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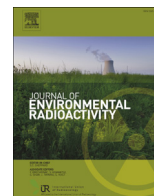
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Approaches to modelling radioactive contaminations in forests – Overview and guidance



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ABSTRACT

Modelling the radionuclide cycle in forests is important in case of contamination due to acute or chronic releases to the atmosphere and from underground waste repositories. This article describes the most important aspects to consider in forest model development. It intends to give an overview of the modelling approaches available and to provide guidance on how to address the quantification of radionuclide transport in forests. Furthermore, the most important gaps in modelling the radionuclide cycle in forests are discussed and suggestions are presented to address the variability of forest sites.

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1. Introduction

Contamination of forests can occur following a nuclear accident, as it happened in Chernobyl in 1986 and in Fukushima in 2011, where forests covered most of the contaminated terrestrial areas (e.g. Hashimoto et al., 2012; Kashparov et al., 2012; Yoschenko et al., 2017). Contamination may also originate from nuclear waste in host rocks at disposal sites or mining activities. Contamination events could lead to radionuclide migration in the soil column and accumulation of radionuclides in surface vegetation and local food sources (Goor and Thiry, 2004; Thiry et al., 2009). Increased external radiological exposure can affect humans who spend time in forests, such as hunters, foresters, mushroom and berry pickers. Internal exposure may result from ingestion of contaminated mushrooms or wild berries (Calmon et al., 2009; Carini, 1999; Gwynn et al., 2013; Steiner et al., 2000, 2002) and game, for example wild boar (Hartmann et al., 2016) or deer.

Forests provide wood for different domestic and industrial purposes, such as biofuel for heat and power plants, paper production, building material and firewood for private homes. The use of contaminated wood and the resulting residues may increase human radiation dose via external exposure and inhalation (Charro et al., 2013; Hubbard, 1997). Secondary contamination several years after the accidental release of radionuclides can be relevant in case of forest fires, which increase the inhalation dose of the exposed population (Yoschenko et al., 2006; Zhou et al., 2016). Secondary contamination may also result from flooding events, e.g. after the Chernobyl accident, when flooding led to the transport of the radionuclides from soil to surface waters (Laptev and Voitsekhovich, 1993).

Modelling the radionuclide circulation in a forest is challenging, because forests are by far more spatially heterogeneous than agricultural lands. Vegetation and soil have both a layered structure. The local soil profile and the contamination levels of the soil horizons can vary at a small scale, depending on the characteristics of the above ground biomass (e.g. type and density of trees) and the proximity to tree stems.

This article summarises the Forest Modelling Handbook which was developed by the Forest Modelling Group working under the EC FP7 project COMET (<http://www.radioecology-exchange.org/content/comet>). It gives an overview about forest modelling approaches and provides guidance for calculating the radionuclide fluxes between the different forest compartments. It is intended for modellers, experimentalists, experts of national authorities and radiation protection advisers.

2. Fundamentals of forest modelling

2.1. Pathways of radionuclides in forests

For modelling the fate of radionuclides in forest ecosystems, the source term must be known. A compilation of typical radionuclide releases from various sources to the atmosphere can be found in UNSCEAR (2000). After the atmospheric transport, radionuclides enter the forest via dry and wet deposition. They are partially intercepted by the canopy (Fig. 1). The amount of intercepted radionuclides depends on physico-chemical forms of the deposited radionuclides, the meteorological conditions (e.g. dry or wet deposition), forest type, tree population density and season (Shaw, 2007). Radionuclides deposited onto vegetation are partially taken up through stomata and cuticle. Then they are translocated from needles and leaves into branches and stems (Fig. 1). The period of 3–5 years after the deposition is characterised by a redistribution of the initial deposits through weathering of radionuclides from the canopy via throughfall, stemflow, litterfall and radionuclide

migration in soil. After the initial rapid infiltration, radionuclides are partially fixed and immobilised through fungal or microbial activity or mineral constituents of forest soil. Correspondingly, the rate of downward migration is considerably reduced. In the organic horizons, this is determined mainly by the decomposition and litter accumulation rates. The downward migration of radionuclides is partially compensated by upward translocation by roots and, notably in the case of radiocaesium, by fungal mycelia (Rafferty et al., 2000). Fungal and microbiological activity are likely to contribute substantially to the long-term retention of radionuclides in organic layers of forest soil (Fig. 1). Bioturbation may also affect the transport of radionuclides in forest soil (Bunzl, 2002). Sorption and complexation with organic and mineral components within the soil are also relevant processes (Berkowitz et al., 2014). In the long-term, root uptake via symbiotic fungi (Smith and Read, 1997) is the dominating factor regarding tree contamination and the local soil is the major radionuclide reservoir (Calmon et al., 2009).

