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15 **Radionuclide biological half-life values for terrestrial and aquatic wildlife**

16 *Abstract*

17 The equilibrium concentration ratio is typically the parameter used to estimate organism
18 activity concentrations within wildlife dose assessment tools. Whilst this is assumed to be fit
19 for purpose, there are scenarios such as accidental or irregular, fluctuating, releases from
20 licensed facilities when this might not be the case. In such circumstances, the concentration
21 ratio approach may under- or over-estimate radiation exposure depending upon the time since
22 the release. To carrying out assessments for such releases, a dynamic approach is needed. The
23 simplest and most practical option is representing the uptake and turnover processes by first-
24 order kinetics, for which organism- and element-specific biological half-life data are
25 required. In this paper we describe the development of a freely available international
26 database of radionuclide biological half-life values. The database includes 1907 entries for
27 terrestrial, freshwater, riparian and marine organisms. Biological half-life values are reported
28 for 52 elements across a range of wildlife groups (marine=9, freshwater=10, terrestrial=7 and
29 riparian=3 groups). Potential applications and limitations of the database are discussed.

30 **1. Introduction**

31 To estimate the uptake of radionuclides by wildlife, the whole organism¹ concentration ratio
32 ($CR_{wo-media}$) is most commonly used (e.g. Beresford et al. 2008a; Hosseini et al. 2008; Strand
33 et al. 2009; Yankovich et al. 2013; Howard et al. 2013; IAEA 2014). This is defined as the ratio
34 of radionuclide activity concentration in the whole organism to that in the surrounding medium:

$$35 \quad CR_{wo-media} = \frac{\text{Activity concentration in whole organism (Bq kg}^{-1}\text{) fresh mass}}{\text{Activity concentration in media}}$$

36 where, media maybe soil (Bq kg⁻¹ dry mass), water (Bq L⁻¹) or air (Bq m⁻³) dependent upon
37 ecosystem and radionuclide.

¹Organism less both gastrointestinal tract contents and fur/feathers

38 The concentration ratio is an aggregated transfer parameter, incorporating within it the
39 physical, chemical and biological factors affecting the uptake of radioelements by biota in an
40 empirical way. The turnover of elements differs depending on, for instance, ingestion or
41 sorption processes, their chemical and biochemical behaviour and the requirements of the
42 organism for the element or its analogue. Instantaneous equilibrium between the organism and
43 the media activity concentrations is assumed in all models that use the $CR_{wo-media}$ concept (e.g.
44 USDOE 2002; Brown et al. 2008; Beresford et al. 2008b).

45 Although some organisms may equilibrate relatively rapidly with radionuclides present in the
46 surrounding media (timescales in the order of days to a few months), there are scenarios
47 whereby equilibrium cannot be assumed. For example, after a short-term pulsed release of ^{99}Tc
48 activity into the marine environment, the activity concentration in lobsters along the dispersion
49 path begins to increase gradually with time. This is because lobsters have a biological half-life
50 for technetium in the range of 60-300 days (Pentreath 1981). Technetium is soluble in seawater
51 and the pulsed release will clear quickly from the area of sea where lobsters live. Therefore, its
52 concentration in seawater will decrease sharply within a few days after a pulse discharge.
53 Lobster specimens sampled from the area within days of the discharge, when water
54 concentrations have declined, may appear to have an anomalously high $CR_{wo-media}$ value
55 because they retain the technetium that they absorbed whilst seawater concentrations were
56 high. Conversely, if sampled on the day of discharge $CR_{wo-media}$ would be low as little uptake
57 would have occurred but seawater concentrations would be high.

58 If the timeframe of interest is long (e.g. years or decades of planned authorised discharges,
59 involving continuous releases or gradual changes in discharge concentrations) then the CR_{wo-}
60 $media$ approach is currently considered to be sufficient (Strand et al. 2009; IAEA 2014).
61 However, if unplanned release scenarios involving abrupt changes in discharge concentrations
62 are being modelled then the $CR_{wo-media}$ approach may be inadequate and dynamic models of
63 radionuclide transfer to biota may be a better assessment tool. This is especially true for
64 organisms that respond slowly to a change in ambient radioactivity concentration (Vives i
65 Batlle 2012), and this has been highlighted in the post-accident assessment of the Fukushima
66 accident (e.g. Kryshev & Sazykina 2011; Buessler et al., 2011). Such dynamic models need
67 to have rate constants, or biological half-life values ($T_{1/2b}$), describing the loss of radionuclides
68 from organisms. Whilst the biological half-life is typically defined to describe the rate of loss
69 of radionuclide from an organism, it is also often used in the estimation of uptake (e.g. Whicker
70 & Schultz 1982; Vives i Batlle et al. 2008).

