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Nuclear reprocessing-related radiocarbon (^{14}C) uptake into UK marine mammals

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ABSTRACT

To evaluate the transfer of Sellafield-derived radiocarbon (^{14}C) to top predators in the UK marine environment, ^{14}C activities were examined in stranded marine mammals. All samples of harbour porpoise (*Phocoena phocoena*) obtained from the Irish Sea showed ^{14}C enrichment above background. Mammal samples obtained from the West of Scotland, including harbour porpoise, grey seals (*Halichoerus grypus*) and harbour seals (*Phoca vitulina*) showed ^{14}C enrichment but to a lesser extent. This study demonstrates, for the first time, enriched ^{14}C is transferred through the marine food web to apex predators as a consequence of ongoing nuclear reprocessing activities at Sellafield. Total Sellafield ^{14}C discharge activity 24 months prior to stranding and, in particular, distance of animal stranding site from Sellafield are significant variables affecting individual ^{14}C activity. ^{14}C activities of West of Scotland harbour porpoises suggest they did not forage in the Irish Sea prior to stranding, indicating a high foraging fidelity.

1. Introduction

During reprocessing of nuclear materials at the Sellafield Ltd. facility (Fig. 1A), low-level radioactive waste, including ^{14}C (half-life 5730 years), is discharged to the Northeast Irish Sea, primarily as dissolved inorganic carbon (DIC; Begg et al., 1992, Cook et al., 1995). Dissolved ^{14}C is subject to solution transport and largely dispersed northwards from the Irish Sea by prevailing currents through the North Channel (Gulliver et al., 2001) and around the Scottish coastline to the North Sea (Gulliver et al., 2004). ^{14}C enters the marine food web via the efficient uptake of soluble ^{14}C in DIC during photosynthesis by primary producing organisms, i.e. phytoplankton (Cook et al., 1995; Cook et al., 1998; Cook et al., 2004; Muir et al., 2017; Tierney et al., 2017). In the UK, Sellafield discharges of ^{14}C have dominated enriched activities in the marine environment. Although Amersham International plc (now GE Healthcare), Cardiff, was an additional source causing localised enriched ^{14}C activities (Cook et al., 1998), the ^{14}C discharge activity from this site was minimal between 2000 and 2010 and negligible since 2010 (RIFE, 2016).

Since the early 1990s there have been significant changes in Sellafield ^{14}C discharges to the Irish Sea as described in detail by Muir et al. (2017). Briefly, the average discharged ^{14}C activity from 1984 to 1993 was 1.78 Tera Becquerels per year (TBq year^{-1}). An increase in the volume of waste reprocessed and a change in discharge policy in 1994, from an atmospheric route to marine discharge routes, resulted in an increase in marine ^{14}C discharges. The annual discharged activity peaked in 2003 at 16.87 TBq and remained high relative to pre-1994 releases with an average of 7.63 TBq year^{-1} until the end of 2015 (RIFE, 2016; Muir et al., 2017).

Recent studies of Sellafield ^{14}C discharges have considered the accumulation of ^{14}C within intertidal environments (Cook et al., 2004; Muir et al., 2015; Tierney et al., 2016) and the biological uptake and transfer of ^{14}C through a major part of the marine food webs of the Irish Sea and West of Scotland (Muir et al., 2017; Tierney et al., 2017). The latter studies reported enriched activities in a range of marine species occupying the lowest (phytoplankton) to middle-upper (e.g. piscivorous fish) trophic levels and described the trophic transfer of Sellafield-derived ^{14}C previously observed for intertidal organisms (Cook et al., 2004). Here we examine ^{14}C activities in marine mammals that occupy the upper trophic levels of the UK marine environment, and which are

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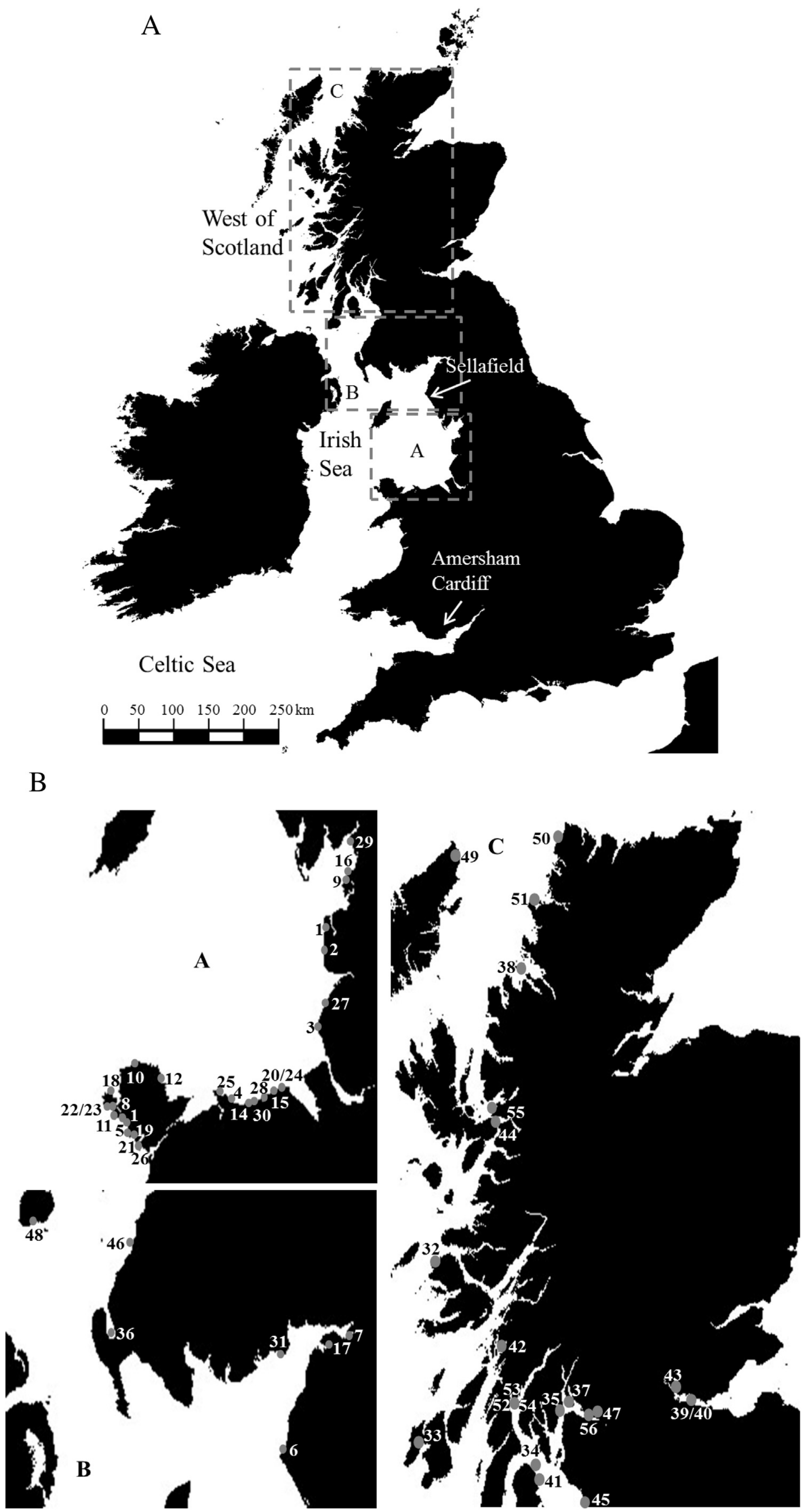

