



This article appeared in a journal published by Elsevier. The attached copy is furnished to the author for internal non-commercial research and education use, including for instruction at the authors institution and sharing with colleagues.

Other uses, including reproduction and distribution, or selling or licensing copies, or posting to personal, institutional or third party websites are prohibited.

In most cases authors are permitted to post their version of the article (e.g. in Word or Tex form) to their personal website or institutional repository. Authors requiring further information regarding Elsevier's archiving and manuscript policies are encouraged to visit:

<http://www.elsevier.com/copyright>

Contents lists available at [SciVerse ScienceDirect](http://www.sciencedirect.com)

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Assessing doses to terrestrial wildlife at a radioactive waste disposal site: Inter-comparison of modelling approaches

M.P. Johansen ^{a,*}, C.L. Barnett ^b, N.A. Beresford ^b, J.E. Brown ^c, M. Černe ^d, B.J. Howard ^b, S. Kamboj ^e, D.-K. Keum ^f, B. Smodiš ^d, J.R. Twining ^a, H. Vandenhove ^g, J. Vives i Batlle ^g, M.D. Wood ^h, C. Yu ^e

^a Australian Nuclear Science and Technology Organisation, Locked Bag 2001, Kirrawee DC, NSW, 2232, Australia

^b Centre for Ecology & Hydrology, Lancaster, UK

^c Norwegian Radiation Protection Authority, Oesteraas, Norway

^d Jožef Stefan Institute, Ljubljana, Slovenia

^e Argonne National Laboratory, IL, USA

^f Korea Atomic Energy Research Institute, Daejeon, Republic of Korea

^g Belgian Nuclear Research Centre, Mol, Belgium

^h University of Salford, Manchester, UK

ARTICLE INFO

Article history:

Received 7 November 2011

Received in revised form 5 April 2012

Accepted 10 April 2012

Available online 10 May 2012

Keywords:

Biota dose modelling

Radionuclides

Transfer

Concentration ratio

Wildlife

ABSTRACT

Radiological doses to terrestrial wildlife were examined in this model inter-comparison study that emphasised factors causing variability in dose estimation. The study participants used varying modelling approaches and information sources to estimate dose rates and tissue concentrations for a range of biota types exposed to soil contamination at a shallow radionuclide waste burial site in Australia.

Results indicated that the dominant factor causing variation in dose rate estimates (up to three orders of magnitude on mean total dose rates) was the soil-to-organism transfer of radionuclides that included variation in transfer parameter values as well as transfer calculation methods. Additional variation was associated with other modelling factors including: how participants conceptualised and modelled the exposure configurations (two orders of magnitude); which progeny to include with the parent radionuclide (typically less than one order of magnitude); and dose calculation parameters, including radiation weighting factors and dose conversion coefficients (typically less than one order of magnitude). Probabilistic approaches to model parameterisation were used to encompass and describe variable model parameters and outcomes. The study confirms the need for continued evaluation of the underlying mechanisms governing soil-to-organism transfer of radionuclides to improve estimation of dose rates to terrestrial wildlife. The exposure pathways and configurations available in most current codes are limited when considering instances where organisms access subsurface contamination through rooting, burrowing, or using different localised waste areas as part of their habitual routines.

Crown Copyright © 2012 Published by Elsevier B.V. All rights reserved.

1. Introduction

Reducing the uncertainty in estimating the radiological dose rates to terrestrial biota has proved to be challenging (Beresford, 2010). Difficulties relate, in part, to the need to make simplifying assumptions when applying dosimetric models to biota of different sizes, shapes and behaviours that are present in various geometric configurations between soil and organism (Taranenko et al., 2004; Vives i Batlle et al., 2010).

However, most variation has been associated with the soil-to-organism transfer of radionuclides (Avila et al., 2004; Beresford, 2010; Higley, 2010). The concept of transfer includes complex biogeochemical and food chain mechanisms (e.g., sorption–desorption, adhesion, ingestion–absorption, metabolism) that vary among different environments, radionuclides, biota and exposure pathways (ingestion, inhalation, direct exposure).

Methods used to estimate transfer, as reviewed by Higley and Bytwerk (2007), rely on three basic approaches: use of concentration ratios, kinetic uptake and loss (compartment) modelling, and allometric-based approaches. Differences in how transfer is modelled can result in substantial variation in modelled dose estimates (Wood et al., 2009). For example, ranges of one–three orders of magnitude were common, and even up to five orders of magnitude, when comparing total dose rate estimates from seven modelling approaches for various organisms in the Chernobyl exclusion zone (Beresford et al., 2010). By comparison,

* Corresponding author. Tel.: +61 2 9717 7206; fax: +61 2 97173686.

E-mail addresses: mathew.johansen@ansto.gov.au (M.P. Johansen), clb@ceh.ac.uk (C.L. Barnett), nab@ceh.ac.uk (N.A. Beresford), justin.brown@nrpa.no (J.E. Brown), marko.cerne@ijs.si (M. Černe), bjho@ceh.ac.uk (B.J. Howard), skamboj@anl.gov (S. Kamboj), dkkeum@kaeri.re.kr (D.-K. Keum), jrt@ansto.gov.au (J.R. Twining), hvanden@sckcen.be (H. Vandenhove), jvbatll@sckcen.be (J. Vives i Batlle), m.d.wood@salford.ac.uk (M.D. Wood), cyu@anl.gov (C. Yu).

when the transfer component of biota dose models was constrained by setting the whole organism tissue concentrations to unity, the variation in dose estimates among a range of model types was limited to about one order of magnitude (Vives i Batlle et al., 2010).

A terrestrial model inter-comparison study for Chernobyl (Beresford et al., 2010), along with an aquatic study for Perch Lake, Canada (Yankovich et al., 2010), were performed under the International Atomic Energy Agency's (IAEA) Environmental Modelling for Radiation Safety Programme (EMRAS), to compare current and developing modelling practices. These studies recommended further assessment of biota dose model variation by considering an extended range of radionuclides, biota types and exposure scenarios. Also recommended, here and elsewhere, was inter-comparison among modellers of varying levels of familiarity with the current model codes (i.e., model developers and informed users) (Beresford et al., 2009; IAEA, in press; Wood et al., 2009).

Concurrent with the above modelling assessments, there has been further development of biota dose modelling codes including use of probabilistic functions to better encompass variability. Some available codes now include capability to enter media (e.g. soil concentration data) and transfer parameters (e.g., concentration ratios) as distributions, instead of single values, for use in Monte Carlo, Latin Hypercube, or other statistically-based methods (Brown et al., 2008; Howard and Larsson, 2008; USDOE, 2004). While this functionality is available, no modelling inter-comparison study has been reported on the probabilistic-based outcomes of various biota dose codes.

Other recent developments include the IAEA's Handbook of Parameter Values for the Prediction of Radionuclide Transfer to Wildlife (Howard et al., in press), and the associated Wildlife Transfer Database (Beresford, 2010; ICRP, 2009). These resources add to previous reference summaries for other biota categories (e.g., IAEA, 2010) and provide opportunity for modellers to more easily access published transfer values. Consistent with the handbook on wildlife transfer, the term *concentration ratio* used in this paper represents the ratio of the equilibrium activity concentration in the whole organism (fresh weight) to that of the reference media, and is denoted as $CR_{wo-soil}$ when considering contaminated soil (dry weight) (Howard et al., in press).

