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Evaluation of extrapolation approaches to provide radioecological parameters

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Executive Summary

The assessment of the exposure of humans and wildlife to ionising radiation for planned, existing and emergency exposure scenarios requires us to make predictions of the transfer of a wide range of radionuclides to a diversity of species and food products. We are unlikely to ever have sufficiently robust measurement data to populate all of the potential parameters required. Therefore, we need robust extrapolation approaches which allow us to make best use of our available knowledge. In this study we have reviewed, developed, tested and validated various extrapolation approaches.

The extrapolation approaches used in the original release of the ERICA Tool are reviewed in the light of the increased data now available. It was found that the extrapolation methodologies were not guaranteed to over-predict 95th percentile values. For instance, for the terrestrial ecosystem the extrapolation methods provide under-predictions as often as they produce over-predictions. In a few cases, the underestimation of $CR_{\text{wo-media}}$ values is substantial, by orders of magnitude, which is clearly unacceptable for a screening assessment. Recommendations from this work were used in the revision of the ERICA Tool.

The transfer coefficient, which is often used to predict radionuclide activity concentrations in farm animal products, is in part determined by dry matter intake. This may give some misleading interpretations for different animal species. The concentration ratio is a more robust parameter which is relatively constant across different species. Concentrations ratios derived for farm animals should also be applicable to wild species; we demonstrate that this is the case for radiocaesium.

Animal mass affects many biological process and the dependence of a biological variable (Y) on body mass (M) is typically characterised by an allometric scaling law (i.e. $Y=aM^b$, where a and b are constants). Radionuclide biological half-life for mammals and birds has been shown to follow allometric scaling for a number of radionuclides with b generally approximating to 0.25. Here we demonstrate that an exponent of 0.25 for biological half-life has a biological basis and propose a method to expand the applicability of the allometric approach to radionuclides for which we have insufficient data to define allometric parameters by model fitting. The approach is demonstrated to work well for a number of radionuclides and species. We also demonstrate that the approach can be adapted to reptiles, though the biological half-life for reptiles shows little dependence on mass. Reasons why biological half-life for some radionuclides (e.g. Pu and Am) does not scale to 0.25 need to be elucidated.

The commonly used (in wildlife assessment) whole-organism to media activity concentration ratio is highly variable and dependent upon site characteristics. For caesium and freshwater fish we demonstrate an alternative approach which is independent of site.

Previously extrapolation approaches have not always made best use of all of our available knowledge. We demonstrate the application of Bayes Theorem to the derivation of probability distribution functions for whole-organism to media activity concentration ratios. The subsequent application of Bayesian approach in the derivation of a revised transfer database for the ERICA Tool is discussed.

Ecological stoichiometry shows potential as an extrapolation method in radioecology, either from one element to another or from one species to another. As the application of approaches such as ICP-MS makes it relatively easy to produce multi-element datasets more, data will become available to test stoichiometric assumptions and theories.

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1. Introduction

The assessment of the exposure of humans and wildlife to ionising radiation for planned, existing and emergency exposure scenarios requires us to make predictions of the transfer of a wide range of radionuclides to a diversity of species and food products (e.g. meat, milk, eggs, fruits etc.).

Documents such as IAEA (2010) and ICRP (2009), which compile human foodstuff and wildlife transfer parameter values respectively, demonstrate that for many of the radionuclide-foodstuff/species combinations that require assessment, we have no empirical data. When empirical data are lacking predictions are often made using transfer parameter values derived using extrapolation approaches (e.g. see Beresford et al. 2004, 2008a; Copplestone et al. 2001; Higley et al. 2003; IAEA 2004; Nordén et al. 2010) though the clarity on the use of extrapolation approaches varies between publications. For instance, where data are lacking for wildlife in the ERICA Tool (Beresford et al. 2008a; Brown et al. 2008), a set of rules was followed to derive default concentration ratio ($CR_{wo\text{-}media}$) values (see Chapter 2 for more details). The same or similar approaches are used in other models or to derive tabulated summaries of recommended values (e.g. Copplestone et al. 2003; IAEA 2004; ICRP 2009; USDOE 2002).

Many, of the extrapolation approaches used do not appear to have not been validated or scientifically assessed. Robust extrapolation techniques are required: (i) to enable initial screening tier assessments for which site-specific data are not available (Brown et al. 2013;2014); (ii) for protected species for which it may be impossible to acquire sufficient data (e.g. Copplestone et al. 2003); and (iii) for the International Commission on Radiological Protection's Reference Animals and Plants (RAPs) which are defined specifically at the taxonomic family level but for which there are relatively few specific data (ICRP, 2009; Copplestone et al. 2013).

In this report, we evaluate a number of the commonly used or suggested extrapolation approaches.

2. Evaluating the extrapolation approaches used in the ERICA Tool

2.1 Introduction

Most commonly applied methodologies for assessing the impact of ionising radiation on wildlife require the derivation of activity concentrations in plants and animals from a starting point of known, or model-derived, activity concentrations in environmental media including water, sediments and soil. The method used in the ERICA Tool (Brown et al. 2008) for making this derivation is the whole-body concentration ratio ($\mathrm{CR}_{\text{wo-media}}$), which, for terrestrial biota, is defined as (Equation 2.1):

$$CR_{wo\text{-}soil} = \frac{A_{b,r}^{biota}}{A_{r}^{soil}} \qquad (2.1)$$

Where:

$\mathrm{A}_{\mathrm{b,r}}^{\mathrm{biota}}$ = Activity concentration of radionuclide ‘r’ in the whole organism of biota ‘b’ (Bq kg^{-1} fresh weight (fw)); $\mathrm{A}_{\mathrm{r}}^{\mathrm{soil}}$ = Activity concentration of radionuclide ‘r’ in soil (Bq kg^{-1} dry weight (dw)). For aquatic organisms activity concentrations in soil are replaced by those in water.

In the case of the ERICA Tool, the collation of data was simplified by acknowledging the impracticability of providing transfer data for every organism type within the earth’s many and varied ecosystems and thus opting to structure data around a set of 38 generic organism groups and three generic ecosystems (freshwater, marine and terrestrial). Even with these simplifications, as the ERICA Tool incorporated radionuclides for 31 elements, a matrix consisting of 1178 radionuclide-organism $\mathrm{CR}_{\text{wo-media}}$ value combinations was required. It was necessary to fill all data gaps because values were required for the initial screening tier in the Tool. Available data for selected radionuclides and organisms were collated through the review of published literature, details of which are given in Beresford et al. (2008a) and Hosseini et al. (2008) for terrestrial and aquatic environments respectively. The collated data were largely direct measurements of organisms and environmental media sampled under field conditions. At the time of publication of the ERICA approach, in 2008, data were available to derive $\mathrm{CR}_{\text{wo-media}}$ values for less than 40% of the required radionuclide-organism combinations. The remaining 60% were derived using a variety of extrapolation approaches:

(1) Use an available $\mathrm{CR}_{\text{wo-media}}$ value for an organism of similar taxonomy within that ecosystem for the radionuclide under assessment (preferred option).

(2) Use an available $\mathrm{CR}_{\text{wo-media}}$ value for a similar reference organism (preferred option).

(3) Use $\mathrm{CR}_{\text{wo-media}}$ values recommended in previous reviews, or derive them from previously published reviews (preferred option).

(4) Use specific activity models for ^{3}H and ^{14}C (preferred option).

(5) Use an available $CR_{wo\text{-}media}$ value for the given reference organism for an element of similar biogeochemistry (neutral option).

(6) Use an available $CR_{wo\text{-}media}$ value for biogeochemically similar elements for organisms of similar taxonomy (neutral option).

(7) Use an available $CR_{wo\text{-}media}$ value for biogeochemically similar elements available for a similar reference organism (neutral option).

(8) Use allometric relationships, or other modelling approaches to derive appropriate $CR_{wo\text{-}media}$ values (neutral option).

(9) Assume the highest available $CR_{wo\text{-}media}$ (least preferred option).

(10) Reference organism in a different ecosystem, (aquatic only - least preferred option)

(11) Combination of approaches.

This list includes ‘preferred’, ‘neutral’ and ‘least preferred’ options. Within each of these 3 specific categories, there is no order of preference. The approach selected depended upon the availability of data/knowledge; if more than one approach could be used based upon a similar degree of knowledge then the value selected was the most conservative (i.e. highest). Depending upon the availability of surrogate data, in a few instances, neutral options were used over preferred.

The ERICA approach is based around a tiered system where the assessor initially applies a screening tier requiring little information and can exit the assessment with a stated high degree of confidence that impacts are negligible if screening criteria are not exceeded. If this is not the case the assessment needs to move to a higher tier assessment where more detailed information is required and more elaborate modelling approaches, such a probabilistic calculations to account for uncertainty, can be used. Therefore, the parameters applied at screening tiers need to provide some assurance that predictions of dose-rate and thereafter the risk quotients based upon these exposure estimates are conservative, i.e. tend to over-predict the actual dose-rate. To this end, the 95th percentile of given parameters and outputs have been used in the derivation of exposure estimates. A similar degree of conservatism is included in the initial screening levels of other models (e.g. USDOE 2002).

The aim of the work described in this chapter (adapted from Brown et al. (2013)) was to test the efficacy of the approaches used to derive extrapolated values in the default ERICA Tool parameter databases (Beresford et al. 2008a; Hosseini et al. 2008).

The fulfillment of this aim was facilitated by the development and population of the ‘Wildlife Transfer Database’ (Copplestone et al. 2013)[1]. This was initially populated using the ERICA Tool databases, following additional quality control. Subsequently, many new (or formerly

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[1] See http://www.wildlifetransferdatabase.org/

unused) data were included in the database, a portion of these covered radionuclide-organism combinations for which no data were available when the ERICA Tool was originally parameterised. Note, given that the primary aim of the default $CR_{wo\text{-}media}$ values in the ERICA Tool is to derive limiting environmental media (soil or water) concentrations for the initial screening tier, 95th percentile values have been compared (these being used to derive the limiting environmental media concentrations). This enabled us to assess how conservative the extrapolation approaches used to derive the ERICA Tool databases were.

There is an 'expectation' that the extrapolated 95th percentile values (based on data gap filling methods) will be more conservative than 95th percentile values based on actual data in most cases. This has mainly arisen from two considerations: (i) When there was more than one extrapolation value to select from, we have tended to opt for the highest available value; and (ii) when using an extrapolated value, we have tended to apply an exponential distribution (thereafter selecting a 95th percentile) which we have assumed would give a more extreme or pessimistic value than an approach using a real distribution and 95th percentile. However, a test as to whether this expectation of conservatism was well-grounded or not has not been undertaken until now.

2.2 Methodology

The testing approach was split into two parts.

Comparing default $CR_{wo\text{-}media}$ values in the ERICA Tool derived using extrapolation approaches with new empirical data

The first step in the process was to identify and extract data for those radionuclide-reference organism combinations where new empirical $CR_{wo\text{-}media}$ data have been collated and where previously values had been derived using extrapolation methods. Newly acquired $CR_{wo\text{-}media}$ data were selected from the Wildlife Transfer Database (Copplestone et al. 2013). Corresponding guidance-based extrapolated data (for the same radionuclide-reference organism combination) were then taken from the ERICA Tool databases. The derivation of these latter values has been reported in Beresford et al. (2008a) and Hosseini et al. (2008). In a few instances, there were differences between the $CR_{wo\text{-}media}$ values incorporated in the Tool databases and the $CR_{wo\text{-}media}$ values reported in these two papers. In such cases, reference has been made to the Beresford et al. and Hosseini et al. articles as the definitive source of information.

The 95th percentile values were derived from the 'extrapolated' (or guidance–based) $CR_{wo\text{-}media}$ values reported in the ERICA Tool database and compared with the derived 95th percentile values from the recently collated empirical datasets of Copplestone et al. (2013).

The probabilistic functionality of Tier 3 of the ERICA Tool was used to derive the 95th percentiles. The ERICA Tool default values which had been derived using extrapolation approaches were assumed to represent the arithmetic mean and the model run assumed that the underlying distribution was exponential; this is compatible with how these values were treated in the derivation of the ERICA Tool Environmental Media Concentration Limit values (EMCLs) when the $CR_{wo\text{-}media}$ values were derived using a guidance (or extrapolation) approach (see Brown et al. 2008; Oughton et al. 2008). For the newly acquired empirical data, the arithmetic mean and standard deviation were entered and the underlying distribution was assumed to be log-normal, once more in line with the approach used by Brown et al. (2008) to derive EMCL values when $CR_{wo\text{-}media}$ values were from empirical data. In this way, it was possible to compare 40 values for the terrestrial ecosystem and 44 values for aquatic systems (36 of which were freshwater and 8 marine). If an empirical value was based on a single observation, then an exponential distribution was assumed; this was only required in 9 cases.

Testing the efficacy of different extrapolation approaches used in ERICA

The element-reference organism combinations for which recent $CR_{wo\text{-}media}$ data have been collated tend to be those cases that originally employed a preferred option such as utilisation of taxonomic analogues, similar reference organisms or previously published review/recommended values. Over 82 % of the tested approaches fell into these preferred options in the initial analyses. Therefore, many of the extrapolation methods could not be considered in the comparison described above. For this reason, in the second part of the present work, attempts were made to give consideration to all the methods that have been previously used when generating values for the ERICA Tool databases. This has been undertaken for the marine ecosystem only, the other two ecosystems having been considered more thoroughly in the initial analysis described above (reflecting the fact that the marine $CR_{wo\text{-}media}$ values have changed the least from those of the ERICA compilation (Howard et al. 2013; Yankovich et al. 2013)).

Radionuclide-reference organism combinations have been selected where the original ERICA Tool default $CR_{wo\text{-}media}$ was based on empirical data (generally with three or more observations). It was then assumed that no data were available and the extrapolation guidance followed to generate a surrogate value. The surrogate value and empirical data were then compared to indicate whether the guidance provided sensible proxy information. Ninety-fifth percentile values were derived using the ERICA Tool as described above.

2.3 Results and discussion

ERICA extrapolated default values versus newly acquired $CR_{wo\text{-}media}$ data from the wildlife transfer database

For the terrestrial datasets (Figure 2.1), approximately 63 % of the $CR_{wo\text{-}media}$ 95th percentile predictions based on extrapolation approaches, fell within one order of magnitude of the 95th percentile empirical values (i.e. 25 of 40 extrapolated 95th percentile values fell in the range 0.1 to 10 times the corresponding empirical values). The extrapolation approaches under-predicted the 95th percentile (21 of 40 values) approximately as often as they over-predicted (19 of 40 values). Therefore, the extrapolation guidance if applied generally across all types of plants and animals does not necessarily ensure conservatism in the estimated value. In view of the requirement to account adequately for uncertainty in impact assessments and the conservative nature of the assessment tiers wherein default $CR_{wo\text{-}media}$ values are applied, this is not satisfactory. Possible explanations as to why discrepancies are large in some cases are given in Brown et al. (2013).

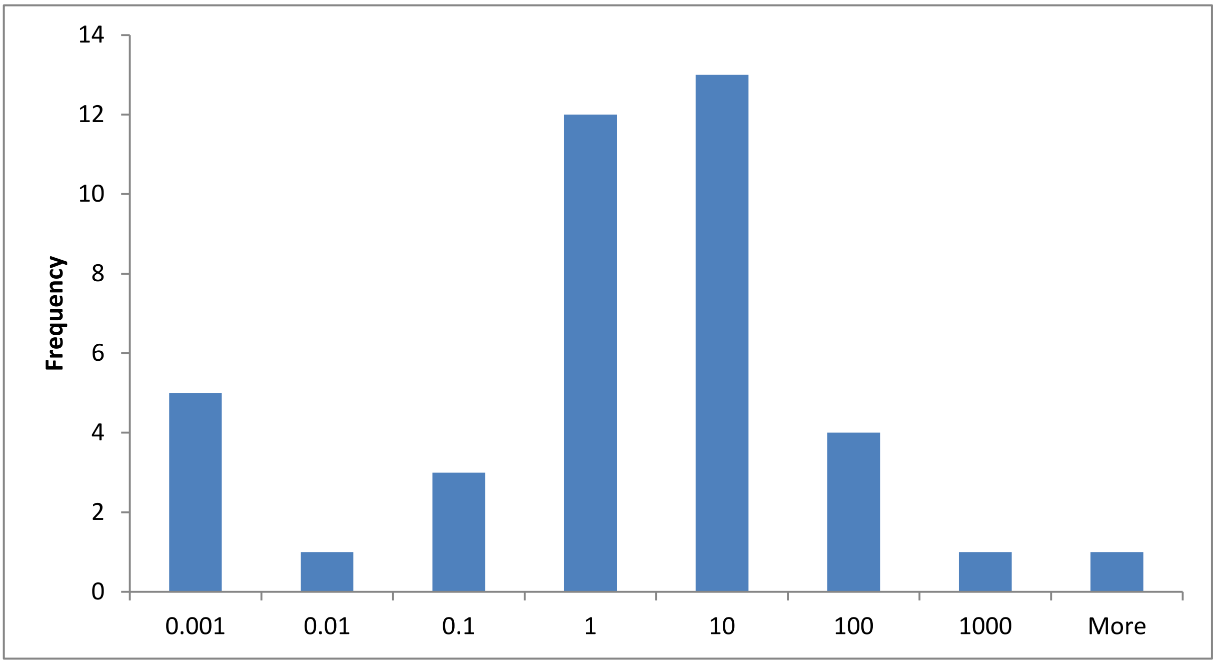

Figure 2.1. Histogram showing distribution of the ratio of predicted (ERICA) / empirical (Copplestone et al. 2013) data for terrestrial organisms. The value of 0.1 corresponds to the interval 0.01 to 0.1 (i.e. an underestimation by a factor between 100 and 10) and the value of 1 corresponds to the interval 0.1 to 1 etc..

For the aquatic (freshwater and marine) ecosystems, the extrapolation approaches used for the ERICA Tool generated 95th percentile $CR_{wo\text{-}media}$ predictions that fell within one order of magnitude of the 95th percentiles for empirical data in approximately 64 % of cases (Figure 2.2). This corresponded to 28 of 44 cases of extrapolated 95th percentile values falling in the range 0.1 to 10 times the corresponding empirical values. Therefore, the application of extrapolation approaches to aquatic ecosystems produced a similar level of efficacy to that observed for the terrestrial ecosystem. However, the guidance, when applied to the aquatic system, had a greater tendency to produce conservative values, with a resultant 27 over-predictions compared to 17 under-predictions. Nonetheless, this is still unsatisfactory for application in an environmental impact assessment in that the guidance is not consistently providing values that are conservative. Conversely, some of the predictions being produced are arguably overly conservative falling at levels 3-4 orders of magnitude above the empirical 95th percentiles. This may lead to unnecessarily restrictive screening assessment results and suggests that the guidance or its application may require refinement. Further details are provided in Brown et al. (2013).

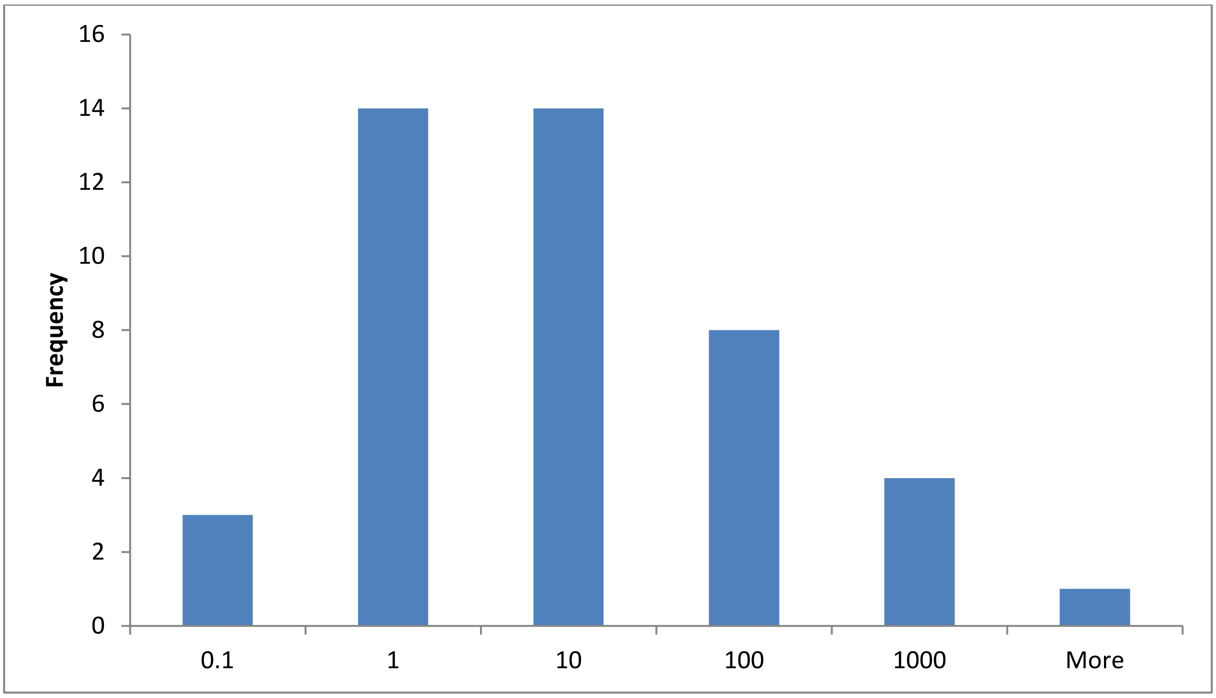

Figure 2.2. Histogram showing distribution of predicted (ERICA)/empirical (Copplestone et al. 2013; Yankovich et al. 2013) data for aquatic ecosystems (comparisons for marine and freshwater ecosystems have been combined).

ERICA extrapolation guidance versus ERICA empirical $CR_{wo\text{-}media}$ data for marine organisms

The following comments can be made:

(1) Use an available $CR_{wo\text{-}media}$ value for an organism of similar taxonomy. This gave 95th percentile predictions for Cs, Pu and Mn in polychaetes (worm) that fell within one order of magnitude of the empirical 95th percentile. Although Cs CRs for vascular plant do not appear to be particularly well represented by Cs CRs for macroalgae, the guidance 95th percentile values are again within one order of magnitude of the empirical 95th percentiles and at least provide a conservative prediction. In 4 of the 5 cases using the taxonomic analogue approach it is not really possible to draw any robust conclusions because the number of observations is low. However, derived values (95th percentile) are generally within one order of magnitude of empirical (95th percentile) CRs although they are not consistently conservative.

