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

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ABSTRACT

Ecosystem uptake and transfer processes of Sellafield-derived radiocarbon (^{14}C) within the Irish Sea were examined. Highly variable activities in sediment, seawater and biota indicate complex ^{14}C dispersal and uptake dynamics. All east basin biota exhibited ^{14}C enrichments above ambient background while most west basin biota had ^{14}C 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 ^{14}C to the west basin and retention therein. Despite ongoing Sellafield ^{14}C discharges, organic sediments near Sellafield were significantly less enriched than associated benthic organisms. Rapid scavenging of labile, ^{14}C-enriched organic material by organisms and mixing to depth of ^{14}C-enriched detritus arriving at the sediment/water interface are proposed mechanisms to explain this. All commercially important fish, crustaceans and molluscs showed ^{14}C 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 (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 ^{14}C, 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. ^{137}Cs$^+$ and actinides from waste effluent, but ineffective at removing most anionic species such as ^{14}CO$_3^{2-}$ (Gulliver et al., 2001). One notable exception is the pertechnetate (^{99}TcO$_4^-$) abatement technology, introduced in 2003, which is efficient at removing anionic ^{99}Tc from the effluent stream (RIFE-9, 2004). Waste radiocarbon (^{14}C) 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 ^{14}N (n, p) ^{14}C reaction (Otlet et al., 1990, 1992). ^{14}C is either stored in waste repositories or discharged, under authorisation, to the environment as low-level waste (LLW).

1.2. Sellafield-derived ^{14}C

Due to the ineffectiveness of the waste treatment plants in removing most anionic species, the pattern of temporal variations in the quantities of ^{14}C discharged to the Irish Sea has differed from most other radionuclides. Up to 1984, aquatic discharges of ^{14}C 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 ^{14}C 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 ^{14}C discharges to the aqueous route, and to a lesser extent to an increase in reprocessing

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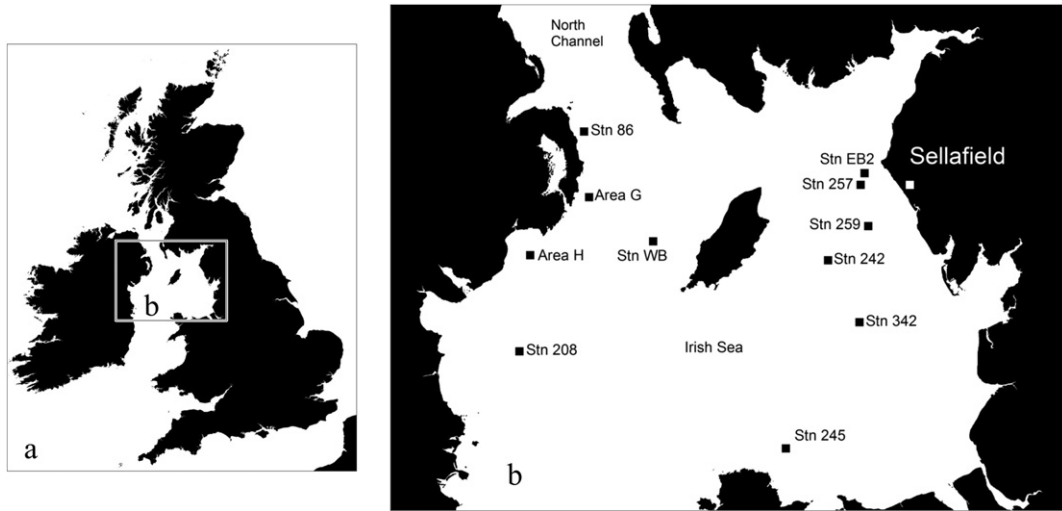*E-mail address:* graham.muir@glasgow.ac.uk (G.K.P. Muir).

