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Päivi Roivainen

Characteristics of Soil-to-Plant Transfer of Elements Relevant to Radioactive Waste in Boreal Forest

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ABSTRACT

The use of nuclear energy generates large amounts of different types of radioactive wastes that can be accidentally released into the environment. Soil-to-plant transfer is a key process for the dispersion of radionuclides in the biosphere and is usually described by a concentration ratio (CR) between plant and soil concentrations in radioecological models. Our knowledge of the soil-to-plant transfer of many radionuclides is currently limited and concerns mainly agricultural species and temperate environments. The validity of radioecological modelling is affected by the accuracy of the assumptions and parameters used to describe soil-to-plant transfer.

This study investigated the soil-to-plant transfer of six elements (cobalt (Co), molybdenum (Mo), nickel (Ni), lead (Pb), uranium (U) and zinc (Zn)) relevant to radioactive waste at two boreal forest sites and assessed the factors affecting the CR values. May lily (*Maianthemum bifolium*), narrow buckler fern (*Dryopteris carthusiana*) and blueberry (*Vaccinium myrtillus*) were selected as representatives of understory species, while rowan (*Sorbus aucuparia*) and Norway spruce (*Picea abies*) represented trees in this study.

All the elements studied were found to accumulate in plant roots, indicating that separate CR values for root and above-ground plant parts are needed. The between-species variation in CR values was not clearly higher than the within-species variation, suggesting that the use of generic CR values for understory species and trees is justified. No linear relationship was found between soil and plant concentrations for the elements studied and a non-linear equation was found to be the best for describing the dependence of CR values on soil concentration. Thus, the commonly used assumption of a linear relationship between plant and soil concentrations may lead to underestimation of plant root uptake at low soil concentrations. Plant nutrients potassium, magnesium, manganese, phosphorus and sulphur were found to have major effects on the soil-to-plant transfer of Co, Mo, Ni, Pb, U and Zn.

Universal Decimal Classification: 504.5, 544.584.4, 546.79, 581.133, 581.19, 628.4.047

CAB Thesaurus: radioactive wastes; radionuclides; cobalt; molybdenum; nickel; lead; uranium; zinc; boreal forests; concentration; transfer; uptake; soil; plants; roots; understory; trees; nutrients

Yleinen suomalainen asiasanasto: radioaktiiviset aineet; radioaktiiviset jätteet; koboltti; molybdeeni; nikkeli; lyijy; uraani; sinkki; boreaalinen vyöhyke; kulkeutuminen; kertyminen; maaperä; kasvit; juuri; juuristo; aluskasvillisuus; puut; kasvinravinteet

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Päivi Roivainen

LIST OF ABBREVIATIONS

CEC	Cation exchange capacity
CR	Concentration ratio
Eh	Redox potential
EU	European Union
GLM	General linear model
GM	Geometric mean
GSD	Geometric standard deviation
HNO_3	Nitric acid
IAEA	International Atomic Energy Agency
ICP-AES	Inductively coupled plasma-atomic emission spectroscopy
ICP-MS	Inductively coupled plasma-mass spectroscopy
ICRP	International Commission on Radiological Protection
Kd	Soil-liquid distribution coefficient
NH_4Ac	Ammonium acetate
OM	Organic matter
RA	Regression analysis
STUK	Säteilyturvakeskus (Radiation and Nuclear Safety Authority)
TEM	Työ- ja elinkeinomisteriö (Ministry of Employment and the Economy)

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 Roivainen P, Makkonen S, Holopainen T and Juutilainen J. Soil-to-plant transfer of uranium and its distribution between plant parts in four boreal forest species. *Boreal Environment Research 16: 158-166, 2011.*

Chapter 3 Roivainen P, Makkonen S, Holopainen T and Juutilainen J. Transfer of elements relevant to radioactive waste from soil to five boreal plant species. *Chemosphere 83: 385-390, 2011.*

Chapter 4 Tuovinen TS, Roivainen P, Makkonen S, Kolehmainen M, Holopainen T and 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: 191–197, 2011.*

Chapter 5 Roivainen P, Makkonen S, Holopainen T and Juutilainen J. Element interactions and soil properties affecting the soil-to-plant transfer of six elements relevant to radioactive waste in boreal forest. Accepted to *Radiation and Environmental Biophysics*.

AUTHORS' CONTRIBUTIONS

Chapter 2 Päivi Roivainen contributed to the designing of the study, field and laboratory work and data analysis, and wrote the paper. Sari Makkonen, Toini Holopainen and Jukka Juutilainen contributed to the designing of the study and writing.

Chapter 3 Päivi Roivainen contributed to the designing of the study, field and laboratory work and data analysis, and wrote the paper. Sari Makkonen, Toini Holopainen and Jukka Juutilainen contributed to the designing of the study and writing.

Chapter 4 Tiina S. Tuovinen contributed to the data analysis and writing. Päivi Roivainen contributed to the designing of the study, field and laboratory work, data analysis and writing. Sari Makkonen and Toini Holopainen contributed to the designing of the study and writing. Mikko Kolehmainen contributed to the writing. Jukka Juutilainen contributed to the designing of the study, data analysis and writing.

Chapter 5 Päivi Roivainen contributed to the designing of the study, field work and data analysis and wrote the paper. Sari Makkonen, Toini Holopainen and Jukka Juutilainen contributed to the designing of the study and writing.

Contents

1 General Introduction

1.1 DESCRIPTION OF RADIOACTIVE WASTE

In general, radioactive waste can be classified into three groups: high level, intermediate level and low level waste (IAEA, 1994). High level waste includes spent nuclear fuel from nuclear power plants and other highly radioactive waste materials (IAEA, 1994; Hu et al., 2010). Intermediate level waste can be characterised as waste that requires shielding due to the radionuclide content but heat dissipation does not cause problems during its handling and transportation (IAEA, 1994). Low level waste has low radionuclide content and can be handled and transported without special shielding (IAEA, 1994). Low and intermediate level waste can be further classified according to the half-lives of the radionuclides they contain. Short-lived waste decays to an acceptably low activity level during the period of expected administrative controls while the decaying of the long-lived waste lasts longer (IAEA, 1994).

Nuclear power plants are important producers of all types of radioactive waste. Low level and intermediate level waste is generated during the operation and maintenance of the reactors, and decommissioning of waste from nuclear power plants can also be included in these groups (IAEA, 2004). High level waste is produced in several processes related to nuclear energy production (IAEA, 1994). The production and testing of nuclear weapons are also sources of high level waste (Hu et al., 2010). Processes related to the mining and milling of uranium generate large volumes of radioactive waste (Hu et al., 2010). The total estimated volume of mill tailings produced worldwide is 938×10^6 m^3 (Abdelouas, 2006). Low and intermediate level waste are also generated in various nuclear applications utilised in medicine and research (IAEA, 2004).

Separate disposal strategies are needed for the different waste types. Storing and exempting can be sufficient treatment for low level waste but with higher radioactivity levels the need for more secure disposal facilities increases, and near-surface disposal facilities are used for low and intermediate level wastes (IAEA, 1994). High level waste has to be isolated from the biosphere, and geological disposal facilities, i.e. repositories, are considered to be a solution for the final disposal of high level radioactive waste and spent nuclear fuel in all countries which have nuclear power plants (IAEA, 1994; Hu et al., 2010). Finland and Sweden are among the first countries where the disposal is planned to take place as the projected year for the repository operation is around 2020 (Ruokola et al., 2004; Hu et al., 2010). In many countries, e.g. Canada, China and Japan, the period 2030–2040 has been suggested for the start of the disposal (Ruokola et al., 2004; Hu et al., 2010).

1.2 MANAGEMENT OF RADIOACTIVE WASTES IN FINLAND

The Ministry of Employment and the Economy (TEM) is responsible for the overall management and supervision of the whole nuclear energy sector in Finland. The other authority is the Radiation and Nuclear Safety Authority (STUK), which supervises the nuclear safety and the use of radiation in Finland.

Radioactive waste produced in Finnish nuclear power plants should be handled according to the Finnish nuclear energy law (990/1987). At the moment there are four nuclear power reactors in operation in Finland, two in Eurajoki and two in Loviisa (TEM, 2011). A fifth reactor is under construction and the Finnish Parliament has also ratified Decision-in-Principle concerning two new nuclear power plants (TEM, 2011). The nuclear power companies are responsible of taking care of their own low and intermediate level wastes and have repositories for them (Ruokola et al., 2004; TEM, 2011). The total amount of these wastes is approximately 300 m^3 per year (STUK, 2011). Additionally, approximately 40 000 m^3 of low and intermediate level wastes will be produced at the time of the decommissioning of the nuclear power plants (STUK, 2011).

The nuclear power companies are also responsible for the handling of spent nuclear fuel. A special fund for nuclear waste management has been created to collect, store and invest the funds that are needed to take care of nuclear waste in the future (TEM, 2011). Spent nuclear fuel from all of the operating reactors, the reactor under construction and also from the future Olkiluoto 4 reactor is planned to be disposed of in a repository built in bedrock on the island of Olkiluoto in the municipality of Eurajoki in south-western Finland (TEM, 2011). If the existing four reactors are in operation for 40 years, they will produce approximately 2600 tons of spent nuclear fuel and the amount increases up to 4000 tons if the operation time is 60 years (STUK, 2011).

Radioactive waste produced in other facilities than nuclear power plants is regulated by the Finnish radiation law (592/1991), according to which the producers are responsible for the handling of radioactive waste. Storage for these kinds of wastes is located in Olkiluoto and contains about 40 m^3 of waste material (Ruokola et al., 2004).

1.3 DISPOSAL OF SPENT NUCLEAR FUEL IN FINLAND

The Finnish nuclear energy law (990/1987) states that nuclear waste should be disposed of in such a way that there is no radiation exposure exceeding the limit that is considered acceptable at the time of final disposal. The law also states that this disposal should be planned so that the long-term safety of this operation requires no surveillance (990/1987). In 2001, the Finnish Parliament ratified the Government's Decision of Principle related to the disposing of spent nuclear fuel in the underground repository in Olkiluoto. The application for the construction of the repository should be filed in 2012, and the repository is planned to be in operation in 2020 (TEM, 2011).

