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Abstract: We will never have data to populate all of the potential radioecological modelling parameters required for wildlife assessments. Therefore, we need robust extrapolation approaches which allow us to make best use of our available knowledge. This paper reviews and, in some cases, develops, tests and validates some of the suggested extrapolation approaches.
The concentration ratio (CRproduct-diet or CRwo-diet) is shown to be a generic (trans-species) parameter which should enable the more abundant data for farm animals to be applied to wild species.
An allometric model for predicting the biological half-life of radionuclides in vertebrates is further tested and generally shown to perform acceptably. However, to fully exploit allometry we need to understand why some elements do not scale to expected values.
For aquatic ecosystems, the relationship between log10(a) (a parameter from the allometric relationship for the organism-water concentration ratio) and log(Kd) presents a potential opportunity to estimate concentration ratios using Kd values.
An alternative approach to the CRwo-media model proposed for estimating the transfer of radionuclides to freshwater fish is used to satisfactorily predict activity concentrations in fish of different species from three lakes. We recommend that this approach (REML modelling) be further investigated and developed for other radionuclides and across a wider range of organisms and ecosystems.
Ecological stoichiometry shows potential as an extrapolation method in radioecology, either from one element to another or from one species to another.
Although some of the approaches considered require further development and testing, we demonstrate the potential to significantly improve predictions of radionuclide transfer to wildlife by making better use of available data.

Making the most of what we have: application of extrapolation approaches in radioecological wildlife transfer models

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Robust extrapolation approaches allowing best use of available knowledge are needed.

Extrapolation approaches are reviewed, developed, tested and validated.

Recommendations for the use of extrapolation approaches and future work are made.

Making the most of what we have: application of extrapolation approaches in radioecological wildlife transfer models

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Abstract

We will never have data to populate all of the potential radioecological modelling parameters required for wildlife assessments. Therefore, we need robust extrapolation approaches which allow us to make best use of our available knowledge. This paper reviews and, in some cases, develops, tests and validates some of the suggested extrapolation approaches.

The concentration ratio ($CR_{product\text{-}diet}$ or $CR_{wo\text{-}diet}$) is shown to be a generic (trans-species) parameter which should enable the more abundant data for farm animals to be applied to wild species.

An allometric model for predicting the biological half-life of radionuclides in vertebrates is further tested and generally shown to perform acceptably. However, to fully exploit allometry we need to understand why some elements do not scale to expected values.

For aquatic ecosystems, the relationship between $\log_{10}(a)$ (a parameter from the allometric relationship for the organism-water concentration ratio) and $\log(K_d)$ presents a potential opportunity to estimate concentration ratios using K_d values.

An alternative approach to the $CR_{wo\text{-}media}$ model proposed for estimating the transfer of radionuclides to freshwater fish is used to satisfactorily predict activity concentrations in fish of different species from three lakes. We recommend that this approach (REML modelling) be further investigated and developed for other radionuclides and across a wider range of organisms and ecosystems.

Ecological stoichiometry shows potential as an extrapolation method in radioecology, either from one element to another or from one species to another.

Although some of the approaches considered require further development and testing, we demonstrate the potential to significantly improve predictions of radionuclide transfer to wildlife by making better use of available data.

1. Introduction

The assessment of the exposure of wildlife to ionising radiation for planned, existing and emergency scenarios requires predictions to be made of the transfer of a wide range of radionuclides to a diversity of species. Most models assessing the exposure of wildlife for regulatory purposes use a simple concentration ratio ($CR_{wo\text{-}media}$) relating the whole organism activity concentration to that in an environmental medium (i.e. soil, air or water) (Beresford et al. 2008a). Other models use radionuclide biological half-lives and transfer from the diet (e.g. USDOE 2002).

For many of the radionuclide-species combinations that require assessment, there are no empirical data. When empirical data are lacking, predictions are often made using transfer parameter values derived using extrapolation approaches, though clarity on the use of such approaches varies between publications/models. For instance, the $CR_{wo\text{-}media}$ value for an organism of 'similar taxonomy' may be assumed (e.g. a mammal value may be used to model birds if data for the latter are lacking) (e.g. Beresford et al 2008b). Brown et al. (2013) recently evaluated how the most commonly used approaches performed against additional data now available (Copplestone et al. 2013). They concluded that the extrapolation approaches commonly used to date have under-predicted the empirical 95th percentile $CR_{wo\text{-}media}$ value as often as they have over-predicted. This highlights the need to treat results from these previous extrapolations with caution, but there have been various recent advances that may bring greater confidence in the application of extrapolation methods. In this paper, we assess our ability to extrapolate radioecological data of relevance to wildlife radiological assessments considering these recent advances and future potential.

2. Transfer from the diet to terrestrial vertebrates is a constant across species

The transfer coefficient was first proposed 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. (1965) reported that this parameter exhibited less variability between individual animals within their experimental herd than when transfer was expressed as the total amount of Cs excreted in milk (represented as a percentage of intake). Ward & Johnson (1965) subsequently defined the meat transfer coefficient as the ratio of the ^{137}Cs activity concentration in boneless meat to the dietary daily ^{137}Cs intake.

Following the publications of Ward and co-workers in the 1960s, 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 1970s to early 1980s, transfer coefficient values were being recommended for most radionuclide-animal product combinations (e.g. Ng 1982; Ng et al. 1977, 1979, 1982) and the International Atomic Energy Agency (IAEA) recommended their use (IAEA 1994). These recommended values have been incorporated into many predictive food chain models (e.g. Brown & Simmonds 1995; Müller & Pröhl 1993; USNRC 1977; Yu et al. 2001).

On the basis of the many studies 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, F_f values for sheep meat recommended by IAEA (2010) for many radionuclides are *circa* one order of magnitude higher than those recommended for beef. Similarly, F_m values for goat milk tend to be one order of magnitude higher than those recommended for cow milk. The use of transfer coefficients has also been suggested for wildlife (e.g. Thomas et al. 1994; MacDonald 1996; Moss & Horrill 1996) and some models use simple food chains to estimate radionuclide concentrations in wildlife (e.g. USDOE 2003).

However, it has been suggested that much of the observed difference in $F_{f,m}$ values is a consequence of differences in dry matter intake, between animals of differing age or species, rather than any difference in radionuclide transfer (e.g. Smith & Beresford 2005; Galeriu et al. 2007; Beresford et al. 2007). The rational for this is that the transfer coefficient is defined as:

$$F_f = \frac{Radionuclide\ activity\ concentration\, in\, meat\ \ (Bq\, kg^{-1})}{Daily\, dry\, matter\, \mathrm{int}ake\ (kg\, d^{-1}) \times Radionuclide\, activity\, concentration\, of\ the\, diet\, (Bq\, kg^{-1}\, DM)}$$

Consequently, the concentration ratio ($CR_{meat\text{-}diet}$) is equal to:

$$CR_{meat-diet} = F_f\ (d\, kg^{-1}) \times\ Daily\, dry\, matter\, \mathrm{intake}\, (kg\, d^{-1})$$

The above equations are for meat but a similar derivation can be performed for milk or eggs.

A between species similarity in *CR* values for animal derived food products should not be surprising, given that the concentrations of many elements in meat, or milk, are similar across species (Mertz 1986; 1987). A particular advantage of being able to assume that the milk or meat *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, the IAEA (2010) summarised milk and meat *CR* values, as well as transfer coefficients.

We would also expect that *CR* values for wildlife would vary little between species (Beresford et al. 2004) and would be similar to those of farm animals. To test this hypothesis, Table 1 presents Cs $CR_{meat\text{-}diet}$ values for seven herbivorous species of wild mammals and birds. The $CR_{meat\text{-}diet}$ values for these species are similar to those for the meat of farm animals in IAEA (2010), which presents a generic value of 0.39 based on data for four farm animal species. Since Cs is relatively homogenously distributed throughout the body tissues (Yankovich et al. 2010a), it can then be assumed that, for Cs $CR_{meat\text{-}diet} \approx CR_{org\text{-}diet}$ (where $CR_{org\text{-}diet}$ is the ratio of the radionuclide activity concentration in the whole organism to that in its diet). Currently there are few data with which to test our hypothesis for the transfer of other radionuclides to wild animals. For many elements other than Cs, distribution is not homogenous throughout the body tissues. However, for such an element an assumption that the distribution within the body was similar across animal species would be reasonable (e.g. Sr accumulates in the bone of all vertebrates).

