of the naturally occurring decay chains, because of the time frame used. Howeer, if a longer time interval after the possible failure and especially human-doses were considered, also ^{226}Ra and ^{231}Pa would be priority nuclides in the safety assessment of final disposal of spent nuclear fuel (Hjerpe et al. 2010).

1.4 UPTAKE OF ELEMENTS BY TERRESTRIAL PLANTS

Uptake of elements by plants is an important step for the transfer of elements into living organisms. Concerning possible failure of a repository of spent nuclear fuel, transfer from soil to plants is crucial. Therefore, this thesis is particularly focused on transfer of elements into terrestrial plants.

1.4.1. Soil concentration

The elements in soil are made up of stable and radioisotopes of the element (Kryshev and Sazykina 1986, Sazykina 1994) and it is from this mixture that radionuclides are taken up by plants and transferred further in the food web. If the stable and

radioisotopes of an element have a similar chemical form, the stable isotope is called the stable analogue of the radioisotope (Sazykina 2000). As organisms are unable to distinguish between these analogues, there is an equal transfer of stable and radioisotopes and therefore the proportion of a radioisotope taken up by organisms is assumed to be the same as the proportion of the radioisotope in the soil (Kryshev and Sazykina 1986, Sazykina 1994). Therefore, this chapter will focus on elements in soil and organisms.

The soil concentrations of some elements relevant to nuclear fuel cycle are given in Table 1. The median cesium concentration in European soils (FOREGS 2005) is within the range of podzol element concentrations combined over the world (Kabata-Pendias 2011), but concentrations of many other elements in Northern Europe (Reiman et al. 2001) are below the ranges given for podzols (Kabata-Pendias 2011). Land use together with the geographical region may explain the difference between the values given, as the Northern European study was restricted to natural forest soils, whereas other European and worldwide studies have concentrated on natural, agricultural and urban soils. A north-south difference in soil element concentrations was also reported by Reiman et al. (2012), who observed lower Co, Ni, Pb, Th and Zn concentrations in northern than southern Europe. Ukonmaanaho et al. (1998) studied heavy metal concentrations in soil water of Finnish boreal forest and concluded that the concentrations of Ni, Pb and Zn were lower in a Finnish boreal forest compared to those found in temperate ecosystems. This is an important finding as the soil element concentration can affect the effectiveness of element uptake by plants.

Table 1. *Median values or ranges of selected total element concentrations in different medias. Soil and plant element concentrations are given as dry weight basis but element concentration of water in wet weight basis. Values are collected from literature.*

Element	**Finnish soil mg kg^{-1}**	**Northern European soil [c] mg kg^{1}**	**European soil [d] mg kg^{-1}**	**World soil [e] mg kg^{-1}**	**European fresh water [d] µg l^{-1}**	**Finnish plants [f] mg kg^{-1}**	**Northern European plants [c] mg kg^{-1}**
U		0.04			0.32		<0.005
Co	0.26-10.4[a]	0.34	7.78	4-65	0.16	<0.05-1.9	0.171[c]
Cs			3.71	2.5-20	0.006		
Mo	0.04-31.7[a]	<0.1	0.62	0.4-6.4	0.22	<0.02-1.5	0.046
Ni	0.70-26.4[a]	0.76	18.0	11-25	1.91	0.05-58	1.97
Pb	4.33-123[a]	6.93	22.6	10-57	0.093	0.05-2.7	0.35
Th		<0.05	7.24		0.009		<0.02
Zn	2.7-74[b]	18.7	52.0	47-63	2.68	8.3-70	32

[a,b] Finnish soil: Ranges of topsoil element concentration by Roivainen et al. 2011a[a] and by Tuovinen et al. 2011[b]

[c] Northern European soil and plants: Median values of topsoil and plant element concentrations by Reiman et al. 2001

[d] European soil and fresh water: Median values of topsoil and fresh water element concentrations by FOREGS 2005

[e] World soil: Ranges of podzols' element concentrations by Kabata-Pendias 2011

[f] Finnish plants: Ranges of leaf/needle element concentrations by Roivainen et al. 2012

1.4.2. Root uptake

The main process influencing element concentration in plants is root uptake from soil and soil pore water (IAEA 2010). Root uptake of elements can be either passive diffusion-induced transfer of ions from the rooting zone, or active uptake that takes place against a chemical gradient and requires metabolic energy. Both processes can be involved in the uptake of an element (Kabata-Pendias 2011). According to Kabata–Pendias (2011), plants use active uptake preferably for Mo and Zn, whereas Pb and Ni gain access to cells by passive uptake.

Plants have an ability to control the uptake of elements by releasing root exudates and forming a symbiosis with soil microbes (Ehlken and Kirschner 2002). The composition of root exudates varies, but normally it consists of low molecular weight acids, although environmental stress can dramatically change the composition of the exudates (Kabata-Pendias 2004). Symbiosis with mycorrhizal fungi is common, as it enhances nutrition uptake (Ehlken and Kirchner 2002). Mycorrhizae also exude low molecular weight acids for mineral dissolution, which is likely to be a major weathering process in Podzols (Lundström et al. 2000a,b). Competitive and inhibitory interactions then control the concentration of trace ions in the soil solution (Ehlken and Kirchner 2002)

In addition to root uptake, vegetation can be contaminated from deposition onto plant surfaces, in which case, small airborne particles are deposited on plant surfaces and transferred through the leaf surface as foliar uptake into plants. Soil and dust particles can also adhere to plant surfaces from where elements can be transferred into plant tissues (IAEA 2010). The significance of foliar uptake and soil adhesion in boreal forest plant species, other than moss and lichen, is considered minor compared to root uptake (IAEA 2010).

The ratio of plant concentration to soil concentration is different for different elements. For example, Zn is efficiently taken up by plants and it accumulates inside plants, leading to higher Zn concentration in plants than in soil, whereas

concentrations of many other elements (e.g., U, Co, Mo, Ni, Pb, Th) in plants have been reported to be lower or similar to soil concentrations (Reimann et al. 2001, Roivainen et al. 2011ab, 2012), as shown in Table 1. However, differences have been observed between different plant species in their abilities to accumulate elements and especially in the accumulation of Zn (Reiman et al. 2001).