The regulatory requirements for the long-term safety of spent nuclear fuel disposal in Finland are given in the Government Decision on the safety of disposal of nuclear waste (TEM, 2008) and in the regulatory guide YVL 8.4 issued by STUK (2001).

YVL 8.4 is currently under revision and a draft dated 22.9.2010 is available (STUK, 2010). The time frame to be considered is several thousand years (STUK, 2010).

The focus of the safety assessments required by the authorities is on the safety of humans in different exposure scenarios, such as using contaminated water or food (STUK, 2010). Unlikely disruptive events that should be considered are those that may arise from thermal, hydrological and chemical processes, and interactions occurring inside the disposal system or external events and processes such as climate change, geological processes and human actions (STUK, 2010).

Consideration of the safety of non-human biota is also required. It should be shown that the final disposal of spent nuclear fuel has no detrimental radiation effects on any plant or animal species (STUK, 2010). Typical radiation doses to organisms living near disposal facilities should be assessed and the estimated radiation exposure should be clearly lower than doses that can cause the loss of biodiversity or any other significant detriment to any population based on the best scientific knowledge available (STUK, 2010). Populations similar to current ones can be considered in these assessments (STUK, 2010).

1.4 ECOLOGICAL RISK ASSESSMENTS RELATED TO RADIOACTIVE WASTE

The Finnish requirements related to the safety of non-human biota reflect the change in international opinions. The focus of radioecology has traditionally been on the protection of humans, while environmental protection has been considered to be of minor importance (Larsson, 2009). However, this view has been questioned during recent years and the International Commission on Radiological Protection (ICRP) has recommended that protection of the environment should also be taken into account (ICRP, 2007). Because of the quite recent change in perspectives, there is still quite limited understanding of the radiation exposure of non-human biota and radiation effects on plants and animals.

Several international projects have been established to fill in the existing data gaps. ICRP has a separate body, Committee 5, to further discuss the safety of non-human biota (Larsson, 2009). The European Union (EU) has also funded three projects related to the protection of the environment from radiation. FASSET (Larsson, 2004) was conducted between 2000 and 2004, ERICA (Larsson, 2008) between 2004 and 2007, and PROTECT (Howard et al., 2010) between 2006 and 2008. The International Atomic Energy Agency (IAEA) has also been actively involved in considering the predictive capability of environmental models with its programmes EMRAS (Environmental Modelling for Radiation Safety) and EMRAS II (Larsson, 2009).

It is impossible to assess the risks to all individual species because of the huge diversity of non-human biota. Therefore, an approach based on reference animals and plants representing marine, freshwater and terrestrial ecosystems has been adopted by the ICRP Committee 5 and EU-funded projects FASSET and ERICA (Larsson, 2008). The possibility to develop radiological environmental protection based on knowledge obtained from chemical environmental protection has been acknowledged by the ICRP (ICRP, 2003) and is taken into account in the EU funded project PROTECT (Copplestone et al., 2009).

The ecological risk assessments carried out in terrestrial environment are necessary since repositories for radioactive waste exist and are planned to be constructed in terrestrial ecosystems. Plants play a key role in the terrestrial ecosystem and understanding uptake by plant roots is critical in all ecological risk assessments carried out in terrestrial environments (McLaughlin, 2002; Johnson et al., 2003). Besides their crucial role as primary producers in ecosystems plants serve as the route through which elements, including radionuclides, can transfer from soil to humans and animals (McLaughlin, 2002; Kabata-Pendias, 2011).

1.5 PLANT UPTAKE OF ELEMENTS

Plants can take up elements from soil by root uptake or absorb them through above-ground plant parts from the air (Denny, 2002; Kabata-Pendias, 2011). Of these two processes, root uptake is considered to be more important (Denny, 2002; Kabata-Pendias, 2011) and it is also relevant when the possible risks related to soilborne radioactive contamination are considered. The importance of foliar uptake is increased in the cases where radioactive fallout occurs after nuclear accidents, for example (IAEA, 2010). Our knowledge concerning root uptake of elements stems mainly from studies on a few elements, especially plant nutrients, but it is assumed to represent the processes in general (McLaughlin, 2002; Kabata-Pendias, 2011). Although plants are effective in acquiring the essential nutrients they need, they are not perfectly selective and also take up toxic elements (Denny, 2002).

Soil solution is the medium for ion movement to the root surface (Taiz and Zeiger, 2006) and three major processes are known to move mineral elements to the root surface. Elements can diffuse along concentration gradients and they can reach the root through root interception or by mass flow driven by transpiration (Marschner, 1995; McLaughlin, 2002). The movement of ions from soil solution to the xylem vessels that are part of the transport system of the plant occurs by two principal processes: apoplastic and symplastic transfer (McLaughlin, 2002; Mauseth, 2003).

In apoplastic transfer, ions move through the free space between cell walls and through cell walls without crossing the plasma membrane inside the cell walls (McLaughlin, 2002; Mauseth, 2003). This uncontrolled diffusion is prevented by root endodermis (McLaughlin, 2002; Mauseth, 2003). Endodermis encircles the conducting tissues in roots and demarcates the stele (Denny, 2002). The endodermal cell wall develops a suberized band, the Casparian strip, that acts as a barrier to the passive movement of ions and makes the uptake selective (Denny, 2002, Mauseth, 2003).

Symplastic transfer means that ions cross the plasma membrane to the cytoplasm of cells in the cortex and move through the plasmodesmata to the stele (McLaughlin, 2002). This pathway is controlled by the selective permeability of the plasma membrane and the presence or absence of molecular pumps (Mauseth, 2003).

In all cases, the bioavailability of elements limits the root uptake since not all forms of elements are available for plants (Sauvė, 2002; Kabata-Pendias, 2011). The binding of elements to soil constituents is an important factor determining the biological availability of elements to plants (Kabata-Pendias, 2011). Several processes related to the properties of both soils and plants affect the bioavailability of elements and thus also the root uptake by plants. These processes will be discussed in the following sections.

1.5.1 Effects of soil properties and other elements on root uptake

Inorganic soil particles can be classified as coarse sand (diameter 0.2–2 mm), fine sand (0.02–0.2 mm), silt (0.002–0.02 mm) and clay (< 0.002 mm) (Mauseth, 2003; Taiz and Zeiger, 2006). The fraction < 0.02 mm (i.e., silt and clay) is known to affect the behaviour of elements in soil (Kabata-Pendias, 2011). Cations in soil are bound to clay particles and have to be dissolved before they are available to plants (Mauseth, 2003, Taiz and Zeiger, 2006). The addition of a cation, such as potassium K^+ or proton H^+, can displace another cation on the surface of a soil particle and make it available for root uptake (Mauseth, 2003; Taiz and Zeiger, 2006). This process is called cation exchange (Taiz and Zeiger, 2006). The cation exchange capacity (CEC) of a soil is the degree to which it can adsorb and exchange ions (Taiz and Zeiger, 2006). Clay minerals are highly variable and their CEC values differ (Kabata-Pendias, 2011). Anions tend to remain dissolved in the soil solution more than cations (Taiz and Zeiger, 2006) but some clay minerals with a positive surface charge are important anion adsorbing components (Koch-Steindl and Pröhl, 2001).

The microbial decomposition of dead plants, animals and microorganisms produces organic soil particles (Taiz and Zeiger, 2006, Kabata-Pendias, 2011). This decaying organic matter (OM) is heterogenous and consists of organic acids, lipids, lignin, and fulvic and humic acids, and there are several possible reactions and interactions between OM and elements (Koch-Steindl and Pröhl, 2001; Kabata-Pendias, 2011). Because OM has a negative surface charge and can hold cations similarly to inorganic soil particles, it can increase the CEC of soils (Mauseth, 2003; Taiz and Zeiger, 2006; Kabata-Pendias, 2011). OM has been found to reduce anion adsorption because of the formation of organic coatings on the surface of anion adsorbing minerals (Koch-Steindl and Pröhl, 2001).

Soil pH is often considered to be among the most important factors affecting root uptake by plants (Denny, 2002; Kabata-Pendias, 2011). When the acidity of soils increases, a greater concentration of protons exists and more cations are released from the soil. However, in very acid soils the cations are released too rapidly (Mauseth, 2003). Protons can also be competitors for metal uptake by roots (McLaughlin, 2002). Soil pH affects the chemical form and thus the solubility of elements (Sauvė, 2002; Mauseth, 2003). In general, a soil pH between 6.5 and 7.0 is considered to be the best for many elements when the solubility of elements is considered (Mauseth, 2003). However, discussing the independent influence of pH is difficult since physicochemical characteristics of soil are involved in interrelated processes (Sauvė, 2002). The redox potential (Eh), which is negatively correlated with soil pH, is also an important factor controlling the kinetics of elements in soils (Koch-Steindl and Pröhl, 2001).

Of the several mineral oxides occurring in soil, Fe and Mn oxides/ hydroxides play the most important role in element behaviour (Koch-Steindl and Pröhl, 2001; Kabata-Pendias, 2011). They are common constituents in soils, are present in various forms and have a high sorption capacity, particularly for trace elements (Kabata-Pendias, 2011). Hydroxides of Al can also adsorb a variety of elements and their role can be significant in some soils (Koch-Steindl and Pröhl, 2001; Kabata-Pendias, 2011).

Iron oxides, in particular, have variable surface charges and can thus also adsorb anions (Kabata-Pendias, 2011). The sorption capacity of Fe oxides for phosphates, molybdates and selenites is high but decreases with increasing pH value (Kabata-Pendias, 2011).

Predicting the effects of soil properties on plant uptake is not straightforward. For example, Watmough et al. (2005) found that soil pH clearly affected the partitioning of metals in soils in Ontario forests but the effects on tree foliage concentrations were less significant. In general, the plant concentration of an element is higher at low soil solution pH (Tyler and Olsson, 2001). However, Ca, Hg, Mg, Mo and S have been found to exhibit the opposite behaviour (Tyler and Olsson, 2001). It is a general trend that the elements adsorbed on clay are most readily available to plants (Kabata-Pendias, 2011). The elements fixed by oxides and bound onto microorganisms are much less available (Kabata-Pendias, 2011).

