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Ecosystem uptake and transfer of Sellafield-derived radiocarbon (^{14}C) part 2: The West of Scotland



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ABSTRACT

Ecosystem uptake and transfer of Sellafield-derived radiocarbon (^{14}C) were examined within the West of Scotland marine environment. The dissolved inorganic carbon component of seawater, enriched in ^{14}C , is transported to the West of Scotland where it is transferred through the marine food web. Benthic and pelagic biota with variable life-spans living in the North Channel and Clyde Sea show comparable ^{14}C activities. This suggests that mixing of ^{14}C within the Irish Sea results in a relatively constant northwards dispersal of activity. Benthic species in the Firth of Lorn have similar ^{14}C enrichments, demonstrating that Irish Sea residual water is the dominant source to this area. Measured ^{14}C activities in biota show some similarity to western Irish Sea activities, indicating that dispersion to the West of Scotland is significant with respect to the fate of Sellafield ^{14}C releases. Activities measured in commercially important species do not pose any significant radiological risk.

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1. Introduction

1.1. Radiocarbon (^{14}C)

Radiocarbon (^{14}C) is estimated to be the largest contributor to the collective dose that the global population receives from the nuclear industry (UNSCEAR, 2008), due to its long half-life (5730 years) and ready uptake into the global carbon cycle. In the UK, the Sellafield nuclear fuel reprocessing facility is authorised to discharge waste ^{14}C to the atmosphere and the marine environment and this ^{14}C is the largest contributor to the collective dose commitment to UK and European populations from the entire nuclear industry (Nuclear Decommissioning Authority, 2015).

Atmospheric releases of ^{14}C from Sellafield largely dominated total discharges until 1994 when a change in discharge policy and, to a lesser extent, an increase in reprocessing activity resulted in increased marine ^{14}C discharges, which have continued since that time. The average marine ^{14}C discharge activity increased from <2 TBq per year, between 1984 and 1993, to approximately 8 TBq per year since 1994 (BNFL, 1985–2004, MAFF, 1992–1995, RIFE, 1996–2015). Low-level radioactive effluent containing ^{14}C is discharged via pipelines that extend 2.1 km offshore into the Irish Sea. ^{14}C is released primarily as inorganic carbon and is incorporated

into the dissolved inorganic carbon (DIC) component of seawater (Begg et al., 1991, 1992, Begg, 1992, Cook et al., 1995). Through fixation of inorganic carbon, marine photosynthesising organisms derive carbon from the DIC pool (Lalli and Parsons, 1993). Marine calcifying organisms, such as molluscs, also utilise DIC to build their exoskeletons (McConnaughey et al., 1997). These two pathways provide routes for the biological uptake of Sellafield-derived ^{14}C ; however, it is the former that allows ^{14}C to be transferred throughout the entire food web.

Sellafield ^{14}C discharges are made in addition to existing “background” inputs of ^{14}C from natural production and fallout from atmospheric testing of nuclear weapons in the 1950s and early 1960s. Cook et al. (1998) and Tierney et al. (2016) presented near-identical background activities for the UK marine environment over 2 decades (248 ± 1.0 and $249 \pm 0.8 \text{ Bq kg}^{-1} \text{ C}$ for the years 1995 and 2014, respectively). Therefore, marine background ^{14}C activity in this area is relatively stable and the latter value is used to define the modern ^{14}C background level in this study. There have been various studies concerned with the fate of Sellafield-derived ^{14}C within the Irish Sea (e.g. Begg et al., 1992, Cook et al., 1995, 1998, 2004, Wolstenholme et al., 1998, Gulliver et al., 2001, Muir et al., 2015, Tierney et al., 2016) and several have identified uptake of Sellafield ^{14}C within the tissues of marine and intertidal organisms, leading to enrichment above ambient background (e.g. Begg et al., 1992, Cook et al., 1995, 1998, 2004), while others have established the role of molluscs in ^{14}C uptake and accumulation within the inter-tidal environment (Cook et al., 2004, Muir et al., 2015, Tierney et al., 2016). However, none have considered the marine ecosystem in detail. There has also been relatively little research on

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Sellafield ^{14}C beyond the Irish Sea. The net northerly movement of water through the North Channel dictates the long-term dilution and dispersion of Sellafield marine discharges (Dunster, 1998), with >99% of Sellafield ^{14}C dispersed through the North Channel (Gulliver et al., 2001). Using the highly soluble caesium isotopes, ^{134}Cs and ^{137}Cs , Jefferies et al. (1973) calculated transit times to the North Channel from Sellafield (then Windscale) in the region of 1.1–1.8 years. A shorter transit time was reported by Kershaw and Baxter (1995) of approximately 1 year, however a study on Sellafield releases of ^{99}Tc observed a further reduced transit time of 3 months (Kershaw et al., 2004). These studies show that Irish Sea circulation is highly variable and it is difficult to estimate a transit time to the North Channel and beyond, for Sellafield releases. Transit times for ^{14}C within the DIC fraction of seawater would, nevertheless, be similar to these relatively conservative radionuclides and, over a period of 3 months to a year, sufficient mixing should occur within the Irish Sea for a relatively homogenous activity (on short time scales) to reach the North Channel. From the North Channel, dissolved ^{14}C is carried around the Scottish coastline by the Scottish Coastal Current (McKay et al., 1986, Hill and Simpson, 1988) and the Fair Isle Current (Turrell and Henderson, 1990) into the North Sea (Gulliver et al., 2004).

Enriched ^{14}C activities in the DIC fraction of seawater and within the tissue of several marine species have been observed along the Scottish coastline (Cook et al., 1998). Similarly, a recent study (Tierney et al., 2016) demonstrated that despite ^{14}C activities reducing with distance from Sellafield due to dilution, mollusc shells on the north-west Scottish coastline were enhanced above ambient background.

This research forms the second half of a two-part study examining the uptake and transfer of Sellafield-derived ^{14}C within the Irish Sea and West of Scotland (WoS) marine ecosystems. In this study, the West of Scotland (Fig. 1) is designated as *far-field*, relative to Sellafield ^{14}C inputs. The *near-field* encompasses the Irish Sea, and is examined in Part 1 (Muir et al., in this issue). Previous studies (Cook et al., 1998, Tierney et al., 2016) have observed Sellafield-derived ^{14}C enhancements in selected offshore and intertidal species in the WoS, therefore it is important to consider ^{14}C uptake within the wider WoS marine ecosystem. The overarching objectives are detailed in Part 1 (Muir et al., in this issue). This study (Part 2. The West of Scotland) investigates, in detail, ^{14}C activities within the biotic and abiotic components of the WoS marine ecosystem, including that of commercially important fish/crustacean species and the transfer of ^{14}C through the marine food web. Due to its continuing discharge from Sellafield, long half-life and high bio-availability, it is important to gain an understanding of the transport,

uptake and ultimate fate of ^{14}C within these marine ecosystems, not only with regard to past, current and future authorised ^{14}C discharges, but also in the unlikely event of a nuclear accident.