The primary objective of this study was to compare the variation resulting from differing biota dose modelling approaches for a range of radionuclides at a waste burial site. The study followed from previous model inter-comparisons (Beresford et al., 2010; Vives i Batlle et al., 2010; Yankovich et al., 2010) in quantifying the variation in model outcomes associated with the transfer parameters and comparing this with the relative variation associated with progeny, exposure configuration and other dosimetric parameters.

Relatively few studies on transfer of radionuclides to biota have been performed in Australia (Johansen and Twining, 2010). Selection of an Australian site challenged participants by including organisms with little, or no, reference data (e.g., wallaby and echidna) along with more commonly referenced organisms (e.g., grass and earthworm). The shallow-buried radiological waste located at the study site provided a previously largely unconsidered source term for biota dose modelling. Study results will be useful in guiding new users, informing future code development and highlighting future research priorities toward improving biota dose assessment methods.

2. Methods

2.1. Model codes and participants

The biota dose assessment codes used in this inter-comparison (Table 1) included the ERICA Tool, FASTER-lite, K-Biota and RESRAD-BIOTA as described in Beresford et al. (2008, 2010), and Vives i Batlle et al. (2010). These codes share basic features of steady-state estimation of radiological tissue concentrations and doses (internal, external, and total) to biological organisms. As applied in this study, the codes differed primarily in their approach for deriving soil-to-organism transfer of radionuclides, and were grouped into three basic approaches (Higley and Bytwerk, 2007) as follows:

- The *concentration ratio approach* to transfer was employed by participants using the ERICA Tool (Brown et al., 2008; Beresford et al., 2008, <http://www.ERICA-tool.com>) and RESRAD-BIOTA (USDOE, 2004, <http://web.ead.anl.gov>). This approach relies upon reference concentration ratios as simplified, integrating factors to estimate the radionuclide concentrations in the whole-organism as a ratio to the host soil, water, or sediment media. In this study, participants differed in their selection of $CR_{wo-soil}$ values from various sources including model defaults, IAEA documents on transfer parameters and associated databases (Howard et al., in press) and published journal references.
- *Kinetic/compartiment modelling approaches* to transfer were used in the FASTER-lite suite of models (Beresford et al., 2010) which included steady-state and dynamic compartment modelling to estimate the inventory of radionuclides in an organism given food-chain uptake, inhalation, retention and excretion parameters. Within this approach, some $CR_{wo-soil}$ values, as well as allometric estimation methods were used for deriving soil-to-diet transfer as part of an organism's food ingestion pathway.

Table 1
Study participants, biota dose modelling codes, and soil-to-organism transfer approaches.

Participant	Code	Method for soil-to-organism transfer
Centre for Ecology & Hydrology, Lancaster, UK (ERICA-CEH)	ERICA Tool (tier 3)	$CR_{wo-soil}$ values from the Wildlife Transfer Database (IAEA, in preparation; http://www.wildlifetransferdatabase.org and IAEA, 2010 for yam and some grass values)
Belgian Nuclear Research Centre, Mol, Belgium (ERICA-SCK)	ERICA Tool (tier 3)	$CR_{wo-soil}$ values from ERICA Tool defaults (Brown et al., 2008), except grass and yam from IAEA (2010)
Jožef Stefan Institute, Ljubljana, Slovenia M. Černe, B. Smodiš (ERICA-JSI)	ERICA Tool (tier 3)	$CR_{wo-soil}$ values from ERICA Tool defaults (Brown et al., 2008)
Norwegian Radiation Protection Authority Oesteraas, Norway (FASTER-lite-NRPA)	FASTER-lite used with ERICA Tool, (used with Eikos, and ECOLOGO)	Dynamic and Steady-state biokinetic transfer based on ingestion (Nagy, 2001) and inhalation/soil ingestion parameters (Brown et al., 2003). Soil-to-diet $CR_{wo-soil}$ values from ERICA-Tool defaults (Brown et al., 2008).
Korea Atomic Energy Research Institute Daejeon, Republic of Korea (K-Biota-KAERI)	K-Biota	$CR_{wo-soil}$ values from: 1) ERICA Tool defaults for grass, tree, earthworm, insect, bird; 2) IAEA (1994) for yam (potato); 3) allometric equation for goanna, echidna, fox, and wallaby after Higley (2007)
Argonne National Laboratory, IL, USA (RESRAD-BIOTA-ANL)	RESRAD-BIOTA (Level 3)	$CR_{wo-soil}$ values from RESRAD-BIOTA defaults (USDOE, 2004)
University of Salford Manchester, UK (RESRAD-BIOTA-UoS)	RESRAD-BIOTA	When run probabilistically, included use of same values as in ERICA $CR_{wo-soil}$ values from RESRAD-BIOTA defaults used except allometric equation for goanna, raven, echidna, fox, and wallaby after USDOE (2004). Designated-MMU in electronic supplements.

• The *allometric approach* (mass dependent) to transfer was used for reptiles, birds and mammals in K-Biota (Keum et al., 2010, 2011) and RESRAD-BIOTA (USDOE, 2004). The allometric equations used in the two model approaches were similar, with some differences in equation components and parameters which were sourced from USDOE (2004) for RESRAD-BIOTA and various published journal and book references such as Whicker and Shultz (1982). Similar to the kinetic method, the allometric approach utilises $CR_{wo-soil}$ values for deriving soil-to-diet transfer as part of an organism's food ingestion pathway.

In some instances, the same model code was used by multiple participants who differed in approach primarily with respect to their selection of $CR_{wo-soil}$ parameters. For example, three participants used the same version of the ERICA Tool and while they used the same radiation weighting factors, progeny assumptions, etc., they differed in their sources of $CR_{wo-soil}$ values as indicated (Table 1). In these cases, the modelled dose estimates among replicate users of the same code were compared to assess the variation in results attributable to their different transfer parameter choices. Consistent with previous recommendations (e.g. Wood et al., 2009), participants included informed modellers with various levels of experience including the developers/custodians of the codes used.

2.2. Scenario basis

The study was based on the Little Forest Burial Ground site, located in New South Wales, Australia (lat. -34.03800 , long. 150.97876), which provided a range of radionuclides, biota types, and exposure pathways. Site contamination is characterised by trace level radionuclides in surface soils associated with the 1960 to 1968 placement of radioactive wastes in a series of 79 shallow trenches. Waste types include fission and activation products derived from research activities and trace radionuclides include alpha, beta and gamma emitters (^{60}Co , ^{90}Sr , ^{137}Cs , ^{232}Th , ^{234}U , ^{238}U , ^{238}Pu , $^{239/240}Pu$, ^{241}Am). The waste trenches are surrounded by adsorptive and poorly transmissive clay soils that are typically unsaturated. The trenches are covered by ~ 1 m of clay-rich soil layer, which in turn is overlain by topsoil and a well-developed and maintained grass-dominated vegetative cover. Due to its relatively dry and clay-rich conditions, only limited transport of radionuclides away from the trenches has been documented by the ongoing site monitoring system (Hughes et al., 2010; Twining et al., 2011).