3. Allometry

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. There are several allometric equations that can be proposed, the simplest being to assume that:

$$Y = aM^b$$

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

Kleiber (1932) found that basal metabolic rate (measured as heat production) across 13 groups of mature animals, ranging from <0.2 to *circa* 680 kg body mass, was proportional to mass to the power 0.74 (i.e. $M^{0.74}$). Kleiber subsequently suggested that ‘metabolic body size’ (now generally referred to as metabolic live-weight) could be determined as $M^{0.75}$ (Kleiber 1947); this has since become known as *Kleiber’s law*.

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). The debate hinges on theoretical considerations, such as whether allometry of the metabolic rate relates to the fractal nature of energy distribution networks (West et al., 1999; Brown et al, 2002) with and an exponent of 0.75, or whether it is based on a consideration of surface area-to-volume ratios with an exponent of 0.67 (Farrer-Gray & Gotelli, 2005).

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 1 demonstrates the relatively small effect of assuming $M^{0.67}$ rather than $M^{0.75}$ over a likely range of vertebrate live masses.

3.1 Allometry in radioecology

Homeothermic vertebrates

Many of the reported allometric relationships, such as dry matter food ingestion rates, water ingestion rates and inhalation rates, have been used in models of the radionuclide transfer to wildlife (e.g. Beresford et al. 2008a; Johansen et al. 2012), including the US Department of Energy’s ‘Graded Approach’ as implemented in the RESRAD-Biota model (USDOE 2002, 2004). However, in the 1970s, radionuclide biological half-life ($T_{1/2B}$) was also shown to scale allometrically (e.g. Stara et al. 1971; Kitchings et al. 1976). The application of allometry to radioecological parameters has received revived attention during the development of models to predict the exposure of wildlife to radionuclides in 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).

USDOE (2002) presents $T_{1/2B}$ allometric relationships for 16 elements in terrestrial/riparian vertebrates. When used in model inter-comparison exercises, allometric relationships from USDOE (2002), and similar models, produced results comparable to models using $CR_{wo\text{-}media}$ values (Beresford et al. 2009; Wood et al., 2009a). Of the allometric expressions describing radionuclide $T_{1/2B}$ from USDOE (2002), a number have an exponent of approximately 0.25 (Cs, Co, Ra, Sb, Sr, U, Zn and Zr). Iodine has an exponent of 0.13 and H of 0.55, but other literature suggests these too should approximate to 0.25 (Galeriu et al. 2003; MacDonald 1996). Therefore, it appears that for most radionuclides, $T_{1/2B}$ is driven by metabolic rate and Beresford & Vives i Batlle (2013) demonstrated that this was logical by considering a simple radionuclide retention model (Sazykina 2000) and Kleibers Law. Notable exceptions are Pu, Am, Ce and Eu, which USDOE (2002) suggest scale to *circa* 0.8.

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 $T_{1/2B}$, *circa* 0.25), then only an estimation of the multiplicand is needed for any given element. Through algebraic derivation, Beresford & Vives i Batlle (2013) proposed a method of determining the multiplicand for the $T_{1/2B}$ relationship:

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

where a_B is the multiplicand for the allometric relationship describing $T_{1/2B}$, a_I is the multiplicand for the allometric relationship describing dry matter intake and f_1 is the fractional gastrointestinal absorption coefficient. By substituting this expression for a_B in the equation describing the $T_{1/2B}$ allometric relationship, Beresford & Vives i Batlle (2013) suggested that an estimate of $T_{1/2B}$ could be derived as:

$$T_{1/2B} = \frac{\ln 2}{a_I f_1} CR_{org-diet} M^{0.25}$$

Values of a_I are relatively well documented for terrestrial vertebrates (e.g. Nagy 2001), f_1 values are collated in publications (e.g. IAEA 2010), and, if we accept that $CR_{org\text{-}diet}$ is a constant across species (see above), then this parameter can also be sourced from IAEA (2010) for many radionuclides.

Beresford & Vives i Batlle tested their hypothesis using data primarily summarised by Whicker & Shultz (1982), which tabulates $T_{1/2B}$ estimates for a number of radionuclides and terrestrial organisms. Data were for mammals ranging from 0.01 kg (harvest mouse) to 80 kg (reindeer) and radionuclides of Co, Cs, I and Sr. A comparison of predicted $T_{1/2B}$ values across all radionuclides made using a_I values appropriate to the feeding type of each species with measured data yielded a linear regression ($p<0.001$) with an R^2 value of 0.58, a slope of 1.4 and an intercept which was not significantly different from zero (Beresford & Vives i Batlle 2013).

Subsequent to Beresford & Vives i Batlle (2013), a review of radionuclide $T_{1/2B}$ values has been conducted (this is contributing to IAEA MOdelling and DAta for Radiological Impact Assessments (MODARIA) programme activities, see: http://bit.ly/1e9Nxxq). Data identified in this review enable us to further test the Beresford & Vives i Batlle (2013) model and to consider a wider range of elements (Ag, Co, Cs, I, Na, Nb, Ru, Se, Sr and Zn) and species. The data used had to meet various criteria:

- animal live-weight mass had to be presented in the source reference;
- only data for adults were used;
- $T_{1/2B}$ values had to be for the whole body except in the cases of I and Cs, for which thyroid and muscle data were also used, respectively, assuming these tissues reflected whole body 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 that these elements behave in a similar manner in the circulatory system after either oral or intravenous administration (Mayes et al. 1996).

A total of 123 $T_{1/2B}$ values have been considered in this expanded model testing (source references are listed in Supplementary Materials); these included data originally considered by Beresford &Vives i Batlle (2013), though source references were consulted rather than relying on the Whicker & Schultz (1982) compilation. Data were largely for mammals (ranging from 8 g to 70 kg body mass), although limited data for ducks were also identified. When multiple components of loss were cited in the source reference, the longest $T_{1/2B}$ value was used for comparison with predictions.

Radionuclide-specific parameter values used to make predictions are presented in Table 2. Estimates of f_1 were taken from IAEA (2010), which cites values from ICRP (2006) for monogastric animals and additionally presents f_1 values for ruminants (the ruminant values were only used here if no monogastric f_1 value was available; Beresford & Vives i Batlle (2013) compare the effect of using ruminant *vs* monogastric f_1 values). Values of $CR_{org\text{-}diet}$ were estimated from $CR_{meat\text{-}diet}$ values presented by IAEA (2010). Whilst IAEA (2010) presents $CR_{meat\text{-}diet}$ values for Co, Cs and I, it does not include a $CR_{meat\text{-}diet}$ value for Sr. Dietary transfer coefficients presented in IAEA (2010) were, therefore, used together with typical dry matter intake rates from IAEA (1994) to estimate an average Sr $CR_{org\text{-}diet}$ across all five species for which F_f data were available (cattle, goat, sheep, poultry and pig) in IAEA (2010). To convert $CR_{meat\text{-}diet}$ values to $CR_{org\text{-}diet}$ values, conversion factors from Yankovich et al. (2010a) were used; if Yankovich et al. contained no data for a given element, then conversion factors were estimated from Coughtrey et al. (1983) or the data of Barnett et al. (2014).

Values of a_I were sourced from Nagy (2001), and both the 'all mammals' value (a_I=0.057 d^{-1} $kg^{0.25}$) and values for the most appropriate taxonomic or feeding strategy group were used: carnivorous mammals (a_I=0.027 d^{-1} $kg^{0.25}$); herbivorous mammals (a_I=0.15d^{-1} $kg^{0.25}$); omnivorous mammals (a_I=0.077 d^{-1} $kg^{0.25}$); omnivorous birds (a_I=0.119d^{-1} $kg^{0.25}$). Values presented by Nagy, in grammes, were converted to kilogrammes for application here, using the multiplicative factor: $(a_I \times 1000^{0.75})/1000$.

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 123 predictions made using the most appropriate feeding group a_I (Table 3). Allometry is a broad indicator only (Higley & Bytwerk, 2007) and hence predictions within an order of magnitude are considered acceptable here.

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 higher than 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 by Beresford & Vives i Batlle (2013). These observations are discussed further below.

When predictions were made using a_I values for specific feeding strategies (see Table 3), the following were observed:

Carnivorous mammals – Using the a_I value for carnivorous mammals increased the predicted $T_{1/2B}$ value compared to that predicted using the 'all mammals' a_I value (Figure 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 $T_{1/2B}$ value compared to that predicted using the 'all mammals' a_I value (Figure 3). There was not a consistent trend 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 for herbivorous mammals resulted 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 $T_{1/2B}$ value compared to that predicted using the 'all mammals' a_I value, but only by *circa* 25%.

