Report Title:

Distinguishing between current & historic radioactive discharges to the marine environment – Further work to validate models and quantify uncertainties

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Contract Title: Distinguishing between the impacts of current & historic discharges into the North Sea from UK nuclear sites

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Abstract (100-200 words as desired): This study followed up on previous work to distinguish between the impacts of current and historic radioactive discharges to the observed concentrations of radionuclides in UK marine environmental media. Additional measurement data was compiled for the period 1997 to 2002 detailing location and species. Model predictions of activity concentrations in the marine environment from two marine models were compared with these data and conclusions drawn on model and measurement variabilities. A literature review for local factors that may impact on modelling in the areas considered was carried out and the results applied to the MARINA II model. Conclusions were drawn on the effect of such changes and the implications of such changes on predicted activity concentrations for future discharges as defined in the UK OSPAR Strategy.

Keywords (5 maximum): OSPAR Strategy, Mathematical modelling, Marine discharges, Radiation monitoring

The results of this work will be used in the formulation of Government policy, but views expressed in this report do not necessarily represent Government policy.
Distinguishing between current & historic radioactive discharges to the marine environment – Further work to validate models and quantify uncertainties

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EXECUTIVE SUMMARY

The project followed up on work carried out in a previous study on distinguishing between the contributions of current and historic liquid radioactive discharges to observed activity concentrations on marine environmental media. This study carried out a more detailed validation of selected combinations of radionuclide, location and media. It also improved the quantification of uncertainties in comparisons between measurement and modelled data.

SUMMARY OF STUDY

Measurement data were collated for the combinations of location, media and radionuclide selected based on a review of the previous study. These combinations were those for which there was least agreement between model predictions and measurement data in the previous study.

To enable study of these combinations in greater detail, measurement data were compiled to allow identification of location, species and time of measurements. These data were then used to carry out further validation of the MARINA II model for the period 1997 to 2002. For the CSERAM model, the runs carried out in the previous study were analysed in further detail to review variability temporally and spatially.

A review was carried out of the impact of local factors on the modelling parameters of sediment partition coefficients (K_d) and concentration factors (CF). This review was then used to refine the MARINA II model runs and evaluate the effectiveness of these factors in improving model representation of environmental activity concentrations through further validation.

MAIN FINDINGS

The measurement data were found to display both natural and systematic variability. Natural variability was seen between years for which there was little change in discharge rates. Differences were also seen between species in the same taxonomic group. These varied from several orders of magnitude in the case of 99Tc in crustaceans, to around one order of magnitude for 137Cs in molluscs.

The temporal and spatial variability in the predicted activity concentrations from the CSERAM model was studied and compared with the variability in measurement data. It was found that the variability between measurement results was in most cases greater than that found in the model output due to additional sources of variability not being considered in the model, e.g. sampling, biological and analytical variability.

In the review of the MARINA II model, differences between measurements of individual species in the same taxonomic class (e.g. between measurements of 99Tc in crabs and lobsters) were observed. The MARINA II model does not distinguish between species in taxonomic classes as these are unlikely to be
important for dose calculation purposes. However, when carrying out extensive model validation, it is important to consider the potential model parameters required for different species or alternatively ensure a suitable average activity concentration is taken.

The review of local parameter data for sediment distribution coefficients and concentration factors was used to investigate the effects of such changes on the MARINA II model predictions. The changes to the $K_d$ improved the comparison of model predictions to measurement data for $^{241}$Am. The changes to CFs resulted in improved comparison of predicted activity concentrations in biota to measurement data in all cases, except for $^{241}$Am in molluscs, although in many cases the improvements were marginal. As a result of the changes, all model predictions fall within a factor of ten of the measurement data, with the majority within a factor of three.

**CONCLUSIONS**

In summary, the study showed that the natural variability in activity concentrations measured in the marine environment is present both temporally and spatially. Based on the limited data available for this study, the combination of uncertainty and variability is likely to lead to differences between model predictions and measurements which are mostly within a factor of three and up to a maximum of a factor of ten. It is important to bear these differences in mind when model predictions are compared with measurement values as the general trend of the measurements should be taken into account rather than individual values.

The MARINA II model predicts activity concentrations on an annual average basis and the spatial resolution is of the order of tens of kilometres. For example, in the vicinity of Sellafield, four sampling locations were identified which fall within the same modelling compartment. It is important to ensure that a consistent approach is adopted to select sampling locations consistent with the modelling and with the approaches used to determine baseline values for comparison with the OSPAR strategy.

Differences between individual species within each taxonomic class were seen during the validation exercise. The implication of these differences for the OSPAR strategy is to ensure that a consistent choice of indicator species is made in determining baselines, and for future comparison against baselines. This choice should not necessarily be one species, but could be a defined approach for averaging across selected species consistently throughout the OSPAR region.

The predicted activity concentrations in 2020 using MARINA II are lower than those estimated for 1998 in all cases considered. Changes made to modelling parameters indicate that the contribution from pre-1998 discharges would be slightly greater in 2020 for $^{239/240}$Pu and $^{241}$Am than estimated previously. A conclusion of the previous study that discharges of $^{239/240}$Pu and $^{241}$Am prior to 1998 are likely to remain the major contributor to activity concentrations in 2020, is therefore strengthened. Overall, the findings of this study support the
conclusions made in the previous study regarding ‘close to zero’ and ‘historical levels’.
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<table>
<thead>
<tr>
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<th>References</th>
</tr>
</thead>
</table>
| 10|            | 84
1 INTRODUCTION

The aim of this study was to carry out further work on distinguishing between the contributions of current and historic discharges to measured activity concentrations of radionuclides in UK marine environmental media. This work continues that carried out under a previous contract for DEFRA (Jones et al., 2003). Specifically, the overall objectives of this study are to:

a More accurately validate the models used in the previous study (Jones et al., 2003) to distinguish between the impacts of current and historic radioactive discharges to seas from UK nuclear sites;
b Improve the quantification of uncertainties and the determination of natural variability in activity concentrations in the marine environment, associated with measurements identified in the previous study (Jones et al., 2003).

Two models to represent the activity concentrations in the marine environment were used, as selected in the previous study. The CSERAM model is a high resolution physically-based model of the Irish Sea. Due to the high spatial and temporal resolution, CSERAM is designed to estimate concentrations over hourly timescales and within grid boxes of approximately 13 km². The MARINA II model is a compartmental model designed to represent the environmental transfer of radioactivity in North European waters over annual timescales. Due to the compartmental nature of MARINA II, model predictions are intended to represent activity concentrations averaged over the year and over the area of the compartment.

2 MEASUREMENT DATA

2.1 Compilation of measurement data

The findings of the previous study (Jones et al., 2003) have been used to help determine appropriate data requirements for this study. Plots of observed activity concentrations against model predictions from the previous study have been used to determine areas for further study. These focus mainly on combinations of location, radionuclide and environmental media where the models predicted lower activity concentrations than those observed. However, combinations where models predicted higher activity concentrations than observed were also considered.

The locations selected for further study are shown in Figure 1. These locations relate to compartments of the MARINA II model (European Commission, 2002) identified as a, e, f and i in the figure (Sellafield, Wylfa, Northern Ireland and Dounreay indicator locations). Principal focus has been made on measurements for the period 1997 to 2002 for $^{137}$Cs, $^{239/240}$Pu and $^{241}$Am, although consideration
was also given to $^3$H and $^{99}$Tc. Details of specific combinations of location, media and radionuclide identified from analysis of the previous study are provided in Table 1.

Additionally, other species were considered within each taxonomic class, where data are available, e.g. cockles and mussels for the mollusc class where the previous study only considered winkles. During monitoring, sufficient samples are collected to enable analysis and provide representative results. Typically, this may involve about 5 individual fish, 10 individual crabs and lobsters, and 100 individual molluscs. For seafood samples, it is the edible fraction (i.e. flesh) rather than the whole sample (i.e. including shell and bones) that is analysed.

For information, the typical mobility of molluscs and crustaceans are indicated below:

**Molluscs:**
- Mussels: 1 m
- Cockles: 1 km

**Crustaceans:**
- Lobsters: 10+ km
- Crabs: 100+ km

### Table 1 Combinations of location, media and radionuclide identified in the previous study. Those for additional review are shown in italics

<table>
<thead>
<tr>
<th>Location</th>
<th>Location</th>
<th>Radionuclides for primary consideration</th>
<th>Radionuclides for secondary consideration</th>
</tr>
</thead>
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<tr>
<td></td>
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<td>$^{239/240}$Pu</td>
<td>$^{241}$Am</td>
</tr>
<tr>
<td>Sellafield</td>
<td>seawater</td>
<td>molluscs</td>
<td>seawater</td>
</tr>
<tr>
<td></td>
<td>molluscs sediment</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>fish</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>crustaceans seaweed</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Northern Ireland</td>
<td>fish</td>
<td>molluscs</td>
<td>seawater</td>
</tr>
<tr>
<td></td>
<td>molluscs sediment</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Wylfa</td>
<td>seawater</td>
<td>molluscs</td>
<td>seawater</td>
</tr>
<tr>
<td></td>
<td>fish</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>molluscs</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dounreay</td>
<td>fish</td>
<td>molluscs</td>
<td>seawater</td>
</tr>
<tr>
<td></td>
<td>molluscs seawater</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Data have been compiled for the Sellafield, Northern Ireland, Wylfa and Dounreay sites based on measurements reported for 1997 – 2002 in the RIFE and Environment Agency series of reports, e.g. (FSA and SEPA, 2002) and (Environment Agency, 2003). These update data for the selected samples and locations reported in the previous study (Jones et al., 2003) and are shown in Table 2. In addition, data have been compiled for materials not reported at these locations in the previous study. These are presented in Section 2.2 which discusses variability in measured environmental activity concentrations.

Data have also been compiled for the Sellafield, Wylfa and Dounreay sites based on measurements reported by operators in their 2001 and 2002 discharge and monitoring reports e.g. (BNFL, 2003), (G Jenkins et al., 2003). These data have been used to assist in the validation of MARINA II (Section 6).

### Table 2 Activity concentration data for samples for additional review

<table>
<thead>
<tr>
<th></th>
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</thead>
<tbody>
<tr>
<td><strong>Sellafield (‘Coastal Area’) Crabs (Bq/kg (wet))</strong></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>$^{99}$Tc</td>
<td>70</td>
<td>51</td>
<td>53</td>
<td>35</td>
<td>60</td>
<td>47</td>
</tr>
<tr>
<td><strong>Sellafield (Nethertown) Winkles (Bq/kg (wet))</strong></td>
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<td></td>
<td></td>
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<td>17</td>
<td>15</td>
<td>15</td>
<td>15</td>
<td>14</td>
<td>13</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>31</td>
<td>31</td>
<td>30</td>
<td>32</td>
<td>38</td>
<td>33</td>
</tr>
<tr>
<td><strong>Sellafield (Maryport) Mud (Bq/kg (dry))</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>680</td>
<td>620</td>
<td>580</td>
<td>204</td>
<td>143</td>
<td>85</td>
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<tr>
<td><strong>Sellafield (St Bees) Seawater (Bq/l)</strong></td>
<td></td>
<td></td>
<td></td>
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<td></td>
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</tr>
<tr>
<td>$^{137}$Cs</td>
<td>0.23</td>
<td>0.17</td>
<td>0.15</td>
<td>0.19</td>
<td>0.13</td>
<td>0.13</td>
</tr>
<tr>
<td><strong>Northern Ireland Winkles (Bq/kg (wet))</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{239/240}$Pu</td>
<td>0.2</td>
<td>0.16</td>
<td>0.16</td>
<td>0.16</td>
<td>0.15</td>
<td>0.16</td>
</tr>
<tr>
<td><strong>Northern Ireland (Ardglass) <em>Fucus vesiculosus</em> (Bq/kg (wet))</strong></td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>$^{99}$Tc</td>
<td>620</td>
<td>400</td>
<td>500</td>
<td>1100</td>
<td>290</td>
<td>590</td>
</tr>
<tr>
<td><strong>Wylfa (‘Pipeline’) Crabs (Bq/kg (wet))</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{99}$Tc</td>
<td>4.8</td>
<td>5.0</td>
<td>7.9</td>
<td>9.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Wylfa (Holyhead) Seawater (Bq/l)</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>0.017</td>
<td>0.019</td>
<td>0.03</td>
<td>0.04</td>
<td>0.01</td>
<td>0.01</td>
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<tr>
<td><strong>Dounreay (Sandside Bay) Winkles (Bq/kg (wet))</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{239/240}$Pu</td>
<td>0.21</td>
<td>0.21</td>
<td>0.11</td>
<td>0.093</td>
<td>0.099</td>
<td>0.10</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>0.12</td>
<td>0.15</td>
<td>0.11</td>
<td>0.089</td>
<td>0.078</td>
<td>0.1</td>
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<tr>
<td><strong>Dounreay (Sandside Bay) Seawater (Bq/l)</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{3}$H</td>
<td>&lt;1.6</td>
<td>2.4</td>
<td>&lt;1.4</td>
<td>&lt;1.3</td>
<td>&lt;1.7</td>
<td>&lt;1</td>
</tr>
</tbody>
</table>
Figure 1 Locations selected for review of measurement and modelling data
Where possible, the data in this report are presented so that the species, type and sampling location of the sample are indicated. Maps of the sample locations are provided in Figure 2 to Figure 5. Two additional sample areas are referred to in subsequent sections. These are the ‘Sellafield Coastal Area’ which extends 15 km to the north and to the south of the Sellafield site from St. Bees Head to Selker and from the coast to 11 km offshore. The second area is the ‘Sellafield Offshore Area’ which is a 1.8 km by 3.6 km rectangle located around 5 km south of the Sellafield pipelines with the long side parallel to the shore.

**Figure 2 Sampling and discharge locations in the eastern Irish Sea**
Figure 3 Sampling and discharge locations on the north Welsh coast

Figure 4 Sampling and discharge locations on the north Scottish coast
2.2 Variability in measured concentration data

This section considers the variability in measured concentration data for the selected sample types and radionuclides (Table 1) and includes consideration of additional monitoring data. Variability is separately considered in terms of species, location and time. This analysis is based only on the RIFE series of data since the use of UKAEA and BNFL data may introduce further variability due to differences in sampling and analytical techniques.

2.2.1 Difference by species

In the previous study (Jones et al., 2003), samples were considered according to the taxonomic groups: fish, crustaceans and molluscs. Table 3 to Table 6 give examples of data for different species within the taxonomic groups which were collected at a similar location. The data also include standard deviations of the results used in calculating the annual mean values and the number of observations within the year. Where only one observation is noted, this may be for a single sample, or it may be for the analysis of a bulk of several samples taken during the year.

Figure 5 Sampling locations in the western Irish Sea and selected discharge locations
Table 3 shows activity concentrations of $^{99}$Tc in crabs and lobsters from Ravenglass. In a given year, values for lobsters are generally more than two orders of magnitude greater than for crabs.