The following ten organism types, representative of the site biota, were assessed:

- grass (*Poaceae* spp.)
- acacia tree (*Acacia longifolia*)
- pencil yam (*Vigna lanceolata*)
- earthworm (*Lumbricidae* spp.)
- grasshopper (*Acrididae* spp.)
- goanna (*Varanus varius*)
- Australian raven (*Corvus coronides*)
- echidna (*Tachyglossus aculeatus*.)
- red fox (*Vulpes vulpes*)
- wallaby (*Wallabia bicolor*)

These organisms were selected as being representative of an extended range of sizes, physiological characteristics and behavioural types. Some organisms have readily available reference data with respect to radionuclide transfer, including grass and earthworm (which are ICRP Reference Animals and Plants; ICRP, 2009). Others have little or no data available with respect to radiological transfer (e.g., wallaby, echidna, goanna) and thus provide opportunity to apply and compare methods that have been suggested for use when data are not available such as allometry (Higley, 2010), or use of $CR_{wo-soil}$ values from similar species (Beresford, 2010). The three mammal infraclasses of monotremes, placentals and marsupials are represented by echidna, fox and wallaby respectively. With the exception of pencil yam, all

organisms were present at the site during 2008–2010 observational studies that included use of motion sensor cameras with infrared capability for nocturnally-active species. Pencil yam was included as it is representative of root tubers generally present in the region.

Due to the relatively small area of the waste trenches (1.1 ha), most animal species are typically not continuously present, but rather visit for periods of varying duration as part of their foraging or other habitual routines. Therefore, a set of occupancy factors (% time spent within a given area; values provided in Electronic supplement 1) were provided for each species based on the observational studies. These occupancy factors were provided for four zones described as:

- zone 1 – the waste material buried within the trenches (1–3 m deep),
- zone 2 – the ground surface and soils above and immediately adjacent (within 4 m) of any trench,
- zone 3 – the ground surface soils further than 4 m from any trench but within the site boundary fence,
- zone 4 – all areas outside of the site boundary.

For any modelled organism, the occupancy factors over the four zones totalled to 100% with occupancy assigned according to field observations (e.g., camera surveys, transect surveys). The area most frequented by the representative organisms studied was the ground surface and soils above, and immediately adjacent to, the trenches (zone 2). Only the acacia (roots) were modelled to access the trench wastes (zone 1) directly. The other organisms with subsurface access either did not penetrate to the depth of the trenches, or were not present in the immediate vicinity of the trenches. For each zone, radionuclide soil activity concentrations were provided (mean, standard deviation, minimum, and maximum) (values provided in Electronic supplement 1). Characterisation of the surface soils at the site was not complete at the time of this study, and for the purposes of the study, some soil concentration data were estimated by extrapolating from known data as indicated in the supplementary data section.

A standard set of scenario information was provided to participants as a common basis for conducting the inter-comparison. This set included: a basic site description, physical attributes of the ten representative organisms, suggested occupancy factors for these organisms relative to the four defined zones, and a list of radionuclides of concern and their activity concentrations for each zone. Study participants were asked to provide dose rate estimates (internal, external and total), whole-body concentrations, as well as documented description of their modelling approaches including transfer parameter sources, progeny assumptions and description of dose parameters. The overall approach sought to examine the types and degrees of variation that emerged as participants used the same common basis of site data, yet choose a variety of modelling codes and approaches including varying sources of model parameters. Site-specific concentrations of radionuclides in animal tissues were not available. Therefore the study design did not attempt to compare modelling outcomes with observed site values, but rather to assess the sources and magnitudes of variation produced among modelling approaches.

2.3. Statistical approach for comparing model results

Model outcomes were evaluated using the following process designed to provide an indication of variation with respect to the approach used, and with respect to the organism being assessed. For each organism, dose rate values, tissue concentrations and $CR_{wo-soil}$ values were normalised to the average prediction across the different participant approaches for a particular radionuclide. The set of $CR_{wo-soil}$ values included the effective $CR_{wo-soil}$ values resulting from the allometric and kinetic compartment modelling approaches. Standard deviations were divided by the averages of all the values for a given radionuclide, including all organisms and approaches, to find which radionuclides had more associated variability.

Since the study design included no observed values for the calculated tissue concentrations and dose rates, for every participating approach we defined a vector $X(i) = (x_1(i), x_2(i), \dots, x_p(i))$ where:

- p is the number of radionuclides (9 for this scenario, from ^{60}Co (1) to ^{241}Am (9));
- i is the index representing a particular approach.

The vector elements $x_1 \dots x_p$ represent the activity concentrations (or total dose rates) for a given approach and radionuclide. This vector is a convenient way to present the quantities to be inter-compared as points in a multi-dimensional space, integrating into a single quantitative value the results of a given participant.

We then calculated a square matrix in which each element d_{ij} represents the geometric mean of the relative half-differences between two points $X(i) = (x_1(i), x_2(i), \dots, x_p(i))$ and $X(j) = (x_1(j), x_2(j), \dots, x_p(j))$ in p -dimensional space, i.e. a measure of the distance between the two points relative to the mid-point, as defined in (Eq. (1)):

$$d_{ij} = \sqrt{\frac{1}{N} \sum_{k=1}^p \left(\frac{x_k(i) - x_k(j)}{x_k(i) + x_k(j)} \right)^2} \quad (1)$$

where N (the number of measurements for which an actual pair $\{x_k(i), x_k(j)\}$ is available: 0 to 9, depending on the number of not reported values), is a weighting factor included to avoid statistical bias. Using relative half-distances instead of simply calculating the Euclidean distance ensures that data for all radionuclides were standardised in order to be considered as non-dimensional, comparable quantities.

Each of the d_{ij} elements takes a value between 0 and 1, and is a measure of the dissimilarity of the approaches i and j in respect of the various radionuclides. Higher values (approaching 1) reflecting greater variability and lower values greater similarity. Note that, in the resulting matrix, all the diagonal elements are zeros because if $i=j$ then $x_k(i) = x_k(j)$. Moreover, $d_{ij} = d_{ji}$ because $(x_k(i) - x_k(j))^2 = (x_k(j) - x_k(i))^2$. This type of matrix is therefore symmetrical (Electronic supplement 2).

In this study, the individual matrix values indicated the extent to which the outcome of a particular participant approach deviated from that of the other approaches. It also indicated if the discrepancies come from doses or from soil-to-organism transfer. Since the comparison works on the basis of relative distances, the units/scales of the dose matrix and the $\text{CR}_{\text{wo-soil}}$ matrix are the same and so these quantities could be directly compared.

Using the geometric mean of all the elements above the diagonal, it was possible to compare among organisms to help illustrate which had closer agreement and which had larger variation in terms of the model outcomes of different approaches.