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

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 H^{14}CO$_3^-$ and ^{14}CO$_3^{2-}$ by primary producing organisms (phytoplankton and macroalgae) during photosynthesis (Lalli and Parsons, 1993) and is thus a bioavailable 'contaminant'
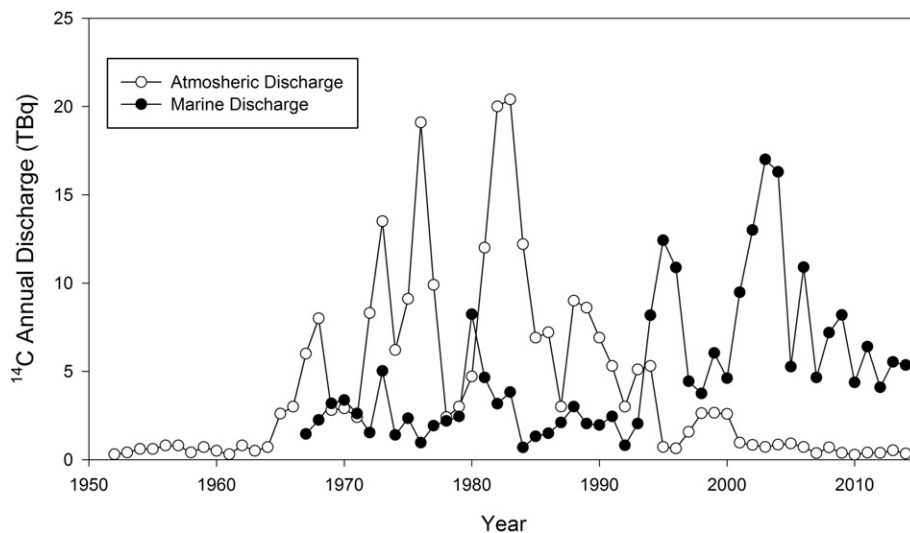

Fig. 2. Sellafield atmospheric and marine ^{14}C discharges (TBq) between 1968 and 2014. Data prior to 1985 are estimated (Cook et al., 2004). Subsequent measured data were taken from BNFL (1985–1989), MAFF (1992–1995) and RIFE (1996–2015) reports.

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, foraminiferans 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 dominated by solution transport of [14]C (Tierney et al., 2016a, 2016b). These studies demonstrate a net overall increase in ^{14}C in the inorganic component of north-east Irish Sea intertidal sediments. Nevertheless, ^{14}C activities in offshore sedimentary carbonates remain low (Gulliver et al., 2001, Gulliver, 2002, Wolstenholme et al., 1998, 1999), in comparison to the organic component of sediment, which has specific ^{14}C activities approximately twice that of the ambient background level (Gulliver, 2002). This provides unambiguous evidence of systematic Sellafield ^{14}C contamination in the Irish Sea (MacKenzie et al., 2004).

Routine monitoring of specific activities of marine organisms close to the discharge pipeline are reported annually by Sellafield (BNFL, 1971-2004, BNGSL, 2005, 2006, Nuclear Decommissioning Authority (Sellafield Ltd.), 2007–2015), the Ministry of Agriculture Fisheries and Food (MAFF, 1967–1995) and then a later partnership of the Food Standards Agency, Food Standards Scotland, the Environment Agency, the Scottish Environmental Protection Agency, Natural Resource Wales and the Northern Ireland Environment Agency (RIFE, 1996-2015).

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 km³, with approximately 80% of this volume lying to the west of the Isle of Man (Dickson and Boelens, 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 predominantly northwards, with inflow through St. George's Channel and outflow via the North Channel (Ramster and Hill, 1969, Howarth, 1982, Gulliver et al., 2001). 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 et al., 2010). As a result of thermal stratification, a density-driven cyclonic gyre forms in deep water (>100 m) in the west basin (Hill et al., 1994) and may have significant implications for the retention time of planktonic larvae (Hill et al., 1996) and juvenile pelagic fish (Dickey-Collas et al., 1997), and the transport and fate of radionuclides and other contaminants (Horsburgh et al., 2000, Dabrowski and Hartnett, 2008). Thermal stratification also occurs in the shallow north-eastern basin but is transient and readily overturned by tidal mixing and storms.

