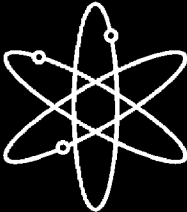


# Alternative Conceptual Models for Assessing Food Chain Pathways in Biosphere Models



**Pacific Northwest National Laboratory**



**U.S. Nuclear Regulatory Commission  
Office of Nuclear Regulatory Research  
Washington, DC 20555-0001**



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## Abstract

This report describes alternative approaches to modeling the key processes in radionuclide transport in the biosphere. Because the focus of the NRC project *Assessment of Food Chain Pathway Parameters in Biosphere Models* is the food-chain models used in performance assessments of radioactive waste disposal facilities, models and approaches applicable over relatively long periods (more than one year) are evaluated, as opposed to approaches detailing radionuclide behavior over the shorter periods applicable to acute, accident-type, calculations. There are a number of important features and processes that all terrestrial biosphere models must address: these include radionuclide behavior in soils, interception of deposition onto vegetation, weathering of intercepted material from plant surfaces, foliar absorption and translocation within plants to other vegetative structures, uptake from soil by plant roots, and transfer from plants to animals and animal products.

The report provides a detailed discussion of possible alternative approaches to modeling the food-chain pathway in biosphere models and recommends the following:

- For *soil*, an annual average model incorporating:
  - Accumulation from irrigation or atmospheric deposition;
  - Leaching to deeper soil, using a  $K_d$ -modulated leach rate;
  - Harvest removal, averaged over crop types
  - Uniform mixing in a reasonable (15-25 cm) upper soil surface layer, implicitly caused by plowing, bioturbation, and leaching;
  - Neglecting radionuclide fixation, because appropriate long-term measurements of distribution coefficient and concentration ratio will already incorporate the effects;
  - Neglecting surface-soil erosion losses, because eroded material from one location may accumulate in another, cancelling any perceived benefit.
- For *resuspension*, a mass-loading approach, because it has the lowest variability and is the most easily defended.
- For *foliar interception*, models incorporating:
  - Dry interception following the basic model suggested by Chamberlain (1967) as updated by Adriano et al. 1982;
  - Wet interception considering one of the algorithms derived from the data of Hoffman et al. (1989) or Prohl and Hoffman (1993).
- For *plant contamination with soil*, an adhesion model applied so that material translocated is not subject to weathering (e.g., IAEA 2003; Wu 2003).
- For *weathering*, a single exponential model with a half-time between about 10 and 20 days, applied to material on the surface of the plant only.
- For *translocation*, a radionuclide-specific (or chemical-class-specific) implementation allowing translocated materials to avoid being removed via weathering processes (e.g., IAEA 2003; Wu 2003).
- For *soil-to-plant transfer*, a radionuclide- and plant-type-specific concentration ratio, where available, with:
  - Site-specific application of additional crops, such as mushrooms;

- The specific-activity model of Peterson and Davis (2002) for elemental and oxide forms of tritium;
- A general specific-activity model for  $^{14}\text{C}$  in air, adapted to the RESRAD family of codes (Yu et al. 2001) model for transfer from irrigation water to air as  $^{14}\text{CO}_2$ ;
- Perhaps site-specific application of specific-activity models for iodine and other micro-nutrient and macro-nutrient elements and their chemical analogues. This is a simplification of the need to allow for homeostatic regulation of some elements.
- Until a simple, robust *Fruit Tree* model is developed, continue to use the concentration ratio approach for fruits.
- For *animal products*, a transfer factor approach applied to average daily feed, water, and soil intake by the animal where data are available.
  - For element/animal combinations where data are lacking, a justified mix of specific-activity and allometric models should be considered.
- For *food processing*, because the data are sparse and the reduction is generally small, modelers are justified in ignoring losses (which is equivalent to using a food-processing transmission factor of 1.0).

This type of information is directly useful in formulating inputs to radioecological and food-chain models used in performance assessments and other kinds of environmental assessment. This food-chain pathway data may be used by the NRC staff to assess dose to persons in the reference biosphere (e.g., persons who live and work in an area potentially affected by radionuclide releases) of waste disposal facilities and decommissioning sites.





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## 1.0 Introduction

The U.S. Nuclear Regulatory Commission's project *Assessment of Food Chain Pathway Parameters in Biosphere Models* has been established to assess and evaluate a number of key parameters used in the food-chain models used in performance assessments of radioactive waste disposal facilities. The objectives of the research program include:

- Provide data and information for the important features, events, and processes of the pathway models for use in biosphere computer codes. These codes calculate the total effective dose equivalent (TEDE) to the average member of the critical group and/or reasonably maximally exposed individual, for example, from radionuclides in the contaminated ground water release scenarios in NRC's performance assessments of waste disposal facilities and decommissioning sites,
- Reduce uncertainties in food-chain pathway analysis from the agriculture scenarios of biosphere models in performance assessment calculations,
- Provide better data and information for food-chain pathway analyses by:
  - Performing laboratory and field experiments, including integral and separate effect experiments, to evaluate the potential pathways and uptake mechanisms of plants and animals contaminated by long-lived radionuclides,
  - Presenting food-chain pathway data and information by regional and local geographical locations,
  - Quantifying uncertainties in the radioactive contamination of food crops and long-term build up of radionuclides in soils with contaminated ground water from water irrigation systems,
  - Determining data on factors affecting radionuclide uptake of food crops including irrigation water processes, soil physical and chemical properties, soil leaching and retention properties near crop roots, soil resuspension factors and other soil and plant characteristics.

The results of this research program will provide needed food-chain and animal product pathway data and information for important radionuclides that may be used by the NRC staff to assess dose to persons who live and work in areas potentially affected by radionuclide releases from waste disposal facilities and decommissioning sites.

The biosphere model is the last model in a series of models used by the NRC staff in performance assessments of waste-disposal facilities and decommissioning sites. The biosphere models used by the NRC staff in its performance assessments are based on conceptual models, their mathematical representation, and the implementing computer codes. The biosphere models used by the NRC, and their implementing computer codes, include hypotheses, assumptions, and simplifications that describe the reference biosphere in the vicinity of the waste-disposal facilities and decommissioning sites. These computer codes consider (1) radionuclide transport through many food-chain pathways such as irrigation-water deposition on vegetation and soil surfaces, (2) crop interception and retention, (3) radionuclide buildup in soils as a result of long-term

irrigation deposition, (4) soil radionuclide leaching and retention mechanisms in plant root zones, (5) resuspension of radionuclide-contaminated soil onto vegetation, and transfer via gaseous phase radionuclides arising from soil, (6) soil to plant uptake via roots, (7) uptake by stems/leaves of soil-derived gaseous contaminants, (8) feed-to-animal product transfer, and (9) individual food product consumption rates. Biosphere models used in performance assessments provide the long-term, time-dependent concentrations of radionuclides in soil, plants, and animal products that could be consumed by future human residents of the biosphere. When coupled with projections of human behavior, they also provide estimates of annual or lifetime radiation dose; that component of the biosphere modeling is not addressed in this report.

Section 1 of this report is this introduction.

Section 2 of this report describes some key concepts in biosphere modeling - important features and processes that all biosphere models must address in some manner. For terrestrial models, these include radionuclide behavior in soils, interception of deposition onto vegetation, weathering of intercepted material from plant surfaces, foliar absorption and translocation within plants to other vegetative structures, uptake from soil by plant roots, and transfer from plants to animals and animal products.

Section 3 of this report provides descriptions of various ways of modeling these important features and processes. For some processes, various methods of different levels of complexity are applied depending on the type of analyses being performed. For other processes, very limited information allows only one or two alternative approaches to the calculations.

Section 4 makes recommendations on the types of modeling approaches that are most applicable for models to be used to assess doses to persons who live and work in areas potentially affected by radionuclide releases from waste disposal facilities and decommissioning sites.

The results are expected to be useful in:

- supporting the development of regulatory criteria (e.g., guidance, technical positions) for food-chain pathway issues involving biosphere models
- providing a basis for evaluating and auditing an applicant's or licensee's biosphere and food-chain pathway data, information, analyses, conceptual models, and computer codes used in license submittals
- providing NRC staff with data and information for resolving biosphere issues involving irrigation pathways, food and animal transfer factors, and groundwater radionuclide release scenarios.

The results of the research program improve the NRC staff's understanding of the features and processes for some important long-lived radionuclides in biosphere modeling of the performance-assessment process.

## 2.0 Concepts in Biosphere Modeling

There are a number of important features and processes that all biosphere models must address in some manner (UNSCEAR 2000). For terrestrial models, these include radionuclide behavior in soils, interception of deposition onto vegetation, weathering of intercepted material from plant surfaces, foliar absorption and translocation within plants to other vegetative structures, uptake from soil by plant roots, and transfer from plants to animals and animal products. Because different models and model types approach these features and processes differently, some of the reasons for the different approaches are discussed in this section in general terms before specific methods of computation are described.

The primary inputs to estimating radiation doses to individuals and populations are the concentrations and availabilities of radionuclides in air, water, soil, and foods, and the level of exposure of the individuals or groups to each radionuclide in each medium. Because it is not possible to completely characterize the radiological environment, and because in many practical cases the concentration of radionuclides in the environment resulting from reactor operations is too low to measure, mathematical models are employed to go from what is known to what needs to be known.

Many different types of models have been developed. The various models are directed to answering different questions. Different models have different levels of detail, different degrees of accuracy, and different temporal or spatial scales. Some are used for retrospective analyses (to analyze what has happened - as for determining compliance with annual radiation dose limits) or for prospective analyses (to project what may happen - as for demonstrating compliance with licensing requirements).

Two general classes of environmental transport models have evolved: dynamic (transient) and equilibrium (steady-state). Both tend to describe the environment in terms of various "compartments" such as soil layers, plant types, and animal types (some environmental media may be described in terms of more than one compartment, such as the roots, trunk, branches, and fruit of trees). The dynamic models consider the time-dependent quantities of radionuclides in various environmental compartments. The structure of the model is represented by a series of coupled differential equations that describe the rate of change of the amount of the radioactive material in each compartment as a function of the various transfer paths into (e.g. irrigation) and out of it (e.g., radioactive decay, harvest). When the equations are evaluated over sufficiently long times with unvarying values of the inputs and rate constants, the ratios of the concentrations of the radionuclides in the various compartments approach constant values. The system then is considered to be in equilibrium or in a steady state. Dynamic models are frequently used as research tools to investigate the detailed phenomena and mechanisms of environmental transport of radionuclides. For example, they are beneficial in evaluating the consequences of pulse inputs to selected compartments, as might happen following an accidental release. However, because of the temporal resolution demanded of the output, a great deal of

information is required as input to this type of model, and extensive computer resources are required for implementation. By using assumptions of quasi-equilibrium (that is, relatively small changes from year to year in local conditions), the dynamic models may be simplified into equilibrium models. The equilibrium models lose the ability to answer certain temporally-based questions, but are generally simpler to use, because many of the detailed rate constants required by the dynamic models can be treated as lumped parameters. Because most regulations dealing with nuclear waste are written in terms of annual doses to people, most routine radiological evaluations are performed using the equilibrium models with annual-average values for the input parameters.



## 3.0 Modeling Key Processes

Alternative approaches to modeling the key processes in radionuclide transport in the biosphere are presented in this section. Because the focus of the NRC project *Assessment of Food Chain Pathway Parameters in Biosphere Models* is the food-chain models used in performance assessments of radioactive waste disposal facilities, models and approaches applicable over relatively long periods (more than one year) are evaluated, as opposed to approaches detailing radionuclide behavior over the shorter periods applicable to acute, accident-type, calculations. International reviews of models of this type that have been performed in the past decade include the IAEA BIOMASS (IAEA 2003) program and the European BIOPROTA program (Albrecht et al. 2005).

### 3.1 Modeling Radionuclides in Soil

Surface soils tend to be compartments for long-term retention and accumulation of radionuclides released into the accessible environment, either from atmospheric or liquid releases or from localized depositions. Soils typically are made up primarily of mineral components with additional organic material incorporated from decomposing plant matter, hosting numerous microorganisms and small invertebrates. The surface layer of soils may be mixed by small animals, and in agricultural systems is usually mixed annually by tillage. Radionuclides may become incorporated in vegetation; the mineral and organic constituents of vegetation cycle in the soil and serve as a reservoir as the organic component decomposes. On small spatial scales, soils may erode, thus moving contamination from one location to another. Percolating water may transport contaminants from shallow to deeper soils.

The concentrations, mobility, and bioavailability of radionuclides in surface and subsurface geologic systems are controlled by numerous hydrologic and geochemical processes. These include hydrologic factors, such as dispersion, advection, and dilution; and geochemical processes, such as aqueous complexation, oxidation/reduction (redox), adsorption/desorption and ion exchange, precipitation/dissolution, diffusion, colloid-facilitated transport, and anion exclusion (Langmuir 1997; Sposito 1989; 1994). Additionally, in the uppermost layer of surface soil, the mobility of radionuclides can also be increased by biological activity and by the drying and subsequent cracking of soils. Colloid-facilitated transport and anion exclusion have received considerable attention recently in that they can enhance the transport of certain radionuclides. However, these processes are hard to quantify, and the extent to which they occur is difficult to determine. The importance of colloid-facilitated migration, especially in aquifer systems that do not involve fracture flow of groundwater, is still a subject of debate.

Some radionuclides, such as technetium, uranium, or plutonium, may be present in more than one oxidation state in the environment. The adsorption and precipitation behavior of different oxidation states of a particular radionuclide are usually very different. For example, in environmental systems, the most stable oxidation states of technetium are +7 and +4 under oxidizing and reducing geochemical conditions, respectively. The chemical

behavior of technetium in these two oxidation states differs drastically. Dissolved Tc(VII) exists as pertechnetate anion,  $\text{TcO}_4^-$ , over the complete pH range of natural waters under oxic conditions. Because the pertechnetate anion is highly soluble and is not strongly sorbed, it is highly mobile in most oxidizing systems. Under reducing conditions, however, Tc(IV) exists as hydrolyzed cations and is relatively immobile in the absence of strongly complexing ligands. Technetium(IV) is highly sorbed and forms the sparingly soluble  $\text{TcO}_2 \cdot n\text{H}_2\text{O}$  solid (Krupka 2003).

Adsorption/desorption (including ion exchange) and precipitation/dissolution are considered the most important processes affecting radionuclide interactions with soils. Precipitation/dissolution is more likely to be an important process where elevated concentrations of dissolved radionuclides exist, such as in the near-field environment of radioactive waste disposal facilities or the spill sites of radionuclide-containing wastes or where steep pH or redox gradients exist. Adsorption/desorption will likely be the key process controlling radionuclide retardation in areas where trace concentrations of dissolved radionuclides exist, such as those associated with far-field environments of disposal facilities or spill sites or in areas of where soils are to be irrigated using radionuclide-contaminated water.

The term “sorption” is used as a generic term devoid of mechanism and used to describe the partitioning of dissolved aqueous-phase constituents to a solid phase. When a radionuclide is associated with a geologic material, however, it is usually not known if the radionuclide is adsorbed onto the surface of the solid, absorbed into the structure of the solid, precipitated as a three-dimensional molecular structure on the surface of the solid, or partitioned into the organic matter (Sposito 1989). The term “sorption” encompasses all of the above processes.

The sorption of radionuclides on soils is frequently quantified by the partition (or distribution) coefficient ( $K_d$ ). The  $K_d$  parameter is a factor related to the partitioning of a radionuclide between the solid and aqueous phases and is defined as the ratio of the quantity of the adsorbate adsorbed per mass of solid to the amount of the adsorbate remaining in solution at equilibrium. Radionuclides that adsorb very strongly to soil have large  $K_d$  values (typically greater than 100 mL/g) compared to those values for radionuclides that are not significantly retarded by adsorption. Radionuclides that do not adsorb to soil and migrate essentially at the same rate as the waterflow have  $K_d$  values near 0 mL/g. The  $K_d$  model is the simplest yet least robust sorption model available. However, the  $K_d$  metric is the most common measure used in hydrologic transport and biosphere codes to describe the extent to which contaminants are sorbed to soils. The primary advantage of the  $K_d$  model is that it is easily inserted into computer codes to quantify the reduction in the extent of transport of a radionuclide relative to groundwater. The  $K_d$  is an empirical unit of measurement that attempts to account for various chemical and physical retardation mechanisms that are influenced by a myriad of variables. As such, the  $K_d$  model is often the subject of criticism (EPA 1999).

Because of recharge from precipitation or irrigation, radionuclides deposited on soils may leach through the surface layer with percolating water, eventually reaching depths below

the surface where they are no longer available for uptake by plants and animals. Sorbed materials may also travel with soil particles as they are physically transported via wind or water erosion.

Deposition of radioactive material from the air or irrigation water onto the earth's surface is a function of surface area. Atmospheric dispersion model outputs generally deal with depletion of plumes or puffs of material in terms of mass per unit area per unit time. Irrigation deals with quantities of water per unit area (e.g., acre-feet) per unit time. Although a fraction of depositing activity usually is intercepted by vegetation, it is generally assumed that all deposition eventually works its way to the soil surface. The initial problem is to convert this initially infinitesimally-thin layer into a bulk concentration in soil, and then to describe its subsequent behavior.

This problem was first addressed in projections of nuclear weapon fallout (Burton 1966; Ng and Thompson 1966). The initial conversion was to assume instantaneous mixing of deposition in a soil "root zone" with a depth of 20 cm and a soil density of 2 g/cm<sup>3</sup>. Here the radionuclides remain except for radioactive decay.

This basic concept was adopted in early radiological models (e.g., Soldat and Harr 1971) which used a 15 cm plow depth and resulting area density of 224 kg/m<sup>2</sup>. This approach was adopted by the NRC in Regulatory Guide 1.109 (1977), with a soil plow layer mixing depth of 15 cm and effective surface density of 240 kg/m<sup>2</sup> (equivalent to a soil density of 1.6 g/cm<sup>3</sup>). This is assumed to represent the depth to which plowing or other agricultural practices mix the soil on an annual basis (so that 1-year's deposition is mixed into the rooting zone and available for plant uptake). This became the default approach for many computer codes written to implement the concepts of this regulatory guide (e.g., FOOD (Baker, Hoenes, and Soldat 1976), AIRDOS (Moore et al. 1979), RSAC (Wenzel 1993)). This basic model became the international standard with the issuance of International Atomic Energy Agency publication Safety Series 57 (IAEA 1982). In the GENII code (Napier et al. 1988), the mixing depth is defined to be a constant 15 cm (6 in.). The code MEPAS (Streng and Chamberlain 1995) allows user inputs, but the defaults are 15 cm (6 in.) for agricultural soils, and 4 cm (1.6 in.) for residential soils. The PATHWAY (Whicker and Kirchner 1987) model uses 25 cm (10 in.). ECOSYS (Mueller and Prohl 1993) uses 10 cm (4 in.) for pastures and 25 cm (10 in.) for plowed soil. The NCRP screening model (NCRP 1999) conservatively uses 5 cm (2 in.). Because of its primary use as a code for remediation of previously contaminated soils, the RESRAD family of codes (Yu et al. 2001) default is 200 cm (79 in.). The calculated soil concentration is inversely related to this assumption. Similarly, the assumed soil density has a direct influence on the estimated concentrations.

It is unlikely that long-lived radionuclides introduced into surface soil would remain in one location forever. Therefore, additional processes of radionuclide migration have been considered in various models. The first mechanism considered is leaching of contaminants out of the root zone and into deeper soils, effectively taking them out of the system. This approach is used in models such as GENII (Napier et al. 1988), RESRAD (Yu et al. 2001), the model of Peterson (1983), and PATHWAY (Whicker and Kirchner

1987). Leaching (as caused by overwatering for example (Schreckhise 1980)) is treated as an average loss rate term, generally using a first-order rate constant. The leaching term is thought to be dependent on the type of soil, the radionuclide, and the amount of water percolating through the surface and into the deeper soil. The format of Baes and Sharp (1981) is frequently used:

$$\lambda_{si} = \frac{P + I - E}{d_s \theta_s \left[ 1 + \frac{\rho_s}{\theta_s} Kd_{si} \right]} \quad (1)$$

where  $\lambda_{si}$  = removal rate constant for activity of radionuclide i in the surface soil layer ( $\text{yr}^{-1}$ )

P = total annual precipitation (cm/yr)

I = total irrigation rate (cm/yr)

E = total evapotranspiration rate (cm/yr)

$d_s$  = depth of soil rooting zone (cm)

$\rho_s$  = surface soil bulk density ( $\text{g}/\text{cm}^3$ )

$\theta_s$  = surface soil volumetric water content ( $\text{mL}/\text{cm}^3$ )

$Kd_{si}$  = surface soil distribution coefficient for radionuclide i ( $\text{mL}/\text{g}$ ).

In general, because precipitation is variable and because it is difficult to calculate the evapotranspiration, the P+I-E term is approximated as a constant “overwatering” term.