71 At higher (more detailed) assessment tiers, the USDOE (2002) RESRAD BIOTA approach
72 incorporates some simple foodchain modelling ability using allometric (or mass) expressions;
73 these include allometric biological half-life relationships for a limited number of radionuclides
74 (Higley et al. 2003). Further exploitation of the allometric $T_{1/2b}$ approach to other radionuclides
75 was not possible because of a lack of $T_{1/2b}$ data from which to derive the relationships.

76 Commonly used assessment tools exploit the $CR_{wo-media}$ model with the tacit assumption that
77 this is generally likely to be conservative. However, it has been noted that wildlife
78 assessment models do not include direct deposition of radionuclides to vegetation surfaces
79 and that under conditions of continuous aerial discharge this may contribute a significant
80 proportion of radioactivity entering food chains (Copplestone et al. 2010). At the time of
81 writing this paper we are aware that the IAEA is working on an assessment approach for
82 wildlife which does include this deposition pathway (see Beresford et al. 2015a), but, which
83 as a consequence, requires some knowledge of the biological half-life of radionuclides in

84 wildlife. Similarly, reported $CR_{wo-media}$ values are, in theory at least, equilibrium values and an
85 increasing number of radioecological studies utilise inductively coupled plasma mass
86 spectrometry (ICP-MS) analyses to derive $CR_{wo-media}$ values from stable element data which
87 should be at equilibrium (e.g. Barnett et al. 2011,2014; Higley 2010; Tagami and Uchida
88 2010; Takata et al. 2010; Sheppard 2013). Application of an equilibrium $CR_{wo-media}$ value to a
89 short-lived radionuclide will over-estimate the resultant whole organism activity
90 concentrations and dose rate. IAEA (2010) propose an approach whereby equilibrium activity
91 concentrations could be corrected (to $CR_{wo-corrected}$) for application to short-lived
92 radionuclides, but this again requires some knowledge of biological half-life:

$$93 \quad CR_{wo-corrected} = CR_{wo-media} \times K \quad (2)$$

94 where:

$$95 \quad K = \frac{T_{1/2p}}{T_{1/2b} + T_{1/2p}} \quad (3)$$

96 where, $T_{1/2p}$ is the physical half-life of the radionuclide under assessment.

97 There are, as demonstrated here, a number of requirements for a comprehensive database of
98 wildlife radionuclide $T_{1/2b}$ values. However, such a database has to date not been available. In
99 this paper we describe the development of a $T_{1/2b}$ database for wildlife.

100 **2. Methods**

101 The work described here was conducted by an international working group under the
102 auspices of the International Atomic Energy Agency's MODARIA programme (see:
103 <http://www-ns.iaea.org/projects/modaria/>). The review and compilation of $T_{1/2b}$ values was
104 divided amongst the group members depending upon their prior expertise (i.e. by ecosystem
105 and/or organism).

106 Prior to beginning the review a recording sheet, in MS Excel™, was designed to allow easy
107 collation of the various components into the final database. The recording sheet entry fields
108 are listed in Table 1. The wildlife group categorisations were broadly compatible with those
109 used in the Wildlife Transfer Database (as described by Copplestone et al. 2013) and
110 subsequently by the IAEA (Howard et al. 2013; IAEA 2014). The 'Changeover time' is
111 defined as the time of intersection of two successive depuration curves which are governed
112 by their respective biological half-lives when the two are plotted as regression lines on a
113 logarithmic scale (this parameter is useful if fractions of loss in each component are not
114 directly reported). No attempt was made to standardise the English common names used;
115 similarly we have not updated Latin species names, but we acknowledge that these may have
116 changed since the original publications.