In addition to the physical and chemical properties of soil, the interactions between chemical elements in soil can affect the root uptake of metals (Ehlken and Kirchner, 2002; Kabata-Pendias, 2011). Competition between different elements in root uptake is an example of these interactions (McLaughlin, 2002; Kabata-Pendias, 2011). In antagonistic interactions the combined physiological effect of two or more elements is less than the independent effects of those elements (Kabata-Pendias, 2011). Synergism occurs if the combined effect is stronger than the independent effects (Kabata-Pendias, 2011). These reactions are highly variable and may occur inside the cells, within the membrane surfaces, and also generally in the rhizosphere, i.e. the immediate microenvironment surrounding the root (Kabata-Pendias 2011). They are controlled by several factors but the mechanisms are still poorly understood (Kabata-Pendias 2011). Calcium, P and Mg are the main antagonistic elements affecting several trace elements (Kabata-Pendias 2011). Usually these effects occur in two ways: macronutrients inhibit trace element absorption and trace elements inhibit macronutrient uptake (Kabata-Pendias 2011). Fe, Mn, Cu and Zn are the trace elements most often involved in antagonistic interactions (Kabata-Pendias

2011). Ehlken and Kirchner (2002) concluded that the concentration of a trace element in plants may depend primarily not on its concentration in the soil-plant system but on the concentration ratio to micro- and macro-nutrients.

1.5.2 Effects of plants on root uptake

The properties of plants are very significant when determining the uptake of trace elements and they can vary even between different genotypes of the same species (Kabata-Pendias, 2011). Plants can modify the chemistry of soil and soil solution in the rhizosphere (McLaughlin, 2002; Taiz and Zeiger, 2006). Courchesne et al. (2008) concluded that the activity of plant roots has a strong effect on the microscale distribution of trace metals in soils and thus the speciation of metals in the rhizosphere differs from that of bulk soil.

The capacity of a given plant to develop an extensive root system with a large absorption area determines its ability to obtain elements (Taiz and Zeiger, 2006). The structure of root systems differs between plant groups and species. Monocots tend to have a fibrous root system with all the roots having the same diameter while the root system of dicots consists of a main root axis and branched smaller roots (Denny, 2002; Taiz and Zeiger, 2006). All active roots have root hairs which are the major sites of absorption of water and elements (Denny, 2002).

The nutrient uptake of many plants is modified by mutualistic symbiotic associations with soil microbes (Denny, 2002; Mauseth, 2003; Taiz and Zeiger, 2006). Mycorrhizal symbioses between fungi and plants are good examples of these symbioses (Denny, 2002; Mauseth, 2003). According to Taiz and Zeiger (2006), 83-% of dicots, 79-% of monocots and all gymnosperms regularly form mycorrhizal associations. The host plant provides carbohydrates to the mycorrhizae and receives nutrients or water from the mycorrhizae (Taiz and Zeiger, 2006). The presence of fungal hyphae improves the capacity of the root system to absorb nutrients because they are much finer than the plant roots and can extend the reach of roots into a wider area (Denny, 2002; Taiz and Zeiger, 2006).

Roots exude substances that are involved in plant uptake of trace elements (Denny, 2002; Kabata-Pendias, 2004). These exudates are composed mainly of various organic compounds, e.g. amino acids and carboxylates, and they vary with plant species, microorganism association and growing conditions (Kabata-Pendias, 2011). They play a key role in various processes occurring in the rhizosphere. Root exudates affect the variation in pH and Eh regimes, the mobility of macro- and micronutrients and the formation of stable complexes (Kabata-Pendias, 2011). The various organic compounds produced by plant roots and associated microorganisms are also effective in releasing elements from firmly fixed species in soil (Kabata-Pendias, 2011).

1.5.3 The significance of the root-to-shoot transfer of elements

The vascular tissue of plants, consisting of phloem and xylem, is responsible for the translocation of elements within plants (Taiz and Zeiger, 2006). Elements taken up by roots are moved upwards to the shoot in the conducting cells of the xylem by the transpiration stream (Denny, 2002; Taiz and Zeiger, 2006). This process is called primary distribution (Marschner, 1995). A higher ionic concentration in the xylem than in the soil water surrounding the roots is maintained by the presence of the Casparian strip (Taiz and Zeiger, 2006). Other important processes related to the redistribution of elements within plants are retranslocation in the phloem, which is selective and takes place mainly from sources to sinks, and transfer from the xylem to the phloem (Marschner, 1995). The immobilisation of elements in roots is an important process governing the translocation to above-ground plant parts (Kabata-Pendias 2011). The Casparian strip plays a role as a barrier for the movement of ions (Denny, 2002). Elements can also be immobilised to mycorrhizae (Leyval et al., 1997).

The translocation in plants is element-specific and varies also between different plant species and growth seasons (Page and Feller, 2005; Kabata-Pendias, 2011). Potassium and P are examples of plant nutrients that are mobile elements while Ca is immobile in plants (Marschner, 1995). Kabata-Pendias (2011) classified trace elements according to their mobility in plants and concluded that Ag, B, Li, Mo and Se are easily transported from roots to shoots, whereas Mn, Ni, Cd and Zn are moderately mobile, and Co, Cu, Cr, Pb, Hg and Fe are strongly bound in root cells.

1.5.4 Characteristics of boreal forest with respect to plant uptake of elements

Boreal forests are located between latitudes N46° and N66° and cover about 11-% of the earth's terrestrial surface (Bonan and Shugart, 1989; Yuan and Chen, 2010). Strong seasonal variation is characteristic of the climate of boreal forests (Bonan and Shugart, 1989). Boreal forests are generally considered to be quite homogenous (Mauseth, 2003). Conifers, which are evergreen and capable of photosynthesising immediately when solar light is available, are the main tree species in boreal forests (Mauseth, 2003). Shrubs and herbs occur but they are not very abundant (Mauseth, 2003).

The cycling of nutrients, especially N, in boreal forests can be limited because of the cold soil temperatures, which result in reduced OM decomposition (Bonan and Shugart, 1989; Yuan and Chen, 2010). Due to nutrient limitations, boreal forest plant species require quite large root systems and tend to have higher root/shoot ratios than plants in other biomes (Yuan and Chen, 2010). Mycorrhizae also play an important role in boreal forests (Kalliokoski et al., 2010).

Podzolic soils with a layered profile are an important feature of boreal forests (Lundström et al., 2000) and this has to be taken into account when assessing the behaviour of elements in soil. Podzolic soils are covered by an organic mor layer (Lundström et al., 2000). Below this layer is a weathered eluvial horizon containing fewer base cations, Al and Fe than the parent material of the soil (Lundström et al., 2000). This horizon is followed by an illuvial horizon which is enriched in Al, Fe and organics (Lundström et al., 2000). The lowest layer, the C horizon, shows relatively little signs of soil formation (Lundström et al., 2000). The pH of podzolic soils is known to increase gradually with depth (Tyler, 2004).

1.6 BASICS OF RADIOECOLOGICAL MODELLING

Radioecological models are used in licensing nuclear facilities, assessing radiological impacts of these facilities when in operation, handling existing nuclear emergencies and also predicting the future impacts of radioactive waste repositories and possible nuclear emergency scenarios (IAEA, 2009). Many of the models give conservative estimates of exposure, and they are needed in demonstrating that the existing facilities are operating in compliance with regulations concerning the dose limits (Kirchner and Steiner, 2008). Decision-making related to emergencies and existing exposure situations requires more realistic modelling (Kirchner and Steiner, 2008).

Different modelling approaches have been suggested since the importance of protecting non-human biota was recognised, and many of the models are still under development (Beresford et al., 2008ab). The transfer of radionuclides is commonly modelled using the compartmentalisation of the ecosystem into discrete and ecologically relevant components, e.g. soil, wood and leaves (Shaw et al., 2003; IAEA, 2010). The radionuclide fluxes between these compartments are usually described with transfer coefficients, often a ratio between the concentrations in the compartments, i.e. the concentration ratio (CR) (Shaw et al., 2003; IAEA, 2010). Although these ratios are simplified models of a complex series of underlying processes they are important because they are easy to obtain and understand (Sheppard, 2005a). They are also empirical and thus self-validating (Sheppard, 2005a). Many of the radioecological models are deterministic approaches including discrete values for specific model parameters (Kirchner and Steiner, 2008). Probabilistic models use distribution functions for parameters (Kirchner and Steiner, 2008).

The complexity of a model is always a compromise (Kirchner and Steiner, 2008).The situation modelled should be adequately described without a high number of uncertain and variable parameters (Kirchner and Steiner, 2008). Thus, the structure of the model should be simple and contain only the processes that contribute significantly to the concentrations in the compartments (Kirchner and Steiner, 2008).

The true heterogeneity of different model parameters is an important source of uncertainty in radioecological models. However, this variability can only be quantified, not reduced (Kirchner and Steiner, 2008). The uncertainties related to the lack of data and inadequate model design are characteristics that can be reduced to improve the quality of the modelling (Kirchner and Steiner, 2008). Evaluation of different models revealed that the parameters describing the transfer between different compartments make major contributions to the variability in the predictions produced by the models, so they warrant further investigation (Beresford et al., 2008b).

Examples of commonly used models are RESRAD-BIOTA, developed to be consistent with the approach of the United States Department of the Environment, and the ERICA Tool that was developed in the ERICA -project in Europe (Beresford et al., 2008b). Both of these models have different levels, which enable both conservative more general assessments and site-specific modelling (Beresford et al., 2008b). Both of these models use CR values, and RESRAD-BIOTA also includes a kinetic-allometric approach to estimate the transfer of radionuclides to animals (Beresford et al., 2008ab). Developing the ERICA Tool was accompanied by thecompiling of a database of CR values (Beresford et al., 2008b).

1.6.1 Soil-to-plant concentration ratio

Soil-to-plant transfer is a typical process described with a CR value between plant and soil concentrations, and the concentration in plant leaves or in edible parts is usually considered (Ehlken and Kirchner, 2002; Higley and Bytwerk, 2007). Other terms, such as transfer factor and bioconcentration factor, are also used for this ratio. Although CR values are simplistic, they represent the complex interrelationships between organisms, ecosystem and the chemical behaviour of the radionuclide of interest (Higley, 2010). CR values, like any other ratio data, tend to be log-normally distributed (Sheppard and Evenden, 1990; McGee et al., 1996; Sheppard, 2005a;

Sheppard et al., 2006; Vandenhove et al., 2009). Consequently, the geometric mean (GM) and geometric standard deviation (GSD) are generally used to describe the distribution of CR values (Sheppard et al., 2006).