Another potential source of contamination could arise from a nuclear waste repository in host rocks. In case of groundwater infiltration into the nuclear waste followed by its dissolution, the contaminant transport is driven mainly by advection along the hydraulic gradient. In host rocks with a low permeability like clay rocks, the contaminant transport is often dominated by diffusion and molecular dispersion processes (GRS, 2007; IAEA, 2011). When contaminated groundwater has reached the root zone of soil (e.g. via capillary rise), radionuclides can be taken up by plants through the interaction with fungal mycelia and directly from the contaminated soil solution. Gaseous transport can lead to inhalation of radioactive pollutants or, in the case of $^{14}\text{CO}_2$, to photosynthetic uptake by plants (Berkowitz et al., 2014).

2.2. How to design a forest model

A radioecological model for forests should be designed in a way that it is as simple as possible but fit for purpose. The purpose of the model and the desired endpoints should be identified at first. Typically, radioecological forest models are used to quantify one or several of the following endpoints:

1. Time-dependent activity concentrations in the affected ecosystem
2. Time-dependent ambient dose rates in the affected ecosystem
3. Doses to humans (equivalent dose to tissues and organs and/or effective dose)
4. Doses to biota (weighted absorbed dose rates)

In the two latter cases, the model should provide conservative dose estimates (e.g. for demonstrating compliance with dose limits), high percentiles of the endpoints (e.g. 95th percentile of dose to a representative person) or the best estimate of human or biota radiation exposure.

A structured approach for the development of radioecological models was published by IAEA (Biomass, 2003). It consists of three major steps:

1. Compilation of a list of Features, Events and Processes (FEPs list).
2. Development of an Interaction Matrix, representing the compartments of the ecosystem to be modelled and the dominant processes.
3. Implementation of the conceptual model, i.e. the Interaction Matrix, into mathematical models.

The FEPs list is a document that compiles all features, events and processes which could be of relevance for the fate, transport and distribution of radionuclides in an ecosystem. The FEPs list helps to

