Ecosystem uptake and transfer of Sellafield-derived radiocarbon (14C).  
Part 1. The Irish Sea

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Ecosystem uptake and transfer processes of Sellafield-derived radiocarbon (14C) within the Irish Sea were examined. Highly variable activities in sediment, seawater and biota indicate complex 14C dispersal and uptake dynamics. All east basin biota exhibited 14C enrichments above ambient background while most west basin biota had 14C activities close to background, although four organisms including two slow-moving species were significantly enriched. The western Irish Sea gyre is a suggested pathway for transfer of 14C to the west basin and retention therein. Despite ongoing Sellafield 14C discharges, organic sediments near Sellafield were significantly less enriched than associated benthic organisms. Rapid scavenging of labile, 14C-enriched organic material by organisms and mixing to depth of 14C-enriched detritus arriving at the sediment/water interface are proposed mechanisms to explain this. All commercially important fish, crustaceans and molluscs showed 14C enrichments above background; however, the radiation dose from their consumption is extremely low and radiologically insignificant.

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1. Introduction

1.1. Sellafield

The Sellafield nuclear complex, situated on the Cumbrian coast of the north-east Irish Sea (Fig. 1), undertakes reactor decommissioning, fuel reprocessing and storage of nuclear materials, including radioactive wastes. During reprocessing operations, radioactive wastes are discharged to the sea and atmosphere, or disposed of as solid waste. During reprocessing operations, radioactive wastes are discharged to the sea and atmosphere, or disposed of as solid waste (Nuclear Decommissioning Authority, 2015). Aquatic radioactive discharges are made via pipelines extending 2.1 km into the north-east Irish Sea. Sellafield (then Windscale) commenced discharges of low-level radioactive waste, including 14C, to the north-east Irish Sea in 1952, with peak discharges for most radionuclides occurring in the early- to mid-1970s (Gray et al., 1995). Extensive research into the fate of discharges to the Irish Sea has demonstrated that an area of fine offshore sediment (known as the Sellafield mudpatch), close to the discharge point, has acted as an important sink for radioactive contaminants (Pentreath et al., 1984, Kershaw et al., 1992, Cook et al., 1997, Hunt, 1985, MacKenzie et al., 1987, 1999, MacKenzie, 2000). Since the 1970s, liquid radioactive discharges have fallen significantly with the introduction of several waste treatment plants that are efficient at removing cationic fission products e.g. 137Cs+ and actinides from waste effluent, but ineffective at removing most anionic species such as 14CO3− (Gulliver et al., 2001). One notable exception is the pertechnetate (99TcO4−) abatement technology, introduced in 2003, which is efficient at removing anionic 99Tc from the effluent stream (RIFE-9, 2004). Waste radiocarbon (14C) is produced during the nuclear fuel cycle from a range of neutron capture reactions, but predominantly from nitrogen impurities in the fuel and cladding involving the 14N (n, p) 14C reaction (Otlet et al., 1990, 1992). 14C is either stored in waste repositories or discharged, under authorisation, to the environment as low-level waste (LLW).

1.2. Sellafield-derived 14C

Due to the ineffectiveness of the waste treatment plants in removing most anionic species, the pattern of temporal variations in the quantities of 14C discharged to the Irish Sea has differed from most other radionuclides. Up to 1984, aquatic discharges of 14C from Sellafield (Fig. 2) were estimated and were subsequently measured between 1984 and 1993 at <2 Tera Becquerels (Tbq) per year, on average, until 1994 when 14C discharges increased by an order of magnitude (BNFL, 1985–1989, MAFF, 1992–1995). This increase was partly due to a change in discharge policy, diverting atmospheric 14C discharges to the aqueous route, and to a lesser extent to an increase in reprocessing

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activity as the thermal oxide reprocessing plant (THORP) began operations (BNFL, 2002). Between 1994 and 2014, Sellafield discharged a total of 166.9 TBq of $^{14}$C to the Irish Sea, with an average annual discharge of 7.9 TBq (MAFF, 1994, 1995, RIFE (1-20)1996–2015). Sellafield $^{14}$C marine discharges contribute 4.8% (2.8 μSv) to the current total dose (58 μSv) received by the critical group of consumers of marine fish and shellfish from the Cumbrian coast, which is small when compared with the average annual dose (2230 μSv) received by an individual in the UK from natural sources of radioactivity (Nuclear Decommissioning Authority, 2015). However, due to the long half-life of $^{14}$C (5730 y) and ready entry into the global carbon cycle, aquatic $^{14}$C discharges are the dominant contributor to collective doses (man Sievert (man Sv)) for UK (0.94 man Sv), European (3.1 man Sv) and World (32 man Sv) populations from total contributions of 1.2, 3.7 and 33 man Sv, respectively, resulting from the effects of all discharges from Sellafield in 2014 (Nuclear Decommissioning Authority, 2015). According to UNSCEAR (2008), liquid $^{14}$C discharges from global fuel reprocessing, between 1998 and 2002, contributed 94% of the total collective dose from all radionuclides. Furthermore, the largest total dose estimates from the nuclear industry remain associated with releases of $^{14}$C.

Sellafield $^{14}$C discharges are additional to an existing 'background' pool of $^{14}$C derived from natural production and atmospheric testing of atomic weapons, primarily during the 1950s and 1960s. This $^{14}$C 'background' level for north-east Atlantic biota and dissolved inorganic carbon (DIC) was measured in 1995 at 248 ± 1 Bq kg$^{-1}$ (Cook et al., 1998). Tierney et al. (2016a, 2016b) redefined this value in 2014, based on analyses of blue mussel (Mytilus edulis) shells collected on the west coast of Ireland, and obtained an almost identical background value of 249 ± 1 Bq kg$^{-1}$, which is the value used in this study.$^{14}$C is discharged to the Irish Sea primarily as dissolved inorganic carbon (DIC) (Begg et al., 1991, 1992, Begg, 1992 and Cook et al., 1995). It is rapidly incorporated into the general DIC component of seawater and is largely dispersed in solution from the Irish Sea through the North Channel by prevailing residual northerly currents (Gulliver et al., 2001). However, a small but poorly characterised component of the $^{14}$C is retained in Irish Sea biota and sediment (MacKenzie et al., 2004). Unlike most other radionuclides, $^{14}$C dispersion cannot be explained by a sorption coefficient ($K_d$). Soluble $^{14}$C in DIC is readily utilised through fixation of $\text{H}^{14}\text{CO}_3^-$ and $^{14}\text{CO}_3^{2-}$ by primary producing organisms (phytoplankton and macroalgae) during photosynthesis (Lalli and Parsons, 1993) and is thus a bioavailable 'contaminant'.

