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1 **Original Paper**

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6 **Estimating the biological half-life for radionuclides in homoeothermic**
7 **vertebrates: A simplified allometric approach**

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Abstract The application of allometric, or mass dependent, relationships within radioecology has increased with the evolution of models to predict the exposure of organisms other than man. Allometry presents a method of addressing the lack of empirical data on radionuclide transfer and metabolism for the many radionuclide-species combinations which may need to be considered. However, sufficient data across a range of species with different masses are required to establish allometric relationships and this is not always available. Here an alternative allometric approach to predict the biological half-life of radionuclides in homoeothermic vertebrates which does not require such data is derived. Biological half-life values are predicted for four radionuclides and compared to available data for a range of species. All predictions were within a factor of five of the observed values when the model was parameterised appropriate to the feeding strategy of each species. This is an encouraging level of agreement given that the allometric models are intended to provide broad approximations rather than exact values. However, reasons why some radionuclides deviate from what would be anticipated from Kleiber's law need to be determined to allow a more complete exploitation of the potential of allometric extrapolation within radioecological models.

Keywords: Allometry, biological half-life, metabolic rate, radionuclide, environmental assessment

Introduction

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

$$Y = aM^b \quad (1)$$

where a and b (the allometric exponent) are constants.

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 liveweight) 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 and Weibel 2005; Higley 2010).

In this paper the use of allometric relationships in radioecological models is explored and in particular a simple solution is suggested to enable their more widespread application to homoeothermic vertebrates.

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. 2008; Johansen et al. 2012) including the US Department of Energy's RESRAD-Biota model (USDOE 2002).

Moreover, there are specific radioecological parameters which have been shown to scale allometrically, with relationships for biological half-life across species having been 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. 2008; Sheppard 2001) and aquatic ecosystems (Vives i Batlle et al. 2007; 2009; Brown et al. 2004). The reason for this attention is the potential of allometry to help address the lack of data for the large number of organism-radionuclide combinations which may need to be assessed (Beresford et al. 2004; Higley et al. 2003; IAEA in-press).

In the marine environment, allometric relationships have been found across plankton, seaweed, fish, crustaceans and molluscs (Vives i Batlle et al. 2007; 2009) for the concentration ratio (CR) of activity concentrations in organisms to those in water ($M^{-0.26 \pm 0.09}$) and also the biological half-life of elimination ($M^{0.16 \pm 0.03}$). The CR scaled better allometrically for particle seeking radionuclides, mainly lanthanides and actinides. A relationship between the independent term of the allometric power function (a) and the sediment-water partition coefficient (k_d) was also observed. This was strongest for particle seeking radionuclides, suggesting the importance of particle-reactive material sorbed onto food and ingested in particulate form relative to conservative radionuclides which tend to stay largely in the aqueous phase.

For terrestrial organisms, allometric relationships have also been suggested 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, Beresford et al. (2004) demonstrated that this was the consequence of the dependence of daily dry matter intake on mass and that the ratio between the activity concentration in the animal and that in feed is independent of mass.

USDOE (2002) presents allometric relationships for the biological half-lives of 16 elements in terrestrial/riparian vertebrates. Many of these have an exponent of approximately 0.25, which 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:

$$\frac{dy}{dt} = -\varepsilon_a \frac{B_r}{M} \left[y - \frac{Q_1^A A_f}{Q_0^A} \right] \quad (2)$$

104

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

$$y = y_0 e^{-\varepsilon_a \frac{B_r}{M} t} + \frac{Q_1^A A_f}{Q_0^A} \left(1 - e^{-\varepsilon_a \frac{B_r}{M} t} \right) \quad (3)$$

113 Where y_0 is the activity concentration of the organism at $t = 0$, *i.e.* at the beginning of
 114 depuration. If $A_f = 0$ and $y_0 \neq 0$ (representing a depuration process) Eq. 3 becomes reduced to
 115 a simple exponential, and applying the definition of biological half-life (ie $y = y_0 e^{-\frac{\ln 2}{T_{B1/2}} t}$)
 116 yields:

$$T_{B1/2} = \frac{M \ln 2}{\varepsilon_a B_r} \quad (4)$$

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

$$T_{B1/2} = \frac{\ln 2}{a \varepsilon_a} M^{0.25} \quad (5)$$

120 This is in agreement with the exponent values quoted by USDOE (2002) for many
 121 radionuclides (note, however, that some radionuclides within USDOE do not scale as
 122 approximately 0.25 as discussed later).

123 The application of allometric biological half-life relationships allows broad approximations to
 124 be made to help address the limitations of the current empirical data for wildlife. However, to
 125 derive such relationships, adequate data are required for a given element and for a number of
 126 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, 0.25), then only an estimation of the multiplicand is needed for any given element. In the following section, a method of estimating this multiplicand is derived and, hence, the applicability of allometric approaches to estimating biological half-life is extended.

