SPATIAL AND TEMPORAL VARIATIONS IN CONCENTRATION FACTORS IN NW EUROPEAN SEAS – SECONDARY USE OF MONITORING DATA

by P. J. Kershaw[1], C. A. McMahon[2], A. L. Rudjord[3], C. Smedley[1] and K. S. Leonard[1]

Introduction

Concentration factors (CFs) are a convenient way of describing the distribution of a radionuclide between an organism and the surrounding water (CF: concentration kg^{-1} biota/concentration l^{-1} seawater). CFs are used to simulate the behaviour of radionuclides in transport and radiological assessment models, and generic values have been recommended [1,2] for use when site-specific data are unavailable. In the calculation it is assumed that the system is in equilibrium, which in practise may be difficult to demonstrate. For radiological purposes the edible fraction is most often used. Despite the general acceptance of CFs as useful tools, there have been relatively few studies in recent years designed to obtain reliable values.

Main coastal sampling locations in Irish coastal waters

Discharges of radionuclides to the coastal waters of NW Europe have taken place since the early 1950s. This led to the establishment of national programmes to monitor radionuclide concentrations in the main marine compartments, with particular emphasis on the human food-chain. Inevitably the greatest focus was on the Irish Sea and adjoining waters, due to releases from the BNFL Sellafield reprocessing plant on the coast of NW England. The region was subject to relatively high direct discharges in the mid- to late-1970s, leading to significant contamination of the seabed sediments [3,4] and widespread contamination of the waters of the NE Atlantic [5,6]. Throughout this period there were developments in waste treatment and other operations on site, with consequent changes in the discharge pattern. Most recently, in the mid-1990s, the introduction of the Enhanced Actinide Removal Plant (EARP) led to both further reductions (e.g. ^{239,240}Pu) and significant increases (e.g. ^{99}Tc) in the quantities of radionuclides discharged [7]. However, from the mid-1980s the seabed sediments have become a more important source of ^{137}Cs and plutonium to the water column, and hence to the biota, as a result of remobilisation [8]. It is possible that the bioavailability of remobilised radionuclides will differ from that of direct releases, as a result of chemical and physical changes in the radionuclide associations.

The data used were obtained as part of routine national monitoring programmes in the UK, Ireland and Norway [9,10,11], conducted for radiological assessment and compliance purposes. They represent a variety of sites and species, and in most cases the series were discontinuous. The challenge was to identify the optimum combination of temporal spread, sampling frequency, location, range of species, seawater data and determinands. The aim of the present study was to see whether concentration data collected for radiological monitoring purposes was suitable for deriving CFs, and whether such a study would reveal patterns of regional or temporal variability which might imply changes in the degree of bioavailability.

Geographical variations

Coastal waters of the UK (eastern Irish Sea)

Despite several tens of thousands of available measurements, from the early 1960s to the present, there were surprisingly few data sets which provided consistent sampling protocols for the purposes of estimating reliable CFs. The main limitation was on the availability of contemporaneous water measurements. Also, the high concentration gradients near the source, time-varying release rates and mobility of some fish species make it difficult to argue that equilibrium conditions had been achieved. Three species were selected as representative of the main human food-chain pathways: edible crab – crustacean *Cancer pagurus*; plaice – fin fish *Pleuronectes platessa*; winkle – mollusc *Littorina littorea*, with 4 radionuclides which were either of current or historic concern (^{99}Tc, ^{137}Cs, ^{239,240}Pu, ^{241}Am). Samples were collected from 3 sites and the data were combined: Sellafield, Chapelcross (SW Scotland) and Wylfa (NW Wales).