For Sellafield, Table 4 shows measured activity concentrations of $^{137}$Cs in winkles and mussels from Nethertown; in winkles, mussels and cockles from Ravenglass; and winkles, mussels and limpets from St Bees. Broadly, the activity concentrations in winkles are about three-times the activity concentrations in mussels at each location, and activity concentrations for limpets and cockles lie in between those for winkles and mussels at the respective locations.

Table 5 shows measured activity concentrations of $^{241}$Am in winkles and mussels from Nethertown; in mussels and cockles from Ravenglass; and winkles, mussels and limpets from St Bees. The variability is less than for $^{137}$Cs. In a given year, activity concentrations in winkles are generally less than twice those in mussels. Activity concentrations in cockles are about twice those in mussels. Unlike the observations for $^{137}$Cs, activity concentrations of $^{241}$Am are slightly higher in mussels than limpets.

### Table 3  Measured activity concentrations of technetium-99 in crustaceans near Sellafield, 1997-2002

<table>
<thead>
<tr>
<th>Location</th>
<th>Species</th>
<th>Year</th>
<th>Average</th>
<th>Standard deviation</th>
<th>No. measurements</th>
</tr>
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<tr>
<td></td>
<td>Crabs</td>
<td>1997</td>
<td>44</td>
<td>23</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1998</td>
<td>28</td>
<td>6.5</td>
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<td>1999</td>
<td>38</td>
<td>5.5</td>
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<td>23</td>
<td>9.7</td>
<td>4</td>
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<td>2001</td>
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<td>2002</td>
<td>24</td>
<td>7.0</td>
<td>4</td>
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<tr>
<td></td>
<td>Lobsters</td>
<td>1997</td>
<td>6218</td>
<td>3262</td>
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<td></td>
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<tr>
<td></td>
<td></td>
<td>2002</td>
<td>2605</td>
<td>374</td>
<td>4</td>
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</tbody>
</table>

Note: Activity concentrations are Bq/kg (wet)

For Wylfa, Table 6 shows activity concentrations of $^{99}$Tc in crabs and lobsters. Comparisons show a similar trend to Sellafield, though the values for lobsters are generally between one and two orders of magnitude greater than for crabs.
### Table 4: Measurements of caesium-137 in molluscs near Sellafield, 1997-2002

<table>
<thead>
<tr>
<th>Location</th>
<th>Species</th>
<th>Year</th>
<th>Average</th>
<th>Standard deviation</th>
<th>No. measurements</th>
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<tr>
<td>Netherton</td>
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<td>17</td>
<td>11</td>
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<td></td>
<td>2002</td>
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<td>Ravenglass</td>
<td>Winkles</td>
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Note: Activity concentrations are Bq/kg (wet)
Table 6  Measured activity concentrations of technetium-99 in crustaceans near Wylfa, 1997-2002

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Notes: Activity concentrations are Bq/kg (wet); NS = no sample

2.2.2  Difference by location

Table 7 to Table 14 give examples of measured activity concentrations where comparisons can be drawn between results for the same species collected at different locations within the general areas considered. Results given in Table 4 to Table 6 are also considered in this comparison though the results are not reproduced in Table 7 to Table 14. As previously, the data include annual mean activity concentrations, standard deviations and numbers of observations used to derive the means.

For Sellafield, (Table 7 to Table 10), the location ‘Sellafield Coastal Area’ includes samples which have been collected from an area which extends 15 km to the north and to the south of Sellafield, from St Bees Head to Selker and 11 km offshore. The location ‘Sellafield Offshore Area’ consists of a rectangle 1.8 km wide by 3.6 km long situated south of the pipelines with the long side parallel to the shore line; it averages about 5 km from the pipeline outlet.

For $^{137}$Cs in molluscs, the difference between the highest and lowest observed activity concentrations in winkles in the area in a given year ranged by a factor of about 2.5. For mussels, the range was about a factor of 2, and for limpets and whelks, it was less than 1.5.

For $^{241}$Am in molluscs, the difference between the highest and lowest observed activity concentrations in winkles in the area in a given year ranged by a factor of about 3. For mussels and whelks, the range was about a factor of 2, and for limpets, it was less than 1.5.

Taking the data for molluscs as a whole however, the difference between the highest and lowest observed activity concentrations of $^{137}$Cs in the area in a given year ranged by about an order of magnitude. For $^{241}$Am, the range was of the order of 5 to 10-times.
For $^{99}$Tc in crustaceans, the difference between the highest and lowest observed activity concentrations in crabs from the area in a given year ranged by a factor of about 2. The range was similar for lobsters, and about 2.5 for nephrops.

Taking the data for crustaceans as a whole however, the difference between the highest and lowest observed activity concentrations of $^{99}$Tc from the area in a given year ranged by more than two orders of magnitude (nearly 400 times in 1997). This was mainly due to the much higher activity concentrations observed in lobsters.

For sediments, activity concentrations of $^{137}$Cs for a number of locations and sediment types are shown in Table 10. Activity concentrations are affected not only by the location, but also the sediment type, the finer sediments (e.g. mud) generally concentrating higher amounts of radioactivity than the coarser sediments (e.g. sand). For sediments in the area, the difference between the highest and lowest observed activity concentrations of $^{137}$Cs in mud in a given year ranged by factors of about 2 between locations. For $^{137}$Cs in sand, there was very little difference between locations. Taking the data for sediments as a whole however, the difference between the highest and lowest observed activity concentrations of $^{137}$Cs in a year ranged by factors of between 5 and 8, mainly due to the higher activity concentrations in mud than in sand.

Figure 6 shows activity concentrations of $^{137}$Cs in sediments at a number of locations in the vicinity of Sellafield in 2001. Higher concentrations were observed around Hambleton and Skippool Creek in the Wyre Estuary and Lytham Yacht Club in the Ribble Estuary. The sediments in the Wyre and Ribble Estuaries are fine-grained muds and silts. These sediment types have a greater affinity for radionuclides and hence lead to higher activity concentrations in these areas. Overall, the figure shows that the type of sediment has a stronger influence than the effect of distance on the measured activity concentrations.

For Northern Ireland, Table 11 gives activity concentrations of $^{99}$Tc in seaweed at 3 locations. Taking seaweeds as a whole, the difference between the highest and lowest observed activity concentrations of $^{99}$Tc in the area in a given year have ranged from 7 to 60 times.

For Wylfa, Table 12 gives activity concentrations of $^{137}$Cs in seawater at locations to the east and west of the site. Concentrations at Llandudno and Prestatyn were generally similar within a given year, with those at Holyhead generally lower. Overall for this area, the difference between the highest and lowest observed activity concentrations in a given year ranged by a factor of about 3.

For Dounreay, Table 13 presents activity concentrations of $^{239/240}$Pu in molluscs at 3 locations. The difference between the highest and lowest observed activity concentrations in winkles from the overall area in a given year have generally been no more than a factor of 2. This was also the case for all molluscs. For $^{241}$Am in molluscs (Table 14), the differences have been more variable, ranging from 1 to 10 times for winkles, and from 1 to 20 times for all molluscs.
## Table 7 Measurements of caesium-137 in Sellafield molluscs, 1997-2002

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Note: Activity concentrations are Bq/kg (wet)
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**Notes:** Activity concentrations are Bq/kg (wet); NS = no sample.
Table 9  Measurements of technetium-99 in Sellafield crustaceans, 1997-2002

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Notes: Activity concentrations are Bq/kg (wet); NS = no sample
### Table 10  Measurements of caesium-137 in Sellafield sediments, 1997-2002

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Note: Activity concentrations are Bq/kg (dry)
Figure 6  Mean activity concentrations of caesium-137 and cobalt-60 in silty sediment with distance from Sellafield in 2001 (Source (Environment Agency, 2003))

Table 11  Measurements of technetium-99 in Northern Ireland seaweeds, 1997-2002

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<td>Strangford Lough</td>
<td>Rhodymenia spp.</td>
<td>1997</td>
<td>104</td>
<td>41</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1998</td>
<td>48</td>
<td>20</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1999</td>
<td>66</td>
<td>32</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2000</td>
<td>19</td>
<td>20</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2001</td>
<td>69</td>
<td>83</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2002</td>
<td>24</td>
<td>6.9</td>
<td>2</td>
</tr>
<tr>
<td>Ardglass</td>
<td>Fucus vesiculosus</td>
<td>1997</td>
<td>617</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1998</td>
<td>397</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1999</td>
<td>495</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2000</td>
<td>1100</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2001</td>
<td>289</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2002</td>
<td>586</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>Carlingford Lough</td>
<td>Fucus spp.</td>
<td>1997</td>
<td>635</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1998</td>
<td>764</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1999</td>
<td>711</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2000</td>
<td>601</td>
<td>1</td>
<td>1</td>
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<tr>
<td></td>
<td></td>
<td>2001</td>
<td>483</td>
<td>1</td>
<td>1</td>
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<tr>
<td></td>
<td></td>
<td>2002</td>
<td>442</td>
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<td>1</td>
</tr>
</tbody>
</table>

Note: Activity concentrations are Bq/kg (wet)
### Table 12: Measurements of caesium-137 in Wylfa seawater, 1997-2002

<table>
<thead>
<tr>
<th>Location</th>
<th>Media</th>
<th>Year</th>
<th>Average</th>
<th>Standard deviation</th>
<th>No. measurements</th>
</tr>
</thead>
<tbody>
<tr>
<td>Llandudno</td>
<td>Seawater</td>
<td>1997</td>
<td>0.07</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1998</td>
<td>0.05</td>
<td></td>
<td>1</td>
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<td></td>
<td></td>
<td>1999</td>
<td>0.06</td>
<td></td>
<td>1</td>
</tr>
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<td></td>
<td></td>
<td>2000</td>
<td>0.07</td>
<td></td>
<td>1</td>
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<td></td>
<td></td>
<td>2001</td>
<td>0.02</td>
<td></td>
<td>1</td>
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<td></td>
<td></td>
<td>2002</td>
<td>0.03</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Prestatyn</td>
<td>Seawater</td>
<td>1997</td>
<td>0.08</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1998</td>
<td>0.06</td>
<td></td>
<td>1</td>
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<td></td>
<td></td>
<td>1999</td>
<td>0.08</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2000</td>
<td>0.06</td>
<td></td>
<td>1</td>
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<td></td>
<td></td>
<td>2001</td>
<td>0.03</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2002</td>
<td>0.03</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Holyhead</td>
<td>Seawater</td>
<td>1997</td>
<td>0.017</td>
<td></td>
<td>1</td>
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<tr>
<td></td>
<td></td>
<td>1998</td>
<td>0.019</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1999</td>
<td>0.03</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2000</td>
<td>0.04</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2001</td>
<td>0.01</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2002</td>
<td>0.01</td>
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<td>1</td>
</tr>
</tbody>
</table>

Note: Activity concentrations are Bq/l

### Table 13: Measurements of plutonium-239/240 in Dounreay molluscs, 1997-2002

<table>
<thead>
<tr>
<th>Location</th>
<th>Species</th>
<th>Year</th>
<th>Average</th>
<th>Standard deviation</th>
<th>No. measurements</th>
</tr>
</thead>
<tbody>
<tr>
<td>Brims Ness</td>
<td>Winkles</td>
<td>1997</td>
<td>0.226</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1998</td>
<td>0.193</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1999</td>
<td>0.099</td>
<td></td>
<td>1</td>
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<td></td>
<td></td>
<td>2000</td>
<td>0.189</td>
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<td></td>
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<td>2001</td>
<td>0.161</td>
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<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2002</td>
<td>0.052</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Sandside Bay</td>
<td>Winkles</td>
<td>1997</td>
<td>0.213</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1998</td>
<td>0.205</td>
<td></td>
<td>1</td>
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<td></td>
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<td>1999</td>
<td>0.106</td>
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<td>2001</td>
<td>0.099</td>
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</tr>
<tr>
<td></td>
<td></td>
<td>2002</td>
<td>0.100</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Echnaloch Bay</td>
<td>Mussels</td>
<td>2000</td>
<td>0.083</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2001</td>
<td>0.079</td>
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<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2002</td>
<td>0.051</td>
<td></td>
<td>1</td>
</tr>
</tbody>
</table>

Note: Activity concentrations are Bq/kg (wet)
Table 14  Measurements of americium-241 in Dounreay molluscs, 1997-2002

<table>
<thead>
<tr>
<th>Location</th>
<th>Species</th>
<th>Year</th>
<th>Average</th>
<th>Standard deviation</th>
<th>No. measurements</th>
</tr>
</thead>
<tbody>
<tr>
<td>Brims Ness</td>
<td>Winkles</td>
<td>1997</td>
<td>0.249</td>
<td>1</td>
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<tr>
<td></td>
<td></td>
<td>1998</td>
<td>0.192</td>
<td>1</td>
<td></td>
</tr>
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<td>0.103</td>
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<td></td>
<td></td>
<td>2000</td>
<td>0.794</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>2001</td>
<td>0.083</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>2002</td>
<td>0.053</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>Sandside Bay</td>
<td>Winkles</td>
<td>1997</td>
<td>0.121</td>
<td>1</td>
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<tr>
<td></td>
<td></td>
<td>1998</td>
<td>0.152</td>
<td>1</td>
<td></td>
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<tr>
<td></td>
<td></td>
<td>1999</td>
<td>0.115</td>
<td>1</td>
<td></td>
</tr>
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<td></td>
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<td>2000</td>
<td>0.089</td>
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</tr>
<tr>
<td></td>
<td></td>
<td>2001</td>
<td>0.078</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>2002</td>
<td>0.097</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>Echnaloch Bay</td>
<td>Mussels</td>
<td>2000</td>
<td>0.041</td>
<td>1</td>
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</tr>
<tr>
<td></td>
<td></td>
<td>2001</td>
<td>0.038</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>2002</td>
<td>0.018</td>
<td>1</td>
<td></td>
</tr>
</tbody>
</table>

Note: Activity concentrations are Bq/kg (wet)

2.2.3  Difference by time
The variability in results within a year can be seen from the values of the standard deviations in Table 3 to Table 14. Additional examples are provided in Figure 7 and Figure 8. Figure 7 shows quarterly measurements of $^{99}$Tc in crabs from the Sellafield Coastal Area whereas Figure 8 shows quarterly measurements in Ravenglass lobsters. These enable further interpretation of the results in Table 3 and Table 9.