117 The review was undertaken to identify studies reporting either $T_{1/2b}$ values (i.e. the time taken
118 for the initial activity concentration in an organism, or tissue, to half) or elimination rate
119 constants from which $T_{1/2b}$ (d) values could subsequently be estimated as:

120
$$T_{1/2b} = \frac{\ln 2}{k} \quad (4)$$

121 Some $T_{1/2b}$ values were estimated from reported percentages of initial activity remaining (A_t)
122 at time t , where:

123
$$k = \frac{\ln\left(\frac{100}{A_t}\right)}{t} \quad (5)$$

124 and $T_{1/2b}$ is subsequently estimated from k as in Equation (4).

125 The review used Web of Knowledge (<http://wok.mimas.ac.uk/>), SCOPUS
126 (<http://www.scopus.com/>), IAEA INIS (<http://www.iaea.org/inis/>) and Pubmed
127 (<http://www.ncbi.nlm.nih.gov/pubmed>). Targeted searches of grey literature catalogues (e.g.
128 the U.S. Department of Energy portal: <http://www.osti.gov/scitech/>) were also conducted.
129 The on-line versions of key journals in the area (e.g. *Journal of Environmental Radioactivity*,
130 *The Science of the Total Environment*, *Radiation Protection Dosimetry*, *Health Physics*,
131 *International Journal of Radiation Biology*, *Journal of Radiological Protection* and *Journal*
132 *of Radiation Research*) were also searched to identify appropriate studies. Search terms
133 included: “biological half-life”, “biological period”, “kinetic transfer modelling”,
134 “accumulation rate”, “deuration rate” and organism names.

135 Reviews of the Japanese and Russian language literature to identify data appropriate for
136 inclusion in the database were also conducted. It was necessary to estimate k and hence $T_{1/2b}$
137 values from some of the Russian language literature by fitting appropriate exponential
138 relationships to reported data using MATCAD software. Some additional Russian language
139 values, for birds, were also obtained from the review of Fesenko et al. (2015).

140 In some instances, previously published reviews were used to identify source references.
141 Where possible the source references were consulted rather than relying on data from the
142 earlier compilation; these reviews are identified below. The reviews were also used as a
143 starting point for additional searches to identify any papers citing them which may have
144 appropriate data. We also benefited from the knowledge of working group members who
145 identified appropriate sources they were aware of, including in-house studies (e.g. PhD.
146 studies funded by the Institute for Radiological Protection and Nuclear Safety and published
147 in French).

148 For the terrestrial ecosystem the earlier key reviews used were those of Kitchings et al.
149 (1976), Whicker & Schultz (1982), Stara et al. (1971) and DiGregorio et al. (1978). The
150 freshwater ecosystem review of Alonzo (2009) was used as the primary source of freshwater
151 information with additional work focussing on identifying appropriate publications
152 subsequent to the year 2000. For the marine ecosystem, the important reviews were those of
153 Vives i Batlle et al. (2007; 2008; 2009) (covering principally ^{99}Tc , ^{129}I , ^{137}Cs , $^{239,240}\text{Pu}$ and
154 ^{241}Am), CIESM (2002) EPA (2013) Gomez et al. (1987) and Phillips & Russo (1978).

155 For the terrestrial environment, whilst they are not wild animals, data from the domesticated
156 dog (*Canis lupus familiaris*) and cat (*Felis silvestris catus*) have been included in the
157 database as these are unlikely to be collated anywhere else and are of relevance to some

158 species of wildlife; farm animal data were excluded from the database with the exception that
159 it is possible that some goose and duck data originate from domesticated species.

160 2.1 *Quality control*

161 An independent quality check was conducted by co-authors not involved in the actual data
162 compilation. As a minimum, 10% of the entered data were confirmed by going back to the
163 source reference. Additionally, for some key data sources all entries were checked. Any
164 issues raised were investigated and corrected as required in consultation with the originators
165 of those entries in the database.

166 Some of the compiled data have already been used in the development of allometric
167 approaches for terrestrial (Beresford & Wood 2014; Beresford et al. 2015b) and marine
168 (Vives i Batlle et al., 2007) wildlife and as such have been subjected to additional quality
169 checking. In the case of the reptile data this involved confirmation of most entries into the
170 database (Beresford & Wood 2014).

