

Enhancing radiological monitoring of ^{137}Cs in coastal environments using taxonomic signals in brown seaweeds

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ABSTRACT

With the rapidly expanding global nuclear industry, more efficient and direct radiological monitoring approaches are needed to ensure the associated environmental health impacts and risk remain fully assessed and undertaken as robustly as possible. Conventionally, radiological monitoring in the environment consists of measuring a wide range of anthropogenically enhanced radionuclides present in selected environmental matrices and using generic transfer values for modelling and prediction that are not necessarily suitable in some situations. Previous studies have found links between taxonomy and radionuclide uptake in terrestrial plants and freshwater fish, but the marine context remains relatively unexplored. This preliminary study was aimed at investigating a similar relationship between brown seaweed, an important indicator in radiological monitoring programmes in the marine environment, and Caesium-137, an important radionuclide discharged to the marine environment. A linear mixed model was fitted using REsidual Maximum Likelihood (REML) to activity concentration data collected from literature published worldwide and other databases. The output from REML modelling was adjusted to the International Atomic Energy Agency (IAEA) quoted transfer value for all seaweed taxa in order to produce mean estimate transfer value for each species, which were then analysed by hierarchical ANalysis Of VAriance (ANOVA) based on the taxonomy of brown seaweeds. Transfer value was found to vary between taxa with increasing significance up the taxonomic hierarchy, suggesting a link to evolutionary history. This novel approach enables contextualisation of activity concentration measurements of important marine indicator species in relation to the wider community, allows prediction of unknown transfer values without the need to sample specific species and could, therefore, enhance radiological monitoring by providing accurate, taxon specific transfer values for use in dose assessments and models of radionuclide transfer in the environment.

1. Introduction

The nuclear energy industry is rapidly expanding in response to low carbon emission targets and energy security concerns. At the end of 2021, worldwide there were 436 operable nuclear power stations with 53 more under construction (World Nuclear Association (WNA), 2022). Currently contributing approximately 10% of the world's electricity production, nuclear power is expected to play an increasingly important role in future energy supply due to concerns over CO₂ emissions and energy security (WNA, 2021). The International Energy Agency (IEA) states that nuclear power will provide an essential foundation for the transition to renewables such as solar and wind, with total energy supply from the industry projected to increase by 15% between 2020 and 2030 in their Stated Policies Scenario (STEPS) (IEA, 2021). Infrastructure such

as geological disposal facilities are also required to aid this expected growth. These large-scale nuclear developments to house new and historic nuclear waste, are vital and contentious parts of the industry. For example, in the UK a number of new nuclear developments, including power stations and supporting infrastructure such as geological disposal facilities, are already underway and with new funding initiatives, more are likely to be built over the next few decades (Department for Business, Energy & Industry Strategy (BEIS), 2021, 2022a; Nuclear Waste Services (NWS), 2022). Concerns over CO₂ emissions have led to the prioritisation of the nuclear power industry in the UK government's most recent energy strategy, leading to an industry that is likely to expand rapidly over the next few decades. (BEIS, 2022b).

Caesium-137 (^{137}Cs ; $t_{1/2} = 30.7$ years) is a significant component of anthropogenic radionuclide releases to the marine environment and an

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important indicator of radioactive contamination (Gwynn et al., 2022). A fission product of uranium, releases to the environment have included authorised discharges from the nuclear fuel cycle (power generation and fuel reprocessing) atmospheric weapons testing and accidents involving nuclear power generation. For example, the Fukushima Dai-ichi Nuclear Power Plant discharge in 2011 was estimated to have released between 19 and 24 PBq of ^{137}Cs into the environment, with 15.5–18.5 PBq of this entering the North Pacific Ocean (Aoyama et al., 2016). The Sellafield reprocessing plant has been discharging ^{137}Cs into the sea since the early 1950s, with peak levels of 5200 TBq in 1975, decreasing to $<10\text{ TBq y}^{-1}$ since 1997 (Hunt et al., 2013). The ^{137}Cs inventory of sub-tidal sediment in the Irish Sea was estimated to be 455 TBq in 2006 (Jenkinson et al., 2014). Monitoring programmes for routine releases of ^{137}Cs from many nuclear facilities not only ensure that doses to flora, fauna and humans are negligible, but also produce the data that is often the baseline for emergency responses. Transported through the marine environment as a simple, soluble cation, ^{137}Cs is bioavailable and extremely mobile, with the Fukushima Dai-ichi ^{137}Cs signal detectable in seawater as far away as the North American coast in the years after the accident (Smith et al., 2015). Caesium-137 can adsorb onto marine sediment, and thus be removed from the water column, but may be subject to remobilisation due to resuspension of historically deposited sediment (Hunt et al., 2013). When combined with its moderate half-life, such behaviour means that ^{137}Cs can remain bioavailable in some marine ecosystems for a relatively long period of time.

