Article (refereed) - postprint

Brown, J.E.; Beresford, N.A.; Hevrøy, T.H.. 2019. **Exploring taxonomic and phylogenetic relationships to predict radiocaesium transfer to marine biota.** *Science of the Total Environment*, 649. 916-928. https://doi.org/10.1016/j.scitotenv.2018.08.343

Title

Exploring taxonomic and phylogenetic relationships to predict radiocaesium transfer to marine biota.

Keywords:

Transfer, Cs-137; Concentration ratio; Residual Maximum Likelihood, taxonomy, phylogeny

Introduction

A common means of quantifying the transfer of radionuclides to human foodstuffs and wildlife in ecosystems is through the application of concentration ratios (also referred to as concentration factors or bioaccumulation factors). When considering wildlife assessment the concentration ratio ($CR_{wo\text{-}media}$) relates the activity concentrations in an organism's habitat (water in the case of aquatic animals), to the activity concentration in the organism using the simple formula:

$$CR_{wo-media} = CR_{j,i} = \frac{C_{j,i}}{C_i^{aq}} \qquad \text{(eq. 1)}$$

Where:

$CR_{j,i}$ = Concentration ratio for organism *j* and radionuclide *i* (dimensionless or l kg^{-1});

$C_{j,i}$ = Activity concentration of radionuclide *i* in the whole organism j (Bq kg^{-1}, fresh mass);

C_i^{aq} = Activity concentration of radionuclide *i* in aqueous phase (Bq l^{-1} or Bq kg^{-1}) - normally filtered water;

Despite certain limitations (Brown et al., 2004; Vives et al., 2008), the CR model constitutes a simple practicable approach that covers a far greater range of radionuclides than any other currently available method for determining transfer. Over the last decades, great effort has been expended on the establishment of CR compendia for wildlife. For the marine environment, this is best exemplified by the work of the IAEA (IAEA, 2004; IAEA, 2014) and work conducted in connection with the ERICA Tool for environmental impact assessment for radioactivity (Hosseini et al., 2008). An international effort has focused on bringing much of the previous work on environmental transfer (for terrestrial and aquatic ecosystems) together culminating in the provision of the 'Wildlife transfer database' (Copplestone et al., 2013). The Wildlife transfer database (WTD) has subsequently been used to help prepare the IAEA's wildlife transfer handbook (Howard et al., 2013; IAEA, 2014), ICRP Publication-

114 (ICRP, 2009) and updating the ERICA Tool (Brown et al., 2016). The WTD provides an online, searchable compilation of $CR_{wo\text{-}media}$ values based on empirical data predominantly from determinations made under field conditions. The database provides a highly valuable resource with which to explore trends in datasets providing the basis for statistical analyses (e.g. Beresford et al., 2013; Wood et al., 2013). The WTD and the IAEA and ICRP compilations summarise data across all isotopes for a given element (i.e. for Cs the summary values will be based on a mixture of ^{137}Cs, ^{134}Cs and stable Cs values). The suitability of using stable Cs CR values as proxies for radiocaesium is evaluated in our analyses below.

Given the large number of organism-radionuclide combinations that may require assessment, it is perhaps not surprising that for many there are no data. In these circumstances, various 'extrapolation' approaches have been suggested to derive suitable values (e.g. Copplestone et al., 2003; Beresford et al., 2008; Brown et al., 2013). One of the key issues that currently confronts assessors working with environmental radioactivity is whether commonly applied 'extrapolation' approaches are suitable and appropriate. Furthermore, the International Commission on Radiological Protection (ICRP, 2008), recommends the use of Reference Animals and Plants, RAPs, to provide the basis for conducting an assessment through the provision of default models and datasets. However, the ICRP also recognizes that for site-specific assessments, 'representative organisms', i.e. defined (regulatory or otherwise) objects of protection, may be of particular interest (ICRP, 2009). This raises the question as to how information for RAPs (i.e. ICRP, 2009) or broad wildlife groups (e.g. IAEA 2014) might be extrapolated to representative organisms. Furthermore, the CR data for RAPs themselves (ICRP, 2009) are often ill defined (due to a lack of specific data) with recourse often having been made to the datasets for more generalized taxonomic groupings. A more analytical means of exploring the validity of this approach by looking at relationships between CRs for various taxa would clearly be advantageous. One potentially useful approach is based upon the hypothesis that some form of underlying taxonomic and/or phylogentic relationship exists in relation to ecological transfer of radionuclides (Beresford et al., 2013).

Research exploring whether elemental or radionuclide bioaccumulation characteristics differ between plant and animal taxa, and whether the degree of difference increases with their period of evolutionary divergence, has been published though predominantly for terrestrial plants. For example, soil-to-plant transfer of elements, that have radioisotopes of radiological interest have been analysed for underlying phylogenetic influences by Willey and co-workers. The authors have shown that relationships between elemental transfer and plant evolutionary history appear to exist for flowering and non-flowering plants for Cs (Broadley et al., 1999; Willey et al., 2005, Beresford and Willey submitted), Sr (Willey and Fawcett, 2005a), Ru (Willey and Fawcett, 2006), Cl (Willey and Fawcett, 2005b), Co (Willey and Wilkins, 2008), U (Willey, 2010) and I (Siasou and Willey, 2015). Willey (2010) suggested that such phylogenetic relationships may present a potential approach to enable predictions of radionuclide transfer for taxonomic groups for which there are data gaps. Turning to the marine environment, Jeffree et al. (2010; 2013) showed that the transfer of a number of radionuclides to marine teleost and chondrichthyan fishes and the amphioxus (fish-like chordate) species *Branchiostoma lanceolatumis* is influenced by phylogeny. However, the work of Jeffree et al. was based upon the results of laboratory studies looking at uptake directly from the water column. Whilst this usefully removes the influences of many confounding factors, food chain transfer was excluded and it is therefore not directly applicable to environmental conditions. Most recently, Beresford et al. (2013; 2016) applied the methodology of Willey et al. to explore underlying relationships between transfer and phylogeny for freshwater fish. Although it was possible to demonstrate differences in Cs transfer to freshwater fish based upon taxonomic groupings, it was not possible to establish a definitive phylogenetic relationship for the Cs transfer to different freshwater species (because of the large number of species and lack of data for most of these). Nonetheless, using model derived from the outputs from Residual Maximum Likelihood (REML) analysis, the authors accurately predicted ^{137}Cs activity concentrations in different species of fish from 27 Finnish and three UK lakes. In effect, the REML model derived by Beresford et al. (2013) removed the effect of site, which is a large contributor to the high degree of observed variability in the available $CR_{wo\text{-}media}$ datasets. A similar REML or taxonomic approach has recently been successfully applied to Pb and terrestrial wildlife (Beresford and Willey submitted).

Phylogenetic analyses using genomic data can reveal relationships between transfer and phylogeny and therefore allow detection of taxonomic patterns for pollution response (Carew et al., 2011; Keck et al., 2016), assuming there is a relationship between response and phylogeny. This approach does however require an understanding of the phylogenetic relationships between species, and simple but practical tools for measuring these relationships. Genomic data like DNA barcoding has been a huge benefit in identifying biomarkers for use in risk management, monitoring and protection of the environment (Carew et al., 2013; Larras et al., 2014;). The mitochondrial Cytochrome c Oxidase I (COI), referred to as a barcode sequence, is widely used as a core for genomic identification of taxa. Its popularity stems from its robustness across almost all animal and phytoplankton phyla, and its substantial divergence between species compared to low variation within species, making it an optimal tool for taxa delimitation (Hebert et al., 2003). The COI sequence been used to successfully identify species across a multitude of phyla (Hebert et al., 2003; Valentini et al., 2009; Freshwater et al., 2010; Bucklin et al., 2011) and in exploring phylogenetic patterns in sensitivity to contaminants (Carew et al., 2011; Guenard et al., 2011; Hammond et al., 2014).

The objective of the study described here was to explore whether taxonomic classifications could be used to characterise variation in the transfer of radiocaesium to a wide range of marine organisms using Residual Maximum Likelihood (REML) mixed-model regression (Willey, 2010). A further objective was to establish whether there were any phylogenetic patterns evident in the REML statistical model output and whether the model presents a useful predictive tool and scientifically based extrapolation approach.

1. Methodology

1.1 Categorisation

Data on caesium CRs were extracted from the Wildlife Transfer Database (WTD) (Copplestone et al., 2013) for all the marine species included in the database. The dataset was first accessed in April 2013 and the extracted data essentially correspond to those data used in compilations of $CR_{wo\text{-}media}$ values made at around that time (i.e. as described by Brown et al. 2016); this remains the current top copy version of the WTD. Species were classified taxonomically using various online resources, primarily the World Register of Marine Species (WoRMs; http://www.marinespecies.org/aphia.php?p=taxdetails&id=106264,). Note; all CR values used were expressed on a fresh mass basis.