Irish Sea surface sediments are dominated by glacial and postglacial material (Pantin, 1977). Muddy sediments occur in areas of low tidal energy and are found off the Cumbrian coast and extensively in the western Irish Sea. Coarser-grained sediments are found to the north and south of the Isle of Man (Kershaw et al., 1999). Muddy north-east Irish Sea subtidal sediments are subject to extensive mixing (MacKenzie et al., 1987, MacKenzie et al., 1998, 2004, Kershaw et al., 1983, 1984) with very low (max. 0.08 cm yr^{-1}) sedimentation rates in this region (Kershaw, 1986, Kershaw et al., 1988). Benthic organisms are responsible for the intensive mixing (bioturbation) observed in the upper 5–10 cm of these sediments (Kershaw et al., 1999) and for the subsequent burial of particle-reactive radionuclides that are strongly associated with fine-grained sediment (MacKenzie et al., 1999). Bioturbation by the echiuran spoon-worm (*Maxmuelleria lankesteri*), for example, has resulted in the redistribution of Sellafield-derived radionuclides to a depth of 140 cm (Kershaw et al., 1983, 1984). Net deposition of fine sediment has been observed in the western basin (Condren et al., 1995) and bioturbation, by *Nephrops norvegicus* (Norway lobster; henceforth referred to as *Nephrops*) in particular and by fishing, are influential physical processes in this area (Kershaw et al., 1999). The diverse habitats of the Irish Sea support many important fishery species including shellfish e.g. *Nephrops*, king scallop (*Pecten maximus*) and queen scallop (*Chlamys operculari*), flatfish 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 northeast 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 (cruise 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 ^{14}C analysis of DIC, DOC, PIC and, 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 N_2 filtration system. GF/F filters have the smallest available pore size (0.7 µm) suitable for ^{14}C 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 ^{14}C analysis. Several 500 ml aliquots of filtrate were collected for DIC analysis in 1 l foil bags (FlexFoil 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
Sampling station details for the Irish Sea.

Sampling station	Date sampled	Station co-ordinates	Sample type(s)
EB2	10 June 2014	54 28.00 N, 03 44.00 W	Seawater, sediment, plankton, benthic organisms
WB	11 June 2014	54 13.00 N, 05 04.00 W	Seawater, sediment, plankton, benthic organisms
86	20 October 2014	54 35.68 N, 05 26.30 W	Fish survey
208	09 October 2014	53 48.84 N, 05 46.40 W	Fish survey
257	13 October 2014	54 24.44 N, 03 45.77 W	Fish survey
259	13 October 2014	54 16.83 N, 03 42.84 W	Fish survey
242	08 October 2014	54 06.04 N, 04 02.39 W	Fish survey
342	08 October 2014	53 55.32 N, 03 49.82 W	Fish survey
245	08 October 2014	53 30.12 N, 04 11.20 W	Fish survey
Area G	16 February 2015	54 20.44 N, 05 24.38 W	Scallop survey
Area H	16 February 2015	54 08.72 N, 05 44.17 W	Scallop survey

liberate DIC from the sample (Burr et al., 2001), thus halting photosynthetic processes and fixing the organic carbon content. Bottles were refrigerated at 3 °C on-board. All analyses of the biogeochemical fractions were undertaken at the SUERC ^{14}C laboratory.

