

# Seafood dose parameters: Updating 210Po retention factors for cooking, decay loss and mariculture

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- 1 Seafood Dose parameters: updating <sup>210</sup>Po retention factors for cooking, decay loss and mariculture
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### 23 Abstract

<sup>210</sup>Po has been identified as one of the main contributors to ingestion doses to humans, particularly

25 from the consumption of seafood. The amount of <sup>210</sup>Po activity concentration data for various types

26 of seafood has increased greatly in recent times. However, to provide realistic seafood dose

27 assessments, most <sup>210</sup>Po data requires correction to account for losses that can occur before the

28 seafood is actually consumed. Here we develop generic correction factors for the main processes

29 associated with reduction of <sup>210</sup>Po in seafood – leaching during cooking, radioactive decay between

30 harvest and consumption, and sourcing from mariculture versus wild-caught.

31 When seafood is cooked, the overall mean fraction of <sup>210</sup>Po retained is 0.74 for all cooking and

32 seafood types, with the means for various seafood types and cooking categories ranging from 0.56

to 1.03 . When considering radioactive decay during the period between harvest and consumption,

34 the overall mean fraction remaining is 0.81 across all seafood processing/packaging types, with

35 estimates ranging from 0.50 (canned seafood) to 0.98 (fresh seafood). Regarding mariculture

influence, the available data suggest marine fish and crustaceans raised with processed feed have
 about one order of magnitude lower (0.10) <sup>210</sup>Po muscle content than wild-caught seafood of the

38 same or similar species.

39 Overall, this study concludes that <sup>210</sup>Po activity concentrations in seafood at the time of ingestion are

40 reduced to only about 55% compared to when it was harvested. Therefore, correction factors must

41 be applied to any data derived from environmental monitoring in order to achieve realistic dose

42 estimates. The data also suggest lower <sup>210</sup>Po ingestion doses for consumers who routinely favour

43 cooked, long shelf-life and farmed fish/crustaceans. However, more data is needed in some

categories, especially for cooking of molluscs and seaweed, and for the <sup>210</sup>Po content in all farmed
 seafood.

- 46
- 47
- 48 Keywords

49 Polonium, farmed seafood, aquaculture, ingestion dose, loss of <sup>210</sup>Po, marine monitoring

50

### 51 **1. Introduction**

52 In marine waters, <sup>210</sup>Po is constantly being replenished via atmospheric deposition of its parent

radionuclides as well as dissolution from sediments (Fowler, 2011; IAEA, 2017; Jeffree et al., 1997). It

54 is therefore ubiquitous in world ocean and coastal waters and is readily available for uptake by

55 marine organisms (Carvalho, 2011, 2018; IAEA, 2017). Seafood typically has much higher <sup>210</sup>Po

56 activity concentrations than foods from terrestrial sources (IAEA, 2021). As <sup>210</sup>Po also has a relatively

57 high ingestion dose conversion factor due to its 5.3 MeV alpha emission (Adult DCF=1.2x10-6; ICRP,

58 2012), the consumption of seafood has been recognised as an important dose pathway for humans

59 (IAEA, 1995, 2021; UNSCEAR, 2008).

60 Given the importance of this radionuclide to ingestion dose, the <sup>210</sup>Po activity concentrations used in

61 seafood dose assessments must be accurate to achieve realistic results. Most of the available <sup>210</sup>Po

62 data comes from environmental monitoring studies which report the Bq kg<sup>-1</sup> present when the

- 63 marine organism is harvested. However, between harvest and consumption, some of this <sup>210</sup>Po is
- 64 typically lost. It may leach during cooking when tissue protein structures are altered or denatured by

- heating (Kondjoyan et al., 2013). In addition, <sup>210</sup>Po is lost due to radioactive decay in the period
- between harvest and consumption, and these losses can be substantial given the relatively short
- 67 <sup>210</sup>Po half-life of 138 days (Delacroix et al., 2002).
- 68 A further factor that should be considered is the difference in the <sup>210</sup>Po content of wild-caught
- 69 seafood as opposed to that from mariculture (farmed or aquaculture in the marine environment as
- 70 opposed to freshwater aquaculture). Mariculture-raised fish may have markedly different
- radionuclide content as compared with wild-caught fish depending on the type of rearing conditions
- such as the use of processed feed types. However, as most available <sup>210</sup>Po data are derived from
- raise environmental monitoring studies that report on wild-caught specimens (e.g., 99.4% of marine fish,
- 74 MARIS database accessed 2022; IAEA, 2022), the use of such data without applying the necessary
- correction factors may overestimate the ingestion dose contribution from <sup>210</sup>Po. Such correction is
   important as finfish mariculture currently provides a substantial proportion of the total marine fish
- supply, approximately 22% in Europe and 11% globally (EUMOFA, 2020; 2018 FOA data summarised
- in NOAA, 2021), with future global supply projections predicted to increase (e.g., 17-27% by 2050:
- 79 Costello et al., 2020; Naylor et al., 2021).
- 80 The need to factor for the effects of cooking, decay, and mariculture has been recognised in previous
- 81 key publications (Fowler, 2011; IAEA, 1995, 2021; UNSCEAR, 2008); and some of these publications
- have provided parameters for correcting the raw <sup>210</sup>Po data (at harvest) for use in dose assessments.
- 83 For example, Aarkrog et al. (1997) proposed the use of 0.6 as a generic decay factor for seafood
- 84 which corresponded to a typical Harvest-to-Consumption delay period of about 90 days. However,
- since the 1997 Aarkrog et al. study, no substantive update to this parameter has been made even
- 86 though there have been major changes in seafood diet patterns (Hallström et al., 2019), in the
- transportation and distribution networks that deliver seafood (Guillen et al., 2019) and in processing
- and packaging technologies (Kontominas et al., 2021). Because of these changes, updated <sup>210</sup>Po
   loss/retention correction factors are needed to support current and future seafood dose
- 90 assessments.
- 91 In this paper, we develop generic seafood <sup>210</sup>Po correction factors for the:
- 92 <sup>210</sup>Po content change during cooking
- 93 Loss of <sup>210</sup>Po from radioactive decay during the period from Harvest-to-Consumption
- 94 <sup>210</sup>Po content differences between mariculture vs wild-caught seafood
- We address these three factors in the order of their relative impact on typical seafood ingestiondose. This paper makes use of global data to provide for broad applicability. However, dose
- 97 assessments are typically conducted on smaller-scale population groups (e.g., national, sub-national
- 98 and localised). Therefore, a further aim of this publication is to clarify best-practice methods for
- 99 deriving <sup>210</sup>Po loss/retention correction factors so that a consistent approach can be adopted in
- 100 these smaller-scale evaluations as well as to update the generic parameters over time as more global
- 101 data become available.

### 102 2. Methods

- 103 The <sup>210</sup>Po activity concentration data available for seafood dose assessments are typically gathered
- 104 from environmental monitoring studies on wild marine organisms and are typically reported for the
- 105 uncooked tissues at the time of harvest (removal from marine waters). However, for calculating
- 106 ingestion dose, the following should be considered: <sup>210</sup>Po losses due to cooking, physical decay
- 107 between harvest and consumption, and the effect of consuming mariculture seafood ("farmed"
- seafood is also used and in this paper always refers to marine conditions). The corrected <sup>210</sup>Po
   activity concentration at the time of ingestion can be represented as:

110 
$$AC_{ingest} = AC_0 \bullet \sum_c f_c(R_{cooking})_c \bullet \sum_d f_d(R_{decay})_d \bullet \sum_f f_f(R_{farmed})_f$$
(1)

111 Where:

112	AC <sub>ingest</sub>	activity concentration at the time of ingestion.
113	$AC_0$	activity concentration at the time of harvest (removal from marine waters).
114	$f_{ m c}$	fraction of seafood consumed using cooking method c.
115	<b>R</b> <sub>cooking</sub>	fraction of $^{210}$ Po activity concentration retained after cooking method <i>c</i> .
116	$f_{\sf d}$	fraction of seafood consumed for preservation method <i>d</i> .
117	<b>R</b> <sub>decay</sub>	fraction of $^{210}$ Po activity concentration remaining after preservation method d.
118	$f_f$	fraction of seafood from source type <i>f</i> (farmed w various feed types, wild-caught).
119	<b>R</b> <sub>farmed</sub>	<sup>210</sup> Po activity concentration ratio of source type $f$ (source: wild-caught, section 3.3).
120		

- 121 When considering the application of the above equation, the R factors used in any assessment
- 122 should match the habits of the population being assessed (or the "representative person" of that
- 123 population; ICRP, 2006). If assessment-specific information is available, it should supersede the use
- of any generic factors. For example, if the period between harvest and consumption is known for a
- specific seafood being assessed, then the standard radioactive decay equation for <sup>210</sup>Po should be
- 126 used instead of the  $R_{decay}$  factors presented here.
- 127 This paper presents broadly applicable factors calculated from available global data. For example,
- 128 the  $f_d$  values used here are world medians of Fresh=44%, Frozen=34%, Canned=11%, Dried=7%,
- Smoked=3%, (NOAA publication (Sun et al., 2022). based on FAO 2018 data). For the R<sub>farmed</sub>
- 130 component, the intent is to provide a more accurate estimate of the <sup>210</sup>Po content of that portion of
- fish and crustaceans sourced from mariculture (e.g., 11% (global) and 22% (Europe) of fish are
- 132 sourced from mariculture (EUMOFA 2020).
- 133 The <sup>210</sup>Po R<sub>cooking</sub>, R<sub>decay</sub> and R<sub>farmed</sub> factors presented in this paper are generic values that have been 134 assembled using the following process: data search on global sources, quality assurance on the 135 source study, categorisation and statistical analysis as described in the following sections and 136 supplementary material, followed by reduction by summary statistics and comparison with 137 previously published values. The parameters are derived from data that often represent stochastic 138 processes (e.g., durations from seafood purchase to ingestion for a population of consumers). Data 139 distribution tests are used as indicated (e.g., Kolmogorov-Smirnov test for normality). However, for 140 some categories where there was little available data and the distributions were indeterminate, the 141 non-parametric Kruskal-Wallis test was used for comparison among data groups. The results tables 142 provide a range of indicators where the data allow (arithmetic mean (AM), standard deviation (SD), 143 geometric mean (GM), geometric standard deviation (GSD), median, minimum, maximum, number 144 of data (N)).
- 145 2.1 Methods for determining <sup>210</sup>Po activity concentration changes during cooking

- 146 The change in <sup>210</sup>Po content during cooking is determined by comparing the measured Bq kg<sup>-1</sup>
- 147 content of cooked samples with that of the corresponding raw samples and can be expressed as a
- 148 retention factor (R<sub>cooking</sub>). While cooking effects on the radionuclide content of foods were identified
- early in the nuclear age (Forrester, 1962; Whicker, 1982), most of the usable data on seafood have
- 150 emerged from research published in recent decades. Data were gathered by searching global
- sources (1992 to 2022) focused on scientific journals, science conference proceedings, industry
- 152 reports, government documents and websites. Studies were assessed for quality assurance of
- 153 methods. Data were grouped by seafood and cooking categories appropriate for dose assessment
- 154 (details in section 3.1.1 and S.1). Some sources reported directly on retention (or loss) of
- radionuclides from cooking, while other sources reported raw vs cooked activity concentrations
- 156 from which retention values were calculated.
- 157 The R<sub>cooking</sub> values were developed using a total of N=63 experimental data, the largest amount of
- which was for the cooking of fish (N=42), followed by crustaceans (N=16) and bivalves (N=5). No data
   were found for <sup>210</sup>Po loss during the cooking of gastropods, cephalopods and seaweed. Key
- references for <sup>210</sup>Po retention were (in order of the amount of data provided): Uddin et al. (2019),
- 161 Díaz-Francés et al. (2017), Roselli et al. (2020), Štrok and Smodiš (2011), Medley and Patterson (2022)
- 162 , Kristan et al. (2015) and the Radioactive Waste Management Center (RWMC), Japan, (1994). When
- 163 compiling data on the effects of cooking, additional reference data were found for the seafood
- 164 cooking losses of other radionuclides that are often important to dose (e.g., <sup>90</sup>Sr, <sup>137</sup>Cs, <sup>210</sup>Pb,
- 165 <sup>226,228</sup>Ra) which are briefly summarized here as they may also be of benefit during dose assessments.
- 166 Unless specifically noted, all activity concentration data are in Bq kg<sup>-1</sup> fresh mass (wet weight).
- 167 The R<sub>cooking</sub> factors are grouped relative to cooking methods with grilling and frying methods
- 168 combined as they have higher heat potential and are applied as a so-called "dry heat." Boiling and
- steaming data are grouped as they share similar heat levels (assumed at standard pressure) and are
- "wet" cooking methods. This categorisation is consistent with past reports (IAEA 1992, 2009). Dry vs
- 171 wet methods may differ in their effectiveness of fragmenting molecules and subsequent dissolution
- and leaching from tissues.
- 173
- 174 2.2 Methods for determining <sup>210</sup>Po retention after radioactive decay from Harvest-to-Consumption
- 175 The standard decay equation for <sup>210</sup>Po decay and ingrowth (from <sup>210</sup>Pb ) is:

176 
$$AC_{t,P_0} = AC_{0,P_0} e^{-\lambda_2 t} + AC_{0,P_0} \frac{\lambda_2}{(\lambda_2 - \lambda_1)} (e^{-\lambda_1 t} - e^{-\lambda_2 t})$$
 (2)

- 177 Where:
- activity concentration of <sup>210</sup>Po at the time of ingestion. 178 AC<sub>t,Po</sub> initial activity concentration of <sup>210</sup>Po (e.g., at the time of harvest).  $AC_{0,Po}$ 179 initial activity concentration of <sup>210</sup>Pb (e.g., at the time of harvest). 180 AC<sub>0,Pb</sub> radionuclide decay constants;  $\lambda_1$  (<sup>210</sup>Pb) = 8.52x10<sup>-5</sup> d<sup>-1</sup>,  $\lambda_2$  (<sup>210</sup>Po) = 0.005 d<sup>-1</sup>. λ 181 t time duration over which decay and ingrowth is being calculated. 182 183 184 The above equation can be used directly if the delay period between harvest and ingestion is known. However, in a typical seafood dose assessment, t may vary substantially across the 185
- assessment population according to the varied types of seafood preservation methods (e.g., fresh,
- frozen, canned, etc.), each of which can have differing durations for transport, processing, and
- 188 shelf-lives. Delay times within categories are also influenced by consumer behaviour, with some

- consumers eating seafood soon after purchase and others waiting until the "use by" date.
   Therefore, when representing populations, delay times for seafood are inherently imprecise and
- 191 can best be estimated stochastically.
- 192

193 The component of the equation (2) for the ingrowth of <sup>210</sup>Po (from <sup>210</sup>Pb) may be relatively minor,

- and may add only an incremental amount, especially for products with short production/storage
- durations. The ingrowth component requires input of the initial activity concentration <sup>210</sup>Pb relative
- to <sup>210</sup>Po (the ratio of <sup>210</sup>Pb to <sup>210</sup>Po activity concentrations at the time of harvest). For this study, a
   generic <sup>210</sup>Pb/<sup>210</sup>Po ratio of 0.16 was used (<sup>210</sup>Po/<sup>210</sup>Pb ratio of 6.3) which was obtained from 770
- generic <sup>210</sup>Pb/<sup>210</sup>Po ratio of 0.16 was used (<sup>210</sup>Po/<sup>210</sup>Pb ratio of 6.3) which was obtained from 770
   paired <sup>210</sup>Po and <sup>210</sup>Pb data after filtering data for edible tissues and considering the seafood types
- and proportions consumed globally (fish=56.8%, Crustacean=14.4%, Bivalve=17.0%,
- 200 Gastropod=2.4%, Cephalopod=3.9% and seaweed=5.4%; Cai and Leung, 2017).
- 201
- Data on the delay durations for seafood were gathered using a search on global sources (through
   2022) focused on scientific journals, industry reports and government documents. Data were sought
- for five generic categories of seafood preservation (Table 1). While these categories are highly
- simplified relative to the numerous types of processing and packaging methods that are available
- 206 (Kontominas 2021), they are fit-for-purpose for dose assessment in that cumulatively they
- 207 represent almost all seafood types available for purchase and that consumption information on
- these generic categories is often available. For dose calculation, a more complex categorisation of
- 209 methods is currently not justified given that more detailed consumption data are typically not
- available (e.g., the diet proportions from different kinds of freezing techniques such as pressure-
- 211 shift vs isochoric freezing).

212	Table 1. For calculating typica	l delay durations.	data were grouped	into five generic	categories:
					00.00000.000

Category	Key descriptors/assumptions
Fresh	Refrigerated/chilled but not frozen or otherwise preserved.
Frozen	Frozen until thawed for consumption.
Canned	Sterilised and packaged for room-temperature storage, includes cans, retort pouches, glass jars and similar packaging that can be sterilised.
Dried	Dehydrated & packaged for room-temperature storage, includes dried, cured and similar products.
Smoked	Refrigerated, cold & hot-smoked processing. Does not include sterilized seafood packaged for long-term room-temperature storage (included in Canned category).

- 213
- 214 The detailed procedure for compiling Harvest-to-Consumption duration data and estimating the
- 215 <sup>210</sup>Po retention factors (R<sub>decay</sub>) is presented in the supplemental (S.2). In brief, for each seafood
- 216 preservation category, available reference data were compiled on the chain of events between
- 217 harvest and consumption. Data were grouped into pre-market termed here "Harvest-to-Market"
- 218 (e.g., harvest, transport to processing location, processing, storage, transport to market), and market
- 219 plus post-market events (storage at market/display, consumer storage) termed here "Market-to-
- 220 Consumption." This grouping is necessary because Harvest-to-Market events are dominated by
- 221 industrial-type production processes while the Market-to-Consumption events are dominated by
- retail and consumer behaviours. Where necessary, bounding data (minimum and maximums) are
- 223 used along with the stochastic distributions indicated by reference studies. For example, for fresh

- 224 seafood, an exponential distribution best represents the Market-to-Consumption delay times as
- retailers desire to sell fresh seafood quickly and most consumers tend to eat fresh seafood within
- two days of purchase (Hicks, 2016; Østli et al., 2013; Roccato et al., 2017). The estimates for total
- delay times (*t*) combine the duration data distributions across the entire Harvest-to-Consumption
- 228 period via Monte Carlo methods (used here via Goldsim<sup>™</sup> software, https://www.goldsim.com).
- Total delay durations were rounded to the nearest day as more precision is not warranted by the available data. See Supplemental for details of each seafood category.
- The above process yielded generic delay times for the five seafood processing types, which, in turn,
- 232 provide <sup>210</sup>Po retention factors  $R_{decay}$  ( $R_{decay}$ =  $AC_{t,Po}$  /  $AC_{0,Po}$  using equation 2). They are stochastic 233 estimates based on limited data and are intended to be updated over time as more data become
- available. These new factors are compared with available previously published values.
- 235 2.3 Methods for determining <sup>210</sup>Po differences between farmed and wild-caught seafood
- 236 Differences in activity concentrations between mariculture-sourced vs wild-caught seafood are
- determined through the straight-forward comparison of measurements in comparative samples. A
- 238 search was conducted focused on scientific journals, science conference proceedings, industry
- reports, government documents and websites (through 2022). In this study, the <sup>210</sup>Po activity
- 240 concentrations of the mariculture vs wild-caught were compared for the same or similar species
- 241 from the same region.
- The available data on <sup>210</sup>Po activity concentrations in the edible tissues of mariculture seafood were few (N=16) and limited to fish and crustaceans. The primary publications for fish were: Heldal et al.
- 244 (2019), Kong et al. (2021) Jalili et al. (2009), Smith (2004). Only limited data were found from one
- study on changes in <sup>210</sup>Po in mariculture vs wild-caught crustaceans. More data are needed for this
- 246 category as they are a major seafood source with approximately 9.4 million tonnes of crustaceans
- 247 harvested from mariculture each year (2018 FAO data reported by (2018 FAO data reported by
- Albalat et al., 2022). Data from bivalve mariculture is not included as processed feeds are mainly
- 249 used only for broodstock/larval stages and not for the period of primary bivalve growth which
- typically is caried out in natural marine settings (Smaal et al., 2018). Similarly, there is no need for
- correction of data for seaweed mariculture where this occurs in natural settings.
- 252 **3. Results and discussion**
- 253 *3.1 <sup>210</sup>Po change during cooking*

## 254 *3.1.1 Data and recommended* R<sub>cooking</sub> factors

255 Recommended cooking correction factors (R<sub>cooking</sub>) have been developed for fish, crustaceans and bivalves (Table 2) but not for other seafood categories due to lack of data. The mean overall 256 retention of <sup>210</sup>Po for all cooking methods and all seafood types is 0.74 (AM; SD=0.21 for combined 257 258 fish, crustaceans and bivalves). When comparing cooking methods across all data, the grilled/fried 259 (dry) retention is somewhat higher (AM=0.78, SD=0.21, N=34) than boiled/steamed retention 260 (AM=0.68, GSD=0.20, N=29) which is consistent with a conceptual understanding of greater leaching of metals from 'wet' cooking methods (e.g., greater loss of Pb in shrimp meat from boiling vs grilling; 261 Abd-Elghany et al., 2020). However, the retention difference between the grilled/fried vs 262 263 boiled/steamed groups is not strong (p=0.02, Kruskal-Wallis test), and one substantive study reported no significant <sup>210</sup>Po retention difference between grilled and boiled methods (Uddin et al., 264 265 2019). Additional data are needed to clarify any difference between these cooking methods.





Figure 1. Histogram of published values of the fraction of <sup>210</sup>Po activity concentration retained after cooking. Data include marine fish, crustaceans, and bivalves (cephalopod, gastropod and seaweed data not available). Data are for the edible tissues, without stock, broth or other leached liquid (see

text). See Table 2 and Table S1.1 for source data summaries and references.

272

273 The variation in <sup>210</sup>Po retention data examining the impact of cooking is substantial. The means of

the various categories range from 0.56 to 1.03 and the source individual data range from 0.19 –

1.24). In particular, significant variation may result from whether and how the digestive tract is

included or treated during preparation, cooking and consumption. Higher <sup>210</sup>Po activity

concentrations occur in seafood where the digestive tract is included compared to the muscle alone(Table S1.1).

279

280 Table 2. R<sub>cooking</sub> fractions of <sup>210</sup>Po activity concentrations retained after cooking (ratios of Bq kg<sup>-1</sup>

281 cooked mass / Bq kg<sup>-1</sup> fresh mass). AM= arithmetic mean, SD= standard deviation, GM= geometric

282 mean, GSD= geometric standard deviation, Med=median, Min= minimum, Max=maximum. See Table

### 283 S1.1 for source data.

	AM	SD	GM	GSD	Med	Min	Max	Ν
All cooking methods:	0.74	0.21	0.70	1 2	0.76	0.10	1 7 4	62
(Fish, Crustacean, Bivalve)	0.74	0.21	0.70	1.2	0.76	0.19	1.24	63
Fish <sup>1,2,3,6</sup>	0.75	0.21	0.72	1.4	0.79	0.19	1.24	42
Crustacean <sup>1,4,6,7</sup>	0.68	0.20	0.65	1.3	0.69	0.35	1.06	16
Bivalve <sup>2,5,7</sup>	0.75	0.26	0.71	1.41	0.62	0.49	1.03	5
<u>Grilled, Fried</u>								
All grilled/fried seafood	0.78	0.21	0.74	1.4	0.78	0.19	1.12	34
Fish <sup>1,2,3</sup>	0.76	0.21	0.72	1.4	0.79	0.19	1.13	25
Crustacean <sup>1,4</sup>	0.81	0.12	0.80	1.2	0.78	0.75	1.06	7
Bivalve <sup>5</sup>	1.03	0.01				1.02	1.03	2
Boiled, Steamed								
All boiled/steamed seafood	0.68	0.20	0.66	1.3	0.69	0.38	1.27	29
Fish <sup>1,6</sup>	0.75	0.21	0.66	1.4	0.66	0.38	1.24	17
Crustacean <sup>1,4,6,7</sup>	0.58	0.20	0.55	1.4	0.54	0.35	1.24	9
Bivalve <sup>2,7</sup>	0.56	0.07	0.56	1.1	0.57	0.49	0.62	3

<sup>1</sup>Uddin et al. (2019) (N=42), <sup>2</sup>Díaz-Francés et al. (2017) (N=9), <sup>3</sup>Štrok and Smodiš (2011) (N=2), <sup>4</sup>Medley and
 Patterson (2022) (N=2), <sup>5</sup>Kristan et al. (2015) (N=2), <sup>6</sup>Roselli et al. (2020) (N=2), <sup>7</sup>Radioactive Waste

286 Management Center (RWMC), Japan, (1994), (N=2).

287 Overall, the retention data are approximately normally distributed (Kolmogorov-Smirnov test; D(63)

= 0.09, p=0.05) and therefore the following recommendations are based on the arithmetic means
 (AMs) of Table 2:

290 When considering different cooking methods (all seafoods; stock/leachate/broth is discarded):

- $R_{cooking}$  (all grilled/fried/boiled/steamed) =0.74 ± 0.21
- 292  $R_{cooking}$  (grilled/fried ) =0.78 ± 0.21
- 293  $R_{cooking}$  (boiled/steamed) =0.68 ± 0.20

### 294 When the seafood and stock/leachate/broth are both fully consumed (no loss):

• R<sub>cooking</sub> =1.0

In practice, some of the <sup>210</sup>Po can be lost to deposition on the dish/cooking implements or otherwise
 may not be eaten, and when data on such losses are known the above generic estimate could be
 refined.