Species were classified in relation to taxonomic family, order, class and phylum. Each data entry was also classified in terms of the location within which the sample was taken (i.e. a 'site' was designated). This procedure was somewhat arbitrary because it largely reflected the way in which sampling locations were reported in the original publications. Nonetheless, some effort was made to systematise this on the basis of approximately commensurable spatial units. So, for example, site codes such as the Greenland Sea, Barents Sea and Kara Sea were used. As will be explored in more detail below when the application of the REML model is described, the 'site' can be considered not only to encompass the notion of a geographical position but also aspects pertaining to methodologies employed within the study and/or study conditions.

For each study site there is a requirement when using the REML analysis, for instance at the family level, that data are available for more than one family and that at least one of these families must occur at another site. If the analysis is run for categories other than family then this requirement changes to

be applicable to those specific taxonomic levels. The data included varies with taxa, for instance, data may only be reported at the order level or a site may have multiple families all belonging to the same order. Excluding data that did not meet the above-mentioned criteria left a total of 592 data (i.e. CR value) entries for the family level. There was no requirement to exclude data for higher levels of biological organization and a total of 600 data entries for order, class and phylum were available for analyses. Seventeen 'sites' were categorized for the analysis at the family level and 18 sites for the higher levels of biological classification. Supplementary data (Table S1) summarises the data available and the analyses applied.

It should be noted that the analyses presented in this paper are based on data from the WTD which does not represent a taxonomic or phylogenetically balanced data set.

1.2 Data regeneration

As discussed elsewhere (Wood et al., 2013), there are problems associated with the derivation of representative data from datasets such as the WTD and also in conducting statistical analyses on those data. For example, mean values and numbers of samples may be presented without a standard deviation in the WTD (as this information was not present in the source publication). Wood et al. (2013) present a way of reconstructing datasets from summarised values, thus allowing an alternative means of characterizing the underlying variability and better enabling statistical evaluation of the data Wood et al. produced a spreadsheet (available from https://wiki.ceh.ac.uk/x/PgC6Cw) to enable data regeneration and this has been used in this study.

A total of 3072 values for the family level and 3080 for order, class and phylum were available for analyses subsequent to processing the data using the approach of Wood et al. (2013) (i.e. of the 592 data entries available at the family level some were means of multiple samples etc.).

1.3 Residual Maximum Likelihood Mixed (REML) model

In this analysis, we are attempting to determine whether taxonomic classification has any influence on Cs transfer to marine organisms, characterised by concentration ratios. Although we have a comprehensive dataset, which includes multiple species and covers a wide range of evolutionary divergence, we are faced with a problem that many samples were taken at different times at different locations within different studies. These latter factors clearly have the potential to introduce variance in the dataset thus making the fitting of correlations problematic.

To get around this we apply a mixed model that includes both fixed and random effects.

We assign the effects of site, (which incorporates variability introduced by time of study, scientific team performing study etc.) to random effects and the effect of taxonomic classification to fixed effects (which accounts for remaining variability). The fixed effects parameters tell how population means differ between taxonomic groups, while the random effect parameters represent the general variability within the taxonomic groups (attributable to their sampling under different conditions etc.).

The model is set up by assigning fixed effects to the taxonomic classification and random effects to the site. The hypothesis we are testing is that the variance for a given taxonomic group (e.g. family, order etc.) over all sites (including study conditions) can be modelled as a random component in a model. In this way, we can reveal what we hypothesise to be differences between taxonomic groups under any specified set of environmental conditions.

IBM SPSS Statistics Version 22 has been used in the analysis. This statistical program allows for nominal datasets, i.e. taxonomical classification and site name in our analyses. The ‘linear Mixed Model’ analysis option was selected in SPSS allowing a REML procedure to be applied to the dataset.

The performance of the models was evaluated through comparisons of various model information criteria (Akaike's Information Criterion; Hurvich and Tsai's Criterion)) for cases where the full REML

196 model (i.e. using fixed and random components) was applied against those where only fixed
197 components of the model were specified (i.e. random components were excluded). These model
198 information criteria were used to determine if the application of the REML model with site as a
199 random factor improved efficacy compared to models where the effect of 'site' was ignored. The
200 analysis was run for both the original dataset as extracted from the WTD and also for the regenerated
201 dataset using the spreadsheet described in Wood et al. (2013).
202
203 *1.4 Data used to blind test predictions*
204
205 Once predictions of relative mean values have been made using the REML analyses it is of importance
206 to establish the efficacy of the procedure (as was successfully conducted for the freshwater model
207 (Beresford et al., 2013; 2016)). An attempt at doing this has involved the comparison of the
208 predictions made using REML means with blind datasets that include some of the taxa for which
209 predictions have been made. Note the outputs of the REML model can be applied to either fresh mass
210 activity concentrations or CR values as the output gives relative values for the different taxa. Three
211 datasets were identified for the purpose of testing model predictions against empirical data these
212 being:
213
214 (i) A dataset providing fresh mass activity concentrations of ^{137}Cs in fish and seafood sampled
215 from Norwegian coastal waters for the period 1991 to 2011 (Heldal et al., 2015) combined
216 with data from a recent annual monitoring report (Skjerdal et al., 2015) for some of the
217 same sea areas. Data were selected for an area covering the Barents Sea, the area around
218 Svalbard and the coast of Finnmark and Troms (this roughly corresponding to the scale of
219 the site units used in setting up the REML model). Data from the period 2007 to 2012
220 inclusive were used, assuming that temporal activity concentration fluctuations (e.g.
221 owing to, among other factors, the ongoing decline of inputs of radiocaesium to the
222 marine environment from sources such as BNFL Sellafield) would be insubstantial over

this time period – an assumption which appears to be reasonably well supported by observation of these data.

(ii) An unpublished dataset included in the WTD by the National Institute for Radiation Safety (NIRS) Japan reporting a range of stable elements in species of macroalgae, crustaceans and molluscs collected from Japanese estuaries that were not used to establish the REML model. An earlier version of the sampling program is described by Takata et al. (2010). Though these data were reported to be from estuaries (and hence not used in the model fitting).. However, concomitant water salinity values (*c*. 25-35 ‰) were specified that were similar to marine waters.

(iii) A dataset covering various species sampled in the Irish Sea in 2014 (Sellafield Ltd., 2015)

To circumvent any problems that may have been introduced by assumptions regarding the normality of the dataset, all data were transformed by taking their natural logarithm. There was justification for doing this based on the observation that the underlying datasets in the WTD tend to be log-normally distributed (Wood et al., 2013). A Grubbs outlier test was performed on the datasets. Running through this test led to the identification of a value on the data from NIRS that did not appear credible, this being for a Gastropod of the family Buccinidae. This single data point was removed before subsequent analyses.

1.5 Relationship between predicted and empirical data

Predictions based on the REML statistical model output were generated by selecting one of the taxonomic groups from the blind dataset and deriving the ratios of the REML-adjusted means for all other groups sampled in the dataset to the selected taxonomic group. The ^{137}Cs (activity concentration or CRs) values were then predicted for all taxonomic groups by multiplying these ratios by the activity concentration (or CRs) for the selected taxonomic group as reported in the blind dataset. For example, using the Barents Sea-Norwegian data (as described in (i) above), the family *Gadidae* was selected because the number of associated data points was relatively high. Ratios were then generated by

dividing the REML-adjusted means for any given family in the data set by the REML-adjusted mean value for *Gadidae*. These ratios were then multiplied by the activity concentration value for *Gadidae* reported in the Barents Sea-Norwegian data to allow prediction for all families that had been sampled. Predictions for the selected taxonomic group, in this example - the family *Gadiadae*, were not included in any comparative analyses. SPSS was used to determine whether the degrees of correlation between predicted and empirical data were statistically significant.

Twelve comparisons between model predication and empirical data have been performed, Thus, there is an increased probability that a significant correlation occurs simply by chance. The Bonferroni correction has been applied to compensate for this by selecting the desired significance level, α, selected as 0.01 for this study in line with convention, and dividing by 'n' the number of hypotheses/tests (i.e. a de facto α of 0.00085 was applied).

1.6 Phylogenetic analyses

We downloaded barcode (mtCOI) sequences for all available marine species included in the REML analysis from the National Centre for Biotechnology Information's Genbank database (http://www.ncbi.nlm.nih.gov/genbank/; last accessed 31/06/2015). Phylogenetic trees were constructed with mitochondrial COI gene sequences from fish, mammals, birds, molluscs, macroalgae and arthropod species. We hypothesised that by mapping the REML residual means of species onto these phylogenetic trees, the existence of any patterns would be revealed.

