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NOTE

Enhancement of tritium concentrations on uptake by marine biota: experience from UK coastal waters

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Abstract

Concentrations of tritium in sea water and marine biota as reported over the last ~10 years from monitoring programmes carried out by this laboratory under contract to the UK Food Standards Agency are reviewed from three areas: near Cardiff; Sellafield; and Hartlepool. Near Cardiff, enhancement of concentration factors (CFs) above an *a priori* value of ~1 have already been studied, and attributed to compounds containing organically bound tritium in local radioactive waste discharges. Further data for Cardiff up to 2006 are reported in this note. Up to 2001, CFs increased to values of more than ~7000 in flounders and ~4000 in mussels, but have subsequently reduced; this variability could be due to changes in the organic constitution of compounds discharged.

Near Sellafield and Hartlepool, enhancements to the tritium concentration factor are observed but they are relatively small compared with those near Cardiff. Near Sellafield, plaice and mussels appear to have a CF for tritium of ~10; in some cases concentrations of tritium in winkles are below detection limits and positively measured values indicate a CF of ~3. The variation could be due to mechanisms of uptake by the different organisms. Near Hartlepool there were only a few cases where tritium was positively measured. These data give a value of ~5 for the CF in plaice (on the basis of two samples); ~15 in winkles (eight samples); and >45 in mussels (two samples). Any differences between the behaviours at Sellafield and Hartlepool would need to be confirmed by improved measurements. Possible causes are the organic composition of the effluent and differences in environmental behaviour and uptake by organisms near the two sites. These potential causes need further investigation.

It is emphasised that results from tritium analyses are heavily method dependent; thus comparison with results from other programmes needs to take this into account. Further, the results for enhancement of CF will also depend on the definition of CF itself.

1. Introduction

Tritium in the environment of the UK derives from both natural sources and discharges of radioactive wastes by nuclear, radiochemical, weapons, medical and research facilities. Doses to the public are low (Environment Agency *et al* 2008, and previous reports in the series) but uncertainty exists in dose assessments, not only in the appropriate dose coefficient (Harrison *et al* 2002, Hunt *et al* 2009) but also in the appropriate concentration factor (CF), mainly if organically bound tritium (OBT) is present (e.g. McCubbin *et al* 2001, Williams *et al* 2001). This note reviews recent empirical data for CFs derived from monitoring of the marine environment near three nuclear sites where positive tritium measurements are observable, and discusses potential causes of variability.

The term 'concentration factor' (CF) is commonly used in radiological assessments in referring to the uptake of a chemical element from sea or fresh water into biota. It is usually defined as the ratio:

$$\frac{\text{(Concentration per unit mass of biota at equilibrium)}}{\text{(Dissolved concentration per unit volume of the ambient water)}}$$

with the dimensions (volume)/(mass), often with units $\mathrm{l\,kg^{-1}}$. The mass of biota usually, but not always, refers to fresh (wet) weight. 'Ambient water' usually refers to filtered sea water. Care is needed in interpretation of CFs, particularly in considering the state of equilibrium between biota and the ambient water, but the term has become accepted as useful for radiological modelling purposes.

Tritium, being an isotope of hydrogen, would be expected to be taken up with the water content of the organism and exhibit a CF of approximately 1, or more precisely 0.8 assuming a typical dry/wet weight ratio of 0.2. In its compilation of CFs for radiological modelling purposes by the International Atomic Energy Agency (IAEA), hydrogen is given a CF of 1 for fish, crustaceans, molluscs, macroalgae, zooplankton and phytoplankton (IAEA 2004). Yet it has been shown that under some conditions there can be enhancement of this factor, sometimes by a significant amount.

In the marine environment off Cardiff, UK, CFs for tritium in biota have been investigated (McCubbin *et al* 2001, Williams *et al* 2001). For flounder (*Platichthys flesus*) and mussels (*Mytilus edulis*) CFs of up to 4×10^3 (fresh weight equivalent) were reported. The significant increase in CF compared with unity has been attributed to uptake of tritium in organically bound forms, due to the existence of organic species of tritium in the mix of compounds in the authorised releases of wastes to the Bristol Channel from the Nycomed-Amersham (now GE Healthcare) radiopharmaceutical plant at Whitchurch, Cardiff. There is no reason for isotopic fractionation of hydrogen as part of the uptake process, being essentially a biochemical one. It was suggested that bioaccumulation of tritium by benthic organisms and demersal fish occurs by absorption of organically bound tritium (OBT) into particulate organic matter and transfer to organisms by the food web (McCubbin *et al* 2001). It needs to be emphasised that resulting radiation doses to the public are small and reducing; for 2007 the reported total dose to adult high-rate fish consumers near Cardiff was 0.012 mSv, with the dose to any unborn children 0.014 mSv (Environment Agency *et al* 2008).