As a consequence of focusing on human risk assessment, most existing data on CR values are from agricultural environments, and fewer data are available for forest plants. For example, default CR values used in the ERICA Tool for shrubs, trees and especially lichens and bryophytes are based on only a few measured values for many of the radionuclides concerned (Beresford et al., 2008c).

There are no standardised methods for studying CR values, so there are certain difficulties in comparing the CR values produced in different studies. Data can be reported based on wet or dry weights, and pre-treatment methods such as washing and peeling can also cause variation between studies (Higley and Bytwerk, 2007). According to the review by Vandenhove et al. (2009), only about 50-% of the reported CR values were accompanied with information about soil type, and data on pH, CEC or OM were even less frequent. This complicates the comparison of different studies and limits the understanding of the effects of soil properties on CR values (Vandenhove et al., 2009).

The CR values of a single radionuclide show large variation even in one plant species (Higley and Bytwerk, 2007; Vandenhove et al., 2009). Sheppard et al. (2006) concluded that this variation coincides with a GSD of the order of 3 to 6. Thus, possible significant differences between plant species are hard to detect (Vandenhove et al., 2009). Lichens, mosses and heather are plant types which normally have higher CR values than other groups, which is probably related to their ability to retain dust (Sheppard et al., 2006). Otherwise the differences between plant types are not consistent from element to element, which might be related to the abilities of plants to regulate the uptake and redistribution of elements (Sheppard et al., 2006). However, there is a tendency that plants consumed by humans have lower CR values than animal forage, native browse, shrubs and trees (Sheppard et al., 2006).

Generic CR values for several plant species have been suggested partly due to the large variation in measured CR values for even a single species (Sheppard et al., 2006; Vandenhove et al., 2009). Another important factor is that there are certain limits for the number of parameters that can be included in one model (Sheppard et al., 2006). The use of generic parameters can be reasonable although ideally CR values are measured for particular plants in contamination scenarios corresponding to the actual cases being investigated (Sheppard and Evenden, 1990; Kabata-Pendias, 2004). In particular, models used to assess the effects of disposal of nuclear waste should be as generic as possible since the contamination scenarios will be relevant in the very distant future, and environmental conditions at that time cannot be precisely known (Sheppard and Evenden, 1990; Sheppard et al., 2006).

An open issue concerning CR values is the assumption of linearity. The calculation of CR values in the traditional way assumes that there is a linear relationship between plant and soil concentrations and that this relationship has a zero intercept (Sheppard and Sheppard, 1985; Simon and Ibrahim, 1987). However, the lack of linearity in plant uptake of elements is well documented in studies on essential plant nutrients (Marschner, 1995) and many heavy metals (Krauss et al., 2002; Han et al., 2006). In the region of low soil concentrations, CR values of essential elements have been reported to decrease with increasing soil concentration towards an asymptotical constant value at higher soil concentrations (Mortvedt, 1994). There is evidence that the assumption of linearity is not valid for radionuclides, either (Simon and Ibrahim, 1987; McGee et al., 1996; Martínez-Aguirre et al., 1997).

Blanco Rodríquez et al. (2002) reported that the linearity assumption of CR values of U, Th and Ra could be verified only if data derived from two distinct areas (a disused uranium mine and a background area) were pooled to create a wide concentration range with observations scattering at both ends of the range. Chojnacka et al. (2005) tested the linearity assumption for CR values of several heavy metals in agricultural plants using different extracting agents. The linearity assumption was valid if soil concentration was analysed after 2-% (w/v) ammonium citrate extraction but not when other extractions were used or the soil total concentration was measured. Vera Tome et al. (2003) reported that the relationship between the CR values of several non-essential elements and their soil concentrations was not linear but different than that between essential elements and their soil concentrations.

Although the CR values intended for assessment purposes are conservative, there is evidence that taking the non-linear behaviour into account could increase the validity of data on the relationship between soil and plant concentrations (Simon and Ibrahim, 1987). McGee et al. (1996) suggested that the theory underlying CR value calculations is inherently flawed because the numerators (plant concentrations of elements) are only slightly variable, while the denominators (soil concentrations of elements) vary widely. They advised that caution be exercised when using any ratio data.

The solid-liquid distribution coefficient (Kd) is a factor closely related to the CR values (IAEA, 2010). The Kd quantifies the degree of radionuclide sorption on the solid phase and can be used to assess the overall mobility and residence times of radionuclides in soils (IAEA, 2010). It is defined as the ratio between the concentration of a radionuclide sorbed on a specified solid phase and the concentration of a radionuclide in a specified liquid phase (IAEA, 2010).

1.7 CHARACTERISTICS OF THE ELEMENTS RELEVANT TO RADIOACTIVE WASTE

The whole nuclear fuel cycle is based on uranium (U) (Abdelouas, 2006). Uranium has ten radioactive isotopes, and U-238 (99.27 %), U-235 (0.72 %) and U-234 (0.0055 %) are the most abundant (Sheppard et al., 2005b). Uranium-235 is the fissionable isotope and its concentration has to be enriched for nuclear fuel (Abdelouas, 2006). Uranium-238 decays to stable Pb-206 through a decay chain that includes Th-234, U-234, Th-230, Ra-226, Rn-222, Pb-210, Bi-210, Po-210 (Mortvedt, 1994).

Possible risks related to U and members of its decay chain can occur during several steps of the cycle from mines to spent nuclear fuel repositories (Abdelouas, 2006; Vandenhove et al., 2007a).

Radionuclides are generated in the nuclear fission and as a result of the activation of reactor core components in operating nuclear power plants (Remeikis et al., 2009). Many of the radionuclides have quite short half-lives and therefore the risks related to those nuclides are relevant mostly during the operation and decommissioning of the nuclear power plants (Ruokola et al., 2004). Examples of these radionuclides are Mn-54, Fe-55, Co-60, Ni-63, Zn-65, Sr-90 and Cs-137 (Mascanzoni et al., 1989; Lindgren et al., 2007; IAEA, 2009; Remeikis et al., 2009).

The focus of radioecological risk assessments is on long-living radionuclides when the risks related to spent nuclear fuel disposal are assessed. Apart from remaining U, spent nuclear fuel contains fission products, Pu and minor actinides (Ruokola et al., 2004; Grambow, 2008; Hu et al., 2010). STUK (2001) sets nuclide-specific constraints for the activity releases to the environment from the disposal facility for the following radionuclides: C-14, Cl-36, Ni-59, Se-79, Zr-93, Nb-94, Tc-99, Pd-107 Sn-126, I-129, Cs-135, Sm-171, Np-237 and the long-living isotopes of Ra, Th, Pa, Pu, Am, Cm and U. Hjerpe et al. (2010) have categorised the key nuclides in the Finnish disposal system into top priority (C-14, Cl-36, I-129), high priority I (Mo-93, Nb-94, Cs-135), high priority II (Ni-59, Se-79, Sr-90) and high priority III (Pd-107, Sn-126).

The following sections describe the soil-to-plant transfer of example elements which have radioactive isotopes (Table 1) in different types of radioactive waste. The elements selected are relevant for the ecological risk assessments of the different steps of nuclear fuel cycle in boreal environments where both mines and nuclear power plants occur. They can be related to environmental problems caused by mining (U-235, U-238, Pb-210), operating nuclear power plants (Ni-63, Co-60, Zn-65) or spent nuclear fuel repositories (Ni-59, Mo-93, U-235, U-238).

Table 1. Radiological properties of radionuclides of Co, Mo, Ni, Pb, U and Zn relevant to radioactive waste.

Nuclide	Half life	Main radiation
Ni-59	7.6×10^4 a	ß
Ni-63	100 a	ß
Co-60	5.27 a	ɣ
Zn-65	244.26 a	ɣ
Mo-93	3.5×10^3 a	electron capture
Pb-210	22.3 a	ß
U-235	7.0×10^8 a	α
U-238	4.5×10^9 a	α

1.7.1 Uranium

Uranium is not known to be an essential element for plants. There is evidence that the cationic uranyl ion is the species most readily taken up by plants (Ebbs et al., 1998). However, this species is present in soil solution only at pH 5.5 or less (Ebbs et al., 1998). Other species preferentially taken up and translocated are uranyl carbonate complexes and $UO_2PO_4^-$ (Vandenhove et al., 2007b).

Uranium accumulates in the plant roots irrespective of plant type (Ebbs et al., 1998; Shahandeh and Hossner, 2002; Thiry et al., 2005; Duquene et al., 2006; Shtangeeva, 2010). Translocation of U in plants differs remarkably between agricultural species: root-to-shoot ratios varied from 28 for Indian mustard to 1330 for maize in a study by Duquene et al. (2006). Shahandeh and Hossner (2002) reported root-to-shoot ratios from 30 to 50 in several agricultural plants. The differences between above-ground plant parts are not that clear. For example, Morton et al. (2002) found that U concentrations in blueberry leaves and stems were similar. Uranium concentrations in twigs and branches of Scots pine were lower than in needles, and the average U content in the foliage increased with the needle age (Thiry et al., 2005). Thiry et al. (2005) also reported that the U concentration in fine roots of Scots pine was higher than in coarser roots.

Soil pH is an important factor affecting the behaviour of U because of the changes in solution speciation and in surface species and surface charge (Ebbs et al., 1998; Echevarria et al. 2001; Vandenhove et al., 2007a). Uranyl cation predominates at low pH where sorption is weak (Ebbs et al., 1998). With rising pH, the number of available binding sites on mineral surfaces increases but also the concentration of carbonate, the most important complexing agent for U, increases (Koch-Steindl and Pröhl, 2001). The mobility of U is increased at pH higher than 6 because of the carbonate complexes (Koch-Steindl and Pröhl, 2001; Tyler and Olsson, 2001) and high total inorganic carbon content has been reported to increase the uptake of U (Vandenhove et al., 2007b). The results of Vandenhove et al. (2007b) suggest that high pH (> 6.9) is related to higher transfer of U into ryegrass (*Lolium perenne* cv Melvina). Tyler and Olsson (2001) found that the root U concentrations of a common grass (*Agrostis capillaris*) were curvilinearly related to soil solution pH, showing a clear U-shaped curve when pH ranged from 5 to 8.