1.2. Study areas

Samples were predominantly collected from 3 areas in the WoS; the North Channel, the Clyde Sea and the Firth of Lorn.

1.2.1. North Channel

The North Channel connects the northern end of the Irish Sea with the northeast Atlantic Ocean via the Clyde Sea. The channel narrows to 30 km in width and the seabed is characterised by rocky outcrops and localised enclosed deeps such as Beaufort's Dyke (Wilding et al., 2005a). Compared to the Irish Sea, the North Channel is relatively deep, averaging 90 m, and the dyke descends to 312 m (Callaway et al., 2011). Flow rates within the Irish Sea are typically low; however the geomorphology of the North Channel means that the tidal flow here can be relatively fast at up to 1.5 m s^{-1} . A net northerly flow though the North Channel exists (Ramster and Hill, 1969, Howarth, 1982, Gulliver et al., 2001), but flow direction is seasonally variable and can be reversed (Dabrowski et al., 2010). Due to high flow rates, the central channel substrate is mainly rock and gravel with finer material deposited to the east, proximate to the Great Plateau (Wilding et al., 2005a). Beaufort's Dyke was used as a munitions disposal ground by the UK military from end of World War I until 1972 (Fisheries Research Services, 1996) and, perhaps as a consequence, there is a lack of studies on the benthic ecosystem within the North Channel.

1.2.2. Clyde Sea

The Arran Basin is the predominant depositional basin within the Clyde Sea, where deep water can become isolated in summer and renewed during winter when currents carrying dense water flow over the Great Plateau (Wilding et al., 2005b). Sills at the entrances of sea lochs north of the Arran Basin produce a similar effect where loch bottom waters can also become isolated (Edwards and Sharples, 1986). As the water is only exchanged intermittently, the deep benthic environment of the Clyde Sea is particularly vulnerable to anthropogenic impacts (Wilding et al., 2005b). The subtidal environment mainly consists of fine grained layered muds which extend from the Arran Basin into the basins of the sea lochs (Moore, 1931, Wilding et al., 2005b). Benthic communities are characterised by high abundances of heart urchin (*Echinocardium cordatum* and *Brissopsis lyrifera*) and brittle stars

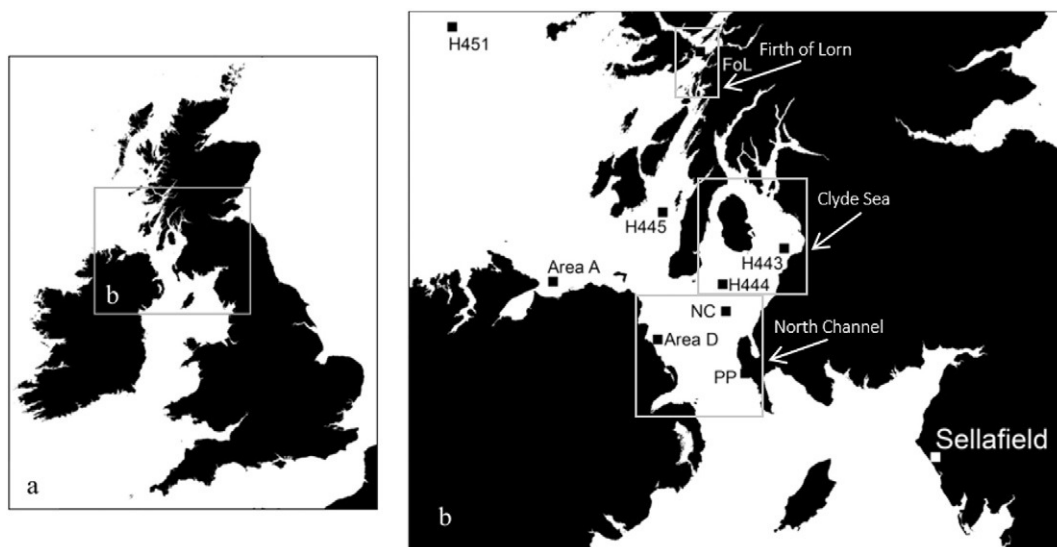


Fig. 1. Map of the British Isles and Ireland (a) with inset (b) showing West of Scotland study sites.

(*Amphiura* spp), while bivalves, polychaetes and the burrowing decapod, *Nephrops norvegicus* (henceforth referred to as *Nephrops*), are also present (Pearson et al., 1986). Fishing pressure has reduced the volume of fish caught in the Clyde Sea significantly (Thurstan and Roberts, 2010). Historically, the area contained important fisheries for herring (*Clupea harengus*), cod (*Gadus morhua*), haddock (*Melanogrammus aeglefinus*), saithe (*Pollachius virens*) and hake (*Merluccius merluccius*) but fishing yields have since collapsed (Thurstan and Roberts, 2010, Heath and Speirs, 2012). Currently, the important remaining fisheries in the Clyde Sea are for *Nephrops* and scallops (*Pecten maximus* and *Chlamys opercularis*) (Thurstan and Roberts, 2010).

1.2.3. Firth of Lorn

The Firth of Lorn is located on the west coast of Scotland to the east and south east of the Isle of Mull. The Corryvreckan whirlpool lies to the south of the Firth of Lorn where a strong tidal race produces flow speeds up to 4.5 m s^{-1} which has a significant impact on local sediment transport (Howe et al., 2015). The central firth area contains depositional basins at 150 m depth and thick sequences of muds are also found in the many sea lochs that surround the firth (Howe et al., 2015). Sediments within the deep basins support dense fields of crinoids (*Leptometra celtica*) and large numbers of northern sea-fans (*Swiftia pallida*) are found on sediment-covered stony sea-beds (Davies, 1999). The diversity of habitats here is reflected in the existence of a number of nationally-rare benthic species (Plaza and Sanderson, 1997). Due to its biological richness, the Firth of Lorn is designated as a Special Area of Conservation (SAC). This has resulted in a ban on scallop dredging in the area, previously an important fishery. *Nephrops*, found in the muddy habitats, is now the single most significant fishery. Although not a major spawning ground, the Firth of Lorn has been identified as an important nursery ground for larval and juvenile fish from a wide range of species (Fox and Lappalainen, 2014).

2. Methodology

Sampling and analytical techniques utilised are described in detail in Muir et al. (in this issue) and summarised here in relation to the sites sampled which are shown in Fig. 1 and detailed in Table 1. Seawater, sediment and benthic organisms were collected in the Firth of Lorn (station FoL) in April 2014, on-board the RV Calanus. Plankton samples were collected at the same site in August 2014, together with additional seawater samples. A sampling campaign within the Irish Sea was conducted in June 2014 on-board the RV Prince Madog, and included the North Channel (station NC) where the same suite of sampling techniques was used. Additional North Channel seawater samples were collected from the coastline near to Portpatrick (station PP) in April 2014

and August 2014. A number of fish and crab samples were collected by the Marine Scotland Science's West of Scotland demersal fish survey, from the RV Scotia during November 2014. These samples were predominantly taken from the Clyde Sea (H443 and H444), however, some were also collected further north, between the Isles of Gigha and Islay (H445) and off the west coast of Tiree (H451). The Agri-Food and Biosciences Institute, Northern Ireland (AFBI-NI) collected shellfish samples from off the northern Irish coastline (Areas A and D) in February 2015.