Figure 7 Activity concentrations of $^{99}$Tc measured in crabs from Sellafield Coastal Area
Figure 8 Activity concentrations of $^{99}$Tc measured in lobsters from Ravenglass

3 DISCHARGE DATA

Annual discharge data for the relevant radionuclides in 2001 and 2002 have been compiled for the sites whose discharges are likely to affect activity concentrations in the specified regions of the UK coast using the RIFE series of reports e.g. (FSA and SEPA, 2002). These data supplement that compiled for the previous study (Jones et al., 2003). Where discharge data are aggregated, e.g. total alpha, a breakdown of specific radionuclides has been obtained directly from the operator. Discharge data have been compiled for the following sites:

a. Dounreay
b. Hartlepool
c. Heysham
d. Hunterston
e. Sellafield
f. Torness
g. Wylfa
h. La Hague (for tritium only)
4 REVIEW OF DATA ON RADIONUCLIDE BEHAVIOUR IN THE MARINE ENVIRONMENT

The sediment distribution coefficient ($K_d$) is a parameter that provides an equilibrium measure of the affinity a radionuclide has for sediment. It is the ratio of the radionuclide activity per unit mass of dry sediment (Bq kg$^{-1}$) to the radionuclide activity per unit volume of seawater (Bq l$^{-1}$). Concentration factors (CFs) are assumed to be an equilibrium measure of the ratio of activity per unit mass of biota (Bq kg$^{-1}$) and unit volume of seawater (Bq l$^{-1}$). When carrying out radiological assessments, it is standard practice to apply the generic parameter values compiled by IAEA (IAEA, 1985). However, it is recognised that in some cases, it is important to use local factors as these may have an influence on the behaviour of radionuclides in the local marine environment. The purpose of this review was to find site-specific $K_d$ values for sediments and CF values for biota that could be used in the calculation of activity concentrations in the Irish Sea (Sellafield, Wylfa, Northern Ireland) and Dounreay areas. These site-specific data were investigated to determine their influence on measured activity concentrations and therefore their appropriateness for implementation in marine models such as CSERAM (Aldridge et al., 2003) and MARINA II (European Commission, 2002).

A literature review was carried out and relevant references were selected. Data obtained from these papers are presented and the data selected for use in this project are presented. Where compartment numbers are described, these refer to the MARINA II model.

4.1 Relevant papers and site-specific parameter values


This paper gives CFs in the Irish Sea from samples taken in 1995 following the initial operation of EARP, mainly from (Busby et al., 1997). Also given are CFs in biota collected from Onsøy, Fredsikstad/Outer Oslofjord, November 1997 which were not used in this study as they are outside the area of interest. A summary of the data from this paper is presented in Table 15.
**Table 15 CFs for selected biota in Irish Sea, 1995 (Bq kg\(^{-1}\) [wet weight] per Bq l\(^{-1}\) [seawater])**

<table>
<thead>
<tr>
<th>Organism</th>
<th>Location</th>
<th>CF</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Seaweed†</td>
<td>Off Sellafield§</td>
<td>(1.19±0.41) 10(^5)</td>
<td>Smith et al 1997</td>
</tr>
<tr>
<td>Mussels</td>
<td>Off Sellafield§</td>
<td>5.0 10(^2)</td>
<td>McCartney and Rajendran (1997)</td>
</tr>
<tr>
<td>Lobster (abdomen muscle*)</td>
<td>Off Sellafield, 1995</td>
<td>7.2 10(^2)</td>
<td>Busby et al 1997</td>
</tr>
<tr>
<td>Lobster (green gland*)</td>
<td>Off Sellafield, 1995</td>
<td>6.5 10(^4)</td>
<td>Busby et al 1997</td>
</tr>
<tr>
<td>Norwegian lobster (abdominal muscle*)</td>
<td>Off Sellafield, 1995</td>
<td>9.7 10(^2)</td>
<td>Busby et al 1997</td>
</tr>
<tr>
<td>Norwegian lobster (hepatopancreas*)</td>
<td>Off Sellafield, 1995</td>
<td>2.3 10(^1)</td>
<td>Busby et al 1997</td>
</tr>
<tr>
<td>Crab (white body meat*)</td>
<td>Off Sellafield, 1995</td>
<td>1.5 10(^2)</td>
<td>Busby et al 1997</td>
</tr>
<tr>
<td>Crab (heart*)</td>
<td>Off Sellafield, 1995</td>
<td>5.4 10(^2)</td>
<td>Busby et al 1997</td>
</tr>
<tr>
<td>Crab (hepatopancreas*)</td>
<td>Off Sellafield, 1995</td>
<td>1.6 10(^2)</td>
<td>Busby et al 1997</td>
</tr>
<tr>
<td>Winkles</td>
<td>St Bees, 1995</td>
<td>3.9 10(^2)</td>
<td>Busby et al 1997</td>
</tr>
<tr>
<td>Mussels</td>
<td>St Bees, 1995</td>
<td>4.2 10(^2)</td>
<td>Busby et al 1997</td>
</tr>
<tr>
<td>Winkles</td>
<td>Ravenglass</td>
<td>6.4 10(^2)</td>
<td>Busby et al 1997</td>
</tr>
<tr>
<td>Mussels</td>
<td>Ravenglass</td>
<td>3.3 10(^2)</td>
<td>Busby et al 1997</td>
</tr>
<tr>
<td>Whelks</td>
<td>Off Sellafield</td>
<td>1.5 10(^2)</td>
<td>Busby et al 1997</td>
</tr>
</tbody>
</table>

† CF dry weight  
§ Assumed  
*CF to body part


This paper gives activity concentrations in filtered seawater and seaweed from samples taken off Balbriggan on the east coast of Ireland and the western Irish Sea. CFs for \(^{99}\)Tc in *Fucus vesiculosus* were calculated using the filtered seawater and seaweed activity concentrations sampled from Balbriggan. Mean values are given for May 1995 – December 1998.

The paper also gives activity concentrations in cod, plaice, whiting, ray, herring and mackerel sampled from Clogherhead and Howth on the east coast of Ireland. Using the mean annual \(^{99}\)Tc activity concentration in seawater from the western Irish Sea and cod, plaice, whiting and ray landed in Clogherhead and Howth, a concentration factor was derived for 1996 – 1998 (Bq kg\(^{-1}\) wet weight, edible flesh: Bq l\(^{-1}\) seawater). Lobsters were sampled from Carlingford, Dundalk Bay, Lough Shinny, Howth and Saltie Islands and activity concentrations given. A mean CF for technetium was calculated for 1997-1998 in lobster tail muscle. Prawns were sampled from Clogherhead, Howth, Castletownbere and Galway from 1996 – 1998 and activity concentrations are given. A mean CF value for technetium for 1996-1998 is estimated. Mussels were also sampled from Carlingford, Bantry, Galway and Killybegs and the activity concentrations are given. A mean CF value is given. Oysters were sampled from Carlingford and activity concentrations given. A mean CF for technetium in edible parts of
oysters is estimated. A summary of the data from this paper is presented in Table 16.

### Table 16  CFs for selected biota in Irish Sea for various years (Bq kg\(^{-1}\) [wet weight] per Bq l\(^{-1}\) [seawater]). Source: Smith et al., 2001

<table>
<thead>
<tr>
<th>Organism</th>
<th>Date of sampling</th>
<th>Location</th>
<th>CF</th>
</tr>
</thead>
<tbody>
<tr>
<td><em>Fucus vesiculosus</em></td>
<td>May 95 – Dec 98</td>
<td>Balbriggan</td>
<td>((1.32 \pm 0.46) \times 10^5)(^{(1)})</td>
</tr>
<tr>
<td>Fish (cod, plaice, whiting and ray)</td>
<td>1996 – 1998</td>
<td>Clogherhead, Howth</td>
<td>(1.25 \times 10^1 \pm 4.90)(^{(2)})</td>
</tr>
<tr>
<td>Lobster (tail muscle)</td>
<td>1997 – 1998</td>
<td>West coast of Ireland</td>
<td>((6.85 \pm 1.30) \times 10^3)</td>
</tr>
<tr>
<td>Prawns (edible portion)</td>
<td>1996 – 1998</td>
<td>West coast of Ireland</td>
<td>((2.80 \pm 0.90) \times 10^3)</td>
</tr>
<tr>
<td>Mussels (edible portion)</td>
<td>1996 – 1998</td>
<td>Carlingford</td>
<td>((9.7 \pm 5.7) \times 10^2)</td>
</tr>
<tr>
<td>Oysters (edible portion)</td>
<td>1996 – 1998</td>
<td>Carlingford</td>
<td>((2.8 \pm 1.2) \times 10^2)</td>
</tr>
</tbody>
</table>

1. Filtered seawater and seaweed activity concentrations.
2. Bq/kg wet wt. Edible flesh: Bq/l.

*The accumulation and retention of \(^{95m}\)Tc by the Norway lobster, *Nephrops norvegicus* L.*  Swift, D. *Journal of Environmental Radioactivity, 57,* (2), p. 139-149, 2001.*

This paper gives the results of laboratory experiments on *Nephrops norvegicus* L (Norway lobster) obtained from commercial landings at North Shields. Results show a CF (whole body) of about \(2 \times 10^3\) Bq kg\(^{-1}\) per Bq l\(^{-1}\) for the tail muscle (edible fraction). For *Nephrops* landed at Whitehaven and activity concentrations of \(^{99}\)Tc in seawater collected at St Bees (using RIFE reports) over 1996 – 1999 gives a mean CF of about \(1.4 \times 10^3\) Bq kg\(^{-1}\) per Bq l\(^{-1}\). A summary of the data from this paper is presented in Table 17.

### Table 17  CF for lobster sampled in Whitehaven (Bq kg\(^{-1}\) [wet weight] per Bq l\(^{-1}\) [seawater]). Source: Swift (2001)

<table>
<thead>
<tr>
<th>Organism</th>
<th>Date of sampling</th>
<th>Location</th>
<th>CF</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nephrops</td>
<td>1996 – 2000</td>
<td>Whitehaven</td>
<td>(1.4 \times 10^3) (^{(1)})</td>
</tr>
</tbody>
</table>

1. Estimated.

This paper contains two tables of mean K\textsubscript{ds} for plutonium, americium, curium and neptunium from samples taken during the period 1977 – 1983. Samples were taken from the surface waters within the Irish Sea, before entering the Irish Sea and after leaving the Irish Sea. The results presented, however, were not used in this study. The paper indicates that there are apparent pitfalls due to adsorption/desorption behaviour in the marine environment and distribution of radionuclides between solid and liquid phases. Also, although K\textsubscript{ds} imply equilibrium has been attained, this may not be the case due to non-constant discharge rates, the chemical forms present and associations made by radionuclides.


This paper gives CFs for various radionuclides in the vicinity of Sellafield in the Irish Sea for fish, crustacea and molluscs and K\textsubscript{ds} for several locations on the Cumbrian coast. A summary of the data from this paper is presented in Table 18 and Table 19.

### Table 18  CFs for fish, crustacea and molluscs from Cumbrian Coast, Irish Sea (Bq kg\textsuperscript{-1} edible fraction per Bq l\textsuperscript{-1} seawater). Source: Goshawk and Clarke (1999)

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Fish</th>
<th>Crustacea</th>
<th>Molluscs</th>
</tr>
</thead>
<tbody>
<tr>
<td>\textsuperscript{3}H</td>
<td>1 10\textsuperscript{0}</td>
<td>1 10\textsuperscript{0}</td>
<td>1 10\textsuperscript{0}</td>
</tr>
<tr>
<td>\textsuperscript{14}C</td>
<td>3.0 10\textsuperscript{3}</td>
<td>6.0 10\textsuperscript{3(1)}</td>
<td>5.0 10\textsuperscript{3(2)}</td>
</tr>
<tr>
<td>\textsuperscript{99}Tc</td>
<td>3.0 10\textsuperscript{1}</td>
<td>9.0 10\textsuperscript{3(1)}</td>
<td>6.0 10\textsuperscript{3(2)}</td>
</tr>
<tr>
<td>\textsuperscript{137}Cs</td>
<td>9.0 10\textsuperscript{1}</td>
<td>3.0 10\textsuperscript{1}</td>
<td>5.0 10\textsuperscript{1(2)}</td>
</tr>
<tr>
<td>\textsuperscript{239/240}Pu</td>
<td>1 10\textsuperscript{-1(4)}</td>
<td>3.0 10\textsuperscript{2}</td>
<td>3.0 10\textsuperscript{2}</td>
</tr>
<tr>
<td>\textsuperscript{241}Am</td>
<td>1 10\textsuperscript{-1(4)}</td>
<td>5.0 10\textsuperscript{2}</td>
<td>3.0 10\textsuperscript{2}</td>
</tr>
</tbody>
</table>

1. Value for lobsters.
2. Value for winkles.
3. Value for crab.
4. Appears to give significant under-prediction of fish activity concentrations.

### Table 19  Site specific K\textsubscript{ds} (Bq kg\textsuperscript{-1} per Bq l\textsuperscript{-1}). Source: Goshawk and Clarke (1999)

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Ribble Estuary</th>
<th>Whitehaven Harbour</th>
<th>Beaches at Seascale</th>
</tr>
</thead>
<tbody>
<tr>
<td>\textsuperscript{3}H</td>
<td>1 10\textsuperscript{0}</td>
<td>1 10\textsuperscript{0}</td>
<td>1 10\textsuperscript{0}</td>
</tr>
<tr>
<td>\textsuperscript{14}C</td>
<td>2.8 10\textsuperscript{3}</td>
<td>3.2 10\textsuperscript{3}</td>
<td>4.0 10\textsuperscript{2}</td>
</tr>
</tbody>
</table>

This paper gives $K_d$s in silt/mud for Whitehaven for relevant radionuclides. A summary of the data from this paper is presented in Table 20.

Table 20  $K_d$s for Whitehaven and Sellafield (Bq kg$^{-1}$ per Bq l$^{-1}$). Source: Barr and Howorth, 1993.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Whitehaven</th>
<th>Sellafield site specific</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Silt/mud</td>
<td>Sand</td>
</tr>
<tr>
<td>$^{99}$Tc</td>
<td>$1.9 \times 10^2$</td>
<td>$2.4 \times 10^2$</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>$2.3 \times 10^2$</td>
<td>$2.3 \times 10^2$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$1.0 \times 10^2$</td>
</tr>
<tr>
<td>$^{239/240}$Pu</td>
<td>$1.8 \times 10^4$</td>
<td>$1.8 \times 10^4$</td>
</tr>
<tr>
<td></td>
<td>$1.8 \times 10^4$</td>
<td></td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>$1.9 \times 10^5$</td>
<td>$2.4 \times 10^5$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$5.0 \times 10^3$</td>
</tr>
</tbody>
</table>

4.2  Conclusions

The aim of this section was to review available data on $K_d$ values for sediments and concentration factor values for biota in the locations of interest for this study. These parameters are used in modelling and reflect the behaviour of radionuclides in the marine environment. The following tables give the sediment distribution coefficients and CFs found together with the value currently used in the MARINA II model.

To determine the site-specific values to be used in the study, emphasis was placed on the radionuclides of primary interest (as indicated in Table 1). Additionally, where several values were identified, the values differing greatest from the current values were selected (appropriate to the sediment conditions being modelled) to illustrate the greatest differences when applied in the models. Table 21 shows that for most radionuclides, the current sediment distribution coefficients used are similar to the site-specific values found. The exceptions are $^{137}$Cs, $^{239}$Pu and $^{241}$Am for which several site-specific values (marked in bold in Table 21) were found that differed by approximately an order of magnitude (with the exception of $^{137}$Cs) from the current values used. These site-specific values were used later in this study as outlined in Section 7.