The highest U concentrations in soil solution can be expected when total inorganic carbon and soil solution K concentration are high and exchangeable Mg, exchangeable Ca and P content in soil solution are low (Vandenhove et al., 2007a). Ebbs et al. (1998) suggest that the complexation of U with phosphate can reduce the bioavailability and toxic effects of U.

However, predicting U uptake based on soil solution U concentration is difficult. Vandenhove et al. (2007b) found no significant correlation between the transfer of U into ryegrass and the U concentration in soil solution or any other soil factor. The presence of OM has been reported to decrease the CR values of U (Mortvedt, 1994; Vandenhove et al., 2007b). Higher CR values have been found on sandy soils than on loamy and, especially, clayey soils (Sheppard and Evenden, 1988; Mortvedt, 1994; Vandenhove et al., 2009).

Shahandeh and Hossner (2002) and Straczek et al. (2010) reported that dicotyledonous plant species accumulate more U than monocotyledonous plant species. In contrast, Chen et al. (2005) and Duquene et al. (2006) found no significant difference between these plant groups. A possible effect of mycorrhizae on U uptake has been suggested by Rufyikiri et al. (2003) but Duquene et al. (2006) found no evidence of differences in U uptake between non-mycorrhizal and mycorrhizal agricultural species. Vandenhove et al. (2007b) reported no significant effect of plant related parameters (dry weight, plant K concentration, plant Ca concentration, plant Mg concentration) on the U uptake of ryegrass.

1.7.2 Lead

Lead has not been shown to play any essential roles in plant metabolism (Kabata-Pendias, 2011). There is great variation in plant Pb concentrations because of the effects of several environmental factors, e.g. the presence of geochemical anomalies and pollution (Kabata-Pendias, 2011). Kabata-Pendias (2011) concluded that Pb is one of the elements that are strongly sorbed by soil particles and not readily transported to above-ground plant parts.

The root uptake of Pb occurs passively (Kabata-Pendias, 2011). In general, roots do not take up large amounts of Pb and it is believed to be the least bioavailable metal to plants (Kabata-Pendias, 2011). The translocation of Pb from roots to the above-ground plant parts is low (McLaughlin, 2002; Johnson et al., 2003; Yoon et al., 2006; Kabata-Pendias, 2011). Soil S concentration has been reported to inhibit this transfer even further, and S- defiency remarkably increases the movement of Pb to shoots because of limited root growth (Jones et al., 1973). Yoon et al. (2006) found a relationship between the translocation of Zn and Pb in a study with 17 native plant species. Root uptake and the translocation of Pb into spruce needles has been reported to be small relative to the total Pb content of needles, which originates mainly from the atmospheric deposition of Pb (Hovmand et al., 2009). The importance of atmospheric deposition to the Pb concentrations of above-ground plant parts have been recognised also for agricultural plants (Pietrzak-Flis and Skowrońska-Smolak, 1995).

Sheppard and Sheppard (1991) found a positive correlation between soil pH and CR values in blueberry but other studies have reported that increasing pH, by liming for example, can reduce Pb uptake (Tyler and Olsson, 2001; Kabata-Pendias, 2011). High OM content of soil decreases Pb uptake (Sheppard and Sheppard, 1991; Kabata-Pendias, 2011). Vaaramaa et al. (2010) found that Pb-210 in Finnish soils is associated with humic substances and oxides of Fe, Al and Mn. Vandenhove et al. (2009), in their review, found no significant correlations between CR values of Pb and soil characteristics (pH, CEC, OM and clay content) in agricultural plants. Soil P concentration is known to inhibit the uptake of Pb because of the formation of insoluble phosphates in soils (Kabata-Pendias, 2011). The reported interference between Cd and Pb concentration in plant uptake might be related to their similar properties as divalent cations (Hardiman et al., 1984; Kabata-Pendias, 2011).

Genetic plant factors, root surface area and root exudates play a role in regulating the uptake of Pb (Kabata-Pendias, 2011). Hovmand et al. (2009) concluded that the root uptake and translocation of Pb to spruce needles is lower than that in agricultural crops. This might be because there are more barriers against the translocation of Pb within the longer transport distance in trees (Hovmand et al., 2009). Birch and juniper have been reported to accumulate more Pb in their tissues compared with pine (Klaminder et al., 2005). Turpeinen et al. (2000) showed that pine roots play an important role in the immobilisation of Pb.

1.7.3 Molybdenum

Molybdenum is an essential micronutrient for plants because of its structural and catalytical functions in enzymes closely involved in nitrogen metabolism (Marschner, 1995; Zimmer and Mendel, 1999; Denny, 2002; Kabata-Pendias, 2011). However, the requirements of plants for Mo are low. Taiz and Zeiger (2006) assumed 0.1 mg kg^{-1} (dw) as an adequate tissue level of Mo in plants while Kabata-Pendias (2011) mentioned a range from 0.2 to 5 mg kg^{-1}. Mo is known to bioaccumulate through the soil-plant-animal food chain which increases its importance in risk assessments (McLaughlin, 2002).

According to Kabata-Pendias (2011), Mo is mobile in soil and readily taken up by plants. Plants take up Mo as molybdate anions, which are the predominant aqueous species at pH values above 4.0 (Zimmer and Mendel, 1999; Kabata-Pendias, 2011). Molybdenum can be classified as moderately mobile in plants, and the molybdate anion is assumed to be a major transport form in xylem and phloem (Marschner, 1995; Kabata-Pendias 2011).

The behaviour of Mo is different from that of other micronutrients since it is least soluble in acid soils and readily mobilised in alkaline soils (Zimmer and Mendel, 1999; Kabata-Pendias, 2011). A significant positive relationship between soil solution pH and Mo concentrations in the shoots of a common grass (*Agrostis capillaris*) was found in a study by Tyler and Olsson (2001). Besides pH the mobility and thus availability of Mo to plants is related to drainage conditions (Kabata-Pendias, 2011). Mo is most available in wet alkaline soils and least available on acid soils with a high Fe oxide levels (Kabata-Pendias, 2011).

The uptake of Mo by plants has been reported to decrease in the presence of high concentrations of sulphate (Zimmer and Mendel, 1999; McGrath et al., 2010; Kabata-Pendias, 2011). These anions might use the same transport system into plants (Marschner, 1995). There are also reports of an interaction between Mo and P which suggests that uptake of Mo can also occur via phosphate binding and transporting sites (Zimmer and Mendel, 1999; Kabata-Pendias, 2011). Other interactions of Mo include those with Mn, Cu and Ca (Kabata-Pendias, 2011).

1.7.4 Nickel

Nickel is required by many plants for the structure and catalytic function of the urease enzyme (Marschner 1995; Denny 2002; Kabata-Pendias, 2011). The Ni content of food plants has been found to vary between 0.06 and 2 mg kg^{-1}; the lowest values are found in apple and highest in cucumber (Kabata-Pendias, 2011). In cereal grains the Ni concentrations have been found to vary between 0.34 and 1.28 mg kg^{-1} (Kabata-Pendias, 2011). Taiz and Zeiger (2006) considered that 0.1 ppm (dw) is an adequate tissue level of Ni in plants.

Nickel is among the elements that are mobile in soil and readily taken up by plants (Kabata-Pendias, 2011). Nickel is absorbed both passively and actively by plant roots and thus plant species and the nutritional status of plants can have major effects on Ni uptake (Pinel et al., 2003). Nickel is mobile in plants and therefore easily translocated to above-ground plant parts (Page and Feller, 2005; Kabata-Pendias, 2011). An effect of plant species on the root-to-shoot transfer of Ni in several agricultural plant species was found for plants grown on acid soil but not for plants grown on rendzina (Pinel et al., 2003).

Kabata-Pendias (2011) considers soil pH to be the most well-known soil factor affecting Ni uptake. A negative correlation between soil solution pH and Ni concentration in the shoots of a common grass (*Agrostis capillaris*) was found in a study by Tyler and Olsson (2001). However, Watmough et al. (2005) found no relationship between soil pH and foliar Ni concentrations in four tree species in an Ontario forest. The CR values of Ni for deciduous tree foliage were higher on peatland sites than on mineral soil sites in southwestern Finland (Aro et al., 2009).

Kabata-Pendias (2011) concluded that, in general, the effect of plant species on Ni uptake is significant. The CR values of mosses were higher than those of evergreens, blueberry, herbs, grasses or lichens in a forest in southwestern Finland (Aro et al., 2009). Pinel et al. (2003) concluded that greater differences in CR values between agricultural species were found in acid soil than in rendzina soil.

Mascanzoni (1989) found that soil Ca concentration is negatively correlated with CR values in wheat (*Triticum aestivum*) and timothy (*Phleum pratense*). The CR values of Ni-63 in wheat and timothy also correlated with the CR values of Sr-90, which indicates that these elements compete with the uptake of Ca in a similar way (Mascanzoni, 1989).

1.7.5 Cobalt

Cobalt is not known to be an essential element for higher plants (Marschner, 1995; Kabata-Pendias, 2011). However, it is essential for cyanobacteria in fixing N_2 and is suggested to have a beneficial effect on plant growth and N-fixing processes (Palit et al., 1994; Kabata-Pendias, 2011). Mean Co concentrations in clovers range from 0.1 to 0.57 mg kg^{-1} and in grasses from 0.06 to 0.27 mg kg^{-1} (Kabata-Pendias, 2011). According to Palit et al.

(1994), normal Co concentrations in plants are within 0.1–10 mg kg^{-1}. Kabata-Pendias (2011) classified Co as mobile in soil and readily taken up by plants. Co bioaccumulates through the soil-plant-animal food chain which increases its importance in ecological risk assessments (McLaughlin, 2002).

The results of Page and Feller (2005) suggest that Co in wheat plants was redistributed within the root system but not or only very slowly released into the shoot. The transporting and storing of Co was different between monocotyledonous wheat and dicotyledonous tomato in a study by Bakkaus et al. (2005). They reported that the concentration of Co in the roots of tomato plants was much higher than in the shoots which could indicate that there is a protecting mechanism involved at high Co exposure.