Within the plant, the Zn concentration was higher in leaves than in the roots, whereas U, Mo, Ni and Pb concentrations were higher in roots than in leaves (Roivainen et al., 2011ab). A likely explanation is that Zn is essential in leaves (see 1.4.3.) and is therefore actively transported there.

1.4.3. Essential and non-essential elements

Elements required by metabolism of an organism are called essential elements. For plants, C, H, O, N, K, Ca, Mg, P and S are essential macronutrients (needed in high amounts) and Cl, Fe, B, Mn, Zn, Cu, Mo, Ni and Co are essential micronutrients (needed in small amounts). All essential elements, excluding airborne O H and C from the air CO_2, are taken up from soil by plant roots (Kabata-Pendias 2011). If one considers the essential elements relevant to the nuclear fuel cycle, then Cl, Co, Cu, Fe, Mn, Mo, Ni, and Zn are listed.

Uptake of elements is dependent on plant nutrient uptake and selectivity (IAEA 2010). Although plants actively take up essential elements, they are not perfectly selective and also non-essential and toxic elements are taken up (Denny 2002). If a radionuclide is a chemical analogue of an essential nutrient, it can be taken up by plants instead of the essential element (IAEA 2010). A deficiency of an analogue essential element in the soil will increase the uptake of the radionuclide whereas an abundance of the analogue essential element will decrease the uptake of the radionuclide.

Potassium (K) is a well-known stable nutrient analogue for ^{137}Cs (Whicker and Schultz 1982). Cesium-137 is taken up and it behaves in plant metabolism like K, but unlike K, the ^{137}Cs concentration usually increases with trophic level, because it is

retained for a longer time in the organism. Lead is an analogue of Ca and can therefore be taken up by plants instead of Ca, and in animal tissues, Pb accumulates into bones similarly as Ca (Pereza et al., 1998).

1.4.4. Factors affecting plant uptake

The mobility of elements affects their uptake by plants, as it increases leaching of elements in soil and their bioavailability for root uptake. The uptake of an element is positively correlated with its available pool at the root surface (Kabata-Pendias 2011). The mobility of elements in soil is controlled by several abiotic processes including transport by flowing water, diffusion within the fluid and physicochemical interactions with the soil particles (IAEA 2010).

Radionuclides are present in the environment in various physicochemical forms (species) varying with respect to size, charge, valence, state of oxidation, structure, morphology and density, water solubility, possibility for exchange or being bound to oxides, carbonates or organic matter or incorporated into minerals (He et al. 2005). The distribution of the element among these different physio-chemical forms is called speciation (Salbu 2009) and this speciation affects the mobility of radionuclides (Salbu and Skipperud 2009). There are several mechanisms which increase the mobility of radionuclides i.e. desorption, dissolution and dispersion. In contrast, hydrolysis, complexation, polymerization, colloid formation and aggregation are processes that decrease the mobility of radionuclides in the environment by increasing the size of particle (Salbu 2009).

For many elements, mobility increases with decreasing soil pH. A low pH value is associated with enhanced chemical weathering, and many trace metals are therefore mobilized in acidic soils (Sohlenius et al. 2013). Soil fauna can also influence the mobility of elements. Mobility and availability of elements have been observed to be increased by non-plant species, especially earthworms (Sizmur and Hodson 2009), which are known to increase chemical weathering in soil. Upward

transport of soil by earthworms also makes it possible for the elements to be transported from the deeper soil to the rooting zone (Müller-Lemans and Van Drop 1996).

Some metals relevant to the nuclear fuel cycle, e.g. Zn, are very mobile and easily bioaccumulated by plants. Cobolt, Mo, and Ni are mobile and readily taken up by plants, whereas Pb is relatively strongly absorbed to soil particles and not readily transported into plants (Kabata-Pendias 2011).

Partitioning of elements between the soil solid phase and soil solution (liquid phase) affects their mobility and availability to plants. The solid-liquid distribution coefficient (K_d) describes the degree of element sorption on a solid phase and this value is calculated as a ratio of element concentration in the solid and liquid phases (IAEA 2010). A high K_d value indicates high sorption and therefore low mobility of an element, whereas a low K_d value is related to high mobility (IAEA2010). Elements with a low K_d value can be easily taken up by plants as well as being leached either to or away from the rooting zone (IAEA 2010).

The element-specific K_d is related to soil type, soil organic matter content, soil pH and the cation exchange capacity of the soil. Vandenhove et al. (2009a) reported that the K_d values for U, Pb and Th were related more to pH than to any other soil characteristics and a similar finding for U was observed by Echevarria et al (2001). International Atomic Energy Agency (IAEA 2010) has estimated K_d values for different soil conditions for modeling purposes. The values given for U vary from 180 to 1200 L kg^{-1} for different mineral and organic soils, and at different pH values e.g. the K_d was ten times higher for U in the pH range 5 - 7 than for pH below 5 or above 7.

Inorganic soils are classified according to the soil particle size. In coarse sand, the particle size is 0.2 – 2 mm, in fine sand 0.02 – 0.2 mm, in silt 0.002 – 0.02 mm and in clay < 0.002 mm (Mauseth 2003). In the soil solid phase, elements are bound around soil particles, and particularly cations are bound to clay particles (Mauseth 2003). As particle surface area (per soil volume) increases with decreasing particle size, there are more binding

sites and therefore less elements in the soluble phase in fine textured soils compared to coarse soils. This explains why the amount of silt and especially its clay content, affects the mobility and bioavailability of elements (Kabata-Pendias 2011).

Sheppard and Evenden (1988) and Vandenhove et al. (2009b) studied the influence of soil properties on the transfer of radionuclides (isotopes of U and Pb among others), and found that transfer was higher in coarse than in fine soils. Roivainen et al. (2012) also reported that the clay content influenced the transfer of U and Pb into native boreal plant species, but the effect was inhibitory only for U, whereas clay actually enhanced uptake of Pb. These opposite effects of clay content are difficult to explain (Roivainen et al. 2012). Kabata-Pendias (2011) suggested that the diversity of clay minerals may partly explain why the variations observed in the clay content affect uptake of elements.

Elements bound to soil particles need to be dissolved in soil solution before they can be taken up by roots and gain access to plant cells. The addition of cations can release the cations bound in the surface of clay particles, as the added cation takes another cation's place on the particle's surface making it available for root uptake (Mauseth 2003). The process described above is called cation exchange, and the cation exchange capacity (CEC) describes the degree to which a soil can absorb and exchange ions (Kabata-Pendias 2011). Although this value is closely related to the surface area of the soil particles, it varies for different clay minerals from 25 to 800 m^2 g^{-1} (Kabata-Pendias 2004).