Table 22 gives the concentration factors found for various biota. Values selected for further study are indicated in bold based on the criteria of required combinations of radionuclide, location and biota as identified in Table 1 and preliminary review of the likely validation of the MARINA II model when such CFs are applied to the model predictions. These site-specific concentration factors
were later applied to the filtered water activity concentrations from the model runs to investigate the effect on predicted activity concentrations in fish, crustaceans, molluscs and seaweed as outlined in Section 7.

### Table 21 Site-specific K_d values for Irish Sea (Bq kg\(^{-1}\) per Bq l\(^{-1}\))

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Area</th>
<th>MARINA II compartments</th>
<th>Current value used(^{(1)})</th>
<th>K_d (2)</th>
<th>K_d (3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^3)H</td>
<td>Ribble Estuary</td>
<td>37</td>
<td>(1 \times 10^0)</td>
<td></td>
<td>(1 \times 10^0)</td>
</tr>
<tr>
<td></td>
<td>Whitehaven Harbour</td>
<td>32</td>
<td>(1 \times 10^0)</td>
<td></td>
<td>(1 \times 10^0)</td>
</tr>
<tr>
<td></td>
<td>Seascale beach</td>
<td></td>
<td></td>
<td></td>
<td>(1 \times 10^0)</td>
</tr>
<tr>
<td>(^{14})C</td>
<td>Ribble Estuary</td>
<td>37</td>
<td>(2 \times 10^3)</td>
<td></td>
<td>(2.8 \times 10^3)</td>
</tr>
<tr>
<td></td>
<td>Whitehaven Harbour</td>
<td>32</td>
<td>(3.2 \times 10^3)</td>
<td></td>
<td>(4.0 \times 10^3)</td>
</tr>
<tr>
<td></td>
<td>Seascale beach</td>
<td></td>
<td></td>
<td></td>
<td>(1 \times 10^0)</td>
</tr>
<tr>
<td>(^{99})Tc</td>
<td>Ribble Estuary</td>
<td>37</td>
<td>(1 \times 10^2)</td>
<td></td>
<td>(1.9 \times 10^2)</td>
</tr>
<tr>
<td></td>
<td>Whitehaven Harbour</td>
<td>32</td>
<td>(2.4 \times 10^2)</td>
<td></td>
<td>(1 \times 10^0)</td>
</tr>
<tr>
<td></td>
<td>Seascale beach</td>
<td></td>
<td></td>
<td></td>
<td>(1 \times 10^0)</td>
</tr>
<tr>
<td>(^{137})Cs</td>
<td>Ribble Estuary</td>
<td>37</td>
<td>(2.3 \times 10^2) (\text{(4)})</td>
<td></td>
<td>(2.3 \times 10^2)</td>
</tr>
<tr>
<td></td>
<td>Whitehaven Harbour</td>
<td>32</td>
<td>(3.0 \times 10^3) (\text{(5)})</td>
<td></td>
<td>(2.3 \times 10^2)</td>
</tr>
</tbody>
</table>
|              | Sellafield | 35 | | | \(1.6 \times 10^2\) \(\text{\(1.57 \times 10^2\)\}}\
|              | Seascale beach | | | | \(1 \times 10^2\) |
| \(^{239/240}\)Pu | Ribble Estuary | 37 | \(1 \times 10^5\) \(\text{(4)}\) | \(\text{\(1.8 \times 10^4\)\}} \(\text{\(1.8 \times 10^5\)\}} \(\text{\(1.8 \times 10^4\)\}} \(\text{\(1.8 \times 10^5\)\}}
|              | Whitehaven Harbour | 32 | | | \(\text{\(1.8 \times 10^4\)\}} \(\text{\(1.8 \times 10^5\)\}}
|              | Seascale beach | | | | \(\text{\(1.8 \times 10^4\)\}}
| \(^{241}\)Am  | Ribble Estuary | 37 | \(2 \times 10^6\) \(\text{(4)}\) | \(\text{\(1.9 \times 10^6\)\}}
|              | Whitehaven Harbour | 32 | \(2.4 \times 10^6\) \(\text{(4)}\) | | \(7.5 \times 10^5\) |
|              | Seascale beach | | | | \(5.0 \times 10^3\) |

2. See Table 19.
3. See Table 20.
4. Site-specific value used for Irish Sea.
5. Refers to silt and clay conditions.
6. Generic value used in MARINA II model.
7. Bold text indicates values to be applied in MARINA II model (see Section 7).
Table 22 Site-specific CFs for the Irish Sea (Bq kg\(^{-1}\) per Bq l\(^{-1}\))

<table>
<thead>
<tr>
<th>Radio-</th>
<th>Area</th>
<th>MARINA II Compart-</th>
<th>Organism</th>
<th>Current value used(^{(1)})</th>
<th>CF</th>
</tr>
</thead>
<tbody>
<tr>
<td>nuclide</td>
<td></td>
<td>ment</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(^{99})Tc</td>
<td>Irish Sea</td>
<td>35</td>
<td>Seaweed(^{(2)})</td>
<td>3 (10^4)</td>
<td>((1.19 \pm 0.41) \times 10^5)</td>
</tr>
<tr>
<td></td>
<td>35</td>
<td>Mussels</td>
<td>1 (10^3)</td>
<td>5.0 (10^2)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>35</td>
<td>Lobster (abdomen muscle)</td>
<td>1 (10^3)</td>
<td>(7.2 \times 10^2)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>35</td>
<td>Norwegian lobster (abdominal muscle(^{(3)}))</td>
<td>1 (10^3)</td>
<td>9.7 (10^2)</td>
<td></td>
</tr>
<tr>
<td>35 Balbriggan</td>
<td></td>
<td>Fucus vesiculosus</td>
<td>3 (10^4)</td>
<td>((1.32 \pm 0.46) \times 10^5) (^{(4)})</td>
<td></td>
</tr>
<tr>
<td>33 Clogherhead, Howth</td>
<td></td>
<td>Fish (cod, plaice, whiting and ray)</td>
<td>3 (10^2)</td>
<td>(1.25 \times 10^1 \pm 4.90 \times 10^0) (^{(5)})</td>
<td></td>
</tr>
<tr>
<td>33 Carlingford</td>
<td></td>
<td>Mussels (edible portion)</td>
<td>1 (10^3)</td>
<td>((9.7 \pm 5.7) \times 10^2)</td>
<td></td>
</tr>
<tr>
<td>33 Carlingford</td>
<td></td>
<td>Oysters (edible portion)</td>
<td>1 (10^3)</td>
<td>((2.8 \pm 1.2) \times 10^2)</td>
<td></td>
</tr>
<tr>
<td>35 Irish Sea</td>
<td></td>
<td>Fish</td>
<td>3 (10^1)</td>
<td>3.0 (10^1)</td>
<td></td>
</tr>
<tr>
<td>35 Irish Sea</td>
<td></td>
<td>Crustacea</td>
<td>1 (10^3)</td>
<td>(9.0 \times 10^3) (^{(6)})</td>
<td>(1.4 \times 10^2) (^{(7)})</td>
</tr>
<tr>
<td>35 Irish Sea</td>
<td></td>
<td>Molluscs</td>
<td>1 (10^3)</td>
<td>6.0 (10^3) (^{(8)})</td>
<td></td>
</tr>
<tr>
<td>(^{95m})Tc</td>
<td>Whitehaven</td>
<td>32</td>
<td>Nephrops</td>
<td>1 (10^3)</td>
<td>(1.4 \times 10^3) (^{(9)})</td>
</tr>
<tr>
<td>(^{3})H</td>
<td>Irish Sea</td>
<td>35</td>
<td>Fish</td>
<td>1 (10^0)</td>
<td>1 (10^0)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>35</td>
<td>Crustacea</td>
<td>1 (10^0)</td>
<td>1 (10^0)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>35</td>
<td>Molluscs</td>
<td>1 (10^0)</td>
<td>1 (10^0)</td>
<td></td>
</tr>
<tr>
<td>(^{14})C</td>
<td>Irish Sea</td>
<td>35</td>
<td>Fish</td>
<td>2 (10^4)</td>
<td>3.0 (10^3)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>35</td>
<td>Crustacea</td>
<td>2 (10^4)</td>
<td>6.0 (10^3) (^{(6)})</td>
<td></td>
</tr>
<tr>
<td></td>
<td>35</td>
<td>Molluscs</td>
<td>2 (10^4)</td>
<td>5.0 (10^3)</td>
<td></td>
</tr>
<tr>
<td>(^{137})Cs</td>
<td>Irish Sea</td>
<td>35</td>
<td>Fish</td>
<td>1 (10^2)</td>
<td>9.0 (10^1)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>35</td>
<td>Crustacea</td>
<td>3 (10^1)</td>
<td>3.0 (10^1)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>35</td>
<td>Molluscs</td>
<td>3 (10^1)</td>
<td>(5.0 \times 10^1)</td>
<td></td>
</tr>
<tr>
<td>(^{239})Pu</td>
<td>Irish Sea</td>
<td>35</td>
<td>Fish</td>
<td>1 (10^2)</td>
<td>(1.1 \times 10^{-1}) (^{(10)})</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>35</td>
<td>Crustacea</td>
<td>2 (10^2)</td>
<td>3.0 (10^2)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>35</td>
<td>Molluscs</td>
<td>3 (10^2)</td>
<td>3.0 (10^3)</td>
<td></td>
</tr>
<tr>
<td>(^{241})Am</td>
<td>Irish Sea</td>
<td>35</td>
<td>Fish</td>
<td>1 (10^2)</td>
<td>(1.1 \times 10^{-1}) (^{(10)})</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>35</td>
<td>Crustacea</td>
<td>5 (10^2)</td>
<td>5.0 (10^2)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>35</td>
<td>Molluscs</td>
<td>2 (10^3)</td>
<td>(3.0 \times 10^3)</td>
<td></td>
</tr>
</tbody>
</table>

2. Dry weight.
3. CF to body part.
4. Filtered seawater and seaweed activity concentrations.
5. Bq/kg wet weight. Edible flesh, Bq/l.
6. Value for lobsters.
7. Value for crabs.
8. Value for winkles.
10. Appears to give significant under-prediction of fish concentration factors.
11. Values in bold selected for later application to the MARINA II model (see Section 7) on basis of required combinations of radionuclide, location and biota (Table 1) and preliminary comparison with measurement data.
5 VARIABILITY IN CSERAM MODEL RUNS

5.1 Analysis of variability in previous model runs (Irish Sea only)

5.1.1 Background
The aim of this section is to determine the magnitude of the variability of predicted radionuclide activity concentrations present in high-resolution computer runs. Within the limits of the model fidelity, the outcome will enable an assessment to be made of the natural variability associated with point observations that are taken to be representative of annual average values over extended spatial areas. Variability of radionuclide activity concentrations in water arises primarily from variations in discharge rates, and tidal and meteorological conditions. Variability of radionuclide concentrations in biota will depend on water activity concentrations, together with a range of possible biological effects including feeding, metabolism, movement (for fauna) and seasonal growth differences. Here, only variability due to non-biological factors is addressed.

Because the CSERAM model simulates the tide and wind driven dynamics of radionuclide transport at a high spatial (3.5km) and temporal (hourly) resolution, it is possible, in principle, to evaluate the variability of radionuclide concentrations in the modelled water and sediment phases. Radionuclides, sites and media where further investigation of the agreement between models and observations was considered desirable are given in Table 1. Existing high resolution output from runs undertaken under the previous study (Jones et al., 2003) was re-analysed to obtain predictions of the spatial and temporal variability at the model sampling points used previously to compare with observations. These model runs include the period 1997 to mid 1999 that has been chosen for detailed study. Some confidence can be attached to the model estimates of temporal variability of water concentrations. Given that the work relies on existing model runs not specifically designed to assess variability, difficulties were found with assessing, in particular, spatial variability. This is discussed further below. With respect to estimating variability of radionuclide concentrations in biota, this ideally requires a biological response model. In the absence of such a validated biological model, an approach has been adopted that will allow the use of concentration factors.

Variability of activity concentrations in water in the CSERAM calculations arise from three principal sources.

a Tidal advection. This will induce a regular 12-hour cycle in values due to tidal advection of concentration gradients past a given point. This is modulated by the effects of the Spring Neap cycle and other variation arising from the combination of tidal components.
MEASUREMENT DATA

b Meteorological conditions. Realistic wind forcing is included which will give rise to changes in transport rates and hence concentrations. For particle reactive radionuclides, changes in activity concentrations in water will occur under storm conditions as radionuclides equilibrate with resuspended bed sediments.

c Discharge profile. For $^{137}$Cs, $^{239/240}$Pu, and $^{241}$Am, higher resolution Sellafield discharge data has been used which give a monthly breakdown of discharges.

While CSERAM can predict the variation in activity concentrations in water, activity concentrations in other media are generally obtained by multiplying activity concentrations in water by appropriate concentration factors. Such a simplistic approach can be justified for estimating annual averages. However, the approach is not likely to be valid for evaluating how biota would respond to short term changes in activity concentrations in water. Nevertheless, the use of concentration factors may be appropriate when applied to intermediate time scales and seasonal (i.e. 3 month) variations. The assumption is therefore made that concentration factors can be used with seasonally averaged activity concentrations in water and that biota will equilibrate over these time scales (Swift, 2001). As outlined in Section 2.1, measurement data is based on aggregated samples in the case of biota, which will have an averaging effect on measured activity concentrations. In seawater, comparisons with an averaged activity concentration in seawater from CSERAM may be less valid due measurements being derived from one sample. Short-term variations can be observed in seawater, but this is less likely to be the case for biota. Thus, for assessing temporal variability of radionuclides in biota, activity concentrations in water predicted using CSERAM were averaged over a 3-month interval and the usual equilibrium concentration factor applied. Seasonal average activity concentrations in water appropriate to each biota were calculated at the grid point closest to the measurement location. These were the same grid locations used to calculate annual averages in the previous study (Jones et al., 2003).

Full spatial output from model runs was archived only at four-month intervals and so spatial variability can only be assessed at these instants. For each model sampling location used to compare against observations, the maximum and minimum value of the activity concentration in water in the surrounding eight grid cells is determined. For biota, the assumption is made that the spatial variation in activity concentrations in water at the given instant is representative of the longer term average difference in activity concentrations in biota, when multiplied by the appropriate concentration factor. Clearly, variability in activity concentrations of biota obtained in this manner needs to be treated with considerable caution since it is based on the spatial variability at a single instance. Given that activity concentrations in biota will depend on the longer term average it is difficult to assess how accurate the resulting estimates are. To resolve this, new computer runs would be required with the specific aim of addressing spatial variability.
5.1.2 Results

Temporal variation in activity concentrations in water are displayed by plotting the 3 month ‘seasonal’ average at the grid point closest to where observational samples were taken. The maximum and minimum values that occurred in the model run during each three-month period are plotted as error bars around the seasonal average. It was not felt appropriate to multiply the maximum and minimum activity concentrations in water by the appropriate concentration factor to obtain an estimate of maximum and minimum activity concentrations as biota will not respond to short-term peaks in the activity concentrations in water. To display spatial variations, the sampled activity concentration is plotted, with the maximum and minimum values in adjacent grid boxes displayed as an error bar.