In some instances, it may be useful to estimate cooking losses by seafood type and recommended
 fractional retention values for three different types of seafood (fish, crustacean and bivalve; Table 2)

301 were calculated from the data for all cooking methods and when the stock/leachate/broth is

- 302 discarded:
- 303 R<sub>cooking</sub> (fish) =0.75 ± 0.21
- 304  $R_{cooking}$  (crustacean) =0.68 ± 0.20

### 305 • R<sub>cooking</sub> (bivalve) =0.75 ± 0.26

- 306 No previous comprehensive summary reference on cooking loss/retention was found for307 comparison of these retention values.
- 308 3.1.2 Variation in cooking correction factors and the need for more data
- 309 Of the highest retention values in this compilation, the majority were in the crustacean and bivalve
- 310 categories, where the hepatopancreas was included during cooking. In crustaceans, the
- hepatopancreas alone typically has 1-2 orders of magnitude higher <sup>210</sup>Po than muscle tissues
- 312 ((Carvalho, 2018; Fowler, 2011; Medley and Patterson, 2022). Uddin found 84% less <sup>210</sup>Po activity
- 313 concentration in shrimps with all of the digestive tract removed vs. whole uncooked shrimp. In some
- cooking methods, the digestive tract/hepatopancreas are included during cooking but are not
   consumed (e.g., a shrimp is cooked whole, then the tail meat alone is consumed). In such situations,
- 316 cross-contamination of the consumed portions may occur when they are cooked along with tissues
- 317 containing higher activity concentrations of  $^{210}$ Po, which can result in retention values > 1.0 relative
- to the edible portion (Medley and Patterson, 2022; Uddin et al., 2019).
- 319 In some seafood cooking methods, the liquid leached from the seafood tissues is retained in the
- 320 stock (broth, gravy). This stock may receive concentrated <sup>210</sup>Po if the digestive tract/hepatopancreas
- 321 are included. For example, 42-110 Bq kg<sup>-1</sup> leached into the stock/broth after one-hour boiling of
- whole prawn samples including digestive tract organs (Medley and Patterson, 2022). Similarly, 20-36
- 323 Bq kg<sup>-1</sup> leached into the stock from shrimp samples with the digestive tract included (Uddin et al.,
- 324 2019). Consumption of such stock will therefore result in higher levels of <sup>210</sup>Po ingestion compared
- 325 with muscle tissues alone.
- 326 Further variation in the cooking effect on <sup>210</sup>Po activity concentrations may derive from differences
- 327 in the methods used by individual studies including differing approaches to compositing samples
- 328 (combined samples vs individual specimens) as well as use of differing heating levels and cooking
- durations (e.g., 20 vs 60 minute boiling durations respectively in (e.g., 20 vs 60 minute boiling
- durations respectively in; Medley and Patterson, 2022; Uddin et al., 2019).
- 331 Overall, more data are needed to better understand the effect of different cooking methods and the
- aforementioned variables, particularly for crustaceans and bivalves where fewer (N=21) studies have
- been published than for fish (N=42). We found no data on the effect of cooking on <sup>210</sup>Po in
- 334 gastropods, cephalopods and seaweed.
- 335

### 337 3.1.3 Retention/loss during cooking of other important radionuclides

338 While this paper focuses on <sup>210</sup>Po, some comparative data were found for other radionuclides that

339 can be important contributors to seafood ingestion dose. The anthropogenic radionuclides <sup>90</sup>Sr and

<sup>137</sup>Cs become important during and after nuclear accidents or other heightened release scenarios

341 (Aoyama et al., 2016; Johansen et al., 2015; Takata et al., 2019; Tateda et al., 2015). Studies on the

342 change of <sup>137</sup>Cs content during frying, baking or grilling of marine fish indicate post-cooking retention

values of: 0.84 (Rantavaara, 1989), 0.72-1.05 (Nabeshi et al., 2013) and 0.8-0.9 (IAEA, 2009).
Reported retention values after boiling or steaming of marine fish were: 0.35-0.68 (RWMC, 1994)

- and 0.2-0.9 (IAEA, 2009). Similar studies on bivalves indicated post-cooking retention values of: 0.8
- 346 (steamed, Quinault, 1989), 0.64-1.30 (boiling) and 0.49 (sautéed) (Masson et al. 1989). For <sup>90</sup>Sr, a
- single retention value of 0.9 was found for "fish: boiling flesh" (IAEA, 2009)).

348 For seaweeds, no conventional cooking effects data were found. However, Maro et al. (2002)

reported on radionuclide loss during the extraction of gelling/thickener products (alginic acid, high

350 milk reactive, satiagum) from seaweed for use in various foods. For <sup>137</sup>Cs and <sup>90</sup>Sr, about 75% and

351 44% were eliminated respectively. The extractions include acid leaching, among other steps, and the

degree of radionuclide elimination exceeded 90% for <sup>106</sup>Ru-Rh, <sup>60</sup>Co, <sup>129</sup>I, <sup>238</sup>Pu, <sup>239+240</sup>Pu, <sup>241</sup>Am and

353 <sup>244</sup>Cm.

354 Other natural radionuclides such as <sup>210</sup>Pb, <sup>228</sup>Ra and <sup>226</sup>Ra also typically contribute to ingestion dose

355 (IAEA 2021). Concentrations of stable Pb decreased in crabs and shrimp after grilling (0.72) and

boiling (0.59) (Abd-Elghany et al., 2020), while increases and decreases (0.77-1.7) were observed in

357 coastal fish of Nigeria after grilling, frying and boiling (Bassey et al., 2014). The retention values from

358 these studies are higher than the values published by the IAEA for Pb retention in shrimp (0.4) and

359 for "lower sea organisms" (0.4-0.5) (IAEA, 2009). For <sup>226,228</sup>Ra, no data on the loss/retention during

360 cooking were readily available.

361

## 362 3.2 <sup>210</sup>Po loss from radioactive decay during Harvest-to-Consumption

Given the relatively short 138-day half-life of <sup>210</sup>Po, seafood dose calculations should correct for the 363 loss of <sup>210</sup>Po due to radioactive decay between the time seafood is harvested to when it is consumed 364 (Harvest-to-Consumption). During this period, substantial decreases in <sup>210</sup>Po activity concentrations 365 366 can occur, especially for preserved seafood with longer processing times and shelf lives. If the delay between harvest and consumption is known, then the standard decay equation for <sup>210</sup>Po should be 367 used. However, most dose assessments are for a varied population of consumers who consume 368 varied types of seafood and therefore a stochastic approach may be used. The recommended R<sub>decav</sub> 369 factors represent the fraction of <sup>210</sup>Po remaining (retained) after the radioactive decay that occurs 370 between harvest and consumption for various seafood processing/preservation types using global 371 372 reference data (through 2022).

# 373 *3.2.1* R<sub>decay</sub> factors for <sup>210</sup>Po in fresh, frozen, canned, dried and smoked seafood

374 For fresh seafood, an R<sub>decay</sub> correction factor of 0.98 is recommended (Table 3, Table S2.1) which

375 corresponds to a mean Harvest-to-Consumption duration of approximately 5 days (rounded from

5.2). As used here, "fresh" seafood includes chilled products, but does not include frozen-thawed

- products (included in the frozen category). Key data references include (Baptista et al., 2020;
- 378 McManus et al., 2014; Østli et al., 2013; Roccato et al., 2017; Simpson et al., 2012). As a reference

- 379 example of Harvest-to-Market timing, fresh farmed salmon were shipped from Chile to the US in 2.7
- 380 days in a study simulating typical transport of fresh seafood (Simpson et al., 2012). After purchase,
- most consumers (>50%) reported a preference for consuming fresh seafood within two days, but 381
- with refrigerated shelf lives of up to 11-14 days possible (McManus et al., 2014; Østli et al., 2013). 382
- 383 Such Market-to-Consumption duration data for fresh seafood (from multiple studies) were best fit
- 384 by an exponential distribution (Hicks, 2016; Østli et al., 2013; Roccato et al., 2017).
- 385 For, frozen, canned, dried and smoked seafood, the respective mean Harvest-to-Market durations are 89, 177, 51 and 13 days (Table 3). These correspond to R<sub>decay</sub> factors of 0.70 for frozen, 0.50 for 386 387 canned, 0.81 for dried and 0.95 for smoked (Table 3 and Tables S2.1, S2.2 for uncertainties and 388 reference information). As an example of frozen seafood Harvest-to-Market information, in China, 389 Xiao et al. (2016) reported a duration of 22 days for a typical transport/storage (cold chain) 390 sequence of events, with the longest link being the transport from processing to market (14 days).
- 391 As an example of current production of canned seafood, Boerder et al. (2018) provided quantitative
- 392 data on tuna captured in the southern Indian Ocean, canned in Texas, US, and then distributed to
- 393 various markets. They estimated that it took about six months on average (18 to 35 weeks) from the
- 394 catch until the canned tuna finally appeared on the shelf for sale. See Table S2.2 for references and
- 395 information summaries.
- 396 Where possible, the above specific factors for fresh, smoked, frozen, canned and dried categories
- should be used proportionally to reflect the diet of the consumer populations being evaluated. 397
- When it is necessary to calculate the radioactive decay of <sup>210</sup>Po in generic seafood (all combined), a 398
- mean R<sub>decay</sub> factor of 0.81 is recommended for use when considering the typical global seafood 399
- consumer diet (the weighted mean using consumption percentages of 44% fresh, 34% frozen, 11% 400
- canned, 7% dried, and 3% smoked (based on 2018 FAO data and reported in NOAA, 2021) 401
- Table 3. Recommended  $R_{decay}$  factors representing the fractions of <sup>210</sup>Po remaining after typical 402
- Harvest-to-Consumption durations for various seafood processing/preservation types (2022 403
- 404 estimates). See Tables S2, S3 for supporting information.

	Total delay, Harvest-to-	Recommended	References <sup>2</sup>
		R <sub>decay</sub> factors	
	50 <sup>°°</sup> (range) <sup>5</sup>	Recommended (range)	
	(days)		
Fresh	5 (0-17)	<b>0.98</b> (0.93-1.0)	1-5
Frozen	89 (1-540)	<b>0.70</b> (0.21-1.0)	6-10
Canned	177 (10-2070)	<b>0.50</b> (0.14-0.96)	9-11
Dried	51 (10-410)	<b>0.81</b> (0.14-0.96)	12-15
Smoked	13 (2-62)	<b>0.95</b> (0.78-0.99)	6,16-22
All seafood	57	0.81	
(weighted) <sup>4</sup>	(0-2070)	(0.14-0.1.0)	

- 405
- <sup>1</sup> Harvest-to-Consumption includes Harvest-to-Market (transport, processing and storage times from the time 406 of removal from the ocean until arrival at retail marketplace) and Market-to-Consumption (warehouse, display 407 and consumer storage times from the time of arrival at the market until consumption).
- 408 <sup>2</sup> References: 1 (USDA, 2022), 2 (Østli et al., 2013), 3 (Simpson et al., 2012), 4 (McManus et al., 2014), 5
- 409 (Baptista et al., 2020), 6 (Roccato et al., 2017), 7 (Tingman, 2010), 8 (Xiao et al., 2016), 9 (Limbo et al., 2009),
- 410 10 (Kontominas et al., 2021), 11 (Tsironi et al., 2016), 12 (Boerder et al., 2018), 13 (Immaculate et al., 2013), 14
- 411 (Obluchinskaya and Daurtseva, 2020), 15 (Foscarini and Prakash, 1990), Various sources (seaveg.com), 168
- 412 (Bienkiewicz et al., 2022), 17 (Afchain et al., 2005), 18 (Chardon and Swart, 2016), 19 (Daelman et al., 2013), 20
- 413 (Pouillot et al., 2007), 21 (Pouillot et al., 2010), 22 (Erkan et al., 2011)

- 414 <sup>3</sup>ranges are minimum maximum for distributions used in this study's Monte Carlo analysis.
- <sup>4</sup>Percentage weighting is world median consumption proportions of: Fresh=44%, Frozen=34%, Canned=11%,
  Dried=7%, Smoked=3%, (NOAA publication (Sun et al., 2022). based on FAO 2018 data).
- 417 The data compiled for this work are drawn from many studies, including review/summary studies.
- 418 Some studies considered various types of seafood (fish, crustaceans, bivalves, etc.) and many studies
- focus on marine fish which is consistent with fish contributing the largest proportion to world
- 420 consumption (Cai and Leung, 2017). However, the available duration data are limited in some
- 421 categories and further data would better define the stochastic distributions of Harvest-to-
- 422 Consumption timings for various seafood processing/packaging products. Such data may emerge
- 423 from source-certification and ecolabelling trends (<u>https://www.fao.org/in-action/globefish</u>), as well
- 424 as advances in technologies, such as Radio Frequency Identification (RFID) on food packages which
- 425 has the potential to generate large datasets that document seafood consumer preferences and shelf
- 426 times (Bibi et al., 2017), Furthermore, such data would allow for a better definition of the
- 427 proportions of processing/packaging types for specific seafood categories (fish, crustaceans, etc.).
- 428 *3.2.2* Comparison with previously published information on <sup>210</sup>Po delay times loss during Harvest-to-429 Consumption
- 430 Comparing the derived time durations of this study (Table 3) with previous estimates (Table S2.3),
- 431 the five days for fresh seafood used in this study is similar to the three days used in (Aarkrog et al.,
- 432 1997). For frozen seafood, the 89 days of this study is comparable to the 90 days value for wild-
- 433 caught frozen seafood in Jones and Sherwood (2009) but higher than the 60 days value for general
- 434 frozen seafood in Aarkrog et al. (1997) (Table S2.3 for more reference information). That the current
- 435 estimate for frozen seafood has a slightly longer delay duration than the 1997 Aarkrog value may
- 436 reflect technological improvements, increased varieties of freezing methods and more complex
- 437 global processing/business arrangements (Kontominas et al., 2021).
- 438 For canned seafood, the recommended delay period in this work (177 days) reflects shorter
- 439 durations than the previous estimate of 365 days in Aarkrog et al. (1997) which may reflect
- 440 improvements in distribution systems (Anderson et al., 2018) and shorter transport times such as
- the air transport as described in Boerder et al. (2018) and Simpson et al. (2012). For smoked
- seafood, Jones and Sherwood (2009) include smoked marine fish in with "fresh" with a
- 443 representative delay estimate of 8-10 days which is slightly shorter, but not substantially different
- 444 than the 13-day recommended estimate of this study. The recommended  $R_{decay}$  in this work (0.50;
- 445 177 days) also includes the ingrowth of  $^{210}$ Po (from  $^{210}$ Pb) which increases the recommended R<sub>decay</sub>
- 446 value of this work (compared with 0.16 in Aarkrog et al. (1997).
- 447 For overall generic seafood, the recommended R<sub>decav</sub> estimate from this work (0.81) includes the ingrowth of <sup>210</sup>Po (from <sup>210</sup>Pb) which results in a slightly higher value than if the ingrowth was not 448 449 included as is the case for past estimates. The overall  $R_{decav}$ =0.81 estimate (as compared with 450 previous dominantly used value of 0.6 (90 days) from Aarkrog et al. (1997)) is also consistent with 451 changing consumer patterns which show a greater proportion of fresh seafood consumed currently 452 (44%, FAO 2018 data) compared with 30% value used be Aarkrog et al. in 1997). Concurrently, 453 canned seafood consumption has decreased to 11% today from 20% used in 1997 (Aarkrog et al., 454 1997). In addition to these changes in consumption patterns, distribution systems have improved 455 allowing faster harvest to market delivery times (e.g., via air transport vs ground transport). The combination of these factors has contributed to shorter overall Harvest-to-Consumption timings 456 resulting in radioactive decay factors (R<sub>decay</sub>) closer to unity (i.e., less loss of <sup>210</sup>Po as a result of 457
- 458 radioactive decay).