Gene sequences were aligned using ClustalW in MEGA 6.0 (Tamura et al., 2013), and then visually checked for errors. Phylogenetic trees were constructed using two methodologies frequently applied in combination in phylogenetic studies (Hall, 2011). The first method was Bayesian Inference (BI), which uses Monte Carlo Markov Chain (MCMC) methodology to run numerous simulations, searching for the best set of trees, outputting the trees with the highest probability. For Bayesian analysis, we used the program MrBayes 3.1.2 (Ronquist and Huelsenbeck, 2003). Two runs were

277 performed simultaneously, each with four Markov chains, which ran for one million steps
278 ("generations"). The first 250,000 generations were discarded from analysis as 'burnin' (values of
279 most parameters change a lot during initial 'burnin' period, before they settle near their most probable
280 values), and every 1000th tree produced from the analyses was sampled to calculate 50% majority-rule
281 consensus tree (the most likely tree), with posterior probabilities for nodes. The second method was
282 Maximum Parsimony (MP) analyses, performed in MEGA 6.0 (Tamura et al., 2013), using the Tree-
283 Bisection algorithm. Maximum parsimony is based on the assumption that the most likely tree is the
284 one that requires the least amount of changes to explain the data (sequences). This analyses operates
285 by selecting the tree or trees that minimise the number of evolutionary steps. The evaluation of nodal
286 support in the MP analyses method is based on 1000 non-parametric bootstrap replicates. Combined,
287 Bayesian and MP analyses give us two values of nodal (tree branching) support; posterior probability
288 (PP) and bootstrap (BS), allowing us to assess the reliability of the output tree. Phylogenetic analyses
289 requires the user to specify a model of evolution that aims to account for the various aspects of ways
290 we think nucleotide substitutions occur during gene evolution. There are numerous types of models of
291 evolution (substitution models), and can be was identified for each dataset (species to phylum level
292 sequences) using the Akaike Information Criterion implemented in jModelTest 2.0 (Posada, 2008). In
293 addition to describing the rate of change from one nucleotide to another, models can include the rate of
294 either variation among sites in sequences through a gamma distributed rate (G), or static, unchanging
295 rate variation (I). Substitution models were as follows: General Time Reversible (GTR) + G for groups
296 Aves and Mollusc, Hasegawa-Kishino-Yano (HKY) + I + G for Mammalia, and GTR + G + I for
297 groups Fish, Arthopoda and macroalgae.

298 A large portion of fish phylogenies are still uncertain (Nelson, 2016), so COI DNA data alone did not
299 provide enough variation to create an understandable phylogenetic tree for the Actinopterygii class
300 (ray-finned fishes) of the Chordate Phylum. However, at selected levels of taxonomy, which we had
301 reliable and large sequence datasets for (multiple species representing multiple families and orders);
302 sensible reference trees could be made. Phylogenetic trees are presented for examples of fish groups
303 where the underlying data permit such an analyses.

1.7 Comparison of CR values based on stable Cs versus radiocaesium

The suitability of using stable Cs CR values as proxies for radiocaesium has been explored by comparing data at the level of 'Order'. This was the lowest level of biological organisation where there was sufficient overlap, in terms of the number of values available, to allow a meaningful statistical comparison to be made. Orders where the number of measurements available were less than three for either stable element or radionuclide derived values were excluded from this analysis. An independent samples student t-test was performed on log-transformed data using SPSS. The analyses were undertaken using the regenerated data set derived through application of the spreadsheet routine described above (Wood et al., 2013).

316

317 ## 2. Results and discussion

318

319 ### *2.1 REML-adjusted means*

320

321 The outputs in the form of REML-adjusted means from the analyses performed for different taxa
322 groupings (from phylum through to family) are presented in Tables 1-3; note the outputs in the three
323 tables all originate from the same analyses, the results have been split into broad organism types
324 purely for ease of presentation. The values presented in the tables are all based on data that have been
325 regenerated using the approach described by Wood et al. (2013).

326

327 The check on model performance by making a comparison of the model with and without a
328 component corresponding to random effects revealed that for all comparisons (at given taxonomic
329 levels) the information criteria (however defined i.e. using Akaike's Information Criterion, Hurvich
330 and Tsai's Criterion etc.) were lower for the model employing mixed and random (i.e. site/study)
331 components. This supports the hypothesis that there would be some efficacy in applying the REML
332 analyses to remove the influence of the variance introduced to CR for a given taxa by sampling site.
333 When considering all taxonomic levels, the application of the REML model to families appeared to
334 provide the most pronounced effect in terms of the difference observed in modelling with and without
335 random effects. The information criteria also demonstrated that the model based at the taxomomic
336 level of the family and including random effects was best.

337

338 ### *2.2 Data regeneration versus no regeneration*

339

340 A comparison of REML-adjusted means for unaltered datasets and datasets for which a reconstruction
341 routine had been run showed, in most cases, small differences in output. Typically (for well over half
342 of the comparisons made), REML-adjusted means for the same taxon differ by less than 20 %.

343 Therefore, for example, the REML-adjusted mean for the family *Gadidae* without data regeneration is
344 84 compared to a value of 72 with data regeneration. However, there are some exceptions to this
345 observation with differences of up to a factor of three. An example exists for the family *Mytilidae*
346 which includes the blue mussel (*Mytilus edulis*), where a value of 20 without data regeneration
347 compares to a value of 61 with data regeneration. This discrepancy for the latter may be explained by
348 the observation that the base data for *Mytilidae* included at least one, large undifferentiated dataset (i.e.
349 included one entry for which mean and standard deviation were reported where n=69).
350
351 *2.3 Testing the REML outputs - Relationship between predicted and empirical data*
352
353 The only significant correlation ($p < 0.05$) between predicted and measured activity concentrations
354 was revealed at the taxonomic level of order when comparing REML analysis output with data from
355 the Barents Sea Region for the period 2007-2012 (Heldal et al., 2015; Skjerdal et al., 2015) as
356 illustrated in Fig. 1. For this single case a Spearman rank correlation coefficient of 0.80 (based on
357 regenerated data) was derived which was significant at the 0.01 level (1-tailed test). The null
358 hypothesis, that the correlation (between predicted and measured values) arose by chance, can
359 therefore be rejected. However, in applying the Bonferroni correction, the correlation is not significant
360 at the 0.01 level. Because multiple comparison between model predictions and empirical data have
361 been performed there is an, albeit, low probability that the correlations unraveled here could have
362 occurred by chance.
363
364 The predictions (based on regenerated data) for the NIRS dataset were particularly poor for all
365 taxonomic classifications. The Pearson correlation coefficient 'r' was -0.082 (Spearman rank 'r' = -
366 0.24) indicating that the correlation is not significant. It is of particular note in this regard that the
367 NIRs data were for stable element measurements and that the REML model was based on very few
368 data for the families for which prediction were being made. For example, the REML model included
369 only four data points for the families *Lessoniaceae* and *Veneridae* with only two data points for the

family *Turbinidae*. The low number of underpinning data entries might plausibly undermine the predictive efficacy of the REML model.

The predictions for the Irish Sea dataset were also poor. For instance, for the family level prediction, he Pearson correlation coefficient 'r' was 0.26 (Spearman rank 'r' = 0.46). Neither of these correlations were significant at the 0.01 level (1-tailed test).

The degree of fit between predicted and measured values for comparisons performed using REML analysis output derived using regenerated and original (i.e. mean) data were similar in the sense that statistically significant correlations between measured and predicted values were generally absent. For the single instance where a convincing correlation was apparent (i.e. REML analyses (order level) for Norwegian monitoring data), the correlation between the REML output (based upon the dataset which had not been regenerated) and measured values was actually more significant than for correlation involving the REML output based on regenerated values (a Spearman rank correlation coefficient of 0.90 was derived for the former which was significant at the 0.01 level (1-tailed test)).

