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Activity concentrations of ^{137}Caesium and ^{210}Polonium in seafood from fishing regions of New Zealand and the dose assessment for seafood consumers

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ABSTRACT

A study was undertaken to determine activity concentrations for ^{134}Caesium, ^{137}Caesium and ^{210}Polonium in New Zealand seafood, and establish if activity concentrations varied with respect to species/ecological niche and coastal region. Thirty seafood samples were obtained from six fishing regions of New Zealand along with a further six samples of two commercially important species (hoki and arrow squid) with well-defined fisheries. ^{134}Caesium was not detected in any sample. ^{137}Caesium was detected in 47% of samples, predominantly in pelagic fish species, with most activities at a trace level. Detections of ^{137}Caesium were evenly distributed across all regions. Activity concentrations were consistent with those expected from the oceanic inventory representing residual fallout from global nuclear testing. ^{210}Polonium was detected above the minimum detectable concentration in 33 (92%) of the analysed samples. Molluscs displayed significantly elevated activity concentrations relative to all other species groups. No significant regional variation in activity concentrations were determined. Two dose assessment models for high seafood consumers were undertaken. Dose contribution from ^{137}Caesium was minimal and far below the dose exemption limit of 1 mSv/year. Exposure to ^{210}Polonium was significant in high seafood consumers at 0.44–0.77 mSv/year (5th–95th percentile). ^{137}Caesium is concluded to be a valuable sentinel radionuclide for monitoring anthropogenic releases, such as global fallout and reactor releases, in the marine environment. ^{210}Polonium is of importance as a natural radionuclide sentinel due to its high contribution to dietary committed dose in seafood consumers.

1. Introduction

With 15,000 km of coastline and a 6.7 million km^2 Exclusive Economic Zone (EEZ), respectively the tenth and sixth largest of any country in the world, the marine environment is a significant and valuable resource to the New Zealand population (Coriolis, 2014). Seafood is collected and harvested on various scales, with a number of quota management systems in place and certain marine species having significant value in terms of trade. Seafood has importance to the New Zealand population as a source of nutrition and is consumed in considerable amounts by some sectors of the community (Tipa et al., 2010; Turner et al., 2005). Chemical contaminants in seafood can therefore lead to significant health burdens to the population and it is an important public health function to identify contaminants of concern and characterise their exposure.

The presence of radionuclides in the environment has been a significant global concern over the last half century. Following the recent accident at the Fukushima Daiichi Nuclear Power Plant, concern has been raised regarding the potential impact of radionuclide release into the Pacific Ocean on seafood, and consequently seafood consumers. Radionuclide monitoring of the marine environment surrounding New Zealand has been limited to date. A much greater focus has been placed on identifying and quantifying terrestrial fallout, through atmospheric dry and wet deposition and through monitoring milk powders from various regions of the country (Matthews, 1993).

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Global release of anthropogenic radionuclides from nuclear weapons testing has also contributed to the levels of radionuclides in the environment. Historically, for the southern hemisphere the most significant contributor to marine anthropogenic radionuclide activities has been nuclear weapons testing. Direct input of global fallout into the South Pacific Ocean (S 30°–60°) has been calculated at 25.8 PBq and 41.3 PBq for ^{90}Strontium (^{90}Sr) and ^{137}Caesium (^{137}Cs) respectively (IAEA, 2005). Oceanic anthropogenic radionuclide activities for four latitudinal boxes of the Pacific and Indian Ocean surrounding New Zealand were estimated for the start of the millennium as being 0.4–0.8 Bq/m^3 for ^{90}Sr and 0.6–1.4 Bq/m^3 for ^{137}Cs (Povinec et al., 2004).

New Zealand is not a nuclear power generating country and its position in the South Pacific places it a considerable distance from nuclear power generating facilities. The closest nuclear reactor is the research reactor (OPAL) at Lucas Heights, Sydney, Australia, approximately 2000 km distant. The closest commercial power reactors are Northern hemisphere plants in Taiwan, Japan and China, all further than 8000 km away, and the closest of the seven Southern hemisphere reactors, in Argentina, is approximately 10,000 km away.

With the input of a significant inventory of radionuclides from the Fukushima-Daiichi nuclear accident into the Northern Pacific, concerns have been raised in many countries surrounding the Pacific as to the potential increase in risks that may result. Modelling of the oceanic distribution suggests that ^{137}Cs from Fukushima-Daiichi will elevate Tasman Sea and South-West Pacific radionuclides by 0.01 Bq/m^3 by 2026, with continuing dispersion through the region over the subsequent 15 years (Nakano and Povinec, 2012).

Certain naturally occurring radioactive materials (NORM) including ^{238}Uranium (^{238}U) and its decay series are present in the marine environment through natural processes, such as terrestrial influx from atmospheric deposition or fluvial transport. As certain minerals can have higher abundances of NORM than are generally present in the crust, the extraction, processing and utilisation of these deposits can lead to concentration of NORM. This technological enhancement of NORM presents a risk for radionuclide entry into the environment from a number of industries which do not involve nuclear technology or nuclear fuel extraction (UNSCEAR, 2000).

A recent survey of radionuclides across the New Zealand diet sampled the muscle tissue of three fish species and three shellfish species for levels of anthropogenic and naturally occurring radionuclides (Pearson et al., in this issue). Of the radionuclides surveyed ^{137}Cs and ^{210}Polonium (^{210}Po) were found mainly in seafood species. ^{137}Cs activity was consistently detected in the higher trophic level fish but was absent in shellfish, while ^{210}Po showed greater variation, with considerably higher levels in shellfish and a roughly hundred-fold difference between tuna and lemonfish. ^{234}Uranium and ^{238}U activities were present in all samples, with significant activities in shellfish, but did not show any elevation in the finfish species over that of terrestrial food sources.