171 A column was added to the database to record which entries had been quality controlled
172 (approximately 40% of entries were quality controlled as described above). The database was
173 also reviewed for duplicate entries and the few identified were removed.

174 Some (n=165), predominantly freshwater, ecological half-life (i.e. parameter describing the
175 overall rate of reduction in radionuclide levels of animals in contaminated environments) data
176 for Cs and Sr were identified during the review and these have been retained within the
177 database (as a separate datasheet). It is important to note that these have not been quality
178 controlled and they are not discussed further below.

179 3. Results

180 The database resulting from the review is freely available (Beresford et al. 2015c;
181 <http://doi.org/10.5285/b95c2ea7-47d2-4816-b942-68779c59bc4d>); this contains all reference
182 details and hence these are not repeated in this paper.

183 The database contains 1907 entry lines split between organisms from three generic
184 ecosystems as follows: marine (n=547); freshwater (n=530); and terrestrial (n=743).
185 Additionally there are 87 entries for riparian organisms which live in both freshwater and
186 terrestrial ecosystems (i.e. amphibians and some species of reptiles and birds). Sixty-five
187 entries from the Japanese literature were for 'brackish water' species, in the database these
188 are classified as marine ('Brackish species' appears within the notes column to identify the
189 entries). Table 2 summarises the available $T_{1/2b}$ values by wildlife group (as defined in
190 Copplestone et al. (2013)) and by generic ecosystem. In some instance entries may be for
191 different tissues from the same study. We should also note that it is likely that the database
192 entries are a mix of mean values and single data.

193 The compiled database contained $T_{1/2b}$ values for a total of 52 elements (27 for freshwater, 48
194 for terrestrial, 24 for marine and 10 for riparian) (Table 2). For all three ecosystems data for
195 Cs were the most numerous entries (Figure 1), with some commonality in the other dominant
196 elements (i.e. Co, Sr, Ru and Mn). Iodine data were relatively numerous within the terrestrial
197 and marine organism data, but not within the freshwater data. Relatively more Co values
198 were available for freshwater and marine organisms than for terrestrial organisms; in the

199 marine ecosystem these data originate primarily for Japanese literature. The large number of
200 data classified as ‘other’ within Figure 1 for terrestrial organisms reflects the greater number
201 of elements for which data are available for this ecosystem (see Table 2) and that few data are
202 available for many of these. For marine organisms, entries for Tc were relatively numerous;
203 no data for the element were available for freshwater or terrestrial organism. Differences
204 between data availability are in-part driven by different radioecological issues between the
205 ecosystems and potentially by the focus of some of the key review sources used to begin to
206 establish the database (e.g. Tc was a focus of the review of marine data by Vives i Batlle et
207 al. (2007)).

208 Data on biological half-lives ranged from single components of loss to up to four components
209 (i.e. four $T_{1/2b}$ values) of loss recorded for some entries. In part, the number of reported loss
210 components is determined by the experimental approach with respect to the total study
211 duration and also the interval between measurements (Fesenko et al. 2015). For instance,
212 short-term studies will not provide estimates of longer components of loss, whilst if sampling
213 is at too large an interval early in the study the shortest components of loss may be missed.

214 Most entries were for the whole organism (n=1417). However, some entries (n=126) do not
215 have a record for the tissue sampled. In most instances, these records are for organisms where
216 it is highly likely that the values are for the whole organism or at least for soft tissues (e.g.
217 terrestrial arthropods, marine mussels, macroalgae). Other entries are for elements where the
218 value can be assumed to be representative of whole organism loss rates (e.g. Cs).

219 Supplementary information entered was, in some instances, sparse (e.g. few data on sex)
220 though many entries recorded study length (n=885), temperature (n=845), live-weight
221 (n=844) and age (n=904). With respect to age, entries were available for different live-stages
222 such as tadpole, fry and larvae.

223 The database is too diverse in terms of organisms, elements, administration routes and how
224 $T_{1/2b}$ values are presented (i.e. number of loss components) to attempt any analyses in this
225 paper.