5.1.2.1 Caesium-137

Sellafield

Temporal variability for $^{137}$Cs concentrations near to Sellafield in water, molluscs and sediment are shown in Figure 9 to Figure 11, respectively. It is important to note that the activity concentration curves in water and biota do not show an identical variation because they refer to different locations. Model output for water is taken at St Bees, molluscs at Nethertown and sediments at Maryport. Figure 10 clearly shows minimum activity concentrations in molluscs in summer, with higher values in autumn and winter. At the location chosen to represent water samples (Figure 9) the general trends are less clear and maximum values occur at different times in different years. For sediments at Maryport, the highest modelled activity concentrations occur in summer. The likely reason for seasonal variations in activity concentrations are the complex interaction between dissolved and particulate phases due to storm resuspension of contaminated bottom sediments. Relative seasonal variations in both water and biota are of the order of 5% of the annual average while extreme values in the water can fluctuate by up to 80% around the annual mean.

Snapshots of the spatial variability for $^{137}$Cs concentrations near to Sellafield in water and biota are shown in Figure 12 and Figure 13. Again, note that the water concentrations used to calculate the results in the two figures come from different locations and so a different distribution is observed. Information on the spatial variability of sediment concentrations was not available from the model run.
Figure 9 Seasonal variability in activity concentrations of $^{137}$Cs in water at St Bees near Sellafield. Three-month average plotted with error bars representing maximum and minimum values that occurred during the averaging period.

Figure 10 Temporal variation in activity concentrations of $^{137}$Cs in mollusc near to Sellafield, based on three-month average of water concentrations.
Figure 11 Seasonal variability in activity concentrations of $^{137}\text{Cs}$ in sediment near to Maryport

Figure 12 Snapshot of spatial variability in activity concentrations of $^{137}\text{Cs}$ in water at St. Bees, near Sellafield. The sample point is plotted with error bars representing maximum and minimum values at that instant taken from the surrounding grid points.
Figure 13 Spatial variability in activity concentrations of $^{137}$Cs in molluscs at Nethertown, near Sellafield calculated by multiplying instantaneous activity concentrations in water by a concentration factor.

**Wylfa**

The results of spatial and temporal variability in activity concentrations of $^{137}$Cs in water at Wylfa (Figure 14 and Figure 15) show relative temporal variations of the order of 20% on a seasonal basis. Extreme values are up to 60% about the annual mean. However, spatial variability is smaller, at around 10%.

Figure 14 Seasonal variability in activity concentrations of $^{137}$Cs in water at Wylfa. Three-month average plotted.
**Figure 15** Snapshot of spatial variability in activity concentrations of $^{137}\text{Cs}$ in water at Wylfa. The sample point is plotted with error bars representing maximum and minimum values at that instant taken from the surrounding grid points.

**5.1.2.2 Plutonium-239/240**

*Northern Ireland*

Activity concentrations in molluscs are considered for this radionuclide at this location (Figure 16 and Figure 17). A seasonal variation averaging 15% around the annual mean is predicted. Instantaneous spatial variations between adjacent grid boxes are around 5%.

**Figure 16** Seasonal variability in activity concentrations of $^{239/240}\text{Pu}$ in molluscs at Northern Ireland locations. Three-month average plotted.
5.1.2.3 Americium-241  
Sellafield  
The seasonal variability in activity concentrations of $^{241}\text{Am}$ in molluscs at Sellafield (Figure 18 and Figure 19) lie in the range 5% - 10%. The lowest activity concentrations occur in summer. The spatial variability is relatively high at about 30%.

Figure 18 Seasonal variability in activity concentrations of $^{241}\text{Am}$ in molluscs at Sellafield. Three-month average plotted.
5.1.2.4 Technetium-99

Sellafield

The seasonal variability in activity concentrations of $^{99}$Tc in crustaceans at Sellafield (Figure 20 and Figure 21) can be up to 30% of the annual mean, although 25% variation is more representative. The relatively large variation between years is due to the change in annual discharge rates. Spatial variability is quite high at about 45% and due to the large gradients in activity concentrations close to the Sellafield source.

Figure 19 Snapshot of spatial variability in activity concentrations of $^{241}$Am in molluscs at Sellafield. Error bars represent maximum and minimum values at that instant taken from surrounding grid points.

Figure 20 Seasonal variability in activity concentrations of $^{99}$Tc in crustaceans at Sellafield. Three-month average with error bars representing maximum and minimum values that occurred during the averaging period.
Figure 21 Snapshot of spatial variability in activity concentrations of $^{99}$Tc in crustaceans near Sellafield. Error bars represent maximum and minimum values at that instant taken from surrounding grid points.

Northern Ireland
Seasonal variability in activity concentrations of $^{99}$Tc in seaweed at the Northern Ireland locations (Figure 22 and Figure 23) can be up to 40% of the annual mean, although 25% variation is more representative. The spatial variability is relatively low due to smearing out of activity concentration gradients at distance from the Sellafield source.

Figure 22 Seasonal variability in activity concentrations of $^{99}$Tc in crustaceans at Northern Ireland locations. Three-month average plotted.
Figure 23 Snapshot of spatial variability in activity concentrations of $^{99}$Tc in seaweed at Northern Ireland locations. Error bars represent maximum and minimum values at that instant taken from surrounding grid points.

Wylfa
The seasonal variability in activity concentrations of $^{99}$Tc in crustaceans at Wylfa (Figure 24 and Figure 25) can be up to 60% of the annual mean, although 25% variation is more representative. The spatial variability is lower again due to smearing out of activity concentration gradients at distance from the Sellafield source.

Figure 24 Seasonal variability in activity concentrations of $^{99}$Tc in crustaceans at Wylfa. Three-month average plotted.
Figure 25 Snapshot of spatial variability in activity concentrations of $^{99}$Tc in crustaceans at Wylfa. Error bars represent maximum and minimum values at that instant taken from surrounding grid points.

5.1.3 Summary and discussion

The results on variability are summarised in Table 23. The relative percentage seasonal variation is the variability around the annual mean averaged over the number of years which is defined to be

$$S_t = \frac{100}{N_y} \sum_i s_i / \bar{X}_i$$

where $s_i$ is the standard deviation over year $i$, 
$\bar{X}_i$ is the annual average of year $i$, 
and $N_y$ is the number of years considered.
The percentage spatial variation is

\[ s_s = \frac{100}{N} \sum_n \max_j \left| \frac{x_j^{(n)} - x_j^{(n)}}{x_j^{(n)}} \right| \]

where 'max' is taken over the points \( x_j^{(n)} \) surrounding the sample point \( x_j^{(n)} \) at time \( n \)

and where \( N \) is the number of times the location is sampled over the period 1997 – 2000.

Because relative changes have been taken, the effect of concentration factors, which scale the activity concentrations in the water column, has been removed. Therefore it is possible to compare results consistently across different media.

Table 23: Summary of variability expressed as a percentage of the seasonal average for all combinations of radionuclides and media considered.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Site</th>
<th>Media</th>
<th>% Seasonal variation</th>
<th>% Spatial variation</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{137}\text{Cs})</td>
<td>Sellafield</td>
<td>Water</td>
<td>5</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>Sellafield</td>
<td>Mollusc</td>
<td>8</td>
<td>40</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sediment</td>
<td>7</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>Wylfa</td>
<td>Water</td>
<td>22</td>
<td>10</td>
</tr>
<tr>
<td>(^{239/240}\text{Pu})</td>
<td>Northern Ireland</td>
<td>Molluscs</td>
<td>15</td>
<td>5</td>
</tr>
<tr>
<td>(^{241}\text{Am})</td>
<td>Sellafield</td>
<td>Molluscs</td>
<td>4</td>
<td>30</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Crustacea</td>
<td>15</td>
<td>45</td>
</tr>
<tr>
<td></td>
<td>Northern Ireland</td>
<td>Seaweed</td>
<td>27</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>Wylfa</td>
<td>Crustacea</td>
<td>30</td>
<td>10</td>
</tr>
</tbody>
</table>

Note: % Seasonal variation is the average of the seasonal variation around the annual mean.

First, comparing the modelling results for \(^{137}\text{Cs}\) and \(^{99}\text{Tc}\), for which variability was calculated near to Sellafield and at a distance, interesting similarities and differences emerge. For \(^{137}\text{Cs}\), the relative (but not absolute variability) is greater at Wylfa than near Sellafield. Near to Sellafield a relatively constant input of \(^{137}\text{Cs}\), mainly remobilised from bed sediments, would be expected to dominate the activity concentrations in water. Further away, activity concentrations in water are controlled by advection from the Sellafield region and controlled by meteorological conditions. For \(^{99}\text{Tc}\), less difference in variability between near distances and far distances is observed, although at far distances, variability is still relatively larger. It can be concluded that the near Sellafield region during the period of interest was dominated by changes in Sellafield inputs.

It is more difficult to draw conclusions from the \(^{239/240}\text{Pu}\) and \(^{241}\text{Am}\) results. However, activity concentrations in water for both radionuclides are dominated by remobilisation from sediments and the near distance results for \(^{241}\text{Am}\) at
Sellafield show less temporal variability than the far distance results for $^{239/240}$Pu in the Northern Ireland region. This is consistent with the sensitivity of far distance activity concentrations being dependent on meteorological conditions.

For spatial variability, the far distance results at Wylfa and Northern Ireland show a significantly smaller variability than the near Sellafield values. This is exactly what would be expected as activity concentration gradients smear out with distance.

In conclusion, the maximum relative seasonal variability is predicted to be around 25% based purely on fluctuations in water activity concentrations and assuming biota respond to the average activity concentrations in the water column on this time scale. Spatial variability was difficult to assess with the present dataset but appears to be of the same magnitude or smaller than the typical seasonal variability. In general, both temporal and spatial variability was assessed to be smaller than the uncertainty in the modelling and parameterisation of key processes.

### 5.2 Comparison of variability between measurements and CSERAM

Where possible estimates of variability obtained from model runs have been compared with the variability in measurement data derived in Section 2.2. Sufficient sample data were not available for all media and radionuclides considered in the previous section, however, all available data are compared here in Figure 26 to Figure 30. In this section, the percentage relative variability in a given year is defined as

\[ \nu = 100 \frac{s}{\bar{x}} \]

where \( s \) is the standard deviation

and \( \bar{x} \) is the annual mean.

In general, modelled variability in biota is less than that observed. This is not surprising as the modelled results only take account of variability arising from physical changes to the water concentrations. It does not account for sampling, analytical or biological factors that may affect measured radionuclide concentrations. Interestingly, the only comparison with non-biological samples, $^{137}$Cs in sediments near Sellafield (Figure 27), showed closer agreement between the observed and modelled variability than most of those involving biological media. This can be expected due to the choice of one sampling location and the removal of any biological factors affecting the measured activity concentration.
Figure 26 Comparison of measured and modelled variability for $^{137}$Cs in molluscs near Sellafield

Figure 27 Comparison of measured and modelled variability for $^{137}$Cs in sediment near Sellafield
Figure 28 Comparison of measured and modelled variability for $^{241}$Am in molluscs near Sellafield

Figure 29 Comparison of measured and modelled variability for $^{99}$Tc in crustaceans near Sellafield
VALIDATION OF THE MARINA II MODEL

The discharge data compiled for this study (Section 3) have been used to extend the MARINA II model runs to 2002. These model runs were compared with the measurement data compiled in Section 2 and in the MARINA II study. This comparison considered additional data for locations previously considered and for wider areas, such as the north coast of Scotland. The regions to be addressed as a priority in the current study were the Irish Sea areas (Sellafield, Wylfa and Northern Ireland indicator locations) and Dounreay.

The validation was carried out for the period 1997 to 2002. Where relevant, the contribution from atmospheric weapons testing and Chernobyl was included. The validation concentrated on those species, radionuclides and locations listed in italics in Table 1.
This validation task consisted of a variety of validation techniques:

a Variation over time for the period 1997 to 2002 – This followed a similar approach to that used in the previous study by presenting annual average activity concentrations derived from measurements and the model results.

b Variation with space – In Section 2, data were compiled of activity concentrations in the regions around the specific locations used in the previous study. This enables the spatial variability of observed activity concentrations to be compared with model predictions in adjacent compartments in the case of MARINA II and adjacent grid points in the case of CSERAM.

c Variation with species – The uptake of radionuclides may differ by species for certain radionuclides, however the previous study used a generic concentration factor for the taxonomic class of flora or fauna that was sampled. One example of the apparent differences can be observed for $^{99}$Tc in lobsters and crabs.

The following section discusses the comparison of modelled activity concentrations with measurement data, with Figure 31 to Figure 53 showing:

a Comparison of measurements with model predictions for each location over time (1997-2002) also showing limited variation with species at the same location.

b Comparison of measurements with model predictions of a single species for spatial variation.

For some radionuclides and locations, further indicators, such as filtered water and sediment, were used in addition to those identified in Table 1. In particular, monitoring data from BNFL and UKAEA have been included. Examples are given in Table 24. These data were used to demonstrate the fit of the model against available measurement data in terms of sedimentation and flow rate data. Additionally, these data provided useful information in deciding which parameters to change, (sediment $K_d$ or biota concentration factor) when refining the model (Section 7).

