Natural Radionuclides in Seafood

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Summary

For some time there has been a growing awareness of the radiological significance of natural radionuclides, particularly ^{210}Po, in marine foodstuffs. In the UK, most attention has, historically, been focussed upon fish and shellfish consumers in the vicinity of Whitehaven, where there are known anthropogenic inputs of natural radionuclides. An accurate knowledge of the ambient baseline concentrations of natural radionuclides is, however, lacking. This investigation was undertaken in an attempt to gain a better understanding of the extent of natural variation in seafood obtained from UK waters.

Samples of commonly consumed seafood were obtained over a three year period from a number of locations around the UK, taking care to avoid areas where natural radioactivity might be enhanced by anthropogenic inputs. Concentrations of ^{210}Pb, ^{226}Ra, U and Th radionuclides in the edible fractions were much lower than those for ^{210}Po. Median ^{210}Po concentrations in individual species ranged from 0.42 Bq kg^{-1} wet weight in cod up to 38 Bq kg^{-1} wet weight in mussels (i.e. a variation of ~90 fold) indicating that the dose to man resulting from seafood consumption is extremely sensitive to variations in diet. ^{210}Po concentrations increased in the order cod ~ whiting < plaice ~ *Nephrops* < lobster ~ shrimp ~ whelk ~ limpet < winkle ~ crab < cockle < mussel. There were, however, marked variations in levels of ^{210}Po between individual samples of the same organisms (typically by ~ 4 fold and up to 26 fold for shrimps).

Assessment of the data to account for the observed variability was difficult. This is because the processes controlling the extent of radionuclide bioaccumulation are complex. Some of the factors, which might be controlling uptake behaviour, have been identified (e.g. different environmental conditions between individual sampling sites, feeding behaviour and seasonal cycling).

The absence of a detailed understanding of the processes and mechanisms controlling ^{210}Po bioaccumulation is of practical, as well as academic, significance. It means there is a lack of any clearly defined framework for a radiological assessment. For example, information from this relatively small scale programme indicates that the practice of using single generic UK wide values for 'background' concentrations of naturally occurring radionuclides in seafood in the RIFE report appears highly questionable. The uncertainties associated with the data should be made more transparent using the range of values derived here (see Table 6). The variability associated with the environmental data indicates that any assessment of the impact of anthropogenic inputs requires detailed site-specific interpretation.

1. Introduction

There has been a growing awareness since the mid 1980's of the radiological significance of natural radionuclides, particularly ^{210}Po, in marine and terrestrial foodstuffs (McDonald *et al*., 1986; Pentreath and Allington, 1988; Carvalho, 1995a; Aarkrog *et al*., 1997; Pollard *et al*., 1998). High bioaccumulation of ^{210}Po in certain marine organisms has been known for many years (Hoffman *et al*., 1974; Heyraud and Cherry, 1979) but it is only in the last decade or so, in estimating dose to man, that concentrations in commonly consumed items of fish and shellfish have become of greater interest (Pentreath *et al*., 1989). On a daily diet of 600 g of fish and 100 g each of crustaceans, molluscs and seaweed (all consumed fresh), an annual dose of 2 mSv would be received from naturally occurring radionuclides (^{210}Po, ^{210}Pb, ^{40}K, ^{87}Rb, ^{226}Ra, U nuclides, ^{14}C) of which 75 % would be attributed to ^{210}Po (Pentreath and Allington, 1988). More recently, a detailed analysis of ^{210}Po and ^{210}Pb in the actual diet of the Portuguese population indicated that the average ingestion rate was 1.2 and 0.47 Bq day^{-1} per capita for ^{210}Po and ^{210}Pb, respectively (Carvalho, 1995a). Seafood was estimated to contribute up to 70% of the ^{210}Po ingestion rate, whereas cereals, vegetables, and meat altogether contributed 79% of the ^{210}Pb ingestion rate. Other ^{210}Po and ^{210}Pb sources, namely inhalation of surface air and cigarette smoke, were estimated to contribute only a small percentage of the absorption of these radionuclides in the blood. The average whole body effective doses for the adult from the Portuguese population were estimated to be about 85 μSv annum^{-1} from ^{210}Po and 170 μSv annum^{-1} from ^{210}Pb absorbed with the diet. The effective dose from ^{210}Po in the diet was shown to be extremely variable ranging from 25 μSv annum^{-1} in a person

consuming no seafood to 120 μSv annum^{-1} in a heavy consumer of sardines, to 1,000 μSv annum^{-1} in a heavy consumer of molluscs.

Enhanced concentrations of natural series radionuclides can occur in discharges from non-nuclear industries, particularly wastes released by phosphate fertiliser plants (Poole *et al*., 1995). In the UK, the large volumes of liquid and solid waste discharged into the eastern Irish Sea at Saltom Bay (near Whitehaven) from the Rhodia Consumer Specialities Ltd. (formerly Albright and Wilson) phosphoric acid production plant between 1954 and 1992 resulted in significantly enhanced levels of natural radionuclides in the local shellfish and hence dose to the local seafood consumers (McCartney *et al*., 1992; Rollo *et al*., 1992; Camplin *et al*., 1996). Enhanced natural series radionuclide concentrations have also been shown to occur in the vicinity of fertiliser plants elsewhere, including the Tagus estuary in Portugal (Carvalho, 1995b) and the estuary formed by the Odiel and Tinto river mouths in the south-west of Spain (Bolivar *et al*., 2000).

In the UK in 1992, operations at the Rhodia Consumer Specialities Ltd. phosphoric acid production plant changed and imported crude phosphoric acid rather than phosphate ore became the raw material. These changes resulted in a large reduction in the volume of waste discharged (Poole *et al*., 1995). Subsequent monitoring indicated that concentrations of natural radionuclides in the local environment decreased accordingly (McCartney *et al*., 2000). The committed effective dose to a group of local seafood consumers via this route was estimated to have decreased from 1.0 mSv annum^{-1} in 1993 to 0.19 mSv annum^{-1} in 1997. It was noted that the latter value was still above the expected background level of 0.1 mSv annum^{-1}. Enhanced

radionuclide concentrations in the local seafood were suggested to be maintained by the reduced, but not insignificant, discharges from the plant and by the presence of phosphate ore in the sediment at Whitehaven Harbour (as a legacy of spillage during the unloading of ore from ships).

The Food Standards Agency (FSA) began operation in April 2000. It is responsible for food safety issues in the UK which were previously undertaken by the Ministry of Agriculture, Fisheries and Food (MAFF). Food produced around UK nuclear sites is monitored for radioactivity by the FSA to check that aquatic foodstuffs are not subject to undue radioactive contamination and is a key component of the Governments strategy to protect the safety of the aquatic food chain, UK fisheries and the marine environment. These results are reported in the annual Radioactivity in Food and the Environment (RIFE) series of reports that the FSA jointly publishes with the Scottish Environment Protection Agency (SEPA). As part of this surveillance effort the FSA has maintained a small monitoring programme to study the importance of natural radionuclides in fish and shellfish to critical groups in the vicinity of Whitehaven. In 1997 the total dose to the most exposed group of local seafood consumers attributable to natural radionuclides was estimated to be 0.25 mSv annum^{-1} (MAFF and SEPA, 1998). Dose estimates may, however, vary significantly based on assumed consumption rates and gut transfer factors. The current ICRP advice is that a gut transfer factor of 0.5 is appropriate for dietary intakes of ^{210}Po by adults (ICRP, 1994). However, a study involving the consumption of crabmeat indicated that the gut transfer factor might be as high as 0.8 (Hunt and Allington, 1993). Consequently, using the conservative assumption that the factor of 0.8 applies to all seafood, the

estimate of 0.25 mSv annum^{-1} for 1997 increases to 0.40 mSv annum^{-1}. A very similar value was also estimated for 2000 (0.47 mSv annum^{-1}, FSA and SEPA, 2001).

Estimates of the impact of anthropogenic natural radionuclide inputs require an accurate knowledge of ambient baseline concentrations and the extent of natural fluctuations. Without this information it would be impossible for the UK to fulfil its responsibilities under the OSPAR convention to ensure that "discharges, emissions and losses of radioactive substances are near background values for naturally occurring radioactive substances and close to zero for artificial radioactive substances". This investigation was undertaken in an attempt to obtain the necessary data for UK seafood, with particular relevance to the RIFE programme. Initially, a literature search was carried out to collate existing data and assess that which were relevant to the present project. Furthermore, a preliminary interpretation of the results was made to inform the direction of the subsequent sampling and analytical work carried out in this investigation. Seafood samples were obtained over a three year period from a number of locations around the UK, taking care to avoid areas which might contain levels of natural radionuclides enhanced by technological inputs.

The information arising from this project has provided an improved knowledge of the baseline content of natural radionuclides in fish and shellfish. It has, perhaps unsurprisingly, also highlighted additional gaps in existing knowledge. Nevertheless, information from this project has enabled a number of preliminary recommendations to be made regarding the assessment of data obtained as part of the FSA monitoring programme.

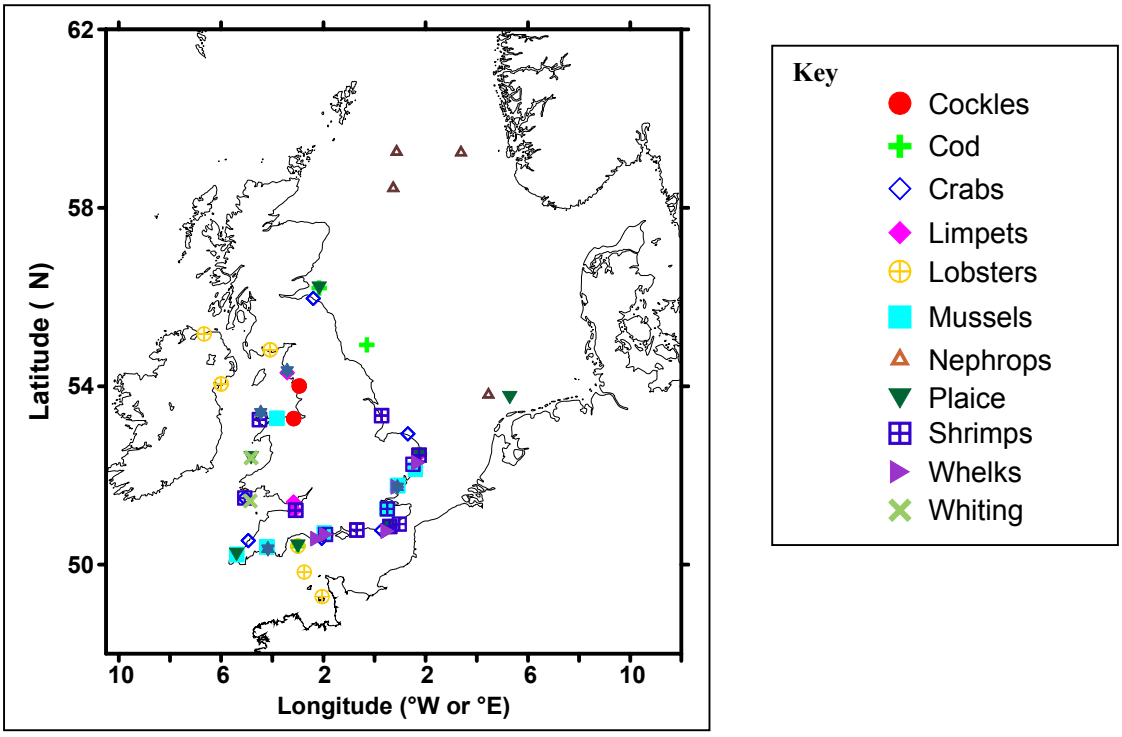
2. Sampling and Analysis

2.1 Sample collection

Samples of twelve different marine species (Table 1) were collected from a wide range of locations (Fig.1) over 3 years, from July 1999 to August 2001. Sampling sites were selected to be distant from known anthropogenic inputs. For a balanced perspective the sampling locations were distributed around the coastline of England and Wales with a small number of additional sites in Northern Ireland, southern Scotland and the North Sea.

Figure 1

Location of individual sampling sites



Catchment geologies from both igneous (e.g. Kirkcudbright on the southern Scottish coastline) and metamorphic regimes (most of England and Wales) were included in the range of sample sites. This is important because, in a survey of radionuclide concentrations from around mainland Britain, McDonald *et al*., 1991 demonstrated that coastal sediments in northern locations contain measurably greater concentrations than those from southern sites. They noted that the difference roughly reflected the geological division of the UK. Although all the sites were selected to be distant from anthropogenic inputs, samples were collected from a site of natural uranium mineralisation (i.e. Carbis Bay a site in Cornwall close to a known outcrop of high-uranium loadbearing rock)

To ensure that samples were representative of the area from which they were collected, approximately 5 kg of fresh live weight, commercially sized individuals of each species were taken from each location. Collection was carried out by local fishermen, fisheries officers, members of the public, and a range of CEFAS staff on field duties. It is acknowledged that it would have been ideal for all sampling to be carried out within a narrow timeframe (one month) to remove the potential for temporal variations to complicate data interpretation (McDonald *et al*., 1991). For practical reasons samples had to be collected throughout the year, although the majority were obtained in summer months.

2.2 Sample preparation

Food preparation practices can have a significant impact upon the radionuclide content, particularly of particulate-associated radionuclides in shellfish (McKay *et al*.,

1997; Jackson and Rickard, 1998). As in the FSA monitoring programme, it was assumed that preparation methods used by critical group consumers do not involve depuration. The processing of samples after collection to separate the edible fraction for radiochemical analysis was carried out using CEFAS Standard Operating Procedures (SOPs), which are accredited to UKAS standards. For clarity, brief details are given below.

Fish samples were generally gutted by the fishermen. All samples were then frozen and dispatched to the CEFAS Lowestoft Laboratory. On receipt samples were allowed to thaw out naturally. Samples were rinsed under a cold running tap to remove fish scales and any extraneous particulate material. They were then filleted. The fillets were again rinsed to remove extraneous material, minced, dried (90°C) to constant weight and ground to produce a homogeneous mixture.

Live crustacean and molluscs were generally dispatched by overnight delivery to the CEFAS Lowestoft Laboratory. With the exception of *Nephrops*, all species considered in this study were cooked (prior to subsequent dissection and analysis) in artificial seawater. Shellfish were dissected to remove all edible material (e.g. in the case of crabs both white and brown meat). Once again, samples were dried (90°C) to constant weight and ground to produce a homogeneous mixture.

An appropriate amount of material was sub-sampled for radiochemical analysis.

2.3 Radiochemical analysis

All of the samples were analysed for ^{210}Po, as this is the single most important contributor to the dose received from ingestion of natural radionuclides in seafood. In addition selected samples were analysed for ^{210}Pb, ^{226}Ra, thorium and uranium nuclides. Decay corrections were applied to account for the time that elapsed between sample collection and analysis. All of the results are given in Bq kg^{-1} for the wet sample, to be consistent with the method used in the RIFE reports. Some results from other investigations have been reported in Bq kg^{-1} dry weight. In order to compare the results with those reported here, it was necessary to make assumptions concerning the dry/wet ratios. Average values for mussels, winkles and prawns, calculated from the samples analysed as part of this programme, were 0.23, 0.29 and 0.25 respectively. It was assumed that it was appropriate to apply these values, in the absence of information for dry/wet ratios.

No counting errors have been quoted for the raw ^{210}Po and ^{210}Pb data (Table 2). This is because environmental variations greatly outweighed counting errors. Very limited numbers of samples were analysed for uranium and thorium radionuclides and ^{226}Ra. Counting errors were high for some samples and therefore the 1σ propagated counting errors associated with the raw radioanalytical data for uranium and thorium radionuclides are also listed in Tables 4 and 5. The quoted uncertainties for ^{226}Ra data (Table 3) are based on multiple extractions of the same sample and represent the $1\ \sigma$ standard deviation associated with replicate measurements

The full details of each radioanalytical procedure are given in CEFAS Standard Operating Procedures (SOPs), which are accredited to UKAS standards. For clarity, brief details are given below.

^{210}Po and ^{210}Pb

Dried samples were weighed and spiked with a ^{209}Po yield tracer and a mixed carrier containing 10mg each of lead and bismuth from which the recovery factor for the ^{210}Pb analysis was derived. Samples were wet-oxidised using a combination of nitric acid and hydrogen peroxide and evaporated to dryness. They were redissolved in 6M hydrochloric acid, filtered (0.45 μm) and again reduced to incipient dryness. Autodeposition onto polished silver disks was accomplished by dissolving samples in 0.5M hydrochloric acid and standing in a plating bath at 90 ± 5°C for 4 hours. The silver disks were removed and α-counted on Si surface barrier detectors. The minimum detectable ^{210}Po activity is about 0.1mBq. Counting errors were generally maintained below 5 % (1σ). Recoveries of the ^{209}Po yield tracer were generally in the range of 75-95 %.

For ^{210}Pb analysis, the remaining plating solutions were stripped of any residual ^{210}Po by anion exchange. Solutions were then re-spiked with ^{209}Po yield tracer and stored for a minimum of three months to allow fresh ingrowth of ^{210}Po from the ^{210}Pb present in the samples. After the ingrowth period expired, solutions were once again analysed for their ^{210}Po content using the method described above in order to derive the ^{210}Pb concentration. Aliquots were removed to determine Pb recoveries by Inductively Coupled Plasma- Atomic Emission Spectrometry (ICP-AES). Recoveries of the stable lead carrier were generally in the range of 75-95 %.