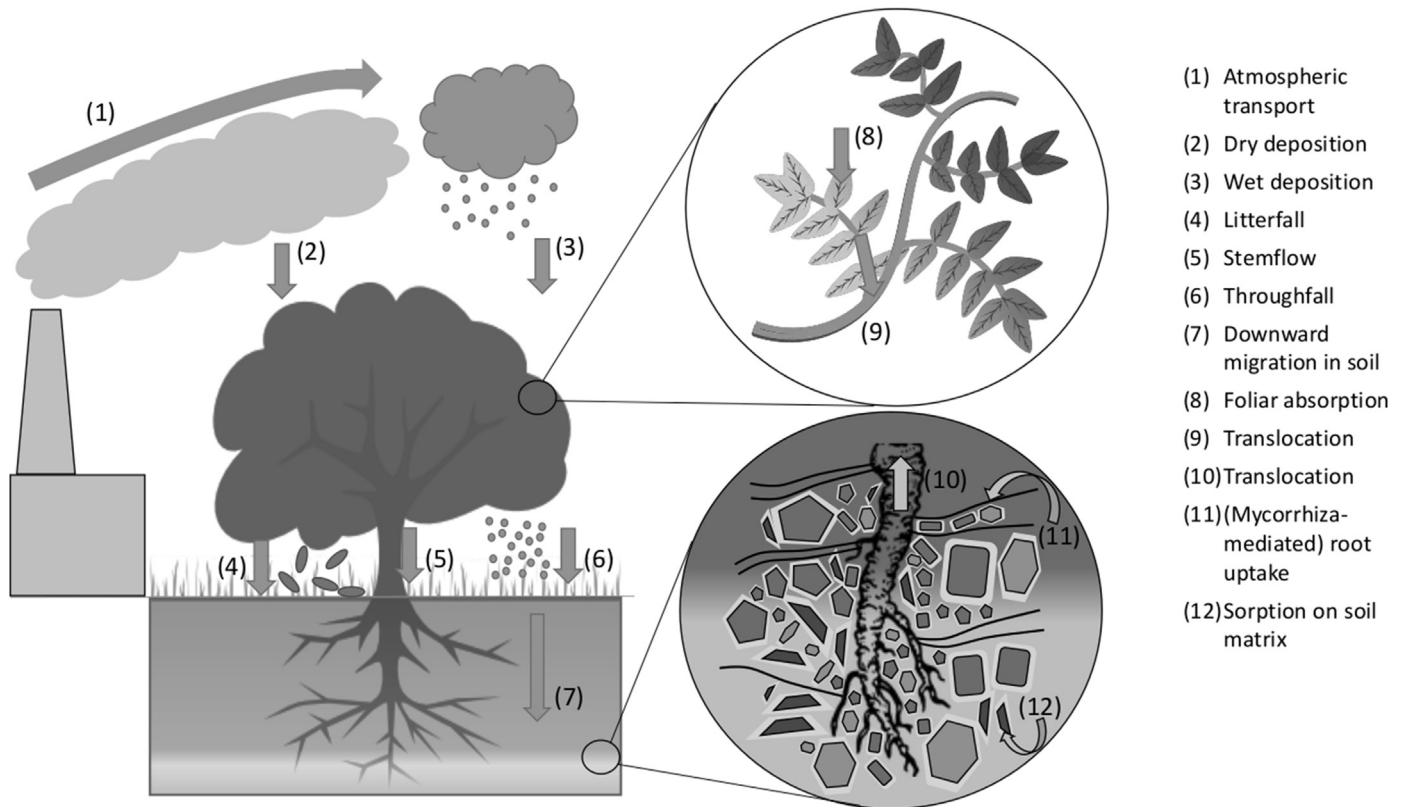


Fig. 1. Overview of the most important processes regarding the radionuclide cycle in forests.

ensure that all important features, events and processes are taken into account in a radioecological model. It also provides a basis for further model development or data acquisition (SKB, 2015). The compilation of FEPs lists has become a common approach in radioecology. In various international projects like IAEA BIOMASS (Biomass, 2003), BIOPROTA (<http://www.bioprot.org/>), and the STAR project (<http://www.radioecology-exchange.org/>), a FEPs list has been developed based on safety studies of nuclear waste repositories and joint efforts of international experts (NWMO, 2011; OECD, 2006; Posiva, 2012; SKB, 2010).

The Interaction Matrix represents the compartments of the ecosystem to be modelled and the radionuclide fluxes between them in a tabular form (e.g. Avila and Moberg, 1999, Fig. 2). The diagonal elements contain the compartments of the ecosystem, while the processes determining the radionuclide fluxes between these compartments are listed in the off-diagonal elements of the Interaction Matrix. The number of diagonal elements, i.e. the size of the Interaction Matrix, expresses the degree of complexity of the conceptual model. An Interaction Matrix is considered complete if each significant interaction between the compartments relevant to the model's purpose is included.

Based on the Interaction Matrix, which represents the conceptual radioecological model, the mathematical implementation is developed. Here, the processes determining the radionuclide fluxes should be mathematically represented with adequate (fit-for-purpose) complexity. One measure for complexity is the number of independent model parameters, which should be as small as possible (Kirchner and Steiner, 2008), in order to keep the model uncertainty reasonably low.

These three 'design' steps are followed by the classical process of model development: coding, parameterisation (attributing values to the model parameters), verification (verifying that the model's

equations are correctly implemented), calibration (varying model parameters within predefined ranges to improve the accuracy of the model) and validation (testing whether model predictions are close to independently measured values).

2.3. Empirical and process-based modelling approaches

There are two different approaches in forest modelling to describe relevant processes, next to hybrid models which consist of both. One is the use of empirical relations, such as aggregated transfer factors (T_{agg}) (Dvornik and Zhuchenko, 1999; Howard et al., 1995; IAEA, 2010) or the solid-liquid partition coefficient (K_d) (IAEA, 2010; SKB, 2009; Whicker et al., 2007). Empirical relations are based on measured data, obtained in real ecosystems or laboratory experiments. They are a simplified quantification of transport and transfer processes and comprise in an aggregated form different physical, chemical or biological processes. The main disadvantage with empirically obtained model parameters is that they should be applied to other forest sites with caution. For example, measured values of the partition coefficient K_d often range between 5 and 6 orders of magnitude (IAEA, 2010). This high variation makes it difficult to choose an appropriate value for a specific study area and hampers model transferability to other areas. The aggregated transfer factors have the main disadvantages that they describe the transfer of radionuclides from soil to plants without temporal considerations and assume equilibrium conditions.