For this validation study, the model uses the parameter values recommended for the MARINA II model for sediment $K_d$ values and biota concentration factors, as used in the previous study (Jones et al., 2003).
Table 24  Measured activity concentrations in molluscs near Sellafield (BNFL data)

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Species</th>
<th>Location</th>
<th>Activity Concentration (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>2001</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>winkles</td>
<td>St Bees-Selker</td>
<td>5.7</td>
</tr>
<tr>
<td></td>
<td>winkles</td>
<td>Parton</td>
<td>2.6</td>
</tr>
<tr>
<td></td>
<td>mussels</td>
<td>St Bees-Selker</td>
<td>2.5</td>
</tr>
<tr>
<td></td>
<td>limpets</td>
<td>St Bees-Selker</td>
<td>5.6</td>
</tr>
<tr>
<td></td>
<td>cockles</td>
<td>Ravenglass</td>
<td>4.5</td>
</tr>
<tr>
<td></td>
<td>whelks</td>
<td>off-shore</td>
<td>1.2</td>
</tr>
<tr>
<td></td>
<td>scallops</td>
<td>off-shore</td>
<td>0.8</td>
</tr>
<tr>
<td></td>
<td>squid</td>
<td>off-shore</td>
<td>1.3</td>
</tr>
<tr>
<td></td>
<td>queens</td>
<td>off-shore</td>
<td></td>
</tr>
<tr>
<td>$^{239/240}$Pu</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>winkles</td>
<td>St Bees-Selker</td>
<td>11</td>
</tr>
<tr>
<td></td>
<td>winkles</td>
<td>Parton</td>
<td>1.7</td>
</tr>
<tr>
<td></td>
<td>mussels</td>
<td>St Bees-Selker</td>
<td>8.3</td>
</tr>
<tr>
<td></td>
<td>limpets</td>
<td>St Bees-Selker</td>
<td>11</td>
</tr>
<tr>
<td></td>
<td>cockles</td>
<td>Ravenglass</td>
<td>8.5</td>
</tr>
<tr>
<td></td>
<td>whelks</td>
<td>off-shore</td>
<td>1.4</td>
</tr>
<tr>
<td></td>
<td>scallops</td>
<td>off-shore</td>
<td>0.4</td>
</tr>
<tr>
<td></td>
<td>squid</td>
<td>off-shore</td>
<td></td>
</tr>
<tr>
<td></td>
<td>queens</td>
<td>off-shore</td>
<td></td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>winkles</td>
<td>St Bees-Selker</td>
<td>18</td>
</tr>
<tr>
<td></td>
<td>winkles</td>
<td>Parton</td>
<td>4.4</td>
</tr>
<tr>
<td></td>
<td>mussels</td>
<td>St Bees-Selker</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>limpets</td>
<td>St Bees-Selker</td>
<td>16</td>
</tr>
<tr>
<td></td>
<td>cockles</td>
<td>Ravenglass</td>
<td>24</td>
</tr>
<tr>
<td></td>
<td>whelks</td>
<td>off-shore</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>scallops</td>
<td>off-shore</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>squid</td>
<td>off-shore</td>
<td></td>
</tr>
<tr>
<td></td>
<td>queens</td>
<td>off-shore</td>
<td></td>
</tr>
</tbody>
</table>

Note: bold text indicates measurement reported as less than value

6.1  Comparison of model predictions with measurements for 1997-2002 showing variation with sample type at the same location

This section considers examples which show the variation of activity concentration between sample types at selected locations.
6.1.1 Sellafield
Measured data in this area were compared with model predictions using discharges from Sellafield only. The contribution from weapons fallout was insignificant in comparison to Sellafield discharges.

6.1.1.1 Caesium-137
Figure 31 to Figure 34 show model predictions and measured activity concentrations in filtered water, molluscs and sediment. Figure 27 shows that the model predictions in the Sellafield local compartment are about a factor of two lower than the measurements from RIFE data and up to seven times lower than BNFL data. One reason for the difference between measurements is that the BNFL concentrations were measured at Sellafield close to the discharge point while the RIFE data were measured at St Bees, which is around 10 km north of the discharge point, but still within the local compartment.

![Activity concentration graph](image)

**Figure 31** Estimated and measured activity concentrations of $^{137}$Cs in seawater near Sellafield

Figure 32 shows that the model predictions for molluscs are closer to the measurements for mussels than for winkles. The observed difference in the model predictions and measurements for filtered water accounts for the lower predictions for molluscs compared to the measured values in mussels. This means that the large difference between measured values in winkles and predicted values is likely to be due to the low mollusc concentration factor used.
This is also shown in Figure 33, where measured activity concentrations in four species of mollusc are compared with predicted data.

**Figure 32** Estimated and measured activity concentrations of $^{137}\text{Cs}$ in molluscs near Sellafield (Measurements at Nethertown)

**Figure 33** Estimated and measured activity concentrations of $^{137}\text{Cs}$ in molluscs near Sellafield (Measurements for Sellafield Coastal Area)
Figure 34 shows that the model predictions for activity concentrations in sediment in the Sellafield local compartment compare closer to the measurements for sand than those for silt. In both cases, the model predictions compare well with measurements for sand and are about a factor of 3 lower than the BNFL measurements in silt from Ravenglass (Eskmeals viaduct) and the RIFE measurements in mud and sand from Ravenglass (Raven Villa). The predictions are approximately a factor of 7 lower than the measurement data in mud from Ravenglass (Carleton Marsh). These differences may indicate that the sediment distribution factor used for the model is too low for mud and silt and more suited for sandy sediments.

![Graph showing activity concentrations of 137Cs in sediment near Sellafield](image_url)

**Figure 34** Estimated and measured activity concentrations of $^{137}$Cs in sediment near Sellafield

### 6.1.1.2 Americium-241

Figure 35 to Figure 37 compare model predictions against measurements of activity concentrations in filtered water and molluscs. Figure 35 shows that the model predictions of activity concentrations in the Sellafield local compartment are a factor of about 20 times lower than the measurements from BNFL data. One reason for the difference is that data were measured at Sellafield close to the discharge point. Additionally, most of the measurements are presented as 'less than' values, indicating that they are not reliable indicators. The actual activity concentration may lie closer to the predicted value than shown in the figure.
DISTINGUISHING BETWEEN CURRENT & HISTORIC RADIOACTIVE DISCHARGES TO THE MARINE ENVIRONMENT – FURTHER WORK TO VALIDATE MODELS AND QUANTIFY UNCERTAINTIES

Figure 35 Estimated and measured activity concentrations of $^{241}$Am in filtered seawater near Sellafield - NB measurements at the limit of detection presented as unfilled points

Figure 37 shows that the model predictions of activity concentrations in molluscs compare closer to those measurements from whelks than from any other species of mollusc. The large difference between measured values in molluscs and predicted values is likely to be due to a combination of the model underestimating activity concentrations in filtered water and the use of a low concentration factor for molluscs.

Figure 36 Estimated and measured activity concentrations of $^{241}$Am in molluscs near Sellafield (Measurements at Nethertown)
**Figure 37** Estimated and measured activity concentrations of $^{241}$Am in molluscs near Sellafield (Measurements for Sellafield Coastal Area)

**6.1.1.3 Technetium-99**

Figure 38 to Figure 40 show model predictions of activity concentrations against measurements in filtered water and crustaceans.

**Figure 38** Estimated and measured activity concentrations of $^{99}$Tc in filtered seawater near Sellafield
Figure 38 shows that model predictions of activity concentrations of filtered water in the Sellafield local compartment compare well with the measured data being within a factor of 2. The value for 2002 is comprised of several observations of which two values were approximately an order of magnitude greater than the other measurements taken during the year. Excluding the outliers, a mean activity concentration of $^{99}\text{Tc}$ in filtered seawater of $0.7 \text{ Bq l}^{-1}$ can be estimated for 2002 compared with the modelling prediction of $0.5 \text{ Bq l}^{-1}$.

Figure 39 and Figure 40 show that the model predictions of activity concentrations for crustaceans are about an order of magnitude greater than the measurements for crabs and about an order of magnitude lower than the measurements for lobsters. This difference is due to the concentration factor used in the model for crustaceans, and not due to the model predictions of activity concentrations in filtered water. In Figure 40, the model predictions of activity concentrations in crustaceans are within about a factor of 2 of the measurements for $\text{Nephrops}$ (large prawns). Both the BNFL and RIFE data refer to sampling areas extending from St Bees Head to Selker. Additional variability may be introduced as a function of sampling and analytical techniques between the two laboratories involved.
6.1.2 Northern Ireland (Marina II compartment 30)
Measured activity concentration data in this region were compared with model predictions due to discharges from the Sellafield site only due to it being the only Irish Sea site discharging the selected radionuclides. The contribution from weapons fallout was also included.

6.1.2.1 Plutonium-239/240
Figure 41 shows that the model predictions of activity concentrations in molluscs are about an order of magnitude greater than the measurements for winkles. The activity concentration due to discharges from Sellafield contributes to around 99% of the modelled activity concentration with the remaining 1% due to atmospheric weapons testing fallout. A previous study (Simmonds et al., 2002) showed that model predictions of activity concentrations in filtered water compare well with measured data in the Northern Ireland region of the Irish Sea. The differences in predictions of activity concentrations in molluscs are likely therefore to be due to a high mollusc concentration factor used in the model. Alternatively, the winkles may have been sampled in a region near the coast, such as an inlet, which differs from the relatively large area represented by the model compartment. The measured activity concentrations in winkles were from samples taken in the Ards peninsula in Northern Ireland. Additional differences between model predictions and actual measurements may be due to local coastal effects such as the majority of the activity flowing through the North Channel rather than reaching the Northern Ireland coastline.
DISTINGUISHING BETWEEN CURRENT & HISTORIC RADIOACTIVE DISCHARGES TO THE MARINE ENVIRONMENT – FURTHER WORK TO VALIDATE MODELS AND QUANTIFY UNCERTAINTIES

Figure 41 Estimated and measured activity concentrations of $^{239/240}$Pu in molluscs for Northern Ireland

6.1.2.2 Technetium-99

Figure 42 shows that the model predictions of activity concentrations in seaweed are approximately an order of magnitude greater than measurement data. A previous study (Simmonds et al., 2002) showed that model predictions of activity concentrations in filtered water compare well with measured data. The differences between predictions of activity concentrations in seaweed and measurements may therefore be due to a high seaweed concentration factor being used. Discharges of $^{99}$Tc from Sellafield contribute 100% of the modelled activity concentration at Northern Ireland.

Figure 42 Estimated and measured activity concentrations of $^{99}$Tc in seaweed for Northern Ireland (Ardglass)
6.1.3 **Wylfa**
Measurement data in this region were compared with model predictions of activity concentrations in the Wylfa local compartment due to discharges from the Wylfa, Sellafield and Heysham nuclear sites. The contribution from weapons fallout was also included.

6.1.3.1 **Caesium-137**
Figure 43 shows that the model predictions of activity concentrations in filtered water for the Wylfa local compartment generally compare well with the measured data. The greatest differences between model predictions and measurement data are seen for 1999 and 2000 when the model predicts values up to a factor of 3 lower than the measurements. The sample location is at Holyhead which lies close to the southern edge of the MARINA II model compartment.

![Figure 43 Estimated and measured activity concentrations of $^{137}$Cs in filtered seawater near Wylfa with breakdown by source of activity](image1)

6.1.3.2 **Technetium-99**
Figure 44 shows that the model predictions of activity concentrations in crustaceans are about an order of magnitude greater than the measurements for crabs. The modelling of activity of $^{99}$Tc in the Wylfa area indicates that it all originates from the Sellafield site. The sample location for the crabs and lobster is the Wylfa pipeline. A previous study (Simmonds et al., 2002) shows that model predictions for activity concentrations in filtered water compare well with
measured data. The differences in predictions in crustaceans are likely therefore to be due to a non-species-specific crustacean concentration factor used in the model. The same was found for the Sellafield location for crabs and lobsters for this radionuclide, indicating that a lower concentration factor should be chosen for crabs and a higher concentration factor for lobsters.

![Figure 44 Estimated and measured activity concentrations of $^{99}$Tc in crustaceans near Wylfa](image)

6.1.4 Dounreay
Measurement data for activity concentrations in this region were compared with model predictions in the Dounreay local compartment due to discharges from the Dounreay, Sellafield, Hunterston (tritium only), and Cap de la Hague (tritium only) nuclear sites as these sites contribute more than 1% of the activity concentration at Dounreay. The contribution from weapons fallout was also included.
6.1.4.1  

**Tritium**

Figure 45 shows that the model predictions for activity concentrations of $^3$H in seawater in the Dounreay local compartment are a between a factor of five to ten times lower than the measurement data. One reason for this difference may be the large number of measurements which are at the limit of detection.

![Figure 45 Estimated and measured activity concentrations of $^3$H in seawater for near Dounreay with breakdown by source of activity - NB measurements at the limit of detection presented as unfilled points](image)

6.1.4.2  

**Plutonium-239/240**

Figure 46 shows that the model predictions for activity concentrations of $^{239/240}$Pu in molluscs are about a factor of five greater than the measurement data for winkles. Additional variability may be introduced by the differences in sampling and analytical techniques employed in the two laboratories. A previous study (Simmonds et al., 2002) showed that model predictions of activity concentrations in filtered water compare well with measured data in this area. The differences between predictions of activity concentrations in molluscs and the measurement data are likely therefore to be due to a high mollusc concentration factor used in the model, as also found for plutonium at the Northern Ireland location.
DISTINGUISHING BETWEEN CURRENT & HISTORIC RADIOACTIVE DISCHARGES TO THE MARINE ENVIRONMENT – FURTHER WORK TO VALIDATE MODELS AND QUANTIFY UNCERTAINTIES

Figure 46 Estimated and measured activity concentrations of $^{239/240}$Pu in molluscs near Dounreay - NB measurements at the limit of detection presented as unfilled points

6.1.4.3 Americium-241
Figure 47 shows that the model predictions for activity concentrations of $^{241}$Am in molluscs are about a factor of 20 times greater than the measurements for winkles. Ninety-nine percent of the modelled activity concentrations are due to discharges from the Dounreay site, with other sites contributing less than one percent. The differences between the model predictions and measurement data are inconsistent with predictions at the Sellafield site, which are much lower than the measurement data in molluscs for this radionuclide. The differences at the Dounreay site may be due to an overestimate of the discharges of $^{241}$Am before 2000. Discharges of $^{241}$Am prior to 2000 were obtained from the ‘total alpha’ category presented in the RIFE series of reports by applying a breakdown of alpha discharges by individual radionuclide (Smith et al., 2002). Whereas, values for the discharge for alpha-emitters between 2000 and 2003 were obtained directly from the Dounreay site (David Lord, 2004). The site also reported that americium was discharged at a lower proportion of alpha discharges than plutonium, while the opposite had been assumed for the pre-2000 discharges. An overestimate of past discharges for $^{241}$Am would mean that the model would overestimate activity concentrations in filtered water and hence activity concentrations in biota for recent years, since most of the activity is due to remobilisation from sediment.
6.2 Comparison of model predictions with measurements for a single sample type at different locations for 1997-2002

This section considers examples which show the variation of activity concentration over distance due to discharges from different sites.

6.2.1 Sellafield

6.2.1.1 Tritium

Figure 48 shows the variation in measurements and predictions of activity concentrations of tritium in filtered water with distance from the Sellafield site for 2002. This shows that the measured activity concentration in water from the local compartment ranges from 7 Bq l\(^{-1}\) at St. Bees to 34 Bq l\(^{-1}\) at Sellafield. A mean activity concentration in local compartment water is presented on the figure with which the MARINA II model prediction compares well. A measured activity concentration in the regional compartment (Cumbrian Waters, see Figure 1) is approximately factor of two lower than the average activity concentration measured in the local compartment. Model predictions generally agree with the measured data when presented in averaged form, but variability can be observed in measurement data between different locations within the model compartment areas.
6.2.1.2 Technetium-99
Figure 49 shows the variation in measurements of activity concentrations of $^{99}$Tc in seaweed with distance from the Sellafield site for 2002, using BNFL data (refer to Figure 1 for compartments). This shows that the activity concentration in seaweed reduces significantly, by a factor of about 15, from the local compartment values at distances up to 30 km from the Sellafield local compartment. Model predictions are generally within a factor of three of the measurement data.