Fig. 1. A. Map of UK and Ireland indicating study areas (Irish Sea and West of Scotland) and the location of the Sellafield nuclear fuel reprocessing facility.

B. Maps of study areas and stranding locations in the southern Irish Sea (A), the transition area between the Irish Sea and West of Scotland (B) and the West of Scotland with additional sites on the Scottish east coast (C).

Table 1
^{14}C results for mammal samples obtained from the Cetacean Stranding Investigation Programme (CSIP). All samples were from Irish Sea sites.

Sample number	CSIP sample code	Species	Approximate stranding location	Year of stranding	Gross ^{14}C activity (Bq kg^{-1}C)	Net ^{14}C activity (background corrected; Bq kg^{-1}C)
1	SW1991/71	Harbour Porpoise	Cymyran Bay	1991	280 ± 2	31 ± 2
2	SW1991/135	Harbour Porpoise	Blackpool	1991	302 ± 2	53 ± 2
3	SW1993/34	Harbour Porpoise	Formby Point	1993	351 ± 2	102 ± 2
4	SW2000/131	Harbour Porpoise	Rhos-on-Sea	2000	338 ± 2	89 ± 2
5	SW2000/191	Harbour Porpoise	Rhosneigr	2000	425 ± 2	176 ± 2
6	SW2002/114	Harbour Porpoise	Whitehaven	2002	674 ± 3	425 ± 3
7	SW2002/159	Harbour Porpoise	Port Carlisle	2002	657 ± 3	408 ± 3
8	SW2002/170	Harbour Porpoise	Trearddur Bay	2002	287 ± 2	38 ± 2
9	SW2005/245	Harbour Porpoise	Morecambe	2005	353 ± 2	104 ± 2
10	SW2006/54	Harbour Porpoise	Cemaes Bay	2006	363 ± 2	114 ± 2
11	SW2006/308A	Harbour Porpoise	Trearddur Bay	2006	315 ± 2	66 ± 2
12	SW2008/37	Harbour Porpoise	Dulas Bay	2008	356 ± 2	107 ± 2
13	SW2008/60	Harbour Porpoise	Blackpool	2008	365 ± 2	116 ± 2
14	SW2010/133	Harbour Porpoise	Abergele	2010	609 ± 3	360 ± 3
15	SW2010/152	Harbour Porpoise	Rhyl	2010	489 ± 3	240 ± 3
16	SW2010/233	Harbour Porpoise	Morecambe	2010	608 ± 3	359 ± 3
17	SW2010/338	Harbour Porpoise	Anthorn	2010	308 ± 2	59 ± 2
18	SW2011/35	Harbour Porpoise	Holyhead	2011	429 ± 2	180 ± 2
19	SW2011/109	Harbour Porpoise	Porth Cwyfan	2011	395 ± 2	146 ± 2
20	SW2012/195	Harbour Porpoise	Rhyl	2012	286 ± 2	37 ± 2
21	SW2013/45	Harbour Porpoise	Aberffraw	2013	296 ± 2	47 ± 2
22	SW2013/273	Harbour Porpoise	Porth Dafarch	2013	524 ± 2	275 ± 2
23	SW2013/327	Harbour Porpoise	Porth Dafarch	2013	315 ± 2	66 ± 2
24	SW2013/381	Harbour Porpoise	Prestatyn	2013	359 ± 2	110 ± 2
25	SW2014/86	Harbour Porpoise	Landudno	2014	428 ± 2	179 ± 2
26	SW2014/272	Harbour Porpoise	Newborough	2014	288 ± 2	39 ± 2
27	SW2014/475	Harbour Porpoise	Ainsdale	2014	326 ± 2	77 ± 2
28	SW2015/6	Harbour Porpoise	Rhyl	2015	285 ± 2	36 ± 2
29	SW2015/123	Harbour Porpoise	Carnforth	2015	325 ± 2	76 ± 2
30	SW2015/224	Harbour Porpoise	Abergele	2015	324 ± 2	75 ± 2

potentially at risk from increased radioactive dose due to uptake of bioavailable contaminant radionuclides.