226 **4. Discussion**

227 There are a number of reasons why $T_{1/2b}$ values are required to improve radiological
228 assessments (as discussed above). The database we describe here represents a significant
229 resource by which we may begin to improve, or test, available assessment approaches. For
230 some elements the database may also be useful to those assessing the exposure of wildlife to
231 metals (e.g. the database contains $T_{1/2b}$ values for Ag, Cr, Hg, Pb, Zn etc.).

232 The application of dynamic models to environmental assessment of radioactivity could be
233 criticised as being overly complex (IAEA, in-press). For many planned release situations it is
234 likely that the $CR_{wo-media}$ model will lead to appropriate assessments (Strand et al. 2009; IAEA
235 2014). Though, as noted above, for routine aerial releases the $CR_{wo-media}$ model may under
236 predict activity concentrations in organisms and the current approaches may not be optimised
237 for all planned release scenarios. Application of equilibrium $CR_{wo-media}$ models to accidental
238 situations or irregular pulsed discharges from licensed facilities is outside of the intended
239 scope of their application. In aquatic ecosystems, immediately after pulsed releases, when
240 media activity concentrations are high, $CR_{wo-media}$ models are likely to over-predict exposure
241 by not taking into account biological half-life (or uptake rate) and the consequent lack of

242 equilibrium. Subsequently they may under-predict as water concentrations decrease more
243 rapidly than those in exposed organisms; in the longer-term the $CR_{wo-media}$ model should
244 adequately predict exposure in the post-accident situation. In the terrestrial ecosystem,
245 immediately after a release the $CR_{wo-media}$ model may again over-predict exposure. However,
246 there is the possibility of under prediction when there is significant direct deposition on
247 vegetation surfaces (i.e. contamination is not dominated by the soil-plant route assumed in
248 the $CR_{wo-media}$ model). Whilst inappropriate, equilibrium $CR_{wo-media}$ models were applied to
249 assess risk to biota following the Fukushima accident (Garnier Laplace et al. 2011; Strand et
250 al. 2014). Although such assessments may have aimed to be conservative, as we demonstrate
251 above, the resultant predictions may not always have been conservative. Some attempts to
252 conduct more relevant assessments in the marine ecosystem using dynamic models have been
253 made (Kryshev et al. 2012; Vives i Batlle et al. 2014). However, more recent analyses
254 suggests that these models over predicted the rates of decline in fish (Johansen et al. 2015).
255 This was potentially because the models assumed single loss component (Vives i Batlle
256 2015); a more robust $T_{1/2b}$ database is expected to improve the parameterisation of such
257 dynamic models.

258 4.1 Use of the database

259 We envisage that the database will be useful in both parameterising and testing models.
260 Indeed some parts of the compiled database have already been used to test allometric models
261 for homoeothermic vertebrates (Beresford & Vives i Battle 2013; Beresford et al. 2015b) and
262 reptiles (Beresford & Wood 2014).

263 Users of the database will need to consider their needs and the suitability of the data that it
264 contains. For instance, we have included $T_{1/2b}$ values derived from studies using a number of
265 exposure routes. However, it is probable that for some elements intravenously administered
266 radionuclides will behave differently to those orally ingested (e.g. Mayes et al. 1996). The
267 study length and experimental protocol (e.g. time of first measurement, frequency of
268 measurements, time taken to reach limits of detection etc.) can influence the resultant $T_{1/2b}$
269 values and the number of components of loss observed. Experimental protocols of
270 insufficient length to enable multiple components of loss to be observed are likely to result in
271 $T_{1/2b}$ values which under-estimate the rate of loss (and consequently over estimate organism
272 activity concentrations) in the short-term, but, over-estimate the rate of loss in the longer-
273 term. Beresford & Wood (2014) present an evaluation of the database with respect to reptiles.
274 They highlighted that some of the entries were based upon only two time points and chose
275 not to use these data in their analyses. Future, users will need to similarly critically evaluate
276 the data. The present database provides them with a valuable resource from which to begin
277 this evaluation.

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Table 1. Parameters included in the database template.