(2) Use an available $CR_{wo\text{-}media}$ value for a similar reference organism. The predictions of CRs derived from this approach provide a similar level of efficacy to that observed for the taxonomic analogue approach. With the exception of Pu in 'Bird', all derived 95th percentile values fall within one order of magnitude of the empirical 95th percentiles. Again the approach does not necessarily appear to provide conservative estimates.

(3) Use $CR_{wo\text{-}media}$ values recommended in previous reviews. The use of published review data (taken from IAEA 2004) would not be expected to generate conservative estimates in the absence of empirically derived data, as the published values will tend to be 'best estimates'. It should also be noted that the empirical ERICA database drew upon some common literature sources with IAEA (2004) though the latter provides a set of "recommended" values with no underlying statistical information. This overlap of source data potentially limits the usefulness of this comparison.

(4) Use specific activity models for ^{3}H and ^{14}C. This approach was not evaluated as these models were only applied in the terrestrial environment.

(5) (6) and (7) approaches based around similar biogeochemistry. The predictions made using these approaches are not particularly robust and 95th percentile predictions are at least one order of magnitude higher or lower than the 95th percentile in approximately half of the cases considered. Using Am as an analogue for Cm provided surprisingly poor predictions in view of the fact that both form (III) valence complexes and are considered to have broadly similar environmental behaviours (and have been consider as such in IAEA (2004) which uses them as biogeochemical analogues in the derivation of transfer parameters). Cerium appears to provide a reasonable analogue for Eu, although the datasets are arguably too small to establish any definitive conclusions. The use of Sr as an analogue for Ra appears to work reasonably well for mollusc but less so for fish, leading to 95th percentile estimates that fall more than one order of magnitude below the 95th percentile based on empirical data. The derived values do not generally provide conservative estimates, in 8 of the 11 examples, the empirical 95th percentile value is greater than the derived 95th percentile value.

(8) Use allometric relationships, or other modelling approaches to derive appropriate CRs. The values derived in applying allometric-biokinetic models for radiocaesium are reasonably close to the mean values from the empirical datasets. However, application of the models for other radionuclides is less than robust, with derived 95th percentile $CR_{wo\text{-}media}$ values often exceeding one order of magnitude above or below the corresponding empirical data. This modelling approach does not produce consistently conservative estimations for CRs. For Po in mammal the approach appears to substantially underpredict whereas for Sr in mammal the derived value is elevated compared to the empirical data. This latter result parallels the analysis undertaken by Beresford et al. (2010), where models based on biokenetic-allometric approaches had a tendency to overpredict the transfer of ^{90}Sr to some bird and small mammal species in terrestrial environments. Nonetheless, in the same study biokinetic-allometric models were considered to perform no worse than $CR_{wo\text{-}media}$ approaches in the derivation of whole body activity concentrations for selected radionuclides in selected biota.

(9) Assume the highest available $CR_{wo\text{-}media}$. The predictions made for CRs using the 'highest available' derived values are generally pessimistic (predicted 95th percentiles are more than 10 times higher than observed 95th percentiles) for Pu and Co but match closely with empirically-based values for Cs and Po. Although it was not a great surprise that the Mammal $CR_{wo\text{-}media}$ prediction for Cs was reasonable having been based on a best estimate for

the relatively closely (phylogenically speaking) related bird[2], the proximity of the mammalian 95th percentile Po $CR_{wo\text{-}media}$ to the corresponding derived value was perhaps more surprising as the latter had been derived from data for zooplankton.

(10) Reference organism in a different ecosystem. This approach was not applied for the marine system although it was applied to generate default freshwater $CR_{wo\text{-}media}$ values. There is little evidence to suggest that using marine data as a proxy for freshwater data is appropriate although admittedly there is no overwhelming evidence to the contrary. A consideration, for example, of the comparison provided by Howard et al. (2013) for molluscs in aquatic ecosystems suggests that for this particular case $CR_{wo\text{-}media}$ values between ecosystems generally fall, with the exception of I, within one order of magnitude of each other. Nonetheless, using marine mollusc CRs for Cs, Sr and Pu as proxies for the corresponding freshwater CRs would lead to some under-prediction and substantial over-prediction for I.

(11) Combination of approaches. The derived value for Pu using the 'combined approach' compared well with the value based on empirical data which is probably more a case of 'luck than judgement' whereas the derived value for Sr differed considerably from the empirical value. It is not possible to draw definitive conclusions based on this analysis but it might be expected that combined approaches will not necessarily produce particularly inferior predictions to many of the other approaches.

2.4 Concluding remarks

We should acknowledge that some of the applied extrapolation methodologies (e.g. using review data) could not be expected to provide conservative values, but an important overall conclusion is that the extrapolation methodologies are not guaranteed to over-predict 95th percentiles. For the terrestrial ecosystem the extrapolation methods provide under-predictions of 95th percentiles as often as they produce over-predictions. In a few cases, when considering all ecosystems, the underestimation of $CR_{wo\text{-}media}$ values is substantial, by orders of magnitude, which is clearly unacceptable for a screening assessment.

A number of recommendations were made in the article of Brown et al (2013) based on the analysis outlined above and other considerations. The recommendations, which have subsequently been used to try to improve the ERICA Tool, are addressed below:

Further refinement of the application of extrapolation approaches to derive surrogate values might be attained through a more elaborate consideration of probability distribution functions (PDFs). An alternative to using a best estimate and exponential PDF as currently employed in the ERICA Tool is to use more expansively the statistics provided by a surrogate dataset, e.g. the arithmetic mean, standard deviation and actual (or assumed) distribution of the

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[2] In retrospect this may have been a case of mistaken categorization as using bird data for mammal would have been more correctly categorized as "similar reference organism".

biochemical analogue or similar organism dataset being used to provide a surrogate 'best estimate' value to which an exponential distribution is then applied (see Chapter 7). This has the advantage of avoiding the use of exponential distributions which tend not to reflect the distributions observed for parameters in natural systems. These tend to more often follow normal or log-normal distributions. Extension of the Central Limit Theorem also leads to the view that CRs and K_d should approach log-normal distributions (Sheppard, 2005). The requirement to adopt this approach has been further promoted through the dialogue between the ERICA Tool developers and end users (see Thorne 2013, Avila et al. 2014). In the latest release of the ERICA Tool, efforts have been made to utilise analogue datasets and apply log-normal distributions as far as practicable.

Some simplification of the various options could be made (e.g. simply use 'similar reference organism' rather than having both 'similar taxonomy' and 'similar reference organism'). Furthermore, on the basis of the above comparison, selecting a $CR_{wo\text{-}media}$ value for a 'similar reference organism' (as redefined above) should be used as a preferred approach to select $CR_{wo\text{-}media}$ values for screening level assessments. This approach is now implemented in the latest version of the ERICA Tool (to be released autumn 2014).

The application of data from different ecosystem types should not be used unless further investigation of this approach can validate its use. For instance, the database described by Copplestone et al. (2013) contains data for estuarine species - these may be appropriate surrogates for other aquatic systems. This advice has been followed as far as practicable in the latest version of the ERICA Tool and estuarine data from the Wildlife Transfer Database have provided a useful supplement for marine transfer values.

3. Transfer coefficient versus dietary concentration ratio

The transfer coefficient was first proposed as a measure of the transfer of radionuclides to animal derived food products by Ward et al. (1965) to describe the transfer of radiocaesium from the diet to the milk of dairy cattle. The authors defined the transfer coefficient as the ratio between the radiocaesium activity concentration in milk and the daily dietary radionuclide intake. Ward et al. reported that this parameter exhibited less variability between individual animals within their experimental herd than expressing transfer as the total amount of Cs excreted in milk expressed as a percentage of intake. The same workers also defined the meat transfer coefficient as the ratio of the ^{137}Cs activity concentration in boneless meat to the dietary daily ^{137}Cs intake (Ward & Johnson 1965).

Following the publications of Ward and co-workers in the 1960's, the transfer coefficient was adopted as the basis for quantifying transfer to milk (F_m, d l^{-1} or d kg^{-1}) and meat and eggs (F_f, d kg^{-1}) for all radionuclides. By the late 1970's - early 1980's, transfer coefficient values were being recommended for most radionuclide-animal product combinations (e.g. Ng 1982; Ng et al. 1977; 1979; 1982). These recommended values were incorporated into many predictive food chain models (e.g. Brown & Simmonds 1995; Müller & Pröhl 1993; USNRC 1977; Yu et al. 2001). The IAEA included tabulated recommended transfer coefficient values for animal products (milk, meat and eggs) in its *Handbook of transfer parameter values for the prediction of radionuclide transfer in temperate environments* (IAEA 1994).

On the basis of the many studies which have been conducted over the approximately 50 years since the transfer coefficient concept was introduced, it has generally been accepted that transfer coefficients for smaller animals are higher than those for larger animals, and that those for adults are lower than those for young (and hence smaller) livestock. For instance, transfer coefficients recommend for sheep meat by IAEA (2010) for many radionuclides are *circa* one order of magnitude higher than those recommend for beef. Similarly, transfer coefficients recommended for goat milk tend to be one order of magnitude higher than those recommended for cow milk (IAEA 2010).

Ward and Johnson (1989) commented that the wide use of F_m in radiological models appeared to be justified and that factors such as stable element intake, soil intake, milk production rate, metabolic rate and inhalation could be ignored for most situations. They acknowledged that these conclusions were based primarily on Cs data for cow milk and noted the lack of critical evaluations for other species and radionuclides (Ward & Johnson 1986). It has subsequently been demonstrated that many factors affect transfer coefficients, including those for radiocaesium, such as dietary source, stable element status, exercise and dry matter intake rate (see review by Howard & Beresford 2001).

3.1 Challenging the transfer coefficient concept

The transfer coefficient is estimated as:

$$F_{f,m} = \frac{Radionuclide\ activity\ concentration\ in\ animal\ product\ (Bq\ kg^{-1}\ or\ Bq\ l^{-1})}{Radionuclide\ ingestion\ rate\ (Bq\ d^{-1})} \qquad (3.1)$$

or

$$F_{f,m} = \frac{Radionuclide\ activity\ concentration\ in\ meat\ or\ egg\ (Bq\ kg^{-1})}{Daily\ dry\ matter\ \mathrm{intake}\ (kg\ d^{-1}) \times Radionuclide\ activity\ concentration\ of\ the\ diet\ (Bq\ kg^{-1}\ DM)} \qquad (3.2)$$

From Equation 3.2 it can be seen that the concentration ratio ($\mathrm{CR}_{\mathrm{product\text{-}diet}}$) is equal to:

$$CR_{product-diet} = F_{f,m}\,(\mathrm{d\,kg}^{-1}) \times Daily\ dry\ matter\ \mathrm{intake}\,(kg\ d^{-1}) \qquad (3.3)$$

Using Equation 3.3, Smith & Beresford (2005) used the recommended milk and meat radiocaesium transfer coefficient values for different farm animals from IAEA (1994) together with recommended dry matter intake rates to estimate $\mathrm{CR}_{\mathrm{product\text{-}diet}}$ values (presented here as Table 3.1). Whilst the transfer coefficient values for meat varied over approximately three orders of magnitude from 10 d kg^{-1} (chicken) to $7.9\mathrm{x}10^{-3}$ d kg^{-1} (beef) the range in estimated $\mathrm{CR}_{\mathrm{product\text{-}diet}}$ values was only two-fold. However, estimated $\mathrm{CR}_{\mathrm{product\text{-}diet}}$ values for milk varied over approximately an order of magnitude (see below and Table 3.2). The similarity in $\mathrm{CR}_{\mathrm{product\text{-}diet}}$ values between species should perhaps not be surprising given that the concentrations of many elements in meat or milk are similar across species (Mertz 1986;1987); Table 3.1 demonstrates this for potassium (a chemical analogue of caesium). Consequently, Smith & Beresford (2005) suggested that much of the observed difference between species in $F_{f,m}$ values was a consequence of differences in dry matter intake rather than any difference in 'radionuclide transfer' between species.

The use of concentration ratios for radionuclides of the macro-elements H, C and S rather than transfer coefficients was suggested by (Galeriu et al. 2007) and Howard et al. (2007). Contents of these elements in meat and milk do not vary significantly with factors, such as milk yield and live-weight, which influence dry matter intake rates and hence estimated transfer coefficient values.

A study of the radiocaesium transfer to groups of female sheep through a breeding cycle (pregnancy – lactation – post-weaning) when analysed using F_f values was found to lead to different conclusions than if analysed using $CR_{meat\text{-}diet}$ (Beresford et al. 2007). This was predominately because variation in dry matter intake rate varied between the groups which influenced the estimates of F_f but not $CR_{meat\text{-}diet}$. This led to the suggestion that the concentration ratio provides a more robust measure of transfer from the diet to animal derived food products than the transfer coefficient.

Table 3.1. Recommended transfer coefficients for radiocaesium and dry matter feed intake rates (IAEA, 1994) and concentration ratios estimated as the product of transfer coefficient and dry matter intake (i.e. using Equation 3.3). Typical potassium concentrations of these products are also shown. Table adapted from Smith and Beresford (2005).

Animal	**Daily dry matter intake (kg d^{-1})**	**F_f (d kg^{-1}) or F_m, (d l^{-1})**	**$CR_{product\text{-}diet}$ [dimensionless]**	**Typical K concentration (mg kg^{-1} FW)**
		Milk		
Cow	1.61E+1	7.9E-3	1.3E-1	1.43E+3
Goat	1.3E+0	1.0E-2	1.3E-2*	1.93E+3
Sheep	1.3E+0	5.8E-2	7.5E-2	1.37E+3
		Meat		
Beef	7.2E+0	5.0E-2	3.6E-1	3.04E+3
Lamb	1.1E+0	4.9E-1	5.4E-1	3.06E+3
Pork	2.4E+0	2.4E-1	5.8E-1	3.765E+3
Chicken	7.0E-2	1.0E+1	7.0E-1	2.570E+3

*Note this value has been corrected from that presented in Smith and Beresford (2005).

In addition to its widespread use for farm animals, some authors have also estimated transfer coefficient values for wildlife (e.g. Thomas et al. 1994; MacDonald 1996; Moss & Horrill 1996). MacDonald (1996) presented mass dependent, or allometric relationships (see Chapter 4) for iodine and caesium transfer coefficients for wild mammals and birds, where for caesium:

$$F_f = 10.2M^{-0.777} \tag{3.4}$$

and for iodine transfer to the thyroid

$$F_f = 4700M^{-0.659} \tag{3.5}$$

were M is mass (kg).

However, Beresford (2003) suggested that the concentration ratio of activity concentrations in the animal to those in the diet ($CR_{wo\text{-}diet}$) would be a constant through the following algebraic derivation using Cs as the example:

$$F_f = \frac{[WB_{Cs}]}{A_f\, I_r} \tag{3.6}$$

Where $[WB_{Cs}]$ is the radiocaesium wholebody activity concentration (Bq kg^{-1} (FW)), A_f is the radiocaesium activity concentration in food (Bq kg^{-1} DM) and I_r is the daily dry matter intake rate of food (kg d^{-1}).

Nagy (2001)[3] presents allometric relationships describing the daily dry matter intake rate of different animals, with that for generic mammals being:

$$I_r = 0.055M^{0.744} \tag{3.7}$$

Equation (3.5) can be rewritten substituting Equations (3.4) and (3.5) for F_f and I_r respectively:

$$10.2M^{-0.777} = \frac{[WB_{Cs}]}{A_f \times 0.055M^{0.744}} \tag{3.8}$$

which can be rearranged to:

$$\frac{[WB_{Cs}]}{A_f} = 10.2M^{-0.777} \times 0.055M^{0.744} \tag{3.9}$$

As the two scaling constants are unlikely to be significantly different to each other then:

$$\frac{[WB_{Cs}]}{A_f} = 10.2 \times 0.055 \approx 0.6 = CR_{wo-diet} \tag{3.10}$$

A suggested $CR_{wo\text{-}diet}$ value across different wildlife species compares relatively well to the available data (see Table 3.4 below).

Beresford et al. (2004) proposed the same hypothesis, that $CR_{wo\text{-}diet}$ would be a constant, but from the allometric expressions for biological half-life and dry matter intake rate.

Concentration ratio a generic parameter

A particular advantage of being able to assume that CR for many radionuclides varies little between species is that generic values can be derived for animals for which no data are currently available. Recognising this, in IAEA (2010) $CR_{product\text{-}diet}$ values were summarised. However, currently transfer coefficient values are more numerous in the literature than CR values.

Tables 3.2 and 3.3 present CR values for milk and meat respectively as presented in IAEA (2010) (see also Howard et al. 2009). It is evident from the tables that there is no consistent ranking between species in CR values as found for $F_{f,m}$ values which are, for instance, consistently higher for sheep and goats compared to cattle (see IAEA 2010). Note that the mean Cs CR for milk (Table 3.2) now various by less than a factor of two compared to the order of magnitude value approximated by Smith and Beresford (2005) (Table 3.1).

As already proposed above (i.e. Equation 3.10) we would also expect that CR values for wildlife would vary little between species and be similar to those of farm animals. Table 3.4

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[3] Note: Beresford 2003 used an earlier allometric relationship for daily dry matter intake as cited by MacDonald (1996)

presents Cs $CR_{meat\text{-}diet}$ values for seven herbivorous species of wild mammals and birds. The CR values for these species are broadly similar to those for the meat of farm animals. As Cs is relatively homogenous distributed throughout the body tissues (Yankovich et al. 2010a) then it can be assumed that $CR_{meat\text{-}diet} \approx CR_{wo\text{-}diet}$.

We should acknowledge that whilst the CR is a more robust and generic parameter than transfer coefficients it is still subject to variation due to a number of the parameters which have been shown to influence transfer coefficients (e.g. bioavailability of the dietary source, concentration of stable/analogue elements in the diet).

Table 3.2. Summarised milk:diet concentration ratios for different animals (kg l^{-1}); adapted from IAEA (2010).

Ele-ment	Cow					Goat					Sheep					Horse	Mean	Ratio
	CR	SD	Min	Max	N	CR	SD	Min	Max	N	CR	SD	Min	Max	N	CR	All species	Min/ Max
Ba	1.3E-2	1.6E-3	1.2E-2	1.5E-2	3	1.2E-1		1.4E-2	2.3E-1	2	6.1E-2				1	3.5E-3	5.0E-2	2.9E-2
Ca	2.5E-1					2.0E-1	8.3E-2	1.3E-1	2.9E-1	4	3.4E-1					1.5E-1	2.4E-1	4.4E-1
Cd	4.3e-2	74E-2	2.7E-5	1.3E-1	3	2.4E-2				1	7.4E-2				1		4.7E-2	3.3E-1
Cl	6.9E-2				1												6.9E-2	
Ce	3.2E-3				1												3.2E-3	
Co	2.5E-3				1	7.6E-3				1	6.2E-3				1		5.4E-3	3.3E-1
Cr	4.0E-2		3.7E-2	4.3E-2	2	4.1E-2				1	3.0E-2				1		3.7E-2	7.2E-1
Cs	1.1E-1	1.2E-1	3.6E-3	6.9E-1	119	1.8E-1	6.5E-2	6.3E-2	3.0E-1	12	1.7E-1	1.3E-1	2.0E-2	5.5E-1	17		1.5E-1	6.4E-1
Fe	1.2E-3	2.4E-4	1.0E-3	1.5E-3	3	3.4E-2				1	5.2E-2				1	9.3E-3	2.4E-2	2.4E-2
I	3.0E-1	2.8E-1	3.0E-3	7.9E-1	44	5.0E-1	5.8E-1	8.4E-2	1.2E+0	3	5.8E-1	2.5E-1	2.5E-1	8.8E-1	5		4.6E-1	5.2E-1
Mn	4.5E-3		8.6E-4	8.2E-3	2	1.5E-3				1	3.6E-3				1	1.6E-3	2.8E-3	3.2E-1
Mo	2.8E-2	1.3E-2	1.9E-2	4.3E-2	3	2.7E-2				1						2.1E-2	2.5E-2	7.5E-1
Na	3.7E-1		2.3E-1	5.0E-1	2	1.8E-1					1.6E-1					6.0E-2	1.9E-1	1.6E-1
Nb	1.0E-5					1.9E-5				1					1		1.5E-5	5.3E-1

Element	Cow					Goat					Sheep					Horse	Mean	Ratio
	CR	SD	Min	Max	N	CR	SD	Min	Max	N	CR	SD	Min	Max	N	CR	All species	Min/ Max
Ni	8.2E-2					2.5E-1				1	4.2E-1						2.5E-1	1.9E-1
P	3.1E-1					4.3E-1					4.7E-1				1	1.8E-1	3.5E-1	3.8E-1
Pb	2.4E-3	1.3E-3	9.9E-4	4.3E-3	7	9.0E-3				1	3.0E-2						1.4E-2	7.9E-2
Po	2.4E-3				1												2.4E-3	
S	1.4E-1				1	6.1E-2	3.0E-2	3.5E-2	1.0E-1	4	2.3E-1						1.4E-1	2.7E-1
Sb	2.7E-3				1												2.7E-3	
Se	5.7E-2	4.5E-2	2.6E-2	1.5E-1	7	3.5E-2											4.6E-2	6.2E-1
Sr	2.3E-2	2.2E-2	5.0E-3	1.4E-1	43	4.4E-2	4.4E-2	1.6E-2	1.2E-1	5						4.4E-2	3.7E-2	5.2E-1
Te	8.0E-3				2	1.2E-2				1							1.0E-2	6.7E-1
U	5.0E-3																5.0E-3	
Zn	7.5E-2	1.6E-2	5.5E-2	9.5E-2	6	9.6E-2					1.2E-1					5.5E-2	8.7E-2	4.6E-1
Zr	1.4E-2				1	1.7E-5				1							1.5E-5	8.3E-1

Table 3.3. Summarised meat:diet concentration ratios for different animals; adapted from IAEA (2010).