Harbour seals (*Phoca vitulina*) are locally resident and typically forage within 40 km of their haul-out sites (Thompson et al., 1998). The foraging range of grey seals (*Halichoerus grypus*) can be much larger but individuals will always return to the same breeding site and they are resident to the British Isles (McConnell et al., 1999). Less is known about the distribution and behaviour of harbour porpoise (*Phocoena phocoena*) in UK waters, however, population structure analysis of the northeast Atlantic has indicated that there is a subpopulation in British waters (De Luna et al., 2012) and ^{137}Cs measurements of their tissues suggest regional residency around the UK (Berrow et al., 1998; Watson et al., 1999). Resident mammals from the Irish Sea and West of Scotland (defined here as the area located to the north of the North Channel) will be susceptible to ^{14}C enrichment as they spend most or all of their foraging time in waters enriched in ^{14}C and consequently, containing prey species enriched in ^{14}C. In these regions, Sellafield ^{14}C will be transferred through the food chain to marine mammals as has been observed for other species (Muir et al., 2017; Tierney et al., 2017).

Harbour porpoise, harbour seal and grey seal are generalist predators, although with some dietary specialisations. Overlaps in their diets have been observed in Irish coastal waters where harbour seals and grey seals predate on a number of the same species, as do grey seals and harbour porpoises (Hernandez-Milian, 2014). However, there may be little direct competition between these mammal species as they target prey of different sizes (Hernandez-Milian, 2014). Dietary differences have been observed between porpoise populations in the Irish Sea and the West of Scotland. Harbour porpoises in Irish Sea waters, for example, show a higher presence of pelagic fish such as herring (*Clupea harengus*) in their diet (Hernandez-Milian, 2014), whereas Scottish coastal harbour porpoises predate more on sandeels (e.g. *Hyperoplus* spp.; Santos et al., 2004), although gadoid species are important prey species for both. This difference in diet could be due to prey availability in different environments but it could also indicate a change in diet during the period between studies. Some genetic research has indicated that the Irish Sea harbour porpoise may be a sub-population within the UK population (Andersen et al., 2001), although a review of recent literature has found no clear evidence for distinct populations on the west coast of Britain (IAMMWG, 2015) and Fontaine et al. (2017) showed that there is a genetic continuum in UK waters.

The samples described in this study come from animals that were found dead, or died at the stranding site. Studies of ^{137}Cs activities (a radionuclide that was historically discharged to the Irish Sea from Sellafield) in marine mammals stranded in the UK and Ireland have shown that Celtic Sea activities are significantly lower than Irish Sea activities (Berrow et al., 1998) and that activities decrease with distance of stranding site from Sellafield (Watson et al., 1999). These findings demonstrate that anthropogenic radionuclides are transferred through the food chain to marine mammals and suggest that stranding sites are approximately within the same region in which the animal has been foraging.

The aims of this study were to: 1) evaluate the transfer of Sellafield-derived ^{14}C to top predators in the UK marine environment; 2) examine the spatial distribution of ^{14}C relative to dilution with distance from Sellafield; and 3) determine the effect of temporal changes in Sellafield ^{14}C discharge activities and subsequent transfer through the food chain to marine mammals.

2. Methods

Access to the Scottish Marine Animal Stranding Scheme (SMASS) and Cetacean Strandings Investigation Programme (CSIP) sample archives provided the opportunity to consider samples from different mammal species at various locations over a relatively long time-period. The species of interest – harbour porpoise, grey seal and harbour seal – were selected as they represent resident UK marine mammal species, of which a number of samples were available. Muscle tissue samples of stranded mammals from the Irish Sea and the West of Scotland (Fig. 1B)

Table 2

^{14}C results for mammal samples obtained from the Scottish Marine Animal Stranding Scheme (SMASS). All samples were from the West of Scotland with the exception of one from the Irish Sea and three from the Scottish east coast.

Sample number	SMASS sample code	Species	Approximate stranding location	Year of stranding	Gross ^{14}C activity (Bq kg^{-1}C)	Net ^{14}C activity (background corrected; Bq kg^{-1}C)
31	M1970/92	Harbour Porpoise	Southerness (Irish Sea)	1992	372 ± 2	123 ± 2
32	M0105/93	Harbour Porpoise	Isle of Mull	1993	255 ± 1	6 ± 1
33	M1106/93	Harbour Porpoise	Islay	1993	257 ± 1	8 ± 1
34	M210/03	Harbour Porpoise	Dunoon	2003	403 ± 2	154 ± 2
35	M197/03	Harbour Porpoise	Isle of Arran	2003	329 ± 2	80 ± 2
36	M186/04	Harbour Porpoise	Stranraer	2004	374 ± 2	125 ± 2
37	M228/04	Harbour Porpoise	Loch Long	2004	368 ± 2	119 ± 2
38	M241/11	Grey Seal	Gruinard river	2011	263 ± 1	14 ± 1
39	M62/12A	Harbour Porpoise	Bo' ness (east coast)	2012	262 ± 1	13 ± 1
40	M87/12	Harbour Porpoise	Bo' ness (east coast)	2012	264 ± 1	15 ± 1
41	M173/12	Harbour Porpoise	Isle of Arran	2012	309 ± 2	60 ± 2
42	M082/13	Harbour Seal	Loch Melfort	2013	260 ± 1	11 ± 1
43	M092/13	Harbour Porpoise	Alloa (east coast)	2013	253 ± 1	4 ± 1
44	M198/13	Harbour Seal	Isle of Skye	2013	254 ± 1	5 ± 1
45	M5/14	Harbour Seal	Troon	2014	284 ± 2	35 ± 2
46	M68/14	Harbour Porpoise	Girvan	2014	238 ± 1	−11 ± 1
47	M134/14	Harbour Porpoise	River Clyde	2014	294 ± 2	45 ± 2
48	M139/14	Harbour Porpoise	Kintyre	2014	398 ± 2	149 ± 2
49	M147/14	Harbour Porpoise	Isle of Lewis	2014	258 ± 2	9 ± 2
50	M279/14	Harbour Porpoise	Kinlochbervie	2014	242 ± 2	−7 ± 2
51	M319/14	Harbour Porpoise	Clacktoll	2014	254 ± 1	5 ± 1
52	M378/14	Harbour Seal	Loch Fyne	2014	262 ± 2	13 ± 2
53	M384/14	Grey Seal	Loch Fyne	2014	271 ± 2	22 ± 2
54	M385/14	Grey Seal	Loch Fyne	2014	265 ± 1	16 ± 1
55	M137/15	Harbour Seal	Isle of Skye	2015	259 ± 2	10 ± 2
56	M147/15	Harbour Seal	River Clyde	2015	254 ± 2	5 ± 2