Parameter
Entry ID
Common name (English)
Latin name
Wildlife group
Ecosystem (Marine, Freshwater, Terrestrial or Riparian)
Radionuclide
Live weight (kg)
Developmental stage (e.g. adult, tadpole etc.)
Compartment (whole organism or specific tissue)
Experiment type
Length of study (d)
Temperature (°C)
Biological half-life (d) (four columns were included to enable recordings of multiple loss components)
Fraction released (four columns, one for each component of loss)
Number of measurements (made in study) to determine $T_{1/2b}$
Measurement interval (d)
Changeover time (d) (repeated for multiple loss components)
Percentage left at time (t)
Time (t) (d) (referring to the above percentage left)
Organism dimensions (length, width, depth) (m)
Sex
Elimination rate (i.e. k ; d^{-1})
Reference
Notes
Comment on if the value has been independently quality controlled

Table 2. A summary by generic ecosystem and wildlife group of the T_{1/2b} values presented in the compiled database.

Wildlife group	Number of entries	Number of species*	Element
<i>Marine</i>			
Annelid	9	≥5	Cd, Cr, Tc
Crustacean	39	≥11	Cd, Co, Cs, I, Pu, Se, Sr, Tc, Zn
Echinoderm [#]	21	>4	Ca, Co, Cs, Mn, Tc, Zn
Fish	192	≥19	Ce, Co, Cr, Cs, Fe, I, In, Mn, Nb, Ru, Sr, Tc, Zn, Zr
Macroalgae	52	25	Am, Ce, Cs, I, Po, Pu, Sr, Tc, U, Zn
Mollusc	227	≥24	Ag, Am, Cd, Ce, Cm, Co, Cs, I, Mn, Pu, Ru, Sb, Se, Sr, Tc, Zn
Phytoplankton	5	n/a	Am, Pu
Sea anemones/true coral	1	1	Cs
Zooplankton	1	n/a	Am
<i>Freshwater</i>			
Algae	22	≥9	Ce, Co, Cs, Fe, P, Sr, Y
Bryophytes [#]	12	2	Ag, Am, Co, Cs, I, Mn, Ru
Crustacean	23	4	Am, Co, Hg, Ra, Ru, W
Fish	246	>23	Ag, Am, Ce, Co, Cs, Fe, H, Hg, I, Mn, P, Ra, Rb, Ru, Sr, Zn
Insect larvae	20	4	Am, Co, Cu, Ni, Pb, Ra, Ru, U, Zn
Mollusc	83	≥9	Ag, Am, Ca, Cd, Ce, Co, Cr, Cs, H, Mn, Na, Ra, Ru, S, Zn
Phytoplankton	22	≥3	Ag, Am, Co, Cs, Mn, Ra, Ru, Zn
Reptile	42	1	Sr, Ra
Vascular plant	24	≥8	Am, Ce, Co, Cs, Na, Sr
Zooplankton	36	1	Ag, Am, Co, Cs, Hg, Mn, Ra, Ru
<i>Terrestrial</i>			
Annelid	56	≥9	Cd, Co, Cs, Cu, Hf, Hg, I, Mn, Pb, Sc, Sr, Tb, U, Zn
Arachnid	11	≥6	Ca, Cs, K, Na, P, Zn
Arthropod	119	>53	As, Ca, Co, Cr, Cs, Cu, Fe, I, Ir, K, Na, P, Pb, Rb, Ru, Sr, W, Y, Zn
Bird	4	2	Cs, I
Mammal	522	≥40	Ag, Am, Au, Be, C, Cd, Ce, Cf, Co, Cr, Cs, Eu, Fe, H, Hg, I, In, Ir, K, Mn, Na, Nb, Np, P, Pa, Pb, Po, Pu, Ra, Rb, Ru, Sb, Sc, Se, Sn, Sr, Tb, Te, Th, U, W, Y, Zn, Zr
Mollusc	2	1	Cs, Na
Reptile	29	6	Co, Cr, Cs, Fe, I, Mn, Na, Rb, Zn
<i>Riparian</i>			
Amphibian	15	6	Co, Cs, I, Mn, Rb, Sr, Zn
Bird	32	≥2	Ba, Co, Cr, Cs, I, Se, Zn
Reptile	40	3	Cs, Sr

[#]The wildlife groups are not included in Copplestone et al. (2013); *where ≥ used some data have been entered with no Latin species name, or as spp. etc..