2.1. Seawater ^{14}C biogeochemical fractions

Surface water was collected to measure ^{14}C activity in the four biogeochemical fractions; dissolved inorganic carbon (DIC), dissolved organic carbon (DOC), particulate inorganic carbon (PIC) and particulate organic carbon (POC). Surface samples from 2 m depth were collected on research vessels at stations NC and FoL by pumping 160 l of seawater on board. Additional 160 l samples were collected, at high tide using 20 l carboys, from the North Channel coastline at Port Patrick. The water was filtered through $0.7 \mu\text{m}$ glass fibre filters with particulate material used for analysis of PIC and POC. Several 500 ml aliquots of filtrate were collected in foil bags for analysis of DIC. Further aliquots of 500 ml were collected in glass bottles for analysis of DOC with 1 ml of (85%) orthophosphoric acid added to liberate DIC and fix the organic carbon content.

2.2. Sediment organic content

Sediment cores were retrieved from stations NC and FoL using an OSIL maxi-corer. Cores were approximately 30 cm in depth and sectioned into 1 cm vertical increments which were initially frozen for storage. The sections were later thawed, oven dried at 40°C and ground into a fine powder. The ^{14}C activity in the organic component of sediment was measured within four depth increments (0–1 cm, 5–6 cm, 10–11 cm and the base of the core) in one core from each site.

2.3. Biota

Beam trawls and Van Veen grabs were utilised to sample the benthic biota at NC and FoL. A range of organisms was collected and identified, where possible, to species level. Fish and shellfish stock surveys (by AFBI and Marine Scotland Science) using bottom trawls provided additional fish and crab samples. Muscle/soft tissue was subsampled and freeze dried. Where more than one individual of a species was collected, samples were proportionally combined and multiple samples were made if six or more individuals were available. Similarly, where several

Table 1
Sampling station details for the West of Scotland.

Sampling station	Study area	Date sampled	Station co-ordinates	Sample type(s)
FoL	Firth of Lorn	April 2014; August 2014	56 23.83 N, 05 36.86 W	Seawater, sediment, plankton, benthic organisms
NC	North Channel	June 2014	55 08.85 N, 05 18.93 W	Seawater, sediment, plankton, benthic organisms
PP	North Channel	April 2014; August 2014	54 50.33 N, 05 06.98 W	Seawater
H443	Clyde Sea	November 2014	55 27.79 N, 04 48.95 W	Fish survey
H444	Clyde Sea	November 2014	55 16.58 N, 05 20.39 W	Fish survey
H445	Sound of Jura	November 2014	55 37.72 N, 05 53.54 W	Fish survey
H451	Outer Hebrides	November 2014	56 27.81 N, 07 50.16 W	Fish survey
Area A	North Channel (north-west)	February 2015	55 14.28 N, 06 49.28 W	Scallop survey
Area D	North Channel	February 2015	54 58.20 N, 05 54.32 W	Scallop survey

relatively analogous species had been collected with few individuals of a specific species, the samples were combined (e.g. starfish). Plankton nets were utilised at NC and FoL to collect phytoplankton (80–270 µm mesh size) and zooplankton (>270 µm). Whole plankton samples were freeze dried.

2.4. ^{14}C analysis procedure

To measure the ^{14}C activity, organic samples (DOC, POC, organic sediment, biota) were combusted in sealed quartz tubes according to the method of Vandeputte et al. (1996) and inorganic samples (DIC, PIC) hydrolysed with HCl (1 M) to liberate CO_2 . The gas was cryogenically trapped and purified and graphite was produced from 3 ml subsamples according to the method of Slota et al. (1987). Sample $^{14}\text{C}/^{13}\text{C}$ 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 ($\delta^{13}\text{C}$) ratios were measured offline on a VG SIRA 11 isotope ratio mass spectrometer for calibration of natural fractionation of the measured ^{14}C . ^{14}C results were calculated relative to the international standard (oxalic acid II, SRM-4990C) as ^{14}C activity ratios (fraction modern, $F^{14}\text{C}$). Fraction modern results were converted to specific activities ($\text{Bq kg}^{-1}\text{ C}$) 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.

3. Results and discussion

3.1. North Channel

3.1.1. Seawater ^{14}C biogeochemical fractions

The ^{14}C values for the biogeochemical fractions of the surface seawater samples from stations PP and NC are presented in Table 2. The DIC component was enriched in ^{14}C at both sites and at all 3 sampling dates, in line with the dissolved inorganic form of ^{14}C discharged from Sellafield. The POC fraction was also enriched at station NC but marginally depleted at station PP for both the April and August sampling periods. The $\delta^{13}\text{C}$ values of the POC fractions at station PP were lower than that at the NC station, which indicates that a greater proportion of the material at station PP was probably terrestrial in origin. This would explain the reduced activities as terrestrially-derived run-off could potentially contain a significant proportion of “old carbon”, resulting in a dilution effect. Surface water collected during June 2014 from station NC was also enriched in ^{14}C within the PIC fraction. This enrichment in the PIC could result from movement of fine, ^{14}C enriched material from the intertidal zone as described by Tierney et al. (2016). The PIC fraction was significantly depleted at station PP and the lower $\delta^{13}\text{C}$ values for these samples indicate that a greater proportion of the material was again terrestrial in origin. The single DOC sample

containing enough dissolved carbon for ^{14}C analysis was significantly depleted and had a $\delta^{13}\text{C}$ value typical of terrestrial carbon, indicating significant run-off of “old carbon” from land. ^{14}C activities measured in the same fractions of seawater from the North Channel in 1989 (Cook et al., 1995) showed similar enrichment in the DIC fraction and depletion in the DOC and POC fractions, despite the increase in Sellafield ^{14}C marine discharges at this period. Also, at this time, North Channel PIC was depleted in comparison to the slight enrichment observed in 2014 at station NC; however, PIC was depleted in 2014 at station PP. The ^{14}C activity of the DIC at station PP was also measured in 1995 at the onset of increased Sellafield discharges (Cook et al., 1998) and was significantly enriched ($430 \pm 4 \text{ Bq kg}^{-1}\text{ C}$) compared to the activities measured in this study. Sellafield discharges of ^{14}C were higher in the 12 months prior to sample collection in 1995 (8.7 TBq) relative to the 2014 sample collection (4.8 TBq). As the transit time of discharges from Sellafield to the North Channel is in the order of 3 months to >1 year (Jefferies et al., 1973, Kershaw and Baxter, 1995, Kershaw et al., 2004), the lower activities described here are most likely a direct result of the lower discharged activities in the preceding months.