were identified formally by CSIP and SMASS respectively (Tables 1 and 2). Three time-periods were significant: 1) Pre-1994, when ^{14}C discharges were relatively low, 2) 2001–2004, which encompasses the period of peak ^{14}C discharge, and 3) 2011–2015, to examine contemporary ^{14}C activities in marine mammals. Most of the samples came from these three time-periods, although additional Irish Sea samples were analysed from other years (Tables 1 and 2). Three samples from the Scottish east coast (Firth of Forth) were also analysed to identify the extent and influence of Sellafield discharges at greater distances from the facility.

The analytical techniques employed are described in detail in Muir et al. (2017) and are briefly summarised here. Muscle tissue samples from each organism were freeze-dried and approximately 15 mg were combusted (850 °C) in sealed quartz tubes, according to the method of Vandeputte et al. (1996) to liberate CO_2 gas. CO_2 was cryogenically purified (under vacuum) and converted to graphite (Slota et al., 1987) prior to $^{14}C/^{13}C$ isotope ratio measurement by accelerator mass spectrometry (AMS). Sub-samples of CO_2 were collected to determine the $\delta^{13}C$ value ($^{13}C/^{12}C$ stable isotope ratio) for calibration of natural fractionation of measured ^{14}C. Reported AMS fraction modern results were converted to specific activities (Bq kg^{-1}C) using the regime described by Mook and van der Plicht (1999). Error bars are omitted from figures, as analytical uncertainties for AMS measurements at SUERC are typically $< 0.5\%$ of the measured activity, and therefore indistinguishable in measured values. Statistical analyses and modelling were conducted using the software package R (R Development Core Team, 2016). Generalised least squares (GLS) regression models were used to identify significant variables and model fit was compared using the corrected Akaike information criterion (AICc; Venables and Ripley, 2002).

Due to natural production of ^{14}C and the legacy of ^{14}C from atmospheric testing of atomic weapons during the 1950s and 1960s, a baseline (or background) activity was quantified to determine enriched activities resulting from Sellafield discharges. Cook et al. (1998) defined the UK ^{14}C coastal marine background as 248 ± 1 Bq kg^{-1}C from west coast of Ireland samples that are free of UK coastal influences, i.e. Sellafield radionuclide discharges. Tierney et al. (2016) presented a new, but near-identical, UK ^{14}C background activity of 249 ± 1 Bq kg^{-1}C which has been used in subsequent studies (Muir et al., 2017; Tierney et al., 2017) and is used here.

3. Results

Analytical results for samples obtained from CSIP and SMASS are listed in Tables 1 and 2, respectively. Of the 56 marine mammal samples analysed, three were from grey seals, six from harbour seals and 47 from harbour porpoises. Two samples (46 and 50) were measured with ^{14}C activities below that of the current UK coastal marine background. These came from porpoises stranded in the West of Scotland in 2014 (238 ± 1 Bq kg^{-1}C and 242 ± 1 Bq kg^{-1}C). The highest measured (gross) activities were from two porpoises (samples 6 and 7) that stranded in the Irish Sea in 2002 (674 ± 3 Bq kg^{-1}C and 657 ± 3 Bq kg^{-1}C). One harbour seal sample and two grey seal samples (52, 53 and 54) from animals that died at approximately the same time in Loch Fyne (West of Scotland) showed similar ^{14}C activities (262 ± 1 Bq kg^{-1}C, 271 ± 1 Bq kg^{-1}C and 265 ± 1 Bq kg^{-1}C), respectively. Conversely, two young male harbour porpoises (samples 22 and 23) that stranded at Porth Dafarch (North Wales) on the Southern Irish Sea coastline at a similar time had a relatively large difference in measured activity (524 ± 2 Bq kg^{-1}C and 315 ± 2 Bq kg^{-1}C). The average ^{14}C activity across all Irish Sea samples was 388 Bq kg^{-1}C, compared to a significantly lower ^{14}C activity of 285 Bq kg^{-1}C for West of Scotland samples. Three samples from the Scottish east coast (39, 40 and 43) also show enriched ^{14}C activities (253 ± 1, 262 ± 1 and 264 ± 1 Bq kg^{-1}C).

No seal samples were obtained for the Irish Sea area and the number of seal samples was low overall, relative to that of harbour porpoise, resulting in a large activity difference between the ranges observed for seal (254–284 Bq kg^{-1}C) and porpoise (238–674 Bq kg^{-1}C) ^{14}C activities. As there was no significant difference between seal and porpoise ^{14}C activity in the West of Scotland, these species were grouped for statistical analysis.

A number of variables were considered to explain the measured ^{14}C activities, including distance (measured as distance from Sellafield by

Table 3
Statistical model descriptions. The AICc scores provide a comparative measurement of goodness of fit where lower values indicate a better model fit.