![Fig. 1. Map of the British Isles (a) and inset of the Irish Sea (b) indicating sampling sites.](image)

![Fig. 2. Sellafield atmospheric and marine $^{14}$C discharges (TBq) between 1968 and 2014. Data prior to 1985 are estimated (Cook et al., 2004). Subsequent measured data were taken from BNFL (1985–1989), MAFF (1992–1995) and RIFE (1996–2015) reports.](image)
which is transferred through the entire marine food chain and will be manifested as particulate organic carbon (POC). Most POC is produced autochthonously from in situ decay of phytoplankton produced in the euphotic zone, and is otherwise comprised of detritus, including faecal pellets, dead organisms and organic aggregates of various types. Thus, $^{14}$C is subsequently transferred from the DIC to the POC reservoir and to the dissolved organic carbon (DOC) reservoir from in situ processes including exudation by phytoplankton, secretion from zooplankton, after-death decay processes and from external terrestrially derived sources (Chester, 1990). Additionally, marine calcifying organisms, e.g. molluscs, foraminiferae and coccolithophorids efficiently utilise DIC directly from the water column during shell (or plate) formation (Chester, 1990, McConnaughey et al., 1997), providing an uptake pathway for inorganic $^{14}$C into the particulate inorganic carbon (PIC) reservoir (Cook et al., 2004, Muir et al., 2015, Tierney et al., 2016a, 2016b). Inorganic carbonate material is broken down after death of the organism, and is removed by settling from surface waters. Eventually, organic and inorganic particulate material will be deposited to some degree into offshore sediment.

Concern over the radiological importance of $^{14}$C has prompted several investigations into the behaviour and distribution of $^{14}$C in the Irish Sea environment (Begg et al., 1991, 1992, Begg, 1992, Cook et al., 1995, 1998, 2004, Wolstenholme et al., 1998, Wolstenholme, 1999, Gulliver et al., 2001, Gulliver, 2002). This research revealed $^{14}$C enrichment in the biogeochemical (carbon) fractions of seawater, sediments and biota. $^{14}$C activities in the biogeochemical fractions (post-1994 discharge policy change) were typically greater than pre-1994 levels (Begg et al., 1992, Cook et al., 1995, 2004). $^{14}$C exhibited conservative behaviour in the DIC component of seawater (Begg, 1992), with some evidence of transfer into the other biogeochemical fractions, although no systematic trend was observed between the DIC activity and that of the other fractions. At this time, incorporation of $^{14}$C was observed in north-east Irish Sea demersal fish and mussels, demonstrating systematic $^{14}$C enrichment relative to $^{14}$C activities in DIC and plankton (Begg et al., 1992, Cook et al., 1995). This anomaly was attributed to mussels and fish integrating $^{14}$C activities, associated with plankton and other organic detritus, over several bloom periods, possibly coinciding with a period of high $^{14}$C discharge (Cook et al., 1995). Significant $^{14}$C enrichments observed in the seawater DIC pool and biota were largely confined to the NE Irish Sea (Cook et al., 1998). Beyond the North Channel, $^{14}$C activities decreased with increasing distance from Sellafield. Differences in $^{14}$C activities between species were attributed to the variability in the kinetic response of biota to changing $^{14}$C discharges, and to the differences in the pools from which biota derived their carbon. These mechanisms have been confirmed by examination of species-specific feeding habits in relation to $^{14}$C activity trends and enhancements in intertidal biota (Cook et al., 2004). Similar trends were evident in shells of Irish Sea intertidal molluscs (Muir et al., 2015) and in molluscs at sites remote from Sellafield (Gulliver et al., 2001). This is consistent with the dispersal of conservative radionuclides (e.g. $^{137}$Cs and $^3$H) which are dominantly conservative radionuclides (e.g. $^{137}$Cs and $^3$H) which are dominantly

Since 1986, and up to the most recent publication for 2015, continued local enhancements in $^{14}$C activity have been observed in north-east Irish Sea organisms due to discharges from Sellafield (MAFF, 1987–1995, RIFE, 1996–2015).

As a consequence of the past and ongoing Sellafield marine $^{14}$C discharges, UK coastal waters present a unique opportunity to investigate the concentration, distribution and environmental behaviour of $^{14}$C. Accurate knowledge of these factors is critical when evaluating historical, contemporary and emergency releases of $^{14}$C to the marine environment in terms of human radiation exposure, via marine food web transfer, and is of fundamental importance for planning, management and regulation of nuclear facilities.

In consideration of Sellafield’s ongoing aqueous $^{14}$C discharges, the work presented here, over two-parts, examines the environmental behaviour and processes of $^{14}$C in two distinct marine ecosystems defined according to geographical location (relative to Sellafield), oceanographic conditions and processes, and the diversity of species found within each setting. Part 1 (this study) investigates processes in the Irish Sea; whilst sites remote from Sellafield, in the west coast of Scotland, are discussed in Part 2 (Tierney et al., 2016a, 2016b).

Specifically, the objectives of this study (Part 1. The Irish Sea) were: i) investigate the marine dispersion of $^{14}$C and aqueous phase and solid/particulate phase partitioning; ii) examine the transfer and extent of $^{14}$C incorporation into biotic (benthic, pelagic and planktonic) and abiotic ecosystem components (sediment/seawater $^{14}$C biogeochemical fractions); iii) explore evidence of trophic transfer of $^{14}$C by examining complex marine food webs and species ecology; and iv) provide an accurate dose rate assessment to critical groups from consumption of $^{14}$C in commercially important organisms. Ultimately, the work presented in this study will contribute to an ongoing novel modelling approach to map the ecological fate of Sellafield-derived $^{14}$C.

1.3. Study area

The Irish Sea (Fig. 1) is a semi-enclosed continental shelf sea measuring ca. 300 km (160 n.m.) in length and 75–195 km (40–105 n.m.) in width, decreasing to 30 km (16 n.m.) at the north end in the North Channel (Bowden, 1980). The total volume is estimated at 2430 km$^3$, with approximately 80% of this volume lying to the west of the Isle of Man (Dickson and Boeens, 1988). An open-ended north/south channel of between 80 and 275 m in depth is located from 5°W with shallower (30–50 m) embayments in the eastern Irish Sea. The north–east Irish Sea basin, the main region of interest in this study, is ca. 30 m in depth. The western channel (or basin) is open ended and connected via St. Georges Channel to the Celtic Sea and the Atlantic Ocean at its broad southern boundary. In the north, the Irish Sea is connected via the North Channel to the Clyde Sea and Malin Shelf, which in turn communicates with the Atlantic Ocean through a narrow section of channel of 20 km (11 n.m.) width (Bowden, 1980). Atlantic water enters the Irish Sea from both channels, forming a complex system where these currents interact, and where wind and density-driven circulation also play important roles (Dabrowski et al., 2012). The long-term circulation is predominately northwards, with inflow through St. George’s Channel and outflow via the North Channel (Ramster and Hill, 1969, Howarth, 1982, Gulliver, 2002, 2007). This is consistent with the dispersal patterns of conservative radionuclides (e.g. $^{137}$Cs and $^3$H) which are dominantly distributed according to the movement of major tides and residual currents (Baxter et al., 1979, McKay and Baxter, 1985, McDonald et al., 1990, Cook et al., 1997). However, the net northerly flow is seasonally highly variable and under certain wind-driven conditions is reversed southwards (Dabrowski et al., 2010). $^{14}$C enhancements observed in intertidal biota both to the north and south of the Sellafield discharge point support this assertion (Begg et al., 1991, 1992, Cook et al., 2004, Muir et al., 2015, Tierney et al., 2016a, 2016b), although it is unclear whether radionuclide transport is forced by long-term net circulation or short-term wind and tide-induced water movement. Dickson and
Boelens (1988) describe a residual southerly drift along the Cumbrian coast which eventually turns west and then north. However, the time-averaged circulation in the Irish Sea is relatively weak and shows no specific directionality over large areas (Nichols et al., 1993). Salinity distributions suggest that exchange between the eastern and western Irish Sea is limited although radionuclide distributions indicate some east to west transport (Leonard et al., 1997), probably to the north of the Isle of Man (Gowen and Stewart, 2005). Well-defined, thermal stratification occurs in summer in regions of weak currents, notably to the west of the Isle of Man where cold dense water left over from the previous winter is trapped in topographic depressions (Dabrowski and Hartnett, 2008). Thermal stratification and the transport and fate of radionuclides and other contaminants (Hill et al., 1996) and juvenile pelagic fish e.g. sole (Solea solea),plaice (Pleuronectes platessa), brill (Scophthalmus rhombus), turbot (Scophthalmus maximus) and gadoids e.g. cod (Gadus morhua), haddock (Melanogrammus aeglefinus) and whiting (Merlangius merlangus) (CEFAS, 2005).