2.1.1.1. PIC/POC. Filter papers containing particulate material from either 40 or 80 l of seawater (depending on the PIC concentration of the seawater) were placed within a reaction vessel under vacuum. Carbon dioxide (CO_2) was generated by acid hydrolysis of PIC material held on the filters using 50 ml of 1 M HCl and subsequently cryogenically trapped and purified prior to ^{14}C analysis. Acidified GF/F filters were retained for ^{14}C analysis of POC material. These were thoroughly washed over a clean GF/F filter paper using ultra-pure water (using vacuum filtration) to remove any traces of acid and subsequently oven dried at 40 °C. Both filters were transferred into quartz tubes for subsequent sealed-tube combustions to liberate CO_2 (Section 2.1.5) (Vandeputte et al., 1996). The CO_2 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 CO_2, according to the method of Bryant et al. (2013). CO_2 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 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 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 SO_2 and the remaining gases (including CO_2) 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 MnO_2 and 2 g CuO added to the vessel to remove traces of HCl and SO_2, oxides of nitrogen and other contaminants. CO_2 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-Corer 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 ^{14}C 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), CO_2 purification, graphitisation and AMS ^{14}C 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 ^{14}C 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 ^{14}C 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 ^{14}C analysis, to provide contemporary information on ^{14}C 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 CO_2.

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 CO_2 for subsequent graphitisation and AMS measurement.

2.1.5. ^{14}C 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 N_2 traps, and 3 ml subsamples of CO_2 converted to graphite according to the procedure of Slota et al. (1987). Sample ^{14}C/^{13}C 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 (δ^{13}C) ratios were measured offline on a VG SIRA 11 isotope ratio mass spectrometer. ^{14}C results were calculated relative to the international standard (oxalic acid II, SRM—4990C) as ^{14}C activity ratios (fraction modern, F^{14}C). Fraction modern results were converted to specific activities (Bq kg^{-1}C) 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 ± 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 ± 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 ± 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 foraminiferans were observed on the PIC/POC filter papers when viewed under a microscope and as such,
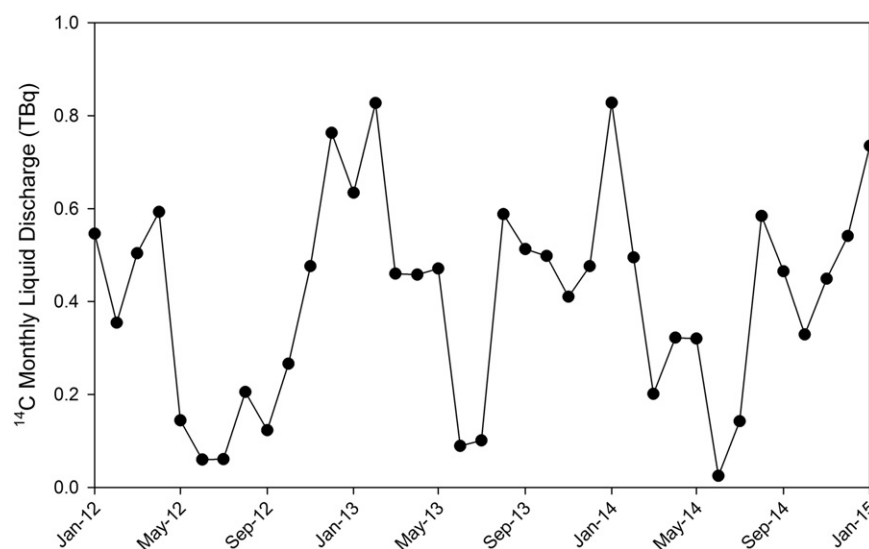
Table 2

Gross and net specific ^{14}C activities (Bq kg^{-1}C $\pm 1\sigma$) and $\delta^{13}C$ (‰ relative to VPDB) values from the four ^{14}C biogeochemical fractions of Irish Sea water samples. Net activities above the ambient background of 249 ± 1 Bq kg^{-1}C are in bold. Samples with values lower than this are marked as 'Depleted'.

Location	^{14}C specific activities (Bq kg^{-1}C $\pm 1\sigma$) $\delta^{13}C$ (‰)			
	DIC	PIC	DOC	POC
Station EB2	546 ± 2	244 ± 1	88 ± 1	471 ± 2
	297 ± 2	**Depleted**	**Depleted**	**222 ± 2**
	(-0.7‰)	($+1.0$‰)	(-30.3‰)	(-21.9‰)
Station WB	264 ± 1	214 ± 1	77 ± 1	259 ± 1
	15 ± 1	**Depleted**	**Depleted**	**10 ± 1**
	($+1.0$‰)	($+1.1$‰)	(na)*	(-20.0‰)

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

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 ± 1 Bq kg^{-1}C).