2.4 Phylogenetic analyses

We have chosen to present data for mammals, birds and fish as examples as they were the most robust phylogenies. The resulting phylogenetic tree topologies for these groups provide genetic distances relative to the length of the branches. For example, within the order Carnivora, the Family *Phocidae* (earless seals including the harbour seal and ringed seal) are more closely related to the family *Ursidae* (includes the Polar bear) than to the family *Otaridae* (eared seals as characterised here by the Northern fur seal). Although the REML-adjusted means (as shown by the number in parenthesis on Fig. 2) for the order *Carnivora* seem, on cursory inspection, to be quite distinctive to those derived for the order *Cetariodactyla*, substantial differences are also apparent within both orders. In other words, the phylogenetic analysis appears to show that large differences in Cs REML values can exist despite there being a close genetic relationship between the given taxa. Concerning mammals, the family

398 *Ursidae* (bear family) are phylogenetically distant from *Monodontidae* (a family which includes the
399 Narwhal and Beluga Whale) but exhibit similar REML-adjusted means representing radiocaesium
400 transfer. Conversely, although *Balaenidae* (Right Whales) are more closely related to *Monodontidae*
401 than *Ursidae* the associated REML-adjusted means are quite different.
402
403 Several explanations beyond phylogenetic relationships may unravel the cause as to why relatively
404 closely related families exhibit quite different transfer of ^{137}Cs. Taking the mammalian order
405 Carnivora, for example, where substantial differences in REML adjusted means are observed (Fig. 2)
406 for the family *Phocidae* compared to the families *Otariidae* and *Ursidae*. While otariids (eared seals)
407 are known for speed and maneuverability, phocids (earless seals) are known for efficient, economical
408 movement. This allows most phocids to forage far from land to exploit prey resources, while otariids
409 are tied to rich upwelling zones close to breeding sites. Ursidae, like Otariids, spend longer periods on
410 land that in the sea, unlike phocids (McLaren, 1984). These factors would undoubtedly have some
411 influence on what these mammals are eating and where they are eating it. Larras et al. (2014)
412 suggested that the large variation in toxin sensitivities between species not related to phylogeny, are
413 likely due to trophic mode and indirectly, on feeding habitats. For radiocaesium, dietary exposure
414 pathways are an important component of exposure, and phylogenetically similar organisms may have
415 quite different diets and be associated with different trophic levels. For example, *Balaenidae*, such as
416 Right whales, consume mainly copepods but also prey upon pteropods and krill, whereas, the closely
417 related family *Monodontidae* have a wide-ranging carnivorous diet and feed on fish, molluscs, and
418 crustaceans. This may dominate any 'signal' allowing the establishment of a phylogenetic relationship
419 for radiocaesium transfer; Cs accumulates up foodchains (Pentreath, 1973) meaning that we could
420 expect higher trophic level species to accumulate higher concentrations of radiocaesium than their
421 prey (see section 3.5.2 for further discussion).
422
423 The observation that closely related species can have considerably different REML adjusted means
424 may have some implication for the ICRP's Reference and Animal Plant approach, which is based
425 around the family level. Users of the approach should be aware of the great variability associated with

426 the transfer parameters associated with any given RAP group. The limitations of the RAPs approach
427 have been discussed before (Bérchignac and Doi, 2009, Bradshaw et al 2014).
428
429 There appears to be a discord between the gene tree and the current taxonomy of the orders
430 Perciformes and Pleuronectiformes, where the Pleuronectiformes order is situated within the
431 Perciformes order (Fig. 3). The same pattern for these orders have been revealed in other studies on
432 fish phylogenies (Kochzius et al., 2010; Near et al., 2012). This illustrates how an analysis based on
433 using taxonomical names alone can be misleading where a group that is considered different based on
434 traditional taxonomy, is actually not a resolved monophyletic group from a phylogenetic perspective.

435 Although the majority of the phylogenetic relationships among birds illustrate correlation with REML
436 values, the REML value for *Stercorarius skua* is an obvious outlier (Fig. 4). This could be due to the
437 fact that *S. skua* are scavengers and could plausibly be scavenging land mammals, such as reindeer,
438 with relatively enhanced levels of Cs-137 (Rissanen et al., 1997). After the Chernobyl accident in
439 1987, high concentrations of Cs-137 have been recorded in reindeer in Norway and Sweden (Skuterud
440 et al., 2005; Åhman et al., 2007) which might explain the Cs bioaccumulation which has followed in
441 scavenger birds.

442 It should be noted that we have not performed a statistical 'phylogenetically-informed trait
443 comparison'. Our analysis has reconstructed the phylogenetic trees but we have not conducted a
444 statistical analysis taking into account phylogenetic distances or evolutionary models. Consequently,
445 whilst we did not detect a phylogenetic effect using the approach described, it does not exclude the
446 existence of such an effect.

447 *2.5* Confounding factors in the application of REML to marine systems
448
449 There are several reasons that may explain why the REML model is not an efficacious predictive tool
450 for the marine system. This can, in-part, be explored further by testing whether stable elements form a

suitable analogue for radiocaesium and considering some of the more important factors that affect radiocaesium transfer in marine environments.

2.5.1 Is stable Cs an efficacious proxy for radiocaesium ?

At least 84 % of the original dataset, used for the REML analysis, was based upon Cs-137 as oppose to stable caesium data. A generally accepted assumption has been that using stable element data provides suitable CR values for application in radiological assessments (Beresford, 2010; Howard et al., 2013; ICRP, 2009). However, more detailed analyses provides evidence that the tacit assumption, that both stable and radionuclide CRs belong to the same data population, may not be tenable in many cases. By way of example, for freshwater fish, Beresford et al. (2013) saw significant differences between $CR_{wo\text{-}water}$ values derived from stable elements and those derived from Cs-137 although other factors such as data provenance and the feeding strategy of the fish included in the stable and radionuclide categories were likely to have confounded this analysis.

As considered by Robertson (1971), radionuclides often enter the oceans in forms completely different from that of their stable isotopes naturally present. If the equilibration between these different forms is slow, the uptake, distribution and behavior of the radionuclides and their naturally occurring stable isotopes by the marine biosphere could show significant anomalies.

The comparison conducted for the marine dataset (Fig. 5) leads to the suggestion that differences between CRs derived from stable elements and those from radiocaesium can be substantial. In fact, the group means for all orders analysed, with one exception, were significantly different at the 0.05 level (2 tailed test). The exceptional case was *Scorpaeniformes* where the absence of any statistically significant difference between group means was undermined by the very low number of observations for stable caesium (where 'n' was equal to 3). Nonetheless, and in line with similar arguments promulgated by Beresford et al. (2013), this observation may be confounded by other factors. All stable element data were extracted from three key studies, two of which were from coastal Japanese

479 locations (Ichikawa and Ohno (1974); Marumo et al., 1998) and one for South African coastal waters
480 (Van As et al., 1975). These locations are atypical for the dataset seen as a whole for which samples
481 were predominantly from the North Atlantic and European coastal environments. Although marine
482 chemistry is not expected to differ significantly even over the global scales involved, there is no way
483 to avoid the speculation that other local factors, such as the species considered, levels of suspended
484 load and sediment type, plus the degree of equilibration that may have occurred following
485 radionuclide release, could be the cause of the observed discrepancy.
486
487 2.5.2 Other factors which may affect radiocaesium transfer
488
489 The physico-chemical form of radionuclides is of paramount importance in the consideration of
490 bioavailability (Salbu, 2007) and subsequent entry to and transfer through food-chains. Nonetheless,
491 for the case of radiocaesium, at least, the physico-chemical form in seawater, once equilibration has
492 occurred, might be expected to be monotonous over large regional scales reflecting the substantial
493 mixing of marine water bodies and the similarity in water chemistry of the world's oceans. Caesium,
494 an alkali metal, forms monovalent Cs+ ions in seawater and in an extensive study, conducted many
495 decades ago, was found to be present as less than 1 % particulate form in open oceanic waters
496 (Robertson, 1971) . Radiocaesium/ is often classified as behaving conservatively in seawater, having a
497 low affinity for particle association and with a behavior consequently controlled by the physics of
498 ocean circulation and mixing (Livingston and Povinec, 2002).
499
500 Differences in radiocaesium transfer within the biotic system might plausibly be related to trophic
501 level in accordance with the generally recognised effect observed in freshwater systems (Fleishman et
502 al., 1994; Kryshev, 1995; Saxen and Koskelainen, 1996). However, evidence for trophic
503 biomagnification is not as clear for marine ecosystems. Data collated on ^{137}Cs activity concentrations
504 in high trophic level fish (e.g. cod (*Gadus morhua*)), at lower trophic level fish, (e.g. herring (*Clupea*
505 *harengus*)), and crustaceans inhabiting northern marine environments (Brown, 2000; Brungot et al.,
506 1999; FSA and SEPA, 2000; Rissanen et al., 1997), do not provide clear evidence for a strongly