Model no.	Description	AICc
Model 1	Best model fit without log-transforming ^{14}C activity. Model included the variable distance ($p > 0.01$) but was not deemed significant.	644.6
Model 2	Log-transformed ^{14}C activity with variables: distance from Sellafield ($p < 0.001$) and year of stranding ($p > 0.01$).	152.6
Model 3	Log-transformed ^{14}C activity with variables: distance from Sellafield ($p < 0.0001$) and pDischarge 12 months prior to stranding ($p > 0.01$).	146.1
Model 4	Log-transformed ^{14}C activity with variables: distance from Sellafield ($p < 0.0001$) and pDischarge 24 months prior to stranding ($p > 0.01$).	143.9
Model 5	Log-transformed ^{14}C activity with variables: distance from Sellafield ($p < 0.0001$) and pDischarge 24 months prior to stranding including delay factor of 12 months for West of Scotland mammals ($p < 0.01$).	139.2
Model 6	Log-transformed ^{14}C activity with variable distance from Sellafield only ($p < 0.0001$).	147.2
Model 7	Log-transformed ^{14}C activity with variable pDischarge only ($p < 0.01$).	166.0

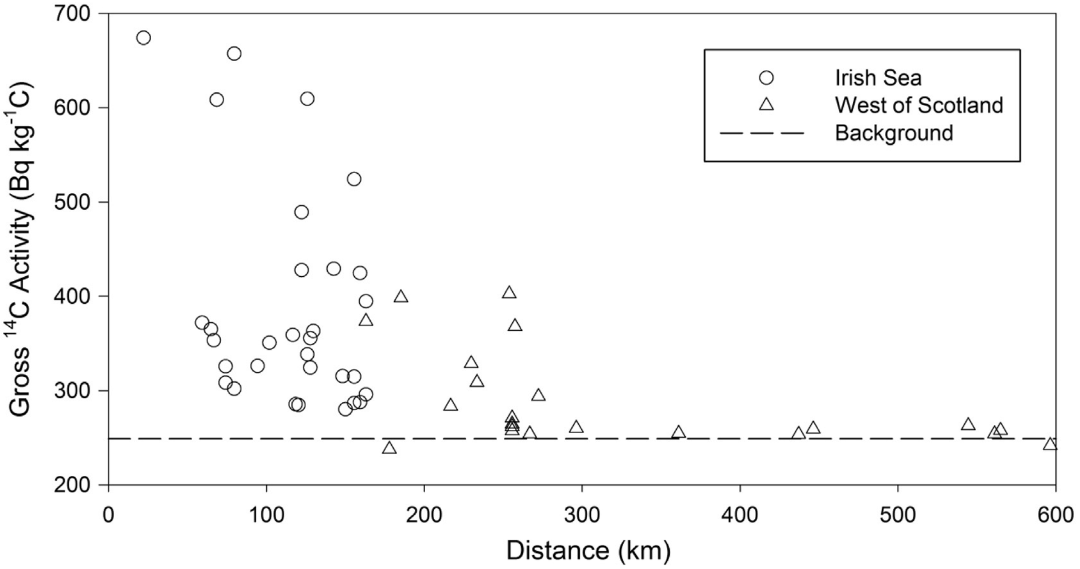

Fig. 2. Sample ^{14}C activities as a function of distance of stranding site from Sellafield by sea (Scottish east coast samples have been omitted for clarity).

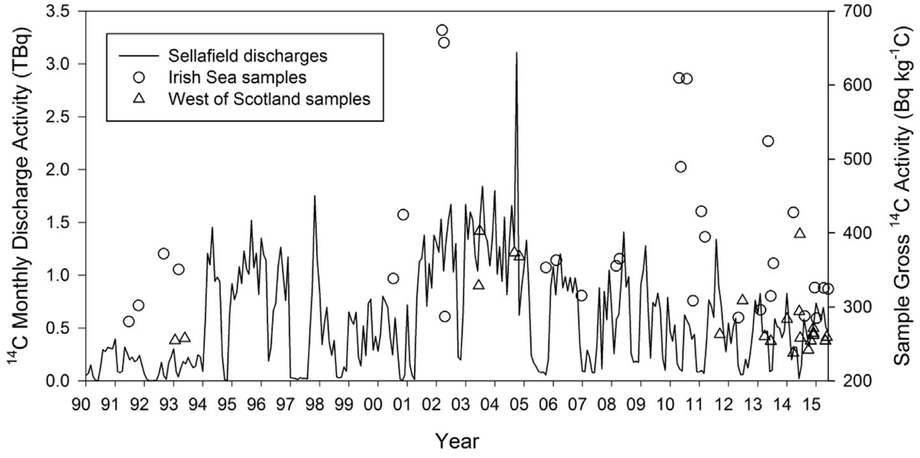

Fig. 3. Sellafield total monthly ^{14}C discharges made to the Irish Sea from January 1990 to June 2015 and sample ^{14}C activities.

sea in km), sex, age class (neonate, juvenile, sub-adult and adult), level of decomposition (freshly dead, slight and moderate decomposition), month of stranding and year of stranding. The data were explored prior to statistical analyses and linear model assumptions checked. As correlation in the data residuals was detected, generalised least squares (GLS) regression was used with a simple correlation structure (AR1) and model descriptions, and AICc scores are given in Table 3. Initial model fitting of ^{14}C activity found the best fit (lowest AICc score) when only including the predictor variable distance (Model 1). However, the relationship between sample activity and distance was not significant ($p > 0.01$). Distance appears to have an exponential influence on activity as stranding site gets closer to Sellafield (Fig. 2). Model fitting of log-transformed ^{14}C activity found distance significantly ($p < 0.001$) affected ^{14}C activity and the model with the lowest AICc score also included year, despite this variable having little significance ($p > 0.01$; Model 2). It is likely that year improved model fit due to the temporal changes in Sellafield ^{14}C discharges having some effect on individual ^{14}C activities, although this is not obvious in Fig. 3. A new variable was considered describing discharge activity prior to stranding (pDischarge), where pDischarge is the sum of total monthly ^{14}C activities discharged from Sellafield (*a*) for a number of months (*n*) prior to the month of stranding (*s*; Eq. (1)). Periods of 6, 12, 24 and 36 months were considered and 12 (Model 3) and 24 months (Model 4) were found to improve model fit, however, pDischarge had little significance ($p > 0.01$).