Materials and methods

Extending the application of allometry in radioecology

If one starts by considering a simple first-order linear retention model with constant input:

$$\frac{dy}{dt} = \frac{f_1 A_f I_r}{M} - \frac{\ln 2}{T_{B1/2}} y \quad (6)$$

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 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 ($t=\infty$), Eq. 6 equals zero and the equilibrium activity concentration in the organism (y_{eq}) is given by:

$$y_{eq} = \frac{A_f f_1 I_r T_{B1/2}}{M \ln 2} \quad (7)$$

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}):

$$CR_{org_diet} = \frac{f_1 I_r T_{B1/2}}{M \ln 2} \quad (8)$$

If it is assumed 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:

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

This gives:

$$CR_{org-diet} = a_B a_I \frac{f_1}{\ln 2} \frac{M^{0.75} \times M^{0.25}}{M} \quad (9)$$

Therefore, mass cancels out and:

$$CR_{org-diet} = a_B a_I \frac{f_1}{\ln 2} \quad (10)$$

If it is accepted that $CR_{org-diet}$ approximates to a constant for a given element across all species as suggested in Beresford et al. (2004) (and later accepted for farm animals in IAEA (2010)) then a solution to a_B can be proposed:

$$a_B = \frac{\ln 2}{a_I f_1} CR_{org-diet} \quad (11)$$

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

$$T_{B1/2} = \frac{\ln 2}{a_I f_1} CR_{org-diet} M^{0.25} \quad (12)$$

Values of a_I are relatively well documented for terrestrial vertebrates (e.g. Nagy 2001). If the ingestion rate is known, then a_I can be substituted by: $(I_r \times M^{0.75})$

Testing the hypothesis

For this test the primary source of $T_{B1/2}$ values was Whicker and Shultz (1982), who tabulated 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 and Staaland (1994). Observed $T_{B1/2}$ values from these sources for Cs, I, Sr and Co are given Table 1. 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 and 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)¹ were used for ‘all mammals’ ($a_I=0.057 \text{ d}^{-1} \text{ kg}^{0.25}$), carnivorous mammals ($a_I=0.027 \text{ d}^{-1} \text{ kg}^{0.25}$) and herbivorous mammals ($a_I=0.15 \text{ d}^{-1} \text{ kg}^{0.25}$); the a_I value for rodents ($a_I=0.059 \text{ d}^{-1} \text{ kg}^{0.25}$) is similar to that of ‘all mammals’. Estimates of f_I have been taken from IAEA (2010) which cites values from ICRP (2006) for monogastric animals and additionally presents f_I for ruminants.

To estimate values of $CR_{org-diet}$ $CR_{meat-diet}$ values presented by IAEA (2010) were used which relate the fresh weight activity concentration in meat (i.e. muscle) to the dry matter activity concentration in the diet of farm animals (Table 1). 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 (i.e. the ratio of the activity concentration of Sr in meat to the daily intake of Sr) presented in IAEA (2010) have been used together with typical dry matter intake rates from IAEA (1994) to estimate the average $CR_{org-diet}$ across all five species for which data are available (cattle, goat, sheep, poultry and pig) in IAEA (2010) (Table 1).

Yankovich et al. (2010) present tissue to wholebody radionuclide activity concentration conversion factors for a range of wildlife groups which could be used to derive wholebody $CR_{org-diet}$ estimates from $CR_{meat-diet}$ values. However, Cs is the only element of interest here for which Yankovich et al. (2010) report a conversion factor for mammals. Therefore, for Sr and Co, conversion factors were estimated using data presented in Barnett et al. (2013) for wood mice and roe deer; a conversion factor for I has been estimated from information presented in Coughtrey et al. (1983) (Table 1).

Results and Discussion

The a_I for ‘all mammals’ was used to predict $T_{B1/2}$ values for all available comparisons (Table 2). All predictions were within an order of magnitude of the observed values with most being within a factor of three. This can be considered to be satisfactory, given that the allometric models are designed to give a broad approximation rather than an exact value. For Cs and I there is a tendency to under-predict, whereas for Co all estimates are over-predictions. If the a_I suggested for carnivorous mammals by Nagy (2001) is used there is a marked improvement in predictions for Cs and I for carnivorous species (Table 2). However, if the a_I

¹ Nagy (2001) presents relationships based upon mass in units of grammes, we have converted these to kilogrammes

applicable to herbivorous mammals is applied, then the predictions for the relatively few herbivores for which there were data are underestimated with the exception of the estimates for Co in laboratory rabbit, and Sr in mule deer (Table 2).

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 2) yields an R^2 value of 0.58 with a slope of 1.4 and an intercept which is not significantly different from zero ($p < 0.001$).

As both mule deer and reindeer are ruminants, predictions for these animals were also made using the ruminant specific f_I 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_I values was 1,000 days compared with the observed value of 228 days.