2. Methodology

2.1. Sampling and analysis

Seawater biogeochemical carbon fractions (DIC, DOC, PIC, POC), sediment, benthic organisms and plankton were collected during extensive sampling surveys undertaken in the Irish Sea on-board the RV Prince Madog in June 2014 from the locations shown in Fig. 1. Sampling station details are presented in Table 1. Sampling was undertaken in the north-east Irish Sea east basin (station EB2) at ca. 28 m depth; and in the west basin (station WB) at ca. 133 m depth. Commercially important fish, molluscs and crustacean samples were collected from nine Irish Sea stations (Table 1) during fish stock surveys (cruse no. CO4114) conducted on-board the RV Corystes by the Agri-Food and Biosciences Institute, Northern Ireland (AFBI-NI) during October 2014 (Fig. 1). The on-board ship sampling procedure and laboratory treatment are described separately for each sample type below.

2.1.1. Seawater biogeochemical carbon fractions

Surface water samples (ca. 2 m depth) were obtained for stations EB2 and WB by pumping 160 l of seawater on-board with an electric pump into pre-washed 20 l containers for subsequent 14C analysis of DIC, DOC, PIC, POC fractions. Each container was rinsed with seawater prior to complete filling. Each 20 l sample was filtered immediately on-board using pre-furnaced (400 °C) 150 mm diameter GF/F (Whatman) glass fibre filters, within a positive pressure N2 filtration system. GF/F filters have the smallest available pore size (0.7 µm) suitable for 14C analysis (to avoid extraneous carbon contamination) and therefore define ‘dissolved’ and ‘particulate’ material in this study. Particulate material from 40 l of seawater was collected on each filter, wrapped in aluminium foil and stored at −20 °C prior to 14C analysis. Several 500 ml aliquots of filtrate were collected for DIC analysis in 1 l foil bags (FlexiFoil PLUS, SKC Inc, USA). Again, these were rinsed with seawater, re-filled and stored at −20 °C. Separate 500 ml aliquots of seawater were collected for DOC analysis. These were transferred into cleaned, pre-furnaced (500 °C) and pre-rinsed glass bottles which were then re-filled and acidified with (85%) orthophosphoric acid to

Table 1
<table>
<thead>
<tr>
<th>Sampling station</th>
<th>Date sampled</th>
<th>Station co-ordinates</th>
<th>Sample type(s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>EB2</td>
<td>10 June 2014</td>
<td>54 28.00 N, 03 44.00 W</td>
<td>Seawater, sediment, plankton, benthic organisms</td>
</tr>
<tr>
<td>WB</td>
<td>11 June 2014</td>
<td>54 13.00 N, 05 04.00 W</td>
<td>Seawater, sediment, plankton, benthic organisms</td>
</tr>
<tr>
<td>86</td>
<td>20 October 2014</td>
<td>54 35.68 N, 05 26.30 W</td>
<td>Fish survey</td>
</tr>
<tr>
<td>208</td>
<td>09 October 2014</td>
<td>53 48.84 N, 05 46.40 W</td>
<td>Fish survey</td>
</tr>
<tr>
<td>257</td>
<td>13 October 2014</td>
<td>54 24.44 N, 03 45.77 W</td>
<td>Fish survey</td>
</tr>
<tr>
<td>259</td>
<td>13 October 2014</td>
<td>54 16.83 N, 03 42.84 W</td>
<td>Fish survey</td>
</tr>
<tr>
<td>242</td>
<td>08 October 2014</td>
<td>54 06.04 N, 04 02.39 W</td>
<td>Fish survey</td>
</tr>
<tr>
<td>342</td>
<td>08 October 2014</td>
<td>53 55.32 N, 03 48.82 W</td>
<td>Fish survey</td>
</tr>
<tr>
<td>245</td>
<td>08 October 2014</td>
<td>53 30.12 N, 04 11.20 W</td>
<td>Fish survey</td>
</tr>
<tr>
<td>Area G</td>
<td>16 February 2015</td>
<td>54 20.44 N, 05 24.38 W</td>
<td>Scallop survey</td>
</tr>
<tr>
<td>Area H</td>
<td>16 February 2015</td>
<td>54 08.72 N, 05 44.17 W</td>
<td>Scallop survey</td>
</tr>
</tbody>
</table>
were weighed, oven-dried at 40 °C and reweighed to obtain a wet:dry ratio. Where samples were discarded to avoid the effects of smearing during extrusion. Samples were immediately extruded and sectioned on-board into 1 cm vertical pieces. Representative DOC concentrations were obtained for Irish Sea water samples by using high-temperature combustion of dried salts (and adhering dissolved organic matter) according to the method of Fry et al. (1996). Some modifications were made to the method and are briefly described as follows. For each station (EB2 and WB), pre-acidiﬁed (orthophosphoric acid) seawater aliquots (500 ml) were transferred to a 1.5 l pre-furnaced (600 °C) high purity quartz glass vessel. Water samples were evaporated to dryness under vacuum according to the method described in Burr et al. (2001). Using this method, samples are never exposed to the atmosphere. The DIC component is removed during drying and the resulting salt contained within the vessel is detached from the evaporation apparatus and transferred to a separate vacuum rig for combustion. The samples, including salts, were combusted using the sealed quartz tube combustion method to liberate CO2 (Section 2.1.5) (Vandeputte et al., 1996). The CO2 was again cryogenically puriﬁed for subsequent conversion to graphite and AMS analysis.

2.1.1.2. DIC. Seawater samples (500 ml) contained in foil bags were thawed in a refrigerator and transferred under vacuum to a reaction vessel. Complete acid hydrolysis of the seawater was performed for each sample by introducing two aliquots of ca. 10 ml (85%) orthophosphoric acid to the vessel. Helium gas was passed through the sample/acid mixture at positive pressure to evolve the CO2, according to the method of Bryant et al. (2013). CO2 was isolated and cryogenically puriﬁed using the regime described in Section 2.1.5 in preparation for graphitisation and AMS measurement.