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. Caesium data were the most numerous (n=55). If we omit an outlying value for rabbit, which was under-predicted by more than an order of magnitude, a linear regression of measured Cs $T_{1/2B}$ values to predictions made using the appropriate feeding group a_I value yields an R^2 of 0.52, a slope of 0.43, and an intercept which is not significantly different to zero. The slope demonstrates a tendency to under-predict the measured values.

There was a tendency to over-predict Sr $T_{1/2B}$ values, some by more than an order of magnitude (Table 3). Those four values which were over-predicted by more than an order of magnitude were all from studies using intravenous administration or radiostronitum. The reported half-lives (see DiGregorio et al. 1978) were comparatively short compared to the two studies reporting Sr $T_{1/2B}$ values following oral administration and also data from farm animal studies (Fesenko et al. 2015).

Predictions for Ag, Co, Se and Zn all had an acceptable level of agreement. Those for Na were within an order of magnitude of the observed $T_{1/2B}$ values, but were all comparatively low.

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, although this is also the case for Ag and Se. 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. This is plausible as IAEA (2010) presents $CR_{meat\text{-}diet}$ values for other elements which vary by 1-2 orders of magnitude between studies. The f_1 values used also impact on the predicted $T_{1/2B}$ values for both Nb and Ru. There are reported f_1 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 $T_{1/2B}$ used here are from Furchner and Drake (1971) and Furchner et al. (1971) respectively; the f_1 values estimated by these authors from experimental data were similar to those used to make our predictions.

There was reasonable agreement between the predicted and measured Zn $T_{1/2B}$ values for ducks; the ratio of predicted to measured values ranged from 0.2 to 1.5. However, $T_{1/2B}$ values varied considerably more for the observed data (28-250 d; n=13) than the predicted values (41-42 d). The low variation in predictions is not surprising given that the mass of the ducks varied little (1.1-1.3 kg).

Overall, 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), $CR_{\text{meat-diet}}$ values are based upon few observations. 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 $T_{1/2B}$ 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 were less than the observed value.

An assumption of the approach described here is that $T_{1/2B}$ scales to the power of 0.25. For five elements in USDOE (2002) (Am, Ce, Eu, Pu and Th), $T_{1/2B}$ scales to the power of 0.8. For all of these elements, International Commission on Radiological Protection 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 important 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 neither essential elements nor their analogues (e.g. U).

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.

For comparison with our predictions, we have used the longest $T_{1/2B}$ value reported where multiple components of loss were observed. In doing so, 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 is questionable whether the longest $T_{1/2\mathrm{B}}$ always best represents metabolic turnover when three and four component loss equations are reported.

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, if the models are correctly parameterised, yield estimates of the equilibrium activity concentration in organisms which are conservative (i.e. they should be over-estimated compared to a model that assumes 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.

Application to reptiles

Although only proposed for homeothermic vertebrates, allometric models for $T_{1/2B}$, such as USDOE (2002), have been used to make predictions of radionuclide activity concentrations in (poikilothermic) reptiles and amphibians (e.g. Wood et al. 2009a; Beresford et al. 2010; Yankovich et al. 2010b; Johanssen et al. 2012).

Beresford & Wood (2014) evaluated to what extent the Beresford & Vives i Batlle (2013) model could be applied to reptiles. Similar criteria as used above for mammal and bird data were applied to identify $\mathrm{T}_{1/2\mathrm{B}}$ values against which to compare predictions. In total 28 $T_{1/2\mathrm{B}}$ values for Cs, three for Sr and two for Ra were identified (see Beresford & Wood (2014)). Data for Cs were 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 $T_{1/2\mathrm{B}}$ 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 $T_{1/2B}$ values.

Using parameter values for mammals, all but one prediction was under-estimated; in the case of Cs, the under-estimates were by 1-2 orders of magnitude.

The exponent in the allometric biological half-life expressions is, in effect, defined by the exponent for the allometric model of metabolic rate (B_r) of *circa* 0.75. Therefore, the exponent for $T_{1/2B}$ is '(1 – the exponent for B_r)'. However, the exponent for the allometric model describing B_r for reptiles has been shown to be in the range *circa* 0.80-0.92 (Nagy 2005; Isaac and Carbone 2010). Therefore, the exponent describing $T_{1/2B}$ for reptiles should be in the range 0.08-0.20. From Nagy (2001), Beresford & Wood (2014) estimated exponents of 0.037 for snakes, derived from information for carnivorous reptiles, and 0.08 for turtles, derived from information for 'all reptiles'. These exponents, together with the relevant values of a_I for reptiles (Nagy 2001) and reptile-specific values of $CR_{org\text{-}diet}$ and f_1 (see Beresford & Wood (2014) for details of derivation), were used to estimate $T_{1/2B}$ values to compare with the available data (Figure 4). Only one of the 33 predictions deviates by more than a factor of six from the measured value; the one 'outlier', a prediction of the Sr $T_{1/2B}$ in

T. scripta scripta, was predicted to be 14 times higher than the measured value. However, $T_{1/2B}$ allometric exponents applicable to reptiles result in a relatively low dependence of $T_{1/2B}$ with mass (see Figures 1 and 4). Over a mass range 0.1 g to 1000 kg, the predicted biological half-lives for reptiles vary by less than a factor of four using an exponent of 0.08, and by less than a factor of two using an exponent of 0.037. For homeotherms, $M^{0.25}$ predicts $T_{1/2B}$ values varying over *circa* 60-fold (Figure 1). Given the small influence of mass on $T_{1/2\,B}$ predictions for reptiles, Beresford & Wood (2014) suggested that if sufficient reported $T_{1/2B}$ values are available for a given element, then it is likely that these would be applicable to any reptile. For instance all of the 28 reported values of Cs $T_{1/2B}$ for reptiles considered by Beresford and Wood (2014), which covered a 50-fold mass range, were within a factor of 5 of the mean.

However, the relatively good agreement between predicted and measured $T_{1/2B}$ in Figure 4 demonstrates that if no reptile data are available for a given radionuclide, then the Beresford & Vives i Batlle model (2013), populated with reptile-specific parameter values, will give reasonable estimates.

There is some evidence that the dry matter intake rate of herbivorous reptiles (namely tortoises) scales to mass, with an exponent of *circa* 0.75 (Franz et al. 2011). Therefore, we may expect the allometric relationship for $T_{1/2B}$ for tortoise species to scale to *circa* 0.25, but we currently have no data to test this.

Aquatic organisms

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). As discussed above for reptiles, for most other aquatic organisms, this would not be a valid approach. However, the application of allometry to derive both biological half-life and *CR* values for marine organisms has been the subject of some studies (see review by Vives i Batlle et al. 2009).

Table 4 presents allometric parameters describing $T_{1/2B}$ for Tc, Cs, Pu and Am in marine organisms. To parameterises these models, data were available for fish, crustaceans, molluscs, macroalgae/seaweed and plankton (Vives i Batlle et al., 2007, 2008). The mean value of *b* for the $T_{1/2B}$ relationships was 0.16 ± 0.03. Vives i Batlle et al. (2009) presented allometric $T_{1/2B}$ expressions for Sr and I from previous studies. However, these expressions were taken from USDOE (2002) and were for terrestrial animals; they are not valid for most marine organisms (although they may be applicable to marine birds and mammals).

For *CR*, the mean (±SD) value of *b* for marine organisms, calculated across all those elements (Ac, Am, Ce, Cm, Mn, Pa, Pm/Eu, Po Pu, Ra, Ru, Th and Zr), was -0.26±0.09 and the fitted relationship had an R^2 value >0.7 (Vives i Batlle et al. 2009) (see Table 5). For the individual radionuclides with an R^2 value in Table 5 greater than 0.7 (i.e. Am, Ce, Cm, Pm/Eu, Pu, Ra, Ru, and Th), Vives i Batlle et al. (2009) also found a significant linear relationship between $\log_{10}(a)$ and $\log_{10}(K_d)$ (i.e. the sediment-water distribution coefficient) (Figure 5), showing particle-reactive (or sediment seeking) radionuclides to have the highest log10(a) values. The relationship between log10(a) and log(K_d) improved when Ru and Pm/Eu were removed though the authors had no scientific justification for doing this. With the addition of Ac, Mn, Pa, Po and Zr (which have R^2 values <0.7 in Table 5), the trend became weaker ($\log_{10}(a) = 0.38 \times \log_{10}(K_d) - 1.6$; $R^2 = 0.50$; n = 13), but the statistical significance remained strong (p=0.007) (Vives i Batlle et al. 2009).

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, assuming K_d is known.