507 pronounced trophic-level effect in marine ecosystems. Fish living as first level predators often exhibit
508 ^{137}Cs activity concentrations commensurate with those fish species living higher up the food-chain at
509 the same location. Although ^{137}Cs was not biomagnified during its transfer to filter-feeding molluscs,
510 as considered by Wang et al. (2000), a trophic transfer factor close to 1 may be reached in high trophic
511 level predators when ingestion of gastropods is high (Wang et al., 2000).
512
513 Activity concentration/CR data for seabirds and mammals are relatively scarce. Data for sea mammals,
514 primarily whales and seals, (e.g. Rissanen et al., 1997; Fisher et al., 1999; Strand et al., 1998; Brown,
515 2000) provide no clear evidence for any trophic level effect, i.e. elevated body activity concentrations
516 of radiocaesium over those observed in prey species. Calmet et al. (1992) concluded that similarity of
517 estimated CRs (30-100) for porpoise with those obtained for fish characterized by the same ethological
518 features, such as tuna, suggests that the mechanisms and rates governing caesium accumulation by
519 these top marine predators may be similar. Biomagnification, however, was not reported explicitly. In
520 contrast, those data that are published in the literature for seabirds (e.g. Rissanen et al., 1997; Fisher et
521 al., 1999) suggest that relatively elevated bioaccumulation of ^{137}Cs may be occurring for seabirds,
522 especially those associated with high trophic levels such as skuas and gulls. Other studies have also
523 led to the suggestion that ^{137}Cs biomagnifies through marine food chains (e.g., Watson et al., 1999;
524 Heldal et al., 2003). Perhaps some of the most convincing evidence for biomagnification effect comes
525 from the work of Kasamatsu and Ishikawa, 1997; wherein the analysis of over 6000 samples from the
526 coastal waters of Japan consisting of fish samples and their stomach contents demonstrated that ^{137}Cs
527 concentration increased with rising trophic level deriving a biomagnification factor of around 2.
528 Andersen et al. (2006) showed that, ^{137}Cs concentration factors for seal species, although often similar
529 in magnitude, are typically higher than those reported for lower trophic levels, which suggests that
530 ^{137}Cs is biomagnified through marine food chains to these consumers. Nonetheless, transfer data for
531 polar bears preying on various seal species (Derocher et al., 2000), did not suggest biomagnification of
532 the radionuclide. Is summary, the diet/feeding strategy of animals appears to influence ^{137}Cs transfer
533 substantially but a distinct biomagnification is not evident across all species.
534

535 It should be acknowledged that a comparison of species from the same marine area, and/or over a
536 number of monitoring years, might not allow suitably robust conclusions to be drawn owing to the
537 migrant nature of many species and fluctuations in ambient radionuclide levels with time. Although
538 the concentration ratio has been commonly applied in both human (IAEA, 2001; 2010) and
539 environmental impact assessments for radioactivity (e.g. Copplestone et al. 2001, Brown et al., 2008;
540 ICRP, 2009; IAEA, 2014), it lends itself most appropriately to steady-state conditions wherein
541 radionuclide levels within the organism have equilibrated with those in water. Such a case might be
542 reasonably represented, for example, by an ecosystem receiving continuous uniform inputs of
543 radioactivity some years after operations from a given nuclear plant have been initiated. In contrast,
544 when radionuclide activity concentrations in the environment are changing rapidly with time,
545 modelling approaches that account for the dynamics of the system may be more appropriately applied
546 (Vives i Batlle et al., 2016). Sampling location and feeding areas may not coincide and questions
547 remain regarding how rapidly biotic and abiotic compartments equilibrate as evidenced by the large
548 temporal variations in ^{137}Cs CR for selected marine locations and given species (Beresford et al.,
549 2003) as shown in Fig. 6.
550
551 The data presented in Fig. 6, which were one of many data inputs to the WTD, demonstrate that the
552 relative CR of different species may vary substantially over time even within the same sea region
553 (synonymous with the site categorisation applied in our study). There seems to be no convincing way
554 that the REML model, as configured in our study, could account for the considerable variance
555 introduced from this source. Although the fixed effects in the model are nominally associated with
556 taxonomic classification only, in practice, the temporal variation in CR for any given species at a site
557 would form an inherent, irreducible component of the fixed effect.
558

3. Conclusions

A REML model, to quantify radiocaesium transfer to various taxa in marine environments, has been successfully configured wherein taxonomic classification has been allocated to fixed effects and site (sampling and environmental conditions) has been allocated to random effects. The inclusion of site as a random factor resulted in a better model than the output of the REML analyses without the inclusion of the random effect. However, the application of this statistical model appears to have limited efficacy. The mapping of ^{137}Cs on phylogenetic trees shows that large differences in REML-adjusted transfer values exist despite there being a close genetic relationship between certain groups of taxa and that difference in habitat and diet may help to explain this. However, we have to acknowledge that we have not conducted a full statistical phylogenetic analysis, as discussed above.

In applying the model to three blind dataset over various levels of taxonomic classification (from family to phyla), a significant correlation was only observed in one instance and even then, if one corrects for multiple correlations, the significance is debatable . The result from this analysis is in contrast to the results from other published works where the REML model was applied, as exemplified by the accurate prediction of ^{137}Cs in fish in freshwater ecosystems (Beresford et al. 2013; 2016) and the promising development of terrestrial wildlife REML models (Søvik et al., 2017; Beresford and Willey submitted).

Reasons that may explain why the REML model application to the marine biological transfer of radiocaesium are numerous. In particular, the fact that phylogenetically similar taxa may have quite diverse life histories and different diets may confound the possibility that any phylogenetic pattern can be revealed. The fact that marine waters exhibit relatively similar chemistry may dampen the influence of site. There are limitations regarding the data used in the blind data tests in the sense that it was only possible to cover a very limited number of taxa. Ideally, it would have been of interest to consider in

586 more detail some of the biota groups for which more extreme radiocaesium were predicted, e.g. some
587 seabirds such as the families Laridae and Stercorariidae. However, for understandable reasons such as
588 their protected status in many countries, the attainment of such data was not practicable. It is also
589 worth noting that the best models for terrestrial and freshwater systems were at the genus or species
590 levels. There is considerable variation between species and genus and therefore, predictions using
591 family level REML means may be poor depending upon the species included in the model fit
592 compared to the species for which predictions are being made. That said if based on sufficient data the
593 family level REML model represents a useful scientifically based extrapolation approach.
594 Other factors that influence ^{137}Cs transfer, such as feeding strategy and/or trophic level, could be
595 explored in a systematic manner during future dataset analyses and REML may provide a suitable way
596 of achieving this objective (i.e. by enable analyses by feeding strategy etc. whilst accounting for the
597 effect of site).
598

Acknowledgements

The input of N.A. Beresford to this work was supported by the TREE project (http://tree.ceh.ac.uk/), funded under the RATE programme by the Natural Environmental Research Council, Radioactive Waste Management Ltd. and the Environment Agency.The Research Council of Norway through its Centre's of Excellence funding scheme, project number 223268/F50, supported this work.