Ele-ment	**Beef**					**Sheep**					**Pork**					**Generic**	**Ratio**
	CR	**SD**	**Min**	**Max**	**N**	**CR**	**SD**	**Min**	**Max**	**N**	**CR**	**SD**	**Min**	**Max**	**N**		**Min/ Max**
Ag						4.3E-4				1						4.3E-4	
Am						1.1E-4				1						1.1E-4	
Ca	2.3E-2		2.1E-2	2.6E-2	2	1.4E-2										1.9E-2	6.0E-1
Cd	1.7E-1	1.5E-1	2.3E-3	3.5E-1	7	1.2E-2		1.3E-3	2.3E-2	2	1.3E-1				1	9.2E-2	6.9E-2
Ce						2.2E-4				1						2.2E-4	
Cl	2.4E-1		4.8E-2	4.3E-1	2											2.4E-1	
Co	3.9E-1		7.2E-3	7.8E-1	2	2.3E-1										3.1E-1	5.9E-1
Cs	2.3E-1	1.7E-1	2.2E-2	7.3E-1	17	6.4E-1	1.0E+0	5.3E-2	7.5E+0	51	9.2E-2	1.0E-1	8.3E-3	2.4E-1	4	3.9E-1	1.4E-1
Fe	2.2E-1	2.5E-1	6.0E-2	7.2E-1	6	2.7E-1				1						2.5E-1	8.2E-1
I	9.5E-2	8.2E-2	3.2E-2	1.9E-1	3					1	9.3E-2		3.5E-2	1.5E-1	2	9.4E-2	9.8E-1
La	1.6E-3	2.4E-4	1.3E-3	1.8E-3	3											1.6E-3	
Mg	1.4E-1		9.4E-2	1.9E-1	2											1.4E-1	
Mn	8.0E-3		4.6E-3	1.1E-2	2											8.0E-3	
Mo	9.6E-2		2.5E-2	1.7E-1	2											9.6E-2	

Ele-ment	Beef					Sheep					Pork					Generic	Ratio
	CR	SD	Min	Max	N	CR	SD	Min	Max	N	CR	SD	Min	Max	N		Min/ Max
Na	9.7E-1				1											9.7E-1	
Nb	6.5E-6				1											6.5E-6	
Ni	8.0E-2				1											8.0E-2	
P	1.3E+0[2]															1.3E+0	
Pb	7.7E-2	1.8E-1	1.0E-3	6.2E-1	11	1.2E-2	4.0E-3	9.2E-3	1.6E-2	3	6.6E-1		2.3E-1	1.1E+0	2	2.5E-1	1.8E-2
Po	1.4E-1	1.3E-1	3.7E-2	4.1E-1	7											1.4E-1	
Pu						3.9E-5	2.4E-5	1.5E-5	6.3E-5	3						3.9E-5	
Ra	1.8E-1	3.8E-1	1.3E-3	1.3E+0	11											1.8E-1	
Rb	3.0E-1				1											3.0E-1	
Ru						5.7E-4				1						5.7E-4	
S						5.0E-1										5.0E-1	
Sb	2.7E-1										1.1E+1				1	2.7E-1	
Se																1.1E+0	
Te	1.8E-1															1.8E-1	
Th	6.2E-3	5.0E-3	1.7E-3	1.2E-2												6.2E-3	

Element	Beef					Sheep					Pork					Generic	Ratio
	CR	SD	Min	Max	N	CR	SD	Min	Max	N	CR	SD	Min	Max	N		Min/ Max
U	3.3E-1	6.1E-1	3.0E-3	1.7E+1												3.3E-1	
Zn	1.7E+1	1.1E+1	4.7E-1	3.2E+1	9	2.1E+1		1.3E+1	2.9E+1	2						1.9E+1	8.2E-1

Table 3.4. Caesium $CR_{meat\text{-}diet}$ values for wildlife; for both references 'diet' concentration is determined from dried stomach contents.

Species Latin	Species common	Mean±SD[*]	n	Reference
Cervus elaphus	Red deer	0.40±0.20	56	Chaplow et al. submitted
Odocoileus virginianus	Whitetail deer	0.53[+]	11	Sheppard 2013
Lepus europaeus	Brown hare	0.46	1	Chaplow et al. submitted
Lepus timidus	Blue hare	0.48±0.06	5	Chaplow et al. submitted
Oryctolagus cuniculus	Rabbit	0.27±0.11	20	Chaplow et al. submitted
Tetrao tetrix	Black grouse	0.40-0.85	2	Chaplow et al. submitted
Lagopus lagopus scotica	Red grouse	0.73±0.40	10	Chaplow et al. submitted

[*] where n=2 the range is presented. [+] Geometric mean as presented in source publication.

4. Allometry

4.1 Introduction

Size affects rates of biological processes from cellular metabolism to population dynamics (Peters 1983; Hoppeler & Weibel 2005). The dependence of a biological variable (Y) on body mass (M) is typically characterised by an allometric scaling law of the form:

$$Y = aM^b \tag{4.1}$$

where a and b (the allometric exponent) are constants, b is dimension-less and a has the units of the variable Y per mass to the power of $-b$.

In the 1930's Kleiber (1932) found that basal metabolic rate (measured as heat production) across 13 groups of mature animals ranging from a ring dove (<200 g body mass) to a steer (about 680 kg body mass) was proportional to mass to the power 0.74. Following further analyses which demonstrated similar exponents Kleiber suggested that 'metabolic body size' (now generally referred to as metabolic live-weight) could be determined as $M^{0.75}$ where M is the mass of the animal (Kleiber 1947); this has since become known as *Kleiber's law*. There have been many compilations of allometric relationships for biological parameters across large mass ranges and a multitude of animal and plant species (e.g. Peters 1983; Hoppeler & Weibel 2005; Higley 2010).

It should be acknowledged that there is considerable debate with regard to the numerical values for the allometric exponent, in particular whether it should be 0.75 or perhaps 0.67 for basal metabolic rate (e.g. West et al. 1997; Hoppeler & Weibel 2005; Isaac & Carbone 2010; Agutter & Tuszynski 2011). It has also been suggested that the scaling exponent may itself be dependent upon body mass (Savage et al. 2008). In discussing this issue with respect to radioecological models, Higley & Bytwerk (2007) suggested that given other uncertainties in radioecological modelling, the exact value of the allometric scaling exponent 'may not be of critical importance' for practical (rather than theoretical) purposes. We support this suggestion and Figure 4.1 demonstrates the relatively small effect of assuming $M^{0.67}$ rather than $M^{0.75}$.

4.2 Allometry in radioecology

Many of the reported allometric relationships are useful in radioecological modelling, for instance, dry matter food ingestion rates, water ingestion rates, inhalation rates, etc. and these have been used in a number of models of the radionuclide transfer to wildlife (e.g. Beresford et al. 2008b; Johansen et al. 2012) including the US Department of Energy's 'Graded Approach' as implemented in the RESRAD-Biota model (USDOE 2002, 2004).

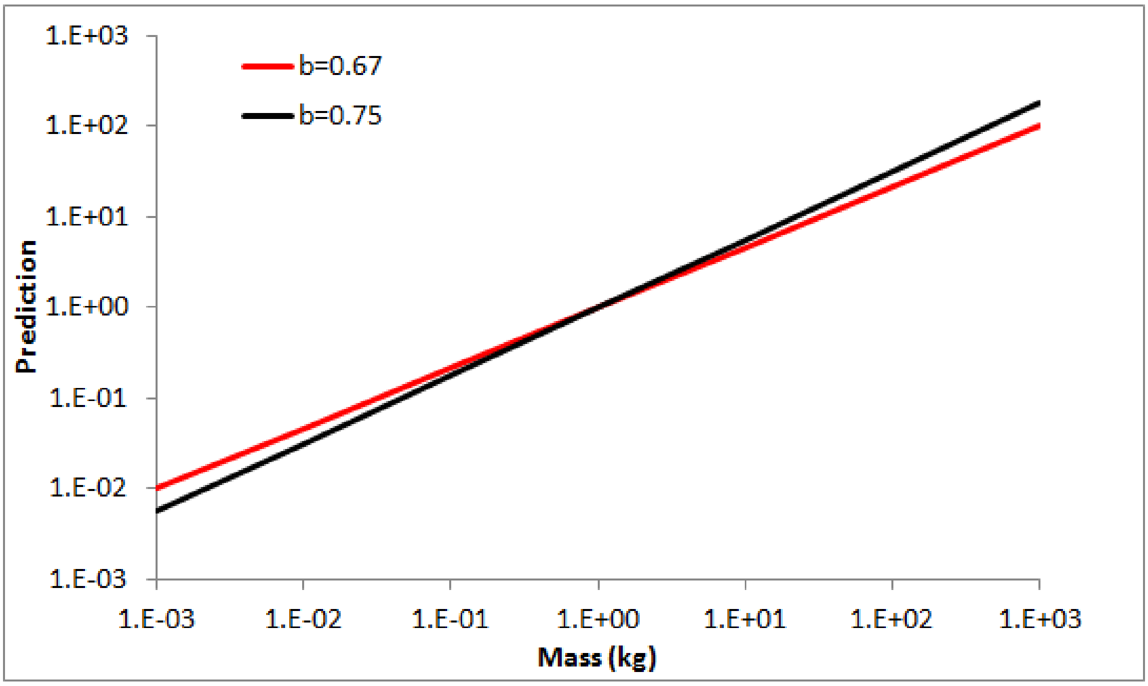

Figure 4.1. A comparison of predictions assuming allometric exponents of 0.67 and 0.75 across a mass range appropriate for terrestrial mammals (predictions are M^b).

Moreover, there are specific radioecological parameters which have been shown to scale allometrically, with relationships for biological half-life across species having been first reported in the 1970's (e.g. Stara et al. 1971; Kitchings et al. 1976). In more recent years, the application of allometry to radioecology has received revived attention during the development of models to predict the exposure of wildlife to radionuclides in both terrestrial (Higley et al. 2003; Higley 2010; Beresford et al. 2004; Sheppard 2001) and aquatic ecosystems (Vives i Batlle et al. 2007; 2009; Brown et al. 2004).

As discussed in Chapter 3, for terrestrial organisms, allometric relationships have also been derived for the dietary transfer coefficient (i.e. the ratio of the activity concentration of a radionuclide in an organism to the daily intake of that radionuclide) (MacDonald 1996). However, as demonstrated above this was the consequence of the dependence of daily dry matter intake on mass and the ratio between the activity concentration in the animal and that in feed is independent of mass (Beresford 2003).

USDOE (2002) presents allometric relationships for the biological half-lives of 16 elements in terrestrial/riparian vertebrates (Table 4.1). When used in model inter-comparison exercises the models using the allometric relationships from USDOE (2002) (or similar) produced results comparable to models using $CR_{wo\text{-}media}$ values (Beresford et al. 2009).

An overview of the application of allometry to aquatic radioecology models is presented in section 4.5 below.

Table 4.1. Allometric constants describing radionuclide biological half-life in terrestrial/riparian vertebrates adapted from USDOE (2002) by Beresford et al. (2004); values of a_B as presented here are for mass in kg converted from a_B' for a mass in g as presented by USDOE using the equation:.$a_B = a_B' \times 1000^b$.

Element	a_B	b	Element	a_B	b
Am	215	0.81	Ra	11.2	0.25
Ce	352	0.8	Sb	2.8	0.25
Cs	18.4	0.24	Sr	645	0.26
Co	13.6	0.24	Tc	4.8	0.4
Eu	352	0.8	Th	888	0.81
H	36.6	0.55	U	5.5	0.28
I	16.7	0.13	Zn	562	0.25
Pu	215	0.81	Zr	562	0.25

The allometric exponent for biological half-life in homeothermic vertebrates

As can be seen in Table 4.1 many of the allometric expressions describing radionuclide biological half-life from USDOE (2002) have an exponent of approximately 0.25. This can be explained on the basis of the relationship between the biological half-life and the metabolic rate as described below.

Taking a simple model, adapted from Sazykina (2000) of intake versus elimination for an adult organism of total mass *M* then the radionuclide activity concentration *y* (Bq kg^{-1}, fresh mass) of the organism changes according to:

$$\frac{dy}{dt} = -\varepsilon_a \frac{B_r}{M}\left[y - \frac{Q_1^A A_f}{Q_0^A} \right] \qquad (4.2)$$

where B_r is the metabolic rate (kg d^{-1}); ε_a is a proportionality constant between the rate of biological loss of a radionuclide from the organism and the metabolic rate of the organism; A_f is the radionuclide activity concentration in food (Bq kg^{-1} dry matter); and Q_1^A, Q_0^A are the total element concentrations in the organism (mg kg^{-1} fresh mass) and in food (mg kg^{-1} dry matter) respectively (here, isotopic equilibrium is assumed, *i.e.* that the ratio of the radionuclide concentration in the organism to that in the diet is the same as the concentration ratio for the total element). This gives the solution:

$$y = y_0 e^{-\varepsilon_a \frac{B_r}{M} t} + \frac{Q_1^A A_f}{Q_0^A}\left(1 - e^{-\varepsilon_a \frac{B_r}{M} t}\right) \tag{4.3}$$

Where y_0 is the activity concentration of the organism at t = 0, *i.e.* at the beginning of depuration. If $A_f = 0$ and $y_0 \neq 0$ (representing a depuration process) Eq. 4.3 becomes reduced to a simple exponential, and applying the definition of biological half-life, i.e.

$y = y_0\, e^{-\frac{\ln 2}{T_{B1/2}} t}$, yields:

$$T_{B1/2} = \frac{M \ln 2}{\varepsilon_a B_r} \tag{4.4}$$

If Kleiber's law is now applied (i.e. $B_r = aM^{0.75}$) then:

$$T_{B1/2} = \frac{\ln 2}{a\varepsilon_a} M^{0.25} \tag{4.5}$$

This is in agreement with the exponent values quoted by USDOE (2002) for many radionuclides (Table 4.1) Note, however, that some radionuclides within USDOE do not scale as approximately 0.25 (Table 4.1; see discussion below).

4.3 A simplified allometric approach for homeothermic vertebrates

The application of allometric biological half-life relationships allows broad approximations to be made to help address the limitations of the current empirical data for wildlife. However, to derive such relationships, adequate data are required for a given element and for a number of species across a range of masses. Sheppard (2001) proposed that, if it is accepted that there is an approximation of the exponent applicable for all elements (i.e. in the case of biological half-life, *c*. 0.25), then only an estimation of the multiplicand is needed for any given element. Here we derive a method of estimating this multiplicand, and hence, extend the applicability of allometric approaches to estimating biological half-life (the derivation of this approach was published as Beresford & Vives i Batlle 2013).

If we start by considering a simple first-order linear retention model with constant input:

$$\frac{dy}{dt} = \frac{f_1 A_f I_r}{M} - \frac{\ln 2}{T_{B1/2}} y \tag{4.6}$$

where y is the fresh mass activity concentration in the whole organism (Bq kg^{-1}), I_r is the dry matter ingestion rate (kg d^{-1}), f_1 is the fractional gastrointestinal absorption coefficient and other terms have been defined above.

Equation 4.6 implies a single component release which is not always observed. However, current allometric relationships predict the long component of loss only (USDOE 2002). At equilibrium (t=∞), Eq. 4.6 equals zero and the equilibrium activity concentration in the organism (y_{eq}) is given by:

$$y_{eq} = \frac{A_f f_1 I_r T_{B1/2}}{M \ln 2} \tag{4.7}$$

This can be rearranged to give the ratio between the activity concentrations in the whole organism (fresh mass) and the diet (dry matter) (CR_{org_diet}):

$$CR_{org-diet} = \frac{f_1 I_r T_{B1/2}}{M \ln 2} \tag{4.8}$$

If we assume that the biological half-life scales allometrically to body mass to the power of 0.25 and that intake rate, which is proportional to metabolic rate, scales allometrically to body mass to the power of 0.75 (see Nagy 2001), then:

$$T_{B1/2} = a_B M^{0.25} \text{ and } I_r = a_I M^{0.75}$$

This gives:

$$CR_{org-diet} = a_B a_I \frac{f_1}{\ln 2} \frac{M^{0.75} \times M^{0.25}}{M} \tag{4.9}$$

Therefore, mass cancels out, and:

$$CR_{org-diet} = a_B a_I \frac{f_1}{\ln 2} \tag{4.10}$$

If we accept that $CR_{org\text{-}diet}$ approximates to a constant for a given element across all species (See Chapter 3 and Beresford et al. (2004)) then a solution to a_B can be proposed:

$$a_B = \frac{\ln 2}{a_I f_1} CR_{org-diet} \tag{4.11}$$

Hence an estimate of $T_{B1/2}$ can be derived for an element if $CR_{org\text{-}diet}$ and f_1 are known:

$$T_{B1/2} = \frac{\ln 2}{a_I f_1} CR_{org-diet} M^{0.25} \tag{4.12}$$

Values of a_I are relatively well documented for terrestrial vertebrates (e.g. Nagy 2001).

Initial hypothesis testing (Beresford & Vives i Batlle 2013)

In Beresford & Vives i Batlle the primary source of $T_{B1/2}$ values used to test this hypothesis was Whicker & Shultz (1982), which tabulates estimates from the literature for a number of radionuclides and terrestrial organisms. For Cs, data were supplemented by values presented in Battison et al. (1991) and Gaare & Staaland (1994). Observed $T_{B1/2}$ values from these sources for Cs, I, Sr and Co are given Table 4.2. The $T_{B1/2}$ values are for the long component of loss, consistent with the allometric $T_{B1/2}$ relationships suggested for use in environmental assessment models (USDOE 2002).

Nagy (2001) fitted allometric relationships to predict dry matter intake rates of terrestrial vertebrates presenting these on the basis of, for example, taxonomic grouping or feeding strategy; a_I values from this source were used here. Reflecting the species for which $T_{B1/2}$ data are available, the a_I values from Nagy (2001) (converted from grammes in Nagy to kilogrammes for application here using the multiplicative factor: $(a_I \times 1000^{0.75})/1000$)) for 'all mammals' ($a_I$=0.057 d^{-1} $kg^{0.25}$), carnivorous mammals (a_I=0.027d^{-1} $kg^{0.25}$) and herbivorous mammals (a_I=0.15d^{-1} $kg^{0.25}$) were used. The a_I value for rodents (a_I=0.059 d^{-1} $kg^{0.25}$) is similar to that of 'all mammals' and hence was not used. Estimates of f_1 were taken from IAEA (2010), which cites values from ICRP (2006) for monogastric animals and additionally presents f_1 for ruminants.

Values of $CR_{org-diet}$ were estimated from $CR_{meat-diet}$ values presented by IAEA (2010) (see Tables 3.2 and 3.3). Whilst IAEA (2010) presents these parameter values for Co, Cs and I, it does not include a $CR_{meat-diet}$ value for Sr. Dietary transfer coefficients presented in IAEA (2010) were used therefore used together with typical dry matter intake rates from IAEA (1994) to estimate the average Sr $CR_{org-diet}$ across all five species for which F_f data were available (cattle, goat, sheep, poultry and pig) in IAEA (2010).

Although Yankovich et al. (2010a) present tissue to wholebody radionuclide activity concentration conversion factors for a range of wildlife groups which can be used to derive wholebody $CR_{org-diet}$ estimates from $CR_{meat-diet}$ values only Cs is included for mammals of the elements considered by Beresford & Vives i Batlle. Therefore, for Sr and Co, conversion factors were estimated using data presented by Barnett et al. (2013, 2014) for *Apodemus sylvaticus* (wood mice) and *Capreolus capreolus* (roe deer); a conversion factor for I has been estimated from information presented in Coughtrey et al. (1983).

All parameter values used in Equation 4.12 by Beresford & Vives i Batlle are presented in Table 4.2.

Using the value of a_I for 'all mammals' to predict $T_{B1/2}$, all predictions were within an order of magnitude of the observed values with most being within a factor of three (Table 4.3). Given that the allometric models are designed to give a broad approximation rather than an exact value this is an acceptable level of agreement. For Cs and I there was a tendency to under-predict, whereas for Co all estimates were over-predicted. If the a_I suggested for carnivorous mammals by Nagy (2001) is used there was a marked improvement in predictions for Cs and I for carnivorous species (Table 4.3). However, applying the a_I applicable to herbivorous mammals, the predictions for the relatively few herbivores considered in the comparison were underestimated with the exception of the estimates for Co in laboratory rabbit, and Sr in mule deer (Table 4.3).

A linear regression of $T_{B1/2}$ values predicted using a_I values appropriate to the feeding type of each species with the measured data (from Table 4.3) yielded an R^2 value of 0.58 with a slope of 1.4 and an intercept which is not significantly different from zero ($p<0.001$).

Table 4.2. Parameter values used in Equation 4.12 to predict the biological half-life of radionuclides in mammals and birds; all data sources are identified within text.

Element	**Monogastric** f_1	**Ruminant*** f_1	$CR_{meat\text{-}diet}$	**Wholebody to muscle correction factor**	$CR_{org\text{-}diet}$
Ag	5E-2	n/r	4.3E-4	1.2E+2	5.2E-2
Co	1E-1	n/r	3.1E-1	3E+0	9.3E-1
Cs	1E+0	8E-1	3.9E-1	1E+0	3.9E-1
I	1E+0	1E+0	9.4E-2	5E+0	4.7E-1
Po	5E-1	n/r	1.4E-1	2E+0	2.8E-1
Sr	3E-1	1.1E-1	2.2E-2	4.0E+2	8.8E+0
Zn	5E-1	n/r	1.9E+0	1.8E+0	3.42E+0
Na	n/a	9E-1$^+$	9.7E-1	1E+0	9.7E-1
Nb	1E-2	n/r	6.5E-6	1.1E+1	7.2E-5
Ru	5E-2	n/r	5.4E-4	1E+0	5.4E-4
Se	8E-1	n/r	1.1E+0	1E+0	1.1E+0

n/a – not available; n/r – not required for this work; *used to derive predictions in Table 4.3 only; $^+$used for all mammals.

Table 4.3. Comparison of measured and predicted $T_{B1/2}$ values as presented by Beresford & Vives i Batlle (2013).