Given the range and magnitude of radionuclides present in seafood it was determined that monitoring of radionuclides in the seafood species may be necessary to provide a complete and accurate portrayal of dietary radionuclide exposure in the New Zealand population. Such an undertaking would complement the current milk monitoring program for terrestrial contamination. To support a seafood monitoring programme, additional research is necessary to better characterise the ranges, and any regional differences, of radionuclides in various New Zealand seafood species including through establishing suitable sentinel radionuclides for monitoring long term trends. The recorded ^{137}Cs and ^{210}Po activities in the analysed seafood samples highlight their suitability as marine monitoring sentinels. ^{137}Cs is a representative anthropogenic radionuclide, with properties such as, significant fission yield, high mobility in the environment and potential for uptake in the marine food chain. ^{210}Po is a marker for NORM sources and also displays the potential for high uptake levels in seafood species. Developing a dataset for ^{137}Cs and ^{210}Po in seafood will also provide a more refined estimate of ionising radiation exposure within seafood diets.

In this paper we detail a monitoring program for ^{137}Cs and ^{210}Po activities in seafood, outlining the sampling from different fishing regions of New Zealand and interpreting the results in the context of the expected behaviour of the radionuclides in the marine environment. Finally the outcome of a dose assessment models for seafood consumers is presented.

2. Materials and methods

2.1. Sampling methodology

The EEZ of New Zealand is divided into ten general fishery management regions (Fig. 1). In order to obtain a wide geographic distribution, the boundaries of these regions were utilised to classify samples as occurring from different sections of the New Zealand coastline. The sampling protocol was designed to capture a range of key seafood species and also analyse for differences between ecological niches.

In addition to targeting niche-specific species in each management region (see sampling protocol in Table 1), two species were specifically targeted as being important commercial catch species for New Zealand. These were the teleost fish, hoki (*Macruronus novaezelandiae*), and the cephalopod mollusc, arrow squid (*Nototodarus* spp. – *Nototodarus gouldi*, *Nototodarus sloanii*). These are deep water species with well-defined fishing stock areas.

All samples specified in the protocol were obtained through

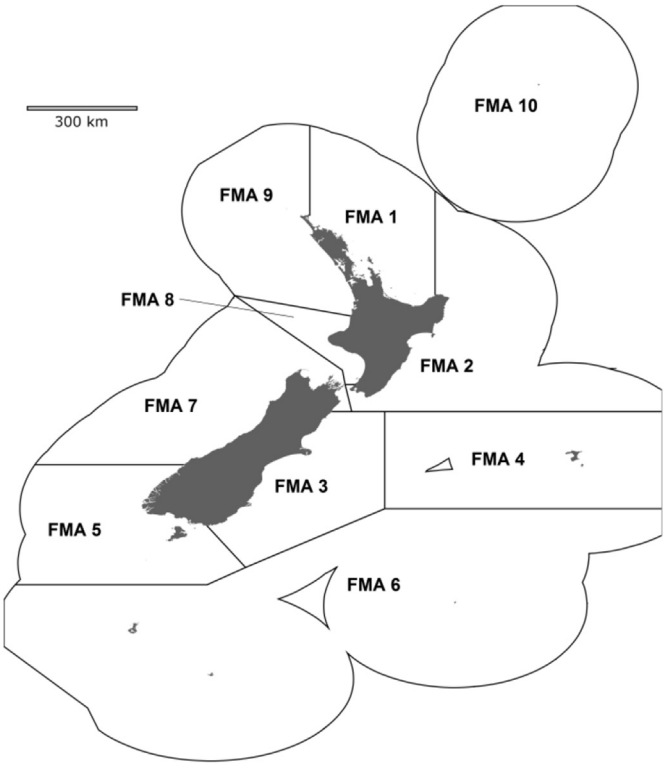

Fig. 1. Outline of New Zealand General Fisheries Management Areas (FMA) *(generated from NABIS (MPI)).*

Table 1
Seafood sampling protocol, with samples obtained, from New Zealand fishing regions for analysis of radionuclide content.

Fishery regions	Target species	Sampled species
Auckland and Kermadec (FMA1, FMA9, FMA10)	Oceanic pelagic fish	Skipjack tuna (*Katsuwonus pelamis*)
	Coastal pelagic fish	Tarakihi (*Nemadactylus macropterus*)
	Demersal fish	Snapper (*Chrysophrys auratus*)
	Non-molluscan invertebrate	Rock lobster (*Jasus edwardsii*)
	Mollusc	Greenshell mussels (*Perna canaliculus*)
Central (FMA2)	Oceanic pelagic fish	Southern bluefin tuna (*Thunnus maccoyii*)
	Coastal pelagic fish	Tarakihi (*Nemadactylus macropterus*)
	Demersal fish	Red cod (*Pseudophycis bachus*)
	Non-molluscan invertebrate	Rock lobster (*Jasus edwardsii*)
	Mollusc	Paua (*Haliotis iris*)
South East (FMA3, FMA4)	Oceanic pelagic fish	Orange roughy (*Hoplostethus atlanticus*)
	Coastal pelagic fish	Tarakihi (*Nemadactylus macropterus*)
	Demersal fish	Flounder (*Rhombosolea leporina*)
	Non-molluscan invertebrate	Rock lobster (*Jasus edwardsii*)
	Mollusc	Queen scallop (*Zygochlamys delicatula*)
Southland (FMA5)	Oceanic pelagic fish	School shark (*Galeorhinus australis*)
	Coastal pelagic fish	Tarakihi (*Nemadactylus macropterus*)
	Demersal fish	Blue cod (*Parapercis colias*)
	Non-molluscan invertebrate	2× Rock lobster (*Jasus edwardsii*)
	Mollusc	2× Bluff dredge oyster (*Tiostrea chilensis*)
Challenger/Central (Egmont) (FMA7, FMA8)	Oceanic pelagic fish	Albacore tuna (*Thunnus alalunga*)
	Coastal pelagic fish	Tarakihi (*Nemadactylus macropterus*)
	Demersal fish	Ling (*Genypterus blacodes*)
	Non-molluscan invertebrate	Kina (*Evechinus chloroticus*)
	Mollusc	Littleneck clams (*Austrovenus stutchburyi*)
Sub Antarctic (FMA6)	3 Oceanic pelagic or demersal fish	Southern blue whiting (*Micromesistius australis*)
		2× Ling (*Genypterus blacodes*)
Specific stock area	Hoki	3× Hoki (*Macruronus novaezelandiae*)
	Squid	3× Arrow squid (*Nototodarus* spp. – *N. gouldi*, *N. sloanii*)