Uranium nuclides

Samples were analysed for the following isotopes of uranium: ^{238}U, ^{235}U and ^{234}U. Dried samples were ashed at 450°C to oxidise organic material. The ashed samples were spiked with ^{232}U yield tracer and wet oxidised using concentrated nitric acid and small quantities of hydrogen peroxide until it was determined that all organic material had been oxidised. Samples were redissolved and filtered (0.45 μm) to remove insoluble residues. As appropriate, two procedures were used to carry out an initial separation of uranium from the bulk of the sample. The first (classical) method was used for large biota samples and involved solvent extraction of uranium from 4 M nitric acid into tributyl phosphate (TBP) in xylene, followed by back-extraction into sodium carbonate solution. The second method was used for small biota samples and involved coprecipitation of uranium onto a mixed ferric hydroxide / phosphate carrier from a boiling solution (Emerson and Young, 1995). Thereafter, a common purification scheme, involving a series of anion exchange separations, was used to separate uranium from remaining radiometric and gravimetric interferences. Finally, stainless steel counting sources were prepared by electrodeposition from a mixed oxalate / sulphate electrolyte plated out for 2 hours at 200 mA. The stainless steel discs were α-counted on Si surface barrier detectors. Counting errors for ^{238}U were generally maintained below 10 % (1σ). The ^{235}U spike recovery was typically in the range 75-85 %. The minimum detectable activity for all three U nuclides was about 0.01 mBq.

Thorium radionuclides

Samples were analysed for the following isotopes of uranium: ^{228}Th, ^{230}Th and ^{232}Th. Dried samples were ashed at 450°C to oxidise organic material. The ashed samples

were spiked with ^{229}Th yield tracer and wet oxidised using concentrated nitric acid and small quantities of hydrogen peroxide until it was determined that all organic material had been oxidised. Samples were redissolved and filtered (0.45 μm) to remove insoluble residues. As appropriate, two procedures were used to carry out an initial separation of thorium from the bulk of the sample. The first method was used for large biota samples and involved coprecipitation of thorium onto calcium oxalate. Careful control over the conditions was required to limit the bulk of the precipitate. The precipitate was then oxidised to calcium oxide by ignition in a platinum crucible. The second method was used for small biota samples and involved coprecipitation of thorium onto a mixed ferric hydroxide / phosphate carrier from a boiling solution (Emerson and Young, 1995). Thereafter, a common purification scheme, involving a series of anion exchange separations, was used to separate thorium from remaining radiometric and gravimetric interferences. Finally, stainless steel counting sources were prepared by electrodeposition from a mixed oxalate / sulphate electrolyte plated out for 5 hours at 200 mA. The stainless steel discs were α-counted on Si surface barrier detectors. Counting errors for ^{228}Th were generally maintained below 10 % (1σ). The ^{229}Th spike recovery was typically in the range 75-85 %. The minimum detectable activity for all three Th nuclides was about 0.01 mBq.

^{226}Ra

^{226}Ra assay was achieved via measurements of its short-lived daughter ^{222}Rn ($t_{1/2}$ ~3.8 days) and associated progeny (^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po; $t_{1/2}$ <30 minutes) using the so-called Rn emanation technique (Lucas, 1957)-it is called an emanation technique because ^{222}Rn exists in a gaseous form. Dried samples were ashed at 450°C to oxidise organic material. The ashed samples were wet oxidised using concentrated

nitric acid and small quantities of hydrogen peroxide until it was determined that all organic material had been oxidised. Samples were redissolved in ~ 75 ml 6 M hydrochloric acid, filtered (0.45 μm) to remove insoluble residues and quantitatively transferred to Dreschel bottles. Sufficient deionised water was added to make the final volume up to 450 ml. Any ^{222}Rn initially present in the Dreschel bottles was swept out with helium and the flasks sealed to allow fresh ingrowth of ^{222}Rn, via decay of ^{226}Ra, to develop over a period of ~7 days. After this time, the activity of ^{222}Rn is ~72 % of the parent ^{226}Ra. The time taken for secular equilibrium to be achieved is ~ 1 month. The Dreschel bottles were connected to a gas extraction vacuum line to allow the in-grown ^{222}Rn to, once again, be swept out with helium and collected on carbon traps at –40°C. The samples were stored to allow freshly ingrown ^{222}Rn to be reanalysed to improve the accuracy of low level measurements. The ^{222}Rn was then transferred into quartz glass scintillation cells when the carbon traps were re-heated to 450°C. The overall extraction and transfer efficiency of this type of Rn extraction system has been reported to be close to 100 % (Mathieu *et al*., 1988). After allowing a period of ~3 hours to elapse to allow ^{222}Rn to achieve secular equilibrium with its very short-lived daughter products, the ^{222}Rn activity (hence ^{226}Ra) was determined by α counting. The uncertainties associated with the ^{226}Ra data were variable ranging from 6-49 %, dependent upon the activity present in individual samples. The minimum detectable activity was about 1mBq. Three samples were below the limit of detection.

3. Results

3.1 Variations in radionuclide accumulation between individual organisms

The raw radioanalytical data are listed in Tables 2-5. These results have been used to calculate the median radionuclide concentrations in all the sampled species (Fig. 2). Assessment of the raw data has been made in terms of median values. It can be misleading to quote a mean value given that i) single high or low values can unduly influence the mean and ii) it is not possible to ascertain from the limited amount of data whether the individual values fit a normal distribution curve. For uranium and thorium nuclides, results have been shown only for ^{238}U and ^{228}Th. To a first approximation data for other uranium and thorium nuclides exhibited similar trends to those shown for ^{238}U and ^{228}Th, respectively. Further details concerning isotopic ratios for these nuclides are given elsewhere.

Figure 2

Median radionuclide concentrations in edible fractions for marine species collected from various UK sites (July 1999-August 2001). Error bars indicate range of values observed in individual samples. For clarity, ranges for selected organisms are listed on the individual figures a) ^{210}Po, b) ^{210}Pb, c) ^{210}Po/^{210}Pb quotient, d) ^{228}Th, e) ^{238}U, f) ^{226}Ra.

Figure 2a

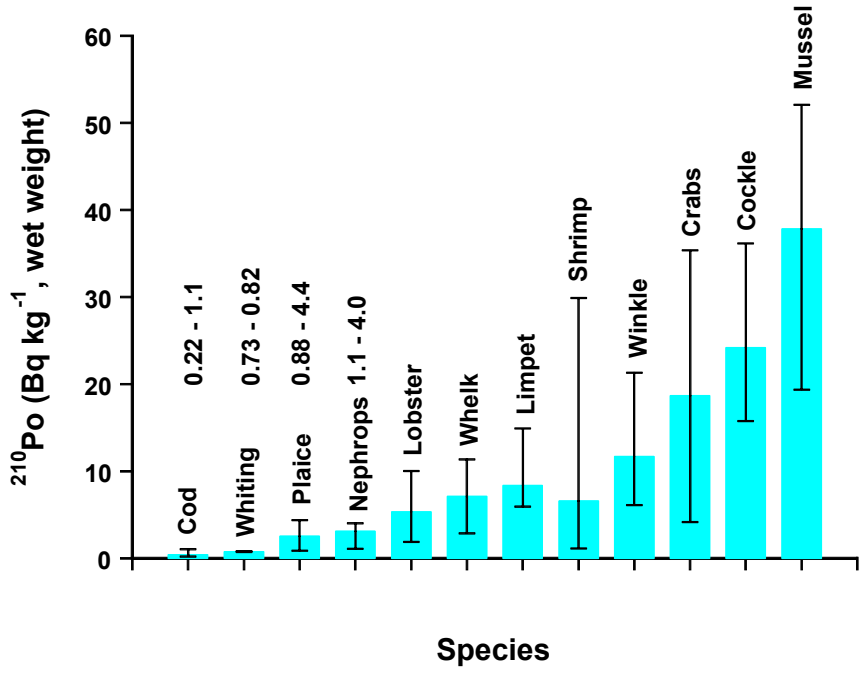


Figure 2b

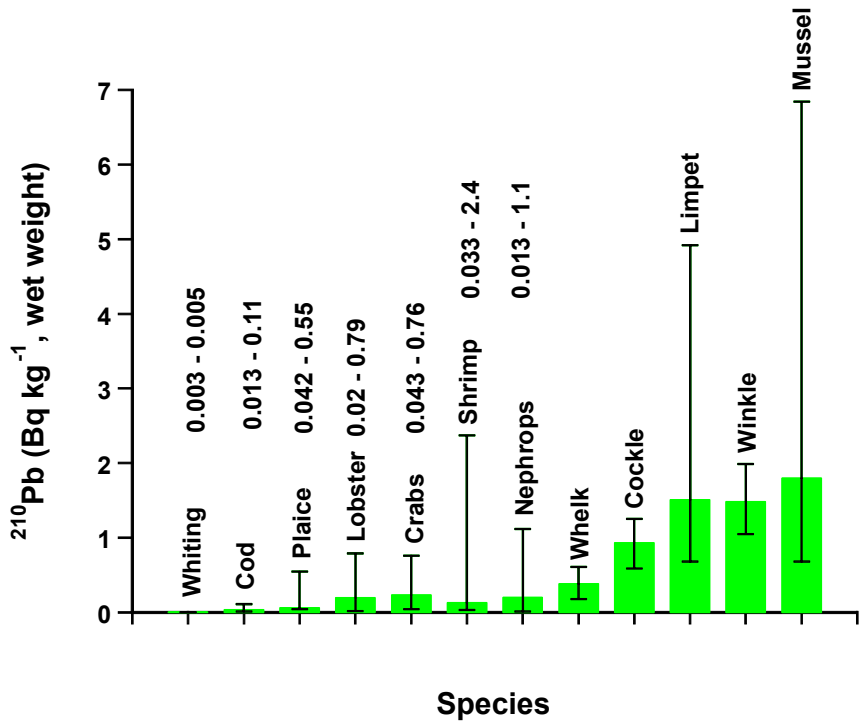


Figure 2c

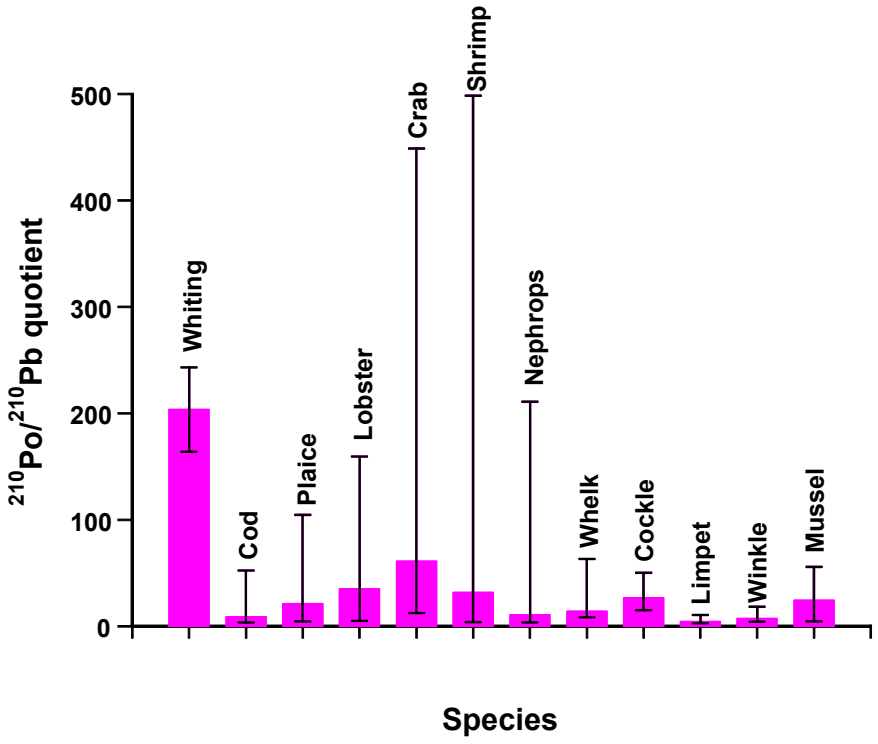


Figure 2d

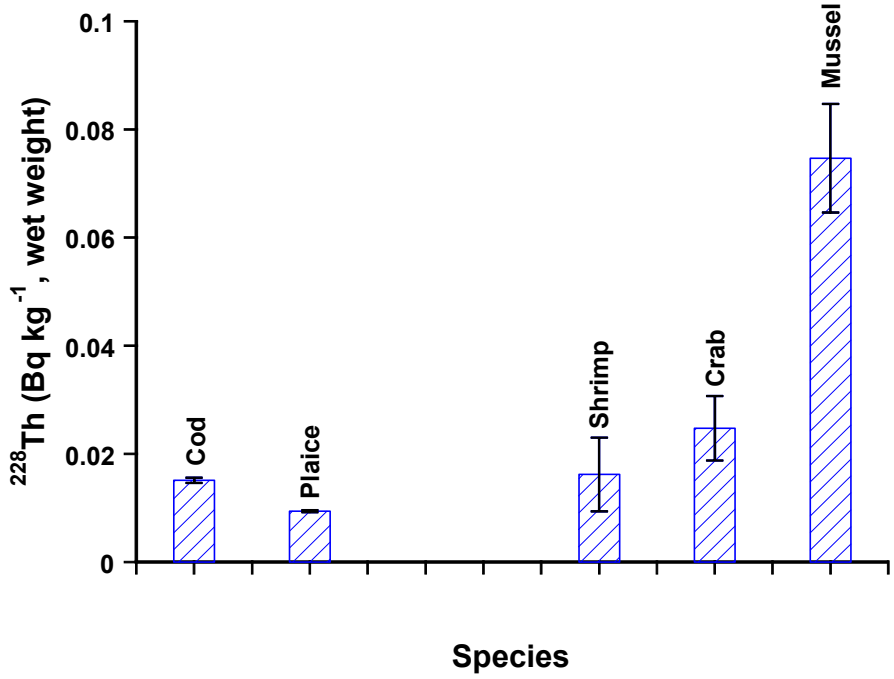


Figure 2e

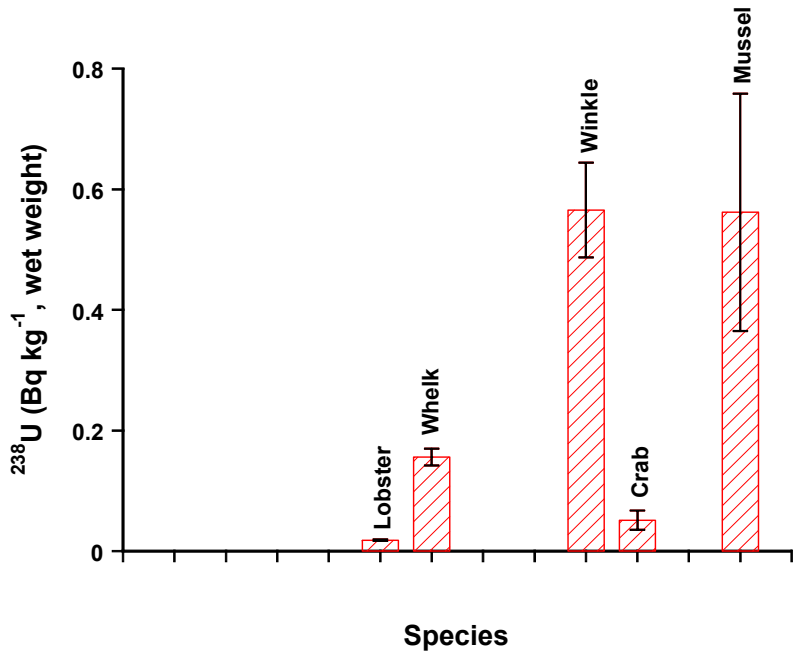
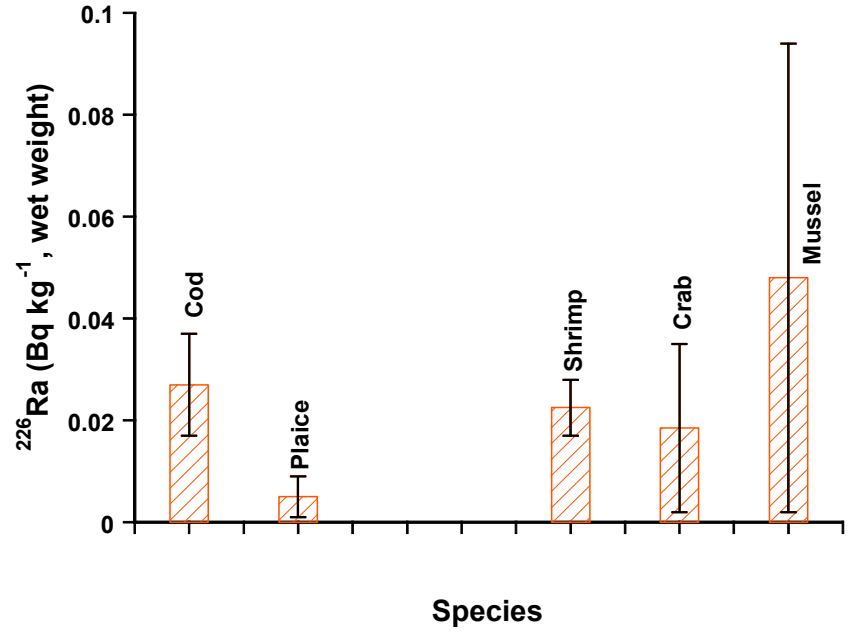


Figure 2f



The data in Fig. 2a indicate median ^{210}Po concentrations in individual species ranged from 0.42 Bq kg^{-1} in cod up to 38 Bq kg^{-1} in mussels (i.e. a variation of ~90 fold). It is clear, therefore, that the dose to man resulting from seafood consumption is sensitive to variations in diet. The error bars show that there was marked variation in the ^{210}Po content between individual samples of the same organisms (typically by ~ 4 fold and up to 26 fold for shrimps). Despite the variability in the data, it is apparent that there were identifiable differences in levels of ^{210}Po between the edible fractions of individual species. ^{210}Po concentrations increased in the order cod ~ whiting < plaice ~ *Nephrops* < lobster ~ shrimp ~ whelk ~ limpet < winkle ~ crab < cockle < mussel. Organisms obtained closed to the shoreline (e.g. whelks, limpets, winkles, cockles and mussels) tended to contain greater concentrations than those obtained offshore (e.g. cod, whiting, plaice and *Nephrops*). In part, the variation is due to the fact that the edible fraction of cod, whiting, plaice and *Nephrops* consists of muscle tissue only whereas the edible fraction of all other species includes the hepatopancreas a known biological hotspot for ^{210}Po accumulation (the distribution of radionuclides between different organs is discussed elsewhere).