Marine macroalgae are important primary producers and habitat builders in marine ecosystems (Steneck et al., 2002). Exposure to Radionuclides due to their coast location and fixed position, as well as bioaccumulation of some radionuclides mean that some Brown Seaweed (Phaeophyceae) species are used as bio-monitors of radioactivity, as well as other contaminants in the marine environment (Szefer, 2002; Zalewska and Saniewski, 2011). Practically, coastal species are often easier to sample, than more open water species, with many found in the littoral or sub-littoral zone. Common coastal brown seaweed species such as *Fucus vesiculosus* and *Fucus serratus* are currently used in several radiological monitoring programmes around the Atlantic and Baltic (e.g. EPA, 2017; Environment Agency, 2010; HELCOM, 2023). One example of this use is as part of the recent OSPAR Radioactivity Substance Committee (RSC) 5th Periodic Evaluation (Gwynn et al., 2022) to demonstrate that the actions and measures taken by OSPAR have been sufficient to meet its objectives under the 2020 Radiological Substance Strategy.

Coastal ecosystems often act as an interface between radionuclide contamination and humans, with activities such as gathering seafood, bait digging, and exposure over sediment in intertidal areas being major exposure pathways (Greenhill et al., 2020). In radioecology, brown seaweeds are primarily known for their high capacity to accumulate radioiodine, but as a K^+ analogue, Cs^+ is also taken up by brown seaweeds (Hagstroem, 2002). The uptake mechanism is not fully understood but considered to be indirectly linked with photosynthesis and metabolism (Gutknecht, 1965). Caesium-137 can therefore be transferred via brown seaweeds to humans through food. However, they can also be useful indicators when monitoring nuclear-derived contamination of the coastal environment. It is listed by the OSPAR commission as an important indicator radionuclide in seaweed associated with nuclear sector along with ^{99}Tc and $^{239,240}\text{Pu}$ (Gwynn et al., 2022). The Transfer Factor (TF) of ^{137}Cs has been shown to increase with trophic level in marine organisms (fish, cephalopods, and crustaceans) (Kasamatsu and Ishikawa, 1997). When investigating Periwinkles (*Littorina littorea*) feeding on *Fucus* sp., Broom et al. (1975) found ^{137}Cs at higher concentrations in higher trophic levels, indicating bioaccumulation up the food chain. Risk to biota itself was not considered. Globally, seaweeds are of great economic importance, with 35.9 million tonnes produced in 2019, 97.38% of which originating in Asia (Zhang et al., 2022). This includes many brown seaweeds, both cultivated and harvested from the wild for products such as chemicals and medicines, as well as a vital food

source (Sanjewa and Jeon, 2018). The potential use of seaweeds as a dietary supplement for cattle has been suggested to help mitigate methane production (Min et al., 2021). Recently, there is increasing interest in seaweed as a source of food in the West.

Radiation risk to humans and other organisms is estimated through modelling radionuclide transfer and calculating subsequent dose. The International Atomic Energy Agency (IAEA) have published a list of empirically derived transfer coefficients often used for this purpose (IAEA, 2014). These values are specific to each element and are calculated for groups of organisms based on field measurements of direct environmental medium-to-organism transfer ratios. These values are useful generalisations, often encompassing large and diverse groups, for example a TF value of 24.0 ± 5.3 for ^{137}Cs is given for all macroalgae (IAEA, 2014). The ERICA (Environmental Risk from Ionising Contaminants: Assessment and Management) tool was developed under a Euratom project funded by the European Commission and is used to assess radiological risk (in the form of estimated dose rates) to the marine environment, and to evaluate potential dose rates to biota for both existing and planned releases (<https://erica-tool.com>). ERICA models radionuclide transfer based on 25 preselected organisms, designed to cover EU protected species, with transfer factors derived from the Wildlife Transfer Database (Brown et al., 2008; Coppelstone et al., 2013). Although this list of organisms is more detailed than the list from the IAEA (TRS479, IAEA, 2014), and the program has the facility to add organisms if the required information is available, measured values are not available for many taxa of interest to specific applications.