Although empirical approaches to radioecological modelling have disadvantages, their application may be justified if so many complex processes are important that a mechanistic model would lead to excessive model complexity. In this case, many uncertain or sometimes even unavailable model parameters may be involved,

Atmosphere	interception rainfall snowfall	interception rainfall snowfall	interception rainfall snowfall	inhalation interception
transpiration burning	Tree translocation	leaves fall weathering interception fertilisation	weathering interception mycorrhizae	ingestion
resuspension	root uptake rainsplash	Soil/litter percolation diffus./adv. litter decomp. sorption/ de- sorption	uptake rainsplash	ingestion
transpiration burning	root uptake (mycorrhizae)	leaves fall weathering interception fertilisation	Understorey translocation root uptake (mycorrhizae)	ingestion
	consumption	fertilisation	consumption	Wild animals internal transfer

Fig. 2. Interaction matrix describing the long-term migration of ^{137}Cs in a forest ecosystem (Avila and Moberg, 1999).

leading to an unacceptably high forecast uncertainty. An example of a state-of-the-art empirical forest model has been published by Calmon et al. (2015), who study the short-term redistribution of radiocaesium deposited onto coniferous forests after the Chernobyl and Fukushima accidents. Key transfer processes, e.g. interception, dry deposition and litterfall, are represented by simplified empirical relations that require only nine generic and five site-specific parameters. This model satisfactorily reproduces the initial interception and the canopy depuration kinetics at two forest sites affected by the Chernobyl accident and the Fukushima event, respectively.

Process-based or mechanistic models are built upon the detailed understanding of key processes in an ecosystem and their explicit mathematical representation. Process-based models have a potentially higher forecasting power and allow for studying the relevance of a specific process (process sensitivity). An example of a process-based forest model has been published by Vives i Batlle et al. (2014), who investigated the cycling of ^{36}Cl in a Belgian Scots pine forest. Their model requires 66 parameters, representing an array of different physical properties such as, for example, forest

stand density, hydraulic properties of the soil or characteristics of the vegetation.

Depending on the purpose of the model and the uncertainty related to the source term, different types of models (empirical, process-based, hybrid) are suitable. For example, a highly uncertain source term will dominate the overall uncertainty of a radioecological model. Correspondingly, a rather simple model will be sufficient.

In general, there is variability between the different forest sites and within an examined forest. The high variability of forest ecosystems within a study area represents a specific challenge for forest models (section 3.2). Variability, i.e. the true spatial and temporal heterogeneity of activity levels, can be characterised and quantified but it cannot be reduced. Available empirical and mechanistic models rarely consider the variability in forest ecosystems. Most radioecological models provide spatially averaged quantities, which do not reproduce highly variable contamination levels. In addition, the seasonal variation might be relevant too.

2.4. Submodels for the radionuclide transfer between forest compartments

2.4.1. Atmospheric transport of radionuclides

To quantify the activity concentration (Bq m^{-3}) in air, atmospheric dispersion models are used. Several atmospheric dispersion models have been developed to take into account different types of sources (point, area, volume), release characteristics (continuous, short-term), different orographic and topographic peculiarities, and different meteorological conditions. These models can broadly be classified as (Gaussian) plume models, (Gaussian) puff models and advanced models, such as the Lagrangian particle models (Holmes and Morawska, 2006; Lutman et al., 2004; Raza and Avila, 2001).

In addition to atmospheric transport, it is necessary to describe the meteorological characteristics influencing the radionuclide uptake by vegetation in forest models. This includes the prevailing conditions of the atmosphere (temperature, pressure, humidity, airborne particulate characteristics, etc.). Some of this information is also required to calculate evapotranspiration, which is the driving force for water (and hence radionuclide) uptake and transport within the vegetation.

2.4.2. Interception of dry deposited radionuclides

Dry deposition is a process of radionuclide transport from the atmosphere onto forest vegetation and ground. Dry deposition is the absorption of gases and particles by surfaces and media in the environment such as vegetation, soil, surface waters or snow. Dry deposition of noble gases such as krypton, xenon or radon, is negligible, since they do not interact with surfaces, but the airborne radon aerosol daughters enter plant leaves to give an internal dose contribution (Vives i Batlle et al., 2011). Reactive gases, such as I_2 or CO_2 , may enter plants through the stomata and undergo chemical reactions with specific compounds in the plant.

The deposition velocity v_g (m s^{-1}) is defined as the ratio between the activity deposited on a specific surface (Bq m^{-2}) and the time-integrated activity concentration of a radionuclide in ground-level air (Bq s m^{-3}). The amount of deposited activity depends on properties of the biosphere, such as surface roughness or canopy structure (Calmon et al., 2015; Pröhl, 2009). The deposition velocity can be determined experimentally. It depends on several factors such as particle size, meteorological conditions, characteristics of the surface-air interface and the chemical form of the deposited contaminant (Sehmel, 1980). Determining an appropriate value for the deposition velocity is difficult and can be a major source of uncertainty. To keep the uncertainty of the deposition velocity acceptably low, information on the size distribution of particulate aerosols is essential (Petroff et al., 2008; Pröhl, 2003).