The factor describing mobility (or transportability) of radionuclides in soil is the Kd. This term is empirical and rarely easily available for specific soils in specific locations. Frequently, measurements may be poor; for very insoluble materials, sometimes experiments may measure precipitation into a solid rather than reversible sorption. The models in GENII, in RESRAD, in MEPAS (Streng and Chamberlain 1995), described in NUREG/CR-5512 (Kennedy and Streng 1992), and Peterson (1983) all follow the same structure. (The MILDOS model [Streng and Bander 1981] is similar, but uses a constant 50-year leaching half-time for all contaminants.) No current biosphere models have addressed solubility limits, which are considered to be potentially important in some circumstances (NRC 2002). The IAEA Safety Series 19 model (IAEA 2001) makes a distinction between anionic radionuclides, isotopes of strontium and cesium, and all other radionuclides. Anions such as  $\text{TcO}_4^-$ ,  $\text{Cl}^-$  and  $\text{I}^-$  are leached quickly; the default value for  $\lambda_s$  is  $0.5 \text{ yr}^{-1}$ . For strontium and cesium the default value is  $0.05 \text{ yr}^{-1}$ . For all other nuclides (also non-anionic Tc) the default value is zero.

Some models have slightly more sophisticated surface soil zone models. The ECOSYS, PATHWAY, and MACCS (Jow et al. 1990) models include a second mechanism for effective removal of radionuclides from the soil for plant uptake. These models include an additional rate constant for “fixation” of contaminants. In these models, the radionuclide is bound to receptor sites in the soil and immobilized. In these models, the radionuclide is no longer mobile, so it is not available for plant uptake, although it still contributes to external exposure.

Measurements of depth profiles of  $^{137}\text{Cs}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Am}$ , and  $^{237}\text{Np}$  in undisturbed soils from both Chernobyl and weapons' fallout indicate that removal times can be derived that are considerably faster than those derived from the distribution coefficient  $K_d$ . It is interesting to note that the residence times of the radionuclides investigated are relatively similar although their chemical properties are different. Apparently, element independent transport mechanisms such as the transport of radionuclides attached to clay particles or bound to soil colloids may also play a role. Since the main transport mechanisms for strongly bound radionuclides may be to a large extent element-independent, the value for the residence half-time for the upper soil layer in the European emergency response code RODOS (Müller, Gering, and Pröhl 2004) is applied for all other elements with high  $K_d$  as zirconium, niobium, ruthenium, cerium and plutonium - a soil half-life of 100 years is assumed.

Another potential mechanism for loss of material from the surface soil is harvest removal (sometimes referred to as *cropping*), the process by which contaminants leave a local system by being harvested with the crop and removed. Hoffman and Baes (1979) proposed a linear rate constant to be applied annually as

$$\lambda_{hi} = \frac{B_{iv} M_h H_n}{P} \quad (2)$$

where

- $\lambda_{hi}$  = the harvest removal rate constant ( $\text{yr}^{-1}$ )
- $B_{iv}$  = the ratio of concentration of radionuclide  $i$  in the harvested plant compared to that in soil ( $\text{Bq kg}^{-1}$  plant per  $\text{Bq kg}^{-1}$  soil)
- $M_h$  = the harvested biomass of vegetation per unit surface area per harvest ( $\text{kg m}^{-2}$ ), the amount of plant material which is removed at harvest and not returned as recycle
- $H_n$  = the number of harvests per year ( $\text{yr}^{-1}$ )
- $P$  = the soil area density discussed above ( $\text{kg m}^{-2}$ ).

The ERB2A model from the IAEA BIOMASS program (IAEA 2003) uses a similar formulation, including averaging over all crop types with the assumption that long-term agriculture would include crop rotation. A similar expression is used also by Willans (2003) in the MONDRIAN code used at British Nuclear Fuels Limited, as well as by EPRI (2002). These models assume that, over long periods of time, crops will be rotated, and so each uses the average of the soil and crop parameters for all of the agricultural pathways assumed.

In the GENII computer codes (Napier et al. 1988; 2002), loss of activity from the surface soil zone by harvest is modeled as a step function applied at the end of each calculational year. The amount of loss is calculated from the plant concentration at harvest, the annual plant yield, and the soil concentrations at harvest. The calculation is represented by the following equation.

$$C_{si}(t_+) = C_{si}(t_-) \left[ (C_{si} - C_{ci}(t) Y_c) / C_{si} \right] \quad (3)$$

where

- $C_{si}(t_+)$  = surface-soil concentration for radionuclide  $i$  after correction for harvest removal at time  $t$  (Bq/m<sup>2</sup>)
- $C_{si}(t_-)$  = surface-soil concentration for radionuclide  $i$  before correction for harvest removal at time  $t$  (Bq/m<sup>2</sup>)
- $C_{ci}(t)$  = crop- $c$  concentration derived from soil uptake for radionuclide  $i$  at time of harvest (Bq/kg)
- $Y_c$  = annual yield of crop  $c$  (kg/m<sup>2</sup>).

The term in brackets represents the average fraction of the contaminant in soil to that in crops over the year; this term is used rather than a simple subtraction of amount harvested because, for short-lived radionuclides, the amount harvested over the year may actually be larger than the amount remaining in the soil at the end of the year. Note that this formulation assumes that harvested materials are removed from the system, and not returned (for instance, as manure or fertilizer).

Under natural conditions, the rate of soil removal by erosion generally is in approximate equilibrium with the rate of soil development from soil forming processes, and under these conditions, soil depth is relatively constant (Troeh et al. 1980). Human activities tend to accelerate the rate of soil removal. The removal of surface soil by erosion would result in the loss of radionuclides attached to the soil particles. Most models ignore this loss, assuming that soils lost from one location would re-accumulate in another. The model used for analyses for the proposed repository at Yucca Mountain, Nevada, which is designed to evaluate conditions over periods of thousands of years, does include a wind-erosion term (Wu 2003). The rate of radionuclide removal from surface soils is quantified in that model using a first-order surface soil erosion rate constant, defined in terms of the surface soil erosion rate as

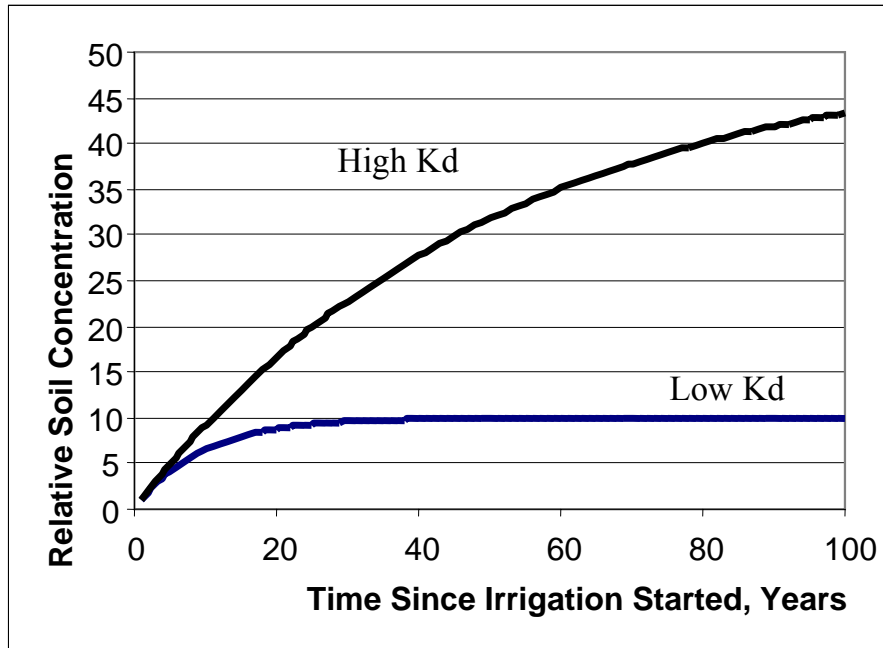
$$\lambda_e = E / \rho_s d_s \quad (4)$$

where

- $\lambda_e$  = the surface soil erosion rate constant (yr<sup>-1</sup>)
- $E$  = the soil surface erosion rate (kg m<sup>-2</sup> yr<sup>-1</sup>)
- $d_s$  = depth of soil rooting zone (m)
- $\rho_s$  = surface soil bulk density (kg/m<sup>3</sup>).

Application of the soil model for the case of continuous irrigation with a contaminated source results in the estimated concentration of contaminant in soil increasing with time until it reaches equilibrium. At equilibrium, the input rate equals the combined loss rates through decay, leaching, erosion, and harvest. For mobile radionuclides (those with small  $K_d$  values), the equilibrium can occur within a few years of the initiation of irrigation, as illustrated in Figure 1. However, for radionuclides with a large  $K_d$ , and a small soil-to-plant uptake (which minimizes harvest loss), the approach to equilibrium soil concentration can take hundreds to thousands of years (Figure 1). This leads to the requirement for an additional input—the length of time irrigation is assumed to occur before the exposure of the individual under consideration. Most codes, such as GENII, MEPAS, or RESRAD, can be set to assume that the irrigation begins either at the time the exposure scenario begins, or at some time before that to allow for buildup. Once

irrigation starts, it is assumed to continue at a constant annual rate (there may be seasonal starts and stops).



**Figure 1.** Idealized Behavior of Radionuclide Concentration in Soil Following Long Periods of Irrigation for Mobile and Immobile Radionuclides

An example application of an irrigation model is provided by the Hanford Site System Assessment Capability (SAC) (Bryce et al. 2002). In the most recent modeling application using the SAC, irrigation is assumed to start at all locations simultaneously at some fixed time following Hanford Site closure and continue indefinitely (Bryce et al. 2002). A slightly different example is provided by the analyses performed for the Yucca Mountain Project. In these analyses, the start time for the irrigation scenario was generated randomly, followed by a random period of time before exposure. Because the Yucca Mountain analyses use pre-calculated Biosphere Dose Conversion Factors, the analyses fit a set of factors generated for different periods of time following initiation of irrigation to an exponential curve. The analyses also used the curve fit as a simple means of evaluating the impacts of long periods of irrigation (Wasiolek 2001). Both the Hanford and Yucca Mountain analyses are projected for very long times into the future (up to 10,000 years). Continuous irrigation using groundwater has taken place for much shorter periods (about 100 years) in the United States. Increasing salinity in soils is a long-term problem. It is still an open question whether irrigation can continue for millennia.

Some radionuclides may be released from soil to air as gasses. This mechanism is only of concern for radionuclides that are gases, produce gaseous progeny, or form gaseous compounds (e.g.,  $^{222}\text{Rn}$  and  $^{14}\text{C}$ ). This removal mechanism is not usually included in

most general models. It would result in non-equilibrium concentrations of decay progeny in the case of radon emanation. The RESRAD model includes estimation of radon fluxes from soils; the result is not used to deplete the soil of radon progeny.

Most of the models described above deal with tillage or plowing in an implicit manner. The surface layer of soil is assumed to be uniformly mixed with the radionuclide. Cultivated soil is assumed to be ploughed, if not every year then every few years. This general concept is also used for residential soils not used for raising crops. Another mechanism for mixing of surface soils is bioturbation, the disturbance of soil layers by biological activity. These biotic pathways include transport of contaminants through various soil horizons by plant root systems and by burrowing insects and small mammals. Bishop (1989) noted that the maximum amount of soil moved by earthworms in established pastures is in the order of 10 kg/m<sup>2</sup>/y (equivalent to a soil depth of around 7 mm). In soil science, bioturbation is modeled as a diffusive process, adopted to avoid quantifying the numerous types of mixing resulting from flora and fauna. The diffusion coefficient describing bioturbation can be determined by fitting results to vertical distributions of natural tracers, radionuclides from fallout, or introduced particles. A more mechanistic approach was developed by McKenzie et al. (1986), which explicitly evaluated the transport of soil from various subsurface layers to the surface soil. The McKenzie et al. (1986) approach provides the amount of soil moved, and the fraction from various depth layers, per year. This is summarized in Table 1 for various environmental conditions, which indicates that even in relatively undisturbed soils without plowing, mixing will occur to substantial depths relatively rapidly.

**Table 1.** Biotic Transport Quantities (adapted from McKenzie et al. 1986)

<u>Depth of Soil Layer</u>	<u>Volume Transported to the Surface (m<sup>3</sup> m<sup>-2</sup> yr<sup>-1</sup>)</u>		
	<u>Arid</u>	<u>Humid</u>	<u>Agricultural</u>
< 0.15 m	9.41E-4	7.48E-4	7.48E-4
0.15 - 0.5 m	7.62E-4	6.73E-4	6.73E-4
0.5 - 1.0 m	1.79E-4	7.18E-5	7.18E-5
1.0 - 1.5 m	1.88E-5	4.49E-6	4.49E-6
1.5 - 2.0 m	7.53E-6	3.74E-8	3.74E-8
> 2.0 m	1.88E-6	3.74E-8	3.74E-8

### 3.2 Modeling Resuspension

Resuspension relates the concentration of contaminants in air above a contaminated surface resulting from losses from that surface to the air. Resuspension occurs when wind exerts a force on surface material or when there is a mechanical action that disrupts the surface (such as agricultural operations or vehicular movement). Modeling resuspension is not easy because of the large number of potential variables. The main ones include:



- Particle size, shape and adherence;
- Wind speed,
- Surface type and cover (measured in terms of “roughness”);
- Time since deposition;
- Intensity of the mechanical action.

To quantify the suspension of contaminated soil/sediment into the atmosphere, three main approaches have been usually used: - resuspension rates, resuspension factors, or mass loading. The resuspension rate is a measure of the fraction of material entering the air per unit time, and has units of  $s^{-1}$ . The resuspension factor relates the radionuclide concentration in soil (per unit area) to the concentration in air and has units of  $m^{-1}$ . The resuspension rate is a mass of soil, assumed to have a contaminant concentration directly related to the source soil, suspended in a volume of air. The mass loading approach essentially assumes that the amount of radioactive contamination in the air is associated with dust having the same concentration as local soil. The resuspension factor for food crop and animal product pathways is representative of conditions on farmland, which may be different from the resuspension factor for the inhalation exposure pathway. Farmland would be expected to be tilled and have soil generally looser than soil for the general residential exposure situations.

The use of resuspension rate approach is based on the transfer of contamination from the soil surface into the air. The resuspension rate is defined to be the ratio between particle flow density and soil contamination, thus, the resuspension rate dimension is a reciprocal time. This method provides a rate constant for loss from the surface soil; an additional model is required to convert this into an air concentration.

The use of resuspension factors in the dose assessments is based on the assumption that the particulate matter in the air has the same activity as the soil at the location. The resuspension factor is defined to be the ratio between air concentration and soil contamination, thus, the resuspension factor dimension is a reciprocal length. This is represented mathematically as follows.

$$C_a = RF C_s \quad (5)$$

where  $C_a$  = air concentration of radionuclide ( $Bq/m^3$ )  
 $RF$  = resuspension factor ( $m^{-1}$ )  
 $C_s$  = average surface soil concentration ( $Bq/m^2$ ).

Resuspension factors have been found to cover a wide range ( $10^{-10}$  to  $10^{-3} m^{-1}$ ), depending on many factors, although this variability is based on short sampling times and long-term variability will be less (Garger, Hoffman, and Thiessen 1997). This approach allows a convenient method of expressing the observed relationship between surface and air contamination, but with a number of limitations. There is an implicit assumption that the air concentrations are a result of the local surface contamination, however, the air concentration usually includes resuspended materials from upwind sources, which may be contaminated at different levels. In addition, the time scale of the measurements (generally short) may not correspond to long-term average needed for annual-averaged models. A decrease in the amount of resuspension has been observed with time, caused

by weathering, erosion, or migration of materials down into the soil. This time dependence has been considered by modelers (e.g., Anspaugh et al 1975; CEC 1995; Linsley 1978) and experimentalists (e.g., Garland et al. 1982). Many authors have described the time dependence using exponential factors, but Garland (1979) found that an inverse power function fitted wind tunnel data over a period of several months. A recent review (Walsh 2002) recommends the Garland approach modified to account for long-term resuspension, given as

$$RF = [1.2 \times 10^{-6} t^{-1} + 10^{-9}] e^{-\lambda t} \quad (6)$$

where  $1.2 \times 10^{-6}$  = resuspension factor at the time of initial deposition to soil ( $m^{-1}$ )  
 $t$  = time after initial deposition of material to soil (d)  
 $10^{-9}$  = resuspension factor after a long time ( $m^{-1}$ ).

The second term in this equation is added based on the assumption that no further measurable decrease in the resuspension factor process occurs after the longest period for which there are data available. In the implementation, an assumption is made that only the top thin layer of soil is available for resuspension.

The atmospheric mass loading of soil in air approach equates the radionuclide concentrations in soil to measured or estimated dust levels in air. Mass loading is used to estimate the resuspension factor as follows:

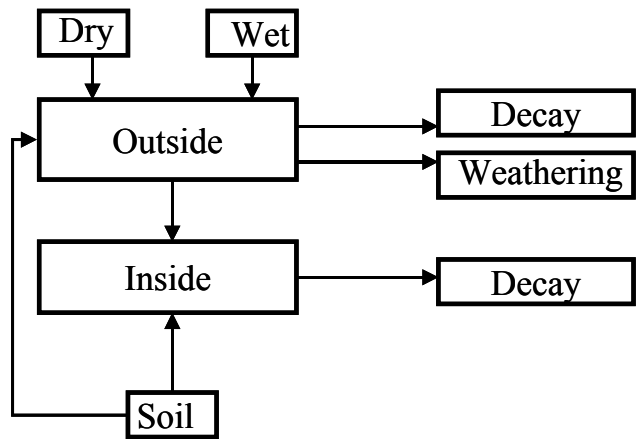
$$RF = \frac{S}{\rho_s d_s} \quad (7)$$

where  $S$  = mass loading of soil in air ( $g/m^3$ ),  
 $\rho_s$  = surface soil density ( $g/m^3$ ),  
 $d_s$  = thickness of surface soil layer (cm).

Different resuspension models may be recommended for different contexts (Garger, Hoffman, and Thiessen 1997). The resuspension rate and resuspension factor models may be better suited for analyses following single acute depositions. The simple mass-loading model may be better suited, and more stable, for analyses of long-term soil accumulation, because it is not based on measurements from single deposition events, as is the resuspension factor approach.

### 3.3 Modeling Radionuclides in Vegetation

Radioactive contamination of vegetation may be a major contributor to human health impacts through both direct ingestion and/or indirect transfers via the food chain. Plants may become contaminated by various processes including direct deposition, external contamination with local contaminated soil via resuspension or splashup, incorporation into edible parts of contamination deposited on leaves or stems, and uptake via the roots. The basic components to be modeled are illustrated in Figure 2.



**Figure 2.** A Basic Model for Contamination of Plants

### 3.3.1 Modeling Foliar Interception and Retention

Deposition of airborne contamination is largely a function of particle size and precipitation. Radionuclides can be present in air as single atoms, as chemical compounds, or as contaminants attached to dust or other aerosol particles. As a general rule, one can expect the deposition of radionuclides to be governed by the physical processes involving aerosol-land surface interactions. The rate of radionuclide deposition from the atmosphere to the ground is proportional to concentrations in air, although the actual deposition is also affected in complex ways by particle characteristics, receiving surface properties, and weather conditions.

Airborne radionuclides can be deposited onto soil or vegetation by “dry” mechanisms such as gravitational settling, surface impaction and electrostatic attraction and by “wet” mechanisms such as washout or rainout during precipitation events (Kinnersley and Scott, 2001). As a general rule, particles with diameters larger than about 20  $\mu\text{m}$  and densities higher than  $1 \text{ g cm}^{-3}$  are affected more by gravity than atmospheric turbulence (Whicker and Schultz, 1982). Deposition of these larger particles can be predicted using Stoke’s law to evaluate a gravitational settling velocity. Smaller particles that are affected more by atmospheric turbulence than gravity can travel great distances before depositing. Deposition of these smaller particles is affected not only by particle size and density, atmospheric turbulence and precipitation, but also by the nature of the plant surfaces that the particles land on.

Contaminants may also be deposited through human activities such as irrigation. The typical irrigation cycle can be from one to many hours. Once plant surfaces are initially wetted, subsequent water flows to soil as run-off. The question is how much of a particular element can be entrained, adsorbed, and/or absorbed during an irrigation event. Nair (Nair et al. 1996) found in rainfall simulation experiments that larger particles tended to adhere to the surface of vegetation and that subsequent rainfall was ineffective in removing what had interacted with the leaf surface during an initial rain event. For anions, like various forms of iodine, the radionuclides followed the water. But, if drying

occurred, then subsequent rainfall was ineffective in removing iodine from the leaf surface. This would likely be true for chemical forms having low to intermediate absorption rates and for elements with low adsorption rates, or with high absorption rates with limited leaf-surface binding sites. It may not be true for elements with exceptionally high bioavailability, as would possibly be the case for technetium and nickel.

Many models, e.g., RESRAD, MEPAS, and CAP88-PC (Chaki and Parks 2000), use a constant dry interception fraction of 0.25. The MILDOS code uses 0.2. The IAEA (2001) Safety Standard model uses a mass interception factor, defined as the fraction of deposited activity intercepted by the edible portion of vegetation per unit mass (or mass interception factor, m<sup>2</sup>/kg), which is set equal to 3 m<sup>2</sup>/kg (dry weight) for forages and 0.3 m<sup>2</sup>/kg (wet weight) for foods consumed directly by humans.