Over the last couple of decades, evidence has demonstrated a relationship between the taxonomy of living organisms and their uptake of heavy metals and radionuclides. Taxonomy can be defined as the classification of organisms based on shared descriptive characteristics, whereas phylogeny uses evolutionary history. A study conducted by Broadley et al. (1999) first developed the approach of using Residual Maximum Likelihood (REML) modelling to investigate this, finding that Cs accumulation in the shoots of flowering plants was influenced by taxonomy. A REML-fitted mixed model for a large dataset of species-specific transfer values minimised the effects of variables that differ from study to study such as sampling method and locational differences. This produced an estimated relative mean for each species, allowing data that has been collected from a variety of different locations to be compared.

Willey et al. (2005) considers evolutionary history, using genetically derived phylogenetic information, to determine that plant taxa do not behave independently for the ^{137}Cs uptake, and that phylogeny influences a significant proportion of soil to plant transfer of $^{134/137}\text{Cs}$. Similar soil to plant phylogenetic effects have also been found in other radionuclides (Willey et al., 2010; Willey, 2014). Willey (2010) uses the IAEA transfer factor values to anchor the relative mean estimate values and produce adjusted soil to plant TFs specific to each species, resulting in more specific values. This method allows the prediction of transfer values in groups of organisms where there are few or no measured values available (Willey, 2010; Beresford and Willey, 2019).

This method has also been applied to other groups of organisms. With significant differences in ^{137}Cs transfer found between freshwater fish species, this method was used to accurately predict transfer in other species using the REML-adjusted means produced (Beresford et al., 2013). However, application of this approach to the wider marine environment has yielded limited success (Brown et al., 2019). Under laboratory conditions there is evidence that radionuclide transfer factors are influenced by phylogeny in Cephalochordates, chondrichthyan and teleost fish, major marine taxa (Jeffree et al., 2010, 2013, 2017). The taxonomic and phylogenetic effects on radionuclide uptake in marine macroalgae has not, however, previously been explored.

This paper aims to investigate the relationship between brown seaweed taxonomy and ^{137}Cs uptake and assess its significance to radiological monitoring of the coastal environment. We examine the following two hypotheses:

- 1) There is significant variation in ^{137}Cs transfer between taxa in the Phaeophyceae ($P = 0.05$).
- 2) That variation in transfer between groups is significant up the taxonomic hierarchy, with greater variation seen at higher taxonomic levels.

2. Methods

The data collected for this study were sourced from published peer-reviewed scientific articles, grey literature, and other databases, with a global scope. Activity concentration data was taken from 51 publications. The main criterion for inclusion of published data was that it contained values for individuals identified at the species level. Values not reported to species level or reported as groups such as “*Fucus* sp.”, “*Fucus* spp.” or merely “Brown Seaweeds” were excluded. Publications were also required to provide the year the samples were collected, and a location. Where available, values for individuals were preferred over mean values. In addition, activity concentration data for selected species collected for UK’s Radioactivity in Food and the Environment (RIFE) report were provided by the UK environmental and food standards agencies: Environment Agency (EA), Food Standards Agency (FSA), Food Standards Scotland (FSS), Natural Resources Wales (NRW), Northern Ireland Environment Agency (NIEA) and Scottish Environmental Protection Agency (SEPA). This, along with data from the Marine Radioactivity Information System (MARIS) (IAEA MARIS, 2020), constituted most of the modelled data.

All model fitting and statistical analysis was conducted in R (R Core Team, 2021). Values for ^{137}Cs were found to be \log_e -normally distributed so were \log_e -transformed prior to modelling. Estimated Relative Means (ERMs) for each species were modelled using a REML-fitted linear mixed-model using the R package lme4 (Bates, 2010). Several iterations of the model were tested to ensure the best results, with fit being measured by REML convergence criteria (Bates et al., 2015). The lower this value, the better fitted the model is to the data. The model was fit at the species level as this also produced the best fit. The random factors were selected to account for variation between groups with some having stronger effects than others. The current model includes taxonomic group as the fixed factor, and location, sample year, and study (each individual publication or data source) as random factors. The lme4 package allows nesting of factors if they are hierarchical. Nesting location within study was seen as a logical approach as some locations are shared across multiple studies and was found to improve model fit. The model accounts for variation in the data due to study, location, and year (i.e., the influence of locational, methodological, and temporal differences), producing an ERM for each species.

In addition to the collated data, two other datasets were also created in order to further test both the model’s effectiveness at removing variation due to the random factors, and the significance of its outputs. Firstly, the collected activity concentrations were assigned to random species from the same radionuclide data creating a scrambled dataset. Secondly, a dataset of random activity concentrations with the same mean, standard deviation, and \log_e -normal distribution, as the specific radionuclide dataset was generated. These scrambled and randomly generated datasets were modelled using the same model structure as the actual collated dataset.