Soil pH has a major effect on the uptake of Co into plants (Palit et al., 1994; Gál et al., 2008; Kabata-Pendias, 2011). Foliar Co concentrations of four tree species in an Ontario forest were highest at the most acidic sites (Watmough et al., 2005). Decreasing CR values with increasing pH have also been found in agricultural crops (endive, maize, wheat, mustard, sugarbeet, potato, Faba bean, rye grass) (Gerzabek et al., 1998). The presence of high concentrations of Mn in soils also inhibits uptake of Co (Palit et al., 1994; Gál et al., 2008; Kabata-Pendias, 2011). The bioavailability of Co in soil is controlled by complexing with organic compounds (Kabata-Pendias, 2011).

Plant properties play an important role in controlling the plant uptake of Co (Palit et al., 1994; Kabata-Pendias, 2011). Gerzabek et al. (1998) concluded that plant-specific factors explained the variation in CR values of Co-60 in agricultural plants better than soil-related factors. CR values in grasses were about 15-fold lower than in other species studied.

1.7.6 Zinc

Zinc is an essential component of several enzymes in plants and its functions in plants are related to the metabolism of carbohydrates, proteins and phosphates (Denny, 2002; Broadley et al., 2007; Kabata-Pendias, 2011). An adequate Zn concentration in plants is considered to be 15–20 mg kg^{-1} (DW)

(Marschner 1995; Taiz and Zeiger, 2006). Background concentrations of Zn in grasses range from 12 to 42 mg kg^{-1} and in clovers from 24 to 45 mg kg^{-1} (Kabata-Pendias, 2011).

Kabata-Pendias (2011) classified Zn as very mobile in soil and easily bioaccumulated by plants. The uptake of Zn can be both an active and a passive process (Broadley et al., 2007; Kabata-Pendias, 2011). Zn^{2+} and Zn complexed with organic ligands are the main forms in which plants take up Zn from soil (Broadley et al., 2007). The mobility of Zn in plants is considered to be high or intermediate (Marschner, 1995; Page and Feller, 2005; Kabata-Pendias, 2011). The average Zn translocation factor between root and shoot in 17 native plant species in Florida has been found to be 0.98 (Yoon et al., 2006). The Zn content of tree roots, especially small lateral ones, is usually higher than that of foliage, branches and trunks (Kabata-Pendias, 2011).

Soil pH is the dominant factor determining the distribution of Zn in soil (Broadley et al., 2007). Soluble Zn increases at low pH (Broadley et al., 2007). Mascanzoni (1989) reported that soil pH correlates negatively with CR values of Zn in wheat and timothy. The effect of soil type (peat, sand, loamy sand, loam, clay loam or clay) did not affect the CR values of Zn in wheat and timothy (Mascanzoni, 1989).

Interactions between Cd and Zn have been reported to have both antagonistic and synergistic effects on the uptake and transport processes of these two elements (Kabata-Pendias, 2011). There is also competition between Zn and Cu, inhibiting the uptake of these elements (Kabata-Pendias, 2011). Yoon et al. (2006) found that the species effective in translocating Cu were also effective in translocating Zn. Soil Ca content has been reported to correlate negatively with CR values of Zn in wheat and timothy (Mascanzoni et al., 1989).

1.7.7 Summary of CR values for Co, Mo, Ni, Pb, U and Zn

As explained in section 1.3.2, CR values are produced with different methods in different studies so comparing the values is not always straightforward. Table 2 summarises the CR values (dw based) for Co, Mo, Ni, Pb, U and Zn. Soil-to-leaf CR values are presented for native plant species while the CR values for agricultural plants describe the overall transfer from soil to leaves and edible plant parts. The calculation of the values presented also differs between the studies referred to. The values reported in Sheppard et al. (2006) and Vandenhove et al. (2009) are GMs of CR values derived from large literature reviews. Other values are from individual studies that report GMs of CR values (Sheppard and Evenden, 1990; Sheppard and Sheppard, 1991), distributions of CR values (Aro et al., 2009) or CR values calculated using medians for plant and soil concentrations (Reimann et al., 2001).

The CR values of Co, Ni, Pb and U have been reviewed in order to produce default CR values for the ERICA Tool (Beresford et al., 2008c). These values were not included in Table 2, because they are calculated for fresh weight-based whole-body activity concentration in biota and have to be converted for comparison with the values in Table 2. The ERICA Tool default values for Co are very limited and based on estimations and assumptions (Beresford et al., 2008c). The values suggest that the soil-to-plant transfer of Co is highest in shrubs (CR 0.75), followed by lichens and bryophytes (0.22), trees (0.018) and grasses and herbs (0.014). More data are available for Ni, Pb and U (Beresford et al., 2008c). The ERICA Tool default CR value of Ni for grasses and herbs (0.2) is higher than the values for lichens and bryophytes (0.09), shrubs (0.03) and trees (0.02) (Beresford et al., 2008c).The default CR value of Pb for lichens and bryophytes is 6.00, which is much higher than the values for shrubs (0.31), trees (0.076) or grasses and herbs (0.067) (Beresford et al., 2008c). The effectiveness of lichens and bryophytes in sorbing and retaining metals from atmospheric deposition is the reason for the high CR value (Berg and Steinnes, 1997; Hovmand et al., 2009). The default CR value of U is also highest for lichens and bryophytes (0.071), followed by values for grasses and herbs (0.015), shrubs (0.0071) and trees (0.0068) (Beresford et al., 2008c).

Table 2. *Summary of the concentration ratios (dry weight based) for Co, Mo, Ni, Pb, U and Zn. The values for native species are soil-to-leaf ratios and those for agricultural plants represent the overall transfer from soil to leaves and edible plant parts.*

Element	**Reference**	**Herbs and grasses**	**Shrubs**	**Deciduous trees**	**Coniferous trees**	**Lichens and bryophytes**	**Agricultural plants**
Co	1		0.02	0.2–0.8	0.03	0.1	
Mo	1		0.1	0.1–0.2	0.02–0.04	0.2	
	2	0.07					
Ni	1		0.07–0.3	0.4–1.0	0.1	0.2	
	3	< 2.0	< 0.5	< 0.5	< 0.5	< 2.0	
Pb	1		0.01	0.01	0.01	0.2	
	4		0.05				
	5						0.02
U	1		0.01	0.01	0.01	0.1	
	5						0.02
	6	0.01	0.007	0.004	0.004	0.3	0.002–0.006
Zn	1		0.3–0.6	2.9–4.8	0.9	0.8	

[1)] Reimann et al., 2001; [2)] Sheppard and Evenden, 1990; [3)] Aro et al., 2009; [4)] Sheppard and Sheppard, 1991; [5)] Vandenhove et al., 2009; [6)] Sheppard et al., 2006

1.8 AIMS OF THE STUDY

The purpose of this study was to investigate the soil-to-plant transfer of elements relevant to radioactive waste in boreal forest plants. Co, Mo, Ni, Pb, U and Zn were the elements chosen for detailed investigation. The study was launched to increase our knowledge concerning the soil-to-plant transfer of radionuclides. Limited data on the soil-to-plant transfer of radionuclides are available for boreal forest environments that can differ greatly from other environments. Thus, the direct use of the results of studies conducted in other kinds of environments, especially in agricultural settings and laboratory conditions, may involve uncertainties. Data are needed for radioecological modelling: the basic assumptions and parameters of the models have to be correct to obtain accurate modelling results representing boreal conditions.

The specific aims of the study were:

1) to determine the CR values of Co, Mo, Ni, Pb and U for five boreal forest plant species representing different growth traits.
2) to investigate the distribution of Co, Mo, Ni, Pb and U in different plant parts (root, stem/ petiole, leaf/needle) and to determine the corresponding CR values for these plant parts
3) to test the validity of the assumption that soil-to-plant transfer is linear in boreal forest species for three essential (Mo, Ni, Zn) and two non-essential (Pb, U) elements
4) to investigate the effects of soil properties and other element concentrations on the soil-to-plant transfer of Co, Mo, Ni, Pb, U and Zn

The results of this work can be utilised in developing radioecological modelling in general and also to make models more suitable to northern conditions. The results related to U and Pb can also be used for risk assessments of possible uranium mines in the boreal zone.

The data for this study were collected from forest sites in Eastern Finland in June 2007. Soil U concentrations were known to be above background concentrations at both sampling sites. Five typical boreal forest plant species representing both understory and tree species were selected for this study based on their availability at the study sites (Table 3). The stable element concentrations were analysed by inductively coupled plasma-mass spectroscopy/atomic absorption spectroscopy (ICP-MS/AES) instead of measuring activity concentrations of specific radionuclides. This allowed measurements of 34 elements in one analysis.

Table 3. *Description of the sampling sites and a summary of the samples collected.*

	Site 1	**Site 2**
Location	Murtolahti, Nilsiä	Puutosmäki, Kuopio
Coordinates	N63°04', E27°54'	N62°42', E27°48'
Forest type	herb-rich, *Oxalis-Maianthemum*	coniferous, *Vaccinium- Myrtillus*
Soil type	no clear distinction between organic and mineral layers	Spodosol
Range and mean of K in soil (mg kg^{-1})	780–4700 (1460)	200–4300 (550)
Range and mean of P in soil (mg kg^{-1})	230–1500 (650)	82–870 (360)
Range and mean of S in soil (mg kg^{-1})	170–4400 (740)	64–1100 (180)
Dominating plant species	May lily, narrow buckler fern, common wood sorrel, common oak fern, grey alder, rowan, Norway spruce	blueberry, Schreber's moss, Norway spruce, Scots pine
Number of sampling points	29	23
Collected plants	May lily (*Maianthemum bifolium*), n = 19 Narrow buckler fern (*Dryopteris carthusiana*), n = 27 Rowan (*Sorbus aucuparia*), n = 28 Norway spruce (*Picea abies*), n = 26	Blueberry (*Vaccinium myrtillus*), n = 23 Norway spruce (*Picea abies*), n = 16

1.9 DATA ANALYSIS

Chemical analyses were carried out in the laboratory of Labtium Ltd. in Espoo, Finland. The laboratory is accredited according to FINAS T025 (EN ISO IEC 17025). The pseudo total concentrations of elements were analysed by ICP-MS/AES after nitric acid (HNO_3) digestion in microwave oven (procedure following US-EPA standard 3051). Ammonium acetate leach (1 M NH_4Ac, buffered at pH 4.5), that extracts the elements chemically adsorbed to soil (Räisänen et al., 1997), was used to obtain an estimate of the mobile fraction of the elements in soil. Plant titanium (Ti) concentration was used as an indicator of soil contamination as plants only take up small amounts of Ti (Cary et al., 1986). The soil properties analysed were pH, OM content and particle size distribution.