For a specific deposition event, the interception fraction is higher the more developed the plant canopy, which is accompanied with an increasing contact surface between the falling rain and the plant surface. There are two principal approaches to parameterize the plant development, the standing biomass per unit soil surface area and the leaf area per unit soil surface area (leaf area index). The advantage of the standing biomass is that it is easy to determine. The sample has simply to be taken from a known area of ground, dried, and the dry mass has to be determined. However, the biomass does not really represent the size of the interface rain/plant, which is the case for the leaf index. Whereas in the first period of growth, in general a good relationship between biomass and leaf area can be observed, the correlation fades away towards the end of growth. Then, the biomass still increases due to growth of storage organs as seeds or tubers while the leaf area already decreases substantially due to dying off of the foliage. The disadvantage of the leaf area index is that quantification is a complicated determination that requires specific optical devices which are usually not available. However, a wide variety of data on leaf area indices for terrestrial systems has recently become available from NASA satellite imagery as a result of their involvement in carbon cycle and climate research, which may simplify this problem.

Models that at least account for biomass have been proposed. An empirical relationship between biomass and interception fraction from atmospheric dry deposition was originally suggested by Chamberlain (1967). This model has been expanded by Pinder et al. (1988) for grasses and other species. The following form of the equation is suggested:

$$r_{dc} = 1 - e^{-AY_c f_c} \quad (8)$$

where

$r_{dc}$  = interception fraction for atmospheric dry deposition to crop type c (dimensionless)

A = empirical constant, about 2.9 for grasses, leafy vegetables, and grains, and 3.6 for fruits and other vegetables

$Y_c$  = standing biomass of the growing vegetation for crop type c (kg wet weight/m<sup>2</sup>)

$f_c$  = dry-to-wet weight biomass ratio for crop type c (kg dry weight per kg wet weight).

The dry-to-wet ratio is required because the Pinder formulations are given in terms of dry biomass. This formulation results in the need to define the growing biomass as well as the harvested yield.

The interception of material in irrigation water is not well studied. Thus, a default value of 0.25 is often used for all materials deposited on all plant types by irrigation. The results described above for irrigation are probably applicable. An empirical equation for the interception fraction,  $r_w$ , used in the Yucca Mountain model ERMYN (Wu 2003) derived from data from Hoffman et al. (1989), which is based on the results of experiments with  $^7\text{Be}$  and  $^{131}\text{I}$ , is expressed as

$$r_{wc} = K_1 Y_c^{K_2} I A_c^{K_3} I^{K_4} \quad (9)$$

where

- $r_{wc}$  = interception fraction of irrigation water for crop type  $c$  (dimensionless);
- $K_1, K_2, K_3,$  and  $K_4$  = empirical constants ( $K_1$  is in units of  $(\text{kg}/\text{m}^2)^{-K_2} (\text{mm})^{-K_3} (\text{cm}/\text{hr})^{-K_4}$ , and  $K_2, K_3$  and  $K_4$  are dimensionless);
- $Y_c$  = standing biomass of crop type  $c$  ( $\text{kg}$  dry weight/ $\text{m}^2$ );
- $I A_c$  = amount of irrigation per application event for crop type  $c$  ( $\text{mm}$ );
- $I$  = irrigation intensity (value in units of  $\text{cm}/\text{hr}$ ).

Because this is a regression equation from experimental data, values for the input parameters must be used in the units specified above. The empirical constants in this equation were developed based on given parameter units for standing biomass, irrigation amount, and irrigation intensity, depend on the plant type and contaminant form. The recommended values (Hoffman et al. 1989) are:

- |   |  |
|---|--|
| $K_1 = 2.29$ for beryllium ( $\text{Be}^+$ );   | $K_1 = 1.54$ for iodine ( $\text{I}^-$ )   |
| $K_2 = 0.695$ for beryllium ( $\text{Be}^+$ );  | $K_2 = 0.697$ for iodine ( $\text{I}^-$ )  |
| $K_3 = -0.29$ for beryllium ( $\text{Be}^+$ );  | $K_3 = -0.909$ for iodine ( $\text{I}^-$ ) |
| $K_4 = -0.341$ for beryllium ( $\text{Be}^+$ ); | $K_4 = -0.049$ for iodine ( $\text{I}^-$ ) |

The interception fraction for wet deposition can be also evaluated as a function of the rainfall rate and standing biomass based on experimental observations of Prohl and Hoffman (1993). The data by Prohl and Hoffman were used to develop relationships for anions, cations, and for insoluble particles for the GENII Version 2 application (Napier et al. 2002). For anions, such as iodide and sulfate, the interception fraction is evaluated as follows:

$$r_{wc} = 2.3 Y_c f_c R^{-0.92} \quad (10)$$

where

$r_{wc}$  = interception fraction from wet deposition to crop type c (dimensionless)

$Y_c$  = standing biomass of the growing vegetation for crop type c (kg wet weight/m<sup>2</sup>)

$f_c$  = dry-to-wet weight biomass ratio for crop type c (kg dry weight per kg wet weight)

$R$  = rainfall rate (mm/d),

and the numbers are regression parameters.

For cations and particles, Prohl and Hoffman (1993) presented the following relationship based on experiments using microspheres (essentially the same data as Hoffman et al. 1989):

$$r_{wc} = 2.95 Y_c f_c R^{-0.191} \quad (11)$$

where terms are as previously defined. In both equations, the numerical term is a fitting parameter with units of m<sup>2</sup>/kg. A potential disadvantage of this approach based on biomass is that it is intended primarily for leafy-type produce; its indiscriminate use could result in the more massive fruits such as apples, melons, tomatoes intercepting more irrigation water than the less massive but more expansive (e.g., surface area, leaf index) lettuce or spinach plant. The selection of which biomass to use is important.

Beyeler et al. (1999) also evaluated the results of Prohl and Hoffman (1993). Beyeler et al. (1999) decided that it was difficult to assign anion/cation status without site-specific knowledge of groundwater chemistry and elected to use a single range of interception fractions. They recommend a central value of about 0.35 within a uniform range of from 0.1 to 0.6 for all crop types.

The ECOSYS code uses a formulation based on Leaf Area Index (LAI) and the water-storage capacity of plant leaves. Buildup of the water film on the leaves during wetting events, the total amount of water deposited, and the radionuclide's ability to be fixed on the leaf (another interpretation of the data of Prohl and Hoffman) are considered. In this formulation, the wet interception fraction is estimated as

$$r_{wc} = LAI_i S_i / R \left[ 1 - \exp\left(\frac{-\ln 2}{3 S_i}\right) R \right] \quad (12)$$

where LAI is the leaf area index of plant i,  $S_i$  is the retention coefficient for plant type i (mm), and R is the amount of rainfall (mm).

If this equation results in an interception fraction greater than one, the value is set to one. Muller and Prohl (1993) provide estimates for the retention coefficient. As for the modified Chamberlain model described above, these vary for grasses, cereals, and corn, and for other plants.

Measured interception fractions for a limited number of radionuclides are becoming available for Asian food types not usually included in American risk assessments such as rice, radish, soybeans, and Chinese cabbage (Choi et al. 1998; 2001; 2002; Lim et al. 2001).

Many investigators have commented that the plant biomass and the interception fraction are directly correlated, and a more stable estimate of interception may be obtained using the quotient  $r/Y$  as a single variable (IAEA 1982). The interception fraction  $r$  rapidly increases with the increasing plant biomass during the first half of the growing period and thereafter it is almost constant or even decreases (Vandecasteele et al. 2001). In contrast to  $r$ , the biomass-normalized interception fraction  $r/Y$  decreases as plants grow from the beginning. The  $r/Y$  values at the early growth stages are conspicuously high, because most of the young leaves are directly exposed to deposition with comparatively low mass-to-area ratios. Accordingly, the  $r/Y$  is a useful parameter in estimating the initial plant concentration after an accidental deposition but it is unnecessary during routine conditions for which such an initial concentration cannot, and need not, be calculated. For assessing continuous deposition, instead, an averaged value of  $r/Y$  can be used, or simply a lumped parameter that is a ratio between the deposition rate and the steady state concentration per gram of plant material.

Kinnersley and Scott (2001) suggest that the complex three-dimensional structure of crop canopies is such that no purely mechanistic model yet exists which can predict the turbulent transport of airborne particulate contamination into, and onto, them. While theoretical understanding of the processes involved in particle dry deposition has advanced considerably in the last decade or so, a gap remains between theory and the practical prediction of particle deposition, particularly under non-laboratory conditions. Bonka and Horn (1983) produced a thorough review of the mechanisms governing deposition of aerosols to vegetation, from which it can be deduced that analytical deposition models are currently limited by uncertainty in the aerosol collection efficiency term. This term must be determined experimentally for canopies consisting of anything but regular arrays of the simplest geometrical shapes. The review describes the model of Slinn (1982), in which the maximum possible deposition velocity (assuming the canopy to be a perfect sink) is attenuated by a term, ultimately empirical in derivation, allowing for the particle collection efficiency of the canopy. Data requirements for this model include a “vegetative hair fraction”, a “small vegetative diameter” and a “large vegetative diameter”. Another well-known model for deposition to vegetation is that of Sehmel and Hodgson (1978). This model takes into account the underlying physics of particle interception in some detail, including amongst its input data requirements details of the turbulence, boundary layer and surface layer resistances, but again it is ultimately dependent on empirical terms determined in a wind tunnel. A third approach has been adopted in the Imperial College (IC) model. This is based on multiple regression analyses of a considerable database of deposition values generated under wind tunnel conditions. The regressions use the three parameters - particle size, turbulence levels and canopy size - which theory suggests should dominate control of the deposition process. These three models give broadly similar results, which are compatible with the few reliable field measurements of deposition to crop canopies which currently exist. Despite data

requirements which can become complex, current deposition models ultimately retain a dependence upon experimentally determined factors. However, there is a lack of reliable data of this sort. This is particularly so for the interception of particles less than 40  $\mu\text{m}$  in diameter (Chamberlain & Garland, 1991) and for larger canopies such as those of fruit crops.

For prolonged deposition to vegetation (as occurred after weapons testing), Chamberlain introduced the concept of normalized specific activity (NSA), defined as the ratio of radionuclide concentration in foliage ( $\text{Bq/kg}$  dry weight) to the rate of deposition to ground ( $\text{Bq/m}^2/\text{d}$ ). A value of  $40 \text{ m}^2/\text{kg}$  was considered appropriate for herbage in good growing conditions; much lower values have been found for grains (Simmonds and Linsley 1982). NSA values are much higher for poor growing conditions and some types of native vegetation (Martin and Coughtrey 1982).

### **3.3.2 Modeling Contamination of Plant Surfaces with Soil**

Plants may acquire surficial contamination in the form of dust or soil particles; the source is the soil in which the plant is growing. Certain types of plants, such as leeks, are frequently associated with soil carryover.

Contaminants sorbed to soil particles may be resuspended by wind, human, animal, mechanical disturbances and redeposit locally on the plant surfaces. Such redeposition occurs when soil is displaced by wind (Anspaugh et al. 1975), animal disturbance (Hinton et al. 1995; Sumerling et al. 1984) or mechanical disturbance (Milham et al. 1976; Pinder and McLeod 1988; 1989). Another physical mechanism for contamination of plant surfaces with soil particles is raindrop splash (Dreicer et al. 1984). In the 30-km zone around Chernobyl, rainsplash of contaminated soil contributed significantly to overall plant contamination, with 60 to 70% of the activity concentrated on the basal parts (10 to 20 cm above the soil surface) of cereals and perennial grasses (Kryshev 1992).

Plant contamination may also be caused by mechanical disturbances that raise dust, such as occur during weeding and harvesting. Adriano et al. (1982) and Pinder and McLeod (1988; 1989) have reported this to be a major source of plutonium contamination in grain grown on contaminated soils. Plants may acquire surficial contamination in the form of dust or splashed-up soil; the source is the soil in which the plant is growing. Soil-to-plant transfer values via the roots for some actinide elements are on the order of  $10^{-6}$  to  $10^{-5}$  ( $\text{Bq/kg}$  dry crop)/( $\text{Bq/kg}$  dry soil). This very low transfer from soil via the roots means that the contamination of a crop due to contaminated soil resuspended or splashed on the leaf surface may easily dominate the plant physiological uptake. A contamination of only 1 mg soil per kg fresh weight crop can cause an apparent transfer value of  $10^{-5}$  for splash-up and resuspension transfer factors. Thus this pathway may be of importance in some analyses of radionuclides that are biologically discriminated against.

The simplest approach is to define a soil adhesion quantity for each type of crop plant. This combines the effects of interception of locally-resuspended material and the splashing of rain and irrigation water droplets, and allows averaging of the effects of



various factors such as height and type of plant canopy, wind, rain, and soil type. This approach is suggested by IAEA (1994) and used by Kennedy and Streng (1992). The later IAEA model (IAEA 2001) combines this mechanism with the soil-to-plant transfer into a single transfer parameter (see below) - to account implicitly for soil adhesion, a minimum value of 0.1 is assigned to the soil-to-plant transfer factor for forages and 0.001 to the transfer factor for human foods. However, this approach is not recommended by the International Union of Radioecologists, who state that “Values of soil adhesion depend so much on environmental conditions that it is not realistic to propose a default value. However, an estimate for a low value of soil adhesion is 4 g soil per kg dry vegetation for leaves and grass taken from 40 cm or more above the soil surface. Below 40 cm, 10 g per kg might be a reasonable estimate for leaves and grass. Considerably lower values are expected for products such as grain, which are protected by plant parts which are not consumed. A reasonable upper limit for soil adhesion is 250 g per kg.”

The processes of suspension and deposition are parameterized as consisting of resuspension of contaminated soil followed by local redeposition in some models. This is the approach taken by the GENII family of codes, as well as the RESRAD family of codes, PATHWAY, and ECOSYS. The resuspension factor for food-crop and animal-product pathways is representative of conditions on farmland, which may be different from the resuspension factor for the inhalation exposure pathway. Farmland would be expected to be tilled and have soil generally looser than soil for the general residential exposure situations.

Many models circumvent the necessity of dealing with soil adhesion by specifying amounts of soil ingested by humans and domestic animals. However, this approach then neglects the potential for transfer from the plant surface to the internal portions of the plant. It also complicates the modeling of weathering of deposited material off of plant surfaces, discussed below.

### **3.3.3 Modeling Weathering from Plant Surfaces**

Material deposited on vegetation may be lost from the plant through a variety of different processes in addition to radioactive decay, such as removal by wind or rain, volatilization, plant senescence (leaf drop), and others. Wind is thought to dominate the field-loss process for particulate contamination, with the contribution of rainfall being significantly less (Kinnersley and Scott 2001). In fast-growing vegetation, a decrease in concentration may be seen as a result of dilution by new vegetative material, and ascribed to a removal process.

Losses from plant surfaces are frequently represented by a weathering rate constant,  $\lambda_{wi}$ . Several processes may be involved. Some measurements of this process have implicitly included plant growth. Volatile materials may evaporate back into the air. If precipitation occurs, or there is heavy irrigation, physical washoff processes may be present. Because of the combination of potential processes, this phenomenon is sometimes called *field loss*.

The lack of detailed measurements of contamination loss over time has led to the use of a simple exponential model for weathering (Chadwick and Chamberlain 1970). The NRC's Regulatory Guide 1.109 (NRC 1977) uses a weathering half-time of 14 days. The default-rate constant in GENII, and in PATHWAY for all radionuclides other than iodines, is based on a weathering half time of 14 days. The values in IAEA Safety Series 57 (IAEA 1982) are 15 days for particulates and 10 days for iodines; that used in the IAEA Safety Reports Series 19 (IAEA 2001) is 14 days. The value in RESRAD (20 year<sup>-1</sup>) is equivalent to a half-time of about 12.6 days. ECOSYS uses a half-time of 25 days; the ECOSYS model attempts to account also for dilution via plant growth in pasture grasses. When combined, the ECOSYS results are in the range of 10 to 16 days. Statistical analysis of data from literature shows a log-normal distribution for half-lives and gives a default value (geometric mean) for estimating the removal of radionuclides from vegetation based on a half-life of 17 days<sup>1</sup>.

Kinnersly and Scott (2001) report that the dominant weathering/field loss processes are the resuspension of particles into the air by atmospheric turbulence, and loss through biological processes such as leaf-loss and cuticular wax-shedding. In recent years much higher resolution studies have been carried out (Ertel, Voigt, and Paretzke 1989; Fraley, Chaveg, and Markham 1993; Kirchner 1994; Kinnersley et al. 1996), and it is now clear that field loss is best described in terms of rapid and slow loss components. The slow loss component can be so slow as to be negligible, such that it can be treated as a fixed fraction of contamination that will not be lost over time, yielding a model (not including radioactive decay) of the form (Kinnersley et al. 1997):

$$A_t = A_r + (A_0 + A_r) e^{-\lambda t} \quad (13)$$

where  $A_0$  is the initial level of contamination,  $A_t$  is the level of contamination at time  $t$ ,  $A_r$  is the fraction of the original deposit not subject to loss, and  $\lambda$  is the time constant for the fraction of initial deposit subject to exponential loss. This particular empirical formulation may also represent the fraction of material that is absorbed and/or translocated by the plant versus that which remains on the surface and is available for weathering (see the next section).

The simple one-exponential approach or the NSA are the easiest to implement in situations with continuing deposition; the Kinnersly et al. (1997) formulation is better suited to single deposition events. Similarly, models such as that in ECOSYS that attempt to account for dilution of contamination in growing crops are better suited for single depositions; a long-term average field loss is better suited for continuous depositions where the intended use is determination of ultimate concentration at harvest.

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<sup>1</sup> E. Leclerc, unpublished data prepared for the IAEA's Environmental Modeling for Risk Assessment coordinated research program.

### 3.3.4 Modeling Foliar Absorption and Translocation

Above-ground parts of plants (leaves, blossoms, fruits, and branches) can directly absorb water and minerals as well as soluble radionuclides. This process is called foliar absorption. Some radionuclides, once absorbed into the body of the plant, will remain at or very near the location from which they were absorbed. Other radionuclides may be transported via the phloem of the plant to other plant structures, organs, or fruits. This process is referred to as translocation.

The processes of absorption and translocation are dependent on the physical and chemical forms of the contaminating radionuclides. Materials incorporated in relatively large particles are not easily absorbed, and so are unlikely to be affected by translocation. Contaminants in ionic form are more likely to be absorbed.

The foliar absorption of  $^{137}\text{Cs}$  that was sorbed to resuspended soil particles was quantified at two sites contaminated by the Chernobyl accident. Although measurable, this foliar absorption was considered inconsequential relative to other plant contamination pathways, which led to the recommendation that it not be considered a critical pathway in routine radionuclide transport models (Hinton et al. 1996).

Once absorbed, the processes of weathering from the plant are changed; transport by wind and rain are much reduced, although physical losses such as leaf drop will remain.

The translocation factor indicates the fraction of total deposition to plant surfaces that is incorporated into edible parts of the plant. This mechanism incorporates two processes - Initial uptake by the leaf and subsequent translocation throughout the plant is dependent on the cuticular properties of the plant, its wetability, strength of absorption onto the cuticle, the radionuclide in question, its solubility, size, speciation, and the time between contamination and harvest. The importance of uptake and translocation will be dependent on what part of the crop is to be eaten and what species of radionuclide has been intercepted.

In many models, e.g., GENII (Napier et al. 1988; 2002), RESRAD (Yu et al. 2002), and CAP88-PC (Cahki and Parks 2000), the translocation factor is defined to be the fraction of total deposition to plant surfaces that is incorporated into edible parts of the plant. A value of 1 is assumed for leafy vegetables and forage crops where the whole above-ground portion of the plant is eaten, and 0.1 for all other vegetation. The value of 0.1 is assigned on the basis of very little information and is assumed to be an upper bound.

The IAEA (1994) provides two alternative definitions for the translocation factor, as

$$T = \frac{\text{activity concentration in the edible parts at harvest (Bq / kg)}}{\text{activity retained on 1 m}^2 \text{ of foliage at the time of deposition (Bq / m}^2 \text{ foliage)}} \quad (14)$$

$$T_a = \frac{\text{activity concentration in the edible parts of 1 m}^2 \text{ at harvest (Bq / m}^2 \text{)}}{\text{activity retained on 1 m}^2 \text{ of foliage at the time of deposition (Bq / m}^2 \text{ foliage)}} \quad (15)$$

Note that each of these formulations has different units. IAEA (1994) recommends using the first approach, normalized to the yield in kilograms, as having less measurement variability. A later IAEA safety series model (IAEA 2001) includes translocation implicitly in the definition of the foliar interception factor used in that (intentionally conservative) model.

The relative rates of absorption of chemical elements from cuticular structures are difficult to quantify. First, absorptive capacity differs from root processes in that there are apoplastic/tricome routes of entry into the foliar interior, which are less discriminating (with respect to species discrimination) than for roots. Secondly, the transfer of individual elements is very dependent on chemical form and stability. For example, free cations and stable anions may be readily absorbed, while reactive or unstable species may not be absorbed, or absorbed to a lesser extent. The transfer process is basically a molecular level interfacial film flow. Thus, only molecular-sized materials can move. Finally, refractory particles, i.e., oxides, carbonates, and hydroxides, of any size will not likely be transferred to the tissue interior unless a significant rate of solubilization is occurring, e.g., complexation agents in irrigation waters. These particles can and will, however, be entrained within the complex cuticular structures associated with most surface foliage.