2.1.1.3. DOC. Representative DOC concentrations were obtained for Irish Sea water samples by using high-temperature combustion of dried salts (and adhering dissolved organic matter) according to the method of Fry et al. (1996). Some modiﬁcations were made to the method and are described as follows. For each station (EB2 and WB), pre-acidiﬁed (orthophosphoric acid) seawater aliquots (500 ml) were transferred to a 1.5 l pre-furnaced (600 °C) high purity quartz glass vessel. Water samples were evaporated to dryness under vacuum according to the method described in Burr et al. (2001). Using this method, samples are never exposed to the atmosphere. The DIC component is removed during drying and the resulting salt contained within the vessel is detached from the evaporation apparatus and transferred to a separate vacuum rig for combustion. The samples, including salts, were combusted at 850 °C for ~2.5 h, where the sulphate contained in the salt provided the oxidant for the organic matter (Fry et al., 1996). Liberated gases were passed through a series of traps: ethanol/dry ice (−78 °C) to remove water vapour, pentane/liquid nitrogen (−130 °C) to remove SO2 and the remaining gases (including CO2) collected using liquid nitrogen (−196 °C). The sample gas, collected using liquid nitrogen, underwent a secondary (closed) combustion at 850 °C (>3 h) with 2 g MnO2 and 2 g CuO added to the vessel to remove traces of HCl and SO2 oxides of nitrogen and other contaminants. CO2 samples were cryogenically puriﬁed for subsequent conversion to graphite and AMS measurement.

2.1.2. Sediment

Several sediment cores were obtained from each sampling site using an OSIL Maxi-Core with 600 kg weight and 8 core boxes, with 600 mm length × 110 mm diameter polycarbonate cores for each. Sediment cores recovered at each deployment were ca. 30–40 cm in length and were immediately extruded and sectioned on-board into 1 cm vertical depth increments. Samples were labelled, bagged and frozen (−20 °C) pending analysis at SUERC.

Core sections were thawed at 3 °C and the outer (approx.) 1 mm was discarded to avoid the effects of smearing during extrusion. Samples were weighed, oven-dried at 40 °C and reweighed to obtain a wet: dry mass ratio and ﬁnally they were gently ground into a ﬁne powder using a mortar and pestle. Organic 14C analysis was undertaken on three samples from single cores selected from stations EB2 and WB, representing the sediment core surface, middle and base horizons. In each instance, the samples were acid-washed with 1 M HCl to remove the carbonate component, rinsed in deionised water, and oven-dried (40 °C) prior to sealed combustion at 850 °C (Vandeputte et al., 1996), CO2 puriﬁcation, graphitisation and AMS 14C analysis. Approximately 100–500 mg of sediment was combusted, depending on the organic content of each sample.

2.1.3. Marine biota

Benthic biota were sampled at each station (EB2 and WB) by two 15 min trawl deployments using a 2 m beam trawl, yielding a wide diversity of organisms at each site (Supplementary Table A.1). Additionally, several deployments were made using a Van Veen grab and Day grab to collect infaunal species. At each site, several specimens of each species were selected (where available) to enable multiple sample 14C analyses to be performed. Specimens were washed in seawater over a sieve, formally identiﬁed and immediately frozen, pending transport to the laboratory. Samples collected from ﬁsh stock surveys by AFBI-NI (supplementary Table A.2) were also immediately frozen after collection and transported to SUERC for 14C analyses.

Samples were thawed in a refrigerator overnight and washed thoroughly with high purity water to remove adhering sediment and debris. Muscle tissue or soft tissue was sub-sampled from each specimen for 14C analysis, to provide contemporary information on 13C uptake in biota relative to the carbon turnover time in each species. Bones and carapaces were not analysed. Tissue sub-samples were weighed and freeze-dried, then re-weighed for wet:dry ratio calculation. Where more than one individual of a species was collected, dried tissue samples were proportionally combined (with the same mass taken from each individual). Multiple samples were made where six or more individuals of a species were available. Small individuals e.g. polychaetes, or analogously similar species e.g. crabs or starﬁsh (inhabiting similar niches) were combined into their higher classiﬁcation groups. Approximately 10–15 mg of each sample was weighed from each species and combusted using the sealed quartz tube combustion method to liberate CO2.

2.1.4. Plankton samples

Plankton samples were collected from each station by deployment of plankton nets. Nets were deployed to the maximum depth at each station then hauled to the surface. Samples were rinsed into the sieve capsule and separated into fractions of ~270 µm mesh size for zooplankton and 80–270 µm mesh size for phytoplankton samples. Specimens were transferred into containers and frozen on-board at −20 °C.

Immediately prior to analysis, specimens were thawed at room temperature, thoroughly washed with deionised water, freeze dried and ca. 10–15 mg of each sample were weighed and combusted using the sealed quartz tube combustion method to produce CO2 for subsequent graphitisation and AMS measurement.

2.1.5. 14C analysis

For all samples collected, carbon dioxide was liberated either by sealed quartz tube combustion (for organic material) or by acid hydrolysis (for DIC and PIC), cryogenically puriﬁed under vacuum with dry ice-ethanol and liquid N2 traps, and 3 ml subsamples of CO2 converted to graphite according to the procedure of Sio et al. (1987). Sample 14C/13C isotope ratios were measured on the SUERC 250 kV SSAMS or the 5 MV tandem AMS (Freeman et al., 2008, 2010) and with quality assurance standards described in Naysmith et al. (2010) and Dunbar et al. (2016). Stable isotope (δ13C) ratios were measured oﬄine on a VG SIRA 11 isotope ratio mass spectrometer. 14C results were calculated relative to the international standard (oxalic acid II, SRM−4990C) as 14C activity ratios (fraction modern, fP/14C). Fraction modern results were converted to specific activities (Bq kg−1C) using the regime for calculating
enhanced activity samples described by Mook and van der Plicht (1999). Uncertainties are typically <0.5% of the measured activity for AMS, and have been omitted from figures for clarity.

3. Results and discussion

3.1. $^{14}$C in biogeochemical carbon fractions of seawater

The liquid $^{14}$C discharge for June 2014 (24.7 GBq) was an order of magnitude less than preceding monthly discharges for 2014 (January to May) which ranged from 201 to 828 GBq, and the lowest monthly $^{14}$C discharge since April 2001. Monthly discharges from January 2012 – December 2014, the period encompassing and immediately preceding this study, are given in Fig. 3.