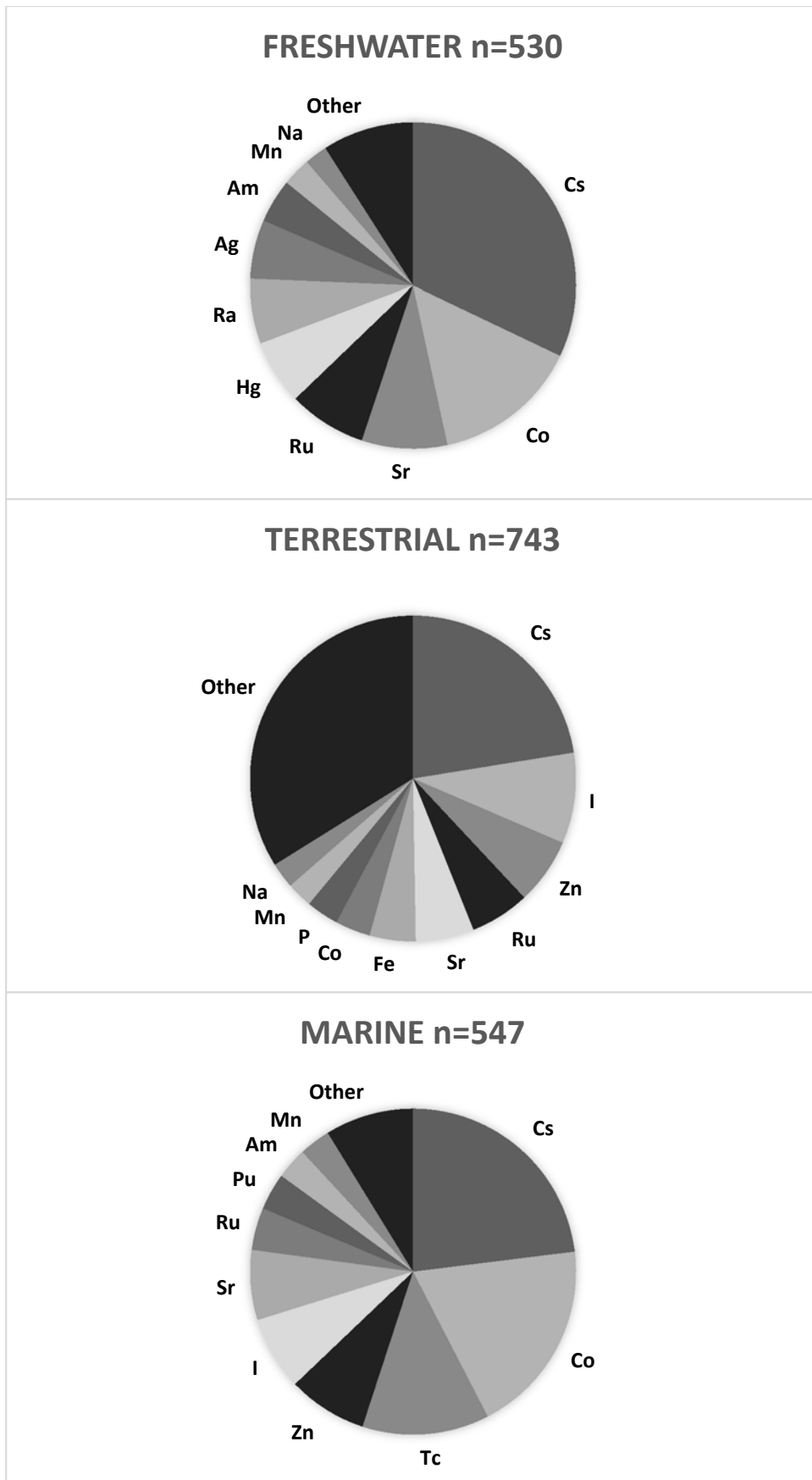


Figure 1. Summary of entries by radionuclide for the three generic ecosystems considered; the 10 elements contributing most entrees for each ecosystem are presented separately.