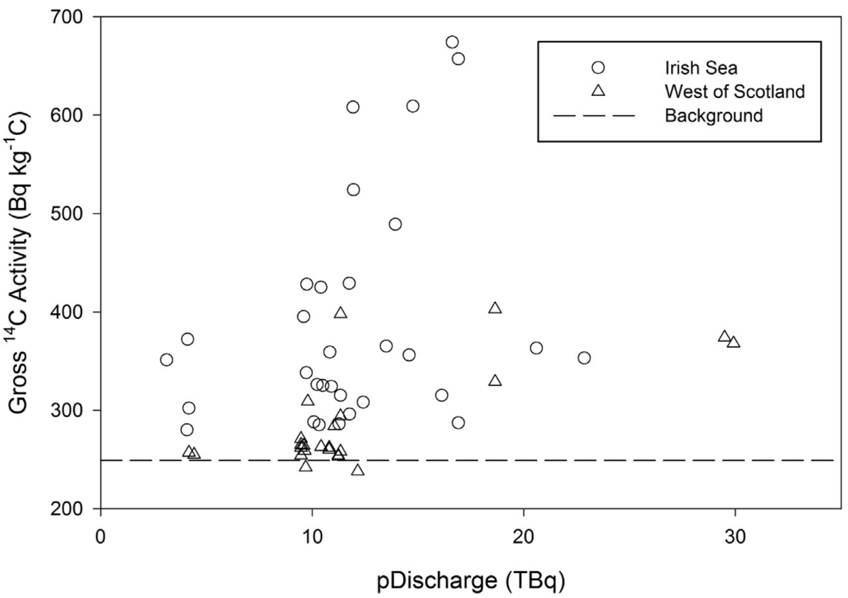

Fig. 4. Sample ^{14}C activities as a function of Sellafield ^{14}C discharge activity for 24 months prior to stranding, including a 12-month delay factor for West of Scotland samples.

$$pDischarge = \sum\nolimits_{i=s}^{n} a_i \tag{1}$$

Sellafield discharges will not reach the West of Scotland environment immediately. Estimates for transit times of other highly soluble radionuclides discharged from Sellafield (^{134}Cs, ^{137}Cs and ^{99}Tc) range from 3 to 18 months (Jefferies et al., 1973; Kershaw and Baxter, 1995; Kershaw et al., 2004). To account for transit time in calculating pDischarge for West of Scotland samples, a delay factor (d) was used (Eq. (2)). Although a number of delay factors were considered, a factor of 12 months was statistically significant ($p < 0.01$), and improved model fit when included with distance ($p < 0.0001$; Model 5). A delay factor of 12 months meant pDischarge for West of Scotland samples was the total ^{14}C discharge activity from 12 to 36 months prior to stranding.

$$pDischarge = \sum_{i=s-d}^{n} a_i \tag{2}$$

Distance alone (Model 6) did not improve model fit and although pDischarge was significant, there is no obvious correlation between ^{14}C activity and pDischarge (Fig. 4) and pDischarge alone (Model 7) did not improve the model fit. Therefore, the overall best model fit for mammal ^{14}C activity included the predictor variables of: distance of stranding from Sellafield and total Sellafield ^{14}C discharge activity 24 months prior to stranding, including a 12-month delay for West of Scotland animals (Model 5). For every kilometre increase away from Sellafield, this model predicts an estimated 0.3% decrease in sample activity. For every TBq increase in discharged ^{14}C activity during the 24 months prior to stranding, the model predicts an estimated 6.5% increase in sample activity. The combined effect of distance and prior Sellafield discharges on sample ^{14}C activity is illustrated in Fig. 5 where distance is normalised to pDischarge and the scatter again indicates an exponential relationship.

4. Discussion

The two West of Scotland samples (46 and 50) with below UK coastal marine ^{14}C background activities (238 ± 1 Bq kg^{-1}C and 242 ± 2 Bq kg^{-1}C) were from young female porpoises that stranded hundreds of kilometres apart (approximately 178 km and 596 km from Sellafield respectively). Similar activities were observed in phytoplankton and zooplankton in the West of Scotland (Tierney et al., 2017). It is not clear why these ^{14}C activities would be below this background value, however, the depleted plankton activities were linked to a possible source of older water, possibly derived from upwelling of deep Atlantic water or another Atlantic source, reducing ambient ^{14}C activities (Tierney et al., 2017). Natural ^{14}C activities are not homogenous and the cited UK coastal marine ^{14}C background activity does not represent the "oceanic background" for the entire northeast Atlantic. This may vary as ^{14}C produced by atomic weapon testing decays in different hydrographic and biogeochemical settings (Scourse et al., 2012). The lower activities could result from the animals previously inhabiting a region with a lower ambient ^{14}C activity before stranding at these sites.