The model output ERMs were transformed to have the relevant IAEA empirically derived TF of 24.0 ± 5.3 as the geometric mean, providing an estimated TF for each species (IAEA, 2014). These estimated TFs for each species were then analysed for taxonomic effect using hierarchical ANalysis Of VAriance (ANOVA) based on the taxonomy of Phaeophyceae.

3. Results and discussion

The data compiled for this research provided an appreciable dataset of ^{137}Cs activity concentrations in brown seaweeds. It included a total of

7484 individual environmental brown seaweed activity concentration measurements including 63 species from 350 different locations worldwide, dating from 1965 to 2021 (Table 1). Much of these data were originally collected as part of regulatory required environmental monitoring. For example, the RIFE monitoring programme and subsequent assessments are used to demonstrate that radioactivity in food and the environment is safe and within UK national dose limits (Dewar et al., 2021, Environment Agency et al., 2022). A large collation of data such as that reported here can provide not only information on environmental

Table 1

The species of Brown Seaweed collated in this database span 15 families and 6 orders. ($n = 7484$).

Order	Family	Species	N	
Desmarestiales	Desmarestiaceae	<i>Desmarestia anceps</i>	2	
		<i>Desmarestia ligulata</i>	2	
		<i>Himantothallus grandifolius</i>	5	
Dictyotales	Dictyotaceae	<i>Padina pavonica</i>	3	
		<i>Dictyota coriacea</i>	1	
		<i>Dictyota dichotoma</i>	8	
		<i>Spatoglossum pacificum</i>	2	
Ectocarpales	Acinetosporaceae	<i>Pylaiella littoralis</i>	2	
		Chordariaceae	<i>Leathesia difformis</i>	1
	<i>Myelophycus caespitosus</i>		16	
	<i>Myelophycus simplex</i>		1	
	<i>Nemacystus decipiens</i>		3	
	<i>Tinoclada crassa</i>		1	
	<i>Ectocarpus siliculosus</i>		1	
	Ectocarpaceae		<i>Colpomenia sinuosa</i>	4
			<i>Petalonia fascia</i>	4
	Scytosiphonaceae		<i>Scytosiphon lomentaria</i>	16
			<i>Analipus japonicus</i>	7
			<i>Ascophyllum nodosum</i>	291
		<i>Fucus distichus</i>	8	
<i>Fucus serratus</i>		1798		
<i>Fucus spiralis</i>		195		
<i>Fucus vesiculosus</i>		4628		
<i>Fucus virsoides</i>		2		
<i>Pelvetia canaliculata</i>		5		
<i>Silvetia babingtonii</i>		2		
Fuciales		Sargassaceae	<i>Cytoseira crinita</i>	6
	<i>Cytoseira ericoides</i>		1	
	<i>Sargassum aquifolium</i>		1	
	<i>Sargassum confusum</i>		2	
	<i>Sargassum fulvellum</i>		6	
	<i>Sargassum fusiforme</i>		45	
	<i>Sargassum horneri</i>		26	
	<i>Sargassum macrocarpum</i>		2	
	<i>Sargassum micracanthum</i>		5	
	<i>Sargassum miyabei</i>		2	
	<i>Sargassum muticum</i>		6	
	<i>Sargassum patens</i>		5	
	<i>Sargassum ringoldianum</i>		6	
	<i>Sargassum siliquastrum</i>		2	
	<i>Sargassum thunbergii</i>		36	
	<i>Sargassum vulgare</i>		6	
	<i>Sargassum yamadae</i>		6	
	<i>Sargassum yezeonce</i>		4	
	<i>Treptacantha barbata</i>		23	
Ishigeales	Seirococcaceae	<i>Cystosphaera jacquinotii</i>	1	
		Ishigeaceae	<i>Ishige foliacea</i>	11
			<i>Ishige okamurae</i>	16
Laminariales	Alariaceae	<i>Ishige sinicola</i>	1	
		<i>Eualaria fistulosa</i>	2	
		<i>Undaria pinnatifida</i>	84	
		<i>Undaria undarioides</i>	2	
		Costariaceae	<i>Costaria costata</i>	3
			Laminariaceae	<i>Kjellmaniella crassifolia</i>
		<i>Laminaria digitata</i>		31
		<i>Saccharina coriacea</i>		1
		<i>Saccharina japonica</i>		18
		<i>Saccharina latissima</i>		8
		<i>Saccharina religiosa</i>		4
Lessoniaceae	<i>Ecklonia cava</i>	1		
	<i>Eisenia arborea</i>	2		
	<i>Eisenia bicyclis</i>	108		

contamination but also valuable radioecological insights into the behaviour of contaminants and the species they interact with.