Species	**Mass (kg)**	$T_{B1/2}$ **reported**	**Predicted** $T_{B1/2}$ **using** a_I **for:**		
			Mammals	**Carnivores**	**Herbivores**
Radiocaesium					
Harvest mouse	1.0E-2	3.7	1.5		
Laboratory mouse	2.0E-2	5.1	1.8		
Whitefooted mouse	2.1E-2	3.5	1.8		
Cotton rat	1.3E-1	8.4	2.8		
Laboratory rat	1.9E-1	6.3	3.1		
Rabbit	1.6E+0	11	5.3		2.0
Arctic fox	4.9E+0	17.5	7	15	

Species	**Mass (kg)**	**$T_{B1/2}$ reported**	**Predicted $T_{B1/2}$ using a_I for:**		
			Mammals	**Carnivores**	**Herbivores**
Silver fox	5.3E+0	25.3	7.1	15	
Coyote	9.5E+0	26	8.3	17	
Red fox	1.0E+1	29	8.4	18	
Coyote	1.2E+1	22	8.7	18	
Dog	1.9E+1	28	9.8	21	
Wolf	3.1E+1	23	11	24	
Mule deer	5.5E+1	14	13		4.8
Reindeer	8.0E+1	14	14		5.3
Radiocobalt					
Whitefooted mouse	2.0E-2	5.2	42		
Laboratory mouse	2.5E-2	4.8	45		
Laboratory rat	4.0E-1	11	89		
Guinea pig	4.7E-1	21	93		
Laboratory rabbit	3.0E+1	13	148		56
Radioiodine					
Laboratory mouse	2.1E-1	5.2	2.2		
Cotton rat	1.1E-1	8	3.3		
Laboratory rat	2.1E-1	2.5	3.8		
Guinea pig	5.0E-1	26	4.8		
Jack rabbit	1.9E+0	5	6.7		2.5
Laboratory rabbit	3.7E+0	13	7.9		3.0
Dog	1.2E+1	17	10	22	
Radiostrontium					
Laboratory mouse	3.0E-2	43	140		
Laboratory rat	2.0E-1	590	240		
Dog	1.0E+1	530	640	1,300	
Mule deer	6.5E+1	190	1,000		380

As both mule deer and reindeer are ruminants, predictions for these animals were also made using the ruminant specific f_1 values from (IAEA 2010) of 0.8 and 0.11 for Cs and Sr respectively. This made little difference to the predictions for Cs (an increase by 25 %) but in the case of mule deer the Sr $T_{B1/2}$ predicted using the herbivorous mammal a_I and ruminant f_1 values was 1,000 days compared with the observed value of 228 days. It is possible that an f_1 value based on agricultural animals receiving diets likely to have excess Ca to requirements is not representative of wild ruminants and that a higher f_1 value would be more applicable; a higher f_1 value would decrease the predicted Sr $T_{B1/2}$.

An extended test of the Beresford & Vives i Batlle model - method

Subsequent to publishing our model to estimate biological half-life, a review of radionuclide biological half-life values has been conducted (this is contributing to IAEA MODARIA programme activities (see: http://bit.ly/1e9Nxxq)). The full data set which considers freshwater and marine species and an expanded range of terrestrial organisms to those considered here will be published in the near future. This has enabled us to expand our test of the model proposed above (i.e. Equation 4.12) to consider a wider range of elements (Ag, Co, Cs, I, Na, Nb, Ru, Se, Sr and Zn) and also, in the case of Zn, to include birds. The data selected for comparison with model predictions had to meet various criteria:

- Animal live-weight mass had to be presented in the source reference
- Data for infants and juveniles were rejected
- $T_{B1/2}$ values had to be for the wholebody except in the cases of I and Cs where thyroid and muscle data were also used respectively, assuming these tissues reflected wholebody loss rates
- With the exception of I, Sr and Cs only data for studies where the radionuclide had been ingested or administered orally were considered; for I, Sr and Cs data from studies using intravenous administration were also used as the weight of evidence suggested these elements behave the same in the circulatory system after both oral and intravenous administration (Mayes et al. 1996).

A total of 123 $T_{B1/2}$ values were considered in the expanded testing of the model; this included data originally considered by Beresford &Vives i Batlle (2013) though source references, or earlier reviews cited by Whicker & Schultz, were consulted rather than relying on the Whicker & Schultz (1982) compilation. Note that in a few instances comparisons used in Table 4.3 were not included in this expanded evaluation as it appears some of the animal masses cited by Whicker & Schultz (1982) may not have been for the actual study animals. When multiple components of loss were cited in the source reference, the longest $T_{B1/2}$ value was selected for comparison with predictions in this assessment. The $T_{B1/2}$ values include study means and in some cases individual values. All data sources used to provide $T_{B1/2}$ values for this comparison are presented in Appendix A.

Parameter values used in Equation 4.12 are all presented in Table 4.2. For Cs, Co, I and Sr the same parameter values as applied in Beresford & Vives i Batlle et al. (2013) were used. To make predictions for birds (*Anas platyrhynchos*, mallard duck) the a_I values from allometric dry matter intake relationship for omnivorous birds (a_I=0.119d^{-1} $kg^{0.25}$) presented by Nagy (2001) was used. In addition the a_I presented by Nagy for omnivorous mammals was used where appropriate (a_I=0.077d^{-1} $kg^{0.25}$). Values of f_1 were available from IAEA (2010) for all elements considered; for Na the f_1 value quoted in IAEA for ruminants was used for all three species considered (rat, laboratory mouse and Rhesus macaque (*Macaca mulatta*)). For the additional elements not considered by Beresford & Vives i Batlle $CR_{meat\text{-}diet}$ values were available from IAEA (2010). These were converted to $CR_{\mathrm{org\text{-}diet}}$ using muscle to wholebody correction factors presented by Yankovich et al. (2010a) for Ag, Po, Zn, Ru and Se. Yankovich et al. does not present correction factors for either Na or Nb. For Na and Nb data for wood mice and roe deer from Barnett et al. (2013, 2014) were used to estimate correction factors of 2 and 11, respectively.

An extended test of the Beresford & Vives i Batlle model - results and discussion

The majority of predictions were within an order of magnitude of the observed data: 97 of 110 predictions using the a_I for all mammals and 107 of the 123 predictions made using the most appropriate feeding group a_I.

Using the a_I for all mammals nine predictions were more than an order of magnitude lower than the observed data; with the exception of one prediction (Cs in rabbit), these large under-predictions were for Nb (n=4) and Ru (n=4). These eight comparisons comprised all of the available data for Ru and Nb. Only four predictions were more than an order of magnitude in excess of the observed data. All of these over predictions were for Sr; these data had not previously been considered in the initial evaluation of the model as presented in Table 4.3. These observations are discussed further below.

When predicted using a_I values for specific feeding strategies then:

Carnivorous mammals – Using the a_I value for carnivorous mammals increased the predicted $T_{B1/2}$ value compared to that predicted using the 'all mammals' a_I value (Figure 4.2). The predictions using the carnivorous mammal a_I value were generally in better agreement with the observed data.

Herbivorous mammals - Using the a_I value for herbivorous mammals decreased the predicted $T_{B1/2}$ value compared to that predicted using the 'all mammals' a_I value (Figure 4.2). This did not have a consistent tendency across all of the elements to improve or not the level of agreement between predictions and observed data (e.g. for I predictions using the a_I value herbivorous mammals were in better agreement with the observed data than those using the 'all mammals' a_I value, whereas the opposite was the case for Zn).

Omnivorous mammals - Using the a_I value for omnivorous mammals decreased the predicted $T_{B1/2}$ value compared to that predicted using the 'all mammals' a_I value but only by *c*. 25 %.

For herbivores and carnivores these observations are in agreement with those made during the initial testing of the model (see Table 4.3); the omnivorous a_I was not applied in the earlier comparison.

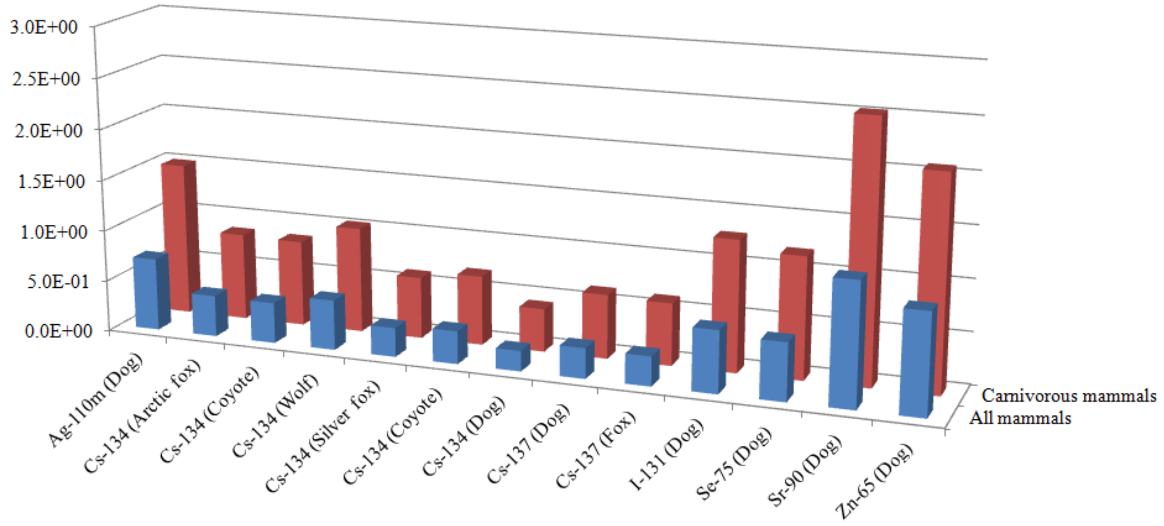

Figure 4.2. The effect of a_I value on the predicted $T_{B1/2}$ values for carnivorous mammals; note three outlying data points (one each for Nb, Ru and Sr) are not shown.

Of those elements previously considered in Beresford & Vives i Batlle (2013; Table 4.3) predictions for I were in good agreement for herbivorous and carnivorous mammals. For omnivores predicted values tended to be <20% of the observed data values. Only two predictions for Co were made with predicted:measured ratios of 0.9 and 6 when the feeding group a_I value was used. Caesium data were the most numerous (n=55) omitting an outlying value for rabbit, under-predicted by more than an order of magnitude, a linear regression of measured $T_{B1/2}$ values to predictions made using the appropriate feeding group a_I yields an R^2 of 0.52 and an intercept (-0.69) which is not significantly different to zero and a slope of 0.43. The slope demonstrates a tendency to under-predict the measured values.

All of the Sr $\mathrm{T}_{B1/2}$ values considered here which were additional to those in Table 4.3 were over-predicted by more than an order of magnitude; those previously consider, which were also included in this evaluation, were not over-predicted to this degree (see Table 4.3). The additional data were all for ^{85}Sr and originate from two studies cited by Kitchings et al. (1976) and have $\mathrm{T}_{B1/2}$ values which are comparatively short compared to the Sr data used in Table 4.3. For instance, the $\mathrm{T}_{B1/2}$ for dog from Moskalev & Buldakov (1968) as cited by Kitchings et al. (1976) and used in our initial model testing (Table 4.3) is 530 d compared to the ^{85}Sr value for dog now included in this comparison (Glad et al. 1960; cited by Kitchings et al.) of18 d. The additional ^{85}Sr considered here appears to show little dependence on mass with $\mathrm{T}_{B1/2}$ ranging from 10-18 d for animals with masses in the range 33 g (mouse) to 10 kg (dog) (Kitchings et al. 1976).

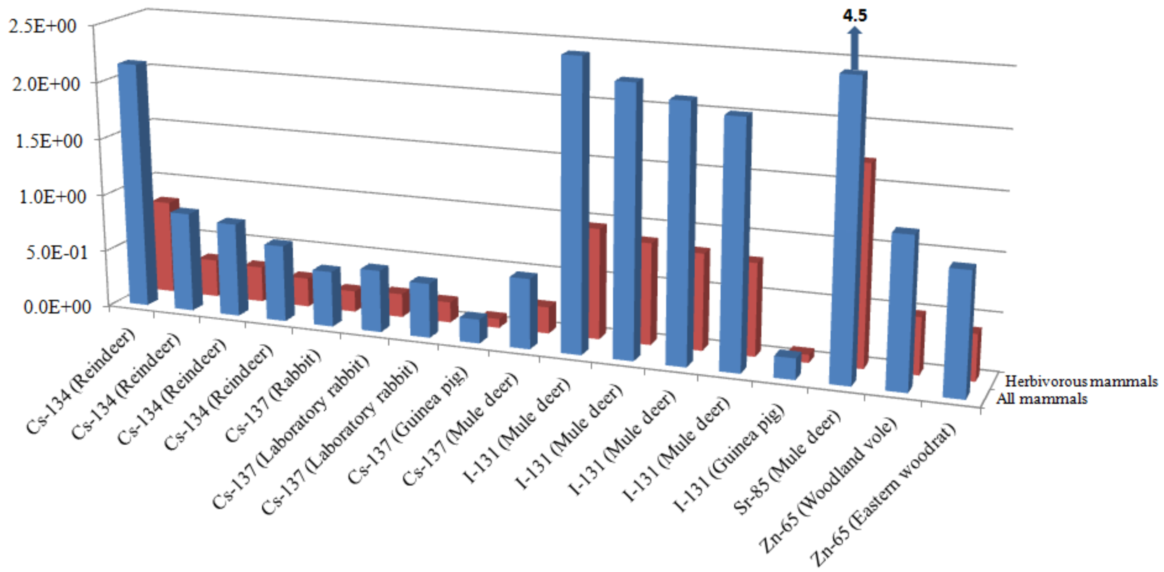

Figure 4.3. The effect of a_I value on the predicted $T_{B1/2}$ values for herbivorous mammals; note two outlying data points (one each for Cs and Sr) are not shown.

A summarised comparison of predicted with observed values for those additional elements which were not considered by Beresford & Vives i Batlle (2013) is presented in Table 4.4. Predictions for Ag, Se and Zn all had an acceptable level of agreement. Those for Na were within an order of magnitude of the observed $T_{B1/2}$ values but were all comparatively low. As already noted above predictions for both Ru and Nb were poor, both elements being considerably under-predicted.

In the cases of Ru, Nb, and Na it is worth noting that the $CR_{meat\text{-}diet}$ values in IAEA (2010) are each based upon one observation only; though this is also the case for Ag and Se. However, to improve the predictions such that they are within an order of magnitude of the observed values the $CR_{meat\text{-}diet}$ for Nb would need to increase by approximately 1-2 orders of magnitude and that for Ru by about two-orders of magnitude. The f_1 values used also impacts on the predicted $T_{B1/2}$ value for both Nb and Ru. There are reported values for these elements which are lower than those we have used in this assessment (Coughtrey & Thorne 1983, Beresford et al. 1998a); application of a lower f_1 would improve the predicted values. However, the Nb and Ru $T_{B1/2}$ used here are from Furchner and Drake (1971) and Furchner et al. (1971) respectively and the f_1 values estimated by these authors from the experimental data were similar to those used to make our predictions.

Table 4.4. A summary comparison of predicted with observed values for those elements not considered by Beresford & Vives i Batlle (2013).

Element	Species compared	N[+]	Predicted:observed using 'all mammal' a_I	Predicted:observed using feeding group a_I
Ag	Mouse, rat, monkey, dog	4	0.79-6.8	1.2-5.0
Na	Mouse, rat, monkey	4	0.13-0.31	0.10-0.23
Nb	Mouse, rat, monkey, dog	4	0.002-0.10	0.002-0.08
Ru	Mouse, rat, monkey, dog	4	0.001-0.02	0.0008-0.03
Se	Mouse, rat, monkey, dog	4	0.40-0.56	0.29-1.2
Zn	Mouse, rat, vole, monkey, dog, duck	29	0.21-5.9	0.15-4.4

[+]Number of comparisons.

Whilst there was reasonable agreement between the predicted and measured Zn $T_{B1/2}$ values for ducks, $T_{B1/2}$ values varied considerable more for the observed data (28-250 d; n=13) than the predicted values (41-42 d). This is not surprising given that the mass of the ducks varied little (1.11-1.25 kg).

It is possible that at least some of the mismatches between allometrically-predicted and literature values could relate to QC issues in the data from the literature that we are trying to predict. In future, we plan to continue this research beyond the present study by sourcing and quality-assuring some of the data which appear to give incongruent values.

Discussion

Our ability to obtain reasonable predictions is in part dependent upon the quality of data available for the required input parameters. For many elements in IAEA (2010), $CR_{\text{meat-diet}}$ are based upon few observations as already highlighted above. Similarly, the correction factors to convert from $CR_{\text{meat-diet}}$ to $CR_{\text{org-diet}}$ are based on relatively few data as exemplified by the need to derive them for Co, I and Sr. Investigation of the data used by Nagy (2001) to derive the allometric dry matter intake relationship for herbivores shows that they are dominated by relatively small species with many of the larger species being marsupials.

It should also be acknowledged that the dry matter intake relationships presented by Nagy (2001) are for animals under field and not laboratory conditions and that field metabolic rates are generally higher than basal metabolic rates determined for housed animals (Nagy 2005). This may result in a tendency to under-predict $T_{B1/2}$ for housed (i.e. experimental) animals.

Overall in the above assessment there was a tendency to under- rather than over-predict; 103 of the 123 predictions using feeding group a_I values being less than the observed value.

An assumption of the approach described here is that $T_{B1/2}$ scales to the power of 0.25. Of the allometric expressions derived for $T_{B1/2}$ for 16 radionuclides by USDOE this is true for eight (Cs, Co, Ra, Sb, Sr, U, Zn and Zr). The mass scaling functions for I and H reported by USDOE are 0.55 and 0.13 respectively. However, other sources suggest that the scaling function for the $T_{B1/2}$ for these two radionuclides should be *circa* 0.25 (Galeriu et al. 2003; MacDonald 1996).

For five elements in USDOE (2002) (Am, Ce, Eu, Pu and Th) biological half-life scales to the power of 0.8. For all of these ICRP publications (ICRP 1979; 1981; 1988) are quoted as the data source but unfortunately we have not been able to find the data within these publications to independently verify the allometric equations presented by USDOE (2002). None of the elements scaling to *circa* 0.8 play an active biological role and hence it could, perhaps, be suggested that there is no reason for them to follow a metabolically driven uptake process (i.e. as signified by a mass scaling function of 0.25). However, we acknowledge that some of the elements which do scale as mass to the power of 0.25 are not essential elements either (e.g. U). The assumption that $CR_{org\text{-}diet}$ is independent of mass for any elements for which $T_{B1/2}$ is proven to not scale allometrically with a value approximating to 0.25 will be invalid (i.e. see Eq. 4.9).Therefore, it is recommended that the reasons for the deviation of some elements from the mass scaling function of 0.25 be explored and/or the relationships presented by USDOE (2002) be independently verified.

As noted above for comparison with our predictions we have used the longest $T_{B1/2}$ value reported where multiple components of loss were observed. In doing this we made no differentiation based on the magnitude of this loss component even though in many instances the long component of loss contributes relatively little to the total loss (<5 %). It may be questionable whether the longest $T_{B1/2}$ best represents metabolic turnover when three and four component loss equations are reported; the component of loss associated with the longest component can be small.

The assumption of a single long-component of loss as currently used in most wildlife assessment models (e.g. USDOE 2002; Avila et al. 2004) should yield estimates of the equilibrium activity concentration in organisms which are conservative (i.e. they should be overestimated compared to a model assuming more than one loss component). However, if used in dynamic models this assumption will predict slower changes in organism activity concentrations than would be observed in reality as a result of changes in activity concentrations in environmental media.

4.4 Applying the Beresford and Vives i Batlle model to the prediction of reptilian biological half-life

Although only proposed for homeothermic vertebrates, we are aware that allometric models for $T_{B1/2}$, such as USDOE (2002), have been used to make predictions of radionuclide activity concentrations in reptiles and amphibians (e.g. Beresford et al. 2010; Yankovich et al. 2010b; Johanssen et al. 2012).

In this sub-section we summarise Beresford & Wood (2014) which evaluated to what extent the Beresford & Vives i Batlle model (i.e. Equation 4.12) could be applied to reptiles. To enable this in Beresford & Wood we first conducted a critical review of $T_{B1/2}$ data for reptiles. Similar criteria as used above for mammal and bird data were applied resulting in 28 $T_{B1/2}$ values for Cs, three of Sr and two for Ra. Data for Cs was predominantly for different species of snake (Staton et al. 1974) with one value for a turtle species (*Trachemys scripta scripta*) (Scott et al. 1986). Data identified for Sr and Ra were all for *T. scripta scripta* (Scott et al. 1986; Hinton et al. 1992). Data covered reptiles with masses ranging from 0.02 to 1.5 kg. Most of the estimated $T_{B1/2}$ values available were longer than the length of the studies from which they were derived. For instance, the maximum radiocaesium half-life determined by Staton et al. (1974) for snakes from their 63 d long study was 430 d. This will undoubtedly add some uncertainty to the reported $T_{B1/2}$ values.

Predicting biological half-life using the Beresford & Vives i Batlle equation

Figure 4.4 compares predicted with measured $T_{B1/2}$ values using Equation 4.2 and input parameter values for mammals. For Ra, not considered above, input values were sourced from IAEA (2010) and Yankovich et al (2010a).

With the exception of one value, all predictions were underestimates; in the case of Cs the underestimates were by 1-2 orders of magnitude (Figure 4.4). There was comparatively less variation in predicted values for a given radionuclide than in the measured values.

It was possible to source values for the constants in Equation 4.12 (i.e. f_1, $CR_{org\text{-}diet}$ and a_I) which are more appropriate for reptiles. Nagy (2001) presents allometric equations for the dry matter intake of reptiles from which a value of a_I for carnivorous reptiles, appropriate for snakes, of 0.0067 can be estimated. Nagy does not give values directly appropriate for *T. scripta scripta* (i.e. omnivorous feeder or testudinata). Therefore the generic reptile a_I value of 0.0064 was used. The Nagy values have been converted from grammes to kilogrammes as described above for mammals. Values of f_1 for reptiles of 0.25, 0.2 and 0.5 for Cs, Ra and Sr respectively have been published (Peters and Brisbin 1996; Hinton and Scott 1990; Hinton et al. 1992). As elemental concentrations in the tissues of reptiles appear broadly similar (Yoshinaga et al. 1992) to those of mammals, the $CR_{meat\text{-}diet}$ values sourced from IAEA (2010) were assumed. Conversion factors from $CR_{meat\text{-}diet}$ to $CR_{org\text{-}diet}$ are described in Beresford & Wood. The $CR_{org\text{-}diet}$ values used were 0.39 for Cs for all reptiles and 2.12 and 23.6 for Ra and Sr respectively, the latter two being derived specifically for turtles.