3. Results and discussion

3.1. Variation due to soil-to-organism transfer

The variation in $\text{CR}_{\text{wo-soil}}$ values used by participants typically ranged over two orders of magnitude for most of the radionuclides and organisms, with maximums of more than three orders of magnitude. For

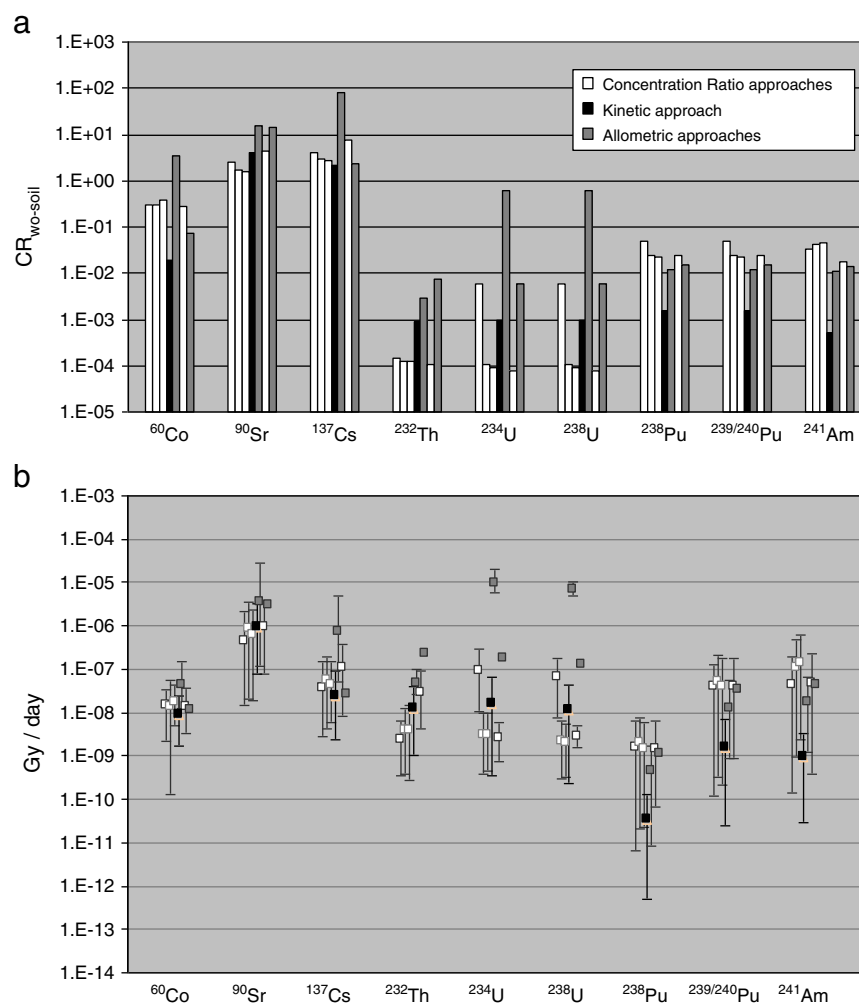


Fig. 1. For wallaby, (a) $\text{CR}_{\text{wo-soil}}$ parameter values, and (b) corresponding modelled total dose rates: mean (\square) with 5th and 95th percentiles indicated. The data order for each set are: ERICA-CEH, ERICA-SCK, ERICA-JSI, FASTer-lite-NRPA, K-Biota-KAERI, RESRAD-BIOTA-ANL and RESRAD-BIOTA-UoS.

Table 2

Overall measure of dissimilarity among modelling results expressed as arithmetic means across all radionuclides of the normalised relative differences between participant approaches. Lower values indicate more agreement among the participant's results, higher values indicate more variation.

	Soil-to-organism transfer ($CR_{wo-soil}$)	Whole-organism tissue conc.	Internal dose rates	External dose rates	Total dose rates
Grass	0.46	0.44	0.65	0.38	0.61
Acacia	0.24	0.41	0.59	0.47	0.52
Pencil yam	0.62	0.60	0.67	0.43	0.64
Earthworm	0.15	0.16	0.49	0.33	0.47
Grasshopper	0.33	0.28	0.54	0.38	0.49
Goanna	0.66	0.63	0.61	0.40	0.57
Raven	0.34	0.57	0.64	0.47	0.61
Echidna	0.57	0.44	0.65	0.38	0.61
Fox	0.58	0.44	0.65	0.38	0.61
Wallaby	0.58	0.44	0.65	0.38	0.61

example, the $CR_{wo-soil}$ values used for wallaby typically varied one to two orders of magnitude for the most radionuclides with more than three orders of magnitude for uranium isotopes (Fig. 1a). Similar variation was observed for most other organisms, with exceptions such as earthworm, which had relatively low variation (Table 2, column 1) likely due to participant's use of the same data sources.

Comparison of the variation in $CR_{wo-soil}$ values (Fig. 1a) with corresponding ranges in total dose rates (Fig. 1b), suggests proportionality between their respective magnitudes. For example, the $CR_{wo-soil}$ values and total dose values for ^{137}Cs both indicate little variation, while for ^{234}U the spread in values is larger in both sets. This apparent proportionality between $CR_{wo-soil}$ values and total dose rates was examined across all radionuclides using linear regression on the measures of dissimilarity values in Table 2, the result of which indicated proportionality between transfer-and-organism concentrations, tissue concentrations-and-internal dose rates and internal-and-total dose rates (Fig. 2a–c).

While these regressions indicate a level of proportionality between $CR_{wo-soil}$ values and tissue concentrations as well as dose rates, they also indicate variation above and below the regression lines which was expected to be due to attributes of specific radionuclide types and was therefore examined further by considering alpha, beta and gamma emitters separately. Using a subsurface tuber (pencil yam) as an example (Fig. 3a), total doses and $CR_{wo-soil}$ values were proportional for alpha emitters (^{232}Th $R^2 = 0.76$ with progeny included, $R^2 = 0.83$ without progeny) and highly proportional for the beta emitter ^{90}Sr ($R^2 = 0.96$). In contrast, the total doses from gamma emitters had very low proportionality to $CR_{wo-soil}$ values such as for ^{60}Co ($R^2 = 0.12$). For (high energy) gamma emitters such as ^{60}Co , these results for pencil yam are consistent with a conceptual model of an organism located in soil which is contaminated with gamma emitting radionuclides in concentrations sufficient for the external dose to dominate over the internal dose associated with transfer to the organism. Results for other organisms living in, or on, the soil were similar with ^{60}Co , followed by ^{137}Cs (the two predominantly gamma emitting radionuclides) having the lowest proportionality between transfer and total dose rates.

Continuing with the pencil yam example, the regression of dose rate against $CR_{wo-soil}$ provided a basis for approximating the range of variation in total dose rates attributed to soil-to-organism transfer. For ^{232}Th , approximately two orders of magnitude of variation in total dose was indicated as taken from the vertical intercepts with the regression line of the lowest and highest ^{232}Th data (Fig. 3a). Residuals above and below regression lines indicate additional variation introduced by other factors, such as inclusion/exclusion of some progeny, and are each considered in sections below. For ^{90}Sr , more than one order of magnitude variation was indicated, and for ^{60}Co less than one order of magnitude was indicated. Using this approach for all organisms, the maximum observed variation was more than three orders

of magnitude for uranium isotopes in wallaby (Fig. 3b). Other organisms indicated maximum variation due to transfer of approximately one to two orders of magnitude.