3.1.2. Sediment organic content

The organic component at all depth increments of station NC sediment were depleted in ^{14}C (Table 3). The measured activities in the top 11 cm were relatively homogenous ($204\text{--}210 \text{ Bq kg}^{-1}\text{ C}$), while the base activity (29–30 cm) was significantly depleted in comparison (135 Bq kg^{-1}). The large variety of benthic species found at this station, (discussed below) demonstrates that this is a highly biologically active site and the relatively homogeneous nature of the surface sediments is likely to be caused to a significant degree by intensive bioturbation. The ^{14}C activity observed in the surface sediment is depleted, which contrasts with the ^{14}C enrichment observed in surface water POC at station NC. POC may be rapidly scavenged from the water column and/or the surface sediment resulting in a very low flux and incorporation of Sellafield-derived ^{14}C into surface sediments. In addition, the build-up of higher activity material could also be masked through effective mixing to depth of ^{14}C -enriched organic material with significant quantities of old, ^{14}C -depleted organic material in the sediments, resulting in a dilution effect (see Muir et al., in this issue). Furthermore, physical transport of ^{14}C enhanced particulate material from this site by currents could also reduce the volume reaching the sediment and this has previously been suggested as an important mechanism for the transport of enhanced fine inorganic material in northern Irish Sea coastal sites (Tierney et al., 2016).

3.1.3. Biota

The ^{14}C activities of sampled biota at station NC are presented in Fig. 2. All species analysed were enriched relative to the ambient background and mostly varied between 280 and 330 $\text{Bq kg}^{-1}\text{ C}$ with whiting (*Merlangius merlangus*) having a significantly higher activity ($413 \pm$

Table 2
Gross and net specific ^{14}C activities ($\text{Bq kg}^{-1}\text{ C} \pm 1\sigma$) and $\delta^{13}\text{C}$ (‰ relative to VPDB) values from the four biogeochemical fractions of North Channel surface water samples from stations NC and PP. Net activities above the ambient background of $249 \pm 1 \text{ Bq kg}^{-1}\text{ C}$ are in bold. Samples less than this are denoted as ‘Depleted’.

Date sampled (station)	^{14}C specific activities ($\text{Bq kg}^{-1}\text{ C} \pm 1\sigma$)			
	DIC	PIC	DOC	POC
April 2014 (PP)	278 ± 2	180 ± 1	n/a	247 ± 1
	29 ± 2 (+1.1‰)	Depleted (−2.0‰)		Depleted (−22.1‰)
June 2014 (NC)	287 ± 1	271 ± 1	106 ± 1	279 ± 1
	38 ± 2 (+3.9‰)	22 ± 1 (+1.1‰)	Depleted (−26.8‰)	30 ± 1 (−19.4‰)
August 2014 (PP)	279 ± 1	153 ± 1	n/a	241 ± 1
	30 ± 1 (+7.5‰)	Depleted (−0.8‰)		Depleted (−22.0‰)

n/a denotes insufficient carbon in sample for analysis.

Table 3

Sediment organic fraction gross ^{14}C activities ($\text{Bq kg}^{-1} \text{C} \pm 1\sigma$) and $\delta^{13}\text{C}$ values (‰ relative to VPDB) in selected horizons for station NC. Values less than ambient background ($249 \pm 1 \text{ Bq kg}^{-1} \text{C}$) are denoted as 'Depleted'.

Horizon depth (cm)	Gross ^{14}C activity ($\text{Bq kg}^{-1} \text{C}$)	$\delta^{13}\text{C}$ (‰)
0–1 (surface)	204 ± 1 Depleted	–21.7
5–6	210 ± 1 Depleted	–21.8
10–11	210 ± 1 Depleted	–21.8
29–30 (Base)	135 ± 1 Depleted	–21.9

$2 \text{ Bq kg}^{-1} \text{C}$). In contrast, phytoplankton ($280 \pm 2 \text{ Bq kg}^{-1} \text{C}$) and zooplankton ($283 \pm 2 \text{ Bq kg}^{-1} \text{C}$) had amongst the lowest ^{14}C activities. Dab (*Limanda limanda*) (samples 1 and 2) showed intra-species variation, where the difference between the two samples, each consisting of 3 individuals, was approximately $42 \text{ Bq kg}^{-1} \text{C}$. Similar intra-species differences have been observed for dab in the Irish Sea (Muir et al., in this issue) while other multiple samples of the same species/species group showed little or no intra-species variation.

Phytoplankton species have a relatively fast carbon turnover rate and short lifespan, and will readily incorporate ^{14}C from DIC during photosynthesis. The ^{14}C activities in DIC, phytoplankton and POC were comparable at station NC illustrating direct uptake of ^{14}C from ambient waters and implying that the POC is predominantly derived *in situ* from phytoplankton. The zooplankton sample from station NC was observed to consist largely of copepods (probably *Calanus finmarchicus* and/or *Calanus helgolandicus* due to their predominance in this area) with some ctenophores and ichthyoplankton. Although some zooplankton species are carnivorous, copepods feed directly on phytoplankton (Meyer-Harms et al., 1999). That zooplankton ^{14}C activity is also similar to that of phytoplankton, reflecting the ^{14}C activity of their primary source of food, and the transfer of ^{14}C through the food chain.

Although phytoplankton and zooplankton ^{14}C activities were amongst the lowest and the whiting activity was the highest, there is

no obvious trend of increasing ^{14}C activity moving up the food chain. This corresponds to a general transfer of ^{14}C from primary producers to higher organisms with no concentration effect, as might be expected. Any variation in ^{14}C activity more likely derives from variations in the food source and the integration period of carbon uptake. Filter feeders, such as the common cockle (*Cerastoderma edule*) will incorporate ^{14}C from the plankton and POC that they ingest (Iglesias et al., 1992). Organisms like polychaete worms, heart urchin (*Echinocardium cordatum*), brittle stars, Calocaris shrimp (*Calocaris macandreae*) and the hermit crab (*Pagurus bernhardus*) are predominantly detritivorous, feeding on the organic material falling from the water column. The relative ^{14}C enhancement above background in these species, confirms the supply of ^{14}C enriched organic material to the sediment surface. Other crab species (*Goneplax rhomboides*, *Atelecyclus rotundatus*, *Inachus* sp.) will predate on smaller organisms as well as feeding on detritus. The sea mouse (*Aphrodita aculeata*), Nephrops, starfish (*Asterias rubens*, *Crossaster papposus*, *Luidia sarsii*, *Asteroidea* sp.) and fish species (dab, sole, dragonet (*Callionymus lyra*), ling (*Molva molva*), whiting) are predatory, feeding on other benthic organisms. These species have higher ^{14}C activities than the plankton groups indicating that organisms occupying higher trophic levels are integrating ^{14}C over a longer period of time, including periods of higher ambient activities. For example, many of the analysed benthic species are only locally mobile (e.g., brittle star, sea mouse and starfish). Therefore, their high activities relative to that of phytoplankton are a result of uptake during a period of higher ambient activity, corresponding to transient ^{14}C enrichment in the DIC fraction of seawater, which was subsequently passed through the food chain. Due to their relatively longer life-span, an integrated higher activity in these species is now observed. Conversely, whiting are highly mobile and the comparatively high ^{14}C activity in this sample probably results from sampling individuals which had previously foraged in the Irish Sea. Demersal fish within the Irish Sea have ^{14}C activities $>400 \text{ Bq kg}^{-1} \text{C}$, as do their prey items (Muir et al., also submitted). Therefore, it is likely that station NC whiting had migrated from the more ^{14}C -enriched Irish Sea.