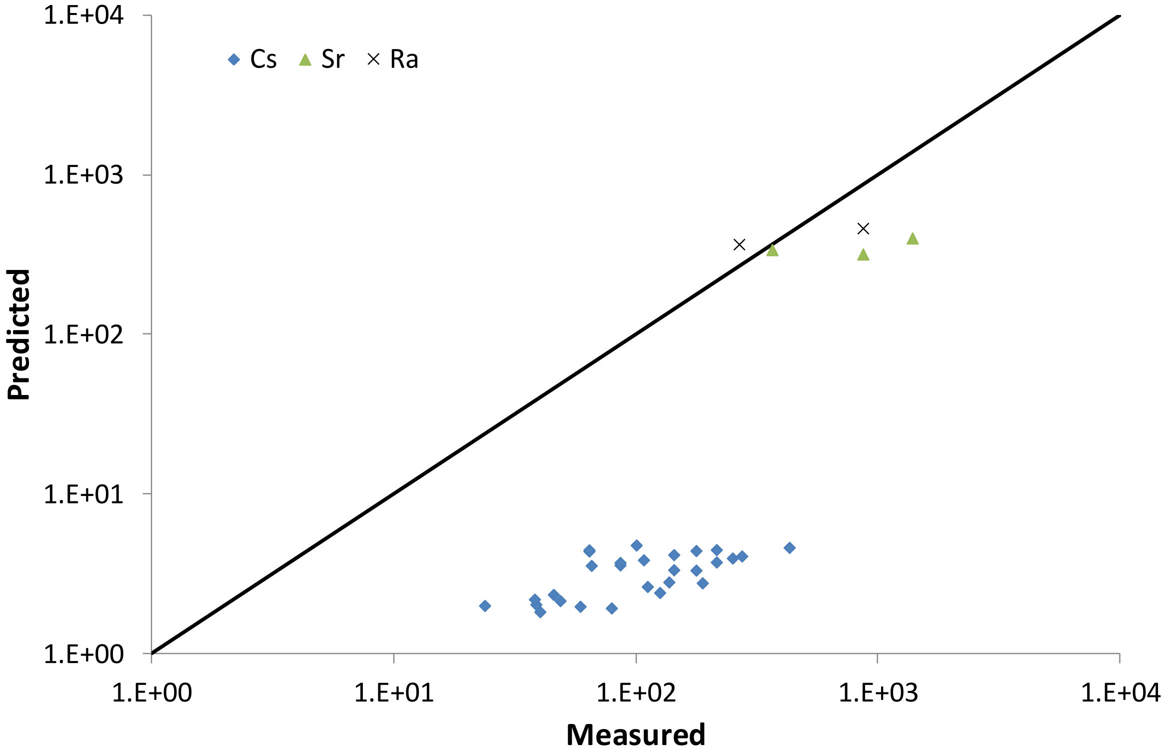

Figure 4.4. A comparison of measured radionuclide biological half-life (d) in reptiles with predictions using parameters for mammals. The line is the 1:1 line.

Predictions made when parameterising Equation 4.12 using the reptile specific values for f_1, $CR_{org\text{-}diet}$ and a_I were an improvement over those using mammalian parameter values (Figure 4.5).

Adapting for reptilian metabolic rate

As demonstrated above (see Equations 4.3 to 4.5) the exponent in Equation 4.12 is defined by the exponent for the allometric model of metabolic rate of *c*. 0.75. In effect the exponent for $T_{B1/2}$ is (1 – the exponent for B_r).

The exponent on the allometric model describing B_r for reptiles has been shown to be in the range *c*. 0.80-0.92 (Nagy 2005, Isaac and Carbone 2010). Therefore, the exponent describing $T_{B1/2}$ should be in the range 0.08-0.20. The exponents of allometric model of dry matter intake presented for reptiles by Nagy (2001) should approximate to those for B_r (Nagy 2001). For snakes an exponent for application in Equation 4.12 of 0.037 can be estimated from the allometric model describing dry matter intake (Nagy 2001). For turtle an exponent of 0.08 is estimated assuming the dry matter intake model proposed for 'all reptiles' (Nagy 2001).

The exponents together with the relevant values of a_I and the reptile specific values of $CR_{org\text{-}diet}$ and f1 described above were used to estimate $T_{B1/2}$ values to compare with the available data. The resultant predictions are presented in Figure 4.6. Predictions are similar to those in

Figure 4.5 (where an exponent of 0.25 was used with reptile specific constant in Equation 4.12) though there is less variation in the predicted values for Cs. Only one of the 33 predictions deviates by more than a factor of 6 from the measured value; the one 'outlier', a prediction of the Sr $T_{B1/2}$ in *T. scripta scripta* was predicted to be 14 times higher than the measured value.

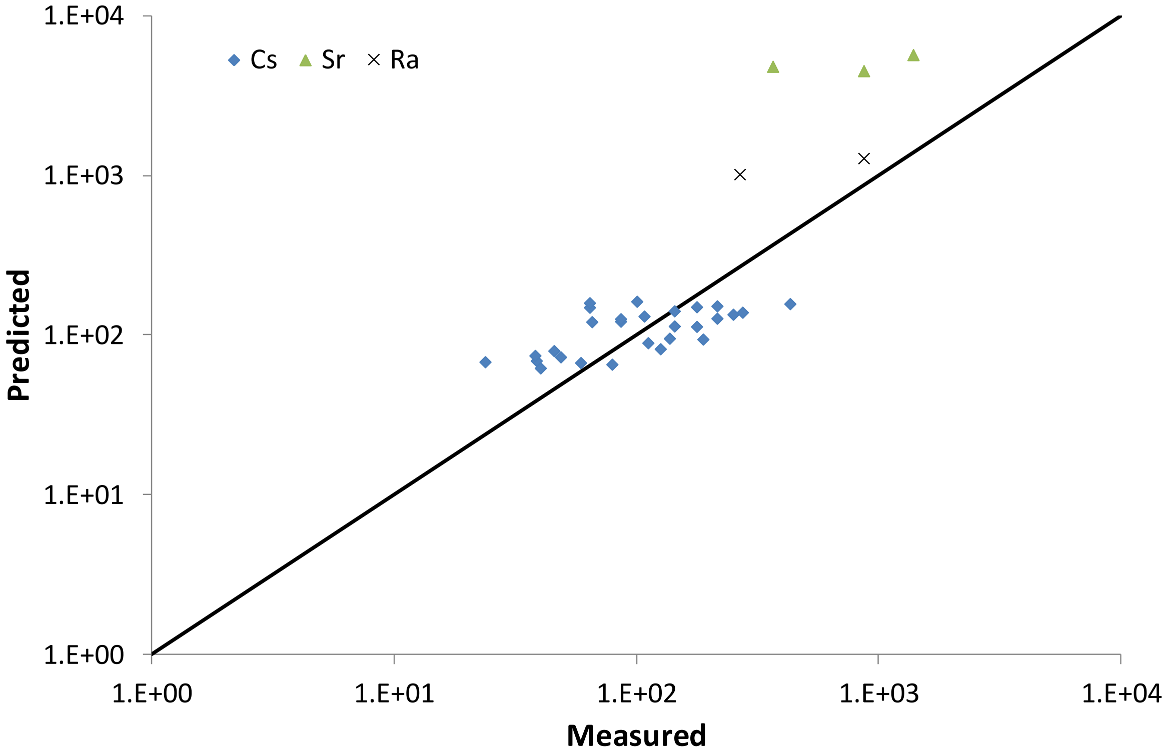

Figure 4.5. A comparison of measured radionuclide biological half-life (d) in reptiles with predictions using Equation 4.12 and reptile specific values of f_1 and $CR_{org\text{-}diet}$. The line is the 1:1 line.

Discussion

Using Equation 4.12 to predict $T_{B1/2}$ with constants derived from mammal data, relatively poor predictions were obtained (Figure 4.4). This demonstrates that existing models for homeothermic vertebrates (e.g. as presented in the RESRAD BIOTA model; USDOE 2004) should not be applied to reptiles.

Reptile-specific parameters for Equation 4.5 were relatively easy to derive, and using these, all predictions were within an order of magnitude of the measured values with the exception of the one prediction for Sr in *T. scripta scripta* for which the prediction was just over an order of magnitude higher than the measured value.

However, as apparent from Figure 4.6, exponents applicable to reptiles result in a relative low dependence of $T_{B1/2}$ with mass. Over a mass range 0.1 g to 1000 kg the predicted biological half-lives for reptiles vary by less than a factor of 4 using an exponent of 0.08 and by less than a factor of 2 using an exponent of 0.037. For homeotherms, $M^{0.25}$ predicts $T_{B1/2}$ values varying over *c*.60 fold.

Given the small influence of mass on $T_{B1/2}$ predictions for reptiles, we suggest that if sufficient reported $T_{B1/2}$ values are available then it is likely that these would be applicable to any reptile. This is demonstrated by the data for Cs used in this assessment. All of the 28 reported values of $T_{B1/2}$ for reptiles, covering a 50-fold mass range, presented in Table 1 are within a factor of 5 of the mean.

However, the relatively good agreement between predicted and measured $T_{B1/2}$ in Figure 4.6 demonstrates that if no reptile data are available for a given radionuclide then Equation 4.12 populated with reptile-specific parameter values will give reasonable estimates.

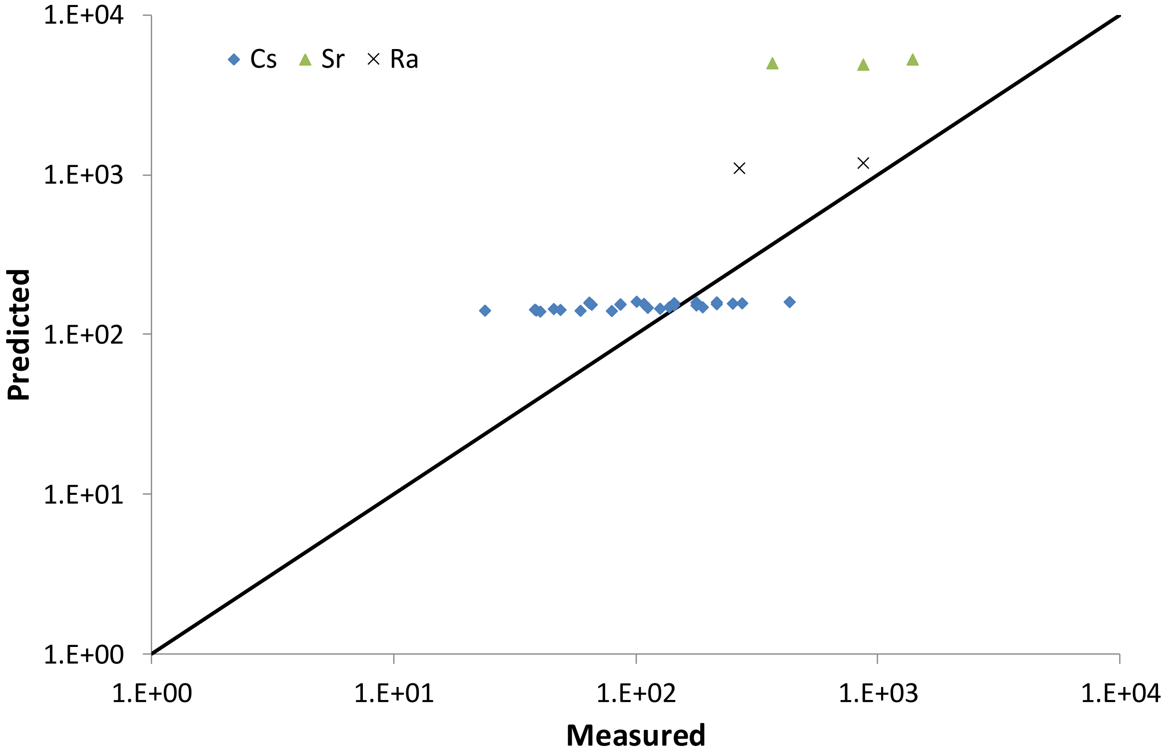

Figure 4.6. A comparison of measured radionuclide biological half-life in reptiles with predictions using reptile specific values for f_1, $CR_{org\text{-}diet}$ and the exponent in Equation 4.12. The line is the 1:1 line.

4.5 Allometry for aquatic organisms

We are not aware of any publications considering allometric radioecology models for organisms residing solely in the freshwater environment. Consequently this section will consider marine organisms only.

For marine mammals the allometric models developed by USDOE (2002) for terrestrial and riparian mammals and birds have been used within radioecological transfer models (e.g. Brown et al. 2004). However, as discussed above for reptiles, for most other aquatic organisms this would not be a valid approach.

Here we review allometric relationships for radionuclides in marine phyla (namely plankton, seaweed, fish, crustaceans and molluscs). This section draws upon the paper of Vives i Batlle et al. (2009) and considers the application of allometry to derive both biological half-life and CR (whole organism to water) values.

Available radioecological allometric parameters for marine organisms

Tables 4.5 presents allometric parameters for marine organisms for CR as estimated by Vives i Batlle et al. (2009). The organisms for which data were available were as follows: cockle, cod, crab, brown seaweed, lobster, mussel, phytoplankton, plaice, winkle and zooplankton, which were grouped into the categories of crustaceans, fish, mollusca, phytoplankton, vascular plant and zooplankton for the allometric analysis. A further 22 elements considered by Vives i Batlle et al. (2009) had no significant allometric relationship for CR. The majority of elements for which allometric relationships were identified were actinides or lanthanides.

Cherry & Heyraud (1991) present exponents (i.e. value of b) for allometric relationships for the CR of Po and Pb in marine organism (cited in Vives i Batlle et al. (2009). The exponents (-0.24 for Po and -0.22 for Pb) were similar to those derived for most elements by Vives i Batlle (2009).

Table 4.5. Allometric parameters for CR ($L\ kg^{-1}$) in marine organisms (adapted from Vives i Batlle et al. 2009); *a* and *b* are the constant and exponent for the allometric model (see Equation 4.1). The authors describe those relationships with R^2 values of <0.7 as ‘potentially approaching statistical significance’. For all elements n = 9.

	Pu	**Am**	**Ru**	**Ce**	**Pm/Eu**	**Ra**	**Th**	**Cm**	**Mn**	**Zr**	**Po**	**Ac**	**Pa**
a	209	251	29	468	1549	85	562	363	4365	269	5495	380	58
b	-0.30	-0.28	-0.46	-0.25	-0.18	-0.11	-0.27	-0.27	-0.13	-0.30	-0.17	-0.18	-0.17
R^2	0.80	0.78	0.75	0.72	0.72	0.79	0.89	0.85	0.46	0.68	0.64	0.58	0.53
p	0.001	0.002	0.002	0.004	0.004	0.001	0.0002	0.0004	0.04	0.007	0.01	0.02	0.03

Note: ^{154,155}Eu and ^{147}Pm were assumed to be biological analogues.

To parameterise the CR and $T_{B1/2}$ relationships derived by Vives i Batlle et al. (2009) (see Table 4.6) a dataset of biokinetic and transfer parameters for Tc, I, Cs, Pu and Am in fish, crustaceans, molluscs, macroalgae/seaweed and plankton (from Vives i Batlle et al. (2007; 2008) was established. The allometric relationships for I were poor ($R^2<0.24$, $p>0.19$), probably due to its complex speciation in the marine environment, and so they are not included in the table below.

Whilst Vives i Batlle et al. (2009) presented allometric $T_{B1/2}$ expressions for Sr and I from previous studies for background information these were from USDOE and hence for terrestrial animals and are not considered here.

Table 4.6. Allometric parameters describing the biological half-life (d) of radionuclides in marine organisms showing sufficiently good correlation (from Vives i Batlle et al. 2009).

	Tc	**Cs**	**Pu**	**Am**
a	98	54	631	251
b	0.15	0.17	0.20	0.13
R^2	0.72	0.92	0.91	0.76
p	0.03	0.04	0.2	0.05
n	6	4	3	5

Discussion

For CR the mean (±SD) value of b calculated across all those elements for which the relationship fitted had an R^2 value >0.7 was -0.26±0.09; that for the five elements in Table 4.5 with an R^2 <0.7 was -0.19±0.06.

Vives i Batlle et al. (2009) found a significant linear relationship between $\log_{10}$(a) and $\log_{10}$ (K_d) (i.e. the sediment-water distribution coefficient) (Figure 4.7) showing sediment-seeking radionuclides to have the highest log10(a) values. Only those elements for which the R^2 on the allometric relationship was in excess of 0.7 were considered (Table 4.5). The relationship between log10(a) and log(K_d) improved if Ru and Eu/Pm were removed. The authors also state that with the addition of Mn, Zr, Po, Ac and Pa the trend becomes weaker ($\log_{10}(a) = 0.38 \times \log_{10}(K_d) - 1.6$; $R^2 = 0.50$; n = 13) but that the statistical significance remained strong (p=0.007).

The available $T_{B1/2}$ data was less than that for CR and as a consequence the statistical significance of the allometric relationships was poorer than those for CR. The mean value of *b* for the $T_{B1/2}$ relationships was 0.16 ± 0.03.

For both CR and $T_{B1/2}$ therefore the exponent of the allometric relationship (i.e. b) approaches a quartile value as observed for many other allometeric relationships (see above). For CR, if we accept that the exponent will approximate to -0.25 for cations then the relationship between $\log_{10}$(a) and log(K_d) observed by Vives i Batlle et al. (2009) presents a potential opportunity to estimate CR values when data are lacking if K_d is known.

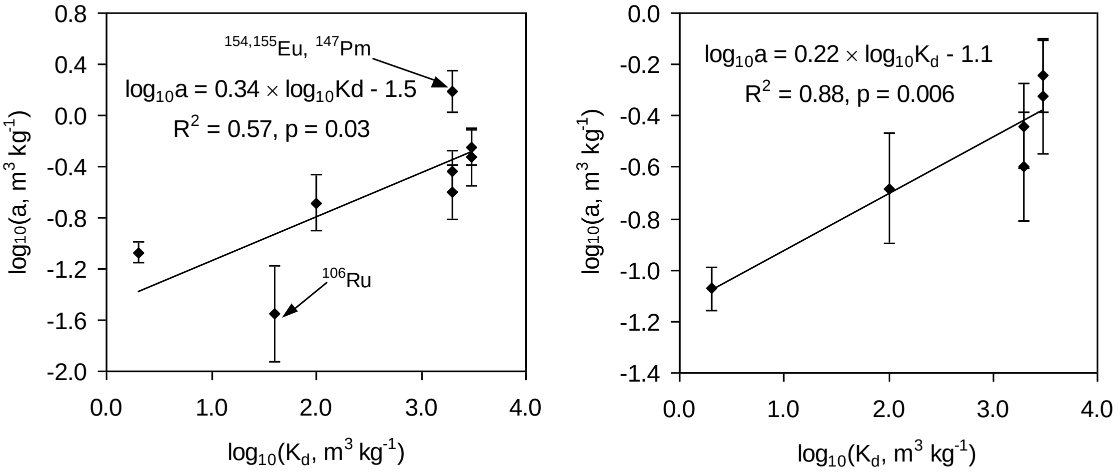

Figure 4.7. Linear trend between $\log_{10}a$ for the CR and $\log_{10}(Kd)$, both with (left) and without (right) Ru, Eu and Pm. Error bars represent the standard error of $\log_{10}(a)$ (reproduced from Vives i Batlle et al. (2009)). Note units of CR in the figure are $m^3 kg^{-1}$ as presented in the original publication.

An anomaly when considering the allometric relationship available for marine organisms with this derived for terrestrial organisms is that the actinide and lanthanide elements scale with an exponent similar to other elements for marine organisms whereas this is not the case for terrestrial organisms (see Table 4.1).

A potential reason for this with respect to CR for marine organisms is the relationship between $\log_{10}(K_d)$ and $\log_{10}(a)$. This infers that the more particle reactive the nuclide is, the more it attaches to organic matter (e.g. food). As the ingestion of food is related to metabolism and hence to body mass via Kleiber's law, the resultant CR will scale allometrically. Obviously this argument applies to animals but not to plants.

Pan & Wang (2008) have previously made similar suggestions with respect to metabolically driven allometry of the ingestion rate and the uptake of Cd and Zn by marine invertebrates. However, Vives i Batlle et al. (2009) suggest that for some elements, the process could also be surface-area driven (e.g. passive sorption of radionuclides on body surfaces, such as mollusc shells). For a perfectly isometrically scaling organism all surface area-based properties change with mass to the power of $^2/_3$ (Galileo, 1638) and this would result in the CR scaling to $M^{-0.33}$ instead of $M^{-0.25}$.

However, currently we cannot propose a reason for apparent differences in allometeric relationships for $T_{B1/2}$ between marine and terrestrial organisms.

5. An alternative to CR for wildlife

As noted in Chapter 1 there are many cases where empirical data to derive $CR_{wo\text{-}media}$ are lacking. Furthermore, the parameter is highly variable as it incorporates many processes and will generally be determined by site-specific characteristics (e.g. Beresford et al. 2008b; Johansen et al. 2012; Wood et al. 2013; Yankovich et al. 2010b).

Soil-to-plant transfer of elements of radiological interest has been related to plant evolutionary history, or phylogeny, for Cs (Broadley et al. 1999; Willey et al. 2005), Sr (Willey and Fawcett, 2005a), Ru (Willey and Fawcett, 2006), Cl (Willey and Fawcett, 2005b), Co (Willey and Wilkins, 2008) and U (Willey, 2010). Such phylogenetic relationships present a potential approach to enable predictions of transfer, with some scientific justification, for taxonomic groups for which there are no data either at the generic or site-specific level (Willey, 2010). The potential to derive phylogenetic relationships for organisms other than plants has been demonstrated by Jeffree et al. (2010; 2013) who suggested that the transfer of a number of radionuclides to marine teleost and chondrichthyan fishes and the amphioxus (fish like chordate) species *Branchiostoma lanceolatumis* is influenced by phylogeny. However, the work of Jeffree et al. was based upon the results of laboratory studies. Whilst this usefully removes the influences of many confounding factors it is not directly applicable to environmental conditions as foodchain transfer was excluded.

Here we begin with exploring if phylogeny can be used to explain variation in the transfer in the environment using data for radiocaesium and freshwater fish species as an example. The work described in the chapter has been published as Beresford et al. (2013).

5.1 Materials and Methods

Data sources

The primary source of data for the analyses was the database on radionuclide transfer to freshwater organisms as described by Yankovich et al. (2013) (see also Copplestone et al. 2013).