Soil organic matter (OM) consists of dead plants, animals and other organisms that are in various stages of decomposition (Kabata-Pendias 2011). Organic matter has a high cation exchange capacity, from 560 to 800 m^2 g^{-1}, and therefore OM increases the CEC of soil (Kabata-Pendias 2011). The organic matter content increases in soil types as follows: sand < loam < clay < organic, and typical values for sandy soil is between 0.5 and 3.0 % and for organic soil over 20 % (IAEA 2010).

Different kinds of acids (organic, fulvic and humic) have important roles in the decay of OM and also in the biochemical weathering of inorganic compounds (Kabata-Pendias 2011). Organic matter provides a high sorption capacity for trace cations and thereby influences transport, leaching and accumulation of cations and supplementation of ions to root surface (Kabata-Pendias 2011).

Acidity or alkalinity of water soluble substances in soil is described by the pH value, which in podzols is ≤ 5.9 according to US Soil Taxonomy (Soil Survey Staff 1998 cited by Lundström et. 2000a). Podzol development is favored by coarsely textured (sandy or sandy till) base-poor medias and coniferous forest (Lundström et al. 2000a,b). The low pH of podzols is related to the presence of organic acids in the soil due to leaching from plants, decomposition of litter by microbes and exudation by roots, fungi and micro-organism (Lundström et al. 2000a,b).

Transfer of U is highly dependent on pH (Echevarria et al. 2001), and a low pH value has been related to high plant uptake of U, with the uptake being highest at pH 5 (Ebbs et al. 1998). According to Rose et al. (1979, cited by Kabata-Pendias 2011), mobility in acidic soil conditions is intermediate for Co, Cs, Mo, Ni and Zn and very low for Th.

1.5 TRANSFER OF ELEMENTS TO ANIMALS

The simple food chain soil – plant – herbivore – carnivore depicts the transfer of elements from primary producers to consumers (Kabata-Pendias 2011). The role of decomposers is critical, as these micro-organisms decompose dead flora and fauna and increase the bioavailability of soil elements, which enables re-entering of elements into the food web (Kabata-Pendias 2011).

The radionuclide concentration in an animal is highly dependent on feeding habits of the animal (IAEA 2010). As diets and therefore also ingested amounts of radionuclides vary between species, the transfer of radionuclides needs to be

described separately for species with different feeding habits (IAEA 2010). Heterogeneous radionuclide contamination in the environment and migration of animals can lead to differences in radionuclide concentrations even between individuals of the same species (IAEA 2010). In addition to diet, other biological factors including age, sex, body condition and reproductive status have been observed to affect the transfer of trace metals to polar bears, seals, and arctic foxes in an arctic ecosystem (Dehn et al. 2006).

In terrestrial ecosystems, transfer of radionuclides from plants to herbivores occurs mainly by ingestion of plant material, but also soil particles on the surface of plants are ingested (IAEA 2010). Concerning animals studied in this thesis, terrestrial snails are herbivorous and detritivorous organisms exposed to contaminated soils by both the digestive and cutaneous routes (Gomot et al. 1989). Snails are also regarded as ecologically key components of terrestrial food webs that may contribute to the transfer of contaminants from soil and plants to top predators (De Vaufleury et al. 2006).

Earthworms are another important group of soil animal species; they are commonly tested in bioaccumulation studies not only due to their high exposure potential to soil contaminants, but also because of their importance as food for a variety of wildlife species (DeForest et al. 2011). Although most trace metals are unlikely to be biomagnified (i.e. the concentration as they move up the food chain), dietary metal exposure can be an important pathway for wildlife, irrespective of their trophic level (DeForest et al. 2011). For animals living in soil, cutaneous uptake is an important route for the transfer of elements into the animal (IAEA 2010).

In aquatic ecosystems, radionuclide contamination has either become deposited from air or been released directly into water bodies, where radioactivity is absorbed by solid particles or dissolved in water (IAEA 2010). Solid particles are typically deposited at the bottom of the lake, but also dissolved contamination can be absorbed by the bottom sediment, and in both cases, radioactivity is no longer mobile in the water column

(IAEA 2010). Thus, radioactivity can be remobilized from the sediment, and become re-available to aquatic organisms (IAEA 2010).

Due to the reversibility of the mobilization and immobilization processes, radioactive contamination is persistent in water ecosystems, and partitioning in solid particles and water is the main process influencing the transport and uptake of radionuclides (IAEA 2010). The partition coefficient between water and sediment is described by the K_d value (IAEA 2010). In water ecosystems, water temperature affects the uptake of elements, as depuration of elements out of the organism is faster in warmer conditions (e.g. in shallow lakes compared to the colder waters of deep lakes) (Forseth et al. 1991).

In a lake ecosystem, soil or sediment is a contaminated media, plant and phytoplankton are primary producers, fishes are consumers or predators and sediment dwelling organisms are decomposers. There are many differences in the feeding habits of freshwater fish species. Benthic fishes eat sediment-dwelling organisms as well as soil particles, in which most of the contamination in water ecosystem has become absorbed (IAEA 2014). Forage fish species feed on primary producers, zooplankton and pelagic invertebrates, and piscivorous fish species consume other fish species (IAEA 2014). Due to differences in feeding habits, the uptake of radionuclides by benthic fish species, piscivorous fish species, and forage fish species should be considered separately (IAEA 2014).

1.6 RADIOECOLOGICAL MODELING

Radioecological models have many applications e.g. in licensing nuclear facilities, assessing radiological impacts of these facilities, handling existing nuclear emergencies and predicting the future impacts of radioactive waste repositories and possible nuclear emergency scenarios (IAEA 2009). Radioecological models made for the purpose of radiation protection are

conservative models preferring rather over- than underestimation of a transfer, effect or risk. These conservative models are used in demonstrating that existing facilities are operating in compliance with legislation whereas more realistic models are used in emergency situations (Kirchner and Steiner 2008).