An anomaly when considering the allometric relationships available for marine organisms with those 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. A potential reason for this with respect to *CR* for marine organisms is the relationship between $\log_{10}(K_d)$ and $\log_{10}(a)$ which implies that the more particle-reactive the nuclide is, the more strongly 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). Supporting this suggestion, it has been shown that dead cells of phytoplankton had the same uptake of transuranic elements as live cells, indicating that the uptake process is passive (Fisher et al., 1983). Similarly, Ginn and Fein (2008) observed that metal adsorption on bacteria was the same for many different bacteria species, suggesting that the surface area rather than the biology of the bacteria is important in metal transfer. For a perfectly isometrically scaled organism, all surface area-based properties change with mass to the power of $^2/_3$ (Galilei, 1638), this would result in the *CR* scaling to $M^{-0.33}$ instead of $M^{-0.25}$.

4. An alternative to the $\mathrm{CR}_{\text{wo-media}}$ model for wildlife?

There are many cases where empirical data to derive $CR_{wo\text{-}media}$ are lacking. For instance, in the revised version of the ERICA Tool (released 2014 http://www.erica-tool.com/news/news-articles/2014/erica-assessment-tool-version-1.2/) (Brown et al. 2008), data were available for only 622 of 1521 required values. Furthermore, this parameter is highly variable as it incorporates many processes and will be largely determined by site-specific characteristics (e.g. Beresford et al. 2008a; Johansen et al. 2012; Wood et al. 2009b; 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 (Willey, 2010), including 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) and Co (Willey and Wilkins, 2008) (Figure 6). 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 radionuclide transfer to other organism types was demonstrated by Jeffree et al. (2010; 2013). Jeffree et al. suggested that the transfer of a number of radionuclides to marine teleost and chondrichthyan fishes, and to the amphioxus (fish like chordate) species, *Branchiostoma lanceolatumis*, was influenced by phylogeny. However, the work of Jeffree et al. was based upon the results of laboratory studies that only considered the exposure pathway from radionuclide contaminated seawater. Although this usefully removes the influences of many confounding factors, it is not directly applicable to environmental conditions as food chain transfer was excluded.

Beresford et al. (2013) used the approach of Willey (2010) to analyse data of Cs transfer to freshwater fish species. Starting with the database on radionuclide transfer to freshwater organisms as described by Yankovich et al. (2013) (see also Copplestone et al. 2013), they compiled a data set of 597 entries covering 53 species of freshwater fish from 67 sites. The

53 fish species all belonged to the class Actinopterygii (ray-finned fishes), with 10 taxonomic orders, 14 families and 33 genera being represented in the dataset.

The Residual Maximum Likelihood (REML) fitting of a mixed-model regression (as described by Willey 2010) was used to analyse the dataset. 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. The REML-adjusted means for different taxonomic groups of freshwater fish are presented in Table 6.

The results demonstrated differences in Cs transfer to freshwater fish based upon phylogenetically-derived taxonomic groupings, though they did not indicate a phylogenetic trend as previously seen for plants (e.g. Figure 6). However, although a relatively large dataset was considered, 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 phylogenetic 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).

Beresford et al. (2013) suggested that the REML-adjusted means potentially provide a more refined approach than the $CR_{wo\text{-}media}$ model. By taking into account inter-site variation, the REML-adjusted means in effect provide a mechanism of accounting for site-specific variables (e.g. K concentrations in water in the case of Cs transfer to fish). To independently test this hypothesis, data from 27 Finnish lakes (STUK, 2012; http://bit.ly/1xDJQu4), which had not been used in the model calibration, were analysed. Data were available for four or more fish species at each lake with a total of 11 fish species being represented. As *Perca fluviatilis* was present at all 27 sites and was also well represented within the dataset used for the REML analysis, it was used as the 'known species' from which to calculate activity concentrations for the other species (treated as 'unknowns'). This allowed 100 predictions to be made.

There was relatively good agreement between predicted and measured values, with a linear regression fit yielding an R^2 of 0.83 ($p<0.001$) and a slope (±standard error) of 0.98±0.04 ($p<0.001$) (Beresford et al. 2013). The intercept was not significantly different to zero.

Predictions were considerably better than if the appropriate feeding group (i.e. benthic feeding, piscivorous or forage feeding), geometric mean $CR_{wo\text{-}water}$ values from Yankovich et al. (2013) are used to predict the ^{137}Cs activity concentrations in the fish. The $CR_{wo\text{-}water}$ values generally resulted in under-predictions, with a linear regression of predicted-to-measured activity concentrations yielding a slope of only 0.31.

In Figure 7, we further test the model of Beresford et al. (2013) against recent data for three lakes in northern England (Barnett et al. 2015). Across the three lakes, data were available for vendace (*Coregonus albula*), Northern pike (*Esox lucius*), ruffe (*Gymnocephalus cernua*), perch (*Perca fluviatilis*), roach (*Rutilus rutilus*) and brown tout (*Salmo trutta*). Replication for each species ranged from 6 to 20, although not all species were sampled in each lake. Perch was present at all sites and was hence used as our 'known' species. Activity concentrations in the other species were then predicted as:

$$Cs\ concentration\ in\ 'unknown'species = \frac{REML-adjusted\ mean\ for\ unknown\ species}{REML-adjusted\ mean\ for\ perch} \times concentration\ in\ perch$$

The results of this comparison were again encouraging, yielding a significant linear regression ($p<0.03$) with a slope of 0.82, an R^2 value of 0.58, and an intercept not significantly different from zero. Water data were not available for these lakes so a comparison of predictions with those using *CR* values was not possible. However, this demonstrates a potential advantage of the alternative ('REML') approach, in that water concentrations, which are prone to comparatively large temporal variations, are not required.

5. Stoichiometry and ionomics

The terms, 'ecological stoichiometry' and 'ionomics', are closely related. Ecological stoichiometry (or ecostoichiometry) is 'the study of the balance of chemical elements in components, interactions, and processes in ecosystems' (Sterner & Elser 2002) and has its origins in aquatic ecology. Ionomics is the study of the total elemental composition of an organism, the ionome (Salt 2004), and the term has been used mostly within terrestrial botany.

5.1 Previous and current uses

Early studies in aquatic ecosystems mainly used ecological stoichiometry to investigate food web dynamics (Lindeman 1942; Lotka 1925), and carbon cycling and energy flows (Odum1960). Redfield (1958) recognised that the elemental composition of seawater and plankton was fairly constant across wide oceanographic areas and that biological processes could be controlled by elements other than C, such as N and P and trace elements. Since then, most studies have continued to focus on C, N and P and primary producers. More recently, ionomics studies focusing on macro- and micronutrients have been used to explore plant phylogenetics and physiology (Broadly et al., 2004; White et al., 2012). Elements other than C, N and P have also been used in the natural sciences to trace the origins of organisms or materials in the environment. For example, elemental signatures have been used to identify the biological origin of raw materials of biodiesels (Paredes et al. 2014) and the geographic origin of a range of food products (Gonzalvez et al. 2009). Multivariate methods (e.g. principal component analysis (PCA) and discriminant analysis), based on elemental composition, have been used to distinguish between ecosystem components and trophic groups in a coastal ecosystem (Kumblad & Bradshaw 2008), different angiosperm families in grassland habitats (White et al., 2012) and freshwater invertebrate taxa (Karimi and Folt, 2006). However, such approaches have not been widely used in radioecology.

In radioecology, it is known from field and experimental data that concentrations of certain elements/radionuclides are closely correlated to each other, to such a degree that they may be used as analogues for each other. The most common examples are radio-Sr/Ca and radio-Cs/K. The concentration of Sr or Cs in fish can be estimated from the water concentrations of Ca or K, respectively (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). Similar relationships between Ca and P (e.g. Hendrixson et al., 2007) or Ra and Ba (e.g. Gonneea et al., 2013) offer potential for further exploration.

Another approach that has been used in radioecology is based on the earlier ecological theory of organic matter (carbon) and energy flows in ecosystems. Radionuclide or element concentrations in ecosystem components are normalised to their C content, i.e. element:C ratios are calculated, 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). 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).

5.2 Natural variation in element composition

A key issue in ecostoichiometry is whether an element is essential or non-essential. The ability of organisms to maintain constant body concentrations, despite changing concentrations in the environment and/or their resource supply, (homeostasis) is generally assumed to be weak for autotrophs and strong for heterotrophs (Sterner & Elser 2002). Therefore, plant and algae stoichiometry is thought to more closely reflect that of the environment than animals, though this is much debated (e.g. Persson et al. 2010). The degree of homeostasis has been shown to vary depending on whether elements are macronutrients, essential micronutrients, or non-essential 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 concentrations in the body.

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; White et al. 2012). 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. A better understanding of these natural ranges would greatly help in our ability to predict element concentrations.