The REML model development for this study builds on the work of Beresford and Willey (2019) to include site nested within study, as well as a further random factor: year. The addition of year as a random factor helps to account for factors that might change over time such as the constituents of radioactive discharges, of climatic factors. Including both these developments were found to explain a greater portion of the variance between taxa, and lead to be better model fit, than study and site alone. The nesting of site within study accounts for the likelihood that the multiple studies may sample from the same site, reducing the risk of pseudo-replication. Study accounted for the most variation in absolute value, followed by location nested within study, with year accounting for a small but still significant proportion of variance, likely due to annual changes in site discharges (Table 2). Only a small amount of the variance could not be attributed to a random factor and was therefore residual. Modelling the scrambled data set and the randomly generated data set produced poor fits, strongly suggesting that the model was explaining a significant proportion of the variation in actual data using the selected factors and model structure. Hierarchical ANOVA on scrambled and random datasets produced no statistically significant effects. We conclude that the ERMs for species when adjusted to the IAEA TF provide useful estimates of TF in the 63 species across 7484 activity concentrations measures at 350 sites.

The IAEA report a geometric mean TF value of 24.0 ± 5.3 for Cs in macroalgae. The range in REML estimated TF values for the 62 species of brown macroalgae in this database is from 1.5 to 132.4, with a geomean of 24 ± 5.3 , as it is anchored to the IAEA value (Fig. 1). The values estimated here, based on probably the most wide-ranging inter-species comparison of ^{137}Cs uptake by the Phaeophyceae yet reported, suggests that the TF for brown seaweed may differ significantly between taxa, and require a range of representative values for different coastal communities. The model used cannot account for all the variation due to the selected factors but has accounted for a significant proportion of it (see Table 2), revealing some significant inter-species differences.

The differences in estimated TFs observed between species is consistent with findings from the few other studies available. For example, in the Aegean Sea, Sawidis et al. (2003) found differences observed in ^{137}Cs uptake between species and attributed it to variation in morphology and life history. Zalewska and Saniewski (2011) measured ^{137}Cs in different species of seaweed from the Baltic Sea and reported that transfer factor differed between species with *Ectocarpus siliculosus* having a higher TF than *Pyaiella littoralis* which in turn had a higher TF than *F. vesiculosus*. This is mirrored in the REML model outputs for species. Both Sawidis et al. (2003) and Zalewska and Saniewski (2011) suggested these differences in TF can be attributed to morphological features such as life history and surface area. As traits such as these inform taxonomic classification, it is feasible that they may contribute to the taxonomic signal in ^{137}Cs uptake reported here.

The estimated TFs reported here indicate that there is significant variation in the uptake of ^{137}Cs between taxa and that taxonomic position might therefore be useful for predicting transfer. Hierarchical ANOVA based on the taxonomy of Phaeophyceae (Bringloe et al., 2020;

Guiry and Guiry, 2023) found significant variation at all taxonomic levels tested to the 0.05 confidence level (Genus $P = 0.024$; Family $P = 0.003$, and Order $P = 0.002$). The taxonomic effect becomes more evident at higher levels of the taxonomic hierarchy, as indicated by the decreasing P-value. Although taxonomic classification is not a strict indicator of evolutionary distance between species, the fact that there is a more significant difference between groups at the order level, compared to family, and more significant difference at the family compared to genus indicates that the more evolutionarily distant a species is, the more likely it is to have a different TF. Organismal traits that are more likely to have different values in more distantly related species are widely analysed in trait-based ecology, particularly in phylogenetic analysis. The phylogeny of a group of organisms is its evolutionary history and can be used to understand the effect of evolutionary distance in the distribution of trait values. The species is a reproductive unit and there is no reason, *a priori*, why variation in many trait values, including TF, should all reside at this level so larger taxonomic units can be used to understand trait values. The data reported here suggests that variation in ^{137}Cs uptake into Phaeophyceae does not all reside at the species level and that membership of a higher taxonomic groups could be used to estimate TFs for species when they are not available. Phylogenetic analysis might help to refine such predictions. A similar observation was made for Cs in terrestrial plants, where taxonomic effects were found at a variety of levels of the taxonomic hierarchy (Beresford and Willey, 2019). In the estimated TFs reported here the effect on ^{137}Cs transfer at the Order level is shown in Fig. 2. One way ANOVA comparison between Orders gives a P-value of 0.031, indicating that, overall, taxonomic Order explains a significant amount of variation between species. Unplanned multiple comparison Tukey-Kramer test was used to compare pairs of Orders within the dataset. Two pairs were found to be significant at the 95% level; Desmarestiales - Dictyotales ($P = 0.044$) and Dictyotales - Ishigeales ($P = 0.017$). If the IAEA TF value is broadly representative of mean transfers for algae, the findings reported here might be used to provide more specific values for any brown seaweed species based on the Orders it is classified in.