Radioecological models are commonly box-models, where ecologically relevant parts of the environment are divided into separate boxes and transfer fluxes between these compartments are predicted with mathematical equations (IAEA 2010). In trophic transfer, the relevant model compartments could be contaminated media, primary producers, herbivores, predators and decomposers. Although air affects transfer, it can be excluded since it is not a major contributor. A model simplifies the complex and multilevel natural phenomena into understandable parameters. The number of parameters should be restricted because of uncertainties inherent in all of the parameters (Kirchner and Steiner 2008). Uncertainty caused by natural variation in the parameters is unavoidable and in fact is an important part of radioecological modeling, whereas uncertainty related to lack of data and poor model design can be reduced (Kirchner and Steiner 2008). It is very important in model design to identify the major contributors (those that significantly influence the process involved), and to model those precisely and correctly (Kirchner and Steiner 2008).

Radioecological modeling includes determining the characteristics of a contamination source, transport of that contaminant in the environment, its uptake by plants, followed by the trophic transfer of the agent in the food chain, dose calculations and risk assessment for exposed populations. In addition, the effects of possible counter-measures for minimizing the effects of radiation can also be modeled.

1.6.1 Dynamic models

Dynamic models are created to predict accumulation during changing conditions, as the conditions in the environment are seldom in a steady-state (AMAP 1998). A dynamic model

includes time-dependent functions that can take into account variations in environmental element concentrations over time (AMAP 1998). In particular, the dynamic model is beneficial, if biological accumulation is modeled immediately after a release or with ongoing emissions into the environment. For example, biological accumulation in a lake ecosystem is a dynamic model with respect to the uptake into organisms – the parameters to be modelled include ingestion rate, assimilation efficiency, and uptake rate from water column - and on elimination rate out of the organisms (Reinfelder et al. 1998). Extensive parameterization, lack of essential parameter values, and poor quality of existing parameter values are problems which may be encountered in dynamic modeling. Ecomod is an example of a dynamic radioecological model for aquatic ecosystems (Sazykina et al. 2000).

1.6.2. Equilibrium-based radioecological models

Equilibrium-based models simplify the transfer of an element into biota by describing it with one parameter called the concentration ratio (CR) or the transfer factor (TF). This parameter is the ratio of an element concentration in an organism to the concentration in the contaminated media. This approach assumes that equilibrium has been reached, and it is suitable for predicting transfer and long-term effects of radiation during relatively stable conditions. For example, possible failure of a spent nuclear fuel repository can be modeled using equilibrium-based models. However, these kinds of models are not suitable for emergency situations where there are rapid and dramatic changes in radionuclide mobility and availability (Howard et al. 2013).

Equilibrium-based models are simple to apply and are widely used, although the assumptions of equilibrium and linearity assumption are questionable, and furthermore the use of a single organism- and element-specific CR does not take into account the influence of environmental factors on the transfer of radionuclides (see sections 1.7 and 1.8). Examples of equilibrium-based radioecological models include the ERICA

tool (Brown et al. 2008), Ecolego (Avila et al. 2003) and CROM (IAEA 2001) for routine releases; and RESRAD-BIOTA (Beresford et al. 2008), MOIRA (Gallego et al. 2000) and Hot Spot (Homan and Aluzzi 2013) for accidental events and emergencies. The ERICA tool specializes in dose-effect relationships (FREDERICA database, Coppelstone et al. 2008), CROM in detailed transfer models, MOIRA in counter-measures whereas Hot Spot is concerned with how environmental contamination can impact on human health.

1.7 CONCENTRATION RATIO

The CR concept used in equilibrium-based radioecological models involves two assumptions. First, this approach assumes that there will be an equilibrium between concentrations in the organism and in the media when enough time has passed after the contamination. In fact, it may well be that no true equilibrium exists in the real world, since the equilibrium is disturbed by changes in several natural factors such as pH and mobility (Kabata-Pendias 2011). Rarely is any information provided on possible effects of non-equilibrium conditions on CRs. Nonetheless, when compared to the wide ranges and uncertainties inherent in CR values, uncertainties related to deviations from equilibrium may be less significant.

Second, the CR approach assumes linearity, i.e., that the element concentration in the organism is zero when its concentration in the medium (soil or water) is zero, and furthermore that the element concentration in the organism increases linearly with an increasing concentration in the medium (IAEA 2010). Under this assumption, a rising straight line originating from zero describes the concentration in the organism as a function of element concentration in the medium, and CR has a constant value regardless of element concentration in the medium (IAEA 2010). The problems of the linearity assumption will be discussed in section 1.8.

In addition to the unproven assumptions related to CR, a single species- and element-specific CR does not take into account the effects of environmental factors, although it is known that these kinds of factors can influence the transfer of elements, and different CR values have been measured in different environments (IAEA 2010, Kabata-Pendias 2011). To overcome this problem, efforts have been made to determine CRs for multiple environments under different conditions (IAEA 2010, 2014, Vandenhove et al. 2009b).

It is clearly impossible to define species-specific CRs for all living plant and animal species, and therefore the concept of the reference organism has been introduced (Beresford 2010, Howard et al. 2013, IAEA 2014). Reference organisms are considered to represent comprehensively ecosystem-relevant functional groups of different trophic levels and ecosystem types (Howard et al. 2013, IAEA 2014). Since the intra-species variation in CR values is wide and in many cases, inter-species differences are undetectable (Vandenhove et al. 2009b), the estimation of generic CRs for validated groups of species has been proposed.

The reference organism is a rather new concept in radioecological modeling, and the most extensive database for reference organisms among wildlife species is available in the Technical Report Series (TRS) of IAEA (2014). Reference organisms are also used in ERICA (Brown et al. 2008). The TRS partly fills data gaps concerning ecologically important species. For example, separate CRs for benthic, piscivorous and forage fish species are given in TRS, whereas one value for all fish species is used in the ERICA Tool (Howard et al. 2013). Uncertainties related to the reference organism concept in terrestrial and aquatic ecosystems have been discussed by Oughton et al. (2008) and Hosseini et al. (2008), respectively.

Concentration ratios have been empirically determined for total elements (e.g. Reiman 2001, Kabata-Pendias 2011) and radionuclides (e.g. IAEA 2010, 2014). Concentration ratios are typically log-normally distributed, and the recommended summarized measures are therefore geometric mean (GM) and

geometric standard deviation (Sheppard and Evenden 1988, 1997). A few high values in log-normally distributed data do not change the geometric mean as much as they would affect an arithmetic mean. Geometric standard deviation (GSD) is a dimensionless multiplicative factor that describes the relative variation around a geometric mean, and GSDs can therefore be directly compared regardless of the GM value.