6. Discussion

We will never have data to populate all of the potential radioecological modelling parameters for wildlife (or indeed human) 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 some of these extrapolation approaches.

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. The concentration ratio ($CR_{product\text{-}diet}$ or $CR_{wo\text{-}diet}$) is a more generic parameter, and should enable the more abundant data for farm animal species to be applied to wild species.

Allometric expressions describing radionuclide biological half-life have previously been proposed for some elements. Beresford & Vives i Batlle (2013) present an approach, based on allometry, to predict acceptable $T_{1/2B}$ for a number of radionuclides and over orders of magnitude range in animal live-weight. However, we acknowledge that for a number of radionuclides (i.e. Am, Ce, Eu, Pu and Th), $T_{1/2B}$ has been reported to scale to *circa* $M^{0.8}$ and not the *circa* 0.25, as assumed in Beresford & Vives i Batlle (2013). To fully exploit the potential of allometric models, the reasons for this need to be explored.

Beresford & Wood (2014) demonstrated that the relationship derived by Beresford & Vives i Batlle (2013) for homoeothermic vertebrates is not directly applicable to poikilothermic reptiles. However, it is possible to parameterise the Beresford & Vives i Batlle model using reptile-specific values to obtain reasonable agreement between measured and predicted $T_{1/2B}$ values. Similarly it is possible to adapt the model parameters for other animal groupings using published information (e.g. as we have demonstrated above for different categories of mammals).

For aquatic ecosystems, the relationship between $\log_{10}(a)$ (on the allometric relationship for CR) and $\log(K_d)$ observed by Vives i Batlle et al. (2009) presents a potential opportunity to estimate CR values when data are lacking, assuming K_d is known.

The commonly used $CR_{wo\text{-}media}$ approach to estimating the radionuclide activity concentrations in wildlife is open to criticism, as $CR_{wo\text{-}media}$ values can be highly variable, largely due to site-specific factors. The analyses of available data using the REML analysis, as demonstrated by Beresford et al. (2013), 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 a new independent dataset well. 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. 2014). However, such data are highly site-specific, potentially limiting their wider applicability. The application of the REML approach to data, such as those presented by Barnett et al. (2014), to derive relative values for different organisms should provide a more generic set of ‘reference data’. 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 focused sampling of species falling within the RAP definitions at a number of terrestrial sites in different countries (Japan, Norway, Spain, UK and Ukraine).

Ecological stoichiometry shows potential as an extrapolation method in radioecology, either from one element to another or from one species to another. 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 analytical 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. Baxter et al. 2007; http://www.ionomicshub.org/home/PiiMS), which can be used to test stoichiometric assumptions and theories, and to develop extrapolation methods. Such 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, such as those developed by the food industry. There, 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 a descriptive or analytical way. 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).

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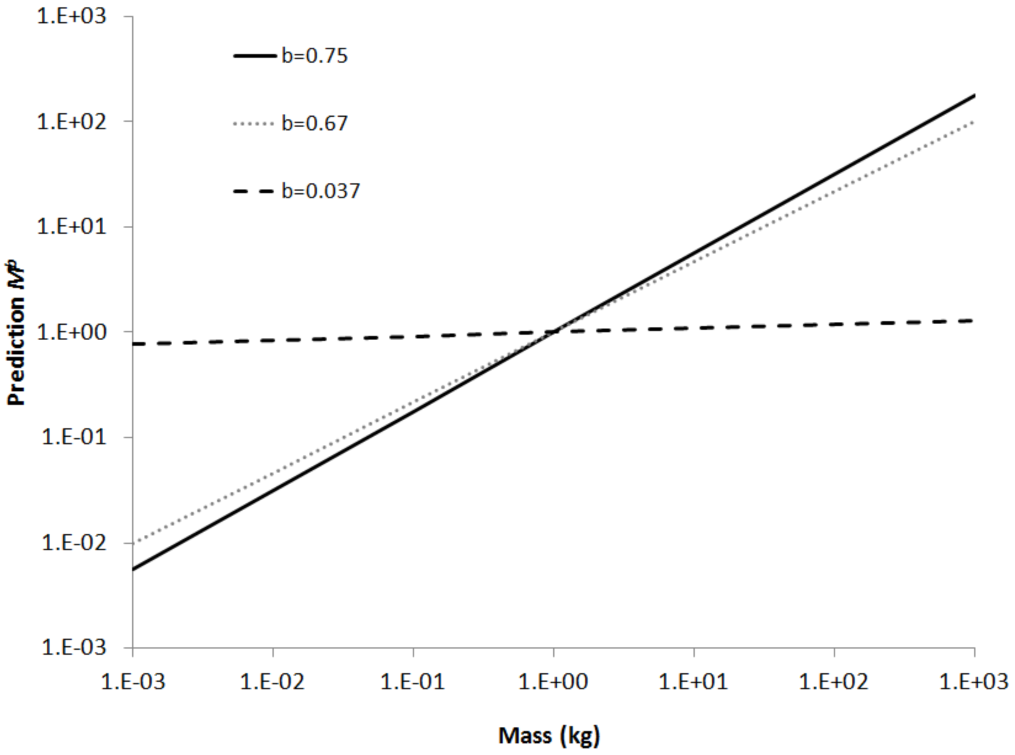

Figure 1. A comparison of predictions assuming allometric exponents of 0.67 and 0.75 across a mass range appropriate for terrestrial mammals (predictions are M^b). Predictions using an exponent of 0.037, appropriate to reptiles, are also shown.

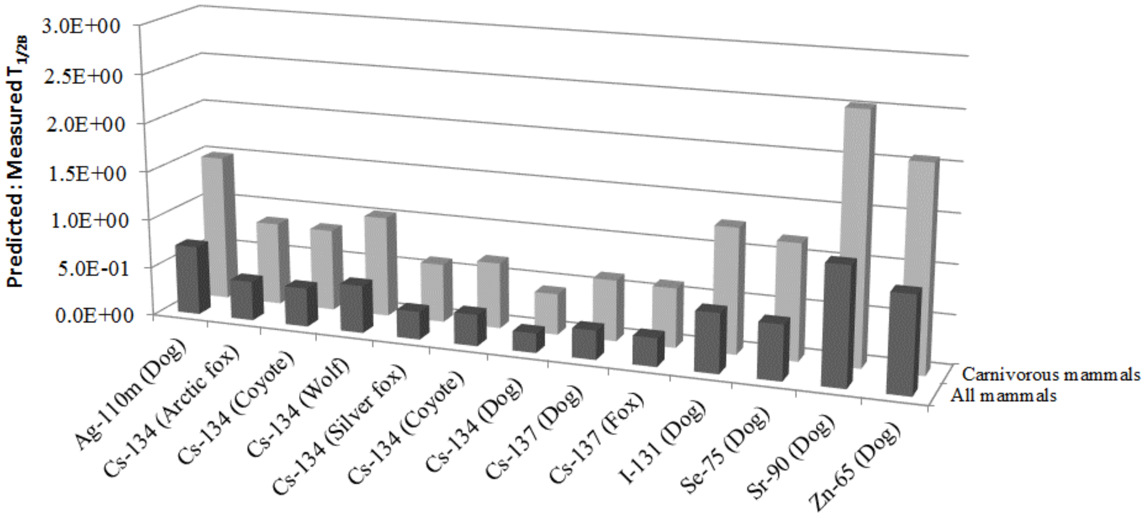

Figure 2. The effect of a_I value on the predicted $T_{1/2B}$ values for carnivorous mammals; note three outlying data points (one each for Nb, Ru and Sr) are not shown. Data represent individual studies/animals for which live-mass and biological half-life values were available.