Small enhancements of the apparent CF for tritium have also been observed in biota from near Sellafield, UK, and reported in the Environment and Food Standards Agencies' annual monitoring reports since 2000 (FSA and SEPA 2000, Environment Agency *et al* 2008 and reports in this series). Values of CF in excess of ~1 have been reported along the French coast of the English Channel (Masson *et al* 2005). The distribution of tritium may be influenced by an affinity for organic matter in suspended particulates and sediments (Turner *et al* 2009). In this note we focus on the current and recent past state of enhancement of the CF for tritium at different locations around the UK, using data from the monitoring programme carried out by this laboratory under contract to the Food Standards Agency.

2. Sites, data and analytical methods

Clearly, useful comparisons using existing data can only be made for locations where tritium concentrations in biota have been positively measured. In the regular monitoring of marine biota carried out by this laboratory the usual limit of detection is 25 Bq kg^{-1} (Environment Agency *et al* 2008). Thus the choice of locations for comparison in this note has been limited to the environs of nuclear sites where the effects of discharges of tritium provide a signal in excess of this value. These sites are effectively (a) Cardiff, referred to above; (b) Sellafield; and (c) the nuclear power station sites operating advanced gas-cooled reactors (AGRs). Of the latter, Heysham and Hinkley point power stations discharge to waters influenced by releases from Sellafield and Cardiff respectively, making comparisons of the relative effects more difficult. Near Dungeness, tritium in marine biota has been undetectable in most cases. At the Scottish AGR sites there is little monitoring of marine biota for tritium. Thus best use has been made of data from near Hartlepool, Cleveland, on the North Sea coast. The locations of the sites are shown in figure 1.

Unlike many other radionuclide analyses, the results for tritium and any organically bound component are heavily dependent on the analytical method used; the determinands are essentially defined by the method. Thus there are uncertainties when comparing results using different techniques. For this reason, to provide consistent data for the purposes of this note, only the results of monitoring by this laboratory under contract to the Food Standards Agency are used here. The method used for analysis of total tritium (tritiated water and OBT) in biota is based on oxidation using chromic acid, distillation and liquid scintillation counting. Some of the samples are also analysed for their organically bound component defined by drying at 40 °C then oxidising the residue with chromic acid, distillation and counting as before. Sea water is analysed by double distillation with suitable hold-back carriers for volatile radionuclides other than tritium, followed by liquid scintillation counting of the distillate for total tritium. It is possible that the sea water method might underestimate the total tritium present if components remain in the residue after distillation. This in turn might overestimate the calculated CF. Further, the sea water is not routinely filtered prior to analysis; this might have the effect of increasing the tritium concentration from the filtered state which is the usual basis for CFs, thus underestimating them. These effects require further investigation, and are underlined here to emphasise the difficulties in comparison of data sets using different methods.

The data from this study are presented in tabular (rather than graphical) form, for three main reasons; first, for traceability; second, in some cases data are sparse, with ‘less than’, ‘more than’ or ‘not analysed’ values; third, time trends are not as important here as the variabilities, means and standard deviations.

3. Monitoring data near Cardiff

The effects of tritium discharges from the Cardiff site have already been studied in some detail (McCubbin *et al* 2001, Williams *et al* 2001). In this note, more recent measurements from the Food Standards Agency’s monitoring programme are presented and observations made mainly as a basis for comparison of data from Sellafield and Hartlepool. Table 1 presents operator discharge data and results of monitoring of sea water, flounder and mussels, from the monitoring programme near Cardiff (Environment Agency *et al* 2007 and previous reports in

Figure 1. Locations of relevant sites.
(This figure is in colour only in the electronic version)

the series). For these materials, monitoring has been reasonably consistently carried out, giving useful time series of data, from 1997 to 2006.