The data used for the analysis reported here is taxonomically unbalanced as studies tend to focus on certain species for reasons such as availability at targeted locations, uses, or site accessibility. The use of scrambled and random data shows that the significant taxonomic effects reported do not arise because of the sampling bias in the data but it is still important to realise that some Orders and Families of Phaeophyceae are not represented here. Only 6 out of 19 brown macroalgae orders are represented, however, those Orders included here represent nearly 89% of all brown seaweed species (Bringloe et al., 2020). Some taxa, such as *Fucus* spp., are over-represented due to their common role as an indicator species in radiological monitoring programmes. This is a commonly encountered challenge to analysing taxonomic effects on species traits but is also a useful guide to sampling gaps that might be targeted for future measurements to refine descriptions of taxonomic effects on variation (Broadley et al., 1999).

The description of inter-species differences in transfer and the taxonomic effects reported above might help improve the interpretation of radioecological monitoring data in coastal environments in several ways. The Fucales is a large order of brown macroalgae that contains many common species, including the genus *Fucus*. *Fucus* species are frequently used as indicators of environmental radioactivity around the coasts of Northwest Europe. Many monitoring efforts solely rely on *Fucus* spp. (in particular, *Fucus vesiculosus* and *Fucus serratus*) to determine radioactivity levels and doses to the coastal marine ecosystem (Environment Agency et al., 2021; EPA, 2017). The model estimated that the TF for Fucales is very similar to the IAEA mean value for macroalgae, indicating that using the IAEA value for dose assessments is likely to produce similar values to those actually occurring. This is likely due the disproportionate representation of *Fucus* spp. within the IAEA data used to calculate this value. The variation between orders reported here suggests that when using *Fucus* spp. as an indicator, it should be

Table 2

The REML criterion and variance from the actual data and two artificially generated datasets indicate that the collated dataset has a stronger model fit and random factor signals.

		Dataset		
		Actual	Scrambled	Randomised
REML Criterion		15970.1	35510.8	46827.9
Variance:	Study	5.5936	0.0000	0.0009
	Study:Location	1.1285	0.0000	0.0000
	Sampling Year	3.2240	0.0000	0.0069
	Redidual	0.3997	6.9110	31.7043