commercial suppliers, with additional rock lobster and Bluff oyster samples also supplied from the Southland fishing region. The coastal pelagic fish was tarakihi (*Nemadactylus macropterus*) which was able to be collected from each coastal fishery region.

The full details of the species sampled from each region are listed in Table 1. A minimum of 140 g of edible tissues was supplied for each species and the date and region of harvest recorded.

2.2. Sample preparation and analysis

Samples were received chilled or frozen and allowed to thaw prior to homogenisation. Fish samples consisted primarily of muscle, with the majority of skin and large bones removed. Crustacean samples were prepared by removing inedible exoskeleton and any non-edible organs prior to homogenisation. For rock lobster this included removing hepatopancreas, which although is consumed, provided insufficient mass to meet analytical requirements. Kina were sampled as pre-removed gonads. Molluscs (excluding the squid) were removed from shells and homogenised. All samples were frozen prior to gamma spectroscopy to delay microbial spoilage during counting.

Samples were transferred to clean 400 ml cylindrical containers and analysed for gamma emissions using CANBERRA high purity germanium semiconductor detectors. Samples were counted for a minimum of 48 h. All spectra were analysed to derive emission counts for ^{134}Caesium (^{134}Cs) and ^{137}Cs. Activity concentrations of any other anthropogenic gamma-emitting radionuclide were recorded if present, and activity concentrations of ^{210}Lead (^{210}Pb) were recorded if above the Minimum Detectable Concentration (MDC) calculated for each assay.

A 5 g fresh weight sub-sample of each homogenised sample was used for the ^{210}Po assay. Samples were acid digested before extraction of ^{210}Po with autodeposition onto silver disks. Plated disks were counted for 23 h using passive implanted planar silicon (PIPS) detectors in a CANBERRA alpha spectrometer. ^{209}Polonium (^{209}Po) was added to sample digests as a yield tracer to estimate method recovery and results were corrected for decay to represent ^{210}Po activity concentrations at time of harvest. Activity concentrations for ^{210}Po were compared against any detectable activity concentrations of ^{210}Pb to determine the proportion supported and unsupported in each sample.

2.3. Data analysis

Emission counts for both isotopes were calculated using the CANBERRA Genie 2000 program. If sufficient activity was present for an isotope a method error based on 2 standard deviations was reported. Trace activity concentrations below the MDC can be detected but not quantified with absolute certainty; the indicative value reported was deemed a trace result. A large proportion of non-detect results complicates the calculation of arithmetic mean activities. To identify the likely ranges of ^{137}Cs mean activities, these have been reported as both lower-bound (LB), whereby all non-detect values were assigned a value of zero, and upper-bound (UB), where all non-detect values were assigned the value of the assay MDC. Trace results were included at the reported value for calculating both UB and LB means. For ^{210}Po all samples had detected or indicative trace activity concentrations and thus means were calculated from all values. Statistical analysis was conducted using the Mann–Whitney *U* test to analyse for differences between species from different ecological niches, and to test for differences between species from different regions. This allowed us to explore the hypothesis that these variables (niche and region) lead to populations with larger activity concentrations.

2.4. Dose assessment for high seafood consumers

To determine the exposure associated with a high or subsistence seafood consumption diet a high diet model consisting of consumption by an individual of 300 g finfish, 200 g crustacean or

squid and 100 g molluscs a week was used. These values were calculated from the mean daily consumption amounts in the 2008 Adult National Nutrition Survey of marine fish (82 g/person), squid (40 g/person) and molluscs (29 g/person) (University of Otago and MOH, 2011). Each commodity was conservatively estimated as being consumed four times over a week, with values rounded to the nearest 100 g. A model was developed in Microsoft Excel 2007 to randomly assign the activity concentrations of one of the samples of each group analysed in the seafood survey to each of the fifty two weeks of a year. A further fully probabilistic model randomly assigned a weekly dietary consumption of between 0 and 1000 g of each of the three seafood types.

Both models then derived an annual dose estimate by summing the total activities and converting through the International Commission on Radiological Protection (ICRP) established dose conversion factors for an adult (^{137}Cs: 0.013 μSv/Bq, ^{210}Po: 1.2 μSv/Bq) (ICRP, 2012). The models were run for 10,000 iterations for both the ^{137}Cs and ^{210}Po seafood analytical datasets. Exposure estimates at the 5th, 25th, 50th, 75th, 95th and 99th percentiles were then calculated.