The DIC and POC 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.

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. δ^{13}C values indicate that the organic material is largely of marine origin but with evidence of a small terrestrially-derived input distributed evenly throughout the core. The relative uniformity in ^{14}C enhancement throughout the core and consistency of δ^{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 sectioning 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 (McGeehin et al., 2001). The ^{14}C analysis revealed that this fraction had an activity of 413 Bq kg^{-1}C (δ^{13}C = −20.8) which is significantly enriched compared to the bulk surface sediment reported in Table 3, and more comparable to the POC fraction (471 Bq kg^{-1}C) at station EB2, demonstrating that the mixing processes at the sediment surface can account, at least in part, for the anomalously low sediment activities relative to the biota. This has implications both for biotic uptake of ^{14}C, depending on the degree of degradation of organic matter in this fraction, and remobilisation processes from sediments and is

the subject of ongoing research. Further build-up of enriched POC in sediments may be prevented by benthic organisms rapidly scavenging labile (^{14}C enhanced) organic matter prior to, or immediately after incorporation into sediments or from oxidative loss (and/or tidal dispersal) of organic material to the water column.

Station WB is depleted in ^{14}C, relative to ambient background, and does not show any evidence of a Sellafield-derived ^{14}C contribution to the sediment. Kershaw et al. (1999) noted that sediments in the west basin are subject to intense physical mixing processes, either by bioturbation and/or fishing and the contribution of Sellafield-derived ^{14}C may be insufficient to cause enrichment of surface sediments. The uniformity in ^{14}C activities and the δ^{13}C profile at station WB could suggest that ^{14}C enhanced material might be rapidly consumed and/or re-worked and homogenised throughout the sediment. δ^{13}C values at station WB are higher than at station EB2, indicating a higher contribution of marine organic material at station WB compared to that of station EB2.

3.3. ^{14}C in north-east Irish Sea benthic and planktonic organisms: station EB2
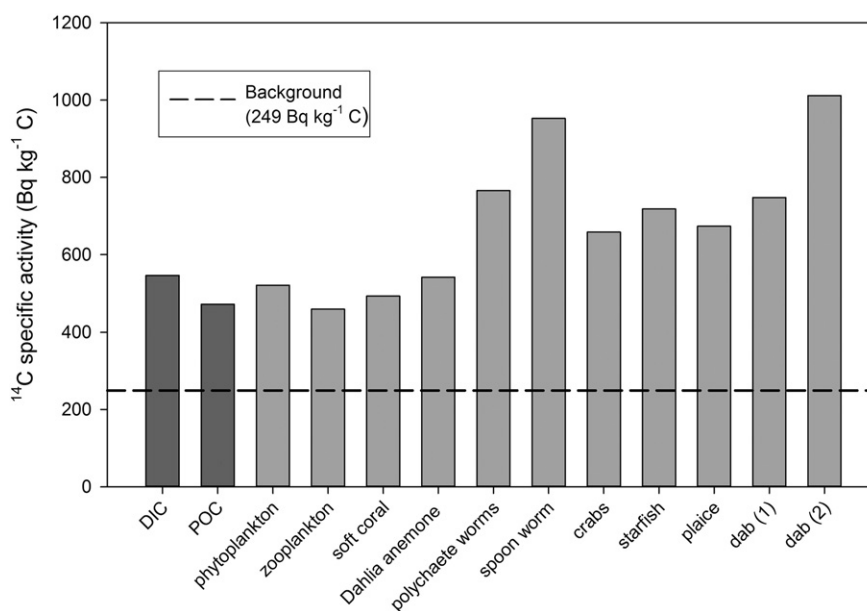
The results from biota collected in June 2014 are presented in Fig. 4. Supplementary (Table A.1) details the number of individuals analysed, the average species size and size range (fish only), and the gross specific ^{14}C activities (Bq kg^{-1}C) for each species. All benthic organisms show ^{14}C 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 seawater. 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}). 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 trophic opportunism in its feeding habits (Migne and Davoult, 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
Sediment organic fraction gross and net ^{14}C activities (Bq kg^{-1}C ± 1σ) and δ^{13}C values (‰ relative to VPDB) in surface, middle and base horizons for stations EB2 and WB. Net activities are in bold. Values lower than ambient background (249 ± 1 Bq kg^{-1}C) are denoted as 'Depleted'.