The database as described by Yankovich et al. (2013)[4] contains 535 $CR_{wo\text{-}water}$ entries for the transfer of caesium to freshwater fish; some entries are mean values and other single data points. The data set includes $CR_{wo\text{-}water}$ based on both radiocaesium and stable caesium values. The $CR_{wo\text{-}water}$ values are categorised by species, feeding strategy (benthic, predatory or forage) and freshwater ecosystems type ('lake' or 'flowing water'). Some of these data were excluded from this analysis as no species information was recorded (e.g. the source reference specified 'freshwater fish' only). The Residual Maximum Likelihood (REML) analysis used in this chapter (see below) requires that, for each study site, data are available for more than one species and that at least one of these species must occur at another site. Excluding data which did not meet these criteria left a total of 248 entries. As we were using the REML model, it was possible to supplement the $CR_{wo\text{-}water}$ values with data from studies

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[4] The database has subsequently been updated see Beresford et al. (2014).

reporting Cs concentrations in fish; these additional data had not been used by Yankovich et al. (2013) as corresponding water concentrations were not available and hence $CR_{wo\text{-}water}$ values could not be calculated. Concentration data had to adhere to the same requirements as the $CR_{wo\text{-}water}$ values to be included in this analysis. An additional 349 data entries reporting activity concentrations which met these criteria were identified (Copeland and Ayers 1972; Copeland et al. 1973; Smith et al. 2003; Andersson pers. comm.[5]). In total, 597 entries were available for 53 freshwater fish species from 67 sites; note that whilst in most instances sites were identified in the source references, in a few cases it was necessary to assume that all the data in a given reference came from one site (these entries represented <10 % of the total dataset). Table 5.1 presents a summary of the available data.

The taxonomy of each species for which data were available was determined with reference to Nelson (2006) and Froese and Pauly (2012). The 53 species for which there were data all belonged to the class *Actinopterygii* (ray-finned fishes) with 10 orders, 14 families and 33 genera being represented in the dataset (Table 5.1). Prior to analyses, orders, families and genera were numerically coded based on the phylogenetic tree presented by Nelson (2006) (Figure 5.1), with approximate timescales for the evolutionary divergence for each order being identified from http://www.timetree.net. The 'oldest' order was defined as '1' and the most recent as '10' (data being available for a total of ten orders) (Figure 5.1). Where orders diverged at the same time (e.g. *Osmeriformes*, *Salmoniformes* and *Esociformes*) the order numbering is simply from left to right on Figure 5.1 and does not reflect differences in evolutionary age. To put some context to the order numbers, the clade containing *Lepisosteiformes* and *Amiiformes* diverged from the other orders considered here >300 million years ago whereas the clade containing *Perciformes* diverged from that containing *Cyprinodontiformes* around 100 millon years ago (see Figure 5.1). Each species was given a 'taxon number' starting with species in the oldest orders, so for the available dataset *Lepisosteus osseus* was defined as *taxon 1* (see Table 5.1).

Table 5.1. Summary of data available for REML analysis to investigate any phylogentic influence on the transfer of Cs to freshwater fish.

Order	Family	Genus	Species	Sites+	Taxon number
Lepisosteiformes	*Lepisosteidae*	*Lepisosteus*	*osseus*	1	1
Amiiformes	*Amiidae*	*Amia*	*calva*	1	2
Anguilliformes	*Anguillidae*	*Anguilla*	*anguilla*	5	3
Clupeiformes	*Clupeidae*	*Dorosoma*	*cepedianum*	1	4
Clupeiformes	*Clupeidae*	*Alosa*	*pseudoharengus*	10	5
Osmeriformes	*Osmeridae*	*Osmerus*	*mordax*	10	6

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[5] Swedish Radiation Safety Authority see: http://www.stralsakerhetsmyndigheten.se/Yrkesverksam/Miljoovervakning/Sokbara-miljodata/ for information on monitoring programme from which data were obtained.

Order	**Family**	**Genus**	**Species**	**Sites+**	**Taxon number**
Salmoniformes	*Salmonidae*	*Coregonus*	*clupeaformis*	2	7
Salmoniformes	*Salmonidae*	*Coregonus*	*hoyi*	2	8
Salmoniformes	*Salmonidae*	*Coregonus*	*artedi*	2	9
Salmoniformes	*Salmonidae*	*Coregonus*	*spp.*	6	10
Salmoniformes	*Salmonidae*	*Oncorhynchus*	*kisutch*	5	11
Salmoniformes	*Salmonidae*	*Oncorhynchus*	*mykiss*	4	12
Salmoniformes	*Salmonidae*	*Oncorhynchus*	*tschawytscha*	3	13
Salmoniformes	*Salmonidae*	*Salmo*	*trutta*	11	14
Salmoniformes	*Salmonidae*	*Salvelinus*	*alpinus*	1	15
Salmoniformes	*Salmonidae*	*Salvelinus*	*fontinalis x namaycush*	1	16
Salmoniformes	*Salmonidae*	*Salvelinus*	*namaycush*	8	17
Salmoniformes	*Salmonidae*	*Salvelinus*	*siscowet*	1	18
Salmoniformes	*Salmonidae*	*Stenodus*	*leucichthys*	1	19
Esociformes	*Esocidae*	*Esox*	*lucius*	38	20
Esociformes	*Esocidae*	*Esox*	*niger*	1	21
Cypriniformes	*Catostomidae*	*Catostomus*	*catostomus*	2	22
Cypriniformes	*Catostomidae*	*Catostomus*	*commersoni*	1	23
Cypriniformes	*Catostomidae*	*Moxostoma*	*aureolum*	1	24
Cypriniformes	*Cyprinidae*	*Abramis*	*brama*	7	25
Cypriniformes	*Cyprinidae*	*Carassius*	*auratus*	3	26
Cypriniformes	*Cyprinidae*	*Carassius*	*carassius*	2	27
Cypriniformes	*Cyprinidae*	*Cyprinus*	*carpio*	3	28
Cypriniformes	*Cyprinidae*	*Notemigonus*	*crysoleucas*	1	29
Cypriniformes	*Cyprinidae*	*Rutilus*	*rutilus*	5	30
Cypriniformes	*Cyprinidae*	*Scardinius*	*erythrophthalmus*	3	31
Cypriniformes	*Cyprinidae*	*Notropis*	*hudsonius*	7	32
Cypriniformes	*Cyprinidae*	*Gobio*	*gobio*	1	33
Cypriniformes	*Cyprinidae*	*Tinca*	*tinca*	7	34
Siluriformes	*Ictaluridae*	*Ictalurus*	*punctatus*	2	35
Siluriformes	*Ictaluridae*	*Ictalurus*	*spp.*	1	36
Perciformes	*Centrarchidae*	*Ambloplites*	*rupestris*	2	37
Perciformes	*Centrarchidae*	*Lepomis*	*gulosus*	1	38
Perciformes	*Centrarchidae*	*Lepomis*	*macrochirus*	3	39
Perciformes	*Centrarchidae*	*Lepomis*	*gibbosus*	1	40
Perciformes	*Centrarchidae*	*Lepomis*	*microlophus*	1	41
Perciformes	*Centrarchidae*	*Micropterus*	*dolomieu*	6	42
Perciformes	*Centrarchidae*	*Micropterus*	*salmoides*	5	43

Order	Family	Genus	Species	Sites+	Taxon number
Perciformes	*Centrarchidae*	*Pomoxis*	*annularis*	1	44
Perciformes	*Centrarchidae*	*Pomoxis*	*nigromaculatus*	1	45
Perciformes	*Moronidae*	*Morone*	*chrysops*	4	46
Perciformes	*Percidae*	*Perca*	*flavescens*	8	47
Perciformes	*Percidae*	*Perca*	*fluviatilis*	28	48
Perciformes	*Percidae*	*Sander*	*lucioperca*	3	49
Perciformes	*Percidae*	*Sander*	*canadensis*	1	50
Perciformes	*Percidae*	*Sander*	*vitreus*	9	51
Perciformes	*Percidae*	*Gymnocephalus*	*cernuus*	1	52
Perciformes	*Sciaenidae*	*Aplodintus*	*grunniens*	1	53

[+]Number of sites species present at.

Data analyses

The Residual Maximum Likelihood (REML) fitting of a mixed-model regression as described by Willey (2010) and originally developed by Broadley et al. (1999, 2001) was used to analyse the data for any phylogenetic influence on Cs transfer. This technique enables the collation of data from different sources and the prediction of values that might be gained if they were all generated under an average set of conditions. The output consists of a mean value for each species on a common scale after REML adjustment (the fixed factor) taking account of the effect of the random factor (i.e. inter-site variation). This provides a method for statistically accounting for as much of the effect of site as possible within the collated data. The mean value output for each species provides a relative scaling value which it had been suggested could subsequently be used to infer *CR* values, or concentrations, from a known value for a given species or a group mean (Willey, 2010) (or indeed site specific activity concentrations if data are available for one species (see below)).

The REML procedure fits the model such that values for each species are made as nearly identical as possible across the studies. Consequently, we were able to include both $CR_{wo\text{-}water}$ values from Yankovich et al. (2013) and concentration data where the criteria specified above were met (i.e. we assumed that the relative difference between Cs concentrations between species at a site will be the same as the relative difference between Cs $CR_{wo\text{-}water}$ values at a site). The REML procedure minimises, as far as possible, variation due to factors such as water chemistry or study methodology (e.g. $CR_{wo\text{-}water}$ values may in some references be related to unfiltered water and in others to filtered water) by treating the 'site' as a random factor.

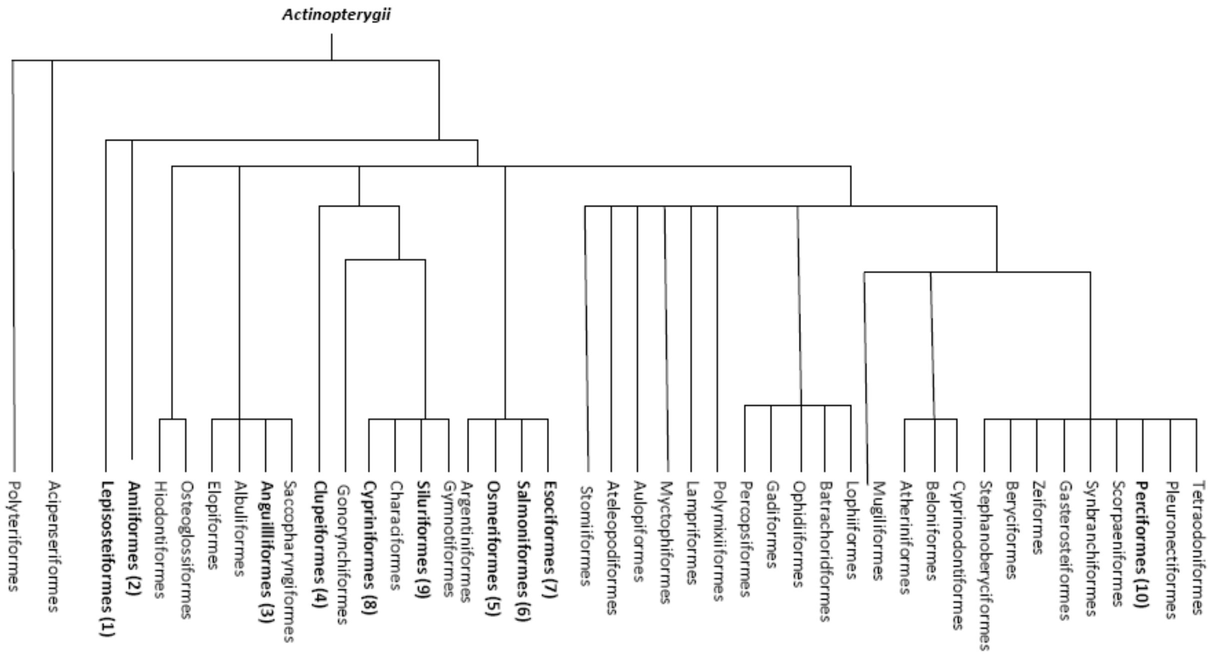

Figure 5.1. Sequence of orders within the class *Actinopterygii* (ray-finned fishes) adapted from Nelson (2006). Orders for which data are available are identified in **bold**; the number in parenthesis denotes that given to the order based upon evolutionary time for statistical analyses.

The REML analysis and associated analysis of variance (ANOVA) was conducted on log-transformed data by adapting the Genstat (http://www.vsni.co.uk) code as presented in Willey (2010) (Beresford et al. (2013), presents the revised Genstat code for the overall REML analysis and hierarchical ANOVA). In addition to outputting REML-adjusted means by species, REML-adjusted means were also estimated at the level of order, family and genus. To determine significant differences between specific taxonomic groupings the standard error of difference was estimated in a pair wise manner for all REML-adjusted means. The *t*-statistic was then calculated as the ratio of the difference between mean pairs and the associated standard error of difference. All other analyses were conducted using the General Linear Model option from the Minitab statistical package (http://www.minitab.com) or linear regression from Microsoft Excel.

5.2 Results and discussion

When all data were considered at the species level, the REML variance component analysis gave a significant (p<0.001) Wald statistic of 116.

The Wald statistic for the analysis at the levels of order, family and genus were 51 (p<0.001), 54 (p<0.001) and 107 (p<0.001) respectively, also indicating significant data fits (Thompson and Welham 2001). Significant variation in REML estimated mean values was, however, explained by hierarchical ANOVA at the order level (ANOVA; $p<0.001$), with little additional variation explained by the effects of family within order or genus within family. REML-adjusted mean values are presented in Table 5.2 for the four different taxonomic levels considered. For each taxonomic level these values should be regarded as relative numbers and not actual estimates of $CR_{wo\text{-}water}$ (see below for examples of application).

From the estimated *t*-statistics *Perciformes* had a significantly higher REML-adjusted mean value than *Anguilliformes*, *Clupeiformes*, *Osmeriformes*, *Salmoniformes* and *Cypriniformes* ($p<0.05$). *Esociformes* also had a significantly higher adjusted mean value than *Anguilliformes*, *Clupeiformes*, *Salmoniformes* and *Cypriniformes* ($p<0.05$). *Anguilliformes* had a significantly lower ($p<0.05$) adjusted mean than all other orders with which statistical comparisons could be made. *Lepisosteiformes*, *Arniiformes* and *Siluriformes* were not considered in statistical tests as they were present at three or fewer sites only. Significant differences, when tested at more refined taxonomic levels, were generally in agreement with those observed at the order level. For instance, at the level of family *Anguillidae* had a significantly lower REML-adjusted mean than *Osmeridae*, *Salmonidae*, *Esocidae*, *Centrarchidae*, *Moronidae*, *Clupeidae*, *Cypinidae* and *Percidae* ($p<0.05$). Similarly, both *Escocidae* and *Percidae* had a significantly higher REML-adjusted means than *Salmonidae*, *Clupeidae* and *Cypinidae* ($p<0.05$). Comparatively few of the potential comparisons at genus and species level could be shown to be significant due to low data availability for some species. Where significant differences were observed ($p<0.05$), these generally involved comparisons which included *Anguilla*, *Esox*, *Perca* or *Sander* species.

Table 5.2. REML-adjusted means for different taxonomic groups. Note these are relative values and not absolute values of $CR_{wo\text{-}water}$.

Order		**Family**		**Genus**		**Species**	
Lepisosteiformes	6.8	*Lepisosteidae*	6.9	*Lepisosteus*	*7.6*	*osseus*	7.4
Amiiformes	3.2	*Amiidae*	2.9	*Amia*	*3.6*	*calva*	3.0
Anguilliformes	1.8	*Anguillidae*	1.9	*Anguilla*	*1.9*	*anguilla*	2.1
Clupeiformes	5.0	*Clupeidae*	4.9	*Dorosoma*	*4.7*	*cepedianum*	4.3
				Alosa	*4.0*	*pseudoharengus*	3.7
Osmeriformes	5.6	*Osmeridae*	5.5	*Osmerus*	*4.5*	*mordax*	4.2
Salmoniformes	5.5	*Salmonidae**	5.5	*Coregonus*	*3.8*	*clupeaformis*	3.9
						hoyi	5.0
						artedi	3.5
						spp.	3.2
				Oncorhynchus	*8.1*	*kisutch*	9.3

Order		Family		Genus		Species	
						mykiss	6.5
						tschawytscha	8.9
				Salmo	*6.0*	*trutta*	5.8
				Salvelinus	*7.8*	*alpinus*	7.9
						fontinalis x namaycush	5.6
						namaycush	8.0
						siscowet	10.8
				Stenodus	*5.4*	*leucichthys*	5.6
Esociformes	8.1	*Esocidae*	8.3	*Esox*	*8.5*	*lucius*	8.8
						niger	3.0
Cypriniformes	4.6	*Catostomidae*	5.3	*Catostomus*	*5.0*	*catostomus*	4.5
						commersoni	4.4
				Moxostoma	*4.7*	*aureolum*	4.2
		Cyprinidae	4.5	*Abramis*	*4.5*	*brama*	4.8
				Carassius	*4.4*	*auratus*	4.3
						carassius	4.9
				Cyprinus	*1.2*	*carpio*	1.2
					6.2	*crysoleucas*	5.7
				Rutilus	*4.8*	*rutilus*	5.0
				Scardinius	*4.4*	*erythrophthaimus*	4.7
				Notropis	*3.9*	*hudsonius*	3.6
				Gobio	*6.2*	*gobio*	6.5
				Tinca	*3.1*	*tinca*	3.2
Siluriformes	7.6	*Ictaluridae*	7.6	*Ictalurus*	*6.2*	*punctatus*	5.7
						spp.	5.1
Perciformes	8.6	*Centrarchidae*	7.0	*Ambloplites*	*14.2*	*rupestris*	13.8
				Lepomis	*4.1*	*gulosus*	5.3

Order		Family		Genus		Species	
						macrochirus	3.7
						gibbosus	3.7
						microlophus	2.9
				Micropterus	*9.6*	*dolomieui*	8.7
						salmoides	8.5
				Pomoxis	*8.3*	*annularis*	7.3
						nigromaculatus	9.3
		Moronidae	7.5	*Morone*	*9.1*	*chrysops*	8.8
		Percidae	9.0	*Perca*	*9.4*	*flavescens*	7.3
						fluviatilis	10.5
				Sander	*10.0*	*lucioperca*	7.8
						canadensis	12.3
						vitreus	11.8
				Gymnocephalus	*1.9*	*cernuus*	2.0
		Sciaenidae	15.9	*Aplodinotus*	*11.2*	*grunniens*	10.3

*The ICRP Reference Trout (the freshwaster fish RAP) is defined as the *Salmonidae* family.

The results of these analyses, therefore, demonstrate differences in Cs transfer to freshwater fish based upon phylogenetically derived taxonomic groupings. Does this then mean that we have demonstrated an evolutionary, or phylogenetic, relationship for the Cs transfer to different freshwater species? On the basis of the data included in our analyses presented here we cannot establish this. For instance, evolutionarily *Lepisosteiformes* are most closely related to *Amiiformes* yet the REML-adjusted means for the two orders differ by a factor of >2 which is more than the difference between *Lepisosteiformes* and *Perciformes*, the most distantly related orders for which the REML-adjusted means are within *c*. 20% of each other). Similarly, whilst the REML-adjusted means for *Salmoniformes* (5.5) and *Osmeriformes* (5.6) are similar, they are considerably lower than that for the order *Esociformes* (8.1) which is in the same clade. Our inability to conclude a ‘phylogenetic effect’ on Cs transfer to freshwater fish is likely due, in part, to the relatively few species and taxonomic groups for which we had data. Whilst we had a relatively large dataset to consider, data were only available for 53 of the total 11952 freshwater species (Nelson, 2006), representing only 10 orders and one class. Earlier analyses which have suggested phylogentic relationships for the transfer of radionuclides to plants (Willey, 2010) and marine fish (Jeffree et al. 2010; 2013) have included species encompassing much wider evolutionary time scales (e.g. >500 million years in the case of marine fish).

Fish within a given taxa are likely to share many characteristics such as feeding strategy. Caesium transfer to piscivorous fish (feeding primarily on smaller fish, but also amphibians, mammals and birds) has been shown to be higher than that to fish with other feeding strategies (e.g. Kryshev 1995, Kryshev et al. 1993; Rowan et al. 1998; Rowan and Rasmussen, 1994; Saxén and Ilus, 2008; Smith et al. 2000; Beresford et al. 2013). The outputs of the REML analysis are in agreement with these observations.

The need for an alternative to the $CR_{wo\text{-}media}$ model

Typically, $CR_{wo\text{-}water}$ values for freshwater fish and other aquatic organisms vary over orders of magnitude, as do $CR_{wo\text{-}media}$ values for organisms in other ecosystem types (e.g. Beresford et al. 2008b; Howard et al. 2013; Wood et al. 2013). This is demonstrated for freshwater fish in Table 5.3 which presents a summary of Cs $CR_{wo\text{-}water}$ values from the compilation of Yankovich et al. (2013) as will be used in a forthcoming IAEA handbook of transfer parameters for wildlife (see Howard et al. 2013). In large part, this variability is due to site factors which influence radionuclide transfer. In the case of Cs and freshwater fish a key site specific factor is the K concentration in water. For instance, Smith et al. (2000) demonstrate approximately two-orders of magnitude variation in $CR_{wo\text{-}water}$ explained by water K concentration. Water pH and Ca concentration have also been suggested to influence Cs transfer (Smith et al. 2002). Consequently, there is often large variation between the outputs of models using $\mathrm{CR_{wo}}$ values to predict activity concentrations in wildlife (Beresford et al. 2008b; Yankovich et al. 2010b; Johansen et al. 2012) and the approach is open to criticism as being too simplistic (ICRP 2009). However, pragmatically the $\mathrm{CR_{wo}}$ approach is easy to apply and has the most comprehensive datasets available, and hence it continues to be recommended in international compilations (e.g. IAEA 2014; ICRP, 2009).

Table 5.3. Summary of Cs $\mathrm{CR_{wo\text{-}water}}$ values for fish by feeding groups (Yankovich et al. 2013).

Feeding group	Arithmetic mean ±SD	Minimum	Maximum	N
Benthic feeding	(1.0±2.0)E+3	1.8E+1	2.0E+4	156
Forage feeding	(9.2±16)E+2	1.7E+1	8.6E+3	125
Piscivorous	(4.5±6)E+3	1.3E+1	8.2E+4	439

Wood et al. (2013) recommend that as a consequence of the uncertainty in $CR_{wo\text{-}media}$ data, summarised $CR_{wo\text{-}media}$ values are used with caution above the initial, highly conservative, screening-level assessments. This is consistent with the recommendation that site-specific data should be used for higher tier assessments (e.g. Brown et al. 2008; USDOE, 2002).