Empirically determined CR values typically vary in wide ranges (Sheppard and Evenden 1988, Ehlken and Kirchner 2002; Higley and Bytwerk 2007, IAEA 2010). This high degree of variation in the CR for an individual radionuclide reflects the complexity of the transfer process (Ehlken and Kirchner 2002). In an attempt to reduce the uncertainty surrounding CR values in terrestrial ecosystems, it has been suggested that soil properties should be characterized to allow CR values to be compared as long as they have been determined in similar environmental conditions (Sheppard and Evenden 1988, Vandenhove et al. 2009b, IAEA 2010).

Such comparisons are, however, difficult because CR values for many specific environments are missing, and information on soil properties is generally not available for existing CRs. Less than half of the studies reviewed by Vandenhove et al. (2009b) reported important soil properties such as soil type, pH, OM and CEC. In addition, the comparability of CRs may be reduced by different pre-treatments, as there is no standardized method with which to generate CRs (Higley and Bytwerk 2007). Measurement of wet or dry weight, mobile or total soil concentration, and differences in methods used for leaching a mobile soil fraction are common methodological differences which can lead to different values in CR studies (Higley and Bytwerk 2007).

1.8 LINEARITY ASSUMPTION

The CR concept includes the assumption that the relationship between an organism and element concentrations in the media

is linear, and the intercept of this relation is zero (Sheppard and Sheppard 1985, IAEA 2010). However, deviation from linearity has been observed in many studies with respect to soil-to-plant CRs. Timperley et al. (1970) suggested that only the uptake of essential elements would be non-linear. However, many investigators have observed non-linearity in the uptake of both essential and non-essential stable elements (Ca, Cd, Co, Hg, Mg, Mn, Pb, Zn) and radionuclides of Cs, K, Po, Ra, Th and U (Sheppard and Sheppard 1985, Simon and Ibrahim 1987, Cook et al. 1994, Mortvedt 1994, Palm 1994, McGee et al. 1996, Martínes-Aguirre et al. 1997, Krauss et al. 2002, Morton et al. 2002, Vera Tome et al. 2003, Han et al. 2006).

The non-linear uptake of elements is characterized by their more efficient uptake at low than at high soil element concentrations. If one considers the situation with boreal forest plant species, the transfers of ^{137}Cs and ^{40}K from soil to *Juncus squarrosus, Calluna vulgaris, Vaccinium myrtillus* and *Vaccinium vitis-idaea* were studied in Ireland and Sweden (McGee et al. 1996). Clearly decreasing CRs with increasing soil activity concentrations were observed uniformly in these two different countries. The authors concluded that there are serious problems with the use of CR for the evaluation of radionuclide transfer. In *Vaccinium palladium*, a decrease in the CR values for U and Th with increasing soil concentrations was reported to be similar in different soil layers in North-America in a location where soil U and Th concentrations were naturally elevated (Morton et al. 2002). Sheppard et al. (1985, 1988, 1992) have repeatedly detected non-linearity of soil-to-plant CRs, mainly of U for several plant species in Canada.

In one study, a linear transfer of heavy metals was observed when the soil concentration was analyzed after 2% ammonium citrate leaching, but the transfer was non-linear when either total concentration or other leaching procedures were applied (Chojnacka et al. 2005). Vera Tomé et al. (2003) found other kinds of inexplicable results i.e. the transfer of essential elements was non-linear and CR values were declining with increasing soil concentrations, but transfer of non-essential elements (Fe,

Li, Na, Ti) was neither linear nor non-linear; for these metals, CRs seemed to describe only wide and random variation.

Non-linearity of transfer has also been observed in animal data. Transfer non-linearity from water to fish was observed by Pyle and Clulow (1997), who reported that activity concentrations of ^{226}Ra, ^{232}Th, ^{230}Th and ^{228}Th in fish tissues did not increase linearly with increasing water activity concentrations in Ontario, Canada.

Sheppard and Evenden (1992) reported non-linearity of soil-to-earthworm CRs of U for *Lumbricus terrestris* in Canada. Studies with two earthworm species (*Lumbricus rubellus, Eisenia andrei*) observed a decrease in CR with increasing soil concentrations for element concentrations of As, Cd, Cu, Pb and Zn (Vermeulen et al. 2009) and for radionuclides of Cs, Sr and Zn (Keum et al. 2013). Di Lella et al. (2005) studied the transfer of U from soil to three different earthworm species (*Lumbricus terrestris, Alloboghora rosea, Nicorilus caliginosus*), and reported more efficient uptake by earthworms at low soil U concentrations than occurring at higher soil concentrations. Nahmani et al., (2007) reviewed studies on earthworms (mainly *Eisenia fetida*) and found that the uptake of several metals was lower at high than at low soil metal concentrations.

Uptake of essential elements from food or medium might be non-linear because there are biological processes intended to maintain element concentrations in tissue at physiologically optimal levels (Windish 2002). With respect to plants, Sheppard and Evenden (1997) suggested that low GSDs of Mn and Zn CRs may be related to physiological regulation of the concentration levels of these elements inside the plant.

However, this is probably a simplified picture, and it is more likely that the behavior of some elements is extremely complex. For example, there is the possibility that biological regulation mechanisms have evolved to avoid excessively high tissue concentrations of elements that are toxic but present in the natural environment (such as Pb). Neuheuser et al. (1995) found that the concentration of Pb in earthworm tissues (mainly *Allolobophora tuberculata Eisen* and *Lumbricus rubellus Hoffmeister)*

was biologically regulated, as were the concentrations of Cd, Cu and Zn but not that of Ni. Furthermore, the uptake of some non-essential elements may be influenced by chemical similarities and interactions with essential elements; this might be possible for Pb because of its structural similarities with Ca (Pereza et al., 1998), which is an essential ion in plant and animal metabolism.