Horizon depth (cm)	Sediment Core			
	Station EB2		Station WB	
	^{14}C activity (Bq kg^{-1}C)	δ^{13}C (‰)	^{14}C activity (Bq kg^{-1}C)	δ^{13}C (‰)
0–1 (surface)	298 ± 1 **49 ± 1**	−23.0	170 ± 1 **Depleted**	−21.9
15–16	295 ± 1 **46 ± 1**	−23.2	152 ± 1 **Depleted**	−21.9
29–30 (station EB2 base)	264 ± 1 **15 ± 1**	−23.0	- -	- -
34–35 (station WB base)	- -	-	155 ± 1 **Depleted**	−21.5

Fig. 4. Gross specific ^{14}C activities (Bq kg^{-1}C) in benthic organisms and DIC/POC seawater biogeochemical fractions collected at station EB2. The dashed line indicates the measured background activity of 249 Bq kg^{-1}C measured in *Mytilus edulis* (blue mussel) shells obtained from the West Coast of Ireland.

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.

Infaunal 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 predator/ 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 detrivores 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 (*Nephtys* 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 planktonic/ 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. [14]C 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 ^{14}C enhancements above ambient background (Fig. 5). Supplementary Table A.1 details the number of individuals analysed for ^{14}C and the gross specific ^{14}C activities for each species. Phytoplankton and zooplankton have the lowest ^{14}C 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, ^{14}C 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' (*Aphrodita aculeata*) are significantly enhanced in ^{14}C above the other organisms collected at station WB. The degree of enhancement in these species is analogous to, or exceeds ^{14}C activities in comparable organisms (e.g. dab, polychaete and starfish) at station EB2. [14]C activities are discussed in relation to the ecology of each species, while those organisms with significant ^{14}C enhancements are considered separately.

Phytoplankton is marginally depleted in [14]C (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 ^{14}C–depleted detrital material being retrieved from the water column during plankton sampling, effectively diluting the sample ^{14}C activity. Zooplankton show slight ^{14}C enrichment (254 Bq kg^{-1}C) reflecting recent time-integrated ^{14}C activities of phytoplankton (and DIC) in the western Irish Sea, and suggesting that the ^{14}C activity of (living) phytoplankton is probably enhanced. Mud-shrimp (*Callianassa subterranea*) have the lowest ^{14}C activity amongst all benthic organisms (mean 268 Bq kg^{-1}C). Mud-shrimp is a sub-surface deposit feeder that can supplement its diet from suspension feeding (Nickell and Atkinson, 1995). Suspension feeders at station EB2 also had notably lower ^{14}C activities, and could explain the limited ^{14}C enhancement in mud-shrimp. In comparison, a second species of mud-shrimp (*Calocaris macandreae*) has a mean ^{14}C activity of 304 Bq kg^{-1}C. *Calocaris macandreae* has several feeding strategies depending upon food availability including filter-feeding, scavenging and predation (Calderon-Perez, 1981), and these would influence the overall ^{14}C activity of this species. *Nephrops* is an opportunistic predator feeding on crustaceans, including *C. macandreae* (Smith, 1988),

molluscs, and to a lesser extent polychaetes and echinoderms. The ^{14}C 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 ^{14}C activity (318 Bq kg^{-1}C) than other organisms. Nevertheless, the homogeneity of the system, in terms of ^{14}C activity, makes interpretation of ^{14}C uptake in most organisms difficult if based on feeding ecology.