Median ^{210}Pb concentrations (Fig. 2b) in individual species ranged from 0.004 Bq kg^{-1} in whiting up to 1.8 Bq kg^{-1} in mussels (i.e. a variation of more than two orders of magnitude). The error bars show that there was also readily detectable differences in ^{210}Pb concentrations between individual samples of the same organisms (typically by ~ 9 fold and by almost two orders of magnitude for shrimps and *Nephrops*). The pattern of bioaccumulation for ^{210}Pb was somewhat different to that observed for ^{210}Po, indicating that the uptake mechanism varies between the two radionuclides.

^{210}Pb concentrations increased in the order whiting< cod < plaice < shrimp < lobster ~ *Nephrops* ~ crab < whelk < cockle < winkle ~ limpet ~ mussel.

^{210}Po/^{210}Pb quotients (Fig. 2c) exhibited a large variation ranging from ~4 in limpets up to ~204 in whiting (i.e. a variation of ~ 46 fold). Nevertheless, it is apparent that ^{210}Po was largely unsupported by ^{210}Pb in all species. The error bars show that there were large variations in the ^{210}Po/^{210}Pb quotients between individual samples of the same organisms (typically by one order of magnitude and by two orders of magnitude for shrimps).

A limited number of species were analysed to provide preliminary estimates of ^{228}Th, ^{238}U and ^{226}Ra concentrations (Figs. 2d-2f) at UK coastal sites free from known anthropogenic inputs. Inspection of the y-axis scales indicates that levels of these nuclides were notably lower than those observed for ^{210}Pb and ^{210}Po. There were measurable variations in bioaccumulation between individual species for all three radionuclides. Levels of ^{228}Th (Fig. 2d) in mussels (~0.075 Bq kg^{-1}) were greater (by ~ 5 fold) compared with those observed in the other organisms (average of 0.016 Bq kg^{-1} in crabs, shrimps, cod and plaice). Assay of ^{228}Th by alpha spectrometry also yielded information for ^{230}Th and ^{232}Th. For a given species, values for both the ^{228}Th/^{230}Th and ^{228}Th/^{232}Th quotients were similar ranging from 1.1 in mussels up to 13 in cod. ^{238}U concentrations (Fig. 2e) varied by ~ 30 fold ranging from 0.02 Bq kg^{-1} in lobster up to ~0.6 Bq kg^{-1} in mussels and winkles. The determination of ^{238}U also yielded information for ^{234}U and ^{235}U. The average ^{234}U/^{238}U and ^{235}U/^{238}U quotients were 1.14 (range1.06-1.18) and 0.039 (range 0.029-0.045). Therefore, in contrast to values for the Th quotients, those for U were remarkably invariant between individual

species. ^{226}Ra concentrations (Fig. 2f) varied by ~ 10 fold ranging from ~0.05 Bq kg^{-1} in mussels down to ~0.005 Bq kg^{-1} in plaice. Concentrations of all three radionuclides were significantly lower than those observed for ^{210}Po (e.g. concentrations of ^{238}U, ^{228}Th and ^{226}Ra in mussels less by ~ 70 fold, 500 fold and 770 fold respectively). The dose to man due to the naturally occurring radionuclides considered in the present study is largely from ^{210}Po.

3.2 Variability in radionuclide concentrations between sampling sites

The radionuclide data in Tables 2-5 were further assessed to examine whether the variations in concentrations between individual samples of the same organisms could be accounted for by differences between separate sampling sites. As mentioned previously, for practical reasons samples had to be collected throughout the year complicating data interpretation. Selected results are provided in Figs. 3a-3b for mussels (species containing largest radionuclide concentrations) and shrimps (species exhibiting largest variation in radionuclide concentrations).

Figure 3.

Variation in ^{210}Po and ^{210}Pb concentrations and the ^{210}Po/^{210}Pb quotient between individual sampling sites (July 1999-August 2001). Dashed arrows indicate overall UK median value from all sites (as shown in Fig. 2a). Error bars indicate range of values observed in individual samples and n indicates number of samples collected at each site: i) Location of sampling sites, ii) ^{210}Po data, iii) ^{210}Pb data, iv) Values for ^{210}Po/ ^{210}Pb quotient; a) mussels; b) shrimps.