3.2. ^{14}C activities in fish and shellfish survey species

Scallops are filter feeders and are, therefore, likely to have a similar ^{14}C activity to phytoplankton and to ambient DIC. The ^{14}C activity of scallops (Fig. 3) collected at Area A ($249 \pm 1 \text{ Bq kg}^{-1} \text{C}$) was identical

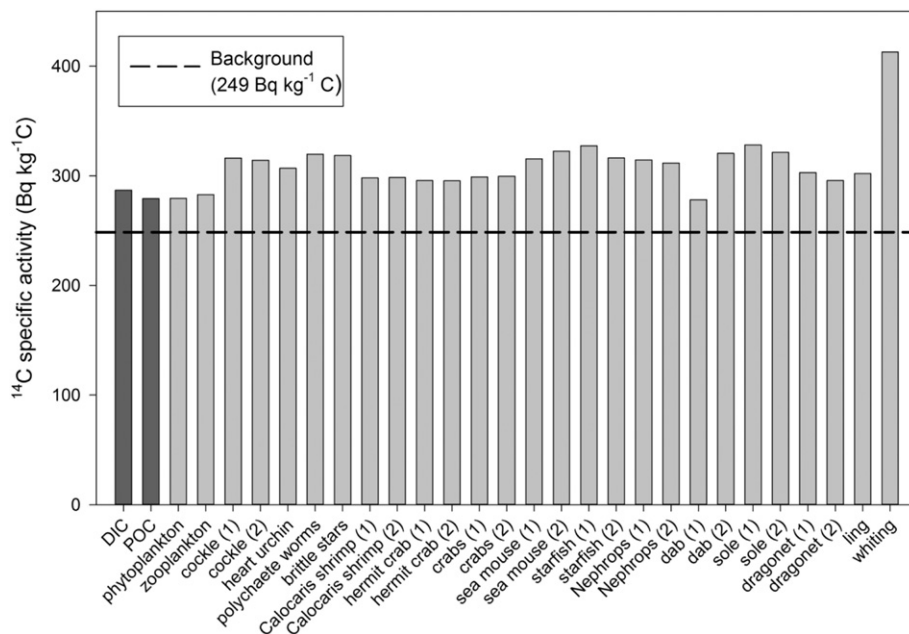


Fig. 2. Gross specific ^{14}C activities ($\text{Bq kg}^{-1} \text{C}$) in benthic biota samples and seawater DIC and POC, collected at station NC. The dashed line indicates the measured background activity of $249 \text{ Bq kg}^{-1} \text{C}$, measured in blue mussel (*Mytilus edulis*) shells obtained from the West Coast of Ireland.

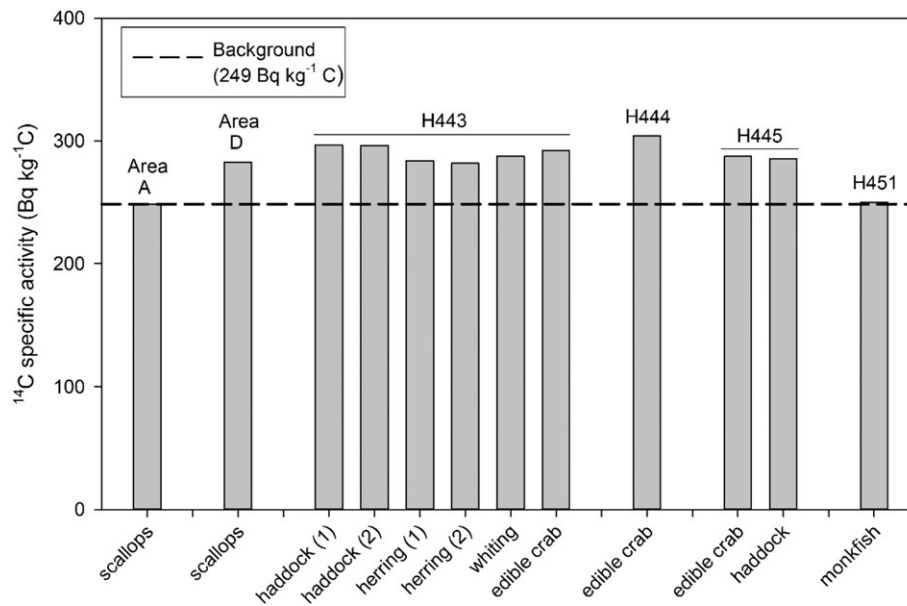


Fig. 3. Gross specific ¹⁴C activities (Bq kg⁻¹ C) in species collected in the Clyde Sea and other areas during fisheries surveys. The dashed line indicates the measured background activity of 249 Bq kg⁻¹ C, measured in blue mussel shells obtained from the West Coast of Ireland.

to background, whereas scallops from Area D were enriched (283 ± 2 Bq kg⁻¹ C). Area A is located on the northern coast of Northern Ireland where the influence of Atlantic Ocean water should be greater and this is demonstrated in the observed ¹⁴C activity being equal to background. Area D, in the North Channel, is affected by a southerly current which carries Atlantic water down the western edge of the North Channel (Bowden, 1980, Dabrowski et al., 2010). Despite there being some Atlantic influence, the effect of Sellafield ¹⁴C discharges is still observable in this sample.

The ¹⁴C activities of fish and crab samples from the Marine Scotland Science surveys are also shown in Fig. 3. Samples from the Clyde Sea area (H443 and H444) were ¹⁴C enriched, although fish samples typically had a lower activity here than fish at station NC. Haddock (*Melanogrammus aeglefinus*), primarily a benthic feeder, and herring (*Clupea harengus*), a planktivore, had relatively similar activities at station H443 (296 ± 2 Bq kg⁻¹ C and 283 ± 2 Bq kg⁻¹ C respectively) indicating little variation in ¹⁴C activity in the water column. In comparison to station NC, whiting at H443 had a significantly lower activity (288 ± 2 Bq kg⁻¹ C), again indicating that station NC whiting had spent time foraging in an enriched area, probably within the Irish Sea. Haddock samples collected further north and west (station H445) were also enriched (286 ± 2 Bq kg⁻¹ C) but had a reduced activity relative to Clyde Sea haddock. Edible crab (*Cancer pagurus*) activity showed little variation between stations H443 (292 ± 2 Bq kg⁻¹ C), H444 (304 ± 2 Bq kg⁻¹ C) and H445 (288 ± 2 Bq kg⁻¹ C). The activities at these stations are similar to those measured in other crustaceans at station NC, suggesting there was little difference in benthic ¹⁴C activity between the North Channel and Clyde Sea.