6.2.1.3 Caesium-137
Figure 50 shows the variation in model predictions and measurements of activity concentrations of $^{137}$Cs in sediment with distance from the Sellafield site for 2002, using BNFL and RIFE data. This shows that the measurements are very dependent on sediment type (e.g. sand or silt) and location (estuary or beach), so it is difficult to establish how measurements vary with distance close to the Sellafield site. Additional variability in the measurement data may be introduced due to different sampling and analytical techniques being employed in two laboratories.
Figure 49  Estimated and measured activity concentrations of $^{99}$Tc in seaweed near Sellafield in 2002

Figure 50  Estimated and measured activity concentrations of $^{137}$Cs in sediment near Sellafield in 2002
6.2.1.4 Americium-241

Figure 51 shows the variation in measurements of activity concentrations of $^{241}$Am in molluscs (winkles) with distance from the Sellafield site for 2002, using BNFL and RIFE data. This shows that the measured activity concentrations in winkles ranges from 9.3 Bq kg$^{-1}$ to 39 Bq kg$^{-1}$ in the Sellafield local compartment, whereas in the Cumbrian Waters compartment measurements range from 13 to 15 Bq kg$^{-1}$. In comparison with the additional measurement data, model predictions are still low for the reasons given in Section 6.1.

Figure 51 Estimated and measured activity concentrations of $^{241}$Am in molluscs near Sellafield in 2002

6.2.2 Dounreay

6.2.2.1 Plutonium-239/240 and americium-241

Figure 52 and Figure 53 show the variation in measurements of activity concentrations of $^{239/240}$Pu and $^{241}$Am in molluscs (winkles) with distance from the Dounreay discharge point for 2001, using UKAEA and RIFE data. All measurements are located within the Dounreay local compartment of the MARINA II model. For the UKAEA data, this shows that the measured activity concentration in molluscs reduces by about a factor of about two at distances over 5 km from the discharge compared with the measurements taken within 5 km of the discharge point. Using the additional measurement data, model predictions are still high for the reasons given in Section 6.1.1.
Figure 52 Estimated and measured activity concentrations of $^{239/240}$Pu in molluscs near Dounreay in 2001

Figure 53 Estimated and measured activity concentrations of americium-241 in molluscs close to Dounreay in 2000
6.3 Summary

In summary, the predicted activity concentrations from the MARINA II model for the different sites were compared with measurements from these regions. Model predictions compared well in some cases and the following observations were made which aided the refinement of the models.

a The model predictions for activity concentrations in filtered water compared well with measured data except for $^{241}$Am for Sellafield and $^3$H at Dounreay. The differences were thought to be mainly due to measurement data which were at the limit of detection. The differences observed for americium may also be due to uncertainties in parameters for sedimentation modelling.

b Discharge data for $^{241}$Am from the Dounreay site may be overestimated before 2000, resulting in an overestimate in predicted activity concentrations in molluscs.

c Model predictions for activity concentrations of $^{241}$Am in molluscs (winkles) were low compared to measured data for Sellafield, possibly due to a combination of a low mollusc concentration factor and underestimation of activity concentrations in filtered water.

d Model predictions for concentrations of $^{239/240}$Pu in molluscs (winkles) were high compared with measured values for Northern Ireland and Dounreay, possibly due to the use of a high mollusc concentration factor.

e Model predictions for concentrations of $^{99}$Tc in crustaceans were high compared to measurements in crabs and low compared to measurements in lobsters at Sellafield and Wylfa sites, due to the choice of the concentration factor.

f Model predictions for concentrations of $^{99}$Tc in seaweed were high compared to measurements in seaweed for Northern Ireland. This is probably due to local effects, since Figure 49 shows a good comparison of measurements with model predictions in the Sellafield area, and hence a reasonable estimate for the seaweed concentration factor. Although the two comparisons use different sets of data, (BNFL and CEFAS).

g Model predictions for concentrations of $^{137}$Cs in molluscs were low compared to measurements in some species of mollusc for Sellafield, due to the choice of the concentration factor.

7 REFINEMENT OF THE MARINA II MODEL

7.1 Introduction

The previous study (Jones et al., 2003) summarised where the MARINA II model was able to accurately represent point measurements in location and time
and, more importantly, highlighted specific instances where the model was not able to reproduce these measurements. Section 4 of the present study suggested changes to both the sediment distribution coefficients (K_d$s) and the concentration factors, as given in the published literature. The K_d$s were altered for $^{137}$Cs, $^{239/240}$Pu and $^{241}$Am; the reasons for choosing these radionuclides and the justification for where these changes were implemented are given below. The concentration factors were either revised generically (for example, molluscs) or made species-specific (for example, winkles) depending on the available literature. The effects of implementing each these changes to the MARINA II model are described separately in the following sections.

7.2 Changing the Sediment Distribution Coefficients

One indicator to suggest that a change in K_d is required is a markedly poorer fit to activity concentrations measured in either filtered seawater or sediment observations. The comparisons of $^{137}$Cs in (Jones et al., 2003) indicated that the model could be improved at Sellafield (seawater underestimated and sediment underestimated) and at Wylfa (seawater underestimated). However, the contribution of $^{137}$Cs measured at Wylfa is dominated by the discharges from Sellafield (see Section 6.1.3). Therefore, it would appear that, in the model, $^{137}$Cs is not sufficiently mobile for these two sites, that is the K_d is too high. Figure 54 and Figure 55 show the changes to the predicted activity concentrations in filtered water and sediment respectively due to a change in K_d from 230 Bq kg$^{-1}$ per Bq l$^{-1}$ to 100 Bq kg$^{-1}$ per Bq l$^{-1}$ (see Table 21) for Sellafield discharges only.

![Figure 54](https://example.com/image54.png)

**Figure 54** Estimated and measured activity concentrations of $^{137}$Cs in filtered seawater near Sellafield. Model estimates are calculated using MARINA II (solid line) and revised (dotted line) K_d$s
The revised $K_d$ gives a marginally worse fit to the measurements of activity concentration in filtered water and also a worse average fit to the measurements taken in sediment. However, as might be expected from using a $K_d$ specific to the Sellafield beach area, the model compares better to measurements of activity concentrations in sand, rather than those in mud. The marginally poorer fit to activity concentrations in filtered water is translated directly to a marginally poorer fit to activity concentrations in molluscs, the indicator used in the model validation (Section 6), as shown in Figure 61. It is possible to improve the fit by decreasing the flow rate to this compartment in the model. However, such a change would compromise the validation of all of the radionuclides used since flow rates have nominally been optimised previously.

The effect of the $K_d$ change on modelled activity concentrations in seawater at Wylfa is shown in Figure 56. Again it can be seen that the model provides a worse fit than the original $K_d$ value, although neither follows the temporal trend particularly well during the five-year period between 1997 and 2002.
Figure 56 Estimated and measured activity concentrations of $^{137}$Cs in filtered seawater near Wylfa. Model estimates are calculated using the MARINA II (solid line) and revised (dotted line) $K_d$ for discharges from Sellafield.

Figure 57 shows the activity concentrations in filtered seawater taken around the Wylfa site at Holyhead and further away at Llandudno (approximately 50 km East of Wylfa) and Prestatyn (approximately 75 km East of Wylfa). All of these measurements were taken by a single organisation (CEFAS) using consistent sampling and analytical techniques and therefore any measurement biases are likely also to be constant over the five-year period. It is likely therefore that the observed decreases in activity concentration, which is present in all three data sets despite the distance between them but which is not reproduced within the model, is in fact a real phenomenon. It maybe that the mechanism which produced this build-up in the environment is local to the Wylfa coastal area and would therefore not be included in a generic model, such as MARINA II.
For $^{137}$Cs therefore, the change in $K_d$ for discharges from Sellafield does not appear to have a positive effect on the comparison to measurements for the indicators that were checked.

The $K_d$s for $^{239/240}$Pu and $^{241}$Am were also changed from the MARINA II values of $1 \times 10^5$ and $2 \times 10^6$ to $1.8 \times 10^4$ and $1.9 \times 10^5$ Bq kg$^{-1}$ per Bq l$^{-1}$, respectively, as given in Table 21. These changes were designed to test measured Sellafield-specific values in the context of the indicators being used in this study. Since all of the modelled activity concentrations at Northern Ireland are derived from discharges at Sellafield, the change in $K_d$ was tested in molluscs at this location. The measured and predicted activity concentrations are shown in Figure 58.

The change in $K_d$ clearly has a negative effect, increasing the difference by nearly a factor of four. The implication of reducing the $K_d$ is that $^{239/240}$Pu would be more freely able to propagate through the water compartments of the model and hence disperse more widely throughout the North European waters. However, it is impossible to ascertain whether this leads to an overall improvement in model estimations without significant further validation by compartment.

Figure 59 and Figure 60 illustrate the comparison of measured and calculated activity concentrations of $^{241}$Am in filtered seawater and molluscs respectively, near the Sellafield site. In both cases, the change in $K_d$ leads to a positive comparison with the activity concentrations that were measured. However, as with $^{239/240}$Pu, this merely suggests a better comparison in one compartment and gives no indication of how the model changes overall.
Figure 58 Estimated and measured activity concentrations of $^{239/240}$Pu in molluscs for Northern Ireland. Model estimates are calculated using the MARINA II (solid line) and revised (dotted line) $K_d$ for discharges from Sellafield.

Figure 59 Estimated and measured activity concentrations of $^{241}$Am in filtered seawater for the Sellafield coastal area. Model estimates are calculated using MARINA II (solid line) and revised (dotted line) $K_d$s. NB – measurements at the limit of detection presented as unfilled points.
Figure 60 Estimated and measured activity concentrations of $^{241}\text{Am}$ in molluscs near Sellafield. Measurements were taken at Nethertown. Model estimates are calculated using MARINA II (solid line) values, a revised concentration factor for molluscs (dashed line) and a revised $K_d$ (dotted line).

It is worth noting that the measurements shown in Figure 59 were taken by a different organisation (BNFL) to those reported in this section of the study, since no appropriate data were available in the RIFE reports. These measurements are therefore likely to have different errors and biases, limits of detection, statistical significance and be taken at different locations and/or times to those reported in RIFE. In the absence of these details however, the measurements are assumed to be no more or less valid than those taken by CEFAS, although a consistent data set would have been more preferable.

### 7.3 Changing the Concentration Factors

The most recent survey of the literature (see Section 4) shows concentration factors to vary by up to two orders of magnitude compared to MARINA II values. It has also been reported that for $^{95m}\text{Tc}$, the concentration factor varied with time below the equilibrium whole-body concentration factor (Swift, 2001). It is therefore possible that measured and published concentration factors vary because the sampled biota were not at this equilibrium point. In addition, species-specific concentration factors have been reported whereas generic values are used with the MARINA II model. To test the significance of these new data, concentration factors were varied, where data exists, and compared to MARINA II values.
7.3.1 Molluscs

Figure 61 Estimated and measured activity concentrations of $^{137}$Cs in molluscs near Sellafield. Measurements were taken at Nethertown. Model estimates are calculated using MARINA II (solid line), values, a revised concentration factor for molluscs (dashed line) and a revised $K_d$ (dotted line).

Figure 61 illustrates the effect of varying the concentration factor for $^{137}$Cs in molluscs from 0.03 to 0.05 Bq kg$^{-1}$ per Bq m$^{-3}$ as suggested in Table 22. The change in value and improvement in this case is small compared to the variation for type of mollusc. In these data sets, the measurements for winkles are almost a factor of three greater than those for mussels. However, the temporal variation, over the five-year period shown here, appears quite stable. It is also worth noting that the errors on the measurements for winkles allows for considerable uncertainty in the actual activity concentrations. No error values were reported for the measurements for mussels.

Figure 60 shows the effect of changing the concentration factor for $^{241}$Am in molluscs from 20 to 3 Bq kg$^{-1}$ per Bq m$^{-3}$ as suggested in Table 22. In this case the revised value in Table 22 does not improve the fit of the model to the measurements. However, it can be seen in Figure 37 that the variation of activity concentration between species of molluscs for $^{241}$Am can be as great as a factor of ten, with temporal variations as great as 40%. It is possible that these variations arise from variations in uptake (either natural or as a function of age and gender of biota) or are time-delayed responses to prior changes in discharges. Such time delays would also be dependent on species.
Figure 62 Estimated and measured activity concentrations of $^{241}$Am in molluscs near Dounreay (Sandside Bay). Model estimates are calculated using the MARINA II (solid line) and revised (dashed line) concentration factor for molluscs.

Figure 62 illustrates the effect of the same change in concentration factor but at the Dounreay site. In this case, the improvement in the comparison of the model to measurements of activity concentrations in molluscs is dramatic. However, it is interesting to note that, although the sources of $^{241}$Am at Sellafield and Dounreay are very different, the accumulated values during this time period are approximately the same. The large difference in the measurements of winkles, taken by the same organisation (CEFAS), is therefore likely to have a real physical interpretation and illustrates real variation as a function of environment and location.

7.3.2 Crustacea

Specific references were found for concentration factors of lobsters, Norwegian lobsters (Nephrops) and crabs. Although traditionally the MARINA II model has been used to calculate activity concentrations in generic "crustacea" as part of dose calculations to individual and collective populations, the specific concentration factors of 0.72 Bq kg$^{-1}$ per Bq m$^{-3}$ for lobster flesh; 9 Bq kg$^{-1}$ per Bq m$^{-3}$ for lobster tail; 1.4 Bq kg$^{-1}$ per Bq m$^{-3}$ for nephrops; and 0.14 Bq kg$^{-1}$ per Bq m$^{-3}$ for crabs given in Table 22 were used to calculate species-dependent activity concentrations, as shown in Figure 63 and Figure 64.
Figure 63 Estimated and measured activity concentrations of $^{99}$Tc in crustacea near Sellafield. Measurements are taken at the Sellafield coastal area. Model estimates are calculated using species-specific concentration factors.

Figure 64 Estimated and measured activity concentrations of $^{99}$Tc in crustacea near Sellafield. Measurements were taken in the Sellafield coastal area. Model estimates are calculated using MARINA II (solid line) and revised, species-specific (dashed line) concentration factors.
In both figures, it is clear that using species-specific concentration factors provides a better approximation to the measured activity concentrations in lobsters, nephrops and crabs for $^{99}$Tc in this instance. For lobsters the concentration factor for lobster flesh provides a better estimate than that for the abdominal muscle (tail) alone. It is also worth noting that the activity concentrations, calculated using the generic "crustacea" concentration factor (labelled MARINA II in Figure 64), varies between 300 and 550 Bq kg$^{-1}$ and provides a good average estimate for all three types of crustacea.

Figure 65 shows the estimated and measured activity concentrations of $^{99}$Tc in crustacea around the Wylfa coastal area. Again the species-specific concentration factor provides a better estimate to the point measurements of activity concentration in crabs than the generic concentration factor. In both of these locations, however, the choice of radionuclide has been limited to $^{99}$Tc; this result should therefore not be generalised to other radionuclides without further work. Typically, much smaller differences are seen in activity concentrations of other radionuclides between crustacean species.