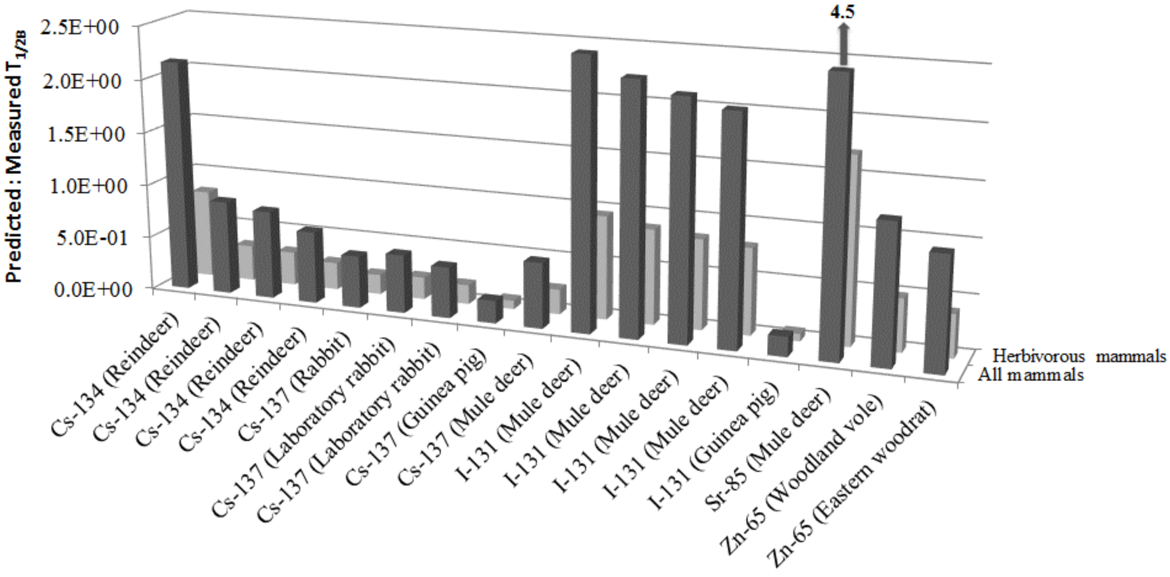

Figure 3. The effect of a_I value on the predicted $T_{1/2B}$ values for herbivorous mammals; note two outlying data points (one each for Cs and Sr) are not shown. Data represent individual studies/animals for which live-mass and biological half-life values were available.

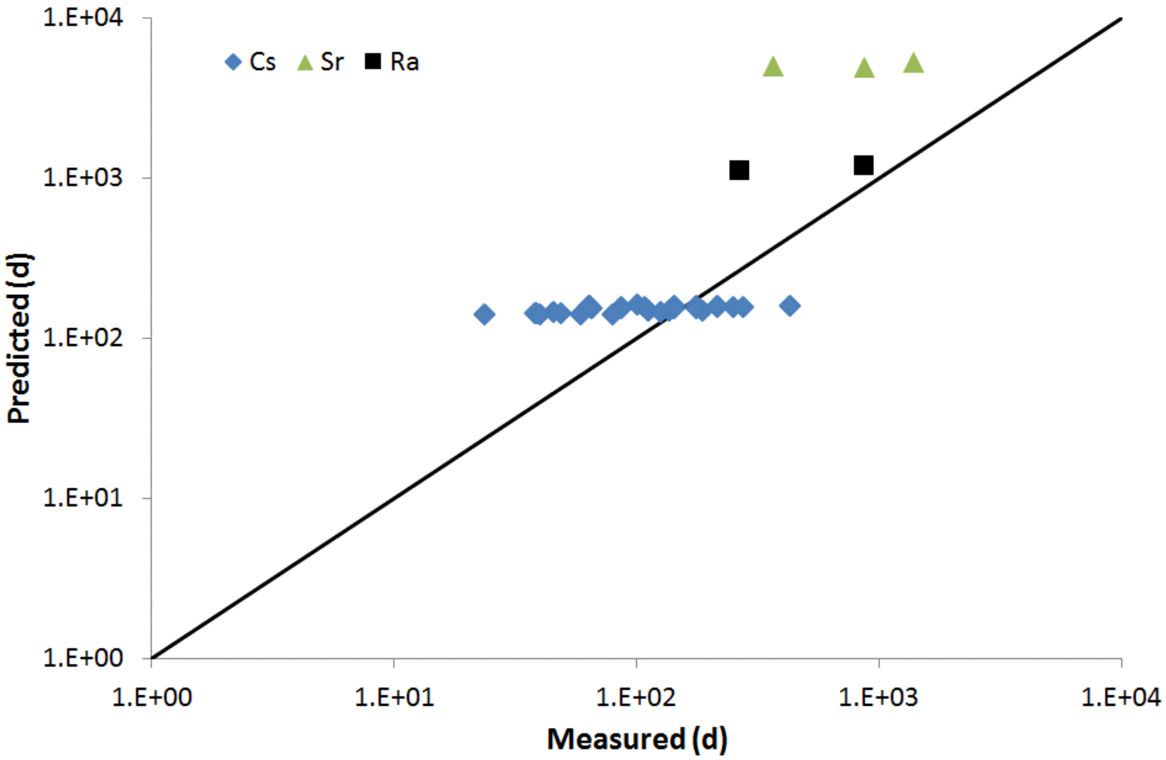

Figure 4. A comparison of measured radionuclide biological half-life (d) in reptiles with predictions using reptile specific parameter values in the model of Beresford & Vives i Batlle (2013) (from Beresford & Wood (2014).

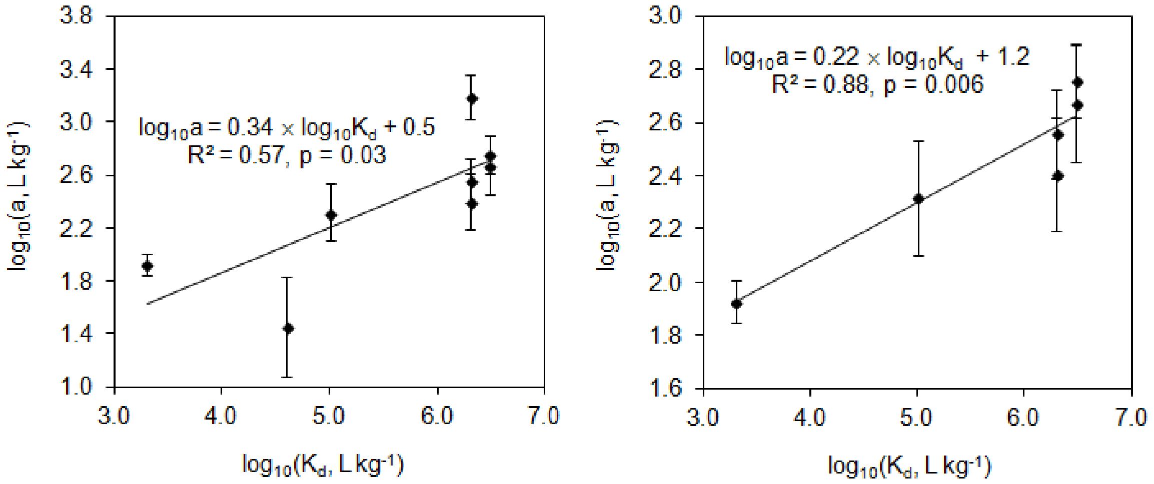

Figure 5. Linear trend between $\log_{10}(a)$ for the *CR* and sediment $\log_{10}(Kd)$ for marine ecosystems, both with (left) and without (right) Ru, Eu and Pm. Error bars represent the standard error of $\log_{10}(a)$ (adapted from Vives i Batlle et al. (2009)).