There has been much research into agricultural chemicals applied to foliage, some of which has been performed with radioactive tracers (e.g. starting with Tukey and Wittwer 1956; Tukey et al. 1956). Potassium and sodium were found to be rapidly absorbed and highly mobile. Phosphorus, sulphur, and chlorine were absorbed at a slower rate, but were also mobile and were transported at a rapid rate. Manganese, zinc, copper, and molybdenum were found to be slightly mobile. Calcium, strontium, barium, iron, and magnesium were readily absorbed but did not move out of the leaf to which they were applied. McFarlane and Berry (1974) reported that the rate at which cations penetrate the cuticle is inversely related to the radius of the hydrated ion, which occurs in the order Cs>Rb>K>Na>Li>Mg>Sr>Ca. Aarkrog (1975) reported that different radionuclides were translocated to a varying rate and degree in the order:

Zn>Fe>Cs>Co>Mn>Na>Cd>Sb>Cr>Sr~Ce~Ru>Ba~Hg~Pb. This pattern may vary with the chemical state of the element and the presence of complexing agents. The solution sprayed on the crops had a pH of 1.6 and solubilities would be different from those in actual rainfall. Also, only the activity rather than the species of the radionuclides applied is known so the results may not be representative of a realistic situation. Bukovac and Wittwer (1957) suggested mobility in the order:

Rb>Na>K>P>Cl>S>Zn>Cu>Mn>Fe>Mo>Ca>Sr>Ba. Distribution of Rb, Na, K, Cl, P, S and possibly Zn appeared proportional to the metabolic activity of various plant tissues. Foliar absorption is time dependent, i.e., the longer a contaminant is retained on a leaf the greater is its absorption. Higher absorption rates have been documented in younger leaves (Hull et al., 1975), and significant percentages of the total cesium intercepted by foliage can be absorbed within a few days (Carini and Bengtsson 2001).

While little data exist to demonstrate or quantify foliar absorption processes and particularly rates of transfer, a list of probable behaviors for priority radionuclides is given in Robertson et al. 2003. This categorization is based on known behavior in soil/plant systems and likely chemical speciation in groundwaters. This listing is based on many assumptions. Although much data exist for nickel and technetium, little is known of the foliar routes of entry for any of these elements. The intermediate category is questionable, but the limited data available would indicate that there is a need for study of particularly uranium, iodine, and americium. In the proposed Low Transfer category, little is known about the behavior of beryllium and niobium.

In the RODOS code (Müller, Gering, and Pröhl 2004), the translocation process is quantified by a translocation factor defined as the fraction of the activity deposited on the foliage being transferred to the edible parts of the plants until harvest. It is dependent on the element, the plant type, and the time between deposition and harvest. In this formulation, only a single deposition is assumed, so the time from deposition to harvest is important. The elements are grouped into two sets, mobile and immobile; the most reliable data for translocation are available for Cs and Sr. Therefore, as long as no other quantitative data are available, the translocation of mobile elements assumes that they behave like Cs, except for Mn, for which translocation factors are assumed to be lower by a factor 0.6 than for Cs. Strontium is considered to be representative for immobile elements. The RODOS assignments are also shown in Table 2; it can be seen that there is some coincidence of interpretation with Robertson et al. (e.g., Tc, Pu, Sr), and some disagreement (e.g., Np). Note that the RODOS implementation tends to focus on radionuclides with shorter half-lives.

**Table 2.** Probable Bioavailability/Cuticular Transfer of Selected Elements. This refers to foliar uptake, and not specifically phloem mobility and redistribution (adapted from Robertson et al. 2003 and Müller, Gering, and Pröhl 2004)

High Transfer	Intermediate Transfer	Low Transfer
Np	Am	Pu
Tc	Cs	Th
Ni	U	Be
	Cm	Nb
	I	Sr
	Se	
	Ag	
	Sn	
Mobile		Immobile
Co, Cs, I, Mn, Mo, Na, Rb, Sb, Tc, Te		Ag, Am, Ba, Ce, Cm, La, Nb, Nd, Np, Pr, Pu, Rh, Ru, Sr, Y, Zr

There is unfortunately no in-depth compilation of the rates of retention nor uptake of radionuclides into the plant via foliar contamination. Kinnersley and Scott (2001) undertook model parameterization for wet and dry deposition processes to plant surfaces. This theoretical parameterization is reasonable, but there is a nearly total lack of supporting data. The IAEA (1994) provides a limited number of rate constants for a few elements for a few crop types.

Translocation and weathering are, in some senses, competing processes for transport of material off of plant surfaces. This is often overlooked in model formulation: material translocated may be affected by the weathering terms if not separately considered. The ERB2A model from the IAEA BIOMASS program (IAEA 2003) and the Yucca Mountain ERMYN model (Wu 2003) are examples that specifically separate these two effects. This is done by developing a fraction of the surface contamination to which weathering processes apply – the difference between the total and the translocation fraction. These models do this by adding additional parameters in the following fashion:

$$C_{c,irrig} = r_{wc} S \left( \frac{(1 - F_{abs}) e^{-\lambda_w T} F_{p3}}{Y_c} + \frac{F_{abs} F_{p2} F_{trans}}{Y_c} \right) \quad (16)$$

- $C_{c,irrig}$  = radionuclide concentration in the edible part of the crop due to irrigation water deposited on crop leaf surface (Bq/kg fresh weight of crop)
- $r_{wc}$  = fraction of radionuclides in spray irrigation water initially deposited on standing biomass (dimensionless)
- $S$  = annual irrigation deposition (Bq m<sup>-2</sup>);
- $F_{trans}$  = fraction of absorbed activity translocated to edible portions of the plant by the time of harvest (translocation fraction)
- $F_{abs}$  = fraction of intercepted radionuclide initially deposited onto plant surfaces absorbed from external surfaces into plant tissues (dimensionless)
- $F_{p2}$  = fraction of internal contamination in the edible parts of plants at harvest that is retained after food processing (dimensionless)
- $F_{p3}$  = fraction of external contamination from interception retained on edible parts of the plant after food processing (dimensionless)
- $\lambda_w$  = removal (weathering) rate for radionuclides deposited on plant surfaces by irrigation (weathering processes include mechanical weathering, wash-off, and leaf fall) (yr<sup>-1</sup>)
- $T$  = interval between irrigation and harvest (yr)
- $Y_c$  = biomass of the crop (kg fresh weight/m<sup>2</sup>).

### 3.3.5 Modeling Soil-to-Plant Uptake

Terrestrial plants, as sessile organisms, have adapted to derive essential nutrients from their environments. Plants absorb nutrients through their roots and transport them via the phloem to active portions of the plant. Biotic factors are likely the source of much of the variability seen in concentrations of contaminants in plants. This variability results from the nature of the sessile terrestrial plant and its relationship with its environment: the need to compete and acquire specific nutrient species from soils (or to avoid uptake of

excesses of potentially toxic materials), the need of individual plant types for specific levels of individual nutrients, and the use of certain metals in tissues as agents to discourage herbivory. Thus, the uptake is affected by the following biotic factors:

1. the plant-available concentration in soils within the rhizosphere (the soil zone that surrounds and is influenced by the roots of plants), which is governed by soil adsorption processes, chemical solubility, microbial/fungal activity, and stability of the chemical complexes
2. the chemical nature and stability of the cation-anion/complex with respect to the plant's capability to metabolically alter and/or absorb the elemental form into the plant
3. a series of plant adaptive/evolutionary processes for survival; this can include, but not be limited to, protective root processes (exclusion or complexation of an ion for detoxification, sequestration within the plant to regulate both ion levels and for detoxification, redox to alter solubility and transport when necessary, organic complexation in the case of all but mono-cationic elements, and uptake capacity being dependent on metabolic needs).

Under these chemical and biotic constraints, uptake can be expected to vary based on source term, kinetics of solubilization/speciation, the relative ability of a plant to view a non-nutrient ion as an analogue to a nutrient species, and the relative need by the individual plant genus/species for specific levels of a particular ion.

Transfer of radionuclides from soils into plants is one of the key mechanisms for long-term contamination of the human food chain. There are several methods that may potentially be used. The most common is through the use of soil-to-plant transfer factors (or concentration ratios), but others may also be used.

#### **3.3.5.1 Development of Soil-to-Plant Transfer Factors**

Research on the relationship between radionuclides in soil and in the plants grown in that soil began soon after the establishment of the major radionuclide production centers of the Manhattan Engineer District. In the early period (through the early 1950s), the potential for root uptake was recognized (Cline and Porter 1951) but not appreciated; the main concern was foliar deposition (Parker 1955). However, some studies were originally undertaken at the Hanford Site to investigate this pathway (e.g., Seiders, Cline, and Rediske 1955). Hanford's Botany Field Station was established in 1949 and used Columbia River water from downstream of the reactor outfalls for an irrigation source. Minor increases in gross radioactivity were noted in crops in 1949 and 1950 (Cline and Porter 1951), but individual radionuclides were not identified. However, by 1953, work had been done at Hanford to determine the *concentration factor* of several key radionuclides from soils contaminated elsewhere by fallout (Rediske and Selders 1953a; 1953b; Selders, Rediske, and Palmer 1953).

Weapons testing was conducted in the United States and elsewhere and a number of countries became concerned about world-wide fallout, particularly of  $^{90}\text{Sr}$ . Results of

surveys of contaminated biota on contaminated islands in the Pacific Proving Grounds were first published in 1956 (Atomic Energy Commission 1956). Meanwhile, agricultural scientists in the U.S. and Great Britain began investigations and field studies on the factors governing uptake of  $^{90}\text{Sr}$  and its transport into food chains that led directly to humans. Concern about uptake through plant roots, as opposed to foliar deposition from fallout, first arose at Oak Ridge in the mid-1950s following the draining of White Oak Lake (Auerbach and Reichle 1999; Stannard 1988). Recognition of root uptake of selected contaminants (primarily related to situations concerning waste disposal in near-surface soils) was wide-spread by the early 1960s; interestingly, however, papers presented at the first major conference on radioecology (Shultz and Klement 1963) contain data on radionuclide concentrations in plants – but no soil-to-plant concentration factors per se. Senate hearings in 1963 focused on radioactive fallout from the weapons tests and food chain contamination (Stannard 1988). The initial attempts to model uptake of radionuclides by plants were driven by unanticipated contamination events following atmospheric nuclear weapons testing.

Menzel (1965) presented results of a literature study of various soil science experiments of soil to plant transfer at the 1965 Hanford Biology Symposium. This is one of the first uses of the term “concentration factor” with regard to plant/soil ratio. Menzel used this ratio to summarize a large amount of data for several elements (in terms of ppm of dry plant per ppm of dry soil). He was more interested in exploring the effects of radiation on the plants and the mixing of radionuclides in soil by plants than on the actual uptake. (At this same Symposium, Mistry et al. (1965) gave detailed measurements of various herbs, shrubs, trees, fruits, and tubers grown in areas of high natural background – but did not provide either concentration ratios or soil measurements from which concentration ratios could be computed.)

The concept of using nuclear explosives for excavation, particularly of a sea-level canal across the Isthmus of Panama, with the potential for associated fallout, led to several attempts to predict the potential environmental affect. Martin et al. (1970, as reprinted in Klement (1982)) proposed a series of physics-based, differential equations to describe the transport of radioactive contaminants resulting from canal-building fallout. This effort included estimation of transfer of radionuclides from soil water into plants that was related to the growth rate of the plant and the stable element content of the plant. No examples are given for this mechanistic approach; it does not appear to have been too successful.

The document series *Prediction of the maximum dosage to man from the fallout of nuclear devices* (Tamplin 1967; Ng and Thompson 1966; Burton and Pratt 1968; Ng et al. 1968) also came about because of concerns about predicting doses from canal-building fallout. This series was intended to provide input to design of nuclear explosives to minimize the impacts of fallout; one segment of the calculation was to develop a “unit-rad” value that related normalized fallout amounts by radionuclide to committed internal doses. While the “unit-rad” calculation did not directly involve estimation of radionuclide transfer from soil to plants, a series of appendices to Ng et al. (1968) did describe how global average concentrations of stable elements in soils and plants could



be ratioed to give an estimate of uptake of radionuclides in plants from contaminated soils. In this description, Ng used the symbol ( $C_p/C_s$ ) for what he called the plant-to-soil concentration ratio. The data in this report became the basis for many later models and calculations, such as NRC Regulatory Guide 1.109 (1977).

The concept pioneered by Ng has been given many names over the years.

- *Plant-to-soil concentration ratio* – a generic term in ecology, used by Whicker and Schultz (1982), Peterson (1983), Faw and Shultis (1999), and others;
- *Soil-to-plant transfer factor* – used to describe the direction of radionuclide migration “from soil into plants”, adopted by Martin Frissell and the International Union of Radioecologists (1989), IAEA (1994), and others;
- *Vegetable/soil transfer factor* – used in documents related to the RESRAD family of computer codes (e.g., Yang, Biwer, and Yu 1993; Yu et al. 2001);
- *Soil-to-plant concentration ratio* – a variant used by NCRP (1984);
- *Soil-to-plant concentration factor* – a variant used by IAEA (1982), Ng, Colsher, and Thompson (1982);
- *Concentration factor* – a variant used by Soldat in Fletcher and Dotson (1971); Miller et al. (1980);
- *Transfer ratio* – Stannard (1988);
- *Concentration ratio* – a commonly-used variant (e.g., Cataldo, Wildung, and Garland 1983; Napier 2003; numerous others);
- *Plant uptake factor* – Eisenbud (1987);
- *Relative ratio* – Dahlman and Van Voris (1976);
- *Uptake ratio* – Grummitt (1976);
- *Plant bioconcentration factor* – a definition more usually used for aquatic uptake processes, but sometimes used with terrestrial vegetation (e.g., Wolterbeek, van der Meer, and Dielemans 2000);
- *Plant bioaccumulation factor* - a definition more usually used for aquatic uptake processes, but sometimes used with terrestrial vegetation (e.g., Wolterbeek, van der Meer, and Dielemans 2000)
- *Discrimination factor* – the name sometimes given to the ratio of the steady state output to input of a compartment model, when applied to the ratio of concentration in soil to concentration in plant, also *accumulation factor* (Peterson 1983; Faw and Shultis 1999).

All of these concepts are the same; the notation generally used is  $B_v$  for the ratio of the concentration of an element in a plant of interest to the concentration in the source soil. The transfer factor applies to long-term, chronic exposures and is ideally measured at equilibrium. Transfer factors are used in risk assessments to estimate the amount of radioactivity that could be present in a food crop based on the calculated concentration in the source soil. By calculating the concentration in the food, the total intake can be estimated and a dose calculated as a result of the annual intake. In terms of radionuclides, the transfer factor is used to calculate how many becquerels per kilogram of soil are transferred to the edible dry plant product (Bq per kg). Although the concept is occasionally questioned for some of its underlying assumptions (e.g., Centofani et al. 2005), it is the most commonly used approach in radiological environmental assessments.

The concentration ratio depends on the radionuclide, the soil type (which may include soil chemistry and concentrations of nutrients and analogues), and the plant type. The transfer factors are empirically derived; they are based on measurements made for various chemical forms of the radionuclide on selected types of plant in selected soil types. Experimental data are not available for all elements for all food types. Frequently, a few measurements on a very limited number of plant types are used to infer a transfer factor for all crops. Often, when no referenceable documents are available, data are derived based on chemical groupings in the periodic table of the elements, such that chemically similar elements are assigned similar values.

In researching concentration ratios, it is necessary to fully understand the nature of the experiment or natural system in which it was measured. The type of plant, the type of soil, the quantity of naturally occurring chemical analogues in the soil, and the stage of growth all influence the amount of contaminant absorbed into the plant through the roots. A large fraction of the observed variability may be due to the fact that analytical methods do not account for bioavailability of materials in soil – how much of the concentration measured in the soil is actually available to the plant and not bound to the soil matrix. In addition, it is important to know whether the concentration measurements are made in terms of wet or dry plant. Vegetation is a large percentage water, and the degree of turgor in the harvested plant has a strong influence in the measured concentration. The intent in most codes is to use a concentration ratio between dry soil and dry plant matter and to adjust to the full mass of the plant with a dry-to-wet mass ratio. When taking transfer factors from the literature, it is important to note whether the values are reported in wet or dry weight. Some researchers differentiate between the concentration ratio for plants consumed as feed by animals (designated as  $B_{v1}$  and given in units using dry weight of vegetation and soil) and for plants consumed fresh by humans (designated  $B_{v2}$  and given in units of fresh [or wet] weight of vegetation and dry weight of soil) (Miller et al. 1980). A conversion of these two concepts is given in Peterson (1983) as

$$B_{v2} = B_{v1} \left( \frac{DW}{FW} \right) \quad (17)$$

where DW is the dry weight and FW is the fresh weight.

Since the handbook prepared by Ng et al. (1968), numerous studies have been undertaken to quantify transfer factors (or concentration ratios) for specific chemical elements as a function of food type. These studies have been compiled in several publications. Most computer codes reference one or more of these compilations as the source of their transfer factors. Several frequently referenced compilations include the International Atomic Energy Agency's Technical Report Series #364, *Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Temperate Environments* (IAEA 1994). This document encompasses a wide variety of plant types and is the result of extensive background investigations. It is based on data compiled by the International Union of Radioecologists. Work is currently underway coordinated by the IAEA to update this document. A second frequently cited reference is the NUREG/CR-5512, *Residual Radioactive Contamination From Decommissioning: Technical Basis for Translating*

*Contamination Levels to Annual Total Effective Dose Equivalent* (Kennedy and Strenge 1992) because of its large set of data and traceable references. Other references include the National Council on Ionizing Radiation and Protection (NCRP) Report #123 (1996), *Screening Models for Releases of Radionuclides to Atmosphere, Surface Water, and Ground*, and the series of documents by Coughtrey and Thorne (1983), *Radionuclide Distribution and Transport in Terrestrial and Aquatic Ecosystems, Vols. 1-6*.

The National Council on Radiation Protection and Measurements developed a set of soil-to-plant transfer factors for screening applications (NCRP 1999), basing it largely on the International Atomic Energy Agency/International Union of Radioecologists (IAEA/IUR) publication (IUR 1989). However, the NCRP specifically assigned uncertainty ranges to the parameters in the form of geometric standard deviations for assumed lognormal distributions. The assigned geometrical standard deviations (GSDs) ranged from 2.5 to 3.0. However, the transfer factor values adopted by NCRP are described as being “close to the top of the measured range for most nuclides,” with higher values used for sparsely vegetated soils. Because the screening intent was to be conservative, the actual uncertainty range could be larger (with a higher possibility of lower values.) Biwer et al. (2000) adopted the NCRP ranges for use in the RESRAD family of codes. Beyeler et al. (1999) independently evaluated the CR values for the NUREG/CR-5512 model. They report GSDs ranging from a minimum of 2.47, based on 1250 measurements of 23 nuclides as reported by Sheppard and Evenden (1990), to as high as 9.5. Most nuclides were assigned the 2.47 default; interestingly, those radionuclides with some conflicting data often had higher GSDs than those with no data. Here, the GSD was used to represent experimental variability, not necessarily uncertainty in an annual average value that is representative of time and space averaging over some defined area. Somewhat different results were obtained using an expert elicitation process (Brown et al. 1997). It is apparent that there is a large potential variability in the CR values as a result of “numerous and complex underlying processes such as climate, growing conditions, plant metabolism, plant rooting traits, soil type, soil moisture, soil texture, and soil pH (Beyeler et al. 1999).”

Some models make additional assumptions involving root uptake and the surface soil layer when a clean non-contaminated layer of soil is applied over contaminated upper soil layer. The GENII family of codes (Napier et al. 1988; Napier 2003) and RESRAD family of codes (Yu et al. 2001) also allow the root uptake transfer factor to be modified if not all of the plants' roots are in the contaminated soil layer. The GENII and RESRAD codes assume that uptake is directly proportional to the fraction of roots growing in the contaminated zone. In GENII, this is accomplished using an input root-penetration factor, usually set to 1.0 for surface contamination and a lower value for contamination covered with a clean surface layer. A similar approach is taken in RESRAD, which derives cover and depth modifying factors as a function of the thickness of the clean overburden above a contaminated layer. Biwer et al. (2000) investigated rooting depths for various types of plants, and suggested that for stochastic simulations this be varied uniformly from 0.3 to 4.0 meters. However, Biwer et al. did not consider the actual appropriateness of the assumption of linearity of uptake with root fraction.

Root uptake is important for biologically active or mobile contaminants; other contaminants are discriminated against by biological systems, and their root uptake is minimal. For these types of radionuclides, other mechanisms may be the dominant ones for vegetation contamination. As discussed above, such mechanisms include simple physical contamination of the external surface of the plant, such as resuspension and deposition of dust or splashup of contaminated particles of soil during rain or irrigation episodes. In the recent IAEA recommendations (IAEA 2001), an aggregated transfer factor was developed that implicitly includes the soil adhesion term. To account implicitly for soil adhesion, a minimum value of 0.1 is assigned to the transfer factor for animal feeds and 0.001 to the concentration ratio for humans. The IAEA's (2001) concentration ratio for human foods is given in terms of wet weight. To differentiate these from the  $B_{v1}$  and  $B_{v2}$  terms defined above, the IAEA gives these the symbols  $F_{v1}$  and  $F_{v2}$ .