Table 1: Summary of CFs from coastal waters of the UK

Species	Radio-nuclide	n	Mean	Std. dev.	Range	IAEA TRS-422*
C. pagurus	^{99}Tc	15	210	98	94-410	1000
	^{137}Cs	66	17	12	4-57	50
	^{239,240}Pu	18	113	84	10-330	200
	^{241}Am	18	1.4 x 10^3	1.5 x 10^3	3.6 x 10^1 - 5.3 x 10^3	400
P. platessa	^{99}Tc	15	78	54	17-220	80
	^{137}Cs	66	35	21	9-160	200
	^{239,240}Pu	18	2	1	0.2-4	200
	^{241}Am	18	8	9	0.6-28	200
L. littorea	^{99}Tc	16	3.1 x 10^3	2.0 x 10^3	1.2 x 10^3 - 7.7 x 10^3	500
	^{137}Cs	79	40	28	5-150	60
	^{239,240}Pu	26	1.8 x 10^3	1.6 x 10^3	1.3 x 10^2 - 5.6 x 10^3	3 x 10^3
	^{241}Am	26	8.7 x 10^3	1.1 x 10^4	1.3 x 10^2 - 3.1 x 10^4	1 x 10^3

* IAEA TRS-422 [2] – CFs are non-species-specific, consult explanatory footnotes

Coastal waters of Ireland

^{137}Cs and ^{99}Tc CFs were calculated for eight species, from the western, southern and eastern Irish coasts: brown seaweed - *Fucus vesiculosus*; mussels - molluscs *Mytilus edulis*; prawns/nephrops - crustacean *Nephrops norvegicus*; and five fish species: plaice - *P. platessa*; cod - *Gadus morhua*; whiting - *Merlangius merlangus*; ray - *Raja* sp.; and mackerel - *Scomber scrombus*. Sampling took place from 1982 to the present. The CFs showed considerable variation but there was no statistically-significant difference between values from the north-east, nearest the source, and those in more remote locations. There were few datasets which could be compared with the eastern Irish Sea. This was largely a reflection of differences in the selection of radionuclides and environmental samples considered of relevance to national monitoring programmes. Comparisons were possible for ^{99}Tc and ^{137}Cs in *P. platessa* in UK and Irish waters and there were no significant differences.

Table 2a: Summary of ^{137}Cs CFs from coastal waters of Ireland

Species	n	Mean	Std. dev.	Range	IAEA TRS-422
F. vesiculosus	172	32	12	7 – 110	4 x 10^3
M. edulis	69	108	61	30 – 360	60
N. norvegicus	115	36	19	4 – 142	50
P. platessa	120	48	43	6 – 260	200*
G. morhua	131	86	71	6 – 42	200*
M. merlangus	132	75	76	3 – 420	200*
Raja sp.	106	60	49	6 – 297	200*
S. scrombus	100	30	34	3 - 200	200*

* IAEA TECDOC-422 [2] does not differentiate between different species of fish.

Table 2b: Summary of ^{99}Tc CFs from coastal waters of Ireland

Species	n	Mean	Std. dev.	Range	IAEA TRS-422
F. vesiculosus	58	3x10^4	2x10^4	8x10^3 - 1x10^5	3 x 10^4
M. edulis	23	2x10^3	1x10^3	4x10^2 – 5x10^3	500
N. norvegicus	26	3x10^3	2x10^3	2x10^2 - 1x10^4	1 x 10^3
P. platessa	17	18	32	3 - 100	80*
G. morhua	15	10	26	4 - 90	80*
M. merlangus	17	18	39	2 - 147	80*
Raja sp.	19	21	17	5 – 58	80*
S. scrombus	7	15	7	5 - 24	80*

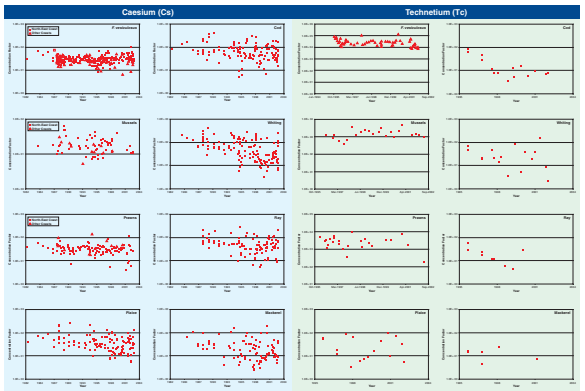
* IAEA TECDOC-422 [2] does not differentiate between different species of fish.