In forest ecosystems, the aggregated deposition velocity is the sum of the deposition velocities of the canopy, tree trunks and soil. Typical estimates of deposition velocities for particles with diameters between 0.1 and 1 μm are 0.0008–0.0025 m s^{-1} for soils, 0.001 m s^{-1} for grass, 0.0005 m s^{-1} for tree trunks and 0.005 m s^{-1} for the crown with mature leaves (Pröhl et al., 1995).

2.4.3. Interception of wet deposited radionuclides

Wet deposition is the transport of radionuclides from the atmosphere to vegetation and soil by precipitation (wash-out). Part of the radionuclides deposited with precipitation is retained by vegetation and the remaining contamination reaches the forest soil. Although the activity retained by vegetation is weathered off to soil with time, interception affects the dynamics of radionuclides significantly. Radionuclides can be rapidly taken up by plant leaves, whereas root uptake from forest soil is considerably slower.

Müller and Pröhl (1993) modelled the interception with an

equation, which consists of five key parameters. The parameter f_w is the interception fraction (Gonze and Sy, 2016; Pröhl, 2009), LAI is the Leaf Area Index, k is an element-specific factor describing the ability of the element to be fixed on the leaves or needles, S is the water storage capacity of the plant surface (mm) and R is the amount of rainfall of the precipitation event considered (mm). Since the plant surface is negatively charged, the absorption of anions is less effective than that of cations (Hoffman et al., 1995; Kinnersley et al., 1997).

This model describes single deposition events. For routine releases, the mean intercepted fraction can be estimated, taking mean values for the leaf area index and the amount of rainfall per rainfall event. For this purpose, a mean rainfall per precipitation event of 5 mm is assumed. This is a typical value for temperate climates with an annual precipitation of 500–1000 mm and approximately 10 days per month with precipitation above 1 mm. For a pine tree with LAI equal to 10 and S with a value of 0.25 mm, an interception fraction of 0.49 is calculated for caesium (Calmon et al., 2015). The model is an example of a semi-mechanistic parameterisation of wet interception, because it includes three parameters of physical or biological relevance (LAI, S and R) but is still based on an empirical functional term and a purely empirical coefficient. In addition, there are some intermediate situations like fog or snow deposition, which require a specific modelling approach (Bernauer, 2015; Katata et al., 2015).

2.4.4. Tree depuration

After deposition, radionuclides are successively removed from the canopy by rain, wind and litterfall. Activity levels further decrease due to biomass growth. Since tree physiology and the meteorological conditions are subject to seasonal variations, the total decrease rate also depends on the season. The total loss is expressed by the weathering half-time T_w , which is empirically derived from the temporal evolution of the radionuclide concentrations in the aerial parts of vegetation, assuming an exponential decrease. In general, higher values of T_w are observed for slowly growing or dormant vegetation (Kirchner, 1994; Miller and Hoffman, 1983). Field experiments with litterfall samplers and water collectors allow to discriminate the respective contributions of throughfall, stem flow and litterfall to the total depuration fluxes.

Data on radiocaesium demonstrates that the evolution of the residual activity in the canopy can be described best by a double exponential function. Experimental investigations on artificially contaminated spruce over one year revealed a short T_w of the order of one week and a longer T_w of the order of several months. During the second year, the weathering rate is further reduced (Bunzl et al., 1989; Sombré et al., 1994). In the case of Fukushima accident, Gonze and Calmon (2017) demonstrated that the depuration kinetics in evergreen coniferous forests followed roughly two distinct characteristic half-lives (about 1 month and about 10 months, respectively). These values are in the range of those observed in coniferous forests contaminated by the Kyshtym and Chernobyl accidents (Bunzl et al., 1989; Tikhomirov and Shcheglov, 1991, 1994). The successive removal of radionuclides from the canopy influences their distribution pattern in forest soil. Throughfall generally leads to a more homogeneous contamination of forest soil underneath the canopy, whereas stem flow affects only a small area close to the tree (Guillitte et al., 1990).