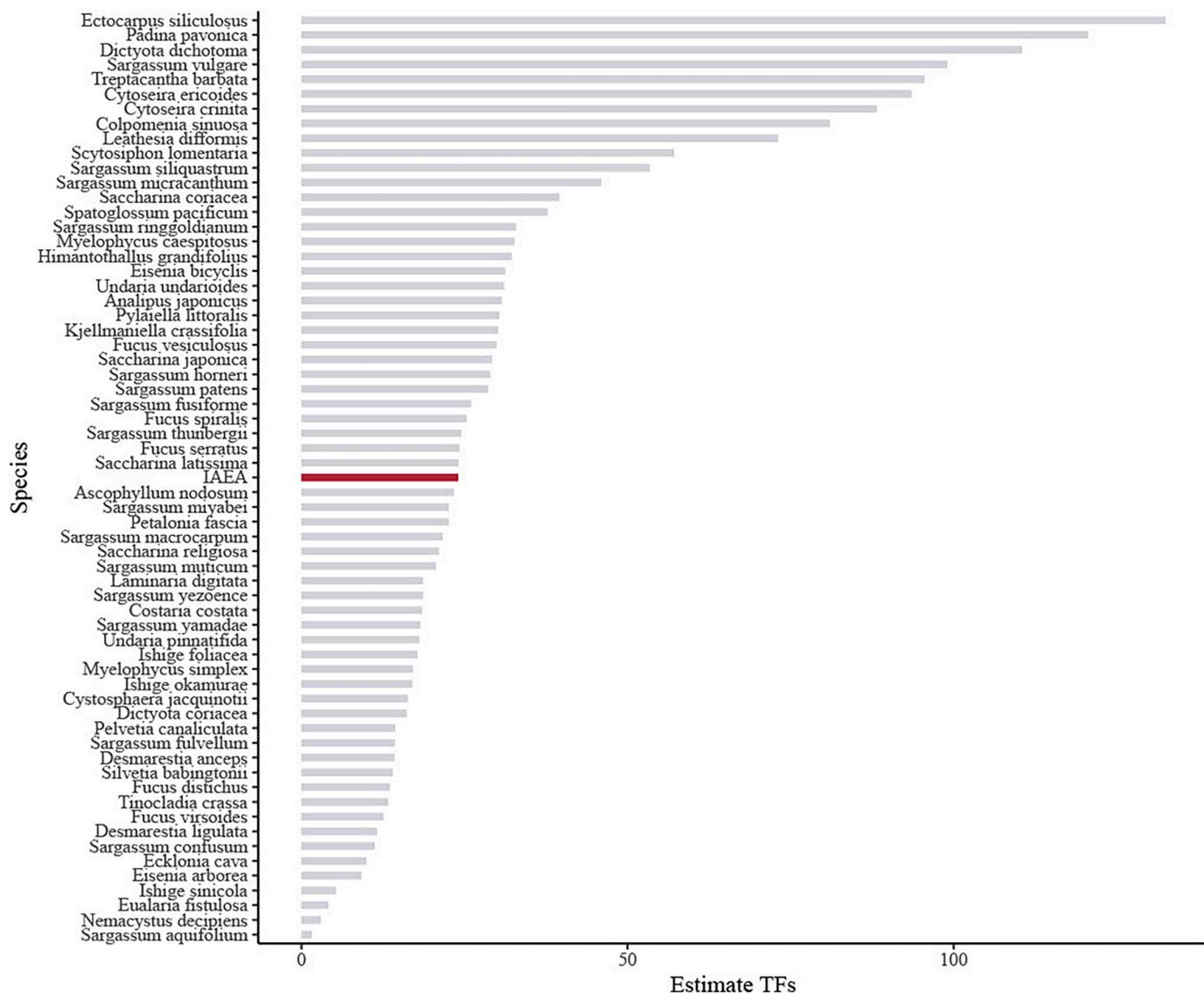


Fig. 1. Estimated transfer factors (TFs) showing the difference in values between brown seaweed species included in the database. TFs were estimated by using a linear mixed-model with species as fixed factor and study, sampling site and year as random factors to produce Estimated Relative Means (ERMs) for species. The ERMs were then adjusted to the IAEA value of 24 for macroalgae (n = 7484 across 350 sites and 63 species). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

realised that it represents an approximate average value for brown seaweeds. However, the use of other species such as those within the Dictyotales or Ishigeales orders, may not represent average values, but instead upper and lower transfers respectively. This is particularly important to note when carrying out monitoring outside of the North Atlantic area in areas where *Fucus* spp. are not as prevalent. The taxonomic effects identified in this study can contribute to better selection of taxa in other parts of the world as well as provide more accurate values for regional seaweed communities globally.

4. Conclusions

Understanding the taxonomic differences in transfer between brown seaweeds for which radioanalytical measurements are available can enrich the interpretation of monitoring data and also enable the prediction of transfer into species for which data is not available. The data reported here also suggests ways in which radiological monitoring in coastal environments using brown seaweeds can be developed for example, the calculation of more accurate transfer values, prediction for

sites and taxa with little available data or physical samples. It seems clear that for some taxa of brown seaweed, TFs can be adjusted from the IAEA mean in order to improve predictions of ¹³⁷Cs transfer. At key sites with particular species, this could provide greater confidence to predicted transfers and doses. At sites with brown seaweeds species for which there are no data the results reported here can provide a guide to selecting an appropriate TF based on taxonomic position. Further, in emergency situations following accidents, estimated TFs could be provided for species for which there are currently no data. Finally, the data reported here could be useful to guide the selection of species for monitoring – the existence of a biomonitor such as *Fucus* spp. at a particular site might not be necessary because the taxonomic position of a species that does occur can be used to understand what its value represents for radionuclide transfer. Describing this inter-species variation in transfer can significantly enrich the interpretation of monitoring data collected from brown seaweed at particular sites – what the activity concentrations in a particular species at a particular site represent can now be better understood within transfer to the Phaeophyceae in general.