### 460 3.3 Differences in the <sup>210</sup>Po activity concentrations in farmed vs wild-caught marine fish

#### 461 3.3.1 Available data and recommended R<sub>farmed</sub> factors

Reported measurements on <sup>210</sup>Po content in the muscle tissues of mariculture fish are few (total 462 summary data N=16 based on N=54 samples) and highly varied (0.003 to 10.4 Bq kg<sup>-1</sup>; Table 4). While 463 few, the data indicate the <sup>210</sup>Po activity concentrations in the muscle tissues of some mariculture fish 464 are very low as compared with wild-caught fish. This difference is clearly indicated when considering 465 farm-raised Atlantic salmon (Salmo salar), with a mean <sup>210</sup>Po activity concentration ((GM=0.02, 466 GMSD=3.7; Heldal et al., 2019; Smith, 2004) of about one order of magnitude lower than that of 467 468 wild-caught fish from the same region (0.14-0.17 Bq/kg; Komperød et al., 2020). Consistent with this, the mean <sup>210</sup>Po/<sup>210</sup>Pb ratio of 0.24 for the maricultured salmon as reported in Heldal et al. (2019) is 469 very low (<sup>210</sup>Po-depleated) compared with wild seafood (mean ratio of 6.3 from 770 paired <sup>210</sup>Po and 470 <sup>210</sup>Pb data after filtering data for edible tissues as described above and in S2). 471

472 In addition to salmon, which are prominent in commercial mariculture production (EUMOFA, 2020),

473 other data from similar industrial-scale operations is available for Gilthead seabream (*Sparus* 

474 *aurata*)(Jalili et al., 2009), and the mean for the combined fish raised on processed feed (GM=0.03

475 Bq kg<sup>-1</sup>, GSD=4.2) is nearly three orders of magnitude lower than the generic global value for wild-

476 caught fish (GM=2.1 Bq kg<sup>-1</sup>, GSD=5.1; global summary, MARIS 2022)(p < 0.01, Kruskal-Wallis test).

477 The main reason for such differences appears to be the use of processed feed in mariculture

478 production facilities. The processed feeds can have lower <sup>210</sup>Po content (compared to natural marine

479 food webs) due to decay during processing/storage time intervals, and the nature of the feed mix

480 sources. The feeds that are typically used in industrial scale mariculture are mainly derived from

481 terrestrial-origin vegetable material (e.g., soy, sunflowers, rapeseed, corn, broad beans and wheat)
482 augmented by smaller proportions of terrestrial animal products (e.g., poultry meal, feather meal,

482 augmented by smaller proportions of terrestrial animal products (e.g., poultry meal, feather meal,
483 blood meal), as well as smaller amounts of marine animal products (e.g., fish meal, crustacean meal,

484 fish oil and salmon oil) and micro-ingredients (e.g., vitamins, mineral mixtures)(Skretting, 2022). In

485 salmon feed, marine protein and oils accounted for only about 14% and 10% by mass, respectively

486 (2016 data; NOFIMA, 2022).

In contrast, other mariculture fish have been reported to have higher <sup>210</sup>Po activity concentrations in
muscle tissues (GM=2.4 Bq kg<sup>-1</sup>, GSD=3.6) (Kong, 2021), that were not distinguishable from generic
wild-caught fish (GM=2.1 Bq kg<sup>-1</sup>, GSD=5.1); global summary, MARIS 2022)(p > 0.05, Kruskal-Wallis
test). In these cases, the fish were raised on "small fish and shrimp" which were marine-sourced
(personal communication Yangin Ji, 24 January 2023), and thus can accumulate <sup>210</sup>Po to levels that

492 are similar to those that feed on natural marine food webs.

For crustaceans, a similar decrease in the <sup>210</sup>Po content in the muscle tissues of mariculture vs wild-493 caught is indicated although the data are few and limited to one study (Table 4; Dong et al. 2018). 494 The R<sub>farmed</sub> factors in Table 4 group into distinct categories that are distinguished by the type of feed 495 496 used to raise the fish and crustaceans. The mariculture fish and crustaceans raised on processed feed mixtures that are mainly derived from of terrestrial plant sources have lower <sup>210</sup>Po content than 497 498 those raised on natural marine-sourced products. Although the data are sparse (and a controlled study has yet to be reported) they suggest an approximate one order of magnitude decrease (R<sub>farmed</sub> 499 of 0.1). This outcome is consistent with previously reported findings that <sup>210</sup>Po is mainly taken up by 500 501 marine organisms via their dietary sources (IAEA, 2017, 2021; Komperød et al., 2020).

Table 4. Summary <sup>210</sup>Po activity concentrations (Bq kg<sup>-1</sup> fresh mass) in the muscle tissues of

504 mariculture fish. Also shown are the recommended R<sub>farmed</sub> values (ratio of <sup>210</sup>Po activity

505 concentrations in muscle of farm-raised vs wild-caught). GM= geometric mean, GSD= geometric

506 standard deviation, AM= arithmetic mean, SD= standard deviation, Min= minimum, Max=maximum.

								Recomm
Species and	Common							ended
reference	name	$GM^1$	GSD	AM	SD	Min	Max	$R_{farmed}$
		Bq kg⁻¹		Bq kg⁻¹	Bq kg⁻¹	Bq kg⁻¹	Bq kg⁻¹	
Fish raised with		0 03	1 20	0.064	0.070	0 003	0 1 9 1	0 1 <sup>2</sup>
processed feed		0.05	4.20	0.004	0.070	0.003	0.101	0.1
Salmo salar <sup>3</sup>	Atlantic Salmon	0.010	2.31	0.013	0.009	0.003	0.023	
Salmo salar <sup>4</sup>	Atlantic Salmon	0.072	2.75	0.094	0.05	0.016	0.13	
Sparus aurata⁵	Gilthead seabream			0.181	0.022			
Fish raised with								6
natural marine-		2.42	4.1	4.26	5.3	0.99	10.4	<b>1.0</b> °
sourced feed								
Larimichthys	Yellow							
polyactis, Pagrus	croaker, Red							
major, Lateolabrax	sea bream,	2.28	2.71	3.92	4.22	0.99	10.4	
japonicus'	Common sea perch							
Crustaceans raised								
with processed		0.88	9.80	3.72	5.87	0.12	10.5	<b>0.1</b> <sup>8</sup>
feed								
Litopenaeus	Whiteleg							
vannamei,	shrimp,	0.88	9.80	3.72	5.87	0.12	10.5	
Penaeus monodon <sup>9</sup>	Tiger prawn							
All Data <sup>8</sup>		0.1	8.7	1.3	3.2	0.003	10.5	<b>0.2</b> <sup>10</sup>

507 <sup>1</sup> The geometric mean values are used as the combined data appear to be log normally distributed and 508 consistent with general practice for environmental activity concentrations.

<sup>2</sup> The ratio (0.1 rounded from 0.13) is relative to the geometric means for farm-raised vs wild-caught *Salmo salar* data (muscle) from the same region (Komperød et al., 2020)).

<sup>3</sup>Heldal 2019, N=7. <sup>4</sup>Smith 2006, N=4. <sup>5</sup>Jalili 2009, N=2.

<sup>6</sup> The ratio reflects that there is no statistical difference between the Kong et al 2018 farm-raised fish (marine
 based feed) vs the global estimate on wild-caught fish data (muscle; GM=2.1 Bq kg<sup>-1</sup>: MARIS 2022).

- <sup>7</sup> Kong 2021, N=3 species summary data based on 41 samples.
- <sup>8</sup> The ratio (0.1 rounded from 0.07) is relative to the geometric means for farm-raised vs wild-caught shrimp
  from the same study (similar species and from the same region (Dong et al., 2018)).
- <sup>9</sup> Dong et al., 2018. N=3 (mariculture), N=5 (wild-caught).

 $^{10} \text{ The ratio (0.2 rounded from 0.21) is the geometric mean of the individual R_{farmed} data from the above categories. }$ 

520

The lowest <sup>210</sup>Po activity concentrations in mariculture fish muscle are for salmon and there may be 521 species-specific influences that contribute to lower <sup>210</sup>Po accumulation (e.g., 12-16 months of 522 523 juvenile growth in fresh water). However, there is a need for more data for other species in order to 524 make any meaningful species-specific comparisons. Much of the available data on non-salmon 525 species comes from a single study with marine-sourced natural feed (Kong 2021). Any species 526 differences in the Table 4 values are more likely as a result of differences in mariculture methods 527 (e.g., feed types) rather than underlying differences between species. Support for this is seen in the 528 data on rainbow trout (Oncorhynchus mykiss), a freshwater species that is sometimes farm-raised in 529 marine waters. This species is distinct from Salmo salar, but when raised in similar marine processed-feed fed operations the reported <sup>210</sup>Po contents were similar to farmed Atlantic salmon 530 531  $(0.020 \pm 0.007 \text{ Bg kg}^{-1})$  (reported by Komperød et al., 2020). The same study attributed differences 532 between mariculture and wild-caught fish as likely due to differing diets (Komperød et al., 2020)

533 When compiling fish and crustacean <sup>210</sup>Po activity concentration data generally (e.g., for use in 534 calculating ingestion doses), the <sup>210</sup>Po content of the edible muscle of any mariculture seafood 535 would ideally be known from measurements and used directly in the dose assessment. Where such 536 data is not available, an alternative approach is to use available wild-caught data, ideally from the 537 same/similar species, and estimate the <sup>210</sup>Po in that portion sourced from mariculture by applying 538 correction factors as follows:

- Where mariculture fish and crustaceans have been raised using processed feed, the
   available data suggest an R<sub>farmed</sub> factor of 0.1 should be applied to wild-caught data of similar
   species.
- Where the feed types used for mariculture fish and crustaceans are varied or not known, the
   available data suggests an R<sub>farmed</sub> factor of 0.2 be applied for the correction of generic wild caught data.
- Where fish or crustaceans are raised using exclusively marine-derived feed, no correction is recommended (R<sub>farmed</sub> = 1.0).
- Where seafood is wild-caught, no correction is needed,  $(R_{farmed} = 1.0)$ .
- The intent of applying these factors in Eqn 1 is to provide a more accurate estimate of the <sup>210</sup>Po content of that portion of seafood sourced from mariculture (e.g., 11% globally).
- 550 *3.3.2 Variation in <sup>210</sup>Po in farmed fish and crustaceans and the need for more data*
- 551 The above correction factors have typical variation when considering the same feed type and species
- (e.g., salmon raised on processed feed GSDs are 2.3-2.7), but have high variation when considering
- data that includes both processed and natural-feed methods. Such mixed data can vary over four
- orders of magnitude (0.003-10.5 Bq kg<sup>-1</sup> range in Table 4) which give rise to elevated GSDs in some
- Table 4 categories. The amount of <sup>210</sup>Po in such processed feeds can vary depending on:
- The percentage of marine-derived ingredients in the feed (e.g., fishmeal and other components that contain marine-derived <sup>210</sup>Po are typically only 15-30% of the feed mass (NOAA, 2022; NOFIMA, 2022).