3. Results and discussion

3.1. Gamma spectroscopy

All 36 seafood samples underwent gamma spectroscopy to determine ^{134}Cs and ^{137}Cs activity concentrations. Previous analysis of ^{137}Cs in New Zealand fish had indicated that activity concentrations were generally at a trace level (Pearson et al., in this issue). As such there was a requirement to obtain low MDCs in the region of 0.1–0.2 Bq/kg to determine the presence of ^{137}Cs. This was primarily achieved by using large analytical masses of 200–500 g and long counting times. Following analysis, all samples had interpretable spectra, which were utilised to calculate activities for ^{134}Cs and ^{137}Cs, and derive MDCs. None of the samples had detectable activity concentrations of ^{134}Cs. Seventeen of the 36 samples analysed had detectable ^{137}Cs activity concentrations. However, of these only two samples had activity concentrations exceeding the assay MDC. The remaining 15 recorded trace activity concentrations for ^{137}Cs, denoting that the isotope was present but the reported value was indicative only. Detected activities, with associated standard error at 95% confidence, and assay MDCs are reported in Table 2.

Detection of ^{137}Cs occurred in all the fishing regions, and of the samples tested was most common in the finfish, with some noted exceptions. Consistent with the absence of ^{137}Cs in shellfish from the previous dietary survey (Pearson et al., in this issue), invertebrates (including squid and shellfish) did not show any ^{137}Cs activity. The sole exception was a single oyster and rock lobster sampled from Southland. The other oyster and rock lobster samples obtained from the Southland fishing region did not have detectable ^{137}Cs activity concentrations. UB and LB means and ranges for each of the ecological niches were calculated (Table 3).

Detections of ^{137}Cs in the three tuna and one shark sampled were consistent with the results from lemonfish and tuna sampled in the radionuclide dietary survey (Pearson et al., in this issue). The higher trophic level of these species likely facilitated bioaccumulation of ^{137}Cs. Similar results have been reported in other surveys of fish. For example, in a study of fish from Oman ^{137}Cs activity was recorded in the predatory pelagic species, such as albacore tuna (0.5 Bq/kg), bluefish (0.18–0.43 Bq/kg) and barracuda (0.25 Bq/kg) (Goddard et al., 2003). However a study of ^{137}Cs activities in fish caught off the south coast of India did not identify any statistically significant trends in terms of the feeding habits of the fish type sampled (Feroz Khan and Godwin Wesley, 2012).

All samples of oceanic pelagic species had detectable activity concentrations of ^{137}Cs with the exception of orange roughy (*Hoplostethus atlanticus*). Orange roughy is a migratory pelagic fish. However it tends to inhabit very deep water, congregating around submerged seamounts, a behaviour which differs from that of the other oceanic pelagic species sampled (Clark, 1999). Concentration factors for mesopelagic species are not considered to differ from those of surface species (IAEA, 2004). The most likely explanation for the lack of detectable ^{137}Cs in this species is the reported decrease in seawater activity concentrations of this isotope in deeper waters of the South Pacific Ocean (Aoyama et al., 2011).

There is no evidence that transfer of ^{137}Cs from Fukushima-Daiichi contaminated water has occurred for the migratory pelagic species with ranges extending to New Zealand. This is further confirmed through the comparable activities that have been found in non-migratory species, such as hoki and tarakihi, and the absence of ^{134}Cs activity concentrations (Table 2).

Residual oceanic contamination from historical nuclear testing is the most probable source for the ^{137}Cs activity concentrations detected in the seafood samples. Extrapolating from ^{137}Cs oceanic activity concentrations prior to the Fukushima-Daiichi accident allows the likely contribution from this source to be determined. The activity concentrations of ^{137}Cs in 2000 in the oceanic regions surrounding New Zealand were 0.6–1.4 Bq/m^3 (Povinec et al., 2004). A further study in 2003–4 undertook analysis for ^{137}Cs in surface waters on a longitudinal crossing of the South Pacific (30–32.5°S), passing through the Tasman Sea and to the North of New Zealand (154–150°E) (Hirose et al., 2007). ^{137}Cs activity concentrations in this transect ranged from 1.2 to 1.7 Bq/m^3. Decay adjusted to 2015 expected ^{137}Cs activity concentrations would therefore range between 0.45 and 1.4 Bq/m^3 or approximately 0.44–1.4 mBq/kg seawater. Concentration factors of 100, 50, 60 and 9 have been published for the transfer of ^{137}Cs from seawater to finfish, crustaceans, molluscs and cephalopods respectively (IAEA, 2004). Considering this the expected ^{137}Cs activity concentration ranges for these species based on legacy global fallout would be 0.045–0.14 Bq/kg for finfish, 0.026–0.07 Bq/kg for crustaceans, 0.027–0.084 Bq/kg for molluscs and 0.004–0.013 Bq/kg for cephalopods. These ranges encompass the majority of the detected activity concentrations for ^{137}Cs found in the tested New Zealand seafood. The absence of detectable activity concentrations for ^{137}Cs in the sampled arrow squid is also supported by the low concentration factors for cephalopods. The lack of activity concentrations in squid may also be a factor of the reported rapid depuration of radio-caesium by cephalopods. A study examining cuttlefish exposed to ^{134}Cs through the diet gave biological half-lives of 16 days for adults and 66 days for juveniles. For juveniles exposed to ^{134}Cs through seawater, depuration was much faster at an average biological half-life of 6 days (Bustamante et al., 2006).

It is therefore highly likely that the primary source of ^{137}Cs in the New Zealand marine region is from legacy nuclear fallout, particularly as ^{137}Cs activity concentrations are present and consistent in species from the more remote Sub Antarctic Islands fishery region. The consistency of the detected activities with the expected concentrations ranges from the historical fallout inventory supports this.