$^{14}$C discharges from January 2012 onwards show large seasonal fluctuations, with peak discharges occurring during the winter months (November to February), coinciding with the periods of limited primary production in the Irish Sea, and reduced discharges in the spring and summer months (May – August). Nevertheless, the gross DIC activity ($546 \pm 2$ Bq kg$^{-1}$C) at station EB2, collected in June 2014, (Table 2) is more than twice the level of ambient background. However, it is difficult to ascertain whether this enhancement is contemporary with June 2014 discharges only, or contains a residual component of DIC from preceding higher monthly discharges retained in the water column. The $\delta^{13}$C values in the DIC (and PIC) fractions are within the range for marine inorganic carbon ($-1.0$ to $+1.0\%$ relative to VPDB), indicating that there is no significant terrestrial influence on these fractions during sampling. The $\delta^{13}$C value for the POC fraction ($-21.9\%$) is commensurate with $\delta^{13}$C values for Irish Sea suspended particulate organic matter of between $-18.0$ and $-22.0\%$ (MacKenzie et al., 2004), compared with terrestrially-derived organic matter of between $-24.0$ and $-32.0\%$. (Gulliver, 2002), and demonstrates that the POC at station EB2 is predominantly of marine origin. The gross activity in the POC fraction ($471 \pm 2$ Bq kg$^{-1}$C) is lower in activity than the DIC fraction but still enriched, confirming that POC is derived largely from the DIC reservoir. Whilst $^{14}$C enhancements observed in the DIC and POC fractions are only a ‘snapshot’ of $^{14}$C activities overlying station EB2, they provide convincing evidence for the mechanistic transfer from dissolved $^{14}$C to particulate organic material. The PIC fraction is slightly depleted at station EB2 ($244 \pm 1$ Bq kg$^{-1}$C). Transfer from DIC to PIC occurs mainly through uptake by molluscs and other calcareous organisms during shell formation. No calcareous foraminifera were observed on the PIC/POC filter papers when viewed under a microscope and as such, demonstrate their low prevalence in the eastern basin during sampling. Therefore, the main process is likely to be gradual erosion of larger shells in the intertidal zone as described in Cook et al. (2004), Muir et al. (2015) and Tierney et al. (2016a, 2016b). This process will occur slowly and will be small when compared to inputs from sedimentary/substrate sources of which old material represents a significant fraction of the carbonate in this system (MacKenzie et al., 2004). The $\delta^{13}$C value ($-30.3\%$) for the DOC sample denotes material derived from a terrestrial organic source, possibly originating from ($^{14}$C depleted) riverine runoff from the Cumbrian coast. Consequently, the $^{14}$C activity is significantly depleted in the DOC fraction at station EB2 (gross activity = $88 \pm 1$ Bq kg$^{-1}$C).

The DIC and PIC fractions at station WB are slightly enhanced and of comparable $^{14}$C activity, supporting the transfer mechanism of $^{14}$C from the DIC to the POC reservoir, predominantly through uptake by phytoplankton, but also by inputs from the non-living components of POC such as faecal matter and other organic detritus. $\delta^{13}$C values in DIC, PIC and POC fractions are consistent with material predominantly of marine origin. The $^{14}$C activity uniformity observed between the DIC and POC reservoirs indicates that the western Irish Sea is receiving relatively homogenous $^{14}$C inputs from the eastern Irish Sea, and would therefore be subject to the same transfer mechanisms discussed above. The PIC fraction at station WB is depleted in $^{14}$C, and similarly to PIC at station EB2, the transfer process from DIC to PIC will be slow and strongly influenced by the presence of a significant pool of old carbonate material. DOC is depleted at station WB, suggesting that the carbon is from an ‘old’ source of possibly re-cycled DOC material.

Table 2
\[
\begin{array}{cccc}
\text{Location} & \text{DIC} & \text{PIC} & \text{DOC} & \text{POC} \\
\hline
\text{Station EB2} & 546 \pm 2 & 244 \pm 1 & 88 \pm 1 & 471 \pm 2 \\
\text{Station WB} & 264 \pm 1 & 214 \pm 1 & 77 \pm 1 & 259 \pm 1 \\
\end{array}
\]

*na: insufficient carbon in sample for $\delta^{13}$C analysis.

![Fig. 3](https://example.com/fig3.png)

Fig. 3. Monthly liquid $^{14}$C discharges from Sellafield (TBq) since January 2012. (Sellafield Ltd., 2015).
Unfortunately there was insufficient carbon present in the sample to perform stable carbon isotope analysis to determine if this material was influenced by terrestrial runoff.

3.2. Sediment

$^{14}$C activities in the organic component of surface sediment (0–1 cm) for stations EB2 and WB are presented in Table 3. Station EB2 shows $^{14}$C enhancements at all depth horizons, indicative of Sellafield $^{14}$C inputs. $\delta^{13}$C values indicate that the organic material is largely of marine origin but with evidence of a small terrigenous-derived input distributed evenly throughout the core. The relative uniformity in $^{14}$C enhancement throughout the core and consistency of $\delta^{13}$C values, suggests that the sediment is subject to intense bioturbation and homogenisation throughout its complete depth, in agreement with the findings of other studies (e.g. Kershaw, 1986, Kershaw et al., 1983, 1984, 1999, MacKenzie and Scott, 1993, MacKenzie et al., 1998). The surface sediment is depleted in $^{14}$C relative to the POC in the overlying seawater, and is contrary to what might be expected in the north-east Irish Sea given the continuous nature of $^{14}$C inputs and transfer processes to the POC fraction. This activity disparity may have resulted from the loss of enriched surface material during core collection, although the Maxi-Corer was deployed to prevent such losses, and no significant re-suspension of material was seen during core recovery or during the core processing process. In the latter case, sectioning at 1 cm increments would effectively dilute enriched sediment with ‘older’ organic material if the enriched material is confined only to the uppermost sediment surface. It is reasonable to assume however, that the principal mechanisms for this activity difference arise from the attenuation of the $^{14}$C enriched POC arriving at the sediment surface, through rapid physical mixing, bioturbation and incorporation of POC into sediment dominated by older organic material. To explore this possibility, a single $^{14}$C analysis was undertaken on the less dense fraction of the surface sediment from station EB2 (0–1 cm), obtained from the bulk sediment using a settling method (Poppe et al., 2001), and assumed to contain organic particles (and clay minerals). A low temperature (~400 °C) combustion was then undertaken to minimise the contribution of ‘old’ clay-bound carbon (McGeethin et al., 2001). The $^{14}$C analysis revealed that this fraction had an activity of 413 Bq kg$^{-1}$C ($\delta^{13}$C = $-20.8$‰) which is significantly enriched compared to the bulk surface sediment reported in Table 3, and more comparable to the POC activity at station EB2 (249 ± 1 Bq kg$^{-1}$C) are denoted as ‘Depleted’.

The results from biota collected in June 2014 are presented in Fig. 4. Supplementary (Table A.1) details the number of individuals analysed, the average species size and size range (fish only), and the gross specific $^{14}$C activities (Bq kg$^{-1}$C) for each species. All benthic organisms show $^{14}$C enhancements above ambient background, indicative of a supply of enriched $^{14}$C to this site. Epifaunal or infaunal organisms feeding on or within sediment respectively, have higher $^{14}$C values than those feeding from the water column. Phytoplankton, zooplankton and suspension feeders e.g. the soft coral (Alcyonium digitatum) and dahlia anemone (Urticina felina) have the lowest $^{14}$C activities amongst the macrobenthos and have $^{14}$C activities broadly consistent with the seawater DIC and POC fractions at the time of sampling. Large inter-species variations are apparent at station EB2 and intra–species variation is also evident in dab (Limanda limanda) samples 1 and 2. $^{14}$C activity variations are discussed here in relation to species-specific ecology and feeding habits. For the purpose of this study, individual samples of infaunal or epifaunal invertebrates (crabs, starfish and polychaetes) were combined into their higher taxonomic groups and are described in these terms.