The REML-adjusted means presented in Table 5.2 potentially provide a more refined approach than the $CR_{wo\text{-}media}$ model. By taking into account inter-site variation, they in effect provide a mechanism of accounting for site specific variables such as the K concentrations in water in the case of Cs transfer to fish as being considered here. Comparison of Tables 5.2 and 5.3 suggests that the variation in the transfer of Cs to fishes between studies/sites (two to three orders of magnitude in Table 5.3) is considerably greater than the likely variation between taxonomic groups at a given site (*circa* one order of magnitude or less in Table 5.2).

5.3 Testing the REML outputs

We propose the hypothesis that *the REML model outputs can be used to predict the radionuclide, in this case radiocaesium, activity concentrations in unknown species from the results of a species which has been sampled at a specific site*. To test this hypothesis we required data which had not been included in the already comprehensive compilation needed to conduct the analysis described above. A large monitoring programme of fish from 590 Finnish lakes has been conducted by STUK since the 1986 Chernobyl accident. Data from this programme have recently been made available via STAR (http://bit.ly/1xDJQu4). These data were not used within the analysis we have described above to establish the REML model and hence provide an opportunity to independently test our hypothesis. The STUK monitoring programme is in part described by Saxén & Koskelainen (2005), Saxén (2007), Saxén & Ilus (2008), Vetikko & Saxén (2010), with a meta data record available on-line (STUK, 2012).

We have selected data from 1988, which were collected from 27 Finnish lakes for which ^{137}Cs activity concentrations were available for four or more fish species. In total data were available for 11 fish species: *A. brama*, *E. lucius*, *P. fluviatilis*, *R. rutilus*, *S. trutta*, *S. lucioperca*, *Coregonus albula*, *Coregonus lavaretus*, *Blicca bjoerkna*, *Leuciscus idus*, *Abramis ballerus* and *Lota lota*. As *P. fluviatilis* was present at all 27 sites and was also well represented within the dataset used for the REML analysis, we selected it as our 'known species' from which to calculate activity concentrations for the other species (treated as unknowns). To calculate the ^{137}Cs activity concentrations, the ratios of the REML-adjusted mean (Table 5.2) for each unknown species to that of *P. fluviatilis* was estimated (e.g. for *R. rutilus* the ratio was 0.48). For each lake the ^{137}Cs activity concentration in different species were then estimated as the product of this ratio and the geometric mean ^{137}Cs activity concentration in *P. fluviatilis* at that site. This approach was possible for *A. brama, E. lucius, R. rutilus, S. trutta* and *S. lucioperca* all of which were in the dataset used for the REML analysis. Although some *Coregonus* species were present within our initial dataset, the two species sampled in Finnish lakes were not. Therefore, the ratio of the REML-adjusted mean for the genus *Coregonus* to that for *Perca* was used to estimate ^{137}Cs activity concentrations in both species; a similar genus based approach was used for *A. ballerus*. No data for the genus *Blicca* or *Leuciscus* were available for our REML analysis. Therefore, as both species are Cypinidae the ratio of the REML-adjusted mean for this family to that of Percidae was estimated and used to predict ^{137}Cs activity concentrations in both *B. bjoerkna* and *L. idus*. No predictions were possible for *L. lota* as it is a Gadiforme and no representatives of this order were present in the database used to establish the REML model. In total this allowed predictions for 100 fish samples across the 27 lakes.

A comparison of predicted ^{137}Cs activity concentrations with measured values is presented in Figure 5.2. There was relatively good agreement between predicted and measured values with a linear regression fit to all 100 data points yielding an R^2 of 0.83 ($p<0.001$) and a slope (±standard error) of 0.98±0.04 ($p<0.001$). The intercept was not significantly different to zero for this or any of the subsequent regressions discussed. Linear regressions were also fitted individually for *A. brama*, *E. lucius, and R. rutilus,* with these three Salmonidae being considered together given there were few observations for them. All regressions yielded R^2 values close to 0.8 (0.76 – 0.84) ($p<0.001$). Slopes (±standard error) were: *A. brama* (0.96±0.12), *E. lucius* (0.81±0.06), *R. rutilus* (1.32±0.12), *S. lucioperca* (0.69±0.07) and Salmonidae species (0.87±0.08) thus suggesting a tendency to under-predict for some species (notably *E. lucius* and *S. lucioperca*) and over-predict for *R. rutilus*.

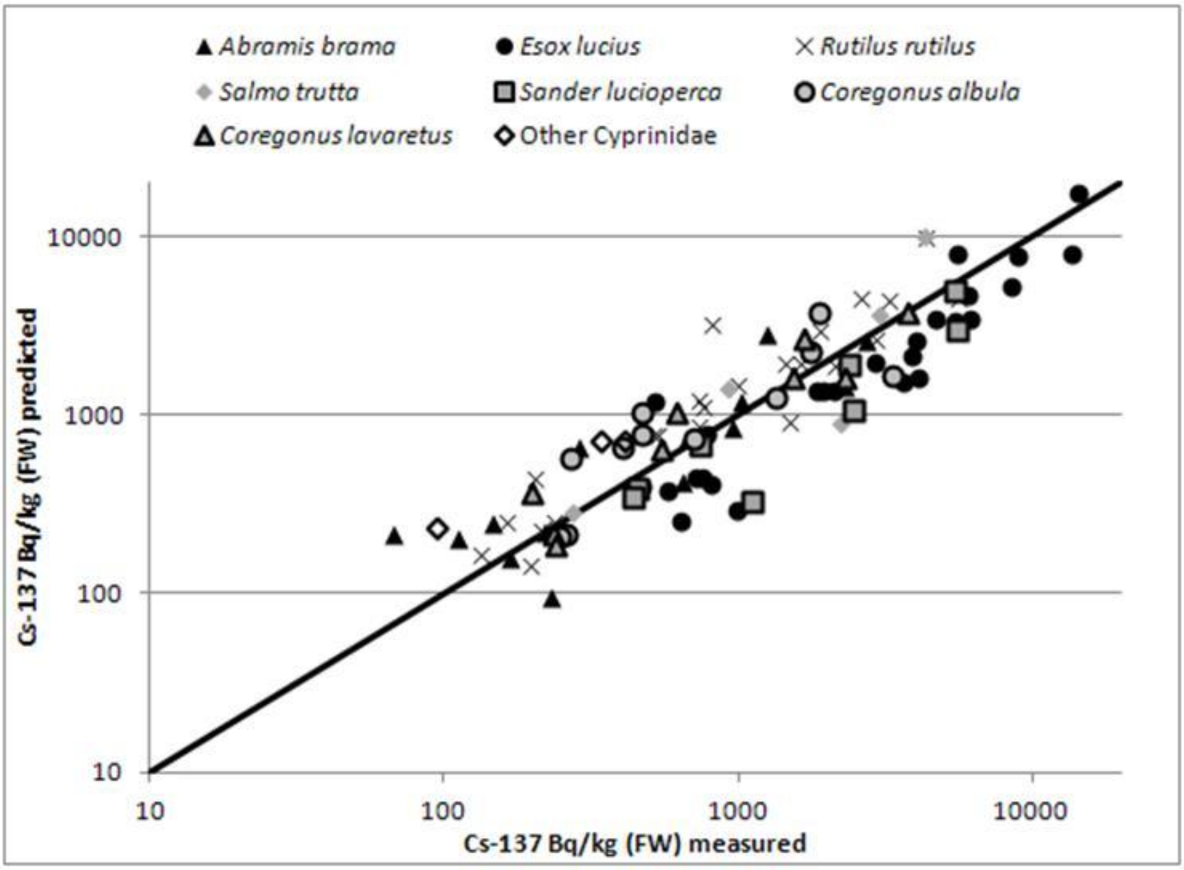

Figure 5.2. Comparison of measured ^{137}Cs activity concentrations in fish collected from 27[6] Finnish lakes in 1988 with predicted activity concentrations using the outputs of the REML analyses and data for *Perca fluviatilis* (line is 1:1 relationship). ‘Other Cyprinidae’ represents single values for *Blicca bjoerkna*, *Leuciscus idus* and *Abramis ballerus*.

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[6] Note the number of lakes quoted on the legend to this figure in Beresford et al. (2013) is in error, value shown here is correct (note data from only 26 lakes are used in Figure 5.3).

The results of this comparison look promising. There is obviously some scope for the results obtained to be influenced by the selection of *P. fluviatilis* as our known species. For instance, *E. lucius* was present in the sample from 26 of the lakes, selecting this species as our known species we could make predictions for 96 fish samples. Whilst all predicted values were within a factor of *c*. 5 of the measured data there was a tendency towards over-prediction (Figure 5.3).

For comparison with our results, if the appropriate feeding group geometric mean $CR_{wo\text{-}water}$ values from Yankovich et al. (2013) are used to predict the ^{137}Cs activity concentrations in fish from the Finnish lakes, there is a general under-prediction with a regression of predicted to measured activity concentrations yielding a slope of only 0.31.

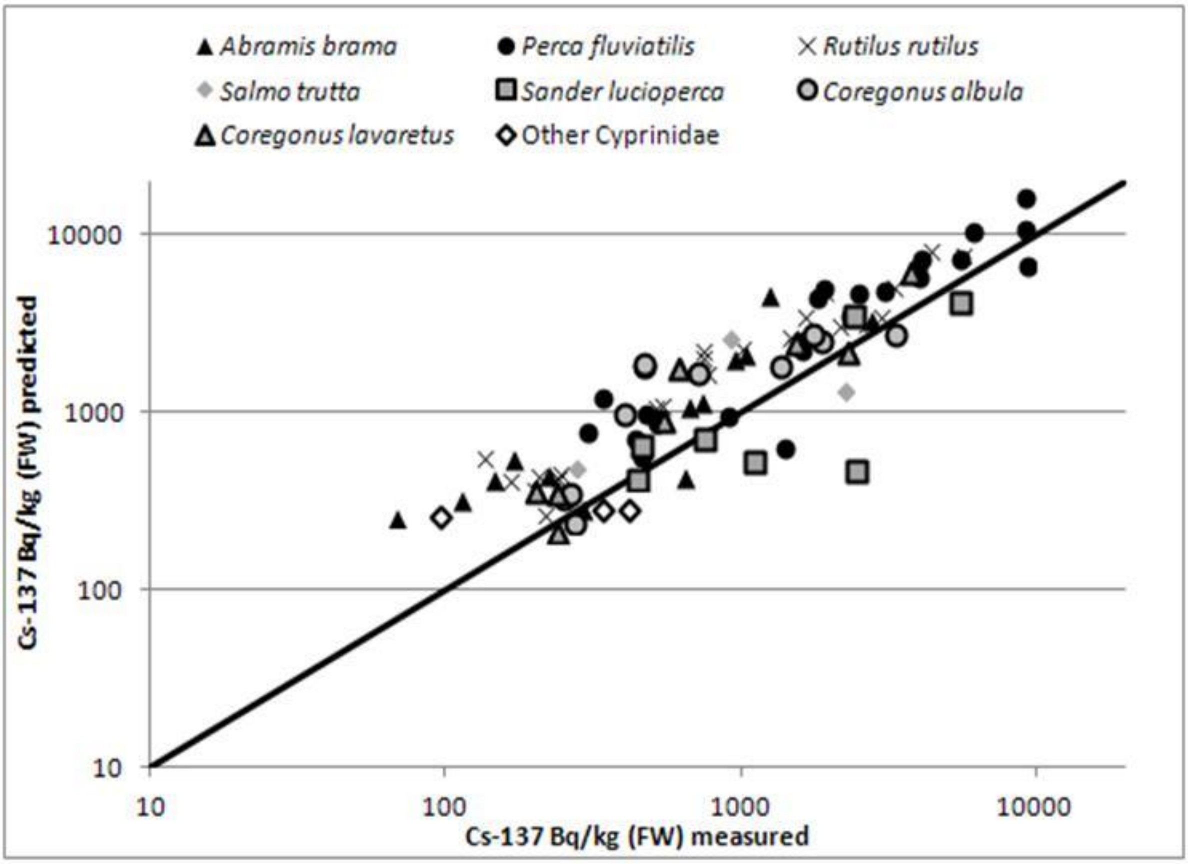

Figure 5.3. Comparison of measured ^{137}Cs activity concentrations in fish collected from 26 Finnish lakes in 1988 with predicted activity concentrations using the outputs of the REML analyses and data for *Esox lucius* (line is 1:1 relationship). ‘Other Cyprinidae’ represents single values for *Blicca bjoerkna*, *Leuciscus idus* and *Abramis ballerus*.

Discussion

The approach tested here should account for site-specific factors, and as demonstrated, appears to work relatively well. However, we should acknowledge some limitations in the available data which may influence the resultant REML adjusted means. There is geographical bias in available data and, therefore, site-specific variability may not be adequately compensated for in some species. For instance, whilst *A. anguilla* data are available from 5 sites (Table 5.1), these were all within Sweden and hence the sites could be relatively similar on a global scale. Similarly, limitations in sample size for the Finnish lakes dataset, which ranged from 1 to >400 fish, may have impacted upon some comparisons of predicted and observed activity concentrations.

Given the variation in biological half-lives across different organisms, the REML approach is unlikely to be applicable in situations where activity concentrations in the environment are rapidly changing. By 1988, water activity concentrations in Finnish lakes as a consequence of deposition from the Chernobyl accident were not changing rapidly; Saxén (2007) reports ecological half-lives for ^{137}Cs in Finnish lake waters of *circa* 1 to 5 years at this time. However, we acknowledge that a lack of equilibrium may have influenced the comparison of our predicted activity concentrations in fish with measured values.

6. Use of (ecological) stoichiometry as an extrapolation method

6.1 What is (ecological) stoichiometry?

Stoichiometry is '*a branch of chemistry that deals with the application of the laws of definite proportions and of the conservation of mass and energy to chemical activity*' (Mirriam-Webster online dictionary; http://www.merriam-webster.com/). This basic concept has been applied in the fields of biology and ecology in what is called ecological stoichiometry, '*the study of the balance of chemical elements in components, interactions, and processes in ecosystems*' (Sterner & Elser 2002).

6.2 Overview of history and examples of current uses of (ecological) stoichiometry

Earlier studies mostly used ecological stoichiometry to investigate food web dynamics (for example, Lindeman 1942; Lotka 1925) and carbon cycling and energy flows (Odum 1959, 1960). Redfield (1958) recognised that the elemental composition of seawater and plankton was fairly constant across wide oceanographic areas (C:N:P 106:16:1 on a molecular mass basis – the '*Redfield ratio*') and that biological processes could be controlled by elements other than C, such as N and P and trace elements.

Since these early works, the majority of ecological stoichiometry studies have continued to focus on C,N and P and primary producers, and how imbalances or mismatches in these element ratios between abiotic/biotic components and between organisms drive ecological processes through element limitation. Most studies have looked at trophic interactions and carbon (energy) and element cycling. More recently, ecological stoichiometry has been put forward as a driving force in evolutionary processes and as a way to integrate ecological dynamics with cellular and genetic mechanisms (Elser et al. 2000, Sterner & Elser 2002).

Elements other than CNP have sometimes been used in the natural sciences to trace the origins of organisms or materials in the environment. For example:

- inter-element (Al/Fe/Ti, P/Ca/Si, Ca/Al, Si/Al) relationships in suspended particulate matter and sediments have been used to distinguish the flux of primary settling matter from resuspended sediments and the extent of riverine inputs (Price et al. 2005).
- Murphy et al. (2008) used element ratios to trace the ocean source of ballast water. They found that the relatively conservative elements Mo, U and V provided little additional information beyond that obtained from salinity, whereas non-conservative Ba, P and Mn offered greater resolution.
- the natal origin of insect pests have been determined using element ratios in combination with isotopic techniques (δ^2H, ^{87}Sr/^{86}Sr, ^{207}Pb/^{206}Pb and ^{208}Pb/^{206}Pb) (Holder et al. 2014). These authors found that the elements of atomic number ≥ Rb were most informative, with Sr, Cs, Ba and Pb, as well as the Pb/Sr elemental ratio giving the best regional separation.

- populations of birds have been distinguished using trace element profiles (Kaimal et al. 2009, Norris et al. 2007).
- across 20 species of fish, whole fish Ca:P ratios were found to have a nearly constant stoichiometry consistent with the chemical signature of bone. Hendrixson et al. (2007) stated that this result, combined with a phylogenetic signal for fish P, indicated that the great stoichiometric variability among fish taxa in P content was derived almost entirely from skeletal investment.
- ^{87}Sr/^{86}Sr ratios are used in archaeology to reconstruct ancient mobility patterns of humans and prehistoric animals, to distinguish between individuals of local and non-local origins at archaeological sites and to establish the provenance of building materials (Slovak & Paytan 2011).

These principles are also used commercially to confirm the source of food and other products. For example, '*normalized ratios of mass fractions found for B, Fe, Cu, Zn, P and S may be used as markers of the biological origin of raw materials of biodiesels... A linear relationship for [S]/[Cu] between these raw samples and their corresponding biodiesels was established*' (Paredes et al. 2014).

By far the most common commercial use is in the food industry, where elemental signatures or profiles are used to trace the origin of food products or ingredients in order to ensure that quality is maintained, or to provide authentication of foods with Protected Designation of Origin, e.g. wines, cheeses, oils, honey, beers, nuts, meats, tea, coffee, etc. (for a review see Gonzalvez et al. 2009). This is sometimes done in combination with isotopic signatures, such as the ratios of stable isotopes ^{15}N/^{14}N and ^{13}C/^{12}C, or the relative amounts of the radioisotopes ^{90}Sr, ^{234}U and ^{238}U (e.g., Pillonel et al. 2003, cited in Gonzalvez et al. 2009). The level of resolution can be quite good; individual wine growing regions or cheese producing regions within a country can be identified. Often only around 10 elements are needed to distinguish geographical origin – these are often elements like Al, Ba, Ca, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Sr, V and Zn. Mineral and trace element content reflect the soil type and environmental growing conditions, while isotopic ratios (^{15}N/^{14}N, ^{13}C/^{12}C) depend on local agricultural practices and animal diets (Gonzalvez et al. 2009). Isotopic methods are more usually used for meat.

In using these methods, multi-element profiles are usually compared against known profiles using a range of pattern recognition methods (mostly multivariate techniques). Neural networks and regression trees have also been used to separate samples by fingerprinting techniques (see Gonzalvez et al. 2009) and many of these can also be used in a predictive rather than an analytical way. Multivariate methods (principal component analysis) based on element composition have been used successfully to separate ecosystem components and trophic groups in a coastal ecosystem (Kumblad & Bradshaw 2008), but such approaches have not been widely used and would be worthwhile exploring further in radioecology. Existing multi-element datasets could be analysed to identify similarities in element profiles between or within taxa, trophic level, functional group, habitat, geographical region, season or year, depending on the temporal and spatial resolution of the data. This would identify the most promising groupings for which extrapolation methods could subsequently be tested, using simple element-specific ratios or predictive multivariate methods.

6.3 Current usage of stoichiometry in radioecology

'Biogeochemically similar' elements were used in the ERICA Tool (Brown et al. 2013) to provide default CR values when data were lacking and when other preferable options were not available (see Chapter 2).

In this context, 'biogeochemically similar' generally means element know to have similar biological/chemical behaviour (typically elements in the same group or period). However, one can question whether using elements in the same period or group of the periodic table always implies that they are similar biogeochemically. This approach probably works better for some chemical groups (e.g. the halogens F-Cl-Br-I or the alkali earth metals Li-Na-K-Rb-Cs) than group 14 (C-Si-Ge-Sn-Pb) or 16 (O-S-Se-Te-Po) whose elements have very different chemical properties and biological roles. One alternative to using periodic group chemical properties may be to use ionic potential (Higley 2010), based on the principle that environmental mobility, and thus bioavailability, is better explained by ionic potential than position in the periodic table (Railsbeck 2003). Classifying elements based on their biological essentiality may also be a useful approach (Bradshaw et al. 2012). However, these ideas have not been explored further in radioecology.

Concentrations of certain elements are known from field and experimental data to be closely correlated to each other, to such a degree that they may be used as analogues for each other. The most common examples in radioecology are (radio) Sr/Ca and (radio)Cs/K. These relationships mean that, for instance: the concentration of Sr or Cs in fish can be quite accurately estimated from the water concentrations of Ca or K (e.g. Kryshev & Ryabov 2000; Smith et al. 2009); and the Sr concentration in milk can be predicted from Ca concentrations in the diet and milk (Beresford et al. 1998b).

Another approach that has been used in radioecology is based on the earlier ecological theory of organic matter (carbon) and energy flows in ecosystems. The approach is based on normalising radionuclide or element concentrations in ecosystem components to their C content, i.e. element:C ratios, based on the assumption that many elements are stoichiometrically related to the carbon content because of their role in metabolism and structural components of the organism (Elser et al. 2000; Bradshaw et al. 2012). Radionuclides/elements can also adsorb to organic surfaces and in this way move through the food chain. Ecosystem models based on carbon flows are thus constructed as the basis for radionuclide/element transfer models, using CR values based on C-normalised element concentrations (Kumblad et al. 2006; Bradshaw et al. 2012; Konovalenko et al. 2014). The theoretical basis for this approach is that energy flows in ecosystems can realistically estimate element transfer in the environment, as modelled uptake is constrained by metabolic rates and the elements available.

6.4 Factors affecting the use of (ecological) stoichiometry in extrapolation

Stoichiometric ratios are not absolute values, varying to some degree with spatial and temporal scale and a range of factors related to these scales. For example, stoichiometric balance can be influenced by irradiance (Finkel et al. 2006), season (Liess & Hillebrand 2005), from year to year and with food web structure (Fitter & Hillebrand 2009). As with other radioecological approaches, it is important to consider whether the system under investigation can be considered to be under equilibrium or non-equilibrium conditions (relative to the temporal scale of interest). Biological, ecological and environmental conditions affect the equilibrium. In the case of radionuclide releases to the environment, the time since release as well as the biogeochemistry of the radionuclide of interest will determine how dynamic the system is, and thus how useful stoichiometry will be.