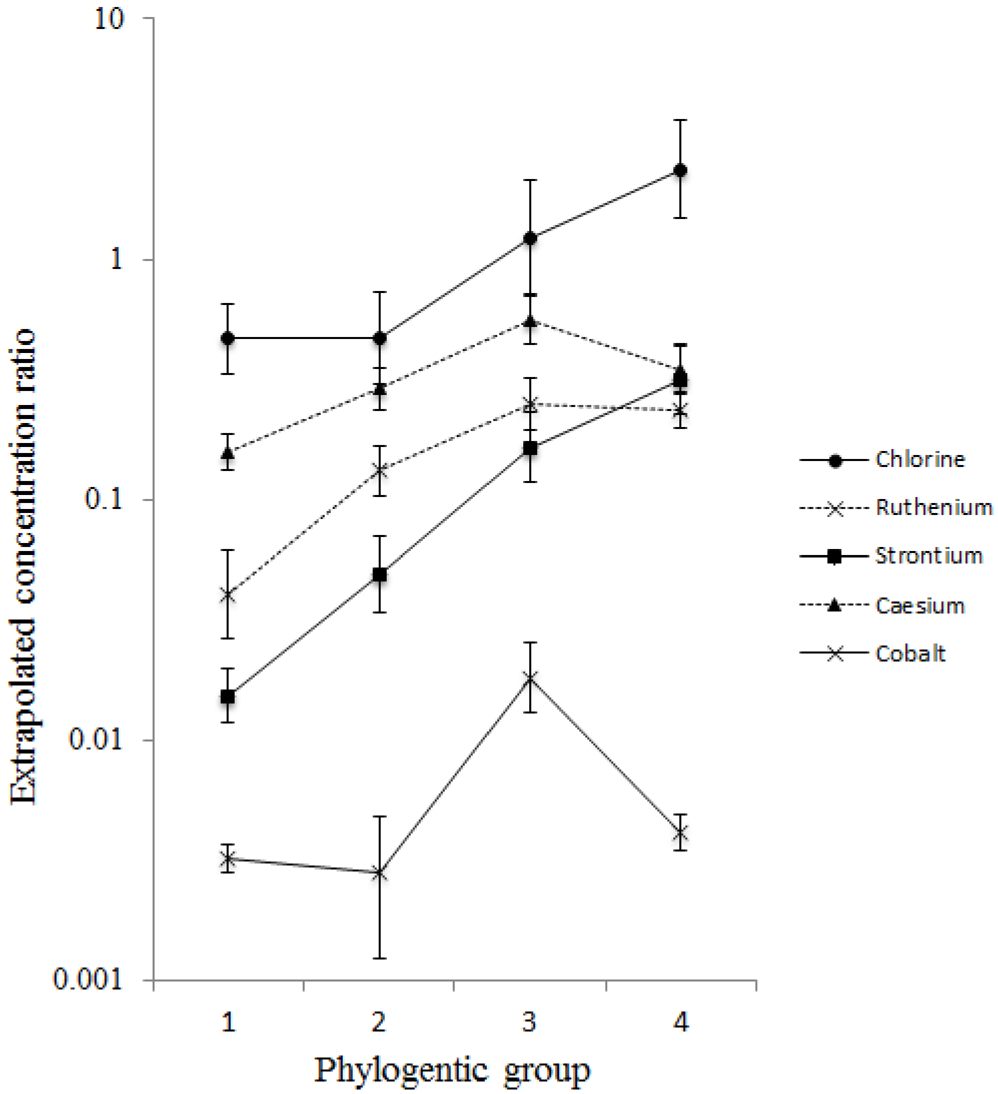

Figure 6. Extrapolated concentration ratios for radionuclides of chlorine, caesium, ruthenium, cobalt and strontium. Published REML-adjusted data (Broadley et al. 1999; Willey and Fawcett, 2005a,b; Willey et al. 2005; Willey and Fawcett, 2006, Willey and Wilkins, 2008), have been transformed to have a geometric mean of IAEA recommended transfer value for 'grass and herbs' (IAEA 2014) then divided into phylogenetic groups of the Angiosperm Phylogeny Group III phylogeny for flowering plants (1=Monocotyledonous plants, 2= Rosids, 3=Caryophyllids, 4= Asterids. Geomeans with 95% confidence Intervals. Cl n=106 species, Ru n=114, Sr n=155, Cs n=256, Co n=241).

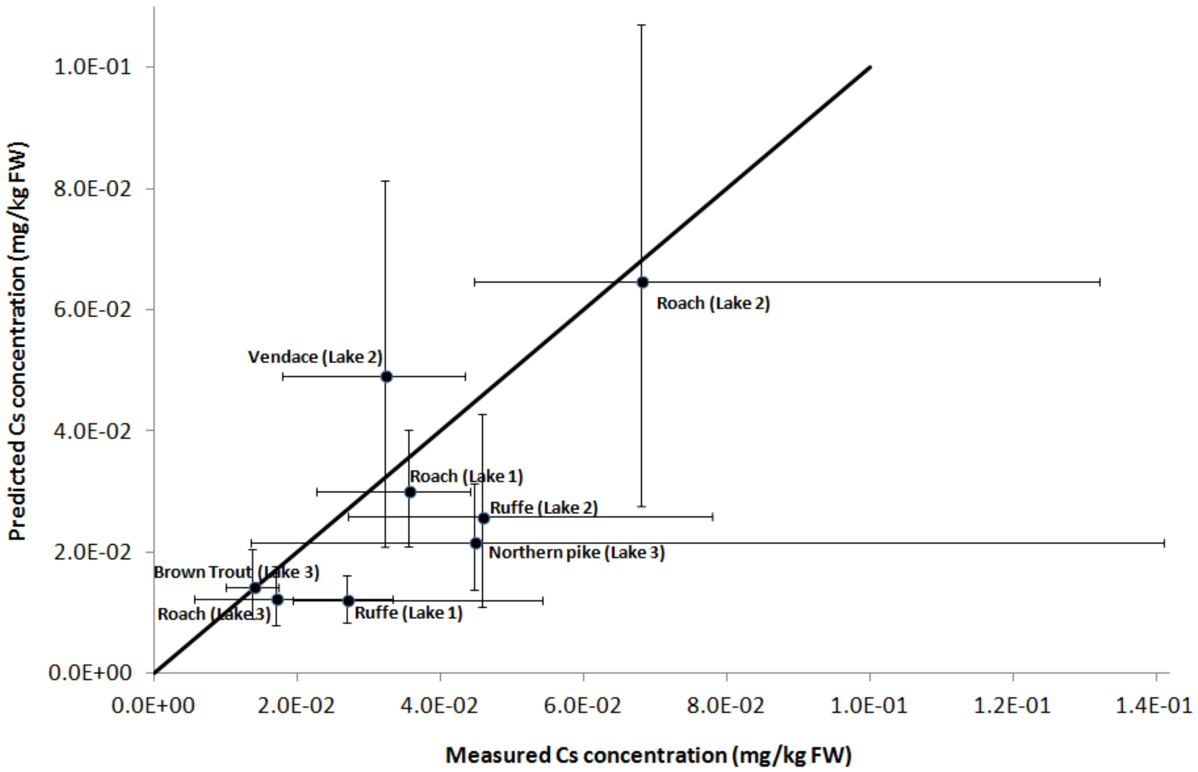

Figure 7. A comparison of measured mean Cs concentrations in fish of different species from three different lakes in northern England with values predicted using measured data for perch and the REML-adjusted means (see Table 6). Error bars are minimum and maximum values, and the solid line represent the 1:1 relationship between predicted and measured values.

Table 1. Caesium $CR_{meat\text{-}diet}$ values for wildlife; for both source 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. 2014
Odocoileus virginianus	Whitetail deer	0.53[+]	11	Sheppard 2013
Lepus europaeus	Brown hare	0.46	1	Chaplow et al. 2014
Lepus timidus	Blue hare	0.48±0.06	5	Chaplow et al. 2014
Oryctolagus cuniculus	Rabbit	0.27±0.11	20	Chaplow et al. 2014
Tetrao tetrix	Black grouse	0.40-0.85	2	Chaplow et al. 2014
Lagopus lagopus scotica	Red grouse	0.73±0.40	10	Chaplow et al. 2014

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

Table 2. Parameter values used to predict the biological half-life of radionuclides in mammals and birds (data sources are cited in text).

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

Table 3. A summary comparison of predicted with observed $T_{1/2B}$ values.

Element	**Animal type compared**[*]	**N**[+]	**Mean (range) predicted:observed using 'all mammal' a_I**	**Mean (range) predicted:observed using feeding group a_I**
Ag	Mouse species, rat, monkey, dog	4	3.1 (0.79-6.8)	2.5 (1.2-5.0)
Co	Mouse, rat	2	(1.2-8.5)	0.85-6.3
Cs	Mouse species, rat, monkey, dog, wild boar, rabbit, guinea pig, deer species, fox species, coyote, wolf	55	0.44 (0.08-2.2)	0.34 (0.03-1.0)
I	Mouse, rat, monkey, dog, guinea pig, deer	11	0.98 (0.14-2.5)	0.50 (0.07-1.3)
Na	Mouse, rat species, monkey	4	0.21 (0.13-0.31)	0.16 (0.10-0.23)
Nb	Mouse, rat, monkey, dog	4	0.04 (0.002-0.10)	0.04 (0.002-0.08)
Ru	Mouse, rat, monkey, dog	4	0.008 (0.001-0.02)	0.01 (0.0008-0.03)
Se	Mouse, rat, monkey, dog	4	0.49 (0.40-0.56)	0.55 (0.29-1.2)
Sr	Mouse, rat, rabbit, dog, deer	6	22 (1.2-50)	21 (1.7-74)
Zn	Mouse species, rat species, vole, monkey, dog, duck	29	1.9 (0.21-5.9)[++]	1.0 (0.15-4.4)[**]

[*] Where 'species' is used data were available for more than one species falling into the broad animal types listed; [+] Number of comparisons; [++] Mammals only (n=16); [**] Includes ducks (n=29).

Table 4. Allometric parameters describing the biological half-life (d) of radionuclides in marine organisms (adapted 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

Table 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. 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.

Table 6. REML-adjusted means for different taxonomic groups; these are relative values and not absolute values of $CR_{wo\text{-}water}$ (reproduced from Beresford et al. 2013).