As activity concentrations of ^{137}Cs in New Zealand resident species were at or below the level of analytical determination, the full ranges for the coastal pelagic and demersal species have not been fully elucidated. This larger proportion of left censored data precluded statistical analysis of the results for species or regional variation, however regional variation of ^{137}Cs activity concentrations in resident finfish species around New Zealand appears low. The results from this current study and the previous dietary survey (Pearson et al., in this issue) indicate that ^{137}Cs activity

Table 2
^{137}Cs and ^{210}Po activity concentrations ± standard error at 95% confidence, with assay MDCs for 36 samples of seafood collected from six New Zealand fishing regions and two species specific stocks.

Fishery regions	Sampled species	^{137}Cs activity concentration (Bq/kg ww)	^{137}Cs MDC (Bq/kg)	^{210}Po activity concentration (Bq/kg ww)	^{210}Po MDC (Bq/kg)
Auckland and Kermadec	Skipjack tuna	0.107 ± 0.043	0.147	32.77 ± 2.29	0.08
	Tarakihi	0.090 ± 0.043	0.135	2.90 ± 0.50	0.15
	Snapper	ND	0.113	0.31 ± 0.16	0.13
	Rock lobster	ND	0.171	0.71 ± 0.24	0.12
	Greenshell mussels	ND	0.081	11.33 ± 1.15	0.06
Central	Southern bluefin tuna	0.125 ± 0.035	0.132	1.11 ± 0.28	0.11
	Tarakihi	ND	0.109	1.07 ± 0.36	0.21
	Red cod	0.052	0.073	0.21 ± 0.14	0.11
	Rock lobster	ND	0.126	0.11 ± 0.13	0.21
	Paua	ND	0.133	2.71 ± 0.47	0.09
South East	Orange roughy	ND	0.11	0.05 ± 0.07	0.13
	Tarakihi	0.069	0.141	1.44 ± 0.38	0.12
	Flounder	ND	0.094	6.00 ± 0.83	0.08
	Rock lobster	ND	0.216	0.28 ± 0.12	0.07
	Queen scallop	ND	0.101	283.54 ± 15.89	0.11
Southland	School shark	0.213 ± 0.055	0.150	0.08 ± 0.08	0.10
	Tarakihi	ND	0.185	1.87 ± 0.38	0.09
	Blue cod	0.145	0.202	0.48 ± 0.18	0.09
	Rock lobster	ND	0.118	2.09 ± 0.37	0.09
	Rock lobster	0.088	0.169	1.50 ± 0.33	0.11
	Bluff oyster	ND	0.079	98.31 ± 5.80	0.09
	Bluff oyster	0.153	0.213	71.82 ± 4.37	0.07
Challenger/Central (Egmont)	Albacore tuna	0.144 ± 0.052	0.133	1.32 ± 0.37	0.09
	Tarakihi	0.060	0.088	2.71 ± 0.57	0.18
	Ling	0.104 ± 0.047	0.138	0.17 ± 0.13	0.12
	Kina	ND	0.057	2.40 ± 0.79	0.31
	Littleneck clams	ND	0.095	21.89 ± 1.62	0.09
Sub Antarctic	Southern blue whiting	ND	0.167	0.96 ± 0.35	0.15
	Ling	0.068	0.115	0.24 ± 0.16	0.16
	Ling	0.068 ± 0.041	0.104	0.38 ± 0.19	0.14
Specific stock area	Hoki	0.126 ± 0.049	0.128	1.91 ± 0.37	0.05
	Hoki	0.052	0.108	0.77 ± 0.29	0.14
	Hoki	0.092 ± 0.048	0.179	1.20 ± 0.30	0.06
	Arrow squid	ND	0.122	1.33 ± 0.74	0.34
	Arrow squid	ND	0.189	2.22 ± 0.97	0.35
	Arrow squid	ND	0.159	2.30 ± 0.96	0.60

ND: Not detected.

concentrations are within the ranges expected from the oceanic inventory of ^{137}Cs from global nuclear fallout.

Modelling of the future elevation of ^{137}Cs in the Pacific Ocean from the dispersion of the oceanic inventory released in the Fukushima-Daiichi accident, has estimated an elevation of upto 0.03 Bq/m^3 could occur in New Zealand coastal waters (Nakano and Povinec, 2012). By using the concentration factors for ^{137}Cs in various seafood groups this would result in an estimated increase of between 0.3 and 3 mBq/kg for species resident to New Zealand waters (IAEA, 2004). Based on the calculated seafood MDC of the gamma-spectroscopy protocol used in this study of 57–220 mBq/kg such an increase is unlikely to be resolvable from the current activity concentration ranges occurring from global nuclear fallout.

3.2. *^{210}Polonium analysis*

Calculated ^{210}Po recoveries ranged from 41 to 89% with a mean recovery of 71%. The mean relative standard deviation (RSD) between duplicates was estimated over five duplicate samples and one quadruplicate sample to determine the reproducibility of result. An average RSD of 20% was calculated. This relatively high value was likely a consequence of the heterogeneous nature of ^{210}Po in shellfish replicates, which likely stemmed from ^{210}Po being bound to retained particulates which were not evenly distributed during sample processing. By removing the three shellfish duplicates the RSD was reduced to 10%.

Detected activity concentrations, with associated standard error

Table 3
Upper-bound (UB) and lower-bound (LB) mean ^{137}Cs activity concentrations and ^{210}Po mean activity concentrations with standard deviation for 36 samples of New Zealand seafood.