It is important to remember that, for all of the different definitions and applications listed here, the modeling of transfer of radionuclides from soils to plants is based entirely on empirical observation. For combinations of plant type and soil type that have not been directly observed, any factor used is at best an approximation.

The model of Baes (1982) and Sheppard (1985) attempted to correlate soil-to-plant concentration ratios with the distribution coefficient  $K_d$ . The approach, introduced by Baes (1982) and further developed by Sheppard (1985), presents the relationship between  $K_d$  and concentration ratio (CR) in the form

$$\ln K_d = a + b (\ln CR) \quad (18)$$

where  $a$  and  $b$  are constants,  $b$  having a negative value. Sheppard found that this model was generally able to estimate the CR to within about an order of magnitude (perhaps because the  $K_d$  is an implicit measure of bioavailability). Its use does require a measurement of the soil  $K_d$ , which is simpler to obtain than a site-specific estimate of the CR values; however, it is not specific to various types of crops. A related model specifically for cesium, introducing even more unknown parameters related to potassium concentrations, was attempted by Absalom et al. (1999). This type of approach has limited utility and has not caught on in the assessment community. However, it does indicate that in when quantifying uncertainty using Monte Carlo sampling procedures, the soil to plant uptake should be correlated with the soil  $K_d$ .

### 3.3.5.2 The Observed Ratio

A different approach to estimating uptake of radionuclides into plants (and animals, see below) is based on the observation that the presence of chemically similar stable elements in soils can have a significant impact on radionuclide uptake in plants. In soils having low concentrations of analogous stable elements, radionuclide uptake by plants may be enhanced, sometimes by orders of magnitude above the levels ordinarily found. Similarly, in soils with high concentrations of analogous elements, uptake may be significantly depressed. This effect has been characterized by the use of elemental ratios to predict the transport of radionuclides in the food chain. The ratio of the radionuclide-

to-stable-element concentration in one medium to the radionuclide-to-stable-element in another medium (for instance, soils and plants) has been named the observed ratio (OR) (Comar et al. 1957). The OR is defined as

$$OR = ([R_s] / [R_p]) ([A_p] / [A_s]) \quad (19)$$

where

- [R<sub>s</sub>] = radionuclide concentration in sample;
- [R<sub>p</sub>] = radionuclide concentration in precursor;
- [A<sub>s</sub>] = analogue concentration in sample; and
- [A<sub>p</sub>] = analogue concentration in precursor.

This equation may be rearranged to yield an equation for radionuclide concentration in the sample based on the observed ratio and the analogue concentrations.

This approach was developed in the 1950s prior to the concentration ratio approach and was applied successfully to strontium and its analogue calcium, and somewhat less successfully to cesium and its analogue potassium. However, it may be made more successful if, instead of using concentrations of radionuclides in soil, the concentration in soil solutions is used instead (see discussion below on non-linear responses).

### 3.3.5.3 Uncommon Food Products

Farm products commonly included in radiological assessment models include leafy vegetables and produce (NRC Regulatory Guide 1.109 ([NRC 1977]; RESRAD family of codes [Yu et al. 2002]), or perhaps leafy vegetables, other vegetables, cereal grains and fruit (used in the GENII family of codes [Napier et al. 1988; Napier et al. 2003] and its derivatives such as the ERMYN code developed for Yucca Mountain [Wu 2003]). Early computer codes developed from the HERMES model (Fletcher and Dotson 1971) such as FOOD (Baker, Hoenes, and Soldat 1976) used a longer list including

- Leafy above-ground vegetables;
- Other above-ground vegetables;
- Potatoes;
- Other below-ground vegetables;
- Berries;
- Orchard fruit;
- Wheat and wheat products; and
- Other grains.

The longer list was reduced because it was observed that soil-to-plant concentration ratios were not available for most of the crop types, and generic values were being used for all foods (e.g., Napier et al. 1988).

The limited number of food crops is beneficial to the analyst, because it minimizes the number of parameters that must be determined. However, this approach may be seen as ethnocentric and biased by members of some groups in the public. In the Columbia River Comprehensive Impact Assessment (CRCIA) (DOE 1997), which attempted to incorporate public inputs in its development, the lack of specific mention of other food types – and in particular, Native American natural foods - and the lack of a wide range of “other” foods was seen as a severe limitation. It was not considered to be useful to

investigate specific ingestion rates of roots, fruits, etc., unless uptake parameters to specific plant parts (roots versus leaves) or specific plant species were available. Medicinal and other uses of plant material also provided reasons to increase ingestion rates of specific plant types. The generic approaches to radionuclide uptake in plants are useful for uncommon foods, but it must be recognized that plant-specific uptake ratios are probably not going to be available, and uncertainty ranges must be expanded to obtain public acceptance.

Following the accidental release of radionuclides at the Chernobyl Power Plant in 1986, the contamination of natural forest products became an issue. Non-cultivated foods such as berries (noted above) and mushrooms were found to be important. Forests are very efficient at trapping atmospheric aerosols. The aerosol radionuclides fall on the aerial parts of trees (leaves, branches and trunks), understory, and on the soil (litter, mosses and soil horizons). Tree canopies can represent large foliar surfaces, up to ten times greater than in agricultural areas. The long residence time of radionuclides in forests implies an increase in potential internal and external doses for decades. The progressive decomposition of leaves fallen onto the soil constitutes the so-called litter that represents a trap that fixes the radionuclides and slows their migration deep into soil. The large numbers of mycorrhizae and the intense microbial activity in forest soils are the main causes of cycling of radionuclides and stability of transfer factors for several years after deposition. The forest products consumed by humans (mushrooms, berries, so-called “tree juices” such as maple syrup, birch juice) can be up to 250 times more contaminated than the equivalent agricultural products<sup>2</sup>. The Chernobyl experience has resulted in the availability of soil-to-plant aggregated transfer coefficients for a few major forest products such as berries and mushrooms, for a limited number of radionuclides (primarily nuclides of cesium and strontium). The aggregated transfer coefficient relates the activity concentration in the forest product (Bq/kg) to the activity of the surface deposit per unit area (Bq/m<sup>2</sup>). The unit is then m<sup>2</sup> kg<sup>-1</sup>. Values for aggregated transfer factors can be notably higher than the equivalent ones for agricultural products. It has to be stressed that it is difficult to get a precise value, because variability is large in such uncontrolled ecosystems. Uncertainties of one order of magnitude higher or lower on the estimations are normal.

The aggregated transfer coefficient for mushrooms is widely variable (up to 3 orders of magnitude). This variability is due to several reasons:

- Species plays a role of prime importance. The high transfers of cesium in some mushrooms species could be related to the existence in the carpophore of substances with a great affinity for cesium. Transfer factors have been related to the species as shown in Table 3.
- The type of mushroom is an indicator of the cesium transfer. Saprophytic mushrooms develop on decomposing materials on the surface of the soil, which means that these kinds of mushrooms will be the first contaminated following deposition. Transfer factors will diminish as the deposit migrates into the soil. The symbiotic mushrooms associate with trees. Due to their extended mycelium, these

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<sup>2</sup> P. Calmon, personal communication of data collected for the IAEA EMRAS Program.

mushrooms bring minerals to the tree and trees provide the mushrooms with organic substrates from photosynthesis. Most of the edible mushrooms are symbiotic ones. Unfortunately, this characteristic does not give any information about the ability to accumulate cesium by mushrooms. The parasitic mushrooms develop at the expense of the trees and are most of the time characterized by low transfer factors.

- Mycelium depth plays a crucial role in the contamination chronology. Mushrooms with surface mycelia will be contaminated quickly after deposition; deep mycelium mushrooms will be contaminated later.

For radionuclides other than cesium, such high transfer coefficients are not observed, nor is there as large a variability in the transfer factors – it is generally comparable to that for vegetative plants.

For berries, there is not as large a variability in the transfer factor for cesium as in the case for mushrooms. However, transfers to berries are higher than those for agricultural plants.

Soil to plant concentration ratios are being measured for a few radionuclides (primarily strontium and cesium) for common Asian food types such as rice, Chinese cabbage, radish, and soybean (Choi et al. 1991; 1992; 1995; 1998). Cultivating systems are an important factor for plant uptake of radionuclides from soil. One of the critical foods for the intake of radionuclides by humans, especially in the monsoon Asian regions, is rice. Rice is usually planted under flooded conditions because they provide the best environment for the rice plants. No crop rotation is necessary, so that rice has been planted in the same places for hundreds of years. Also, the crop can be grown continuously twice a year in the same paddy fields in tropical or subtropical areas and in some parts of the temperate zones. Because rice is essentially emergent (from the water) vegetation, the main uptake route may not be through the roots. In a tropical area, cropping five times in two years or triple cropping in one year is possible if irrigation water is available.

**Table 3. Aggregate transfer coefficient for cesium for 4 mushroom classes** (Calmon, personal communication of data collected for the IAEA EMRASW Program)

Description	Value ( $m^2 \cdot kg^{-1}$ ) fresh weight	Mushroom species in the class
Mushrooms with low transfer coefficient for cesium	$1 \cdot 10^{-2}$	<i>Agaricus arvensis</i> , <i>Agaricus sylvaticus</i> , <i>Armillaria mellea</i> , <i>Boletus appendiculatus</i> , <i>Boletus elegans</i> , <i>Cantharellus cornucopiensis</i> , <i>Leccinum aurantiacum</i> , (Macro) <i>Lepiota procera</i> , <i>Lepista nuda</i> , <i>Lepista saeva</i> , <i>Lycoperdon perlatum</i> , <i>Psalliota campestris</i> , <i>Sarcodon imbr.</i>
Mushrooms with medium transfer coefficient for cesium	$5 \cdot 10^{-2}$	<i>Boletus aestivalis</i> , <i>Boletus edulis</i> , <i>Cantharellus cibarius</i> , <i>Cantharellus palens</i> , <i>Clitocybe nebularis</i> , <i>Collybia buthyracia</i> , <i>Collybia confluens</i> , <i>Collybia dryophylla</i> , <i>Collybia maculata</i> , <i>Collybia peronata</i> , <i>Hydnum repandum</i> , <i>Kuehneromyces mutabilis</i> , <i>Lactarius deterrimus</i> , <i>Lactarius helvus</i> , <i>Lactarius odoratus</i> , <i>Lactarius picinus</i> , <i>Leccinum sp.</i> , <i>Leccinum versipelle</i> , <i>Lepiota naucina</i> , <i>Oudemansiella sp.</i> , <i>Oudemansiella radicata</i> , <i>Pholiota aegerita</i> , <i>Russula decolorans</i>
Mushrooms with high transfer coefficient for cesium	$1 \cdot 10^{-1}$	<i>Boletus cavipes</i> , <i>Cantharellus lutescens</i> , <i>Cantharellus tubaeformis</i> , <i>Clitocybe infundibuliformis</i> , <i>Lactarius lignyotus</i> , <i>Lactarius quietus</i> , <i>Lactarius torminosus</i> , <i>Lactarius turpis</i> , <i>Leccinum scabrum</i> , <i>Russula nigricans</i> , <i>Suillus grevillei</i> , <i>Tricholoma aurata</i> , <i>Trichomolopsis rutilans</i>
Mushrooms with very high transfer coefficient for cesium	$5 \cdot 10^{-1}$	<i>Clitocybe cavipes</i> , <i>Dermocybe sp.</i> , <i>Hebeloma sp.</i> , <i>Hygrophorus sp.</i> , <i>Hygrophorus olivaceoalbus</i> , <i>Laccaria amethystina</i> , <i>Laccaria laccata</i> , <i>Laccaria proxima</i> , <i>Lactarius sp.</i> , <i>Lactarius camphorates</i> , <i>Lactarius necator</i> , <i>Lactarius porninsis</i> , <i>Lactarius rufus</i> , <i>Lactarius theiogalus</i> , <i>Lactarius trivialis</i> , <i>Rozites caperata</i> , <i>Russula sp.</i> , <i>Russula badia</i> , <i>Russula erythropoda</i> , <i>Russula ochroleuca</i> , <i>Russula turci</i> , <i>Suillus bovinus</i> , <i>Suillus granulatus</i> , <i>Suillus luteus</i> , <i>Suillus variegatus</i> , <i>Xerocomus badius</i> , <i>Xerocomus chrysenteron</i> , <i>Xerocomus subtomentosus</i>

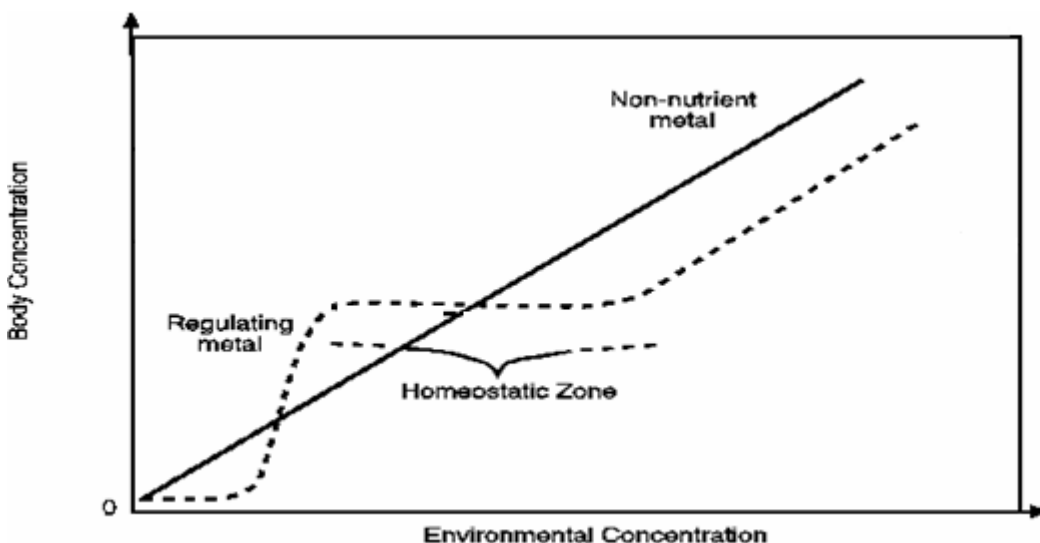
(Data from Amundsen, Gulden, and Strand 1996; Baeza et al. 2000; Bakken and Olsen 1990; Barnett et al. 1999; Battiston et al. 1989. Borio et al. 1991; Byrne 1988; Djingova and Kuleff 2002; Fraiture, Guillitte, and Lambinon 1990; Gaso et al. 1998; Gaso et al. 2000; Giovani, Nimis, and Padovani 1990; Grueter 1971; Guillitte, Fraiture, and Lambinon 1990; Heinrich 1992; Heinrich 1993; Horyna and Randa 1988; Kammerer, Hiersche, and Wirth 1994; Kirchner and Daillant 1998; Lambinon et al. 1988; Mascanzoni 1990; Mietelski et al. 2002; Pietrzak-Flis et al. 1996; Randa et al. 1990; Rantavaara 1990; Roemmelt, Hiersche, and Schaller 1988; Rømmelt et al. 1990; Rühm et al. 1998; Seeger and Schweinshaut 1981; Steiner, Linkov, and Yoshida 2002; Svadlenkova, Konecny, and Smutny 1996; Tsukada, Shibata, and Sugiyama 1998; Vinichuk, Johanson, and Taylor 2003)



### 3.3.5.4 Potential Non-Linear Uptakes

A key assumption of the concentration-ratio approach is that the CR is a constant as a function of concentration; that is, the uptake is linear. This may not be true for contaminants that are nutrients or are chemical analogues for them. Non-linear responses may be possible if plants scavenge essential elements at low concentrations but maintain a homeostatic balance at higher soil concentrations (Sheppard and Sheppard 1985). The assumption of linearity may be appropriate for elements that are not essential to biological function, are not analogues of such elements, or are not absorbed by organisms via nutrient pathways. These latter elements seldom exhibit linearity at very low environmental concentrations (Figure 3). For such elements (Table 4), organisms often are able to homeostatically regulate their tissue concentrations over a range of environmental concentrations. In some cases, these concentrations may be 40 to 200 times greater than the amount needed to sustain life (Förstner and Wittmann 1981). There is generally little information to evaluate this concept except for a few radionuclides such as cesium, which mimics potassium, or strontium, which mimics calcium.

The differences in predicted effects between these two models can be substantial. Experience in assessing risks of metals in sediments and groundwater indicates that, for certain metals and species, the linear model can overestimate exposure by up to six orders of magnitude (DOE 1997).



**Figure 3.** Relationships are shown between tissue concentrations and environmental concentrations for non-nutrient and nutrient compounds (adapted from personal communication with C. A. Brandt).

**Table 4.** In a biological classification of metals (modified from Beeby 1991) shaded cells are non-nutrient analogs; bold cells are nonessential metals.

Period	Macronutrient		Micronutrient					Non-essential	
3	Na	Mg							
4	K	Ca	Cr	Mn	Fe, Co, Ni	Cu	Zn		
5		<b>Sr</b>					<b>Cd</b>		
6	<b>Cs</b>							<b>Hg</b>	<b>Pb</b>
7								<b>Eu, U</b>	<b>Np</b>

(a) Period is from the Periodic Table of the Elements.

To date, few models have been developed to deal with this problem. A possible alternative for radionuclides with this behavior would be to treat them with a specific-activity type model, wherein the atom ratio of the radioactive to stable atoms of an element are assumed to be the same in the plant as in the soil (see below). The model proposed by Norden et al. (2005) is essentially a specific-activity type model; they suggest that transfer rates of contaminants would mimic analogue nutrient transfer rates. Their suggestion is that for the case of radionuclides that are analogues of macronutrients, it can be assumed that the plant uptake of the nutrient modulates the uptake of the radionuclide. This means, that the radionuclide and the corresponding analogue nutrient are taken up by plants in an identical manner. Furthermore, assuming that only ions in the soil solution near the roots are available for transition into the roots, the transition of the radionuclide from soil to plant roots can be represented as an independent Poisson process with the following rate:

$$TR(RN) = \frac{[RN]_{ss}}{[RN]_{ss} + [A]_{ss}} TR(A) \quad (20)$$

where,

$TR(RN)$  is the transition rate of the radionuclide from the soil solution into the roots [mol/yr],

$TR(A)$  is the overall transition rate of the analogue from the soil solution into the roots [mol/yr]

$[RN]_{ss}$  is the radionuclide concentration in the soil solution near the roots [mol/m<sup>3</sup>]

$[A]_{ss}$  is the analogue nutrient concentration in the soil solution near the roots [mol/m<sup>3</sup>].

Because  $[RN]_{ss}$  is generally very small with respect to  $[A]_{ss}$ , the overall transition rate from the soil solution into the roots equals the analogue uptake rate. These authors also suggest that a selectivity coefficient may be added to this equation if the radionuclide has a slightly lower uptake than the analogue. This model is very similar in concept and application to the Observed Ratio, except that it is based on soil solutions rather than soil concentrations.

### 3.3.5.5 Specific-Activity Models

Some key biologically-active elements are modeled in a separate way through the use of specific activity models. The behavior of the radionuclides tritium and  $^{14}\text{C}$  in exposure pathways must be handled in a special manner, because the behavior of these environmentally mobile radionuclides mimics that of stable hydrogen and carbon in biological processes, respectively. The concentrations of tritium or  $^{14}\text{C}$  in environmental media (soil, plants, and animal products) are assumed to have the same specific activity (curies of radionuclide per kilogram of soluble element) as the contaminating medium (air or water). The fractional content of hydrogen or carbon in a plant is then used to compute the concentration of tritium or  $^{14}\text{C}$  in the food product under consideration. The hydrogen content in both the water and the nonwater (dry) portion of the food product is used when calculating the tritium concentration. For airborne releases, it is assumed that plants obtain all their carbon from airborne carbon dioxide.

In the simplest implementation, tritium in the environment is assumed to be in the oxide (HTO) form, and to behave in a manner identical with water. For HTO releases, concentrations in air moisture ( $\text{Bq L}^{-1}$ ) are found by dividing the concentration in air ( $\text{Bq m}^{-3}$ ) by the absolute humidity ( $\text{kg or L m}^{-3}$ ). The concentration of tritium in vegetation is calculated (Baker, Hoenes, and Soldat 1976) as

$$C_{Tp} = 9 C_{wT} F_{Hp} \quad (21)$$

where  $C_{Tp}$  = the concentration of  $^3\text{H}$  in plant type  $p$  ( $\text{Bq/kg}$ );  
 $C_{wT}$  = the concentration of tritium in the environmental water ( $\text{Bq/L}$ ); and  
 $F_{Hp}$  = the fraction of hydrogen in total vegetation of plant type  $p$ .

The coefficient, 9, converts tritium concentration in environmental water to concentration in hydrogen (this is approximate, but applicable to water with dilute solutions of HTO). Note that 1 kg of water equals 1 L in the above equation. The fractions of hydrogen in various food types are given in Table 5. (Some additional parameters are included in Table 4 for the discussion of animal products given below).