2.4.5. Translocation

Translocation is the transport of radionuclides within the plant subsequent to foliar absorption or root uptake. This process influences the distribution of radionuclides between different parts of the plants, which could be used as food or, in case of wood, for different domestic products (Thiry et al., 2016). Translocation in

trees following root uptake and foliar absorption can be quantified by means of translocation factors, i.e. the ratio between the activity concentration of a radionuclide in the peripheral parts of the plant (leaves, needles or the whole crown layer) and the activity concentration in trunk wood (Römmelt et al., 1990; Soukhova et al., 2003). Other authors, e.g. Barci-Funel et al. (1995) and NRC (2014), relate the translocation factor to the contamination of newly grown trunk wood due to root uptake, which is estimated for the annual wood growth increment. This activity can be added to the radionuclide pool from past uptake. Calmon et al. (2015) use a translocation parameter, which relates the resorbed activity of radiocaesium to the total amount of caesium deposited onto leaves. A more process-based representation of translocation is to consider the water and element transport, which is mainly based on (a) ascent of xylem across a hydraulic potential gradient, and (b) descent of phloem along an osmotic pressure gradient which depends on the loading and unloading of plant sugars. A more process-based representation should also include the passive (diffusion) or active (transport-driven) transfer of radionuclides through root membranes.

2.4.6. Migration in soil

Vertical migration of radionuclides in forest soil is often modelled using compartment models (Mamikhin, 1995; Rühm et al., 1996; Schell et al., 1996a, 1996b). For this purpose, forest soil is subdivided into soil layers and the net transport of radionuclides between different layers is described by various means. The simpler of these means is rate coefficients or ecological half-lives attributed to specific soil layers. This approach is adequate to predict the time-dependent radionuclide concentration in different horizons of forest soil, provided that the soil layers are defined according to the natural horizontation (Rühm et al., 1996). In fact, the rate of downward migration in the organic horizons is determined mainly by the rates of decomposition and litter accumulation in the specific horizons. It is partially compensated by an upward translocation in roots and, notably in the case of radiocaesium, in fungal mycelia (Rafferty et al., 2000). Radionuclides may be available for root uptake via symbiotic fungi, but are supposed to be hardly leached to deeper soil layers (Smith and Read, 1997). The compartmental description of radionuclide transport in forest soil does not account for processes on a microscopic level and has to be calibrated for each site individually.

Other approaches to modelling the vertical migration of radionuclides in forest soil explicitly consider advection, diffusion (i.e. mechanical dispersion, molecular diffusion) and absorption/desorption processes (Konshin, 1992a, 1992b; Kurikami et al., 2017; Mishra et al., 2016; Olondo et al., 2017). Furthermore, intermediate approaches exist that rely on a multi-layered compartmental assumption (neglecting diffusion and not solving for the hydrological balance) but explicitly account for advection and other biophysical-chemical processes (Ota et al., 2016). The transport of radionuclides via the soil solution, however, is of minor importance in the organic horizons of forest soil compared to the processes explained above. In its fullest extent, the problem is treated by numerically solving the Richard's equation for transport of water across the soil, coupled with the transport (advection and dispersion) equation for the solutes. This approach necessitates quite an advanced degree of knowledge regarding the hydrological properties of the different soil horizons and the geochemical reactions involved in radionuclide migration through soil. The key parameters in this respect are the soil hydraulic conductivity and K_d , which is not a constant but a function of the soil physico-chemical properties (e.g. soil type, organic matter content, porosity, etc.).

2.4.7. Root uptake from soil

Forest soil may be contaminated through atmospheric deposition following accidental releases of radionuclides, input of radionuclides from nuclear waste repositories (IAEA, 2011) or mining activities. In view of the limited knowledge about the mechanisms and processes involved in the mobilisation, translocation and uptake of radionuclides, empirical transfer factors and concentration ratios are usually used to quantify the transfer of radionuclides from forest soil to vegetation. Common definitions of these parameters relate to the total inventory in soil per area or the activity concentration within the uppermost soil layer of a standardised thickness.

Soil-plant concentration ratios are defined as the ratio of the activity concentration in plant (Bq kg^{-1} fresh or dry weight) and the activity concentration in the top soil layer (Bq kg^{-1} dry weight). This concept has been introduced for arable land, where radionuclides are homogeneously distributed within the rooting zone. Undisturbed forest soils, however, have a multi-layered structure and show a distinct vertical profile of activity concentration (Tuovinen et al., 2016). In order to address this fact in a simplified way, aggregated transfer factors have been suggested, relating the activity levels in vegetation (Bq kg^{-1} fresh or dry weight) to the total inventory in soil per area (Bq m^{-2}). The main disadvantage of the traditional empirical approaches is their large uncertainty and site-specificity. Aggregated transfer factors for radiocaesium and mushrooms, for example, vary by more than four orders of magnitude (IAEA, 2010).