Figure 3ai-Mussels

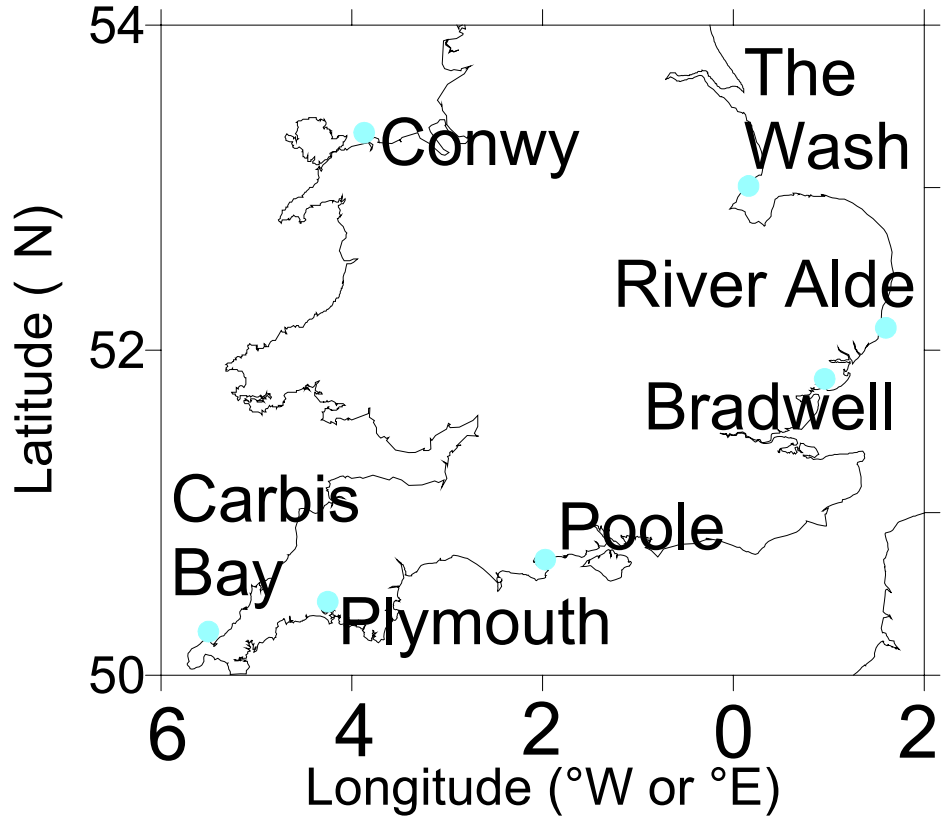


Figure 3aii-Mussels

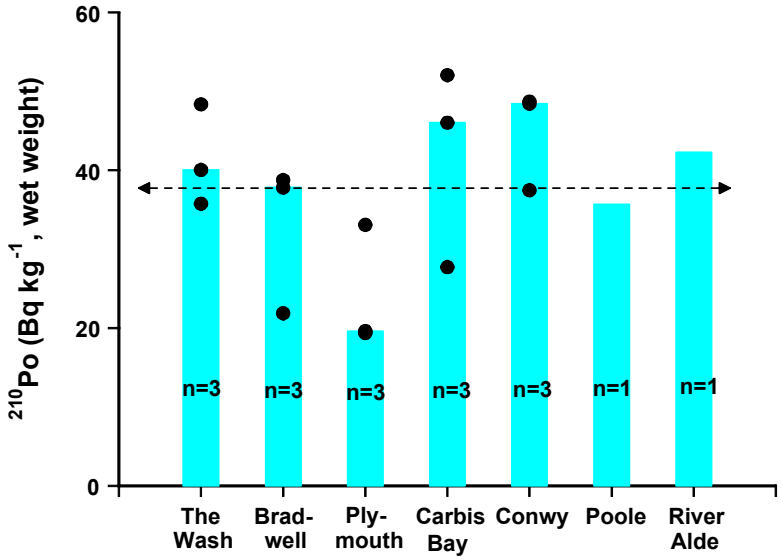


Figure 3aiii-Mussels

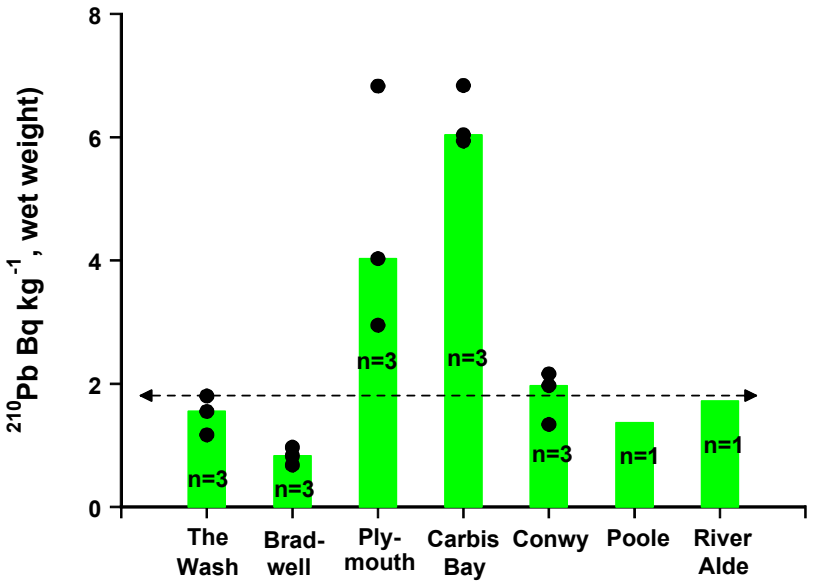


Figure 3aiv-Mussels

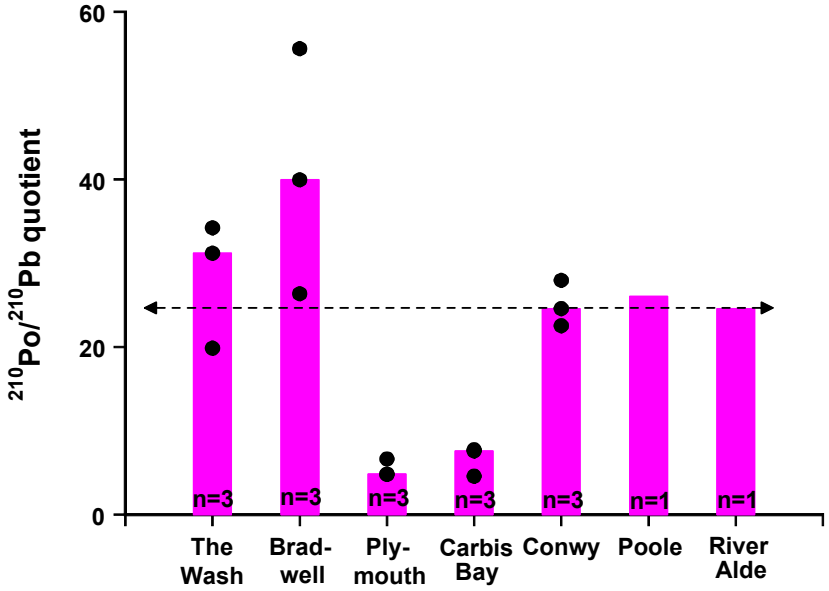


Figure 3bi-Shrimps

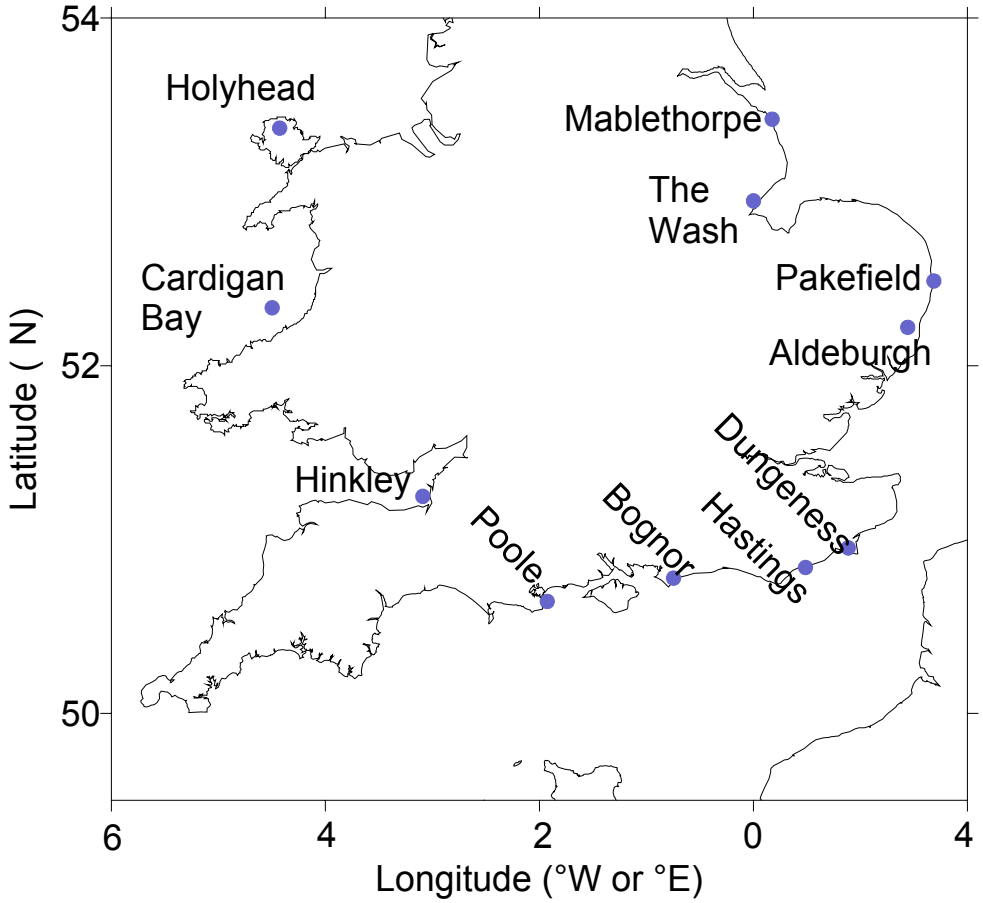


Figure 3bii-Shrimps

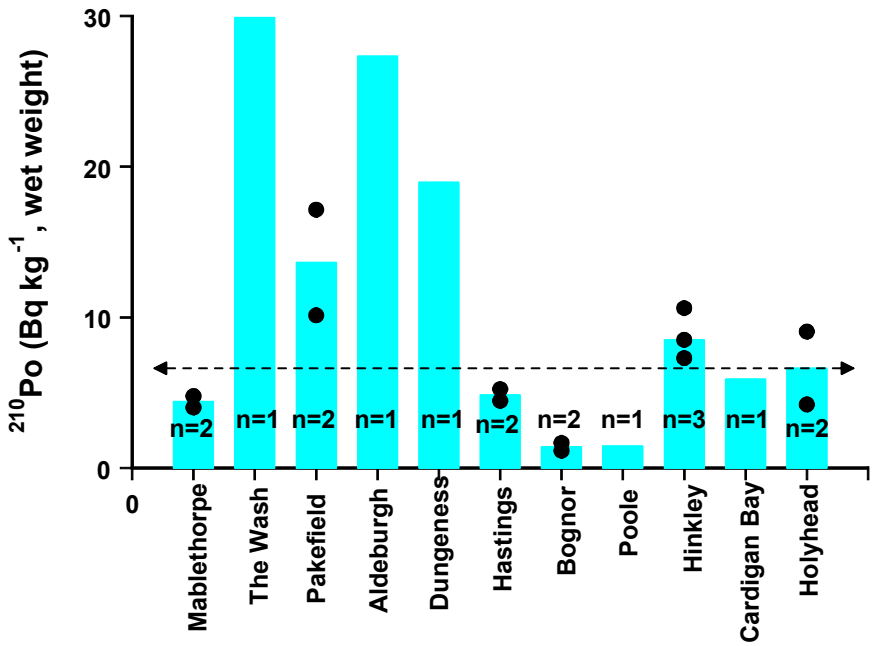


Figure 3biii-Shrimps

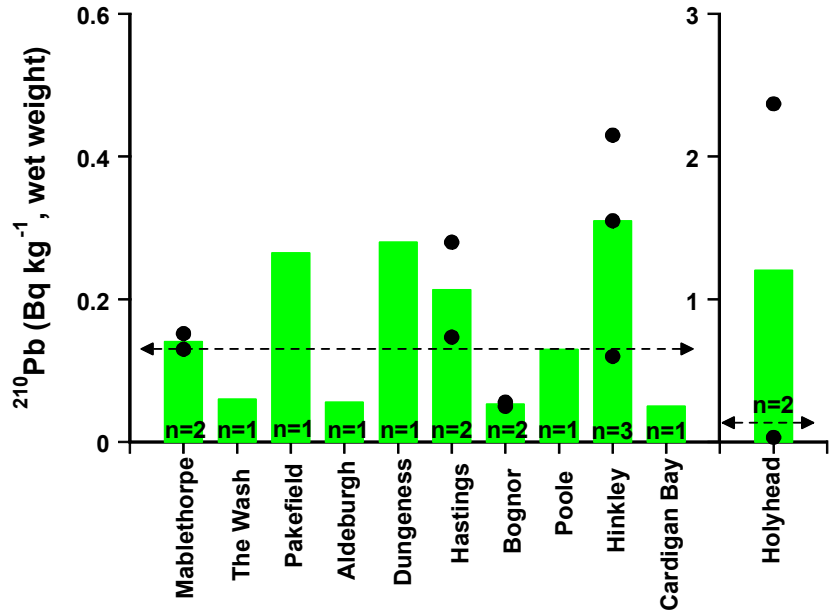
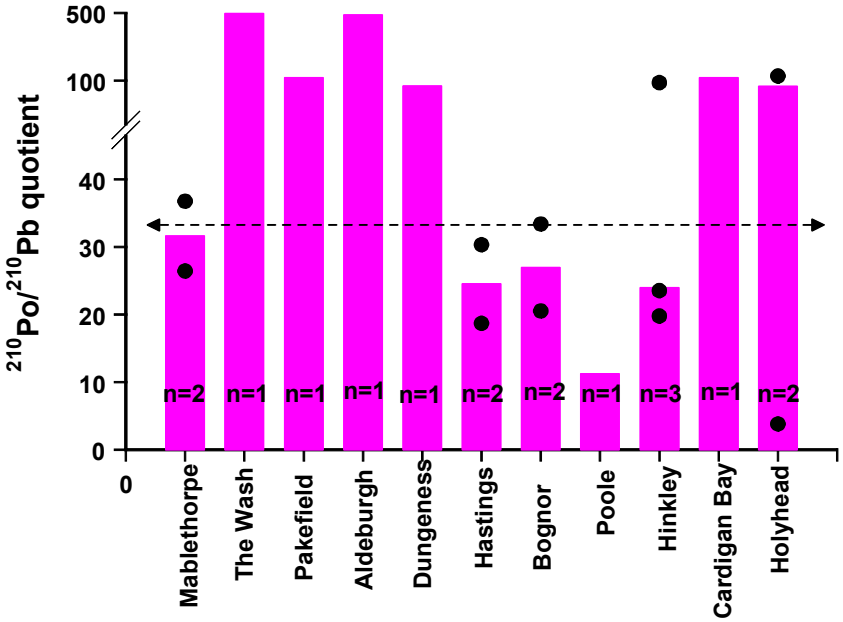


Figure 3biv-Shrimps



The data in Fig. 3aii indicate that, at all sampling sites, ^{210}Po mussel concentrations varied measurably (typically by 1.6 fold). The scatter in the mussel data at an individual site was, therefore, only slightly reduced compared with that observed in the pooled data for all sampling sites (variation between maximum and minimum mussel values was ~2.7 fold, Fig. 2a). Given the variability in the data, and the very limited numbers of samples analysed, it is impossible to draw any firm conclusions regarding variations in ^{210}Po mussel concentrations between sampling sites, although it is possible that levels at Plymouth were marginally lower than those observed at other locations. A similar pattern of behaviour was noted for cockles, limpets and whelks (data not shown). The variability in ^{210}Po concentrations at the individual sites where more than one sample was collected was typically two fold, compared with a scatter of 3-4 fold observed in the pooled data for all sampling sites.

The ^{210}Pb mussel data (Fig. 3aiii) differ somewhat from the ^{210}Po data in that the scatter in the ^{210}Pb data from individual sites (difference < 2.3 fold) was markedly reduced compared with that observed in the pooled data for all sampling sites (variation between maximum and minimum mussel values was ~10 fold, Fig. 2b). Mussels from the Wash, River Alde, Bradwell (North Sea coastline), Poole (English Channel coastline) and Conwy (Wales) contained a median of 1.4 Bq kg^{-1} ^{210}Pb compared with ~6.0 Bq kg^{-1} at Carbis Bay (a site in Cornwall close to a known outcrop of high-uranium loadbearing rock) and 4.0 Bq kg^{-1} at Plymouth. These results indicate readily detectable variations in ^{210}Pb concentrations were observed between individual sampling sites. The difference does not appear to be due to variable sampling time. ^{210}Pb levels at Carbis Bay were greater (by ~4 fold) compared with those observed at the Wash, River Alde, Bradwell, Poole and Conwy.

The $^{210}Po/^{210}Pb$ quotients (Fig. 3aiv) ranged between 20 and 56 at the Wash, Bradwell, River Alde, Poole and Conwy compared with 5 and 8 at Plymouth and Carbis Bay. The ^{210}Po is, therefore, largely unsupported at all sites.

^{210}Po shrimp concentrations (Fig. 3bii) at the individual sites, where more than one sample was collected, exhibited a typical variability of 1.5 fold which was similar to that noted for mussels (1.6 fold). The scatter in the shrimp data for the individual sites was, however, markedly reduced compared with that observed in the pooled dataset (variation between maximum and minimum shrimp values was ~26 fold, Fig. 2a). The assessment of differences in ^{210}Po shrimp bioaccumulation between individual sampling sites is, however, complicated by the paucity of data. The two highest values (observed at the Wash and Aldeburgh) are unfortunately based on the analysis of single samples. Excluding these and other sites from which single samples were collected, ^{210}Po shrimp concentrations decreased in the order Pakefield (Lowestoft) >Hinkley~Holyhead>Hastings~Mablethorpe>Bognor. Insofar as it is possible to interpret this small dataset, the site differences do not appear to be related to sampling times. The variation between the median value at Pakefield and Bognor was almost 10 fold. These results provide a preliminary indication that ^{210}Po shrimp concentrations may vary significantly between separate sampling sites. It is also possible that differences in ^{210}Po content, between individual sampling sites, may also occur for crabs and winkles. ^{210}Po concentrations in crabs from along the eastern coastline at Cromer and Torness (median of 8.1 Bq kg^{-1}, range 4.1-9.8 Bq kg^{-1}) were less (by ~ 2 fold) compared with those observed at the other sampling sites in Wales (Conwy and Cardigan) and the English Channel (Hastings, Eastbourne and Chapmans Pool). Crabs at the latter sites contained a median of 20 Bq kg^{-1} (range 15-35 Bq kg^{-}

^{1}). ^{210}Po concentrations in winkles from Plymouth (average of 7.4 Bq kg^{-1}) were less (by ~ 2 fold) compared with those observed at the other sampling sites along the North Sea (Bradwell) and Irish Sea (Cemaes Bay). Winkles at these sites contained a median of 14 Bq kg^{-1} (range 10-21 Bq kg^{-1}). The intrasite variations in ^{210}Po concentrations for crabs and winkles were noticeably less than those for shrimps. More data is, however, required to confirm these preliminary indications that differences in ^{210}Po crab/winkle bioaccumulation may occur between individual sampling sites.

The data in Fig. 3biii indicate that ^{210}Pb shrimp concentrations at the individual sites were variable. The highest and lowest concentrations (0.03 Bq kg^{-1} and 2.37 Bq kg^{-1}, respectively were both observed at Holyhead). Comparison with the other data indicates that the highest value may be an outlier and should, therefore, be regarded with caution. The median concentration at Mablethorpe, the Wash, Aldeburgh, Bognor, Poole and Cardigan Bay (~0.06 Bq kg^{-1}) was less (by ~5 fold) than that observed at Hinkley and Pakefield. It is, however difficult to draw any firm conclusions regarding variations in ^{210}Pb concentrations between individual sampling sites from these limited amount of data. The ^{210}Po/^{210}Pb quotients ranged from ~11 at Poole up to 498 at the Wash (excluding the very low value of 4 observed at Holyhead which may be an outlier). The median ^{210}Po/^{210}Pb quotient was 32.

The results in Figs 3a-3b indicate that the practice of using generic UK wide values for ‘background’ concentrations of naturally occurring radionuclides in seafood may not be appropriate. These data support the point drawn from another study (Hamilton, 1998) that in order to understand the behaviour of radionuclides in marine and

estuarine systems, site-specific characteristics (as discussed in more detail below) must be taken into account.

4. Discussion

4.1 Comparison of present data with literature values

As mentioned previously, a literature review was carried out as part of the present study to compile all the relevant data. It has, therefore, been possible to use some of results reported elsewhere to compare and contrast with the data obtained in this investigation (Fig. 4).

Figure 4

Comparison of median radionuclide concentrations in marine species reported in literature (Appendix A) with those obtained in present study. Error bars indicate range of values observed in individual samples. a) ^{210}Po, b) ^{210}Pb, c) ^{238}U, d) ^{226}Ra.

Figure 4a

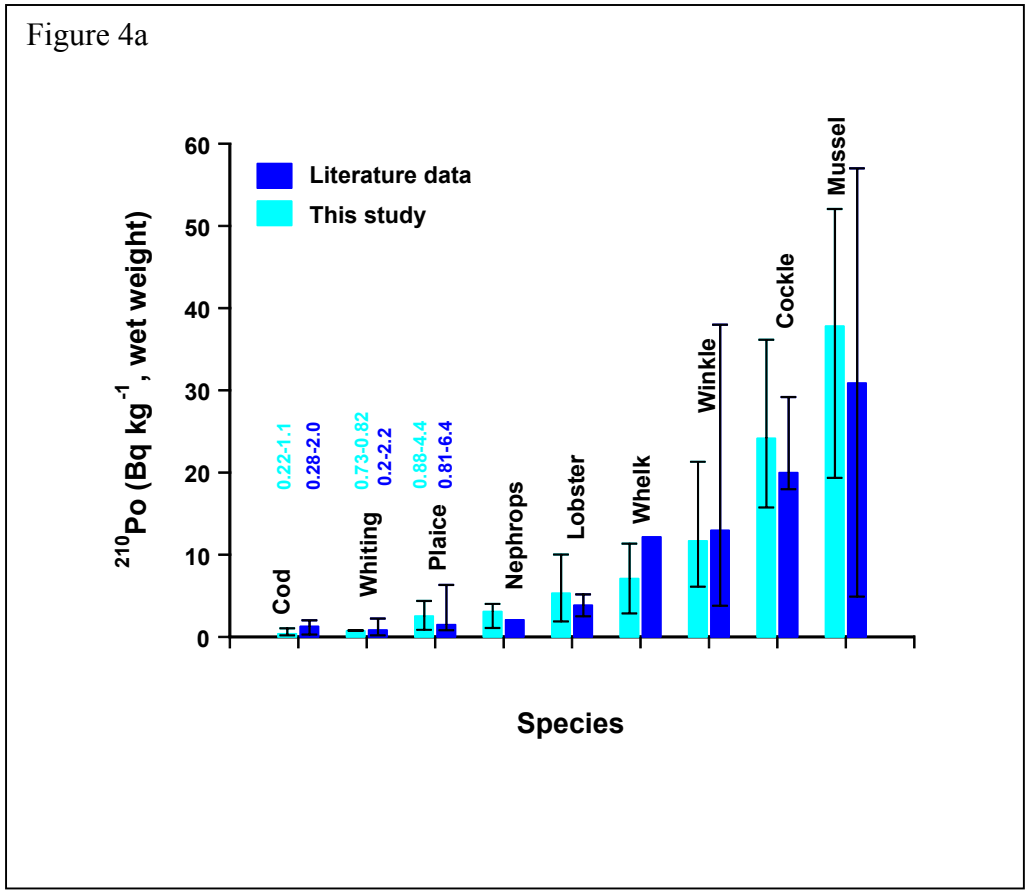


Figure 4b

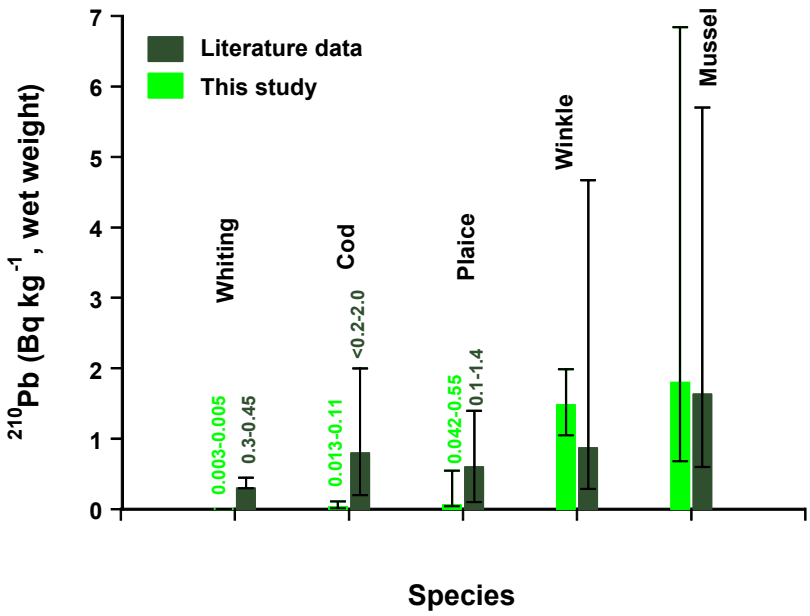


Figure 4c

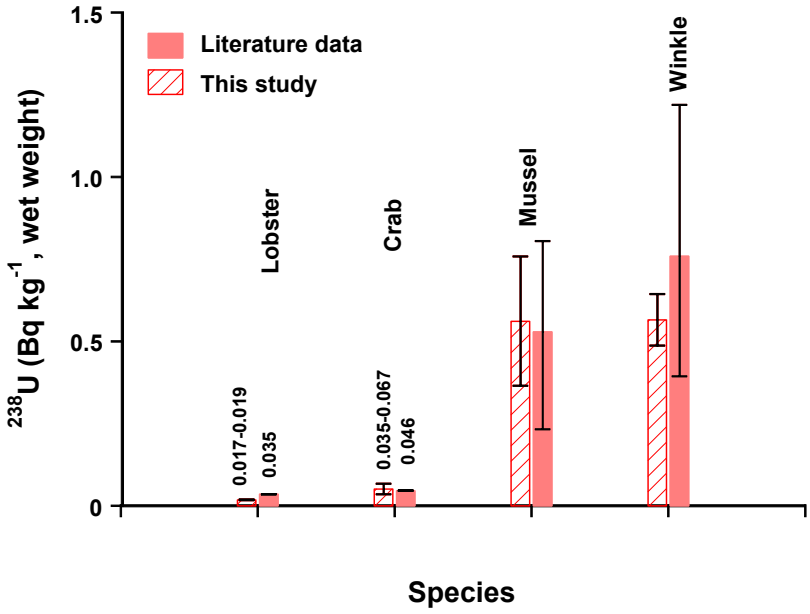
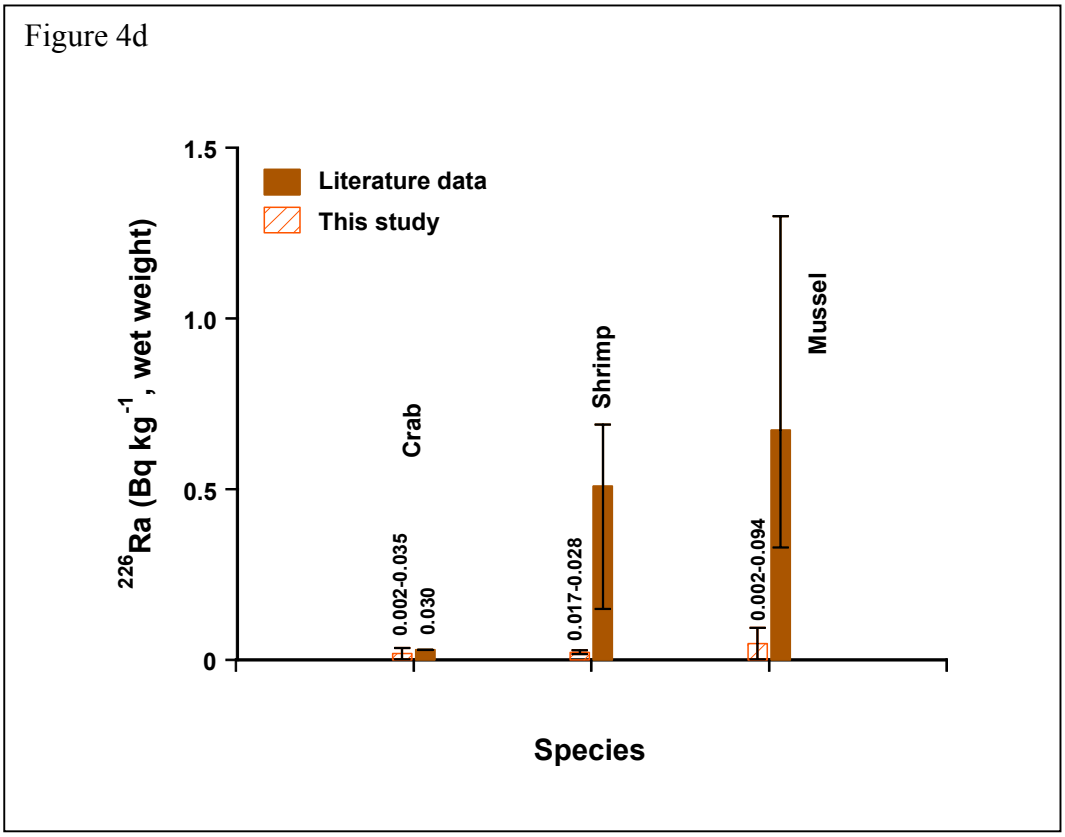