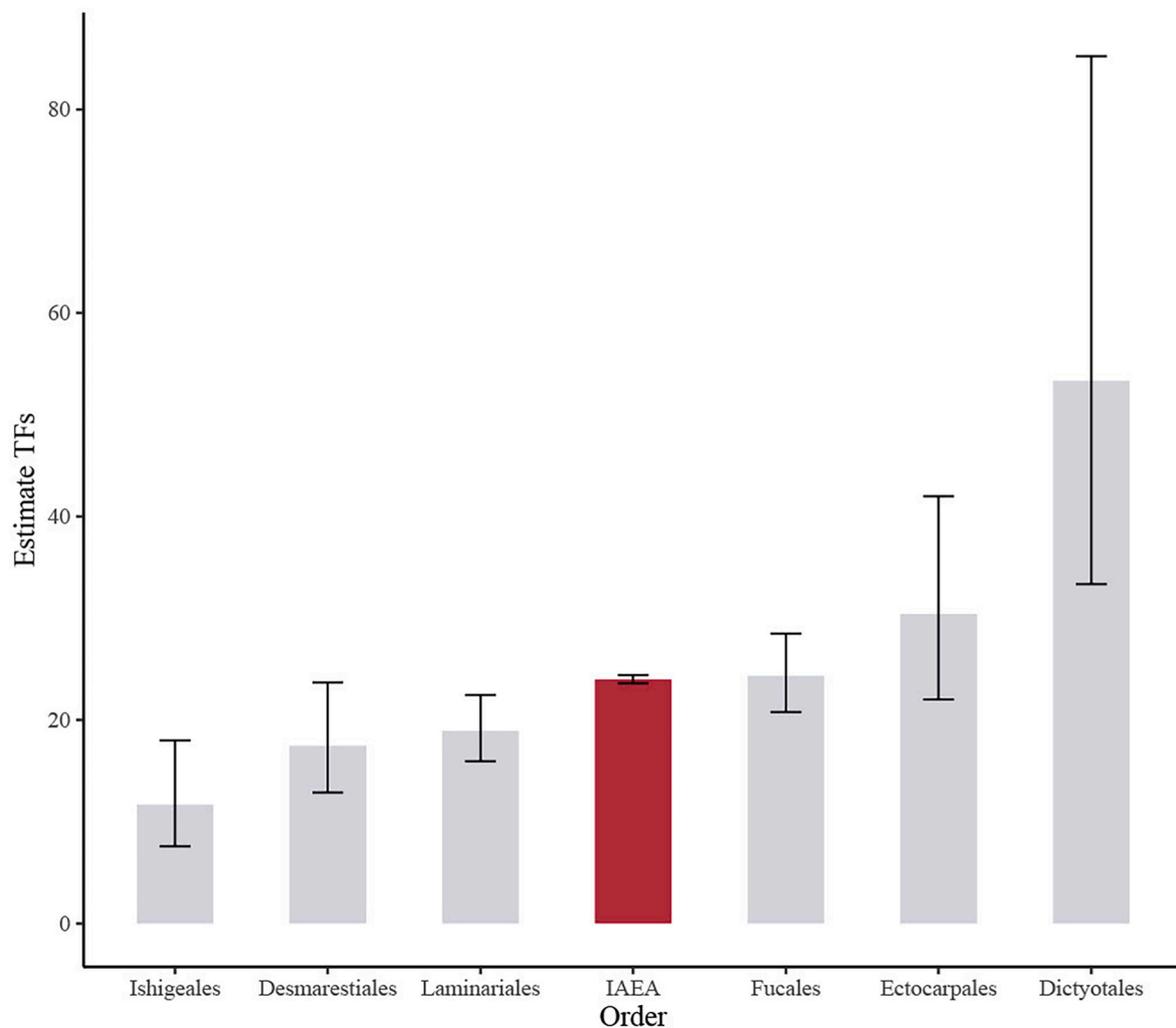


Fig. 2. Geometric mean estimated transfer factors (TFs) for brown seaweed Orders (error bars for orders = 95% geometric confidence interval, for ANOVA $P = 0.031$, error bar for IAEA = 95% confidence interval). TFs were estimated using a linear mixed-model with species as fixed factor and study, site and year as random factors to model estimated relative means for species and then anchoring them to the IAEA value of 24 for macroalgae. ($n = 7484$ across 350 sites and 63 species). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

We conclude that it's likely that there are taxonomic differences in the transfer of ^{137}Cs to brown seaweeds. These are significant up the taxonomic hierarchy, can enrich the interpretation of current data, provide improved estimates of TF, and predict values for brown seaweeds in which ^{137}Cs transfer has never been measured. This approach can provide greater confidence in the TF values used in dose assessment models such as ERICA, helping to refine radiological monitoring conducted in coastal environments. Brown seaweeds have a global distribution, primarily in non-tropical waters (Assis et al., 2020) and the work described here might be helpful with monitoring and assessment in environments for which there are few values for brown seaweeds. Further work, which might extend and refine the suggestions made above, should focus on extending the taxonomic range of the data set to provide a more balanced dataset, and on carrying phylogenetic analysis on the data, as detailed in Willey (2010). Phylogenies constructed from genetic information are based on evolutionary history and can be used to enhance predictions made from taxonomies (which do not represent evolutionary distance as strictly as do phylogenies). Given the radiological importance of coastal environments, it might be very useful to extend this work to other radionuclides and other groups of organisms.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Supplementary file is included containing references to all data sources.

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