Temporal variations

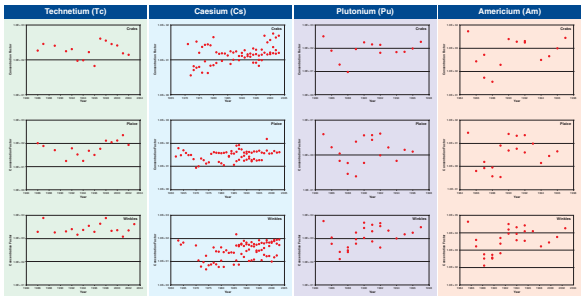
Time-series of CFs were compiled using data from the Irish Sea and Irish coastal waters influenced by the BNFL Sellafield discharges, to look for changes in bioavailability. For nephrops, winkles and *Fucus*, in Irish coastal waters, there were no statistically-significant, time-dependent changes in the ^{137}Cs and ^{99}Tc CFs. For the 5 commercial fin fish species studied there was an apparent decrease in the CF for ^{137}Cs in whiting and ray, and for ^{99}Tc in cod and ray. The CFs estimated for the 12 radionuclide-biota pairs from the eastern Irish Sea showed considerable variability. However, there appeared to be a significant correlation for ^{137}Cs in crab and winkles, i.e. the CF increased in the 1960-2000 sampling period. We do not have an explanation for these apparent changes in Cs bioavailability.

Table 3: Correlation coefficients between the year of sampling and estimated CF, compiled from 3 sites in the eastern Irish Sea (Sellafield, Chapelcross SW Scotland, Wylfa Wales).

species	radionuclide	r	n	> 95% probability
crabs	^{99}Tc	0.04	15	no
C. pagurus	^{137}Cs	0.30	66	yes
	^{239,240}Pu	- 0.11	18	no
	^{241}Am	- 0.11	18	no
plaice	^{99}Tc	0.47	15	no
P. platessa	^{137}Cs	0.21	66	no
	^{239,240}Pu	- 0.04	18	no
	^{241}Am	- 0.06	18	no
winkles	^{99}Tc	- 0.07	16	no
L. littorea	^{137}Cs	0.29	79	yes
	^{239,240}Pu	0.21	26	no
	^{241}Am	0.07	26	no



CFs for ^{137}Cs and ^{99}Tc in seaweed (F. vesiculosus), mussels (M. edulis), nephrops (N. norvegicus), plaice (P. platessa), cod (G. morhua), whiting (M. merlangus) ray (Raja sp.) and mackerel (S. scrombus) from the coastal waters of Ireland



CFs for ^{99}Tc, ^{137}Cs, ^{239,240}Pu and ^{241}Am in crabs (C. pagurus), plaice (P. platessa) and winkles (L. littorea) from the Irish Sea

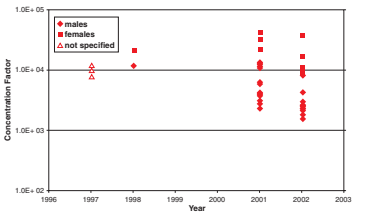
^{99}Tc in lobster (*H. gammarus*), Norwegian waters

There have been a several studies of ^{99}Tc uptake in the lobster (*Homarus gammarus*), as result of the EARP releases and the relatively high bioavailability which led to relatively high ^{99}Tc concentrations. CFs were obtained for a number of biota/sea water samples collected at coastal stations in the southern part of Norway. It was observed that CFs for ^{99}Tc in female lobsters were significantly higher than for males, confirming the findings from the Irish Sea in 1997 of Swift and Nicholson [12]. In the latter study the authors reported considerable variability in concentration, despite trying to control for size, gender and location. This is likely to mask more subtle temporal trends in bioavailability. The mean CF did not vary during the sampling period but the gender difference was persistent.

Table 4: Summary of ^{99}Tc CFs for H. gammarus (lobster) from coastal waters of Norway

Species	n	Mean	Std. dev.	Range	IAEA TRS-422
H. gammarus male	50	6 x 10^3	4 x 10^3	2 x 10^3 - 1 x 10^4	1 x 10^4*
H. gammarus female	12	2 x 10^4	1 x 10^4	1 x 10^4 - 4 x 10^4	1 x 10^4*