The relatively high ^{14}C activities (674 ± 3 and 657 ± 3 Bq kg^{-1}C) observed in harbour porpoises stranded close to Sellafield in 2002 (samples 6 and 7) coincides with the period of peak ^{14}C discharges. Although peak ^{14}C discharge occurred in 2003 (17 TBq), cumulative increases in ^{14}C discharge were made to the Irish Sea in both 2001 (9.5 TBq) and 2002 (13 TBq). Similarly, higher activities (608 ± 3 Bq kg^{-1}C and 609 ± 3 Bq kg^{-1}C) were measured in two harbour porpoises that stranded in different areas of the Irish Sea in 2010 (samples 14 and 16). Although the 2010 annual discharge (4.4 TBq) was low relative to the peak discharges, the 2009 annual discharge (8.2 TBq) was the highest between 2006 and 2015. However, a porpoise (sample 17) that stranded relatively close to Sellafield in 2010 (74 km away) had a comparatively low activity (308 ± 2 Bq kg^{-1}C). This individual died from starvation and an extended period of limited foraging with little or no food intake from the eastern Irish Sea may help explain this individual's anomalously low ^{14}C activity.

It is reasonable to assume that samples obtained from animals of the same or similar species that stranded in the same location at the same time of year, would have comparable ^{14}C activities. This was observed in three seal samples (52, 53 and 54) from Loch Fyne (West of Scotland; 262 ± 1 Bq kg^{-1}C, 271 ± 1 Bq kg^{-1}C and 265 ± 1 Bq kg^{-1}C). The fact that two porpoise samples (22 and 23) from Porth Dafarch (North Wales) had significantly different activities (524 ± 2 Bq kg^{-1}C and 315 ± 2 Bq kg^{-1}C) illustrates the difficulty in making assumptions based on stranding location alone. Animals that inhabit different areas might strand in the same area due to a number of variables including changes in water masses, wind patterns, and bloating of animal carcasses prior to stranding. However, diet source will directly affect the individual's ^{14}C activity. Studies of shark age, using the radiocarbon bomb peak (from atomic weapons testing), found that changes in diet could affect shark vertebrae ^{14}C activity (Campana et al., 2002). Sharks feeding on longer-lived species during the bomb peak could have a relatively lower ^{14}C activity because of integration with lower activities

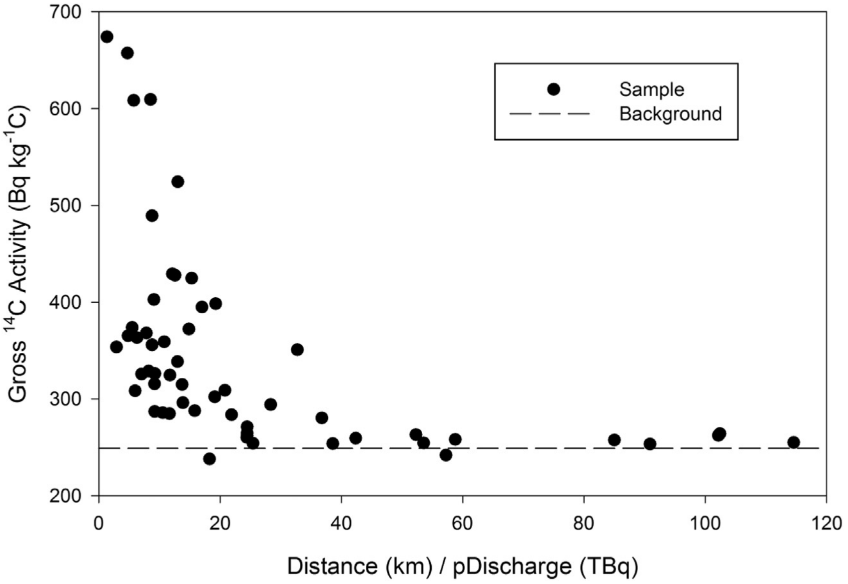

Fig. 5. Sample ^{14}C activity as a function of stranding distance from Sellafield, normalised by the discharge activity for the 24 months prior to individual stranding (note that no delay factor for West of Scotland samples is included here).

from before the bomb peak (Campana et al., 2002; Kerr et al., 2006). Integration of higher ^{14}C activities from Sellafield discharges have also been linked to higher activities in longer-lived species at specific sites in the Irish Sea (Muir et al., 2017). Of the two Porth Dafarch porpoise samples, the higher ^{14}C activity came from a juvenile porpoise, whereas the lower activity was from a neonate. The ^{14}C activity of the neonatal porpoise is likely a result of transfer from mother to calf. It could be inferred, therefore, that its mother had been foraging in an area of lower ambient activity relative to the sampled juvenile. It is equally possible that the mother of the neonatal porpoise had a lower ^{14}C activity due to a longer integration period covering a previous period of lower prey ^{14}C activity and this was transferred to the calf.

During data analysis, species type did not have an impact on sample ^{14}C activity. However, harbour seal and grey seal samples were only available from the West of Scotland where there was little observed variation in ^{14}C activities relative to Irish Sea mammals, and concurs with the relative homogeneity in ^{14}C activities of other marine species in this area (Tierney et al., 2017). In order to determine whether diet and life history influence mammal ^{14}C activities between species, it would be necessary to analyse seal samples from the Irish Sea.

Across all the data, several other variables including sex, age class (neonate, juvenile, sub-adult and adult) and level of decomposition (freshly dead, slight and moderate decomposition), showed no significance with ^{14}C activity. However, distance of stranding site from Sellafield and the Sellafield ^{14}C discharge activity prior to stranding (pDischarge) were significant. The best model fit predicted that for every 1 km increase in distance away from Sellafield there would be an estimated 0.3% decrease in mammal ^{14}C activity. This is significant as the samples analysed in this study came from mammals that stranded in the range of 10–1000 km from Sellafield. It indicates that stranding site is a reasonable approximation for the area an individual has been foraging in, as ^{14}C activities in the UK marine environment reduce with distance from Sellafield (Begg et al., 1992, Cook et al., 1998, Cook et al., 2004., Muir et al., 2015., Tierney et al., 2016, Muir et al., 2017., Tierney et al., 2017). Within the Irish Sea and, in particular, at stranding distances of $<$ 200 km from Sellafield, there was a wide range of ^{14}C activities (280–674 Bq kg^{-1}C). At distances $>$ 200 km a general tail of decreasing ^{14}C activity is apparent and a distinct reduction in maximum activity exists between Irish Sea and West of Scotland samples. Three samples from the Scottish east coast show slight enrichments above ^{14}C background activity (253–264 ± 1 Bq kg^{-1}C). This could indicate the long distance dispersion of ^{14}C from Sellafield to the North Sea, as has been noted before (Cook et al., 1998; Gulliver et al., 2004). The reduction in ^{14}C activity with distance from Sellafield is due to dilution and subsequently lower activities within prey species (Tierney et al., 2017) as discussed below.