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
						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

Order		**Family**		**Genus**		**Species**	
				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
						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

Responses to reviewers comments are detailed below.

Reviewer #1 comments:

The objective of the paper is to be applauded with its virtues of good 'radioecological housekeeping' both in the context of economic and eco-ethical cost-effectiveness. It will be a valuable contribution to the radioecological literature.

RESPONSE> We thank the referee for their comments.

A few issues are raised below but these are minor compared to this reviewer's very positive opinion of the paper.

a) For the assessment of the adequacy of the extrapolation approaches used it would be helpful to indicate *a priori* the levels of acceptability which are being adopted. The +/- 'order of magnitude' level of acceptability is indicated *a posteriori.* But what is the justification for this 'acceptable' degree of difference between predicted and empirical values? Modelling convention or radiological significance?

RESPONSE> Within the paper we highlight those predictions which fall more than an order of magnitude outside the measured values. This is used as a pragmatic approach to reviewing our predictions given the uncertainties (variation) in radiological parameters. It is generally accepted that allometry produces 'order of magnitude estimates and this is now stated and referenced.

b) The paper does set out to critically evaluate extrapolation techniques for radioecological transfer models using existing databases, but one is still left wondering about the adequacy of the biodiversity coverage in these databases. For example, only ducks represented wild birds, and it is known that passerines (not represented) have basal metabolic rates (BMR) and maximal existence metabolism that are 1.3 times higher than non-passerines (Gavrilov, 2014: QRB, 89 (2). Some additional comments from the authors on this concern would be useful.

RESPONSE> If coverage in databases was complete there'd be little need for the approaches we are discussing! That said we have included variation in metabolic rate by using group specific (e.g. herbivorous mammal etc.) parameters. This is of course best demonstrated for reptiles within our paper. A sentence has been added to the Discussion.

c) The REML approach needs some further clarification as to how and why it predicts, radioecologically.

RESPONSE>We are unclear as to what the reviewer want here with respect to 'how and why it predicts, radioecologically'. As described in the paper the output is a mean value for each species on a common scale having accounted for the effect of site as much as possible. This provides a relative scaling value.

d) With regard to the ecological stoichiometry section of the paper, the authors set out a promising research program for the future rather than a critical evaluation of existing databases. As such this component is not fully aligned with the thrust of the paper, which is to make the best of existing knowledge. Moreover, metabolic model and analogue pairing is not really a new idea, although its full value as an investigative and extrapolative tool has probably not been realised to date. Having said that I look forward to reading the outcomes of these radioecological explorations.

RESPONSE> The reviewer is correct in that this section is a little different than the others. However, the aim of our paper included to consider 'future potential' (see section 1) – though we have clarified the abstract on this respect. We make no claim that stoichiometry is new and indeed cite references as far back as 1925! The reviewer is correct that the full value as an extrapolative tool, for radioecology, has not been realised (or indeed properly considered). This section of our paper indeed highlights the potential of the approach and need for proper consideration.

Reviewer #2 comments:

Line 17. Consider adding "transfer" somewhere in the first paragraph. I think the paper is on extrapolation approaches for transfer parameters.

RESPONSE> Given the paper considers biological half-life in addition to CR the scope is broader than 'transfer' and we have not amended.

Line 18. Should read ". . . is shown"

RESPONSE> Corrected

Line 61. What "same workers?"

RESPONSE> Ward et al. …… have amended.

Line 64. Check "d kg^{-1}" for milk.

RESPONSE> d kg^{-1} is correct.

Line 81. Unclear. Do you mean the differences in dry matter intake that are inherent among wildlife types (a large snake eats less dry matter in a month than a rabbit of equivalent mass due to inherent food preference, absorption, and metabolic differences). If so, then these differences should be appropriately reflected in transfer. (Or perhaps you mean the differences are artefacts of experiments).

RESPONSE> We simply mean different species have different dry matter intake rates; sentence has been amended and this hopefully clarifies for all readers.

Line 95. What kind of CR? I think you mean $CR_{org\text{-}diet}$.

RESPONSE> We meant milk or meat CRs and text is now clarified accordingly.

Line 100. Yes, Cs is relatively homogeneous in organisms. However, this is not the case for many other radionuclides (e.g. Sr, Pu) as indicated in same reference (Yankovich et al. 2010)

RESPONSE>Accepted, though we are discussing data for Cs here as stated in the text. However, we have expanded the paragraph to comment on other elements.

Line 112. Shouldn't the full stop be after mass? b is dimensionless so adds nothing to defining the units of a.

RESPONSE> The text in the paper is correct.

Line 114. Check Journal style. Is usage correct for *circa* relative to mass (and other non-date values later in text)?

RESPONSE> To my knowledge (and I have checked the instructions to authors) the journal has no policy on the use of '*circa*'. However, we have standardise to *circa* rather than mixing *circa* and *c*. as in our earlier submission.

Line 175. Which test for ($p<0.001$)?

RESPONSE> This was a linear regression as stated – we have moved the '$p<0.001$' within the sentence to avoid any confusion.

Line 178. What is MODARIA?

RESPONSE> Website is provided for this.

Line 274. Consider "highly" instead of "in part."

RESPONSE> 'in part' is correct

Lines 307-310. Consider rewording. Both one component, and multi-loss component models, can either over- or under-predict depending on how they are parameterised.

RESPONSE> The assumption of a single long-term loss component for something which shows multi-components of loss should yield predicted activity concentrations which are conservative. Sentence has been amended to improve clarity to reader.

Lines 376-381. Something doesn't follow in Lines 376-381 which refer back (I think) to the marine organism data in the previous paragraph. But the *a* values (for $\log_{10}(a)$) were for Tc, Cs, Pu and Am (line 369) from which Ru, Eu, and Pm are removed? So, it appears that statistics and information in these two paragraphs (369-384) are being presented on subsets of a larger marine organism data set. Need to clarify.

RESPONSE> The two paragraph sequentially discuss $T_{12/B}$ (first paragraph) and then CR (second paragraph). There was no larger dataset within the Vives I Batlle et al. papers which requires description.

Lines 407-478. This section effectively introduced the REML approach as an alternate to (adjustment of) the standard CR approach. However, if the authors are intending to demonstrate an improvement of the alternate REML approach over the standard CR approach, the section falls short. The figures and tables do not compare the two approaches, rather they present REML adjusted values alone, or, in some cases, compare these with measured values. Lines 461 and 467 mention an improvement on the predicted-measured slope of the REML values compared with the standard CR values for one set of data. However the R^2 values are not provided for both, and a comparison showing some improvement of the REML values aren't presented in a figure or table or otherwise elaborated.

RESPONSE> We cannot do this as water data are not available for these lakes. Have amended paer to make this clear and also suggest this as a possible advantage (i.e. can make predictions with no water data).

Line 478. Agree that R^2 of 0.58 is encouraging. Suggest it also warrants discussion of why the correlation isn't higher.

RESPONSE> Any discussion on this would be purely speculation, though note there are only 8 points compared in this figure. However, we have added the level of significance of the linear relationship which was missing from the earlier manuscript.

Line 564. Suggest reword “we have to.”

RESPONSE> Reworded

Line 565. *Circa* is spelled out here, but c. before. See earlier comment on *circa*.

RESPONSE> See response above

Line 575. Suggest the authors should acknowledge the high uncertainty associated with Kd values, as well as discuss the fundamental issue of combining the highly variable Kds (an equilibrium concept that is arguably almost never found in equilibrium in open water bodies in nature), with the highly variable CRs (which is also an equilibrium concept that is also arguably almost never found in equilibrium in nature).

RESPONSE> Variability in CR is acknowledged in the first paragraph of section 4 and indeed is a justification for suggesting our alternative (REML) approach. There is likely a need for a critical evaluation of Kd’s – but we do not feel our paper is the place for this.

Lines 599-620. Is this entire paragraph on stoichiometry? It seems to move to other topics. Consider clarifying or breaking it up.

RESPONSE> This paragraph only discusses stoichiometry – but minor amended made to increase readability.

Figures 2 and 3. There are duplicates along the x axis that aren’t explained.
Figures 2 and 3. It is not clear what the y values represent.

RESPONSE> Data are presented for individual studies (hence 4x Cs-134 reindeer in Figure 3). We have clarified the figure legend. Y-axis legend added to both figures.

Figure 5. What kind of samples, media, etc. are the *a* values for? What kind of ecosystem is this figure about?

RESPONSE> Title amended.

Figure 6. Suggest rewording the title to better connect the first and second sentences. Also, provide ref if these are published data.

RESPONSE> Title amended.