Station H451 (situated approximately 74 km to the west of the Outer Hebrides) was the most remote sampling area from Sellafield in this study. Only monkfish (*Lophius piscatorius*) were sampled from this station and the measured ¹⁴C activity (250 ± 1 Bq kg⁻¹ C) is indistinguishable from ¹⁴C background and reflects the influence of Atlantic waters. Irish Sea residual waters (with enriched ¹⁴C activity) are carried northwards by currents which closely hug the Scottish coastline (McKay et al., 1986, Hill and Simpson, 1988). This result signifies that Sellafield ¹⁴C has little influence in offshore waters dominated by Atlantic water to the west of the Scottish Coastal Current, at least in the benthic environment that monkfish inhabit, and at the time of sampling.

3.3. Firth of Lorn

3.3.1. Seawater ¹⁴C biogeochemical fractions

Surface water sample activities (Table 4) showed an enriched DIC ¹⁴C activity at station FoL during the period when benthic biota were sampled in April 2014 and when plankton were sampled in August 2014. In contrast, PIC and POC activities were depleted at both sampling periods. The reduction in DIC activity between April and August demonstrates that the ambient ¹⁴C activity is changeable over short periods of time despite being distant from the discharge source. This could be due to the variation in discharged activity but local current direction influencing the sources of local water will also have an impact on ambient activities. However, station FoL DIC activities are similar to those observed at stations NC and PP. This suggests that at the time of sampling, there was relatively little dilution of DIC ¹⁴C between these two sites after mixing between Atlantic and Irish Sea water in the North Channel, and that there was little further input of Atlantic water to this water mass. DIC sampled at a coastal site further north in Scotland in 1995 (Cook et al., 1998) showed a comparable ¹⁴C activity (272 ± 4 Bq kg⁻¹ C), despite higher discharges in the year preceding sampling. The observed activity could arise from further dilution of residual Irish Sea water with Atlantic water. Although the small dataset from both these studies makes it difficult to draw any firm conclusions on hydrodynamics, it does highlight a general reduction in ¹⁴C activity with distance from Sellafield and the possibility of using Sellafield-derived ¹⁴C as a tracer for water masses in the north of Scotland.

In the case of the PIC, carbon derived from terrestrial sources could affect the activity although the $\delta^{13}\text{C}$ indicates that the material is predominantly of marine origin. This would imply a dilution by PIC of pre-Sellafield origin as described for the intertidal environment in Tierney et al. (2016). In the case of the POC, the April 2014 sample is much more depleted than the August 2014 sample. The April sample has a $\delta^{13}\text{C}$ value that is 3.1‰ lower than the August sample and would therefore imply a greater effect from carbon derived from the terrestrial environment in early spring. Also, the increase in POC activity between April and August probably results from increased primary production over the spring and summer months, converting more ¹⁴C enriched DIC into phytoplankton with subsequent transfer through the food chain resulting in a higher POC activity.

Table 4

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

Date	^{14}C specific activities ($\text{Bq kg}^{-1} \text{C} \pm 1\sigma$)			
	DIC	PIC	DOC	POC
April 2014	298 ± 2	183 ± 1	n/a	198 ± 1
	49 ± 2 (0‰)	Depleted (−2.7‰)		Depleted (−24.5‰)
August 2014	277 ± 1	196 ± 1	n/a	234 ± 1
	28 ± 1 (+7.7‰)	Depleted (−3.4‰)		Depleted (−21.4‰)

n/a denotes insufficient carbon in sample for analysis.

3.3.2. Sediment organic content

Organic sediment ^{14}C activities at station FoL are all below the ambient background (Table 5). They are again uniform throughout the top 11 cm (208–209 $\text{Bq kg}^{-1} \text{C}$) with a small decrease in activity at the core base, and very similar to station NC for the top 11 cm (204–210 $\text{Bq kg}^{-1} \text{C}$) (Table 4). Again, sediment homogeneity is consistent with intensive bioturbation as a result of the high biological activity at this site. Surface sediment ^{14}C activity falls between the depleted activities measured in surface water POC in April and August 2014 indicating that this POC material is deposited in the sediment.

3.3.3. Biota

Phytoplankton at station FoL were slightly depleted below background ($241 \pm 1 \text{ Bq kg}^{-1} \text{C}$) and zooplankton were significantly depleted ($227 \pm 1 \text{ Bq kg}^{-1} \text{C}$). All other samples were enhanced and relatively uniform in activity (Fig. 4). The reduced phytoplankton ^{14}C activity at FoL questions the previously stated convention that phytoplankton activity would be expected to reflect DIC activity as is observed at station NC. This could be explained by the phytoplankton sample containing some POC with a depleted ^{14}C activity. However, the zooplankton ^{14}C activity, which is significantly depleted relative to that of phytoplankton, suggests that there may be a more complex explanation as this sample, like its equivalent at station NC, was comprised almost totally of copepods which feed directly on phytoplankton. This aspect of the study will require further sampling and analysis in the future as the value of $227 \text{ Bq kg}^{-1} \text{C}$ is difficult to reconcile with general ocean ambient background activities, although upwelling of older deep water could provide a source for below background ^{14}C activities.

As benthic organisms were enriched in ^{14}C in April 2014, it follows that their food source must have also been enriched. Phytoplankton, at the base of the food chain, and zooplankton (dominated by copepods), as grazers of phytoplankton, were therefore likely to be enriched in ^{14}C prior to when plankton were sampled and ^{14}C subsequently

transferred along the food chain to other organisms. It is possible that plankton and water samples were taken shortly after a change in water mass. If, after a period of stronger Atlantic influence, the water mass became increasingly dominated by Irish Sea residual water, an increase in DIC ^{14}C activity from background or potentially from below background (depending on the age and ^{14}C activity of the Atlantic water) to above background would follow. Although this would result in an increase in plankton activity, this would take time to develop, and therefore result in the ^{14}C enrichment observed in the DIC fraction and depletion in plankton activities at the time of sampling. The fact that other organisms were all enriched in ^{14}C indicates that, in the long-term, the Firth of Lorn is dominantly supplied by residual Irish Sea water, resulting in these longer-lived organisms integrating ^{14}C over relatively long periods.