* IAEA TRS-422 [2] states a higher value should be applied to lobster



CFs for ^{99}Tc in lobster (H. gammarus) from the coastal waters of Norway

Conclusion

It was possible to make secondary use of radiological monitoring data, of radionuclide concentrations in seawater and biota, to estimate CFs. However, in many cases inconsistencies in sampling protocols over periods of several decades did limit the utility of the data, particularly the lack of contemporaneous seawater concentrations. Differences in the priorities of national programmes meant that regional comparisons were very limited. Where they were possible then no differences were observed. In most cases there were no statistically-significant variations in CFs with time, except for ^{137}Cs in winkles and crabs from the Irish Sea. For the radionuclides studied the observed CFs were similar to the most recent IAEA recommendations with the following exceptions: ^{99}Tc – *C. pagarus*, *M. edulis*, *L. littorea*, *H. gammarus*; ^{137}Cs – *F. vesiculosus*; ^{239,240}Pu – *P. platessa*; ^{241}Am – *C. pagarus*, *P. platessa*. This demonstrates the continuing importance of obtaining site-specific and species-specific data, particularly in situations leading to significant human exposure.

Acknowledgements

The authors wish to acknowledge the efforts of many individuals responsible for carrying out the monitoring programmes. We are grateful for the support of the RPII, NRPA and Defra (UK Department for Environment, Food & Rural Affairs, project C1061). This study was conducted as part of the EU FP5 Remotrans programme (*Processes regulating remobilisation, bioavailability and translocation of radionuclides in marine sediments*).

References

[1] IAEA (1985). Sediment Kds and Concentration Factors for radionuclides in the Marine Environment. (Technical Reports Series Number 247, International Atomic Energy Agency, Vienna, Austria).

[2] IAEA, Sediment Distribution Coefficients and Concentration Factors for Biota in the Marine Environment (Technical Reports Series Number 422, International Atomic Energy Agency, Vienna, Austria, 2004) 95pp.

[3] Kershaw PJ., Denoon D.C. and Woodhead D.S., *J. Environ. Radioactivity* (1999) **44** 191-221.

[4] Condren O.M., Mitchell P.I., Leon Vintro L. and Downes A.B., "Plutonium and radiocaesium in western Irish Sea sediments: origin, bioavailability and ultimate fate", Irish Marine Science Symposium (Royal Irish Academy, Galway, 1995) pp. 567-583.

[5] Kershaw P.J. and Baxter A., *Deep-Sea Res. Part II*, (1995) **42** (6) 1413-1448.

[6] AMAP, Assessment 2002: Radioactivity in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

[7] Leonard K.S., McCubbin D., Brown J., Bonfield R. and Brooks T., *Mar. Poll. Bull.* (1997) **34** (8) 628-636.

[8] Hunt G.J. and Kershaw P.J., Remobilisation of artificial radionuclides from the sediments of the Irish Sea. *J. Radiol. Protection* (1990) **10** (2) 147-151.

[9] RIFE, Radioactivity in food and the environment. (RIFE report no. 3. Ministry of Agriculture, Fisheries & Food (MAFF) and the Scottish Environment Protection Agency (SEPA), London, 1997) 162pp.

[10] Ryan T.P., McMahon C.A., Dowdall A., Fegan M., Sequeira S., Murray M., McKittrick L., Hayden E., Wong J., Colgan P.A., Radioactivity monitoring of the Irish Marine Environment, 2000 and 2001, report RPII-03/3. Dublin: (Radiological Protection Institute of Ireland, Dublin, 2003).

[11] Gilhert, T, Fayn, L, Brungot, A L, Kolstad A K, Lind B, Christensen G C, Strålberg E, Drefvelin J, Rudjord A L. Radioactivity in the Marine Environment 2000 and 2001. Results from the Norwegian National Monitoring Programme StrålevernRapport 2003: 8 Norwegian Radiation Protection Authority, Østerås, 2003

[12] Swift D.J. and Nicholson M.D., *J. Environ. Radioactivity* (2001) **54** 311-326.

[1] The Centre for Environment, Fisheries and Aquaculture Science (CEFAS), Lowestoft Laboratory, Pakefield Road, Lowestoft, Suffolk NR33 0HT, UK. http://www.cefas.co.uk p.j.kershaw@cefas.co.uk

[2] Radiological Protection Institute of Ireland (RPII), Dublin, Ireland [3] Norwegian Radiological Protection Authority (NRPA), Østerås, Norway

© Crown Copyright 2004