The discharges have progressively reduced in activity terms, by a factor of ∼20 over the period. This has been reflected in decreasing concentrations of tritium in sea water. The normalised concentration, calculated as $\mathrm{Bq\,l^{-1}}$ per $\mathrm{TBq\,y^{-1}}$, appears to have remained reasonably constant to 2002, increasing from 2003 (significant on the basis of a t-test, $p \sim 0.03$). The increase is by about a factor of 2. Apparent increases in normalised concentration can occur if there is remobilisation from sediment in a period of reducing discharges, as occurred at Sellafield for radiocaesium discharges post-1983 (Hunt and Kershaw 1990); however for tritium from Cardiff, discharges were also reducing from 1997 to 2002, when observed dilution factors were fairly constant. A possible explanation of the observed increases in normalised concentration may be a change post-2003 to the chemical form of compounds of tritium, decreasing its affinity for adsorption on sediment.

Table 1 next presents total tritium concentrations in flounders from the discharge area. From 1997 to 2000 these increased by a factor of 2.8 to 54 $\mathrm{kBq\,kg^{-1}}$ despite a reduction in discharges during this time by a factor of 5.4. There were then decreases in tritium in

Table 1. Cardiff: monitoring data and concentration factors. (Note: (n) indicates number of samples, NA = not analysed.)

Year	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	Mean and range where relevant
^{3}H(total) discharge, TBq	473	277	105	87.2	67.2	59.5	30.2	44.4	40.4	24.8	
^{3}H(total) in sea water, Orchard Ledges East, Bq l^{-1}	53.0(2)	28.0(2)	9.2(2)	8.1(2)	6.0(2)	4.9(2)	5.9(2)	5.3(2)	10.0(2)	5.0(2)	
Normalised concentration, Bq l^{-1} per TBqy^{-1}	0.11	0.10	0.09	0.09	0.09	0.08	0.20	0.12	0.25	0.20	0.13, 0.08–0.25
^{3}H(total) in flounder, Bq kg^{-1} (wet)	19 000(4)	31 000(6)	23 000(3)	54 000(14)	46 000(2)	30 000(2)	15 000(2)	6600(2)	11 000(3)	4400(4)	
OBT in flounder, Bq kg^{-1} (wet)	NA	NA	16 000(3)	51 000(14)	NA	27 000(2)	14 000 (2)	7600(2)	9700(3)	4000(4)	
Fraction of OBT in ^{3}H(total), flounder	NA	NA	0.70	0.94	NA	0.90	0.93	1.15	0.88	0.91	0.92, 0.70–1.15
Concentration factor, ^{3}H(total) flounder	358	1107	2500	6667	7667	6122	2542	1245	1100	880	3000, 360–7700
^{3}H(total) in mussels, Bq kg^{-1} (wet)	NA	41 000(2)	26 000(3)	27 000(14)	24 000(3)	14 000(2)	19 000 (2)	5700(2)	3300(2)	2500(2)	
OBT in mussels, Bq kg^{-1} (wet)	NA	NA	20 000(3)	24 000(14)	25 000(3)	12 000(2)	19 000 (2)	7900(2)	3000(2)	2300(2)	
Fraction of OBT in ^{3}H(total), mussels	NA	NA	0.77	0.89	1.04	0.86	1.00	1.39	0.91	0.92	0.97, 0.77–1.39
Concentration factor, ^{3}H(total) mussels	NA	1464	2826	3333	4000	2857	3220	1075	330	500	2300, 330–4000

flounders to 2006 by a factor of ~12; discharges over this time only decreased by a factor of 3.5. This anomalous behaviour is referred to later in connection with the CFs. Also presented is the concentration of organically bound tritium (OBT) in flounders measured as described in section 2, and the fraction of OBT in the total tritium. This was high, ranging from 0.7 to 1.15; values >1 are due to analytical variations. Next is given the CF in flounders for total tritium, calculated from the measurements in fish and sea water. These increased significantly from 1997 to 2001 by a factor of >20 to ~7700, then decreased to 2006 by a factor of ~9. On the assumption that enhanced uptake of tritium occurs in biota when in an organically bound state, this behaviour could be due to organic status of the tritium over the period and would be consistent with the presence in the effluent up to ~2001 of organic components more readily taken up by biota, followed by a decrease in such components. Data for mussels are also presented, and the behaviour of the CFs is generally similar to flounders although the increases are not as pronounced, and the concentrations themselves show a continuing decrease from 41 kBq kg^{-1} measured in 1998.

4. Monitoring data near Sellafield

Table 2 presents operator discharge data and results of the Food Standards Agency's monitoring of sea water, plaice (*Pleuronectes platessa*), mussels and winkles (*Littorina littorea*) from the monitoring programme at Sellafield (Environment Agency *et al* 2007 and previous reports in the series). For these materials, monitoring has been reasonably consistently carried out, giving useful time series of data from 1999 to 2006; OBT was not included until 2001.