When comparing results for wallaby from the three ERICA-Tool users, total dose estimates grouped together for some radionuclides such as for ^{60}Co , ^{90}Sr and ^{137}Cs , (first three data in Fig. 1: ERICA-CEH, ERICA-SCK, ERICA-JSI), and diverged for others such as the approximately two orders of magnitude difference in ^{234}U and ^{238}U isotopes. This difference was mainly due to variation of $CR_{wo-soil}$ values sourced by the different ERICA-Tool users (Fig. 1a) as the other model parameters used were very similar. Overall, when considering variation from transfer in just the set of ERICA-Tool results, the largest variation in total dose rates was the approximately two orders of magnitude for ^{238}Pu and $^{239/240}Pu$ in pencil yam. Variation of one to two orders of

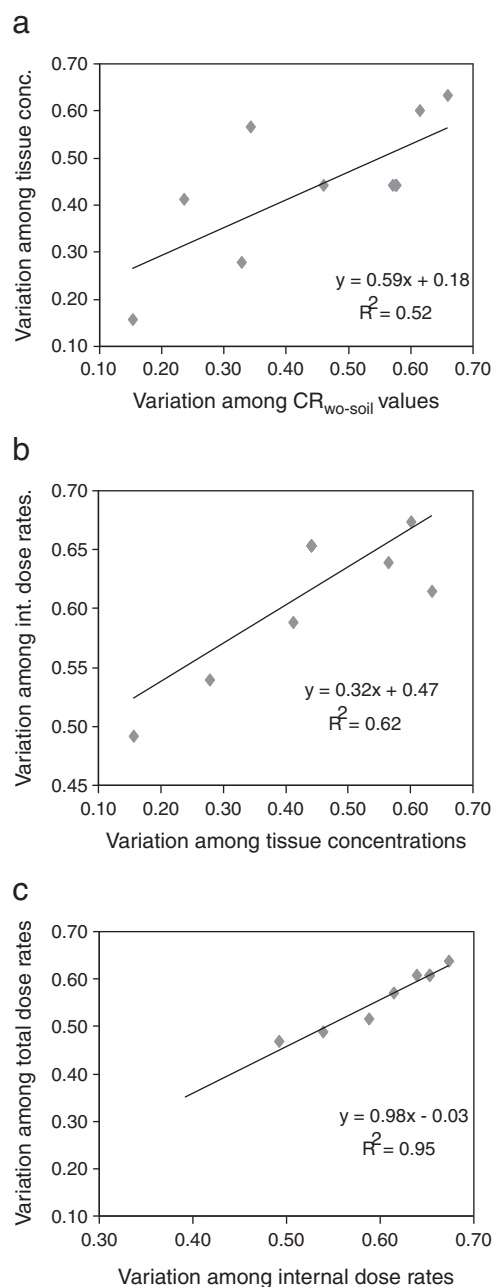


Fig. 2. Linear regressions between matrix variation values (unitless) for (a) $CR_{wo-soil}$ values v. whole-organism tissue concentrations, (b) whole organism tissue concentrations v. internal dose rates, and (c) total v. internal dose rates. Each point is for a study organism and represents a measure of the variability (0–low, 1–high) combined across all radionuclides as indicated in Table 2. Note some data are superimposed.

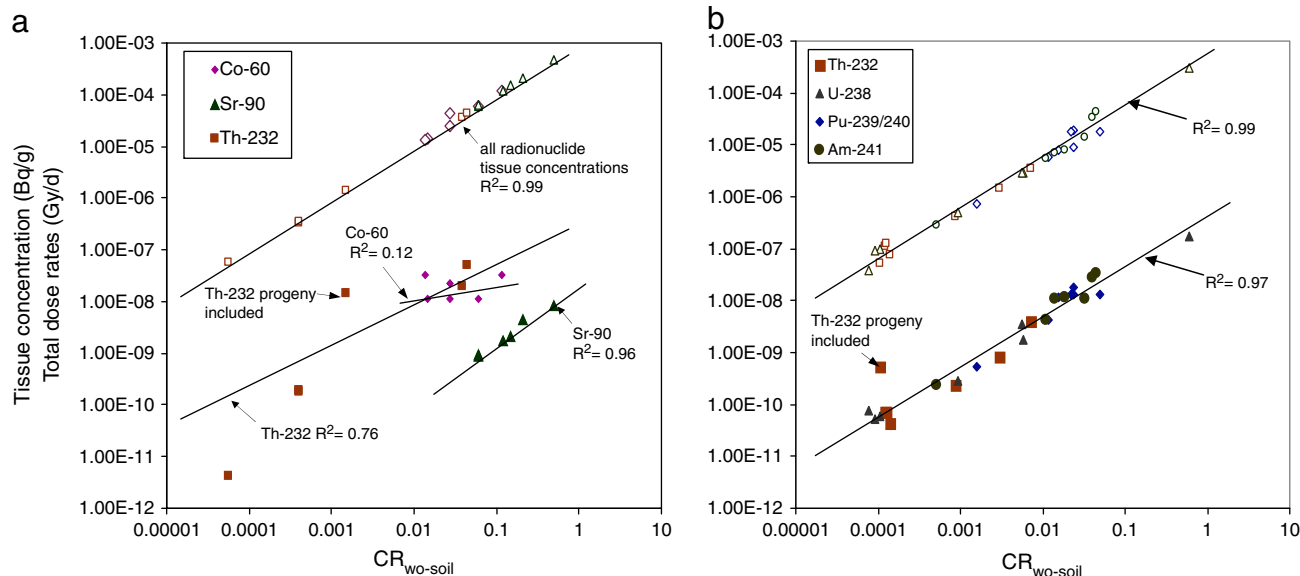


Fig. 3. Predicted tissue concentrations (open symbols) and total dose rates (closed symbols) v. $CR_{wo-soil}$ for (a) gamma, beta, and alpha emitters in pencil yam, and (b) alpha emitters in wallaby. Values have been normalised relative to soil concentrations of 1 Bq kg^{-1} .

magnitude was common in many organisms, and consistently corresponded to differences among the $CR_{wo-soil}$ reference sources (Electronic supplement 3).