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

3.5. [14]C 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 ^{14}C, the average species size and size range (fish only), and the gross specific ^{14}C activities for

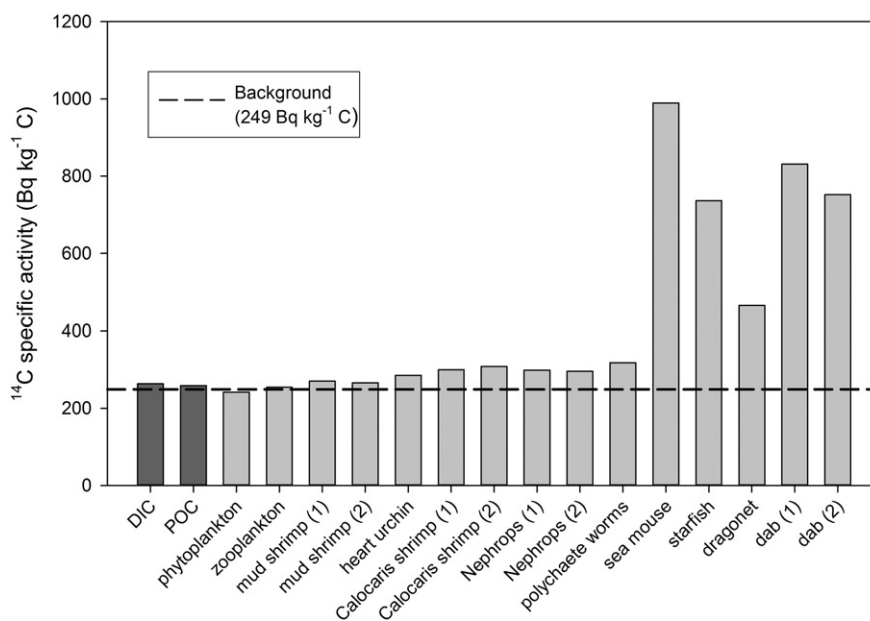**Fig. 5.** Gross specific ^{14}C activities (Bq kg^{-1}C) in benthic biota and DIC/POC seawater biogeochemical fractions, collected at station WB. The dashed line indicates the measured background activity of 249 Bq kg^{-1}C, measured in *Mytilus edulis* (blue mussel) shells obtained from the West Coast of Ireland.

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 foraging 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}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 (*Ammodytes 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}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,

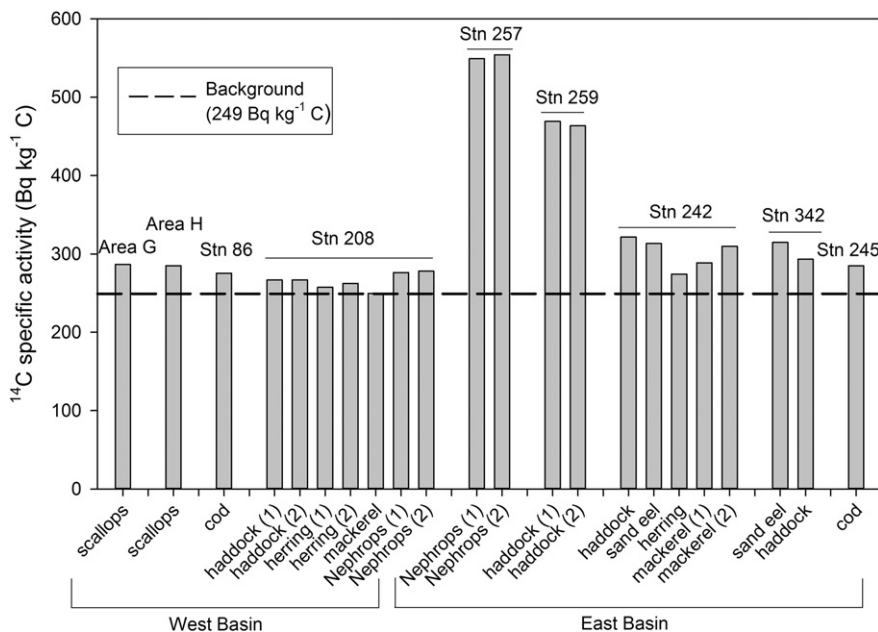**Fig. 6.** Gross specific ^{14}C activities (Bq kg^{-1}C) in commercially important Irish Sea fish, molluscs and crustaceans obtained during the fish and scallop stock surveys (conducted by AFBI-NI.). The dashed line indicates the measured background activity of 249 Bq kg^{-1}C, measured in *Mytilus edulis* (blue mussel) shells obtained from the West Coast of Ireland.