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 (e.g. I is based upon six studies and Co on three) and the value for Sr had to be estimated as described 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-prediction of $T_{B1/2}$ for housed (i.e. experimental) animals as was observed for Cs and I (Table 2).

An assumption of the approach developed 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 Publication 30 (ICRP 1979; 1981; 1988) is quoted as

the data source but unfortunately it not possible to find the data within this publication to independently verify the allometric equations presented by USDOE (2002). None of these elements 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, it is noted 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 $CR_{org-diet}$ 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. 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.

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 and Weibel 2005; Isaac and Carbone 2010; Agutter and 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 and 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, and this suggestion is supported here.

Conclusions

Based upon the above evaluation, Eq. 12 could be applied to make predictions of $T_{B1/2}$ values for application in wildlife assessment models expanding upon the limited range of radionuclides for which allometric $T_{B1/2}$ equations are currently available (USDOE 2002). The approach presented requires that values of $CR_{org-diet}$ and f_I are available for the radionuclide of interest. These parameters are presented in, or can be estimated from, existing compilations for many radionuclides (e.g. IAEA 2010); a basic premise of the suggested model is that the two parameters are not species specific. However, reasons why some radionuclides deviate from what would be anticipated from Kleiber’s law (see Eq. 5) need to

be determined to enable more complete exploitation of the potential of allometric extrapolation within radioecological models.

The current assumption of a single long-component of loss in some wildlife assessment models (e.g. USDOE 2002; Avila et al. 2004) should yield estimates of the equilibrium activity concentration in organism 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. Theoretically it is possible to generalise an expression to suggest that both the short and long-term biological half-lives have an allometric exponent of 0.25 although this requires validation with suitable data.

Although not tested here it is recommended that the application of Eq. 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.

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Table 1 Values used to parameterise Eq. 12; conversion factor is defined as ratio of the fresh mass wholebody activity concentration to that of muscle (meat) (Barnett et al. 2013; Coughtrey et al. 1983; IAEA 1994, 2010; Yankovich et al. 2010).

Element	f_1	$CR_{meat-diet}$	Conversion factor	$CR_{org-diet}$
Co	0.1	3.1×10^{-1}	3	9.3×10^{-1}
Cs	1	3.9×10^{-1}	1	3.9×10^{-1}
I	1	9.4×10^{-2}	5	4.7×10^{-1}
Sr	0.3	2.2×10^{-2}	400	8.9

Table 2 Comparison of reported $T_{B1/2}$ values with estimates from Eq. 12 assuming different a_I values.

Speices	Mass (kg)	T _{B1/2} reported	Predicted T _{B1/2} using <i>a_I</i> for:			Reference ID
			All mammals	Carnivores	Herbivores	
<i>Radiocaesium</i>						
Harvest mouse	1.0x10 ⁻²	3.7	1.5			1
Laboratory mouse	2.0x10 ⁻²	5.1	1.8			1
Whitefooted mouse	2.1x10 ⁻²	3.5	1.8			1
Cotton rat	1.3x10 ⁻¹	8.4	2.8			1
Laboratory rat	1.9x10 ⁻¹	6.3	3.1			1
Rabbit	1.6	11	5.3		2.0	2
Arctic fox	4.9	17.5	7	15		3
Silver fox	5.3	25.3	7.1	15		3
Coyote	9.5	26	8.3	17		3
Red fox	10	29	8.4	18		1
Coyote	12	22	8.7	18		3
Dog	19	28	9.8	21		1
Wolf	31	23	11	24		3
Mule deer	55	14	13		4.8	1
Reindeer	80	14	14		5.3	1
<i>Radiocobalt</i>						
Whitefooted mouse	2.0x10 ⁻²	5.2	42			1
Laboratory mouse	2.5x10 ⁻²	4.8	45			1
Laboratory rat	4.0x10 ⁻¹	11	89			1
Guinea pig	4.7x10 ⁻¹	21	93			1
Laboratory rabbit	3.0	13	148		56	1
<i>Radioiodine</i>						
Laboratory mouse	2.1x10 ⁻²	5.2	2.2			1
Cotton rat	1.1x10 ⁻¹	8	3.3			1
Laboratory rat	2.1x10 ⁻¹	2.5	3.8			1
Guinea pig	5.0x10 ⁻¹	26	4.8			1
Jack rabbit	1.9	5	6.7		2.5	1
Laboratory rabbit	3.7	13	7.9		3.0	1
Dog	12	17	10	22		
<i>Radiostrontium</i>						
Laboratory mouse	3.010 ⁻²	43	140			1
Laboratory Rat	2.010 ⁻¹	590	240			1
Dog	1	530	640	1,300		1
Mule deer	65	190	1,000		380	1

References: (1) Whicker and Shultz (1982); (2) Battiston et al. (1991); (3) Gaare and Staaland (1994).