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Figures

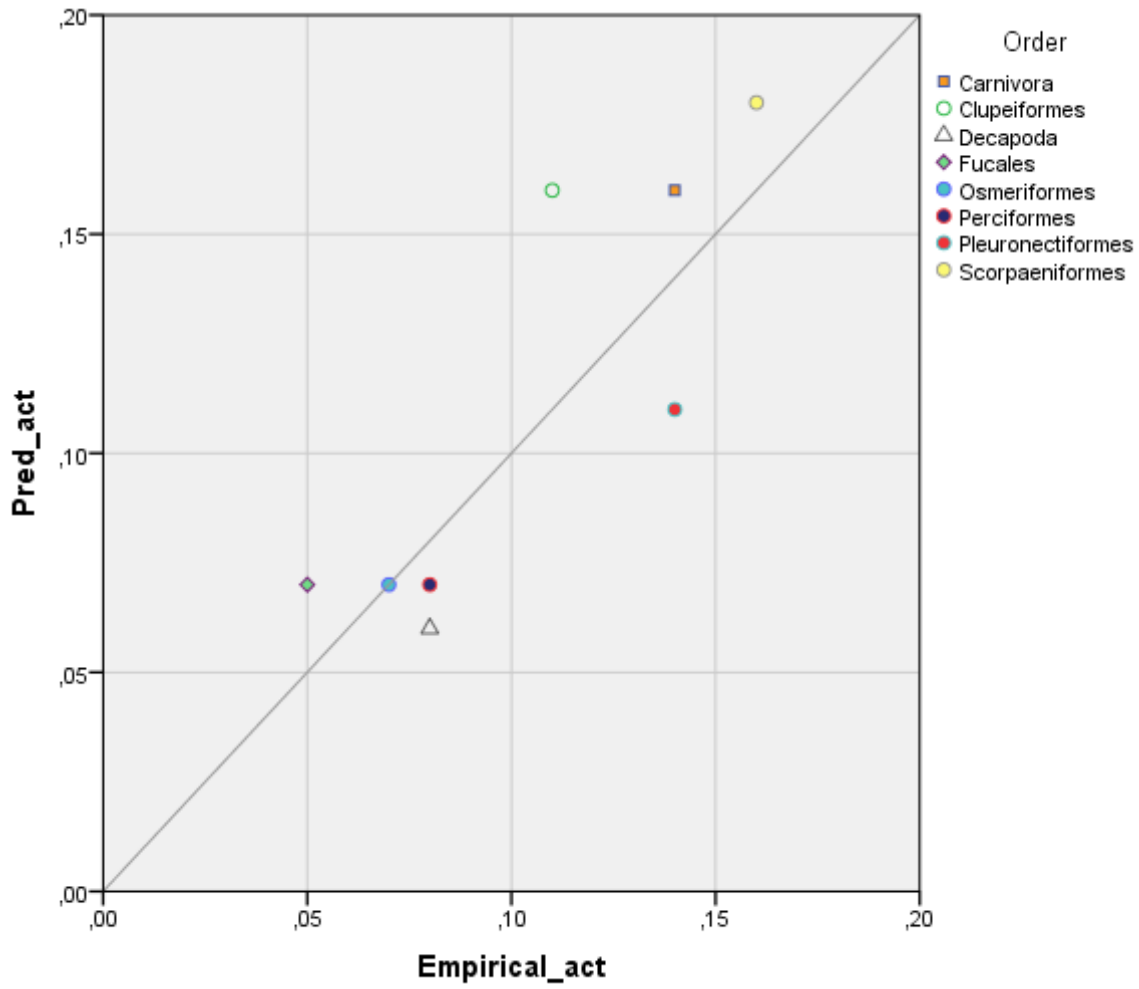

Fig 1. Comparison of measured ^{137}Cs activity concentrations in samples (categorised by order) collected from locations within the Barents Sea Region in the period 2007-2012 (Heldal et al, 2015; Skjerdal et al., 2015) with predicted activity concentrations using the outputs of the REML analyses (order level) and data for *Gadiformes* (line is 1:1 relationship).

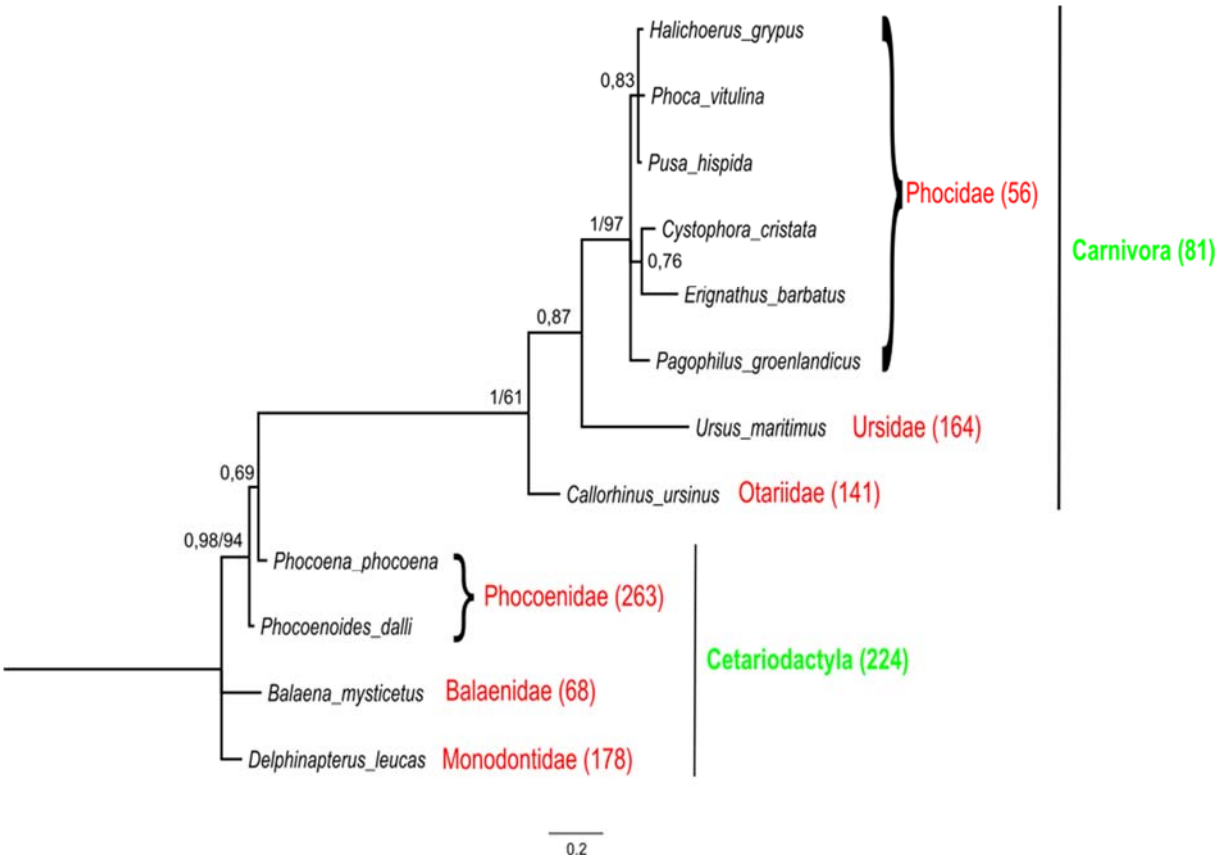

Fig 2. Phylogenetic reconstruction of Class Mammalia based on a 655 basepair alignment of the mitochondrial COI gene. Nodal support shown on branches, values are Bayesian inference posterior probabilities (PP:0.5–1.0) and bootstrap support (BS:50–100). REML analyses residual means shown in brackets after Family (red) and Order (green) groupings.