All the data used in this thesis were corrected to represent dry matter content. If the measured concentration was under the detection limit, the value corresponding to the half of the detection limit was used in calculations and statistical analyses (Chapters 2, 3 and 5) or the case was excluded from analysis (Chapter 4). The plant and soil element concentrations and CR values tended to be log-normally distributed, and therefore GMs and GSDs are used to describe the distributions throughout the thesis.

The statistical comparisons of the CR values for different plant species and plant parts were carried out with original data and non-parametric tests for U (Chapter 2). For Co, Mo, Ni and Pb the log-transformation of the CR values normalised the data, and One-Way ANOVA was used for these comparisons (Chapter 3). The effects of soil- and plant-related properties on CR values were studied by general linear model (GLM) for all the elements studied. Again, original data were used for U (Chapter 2) and log-transformed data for Co, Mo, Ni and Pb (Chapter 3).

Original data were used for fitting with linear and non-linear equations when the validity of the linearity assumption was investigated (Chapter 4). Linear regression analyses (RA) for investigating the effects of soil element concentrations and soil properties on CR values of Co, Mo, Ni, Pb, U and Zn were carried out with ln-transformed data (Chapter 5). The ln-transformation approximately normalised the distributions of CR values and also linearised the observed nonlinear dependency of CR values on soil concentration. To facilitate the interpretation of the results of the RA, principal component analysis (PCA) for soil properties was carried out. Variance scaling was performed for the variables before PCA to diminish the variability in the data and also to equalise the effect of different variables.

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2 *Soil-to-plant transfer of uranium and its distribution between plant parts in four boreal forest species*

Roivainen P, Makkonen S, Holopainen T and Juutilainen J (2011)

Boreal Environment Research 16: 158–166

3 Transfer of elements relevant to radioactive waste from soil to five boreal plant species

Roivainen P, Makkonen S, Holopainen T and Juutilainen J (2011)

Chemosphere 83: 385–390

4 Soil-to-plant transfer of elements is not linear: results for five elements relevant to radioactive waste in five boreal forest species

Tuovinen TS, Roivainen P, Makkonen S, Kolehmainen M, Holopainen T and Juutilainen J (2011)

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5 Element interactions and soil properties affecting the soil-to-plant transfer of six elements relevant to radioactive waste in boreal forest

Roivainen P, Makkonen S, Holopainen T and Juutilainen J

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6 General discussion

6.1 COMPARISON OF ELEMENT CONCENTRATIONS IN ROOTS AND LEAVES

Root concentrations of Co, Mo, Ni, Pb and U were higher than the corresponding leaf/needle concentrations (Chapters 2 and 3). This was consistent for all elements in all plant species studied, both understory and trees, except for Mo in blueberry. In general, the translocation of elements to the foliage of deciduous rowan seemed to be higher than to the foliage of coniferous Norway spruce and this was statistically significant for Mo and Pb (Chapter 3). Uranium was an exception to this trend, as the root-to-shoot ratio of U was higher in rowan than in Norway spruce (Chapter 2). Fern was the understory species that had the highest root-to-leaf ratios, except for Co (Chapters 2 and 3).

Accumulation in roots has been commonly reported for many elements (Shahandeh and Hossner, 2002; Johnson et al., 2003; Shtangeeva, 2010; Kabata-Pendias, 2011). However, the data on CR values for roots of native plants are limited. This is probably due to the fact that the studies have generally focused on the protection of humans and have therefore addressed mainly the edible parts of plants. Further, sampling above-ground plant parts is less laborious than collecting representative root samples, especially from the fine root fraction.

The results suggest that plant- and site-specific factors affect the translocation of Mo more than that of other elements; the root-to-leaf ratios of Mo differed significantly between all understory species and all trees. The root-to-leaf ratios of Pb varied significantly between the tree species studied and also between Norway spruces growing at different sites. Although the amount of atmospheric Pb has declined because of the use of unleaded petrol, foliar uptake from atmospheric deposition may have affected the results for Pb in addition to the factors affecting root uptake (Hovmand et al., 2009).

Higher root-to-leaf ratios for Norway spruce were consistently found at the site where the element concentration in soil was higher (Chapter 3). The same trend was found in the understory data for Co and Mo (Chapter 3). These results are consistent with an active role of the roots as barriers preventing the translocation of elements. This is an expected finding, as the elements studied are required by plants only in small amounts (Co, Mo, Ni) or not at all (Pb, U) (Taiz and Zeiger, 2006; Kabata-Pendias, 2011). Elements are known to accumulate in roots because of the endodermal Casparian strips, which prevent the free movement of elements in cells (Denny, 2002; Mauseth, 2003, Seregin and Kozhevnikova 2008). Accumulation of cations also occurs due to binding into negatively charged root cell walls and membranes (Meychik and Yermakov, 2001; Seregin and Kozhevnikova 2008). Some fraction of the elements measured might have accumulated in the associated mycorrhizae since mycorrhizal fungi have been found to protect the associated plants by accumulating metals within the fungal component of the mycorrhizal root system (Leyval et al., 1997; Hall, 2002; Denny, 2002).

The possible incomplete cleaning of adhering soil particles from the plant samples was evaluated using CR values for Ti, as plants are assumed to take it up only in very small amounts (Cary et al., 1986; Berrow, 1988). Considerable soil contamination was found in a few cases but generally the effect of soil contamination was considered to be minor (Chapter 3). The effect of soil contamination was strongest for root concentrations so the actual differences between root and leaf concentrations may not be as large as the observed root-to-leaf ratios showed.

The results of the present study suggest that the accumulation of elements in roots should be taken into account when modelling the soil-to-plant transfer of elements, especially when the focus is on the radiation protection of plants. There are also many animals that feed on plant roots so the elements can transfer to animals through that route. The importance of the root fraction has been emphasised also in assessing the risks of inorganic, non-radioactive contaminants (Johnson et al., 2003; Courchesne et al., 2008). Johnson et al. (2003) particularly stressed the importance of fine roots in assessing possible negative belowground impacts of metals. Higher element concentrations in fine roots than in coarse roots of trees were also found in the present study (Chapters 2 and 3).

6.2 COMPARISON OF THE CR VALUES FOR DIFFERENT SPECIES AND SITES

The need for distinct, even site-specific, CR values for different plant species is an important issue in modelling. The ideal situation would be that generic CR values could be used for various plant species and growth sites because this simplifies modelling. Sheppard (2005), Vandenhove et al. (2009) and Higley (2010) have proposed that generic CR values are appropriate substitutes for site-specific data. Sheppard (2005) concluded that site-specific properties usually do not affect the transfer so much that the increased accuracy from using site-specific CRs would override the advantages of using generic data based on higher numbers of observations.

The results of the present study also support the use of generic CR values instead of plant-specific values. The present study included plant species representing different growth traits. The understory species selected were a monocotyledonous herb (May lily), a fern (narrow buckler fern) and a dicotyledonous dwarf shrub (blueberry). The variation in CR values between species was not clearly higher than the within-species variation.

The results of the present study do not unambiguously support the use of different CR values for the deciduous rowan and the coniferous Norway spruce. Although some significant differences between the CR values for these two species were found (e.g. soil-to-leaf/needle CR values of Co and Mo; see Chapter 3), significant differences were also found between the CR values of Norway spruce grown at different sites (e.g. soil-to- needle CR values of Mo and Ni).

The present study included a herb-rich forest site with higher soil fertility and a more barren coniferous site (Table 3). The effect of sampling site was most evident for the CR values of Pb: the values were higher at the herb-rich forest than at the coniferous forest site (Chapters 3 and 5). Significant differences between sites were also found for Ni when CR values for Norway spruce grown at different sites were compared (Chapter 3), and when the non-linear equation was fitted with the data (Chapter 4). In contrast to the values of Pb, the CR values of Ni for Norway spruce were higher at the coniferous site than at the herb-rich site (Chapter 3). The effects of site-specific factors on the soil-to-plant transfer of Co and Mo were smaller, which supports the use of generic CR values.

Large GSDs due to within-species variation are characteristic of CR values (Sheppard et al., 2006). In the present study, the GSDs of CR values calculated individually for one plant species ranged from 1.33 to 5.74 and the range of GSDs of the pooled CR values was not markedly higher (from 1.81 to 5.13) (Chapters 2 and 3). The high within-species variation may have hindered the detection of between-species differences. However, as shown by the large GSDs, the CR values include considerable uncertainty, and the use of specific CR values for different plant species would not essentially reduce this uncertainty.

All the CR values calculated were above 0.001, which Sheppard et al. (2010) suggest as a detection limit for CR values to represent root uptake. The CR values for U were consistent with the values found in the literature (Chapter 2). Smaller numbers of published CR values were found for the other elements studied, and the comparisons are therefore not that extensive. After converting to a fresh weight basis to allow comparison, the values for Pb and Ni in the present study were found to be about ten times lower than those used in the ERICA Tool (Beresford et al., 2008). The CR values in the ERICA Tool are based on studies conducted on 15 native plants in southwestern Canada (Mahon and Mathewes, 1983), on grass in New Mexico, USA (Lapham and Millard, 1989), on agricultural plants in Poland (Pietrzak-Flis and Skowrońska-Smolak, 1995), and on a review of field and greenhouse studies including various plant species and sampling sites (Efroymson et al., 2001). The differences between the CR values in this study and those in the ERICA Tool may have resulted from differences in growth conditions. This is supported by the fact that the CR values for Ni found in the present study were consistent with the values found in southwestern Finland (Aro et al., 2009). However, it should be noted that the values in the ERICA Tool are based on only a few studies (Beresford et al., 2008), and are therefore uncertain. Concentration ratios for Co and Mo have been previously determined by Reimann et al. (2001) within a large area in Northern Europe. In general, the CR values found in the present study for Co were similar to those reported by Reimann et al. (2001), while the values for Mo were higher.

6.3 CONSIDERATIONS RELATED TO THE LINEARITY ASSUMPTION

A lack of linearity between plant and soil concentrations was evident in the data of the present study (Chapter 4). One important finding was that there was no difference between the behaviour of essential and non-essential elements (Chapter 4). This systematic variation in CR values with soil concentration might explain a considerable fraction of the variation in published CR values. The error caused by omitting non-linearity is largest at low soil concentrations, and might lead to the underestimation of plant uptake in radioecological modelling, as discussed in Chapter 4.