Ecological niche/species	Mean ^{137}Cs LB:UB (Bq/kg ww) (range)	Mean ^{210}Po ± SD (Bq/kg ww) (range)
Oceanic pelagic fish	0.10–0.14 (0.11–0.21)	6.05 ± 13.10 (0.05–32.77)
Coastal pelagic fish	0.08–0.10 (0.06–<0.19)	2.00 ± 0.79 (1.07–2.90)
Demersal fish	0.05–0.09 (0.05–0.15)	1.11 ± 2.16 (0.21–6.00)
Non-molluscan invertebrate	0.01–0.13 (<0.06–<0.22)	1.18 ± 0.96 (0.11–2.40)
Mollusc	0.03–0.11 (<0.08–0.15)	81.60 ± 105.72 (2.71–283.54)
Hoki	0.09 (0.05–0.13)	1.29 ± 0.58 (0.77–1.91)
Arrow squid	0.00–0.16 (<0.12–<0.19)	1.95 ± 0.54 (1.33–2.30)
All samples	0.05–0.12 (0.05–<0.22)	15.57 ± 50.22 (0.05–283.54)

at 95% confidence, and assay MDCs for ^{210}Po are reported in Table 2. Higher MDCs were recorded in squid samples in comparison to the rest of the samples due to the one year time period between reported catch date and analysis, a factor that increased the analytical uncertainty. For the remaining samples analysis was undertaken 1–2 months after catch date thus providing an accurate estimate of ^{210}Po in the freshly caught sample. A total of 92% of the 36 samples analysed contained ^{210}Po activity concentrations above the MDC, with the remaining samples reporting trace activity concentrations below the MDC. Samples with trace activity concentrations included a shark species. This was not unexpected as the activity concentrations in lemonfish (rig shark) from the previous dietary survey research would have fallen below the MDC of this method (mean: 0.044 Bq/kg ww) (Pearson et al., in this issue).

The activity concentrations of ^{210}Po in species within the same ecological niches were generally consistent (Table 3). Statistical analysis indicated no significant differences ($p > 0.05$) between all fish groups (oceanic pelagic, costal pelagic and demersal) and non-molluscan invertebrates. However the mollusc population was significantly elevated in ^{210}Po activity concentrations relative to oceanic pelagic fish (Mann–Whitney U test, $Z = -2.322$, $N = 12$, $p = 0.02$), coastal pelagic fish ($Z = 2.374$, $N = 11$, $p = 0.018$), demersal fish ($Z = -2.786$, $N = 13$, $p = 0.005$) and non-molluscan invertebrates ($Z = -2.8$, $N = 12$, $p = 0.005$).

In the 2013/14 survey of radionuclides in the New Zealand diet, ^{210}Po activity concentrations were highest in shellfish species (Pearson et al., in this issue). This finding was confirmed in the current study, however the magnitude of the accumulation, and the variability between individuals, was much greater than we have previously shown, with accumulation varying by more than two orders of magnitude (Table 2). This variation is consistent with that reported overseas for shellfish (Table 4) and may relate to several factors such as species, season and region. For example, diverse species of molluscs have been reported to have differing susceptibility to accumulate ^{210}Po. Among molluscs collected in the intertidal zone of a region of the Portuguese coast of the North Atlantic activities ranged from 5.8 Bq/kg for common cockle (*Cerastoderma edule*) to 283 Bq/kg in common periwinkle (*Littorina littorea*) (Carvalho, 2011). A comparable range is visible in the New Zealand results with paua reported at 2.7 Bq/kg and queen scallops with an activity concentration of 283.5 Bq/kg.

Seasonal variation in the activity concentrations of ^{210}Po is common. The ^{210}Po activity in Mediterranean mussels (*Mytilus galloprovincialis*) increased four-fold from summer to winter (Aközcan and Uğur Görgün, 2013). The increase was correlated to the increased rainfall during autumn and winter with the source being increased land runoff of ^{210}Po. Another study examining temporal variation in ^{210}Po indicated that activity increased as a result of phytoplankton blooms (Wildgust et al., 1998), whereby increased scavenging of soluble ^{210}Po occurred due to the higher organic matter content in the water, with subsequent increased uptake by marine organisms. Activity is also expected to increase following spawning as body weight is lost causing a relative increase in concentrations in the soft tissue of the mollusc (Wildgust et al., 1998). Addressing seasonal variation in commercial seafood species is difficult due to the limited harvesting times of many of the species. The seafood samples in this survey were primarily obtained in the first half of the year from January to June. Consequently, based on the limited data, no trend can be analysed in the results to indicate if seasonal variation is significant.

The species analysed in the present study contained ^{210}Po activities that were generally consistent throughout the different fishing regions. This included tarakihi caught as the coastal pelagic species from all of the coastal regions and the benthic herbivorous species of rock lobster and kina. Intra-species variation between squid and hoki samples was also low. If the shellfish activity concentrations are excluded from the results, the mean seafood activity concentration of ^{210}Po is 2.4 Bq/kg with a standard deviation of 5.9 Bq/kg. For the largely sedentary or non-migratory species of fish and invertebrates the low regional variation is evidenced by the mean activity concentrations of these groups being 1.5 Bq/kg with a standard deviation of 1.3 Bq/kg.

No statistically significant differences ($p \geq 0.05$) were noted in ^{210}Po activity concentrations between any of the different regions. Overseas, regional differences in activities have been recorded. A comparison of Mediterranean mussels from two regions of the Turkish coast of the Aegean Sea showed a difference of 5–40 times the activity of ^{210}Po between the sites (Aközcan and Uğur Görgün, 2013). The higher activities in one site were considered to result from the discharge of a river system giving an influx of ^{210}Po from land runoff into the area. Regions containing industries that release NORM can also influence ^{210}Po in molluscs. Water in the vicinity of a coal powered power plant in Malaysia was noted by the authors as having higher ^{210}Po activity than other locations around the coast (Alam and Mohamed, 2011).

In the current study the highest recorded ^{210}Po value was in

Table 4
Variation in ^{210}Po activity concentrations reported in studies of shellfish overseas compared with New Zealand harvested shellfish.