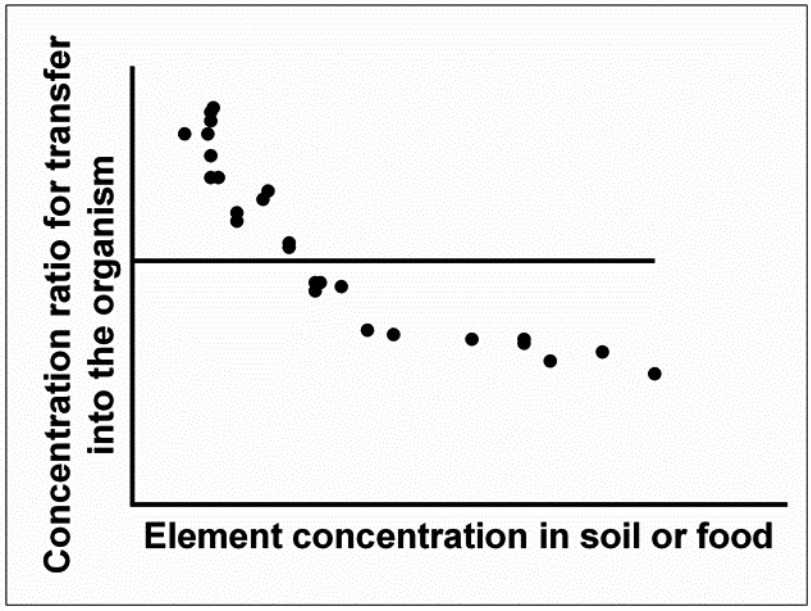

Figure 1. *Schematic diagram showing transfer into organisms as a function of element concentration in soil or food. The "observed" (non-linear) transfer pattern is marked with dots and the linearity assumption (constant concentration ratio) with a solid line*

The non-linear transfer of elements has been observed in many studies for different isotopes, elements, species and trophic levels. The pattern of this non-linearity in different studies displays similarities: the transfer of elements decreases with an increasing element concentration in soil or in food (Fig. 1). If this kind of non-linear transfer is modeled by a linear model, the transfer of elements will be underestimated if there is a low soil or food element concentration and overestimated at high soil or food element concentrations (Sheppard and Sheppard 1985, Pyle and Clulow 1997). According to Sheppard and Sheppard (1985), transfer of U to plants is underestimated

at soil concentrations < 20 mg kg^{-1}, which is the situation in boreal forest where the prevailing soil U concentration is 0.04 mg kg^{-1}. Simon and Ibrahim (1987) suggested that more realistic transfer estimations might be obtained by devising non-linear equations that describe uptake as a function of soil element concentration.

Sheppard and Evenden (1997) considered non-linearity to be just one of the many sources of variability in CR values, and argued that a non-linear relationship has usually been reported when only narrow ranges in soil concentration have been studied. Due to the high variation of CRs, they proposed that soil concentrations would need to differ by several orders of magnitude before a statistically significant relationship between plant and soil concentrations could be detected. In fact, soil concentrations in boreal forest are low and the range of background levels is narrow (Reiman et al. 2001) compared to the studies of Sheppard and Evenden (1997), which included transfer data related to uranium mines. Nonetheless, from the point of view of risk assessment, as the difference between linear and non-linear transfer is most critical at low soil concentrations, it is necessary to study the transfer of elements in low, naturally prevailing soil concentrations.

1.9 AIMS OF THE STUDY

This study aimed at determining the transfer of elements relevant to the nuclear fuel cycle in boreal ecosystems. The study focused on the linearity of the transfer of radionuclides into plants and animals and the influence of non-linear transfer on radioecological modeling. The detailed aims of the study were:

1) To investigate the validity of the linear assumption for uptake of essential and non-essential elements into boreal forest plants

2) To examine the possible non-linearity of transfer of ^{137}Cs into several fish species with different trophic levels in northern lakes
3) To confirm earlier observational results of non-linearity of uptake into plants in a controlled experimental system
4) To determine the transfer from soil or food to animals in a terrestrial food chain consisting of boreal forest species
5) To evaluate the impact of non-linear transfer in radioecological modeling.

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6 Discussion

6.1 METHODOLOGICAL ISSUES

6.1.1 Total versus mobile element concentration

The total element concentration in soil is measured after HNO_3 leaching, whereas the mobile fraction of the total concentration can be assayed by using different leaching methods. The majority of published CR values have been based on total element concentrations in soil (IAEA 2014), although it is commonly assumed that only the mobile fraction is biologically available for plants, and that uptake is more directly associated with the mobile than on total element concentration in the soil (Ehlken and Kirchner 2002, Chojnacka et al. 2005, Blanco Rodríguez et al. 2006). Therefore, it has been postulated that CRs would be better defined based on soil mobile rather than on the total concentration. However, there is an open question about how the bioavailable fraction should be defined, as there is no consensus about what would represent a universally valid method to determine this fraction (Ehlken and Kirchner 2002, Kabata-Pendias 2004, Vandenhove et al. 2007a).

In this study, both total and mobile soil concentrations were determined in Chapter 2. Total concentration was analyzed after nitric acid digestion (HNO_3) and the method used in this study to extract mobile soil concentration was 1M ammonium acetate (NH_4Ac) leach buffered at pH 4.5. This leach extracts those elements chemically adsorbed to the soil (Räisänen et al. 1997) and is suitable for simulating metal behavior in close proximity to the root (Schultz et al. 2004). Nonetheless, the results revealing a non-linearity of transfer were similar when both methods were used to extract the soil samples (Chapter 2). Furthermore, earlier studies have indicated that soil mobile concentration was no better than the total concentration for predicting the actual concentration in plants, and the variations

in the CR values were not decreased when they were based on mobile soil concentrations (Roivainen et al. 2011a,b). As there appeared to be no advantage from the adoption of the mobile fraction in soil, the total element concentration was used as the only measure of soil concentration in Chapters 4 and 5. The application of different extraction methods would also increase the complexity of modeling, and it has been recommended to keep the parameters as simple as possible in radioecological risk assessment models (Ehlken and Kirchner 2002).

6.1.2. Environmental factors affecting transfer of elements

Several environmental factors including K_d (Vandenhove et al. 2007b, 2009), pH (Echevarria et al. 2001), fertilization (Ryfyikiri et al. 2006), interactions of elements, OM, clay content (Roivainen et al. 2012, Vandenhove et al. 2007c) and rhizosphere processes (Ehlken and Kirchner 2002) may affect the transfer of elements from soil to plants. The pH, OM and clay content of soil were measured in this study (Chapters 2 and 4). As all environmental factors cannot be controlled in field conditions, transfer studies were conducted also in experimental mesocosms in an attempt to reduce the effects of uncontrolled variables influencing the transfer of elements. The main finding of the boreal forest study (Chapter 2) - non-linear transfer of elements from soil to plants - was similar at two forest sites and then confirmed in the experimental system (Chapter 4). It therefore appears to reflect true non-linearity in the uptake of the elements into plants rather than confounding by uncontrolled environmental factors.