Murphy (1986) and Raskob (1994) have developed models designed specifically to calculate doses from chronic releases of elemental tritium (HT) and HTO, taking into account the formation of organically-bound tritium (OBT). Both models are similar in that they model dispersion of HT and HTO, deposition of HT and HTO through dry deposition, and deposition of HTO through wet deposition. Treatment of the soil compartment includes such processes as diffusion and resuspension. In NORMTRI (Raskob 1994), air concentrations due to resuspension are calculated by treating the area of deposition as an array of ground-level area sources. For an HT release, 20% of the deposited tritium is assumed to remain in the soil; the rest is re-emitted. In the Murphy model, HT is oxidized in the leaf as well as in the soil. Both models account explicitly for the transfer of OBT in plants to OBT in animal products. However, both of these models require extensive information and operate on a limited time scale. For assessments of a more general nature, the model of Peterson and Davis (2002) that includes both HTO and HT is useful.

**Table 5.** Weight Fractions of Hydrogen and Carbon in Environmental Media, Vegetation, and Animal Products (Napier et al. 2003)

Food or Fodder	Water $f_w$	Carbon dry $f_c$	Hydrogen dry $f_h$	Carbon <sup>a</sup> wet $F_{cv}, F_{ca}$	Hydrogen <sup>b</sup> wet $F_{hv}, F_{ha}$
Fresh fruits, vegetables, and grass	0.80	0.45	0.062	0.090	0.10
Grain and stored animal feed	0.12	0.45	0.062	0.40	0.068
Eggs	0.75	0.60	0.092	0.15	0.11
Milk	0.88	0.58	0.083	0.070	0.11
Beef	0.60	0.60	0.094	0.24	0.10
Poultry	0.70	0.67	0.087	0.20	0.10

<sup>a</sup>  $F_{cv}$  or  $F_{ca} = f_c (1 - f_w)$ .  $F_{cv}$  applies to plants and  $F_{ca}$  applies to animal products.

<sup>b</sup>  $F_{hv}$  or  $F_{ha} = (f_w / 9) + f_h (1 - f_w)$ .  $F_{hv}$  applies to plants and  $F_{ha}$  applies to animal products.

To estimate HTO concentrations in plant products, Peterson and Davis (2002) assumed conservatively that the concentration of HTO in plant water equals 0.9 times the HTO concentration in air moisture ( $C_{am}$ ) for leafy vegetables and pasture and 0.8  $C_{am}$  for fruit, root crops and other vegetables, and grain. These reduction factors account for dilution from soil water, which has a lower tritium concentration than does air moisture and which affects fruit and root crops more than leaves. OBT concentrations are estimated from a plant water concentration of 0.9  $C_{am}$  for all plant types, since OBT is assumed to form exclusively in the leaves and to be translocated to other plant parts. Then, isotopic discrimination occurring in the formation of OBT (Table 6) is assumed conservatively to result in an OBT concentration 0.9 times the concentration of HTO in plant water.

The Peterson and Davis (2002) equation for calculating concentrations of HTO in plant products ( $Bq\ kg^{-1}\ fw$ ) is:

$$C_{pp\_HTO} = RF_{pp} C_{am} F_{fw\_pp} \quad (22)$$

where

- $C_{pp\_HTO}$  = concentration of HTO in plant products ( $Bq\ kg^{-1}$  fresh weight)
- $F_{fw\_pp}$  = average fresh weight (fw) fraction of plant products ( $kg\ kg^{-1}$ ) (Note, this is essentially the same as  $f_w$  in the preceding equation)
- $RF_{pp}$  = reduction factor that accounts for low soil water concentrations relative to concentrations in air moisture and varies by vegetable type
- $C_{am}$  = HTO concentration in air moisture ( $Bq\ L^{-1}$ )

**Table 6.** Parameter values for calculating HTO and OBT concentrations in plant products (adapted from Peterson and Davis 2002).

	Leafy vegetables	Fruit	Root and other vegetables	Grain	Pasture
Reduction factor (RF <sub>pp</sub> )	0.9	0.8	0.8	0.8	0.9
Reduction factor for leaves (RF <sub>l</sub> )	0.9	0.9	0.9	0.9	0.9
Isotopic discrimination factor (ID <sub>pp</sub> )	0.9	0.9	0.9	0.9	0.9
Fresh matter fraction (F <sub>fw_pp</sub> )	0.906	0.853	0.824	0.117	0.8
Dry matter fraction (F <sub>dm_pp</sub> )	0.094	0.147	0.176	0.883	0.2
Water equivalent factor (W <sub>eq_pp</sub> )	0.6	0.59	0.58	0.577	0.616

The Peterson and Davis (2002) equation for calculating concentrations of OBT in plant products (Bq kg<sup>-1</sup> fw) is:

$$C_{pp\_OBT} = RF_l ID_{pp} C_{am} F_{dm\_pp} W_{eq\_pp} \quad (23)$$

where

RF<sub>l</sub> = reduction factor for plant leaves

ID<sub>pp</sub> = isotopic discrimination factor for plant products

C<sub>pp\_OBT</sub> = concentration of OBT in plant products (Bq kg<sup>-1</sup> fresh weight)

F<sub>dm\_pp</sub> = average dry matter fraction of the plant products (kg dry mass kg<sup>-1</sup> fresh mass)

W<sub>eq\_pp</sub> = water equivalent factor of plant products (L kg<sup>-1</sup>)

HT is a biologically inert gas and imparts an extremely low dose relative to tritiated water. However, HT is converted in the environment to HTO, in which form it moves through the food chain to dose. Therefore, a model of dose from HT releases is necessary. Ratios of the HTO concentrations in soil, air and plants to the HT concentration in air recommended for screening level assessments by Peterson and Davis (2002) are 8 Bq L<sup>-1</sup> / (Bq m<sup>-3</sup>) for the ratio of HTO in air moisture to HT in air, and 12 Bq L<sup>-1</sup> / (Bq m<sup>-3</sup>) for the ratio of HTO in plant water to HT in air. Doses from HT are then modeled using the recommended HTO / HT ratios for air moisture and plant water. Concentrations in air moisture are calculated by multiplying the HT concentration in air (C<sub>aHT</sub>; Bq m<sup>-3</sup>) times eight. Concentrations of HTO and OBT in plant and animal products are calculated for releases of HT by replacing RF<sub>pp</sub> in the above equations with a factor of 1.5. Thus the plant water concentration in these equations becomes 1.5 C<sub>am</sub> = 12 C<sub>aHT</sub>.

Using the specific activity model for <sup>14</sup>C in air, the concentration of <sup>14</sup>C in crops from atmospheric contamination C<sub>Cp</sub> is calculated (Baker, Hoenes, and Soldat 1976) as

$$C_{Cp} = C_{a,C} F_{Cp}/P_C \quad (24)$$

where  $C_{a,C}$  = the concentration of  $^{14}\text{C}$  in air ( $\text{Bq}/\text{m}^3$ );  
 $F_{Cp}$  = the fraction of carbon in the plant (dimensionless); and  
 $P_C$  = the concentration of carbon in air ( $\text{kg}/\text{m}^3$ ).

Plants obtain most of their carbon via photosynthesis of  $\text{CO}_2$  from the air. Therefore, it is difficult to estimate the transfer of radioactive  $^{14}\text{C}$  from irrigation water or soil to plants. The approach adopted by Napier et al. (1988) is to assume that plants obtain a small fraction of carbon from soil, and to use that activity ratio. The approach used by Yu et al. (2001) is to assume that carbon in the soil slowly converts to  $\text{CO}_2$  and to then estimate a  $^{14}\text{CO}_2$  concentration in the air near crops. Due to the volatility of  $^{14}\text{C}$  in soil, it is quickly released via gaseous emission as  $^{14}\text{CO}_2$ . Sheppard et al. (1991) measured the rate of  $^{14}\text{C}$  loss from soil in outdoor lysimeter experiments. Carbon loss from the soil, measured by the emission rate (the “evasion rate” in RESRAD), is 12/yr for clay and loamy soils, and 22/yr for sandy and organic soils (Yu et al. 2001). Thus,  $^{14}\text{C}$  concentrations in surface soil reach equilibrium within 1 to 2 months. Emission is the dominant mechanism for removing  $^{14}\text{C}$  from the soil. In comparison, losses due to leaching, radioactive decay, and soil erosion are slight.

Specific activity models have been recommended for other radionuclides. Attributes that lead to this type of recommendation are long half-life, high mobility, biologically essential element, and massive isotopic dilution in the geosphere and biosphere.  $^{129}\text{I}$  and  $^{36}\text{Cl}$  are some of the most critical radionuclides to consider in intermediate- and high-level nuclear-waste management, and both have these attributes. Uptake in plants can be simulated using equations similar to those above for tritium or  $^{14}\text{C}$ , using the measured concentrations of the stable elements in local soil and plants. The specific-activity approach has also been suggested for  $^{35}\text{S}$ ; however, its half-life is only about 3 months, and so is unlikely to be present in radioactive wastes.

### 3.3.5.6 Uptake in Fruit Trees

The standard uptake model described above using concentration ratios was originally developed for annual crops (leafy vegetables, root vegetables, forages, and grains). The vast majority of available observations, either laboratory or field studies, are for a limited number of crop types. In addition, observations are not available for all crop types for all chemical elements, and most compendiums of transfer factors use surrogates, relying on similar plant types, soil types, or chemical behavior to fill gaps in the knowledge (e.g., Staven et al. 2003; Yu et al. 2001).

The standard model is also used for perennial plants, including fruit trees. The model may not be appropriate for trees, considering their longer life and potential for accumulating contaminants in roots, trunks, and leaves, with transport to fruits possibly delayed for periods of over 1 year. The British Ministry of Agriculture, Fisheries, and Food (MAFF, since 2001 called the Department for Environment, Food, and Rural Affairs) has noted that such “models for fruit are extremely conservative... Extensive research is underway to produce more appropriate data. However, a review for MAFF concludes that no better fruit models currently exist” (MAFF 1999).

The International Union of Radioecologists and the International Atomic Energy Agency jointly prepared a major review of soil-to-plant transfer factors (IAEA 1994), which is used as a basic reference world-wide. In this review, soil-to-plant transfer factors are presented for many radionuclides and crop types. *None* of them refer to fruit or nut trees. A more recent compilation for tropical ecosystems does include some fruit trees (primarily apples), but the only nut is the coconut (IAEA-IUR 1997, quoted in Carini 2001). This state of affairs has been discussed in the international arena for several years; the IAEA's Validation of Model Predictions (VAMP) Program Multiple Pathways Assessment Working Group noted in the early 1990s that many participants overestimated concentrations of  $^{137}\text{Cs}$  in fruit trees, and that models for predicting the contamination of fruit were in need of further improvement (IAEA 1995). The sequel to VAMP, the IAEA's Biosphere Modeling and Assessment (BIOMASS) Program, included a Fruits Working Group (IAEA 1996). As part of the work of the Fruits Working Group, a review was undertaken of the experimental, field and modeling information on the transfer of radionuclides to fruit. The results of this work were published as a special issue of the *Journal of Environmental Radiation* (Ventner 2001).

In this special issue, Mitchell (2001) reports that there are three generic types of models that are applicable to fruit trees:

- Simple mathematical functions describing declining concentration in fruit based on observations following deposition (e.g., Antonopoulos-Domis et al. 1990);
- Models that attempt to predict temporal distribution in soil-plant systems through descriptions of the processes involved, e.g. a model postulated by Frissell at the 1994 VAMP meeting in Vienna;
- Radiological dose assessment models that use a mixture of equilibrium and/or dynamic modeling approaches to predict concentrations in edible products, e.g. SPADE (Thorne and Coughtrey 1983).

Antonopoulos-Domis et al. (1990) developed a model structure for perennial fruit trees describing distribution, retention, transfer and rejection of radionuclides, based on experimental determinations of  $^{137}\text{Cs}$  in apricot fruit trees. The original concept for the model was based on the fact that the leaves and fruits developing each year are only contaminated by a portion of the  $^{137}\text{Cs}$  in the body of the tree. A fraction of this available reservoir is removed each year, part is lost from the tree through leaves and fruit and part becomes irretrievably associated with the body of the tree. This model requires knowledge of the contaminant inventory in the soil and the tree, as well as the deposition. The model is not immediately transferable to other types of trees, radionuclides, and locations, but it does indicate that at least two compartments are probably necessary to adequately describe the long-term accumulation and transfer of contaminants from trees into fruits.

A model for radiocesium transfer to tree fruit described by Frissel (1994) considers the homeostatic control of potassium within fruit trees. The model structure has four compartments and was designed to consider the long-term fate of cesium in soil as affected by changes in the supply of potassium to soil. The four compartments are soil, the easily accessible part of a tree, the poorly accessible woody part, and the fruit or leaf.

The model is homeostatic, i.e. all cesium concentrations and fluxes are controlled by potassium concentrations and fluxes, respectively. In determining the various transfer parameters, it is assumed that there is no difference in the behavior of potassium and cesium, but that discrimination occurs between the compartments. The loss of plant material, termed debris by Frissel, via branches, leaf fall and fruit loss is included and returned to the soil, but because uptake is homeostatically controlled, this has minimal influence on the tree contents. Frissel (1994) concluded that the model was probably not sufficient to describe cesium transfer to fruit. In particular, the use of three compartments was not sufficient to model availability within the plant. The model results do indicate that important processes are likely to be the biological half time of cesium in wood, the discrimination between cesium and potassium, cycling of potassium (through falling leaves, etc.), and uptake of potassium.

The fruit plant model in the SPADE computer code (Thorne & Coughtrey 1983) has six compartments, representing internal leaf, external leaf, stem, fruit, storage organs and root. Movement of radionuclides within the plant model is controlled by empirically derived rate constants and parameters are derived for three broad categories of fruit plant: herbaceous, shrub and tree. Foliar absorption is represented by transfers between the external leaf and internal leaf compartments. Interception by plants takes account of changes in plant biomass with season. The original default parameters were based largely on data for cereals but were modified in the case of tree and shrub fruits to allow for more rapid transfer from stem to root so that the root store could serve as a reservoir through subsequent seasons. Loss of radionuclides from external plant surfaces to the soil is modeled as transfer to the surface layer of the soil model. The process of root uptake is modeled as the transfer of radionuclides from soil solution to the plant root compartment. The transfer rate is also assumed to vary with soil layer depth, both as a function of the root distribution throughout the soil profile and as a function of the deposit distribution in soil. Consequently, the transfer of radionuclides from the soil solution to root is represented by a discrete transfer from each of the 10 layers in the soil model. The soil solution to root rate constant in each soil layer is a function of the root uptake rate constant and the assumed distribution of root activity in each layer. Three plant absorption mechanisms are responsible for the transfer of radionuclides at the soil-root interface: plant-base absorption, main root system absorption and tap root absorption. The actual value of the soil solution to root transfer coefficient for each root layer corresponding to the soil layer of the soil model is calculated as the product of the specified rate coefficient and the normalized root shape modifier.

None of these models appears to be suitable for generic use in long-term radiological assessments without substantial modification and simplification, and all require additional development of parameters before general use. Again, it was noted that measurements of radionuclide uptake in trees were lacking (Carini 2001), and recommended that “There is a need for research on the behavior of radionuclides in fruit crops to drive model development, not simply to parameterize existing models. Research should focus on understanding the key processes” (Coughtrey et al. 2001). As a result of these recommendations, the IAEA initiated the Environmental Modeling for Radiation Safety (EMRAS) program in 2003. This program has a Working Group on Revision of



IAEA Technical Report Series No. 364 “Handbook of parameter values for the prediction of radionuclide transfer in temperate environments” and attempted to establish another Fruits Working Group. However, the participation in the Fruits group was so low that these participants joined with the Handbook group.

### 3.4 Modeling the Uptake of Radionuclides in Animal Products

In general, animals and animal products may become contaminated through a wide variety of pathways, in ways similar to humans. Pathways by which animal products may become contaminated include animal ingestion of plants, water, and soil (the animals’ inhalation is usually neglected as contributing little to the total as discussed below). However, as with people, the source of the foods and water may be quite varied, and include contributions from contaminated soil as well as, for predators, other potentially-contaminated animals.

Absorption of radionuclides by animals via inhalation is of limited importance. Hvinden et al. (1964) showed clearly that the intake of radionuclides through inhalation was at least three orders of magnitude less than that from pasture. Since the respiratory tract is not usually an edible product for man, the significance of the inhalation route for food chain contamination is further reduced.

#### 3.4.1 Feed to Animal Product Transfer Factors

The various routes of ingestion by animals (plants, water, prey) can be combined through the use of a transfer coefficient. The transfer coefficient is widely used in the literature to predict the transfer of radionuclides to animal food products. Ward and colleagues first used the transfer coefficient to describe the relationship between  $^{137}\text{Cs}$  in animal feed and its concentration in milk of dairy cows (Ward and Johnson 1965; Stewart et al. 1965; Johnson et al. 1968). They subsequently described the validation of this term as conceived (Ward and Johnson 1986). Ward and colleagues expressed their results as a ratio of the daily  $^{137}\text{Cs}$  intake (Bq/day) to the concentration of  $^{137}\text{Cs}$  per liter of milk (Bq/L). This term was used to successfully describe the relationship of  $^{137}\text{Cs}$  levels in feed and milk under varying environmental management conditions. Subsequently, this approach was found to provide a convenient method to describe the relationship between  $^{137}\text{Cs}$  in feed and meat products (Johnson et al. 1969).

A detailed mathematical derivation of the animal product transfer function is provided in Peterson (1983). Much of the early development of this function was done by Ng and associates (Ng and Thompson 1968; Ng et al. 1977; Ng, Colsher, and Thompson 1979; Ng 1982; Ng, Colsher, and Thompson 1982a; Ng, Colsher, and Thompson 1982b). This relationship ( $F_m$ ) has subsequently been expressed in the literature by the following equation:

$$F_m \text{ (day / L)} = \frac{\text{Radionuclide Milk Concentration (Bq / L)}}{\text{Feed Concentration (Bq / kg)} \times \text{Feed Intake (kg / day)}} \quad (25)$$

For appropriate estimations, the feed concentration and the feed intake should be long-term averages of what is eaten rather than point measurements. Since the inception of this concept, transfer coefficients have been applied to many of the radionuclides; nevertheless, Ward and Johnston (1986) cautioned regarding the general applicability of the use of transfer coefficients for a wide number of radioelements and animal species. Others argue about the use and abuse of transfer coefficients, but according to Shaw (2001), “no one has yet come up with a better idea” for expressing this relationship. The same relationship is used for other animal products such as meat and eggs.

Transfer to animal products has been expressed using various quantitative terms, sometimes with different dimensional units. These have included terms such as bioconcentration factor, transfer coefficients, transfer factor, concentration factor, concentration ratio, and aggregated transfer coefficient (ICRU 2001). In an effort to simplify and standardize this widely differing terminology, the International Commission on Radiation Units and Measurements has adopted a broad term called the concentration ratio, or  $C_r$ , which is defined as the ratio of the activity density of a radionuclide in the receptor compartment to that in the donor compartment (ICRU 2001). In the case of plant to animal transfers, the concentration ratio in this case is defined as the steady-state concentration in an animal tissue divided by the average concentration in feed.

The concept of the transfer factor may be applied to essentially all routes of ingestion by animals: soil-to-animal, plant-to-animal, water-to-animal, and prey animal-to-predator animal.

Some dynamic models such as PATHWAY( Whicker and Kirchner 1987) use assimilation fractions for ingested materials and biological elimination rates; however, these are also directly derived from the steady state or equilibrium transfer factors.

As with radionuclide uptake into plants, some elements in animals may be homeostatically controlled. Very little work has been done to quantify the effect that this might have on animal product transfer factors.

### **3.4.2 The Observed Ratio in Animal Products**

A different approach to estimating uptake of radionuclides into animals is based on the observation that the presence of chemically similar stable elements in feed can have a significant impact on radionuclide uptake in animals. The ratio of the radionuclide-to-stable-element concentration in one medium to the radionuclide-to-stable-element in another medium (for instance, feed and animals) has been named the observed ratio (OR). Early work developed the OR to predict the transfer of  $^{90}\text{Sr}$  uptake by milk (Comar et al. 1961; Comar et al. 1966). In this sense, the OR is described as the ratio of strontium to calcium in the milk following consumption of  $^{90}\text{Sr}$ -contaminated feed. Likewise, the cesium/potassium ratio was used to describe the uptake of  $^{137}\text{Cs}$  in milk, assuming that the relationship would be similar for cesium and potassium as for strontium and calcium.

Although this approach has not been widely used for many years, it could potentially be used to simulate the uptake of radionuclides that are analogues of stable elements that are homeostatically regulated in animals. Homeostatic control via major routes allows the animal to regulate the tissue content of the various elements taken into the body, mostly via ingestion. These control systems include gastrointestinal absorption, urinary excretion, tissue deposition (storage pools or organ uptake such as the thyroid), secretion into milk, and endogenous excretion via feces. Some radionuclides have dietary essential stable isotopes (such as iodine) or analogues (such as calcium in the case of strontium), which are under homeostatic control by the animal. For other nuclides, control may be by the mechanisms controlling other essential elements. The intestine, thyroid gland, and mammary gland all have unique mechanisms for controlling many elements. For elements that are homeostatically controlled, the uptake and excretion is set to maintain the plasma or milk concentration. In this case, the transfer coefficient will not be constant over a wide range of conditions, and milk or tissue concentration may not vary as the intake changes. An increase in feed concentration of a radionuclide may not equate to an increase in tissue concentration for these contaminants.