Country of origin	Species sampled (Latin name)	^{210}Po activity concentration range (Bq/kg ww)	Reference
New Zealand	Bluff oyster (*Tiostrea chilensis*), Greenshell mussel (*Perna canaliculus*), Littleneck clam (*Austrovenus stutchburyi*), Paua (*Haliotis iris*), Queen scallop (*Zygochlamys delicatula*)	2.7–283.5	This study
New Zealand	Greenshell mussel (*Perna canaliculus*), Littleneck clam (*Austrovenus stutchburyi*), Pacific oyster (*Crassostrea gigas*)	20.8–29.4	Pearson et al., in this issue
Croatia	Mediterranean mussel (*Mytilus galloprovincialis*)	22.1–207.0	Rozmaric et al., 2012
Kuwait	Clams (*Marcia marmorata*, *Circe intermedia*, *Marcia opima*), Cockle (*Fulvia fragile*), Sea snail (*Stomatella auricular*, *Cerithium scabridum*)	2.7–53.3	Uddin and Bebhehani, 2014
India	Periwinkle (*Cerithium scabridum*)	13.5–58.9	Sunith Shine et al., 2013
India	Mussel (*Perna indica* & *Perna viridis*)	31–212.0	Feroz Khan et al., 2014
Slovenia	European flat oyster (*Ostrea edulis*), Mediterranean mussel (*Mytilus galloprovincialis*)	51.2–124.6	Štrok and Smodiš, 2011
Taiwan	Oyster (*Crassostrea gigas*)	23.4–126.0	Lee and Wang, 2013
UK	Blue mussel (*Mytilus edulis*), Common cockle (*Cerastoderma edule*), Common limpet (*Patella vulgata*), Common periwinkle (*Littorina littorea*), Whelk (*Buccinidae* spp.)	2.9–52.1	Young et al., 2002

queen scallops obtained from remote fishery areas in deeper waters off the Otago Peninsula. Influence from land based sources to this fishery is unlikely. Higher accumulation than in molluscs residing at shallower depths may be due to sediment contact or higher mobilisation of ocean-deposited ^{210}Pb into the deeper water. The gamma spectra for this sample showed gamma emission activity consistent with other ^{238}U decay nuclides, such as ^{234}Thorium, ^{214}Bismuth and ^{214}Lead, which would indicate that some degree of contribution of radionuclides from the surrounding sediment is occurring.

The most likely source of the ^{210}Po activities in the shellfish sampled in the current study is through their diet. This can be confirmed by using the activities ratio of ^{210}Pb and ^{210}Po to derive the contribution from the parent radionuclides in the uranium decay. As both ^{210}Po and its grandparent ^{210}Pb are present in the marine environment, accumulation of ^{210}Po in seafood species can derive either from direct accumulation of the radionuclide (unsupported ^{210}Po) or through accumulation of ^{210}Pb and its subsequent decay to ^{210}Po *in situ* (supported ^{210}Po). ^{210}Pb activity can be quantified from its gamma emission at 46 KeV. However, as the gamma emission has a low decay intensity (5%) the method sensitivity is poor, resulting in a higher MDC. As a consequence in this study only two shellfish samples had detectable ^{210}Pb activity concentrations. Comparison of the ^{210}Pb activity concentrations with those of the ^{210}Po in the two shellfish samples with reported ^{210}Pb activity concentrations gives an estimation of the supported ^{210}Po. The comparison of ^{210}Pb:^{210}Po ratio determined 97–98% of the ^{210}Po activity concentration was unsupported. The low percentage of ^{210}Po present from decay of grandparent ^{210}Pb indicates that the primary source is direct uptake of ^{210}Po from the environment. ^{210}Po is particle reactive in the marine environment and readily binds to organic matter. The binding of ^{210}Po to organic matter results in concentration factors as high as 30,000 into zooplankton and 70,000 into phytoplankton (IAEA, 2004), and thus the consumption of these primary consumers is likely to be the source of ^{210}Po in species higher up the food chain.

A single high activity concentration of ^{210}Po, 32.77 Bq/kg, was noted in a skipjack tuna (*Katsuwonus pelamis*) sample. This was considerably higher than the other species of tuna (albacore and southern bluefin) and the other recognised oceanic pelagic species analysed. Tuna data from other studies indicate that muscle ^{210}Po levels can vary considerably, ranging from 1.7 Bq/kg ww in the Eastern Pacific to 137 Bq/kg in the Mediterranean (Ruelas-Inzunza et al., 2012; Heyraud and Cherry, 1979). Yellowfin tuna from the south coast of India had a mean activity concentration of 19.9 Bq/kg (Feroz Khan and Godwin Wesley, 2012). What is notable about this latter study is that the activities of ^{210}Po reported in pelagic planktivorous species (e.g. anchovy, shad and sardine) were the highest of the finfish species sampled. Reported ^{210}Po activities in the pelagic planktivores ranged from 32.5 Bq/kg to 46.8 Bq/kg. The noted variation in ^{210}Po activity concentrations between tuna species and in comparison to planktivores supports the hypothesis that ^{210}Po activities reflect the diet of the species. As soluble ^{210}Po is scavenged to organic matter particulates, such as the small bacteria and algae present in plankton (Carvalho, 2011), planktivorous species take up more ^{210}Po than other species. Consequently the preference for consuming these species in an animal from a higher trophic level would influence the ^{210}Po uptake and retention in the tissues of the consumer.