From 1999 to 2004, discharges of tritium varied within a factor of about 1.5, depending on fuel throughput in the Magnox and THORP reprocessing plants; subsequently a reduction of throughput in THORP resulted in lower overall tritium discharges. The standard sea water monitoring location is at St Bees, some 10 km from the discharge point but still within the area of the main tidal excursion. Concentrations of tritium in sea water are representative of those in this area for calculation of CFs. These concentrations have varied throughout the period broadly in line with the discharges, and the normalised concentration (Bq l^{-1} per TBq y^{-1}) varies within a factor of ~2.5; the average value (0.0072 Bq l^{-1} per TBq y^{-1}) is in reasonable agreement with 0.0085, established for ^{137}Cs during the period of relatively high but stable discharges from 1977 to 1982 (Hunt and Kershaw 1990).

Table 2 next presents total tritium concentrations in plaice from the Sellafield tidal area, and OBT when analyses were included from 2002. As for Cardiff, the fraction of OBT in total tritium is high, ranging from 0.75 to 0.95. Calculated CFs for total tritium in plaice range from 6.3 to 18 with a mean of 9.6, showing a distinct enhancement from the *a priori* assumption of ~1. The upper values in 1999 and 2006 reflect the low measured concentrations of tritium in sea water for those years.

In mussels, the behaviour is similar to that in plaice, with a high OBT fraction in total tritium and CFs from 5.1 to 12 with a mean of 8.5. However, winkles appear to exhibit much lower concentrations of total tritium and OBT, in many cases falling below detection limits. This difference could be a reflection of the nature of the food intake of the gastropod winkle by a grazing process rather than uptake of more mobile organic matter by the mussel and plaice. The CF in winkles ranges from <1.1 to 4.3, with a mean of 2.8 using positive measurements.

5. Monitoring data near Hartlepool

Table 3 presents operator discharge data and results of the Food Standards Agency's monitoring of sea water, plaice, mussels and winkles from near Hartlepool (Environment Agency *et al* 2007

Table 2. Sellafield: monitoring data and concentration factors. (Note: (n) indicates number of samples analysed, NA = not analysed.)

Year	1999	2000	2001	2002	2003	2004	2005	2006	Mean and range where relevant
^{3}H(total) discharge, TBq	2520	2260	2560	3320	3900	3170	1570	1090	
^{3}H(total) in sea water, St Bees, Bq l^{-1}	12(12)	17(12)	17(10)	23(12)	21(12)	23(12)	19(12)	9.0(12)	
Normalised concentration, Bq l^{-1} per TBq y^{-1}	0.0048	0.0075	0.0065	0.0069	0.0054	0.0073	0.0121	0.0083	0.0072, 0.0048–0.012
^{3}H(total) in plaice, Bq kg^{-1} (wet)	212(4)	140(3)	108(4)	168 (6)	132(4)	205(4)	161(4)	122(4)	
OBT in plaice, Bq kg^{-1} (wet)	NA	NA	NA	145(4)	103(4)	153(4)	153(4)	105(4)	
Fraction of OBT in ^{3}H(total), plaice	NA	NA	NA	0.86	0.78	0.75	0.95	0.86	0.84, 0.75–0.95
Concentration factor, ^{3}H(total) plaice	18	8.2	6.5	7.3	6.3	8.9	8.5	14	9.6, 6.3–18
^{3}H(total) in Nethertown mussels, Bq kg^{-1} (wet)	NA	NA	85(2)	130(2)	191(2)	210(2)	230(2)	91(2)	
OBT in Nethertown mussels, Bq kg^{-1} (wet)	NA	NA	71(2)	130(2)	182(2)	202(2)	190(2)	78(2)	
Fraction of OBT in ^{3}H(total), mussels	NA	NA	0.84	1.00	0.95	0.96	0.83	0.86	0.91, 0.83–1.0
Concentration factor, ^{3}H(total) mussels	NA	NA	5.1	5.7	9.1	9.1	12	10	8.5, 5.1–12
^{3}H(total) in Nethertown winkles, Bq kg^{-1} (wet)	52 (2)	<25(2)	<25(2)	<25(2)	<33(2)	41(3)	37 (2)	29(2)	
OBT in Nethertown winkles, Bq kg^{-1} (wet)	NA	NA	<26(2)	<25(2)	<46(2)	<27(3)	<32(2)	<25(2)	
Concentration factor, ^{3}H(total) winkles	4.3	<1.5	<1.5	<1.1	<1.6	1.8	1.9	3.2	2.8 (mean of 4 positives), <1.1–4.3