### **3.4.3 Unusual Animal Products**

The animal products commonly modeled include meat, milk, poultry, and eggs (e.g., GENII [Napier et al 2003], ERMYN [Wu 2003]). Beef is frequently the representative animal product for all meat (including beef, pork, wild game, and other meat), cows are the representative milk-producing animal, and chicken is the representative poultry animal and egg producer. Some compilations of transfer factor values are available for other animals, such as pork and lamb (e.g., Ng, Colsher, and Thompson 1982). As a general rule, transfer factors are not explicitly available for other meat animals such as deer, goats, rabbits, wild game birds, or other animals occasionally consumed by humans. A limited amount of data are available for transfer of contaminants to goat's milk (primarily for iodine). In an appendix to their report, Ng, Colsher, and Thompson (1982) provide information on scaling reported values of the transfer factor for animals with characteristics different from those of the reference livestock. Such information could be used to scale estimates of transfer factors for other types of animal product, such as eggs from birds like ostriches or emus (see discussion below on allometric approaches).

Arctic mammals such as seals, walrus, and whales are a unique subset of animals consumed by humans. A review by Layton et al. (1997) provides transfer factors for a few natural and artificial radionuclides for these types of animals.

Some nomadic societies consume products from herded sheep and horses. Some limited data are available for actinides and long-lived fission products in these animals from measurements made at the Semipalatinsk Test Site in Kazakhstan. The Semipalatinsk Test Site, known as the "polygon", is a 19,000 km<sup>2</sup> zone located in the northeast of Kazakhstan, 800 km north of the capital Almaty. Between 1949 and 1989, the former USSR conducted about 456 nuclear explosions at STS. Until 1963, most of the explosions were carried out on the surface and in the atmosphere (including 126 atmospheric tests with 30 surface bursts). During the 40 years of testing, the total energy released in the testing was equivalent to 17.4 megatons of TNT. After the breakup of the

Soviet Union, effective control over the area by local governments was lost. The region is a combination of arid plains and lightly forested mountains. Between 30,000 and 40,000 people live near the site, and a small number of people actually live on the site, primarily semi-nomadic herders and farmers. Following the breakup of the Soviet Union, the Russian military moved out of the STS, and took nearly all records of activities and resulting contamination with them. The National Nuclear Center, Institute of Radiation Safety and Ecology (IRSE), in the city of Kurchatov, Kazakhstan, carries out research on radiation safety at the STS and radioecology of plants and animals. Preliminary contacts (personal communication, Dr. Larisa Ptitskaya to Bruce Napier) indicate that information may be available for the radionuclides of tritium, cobalt, strontium, cesium, europium, plutonium, americium, and radon.

A few other animal products consumed by humans are not amenable to estimation using transfer factors. An example is honey from honey bees (e.g., Bonazzola et al. 1991; Fulkery et al. 1998). Accumulation of radionuclides in honey has been reported; however, no models for estimating concentration from environmental contamination levels are known. Bonazzola et al. (1991) reported that honey can be contaminated by radionuclides accumulating in soils or plants. The primary pathway of contamination is plant nectar; pollen did not appear to be a dominant contributor. However, the radionuclide content did not appear to be correlated between radionuclides. In addition, the plant concentration varied sufficiently that multifloral honeys could not be traced. These researches concluded that consideration of the botanical sources of the honey was also insufficient to allow use of honey as a bioindicator.

### 3.4.4 Specific Activity Models for Animal Products

Some key biologically-active elements are modeled in a separate way through the use of specific activity models. The behavior of the radionuclides tritium and  $^{14}\text{C}$  in exposure pathways must be handled in a special manner, because the behavior of these environmentally mobile radionuclides mimics that of stable hydrogen and carbon in biological processes. The concentrations of tritium or  $^{14}\text{C}$  in environmental media (soil, plants, and animal products) are assumed to have the same specific activity (curies of radionuclide per kilogram of soluble element) as the contaminating medium (air or water). The fractional content of hydrogen or carbon in an animal product is then used to compute the concentration of tritium or  $^{14}\text{C}$  in the food product under consideration. The hydrogen content in both the water and the nonwater (dry) portion of the food product is used when calculating the tritium concentration. For airborne releases, it is assumed animals obtain all their carbon through ingestion of plants.

The simplest model for tritium in animal products (Baker, Hoenes, and Soldat 1976) defines the concentration of tritium in the animal product as

$$C_{Ta} = \frac{C_{Tp} Q_f + C_{Tw} Q_w}{F_{hf} Q_f + Q_w / 9} F_{ha} \quad (26)$$

where  $C_{Ta}$  = the concentration of  $^3\text{H}$  in animal product (Bq/kg or Bq/L);  
 $C_{Tp}$  = the concentration of  $^3\text{H}$  in animal product feed crop p (Bq/kg);

- $C_{Tw}$  = the concentration of  $^3\text{H}$  in animal drinking water (Bq/L);  
 $F_{hf}$  = the fraction of hydrogen in animal feed (dimensionless);  
 $F_{ha}$  = the fraction of hydrogen in animal product m (dimensionless);  
 $Q_f$  = the quantity of feed consumed per day (kg/d); and  
 $Q_w$  = the quantity of water consumed by the animal per day (L/d)  
 9 = coefficient converting concentration of tritium in water to concentration of tritium in hydrogen, valid for dilute solutions.

For the variant of this model proposed by Peterson and Davis (2002), the generalized equations for calculating HTO and OBT concentrations in animal products ( $\text{Bq kg}^{-1}$  fw) are:

$$C_{ap\_HTO} = [RF_{pp} F_{fr} + RF_l IDpp Dfr + W_{fr} RF_{dw} + ISA_{fr}] C_{am} F_{fw\_ap} \quad (27)$$

where

- $F_{fr}$  = water fraction contributed to the diet by the water fraction of food;  
 $D_{fr}$  = water fraction contributed to the diet by the dry matter fraction of food;  
 $W_{fr}$  = water fraction contributed to the diet by drinking water;  
 $RF_{dw}$  = fraction of HTO concentration in drinking water relative to that in air moisture;  
 $ISA_{fr}$  = water fraction contributed to the diet by inhalation and skin absorption, and the other parameters are as defined for plants.

The equation to calculate OBT in animal products assumes the specific activity in organic material equals the specific activity in the aqueous phase apart from a discrimination factor ( $W_{eq\_ap}$ ).

$$C_{ap\_OBT} = [RF_{pp} F_{fr} + RF_l IDpp Dfr + W_{fr} RF_{dw} + ISA_{fr}] C_{am} F_{dm\_ap} W_{eq\_ap} \quad (28)$$

Recommended default values for parameters in these equations are presented in Table 7. Values for fresh and dry matter fractions and for water equivalent factors are from Geigy (1981).

As with Peterson and Davis' (2002) model for HT in plants, the ratios of the HTO concentrations in soil, air and plants to the HT concentration in air recommended for screening level assessments by Peterson and Davis (2002) are  $8 \text{ Bq L}^{-1} / (\text{Bq m}^{-3})$  for the ratio of HTO in air moisture to HT in air, and  $12 \text{ Bq L}^{-1} / (\text{Bq m}^{-3})$  for the ratio of HTO in plant water to HT in air. Doses from HT are then modeled using the recommended HTO / HT ratios for air moisture and plant water. Concentrations in air moisture are calculated by multiplying the HT concentration in air ( $C_{aH}$ ;  $\text{Bq m}^{-3}$ ) times eight. Concentrations of HTO and OBT in animal products are calculated for releases of HT by replacing  $RF_{pp}$  in the above equations with a factor of 1.5.

**Table 7.** Suggested parameter values for calculating HTO and OBT concentrations in animal products (adapted from Peterson and Davis 2002).

	Eggs	Milk	Beef	Pork	Poultry
Fraction water ingested from fresh weight food ( $F_{fr}$ )	0.034	0.371	0.409	0.031	0.034
Fraction water ingested from dry weight food ( $D_{fr}$ )	0.149	0.065	0.074	0.135	0.149
Fraction water ingested from drinking water ( $W_{fr}$ )	0.781	0.544	0.490	0.782	0.781
Fraction water from inhalation and skin absorption ( $ISA_{fr}$ )	0.036	0.021	0.028	0.052	0.036
Drinking water HTO / air moisture HTO ( $RF_{dw}$ )	0.5	0.5	0.5	0.5	0.5
Fresh weight fraction ( $F_{fw\_ap}$ )	0.74	0.897	0.668	0.50	0.67
Dry matter fraction ( $F_{dm\_ap}$ )	0.26	0.103	0.332	0.50	0.33
Water equivalent factor ( $W_{eq\_ap}$ )	0.835	0.669	0.795	0.904	0.796

The concentration of  $^{14}\text{C}$  in animal products is calculated as

$$C_{ca} = \frac{C_{cp} Q_f + C_{cw} Q_w}{F_{cf} Q_f + F_{cw} Q_w} F_{ca} \quad (29)$$

where  $C_{ca}$  = the concentration of  $^{14}\text{C}$  in animal product (Bq/kg or Bq/L);  
 $C_{cp}$  = the concentration of  $^{14}\text{C}$  in crop used for animal feed (Bq/kg);  
 $C_{cw}$  = the concentration of  $^{14}\text{C}$  in animal drinking water (Bq/L);  
 $F_{cf}$  = the fraction of carbon in animal feed (dimensionless) ( $F_{cv}$  in Table 5);  
 $F_{cw}$  = the fraction of carbon in animal drinking water (dimensionless);  
 $F_{ca}$  = the fraction of carbon in animal product m (dimensionless) (Table 5);  
 $Q_f$  = the quantity of feed consumed per day (kg/d); and  
 $Q_w$  = the quantity of water consumed by the animal per day (L/d).

### 3.4.5 Kinetic Modeling for Animals

Animals may be exposed to a contaminant via ingestion of contaminated water, soil, and food. The ECEM (Ecological Contaminant Exposure Model) is a food-web based ecological exposure assessment tool (Eslinger et al. 2004). ECEM is a detailed, food-web based, chronic exposure model intended for use in situations where an organism's exposure to contaminants in the environment are of sufficient duration to result in contaminant uptake by the organism. This model was developed to allow estimation of concentrations and doses to biota for both hazardous chemicals and radionuclides; however, its structure allows its use in estimating concentrations in animals that humans might consume as well. The ECEM equations to estimate tissue concentration are described below for ingestion (similar sets of equations are also used to estimate animal

exposures from dermal contact and inhalation – these are not regularly used for radionuclides). The basic ECEM structure is predicated on equilibrium absorption and loss rates from the animal.

**Water Ingestion:** The equilibrium contaminant tissue concentration in species *i* from ingestion of water is calculated from the following expression:

$$C_{ingwi} = D_{ingwi} \left( \frac{\alpha_{i,water}}{K_i} \right) \quad (30)$$

where

- $C_{ingwi}$  = tissue concentration in species *i* from ingestion of water ( $\mu\text{g}/\text{kg}$  wet tissue or pCi/kg wet tissue)
- $D_{ingwi}$  = applied daily dose to species *i* from water ingestion ( $\mu\text{g}/\text{kg}$  wet tissue/day or pCi/kg wet tissue/day)
- $\alpha_{i,water}$  = ingestion absorption factor for species *i* from water ingestion (unitless)
- $K_i$  = loss rate of contaminant for species *i*, including depuration and metabolism (1/day).

The applied daily dose to species *i* from water ingestion is calculated from the equation (modified from EPA [1993]):

$$D_{ingwi} = \theta_i \psi_i \left[ \frac{EC_{water} WI_i}{W_i} \right] \quad (31)$$

where

- $D_{ingwi}$  = applied daily dose to species *i* from water ingestion ( $\mu\text{g}/\text{kg}$  wet tissue/day or pCi/kg wet tissue/day)
- $EC_{water}$  = concentration in surface water ( $\mu\text{g}/\text{L}$  or pCi/L)
- $WI_i$  = water ingestion rate of species *i* (L/day)
- $W_i$  = body weight of species *i* (kg wet tissue)
- $\theta_i$  = area use factor for species *i* (ratio of contaminant area to home range – unitless)
- $\psi_i$  = seasonality factor for species *i* (fraction of year spent at the contaminated site – unitless)

**Soil Ingestion:** The contaminant body burden to species *i* from ingestion of soil is calculated from the equation:

$$C_{ingsi} = D_{ingsi} \left( \frac{\alpha_{i,soil}}{K_i} \right) \quad (32)$$

where

- $C_{ingsi}$  = contaminant tissue concentration in species *i* from ingestion of soil ( $\mu\text{g}/\text{kg}$  wet tissue or pCi/kg wet tissue)
- $D_{ingsi}$  = applied daily dose to species *i* from soil ingestion ( $\mu\text{g}/\text{kg}$  wet tissue/day or pCi/kg wet tissue/day)

- $\alpha_{i,soil}$  = ingestion absorption factor for species  $i$  from soil ingestion (unitless)  
 $K_i$  = loss rate for species  $i$ , including depuration and metabolism ( $\text{day}^{-1}$ ).

The applied daily dose to species  $i$  from soil ingestion is calculated from the equation (modified from EPA [1993]):

$$D_{ingsi} = EC_{soil} SI_i NIR_{totali} F_{dwi} \theta_i \psi_i \quad (33)$$

where

- $D_{ingsi}$  = applied daily dose to species  $i$  from soil ingestion ( $\mu\text{g}/\text{kg}$  wet tissue/day or  $\text{pCi}/\text{kg}$  wet tissue/day)  
 $EC_{soil}$  = concentration in soil ( $\mu\text{g}/\text{kg}$  dry or  $\text{pCi}/\text{kg}$  dry)  
 $SI_i$  = soil ingestion rate of species  $i$  ( $\text{kg}$  soil ingested/ $\text{kg}$  dry diet)  
 $NIR_{totali}$  = total normalized ingestion rate for species  $i$  ( $\text{kg}$  prey wet tissue/ $\text{kg}$  predator wet tissue/day). “Prey” may include plant species in this definition.  
 $F_{dwi}$  = conversion factor, dry diet to wet diet for species  $i$  ( $\text{kg}$  dry tissue/ $\text{kg}$  wet tissue)  
 $\theta_i$  = area use factor for species  $i$  (ratio of contaminant area to home range – unitless)  
 $\psi_i$  = seasonality factor for species  $i$  (fraction of year spent at the contaminated site – unitless)

The total normalized ingestion rate for species  $i$  is calculated from the equation (EPA 1993):

$$NIR_{totali} = \frac{FMR_i}{W_i \sum_{j \neq i} (P_{ij} ME_j)} \quad (34)$$

where

- $NIR_{totali}$  = total normalized ingestion rate for species  $i$  ( $\text{kg}$  prey wet tissue/ $\text{kg}$  predator wet tissue/day)  
 $FMR_i$  = free-living metabolic rate of predator species  $i$  ( $\text{kcal}/\text{day}$ )  
 $W_i$  = body weight of species  $i$  ( $\text{kg}$  wet tissue)  
 $P_{ij}$  = wet weight or volume fraction of  $i$ 's diet consisting of prey  $j$  (unitless)  
 $ME_j$  = metabolizable energy from prey  $j$  ( $\text{kcal}/\text{kg}$  prey wet tissue)

The metabolizable energy from prey  $j$  is calculated from (EPA 1993):

$$ME_j = GE_j AE_j \quad (35)$$

where

- $ME_j$  = metabolizable energy from prey  $j$  ( $\text{kcal}/\text{kg}$  prey wet tissue)  
 $GE_j$  = gross energy from prey  $j$  ( $\text{kcal}/\text{kg}$  wet tissue)  
 $AE_j$  = assimilation efficiency of prey  $j$  (unitless)



**Food Ingestion:** The equilibrium contaminant tissue concentration in species  $i$  from ingestion of food is calculated from the following expression:

$$C_{ingfi} = D_{ingfi} \left( \frac{\alpha_{ij}}{K_i} \right) \quad (36)$$

where

- $C_{ingfi}$  = equilibrium tissue concentration in species  $i$  from ingestion of food ( $\mu\text{g}/\text{kg}$  wet tissue or  $\text{pCi}/\text{kg}$  wet tissue)
- $D_{ingfi}$  = applied daily dose to species  $i$  from food ingestion ( $\mu\text{g}/\text{kg}$  wet tissue/day or  $\text{pCi}/\text{kg}$  wet tissue/day)
- $\alpha_{ij}$  = ingestion absorption factor for species  $i$  from ingestion of food species  $j$  (unitless)
- $K_i$  = loss rate of contaminant for species  $i$ , including depuration and metabolism (1/day)

The applied daily dose to species  $i$  from food ingestion is calculated from the equation (modified from EPA [1993]):

$$D_{ingfi} = \theta_i \psi_i \sum_j (C_j \text{ NIR}_j) \quad (37)$$

where

- $D_{ingfi}$  = applied daily dose to species  $i$  from food ingestion ( $\mu\text{g}/\text{kg}$  wet tissue/day or  $\text{pCi}/\text{kg}$  wet tissue/day)
- $\theta_i$  = area use factor for species  $i$  (ratio of contaminant area to home range – unitless)
- $\psi_i$  = seasonality factor for species  $i$  (fraction of year spent at the contaminated site – unitless)
- $C_j$  = average contaminant concentration in  $j^{\text{th}}$  food item ( $\mu\text{g}/\text{kg}$  wet tissue or  $\text{pCi}/\text{kg}$  wet tissue)
- $\text{NIR}_j$  = normalized ingestion rate of  $j^{\text{th}}$  food type on a wet-weight basis ( $\text{kg}$  prey wet tissue/ $\text{kg}$  predator wet tissue/day)

The normalized ingestion rate of  $j^{\text{th}}$  food type on a wet-weight basis is calculated (EPA 1993) as:

$$\text{NIR}_j = P_{ij} \text{ NIR}_{totali} \quad (38)$$

where

- $\text{NIR}_j$  = normalized ingestion rate of  $j^{\text{th}}$  food type on a wet-weight basis ( $\text{kg}$  prey wet tissue/ $\text{kg}$  predator wet tissue/day)
- $P_{ij}$  = wet weight or volume fraction of  $i$ 's diet consisting of prey  $j$  (unitless)
- $\text{NIR}_{totali}$  = total normalized ingestion rate for species  $i$  ( $\text{kg}$  prey wet tissue/ $\text{kg}$  predator wet tissue/day)

The term  $P_{ij}$  is a large matrix describing which animal eats which plant or other animal.

**Total Terrestrial Animal Burden:** The equilibrium tissue concentration for a contaminant in terrestrial animal species  $i$  is then calculated from the sum of the dermal, inhalation, and ingestion exposures. As can be seen from these equations, this model requires a great deal of information, including the absorption and depuration rates and the predation (food source) matrix. Much of this can be approximated, or estimated using allometric analyses (see below).

### 3.4.6 Allometric Models for Uptake by Animals

A key limitation to the use of transfer factors to relate intake of contaminants by various animal types to concentrations in animal products is the lack of measurements of various radionuclides in most animals. A developing technique for estimating the transfer of contaminants in animal products is allometry (Higley, Domotor, and Antonio 2003).

The application of transfer factors for many types of animal products is problematic because of the wide range of organisms potentially exposed. For example, there are very limited data available for riparian and terrestrial animals (i.e. very few terrestrial animal:water and riparian animal:sediment concentration ratios). An alternative approach, called the kinetic/allometric method, was developed. Among other objectives, this work filled in data gaps in the literature on transfer factors for specific riparian and terrestrial animal receptors.

Allometry, or the biology of scaling, is the study of size and its consequences. There are several allometric equations that relate body size to many parameters, including ingestion rate, life span, inhalation rate, home range and more (Schmidt-Nielsen 1977; West et al. 1997; Wilkie 1977). The most common form of allometric equation is the power function

$$Y = \alpha X^{\beta} \quad (39)$$

where  $Y$  and  $X$  are size related measures, and  $\alpha$  and  $\beta$  are constants. Allometric equations provide a way to estimate a biological process given a specific set of parameters. While these equations were originally derived from empirical observations, there is a growing body of evidence to suggest that these relationships have their origins in the dynamics of energy transport mechanisms. For example, allometry has been used to estimate the daily rate of consumption by an animal in relation to its body size. Metabolic rate is known to scale to body mass to the  $3/4$  power (Calder 1984; Reiss 1989; West et al. 1997). This scaling has been found to extend to poikilothermic and ectothermic animals (Bennett and Dawson 1976) as well as unicellular ones (Wilkie 1977). Intraspecific scaling of metabolism has been found to vary somewhat, with the exponent of body weight larger for juveniles than adults (Reiss 1989). However, allometric equations are meant to represent qualitative trends over orders of magnitude of body mass. Consequently, these general allometric relationships can be used as general predictors regardless of age, or phyla to provide species-independent estimates of uptake.