3.3. Dose assessment for high seafood consumers

Seafood represents a significant source of nutrition for much of the population in New Zealand. The consumption of seafood in the diet in combination with the higher natural levels of ^{210}Po present likely makes it a high contributor to total dietary ionising radiation. Some sub-populations in New Zealand can be much higher consumers of seafood than the population average. For example where fish consumption is culturally important, those undertaking a large proportion of recreational fishing and seafood collection, and those relying on fishing and shellfish collection for sustenance due to economic reasons (Tipa et al., 2010; Turner et al., 2005). In the latest New Zealand adult nutrition survey, conducted in 2008, the mean consumption values for molluscs were 29 g/day, however consumers at the 97.5th percentile ingested 240 g/day (University of Otago and MOH, 2011). This sub-population is potentially exposed to higher levels of ionising radiation due to the presence of foods with greater natural radionuclide activity concentrations.

To estimate the ranges of exposure a high seafood consumer may be receiving from natural and anthropogenic sources through their diet a high consumption model was developed. Data from the seafood assays (Table 2) was used to assign likely activities for both ^{210}Po and ^{137}Cs occurring in the diets of a high seafood consumer over the course of a year. The model was run both semi-probabilistically, through the use of set weekly consumption values, and fully probabilistically, through assigning a random weekly dietary consumption for each seafood type of between 0 and 1 kg. Estimates at a range of percentiles for ingested dose due to ^{137}Cs and ^{210}Po in seafood were obtained from the high consumer model (Table 5).

The results of the dietary modelling show the current ^{137}Cs activity concentrations in seafood represent a minimal contribution to the total dose for high seafood consumers. This exposure differs little with the types of seafood species consumed over a year with a negligible difference in dose between the 5th and 95th percentiles. When compared to the current ICRP dose limit of 1 mSv, for exposure of the general public to radiation above background, ^{137}Cs in New Zealand seafood currently represents a negligible concern (ICRP, 2007).

In comparison, the dose from natural ^{210}Po exposure is approximately 10,000 times higher, ranging up to a 99th percentile exposure of 0.78 mSv in the semi-probabilistic model and 4.05 mSv in the probabilistic model. The ^{210}Po dose varied considerably depending on the composition of seafood making up the diet, with the range between the 5th and 95th percentiles encompassing 0.28 mSv (0.44–0.72 mSv) in the fixed consumption and 1.7 mSv (1.87–3.61 mSv) in the random consumption models. This range can be expected given the large variation in the activities between different species of each seafood group. Individual consumers with a large intake of shellfish, in particular scallops and oysters, are likely to receive the highest dietary doses of radionuclides across the New Zealand population.

The large contribution of ^{210}Po activity concentrations in seafood to dietary doses has been reported for a range of other countries, including India and Lebanon (Feroz Khan and Godwin Wesley, 2011; Aoun et al., 2015). In a further study in India, average consumption of values of 25 kg/yr for each seafood type were determined to lead to annual committed doses of 0.59 mSv, 0.61 mSv and 3.36 mSv from ^{210}Po in marine finfish, prawns, and crabs, respectively (Kannan et al., 2001). In this later study despite seafood species accounting for only 3% of the composition of the Indian diet model they accounted for 81% percent of the dietary dose of ^{210}Po.

The dose from ingestion of ^{210}Po is part of the variation in natural background radiation between diets. Of interest is an estimate of the dose prehistoric coastal dwellers of South Africa, the Khosian, received from a characterised high shellfish diet (Heyraud et al., 1994). Based on modern day monitoring of shellfish from the Cape of Good Hope it was calculated that the Khosian would have received an annual dose of 4 mSv as a result of ^{210}Po. This dose is

Table 5
Probabilistic and semi-probabilistic exposure estimates for high seafood consumers of the annual committed dose through recorded activity concentrations of ^{137}Cs and ^{210}Po in New Zealand seafood.

Probabilistic model	Radionuclide	High seafood consumer dietary radionuclide exposure at *n*th percentile (μSv/person/year)					
		5th	25th	50th	75th	95th	99th
Semi	^{137}Cs	0.046	0.047	0.048	0.049	0.051	0.052
Full	^{137}Cs	0.11	0.12	0.12	0.12	0.13	0.14
Semi	^{210}Po	443	520	576	635	721	784
Full	^{210}Po	1874	2312	2652	3028	3605	4047

comparable to the maximum obtained in the full probabilistic modelling for New Zealand high seafood consumers and suggests that doses of this magnitude are ubiquitous for seafood consuming populations.

4. Conclusions

This study of activities of ^{137}Cs and ^{210}Po in New Zealand seafood has determined their respective current activity concentration ranges. ^{137}Cs activity was present predominantly in finfish, but all detected activity concentrations were very low and occurred in a small range above and below the MDC. No apparent variation was present between regions which is consistent with the source of ^{137}Cs being the diffuse residue from historical nuclear weapons fallout. No evidence of contribution to ^{137}Cs activities from the Fukushima-Daiichi nuclear accident has been noted in either migratory or non-migratory species. Forecasting future influx of ^{137}Cs, based on published modelling, it is unlikely that any elevation in New Zealand resident seafood species could be measured using the current analytical methods.

^{210}Po was present in the majority of seafood samples. Activities for molluscs showed a large range from 2 to 283 Bq/kg and this ecological niche was significantly elevated over the other seafood groupings. Ranges of ^{210}Po activity concentrations for other seafood species tended to be less diverse. No significant regional variation was recorded. Analysis of ^{210}Pb in two mollusc samples suggests that the ^{210}Po present is primarily accumulated through the diet.

Dose estimates for high seafood consumers based on the derived activities indicate that current activities of ^{137}Cs represent a minimal dietary concern. ^{210}Po activities, however, can contribute significantly to the dietary dose of ionising radiation for high seafood consumers, although the magnitude varies considerably depending on the composition of seafood species consumed. Due to the activities of ^{210}Po, the high seafood consuming sub-population is likely to receive the highest dietary ionising radiation doses for the New Zealand population.

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