Table 4

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

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

for comparison, the dose rate received from natural/ weapons testing ^{14}C inputs is included. Dose rates and the net ^{14}C 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 ^{14}C activities from Bq kg^{-1}C to Bq kg^{-1}C fresh (wet) weight. The critical consumer group (Sellafield Fishing Community) 5-year average consumption rates (kg y^{-1}) were obtained from the CEFAS radiological habits survey (Garrod et al., 2015) of 14.8 kg y^{-1} cod, 31 kg y^{-1} other fish; 8.9 kg y^{-1} crabs, 6.9 kg y^{-1} lobsters; 12 kg y^{-1} other crustaceans; 7.4 kg y^{-1} winkles, 6.4 kg y^{-1} other molluscs. The dose per unit intake by ingestion of [14]C (5.8×10^{-10} Sv Bq^{-1}) was taken from ICRP–72 (ICRP, 1996). Most critical group species were not available from the north-east Irish Sea during sampling; therefore, dose calculations were based on 'worst-case' scenarios for consumption of commercial species with the highest observed ^{14}C activities in this study, e.g. dab, plaice, haddock and *Nephrops*. Total fish consumption (i.e. 45.6 kg) was calculated for 100% consumption each of dab, plaice and haddock. For the crustaceans, 100% (i.e. 27.8 kg) was assigned to *Nephrops*. Molluscs were omitted from the dose calculation.

The maximum dose from Sellafield-derived ^{14}C to the critical consumer group for this study (dab + *Nephrops*) of 2.05 μSv is in excellent agreement with the (mollusc subtracted) dose of 2.07 μSv (total dose: 2.8 μSv) reported by Sellafield in their 'summary of doses associated with marine discharges for 2014' (Nuclear Decommissioning Authority, 2015). The combined Sellafield and natural production/ weapons testing dose of 2.90 μSv represents <0.3% of the annual permitted dose limit to a member of the general public.

4. Conclusions

Highly variable ^{14}C 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 ^{14}C to subsequent biological uptake and transfer throughout the marine food chain. [14]C 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 and ^{14}C activity of plankton and POC are important factors in the transposition of ^{14}C to higher organisms.

In terms of ^{14}C 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 ^{14}C 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 ^{14}C 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 ^{14}C enriched organic material arriving at the sediment/ water interface, resulting in dilution with older organic material. Further build-up of ^{14}C in sediment would be limited through benthic organisms rapidly scavenging the more labile ^{14}C 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 ^{14}C uptake and enrichment above ambient background. Whilst the degree of ^{14}C enrichment appears to be controlled by proximity to Sellafield, it is not exclusive to the eastern Irish Sea. Significant ^{14}C activities were observed in several western basin organisms, equal to, or in excess of ^{14}C activities observed in comparable east basin organisms. The western gyre is suggested as a possible mechanism for the ^{14}C transfer, retention and uptake in these organisms, but this remains, as yet, unconfirmed.

All Irish Sea pelagic and demersal fish have ^{14}C enhancements above background. Distinctions in ^{14}C 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 ^{14}C activities are observed in planktivorous and migratory species.

This study demonstrates the pervasive nature of ^{14}C throughout the Irish Sea, coinciding with continuing nuclear re-processing activities and ^{14}C discharges from Sellafield. Nevertheless, it is important to re-state that current Sellafield ^{14}C 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).

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.marpolbul.2016.10.072.

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