and previous reports in the series). For sea water, plaice and winkles, monitoring for tritium and OBT was reasonably consistently carried out from 2000 to 2006, however mussels were analysed for these determinands only in 2005. The monitoring was carried out on a twice-yearly basis, and in many cases for the biota, measurements were below limits of detection. So to make best use of the data for this site, especially where positive results were obtained, individual half-years' measurements are presented. The dates and times of sea water and biota samples taken are close together being dictated by the sampling campaigns, thus the biota and sea water for a given half-year may be reasonably compared to derive CFs. However, discharges of tritiated water, largely originating from the power station's coolant gas driers, can produce fluctuations in instantaneous concentrations near the outfall (Camplin *et al* 1990) and this may explain some fluctuations. It is a reasonable assumption that the sea water at the sampling point (North Gare) is representative of the tidal area from which biota is sampled.

Table 3. Hartlepool: monitoring data and concentration factors (individual measurements). (Note: NA = not analysed, ND = not determinable due to detection limits.)

Year	2000		2001		2002		2003		2004		2005		2006		Mean and range where relevant
^{3}H(total) discharge, TBq	411		386		411		360		229		242		238		
Measurement no.	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)	
^{3}H(total) in sea water, North Gare, Bq l^{-1}	2.0	4.1	4.9	6.8	1.7	15.6	3.2	6.2	3.1	1.6	2.4	<1.5	5.4	<1.5	
Normalised concentration, Bq l^{-1} per TBq y^{-1}	0.005	0.010	0.013	0.018	0.004	0.038	0.009	0.017	0.014	0.007	0.010	<0.006	0.023	<0.006	<0.013, <0.006–0.038
^{3}H(total) in plaice, Bq kg^{-1} (wet)	NA	<25	<25	<25	<25	<25	<25	27	<25	<25	<25	25	27	<25	
Concentration factor, ^{3}H(total) plaice	NA	<6	<5	<4	<14	<1.6	<8	4.3	<8	<15	<10	>16	5.0	ND	5 based on 2 positives, range <1.6–>16
^{3}H(total) in mussels, Bq kg^{-1} (wet)	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	65	95	NA	NA	
OBT in mussels, Bq kg^{-1} (wet)	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	43	68	NA	NA	
Fraction of OBT in ^{3}H(total), mussels	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	0.66	0.72	NA	NA	
Concentration factor, ^{3}H(total) mussels	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	27	>63	NA	NA	>45, 27–>63
^{3}H(total) in winkles, Bq kg^{-1} (wet)	NA	105	94	48	52	31	33	43	<25	<25	49	<25	<25	<25	
OBT in winkles, Bq kg^{-1} (wet)	NA	95	89	46	48	27	<25	37	<25	<25	31	<25	<25	<25	
Fraction of OBT in ^{3}H(total), winkles	NA	0.90	0.95	0.96	0.92	0.87	<0.7	0.86	ND	ND	0.63	ND	ND	ND	Range <0.7–0.96
Concentration factor, ^{3}H(total) winkles	NA	26	19	7	30	2	10	7	<8	<15	20	ND	<5	ND	15 based on 8 positives, 2–20

Of relevance near Hartlepool is the potential background effect due to discharges from other establishments, mainly Sellafield. During the period of relatively high radiocaesium discharges from Sellafield in 1977–80, the concentration of ^{137}Cs measured in sea water of the North Sea in the Hartlepool region was about 0.2 Bq l^{-1} (e.g. Hunt 1982) compared with an average at St Bees of 29 Bq l^{-1} (Hunt and Kershaw 1990) giving a dilution factor of ∼150. A similar value may be derived from measurements of ^{99}Tc at St Bees (1.1 Bq l^{-1} in 1996 (MAFF and SEPA 1997)) and in the Hartlepool vicinity (7.2 mBq l^{-1} in late 1996 (McCubbin *et al* 2002)). Thus the distant effect of Sellafield discharges during the period 2000–2006, based on ∼20 Bq l^{-1} observed at St Bees, might be expected to contribute $\sim 20/150 = 0.13$ Bq l^{-1}. The contribution due to tritium in fallout, based on measurements in the western English Channel (Maro *et al* 2005), is ∼0.15 Bq l^{-1}. Any effects from the La Hague reprocessing plant would be likely to be very small, the concentrations of tritium being low and the main stream remaining close to the European coast (Bailly du Bois 1996). Thus the measured concentrations of tritium in sea water near Hartlepool, which averaged ∼5 Bq l^{-1} from 2000 to 2004, may mostly (∼95%) be attributed to discharges from Hartlepool itself.