- Feed is tailored to certain species, and even for the same species, variation in feed could
   occur in processed batches from either the same supplier or different suppliers operating
   across various global regions (Glencross et al., 2020).
- The time delay from when any marine-derived ingredients were extracted from the marine environment until their use as feed. During such delays, the amount of unsupported <sup>210</sup>Po decreases through natural decay (x2.6 decrease with each year of storage).
- Potential processing losses (e.g., leaching or volatilisation when heated).
- The additional amount of <sup>210</sup>Po in the feed mix due to terrestrial sources, including any 566 • mineral additives. Teien (2022) reported uranium-series radionuclides (hence <sup>210</sup>Po progeny) 567 in commercial fish feed for Atlantic salmon (Salmo salar) that are associated with the 568 phosphate compounds added to enhance P nutrient content. Smith (2004) measured 569 substantially elevated <sup>210</sup>Po levels in fish feed for Atlantic salmon (GM=10.3 Bq kg<sup>-1</sup>, GSD=2.8, 570 range of 2.8-25 Bq kg<sup>-1</sup>). However, despite this added source, the <sup>210</sup>Po content remained 571 relatively low in the salmon muscle measured in the same study (GM=0.7 Bg kg<sup>-1</sup>, range of 572 573 0.02 to 0.16).
- 574 Given the potential for large variation of <sup>210</sup>Po in processed feeds, more data is needed to
- 575 refine/improve the summary in Table 4 which have relatively few data in all categories (N=19). In

576 particular, more data are needed on mariculture vs wild-caught crustaceans as a greater proportion

of shrimp/prawn seafood is produced by mariculture than for fish (43% in Europe, EUMOFA, 2020).

578

### 579 4.Conclusions

- 580 Global data through 2022 were used to develop factors to correct for the <sup>210</sup>Po change in seafood 581 during cooking (R<sub>cooking</sub>), the <sup>210</sup>Po loss from radioactive decay during the period between harvest and
- during cooking (R<sub>cooking</sub>), the <sup>210</sup>Po loss from radioactive decay during the period between harvest an
   seafood consumption (R<sub>decay</sub>) and differences in <sup>210</sup>Po content of farmed vs wild-caught fish and
- 583 crustaceans ( $R_{farmed}$ ). The mean retention factor ( $R_{cooking}$ ) of <sup>210</sup>Po in seafood after cooking was 0.74
- with a range of 0.56 to 1.03 depending on the seafood categories and cooking methods used (these
- values do not include the consumption of the stock/broth). For the fraction of <sup>210</sup>Po remaining in
- 586 seafood following radioactive decay during the period from Harvest-to-Consumption (R<sub>decay</sub>), our
- recommended estimate for generic seafood is 0.81 with a range of 0.50 for long-storage seafood
   (e.g., canned) to 0.98 for fresh seafood. These R<sub>decay</sub> values are slightly higher than previously
- 589 published values due to inclusion of <sup>210</sup>Po ingrowth (from <sup>210</sup>Pb) and are consistent with the changes
- 590 in seafood processing and distribution systems in recent times.
- 591 When considering seafood from mariculture, the available data suggest marine fish and crustaceans 592 raised with processed feed have about one order of magnitude lower <sup>210</sup>Po muscle content than wild 593 fish of the same species (R<sub>farmed</sub> of 0.10). However, mariculture practices vary, and fish raised with 594 marine-sourced feed (e.g., small marine fish) have <sup>210</sup>Po content similar to wild-caught fish. When
- 595 feed types vary or are unknown, the limited data available suggest a generic R<sub>farmed</sub> of 0.2 that would
- take into account the variation in mariculture operations and practices around the world.
- 597 The potential losses of <sup>210</sup>Po associated with the cooking, decay and mariculture influences can be
- significant. Overall, the data suggest that, at the time of ingestion, seafood often has only about 55%
- 599 of <sup>210</sup>Po content as compared to when it was harvested (using the means/recommended values of
- 600 R<sub>cooking</sub>= 0.75, R<sub>decay</sub>= 0.81, and R<sub>farmed</sub>= 0.2 for 11% of data; see supplement S.4). Given the
- 601 importance of <sup>210</sup>Po to ingestion dose, if these factors are ignored, any calculated doses for seafood
- 602 ingestion may be substantially overestimated. The data also suggest that lower <sup>210</sup>Po seafood

- 603 ingestion doses will occur for consumers who routinely favour farmed fish, long shelf-life products
- and cooked seafood without stock/broth. Conversely, higher <sup>210</sup>Po seafood doses are implied to
- those who favour fresh, wild-caught and uncooked seafoods.
- 606 This paper utilised global data to provide broad applicability. However, the amount of supporting
- 607 data varied, and more data is required, particularly with regard to the loss/retention of <sup>210</sup>Po when
- 608 cooking molluscs (bivalves, gastropods and cephalopods) and on the <sup>210</sup>Po content of seafood from
- the varied range of world mariculture practices, especially for crustaceans.

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- 618

#### 619 References

620

- Aarkrog, A., Baxter, M.S., Bettencourt, A.O., Bojanowski, R., Bologa, A., Charmasson, S., Cunha, I.,
  Delfanti, R., Duran, E., Holm, E., Jeffree, R., Livingston, H.D., Mahapanyawong, S., Nies, H., Osvath, I.,
  Pingyu, L., Povinec, P.P., Sanchez, A., Smith, J.N., Swift, D., 1997. A comparison of doses from 137Cs
- and 210Po in marine food: A major international study. J. Environ. Radioact. 34, 69-90.
- 625

- some heavy metal residues in crabs and shrimps from the Mediterranean Coast at Damietta region
  with their probabilistic health risk assessment. Journal of Food Composition and Analysis 93, 103606.
- 629

Afchain, A.L., Derens, E., Guilpart, J., Cornu, M., 2005. Modelling of cold-smoked salmon

- temperature profiles for risk assessment of listeria monocytogenes. Acta Hortic. 674, 383-388.
- 632
- Albalat, A., Zacarias, S., Coates, C.J., Neil, D.M., Planellas, S.R., 2022. Welfare in Farmed Decapod
   Crustaceans, With Particular Reference to Penaeus vannamei. Frontiers in Marine Science 9.
- 635

Anderson, J.L., Asche, F., Garlock, T., 2018. Globalization and commoditization: The transformation
of the seafood market. Journal of Commodity Markets 12, 2-8.

638

- Aoyama, M., Kajino, M., Tanaka, T.Y., Sekiyama, T.T., Tsumune, D., Tsubono, T., Hamajima, Y.,
  Inomata, Y., Gamo, T., 2016. 134Cs and 137Cs in the North Pacific Ocean derived from the March
  2011 TEPCO Fukushima Dai-ichi Nuclear Power Plant accident, Japan. Part two: estimation of 134Cs
- and 137Cs inventories in the North Pacific Ocean. Journal of Oceanography 72, 67-76.
- 643
- Baptista, R.C., Rodrigues, H., Sant'Ana, A.S., 2020. Consumption, knowledge, and food safety
   practices of Brazilian seafood consumers. Food Research International 132, 109084.

646

Bassey, F.I., Oguntunde, F.C., Iwegbue, C.M., Osabor, V.N., Edem, C.A., 2014. Effects of processing on
the proximate and metal contents in three fish species from Nigerian coastal waters. Food Sci Nutr 2,
272-281.

Abd-Elghany, S.M., Zaher, H.A., Elgazzar, M.M., Sallam, K.I., 2020. Effect of boiling and grilling on

- Bibi, F., Guillaume, C., Gontard, N., Sorli, B., 2017. A review: RFID technology having sensing
- aptitudes for food industry and their contribution to tracking and monitoring of food products.
- Trends in Food Science & Technology 62, 91-103.
- 654
- 655 Bienkiewicz, G., Tokarczyk, G., Biernacka, P., 2022. Influence of Storage Time and Method of 656 Smoking on the Content of EPA and DHA Acids and Lipid Quality of Atlantic Salmon (Salmo salar)
- 657 Meat. Int J Food Sci 2022, 1218347.
- 658
- Boerder, K., Miller, N.A., Worm, B., 2018. Global hot spots of transshipment of fish catch at sea.Science Advances 4, eaat7159.
- 661
- 662 Cai, J., Leung, P.S., 2017. Short-term projection of global fish demand and supply gaps. FAO, Rome.
- 663
- 664 Carvalho, F.P., 2011. Polonium (Po-210) and lead (Pb-210) in marine organisms and their transfer in 665 marine food chains. J. Environ. Radioact. 102, 462-472.

667 Carvalho, F.P., 2018. Radionuclide concentration processes in marine organisms: A comprehensive 668 review. J Environ Radioact 186, 124-130.

669

- 670 Chardon, J., Swart, A., 2016. Food Consumption and Handling Survey for Quantitative
- 671 Microbiological Consumer Phase Risk Assessments. J Food Prot 79, 1221-1233.

672

- 673 Costello, C., Cao, L., Gelcich, S., Cisneros-Mata, M.Á., Free, C.M., Froehlich, H.E., Golden, C.D.,
- Ishimura, G., Maier, J., Macadam-Somer, I., Mangin, T., Melnychuk, M.C., Miyahara, M., de Moor,
- 675 C.L., Naylor, R., Nøstbakken, L., Ojea, E., O'Reilly, E., Parma, A.M., Plantinga, A.J., Thilsted, S.H.,
- Lubchenco, J., 2020. The future of food from the sea. Nature 588, 95-100.

677

Daelman, J., Jacxsens, L., Membré, J.-M., Sas, B., Devlieghere, F., Uyttendaele, M., 2013. Behaviour
of Belgian consumers, related to the consumption, storage and preparation of cooked chilled foods.
Food Control 34, 681-690.

681

682 Delacroix, D., P. Guerre, J., Leblanc, P., Hickman, C., 2002. RADIONUCLIDE AND RADIATION
683 PROTECTION DATA HANDBOOK 2002. Radiat. Prot. Dosim. 98, 1-168.

684

Díaz-Francés, I., Díaz-Ruiz, J., Manjón, G., García-Tenorio, R., 2017. 210Po Activity Concentrations in
Cooked Marine Food. Journal of FisheriesSciences.com.

687

Dong, X., Chen, L., Pan, J., Wang, J., Zhou, S., Cao, Z., Pan, Z., 2018. 210Po level in five kinds of typical
aquatic products from the Yello Sea of China. J. of Nuclear and Radiochemistry (China).

690

- 691 Erkan, N., Üretener, G., Alpas, H., Selçuk, A., Özden, Ö., Buzrul, S., 2011. The effect of different high
- 692 pressure conditions on the quality and shelf life of cold smoked fish. Innovative Food Science &693 Emerging Technologies 12, 104-110.

695 696 697	EUMOFA, 2020. The EU Fish Market, 2020 Edition. European Union, Luxembourg Forrester, J., 1962. Effects of three cooking methods on cesium-134 content of beef from orally dosed steers. Food Technol. 16, 110-112.
698 699 700	Foscarini, R., Prakash, J., 1990. Handbook on Eucheuma seaweed cultivation in Fiji. Food and Agriculture Organization of the United Nations.
701 702 703	Fowler, S.W., 2011. 210Po in the marine environment with emphasis on its behaviour within the biosphere. J Environ Radioact 102, 448-461.
704 705 706 707	Glencross, B.D., Baily, J., Berntssen, M.H.G., Hardy, R., MacKenzie, S., Tocher, D.R., 2020. Risk assessment of the use of alternative animal and plant raw material resources in aquaculture feeds. Reviews in Aquaculture 12, 703-758.
708 709 710	Guillen, J., Natale, F., Carvalho, N., Casey, J., Hofherr, J., Druon, JN., Fiore, G., Gibin, M., Zanzi, A., Martinsohn, J.T., 2019. Global seafood consumption footprint. Ambio 48, 111-122.
711 712 713	Hallström, E., Bergman, K., Mifflin, K., Parker, R., Tyedmers, P., Troell, M., Ziegler, F., 2019. Combined climate and nutritional performance of seafoods. Journal of Cleaner Production 230, 402-411.
714 715 716 717	Heldal, H.E., Volynkin, A., Komperød, M., Hannisdal, R., Skjerdal, H., Rudjord, A.L., 2019. Natural and anthropogenic radionuclides in Norwegian farmed Atlantic salmon (Salmo salar). J Environ Radioact 205-206, 42-47.
718 719	Hicks, D.T., 2016. Seafood Safety and Quality: The Consumer's Role. Foods 5, 71.
720 721 722 723	IAEA, 1995. Sources of Radioactivity in the Marine Environment and their Relative Contributions to Overall dose Assessment from Marine Radioactivity (MARDOS). INTERNATIONAL ATOMIC ENERGY AGENCY, Vienna.
724 725 726	IAEA, 2009. Quantification of Radionuclide Transfer in Terrestrial and Freshwater Environments for Radiological Assessments. IAEA, Vienna Austria.
727 728 729	IAEA, 2017. The environmental behaviour of polonium. International Atomic Energy Agency, Vienna, Austria.
730 731 732	IAEA, 2021. Exposure due to Radionuclides in Food Other Than During a Nuclear or Radiological Emergency. IAEA, Vienna, Austria.
733 734 735	IAEA, 2022. IAEA Marine Radioactivity Information System (MARIS), IAEA Environmental Laboratories, Monaco. <u>https://maris.iaea.org/home</u> .
736 737 738	ICRP, 2006. Assessing Dose of the Representative Person for the Purpose of the Radiation Protection of the Public. International Commission on Radiological Protection.