7.3.3 Seaweed

For this comparison, an average, dry weight concentration factor for seaweed, of 25.1 Bq kg$^{-1}$ per Bq m$^{-3}$ for $^{99}$Tc, was calculated based on the average of the revised literature values given in Table 22 compared with the MARINA II value of
30 Bq kg\(^{-1}\) per Bq m\(^{-3}\). This includes the conversion to an equivalent, wet weight concentration factor using a conversion factor for seaweed of 0.2 (IAEA, 1985). The estimated and measured activity concentrations of \(^{99}\)Tc in seaweed at Northern Ireland are shown in Figure 66.

![Figure 66 Estimated and measured activity concentrations of \(^{99}\)Tc in seaweed for Northern Ireland. Model estimates are calculated using the MARINA II (solid line) and revised (dashed line) concentration factor for seaweed.](image)

The change in concentration factor gives a better approximation to measured activity concentrations, although calculated values are still much higher than measured values.

7.3.4 Derived Concentration Factors

One outcome from this study was the clear lack of available data for particular radionuclides and species of biota. Preliminary work was hence carried out to test whether or not it was possible to derive concentration factors using measurements of activity concentration in filtered seawater and in biota.

Measurements taken by CEFAS of activity concentrations of \(^{137}\)Cs in filtered seawater, winkles, mussels and limpets at St. Bees were used to derive concentration factors for these biota. Values calculated were 0.20 Bq kg\(^{-1}\) per Bq m\(^{-3}\) for winkles and 0.12 Bq kg\(^{-1}\) per Bq m\(^{-3}\) for both mussels and limpets. The calculated activity concentrations in these biota were then compared to measured activity concentrations of biota, measured by CEFAS but in the Sellafield coastal area. The comparison is shown in Figure 67.
Distinguishing between current & historic radioactive discharges to the marine environment – further work to validate models and quantify uncertainties

Figure 67 Estimated and measured activity concentrations of $^{137}$Cs for molluscs near Sellafield. Model estimates are calculated using the MARINA II (solid line) and derived, species-specific (dashed line) concentration factors.

The derived concentration factors typically give higher activity concentrations than the measured values. This is may be because the derivation is inaccurate: for example, the activity concentrations in filtered seawater may have been measured at different times in the year, and therefore different geographical locations also, to the corresponding measurements in biota. However, it is also possible that the derivation does not work well in this compartment or for these measurements (the Sellafield coastal area incorporates different physical environments, for example sandy beaches and silt-rich regions). These point measurements may not be accurately represented in this case using compartment-averaged activity concentrations. Further work to ascertain the feasibility of such an approach is clearly necessary, should species-specific concentration factors be required.

7.4 Conclusion

It is difficult to draw definite conclusions from comparisons of single point measurements in location and time to the compartment and annual averaged model calculations from MARINA II, since local features will not be represented in the model and biases will undoubtedly be introduced. However, with this caveat in mind, it is possible to suggest that the effects of changing the $K_d$ value of $^{137}$Cs and $^{239/240}$Pu generally lead to poorer model fits to measurements of activity concentrations in filtered water and biota for the locations considered on the basis of the comparisons presented. A better comparison was gained for activity concentrations in sediment at Sellafield and this result could be exploited in the model more fully if site- or compartment-specific $K_d$ values were known, or possibly be estimated from the composition of the sediment at each location (assuming this was known). The change in $K_d$ for $^{241}$Am led to improved...
estimates of activity concentrations in molluscs at Sellafield. However, it is unclear from this study whether or not this improvement would be seen throughout the model, that is, in other compartments. Further work would be required to test this.

The change in concentration factors, and the inclusion of species-specific values from recent literature publications, led to improved fits to measured activity concentrations in biota in all cases, with the exception of $^{241}$Am in molluscs at Sellafield. However, in many cases, the improvement was marginal. Further work to validate these values should be undertaken for different compartments, however, before these values can be adopted.

No clear trend in variability of activity concentration in biota by species was observed for all radionuclides. However, measurements will undoubtedly show a combination of natural variation (including variation by age and gender) and variation by uptake, which depends on whether or not each sampled biota had reached an equilibrium value. It is likely to be very difficult to distinguish these effects practically.

Additionally, it was possible to estimate concentration factors for three types of molluscs, based on separate measurements of activity concentrations in both filtered seawater and biota. The values derived were found to be a factor of between two and three greater than the revised generic concentration factor for molluscs. The subsequent comparison to measured activity concentrations were shown to be poorer than those comparisons using published site-specific values but reasonable as a first approximation.

Finally, the MARINA II model has been shown in previous studies to be an effective tool for radiation protection calculations on average. In this study, it has been shown that revising the concentration factors, where appropriate data are available, may further enhance this capability for some species.

### 8 IMPLICATIONS ON OSPAR TARGETS

This study has reviewed the literature for revised site-specific data to be used in support of modelling the activity concentrations for the period 1997 to 2002. The effects of adopting these revised values in MARINA II were reviewed in the previous section in comparison with measurement data and conclusions drawn as to the effectiveness of the changes. In this section, the implications of these changes on the modelling of activity concentrations to 2030 have been examined for two radionuclides discharged from the Sellafield site: $^{239/240}$Pu and $^{241}$Am (plus in-growth from $^{241}$Pu) for which revised modelling parameters have been derived. In all cases, activity concentrations due to weapons testing fallout contributes less than one percent and have therefore not been presented.

The same approach as the previous study (Jones et al., 2003) was used to estimate annual discharges of the two radionuclides (plus $^{241}$Pu) to 2020. This
was calculated by taking the ratio of the specific radionuclide discharge in 2000 to the discharge by aggregated category (e.g. total alpha) estimated in the UK Discharge Strategy for 2000. The calculated ratio was then applied to subsequent UK Discharge Strategy estimations up to 2020.

### 8.1 Plutonium-239/240

Although the revised K\textsubscript{d} value for \textsuperscript{239/240}Pu gave a worse fit to the measurements for the Northern Ireland location in the previous section, an improved fit was found for molluscs near the Sellafield site. This is shown in Figure 68. This shows the importance of fully validating changes to modelling parameters before adopting them throughout the model. The revised modelling predicts activity concentrations approximately a factor of three higher than those predicted in the previous study. The overall trend of activity concentrations decreasing from 1998 to 2030 is however unchanged. Discharges during 2002 were higher than those estimated by the UK Discharge Strategy, hence the slight increase in activity concentrations during 2002. Measurement data, as used in the previous study, are included for comparison purposes.

**Figure 68 Estimated and measured activity concentrations of \textsuperscript{239/240}Pu for molluscs in the Sellafield coastal area using MARINA II model parameters from the current and previous studies for estimated discharges to 2020 based on the UK Discharge Strategy**
8.2 Americium-241

The effects of the reduced K_d for ²⁴¹Am in the Irish Sea is shown for molluscs near the Sellafield site in Figure 69. The revised modelling predicts activity concentrations approximately a factor of eight higher than those predicted for the previous study. The overall trend of activity concentrations decreasing from 1998 to 2030 is again unchanged. Measurement data from the current study, for a variety of species, are included for comparison purposes.

![Figure 69 Estimated and measured activity concentrations of ²⁴¹Am for molluscs in the Sellafield coastal area using MARINA II model parameters from the current and previous studies for estimated discharges to 2020 based on the UK Discharge Strategy](image)

The effects of the changes to the K_d of ²⁴¹Am have also been investigated for molluscs at the Wylfa and Northern Ireland indicator locations as shown in Figure 70 and Figure 71. The previous study found that, in both cases, the activity concentrations in 2020 were greater than those predicted for 1998. The changes result in the peak activity concentration being predicted in 2004 for Wylfa and 2008 for Northern Ireland. In both cases, the predicted activity concentrations using the revised K_d are between a factor of two or three higher than the values estimated in the previous study. The changes are due to the reduced K_d value increasing the activity present in the water column (due to direct discharge and remobilisation) and therefore the transfer across the Irish Sea occurs over a shorter time period, with the peak activity concentrations occurring earlier.
Figure 70 Estimated activity concentrations of $^{241}$Am for molluscs at Wylfa using MARINA II model parameters from the current and previous studies for estimated discharges to 2020 based on the UK Discharge Strategy.

Figure 71 Estimated activity concentrations of $^{241}$Am for molluscs in Northern Ireland using MARINA II model parameters from the current and previous studies for estimated discharges to 2020 based on the UK Discharge Strategy.
8.3 Overall implications

Two radionuclides (\(^{239/240}\)Pu and \(^{241}\)Am) were modelled following the UK Discharge Strategy estimates up to 2020 using revised \(K_d\) values as considered in Section 7. In both cases, the predicted activity concentrations were greater than those presented in the previous study, however the overall trend of activity concentrations decreasing to 2020 was observed. For \(^{241}\)Am, the effects of the changes at Wylfa and Northern Ireland were studied. For both of these locations, the predicted activity concentrations were estimated to increase from those calculated in the previous study. In addition, the activity concentrations peak prior to 2020 and are estimated to have decreased below 1998 activity concentrations by this time.

9 CONCLUSIONS

The project investigated, in further detail, the findings from a previous study to distinguish between the impacts of historical and current discharges of radioactivity to the UK marine environment. This study had two main objectives:

a. To more accurately validate the models used in the previous study against environmental measurements
b. To improve the quantification of uncertainties and determination of natural variability associated with measurements in the marine environment

The period 1997 to 2002 was selected to allow greater focus on the validation and uncertainty tasks. In addition, specific combinations of radionuclide, location and environmental media were chosen to restrict the scope of the study. These combinations were selected on the criteria of those comparisons where greatest difference between measurements and model predictions were observed in the previous study.

To fulfil the two objectives, a series of measurement data were compiled from the RIFE dataset for the selected indicator combinations. These data were provided in greater detail than for the previous study, in that species, time within the year, and specific locations were clearly identified.

The measurement data display both natural variability and systematic variability. Natural variability can be seen between years for which there was little change in annual discharges. Further review of variation within a year was undertaken for measured activity concentrations of \(^{99}\)Tc in crabs and lobsters near Sellafield. The measurements showed fluctuation of up to a factor of three from the mean within a year, however given the relatively small number of measurements, actual variation may be higher. Differences can also be seen between species in the same taxonomic group. From the measurements available, these differences vary from several orders of magnitude for \(^{99}\)Tc in crustacea to around one order
of magnitude for $^{137}$Cs in molluscs, although true variation may be higher. In general, the variability between species within a taxonomic class was similar to the variability by location within a few tens of kilometres, at around one order of magnitude based on the measurements available for this study.

The high-resolution CSERAM output generated for the previous study was reviewed in greater detail to examine the spatial and seasonal variability. Variability in the model is due to the physical processes being modelled on an hourly basis, such as meteorological data and tides. The spatial resolution of the model enables differences to be investigated between locations a few kilometres apart. The level of variability in the CSERAM model was similar spatially to that predicted seasonally. As can be expected, this variability was several factors less than that seen in the measurement data. This is due to additional factors affecting measurement variability that are not represented in the model, such as sampling, analysis and biological factors. It should be emphasized that there are inherent uncertainties in both CSERAM and MARINA II due to the inability to fully represent and quantify the processes in the physical environment. These uncertainties are much greater than the variability demonstrated in model runs.

Further validation of the MARINA II model was carried out for the period 1997 to 2002. This validation compared model output with the compiled data detailing locations and species of specific samples. From the validation, several areas were identified for further investigation. These include the relatively poor comparison of the model to measurement values for $^{241}$Am (model estimates generally lower than the measurement values at Sellafield), $^{99}$Tc (for which the model did not represent the clear inter-species differences) and $^3$H and $^{239/240}$Pu at Dounreay. Some local factors were considered to result in differences between the model predictions and measurement data. For example, MARINA II model predictions were compared with measurements of activity concentrations in seaweed harvested from inlets on the Northern Ireland coast. These comparisons showed the model to predict values higher than the measured values. The model does not represent the inlets, but provides an average value for the Northern Ireland coastline that could result in local differences between predictions and measurements.

Changes were made to MARINA II modelling parameters following a literature review of local data on sediment distribution coefficient ($K_d$) and concentration factor (CF) values. Such changes were investigated for the specified locations, although further validation would be required to adopt the values more generally. Improvement in the model predictions were seen after application of a revised $K_d$ for $^{241}$Am. The changes to CFs resulted in improved modelling of biota in all cases except for $^{241}$Am in molluscs. As a result of the changes, the model predictions all fall within a factor of ten of the measurements with the majority within a factor of three. These changes have been applied to the discharges described in the UK Discharge Strategy for the Sellafield site up to 2020. The activity concentrations of $^{239/240}$Pu in molluscs at Sellafield show a similar decreasing trend as observed in the previous study, however the absolute values are approximately three-times higher. For $^{241}$Am in molluscs near Sellafield, the changes again resulted in a similar trend to the previous study, but the absolute
activity concentrations were approximately a factor of eight higher than the previous study. The effects of the changes to the $K_d$ for $^{241}$Am were investigated at the Wylfa and Northern Ireland locations, as the previous study suggested the activity concentrations slowly increased to 2020 and beyond. With the proposed changes to the $K_d$, the activity concentrations peak in around 2004 for Wylfa and 2008 for Northern Ireland. For both sites, the MARINA II model predicts lower activity concentrations in 2020 than present in 1998. This differs from the previous study, in which activity concentrations were predicted to be higher in 2020 than 1998.

In summary, the study showed that the natural variability in activity concentrations measured in the marine environment is present both temporally and spatially. Based on the limited measurement data used in this study, the combination of uncertainty and variability is likely to lead to differences between model predictions and measurements which are mostly within a factor of three and up to a maximum of a factor of ten. It is important to bear these differences in mind when model predictions are compared with measurement values as the general trend of the measurements should be taken into account rather than individual values. The MARINA II model predicts activity concentrations on an annual average basis and the spatial resolution is of the order of tens of kilometres. For example, in the vicinity of Sellafield, four sampling locations were identified which fall within the same modelling compartment. It is important to ensure that a consistent approach is adopted to select sampling locations consistent with the modelling and with the approaches used to determine baseline values for comparison with the OSPAR strategy.

Differences between individual species within each taxonomic class were seen during the validation exercise. This can, in some instances, be modelled using an species specific concentration factor such as for the case of $^{99}$Tc in lobsters and crabs. However, this can only be achieved where there are sufficient published data on the subject. An alternative approach is to derive concentration factors for individual species based on measurement data as a first approximation. The implication of these differences between species for the OSPAR strategy is to ensure that a consistent choice of indicator species is made in determining baselines, and for future comparison against baselines. This choice should not necessarily be one species, but could be a defined approach for averaging across selected species consistently throughout the OSPAR region.

The predicted activity concentrations in 2020 are lower than those estimated for 1998 in all cases considered. Changes made to modelling parameters indicate that the contribution from pre-1998 discharges would be slightly greater in 2020 for $^{239/240}$Pu and $^{241}$Am than estimated previously. A conclusion of the previous study that discharges of $^{239/240}$Pu and $^{241}$Am prior to 1998 are likely to remain the major contributor to activity concentrations in 2020, is strengthened. Overall, the findings of this study therefore support the conclusions made in the previous study regarding ‘close to zero’ and ‘historical levels’.
10 REFERENCES