The gross phytoplankton $^{14}$C activity (520 Bq kg$^{-1}$C) is similar, at the time of sampling, to the gross DIC $^{14}$C activity (546 Bq kg$^{-1}$C) of sea-water. The gross $^{14}$C activity of the zooplankton (458 Bq kg$^{-1}$C) is similar to the POC activity at station EB2 (471 Bq kg$^{-1}$C), but lower than their principal food phytoplankton (520 Bq kg$^{-1}$C). Zooplankton samples at station EB2 were dominated by copepods which feed exclusively on phytoplankton. Nevertheless, zooplankton will be integrating carbon over a longer period of time than phytoplankton, which has a relatively fast carbon turnover rate and will readily ‘capture’ transient $^{14}$C enhancements in DIC from the water column. Seawater samples were collected from the surface (2 m depth) while plankton samples were collected from the whole water column, and any depth $^{14}$C activity stratification would influence overall plankton activities. The apparent activity disparity between zooplankton and phytoplankton underlines the spatial and temporal variability of $^{14}$C uptake in planktonic organisms.

Soft coral is a passive suspension feeder on both phytoplankton and zooplankton (Roushdy and Hansen, 1961, Fabricius et al., 1995) and although feeding preferentially on zooplankton, soft coral shows tropic opportunism in its feeding habits (Migne and Davout, 2002). The soft coral $^{14}$C activity (492 Bq kg$^{-1}$C) lies between the end members of its immediate food sources of zooplankton and phytoplankton at 458 and 520 Bq kg$^{-1}$C, respectively, and is in good agreement with the mean planktonic $^{14}$C activity of 489 Bq kg$^{-1}$C. The dahlia anemone (Urticina felina) is a carnivorous feeder, typically consuming small fish, crustaceans, molluscs, shrimps, and urchins. The $^{14}$C activity of this species (542 ± 3 Bq kg$^{-1}$) is more akin to that of planktonic species in the

Table 3

| Horizon depth (cm) | Station EB2 | | Station WB | |
|--------------------|-------------|-----------------------------|-----------------------------|
|                    | $^{14}$C activity (Bq kg$^{-1}$C) | $\delta^{13}$C (%) | $^{14}$C activity (Bq kg$^{-1}$C) | $\delta^{13}$C (%) |
| 0–1 (surface)      | 298 ± 1     | -23.0 | 170 ± 1 | -21.9 |
| 15–16              | 295 ± 1     | -23.2 | 152 ± 1 | -21.9 |
| 29–30 (station EB2 base) | 264 ± 1     | -23.0 | - | - |
| 34–35 (station WB base) | - | - | 155 ± 1 | -21.5 |

The values in bold indicate 'Depleted'.
east basin, indicating that it is deriving the majority of its food from a lower $^{14}$C activity source, quite possibly from planktonic crustaceans (Rasmussen, 1973) and to a lesser extent from other benthic biota. The comparable $^{14}$C activities of the soft coral and dahlia anemone with that of plankton suggest that these organisms are principally feeding from the water column. However, both these species are long-lived with the life-span of the dahlia anemone exceeding 50 years (Jackson and Hiscock, 2008) and the soft coral exceeding 20 years (Hartnoll, 1998), demonstrating that they will integrate $^{14}$C over a much longer period than comparably short-lived plankton.

A specific activity increase of $>285$ Bq kg$^{-1}$C is observed between the mean $^{14}$C activity of planktonic species and those organisms feeding from the water column to that of benthic organisms occupying higher trophic levels at station EB2, implying that these groups are reasonably distinct in their carbon sources. This also presents an activity paradox between the higher $^{14}$C activities observed in (non-suspension feeding) benthic organisms and the relatively low $^{14}$C activities observed in sediment, planktonic organisms and seawater biogeochemical fractions. The ecology of these benthic organisms is discussed in relation to species-specific $^{14}$C measurements.

Infraunal invertebrates, living either partly or wholly within sediment, are dominated at station EB2 by polychaete worms, which form the largest and most diverse community in soft subtidal sediment. Polychaetes may be carnivorous but may also consume algal matter. Many burrowing species or tube dwellers are deposit feeders that consume directly any organic particles from sand or mud, or extend tentacle-like structures to the sediment surface and convey detritus to the mouth (Lalli and Parsons, 1993). Crabs are predominantly omnivorous predators/ scavengers and deposit-feeders, whereas starfish prey upon a wide range of living organisms and carrion that include molluscs, polychaete worms and other echinoderms, small crustaceans, anemones and carrion, which may reflect their higher $^{14}$C activity. The spoon-worm (*Maxmuelleria lankesteri*) was analysed separately for $^{14}$C, as this species forms a major component of the burrowing megafauna of the north-east Irish Sea and is postulated to significantly affect the distribution of radionuclides in bottom sediments (Hughes et al., 1996a, 1996b, Kershaw et al., 1983, 1984, 1999). The spoon-worm is found at high densities in the Irish Sea (up to 35 m$^{-2}$) (Williams et al., 1981, Swift, 1993) and is a sedentary deposit feeder of sediment which it obtains by extending its proboscis from the burrow to graze the accessible sediment surface (Hughes et al., 1993, 1994). The $^{14}$C activity (953 Bq kg$^{-1}$C) infers that spoon-worms are selectively feeding on a $^{14}$C enhanced carbon source arriving at the surface sediment. Rapid scavenging (and bioturbation) caused by spoon-worms, polychaetes and other benthic detritivores of $^{14}$C enhanced organic material is, therefore, likely to be a significant reason for the depleted activities measured in organic sediments.

North-east Irish Sea plaice preferentially consume polychaetes (*Nephys* spp.) and bivalves (*Abra alba*) (Johnson et al., 2015). The $^{14}$C activity of plaice (673 Bq kg$^{-1}$C) is of the same order as crab, starfish and dab (sample 1), indicating that these species are consuming organisms with broadly similar $^{14}$C activities. In contrast to plaice, dab have a wide-ranging diet of larger and more energy-rich prey, primarily favouring crustaceans e.g. mud-shrimps (*Callianassa subterranea* and *Jaxea nocturna*) and angular crab (*Goneplax rhomboides*) (Johnson et al., 2015). This could explain their relative enhancement over plaice, although the former two prey species were not obtained at station EB2 and therefore, it was not possible to test this assertion. The highest $^{14}$C activity was recorded for dab (2) at 1012 Bq kg$^{-1}$C. It is unclear whether this enhancement is due to prey selection, one or more individuals feeding from an area that is highly enhanced in $^{14}$C or if individuals in the lower activity sample (dab sample 1) are feeding in a $^{14}$C depleted area. Age and size differences between individuals may influence both dietary preference and $^{14}$C activity, however, the relatively narrow size range observed between dab individuals (80–130 mm; average 110 mm) tends to preclude this argument. Whilst predator-prey interaction could explain some $^{14}$C enhancement observed in benthic species occupying higher trophic levels, it does not explain fully the increase noted between planktomic/suspension feeding organisms and higher trophic level organisms. The integration period over which an organism has consumed $^{14}$C-enriched material and the carbon turnover rate within each organism are factors that will influence the overall species activity. The lower $^{14}$C activity observed in plankton is due to their fast carbon turnover rate and, as such, is representative of the relatively low $^{14}$C discharges occurring from Sellafield immediately before or during the sampling period (June 2014). The higher $^{14}$C activities observed in higher organisms suggest that they are integrating $^{14}$C over a longer time period, coinciding with periods of higher $^{14}$C discharge.
3.4. 14C in north-west Irish Sea benthic and planktonic organisms: station WB