740 ICRP, 2012. Compendium of dose coefficients based on ICRP Publication 60. International Commision 741 on Radiologial Protection Publication 119. Ann. ICRP 41 (Suppl.). 742 743 Immaculate, K., Sinduja, P., Velammal, A., Patterson, J., 2013. Quality and shelf life status of salted 744 and sun dried fishes of Tuticorin fishing villages in different seasons. international food research 745 journal 20, 1855-1863. 746 747 Jalili, A., López-Pérez, M., Karlsson, L., Hernández, F., Rubio, C., Hernández-Armas, J., Hardisson, A., 748 2009. Radiometric analysis of farmed fish (sea bass, gilthead bream, and rainbow trout) from 749 Tenerife Island, Spain. J Food Prot 72, 1941-1947. 750 751 Jeffree, R.A., Carvalho, F., Fowler, S.W., Farber-Lorda, J., 1997. Mechanism for Enhanced Uptake of 752 Radionuclides by Zooplankton in French Polynesian Oligotrophic Waters. Environ. Sci. Technol. 31, 753 2584-2588. 754 755 Johansen, M.P., Ruedig, E., Tagami, K., Uchida, S., Higley, K., Beresford, N.A., 2015. Radiological Dose 756 Rates to Marine Fish from the Fukushima Daiichi Accident: The First Three Years Across the North 757 Pacific. Environ. Sci. Technol. 49, 1277-1285. 758 759 Jones, A.L., Sherwood, J.C., 2009. Delay times between harvesting or collection of food products and 760 consumption for use in radiological assessments. Journal of Radiological Protection 29, 377. 761 762 Komperød, M., García, F.P., Guðnason, K., Jensen, L.K., Kämäräinen, M., Roos, P., Skjerdal, H.K., 763 2020. Natural Radioactivity in Nordic Fish and Shellfish – Final report. NKS-435. NKS, Nordic Nuclear 764 Safety Research, Roskilde, Denmark 765 Kondjoyan, A., Oillic, S., Portanguen, S., Gros, J.-B., 2013. Combined heat transfer and kinetic models 766 to predict cooking loss during heat treatment of beef meat. Meat Science 95, 336-344. 767 768 Kong, X., Qian, Y., Zheng, Q., Ji, Y., 2021. Levels and Distributions of (210)Pb and (210)Po in Selected 769 Seafood Samples in China and Assessment of Related Dose to Population. Int J Environ Res Public 770 Health 18. 771 772 Kontominas, M.G., Badeka, A.V., Kosma, I.S., Nathanailides, C.I., 2021. Recent Developments in 773 Seafood Packaging Technologies. Foods 10. 774 775 Kristan, U., Planinšek, P., Benedik, L., Falnoga, I., Stibilj, V., 2015. Polonium-210 and selenium in 776 tissues and tissue extracts of the mussel Mytilus galloprovincialis (Gulf of Trieste). Chemosphere 119, 777 231-241. 778 779 Limbo, S., Sinelli, N., Torri, L., Riva, M., 2009. Freshness decay and shelf life predictive modelling of 780 European sea bass (Dicentrarchus labrax) applying chemical methods and electronic nose. LWT -781 Food Science and Technology 42, 977-984.

782

783 784	McManus, A., Hunt, W., Storey, J., McManus, J., Hilhorst, S., 2014. Perceptions and preference for fresh seafood in an Australian context. International Journal of Consumer Studies 38, 146-152.
785 786 787	Medley, P., Patterson, S., 2022. Determining baseline radiation levels in marine biota – A comparison of SE Queensland commercial species. Journal of Environmental Radioactivity 255, 107032.
788 789 790	Nabeshi, H., Tsutsumi, T., Hachisuka, A., Matsuda, R., 2013. [Reduction of radioactive cesium content in pond smelt by cooking]. Shokuhin Eiseigaku Zasshi 54, 303-308.
791 792 793 794	Naylor, R.L., Hardy, R.W., Buschmann, A.H., Bush, S.R., Cao, L., Klinger, D.H., Little, D.C., Lubchenco, J., Shumway, S.E., Troell, M., 2021. A 20-year retrospective review of global aquaculture. Nature 591, 551-563.
795 796 797 798	Newsome, R., Balestrini, C.G., Baum, M.D., Corby, J., Fisher, W., Goodburn, K., Labuza, T.P., Prince, G., Thesmar, H.S., Yiannas, F., 2014. Applications and Perceptions of Date Labeling of Food. Compr Rev Food Sci Food Saf 13, 745-769.
799 800	NOAA, 2021. Fisheries of the United States 2019. NOAA, Silver Spring, MD, USA.
801 802	NOAA, 2022. Feeds for Aquaculture. National Oceanic and Atmospheric Administration.
803 804 805	NOFIMA, 2022. Utilization of feed resources in Norwegian farming of Atlantic salmon and rainbow trout in 2020. NOFIMA, Norway.
806 807 808	Obluchinskaya, E., Daurtseva, A., 2020. Effects of air drying and freezing and long-term storage on phytochemical composition of brown seaweeds. Journal of Applied Phycology 32, 4235-4249.
809 810 811	Østli, J., Esaiassen, M., Garitta, L., Nøstvold, B., Hough, G., 2013. How fresh is fresh? Perceptions and experience when buying and consuming fresh cod fillets. Food Quality and Preference 27, 26-34.
812 813 814	Pouillot, R., Lubran, M.B., Cates, S.C., Dennis, S., 2010. Estimating parametric distributions of storage time and temperature of ready-to-eat foods for U.S. households. J Food Prot 73, 312-321.
815 816 817 818	Pouillot, R., Miconnet, N., Afchain, A.L., Delignette-Muller, M.L., Beaufort, A., Rosso, L., Denis, J.B., Cornu, M., 2007. Quantitative risk assessment of Listeria monocytogenes in French cold-smoked salmon: I. Quantitative exposure assessment. Risk Anal 27, 683-700.
819 820 821 822	Roccato, A., Uyttendaele, M., Membré, JM., 2017. Analysis of domestic refrigerator temperatures and home storage time distributions for shelf-life studies and food safety risk assessment. Food Research International 96, 171-181.
823 824 825	Roselli, C., Meli, M.A., Fagiolino, I., Desideri, D., 2020. Bioaccessibility assessment of stable elements and 210Po in food. PLoS One 15, e0236871.

826 827 828	RWMC, 1994. Removal of Radionuclides during Food Processing and Culinary Preparation. Radioactive Waste Management Center, Japan, Japan.
829 830 831 832	Simpson, R., Almonacid, S., Nunez, H., Pinto, M., Abakarov, A., Teixeira, A., 2012. TIME- TEMPERATURE INDICATOR TO MONITOR COLD CHAIN DISTRIBUTION OF FRESH SALMON (SALMO SALAR). Journal of Food Process Engineering 35, 742-750.
833 834	Skretting, 2022. What ingredients are in Skretting feeds? . Skretting, a Nutreco Company.
835 836 837	Smaal, A.C., Ferreira, J.G., Grant, J., Petersen, J.K., Strand, Ø., 2018. Goods and Services of Marine Bivalves, Cambridge International Law Journal.
838 839	Smith, B., 2004. The Radiological impact of farmed fish. CEFAS, UK.
840 841	Smith, B., 2006. The Radiological Impact of farmed fish in the UK. CEFAS.
842 843 844	Štrok, M., Smodiš, B., 2011. Levels of 210Po and 210Pb in fish and molluscs in Slovenia and the related dose assessment to the population. Chemosphere 82, 970-976.
845 846 847	Sun, L., Engle, C., Kumar, G., van Senten, J., 2022. Retail market trends for seafood in the United States. Journal of the World Aquaculture Society n/a.
848 849 850 851	Takata, H., Johansen, M.P., Kusakabe, M., Ikenoue, T., Yokota, M., Takaku, H., 2019. A 30-year record reveals re-equilibration rates of 137Cs in marine biota after the Fukushima Dai-ichi nuclear power plant accident: Concentration ratios in pre- and post-event conditions. Sci. Total Environ.
852 853 854 855	Tateda, Y., Tsumune, D., Tsubono, T., Aono, T., Kanda, J., Ishimaru, T., 2015. Radiocesium biokinetics in olive flounder inhabiting the Fukushima accident-affected Pacific coastal waters of eastern Japan. J Environ Radioact 147, 130-141.
856 857 858	Teien, HC., 2022. Uranium in fish feed and transfer to farmed Atlantic salmon (Salmo salar L.), International Conference on Radioecology & Environmental Radioactivity, 2022, Oslo, Norway.
859 860 861	Tingman, W., 2010. Fish product quality evaluation based on temperature monitoring in cold chain. African Journal of Biotechnology 9, 6146-6151.
862 863 864 865	Tsironi, T., Giannoglou, M., Platakou, E., Taoukis, P., 2016. Evaluation of Time Temperature Integrators for shelf-life monitoring of frozen seafood under real cold chain conditions. Food Packaging and Shelf Life 10, 46-53.
866 867 868	Tyworth, J.E., O'Neill, L., 1997. Robustness of the normal approximation of lead-time demand in a distribution setting. Naval Research Logistics (NRL) 44, 165-186.

- Uddin, S., Behbehani, M., Fowler, S.W., Al-Ghadban, A., Dupont, S., 2019. Assessment of loss of
- 210Po from fish and shrimp by cooking and its effect on dose estimates to humans ingesting
- 872 seafood. Journal of Environmental Radioactivity 205-206, 1-6.

873

UNSCEAR, 2008. Sources, Effects and Risk of Ionizing Radiation. . United Nations, New York.

875

USDA, 2022. Food Safety, <u>https://www.fsis.usda.gov/food-safety/safe-food-handling-and-</u>
 preparation/food-safety-basics/shelf-stable-food. USDA.

878

Whicker, F.W., 1982. Radioecology : nuclear energy and the environment, in: Schultz, V. (Ed.). CRC
Press, Boca Raton, Fla. :.

- Xiao, X., He, Q., Fu, Z., Xu, M., Zhang, X., 2016. Applying CS and WSN methods for improving
- efficiency of frozen and chilled aquatic products monitoring system in cold chain logistics. FoodControl 60, 656-666.
- 885
- 886
- 887
- 888
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#### 890 Supplemental

- 891 S.1 <sup>210</sup>Po change during cooking
- 892 S.2 <sup>210</sup>Po loss from radioactive decay during Harvest-to-Consumption
- 893 S.3 <sup>210</sup>Po differences in farm-raised vs wild-caught marine fish
- 894 S.4 Example of typical <sup>210</sup>Po reduction using the recommended correction factors
- 895

### 896 S.1 <sup>210</sup>Po change during cooking

The change in  $^{210}$ Po content during cooking is determined by comparing the measured Bq kg<sup>-1</sup> of 897 cooked samples with that of the corresponding pre-cooked raw samples and can be expressed as a 898 899 correction factor (R<sub>cooking</sub>). While cooking effects on the radionuclide content of foods were identified early in the nuclear age (Forrester, 1962; Whicker, 1982), most of the usable data on seafood have 900 emerged from research published in recent decades. Data were gathered by searching global 901 902 sources (1992 to 2022) focused on scientific journals, science conference proceedings, industry reports, government documents and websites. In gathering data on <sup>210</sup>Po change from cooking, 903 904 various search terms were used: seafood (marine fish, crustacean, bivalve, gastropod, cephalopod, 905 seaweed), cooked seafood, heat leaching from tissues, radionuclide loss from cooking, radionuclides, dose, radioactive polonium, <sup>210</sup>Po. Studies were assessed for guality assurance of methods. Data 906 were grouped by seafood and cooking categories appropriate for dose assessment (details in section 907 908 3.1.1 and S.1). Some sources reported directly on retention (or loss) of radionuclides from cooking, while other sources reported raw vs cooked activity concentrations from which retention values 909 910 were calculated.

912	Table S1.1. Fractions of <sup>210</sup> Po activity concentration retained after cooking (relative to the raw
913	tissues) including summarised source data.