Two participants used similar allometric approaches to derive $CR_{wo-soil}$ values but utilised sometimes differing assumptions within the specific allometric equation parameters. In many instances, the resulting dose rates were similar (e.g., Fig. 1: ^{238}Pu , $^{239/240}\text{Pu}$ and ^{241}Am for the allometric approaches used by K-Biota-KAERI and RESRAD-Biota-UoS). In other instances, however, they diverged up to two orders of magnitude (e.g. ^{234}U , ^{238}U). The dose rate results of the kinetic-compartment modelling approach (FASTER-lite-NRPA), which also made use of allometric relationships in its food ingestion pathways, were typically near the average of the group results. The allometric equations required multiple parameters regarding radionuclide biological half-life, food and soil ingestion, metabolism, assimilation and inhalation. In this study, most allometric parameters used by the participants were similar, with the largest differences in transfer from soil to the food items consumed by the vertebrate organisms, such as the concentration ratios for soil-to-grass which was then consumed by wallaby. The allometric approach provides for parameterising specific exposure pathways. For example, inhalation factors were important when considering transfer to echidna which feeds on ants and termites in often dusty conditions with one participant's hypothetical model results suggesting up to 38% of the whole-organism activity concentrations resulted from dust inhalation.

Overall, the assessment of soil-to-organism transfer indicated that varying choices of transfer parameters and modelling approaches resulted in up to three orders of magnitude variability in total dose rates, with one to two orders of magnitude frequently indicated. Proportionality between transfer and dose was suggested for most radionuclides, such as the $R^2 = 0.79$ for alpha emitters relative to wallaby, but not all, such as the $R^2 = 0.12$ for the high energy gamma emitter ^{60}Co relative to pencil yam (Fig. 3). This is consistent with a conceptual model of total dose being derived largely from internal exposure after transfer of radionuclides from soil, with potentially important exceptions where the external dose from some gamma emitters dominates over internal dose rates.

3.2. Variation due to exposure configuration

Spatial configurations were specified by the modeller (e.g., organism on soil surface, organism 10 cm below soil surface, etc.). In most

instances, all participants used similar spatial factors to define which zones of contamination were accessed by a particular organism. The primary exception was acacia, which had a portion of its roots directly accessing the subsurface waste (comparatively high contaminant levels), and its other roots accessing less-contaminated soil (Electronic supplement 1). This exposure configuration represents a realistic condition at shallow waste sites where organisms, both stationary and mobile, can access zones with differing contamination levels through rooting, burrowing, or regular movement across localised waste areas as part of their habitual routine. While realistic, such exposure configurations are not generally available in models which typically assume steady-state access to homogeneously contaminated soil.

Given the non-standard scenario for acacia tree, participants used varying approaches to derive whole-organism total dose including a conservative approach of assuming the tree was rooted completely in the highest concentration soil. Other approaches included dividing the maximum modelled dose from the waste source by the fraction of roots exposed, as well as summation of separate contributions from each of the different soil sources weighted by their respective mass fractions. As a result, even though participants generally used very similar $CR_{wo-soil}$ values (relatively low dissimilarity value for acacia of 0.24 in Table 2), their varied approaches to conceptualising and modelling the exposure yielded variation in total dose rates for acacia that was comparable to other species (dissimilarity value of 0.52 in Table 2). One participant examined dose rates to the roots alone modelled as a sub-surface organism, compared with dose rates to the trunk modelled in the more conventional approach of a standard plant experiencing uptake from the soil. These two approaches resulted in total dose differences of typically two orders of magnitude across study radionuclides.

Modelled dose rates of other organisms (e.g., goanna and raven) also included variation associated with differing exposure configuration assumptions related to the proportion of time spent on the ground exposed to soil contaminants, versus time spent in trees away from contaminated soil. These results demonstrate how realistic exposure configurations may not easily fit into some current models, and that differing interpretations of exposure by model users can lead to order-of-magnitude variation in dose estimates.

Overall, the variation in the total dose rate attributed to differences in how participants conceptualised and modelled exposure configurations was typically less than one order of magnitude for most organism types, but up to two orders of magnitude for acacia.

Table 3

Relative standard deviation of normalised model results treating all of the study species data for each radionuclide as a set. Higher values indicate more variation among participant results.

	Soil-to-organism transfer (CR _{wo-soil})	Whole-organism tissue conc.	Internal dose rates	External dose rates	Total dose rates
⁶⁰ Co	1.15	1.03	1.09	0.41	0.68
⁹⁰ Sr	1.40	1.35	1.36	1.59	1.26
¹³⁷ Cs	1.23	1.16	1.19	0.42	1.05
²³² Th	1.39	1.30	1.55	0.50	1.54
²³⁴ U	1.66	1.61	1.64	0.48	1.64
²³⁸ U	1.55	1.61	1.65	1.53	1.65
²³⁸ Pu	0.65	0.77	0.84	0.95	0.84
^{239/240} Pu	0.69	0.76	0.83	0.54	0.83
²⁴¹ Am	0.97	1.12	1.11	0.53	1.11

3.3. Variation due to progeny assumptions

Results indicated that up to one order of magnitude variation in total dose rates was associated with differing assumptions of which progeny to include along with the parent radionuclides. Of the radionuclides tested, ²³²Th doses had relatively high variation ($\sigma = 1.54$, Table 3) associated with varying progeny assumptions among participant models. The key progeny ²²⁸Ra, ²²⁸Ac and ²²⁸Th have half-lives that are much less than that of the parent ²³²Th (1.4E10 yrs), and much less than the ~40 years the contamination has been in place at the waste site as well, indicating that the ²³²Th progeny can be considered as being in equilibrium and therefore contribute to overall dose. Whether or not participants included or excluded the ²³²Th progeny appeared to depend mainly upon model code defaults, with the RESRAD-BIOTA code users including progeny (default when progeny half-lives < 100-yr), and others excluding them (e.g., progeny with half-lives > 10-day are excluded in ERICA). When the progeny were excluded, the resulting ²³²Th dose estimates were approximately one order of magnitude lower.

These results indicate the potential for order-of-magnitude variation associated with progeny assumptions for a typical waste site, which may vary depending on the specific radionuclides and time-frames involved. Some currently available codes include progeny, but have differing default assumptions, and code users may benefit from improved prompts within the user interface, or similar means, to highlight the default assumptions being used and to provide guidance during development of each modelling case.

3.4. Variation from dose calculation parameters

The dose calculation parameters considered included radiation weighting factors and dose conversion coefficients (DCCs). Most participants used radiation weighting factor values of 10, 3 and 1 for alpha, low-energy beta and high-energy beta plus gamma emissions respectively (defaults in the ERICA Tool), with two participants using 20, 1, 1 (defaults in RESRAD-BIOTA). These weighting factors apply linearly to internal dose rates and, therefore, when considering the above two approaches, the respective differences on total dose variation are expected to be: a factor of two for alpha, a factor of three for low energy-beta, and no difference for gamma.

The present study did not separate the relative contributions to dose variation attributed to DCCs versus that from radiation weighting factors. The variation in total dose that remained as residual after excluding effects of transfer, configuration and progeny was attributed to dose calculation parameters and was less than one order of magnitude across all organisms.

This degree of variation observed in the present study is consistent with recent studies of currently available biota dose modelling codes (Vives i Batlle et al., 2007, 2010), indicating a variation of $\pm 30\%$ for internal dose rates and within one order of magnitude for external dose rates, over a range of DCCs for 74 radionuclides.

While additional factors, such as soil density differences, skin/fur shielding and organism geometry assumptions have the potential to alter dose estimates, they were not evident as causes of variation in this evaluation.

