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

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HIGHLIGHTS

- A Residual Maximum Likelihood, REML, model has been applied to marine Cs transfer data.
- Differences in ^{137}Cs REML-derived values exist despite close genetic relationships.
- A phylogenetic pattern defining ^{137}Cs transfer to marine biota is virtually absent.
- Other parameters, e.g. life history traits, may affect transfer more than phylogeny.
- This finding contrasts with REML application to freshwater and terrestrial systems.

GRAPHICAL ABSTRACT
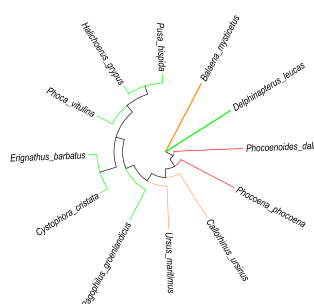
Phylogenetic tree of Mammalia with REML residual means of Cs137 tranfser.

0 - 75
75 - 150
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ABSTRACT

One potentially useful approach to fill data gaps for concentration ratios, CRs, is based upon the hypothesis that an underlying taxonomic and/or phylogenetic relationship exists for radionuclide transfer. The objective of this study was to explore whether these relationships could be used to explain variation in the transfer of radiocaesium to a wide range of marine organisms. CR data for ^{137}Cs were classified in relation to taxonomic family, order, class and phylum. A Residual Maximum Likelihood (REML) mixed-model regression modelling approach was adopted. The existence of any patterns were then explored using phylogenetic trees constructed with mitochondrial COI gene sequences from various biota groups and mapping the REML residual means onto these trees. A comparison of the predictions made using REML with blind datasets allowed the efficacy of the procedure to be tested. The only significant correlation between predicted and measured activity concentrations was revealed at the taxonomic level of order when comparing REML analysis output with data from the Barents Sea Region. For this single case a correlation 0.80 (Spearman rank) was derived which was significant at the 0.01 level (1-tailed test) although this was not the case once a (Bonferroni) correction was applied. The application of the REML approach to marine datasets has met with limited success, and the phylogenetic trees illustrate complications of using predictions based on values from different levels of taxonomic organization, where predicted values for the order level can mask the values at lower taxonomic levels. Any influence of taxonomy and phylogeny on transfer is not immediately conspicuous and categorizing marine organisms in this way is limited in providing a potentially robust prognostic extrapolation tool. Other factors may plausibly affect transfer to a

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much greater degree in marine systems, such as quite diverse life histories and different diets, which may confound any phylogenetic pattern.

1. 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:

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

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 i Batlle 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 phylogenetic 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 lanceolatum* 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. While 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.,