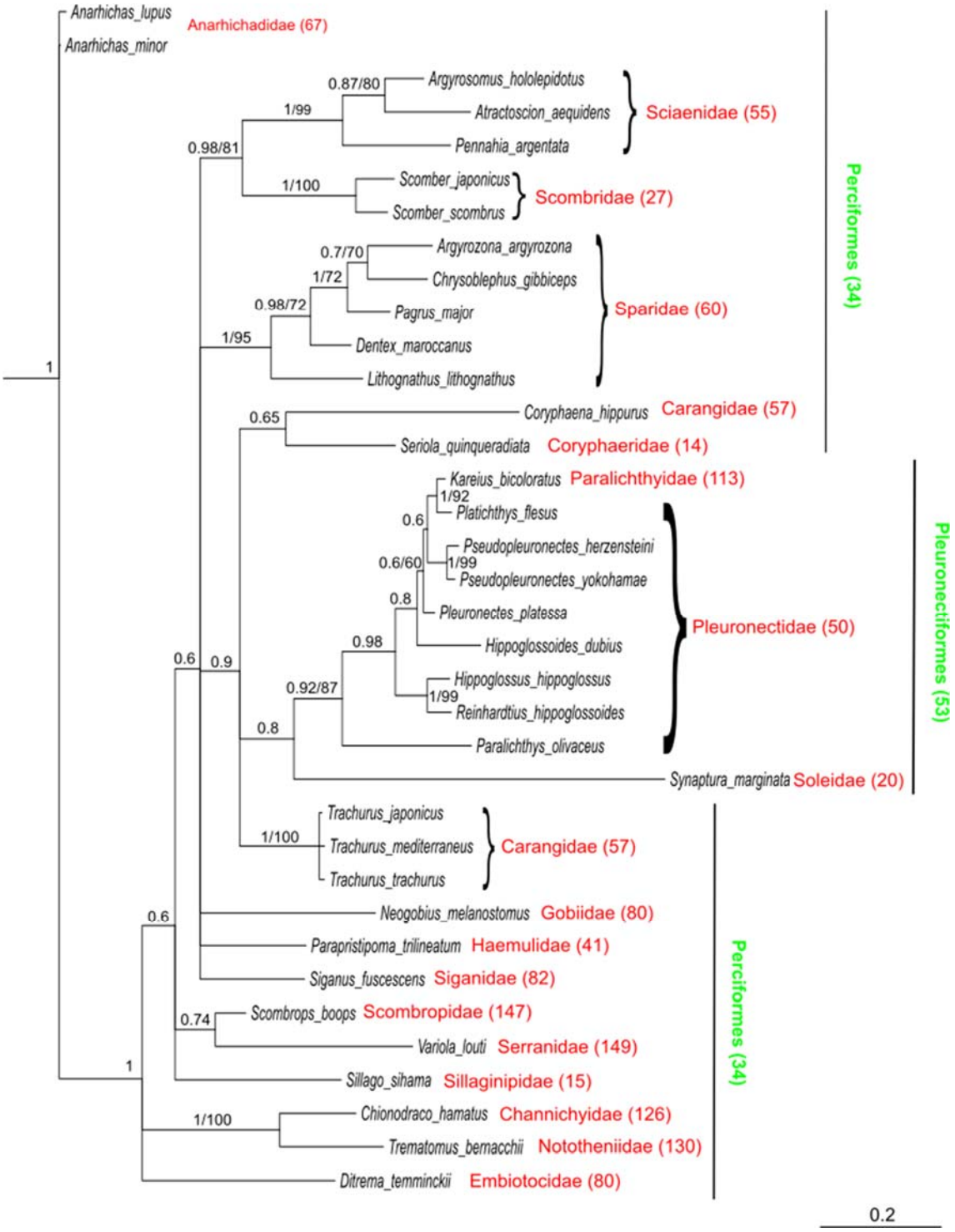

Fig 3. Phylogenetic reconstruction of Orders Perciformes and Pleuronectiformes based on a 655 basepair alignment of the mitochondrial COI gene. Nodal support shown on branches, values are Bayesian inference posterior probabilities (PP: 0.5–1.0) and/or maximum parsimony bootstrap support (BS: 50–100). REML analyses residual means shown in brackets after Family (red) and Order (green) groupings.

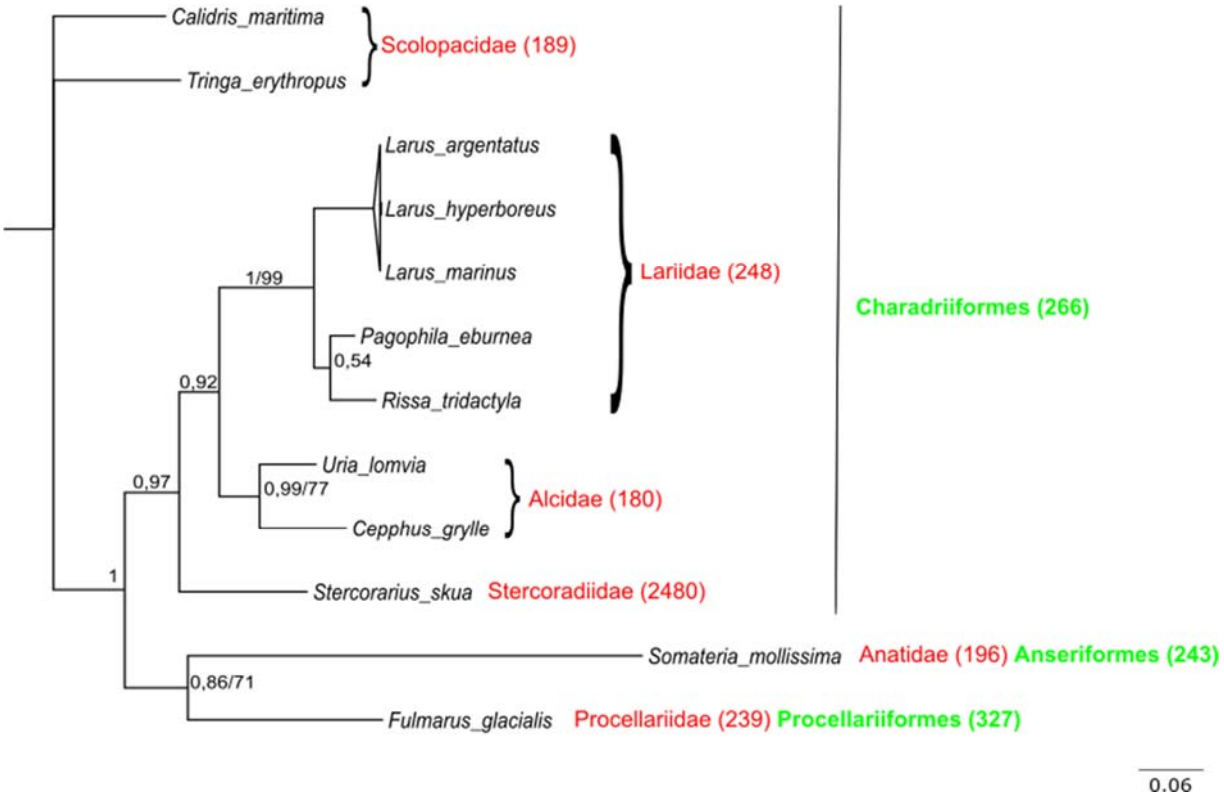

Fig 4. Phylogenetic reconstruction of Aves based on a 653 basepair alignment of the mitochondrial COI gene. Nodal support shown on branches, values are Bayesian inference posterior probabilities (PP: 0.5–1.0) and maximum parsimony bootstrap support (BS: 50–100). REML analyses residual means shown in brackets after Family (red) and Order (green).

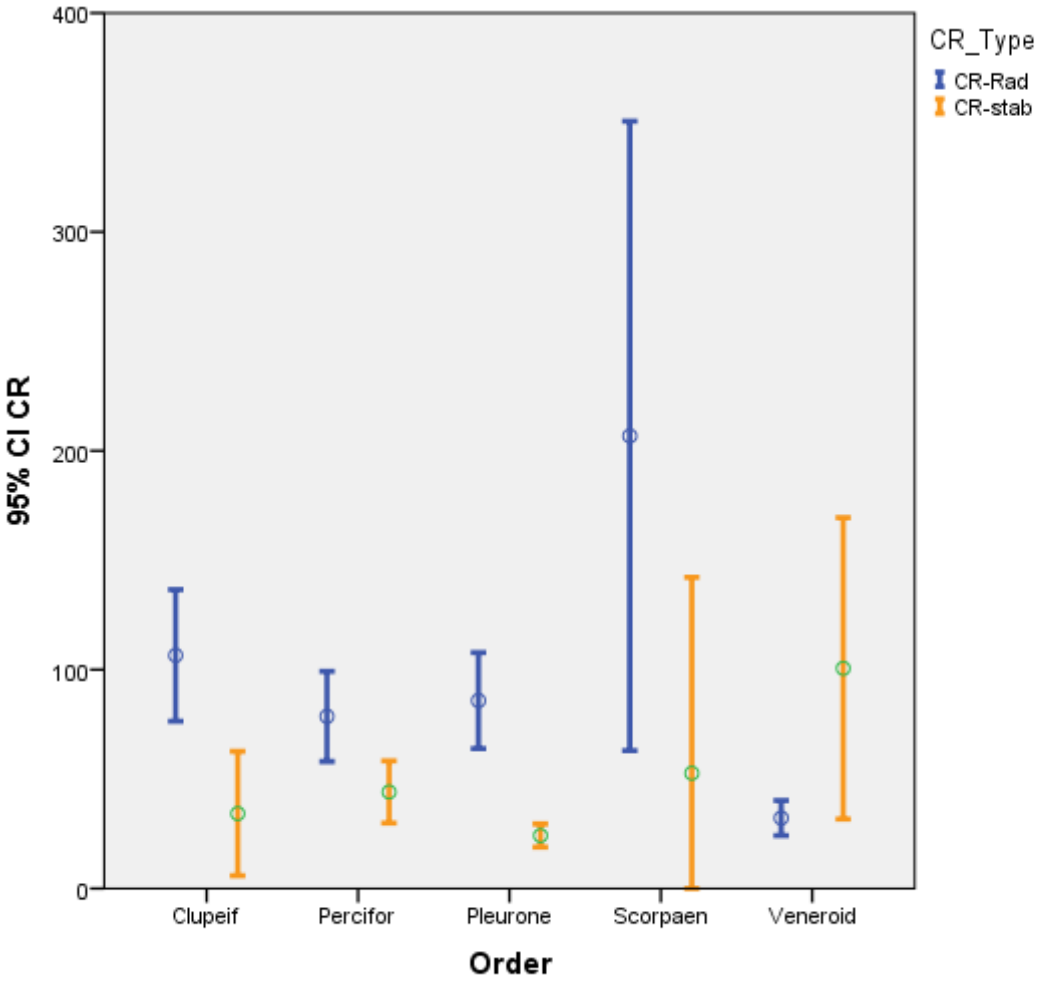

Fig 5. Comparison of ^{137}Cs CR values derived from radionuclides (CR-Rad) and stable element (CR-stab) for selected orders of marine organisms. The circles provide the mean value and the whiskers the 95 % confidence intervals of the datasets.