Figure 4d



For all species, the median ^{210}Po values reported in the literature (Fig. 4a) are similar to those observed in the present study. However, the range of literature values tends to be somewhat broader than that found here (e.g. 11 fold in mussels compared with 3 fold in the present study). Relevant literature values for ^{210}Pb are sparse (Fig. 4b). The range of values for plaice, winkles and mussels are broadly comparable. In contrast, literature values for cod and whiting are one to two orders of magnitude greater than those reported here. Ultra low levels (down to 0.003 Bq kg^{-1}) were found in cod and whiting in the present study due to the use of large sample sizes and extended count times. The literature values may provide an overestimate, although further sampling is required to confirm the new values reported here. The data in Fig. 4c show that ^{238}U literature values were similar to those observed in the present study. In contrast, levels of ^{226}Ra found here for shrimps and mussels were more than an

order of magnitude lower than those reported elsewhere. Once again, additional sampling is required to better inform the most appropriate explanation for the discrepancy.

The values currently used in the RIFE report (FSA and SEPA, 2001) for 'background' concentrations of ^{210}Po and ^{210}Pb are shown in Fig. 5, together with data obtained in the present study.

Figure 5

Comparison of 'background' values used in RIFE reports with median radionuclide concentrations observed in present study. Error bars indicate range of values observed in individual samples. a) ^{210}Po, b) ^{210}Pb.

Figure 5a

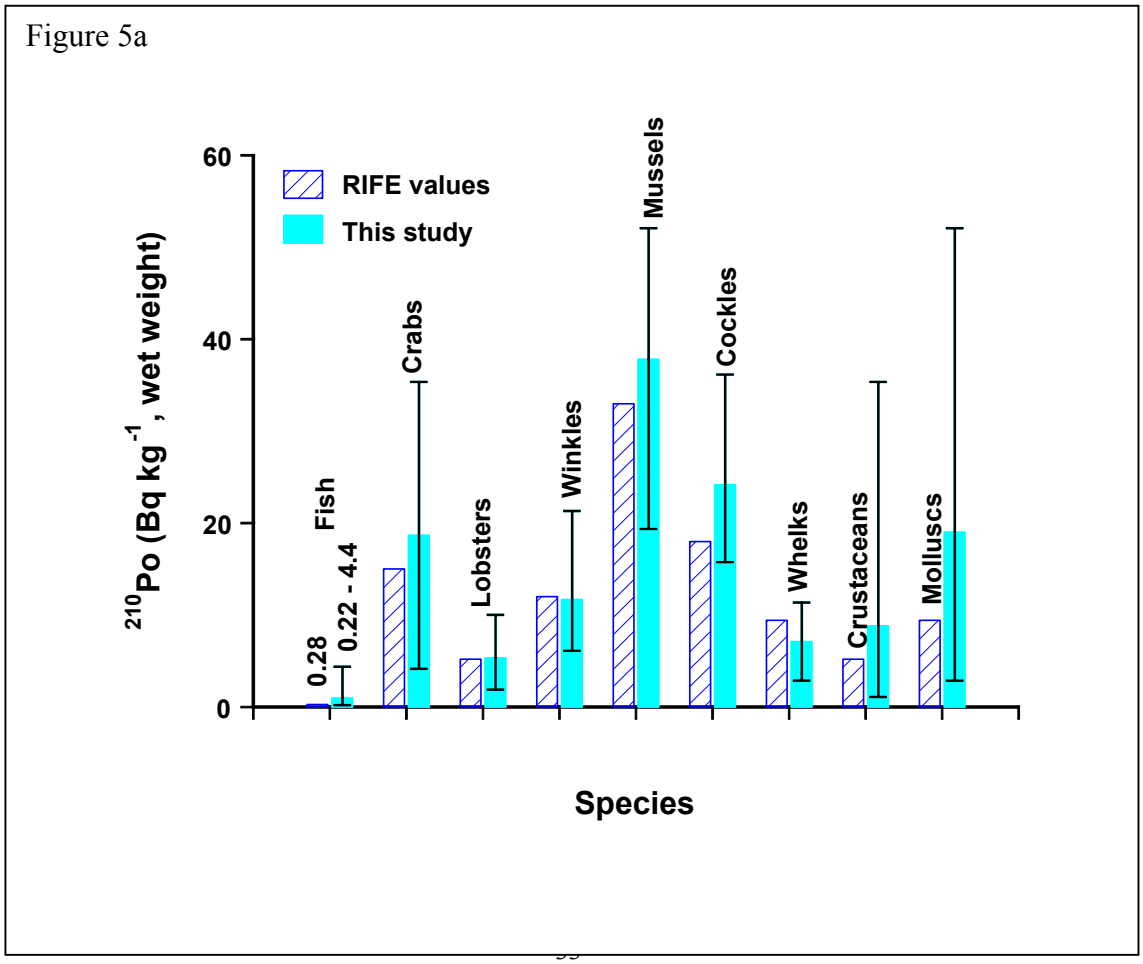
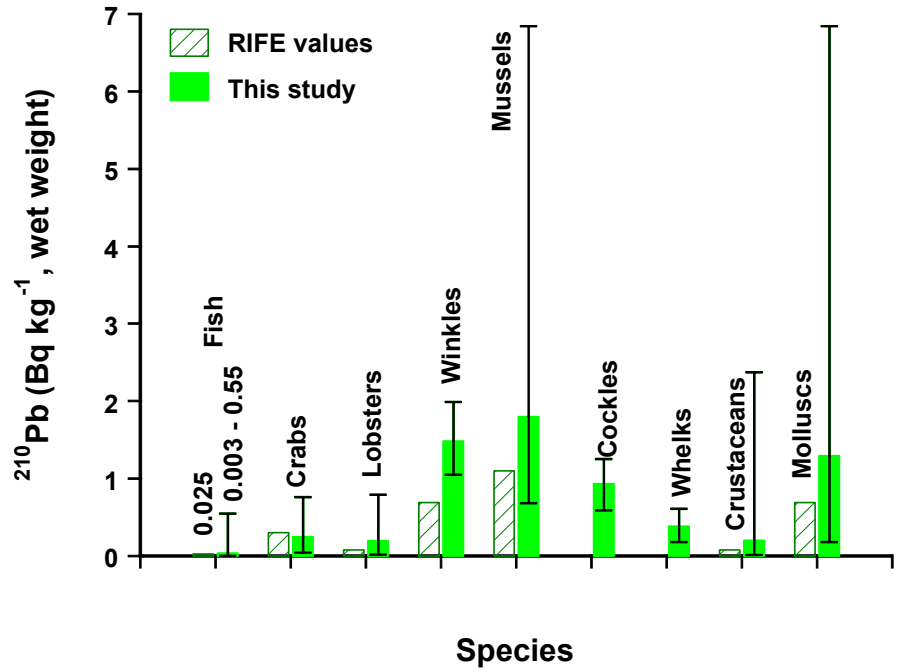


Figure 5b



With the exception of fish and molluscs, the background ^{210}Po values currently used in the RIFE report are within 40% of the median concentrations observed in the present study (Fig. 5a). The value for fish used in RIFE-6 (0.28 Bq kg^{-1}) is, however, lower (by ~ 3 fold) than the overall median of the median values for cod, whiting and plaice (0.78 Bq kg^{-1}). In the event that a wider range of fish species had been analysed, the difference would most likely have been even greater. For example, a survey of ^{210}Po concentrations in the edible muscle tissue of 36 species of marine fish indicated concentrations ranged from 0.27 - 27 Bq kg^{-1} wet weight (Al-Masri *et al*., 2000). The background ^{210}Pb values currently used in the RIFE report are typically 2 fold less compared with the median concentrations observed in the present study (Fig. 5b). Given the disparity in ^{210}Po and ^{210}Pb concentrations between individual fish species (Fig. 2a), the use of generic values for fish in RIFE is of limited value.

Similar considerations apply to the use of generic values for crustacean and mollusc. To accurately assess the extent of enhanced dose due to anthropogenic inputs of naturally occurring radionuclides requires detailed knowledge of local dietary habits and levels in all the individual items.

4.2 Bioaccumulation mechanisms

An essential prerequisite for the providing a credible interpretation of the data reported here, and subsequent recommendations, requires an understanding of the pathways by which radionuclides accumulate in marine organisms. Relevant general considerations are outlined in section 6.2.1. The data in Figs. 2a-2f, illustrate that the significant variability in uptake behaviour between different species. The specific considerations for individual organisms are given in section 6.2.2.

4.2.1 General considerations

To a first approximation biological uptake can occur via three possible pathways: (i) direct uptake of dissolved species from seawater; (ii) ingestion of sediment; (iii) ingestion of contaminated prey items. Benthic shellfish, and fish feeding on these species (e.g. cockles and plaice), incidentally ingest sediment. Radionuclides in sediments are less available than their concentrations might imply, although measures of this availability have been generally lacking (Mayer *et al*., 1996; Turner and Olsen, 2000). Ingested sediment is exposed to a unique chemical environment controlled by the digestive chemistry of the organism. The bioavailability of sediment bound radionuclides is dependent upon gut residence time and levels of solubilising agents,

such as amino acids for metals and surfactants for PAH's, in the digestive fluids. Bioavailability is also dependent upon the association of contaminants between different geochemical fractions in sediment particles. Studies carried out to measure solubilization of non-radioactive sedimentary contaminants (Cu, Pb, and polycyclic aromatic hydrocarbons (PAH)) by digestive fluids extracted from marine invertebrates indicated that the bioavailability of these contaminants is a small fraction of total contaminant loading (typically 1-10 %). Digestive fluids from different animal species solubilise different amounts of metals, indicating that bioavailability varies among species even under constant mode of uptake. Biological factors affecting the assimilation efficiencies of radionuclides from ingested food include food quantity and quality, partitioning of radionuclides in the food particles, as well as the digestive physiology of individual organisms (Wang and Fisher, 1999). It is an extremely challenging task to quantitatively assess the relative importance of individual uptake pathways for a given species since it requires measurements of several physiological parameters, including assimilation efficiency (AE) from ingested food, uptake rate from the dissolved phase, and efflux rates (physiological turnover rates).

4.2.2 *What are the likely uptake pathways for the organisms considered in the present study?*

Uptake of ^{210}Po by marine organisms is thought to primarily occur from food (Carvalho and Fowler, 1994). This is, in part, due to the fact that ^{210}Po is very insoluble in seawater (Tanaka *et al*., 1983), therefore the majority of ^{210}Po in the water column is bound to suspended particulate material. Given the importance of the

food consumption pathway in ^{210}Po accumulation, it is essential to consider their individual feeding behavior. Mussels and cockles are filter and suspension feeders, respectively, and graze on phytoplankton and other suspended matter in the near bottom water (Boyle, 1981; McLusky, 1981). The ventilation rate of the mussel is high (~ 1.5 litre hr^{-1} for a typical 5 cm organism). This and the fact that levels of ^{210}Po in the digestive gland of (Colwyn Bay) mussels have been shown to be strongly correlated with changes in levels on suspended particulate material (Wildgust *et al*., 1998) suggests that the majority of ^{210}Po in these organisms is derived by leaching from sedimentary particles. Crabs (and shrimps) are opportunistic feeders and indiscriminately take food from the benthic zone (Warner, 1977). Indeed, it has been reported that there appears to be a clear relationship between diet and ^{210}Po concentrations in penaeid and carid shrimps from the north-eastern Atlantic (Heyraud *et al*., 1988). Variations in available food sources between different sampling sites could perhaps account for some of the variability in radionuclide concentrations in shrimps (Figs.2a and 2b).

The relationship between the feeding behavior of the winkle and ^{210}Po is not so direct. The diet of the winkle is largely herbivorous. It feeds on seaweed, preferably green alga (Chlorophyceae) such as *Ulva* and *Enteromorpha* (Watson and Norton, 1985). However, data obtained from a study of the temporal variation in levels of ^{210}Po in winkles and *Ulva* (at Colwyn Bay) did not indicate that seasonal changes in concentrations were correlated (Wildgust *et al*., 1998). It was, therefore, suggested that the majority of the ^{210}Po in winkles was derived from sources other than seaweed. Other items in the winkle's diet include microorganisms and detritus on solid surfaces (Fretter and Graham, 1962; Newell, 1979). Moreover, the passage of water through

the mantle cavity of the winkle (albeit small in volume compared with the mussel) does result in the ensnaring of some detrital particles (McDonald *et al*., 1993). It is apparent that identification of uptake pathways may be complex.

Bioaccumulation in the fish considered in this study (cod, whiting and plaice) were comparatively low (more than an order of magnitude less than that observed in mussels). In part, this is due to differences in absorption efficiency. Carvalho and Fowler (1994) observed that the ^{210}Po absorption efficiency for prawns and fish was approximately 0.35 and 0.05, respectively, and roughly corresponded to the assimilation efficiencies of proteins from food. More subtle variations between levels of ^{210}Po were observed between the individual fish species considered in the present study, presumably as a result of differing diet. The prime component of adult cod's diet is fish; it will feed on almost any suitable species present in numbers. Herring, sand eels, capelin, haddock and codlings are generally the most important. Whiting is an active predator that feeds mostly in mid-water or just off the bottom. In common with cod, the prime component of adult whiting is fish (sand eels, sprats, younger whiting, poor-cod, Norway pout). They also eat benthic organisms such as shrimps, swimming crabs and hermit crabs. Plaice feed on a range of benthic prey species, which are taken into the mouth by suction. Their diet usually includes large numbers of bivalve molluscs. The elevated concentrations in plaice (by ~4 fold) compared with cod and whiting are probably caused by its preference for bivalves and these have a relatively high ^{210}Po content.

More generally, Shannon (1973) reported that pelagic fish (those that live and feed in the water column such as mackerel) contain roughly five times more ^{210}Po than

demersal species (those that live and feed on the bottom such as plaice). Pelagic fish were not considered here. To complement the dataset generated in the present study, additional sampling and analyses are required to assess variations in these species.

4.2.3 Depuration and food preparation

Information concerning bioaccumulation mechanisms is helpful in understanding why food preparation methods can have a significant impact upon the radionuclide content of seafood. As mentioned previously, in monitoring programmes it is assumed that preparation methods used by critical group consumers do not involve depuration, although it is routinely advised that the preparation of molluscs should include a period of immersion in clean water, allowing non-edible material to be cleared from the gut (Good Housekeeping Institute, 1989). Studies of artificial radionuclide behaviour have shown that a high proportion of radioactivity in fresh shellfish may be associated with sediment as opposed to shellfish flesh. For example an investigation by McKay and Fox (1991) found that sediment in the gut of winkles accounted for ~95% of the total ^{137}Cs, $^{239+240}$Pu and ^{241}Am activity. In a detailed study of the depuration of $^{239+240}$Pu and ^{241}Am from Cumbrian winkles into uncontaminated seawater, Swift *et al*. (1995) found that loss of both radionuclides was initially rapid and similar and could be described by a mathematical two compartment model. The biological half-time for the initial elimination was about one to two days and accounted for about 70 to 90% of the activity within the winkles when collected. The estimated radionuclide assimilated fractions for winkle flesh ranged from 5% to 40% of the activity content when collected, corroborating the previous finding that a very large fraction may initially be associated with sediment in the gut. Swift *et al*. (1995)

concluded that depuration of winkles before cooking would reduce their $^{239+240}$Pu and ^{241}Am activity content by about 40 to 50% resulting in a significant reduction in the internal radiological dose for consumers.