The suitability of aggregated transfer factors depends on the purpose of the radioecological assessment. In the case of an accidental release of radionuclides, when activity levels have to be estimated very quickly, aggregated transfer factors can be a good choice to assess the contamination levels in plants, mushrooms and game, if radionuclide fluxes have stabilised (Calmon et al., 2009). A recent modelling approach using non-linear equations to describe the local soil-to-plant transfer showed encouraging results (Tuovinen et al., 2016). The time-dependent contamination of mushrooms has successfully been quantified using modified concentration ratios, explicitly referring to the soil layer exploited by the fungal mycelium (Rühm et al., 1996; Steiner et al., 2002).

2.4.8. Radionuclide uptake by animals

Contamination of forest animals is often modelled using concentration ratios or aggregated transfer factors. The ERICA Tool (Larsson, 2008), for example, applies concentration ratios to assess doses to wildlife. The element-specific concentration ratio is defined as the ratio of the activity concentration in an organism (Bq kg^{-1} fresh weight) to that in an environmental medium (Bq kg^{-1} fresh or dry weight; Bq L^{-1}). Implicitly it is assumed that the transfer processes between medium and organism are at equilibrium. If only the soil inventory of a radionuclide is known but not its activity concentration, aggregated transfer factors can be used (Johanson and Bergström, 1994). Aggregated transfer factors are defined as the ratio of the activity concentration in an organism (Bq kg^{-1} fresh weight) to the area-related soil inventory (Bq m^{-2}).

2.4.9. Examples of innovative modelling approaches

Most innovative modelling approaches can be grouped into two categories. Either traditional empirical approaches are modified to consider basic ecological and biological facts or key processes are explicitly modelled. An example of the first category is the Wood Immobilisation Potential (WIP). It has been developed to estimate the contamination of trunk wood, taking into account the absorption of radionuclides in newly grown wood and the observed radial transport of radionuclides between the annual growth rings. The WIP was suggested to quantify the radionuclide flux from soil to

trunk wood. It is defined as the linear extrapolation of the cumulated ^{137}Cs activity in trunk wood as a function of the cumulated trunk volume (BIOMASS, 2002; Thiry et al., 2000). It is considered to provide a suitable estimate of annual cation fluxes to trunk wood via root uptake more than a decade after the deposition event.

An example of the second category is the process-based modelling of the radiocaesium contamination of wild boar. These animals may show extremely variable contamination levels that cannot satisfyingly be quantified using aggregated transfer factors. The observed contamination pattern can be attributed to the irregular consumption of highly contaminated deer truffles (*Elaphomyces granulatus*) by the animals (Hohmann and Huckschlag, 2005). If the stochastic nature of the radiocaesium intake via deer truffles is taken into account, the activity A of ^{137}Cs in wild boar and its variability can be reproduced using a one-compartment model (Hartmann et al., 2016). The parameters used are the activity $A(t)$ of ^{137}Cs in wild boar (Bq), f is the fraction of ^{137}Cs absorbed in the gastro-intestinal tract, $I(t)$ is the intake rate of ^{137}Cs (Bq d^{-1}), λ_{bio} is the mass-dependent excretion rate of ^{137}Cs (d^{-1}) and λ_{phys} is the rate constant for the physical decay of ^{137}Cs (d^{-1}). The time-dependent intake rate of ^{137}Cs is modelled as the sum of stochastic intake rate of deer truffles and the contribution of all other food items. The predicted distribution of radiocaesium levels in wild boar agrees well with the measured values (Hartmann et al., 2016).

3. Specific challenges of forest modelling

3.1. Current knowledge gaps about the radionuclide cycle in forests

Many processes that determine the radionuclide cycling in forests are not yet fully understood and thus cannot be integrated into a generally applicable forest model. Fungi, for example, play a key role for mobilisation, uptake and translocation of radionuclides. They are the primary sources of enzymes necessary to degrade the litter, can directly bind or precipitate radionuclides and thus affect radionuclide speciation and mobility in forest soils. Experimental results for forest soils revealed that the fraction of radiocaesium in the soil solution is low, but nevertheless radiocaesium is highly available for uptake by fungi and green plants. The decomposition of organic material by microbial and fungal activity has been identified as the reason for the mobilisation of radionuclides in organic layers (Agapkina et al., 1998; Desmet et al., 1991; Rafferty et al., 2000). Moreover, Smith and Read (1997) argue that mycorrhizae, not roots, are the chief organs of nutrient uptake in forests. Despite their importance for the cycling of radionuclides in forest ecosystems, fungal interactions are rarely taken into account in forest models. Filling this knowledge gap requires intensive field research, since laboratory results cannot easily be extrapolated to the field.