0.74	0.21	0.70						
		5.70	1.2	0.76	0.19	1.24	63	This study
0.75	0.21	0.72	1.4	0.79	0.19	1.24	42	This study
0.68	0.20	0.65	1.3	0.69	0.35	1.06	16	This study
0.75	0.26	0.71	1.41	0.62	0.49	1.03	5	This study
0.78	0.21	0.74	1.4	0.78	0.19	1.12	34	This study
0.76	0.21	0.72	1.4	0.79	0.19	1.13	25	This study
0.74	0.14	0.73	1.2	0.78	0.45	0.94	16	Uddin 2019
0.73	0.36	0.62	2.0	0.80	0.19	1.13	7	Díaz-Francés 2017
0.93	0.05				0.89	0.96	2	Strok 2011
	0.75 0.68 0.75 <b>0.78</b> <b>0.76</b> 0.74 0.73 0.93	0.75 0.21 0.68 0.20 0.75 0.26 0.78 0.21 0.78 0.21 0.74 0.14 0.73 0.36 0.93 0.05	0.75         0.21         0.72           0.68         0.20         0.65           0.75         0.26         0.71           0.78         0.21         0.74           0.76         0.21         0.72           0.74         0.14         0.73           0.73         0.36         0.62           0.93         0.05	0.75       0.21       0.72       1.4         0.68       0.20       0.65       1.3         0.75       0.26       0.71       1.41         0.78       0.21       0.74       1.4         0.76       0.21       0.72       1.4         0.74       0.14       0.73       1.2         0.73       0.36       0.62       2.0         0.93       0.05       0.55	0.75       0.21       0.72       1.4       0.79         0.68       0.20       0.65       1.3       0.69         0.75       0.26       0.71       1.41       0.62         0.75       0.26       0.71       1.41       0.62         0.78       0.21       0.74       1.4       0.78         0.76       0.21       0.72       1.4       0.79         0.74       0.14       0.73       1.2       0.78         0.73       0.36       0.62       2.0       0.80         0.93       0.05       0.05       0.05       0.05	0.75       0.21       0.72       1.4       0.79       0.19         0.68       0.20       0.65       1.3       0.69       0.35         0.75       0.26       0.71       1.41       0.62       0.49         0.78       0.21       0.74       1.4       0.78       0.19         0.76       0.21       0.72       1.4       0.79       0.19         0.74       0.14       0.73       1.2       0.78       0.45         0.73       0.36       0.62       2.0       0.80       0.19         0.93       0.05       0.89       0.89       0.89	0.75       0.21       0.72       1.4       0.79       0.19       1.24         0.68       0.20       0.65       1.3       0.69       0.35       1.06         0.75       0.26       0.71       1.41       0.62       0.49       1.03         0.78       0.21       0.74       1.4       0.78       0.19       1.12         0.76       0.21       0.72       1.4       0.79       0.19       1.13         0.74       0.14       0.73       1.2       0.78       0.45       0.94         0.73       0.36       0.62       2.0       0.80       0.19       1.13         0.93       0.05       0.89       0.96	0.75       0.21       0.72       1.4       0.79       0.19       1.24       42         0.68       0.20       0.65       1.3       0.69       0.35       1.06       16         0.75       0.26       0.71       1.41       0.62       0.49       1.03       5         0.78       0.21       0.74       1.4       0.78       0.19       1.12       34         0.76       0.21       0.72       1.4       0.79       0.19       1.13       25         0.74       0.14       0.73       1.2       0.78       0.45       0.94       16         0.73       0.36       0.62       2.0       0.80       0.19       1.13       7         0.93       0.05       0.89       0.96       2

Crustacean	0.81	0.12	0.80	1.2	0.78	0.75	1.06	7	This study
Shrimp (various)	0.78	0.03	0.78	1.0	0.78	0.75	0.82	5	Uddin 2019
Prawn (whole)	0.88	0.25				0.70	1.06	2	Medley 2022 <sup>1</sup>
Bivalve	1.03	0.01				1.02	1.03	2	Krystan 2015
Boiled, Steamed									
All boiled/steamed seafood	0.68	0.20	0.66	1.3	0.69	0.38	1.27	29	This study
Eich	0.75	0.21	0.66	1.4	0.66	0.38	1.24	17	This study
F1511	0.72	0.15	0.00	12	0.00	0.30	0.02	16	- Uddin 2010
	1.24	0.13	0.71	1.5	0.75	0.56	0.92	10	
	1.24							T	Rosenii 2020
Crustacean	0.58	0.20	0.55	1.4	0.54	0.35	1.24	9	This study
Shrimp (various)	0.50	0.08	0.49	1.2	0.46	0.41	0.62	5	 Uddin 2019
Prawn (whole)	1.04					0.82	1.03	2	Medley 2022 <sup>1</sup>
Mantis shrimp (meat)	0.66							1	Roselli 2020
Small shrimp	0.35							1	RWMC 1994
Bivalve	0.56	0.07	0.56	1.1	0.57	0.49	0.62	3	This study
	0.55	0.09				0.49	0.62	2	Díaz-Francés 2017
	0.57							1	RWMC 1994

- 917
- 918

S.2 <sup>210</sup>Po loss from radioactive decay during Harvest-to-Consumption 919

920 Within each seafood production category, there exists a chain of events (e.g., harvest, transport, 921 processing, transport to market, market storage, home storage consumption). In this study, the 922 duration data for these events are combined into two groups termed here "Harvest-to-Market" and "Market-to-Consumption". This is necessary because Harvest-to-Market events are dominated by 923 924 industrial-type production processes while the Market-to-Consumption events are dominated by 925 retail and consumer behaviours. These two groups of events are therefore expected to have 926 differing data sources, data ranges and potentially differing data distributions. The following process 927 was used to estimate delay durations:

- 928 Seafood categories of fresh, frozen, canned, dried and smoked are considered separately 929 given their potentially different storage delay times. These simplified categories serve the 930 purpose for use in typical dose assessments. More detailed categorisation is not currently 931 justified given that most dose assessments do not have access to more detailed data on 932 receptor preferences.
- 933 For each category, duration data are gathered and grouped into the two types of Harvest-to-• 934 Market and Market-to-Consumption. In gathering duration data, various combinations of 935 the following search terms were used: seafood, fresh seafood, marine fish, 936 chilled/refrigerated, smoked, frozen, canned, retort pouch, dried, cured, "use by" date, shelf 937 life, storage time, radionuclides, dose, radioactive polonium, 210Po.
- 938 The Harvest-to-Market duration ranges (from harvest until arrival at market) are typically • 939 sourced from industry data (e.g., harvest, production and process studies), science 940 publications (e.g., minimum and maximum observed data for various seafoods).
- 941 The Market-to-Consumption duration ranges (from arrival at market until consumption. 942 Includes warehouse, display/shelf time, transport, home storage) are typically sourced from 943 industry data, science publications and government reports.
- 944 Within each of the above ranges, data distributions are applied that are consistent with observed data and reference information. Distributions may vary depending on the 945 946 category. See Table S2.1 and text on distributions for specific seafoods.
- 947 The overall delay estimates for each seafood category are calculated by combining the 948 Harvest-to-Market and Market-to-Consumption distributions using Monte Carlo analysis 949 (used here via Goldsim<sup>™</sup> software). The Monte Carlo methods used common parameters (e.g., latin hypercube sampling over 10,000 realisations) and, where appropriate, ustilised 950 951 distributions that were truncated to the minimum and maximum data (Table S2.1).

952 Applying the above procedure to fresh seafood, using the available data up through 2022, the range 953 of Harvest-to-Market durations is typically 0-6 days (Table S2.1). Within this range, it is assumed the 954 durations can be approximated by a normal distribution (mean=3.0 days), consistent with typical 955 production-transport supply chains (e.g., Tyworth and O'Neill, 1997). After the fresh seafood is 956 purchased, data reflect a preference for consumption with 1-3 days, but some durations ranged up 957 to 12 days (Baptista et al., 2020; McManus et al., 2014; Østli et al., 2013; Roccato et al., 2017; 958 Simpson et al., 2012). These durations are consistent with the desire of producers to sell fresh 959 products quickly and the preferences of consumers to for seafood products that are fresh, appealing

- and considered to be safe (Newsome et al., 2014). The data on Market-to-Consumption durations of
  fresh seafood (from multiple studies) were best fit by an exponential distribution (Roccato et al.,
  2017). This same study reported that the mean storage time was approximated by dividing the useby date by four (Roccato et al., 2017). The two separate data distributions, Harvest-to-Market
  (normal) and Market-to-Consumption (exponential), were then combined using Monte Carlo
- 965 methods to arrive at an overall distribution of Harvest-to-Consumption durations (total delay966 estimates, Table S2.1).

967 Applying the above procedure to packaged seafood (frozen, , frozen, canned, dried and smoked), the 968 typical Harvest-to-Market duration ranges are provided in Table S2.1 based on reported data 969 (Bienkiewicz et al., 2022; Boerder et al., 2018; Immaculate et al., 2013); Kontominas et al. (2021); 970 (Limbo et al., 2009; Obluchinskaya and Daurtseva, 2020; Østli et al., 2013; Roccato et al., 2017; 971 Tingman, 2010; Tsironi et al., 2016; USDA, 2022; Xiao et al., 2016). The distributions within these 972 Harvest-to-Market ranges were approximated using normal distributions (e.g., Tyworth and O'Neill, 973 1997). For the Market-to-Consumption period, the durations ranged up to more than 5 years for 974 canned seafood in reference publications (Boerder et al., 2018; Kontominas et al., 2021; Tsironi et 975 al., 2016). Within the Market-to-Consumption ranges for seafood packaged/preserved for longer 976 shelf lives (frozen, , frozen, canned, dried), the reported quantitative reports and qualitative 977 observations suggest the data are best represented by lognormal distributions. Compared with fresh 978 seafood, there is less urgency for quick sale of the packaged/preserved products. However, retailers 979 tend to avoid long storage times and, after purchase, much of the packaged seafood is consumed in 980 a timely manner as many consumers are aware of the potential for degradation of quality and tastes 981 over time and also have a perception of increased risk with delayed usage (Newsome et al., 2014). A 982 small percentage of products may have long shelf storage delays before use consistent with a 983 lognormal distribution. As with fresh seafood, the overall estimated distribution of Harvest-to-984 Consumption durations for packaged/preserved seafood were derived using Monte Carlo (Goldsim™ 985 software; Ref) to combine the Harvest-to-Market (normal distributions) and Market-to-Consumption 986 (log-normal distributions) estimates. Total delay durations were rounded to the nearest day as more 987 precision is not warranted by the scarce data.

988 For smoked seafood, the Market-to-Consumption durations were, similar to fresh seafood,

- approximated using an exponential distribution consistent with reference publications (Afchain et
- al., 2005; Chardon and Swart, 2016; Pouillot et al., 2010) and using duration data from (Afchain et
- al., 2005; Chardon and Swart, 2016; Daelman et al., 2013; Pouillot et al., 2010; Roccato et al., 2017)

993 Table S2.1. Recommended R<sub>decay</sub>, correction factors (fraction <sup>210</sup>Po retained) derived from <sup>210</sup>Po delay

- 994 times between harvest and consumption for various seafood processing/preservation types (2022
- 995 estimates).

		Market-to-	Total delay Harvest-	B	
	Harvest-to-Market <sup>1</sup>	Consumption <sup>2</sup>	to-Consumption	correction factors	
	Distribution mean (range) (days)	Distribution mean (range) (days)	50 <sup>th</sup> (range) (days)	Recommended value (range)	
Fresh	Normal 3(0-6) <sup>1,2</sup>	Expon. 3(0-12) <sup>1-5</sup>	5 (0-17)	0.98 (0.93-1.0)	
Frozen	Normal 65.5(1-130) <sup>6-9</sup>	LogNorm(t) 20.2(0-410) <sup>6-10</sup>	89 (1-540)	0.70 (0.21-1.0)	
Canned	Normal 127.5(10-245) <sup>9,10</sup>	LogNorm(t) 42.7(0-1825) <sup>10,11</sup>	177 (10-2070)	0.50 (0.14-0.96)	
Dried	Normal 30.5(10-51) <sup>12-14</sup>	LogNorm(t) 19.1(0-365) <sup>12-15</sup>	51 (10-410)	0.81 (0.26-0.96)	
Smoked	Normal 5.5(2-9) <sup>16</sup>	Expon. 11(0-53) <sup>6,17-22</sup>	13 (2-62)	0.95 (0.78-0.99)	
All seafood (weighted)				0.81 (0.14-0.1.0)	

<sup>1</sup> Harvest-to-market includes all transport, processing and storage times from the time of removal from the
 ocean until arrival at retail marketplace.

998 <sup>2</sup> Market-to-Consumption includes all warehouse, display and consumer storage times from the time of arrival
 999 at the market until consumption.