Given the highly particle-reactive nature of ^{210}Po, it is likely that depuration of Cumbrian winkles prior to cooking would result in significant losses of similar magnitude to those reported for $^{239+240}$Pu and ^{241}Am. However, due to the environmental variability of sediment loads in marine waters, the proportion of radioactivity associated with sediment and shellfish flesh is itself likely to be variable (hence ^{210}Po loss during depuration).

4.3 Radionuclide distribution between different organs

Heyraud and Cherry (1979), Skwarzec (1988), Swift *et al*. (1994), Wildgust *et al*., (1998) have all reported that ^{210}Po is non-uniformly distributed within fish and benthic organisms. The highest ^{210}Po concentrations occur in organs involved in digestion and metabolism such as the hepatopancreas of marine invertebrates (Swift *et al*., 1994) and the intestines, stomach, spleen and pyloric caecal of fish (Skwarzec, 1988). The lowest levels are found in muscle and bone. This information is important when comparing data between different studies where sample preparation techniques may vary.

The extent of radionuclide concentration in the digestive organs varies between different species. For example, ^{210}Po levels in the shrimp hepatopancreas are two to

three orders of magnitude greater than that found in tail muscle (Swift *et al*., 1994). Although the shrimp hepatopancreas constitutes only a minor fraction (~5%) of the whole body weight, it necessarily contains a large fraction of the total ^{210}Po inventory. Concentrations in the brown meat of crabs are roughly 50 times greater than those in claw muscle. The disparity in the distribution of ^{210}Po between mussel tissues is less marked (e.g. levels in the viscera just ~ 5 fold greater than those in muscle; McDonald *et al*., 1993). The digestive gland of the mussel accounts for approximately 10% of the total soft tissue weight but contains between 15 and 36% of the ^{210}Po soft tissue inventory (Wildgust *et al*., 1998). These authors suggested that environmental fluctuations of ^{210}Po are better reflected in the digestive gland than the whole soft tissue because this organ is the major entry point of particle-bound ^{210}Po.

Levels of ^{210}Po found in the digestive organs of fish tend to be correlated with the degree of stomach repletion and thus decrease if food is lacking (Skwarzec, 1988). This was illustrated by data indicating that the digestive organs and muscle of a replete cod constituting ca 11% and 45% of the wet body weight, respectively, contained ca 73% and 3% of the total ^{210}Po fish inventory. In contrast, the digestive organs and muscle of non-replete flounder constituting ca 5% and 58% of the wet body weight, respectively, contained ca 18% and 51% of the total ^{210}Po fish inventory. It was concluded that the residence time of ^{210}Po within the digestive system of fish was short (in the order of days) resulting in a rapid decrease in ^{210}Po content of the liver and intestine when the stomach is empty (a situation which occurs during spawning).

An explanation and the implications of, the non-homogeneous distribution requires a consideration of the mechanisms for bioaccumulation. As mentioned previously, uptake of ^{210}Po by fish appears to occur via feeding. Food is usually broken down in the stomach of carnivorous fish through a combination of muscular contractions of the stomach wall and enzymatic action. Breakdown products are expelled from the stomach through the pyloric sphincter into the small intestine in a process called gastric evacuation. Digestion and food absorption are completed in the small intestine and rectum. The elevated concentrations in the digestive organs relative to muscle and bone indicate a mechanism must exist to result in the rapid transport of solublised ^{210}Po into, and binding within, the cells of the stomach wall. This may be due to profuse binding of $^{210}Po^{2+}$ ions to negative charge sites on the mucosal side of the gut wall and the low permeability coefficient of divalent cations through the lipid bilayers of membranes (Farmanfarmaian and Socci, 1984). Moreover, it has been reported that ^{210}Po has a high affinity for sulphur containing compounds (e.g. certain amino acids; Heyraud *et al*., 1987). This behaviour is typical of so called class B metals which bind readily with groups such as sulphydrl (-SH), disulphide (-S-S-), thioether (-SR) and amino ($-NH_2$) functional groups. The implication is that, if ^{210}Po does behave as a class B metal, then the accumulation of ^{210}Po by marine organisms may be affected by other class B metals. Exposure to class B metals such as cadmium induces metal binding proteins called metallothioneins and could result in enhanced ^{210}Po accumulation.

The application of information concerning the distribution of ^{210}Po is of more than academic interest. In a recent CEFAS report to MAFF (Transfer of Radioactivity from Fishmeal in Animal Feeding Stuffs to Man, CEFAS (1999)) analyses of

fishmeal samples from a variety of sources were undertaken to assess the content of both natural and anthropogenic radionuclides. Fishmeal is a high protein animal feed which is used to supplement the food intake of a number of animal species including ruminants, poultry and fish. The fishmeal is produced by processing (rendering) fish caught at sea. The species used comprise those for human consumption as well as those which are not (industrial fish). Only the trimmings remaining from fish for human consumption tend to be used in fishmeal whereas for industrial fish species, all of the fish is included. The radioactivity present in fish used to make fishmeal can ultimately reach man when he consumes meat products derived from animals which have been fed fishmeal-based feedstuffs. The observed radionuclide concentrations in the fishmeal samples were consistent with reported concentrations in fish except for ^{210}Po, ^{226}Ra, and uranium radionuclides, which were more than an order of magnitude higher. Two explanations were provided to account for the discrepancy: 1) that fishmeal is prepared from whole fish, not just the edible parts which tend to have lower activity concentrations or; 2) fishmeal is derived from fish containing enhanced levels of natural nuclides. Given the enhanced ^{210}Po concentrations which may occur in the digestive organs of replete fish and the variability between individual fish species either explanation is plausible.

The assessment of the potential radiological significance of fishmeal in the human food chain indicated the dose commitment was significant. For example individual doses derived for high rate food consumers ranged from 450 μSv y^{-1} for adults to 1255 μSv y^{-1} for 1 year olds. Doses are dominated by ^{210}Po, the main pathway being consumption of farmed salmon and trout. Additional research has been commissioned by the FSA to investigate this pathway further (project RO2015).

4.4 Variations between individual sampling sites

The conclusion from the present study that ^{210}Po concentrations in shrimp (Fig. 3bii) may vary between sites is clearly tentative, resulting from the limited amount of data. There is, however, some indication from other studies (e.g. Heyraud and Cherry, 1979; Cherry and Heyraud, 1981) that levels may vary concomitant with the prevailing environmental conditions. In a study of the ^{210}Po content of the marine shrimp, Cherry and Heyraud (1981) noted that there appeared to be a steady increase in levels on going from estuarine to coastal to pelagic to deep sea species. The extent of the increase between estuarine and coastal environments was in the order of 3 fold. A plausible explanation to account for the increase in ^{210}Po concentrations between estuarine and coastal environments was provided in terms of changes in the chemical form of dissolved ^{210}Po (Cherry and Heyraud, 1981). Although the immediate source of ^{210}Po in shrimp was suggested to be the food they eat, the ultimate source of ^{210}Po in the food was ^{210}Po in the water. Given that variations in the ^{210}Po content of seawater are relatively small it was further suggested that the ultimate availability of ^{210}Po to the foodchain depends on its form in seawater. This was related to the fact that certain metals are accumulated more strongly by phytoplankton and filter feeding organisms when they exist as inorganic species than when they are organically bound. Many trace elements are organically bound in estuarine environments and if this was true for ^{210}Po as well then the degree of uptake of ^{210}Po from water (by whatever food or organisms are responsible for the initial entry of ^{210}Po into the food chain) was suggested to be less than in a 'truly marine' environment.

An assessment of differences in bioaccumulation behaviour is complicated by the fact that the variability in ^{210}Po concentrations is large, even among individuals of the same species. For example, Heyraud and Cherry (1979) found considerably higher levels in *Sergestes* spp. compared to *Pasiphaea* spp. although these two shrimps live at similar depths and occupy the same niche. Charmasson *et al*. (1998) subsequently suggested that, given the relatively short ^{210}Po half-life, the ageing of food might account for the discrepancy since the feeding of these benthopelagic amphipods is irregular. More evidence is clearly required to substantiate the explanation (i.e. variations in food quality between sampling locations) previously provided to account for the large variation (Figs. 2a and 2b) in shrimp bioaccumulation observed in the present study.

With respect to species other than shrimps, ^{210}Po concentrations in crabs from along the eastern coastline appear to be less (by ~ 2 fold) compared with those from Wales and the English Channel. The disparity may be related to the fact that edible crabs show strong geographical (and seasonal) differences in size composition (Bannister, 1999). Crabs show significant regional differences in growth rate, which is much higher in the western Channel than elsewhere, but probably also varies between other parts of the coast. This causes regional differences in the maximum size range available for capture, and also in the minimum landing size. About 75% of hen crabs mature at 115 mm carapace width (CW) in Norfolk, and at 110 mm CW in Yorkshire. In contrast, in the English Channel most hens mature at 140 mm CW. Another factor which complicates any simplistic analysis of radionuclide data is that female crabs migrate during summer and autumn, when they are heavily exploited by the fisheries.

The variability in the data, and the very limited numbers of samples analysed meant that it proved impossible to draw any firm conclusions regarding variations in ^{210}Po mussel concentrations between sampling sites in the present study (Fig. 3bii). However, results from another study (McDonald *et al*., 1986) indicate that concentrations can vary between different sites. In a pilot study in which spot samples were collected from six sites coastal sites between Scotland, England, France and Monaco it was found that levels of ^{210}Po ranged from 111-459 Bq kg^{-1} dry weight in soft tissues. The highest concentrations were found in the Mediterranean mussels collected from Monaco. It is possible that intrasite variations in the UK could have been found if a wider range of sampling sites had been considered.

4.5 Seasonal cycling

For practical reasons, samples analysed in the present study were collected throughout the year from February until November. It is important, therefore, to consider what, if any, impact this might have upon the interpretation of variability reported here. Unfortunately, information in the literature concerning the extent of any temporal variations in ^{210}Po levels is sparse. Results from a study of ^{210}Po concentrations in the common shrimp and edible crab from the North Sea indicated that although levels were significantly variable, it was impossible to correlate the changes with time (Swift *et al*., 1994). Similarly, Germain *et al*. (1992) and Ryan *et al*. (1997) were unable to find any consistent temporal change in levels of ^{210}Po by whole soft tissues in mussels. Interpretation of results from these studies is, however, hampered by a lack of supporting information. To assess the impact of environmental fluctuations it

is essential to carry out ancillary measurements to quantify seasonal changes arising from phytoplankton blooms, physico-chemical parameters such as temperature and salinity and biological variables such as the reproductive cycle.

The only attempt to quantify the impact of some of the relevant environmental fluctuations appears to that of Wildgust *et al*., 1998. They observed significant variations (range ~5 fold) in levels of ^{210}Po in the mussel digestive gland over a 12 month period. Levels were highly correlated with those on suspended particles. This is to be expected given that mussels feed by filtering suspended particles. Levels of ^{210}Po in winkles appear to remain relatively constant throughout the year except for a peak between May and August (Rollo *et al*., 1992; Swift *et al*., 1995; Wildgust *et al*., 1998). Wildgust *et al*. (1998) suggested the increase is due to a significant drop in body weight (hence increased ^{210}Po concentration) bought about by spawning. If so, then the seasonal changes in ^{210}Po concentration are likely to be variable between sites (and indeed from year to year) as the release of gametes are thought to be controlled by temperature. Given the paucity of available information it is impossible to assess what was the impact, if any, upon the variability of the data reported in the present study from using samples collected throughout all four seasons.

5. Conclusions

Concentrations of ^{210}Pb, ^{226}Ra, U and Th radionuclides were much lower than those for ^{210}Po.

Median ^{210}Po concentrations in individual species ranged from 0.42 Bq kg^{-1} in cod up to 38 Bq kg^{-1} in mussels (i.e. a variation of ~90 fold). Concentrations increased in the order cod ~ whiting < plaice ~ *Nephrops* < lobster ~ shrimp ~ whelk ~ limpet < winkle ~ crab < cockle < mussel.

Median ^{210}Pb concentrations ranged from 0.004 Bq kg^{-1} in whiting up to 1.8 Bq kg^{-1} in mussels (i.e. a variation of more than two orders of magnitude). The significantly lower ^{210}Pb concentrations indicate the ^{210}Po is almost entirely unsupported.

The dose to man resulting from seafood consumption is, therefore, extremely sensitive to variations in diet.

Marked variations occurred in levels of ^{210}Po and ^{210}Pb between individual samples of the same organisms (typically by ~ 4 fold and ~ 9 fold for ^{210}Po and ^{210}Pb, respectively). The greatest variability was observed in shrimps (up to 26 fold and 72 fold for ^{210}Po and ^{210}Pb, respectively) and may be due to differences in environmental conditions (e.g. food availability) between sampling locations.

Although excellent progress has been made in the present study in achieving the stated objectives, much work remains to be done. Assessment of the data to account

for the observed variability was difficult. This is because the processes controlling the extent of radionuclide bioaccumulation are complex. Some of the factors, which might be controlling uptake behaviour, have been identified (e.g. different environmental conditions between individual sampling sites, feeding behaviour and seasonal cycling).

The absence of a detailed understanding of the processes and mechanisms controlling ^{210}Po bioaccumulation behaviour is of practical, as well as academic, significance. It questions whether there is a clearly defined framework for a radiological assessment. For example, the information from this relatively small-scale programme has highlighted the uncertainties associated with the use of generic UK wide values for 'background' concentrations of naturally occurring radionuclides in seafood in the RIFE report. Variations occur concomitant with changes in catchment geology, estuarine and coastal areas and seasonal cycling. The noise associated with the environmental data indicates that any assessment of the impact of anthropogenic inputs requires a detailed site-specific interpretation.

6. Recommendations

The 'background' concentrations of ^{210}Po and ^{210}Pb in seafood in the RIFE report (Table A6.1) should be replaced with the median values observed in the present study (Table 6). In addition, ranges should be quoted to indicate the extent of natural variation. Insufficient data is available for nuclides other than ^{210}Pb and ^{210}Po to recommend further changes.

Extreme caution needs to be applied in the use of generic values for fish, crustacean and mollusc, given the wide variation in bioaccumulation between individual species. Additional sampling and analyses are required to establish range of radionuclide concentrations in species not considered in the present study (e.g. pelagic fish species such as mackerel and herring)

Site-specific assessments, including spatial and species specific data, are required to assess the impact of any anthropogenic input

More information is required to credibly assess the impact of radionuclides in fishmeal as a pathway to man.

Given the extent of natural variation, information solely concerning radionuclide concentrations in seafood is of limited value. In the event that resources were available for follow up work, it would be desirable to carry out ancillary measurements to quantify environmental conditions under which samples are collected and thereby obtain a more holistic picture. Although not directly related to food, ancillary information would add value given that interpretation of the seafood data is dependent upon an understanding of the processes controlling radionuclide behaviour.

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8. References

Aarkrog, A., Baxter, M.S., Bettencourt, A.O., Bojanowski, R., Bologa, A., Charmasson, S., Cunha, I., Delfanti, R., Duran, E., Holm, E., Jeffree, Livingston, H.D., Mahapanyawong, S., Nies, H., Osvath, I., Pingyu, Li, Povinec, P.P., Sanchez, A., Smith, J.N. and Swift, D. (1997). A comparison of doses from ^{137}Cs and ^{210}Po in marine food: a major international study. J. Environ. Radioact., 34(1): 69-90.

Al-Masri, M. S, Mamish, S., Budeir, Y and Nashwati, A. (2000). ^{210}Po and ^{210}Pb concentrations in fish consumed in Syria. J. Environ. Radioact., 49(3): 345-352.

Bannister, C. R. (1999). A review of shellfish resources and their management. Dr Walne memorial lecture, 30th annual shellfish conference, May 18th 1999, Fishmonger's Hall, London. (http://intranet/cefaswww/publications/walne.htm)

Bolivar, J.P, Garcia-Tenorio, R. and Vaca, F. (2000). Radioecological study of an estuarine system located in the south of Spain. Water Res., 34(11): 2941-2950.

Boyle, P. R. (1981). Molluscs and Man. The Institute of Bilogy's studies in biology, No. 134, 60 pp. Edward Arnold, London.

Camplin, W.C; Baxter, A.J. and Round, G.D., (1996). The radiological impact of disposals of natural radionuclides from a phosphate plant in the United Kingdom. Environ. Int., 22, Suppl. 1: 5259-5270.

Carvalho, F.P.(1995a). ^{210}Po and ^{210}Pb intake by the Portuguese population: The contribution of seafood in the dietary intake of ^{210}Po and ^{210}Pb. Health Physics, 69(4): 469-480.

Carvalho F.P. (1995b). ^{210}Pb and ^{210}Po in sediments and suspended matter in the Tagus esturay, Portugal. Local enhancement of natural levels by wastes from phosphate ore processing industry. Sci. Tot. Environ., 159(2-3): 201-214.

Carvalho, F.P. and Fowler, S. W. (1994). A double tracer technique to determine the realtive importance of water and food as sources of polonium-210 to marine prawns and fish. Mar. Ecol. Prog. Ser. 103: 251-264.

CEFAS (1999). Transfer of Radioactivity from Fishmeal in Animal Feeding Stuffs to Man (Contract Leader B. D. Smith). Report for MAFF, Radiological Safety and Nutrition Division , CEFAS Contract No. C0608.