Non-linearity has been recognised earlier but its significance in modelling has been questioned since many other factors also affect the CR values (Sheppard and Evenden, 1988; Mortvedt 1994). Sheppard (2005), for example, concluded that the soil concentration is just another source of variation in CR values and can be taken into account as a site-specific factor. Although different approaches, e.g. a curvilinear function (Simon and Ibrahim, 1987) and a Freundlich-type function (Krauss et al., 2002), have been proposed for describing soil-to-plant transfer, there have not been many serious attempts to handle non-linearity in modelling. McGee et al. (1996) strongly criticised the use of ratios of any kind but did not propose any alternative approaches.

Including non-linearity in modelling is not a trivial task. Using Langmuir-type equations (Chapter 4) would not be essentially more difficult than using constant CR values, but it is still unclear how the parameters for the equation should be created so that they could be generalised to different sites. As with the use of traditional CR values, it is not realistic to assume that extensive site-specific measurements would be done for every assessment where the equation is applied.

However, it will certainly be useful to build a model based on the Langmuir-type equation to find out how much the predicted plant concentrations and the associated uncertainty will change in comparison with the traditional linear approach. It will also be important to fit the equation with other data-sets to obtain data of the variation of the equation parameters at different sites and in different plant species.

6.4 EFFECTS OF SOIL PROPERTIES AND INTERACTING ELEMENTS ON CR VALUES

The general linear model (GLM) (Chapters 2 and 3) and regression analysis (RA) (Chapter 5) gave somewhat different results with respect to the effects of soil properties on CR values (Table 4). In general, the results of the RA can be considered to be more reliable, since the soil concentration of the element studied was included in the analysis and the observed non-linearity of element uptake (Chapter 4) was thereby taken into account. The results of the RA were also based on higher numbers of observations, as the data from two sampling sites were pooled. Plant species and sampling site were included as explanatory variables in the RA which reduced the variation caused by pooling. However, the correlations between explanatory variables could have affected the results of the RA even though sensitivity analysis using the results of principal component analysis was carried out. In particular, the effect of

OM on the CR values of Mo and Pb seemed to be confounded by soil S concentration (Chapter 5). The negative effect of soil OM content on the CR values of Pb found by GLM is more consistent with existing knowledge (Sheppard and Sheppard, 1991; Kabata-Pendias, 2011) than is to the positive effect found by the RA.

Table 4. *The soil properties (pH, organic matter (OM) content, clay content, silt content) found to have a significant positive or negative effect on the soil-to-plant transfer of Co, Mo, Ni, U and Zn in the general linear model (GLM) or regression analysis (RA).*

Element	Positive effect	Negative effect
Co	silt (GLM)	
Mo	OM (RA)	pH (GLM)
	silt (GLM)	clay (GLM)
Ni		clay (GLM)
Pb	OM (RA)	OM (GLM)
	clay (RA)	pH (GLM)
U		pH (GLM)
		clay (RA)
Zn	n.s.	n.s.

n.s. = no significant effects found

A significant effect of soil pH on the CR values of Mo, Pb and U was found by GLM but not by RA, which is also a clear difference between these two methods. Soil pH is commonly described as one of the most significant soil properties affecting the behaviour of elements (Efroymson et al., 2001; Koch-Steindl and Pröhl, 2001; Kabata-Pendias, 2011). However, the soils used in this study represented quite a narrow range of pH values (3.7–5.1), which may have limited the effect of pH. It is also possible that pooling the data from two sites for RA have had a contributing factor. Also, soil Mg concentration was included as a variable in the RA but not in the GLM. This may have affected the results, as soil Mg concentration is associated with soil pH (Chapter 5).

Inconsistency between the two analysis methods was also seen in the effects of interacting elements. When the effects of Ca, P, K and S on the CR values of Co, Mo, Ni and Pb were studied using the GLM, no significant effects were found (Chapter 3). However, the RA showed that there were significant effects of these elements, especially in the case of P and S (Chapter 5). Again, the results of the RA can be considered to be more reliable because of the above-mentioned reasons.

The literature contains very limited information about the effects of soil properties and other elements on soil-to-plant transfer in forest ecosystems. One important reason for this is that soil properties are not always measured when CR values are investigated (Vandenhove et al., 2009). Furthermore, most data are from studies in laboratory settings and agricultural plant species.

The complexity of models is always a compromise and including many parameters explaining the soil-to-plant transfer contradicts the principle of simplicity (Kirchner and Steiner, 2008). From this point of view, it is worth noting that quite a few among the 23 elements studied affected the soil-to-plant transfer of other elements (Chapter 5). In general, the elements which had major effects were plant nutrients (K, Mg, Mn, P and S). This is consistent with existing knowledge (Ehlken and Kirchner, 2002; Kabata-Pendias, 2011). Cd was an exception and its effect was probably related to competition in the uptake of divalent cations, as the effect was most evident for the soil-to-plant transfer of Pb and Zn (Chapter 5). Thus, radioecological models might be improved by including only a few affecting elements. In practise, this could be done using probability distributions of soil concentrations of the elements of interest. The fact that these elements are nutrients simplifies this task as investigations on the concentrations of these elements in soils are numerous.

6.5 BIOAVAILABILITY

Total element concentration in soil is still generally used for deriving CR values, although it is commonly accepted that the uptake in plants more directly depends on bioavailable concentration (Ehlken and Kirchner, 2002; Chojnacka et al., 2005;

Blanco Rodríguez et al., 2006). One reason for this is that, – due to the wide range of soil- and plant-related factors involved, - there is no consensus on how the bioavailable fraction should be measured (Kennedy et al., 1997; Ehlken and Kirchner, 2002; Kabata-Pendias, 2004).

In the present study, leach with 1 M ammonium acetate (NH_4Ac) buffered at pH 4.5 was used as an estimate of the mobile fraction of elements in soil. This leach extracts elements chemically adsorbed to the soil (Räisänen et al., 1997) and can be used to simulate metal behaviour near the roots (Schultz et al., 2004). The mobile concentrations measured by this method were generally 1–50 % of the pseudo total concentration in soil (Chapters 2 and 3). The exceptions were U, with mobile fractions up to 90 % of the total concentration, and Mo, for which the mobile fraction was in most cases under the detection limit.

The results of the present study do not unambiguously support the use of mobile fraction analysed after NH_4Ac leach as the substrate concentration when assessing soil-to-plant transfer. Co and U were the only elements studied for which the plant concentrations correlated significantly better with the mobile fraction than with the total concentration (Chapters 2 and 3). Plant uptake was similarly non-linear regardless of the measure of soil concentration (Chapter 4). The soil-related factors that were found to affect plant uptake were mostly the same in analyses based on soil total and soil mobile concentration (Chapter 5).

Measuring Kd values to assess element sorption by soil particles would have revealed more of the bioavailability of elements, and it is advisable in future investigations. There is usually a negative correlation between Kd and CR values (Watmough et al., 2005; Vandenhove and Van Hees, 2007), indicating that high solubility in soil water (low Kd) is associated with greater potential for plant uptake.

It is also generally accepted that the rhizosphere affects the bioavailability of elements (Ehlken and Kirchner, 2002; Courchesne et al., 2008). The effect of plant-mycorrhizae associations (Ehlken and Kirchner, 2002; Courchesne et al., 2008) has probably influenced the availability of elements also in the present study. The effects of these factors could be considered in more detail in future studies in order to facitiliate more accurate interpretation of soil-to-plant transfer of elements, although it would be extremely difficult to include these factors in radioecological models.

6.6 CONCLUSIONS

The present study produced CR values describing the soil-to-plant transfer of elements relevant to radioactive waste (Co, Mo, Ni, Pb and U) in boreal forests. The values for Co, Mo and Ni are particularly valuable, as existing data for these elements are quite limited for any environment. Although more data are generally available for Pb and U, the CR values produced in the present study allowed comparison with existing data collected in more temperate conditions.

The results of the present study suggest that the CR values for three understory species used (May lily, narrow buckler fern, blueberry) can be pooled to obtain a generic CR value to represent the soil-to-plant transfer in boreal conditions. The same can be done for the CR values of trees (Norway spruce and rowan). Site-specific factors affected especially the transfer of Pb and Ni. Hence, special attention should be given to the selection of suitable CR values for these elements when modelling their transfer. The CR values found in the present study for Pb and Ni were generally lower than previously published values , which mainly represent temperate environments. The CR values of U suggest very low soil-to-plant transfer and are consistent with the values found in the literature. The transfer of Co is consistent with the few observations available in the literature, while the transfer of Mo is higher.

The importance of the root fraction of plants was emphasised as the elements studied tended to bind there. Root fraction and above-ground plant parts should be considered separately when conducting risk assessments focusing on the safety of plants and animals.

The results of the present study do not invalidate the common use of CR values based on total soil concentration in modelling. The CR values based on mobile soil concentration (measured after NH_4Ac leach) were not markedly better in describing the soil-to-plant transfer than those based on pseudo total concentrations (measured after HNO_3 digestion).

The results suggest that the accuracy of radioecological modelling could be improved by using non-linear models to describe the soil-to-plant transfer instead of traditional CR values which assume a linear relationship between plant and soil concentrations. A non-linear function based on the Langmuir equation was suitable for describing the transfer, which allows the development of non-linear models and comparison of their predictions with those of the traditional linear models.

The predictive power of radioecological models might also be enhanced by including the effects of soil properties and interacting elements in soils. Plant nutrients, namely K, Mg, Mn, P and S, were found to affect the transfer of Co, Mo, Ni, Pb, U and Zn. As data on the concentrations of these nutrients are available for many soils, it is worthwhile investigating whether these elements can be included in radioecological models without making them too complex.

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Päivi Roivainen

Characteristics of Soil-to-Plant Transfer of Elements Relevant to Radioactive Waste in Boreal Forest

This thesis reports on the soil-to-plant transfer of a series of elements relevant to radioactive waste in boreal forests. This data are needed for ecological risk assessments of radioactive waste disposal. The thesis presents results from a field study conducted at two forest sites in Eastern Finland. This study produced parameters for modelling the transfer of elements to typical boreal forest plant species, and increased the general understanding of processes underlying the soil-to-plant transfer.

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