A wide range of benthic organisms were collected at station FoL and results are presented in Fig. 4. The sea pen (*Funiculina quadrangularis*) is a colonial planktivorous, passive suspension feeder and its ^{14}C enrichment ($267 \pm 2 \text{ Bq kg}^{-1} \text{C}$) confirms that primary producers in this area were enriched in ^{14}C in the period prior to sampling. The edible sea urchin (*Echinus esculentus*) is an omnivore and could integrate ^{14}C from grazing on algae or preying upon other invertebrates. A number of species which are predominantly detritivorous were sampled, including the heart urchin, sea cucumber (Holothuroidea sp.), ribbon worm (*Nemertea* sp.), polychaete worms, spoon worm (*Maxmuelleria lankesteri*), brittle star species (*Ophiocoma nigra*, *Ophiothrix fragilis* and *Ophiura albida*), burrowing shrimp species (*Jaxea nocturna*, *Callianassa subterranea*), and crab species (*Pagurus prideaux*, *Ateacyclus rotundatus*, *Inachus* sp.). The enriched ^{14}C activities measured in these organisms proves that ^{14}C enhanced organic material is supplied to the sediment surface and is being scavenged by the large number of detritivores inhabiting this area. The overall extent of Sellafeld ^{14}C ecosystem uptake and biological transfer is shown by the enriched activities found in predatory organisms from station FoL, including the common whelk (*Buccinum undatum*), sea mouse, starfish species (*Asterias rubens*, *Crossaster papposus*, *Luidia sarsii*), squat lobster (*Munida rugosa*), *Nehrops* and megrim (*Lepidorhombus whiffiagonis*).

Small reductions in ^{14}C activities of biota are observed at station FoL compared to those of station NC, with an average reduction in the enhancement over ambient background of 13%. This confirms that Irish Sea residual water is the dominant source of water to station FoL and so we might expect to see similar enhancements in biota beyond station FoL until significant dilution of the Scottish Coastal Current occurs. Clyde Sea samples were similar to station NC but fish ^{14}C activities were generally lower. At each site, the activities of organisms are relatively homogenous, despite the large variation in species. The range in species covers significantly different lifespans, different metabolic rates and different feeding behaviours and suggests that the overall Sellafeld effect at these sites, particularly at station FoL, is relatively constant. Variable mixing patterns of seawater in the West of Scotland with residual Irish Sea waters, at any given time, may cause small changes in the overall ^{14}C DIC activity. However, any short-term variations in ambient DIC ^{14}C activity are likely to be minor in comparison to sites closer to

Table 5

Sediment organic fraction gross and net ^{14}C activities ($\text{Bq kg}^{-1} \text{C} \pm 1\sigma$) and $\delta^{13}\text{C}$ values (‰ relative to VPDB) in selected horizons for station FoL. Values less than ambient background ($249 \pm 1 \text{ Bq kg}^{-1} \text{C}$) are denoted as 'Depleted'.

Horizon depth (cm)	Gross ^{14}C activity ($\text{Bq kg}^{-1} \text{C}$)	$\delta^{13}\text{C}$ (‰)
0–1 (surface)	208 ± 1 Depleted	−22.0
5–6	208 ± 1 Depleted	−21.9
10–11	209 ± 1 Depleted	−21.8
33–34 (Base)	200 ± 1 Depleted	−21.6

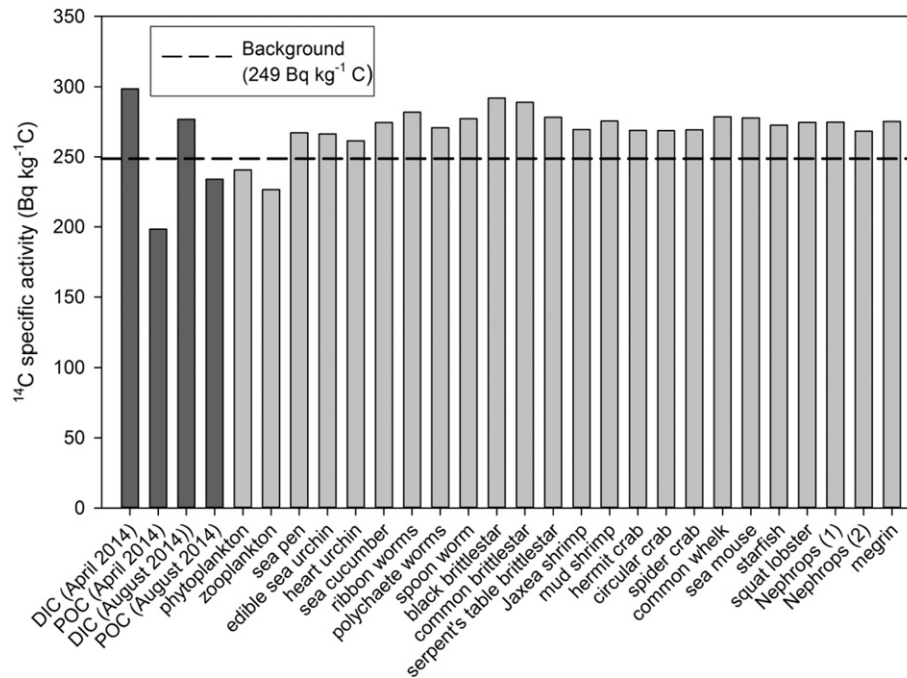


Fig. 4. Gross specific ^{14}C activities ($\text{Bq kg}^{-1}\text{C}$) in benthic biota samples and seawater DIC and POC, collected at station FoL. The dashed line indicates the measured background activity of $249\text{ Bq kg}^{-1}\text{C}$, measured in blue mussel shells obtained from the West Coast of Ireland.

Sellafield, particularly in the north-east Irish Sea which presents greater heterogeneity.

4. Comparison of far-field (West of Scotland) and near-field (Irish Sea) results

Comparing the results presented here with data from the Irish Sea (Muir et al., also submitted) allows us to better understand the scope of transport of ^{14}C to the WoS and ecosystem uptake in this region. A general reduction in DIC ^{14}C activity with increasing distance from Sellafield is observed when comparing measurements from the north-east Irish Sea ($546 \pm 2\text{ Bq kg}^{-1}\text{C}$) to WoS results. The relative decrease in western Irish Sea DIC activity ($264 \pm 1\text{ Bq kg}^{-1}\text{C}$), compared to WoS

sites, supports previous work which showed that $>99\%$ of discharged ^{14}C leaves the Irish Sea through the North Channel (Gulliver et al., 2001). Organic sediment activities at stations NC and FoL are depleted, whereas enriched activities are observed in the north-east Irish Sea (e.g. $298 \pm 1\text{ Bq kg}^{-1}\text{C}$ in surface sediment). This shows the greater flux of enriched material to the sediment in the north-east Irish Sea as would be expected at a site much closer to the ^{14}C source.