Charmasson, S., Germain, P. and Leclerc, G. (1998). ^{210}Po as a tracer of trophic input to deep-sea benthic ecosystems: A study of the deep-sea amphipod *Eurythenes Gryllus* from the tropical Atlantic. Rad. Prot. Dosim. 75(1-4): 131-138.

Cherry, R.D. and Heyraud, M. (1981). Polnium-210 content of marine shrimp: Variation with biological and environmental factors. Mar. Biol. 65: 165-175.

Emerson H.S.. and Young A..K. (1995). Method Development for the extraction of naturally occurring radionuclides in marine sediments., Sci. Total. Environ., 173/174: 313-322.

Farmanfarmaian, A. and Socci, R. (1984). Inhibition of essential amino acid absorption in marine fishes by mercury. In: Responses of Marine Organisms to Pollutants (Ed., Stegeman, J.J.). Vol. 14 (1-4): 185-199.

Fretter, V. and Graham, A. (1962). British Prosobranch Molluscs. The Ray Society, London.

FSA and SEPA (2000). Radioactivity in food and the environment, 1999. RIFE report 5. FSA and SEPA, London, UK.

Germain, P., LeClerc, G. and Simon, S. (1992). Distribution of ^{210}Po in *Mytilus edulis* and *Fucus Vesiculosis* along the channel coast of France; influence of industrial releases in the Seine river and estuary. Radiation Protection Dosimetry 45: 257-260.

Good Housekeeping Institute (1989). Good housekeeping cooking. Ebury Press, London.

Heyraud, M. and Cherry, R.D. (1979). ^{210}Po and ^{210}Pb in marine food chains. Mar. Biol. 52: 227-236.

Heyraud, M., Cherry, R.D. and Dowdle, E.B. (1987). The subcellular localisation of natural ^{210}Po in the hepatopancreas of the Rock Lobster (*Jasus Calandii*). J. Environ. Radioactivity 5: 249-260.

Heyraud, M., Domanski, P., Cherry, R.D. and Fasham, M.J.R. (1988). Natural tracers in dietrary studies for ^{210}Po and ^{210}Pb in decapod shrimp and other pelagic organisms in the north-east Atlantic Ocean. Mar. Biol. 97: 507-519.

Hoffman, F.L., Hodge, V.F. and Folsom, R.R. (1974). ^{210}Po radioactivity in organs of selected tunas and other marine fish. J. Radiation Res. 15: 103-106.

Hunt, G.J. and Allington, D.J. (1993). Absorption of environmental polonium-210 by the human gut. J. Radiol. Prot., 13(2): 119-126.

ICRP (1994). Age-dependent doses to members of the public from intake of radionuclides: Part 2 Ingestion dose coefficients. Annal ICRP 23(3/4). Pergamon Press Oxford, 168pp. (ICRP Publ. (67)).

Jackson, D. and Rickard, A. (1998). The influence of body-size and food prepartion practices on the uptake and loss of radionuclides in Cumbrian winkles. Radiat. Prot. Dosim., 75(1-4): 155-159.

Lucas, H.F., 1957. Improved low-level alpha-scintillation counter for radon. The Review of Scientific Instruments., 9: 680-683.

Mayer, L.M., Chen, Zhen, Findlay, R.H., Fang, Jiasong, Sampson, S., Self, R.F.L., Jumars, P.A., Quetel, C., and Donard, O.F.X. (1996). Bioavailability of sedimentary contaminants subject to deposit-feeder digestion. Environ. Sci. Technol., 30(8): 2641-2645.

McCartney, M., Kershaw, P.J., Allington, D.J., Young, A.K., and Turner, D. (1992). Industrial sources of naturally-occurring radionuclides in the eastern Irish Sea. Rad. Prot. Dosimetry, 45:711-714.

McCartney, M., Davidson, C.M., Howe, S.E., and Keating, G.E. (2000). Temporal changes in the distribution of natural radionuclides along the Cumbrian coast following the reduction of discharges from a phosphoric acid production plant. J. Environ. Radioact., 49(3): 279-291.

McDonald, P., Fowler, S.W., Heyraud, M. and Baxter, M.S. (1986). Polonium-210 in mussels and its implications for environmental alpha-autoradiography. J. Environ. Radioact. 3: 293-303.

McDonald, P., Cook, G.T. and Baxter, M.S. (1991). Natural and artificial radioactivity in coastal regions of the UK. In: Radionuclides in the Study of Marine Processes, eds P.J. Kershaw and D.S. Woodhead. Elsevier Applied Science, London, pp. 329-339.

McDonald, P., Baxter, M.S. and Fowler, S.W. (1993). Distribution of radionuclides in mussels, winkles and prawns. Part 1. Study of organisms under environmental

conditions using conventional radio-analytical techniques. J. Environ. Radioact., 18(3), 181-202.

McKay, W.A. and Fox, A.A. (1991). Particulate-associated nuclides in Cumbrian winkles-Implications for assessment of dose to man. J. Environ. Radioact. 14: 1-21.

McKay, W.A, Halliwell, C.M. and Rose, C. (1997). The impact on dose of particulate-associated radionuclides in Irish Sea shellfish. J. Radiol. Prot.,17(2): 115-117.

McLusky, D. S. (1981). The estuarine ecosystem-(Tertiary level biology). 1. Estuarine ecology. Blackie & Son Ltd., Glasgow, UK.

Newel, R.C. (1979). Biology of intertidal animals. Marine ecological surveys, LTD Kent.

Pentreath, R.J and Allington, D.J. (1988). Dose to man from the consumption of marine seafoods: A comparison of naturally-occurring ^{210}Pb with artificially produced radionuclides. In 'Proceedings of VII international IPRA congress', Sydney, 10-17 April, 3: 1582-1585.

Pentreath, R.J., Camplin, W.C. and Allington, D.J. (1989). Individual and collective dose rates from naturally-occuring radionuclides in seafood. pp 297-300. In: E.P. Goldfinch (Ed) , 'Radiation Protection Theory and Practice, Proc. 4th Int. Symp. Soc. Radiol. Prot., Malvern, 4-9 June 1989'. Institute of Physics, Bristol and New York.

Pollard, D., Ryan, T.P. and Dowdall, A. (1998). The dose to Irish seafood consumers from ^{210}Po. Rad. Prot. Dosim. 75(1-4): 139-142.

Poole, A.J., Allington, D.J., Baxter, A.J. and Young, A.K. (1995). The natural radioactivity of phosphate ore and associated waste products discharged into the eastern Irish Sea from a phosphoric acid production plant. Sci. Total. Environ. 173/174: 137-149.

Rollo, S.F.N., Camplin, W.C., Allington, D.J. and Young, A.K. (1992). Natural radionuclides in the UK marine environment. In: 'Proceedings of the fifth International Symposium on Natural Radiation Environment, Salzburg, September 22-28, 1991'. Rad. Prot. Dosim., 45(1-4): 203-210.

Ryan, T.P., Germain, P., Dowdall, A.M., LeClerc, G. and Pollard, D.A. (1997). A comparison of ^{210}Po in *Mytilus edulis* and *Fucus Vesiculosis* in French and Irish coastal waters. . Radioprotection-Colloques, 32(C2):

Shannon, L.V. (1973). Marine alpha-radioactivity off Southern Africaa, Polonium-210 and lead-210. Investl. Rep. Div. Fish. S.Africa, 100:1-34.

Skwarzec, B. (1988). Accumulation of ^{210}Po in selected species of Baltic Fish. J. Environ. Radioactivity 8: 111-118.

Swift D.J.,. Smith, D.L., Allington, D.J., and Ives, M. J. (1994). The ^{210}Po content of North Sea edible crab, *Cancer pagurus* L., and common shrimp, *Crangon crangon* L. and the potential radiological impact. J. Environ. Radioactivity, 23: 213-230.

Swift D.J.,. Smith, D.L., Allington, D.J., and Winpenny, K. (1995). A Laboratory and Field Study of ^{210}Po Depuration by Edible Winkles, *Littorina littorea* L., from the Cumbrian Coast (North Eastern Irish Sea). J. Environ. Radioactivity, 26: 119-133.

Tanaka, N., Takeda, Y. and Tsunogai, S. (1983). Biological effects on the removal of Th-234, Po-210 and Pb-210 from surface waters on Funka Bay. Geochim. Cosmochim Acata 47: 1783-1790.

Turner, A. and Olsen, Y.S. (2000). Chemical versus enzymatic digestion of contaminated estuarine sediment: Relative importnace of iron and manganese oxides in controlling trace metal bioavailability. Estuarine, Coastal Shelf Sci., 51: 717-728.

Wang, Wen-Xiong and Fisher, N.S. (1999). Assimilation efficiencies of chemical contaminants in aquatic invertebrates: A synthesis. Environ. Toxicol. Chem. 18(9): 2034-2045.

Warner, G. F. (1977). The Biology of crabs, Paul Elek (Scientific Books), London.

Watson, D. C. and Norton, T. A. (1985). Dietary preferences of the common periwinkle, *Littorina littorea* (L.). Exp. Mar. Biol. Ecol. 88: 193-211.

Wildgust, M.A., McDonald, P. and White, K.N. (1998). Temporal changes of ^{210}Po in temperate coastal waters. Sci. Tot. Env. 214: 1-10.

Table 1.
Species types sampled

Common Name	Latin Name
Winkles	*Littorina littorea*
Mussels	*Mytilus edulis*
Crab	*Cancer pagerus*
Lobster	*Homarus gammarus*
Nephrops/ Dublin Bay Prawn Norway Lobster	*Nephrops norvegicus*
Limpets	*Patella vulgata*
Shrimps	*Crangon crangon*
Cockles	*Cerastoderma (cardium) edule*
Whelks	*Buccinidae*
Whiting	*Merlangius merlangus*
Cod	*Gadus morhua*
Plaice	*Pleuronectes platessa*

Table 2.
^{210}Po and ^{210}Pb in fish and shellfish

Location	Species	^{210}Po (Bq kg^{-1} wet weight) and *Collection Date*			^{210}Pb (Bq kg^{-1} wet weight) and *Collection Date*		
MOLLUSCS							
Middleton Sands	Cockles		15.78 *23/07/00*	18.69 *03/03/01*		0.79 *23/07/00*	1.25 *03/03/01*
Poole	Cockles		36.15 *07/10/00*	27.44 *10/05/01*		0.72 *07/10/00*	0.94 *10/05/01*
River Dee	Cockles		16.94 *09/08/00*	29.35 *15/05/01*		0.59 *09/08/00*	1.04 *15/05/01*
The Wash	Cockles		24.71 *27/07/00*	23.69 *27/07/01*		1.13 *27/07/00*	0.93 *27/07/01*
Hinkley	Limpets		6.75 *14/06/00*			1.51 *14/06/00*	
Lavernock Point	Limpets		5.92 *06/08/00*	14.91 *01/05/01*		1.34 *06/08/00*	4.92 *01/05/01*
Lowestoft	Limpets		7.11 *14/07/00*	12.53 *16/05/01*		0.68 *14/07/00*	2.54 *16/05/01*
Tarn Bay	Limpets		8.36 *24/07/00*	9.75 *11/05/01*		1.36 *24/07/00*	3.34 *11/05/01*
Conwy	Mussels	37.46 *23/07/99*	48.44 *04/08/00*	48.69 *25/07/01*	1.34 *23/07/99*	1.97 *04/08/00*	2.16 *25/07/01*
R. Alde	Mussels			42.26 *14/06/01*			1.72 *14/06/01*
Carbis Bay	Mussels	52.05 *08/09/99*	27.72 *01/08/00*	46.02 *26/06/01*	6.84 *08/09/99*	6.04 *01/08/00*	5.94 *26/06/01*
The Wash	Mussels	48.35 *15/07/99*	40.04 *04/09/00*	35.75 *10/04/01*	1.55 *15/07/99*	1.17 *04/09/00*	1.8 *10/04/01*
Poole	Mussels	35.69 *15/07/99*			1.37 *15/07/99*		
Bradwell	Mussels	38.76 *12/10/99*	21.88 *24/08/00*	37.81 *07/08/01*	0.97 *12/10/99*	0.83 *24/08/00*	0.68 *07/08/01*
Plymouth	Mussels	19.62 *01/08/99*	19.37 *06/08/00*	33.08 *13/05/01*	2.95 *01/08/99*	4.03 *06/08/00*	6.83 *13/05/01*
Bradwell	Whelks		6.54 *05/04/00*	3.14 *14/06/01*		0.49 *05/04/00*	0.38 *14/06/01*
Dungeness	Whelks		2.89 *15/09/00*	7.64 *12/02/01*		0.27 *15/09/00*	0.39 *12/02/01*
Dunwich	Whelks		11.07 *06/08/00*	9.03 *15/06/01*		0.53 *06/08/00*	0.61 *15/06/01*
Poole	Whelks		4.03 *24/10/00*	9.71 *17/03/01*		0.34 *24/10/00*	0.41 *17/03/01*

Weymouth Bay	Whelks		3.43 *16/10/00*	11.38 *09/02/01*		0.34 *16/10/00*	0.18 *09/02/01*
Bradwell	Winkles		13.18 *07/07/00*	10.2 *28/04/01*		1.49 *07/07/00*	1.65 *28/04/0*
Cemaes Bay	Winkles		14.96 *07/08/00*	21.33 *17/05/01*		1.48 *07/08/00*	1.17 *17/05/01*
Plymouth	Winkles		6.12 *06/08/00*	8.74 *13/05/01*		1.05 *06/08/00*	1.99 *13/05/01*
Ravenglass[1]	Winkles		25.21 *24/07/00*	25.35 *31/05/01*		2.29 *24/07/00*	2.61 *31/05/01*
CRUSTACEA							
Cardigan Bay	Crabs	28.56 *17/09/99*	16.74 *19/09/00*		0.2 *17/09/99*	0.61 *19/09/00*	
Chapmans Pool	Crabs	18.69 *25/07/99*	14.75 *21/07/00*	18.61 *03/04/01*	0.2 *25/07/99*	0.14 *21/07/00*	0.23 *03/04/01*
Conwy	Crabs	26.05 *16/09/99*	26.72 *16/10/00*	35.35 *10/02/01*	0.31 *16/09/99*	0.21 *16/10/00*	0.76 *10/02/01*
Cromer	Crabs	9.78 *17/07/99*	6.59 *25/07/00*	8.63 *24/05/01*	0.3 *17/07/99*	0.26 *25/07/00*	0.69 *24/05/01*
Eastbourne	Crabs		31.58 *21/08/00*			0.51 *21/08/00*	
Hastings	Crabs			19.14 *25/06/01*			0.18 *25/06/01*
Padstow	Crabs	25.74 *08/11/99*	20.31 *01/08/00*	14.96 *14/05/01*	0.24 *08/11/99*	0.4 *01/08/00*	0.56 *14/05/01*
Torness	Crabs	7.65 *20/07/99*	9.64 *28/07/00*	4.14 *10/06/01*	0.18 *20/07/99*	0.16 *28/07/00*	0.12 *10/06/01*
Alderney	Lobster			10.03 *11/06/01*			0.79 *11/06/01*
Jersey	Lobster		1.88 *15/09/00*			0.38 *15/09/00*	
Kilkeel (N.Ireland)	Lobster		5.14 *15/10/00*	3.07 *01/08/01*		0.06 *15/10/00*	0.02 *01/08/01*
Kirkcudbright	Lobster		2.69 *31/07/00*	5.92 *11/06/01*		0.05 *31/07/00*	0.57 *11/06/01*
Plymouth	Lobster			9.56 *25/04/01*			0.06 *25/04/01*
Portrush (N.Ireland)	Lobster		5.52 *15/07/00*			0.33 *15/07/00*	
Northern North Sea	*Nephrops*	1.1 *16/09/99*	3.48 *04/09/00*		0.013 *16/09/99*	0.23 *04/09/00*	

Southern North Sea	*Nephrops*	2.74 *21/08/99*					
Off Aldeburgh	Shrimps	27.34 *29/09/99*			0.056 *29/09/99*		
Off Bognor rocks	Shrimps	1.15 *10/08/99*		1.67 *31/07/01*	0.056 *10/08/99*		0.05 *31/07/01*
Cardigan Bay	Shrimps			5.89 *26/06/01*			0.05 *26/06/01*
Dungeness	Shrimps			18.97 *19/03/01*			0.28 *19/03/01*
Off Hastings	Shrimps	4.46 *27/08/99*	5.24 *24/08/00*		0.15 *27/08/99*	0.28 *24/08/00*	
Off Hinkley	Shrimps	10.62 *27/07/99*	8.51 *01/08/00*	7.3 *30/05/01*	0.12 *27/07/99*	0.43 *01/08/00*	0.31 *30/05/01*
Holyhead Bay	Shrimps	4.22 *27/09/99*	9.06 *19/09/00*		0.033 *27/09/99*	2.37 *19/09/00*	
Off Mablethorpe	Shrimps	4.02 *08/09/99*	4.78 *27/07/00*		0.15 *08/09/99*	0.13 *27/07/00*	
Off Pakefield Beach	Shrimps		10.14 *09/08/00*	17.15 *26/07/01*		0.45 *09/08/00*	0.08 *26/07/01*
Off Poole	Shrimps	1.46 *05/08/99*			0.13 *05/08/99*		
The Wash	Shrimps			29.9 *30/08/01*			0.06 *30/08/01*
FISH							
North Sea	Cod	0.68 *24/08/99*			0.013 *24/08/99*		
Off Hastings	Cod	0.38 *05/07/99*			0.11 *05/07/99*		
Off Lowestoft	Cod	0.42 *14/07/99*			0.048 *14/07/99*		
Off Plymouth	Cod	0.22 *20/07/99*			0.035 *20/07/99*		
Off Torness	Cod	1.05 *20/07/99*			0.025 *20/07/99*		
North Sea	Plaice	4.4 *21/08/99*			0.042 *21/08/99*		
Cardigan Bay	Plaice	3.11 *16/09/99*			0.16 *16/09/99*		
Conwy Bay	Plaice	2.88 *22/10/99*			0.054 *22/10/99*		
Off Hastings	Plaice	1.48 *05/07/99*			0.062 *05/07/99*		
Off Lowestoft	Plaice	0.88 *14/07/99*			0.042 *14/07/99*		

Off Plymouth	Plaice	2.53 *20/07/99*	0.55 *20/07/99*
Off Torness	Plaice	1.69 *20/07/99*	0.095 *20/07/99*
Bristol Channel	Whiting	0.82 *16/09/99*	0.005 *16/09/99*
Cardigan Bay	Whiting	0.73 *16/09/99*	0.003 *16/09/99*

(1) Data for Ravenglass winkles excluded from all calculations, graphs and other Tables in report. This is because seafood items at this site could contain elevated levels as a legacy of the discharges from the Rhodia Consumer Specialities Ltd. (formerly Albright and Wilson) phosphoric acid production plant at Saltom Bay (near Whitehaven, Cumbria).