3.5. Considerations in understanding and managing variation in biota dose models

In order to encompass the variation in data and parameters, most participants made use of model code capability to enter values as probabilistic distributions. For example, some codes accepted distribution input for soil concentrations and CR_{wo-soil} values. In these cases, participants reported their resulting dose estimates as distributions. These were compared using relative distributions (5th to 95th percentiles) for each model outcome. The resulting ranges (5th to 95th) of total doses for alpha emitters were typically two to three orders of magnitude with some reaching three to four orders of magnitude. The beta emitter (⁹⁰Sr) had relatively large ranges, approximately three orders of magnitude, but included spreads of up to five orders of magnitude. Distribution spreads were smaller for gamma emitters, typically one to two orders of magnitude on average, with some instances of up to three orders of magnitude. The spread in total dose distributions partly reflected the variability in soil data used in the scenario.

The cumulative probability distributions for total dose rates were summed for all radionuclides and were compared with Derived Consideration Reference Levels (DCRLs) (ICRP, 2008), which were used here as screening dose rates to indicate which organisms and exposure conditions would need more detailed assessment. The ICRP define DCRLs as an order of magnitude dose rate band in which there is likely to be some deleterious effect of ionising radiation. In this study, the total dose rate estimates (mean and 95th percentiles) for all organisms were well below their closest reference DCRLs with the exception of acacia tree, which had means below, but more than half of the 95th percentile estimates were within the DCRL range for reference pine tree (Fig. 4). The acacia tree was the only organism that had direct access to the buried waste. This suggests that biota that habit the Little Forest site in its current condition (based on 2007–2010 data) will not receive doses of concern unless they directly access the buried waste. In this instance, the probabilistic capabilities of the model codes helped interpret site uncertainties and provided guidance on a protective approach to site assessment.

During this study, data quality assessment and control was performed primarily by comparing draft model outcomes, identifying and questioning outliers, then providing participants with the opportunity

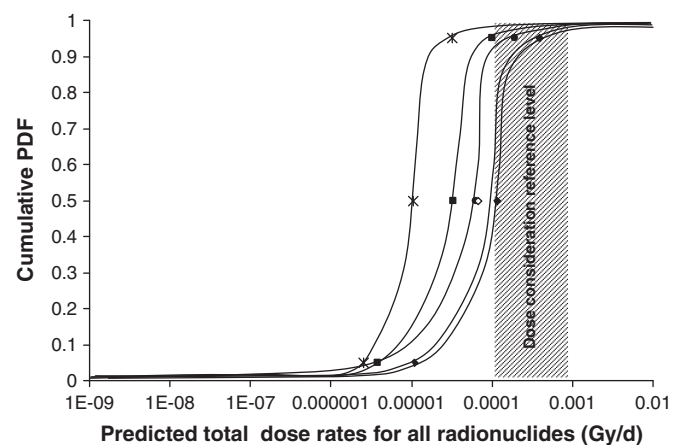


Fig. 4. Predicted cumulative probabilistic distributions for total dose rates from all radionuclides to acacia from the various participant modelling approaches with data indicated for 5th, mean, and 95th percentiles. Lowest to highest are (x) K-Biota-KAERI, (■) ERICA-SCK, (-) RESRAD-BIOTA-UoS, (◇) RESRAD-BIOTA-ANL, (no symbol) ERICA-JSI, and (◆) ERICA-CEH.

to correct any inadvertent errors where appropriate. This assessment was intended to prevent the inadvertent errors from confounding the results; it also provided instructive insights into the types of potential issues that arise during biota dose modelling. Of these issues, the most frequently observed were transcription errors that occurred when large amounts of data were generated over sequential model runs, were then transcribed from model output file formats (e.g. HTML documents) to other formats (e.g. spreadsheets) used for comparing and further analysis of results. Quality control issues appeared to be more common for, but not exclusive to, the less experienced participants, although this was not statistically tested. Participant feedback indicated the probabilistic outputs of some models are not yet presented in a user friendly manner.

Other 'user' issues encountered were: use of $CR_{wo-soil}$ values that were derived from organism dry weight calculations, instead of from organism fresh (wet) weights as required by model codes; use of a cylindrical volume calculation for an organism instead of the ellipsoid volume required by model codes; inadvertent selection of a model exposure geometry option that was not well-suited for the organism and use of an unintended set of soil concentrations (given the scenario's use of multiple contaminant zones); and poor description of some model functions within accompanying help files. Awareness of these issues encountered during this model inter-comparison study will be of use generally in performing model quality assessment and control processes and in training model users.

4. Conclusions

Assessment of the factors causing variation in modelled total dose rates to terrestrial biota that are associated with soil contamination at a low-level waste site, indicated the following (in decreasing order of influence):

1. The dominant factor was soil-to-organism transfer which typically contributed one to two orders of magnitude, and occasionally more than three orders of magnitude, to total dose rate variation. Strong proportionality between transfer values and total dose rates was observed for the beta and alpha emitting radionuclides. However, little proportionality between transfer and dose rates were observed for some gamma emitters such as ^{60}Co where the external dose rates from the surrounding soil dominated over the internal dose rates within the organism.
2. The varied exposure configuration assumptions used by participants resulted in up to two orders of magnitude variation in total dose rates. The acacia tree scenario, with a portion of its roots directly accessing the buried waste, provided most variation in how modellers conceptualised and parameterised exposure configuration and suggested the geometries and exposure scenarios available for plants within available models (and the ability to interpret the subsequent results) can be improved.
3. The differences in progeny assumptions among models contributed up to one order of magnitude variation in the present scenario, primarily from differing treatment of progeny ingrowth for ^{232}Th .
4. Dose calculation parameters, including radiation weighting factors and DCCs, had maximum variation in total dose rates of up to, but typically less than, one order of magnitude with variation being the greatest for alpha emitting radionuclides (largely because of variation in radiation weighting factors used).

This study indicated that the largest contribution to variability in biota dose estimation was parameterisation of the transfer component, as embodied in empirical concentration ratios which are aggregated parameters that typically do not take into account individual, species, and site-specific differences. These results highlight the need for continued evaluation of the underlying mechanisms governing soil-to-organism transfer and variation associated with specific types of ecosystems. Additional empirical research is needed to improve transfer

data for less well-studied organisms, and for improving transfer rate approaches when data are lacking for species of interest. The exposure pathways and configurations available in current codes are limited when considering instances where organisms, both stationary and mobile, can access different contamination zones through rooting, burrowing, or periodic use as part of their habitual routines. Probabilistic capabilities of current model codes provide for describing the variability in results that derive from the uncertainties of the transfer, dose calculation inputs and user inputs and allow for use of confidence intervals when assessing dose estimates to wildlife.

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.scitotenv.2012.04.031>.

Acknowledgements

The authors would like to thank all members and scientific secretary, Sergey Fesenko, of the IAEA EMRAS II programme Biota Modelling Group (Working Group 4, <http://www-ns.iaea.org/projects/emras/emras2/working-groups/working-group-four.asp>) for their comments on this work during the group's workshops. This study, and other investigation work at the Little Forest Burial Ground was conducted under the management and support of Timothy Payne and Henk Heijnis, Nuclear Methods in Earth Sciences Project, Australian Nuclear Sciences and Technology Organisation, Sydney Australia.