Another important process that also affects root uptake is the sorption of radionuclides in the mineral horizons of forest soil. Radionuclide bonding to minerals is a complex interaction depending on soil type, pH, redox potential, sorption capacity, clay content, organic matter, and management practice. Although these factors are qualitatively known, they are hard to quantify. Typically, the experimentally determined partition coefficient K_d is used to model the distribution of a radionuclide between the solid and liquid phase but this empirical parameter has a high uncertainty (Bossey and Kirchner, 2004; Kirchner et al., 2009).

The K_d has a different value in the water table (anoxic conditions) compared with the unsaturated zone (oxic conditions). In principle, variations in redox potential should be modelled explicitly as they affect the degree of sorption, requiring the coupling of a biogeochemical model to the hydrological model.

However, the redox potential is closely related to the water content of the soil as it affects directly the ionic strength, being positive well above the water table and negative below the water table, with the transition between these two states occurring mainly across the capillary fringe (Perez-Sanchez et al., 2012; Wheater et al., 2007). Hence, the effects of redox variations are represented indirectly in a simpler way by making the distribution coefficient and soil volatilisation rate dependent on the water content of the soil.

A more advanced model could apply K_d values for the radionuclide retardation functions by using current nutrient concentrations as chemical analogues and some soil properties. For example, the K_d for Cs can be calculated from the parameter RIP (radiocaesium interception potential), which defines the soil exchange capacity and K and NH_4 concentrations (Wauters et al., 1994). For Sr it is calculated from CEC (cation exchange capacity in soil) and Ca and Mg concentrations in soil solution (Rauret et al., 1996). An even more advanced representation would be a parametric K_d -approach, for which a good methodology is provided elsewhere (Vidal et al., 2009). There are good reviews of K_d regression relationships for many elements (Sheppard, 2011), all of which commonly take the form of a nonlinear polynomial expression with input parameters like pH-value and the amount of organic carbon and clay. This approach obviates the need for complex geochemical model, and reduces the number of parameters required by the model to the level of what is available in environmental situations. However, the inherent uncertainties involved in these relationships are only suitable for a limited range of soil properties.

The last major knowledge gap about radionuclide cycling in forests is the translocation of radionuclides in plants. In its simplest form, the problem is addressed by means of transfer ratios within plant compartments. A more advanced approach is using kinetic rates that give an indication of the differential mobility of radionuclides compared with the transport of key nutrients or water within the plant. This is expressed as multiplicative coefficients for the fluxes, following the concept introduced by Casadesus et al. (2008), in which the transfer of radionuclides between two compartments is calculated as the transfer of a known element (e.g. a nutrient), multiplied by the ratio of concentrations of radionuclide to nutrient, corrected by a selectivity coefficient.

3.2. Challenge of variability

Spatial and temporal variability in forests pose several challenges. These are mainly related to the design of monitoring campaigns, of radioecological models and their application.

Representative data are the basis for the development, calibration and validation of robust radioecological models. Samples need to be collected with a proper and structured methodology (Ramsey and Hewitt, 2005), taking into account the purpose of the sampling campaign and the expected variability. For the calibration and validation of radioecological forest models, extensive data sets characterising both, the status of the forest ecosystem and the concentration levels of radionuclides in different media, are necessary. If a radioecological model is designed to provide the variability of its endpoints, data for model validation have to be collected accordingly (sample size, representativeness). Seasonal changes and specific climatic conditions often require frequently sampled data. Practical problems, e.g. restricted access to the area of interest and limited resources, may complicate an optimum monitoring strategy.

Forest ecosystems change in time scales of several years to decades due to external influences (e.g. forest fires, deforestation) or factors which are inherent to the ecosystem (e.g. tree age, shifting

ecologic equilibrium). Whenever working with historical data for model calibration and validation, such effects need to be considered.

3.3. Available experimental data

Two important events, which contaminated large forested areas, provide most of the experimental data: the Chernobyl accident in 1986 and the Fukushima event in 2011. After both accidental releases of man-made radionuclides, radiocaesium and other radionuclides (e.g. strontium and americium) have been monitored in forest ecosystems, allowing for the comparison of predicted concentration levels at specific locations with measurements (Hashimoto et al., 2012). Comparative studies using data from both events can lead to more confidence in the validity of the models in question (Calmon et al., 2015).

4. Conclusion

Modelling the radionuclide cycle in forests is a challenge because of many complex processes and the heterogeneity of the ecosystem. Radioecological models should cover the key processes but should be kept as simple as possible with respect to their specific purposes. Models with a reasonably simple mathematical structure and a small number of parameters should be preferred to quantify the activity levels in forests and the radiation exposure of humans and the environment. A strong interaction between experimentalists and modellers helps to obtain well-documented high-quality data as the basis for model development, calibration and validation.

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