In contrast to the heterogeneity observed at station EB2, most benthic biota at station WB show small 14C enhancements above ambient background (Fig. 5). Supplementary Table A.1 details the number of individuals analysed for 14C and the gross specific 14C activities for each species. Phytoplankton and zooplankton have the lowest 14C activities at 242 and 254 Bq kg\(^{-1}\)C, respectively, comparable with DIC (264 Bq kg\(^{-1}\)C) and POC (259 Bq kg\(^{-1}\)C) activities at the time of sampling. For most organisms, similarly to station EB2, 14C activities exceed that of the surrounding sediment, planktonic organisms and seawater biogeochemical fractions. Dab, starfish (Asteroidea spp.), dragonet (Callionymus lyra) and the polychaete worm or ‘sea mouse’ (Aphroditida aculeata) are significantly enhanced in 14C above the other organisms collected at station WB. The degree of enhancement in these species is analogous to, or exceeds 14C activities in comparable organisms (e.g. dab, polychaete and starfish) at station EB2. 14C activities are discussed in relation to the ecology of each species, while those organisms with significant 14C enhancements are considered separately.

Phytoplankton is marginally depleted in 14C (242 Bq kg\(^{-1}\)C) in comparison to the marine ambient background activity. This may be caused by the presence of a small fraction of old 14C-depleted detrital material being retrieved from the water column during plankton sampling, effectively diluting the sample 14C activity. Zooplankton show slight 14C enrichment (254 Bq kg\(^{-1}\)C) reflecting recent time-integrated 14C activities of phytoplankton (and DIC) in the western Irish Sea, and suggesting that the 14C activity of (living) phytoplankton is probably enhanced. Mud-shrimp (Callianassa subterranea) are significantly enhanced in 14C above the other organisms collected at station WB. The dashed line indicates the measured background activity of this species.

Molluscs, and to a lesser extent polychaetes and echinoderms. The 14C activity of Nephrops (297 Bq kg\(^{-1}\)C) is comparable to that of C. macandreae and its other prey species. Similarly to station EB2, polychaete worms are proportionally higher in 14C activity (318 Bq kg\(^{-1}\)C) than other organisms. Nevertheless, the homogeneity of the system, in terms of 14C activity, makes interpretation of 14C uptake in most organisms difficult if based on feeding ecology.

Four species (including grouped starfish) show 14C enhancements at station WB ranging from ca. 2–4 times the ambient background level, which is unexpected given that station WB is approximately 130 km. (70 n.m.) from the Sellafield discharge outfall. Interpretation is also problematic as it encompasses several species and both highly mobile (dab and dragonet) and slower-moving organisms (sea mouse and starfish). Dab are migratory, moving from shallow inshore water to deeper offshore areas, on a seasonal basis, especially as juveniles (Ortega Salas, 1988, Rijnsdorp et al., 1992). Therefore, they could feasibly migrate and feed in an area with enhanced 14C activities. However, for less mobile species such as the sea mouse and starfish, the western basin gyre (Section 1.3) could provide a mechanism for transfer and retention of enriched 14C material at depth, facilitating a pathway for limited or species-specific 14C uptake in benthic organisms. This possibility requires greater investigation. It is equally justifiable to question why other benthic organisms collected at the same site show no such enhancement.

Overall, 14C activities in benthic organisms, both at station EB2 and WB, appear to be driven by a combination of mechanisms: i) the quantity, ‘bioavailability’ and 14C activity of organic matter supplied to sediments; ii) feeding behaviour: mobility, scavenging/feeding proficiency and selectivity for 14C-enriched organic material; iii) the assimilation and integration period for 14C enriched food, and carbon turnover rate of each species; and iv) trophic-level transfers of 14C through predator-prey interaction. Micro-, or even nano-scale processes could conceivably influence the transfer and uptake of 14C in organisms, but were beyond the scope of this study.

3.5. 14C in commercially important fish, molluscs and crustaceans

Results from samples obtained during fish and scallop stock surveys conducted by AFBI-NI are presented in Fig. 6. Additional information detailing the number of individuals analysed for 14C, the average species size and size range (fish only), and the gross specific 14C activities for
each species is given in Supplementary Table A.2. Fish species can have complex movement patterns as well as diverse feeding behaviours. Consequently, the $^{14}$C uptake mechanisms affecting these organisms will be driven by several factors including their proximity to, and time spent within feeding/spawning grounds enriched in Sellafield-derived $^{14}$C. Additionally, for migratory species such as the Atlantic herring (Clupea harengus) and Atlantic mackerel (Scomber scombrus), the location within and time spent transiting the Irish Sea will also be factors. $^{14}$C uptake and removal will be affected by feeding behaviour, food availability/source and subsequent transfer through the food chain via predator-prey interactions, as well as the carbon turnover rate of each species. $^{14}$C activities in other commercially important species e.g. dab and plaice have been discussed in context with the ‘ecosystem’ $^{14}$C activities observed at stations EB2 and WB (Sections 3.3 and 3.4).

All Irish Sea organisms collected during the fish and scallop stock surveys have $^{14}$C activities above the ambient background (Fig. 6). Generally, higher $^{14}$C activities can be observed in organisms from the eastern Irish Sea compared with those from the west, corresponding to their proximity to Sellafield. With the exception of the $^{14}$C enhancements noted in station WB benthic organisms, western Irish Sea organisms are relatively uniform in $^{14}$C activity in comparison to those found in the eastern Irish Sea, implying west basin organisms are feeding in a more homogeneous environment with respect to $^{14}$C activity than those foraging in the east. Organisms such as the King scallop (Area G and H – west basin) whose main food source is phytoplankton and POC, and Nephrops (station 208 – west basin) have elevated $^{14}$C activities compared with all fish species in the west although scallops were collected later in the year than the other organisms. However, their enhancement indicates that their time-integrated food source is enriched in $^{14}$C relative to that of fish, whose mobility allows foraging and feeding from relatively $^{14}$C depleted areas. None of the western Irish Sea samples showed $^{14}$C anomalous enrichment to the degree observed in station WB benthic organisms, from June 2014. Amongst the fish, cod (station 86 – west basin) and haddock (station 208 – west basin) have small $^{14}$C enhancements over mackerel and herring at station 208 – west basin, possibly from feeding exclusively within the Irish Sea. Also, cod and haddock consume a number of species including Nephrops (Howard, 1989); mud-shrimps including Calocaris macandreae (Buchanan, 1963) and spoon-worms (Rachor and Bartel, 1981), and exhibit comparable $^{14}$C activities to those species found at station WB.

Herring are facultative zooplanktivorous filter-feeders (Blaxter, 1990), feeding mainly on copepods (Holst et al., 2004); whilst mackerel feed on small fish and crustaceans, crustacean larvae and other zooplankton (Collette and Nauen, 1983). In addition, the migratory nature of these fish, and consequent consumption of food outside the confines of the Irish Sea, will reduce their overall tissue $^{14}$C activity and explain their near-background activity relative to enhanced activities found in other species at station 208.