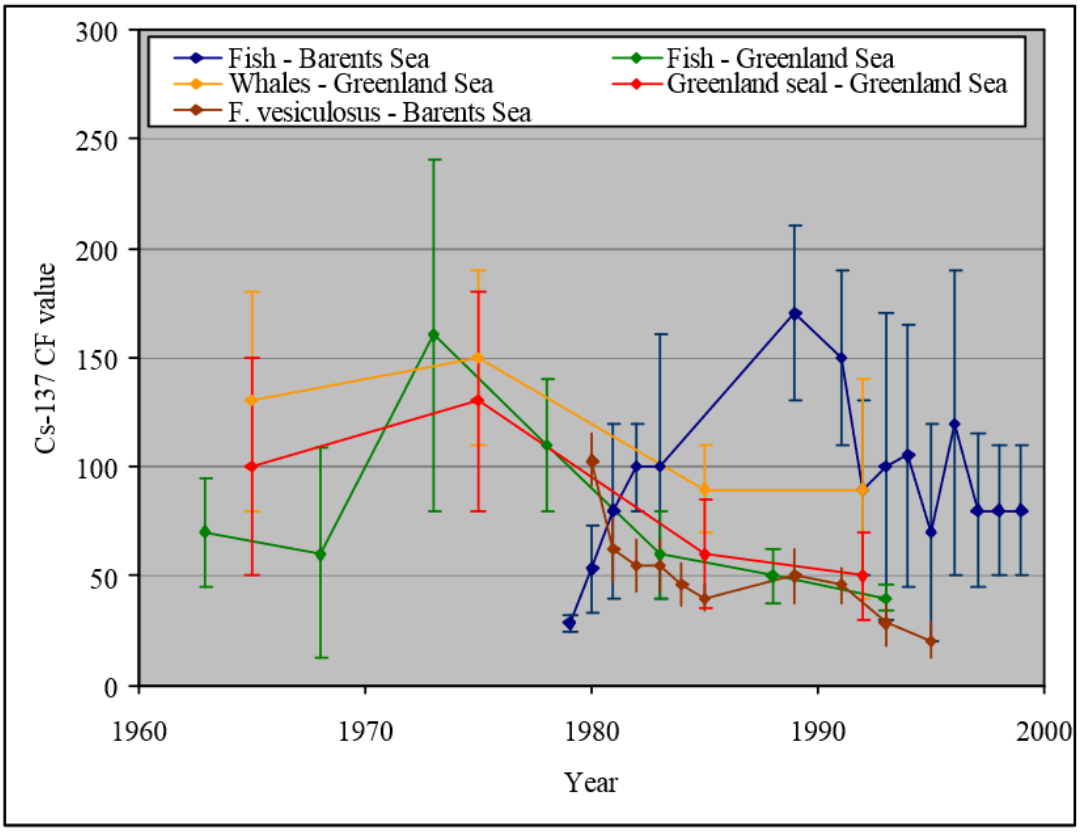

Fig 6: Temporal variation in ^{137}Cs CF (Concentration factor, equivalent to CR) values (FW) for: *Gadus morhua* in the Barents Sea (annual average ± standard deviation); fish in the Greenland Sea (five year averages ± standard deviation); whales and Greenland seals in the Greenland Sea (ten year average ± standard deviation); and *Fucus vesiculosus* in the Barents Sea (annual average ± standard deviation). Reproduced from EPIC D2 (Beresford et al., 2003).

Tables.

Table 1 REML-adjusted means for different vertebrates. Note these are relative values and not absolute values (the values are also relative to the values in Tables 2 & 3).

Phylum		Order		Class		Family	
Chordata	71	Actinopterygii	54	Beloniformes	47	Exocoetidae	65
				Clupeiformes	80	Clupeidae	85
						Dussumieriidae	78
						Engraulidae	56
				Gadiformes	76	Gadidae	72
						Lotidae	103
						Merlucciidae	119
				Lophiiformes	38	Lophiidae	48
				Mugiliformes	19	Mugilidae	25
				Ophidiiformes	76	Ophidiidae	97
				Osmeriformes	35	Osmeridae	33
				Perciformes	34	Anarhichadidae	67
						Carangidae	57
						Channichthyidae	126
						Coryphaenidae	14
						Embiotocidae	80
						Gobiidae	80
						Haemulidae	41
						Nototheniidae	130
						Sciaenidae	55
						Scombridae	27
						Scombropidae	147
						Serranidae	149
						Siganidae	82
						Sillaginidae	15
						Sparidae	60
				Pleuronectiformes	53	Paralichthyidae	113
						Pleuronectidae	50
						Soleidae	20
				Salmoniformes	56	Salmonidae	55
				Scorpaeniformes	87	Cottidae	47
						Hexagrammidae	124
						Liparidae	622
						Sebastidae	85
						Triglidae	111
				Tetraodontiformes	15	Monacanthidae	20
		Aves	277	Anseriformes	243	Anatidae	196
				Charadriiformes	266	Alcidae	180
						Laridae	248
						Scolopacidae	189
						Stercorariidae	2480
				Procellariiformes	327	Procellariidae	239
		Elasmobranchii	57	Rajiformes	65	Dasyatidae	19
						Rajidae	60
		Mammalia	127	Carnivora	81	Otariidae	141
						Phocidae	56
						Ursidae	164
				Cetartiodactyla	224	Balaenidae	68
						Monodontidae	178
						Phocoenidae	263

Table 2 REML-adjusted means for different plants and macroalgae. Note these are relative values and not absolute values (the values are also relative to the values in Tables 1 & 3).

Phylum		**Order**		**Class**		**Family**	
Angiosperms	3	Monocots	3	Alismatales	3	Zosteraceae	3
Chlorophyta	36	Bryopsidophyceae	21	Bryopsidales	25	Bryopsidacea	35
		Ulvophyceae	38	Cladophorales	56	Cladophoracaea	56
				Ulvales	35	Ulvaceae	42
Ochrophyta	36	Phaeophyceae	33	Desmarestiales	9	Desmarestiaceae	13
				Dictyotales	37		
				Fucales	37	Fucaceae	34
						Sargassaceae	60
				Laminariales	39	Alariaceae	43
						Chordacea	119
						Costariaceae	34
						Laminariaceae	38
						Lessoniaceae	61
Rhodophyta	51	Bangiophycidae	15	Bangiales	14	Bangiaceae	15
		Florideophyceae	53	Ceramiales	45	Ceramiaceae	61
						Rhodomelaceae	50
				Corallinales	134		
				Gelidiales	19	Gelidiacea	25
				Gigartinales	55	Dumontiaceae	82
						Gigartinaceae	48
				Palmariales	108	Palmaiaceae	102

Table 3 REML-adjusted means for different invertebrates. Note these are relative values and not absolute values. .(the values are also relative to the values in Tables 2 & 3).

Phylum		**Order**		**Class**		**Family**	
Annelida	62	Polychaeta	58	Capitellida	10	Arenicolidae	10
				Phyllodocida	206	Nephtyidae	203
				Spionida	54	Chaetopteridae	53
				Terebellida	59	Pectinariidae	57
Arthropoda	32	Branchiopoda	378	Diplostraca	394	Podonidae	547
		Malacostraca	28	Amphipoda	25	Gammaridae	24
				Decapoda	31	Cancridae	15
						Lithodidae	114
						Nephropidae	38
						Oregoniidae	20
						Paguridae	20
						Palinuridae	38
						Pandalidae	35
						Penaeidae	19
						Portunidae	25
				Isopoda	36	Chaetiliidae	35
		Maxillopoda	73	Calanoida	289	Calanidae	363
						Eucalanidae	324
						Paracalanodae	382
						Pontellidae	248
						Temoridae	935
				Sessilia	9	Balanidae	9
Chaetognatha	18	Sagittoidea	15	Aphragmophra	13	Sagittidae	17
Cnidaria	14	Anthozoa	13	Actiniaria	14	Actiniidae	15
Mollusca	50	Bivalvia	52	Arcoida	28	Arcidae	30
				Carditoida	32	Astaridae	23
				Myoida	57	Myidae	61
				Mytiloida	66	Mytilidae	61
				Nuculanoida	32	Yoldidae	26
				Pectinoida	49	Pectinidae	48
				Veneroida	43	Arcticidae	32
						Cardiidae	27
						Donacidae	25
						Tellinidae	27
						Veneridae	222
		Gastropoda	36	Haliotoidea	3	Haliotidae	4
				Littorinimorpha	41	Littorinidae	39
						Naticidae	15
				Neogastropoda	33	Buccinidae	30
						Muricidae	27
				Vetigastropoda	17	Turbinidae	21
Sipuncula	136	Sipunculidea	127	Golfingiiformes	138	Golfingiidae	121