



## APPROACHES TO MODELLING RADIONUCLIDE TRANSFER IN AGRICULTURAL SYSTEMS

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### 1. Introduction

Radiological dose assessment requires information describing the concentration and distribution of radionuclides in the environment. This information can be obtained from monitoring but is also evaluated with the aid of mathematical models. In such models the pathways of radionuclides from the release point to man are described in terms of transfer between compartments.

The main pathways to be considered include: deposition to vegetation and soils; transfer from soil-to-plant; uptake and turnover in domestic animals; and, intake by man. The development of mathematical models for simulating transfer via these pathways depends on: an understanding of the system under study, in particular for those processes that are most important in the overall transfer to man; the availability of data to determine the structure and parameters for the model; the computing systems available; the knowledge of the user of the model; and, the application of the model.

### 2. Equilibrium versus dynamic models

Deterministic models describing food chain transfer of radionuclides fall into two general categories of equilibrium and dynamic models. Selection of an appropriate model for assessing ingestion dose depends largely on the type of release and available input data but selection of transfer parameters depends on the purpose of the assessment (e.g. generic or specific use, best or conservative estimates).

Equilibrium models, such as those described by IAEA (1982), are used to assess the radiological impact of routine releases from nuclear plant. Transfer parameters often represent the ratio of concentrations in two compartments for equilibrium conditions and parameter values are often selected to provide a conservative estimate of dose. Such models also tend to conserve activity within compartments; for example, it may be assumed that repeated cropping does not reduce radionuclide concentration in soil, or alternatively,

deposition to soil is not reduced by the amount that is intercepted by crop foliage. The soil-plant transfer factor is used by equilibrium models.

The main advantages of equilibrium models are as follows: simple in concept and in theory, easy to understand; the parameter set is well defined (deposition velocity, interception, weathering half-life, soil-to-plant and plant-to-animal or aggregate transfer coefficients); and, the models are computationally fast to solve and amenable to probabilistic methods. Disadvantages include: the results are only applicable to timescales relating to the longest equilibrium period; the models require a comprehensive databank for all environmental conditions; and, the approach does not allow interpretation of underlying mechanisms.

There are several advantages to dynamic models. They attempt to model the time-dependence of foodchain transfer and can therefore be used to assess both operational and accidental releases. The models are able to account for interacting transport mechanisms and processes resulting in the contamination of food products. The models can therefore be applied to discharges that vary as a function of time and be used to analyse pathways and environmental conditions. However, such models require a larger parameter database that is not always available and frequently cannot be derived directly from the substantial body of data reported as concentration ratios without making several major assumptions (e.g. yield, soil bulk density). A further drawback of dynamic models which calculate only content ( $\text{Bq m}^{-2}$ ) in the edible plant part from a known soil content ( $\text{Bq m}^{-2}$ ), is the need for additional input data on plant yield to calculate concentrations.

There are models that fall partly between the two types discussed above, these use equilibrium data to drive a dynamic model such that equilibrium concentrations are set up within a defined time period. This approach is valid only when the model is applied consistently with the time periods upon which transfer parameters are calculated. Alternatively, there are methodologies that use both modelling approaches.

There is a mis-conception that equilibrium models tend to be simple and generic, and that dynamic models are complex and site specific. Such characterisations are invalid and subjective. Model complexity is dependent on the design requirement and site-specificity is dependent on the availability of relevant information with which to apply the model in a given situation.

The parameters for a simple model can be selected either by taking an average for each parameter from all observed values or by selecting values based on a detailed knowledge

of those factors that influence each parameter. Where the user attempts to apply the model to a specific situation a decision support facility is needed to assist with the selection of appropriate parameters. With increasing model complexity the number of parameters that must be selected increases. However, if the more complex model is intended to simulate underlying processes then the number of factors that need to be considered when selecting each parameter value should decrease.

The available data that can be used to construct a dynamic model is limited, partly by the trend to report data as equilibrium transfer values but also because non-equilibrium data are often reported in the same way. The last point concerning different models is that individual dynamic models tend to have a specific compartmental structure. There are in general two approaches when specifying a model, these are to take either a generalised structure that represents the system and provide a fit to available data or to select a compartmental structure that strikes a balance between the available data and the design requirement. In both cases predictive uncertainty would be reduced if data on radionuclide fluxes were more readily available.

The SPADE models are an example of dynamic foodchain transfer models. The models are mostly of the compartmental type, where the transfer from compartment A to compartment B is simulated according to first order kinetics, ie.

$$A(t) = A_0 e^{-k(a,b)t}$$

where  $A(t)$  is the content of compartment A at time  $t$ ,  $A_0$  is the original content of compartment A and  $k(a,b)$  is the effective transfer from compartment A to compartment B. It is important to note that  $k(a,b)$  is assumed to be: a function of the content of compartment A; time-independent; and, independent of the size of either compartment A or compartment B. However, in biological systems  $k(a,b)$  is: dependent on the concentrations in compartments A and B and often a function of physiological factors and therefore of time. These factors can be very important for young growing animals or in the case of radionuclides such as  $^{14}\text{C}$  and  $^3\text{H}$  that are incorporated in organic compounds.

### 3. Identification of suitable data

There are several sources of information that can be used for radionuclide transfer model parameter databases. These include data obtained direct from field and experimental investigations, published literature, and existing databases of parameters.