In chapter 4, transfer of elements to animals was studied in experimental systems, to avoid some of the environmental factors which cannot be controlled in field experiments. Mesocosm experiments were not totally controlled, as two different kinds of soils were included in the mesocosms. However, the confounding effect of the two soils was excluded in the microcosm studies, where identical soil (sand) was used. The results of the mesocosm and microcosm experiments (Chapter 4, Table 5) were comparable, indicating that different

soils did not affect transfer of elements to animals in the mesocosms. The only exception to the similarity of results was found for the uptake of U from uranium-rich soil, as microcosm results showed that soil contact of snails to uranium-rich soil had a major impact on the transfer of U into the snails.

Lack of measurements of K_d and CEC is a weakness of this study, as these properties are closely related to the mobility of elements, which may affect uptake of elements into plants. In other studies, high solubility, i.e. low K_d value, has been related to higher potential plant uptake, as a negative correlation was revealed between K_d and CR (Watmough et al. 2005, Vandenhove and Van Hees 2007). The availability of K_d and CEC would have increased comparability of the results of this study (Chapter 2 and 4) with data in the literature.

6.1.3. Total element versus radionuclide transfer

In this study, total element concentrations were measured in order to increase our understanding of the uptake of radionuclides into plants and animals. This approach is often used as it can be assumed that stable and radioisotopes of the same element are taken up similarly (Baes et al. 1984, cited by Sheppard and Evenden 1988; IAEA 2010; Kryshev and Sazykina 1986; Sazykina 1994). However, equal uptake may not be always true, as differences in speciation may influence the relative bioavailabilities of stable vs. radioisotopes (Salbu and Skipperud 2009). As the focus of this study was on modeling of future contamination situations, the relative bioavailabilities of stable vs. radioisotopes are not known. It is therefore a reasonable assumption that there would be equal uptake of stable and radioisotopes and this was used here. This assumption does not reduce the usability of the results of the present study. As pointed out in Chapter 5, the modeler can easily use alternative assumptions (by using an uptake coefficient lower or higher than 1.0 for the radioisotope), if there is a good basis for assuming that the bioavailability of a radioisotope would differ from that of the stable element.

6.1.4 Equilibrium

The novel modeling approach created in Chapter 5 for modeling non-linear transfer of elements from soil to plant was based on equilibrium as is the CR concept and the non-linear model. As the models discussed in this study are intended for modeling radionuclide concentrations in the far future, it can be assumed that equilibrium will have been reached. This is a common assumption in radiological modeling and it has been stated that equilibrium based models are acceptable when long time intervals are being modelled (Howard et al. 2013). Of course, deviations from equilibrium can occur, but as their size and direction in future conditions cannot be known, equilibrium can be considered as the best alternative.

6.2 IS UPTAKE INTO ORGANISMS ALWAYS NON-LINEAR?

It has been proposed that non-linear transfer of elements is a characteristic seen only with essential elements due to the necessity of plants to control nutrient concentrations (Kabata-Pendias 2011), with the corollary, that uptake of non-essential elements should be linear (Timperley 1970, Vera Tomé et al. 2003). However, non-linear uptake has been observed also for non-essential elements (Sheppard and Sheppard 1985, Sheppard and Evenden 1988, Krauss et al. 2002, Han et al. 2006). This, may be related to the ability of plants to inhibit concentrations of those elements that are needed in small quantities but deleterious at higher doses (Kabata-Pendias 2011, Palmer and Guerinot 2009). In this study, non-linearity of transfer was not related to essentiality of elements, as non-linearity was similarly observed for both essential (Co, Mo, Ni, Zn) and non-essential (U, Pb, Th) elements (Chapters 2 and 4).

This study included several plant species displaying different growth characteristics. The understory species selected were a monocotyledonous herb (May lily), a fern (narrow buckler fern), a dicotyledonous dwarf shrub (bilberry) and a perennial grass (Scandinavian small-reed), and the tree species selected were

coniferous Norway spruce and deciduous rowan and downy birch with two different genotypes. Regardless of growth characteristics, the transfer of elements from soil to plant species was non-linear (Chapters 2 and 4). Transfer of elements was similarly non-linear also for two different genotypes of downy birch, one of which was known to efficiently accumulate heavy metals (Chapter 4). In field layer plants, the plant parts studied were leaf, stem or petiole, and root. Leaf or needle, coarse root and fine root were investigated in the tree species (Chapter 2). It was found that the transfer of elements was non-linear into all plant parts studied (Chapter 2). As non-linear uptake was observed independently of whether the mobile or total concentration in soil was measured, and in both field and experimental study designs (see section 6.1), the results of this study indicate that the transfer of most elements, from soil to plants is non-linear.

Transfer of ^{137}Cs from water to fish was also exhibited a similar non-linear pattern as the uptake of elements from soil to terrestrial plants. As transfer from water to plankton was not studied, it is not known whether the results reflect non-linear uptake from water to plankton or from plankton to plankton-eating fish species, or both. Given the observations that uptake from soil to plants was nonlinear and transfer from prey fish to predator fish was linear, it would have been tempting to hypothesize (as in chapter 3) that transfer of elements is nonlinear only at the first trophic level (e.g., from soil to plants or from water to phytoplankton).

The mesocosm experiments, however, showed that transfer of elements can be non-linear also at higher trophic levels. Transfer of four essential elements (Co, Mo, Ni, Zn) into earthworms and snails was non-linear: it was more efficient at low than at high soil or food element concentrations (Chapter 4). On the contrary, the transfer of a non-essential element (U) into earthworms and snails was found to be nearly linearly related to element concentration in soil or food (Chapters 3 and 4). For another non-essential element (Pb), however, transfer into earthworms and snails was found to be non-linear, which

indicates that the linearity of transfer into animals cannot be simply predicted by deciding whether the element is or is not essential. Nonlinear uptake of Pb may be related to its chemical similarities with Ca (Pereza et al. 1998), i.e., Pb may be transferred as an analogue of Ca.