References

- Avila R, Beresford NA, Agüero A, Broed R, Brown J, Iospje M, et al. Study of the uncertainty in estimation of the exposure of non-human biota to ionizing radiation. *J Radiol Prot* 2004;24:A105–22.
- Beresford NA. The transfer of radionuclides to wildlife. *Radiat Environ Biophys* 2010;50:504–8.
- Beresford NA, Balonov M, Beaugelin-Seiller K, Brown J, Copplestone D, Hingston JL, et al. An international comparison of models and approaches for the estimation of the radiological exposure of non-human biota. *Appl Radiat Isot* 2008;66:1745–9.
- Beresford NA, Barnett CL, Beaugelin-Seiller K, Brown JE, Cheng J-J, Copplestone D, et al. Findings and recommendations from an international comparison of models and approaches for the estimation of radiological exposure to non-human biota. *Radioprotection* 2009;44(5):565–70.
- Beresford NA, Barnett CL, Brown JE, Cheng J-J, Copplestone D, Gashchak S, et al. Predicting the radiation exposure of terrestrial wildlife in the Chernobyl exclusion zone: an international comparison of approaches. *J Radiol Prot* 2010;30:341–73.
- Brown JE, Strand P, Hosseini A, Børretzen P, editors. Handbook for assessment of the exposure of biota to ionising radiation from radionuclides in the environment. Deliverable Report for the EC Project FASSET (Contract No. FIGE-CT-2000-00102) Østerås, Norway: Norwegian Radiation Protection Authority; 2003. p. 101.
- Brown JE, Alfonso B, Avila R, Beresford NA, Copplestone D, Pröhl G, et al. The ERICA Tool. *J Environ Radioact* 2008;99:1371–83.
- Higley KA. Estimating transfer parameters in the absence of data. *Radiat Environ Biophys* 2010;49:645–56.
- Higley KA, Bytwerk DP. Generic approaches to transfer. *J Environ Radioact* 2007;98:4–23.
- Howard BJ, Larsson CM. The ERICA Integrated Approach and its contribution to protection of the environment from ionising radiation. *J Environ Radioact* 2008;99:1361–3.
- Howard BJ, Beresford NA, Copplestone D, Telleria D, Proehl G, Fesenko S, Jeffery R, Yanovich T, Brown J, Higley K, Johansen MP, Mulye H, Vandenhove H, Gashchak S, Wood MD, Takata H, Andersson P, Dale P, Ryan J, Bollhöfer A, Doering C, Barnett CL, and Wells C. (in press) The IAEA Handbook on Radionuclide Transfer to Wildlife. *J Environ Radioact*. <http://dx.doi.org/10.1016/j.jenvrad.2012.01.027>.
- Hughes CE, Cendon DI, Harrison JJ, Hankin S, Johansen MP, Payne TE, et al. Movement of a tritium plume in shallow groundwater at a legacy low-level radioactive waste disposal site in eastern Australia. *J Environ Radioact* 2010;102:943–52.
- IAEA (International Atomic Energy Agency). Handbook of parameter values for the prediction of radionuclide transfer in temperate environments. Technical Report No. 364. Vienna: IAEA; 1994.
- IAEA (International Atomic Energy Agency). Handbook of parameter values for the prediction of radionuclide transfer in terrestrial and freshwater environments. IAEA-TRS Vienna: IAEA; 2010.
- IAEA (International Atomic Energy Agency). Modelling radiation exposure and radionuclide transfer for nonhuman species, Report of the Biota Working Group of the Environmental Modelling for Radiation Safety (EMRAS) (<http://www-ns.iaea.org/downloads/rw/projects/emras/final-reports/biota-final.pdf>) IAEA Vienna; in press.
- IAEA (International Atomic Energy Agency). Handbook of parameter values for the prediction of radionuclide transfer in wildlife. IAEA, Vienna; in preparation.
- ICRP (International Commission on Radiological Protection). Environmental protection: the concept and use of reference animals and plants. *Annals of the ICRP, Publication 108*. Ann. ICRP Amsterdam: Elsevier; 2008.

- ICRP. Environmental protection: transfer parameters for reference animals and plants. ICRP Publication 114. Ann. ICRP Amsterdam: Elsevier; 2009.
- Johansen MP, Twining JR. Radionuclide concentration ratios in Australian terrestrial wild-life and livestock: data compilation and analysis. *Radiat Environ Biophys* 2010;49: 603–11.
- Keum DK, Jun I, Lim K-M, Choi Y-H. Absorbed internal dose conversion coefficients for domestic reference animals and plant. *Nucl Eng Tech* 2010;42:89–96.
- Keum DK, Jun I, Lim K-M, Choi Y-H. Approach to non-human species radiation dose assessment in the republic of Korea. *Radiat Prot Dosimetry* 2011;146:299–302.
- Nagy KA. Food requirements of wild animals: predictive equations for free-living mammals, reptiles and birds. *Nutr Abs Rev* 2001;71:21–31.
- Taranenko V, Pröhl G, Gómez-Ros JM. Absorbed dose rate conversion coefficients for reference terrestrial biota for external photon and internal exposures. *J Radiol Prot* 2004;24:A35–62.
- Twining JR, Hughes CE, Harrison JJ, Hankin S, Crawford J, Johansen MP, et al. Biotic, temporal and spatial variability of tritium concentrations in transpirate samples collected in the vicinity of a near-surface low-level nuclear waste disposal site and nearby research reactor. *J Environ Radioact* 2011;102:551–8.
- USDOE (United States Department of Energy). User's guide: RESRAD-BIOTA: a tool for implementing a graded approach to biota dose evaluation. ISCORs Technical Report 2004–02:DOE/EH-0676; 2004.
- Vives i Batlle J, Balonov M, Beaugelin-Seiller K, Beresford NA, Brown J, Cheng J-J, et al. Inter-comparison of unweighted absorbed dose rates for non-human biota. *Radiat Environ Biophys* 2007;46:349–73.
- Vives i Batlle J, Beaugelin-Seiller K, Beresford NA, Copplestone D, Horyna J, Hosseini A, et al. The estimation of absorbed dose rates for non-human biota: an extended inter-comparison. *Radiat Environ Biophys* 2010;50:231–51.
- Whicker FW, Shultz V. Radioecology: nuclear energy and the environment. Boca Raton, Florida, USA: CRC Press; 1982.
- Wood MD, Beresford NA, Barnett CL, Copplestone D, Leah RT. Assessing radiation impact at a protected coastal sand dune site: an intercomparison of models for estimating the radiological exposure of non-human biota. *J Environ Radioact* 2009;100:1034–52.
- Yankovich TL, Vives i Batlle J, Vives-Lynch S, Beresford NA, Barnett CL, Beaugelin-Seiller K, et al. An international model validation exercise on radionuclide transfer and doses to freshwater biota. *J Radiol Prot* 2010;30:299–340.