Table 3.
^{226}Ra in fish and shellfish

Location	Species	^{226}Ra (Bq kg^{-1} wet weight)
Conwy	Mussels	< 0.002
Plymouth	Mussels	0.094 ± 0.007
Chapmans Pool	Crabs	0.035 ± 0.017
Torness Outfall	Crabs	< 0.002
Off Plymouth	Cod	0.037 ± 0.013
Off Torness	Cod	0.017 ± 0.001
Off Plymouth	Plaice	< 0.001
Off Torness	Plaice	0.009 ± 0.001
Off Hinkley	Shrimps	0.028 ± 0.002
Off Mablethorpe	Shrimps	0.017 ± 0.002

Quoted uncertainties are the ± 1σ standard deviation on multiple counts of the same sample

Table 4.
Thorium radionuclides in fish and shellfish

Location	Species	Concentration (Bq kg^{-1}, wet weight)		
		^{232}Th	^{230}Th	^{228}Th
Plymouth	Cod	0.0012 ± 0.0001	0.0012 ± 0.0002	0.0156 ± 0.0004
Torness	Cod	0.0025 ± 0.0001	0.0014 ± 0.0001	0.0146 ± 0.0003
Plymouth	Plaice	< 0.001	<0.001	0.0096 ± 0.0002
Torness	Plaice	0.0011 ± 0.0001	0.0009 ± 0.0001	0.0092 ± 0.0002
Torness	Crab	0.0118 ± 0.0004	0.0087 ± 0.0004	0.0307 ± 0.0006
Chapmans Pool	Crab	0.0022 ± 0.0003	0.0022 ± 0.0004	0.0188 ± 0.0006
Hinkley	Shrimps	0.0045 ± 0.0003	0.0072 ± 0.0005	0.0230 ± 0.0007
Mablethorpe	Shrimps	0.0013 ± 0.0002	0.0025 ± 0.0003	0.0094 ± 0.0003
Conwy	Mussels	0.0220 ± 0.0006	0.0291 ± 0.0008	0.0646 ± 0.0011
Plymouth	Mussels	0.0715 ± 0.0015	0.0740 ± 0.0015	0.0847 ± 0.0016

Quoted uncertainties are ±1σ propagated counting uncertainty only

Table 5.
Uranium radionuclides in fish and shellfish

Location	Species	Concentration (Bq kg^{-1}, wet weight)		
		^{238}U	^{235}U	^{234}U
Plymouth	Mussels	0.758 ± 0.015	0.028 ± 0.001	0.841 ± 0.017
Bradwell	Mussels	0.365 ± 0.008	0.016 ± 0.001	0.417 ± 0.009
Bradwell	Winkles	0.644 ± 0.016	0.025 ± 0.002	0.739 ± 0.018
Plymouth	Winkles	0.487 ± 0.011	0.019 ± 0.002	0.570 ± 0.013
Cromer	Crabs	0.035 ± 0.001	0.001 ± 0.0002	0.037 ± 0.001
Chapmans Pool	Crabs	0.067 ± 0.002	0.003 ± 0.0004	0.075 ± 0.002
Dungeness	Whelks	0.142 ± 0.004	0.005 ± 0.001	0.160 ± 0.005
Poole Bay	Whelks	0.170 ± 0.005	0.007 ± 0.001	0.201 ± 0.005
Kilkeel	Lobster	0.019 ± 0.001	< 0.001 ±	0.022 ± 0.001
Kirkcudbright	Lobster	0.017 ± 0.001	< 0.001 ±	0.020 ± 0.001

Quoted uncertainties are ± 1 sd propagated counting uncertainty only

Table 6
Summary of median ^{210}Po and ^{210}Pb data observed in present study

	Radionuclide concentration (Bq kg^{-1}, wet weight)									
	Fish	**Crustaceans**	**Molluscs**	**Crabs**	**Lobsters**	**Cockles**	**Limpets**	**Mussels**	**Whelks**	**Winkles**
^{210}Pb	0.05 (0.003-0.55)	0.20 (0.013-2.4)	1.2 (0.18-6.8)	0.24 (0.04-0.76)	0.20 (0.02-0.79)	0.94 (0.59-1.3)	1.5 (0.68-4.9)	1.8 (0.68-6.8)	0.39 (0.18-0.61)	1.5 (1.1-2.0)
^{210}Po	1.0 (0.22-4.4)	8.8 (1.1-35)	18 (2.9-52)	19 (4.1-35)	5.3 (1.9-10)	24 (16-36)	8.4 (5.9-15)	38 (19-52)	7.1 (2.9-11)	12 (6-21)

Data in brackets indicate maximum and minimum values

Appendix A
Radionuclide data in fish and shellfish reported elsewhere

Site	Species	Radionuclide (Bq kg^{-1}, wet weight)				Date	Ref No.
		^{210}Po	^{210}Pb	^{238}U	^{226}Ra		
Ribble Estuary	Cockles	29				87-90	5
Wash	Cockles			0.53		87-90	5
Ribble Estuary	Cockles	21				1991	9
Wirral	Cockles	18				1991	9
Dee Estuary	Cockles	19				1991	27
	Median	20					
North Sea ICES area IV	Cod	0.44-2.0				1978	3
Irish Seafood	Cod	0.37-2.0	<0.2-2.0			1995	4
Mid-North Sea	Cod	0.28		0.004		1991	9
Western English Channel	Cod	1.3				1979	22
	Median	0.9					
Torness	Crabs	17		0.046	0.030	1991	9
Hartlepool	Crabs	8.5				1993	27
	Median	13					
Irish Seafood	Lobster	2.5				1995	4
Torness	Lobster	5.2		0.035		1991	9
	Median	3.9					
Tongue	Mussels	45	1.8	0.48		Sep-89	1
Poolewe	Mussels	39	3.1	0.67		Sep-89	1
Stonehaven	Mussels	36	4.0	0.81		Sep-89	1
Sandyhills	Mussels	33	1.2	0.26		Sep-89	1
Ravenglass	Mussels	37	1.5	0.63		Sep-89	1
Hartlepool	Mussels	24	1.7	0.37		Sep-89	1
Colwyn Bay	Mussels	24	0.6	0.23		Sep-89	1
Lowestoft	Mussels	43	1.1	0.79		Sep-89	1
Southern North Sea	Mussels				0.39	1982	3
Southern North Sea	Mussels				1.3	1983	3
Southern North Sea	Mussels	28				1984	3
Central North Sea	Mussels	22				1984	3
Irish Sea	Mussels	12				1984	3
Southern North Sea	Mussels	38				1985	3
Irish Seafood	Mussels	39	0.6			1995	4
Cleethorpes Pier	Mussels	26				87-90	5
Wash	Mussels	48				87-90	5
Lowestoft	Mussels	25				87-90	5
Newlyn	Mussels	23	1.5			87-90	5

Carbis Bay/Cornwall	Mussels	31			0.33	87-90	5
Holyhead Penrhos	Mussels	43				87-90	5
Portling Bay/Scotland	Mussels	54	2.1			87-90	5
Kinlochleven	Mussels	55				87-90	5
Fort William Pier	Mussels	41				87-90	5
Newlyn	Mussels	19				Oct-87	16
Lowestoft	Mussels	5				Jun-86	16
Lowestoft	Mussels	23				Sep-87	16
Carbis Bay/Cornwall	Mussels	14				Aug-88	16
Portling Bay/Scotland	Mussels	24				Aug-88	16
Western English Channel	Mussels	19				1988	21
Western English Channel	Mussels	18				1988	21
Hunterston	Mussels	26				1984	26
Firth of Forth	Mussels	57	5.7			1985	26
Cleethorpes	Mussels	26				1990	27
Southern North Sea	Mussels	50				1990	27
Southern North Sea	Mussels	56				1991	27
Southern North Sea	Mussels	43				1990	27
	Median	31	1.6	0.55	0.39		
North Shields	Nephrops	2.1				Feb-87	16
North Sea ICES area IV	Plaice	0.81-3.2				1978	3
Irish Seafood	Plaice	1.3-6.4	<0.1-1.4			1995	4
Western English Channel	Plaice	1.5				1979	22
	Median	1.5	0.8				
Southern North Sea	Shrimp				0.52	1982	3
Southern North Sea	Shrimp				0.69	1983	3
Southern North Sea	Shrimp	43			0.52	1984	3
Southern North Sea	Shrimp	52			0.64	1985	3
Ribble Estuary	Shrimp				0.15	1996	14
Ribble Estuary	Shrimp				0.53	1997	15
Lowestoft	Shrimp	1.2				Oct-88	16
Lowestoft	Shrimp	0.4-4.1				90-91	23
Plymouth	Shrimp	31	0.6			77-80	28
	Median	37			0.53		
Carbis Bay/Cornwall	Whelks	12			0.05	87-90	5
Irish Seafood	Whiting	0.2-2.2	0.3-0.45			1995	4
	Median	1.2	0.38				
Tongue	Winkles	23	4.7	1.01		Sep-89	1

Aberdeen	Winkles	19	0.3	0.75		Apr-90	1
Stonehaven	Winkles	15	1.0	0.71		Sep-89	1
Anstruther	Winkles	4	0.3	0.39		Sep-89	1
Sandyhills	Winkles	8	1.3	0.79		Sep-89	1
Ravenglass	Winkles	17	1.5	0.63		Sep-89	1
Colwyn Bay	Winkles	17	0.8	0.60		Sep-89	1
Plymouth	Winkles	15	0.6	0.78		Sep-89	1
Sandside Bay	Winkles	20				87-90	5
Torness	Winkles	11		0.84		87-90	5
Lynemouth	Winkles	7				87-90	5
Tynemouth	Winkles	9				87-90	5
Hartlepool	Winkles	7			0.18	87-90	5
Cromer	Winkles	13				87-90	5
Newlyn	Winkles	9				87-90	5
Tarn Bay	Winkles	29				87-90	5
Southerness	Winkles	24		1.22	0.66	87-90	5
Kinlochleven	Winkles	38				87-90	5
Southerness	Winkles	25				1991	9
Tarn Bay	Winkles	23				1991	9
Torness	Winkles	13		0.62	0.08	1991	9
Sandside Bay	Winkles	18				1991	9
Fort William Pier	Winkles	30				1991	9
Kinlochleven	Winkles	38				1991	9
Tarn Bay	Winkles	19				1992	10
Tarn Bay	Winkles	13				1993	11
Tarn Bay	Winkles	12				1994	12
Tarn Bay	Winkles	10				1995	13
Tarn Bay	Winkles	13				1996	14
Tarn Bay	Winkles	18				1997	15
Newlyn	Winkles	8				Nov-87	16
Cromer	Winkles	7				Jul-87	16
Ravenglass	Winkles	9				1985	17
Colwyn Bay	Winkles	13-22				Jan-96-Jan-97	19
Western English Channel	Winkles	8				1988	21
Western English Channel	Winkles	8				1988	21
Median		13	0.87	0.75	0.18		

Ref No.	Authors	Date	Title
1	McDonald-P, Cook-G.T, Baxter-M.S.	1991	Natural and artificial radioactivity in coastal regions of UK. Radionuclides in the Study of Marine Processes. Elsevier Applied Science. 1991. pp 329-339
2	Ryan-T.P, Germain-P, Dowall-A.M, Leclerc-G, Pollard-D	1997	A comparison of ^{210}Po in *Mytilus edulis* and *Fucus vesiculosis* in French and Irish coastal waters Radionuclides in the Oceans. Radioprotection-colloques.1997. Vol. 32.C2.345-352

3	Camplin-W.C, Aarkrog-A,.	1989	Radioactivity in North European waters: Report of Working Group 2 of CEC Project MARINA Fisheries Research Data Report No.20 120 pp.
4	Pollard-D, Ryan-T.P, Dowdall-A.	1998	The Dose to Irish Seafood Consumers from ^{210}Po. Rad. Prot. Dosim. Vol 75, Nos 1-4, pp 139-142
5	Rollo-S.F.N, Camplin-W.C, Allington-D.J, Young-A.K.	1992	Natural radionuclides in the UK marine environment. Rad. Prot. Dosim. Vol 45 (1992) no1-4 203-209
6	Hunt-G.J	1988	A.E.M.R. Number 21
7	Hunt-G.J	1989	A.E.M.R. Number 23
8	Camplin-W.C.	1990	A.E.M.R. Number 29
9	Camplin-W.C.	1991	A.E.M.R. Number 34
10	Camplin-W.C.	1992	A.E.M.R. Number 38
11	Camplin-W.C.	1993	A.E.M.R. Number 42
12	Camplin-W.C.	1994	A.E.M.R. Number 45
13	Camplin-W.C.	1995	RIFE-1
14		1996	RIFE-2
15		1997	RIFE-3
16	Pentreath-R.J, Camplin-W.C, Allington-D.J.	1989	Individual and collective dose rates from naturally-occuring radionuclides in seafood. Radiation Protection-Theory and Practice. 4th Int. Symp. Malvern, Jun'89 pp 297-300.
17	McDonald-P, Baxter-M.S, Fowler-S.W.	1993	Distribution of Radionuclides in Mussels, Winkles and Prawns. Part 1. Study of Organisms under Environmental Conditions using Conventional Radio-analytical Techniques. J. Environ. Radioact. 18 (1993) 181-202
18	Skwarzec-B.	1988	Accumulation of ^{210}Po in Selected Species of Baltic Fish. J. Environ. Radioact. 8 (1988) 111-118
19	Wildgust-M.A, McDonald-P, White-K.N	1998	Temporal changes of ^{210}Po in temperate coastal waters Sci. Tot. Environ. 214 (1998) 1-10
20	Szefer-P, Wenne-R.	1987	Concentration of uranium and thorium in molluscs inhabiting Gdansk Bay, Baltic Sea. Sci. Tot. Environ 65 (1987) 191-202
21	Pentreath-R.J, Allington-D.J.	1988	Dose to man from the consumption of marine foods: A comparison of the naturally-ocurring ^{210}Po with artificially-produced radionuclides. Proceedings of 7th Int. Cong. of I.R.P.A., Sydney, April 10-17, 1988
22	Pentreath-R.J, Lovett-M.B, Harvey-B.R, Ibbett-R.D.	1979	In Biological Implications of Radionuclides Released from Nuclear Industries.Vol II, IAEA-SM-237, Vienna, (ST1/PUB/522) 2: 227-245
23	Swift-D.J,Smith-D.L, Allington-D.J. Ives-M.J.	1994	The ^{210}Po Content of North Sea Edible Crab, *Cancer pagerus* L., and Common Shrimp, *Crangon crangon* L. and the Potential Radiological Impact. J. Environ. Radioact. 23 (1994) 213-230

24	Aarkrog-A et al.	1997	A Comparison of Doses from ^{137}Cs and ^{210}Po in Marine food: A Major International Study. J. Environ. Radioact. 34 No.1 (1997) 69-90
25	Dahlgaard-H.	1996	Polonium-210 in Mussels and Fish from the Baltic-North Sea Estuary. J. Environ. Radioact. 32 Nos.1-2 (1996) 91-96
26	McDonald-P, Fowler-S.W. Heyraud-M, Baxter-M.S	1986	Polonium-210 in Mussels and its Implication for Environmental Alpha-Autoradiography J. Environ. Radioact. 3 (1986) 293-303
27			CEFAS Archived data
28	Cherry-R.D, Heyraud-M.	1981	Polonium-210 Content of Marine Shrimp: Variation with Biological and Environmental Factors. Mar. Biol. 65 (1981) 165-175