Higley, Domotor, and Antonio (2003) have developed equations to predict concentrations of radionuclides in tissues for terrestrial and riparian animals exposed to contaminated soil, sediment or water. The equations were developed as a means to provide estimates of transfer factors in the absence of data for particular species for radionuclides in soil, water or sediment. The allometric approach provides a potentially useful method to extrapolate radionuclide behavior across multiple species. While the method is simplistic, variations of this technique can be used to roughly estimate tissue concentrations, biological elimination rates, and uptake factors across a wide range of organisms. In the absence of measured data it can provide a starting point for modeling environmental exposure to contaminants.

### **3.5 Modeling the Effects of Food Processing on Radionuclide Concentrations**

Changes in concentration in foods as a result of food preparation practices and activities are not commonly included in assessment models. Although such processes are routinely acknowledged, there is a relative scarcity in the literature of information as to how they should be handled. Evidence exists that soaking, peeling, boiling, or baking can effectively halve concentrations in consumed food. (NRPB 1995; Watterson and Nicholson 1995; Jackson and Rickard 1998). For instance, the EPA Exposure Factors Handbook does not even include a section about this process (EPA 1997). “Dose calculations do not normally make allowance for such reductions, partly on the basis that considerable variability may exist between individuals and that, for instance, water or fats used for cooking may themselves be contaminated, so that any dose saving overall would be marginal” (Jackson et al. 1998). In addition to that, there is a growing segment of the general population that tend to reduce its consumption of the heavily processed foodstuffs, as well as meat, poultry and other products.

Most radiological assessment codes use a simple approach to determining contaminant concentrations in food crops; the concentrations in the crops are determined using a surface-deposition-to-concentration translocation factor for above-ground deposition, and a soil-to-edible-plant concentration ratio for growth in contaminated soils. This differs somewhat from the EPA approach to the same problems for dioxins and other lipophilic contaminants (EPA 1994, EPA 1998). The EPA approach uses a soil-water-to-bulk contamination ratio rather than a soil-to-bulk ration, and uses a  $K_d$  to relate soil contamination to soil-water concentrations. A further assumption in the EPA method for these contaminants is that these compounds do not transfer from contaminated surfaces (leaves or roots) into the vegetation, but are sorbed onto the surface. They then define an “empirical correction factor” (VG) that is essentially the ratio of the mass of the skin of the vegetable to the mass of the whole vegetable, further simplified with the assumption that the densities of the skin and bulk volume are the same, so that the ratio is of the volume of skin to total volume of vegetable. This correction factor is then adjusted for washing and peeling. The justification given in EPA (1994) is “Additional reductions in concentration result from peeling, cooking, or cleaning, for example. Wipf, et al. (1982) found that 67% of unwashed carrot residues of 2,3,7,8-TCDD came out in wash water, and 29% was in the peels. A peeled, washed carrot correction factor might ... be, ... 0.04,

(... 0.04 = 100% - 67% - 29%). A 96% reduction in the estimated VG for the potato (the potato is cleaned and the skin is not eaten; additional reductions possibly when cooking the potato) would equal 0.01. In a site-specific application, the type of vegetation, preparation, and so on, should be considered.” The adjustment given by EPA is thus a combination of the washing and peeling and the assumption that nearly all contamination is in the skin. This sort of correction factor should not be used with the soil-to-edible-plant concentration ratio models, because the CR used should already contain the assumption of low translocation and soil-to-plant transfer. Only the fraction of the reduction due to washing is always pertinent, and the peeling portion should be considered.

The NATO Manual FM-8-10-7 (Army 1993), Appendix F, gives decontamination procedures for foods contaminated by nuclear, chemical, and biological (NBC) agents. The procedures are designed for contamination resulting from NBC combat, and the decontaminated foods are intended for consumption by soldiers or civilians in a post-combat situation; however, the processes and techniques are common ones that also could apply to home uses. As described in this manual, “some products can be decontaminated by washing, peeling, or trimming the outer skin or leaves. Decontaminate potatoes and hard-skinned fruits and vegetables by washing or scrubbing under running water, followed by peeling or scraping, then washing again. Potatoes, carrots, beets, and turnips can be washed at the supply depot. However, do not wash beans, rice, and onions until they are delivered to the field kitchen; washing reduces their storage quality and shelf life. Citrus fruits, pineapples, corn, peas, beans, melons, pumpkins, cabbage, and nuts can be peeled. Decontaminate cucumbers, tomatoes, cherries, cranberries, grapes, pears, plums, and thin-skinned squash by soaking in a water or detergent solution and rinsing with vigorous agitation or brushing. Apricots, peaches, most berries, asparagus, broccoli, and leafy vegetables cannot be satisfactorily decontaminated because of fuzzy surfaces, irregular shapes, or small size which makes washing difficult. It is not practical to decontaminate food contaminated through the food-chain. Meat and milk are the two most common foodstuffs contaminated in this way.

“Milk may be decontaminated to a safe level by a complicated ion exchange process. The I-131 activity will decline rapidly during storage of milk and milk-products, although the Cesium and Strontium activity will remain almost constant for years. In an area with high-level fallout, milk is withdrawn from human consumption. The duration of withdrawal will be dependent upon the type of fallout and levels. Meat may be decontaminated to a safe level by soaking in water or brine. Cesium is loosely bound in the meat. By repeated soaking of meat cut in small pieces, most of the Cesium activity will be removed. Traditional meat preserving, such as salting with brine, will remove up to 60 to 70 percent of the Cesium activity.” (Army 1993).

A fairly extensive review of available literature was conducted in association with the VAMP program of the IAEA (Noordijk and Quinalt 1992). This reference introduces a food processing retention factor,  $F_r$ , which is defined as the total amount of a contaminant in processed food divided by the total amount of contaminant in the original raw food. This report also introduces a related parameter, the processing efficiency  $P_e$ , which is the

mass of prepared product divided by the mass of raw material. Numerous tables of these values for various foods processed via various methods are presented. Averages and ranges are presented in these tables. The ranges tend to be broad, and the average  $F_r$  values appear to be around 0.5.

Noordijk and Quinalt (1992) stress that all  $F_r$  values referring to extraction procedures such as cooking, frying, etc., are only valid when the extraction liquid is removed and not used for other culinary purposes. The same applies to fractionation processes which take place, such as in the dairy industry or during the milling of grains. For example, the by-product of cheese making, whey, was formerly regarded as a waste or used to feed animals. A large part of produced whey today is used as an additive for food for humans. Similarly, all fractions of the milling process are usually used to produce food. In these cases, an  $F_r$  value of 1.0 should be used.

Noordijk and Quinalt (1992) summarize that the effects of processing on the behavior of contaminants depends on the contaminant, on the type of product, and on the method of processing. Processing effects are rather small for root crops, whereas milling cereal grains to flour will often remove about 70% of the contamination. The effects of processing on vegetables and fruits are rather unpredictable, especially when the contaminants are adsorbed on the surface of the plants.

Green and Wilkins (1995) extend the efforts of the IAEA study by Noordijk and Quinalt (1992). This review of the literature uses the same concepts (while renaming  $F_r$  as  $R_i$ ) and adds additional references. This review also has reasonably detailed discussions of the applicability of the reported values for application to critical group analyses. In general, Green and Wilkins recommend values of 1.0 because of uncertainties in whether liquids used in food processing would be consumed.

A possible method of handling the complexities of the food processing retention factor was developed for this report. The method incorporates the uncertainties of both the retention in food of the contaminants, the variabilities in retention factor  $F_r$  for different types of cooking technique, as well as the uncertainties in whether the cooking liquors are consumed. The concept can be described as:

$$C_{eaten} = C_{raw} [ F_r + ( 1 - F_r ) S ] \quad (40)$$

where  $C_{eaten}$  = the concentration in cooked or processed foods,  
 $C_{raw}$  = the concentration in raw or unprocessed foods, and  
 $S$  = the fraction of cooking liquids and sauces that is consumed.

This formulation assumes that the contaminants lost in the food preparation process are released into the cooking liquids.

A complication to the process is that there are different types of food preparation processes. In evaluation of the data in the various reports, it appears that “dry cooking” techniques such as broiling, baking, or frying have different removal efficiencies than

“wet cooking” techniques such as boiling, steaming, or stewing. In addition, different types of foods will have different proportions of each preparation technique. These can be accommodated by expanding the general equation for specific applications, as:

$$C_{eaten} = C_{raw} [ f_{dry} \{ F_{r\_dry} + (1 - F_{r\_dry}) S_{dry} \} + f_{wet} \{ F_{r\_wet} + (1 - F_{r\_wet}) S_{wet} \} + f_{raw} ] \quad (41)$$

In this formulation, the total intake of each type of food is subdivided into raw or unprocessed, wet cooked, and dry cooked. The approach may also be expanded to include other types of processing, as for milk, which includes fresh milk, cheeses, and whey.

It seems apparent that this approach is impractical, because it has introduced 3 fractions, two processing retention factors, and a fractional consumption of cooking liquors for each food type. It is unlikely that data will be available to populate such a model, although there might be data available in the 1977 National Food Consumption Survey to begin defining such parameters. However, in a stochastic model, wide ranges of uncertainty may be combined into a single compound distribution.

A Crystal Ball® enabled EXCEL spreadsheet was used to evaluate the possible distributions that might be encountered. The fractions of foods eaten in raw, wet-cooked, and dry-cooked states were broadly estimated by the author. The ranges for each of the possibilities were assigned uniform distributions. Most are wide enough to accommodate nearly all eventualities; none of the results are overly sensitive to the assumptions. The total fraction of foods consumed for each type was normalized to 1.0; this ensures conservation of mass, but it also tends to minimize the sensitivity of the answer to the specific distributional assumptions of the consumption fractions. The ranges of food retention parameters are taken from Green and Wilkins (1995). Using the above general equation, food processing retention factor distributions were generated for the common food types used in risk assessments: leafy vegetables, other vegetables, fruits, grains, meat, and milk.

The results of the computations confirm that for deterministic, maximum individual or critical group calculations, a processing retention of 1.0 is a reasonable and not overly-conservative assumption. A value of 1.0 is always possible, and mean values for most food types are around 0.8 to 0.9. For leafy vegetables and other vegetables, the mean is about 0.8, ranging from 0.4 to 1.0. For fruits and grains, the mean is nearly 0.95, and the range is well under a factor of 2. The range for meat is from 0.3 to 1.0, with a mean of about 0.8. Only for milk is there a substantial variability and non-linearity: the result depends heavily on how much cheese is eaten in proportion to milk, because the making of cheese tends to reduce the contaminant concentrations.

As a result of the investigation, the distributions developed for the analysis are available for use. All of them are quite stable, except for the milk-processing factor. The results are summarized in Table 8. All distributions are simple – either uniform or triangular. Several approach right triangles, where the mode equals the maximum at 1.0. For milk, a

triangular distribution is suggested, but the mode (most likely) value is essentially equivalent to the adjusted fraction of milk intake assigned to fresh milk; i.e., if 80 percent of milk intake is fresh, the mode of the triangle should be at 0.8, and if 50 percent of milk intake is fresh, the mode of the triangle should be at 0.5. This is the only parameter with sufficient sensitivity to warrant case-specific consideration in developing of exposure scenarios.

**Table 8.** Recommended Values of Food Processing Retention Factors for Most Contaminants

Food Type	Distribution Type	Minimum	Mode	Maximum
Leafy Vegetables	Uniform	0.5	0.85	1.0
Other Vegetables	Triangular	0.6	0.95	1.0
Fruits	Triangular	0.9	1.0	1.0
Grains	Uniform	0.9	-	1.0
Meat	Triangular	0.5	0.95	1.0
Milk	Triangular	0.25	0.8*	1.0

\* Distribution for milk is dependent on the fraction of milk intake assigned to cheese. The case here assumes 20 percent of milk intake results from consumption of locally-made cheese.

The radionuclide tritium ( $^3\text{H}$ ) is a special case. The food processing retention factors for tritium could be lower, because cooking tends to drive off water in foods, wherein most of the tritium resides. The spreadsheet calculations described above for other contaminants were modified and performed for tritium. The results are presented in Table 9. Note that there is relatively little difference from Table 1, indicating that it may not be necessary to have any contaminant-specific inputs.

**Table 9.** Recommended Values of Food Processing Retention Factors for Tritium

Food Type	Distribution Type	Minimum	Mode	Maximum
Leafy Vegetables	Uniform	0.5	0.85	1.0
Other Vegetables	Triangular	0.6	0.9	1.0
Fruits	Triangular	0.75	1.0	1.0
Grains	Uniform	0.99	-	1.0
Meat	Triangular	0.6	0.9	1.0
Milk	Triangular	0.5	1.0	1.0

## 3.6 Modeling Other Terrestrial Exposure Pathways

In the preparation of this report, a few unique examples of terrestrial human exposure pathways were identified. Although not part of standard food-chain pathways, these additional pathways deserve attention.

### 3.6.1 Unique Direct Pathways

The pathway of showering with contaminated water, leading to inhalation exposure, is common to non-radiological assessments dealing with volatile chemical compounds (US EPA 1997); however, with the notable exception of radon, it is infrequently examined in radiological assessments because most radionuclides are not considered to be volatile. Even non-volatile contaminants can become entrained in water droplets and inhaled. For some radionuclides with very low GI-tract uptake, lung doses from this mechanism can be higher than other organ doses from direct drinking of the water. The concentration of radionuclides in the air of the shower can be related to the quantity of mist droplets generated, using the absolute humidity of the air (Andelman 1990). As a general rule, radionuclide intakes via this pathway tend to be small. However, tritiated water can transpire through the skin, and some models (e.g. GENII) sometimes multiply the nominal inhalation dose conversion factor by a factor of 1.5 to account for it (Napier et al. 1988).

A similar exposure pathway is Sweat bathing as part of the Sweat Lodge Ceremony, a traditional Native American custom (DOE 1997; Harris and Harper 1997). Based on tribal descriptions, between 0.5 and 3 hours/day on a weekly basis is assumed to be spent inside a sweat lodge kept at 60 - 80 degrees Centigrade (145 - 180 degrees Fahrenheit). No or very limited clothing is worn by the Ceremony participants. A large amount of wood is used to heat up the rocks in an open outside fire adjacent to the Lodge. The sweat lodge is heated with these hot rocks, onto which water is poured to create steam. Air inside the sweat lodge is assumed to be saturated with water (equivalent to 0.3 kilograms of water per m<sup>3</sup> of air, and 0.3 L/m<sup>3</sup> of semivolatiles and 2.5 L/m<sup>3</sup> of volatiles), which are then available for inhalation and dermal absorption over the entire body. During the 1 hour of use, 4L of water is used. Doses resulting from intakes of some radionuclides via this pathway can be larger than the associated doses via drinking water if the radionuclides have low GI-tract absorption, since this is a mechanism for inhalation of large amounts of water.

Evaporative coolers are commonly used in the hot, arid American West. Evaporative coolers cool air by evaporating a stream of water into a stream of air. These coolers might transfer water-borne contaminants to the indoor air. Thus, modeling must include an estimate of the radionuclide concentrations in indoor air when evaporative coolers are in operation so that the radiation dose for the human receptor who inhales the contaminated air can be evaluated. Based on how evaporative coolers operate and the conservation of radioactivity (i.e., activity transferred to air is equal to the loss of activity from water), radionuclide concentrations in indoor air are estimated for this pathway by Wu (2003) as



$$Ca_{ei} = f_{evap} \frac{M_{water}}{F_{air}} Cw_i \quad (42)$$

where

$Ca_{ei}$	=	activity concentration of radionuclide $i$ in the air resulting from the operation of an evaporative cooler (Bq/m <sup>3</sup> )
$f_{evap}$	=	fraction of radionuclides in water transferred to indoor air (dimensionless)
$M_{water}$	=	water evaporation rate (water use) for an evaporative cooler (m <sup>3</sup> /hr)
$F_{air}$	=	air flow rate for an evaporative cooler (m <sup>3</sup> /hr)
$Cw_i$	=	activity concentration of radionuclide $i$ in the groundwater (Bq/m <sup>3</sup> ).

The fraction of radionuclides that remain in the reservoir, bleed-off water, or in the pads of evaporative cooler are not further modeled because it becomes a regular water source for irrigation or sewage, or is removed when the pads are replaced, and therefore may not cause direct exposure (although for the case of continuous deposition of gamma emitters on the pad when the pad is replaced only once in every several years, there might be some concern of the long-term continuous direct exposure to the radionuclides deposited on the pad, depending on the location of the evaporative cooler inside the dwelling). Evaporation and air flow rates are estimated based on specifications of residential evaporative cooling units. The typical evaporation rate is about 20 L/hr, and typical air flow rates range from 2,000 to 10,000 m<sup>3</sup>/hr (BSC 2003). The fraction of radionuclides transferred from the water to the indoor air is an important parameter that is not available in the literature, but the theoretical range is from 0 to 1 (BSC 2003). The calculation of activity concentrations in the air resulting from evaporative coolers does not include consideration of radionuclide buildup in the indoor air, because the air flow associated with the use of these coolers is such that during operation the windows of the building remain open and the number of air exchanges per hour is quite large. The calculated indoor air concentrations are usually low because the air flow rate of the coolers is so high.

### 3.6.2 Unique Indirect Pathways

Contamination of trees in Europe with <sup>137</sup>Cs following the Chernobyl accident has led to detectable concentrations of radionuclides in wood. In villages in Ukraine and Belarus, wood is a common kitchen fuel. Wood ashes have been found to routinely exceed 50,000 Bq/kg. Besides being a source of direct external exposure, the wood ashes were being distributed in kitchen gardens as soil amendments. The total administration of ashes in one year was equivalent to about 25% of the total <sup>137</sup>Cs deposition on the garden from the initial accident. Over a period of many years, this process resulted in reconcentration of cesium and creation of an ingestion pathway (Dubreuil et al. 1999) that could be multiples of 2 or 3 greater than the initial fallout soil contamination in the gardens.

Groundwater beneath the Hanford Site in eastern Washington State is contaminated with tritium at levels several times that of the drinking water standards (Poston, Hanf, and Dirkes 2005). Trees near the Columbia River have roots that penetrate to the groundwater. The trees have been found to not only contain tritium, but to transpire it to the atmosphere. A similar source was documented at the Oak Ridge Site under Solid

Waste Storage Area Five. In essence, the trees are a source of atmospheric release of groundwater tritium. However, the resulting atmospheric concentrations are low because of the large dispersion.

## 4.0 Discussion

A number of alternative conceptual models for the key processes and mechanisms related to modeling the transport of radionuclides through the terrestrial biosphere have been presented. Several of the processes are not well known (e.g., foliar absorption and translocation, non-linear uptake of nutrient analogues, numerous transfer factors for understudied radionuclide plant/animal combinations) and offer areas for fruitful research.

As a summary of the presentations, the following currently available techniques are the most likely to be successful in generic applications.

- For *soil*, an annual average model incorporating:
  - Accumulation from irrigation or atmospheric deposition;
  - Leaching to deeper soil, using a  $K_d$  -modulated leach rate;
  - Harvest removal, averaged over crop types
  - Uniform mixing in a reasonable (15-25 cm) soil surface layer, implicitly caused by plowing, bioturbation, and leaching;
  - Neglecting radionuclide fixation, because appropriate long-term measurements of distribution coefficient and concentration ratio will already incorporate the effects;
  - Neglecting surface-soil erosion losses, because eroded material from one location may accumulate in another, cancelling any perceived benefit.
- For *resuspension*, a mass-loading approach, because it has the lowest variability and is the most easily defended.
- For *foliar interception*, models incorporating:
  - Dry interception following the basic model suggested by Chamberlain (1967) as updated by Adriano et al. 1982;
  - Wet interception considering one of the algorithms derived from the data of Hoffman et al. (1989) or Prohl and Hoffman (1993).
- For *plant contamination with soil*, an adhesion model applied so that material translocated is not subject to weathering (e.g., IAEA 2003; Wu 2003).
- For *weathering*, a single exponential model with a half-time between about 10 and 20 days, applied to material on the surface of the plant only.
- For *translocation*, a radionuclide-specific (or chemical-class-specific) implementation allowing translocated materials to avoid being removed via weathering processes (e.g., IAEA 2003; Wu 2003).
- For *soil-to-plant transfer*, a radionuclide- and plant-type-specific concentration ratio, where available, with:
  - Site-specific application of additional crops, such as mushrooms and berries;
  - The specific-activity model of Peterson and Davis (2002) for elemental and oxide forms of tritium;

- A general specific-activity model for  $^{14}\text{C}$  in air, adapted to the RESRAD (Yu et al. 2001) model for transfer from irrigation water to air as  $^{14}\text{CO}_2$ ;
- Perhaps site-specific application of specific-activity models for iodine and other micro-nutrient and macro-nutrient elements and their chemical analogues. This is a simplification of the need to allow for homeostatic regulation of some elements.
- Until a simple, robust *Fruit Tree* model is developed, continue to use the concentration ratio approach for fruits.
- For *animal products*, a transfer factor approach applied to average daily feed, water, and soil intake by the animal where data are available.
  - For other element/animal combinations, a justified mix of specific-activity and allometric models should be considered.
- For *food processing*, because the data are sparse and the reduction is generally small, modelers are justified in ignoring losses (which is equivalent to using a food-processing transmission factor of 1.0).

Site-specific models for additional little used or unusual pathways (e.g., honey, evaporative cooling systems) may need to be developed, justified, and used as appropriate for the system and environment being analyzed.

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