Table 3 shows that annual tritium discharges from Hartlepool have reduced by almost a factor of 2 over the period, essentially during 2002–2004. Concentrations in sea water have similarly reduced, such that two observations in 2005–2006 were below detection limits. The normalised tritium concentrations in sea water fluctuated markedly, by a factor of more than 6, and averaged less than 0.013 Bq l^{-1} per TBq y^{-1}. Total tritium concentrations in plaice were only measured above detection limits in 3 samples out of 13 during the 7 year period. CFs ranged from <1.6 to >16 with a mean of ∼5 for the positively measured values. Thus there is evidence for a small enhancement from the *a priori* value of ∼1. Concentrations of OBT in plaice were measured (Environment Agency *et al* 2007 and previous reports) but are not given in this note as they are all ‘less than’ values and do not provide new information.

Mussels were analysed for tritium and OBT in 2005 only, giving measurable concentrations. There was, as for other sites, a high fraction of OBT in the total tritium, around 0.7. The CF averaged >45 based on two measurements of 27 and >63; the latter value is the result of a ‘less than’ concentration in the local sea water.

Winkles have been regularly measured, but gave ‘not detected’ tritium concentrations in the post-2003 period characterised by lower discharges. Where measurable, there were again relatively high fractions of OBT in total tritium, from <0.7 to 0.96. CFs based on positive measurements were in the range 2 to 20 with an average of 15, again evidencing an enhancement above the *a priori* value of ∼1.

6. Conclusions

First, it is emphasised that enhancement of tritium CFs above the *a priori* factor of ∼1 is reported here on the basis of the analytical methods used, as described in section 2. Further investigation is needed of the effects of adjusting methods, including for the purpose of representing ‘CF’ in the case of tritium.

Enhancement of tritium CFs in marine biota in excess of a factor of ∼1 has previously been reported in connection with liquid discharges from the radiopharmaceutical plant at Cardiff and attributed to components of the effluent containing OBT compounds. Data for tritium in flounders over the period 1997–2006 shows that CFs increased significantly to 2001 by a factor of >20 to ∼7700, then decreased to 2006 by a factor of ∼9. This may be due to a change in chemical form of the tritium over the period and would be consistent with the presence in the effluent up to ∼2001 of organic components more readily taken up by biota, followed by a decrease in such components.

At Sellafield and Hartlepool, although organic materials are used on both sites, there is no operational manufacture of organic complexes as at Cardiff. Yet, on the basis of the methodology used, there does appear to be a relatively small enhancement of CF for marine biota at Sellafield and Hartlepool, deserving investigation. At Sellafield, plaice and mussels appear to have a CF of about 10, whilst positively measured values for winkles exhibit a CF of about 3. The differences might be attributable to the way in which these organisms feed. At Hartlepool, it is difficult to draw firm conclusions because of small numbers of samples with positively measured data. With current data, plaice appear to have an enhanced CF of ∼5 (2 measured samples); mussels appear to have a larger CF than plaice of >45 (2 samples) whilst winkles (in contrast to behaviour near Sellafield) appear to show a CF of 15 (8 samples). More observations are needed especially near Hartlepool and with improved detection limits.

The observed enhancement of CFs for tritium in the marine environment near Sellafield and Hartlepool could be due to formation of organically bound complexes of tritium either in the effluent prior to discharge or in the marine environment due to natural processes. Formation of OBT complexes in effluents is a potential mechanism due to contact of tritium in waste materials with organic materials used on both sites. In the environment, there could be a mechanism for organic complexation, possibly on particulate matter as part of biochemical action (e.g. Turner *et al* 2009) then subsequent transfer within food webs. Characterisation of effluents in terms of OBT would assist in identifying which process, i.e. organic complexation within effluents or in the environment, is important. Measurements on sediments and suspended particulates are needed to gauge the degree of enhancement in pathways involving sediment uptake. Improved detection limits are also needed to provide effective measurements of the degree of enhancement of CFs. However, as discussed above, it is also essential to investigate the effects and uncertainties of different analytical techniques before comparing results of different programmes.

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