A key issue in ecostoichiometry is whether an element is essential or non-essential to the organism(s) of interest. The ability of organisms to maintain constant body concentrations despite changing concentrations in the environment and/or their resource supply is known as homeostasis (Kooijman 1995), and is generally assumed to be weak for autotrophs and strong for heterotrophs (Sterner & Elser 2002), so that plant and algae stoichiometry is thought to more closely reflect that of the environment than animals, though this is much debated. The degree of homeostasis has been shown to vary depending on whether elements are macronutrients, essential micronutrients, or nonessential elements (Karimi & Folt 2006; Bradshaw et al. 2012). Organisms often take up the necessary amounts of trace elements from their food within the '*window of essentiality*' (Hopkin 1989) to ensure essential levels but avoid toxic body concentrations.

These principles could be made use of when using ecostoichiometry in extrapolation. For example, for autotrophs, extrapolation from environmental concentrations or ratios is probably more appropriate, whereas for heterotrophs extrapolation within taxonomic groups may be more relevant (Karimi & Folt 2006). Higher trophic levels may have a more similar elemental composition to their food than lower trophic levels, and stoichiometric ratios may shift between abiotic-biotic components and from primary producers to primary consumers (Bradshaw et al. 2012). In cases of homeostasis, element ratios will be constrained by the biology and ecology of the organism/ecosystem. There will be ranges outside of which ratios are impossible; at one end of the spectrum organisms/ecosystems will die/collapse due to element deficiency, and at the other end elements will not be taken up since either organisms already have sufficient of them, or an increased level will become toxic. A better understanding of these natural ranges would greatly help in our ability to predict element concentrations within a feasible range.

It should be mentioned that element ratios do not take into account the form of the element (e.g. chemical form, speciation) or potential isotope-specific differences, although it is well known that this can be important, particularly for elements with lower molecular masses where this is in fact exploited as a means of studying ecological / biological processes (e.g. stable isotope ratios (delta C and N).

Care needs to be taken to compare relevant concentrations/ratios. For instance, for smaller organisms, element concentrations and ratios are given for the whole organism, while for larger organisms muscle tissue is often analysed. Likewise, water samples may be filtered or unfiltered.

6.5 Final remarks

(Ecological) stoichiometry shows potential as extrapolation method in radioecology, either from one element to another or from one species to another. As for all extrapolation methods, this will be most successful when using data from elements, organisms (taxa, functional groups) and/or ecosystems that are as similar as possible to each other. At the very least, stoichiometry could be used to set limits on the range of possible element concentrations; there are biological and ecological limits to many element concentrations / ratios, even if there is variability. Multielement datasets are becoming more common as analysis techniques (e.g. ICP-MS) become easier, faster and cheaper. In the near future we will have access to a much larger amount of data (e.g. see http://www.ionomicshub.org/home/PiiMS) on which to test stoichiometric assumptions and theories and develop extrapolation methods.

7. Bayesian approach: an alternative means for derivation of PDFs characterising concentration ratios

7.1 Introduction

A common step in data collations required to populate CR (and other) databases (e.g. Beresford et al. 2008a; Hosseini et al. 2008; IAEA 2010; Howard et al. 2013) is the pooling or combining of data to produce more precise estimates for the parameters of interest. However, the combined data are usually extracted from different studies with variable sample sizes and different measures of central tendency and dispersion. Another common issue is the application of different extrapolation approaches to derive missing transfer parameter values (e.g. see Brown et al. 2013).

In the course of analysing data for the development of such transfer databases, some consideration has been given to how data will be used in a more robust risk characterisation where not only the severity, but also the probability of occurrence of the exposure needs to be considered (e.g. Brown et al. 2008). The requirement relates not only to detailed, site specific risk characterisation but also to screening assessments where high percentile CRs, that can only be derived from parameters with characterised probability density functions (PDFs), are often utilised in the derivation of screening criteria, such as limiting media concentrations (e.g. see Brown et al. 2013; USDOE 2002). Furthermore, by providing statistical information, the uncertainty associated with the calculation of exposure can be propagated through the assessment using approaches such as Monte Carlo simulation (e.g. see Vose, 1996). Hence, there are clear needs to acquire statistical information, particularly in relation to the assignment of PDFs to parameters within the datasets underpinning the assessment.

The approach that can be applied for the derivation of a PDF characterising a CR (or other required parameter) that is representative for a case of interest will depend on the availability of representative data. Firstly, data either exist or they do not. In cases where there are no data, some reference has to be made to predefined approaches for deriving the distributions, such as those based on extrapolation methods. Typical examples include applying the CR distribution for an analogue, such as data that are available for a similar reference organism or biogeochemical element, to the case of interest (see Chapter 2) (Brown et al. 2013). However, in cases where we have data, we have to differentiate between instances where the data coverage is sufficient and where it is limited. For the latter, we will face different situations depending on what kind of statistical data we have and whether other relevant data (in the literature or as a result of using extrapolation approaches e.g. data for the given radionuclide may be available for a similar organism) can be found or not.

This kind of consideration is especially relevant for many cases in the ERICA Tool[7] $CR_{wo\text{-}media}$ databases where initially, due to lack of data, extrapolation methods were used

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[7] STAR is contributing to the development of a revised ERICA Tool.

extensively (Beresford et al. 2008a; Hosseini et al. 2008). However, now more empirical data (see Copplestone et al. 2013) have become available. It is clear that application of extrapolation approaches is based on the assumption of some kind of similarity or commonality between the missing data and the data being extrapolated. This means we believe that the surrogate data contain some information about the missing value. However, the relevant questions to ask here are: (i) what status do these surrogate data have once some new empirical data for the desired parameter become available? (ii) do they become irrelevant or can they still play a role in our estimation of the unknown parameter? These questions are especially relevant in cases where there are few newly acquired empirical data.

Sheppard (2005) discussed cases where limited site-specific data were available for the case of interest and advised against basing assessments solely on these data. He argued that given the inherent large variability of transfer parameters using a few on-site data to the exclusion of many generic data may decrease accuracy due to the potential error resulting from too few measurements. However, guidance was not provided on how to combine both site-specific and generic data such that all available information could be taken into consideration without imposing unnecessary bias into any subsequent calculations.

Hosseini et al. (2013) suggested an alternative approach that allows for the utilisation of the various related datasets/information that are often available in addition to sets of values that are specific to an organism grouping, site or element. The suggested methodology is based on application of Bayesian statistics. Given that the prior knowledge (external data/information) is valid, this approach provides more robust parameter estimates as compared to when only limited site or study specific empirical data are used. A full description of the methodology presented here, and discussion of its application within radioecology, can be found in Hosseini et al. (2013).

In this chapter, Bayes Theorem is introduced along with a brief description of situations where it can be applied in the context of derivation of PDFs for CRs. Thereafter, the practical application of a Bayesian approach for derivation of PDFs in connection to the updating of the ERICA Tool CR databases is introduced and discussed.

7.2 Estimation of distribution parameters using Bayesian inference

Based on different concepts of probability, statistics may be divided into two main schools (Suter, 2007): *Frequentist* and *Bayesian*. Probability from a Frequentist (or relative-frequency) point of view is understood as an expression of frequency whereas Bayesian statistics: (i) defines probability as a conditional measure of uncertainty and (ii) provides a method for modification of probability in the light of new evidence.

If we consider the situations where inferences are to be drawn on the unknown parameter (or parameter vector) θ in light of vector of independent and identically distributed empirical data values $\boldsymbol{y} = y_1, \ldots, y_n$ and a prior probability distribution $p(\theta)$. Then Bayes' theorem (Bayes, 1763) provides the means for combining information from the prior and the likelihood to produce the posterior density of the parameter conditioned on the data:

$$p(\theta \mid y) = \frac{p(y \mid \theta)\, p(\theta)}{\int_{-\infty}^{\infty} p(y \mid \theta)\, p(\theta)\, d\theta} \qquad \text{or} \qquad \text{posterior} = \frac{\text{likelihood} \times \text{prior}}{\int_{-\infty}^{\infty} (\text{likelihood} \times \text{prior})\, d\theta} \qquad (7.1)$$

Here, the posterior distribution describes our state of knowledge about the parameter θ after considering the data. The likelihood function describes how probable the current data are given the parameter θ. The prior represents the present state of our knowledge based on an initial consideration of the parameter θ. The denominator is the probability of the data, a normalising constant. Hence, the combined (posterior) probability distribution of the parameter given the empirical data is proportional to the prior probability distribution times the likelihood function of the empirical data values:

$$p(\theta \mid y) \propto \prod_{i=1}^{n} p(y_i \mid \theta) \times p(\theta) \qquad (7.2)$$

A crucial step in application of the Bayesian approach to update, for instance, the ERICA Tool CR databases is to define a suitable prior such that it allows for the external information from the extrapolation approaches to be incorporated without overruling the importance of the empirical data which is the main source of information.

Below we first introduce two different types of priors, conjugate and semi-conjugate priors, before describing how the Bayesian approach has been applied in the process of the updating of the ERICA Tool CR databases.

A joint prior distribution for the mean and variance

The first Bayesian inference method considered here for estimation of the distribution parameters is based on the assumption that the available external data used for deriving the prior distribution, and the available empirical data for the case of interest are exchangeable, i.e. can be considered as being from the same population. In this case, the external data carries information on both the mean and variance of the distribution and can be used to define a joint *conjugate prior distribution* (Gelman et al. 2004). A prior distribution is said to be ‘conjugate’ to the measurement model if the resulting posterior distribution is of the same functional form as the prior, but with new parameters.

Independent prior distributions for the mean and variance

The second Bayesian updating technique considered here can be applied in situations where, in addition to data for the case of interest, there is available relevant external data, which does not carry information about the variance of the case of interest, or if other (qualitative) information rather than data is available for the variance. In such a situation, the prior distributions of mean and variance are specified independently. If no prior information is available for the variance, a so called non-informative prior for the variance can be used. With these prior distributions the *conditional* posterior distributions of the mean and variance attain the same functional form as the prior, but the joint conjugate posterior does not. Therefore, these prior distributions are often referred to as *semi-conjugate* prior distributions.

7.3 Updating ERICA CR database using Bayesian approach

Upon its release in 2007, the ERICA Tool (Brown *et al*. 2008) was accompanied with the most comprehensive $CR_{wo\text{-}media}$ database available for wildlife. However, recent developments including a new collation of concentration ratio data (the Wildlife Transfer Database – WTD (Copplestone *et al.* 2013)) has called for the need to update parameter values in the Tool's databases. The WTD values as of December 2013 have been used to derive an updated set of default CR values for a revision of the ERICA Tool (to be released late 2014).

Upon updating the ERICA Tool CR databases some decisions have to be made in dealing with cases where available data were limited. It was decided to look closer to cases where the number of available data were equal to or less than 5. It was assumed that any parameter derived in these cases suffers a lack of credibility as they are based on very few data points. To improve the situation in terms of deriving more robust parameter estimates the Bayesian approach was applied. After identifications of such cases ($N \leq 5$), the updated gap-filling options, available in the ERICA Tool, were consulted to discern which analogues would have been used if no data were available for these cases. In this way a surrogate organism or radionuclide has been assigned for each case where possible. Hence, depending on the availability of a surrogate dataset as well as statistical information, different situations (Table 7.1) had to be dealt with.

Table 7.1. An overview of cases ($N \leq 5$) that had to be dealt with in updating ERICA $CR_{wo\text{-}media}$ databases.

Statistical data for organism		**Surrogate dataset**	**Approach**
Mean	***SD***		
A[*]	*A*	*A*	*Semi – conjugate (non-informative)*
A	*A*	*NA*[**]	*Using data as it is (assume lognormal PDF)*
A	*NA*	*A*	*Using ratio of SD to mean for the surrogate to derive the missing SD from the data mean*
A	*NA*	*NA*	*Using data as it is (assume exponential PDF)*

[*] A – available; [**] NA - not available.

For the discussion below we only concentrate on the first case, as shown in Table 7.1, because this is the only instance where we have applied a Bayesian approach. In these cases the existing surrogate data have been used as priors in the process of updating.

Table 7.2 illustrates some examples from the revised ERICA Tool marine $CR_{wo\text{-}water}$ database where new parameter estimates have been derived based on the application of the Bayesian semi-conjugate method.

As the last column of Table 7.2 shows, considering the prior knowledge results in larger SDs. This extra uncertainty can be viewed as a more robust characterisation of the derived parameter values in cases where the empirical data are very few and the uncertainty very large. This is especially true given the primary purpose of the default ERICA Tool database is to derive conservative $CR_{wo\text{-}media}$ estimates (i.e. 95th percentile).

Table 7.2. Examples of the ERICA marine $CR_{wo\text{-}water}$ (L kg^{-1}) database where the Bayesian approach has been applied to derive new parameter estimates. Shaded rows represents surrogate species or radionuclides which have been used as priors.

Radionuclide	**Species**	**N**	**Mean**	**SD**	***New estimates***	
					Mean	***SD***
Ca	Fish	3	6.2E+0	5.5E+0	*8.24E+0*	*8.87E+0*
Sr	Fish	118	2.5E+1	3.9E+1		
Cd	Crustacean	5	9.6E+3	5.0E+3	*1.0E+4*	*5.79E+3*
Cd	Mollusc - bivalve	64	1.1E+5	4.8E+5		
Co	Polychaete worm	3	8.3E+3	1.0E+4	*8.03E+3*	*1.20E+4*
Co	Mollusc - bivalve	42	5.3E+3	1.5E+4		
Co	Sea Anem./True Coral	4	3.3E+2	5.2E+2	*6.08E+2*	*1.29E+3*
Co	Mollusc - bivalve	42	5.3E+3	1.5E+4		
Co	Vascular plant	3	5.2E+1	5.9E+1	*3.28E+2*	*1.06E+3*
Co	Macroalgae	130	1.7E+3	3.2E+3		
Eu	Macroalgae	4	1.4E+3	1.0E+3	*1.66E+3*	*1.37E+3*
Eu	Phytoplankton	11	1.1E+4	2.2E+4		
Pu	Polychaete worm	3	1.51E+3	2.25E+3	*1.53E+3*	*2.53E+3*
Pu	Mollusc - bivalve	164	1.1E+3	1.4E+3		

7.4 Discussion

Bayesian approaches require specification of prior distributions for parameter values. In the context of the work discussed as an example here, priors have been used to take into account the existing external information/data and also to describe our 'belief' about the relevancy of these data for our case of interest. As priors represent additional knowledge which would not be considered if we were only using likelihood function, considering them is not only a necessary step in the process of learning and acquiring knowledge, but also a crucial element for coming to the right conclusion (Kruschke, 2010).

To individuals not familiar with Bayesian methods the inclusion of information based upon belief about the relevancy of data may seem overly subjective. However, this is exactly the process that is employed during pooling of datasets using classical statistical methods wherein, for instance, knowledge/belief about congruity of $CR_{\text{wo-media}}$ values between taxonomically similar organism groups is used to delineate the extent to which data are combined. For example, in the Wildlife Transfer Database (Copplestone et al. 2013), there are some tacit assumption that there is some rationale in grouping, for example, mammals in terms of feeding strategy, this assumption in part being based upon prior knowledge/belief regarding the importance of the ingestion pathway in determining internal radionuclide body burdens and similarities in physiology dictating uptake. Hence, while the 'classical' approach implicitly uses judgment, the Bayesian approach explicitly acknowledges the role of judgments made.

In the process of updating the ERICA Tool $CR_{\text{wo-media}}$ databases the Bayesian semi-conjugate approach has been used to derive more robust parameter values. When updating the parameters with independent (semi-conjugate) prior distributions we are interested in deriving a PDF for a given organism by using the available empirical data and relevant information from other sources. The belief/knowledge we are expressing in using this method is that although the given organism type may share common traits with regards to radionuclide transfer with another organism, there is a substantial likelihood that the organism under consideration expresses its own CR values thus rendering mere pooling of data or conjugate updating inappropriate. The independent priors offers the means of placing emphasis on the species-specific data whilst linking the data to what is known about related generic datasets in a mathematically structured way.

A Bayesian approach has also been applied by Norden et al. (2010) to derive CR and K_d values using both site-specific and literature data. Bayesian approaches have a much wider applicability to radioecological problems and are being considered within the COMET (www.comet-radioecology.org) project.

8. Summary

We will never have data to populate all of the potential radioecological modelling parameters required for human and wildlife assessments. Therefore, we need robust extrapolation approaches which allow us to make best use of our available knowledge. In this study we have reviewed, developed, tested and validated various extrapolation approaches.

Our report starts with an evaluation of the extrapolation approaches used to populate the $CR_{wo\text{-}media}$ database of the original version of the ERICA Tool (Chapter 2; Brown et al. 2013). We have shown that these were not guaranteed to result in conservative values as required to populate a database primarily designed for initial protective screening tier assessments. Recommendations from this assessment have been taken forward in the STAR supported derivation of revised $CR_{wo\text{-}media}$ values for the ERICA Tool (see Brown et al. 2014; Beresford et al. 2014)[8] including a more robust consideration of available knowledge and PDFs using Bayesian statistics (Chapter 7; Hosseini et al. 2013).

Application of the transfer coefficient concept has been widely adopted in radioecology to estimate activity concentrations in farm animal products; applicability to wildlife assessments has also been proposed. However, this parameter is, in part, dependent upon the dry matter intake of animals and this gives a false impression of differences in transfer between different species (Chapter 3). The concentration ratio ($CR_{product\text{-}diet}$ or $CR_{wo\text{-}diet}$) is a more generic parameter and also enables the more abundant data for farm animal species to be applied to wild species.

Allometric relationships are used in some radioecological models to derive default $CR_{wo\text{-}soil}$ values or to enable dynamic predictions. Allometric expressions describing radionuclide biological half-life have been proposed for some elements. Of these many scale to circa $M^{0.25}$. We have shown that for mammals and birds allometric expressions with $M^{0.25}$ have a biological basis. Previously it had been suggested that if you could assume $M^{0.25}$, then for those elements/radionuclides for which allometric expressions had not be derived (i.e. due to lack of data) all that was required was an estimate of the constant '*a*' (i.e. where $T_{B1/2}=aM^{0.25}$). We have derived an approach to estimate '*a*' for birds and mammals which is based upon generally available parameters (Chapter 4; Beresford & Vives i Batlle 2013). This has been shown to predict generally acceptable $T_{B1/2}$ for a number of radionuclides and over orders of magnitude range in animal live-weight.

However, we have to acknowledge that for a number of radionuclides (i.e. Am, Ce, Eu, Pu and Th) $T_{B1/2}$ have been shown to scale to *circa* $M^{0.8}$. To fully exploit the potential of allometric models the reasons for this need to be explored.

We demonstrate that application of the relationship derived for homoeothermic vertebrates is not directly applicable to poikilothermic reptiles. However, it is possible to parameterise

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[8] The revised version of the ERICA Tool will be released autumn 2014.

Equation 4.12 with reptile specific values and obtain reasonable agreement between measured and predicted $T_{B1/2}$ values (Chapter 4; Beresford & Wood, 2014). It should be possible to populate Equation 4.12 to predict biological half-life for other types of organism assuming the required relationships and parameter values are available. However, it is likely that such information may be sparse for some organisms.

Although not assessed here it is recommend that the application of Equation 4.12 to make approximations of $T_{B1/2}$ for edible tissues of farm animals, a relatively poorly studied parameter for many radionuclides, be tested against available data.

The commonly used $CR_{\text{wo-media}}$ approach to estimating the radionuclide activity concentrations in wildlife is open to criticism, as $CR_{\text{wo-media}}$ values can be highly variable, largely due to site-specific factors. The analyses of available data using the REML analysis, as demonstrated here (Beresford et al. 2013; Chapter 5), should compensate for inter-site variation, assuming sufficient data are available for the analysis. For freshwater fish the outputs of the REML analysis predicted ^{137}Cs activity concentrations in an independent dataset well. Caesium was selected here as an example because there was a relatively large amount of data available. We recommend that this approach of producing relative values be further investigated and developed for other radionuclides and across a wider range of organisms and ecosystems. A disadvantage of the approach is that it requires relatively large datasets which must meet specific criteria. However, recent data compilations (Howard et al. 2013; Yankovich et al. 2013; Copplestone et al. 2013) should enable similar analysis to be conducted for a number of elements for terrestrial, marine and freshwater species.

The ICRP (2009) suggested identifying a series of terrestrial, freshwater and marine sites from which samples of their Reference Animals and Plants (RAPs) could be sampled and analysed to serve as ‘points of reference’. Such studies have been initiated and results are starting to be published (Barnett et al. 2013). However, such data are highly site specific, potentially limiting their wider applicability. The application of the REML approach to data such as that presented by Barnett et al. (2013) to derive relative values for different organisms should provide a more generic set of ‘reference data’. In taking the REML approach forward if will be beneficial to target studies to provide data that will fill gaps in the input data reducing uncertainties and biases in the REML outputs. A collaboration of the TREE (http://www.ceh.ac.uk/tree) and COMET (www.comet-radioecology.org) projects is taking this area of research forward, including the focussed sampling of species falling within the RAP definitions at a number of terrestrial sites in different countries (Norway, Spain, UK, Ukraine and Japan).

Ecological stoichiometry shows potential as extrapolation method in radioecology, either from one element to another or from one species to another (Chapter 6). This will be most successful when using data from elements, organisms (taxa, functional groups) and/or ecosystems that are as similar as possible to each other. At the very least, stoichiometry could be used to set limits on the range of possible element concentrations; there are biological and ecological limits to many element concentrations / ratios, even if there is variability. Multi-element datasets are becoming more common as analysis techniques (e.g. ICP-MS) become easier, faster and cheaper. In the near future we will have access to a much larger amount of data (e.g. see http://www.ionomicshub.org/home/PiiMS) on which to test stoichiometric assumptions and theories. The TREE project (http://www.ceh.ac.uk/tree) is taking forward this area of research, primarily for crop plants but with some consideration of wildlife and potentially farm animals.

We have made the case for the application of Bayesian statistics in the derivation of transfer parameters (Chapter 7; Hosseini et al. 2013). The approach has now been used to help populate the revised parameter database for the ERICA Tool. However, there is a need to investigate and foster the wider application of Bayesian statistics in radioecology and the COMET (www.comet-radioecology.org) project is now doing this.

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Appendix A: References used to source $T_{B1/2}$ values for the extended evaluation of Beresford & Vives i Batlle model in Chapter 4

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