Fallout from the atmospheric testing of nuclear weapons provided the impetus to acquire data on the behaviour of radionuclides in the environment. Many data were obtained during the late 1950s and early 1960s on the fate of some radionuclides in particular ecosystems in order to determine the risk to man resulting from fallout (Russell, 1963). The amount of fallout radioactivity entering food chain pathways was found to vary throughout Europe due to various factors including rainfall, latitude and soil-plant interactions (Pavlotskaya, 1971). Monitoring programmes were able to identify groups of individuals that were at greatest risk from specific combinations of radionuclide and food chain pathway.

The pasture grass-cow-milk (Garner, 1969) and lichen-deer (Rahola & Miettinen, 1977) pathways received much attention. Initially, experimental studies considered those factors which could influence the long-term consequences of fallout, in particular, whether ameliorative techniques could be used to reduce the risk to man (Howorth & Sandalls, 1987). Detailed investigations were limited to a small number of radionuclides ( $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{131}\text{I}$ ) and in most cases to a limited number of crops (forage species and cereals). Those properties of soils and crops which influenced radionuclide transfer were investigated using a variety of different growing media such as culture (nutrient) solution, split pot techniques, clay suspensions, disturbed and undisturbed soil in various sized containers and soils under field conditions (Cawse & Turner, 1982). Such investigations provide some data of use for equilibrium models but very little of use for dynamic soil-plant models.

The use of radio-tracers in agricultural research as analogues of plant nutrients provides additional information on radionuclide fate in agro-ecosystems (Newbould & Taylor, 1965). Other useful sources of information include stable element studies (Lisk, 1972) and investigations related to metal ore mining (Schüttelkopf & Kiefer, 1982) and processing industries (Crecelius *et al.*, 1974) which release heavy metals into the environment.

Extensive data come from studies performed by researchers in the United States of America. These have been carried out over a wide range of ecosystems in the vicinity of nuclear bomb test sites (Gilbert *et al.*, 1988), nuclear waste disposal sites (Auerbach, 1987), fuel reprocessing complexes (Garland *et al.*, 1983) and in regions where radionuclide levels in the human diet were elevated due to a combination of soil and climatic factors (Gamble, 1971). Although there are obvious similarities between some of the crops, climates and soils studied, and those found under European conditions, the soil-plant transfer data from work in the USA provide unreliable estimates for use in predicting the risk to the European population (Anon., 1982).

In the early 1980s the International Union of Radioecologists (IUR) formed a largely European working group to coordinate research into soil-to-plant transfer of radionuclides (Anon., 1982). It aimed to improve the estimates of risk to the population from the application of nuclear technology in Europe by compiling a data bank of reliable soil-plant transfer factors. The definition of a soil-to-plant 'transfer factor' can vary and it is important that the same definition is used when comparing values. The IUR working group defined a transfer factor for arable crops as follows:

$$\text{Transfer factor} = \frac{\text{Bq kg}^{-1} \text{ dw plant material}}{\text{Bq kg}^{-1} \text{ dw soil (0-20 cm depth)}}$$

and provides guidelines for the experimental protocol. The guidelines aim to ensure that exchanges between soil exchange sites and soil solution are at equilibrium. Values obtained from reviews of the literature (Ng *et al.*, 1982; Coughtrey & Thorne, 1983a&b) are not always adequately defined and are subject to variations in experimental conditions, such as pot size, plant maturity at harvest, fertilizer additions, radionuclide placement and chemical form, soil characteristics, use of irrigation and climatological conditions. The data base that the IUR constructed, which is reviewed and updated periodically, contains several thousand transfer factors with a description of the experimental basis for the derivation of each value (Anon., 1987). The data base provides an important reference for dose assessment using equilibrium-type models.

There remains a heavy bias in the literature world-wide towards transfer factors for certain combinations of radionuclides ( $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{239}\text{Pu}$ ), crops (cereals and pasture grasses) and soil types (clays, loams and sandy soils). Although in recent years research has diversified, it is still the case that limited data are available for many radionuclides (including  $^{106}\text{Ru}$ ,  $^{144}\text{Ce}$ ,  $^{125}\text{Sb}$ ,  $^{237}\text{Np}$  and  $^{244}\text{Cm}$ ), crops (soft fruits, sugar beet and tomatoes) and soils (silts and organic soil types). Evaluation of the immediate radiological impact of abnormal events in nuclear power plants requires dynamic models supported by experimental programmes that are designed to provide relevant model parameters (Thorne & Coughtrey, 1983).

#### 4. Conclusions

The development of time-dependent compartmental models requires data on the amount of a substance transferred between defined compartments over a known time period, supporting data are also required on the volume or mass of donor or receiving compartments to determine concentrations. Although the types of information sources described above will provide new data for dynamic models, much of the existing data produced for equilibrium-type models and are rarely relevant to time-dependent modelling. However, in the last five

years since the Chernobyl accident, more and more data have been generated on the dynamics of radionuclide transfer in terrestrial ecosystems. On-going literature reviews identify information from CD-ROM searches, Current Contents and CAB Abstracts. Where reported data look relevant but are not reported in sufficient detail authors are approached for further information. The quality of the data is a major factor that must be taken into account when constructing models. The IUR attempted to define an experimental protocol for equilibrium transfer studies but this has not been achieved for data relevant to dynamic models. Data must therefore be screened or graded to ensure that emphasis is given to better quality data.

It is important to maintain rigorous quality assurance in both experimental and modelling studies. An experiment may only involve a single measurement or a model parameter may be an informed guess but they are both valid as long as proper records are maintained and the basis for the data is transparent.

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