The best model fit also predicted that for every 1 TBq increase in total Sellafield ^{14}C discharge activity for the period of 24 months prior to stranding, mammal ^{14}C activity would increase by an estimated 6.5%. This confirms that Sellafield is the source of ^{14}C enrichment in these samples. Furthermore, it also indicates the complex nature of ^{14}C transfer to these animals through the food web and shows the persistence of enriched ^{14}C within the marine environment, despite dispersion and dilution. Adding a delay factor of 12 months for West of Scotland samples improved the overall model fit. It suggests that the sampled West of Scotland mammals have spent little or no time foraging in the Irish Sea during the 12 months prior to stranding. This is expected of harbour seals as they typically only forage within 40 km of their haul-out site (Tierney et al., 2016). However, the number of harbour seal samples analysed was low (6) so this increased significance is unlikely to be attributable to these samples alone. In addition, few grey seal samples were analysed (3), therefore it is likely that the increased model significance is proportionally weighted toward the porpoise samples, which made up the bulk of the samples analysed (47). By removing the seal samples from the model fitting process, a similar level of significance for pDischarge ($p < 0.01$) was found. The model fit suggests that the sampled West of Scotland porpoises fed mainly in areas *other* than that of the Irish Sea for (at least) 12 months prior to stranding. This indicates a high foraging fidelity for harbour porpoises in the West of Scotland.

Herring, sandeel and gadoid species such as haddock (*Melanogrammus aeglefinus*) and whiting (*Merlangius merlangus*) are important prey for harbour porpoise (Santos et al., 2004; Hernandez-Milian, 2014). Herring activity (from a bulk sample) in the eastern Irish Sea in 2014 was reported at 274 ± 1 Bq kg^{-1}C, sandeel 314 ± 1 Bq kg^{-1}C and Irish Sea haddock ranged between 293 and 469 Bq kg^{-1}C (Muir et al., 2017). The average ^{14}C activity of the six Irish Sea porpoise samples from animals that stranded between 2014 and 2015 was 329 Bq kg^{-1}C (range 288–428 Bq kg^{-1}C) and agrees well with the ^{14}C activities of their prey species. This is expected as ^{14}C is transferred through the food web without any bioaccumulation or concentration effect. Measurements of West of Scotland fish demonstrated ^{14}C activity ranges of 282–284 Bq kg^{-1}C in herring, 286–296 Bq kg^{-1}C in haddock, and 288–413 Bq kg^{-1}C in whiting (Tierney et al., 2017). These activities fit reasonably well with the range of porpoise and seal activities between 2014 and 2015 porpoise

(254–398 Bq kg^{-1}C), after exclusion of the two individuals with below-background activities. The average mammal ^{14}C activity of these ten samples, 280 Bq kg^{-1}C, is at the lower end of the prey species activity range. However, the mammal samples come from a much wider area, including north of the fish sample sites, where ^{14}C activities in other benthic species are lower (Tierney et al., 2017). It is apparent that trophic transfer of enriched ^{14}C from prey species is the cause for enriched activities found in mammals. The significant relationship that exists between Sellafield ^{14}C discharges and mammal ^{14}C activity, and the comparable activities between predator and prey, demonstrate the transfer pathway in its entirety as a trophic level flow of ^{14}C from source to top marine predators.

5. Conclusions

Enriched ^{14}C activities were found in almost all marine mammal samples from the west coast of the British Isles. The highest activities were from harbour porpoises that stranded within the Irish Sea, although enriched ^{14}C activities were also observed in the West of Scotland and in three samples from the Scottish east coast. ^{14}C activities vary greatly both temporally and spatially. They correlate significantly with: 1) the distance the animal stranded from the Sellafield nuclear fuel reprocessing facility; and 2) the total ^{14}C activity discharged from Sellafield to the Irish Sea for a period of 24 months prior to stranding.

West of Scotland marine mammal ^{14}C activities correlate significantly with discharges made between 12 and 36 months prior to the animal stranding. This indicates the time taken for Sellafield ^{14}C discharges to be transported to the West of Scotland environment and become fully integrated in prey species. The model fit also suggests that West of Scotland harbour porpoises did not forage in the Irish Sea and have a high foraging fidelity to the West of Scotland.

^{14}C activities in samples from 2014 and 2015 are similar to ^{14}C activities measured in typical prey species showing that transfer of enriched ^{14}C from prey to predator occurs without any concentration or bioaccumulation effect. Although the ^{14}C activities presented in this study do not pose any radiological risk to the individual, it is clear that ^{14}C enrichment in marine mammals result from ^{14}C transfer from prey species, and that distance and discharge activity from Sellafield are key factors in determining an individual's muscle ^{14}C activity.

Sellafield is one of a number of facilities that continue to release low-level radioactive material, such as ^{14}C, into the marine environment. This study demonstrates, for the first time, the transfer of nuclear industry derived, ^{14}C through the entire marine food web to top predators, and highlights the necessity for continual monitoring of the fate of ^{14}C and other bioavailable radionuclides in marine ecosystems. Future work includes measuring ^{14}C concentrations in seals from the Irish Sea and addressing differences in ^{14}C transfer in relation to dietary preferences.

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