### 1000 Table S2.2. Expanded reference descriptions for Table S2.1.

1	(USDA, 2022). US Department of Agriculture (https://www.fsis.usda.gov/food-safety/safe-food- handling-and-preparation/food-safety-basics/shelf-stable-food)
2	(Østli et al., 2013). How fresh is fresh? Perceptions and experience when buying and consuming fresh cod fillets (Norway). Summarizing, when elder respondents were asked which was the last day they would be willing to buy cod after capture, there was a 75% probability that this would be approximately 3 days(Fig. 2); for young respondents this time was approximately 5 days. There was a 75% probability that these consumers would have the fish approximately 1 day at home before cooking and eating it. Using a rejection probability of 25% results in a shelf life of approximately 11 days when no capture date information was given.
3	(Simpson et al., 2012). Time-temperature indicator to monitor cold chain distribution of fresh salmon (Salmo Salar). Fresh salmon (farmed) were shipped from Chile to the US in 65 hours total time (truck transport, air flight, final transport) in a TTI study simulating typical durations for fresh fish Harvest-to-Market.
4	(McManus et al., 2014). Perceptions and preference for fresh seafood in an Australian context. 91.3% of respondents reported storing thawed seafood in the refrigerator for 3 days or less with 2 weeks being the maximum storage period reported." (Australia). 57.8% of respondents stating that 'fresh' meant that the seafood or fish displayed in retail outlets was caught the same day as displayed for sale. Four percent (n = 7) thought 'fresh' meant caught within a few days.
5	(Baptista et al., 2020). Consumption, knowledge, and food safety practices of Brazilian seafood.
6	(Roccato et al., 2017). Analysis of domestic refrigerator temperatures and home storage time distributions for shelf-life studies and food safety risk assessment. The storage time was described by an exponential distribution corresponding to the use-by date period divided by 4.
7	(Tingman, 2010). Fish product quality evaluation based on temperature monitoring in cold chain Maximum recommended total time from Harvest-to-Consumption of 330 days (shorter durations if temperatures not maintained throughout cold chain).
8	(Xiao et al., 2016). Typical frozen cold-chain from Harvest-to-Market for farmed fish in China was

	approximately 22 days total (Harvest-to processing 1.4 d, processing 1.3 d, transport to market 15 d, market 4.3 d).
9	(Limbo et al., 2009). Freshness decay and shelf life predictive modelling of European sea bass ( <i>Dicentrarchus labrax</i> ) applying chemical methods and electronic nose. Maximum recommended total time from Harvest-to-Consumption of 393 days.
10	(Kontominas et al., 2021). Recent Developments in Seafood Packaging Technologies.
11	(Tsironi et al., 2016). Evaluation of Time Temperature Integrators for shelf-life monitoring of frozen seafood under real cold chain conditions (i.e. production warehouse, distribution centre, retail and domestic storage).
12	(Boerder et al., 2018). "It thus takes about half a year on average (18 to 35 weeks) from the Harvest of albacore tuna to the canned final product on the shelf. Along the entire supply chain, the fish have travelled an average 17,000 km"
13	(Immaculate et al., 2013). Quality and shelf life status of salted and sun dried fishes of fishing villages in different seasons dried fish is that if properly stored, the product can maintain quality for up to 3-6 months
14	(Obluchinskaya and Daurtseva, 2020). Effects of air drying and freezing and long-term storage on phytochemical composition of brown seaweeds. In the course of the experiment on long-term storage of samples of these algae, the similarity in phytochemical composition in dry and frozen states remained up to 365 days of storage.
15	(Foscarini and Prakash, 1990), Various sources (seaveg.com) Storage times vary depending on residual moisture content, treatments and storage conditions. In one example (FAO 199)) "Well dried seaweed with salt crystals on its surface can be stored for a long time; up to 2 years without getting spoiled. The salt covering the seaweed prevents the spoilage of the carrageenan. Of course it is important that seaweed is stored properly." However, many products will not be salt encrusted. For most commercial products, we estimated a practical shelf life limit of about one year.
16	(Bienkiewicz et al., 2022) The available literature contains a lot of information on microbiological safety or freshness level indicators, such as TVB-N. This experiment showed that the 28-day storage period of the hot and cold smoked product did not result in exceeding the TVB-N limits. However, in the case of raw salmon, on day 21 of storage, a TVB-N content limit was found.
17	(Afchain et al., 2005) Proposes a statistical approach that utilises an approximate 12-day total storage time (retail + consumer) for smoked salmon.
18	(Chardon and Swart, 2016) Consumer survey data. The smoked fish consumer storage data were represented by and exponential distribution with a mean of 3.29 days (home refrigerated storage).
19	(Daelman et al., 2013) One fifth (19.8%) of the REPFEDs (chilled goods including smoked fish) were consumed on the day of purchase, little over half of the products (52.9%) were estimated as consumed within the 2 days after purchase and 93.7% was estimated as consumed within the first week after purchase.
20	(Pouillot et al., 2007) Utilises/updates estimates in Afchain et al., 2005. Proposes a statistical approach that utilises an approximate 12-day total storage time (retail + consumer) for smoked fish. Proposes exponential distributions for various components of the retail and consumer storage times.
21	(Pouillot et al., 2010) Proposed a Fitted distribution of time to first consumption of smoked seafood, exponential with mean of 4 days.
22	(Erkan et al., 2011) Based on the sensory and microbiological results, the control samples were acceptable only up to 6 weeks, compared to 8 weeks in HP treatment cold smoked salmon samples, extending the shelf-life by 2 weeks.

Table S2.3. Results from this study (bolded) compared with previously published estimates on <sup>210</sup>Po 1003 1004 delay times between harvest and consumption for various seafoods.

		Total duration		
		Harvest-to-	R <sub>decay;</sub>	
		Consumption	correction	
		(days)	factors <sup>1</sup>	Reference
Fresh				
	Seafood	5 (0-17)	<b>0.98 (0.93-1.0)</b> <sup>1</sup>	This study (Table S3)
	Seafood	3	0.99	Aarkrog et al. (1997)
	Wild-caught fish	8 (2-12 )	0.96	Jones and Sherwood (2009)
	Farm-raised fish	2 (1-5 )	0.99	Jones and Sherwood (2009)
	Bivalve/crustacean	7 (1-9 )	0.97	Jones and Sherwood (2009)
Frozen				
	Seafood	89 (1-540)	<b>0.70 (0.21-1.0)</b> <sup>1</sup>	This study (Table S3)
	Seafood	60	0.74	Aarkrog et al. (1997)
	Wild-caught fish	90 (60-365 )	0.64	Jones and Sherwood (2009)
	Farm-raised fish	14 (2-60 ) <sup>2</sup>	0.93	Jones and Sherwood (2009)
	Bivalve/crustacean	7 (4-548 ) <sup>2</sup>	0.97	Jones and Sherwood (2009)
Canned				
	Seafood	177 (10-2070)	<b>0.50 (0.14-0.96)</b> <sup>1</sup>	This study (Table S3)
	Seafood (canned)	365	0.16	Aarkrog et al. (1997)
Dried	Seafood	51 (10-410)	<b>0.81 (0.26-0.96)</b> <sup>1</sup>	This study (Table S3)
Smoked	Marine fish	13 (2-62)	<b>0.95 (0.78-0.99)</b> <sup>1</sup>	This study (Table S3)
All types				
	Seafood <sup>3</sup>		0.81	This study (Table S3)
			(0.14-0.1.0) <sup>1,3</sup>	
	Seafood	90	0.6	Aarkrog et al. (1997) <sup>4</sup>

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For reference data, the correction factors are calculated here from the published delay estimates using a 1005 standard decay equation without ingrowth of <sup>210</sup>Po from <sup>210</sup>Pb, as is consistent with past approaches. For this study, the R<sub>decay</sub> factors include the ingrowth of <sup>210</sup>Po from <sup>210</sup>Pb and are therefore slightly higher 1007 1008 even when considering similar delay durations.

2 1009 The reference does not explain or support this comparatively short duration.

1010 3 Percentage consumption weighting is world median Fresh=44%, Frozen=34%, Canned=11%, Dried=7%, 1011 Smoked=3%, (based on FAO 2018, (Sun et al., 2022)).

Aarkrog et al., 1997 report correction factors of 0.6 in the text (results section) and also reported use of a 1012 1013 value of 0.5 in calculations.

Part of the difference in R<sub>decav</sub> factors from this study vs previous is that the values presented here 1014 include the <sup>210</sup>Po ingrowth from the <sup>210</sup>Pb that is in the seafood (as measured at the time of harvest; 1015 Eqn (2)). This ingrowth is relatively minor because the <sup>210</sup>Pb activity concentrations in seafood are 1016 typically relatively low compared to <sup>210</sup>Po which is mainly derived via the diet of the organism. In 1017 addition, the <sup>210</sup>Po ingrowth is relatively slow (about 2.3 years to equilibrate). For Fresh and Smoked 1018 1019 categories, the ingrowth adds only 0.01 to the fraction retained. For the longest category of Canned, 1020 the ingrowth adds 0.09 to the fraction estimate. These are minor, but not insignificant increases in <sup>210</sup>Po, 1021

The ingrowth component of equation (2) requires input of the initial activity concentration <sup>210</sup>Pb 1022 relative to <sup>210</sup>Po (the ratio of <sup>210</sup>Pb to <sup>210</sup>Po activity concentrations at the time of harvest). For this 1023 study, a generic <sup>210</sup>Pb/<sup>210</sup>Po ratio of 0.16 was used (<sup>210</sup>Po/<sup>210</sup>Pb ratio of 6.3) which was obtained from 1024

- 770 paired <sup>210</sup>Po and <sup>210</sup>Pb data after filtering data for edible tissues and considering the seafood 1025 types and proportions consumed globally (fish=56.8%, Crustacean=14.4%, Bivalve=17.0%, 1026 1027 Gastropod=2.4%, Cephalopod=3.9% and seaweed=5.4%; Cai and Leung, 2017). In deriving the generic <sup>210</sup>Pb/<sup>210</sup>Po ratio of 0.16, the <sup>210</sup>Pb and <sup>210</sup>Po activity concentration data were screened for 1028 relatively high ± uncertainties, and removed if their ± uncertainty values were greater than 50% of 1029 the reported activity concentrations. The same test was applied to the propagated uncertainties on 1030 the <sup>210</sup>Pb/<sup>210</sup>Po activity concentration ratios. The application of these screens did not introduce 1031 1032 substantive bias across the distribution ranges (e.g., data were removed relatively evenly across the 1033 entire range of activity concentrations) and therefore the estimates on the distribution means did not change substantially. This screening provided greater confidence in the <sup>210</sup>Pb/<sup>210</sup>Po ratio used in 1034 1035 equation (2).
- 1036

1037 S.3 <sup>210</sup>Po differences in farm-raised vs wild-caught marine fish

Differences in the activity concentrations between mariculture-sourced vs wild-caught seafood are 1038 1039 determined through the straight-forward comparison of measurements in comparative samples. A 1040 search was conducted focused on scientific journals, science conference proceedings, industry 1041 reports, government documents and websites (through 2022). Various search terms were used: mariculture, aquaculture, seafood, marine fish, fish, crustaceans, cephalopods, farmed, farm-raised, 1042 pen-raised, , artificial feed, radionuclides, radiological dose, polonium, <sup>210</sup>Po. In this study, the <sup>210</sup>Po 1043 activity concentrations of the mariculture vs wild-caught were compared for the same or similar 1044 1045 species from the same region.

1046	Table S3.1. Summary <sup>210</sup> Po activity concentrations in the muscle tissues of farm-raised marine fish
1047	grouped by species. Note that the feed types are different among studies (see main text).

		<sup>210</sup> Po			
Species and reference	Common name	Bq kg⁻¹ (ww)	±	Ν	Reference
Fish raised with					
processed feed					_
Salmo salar <sup>-</sup>	Atlantic Salmon				_
		0.003	0.001	1	Heldal et al., 2019)
		0.004	0.001	1	u
		0.013	0.003	1	"
		0.02	0.008	1	- <i>u</i>
		0.022	0.007	1	- <i>u</i>
		0.008	0.002	1	"
		0.023	0.008	1	"
		0.016	NA		(Smith, 2006) <sup>2</sup>
		0.13	NA		"
		0.1	NA		"
		0.13	NA		- "
Sparus aurata	Gilthead seabream	0.181	0.022	2	(Jalili et al., 2009)
Fish raised with					-

natural marine- sourced feed	-				-
Larimichthys polyactis,	Yellow croaker	1.39	0.15	18	(Kong et al., 2021)
Pagrus major	Red sea bream	10.4	1.2	12	- "
Lateolabrax japonicus	Common sea perch	0.99	0.12	11	u
Crustaceans raised with processed feed					
Litopenaeus vannamei,	Whiteleg shrimp	0.117	0.019	1	(Dong et al., 2018)
Litopenaeus vannamei,	Whiteleg shrimp	0.562	0.099	1	u
Penaeus monodon	Tiger prawn	10.5	0.4	1	"

1048 <sup>I</sup>. From commercial/industrial-scale operations.

1049 <sup>2</sup> The available data from Smith 2006 were the minimum and maximums for two time/sampling periods.

The available data on <sup>210</sup>Po activity concentrations in the edible tissues of mariculture seafood were 1050 few (N=19) and limited to fish and crustaceans. The primary publications were: Heldal et al. (2019), 1051 1052 Kong et al. (2021) Jalili et al. (2009), Smith (2004). Data from bivalve aquaculture is not included as 1053 processed feeds are mainly used for broodstock/larval stages and not for the period of primary 1054 bivalve growth which typically is caried out in natural marine settings (Smaal et al., 2018). Only limited data were found from one study on changes in <sup>210</sup>Po in mariculture vs wild-caught 1055 crustaceans. More data are needed for this category as they are a major seafood source with 1056 1057 approximately 9.4 million tonnes of crustaceans harvested from mariculture each year (2018 FAO 1058 data reported by (2018 FAO data reported by Albalat et al., 2022).

1059

1060 S.4 Example of typical <sup>210</sup>Po reduction using the recommended correction factors

1061 The potential losses of <sup>210</sup>Po associated with the cooking, decay and mariculture influences can be 1062 significant. If for example, when the mean (or recommended) correction factors are used, the data 1063 suggest that, at the time of ingestion, seafood often has only about 55% of <sup>210</sup>Po content as 1064 compared to when it was harvested. The example calculation is:

1065 Overall generic correction factor = (1 X 0.74) \* (1 X 0.81) \* ((0.89 X 1) + (0.11 X 0.1)) = 0.55.

1066 Table S4.1 Recommended typical correction factors.

Factor	Value	Section reference
F <sub>c</sub> (all cooking)	1	3.1.1
R <sub>cooking</sub> (all seafood)	0.74	
F <sub>d</sub> (all methods)	1	3.2.1
R <sub>decay</sub>	0.81	
F <sub>f</sub> (wild caught)	0.89	3.3.1
R <sub>farmed</sub> (wild caught)	1	
F <sub>f</sub> (mariculture)	0.11	
R <sub>farmed</sub> (mariculture)	0.2	
Generic Global correction		
factor that considers cooking,	0.55	

delay-decay and mariculture	
corrections.	