6.3 IMPACTS OF NONLINEAR TRANSFER ON THE PREDICTIONS OF RADIOECOLOGICAL MODELS: COMPARISON OF THREE MODELS

This study has focused on examining the linearity of transfer, which is a fundamental assumption in equilibrium-based models that use CR to describe the transfer of elements into organisms. Both field studies and mesocosm experiments demonstrated that the uptake of elements into plants did not fit with the assumption of linear uptake. Modelling transfer with a non-linear function was then investigated, and finally a novel, simple model was proposed, taking into account the observed non-linearity of transfer from soil to plants.

The results of this study indicate that, at least for the elements studied, the concentration in plants increases steeply at low soil concentrations, and then reaches a plateau, with no further increase even though the soil concentration continues to increase. If the concentration ratio is plotted as a function of concentration in soil, CR is seen to systematically decrease with soil concentration. This pattern of transfer into plants was found to be in agreement with the non-linear equation proposed by Irving Langmuir in 1819 for adsorption of gases.

$$C_p = abC_s/(1+bC_s), \quad (1)$$

where a and b are experimentally determined parameters, C_s is the element concentration in soil, and C_p is the element concentration in plant. The Langmuir function has been previously used to describe the transfer of elements from soil into plants by Palm (1994), Wenger et al. (2002), Han et al (2006)

and Redjala et al. (2010). However, although the real uptake process in nature appears to be non-linear, the introduction of a Langmuir-type equation in radioecological modeling would not be straightforward. Instead of one empirically determined parameter (CR) which is used in linear modeling, models based on a Langmuir-type equation would require details of two empirically determined parameters (*a* and *b* in the Langmuir equation) for many elements, species and different environments. Although it would be theoretically possible to determine these values, they are not currently available, and it would represent a considerable effort to compile an extensive database. Furthermore, the uncertainty of parameter *b* appeared to be very high, exhibiting standard errors that were much larger than the estimated value of *b* (Chapter 5, Table 1).

The novel model proposed in this study (Chapter 5) was based on the observation that element concentrations in plants were constant in practice. This can be understood to indicate that in all of the cases studied, element concentrations in soil were sufficiently high so that the plateau in plant concentration had been reached. At very low soil concentrations, the concentration in plants must of course increase with the concentration in soil (as would occur in the rising part of the Langmuir function), since the plant concentration must be zero when the soil concentration is zero. Such very low concentrations in soil were not observed in this study, and are apparently not common. In a hypothetical situation with a very low concentration of the element of interest in the target soil, the new model would not correctly describe uptake. Importantly, however, it would not underestimate the transfer of the element. Conservative assumptions are generally preferred in radioecological modeling. The advantage of the new model, compared to a model using non-linear equations, is its simplicity. Data needed for model parameters (plant and soil element concentrations) have also been widely published.

It was decided to compare the conventional linear model, a non-linear model based on the Langmuir equation, and the novel constant plant concentration model. The results of these

comparisons have been examined in Chapter 5 where the models were used to predict ^{234}U, ^{59}Ni and ^{210}Pb concentrations in spruce needles. The element concentrations and model parameter values used in the model calculations were obtained from previous studies in boreal forest (Roivainen et al. 2011a,b), and the ranges of assumed radionuclide concentrations in soil were worst case concentrations that could be released from the storage of spent nuclear fuel, according to Jones et al. (2003). Predictions emerging from the non-linear model and the new constant plant concentration model were similar, with only slightly lower radionuclide concentrations predicted by the constant plant concentration model. The predictions of these two models differed clearly from the conventional linear model when the soil total element concentration was either low or high. When compared to these models, transfer of radionuclides from soil to plants was underestimated by the linear model at low total element concentrations in soil. A low total element concentration is often the prevailing condition in boreal forest, as shown for Zn and Ni in Chapter 2 (Fig. 3.) and for Mo and Pb in Chapter 5 (Fig. 1.).

The constant plant concentration model can potentially decrease the uncertainty of the predictions from the radioecological models. The very wide ranges of published CR values may be partly explained by systematic variation of CR with soil concentration, as the empirically determined CR values have been measured in many different environments with varying soil characteristics. As empirically determined CR values are not used in the proposed new model, this additional source of uncertainty can be avoided.

Importantly, the new constant plant concentration model is based on observations on element concentrations in plants as a function of the concentration in soil, and it therefore reflects the natural uptake process of elements from soil to plants more realistically than the linear model. Although modeling is always a simplification of the true natural phenomena, fundamental processes should be modeled as realistically as possible. The transfer of elements from soil to plants is a key process, as

transfer further throughout the food web is based on this initial transfer process.

6.4 CONCLUSIONS

The present study investigated the linearity of element transfer into organisms at different trophic levels in boreal ecosystem species. The study focused on elements related to the nuclear fuel cycle. Element-specific CRs were calculated at both lower and higher media (or food) element concentrations in order to determine whether CR was constant irrespective of the media (or food) concentration, as should be the case if the transfer into organisms was linear.

The results showed that transfer was generally more effective at low than at high media or food element concentrations. This kind of non-linear transfer was observed in all understory and tree species and for all elements studied, both in natural boreal forest and in experimental systems. Linear or nearly linear transfer from food to animals was observed in some cases (e.g. for the transfer of uranium into snails), but the results did not support the hypothesis that uptake into animals would be non-linear only for essential elements. Overall, linear (or nearly linear) uptake of elements into organisms may be an exception (in terrestrial ecosystem studies it was observed only for the uptake of U into snails and earthworms), and more research is needed to determine the specific elements, organisms and other conditions for which linear transfer can be assumed.

The proposed novel modelling approach was based on the observation that element concentrations in plants were in practice constant, and this was introduced as a simple solution to incorporate non-linearity of transfer into radioecological modelling. This approach reflects true uptake processes more realistically than the linear model, and can potentially improve the precision of model predictions.

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In radioecological modeling, transfer of radionuclides from soil or water to organisms is commonly assumed to be a linear phenomenon that can be described by constant concentration ratios. This study reports findings showing that transfer into the biosphere is generally non-linear, as organisms take up elements more effectively when concentration in the medium is low. Implications of these findings to radioecological modeling are discussed, and a novel modeling approach is introduced.

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