In comparison with organisms obtained from the north-east Irish Sea (station EB), it is apparent that ^{14}C activities in West of Scotland organisms are significantly reduced (Fig. 5). Although high ^{14}C activities have been observed in some western Irish Sea (station WB) organisms, most results at station WB are comparable to, or below the activities observed for the North Channel, Clyde Sea and Firth of Lorn stations. The

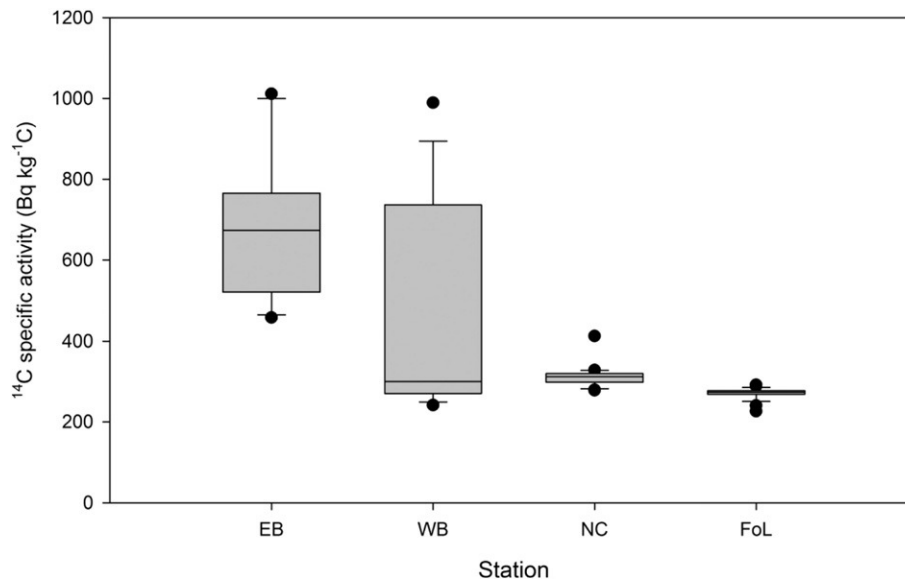


Fig. 5. Boxplot of ^{14}C activities ($\text{Bq kg}^{-1}\text{C}$) from all biota samples measured in Irish Sea stations EB and WB (from Muir et al., also submitted) and West of Scotland stations NC and FoL. Boxes describe respective the interquartile range and whiskers describe the 5th and 95th percentiles. The solid black line within boxes indicates respective median values. Each black dot represents a singular outlier with respect to the majority of the data for that site.

high activity of whiting collected at station NC is clearly identified as an outlier, however, the median activity in benthic biota from station WB is actually less than the equivalent at station NC. This indicates that the northern extremity of the North Channel is receiving similar or higher ^{14}C -enriched DIC inputs than the western Irish Sea, as confirmed by the DIC data. Results from the Irish Sea show a much wider range of activities to that of the West of Scotland. It is likely this is due to monthly changes in ^{14}C discharges from Sellafield having a greater impact on the Irish Sea ambient ^{14}C activity, ultimately increasing the variability. Transfer and mixing processes within the Irish Sea result in a more homogeneous activity being transferred through the North Channel and northwards along the West of Scotland where mixing with Atlantic water can reduce the ambient activity with distance from Sellafield (Cook et al., 1998, Gulliver et al., 2001, Tierney et al., 2016).

Relatively low ^{14}C activities measured in plankton groups both in the Irish Sea and West of Scotland are identified as being statistical outliers and are depicted in Figure 5 as black dots below the interquartile range. In the Irish Sea this most likely occurs as a consequence of the very low Sellafield ^{14}C discharge during the sampling period (Muir et al., also submitted). In recent years it appears that highest monthly discharges of ^{14}C coincide with autumn-winter months (Muir et al., also submitted). Discharges coinciding with plankton blooms during the spring and summer could result in higher organic activities and increased ^{14}C transfer through the food chain (Cook et al., 1995). It is not clear if the recent discharge policy has followed this protocol or is coincidental, as previously there were no trends in discharge activity. Discharging more ^{14}C in periods of low primary production will probably result in a net reduction in the overall ecosystem ^{14}C uptake within the north-east Irish Sea. Due to intensive mixing within the Irish Sea, and the time taken for ^{14}C to be transported northwards, it is unlikely this would have a similar impact beyond the North Channel. However, it is conceivable that due to reduced uptake within the Irish Sea, higher activity water will be transported north, potentially resulting in increased activities in West of Scotland biota, although this effect remains unconfirmed.

Radiation dose rates have been calculated for the Sellafield critical consumer group for ^{14}C activities measured in the north-east Irish Sea (Muir et al., also submitted). These dose rates are negligible for ^{14}C activities which are significantly higher in comparison to the ^{14}C activities observed in commercially important species from the WoS sites. Assuming a WoS critical consumer group has the same consumption rates as the Sellafield critical consumer group (Garrod et al., 2015) and by using the highest activities measured in the WoS for fish (whiting $413 \pm 2 \text{ Bq kg}^{-1} \text{ C}$) and *Nephrops* ($315 \pm 2 \text{ Bq kg}^{-1} \text{ C}$), the maximum dose received would be $0.59 \mu\text{Sv}$. This dose is 71% less than the maximum dose measured for the Sellafield critical consumer group ($2.05 \mu\text{Sv}$) from ^{14}C discharges (Muir et al., also submitted) and does not pose any radiological risk to the public.

5. Conclusions

Sellafield-derived ^{14}C is transported to the north-west of Scotland in the form of DIC. ^{14}C is highly bioavailable as demonstrated from the widespread ^{14}C enrichments observed in marine organisms. Although sediment activities are depleted in ^{14}C relative to ambient background, there is a clear pathway of uptake of ^{14}C by phytoplankton during photosynthesis, followed by transfer to planktivorous organisms and deposition of enriched particulate material. This organic material is rapidly consumed by detritus feeders and subsequently, ^{14}C is transferred through the entire benthic food web. It is apparent that sedimentation processes must be examined in more detail to determine the fate of ^{14}C at the sediment-water interface. Any future work should focus on the different organic carbon fractions within the sediment to better understand both the pathways for ^{14}C re-entry into the marine food and deposition of ^{14}C .

The extent of ^{14}C transport and ecosystem uptake is revealed by enriched activities at the Firth of Lorn (approximately 260 km from Sellafield). Although any increase in Atlantic water influence in the Firth of Lorn will reduce ambient ^{14}C activity, as shown by plankton activities, this area is dominated by inputs from a residual water component from the Irish Sea. However, the overall effect of dilution with Atlantic water is clear and ^{14}C activities reduce with distance from Sellafield, though a similar order of magnitude of ^{14}C enrichments in biota can be expected until there is significant dilution of the Scottish Coastal Current. The small dataset of surface water ^{14}C activities presented in this study shows the potential use of ^{14}C as a tracer for Irish Sea water and mixing processes in the UK marine environment.

Many of the organisms measured are commercially important species and the findings in this study suggest that ^{14}C enrichment is likely to be found in other unmeasured species from the same areas. It must be re-stated that the potential ^{14}C dose received from consumption of seafood in the WoS is negligible, and does not pose any radiological risk to consumers or local populations in the west of Scotland. However, due to its long half-life, high bioavailability and continued release, continued assessment of the fate of ^{14}C in the environment is important. To this purpose, ongoing work is utilising the data collected across this study to develop a predictive ecosystem model tracing the biological fate of ^{14}C released into the marine environment which, unlike other discharged radionuclides, cannot be described using a distribution coefficient.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.marpolbul.2016.11.014>.

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