A north to south decrease in $^{14}$C activity is apparent in the eastern Irish Sea. Nephrops (station 257 – east basin) have the highest activity (mean: 552 Bq kg$^{-1}$ $^{14}$C) corresponding to their close proximity to Sellafield (and to station EB2). Haddock were collected from three stations in the eastern Irish Sea (stations 259, 242 and 342 – east basin) and one from the west (station 208). $^{14}$C activities reduce with distance from Sellafield implying that their foraging/feeding behaviour is area-specific, at least in the immediate months preceding sampling. The low $^{14}$C activity in herring in the eastern Irish Sea is consistent with correspondingly low $^{14}$C activities amongst their planktonic food source and so are possibly feeding in areas remote from Sellafield. Sand eel (Ammodites tobianus) adults feed on zooplankton and some large diatoms (Bauchot, 1987) and their higher or comparable activity over other species at the east basin stations (242 and 342) may arise from more localised feeding as a result of a limited foraging range. Despite this, sand eel activity does not notably change between stations 242 and 342 (ca. 28 km (15 n.m.) apart) where the activity in haddock (a more mobile species) decreases. Mackerel samples 1 and 2 (station 242 – east basin) show intra-species variation (289 and 310 Bq kg$^{-1}$ $^{14}$C respectively). Given the small amount of data it is difficult to conclude if this is due to one or more individuals feeding in an area of high activity or conversely, from a $^{14}$C depleted area. The size range for mackerel was narrow (220–240 mm; mean 230) and argues against age (and size), and hence dietary preference, influencing the $^{14}$C activity differences in this species. However, the high mobility of mackerel, with a range extending beyond the Irish Sea may result in high $^{14}$C variability.

3.6. Dose from $^{14}$C to critical consumers of seafood from the Irish Sea

Dose rates (μSv) received by the critical consumers of seafood in the Irish Sea (Sellafield Fishing Community) are presented in Table 4 and,
for comparison, the dose rate received from natural/weapons testing 14C inputs is included. Dose rates and the net 14C activities were determined for the highest activities observed in commercially important species e.g. dab, plaice, haddock and Nephrops obtained for the NE Irish Sea. The average wet: dry weight ratios and percentage carbon content values for each species were used to convert 14C activities in plankton and POC are important factors in species e.g. dab, plaice, haddock and

<table>
<thead>
<tr>
<th>Sample type/ station</th>
<th>Consumption rate (kg)</th>
<th>Average wet: dry ratio</th>
<th>Average % carbon</th>
<th>Dose from Sellafield 14C (μSv)</th>
<th>Dose from natural/weapons testing 14C (μSv)</th>
<th>Total dose Sellafield and natural/weapons 14C (μSv)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dab (EB2)</td>
<td>45.8</td>
<td>4.8</td>
<td>39</td>
<td>1.67</td>
<td>0.54</td>
<td>2.21</td>
</tr>
<tr>
<td>Plaice (EB2)</td>
<td>45.8</td>
<td>4.4</td>
<td>42</td>
<td>1.07</td>
<td>0.63</td>
<td>1.69</td>
</tr>
<tr>
<td>Haddock (259)</td>
<td>45.8</td>
<td>4.6</td>
<td>43</td>
<td>0.54</td>
<td>0.62</td>
<td>1.16</td>
</tr>
<tr>
<td>Nephrops (257)</td>
<td>27.8</td>
<td>5.0</td>
<td>38</td>
<td>0.38</td>
<td>0.31</td>
<td>0.69</td>
</tr>
<tr>
<td>Total</td>
<td>(Dab + Nephrops)</td>
<td>45.8 + 27.8</td>
<td>–</td>
<td>2.05</td>
<td>0.85</td>
<td>2.90</td>
</tr>
<tr>
<td>(Plaice + Nephrops)</td>
<td>45.8 + 27.8</td>
<td>–</td>
<td>–</td>
<td>1.45</td>
<td>0.94</td>
<td>2.38</td>
</tr>
<tr>
<td>(Haddock + Nephrops)</td>
<td>45.8 + 27.8</td>
<td>–</td>
<td>–</td>
<td>0.92</td>
<td>0.93</td>
<td>1.85</td>
</tr>
</tbody>
</table>

Table 4

Dose rates (μSv) to the Sellafield Fishing Community critical consumer group of seafood from Sellafield-derived 14C discharges and from natural/weapons testing 14C. ‘Worst case’ total dose scenarios are presented for critical consumers of fish and crustaceans with the highest 14C activities observed in this study (dab, plaice, haddock and Nephrops).

4. Conclusions

Highly variable 14C activities across sediment, water and pelagic, demersal and benthic organisms indicate complex dispersal dynamics in the marine ecosystem of the Irish Sea, from initial discharge and transport as inorganic 14C to subsequent biological uptake and transfer throughout the marine food chain. 14C enhancements observed in the biogeochemical fractions of seawater and planktonic organisms substantiate a mechanistic transfer from the aqueous (dissolved) phase to the particulate phase, via DIC → plankton → POC, and that the constancy of supply 14C activity of plankton and POC are important factors in the transposition of 14C to higher organisms.

In terms of 14C incorporation into organisms, planktivorous species and those organisms predominantly feeding from the water column have markedly lower activities than benthic organisms occupying higher trophic levels at the east basin sampling station (EB2). Organism-specific 14C uptake and transfer, dictated by feeding behaviour, the carbon integration period and turnover rate, as well as ensuing trophic-level transfer through predator–prey interactions are key concepts in this activity difference.

Notably, organic sediments, in which benthos live and feed located near to Sellafield show only modest 14C enhancements over the total depth analysed (30 cm), and less than that of the benthic organisms found in surface sediments. It is proposed that the apparent ‘loss’ of enriched organic matter principally occurs through intensive physical mixing and bioturbation to depth of 14C enriched organic material arriving at the sediment/water interface, resulting in dilution with older organic material. Further build-up of 14C in sediment would be limited through benthic organisms rapidly scavenging the more labile 14C enriched organic material from surface sediments, and oxidative loss from the water column.

Most biotic and abiotic components of the ecosystem at station EB2 exhibit 14C uptake and enrichment above ambient background. Whilst the degree of 14C enrichment appears to be controlled by proximity to Sellafield, it is not exclusive to the eastern Irish Sea. Significant 14C activities were observed in several western basin organisms, equal to, or in excess of 14C activities observed in comparable east basin organisms. The western gyre is suggested as a possible mechanism for the 14C transfer, retention and uptake in these organisms, but this remains, as yet, unconfirmed.

All Irish Sea pelagic and demersal fish have 14C enhancements above background. Distinctions in 14C activities in fish species can be made between those feeding in the eastern Irish Sea from those sampled in the west. Area-specific foraging/feeding behaviour can be seen in some east basin species e.g. in haddock. The lowest 14C activities are observed in planktivorous and migratory species.

This study demonstrates the pervasive nature of 14C throughout the Irish Sea, coinciding with continuing nuclear re-processing activities and 14C discharges from Sellafield. Nevertheless, it is important to restate that current Sellafield 14C discharges contribute only a small dose to critical consumers of seafood from the Cumbrian coast. Dose rates presented here are comparable to those reported by Sellafield for 2014, and are negligible when compared with the annual UK dose limit of 1000 μSv to members of the public from all man-made sources of radiation (other than medical exposure); and the average annual (UK) dose received by an individual from natural sources of radioactivity (2230 μSv).

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