



1	Artificial Radionuclides in Squid from northwestern Pacific in 2011 following the
2	Fukushima accident
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8	Abstract:
9	In order to better understand the impact of Fukushima Nuclear Power Plant (NPP) Accident on
10	commercial marine species, squid (Ommastrephe bartrami) samples, obtained from the
11	northwestern Pacific in November 2011, were analyzed for a range of artificial and natural
12	radionuclides (Cs-134, Cs-137, Ag-110m, U-238, Ra-226 and K-40). Short-lived radionuclides Cs-
13	134 and Ag-110m released from Fukushima NPP Accident were found in the samples, with an
14	extremely high water-to-organism concentration ratio for Ag-110m (> 2.9E+04). The radiological
15	dose rates for the squid from the radionuclides measured were far lower than the relevant benchmark
16	of 10 $\mu Gy\ h^{\text{-1}}.$ For human consumers ingesting these squid, the dose contribution from natural
17	radionuclides (>99.9%) including Po-210, was far greater than that of Fukushima-accident
18	radionuclides (<0.1%). The whole-body to tissue and whole-body to gut concentration ratios were
19	calculated and reported, providing a simple method to estimate the whole-body concentration in the
20	environmental monitoring programs, and filling the data gap of concentration ratios in cephalopods.
21	Our results help fill data gaps on uptake of NPP radionuclides in the commercially important
22	Cephalopoda class and add to scarce data on open-ocean nekton in the northwestern Pacific soon
23	after the Fukushima accident.
24	Key words:
25	Fukushima NPP Accident; squid; concentration ratios; radiological dose; Silver-110m.
26	

27 1. Introduction

28 The Fukushima Daiichi Nuclear Power Plant (NPP) Accident, which was caused by the combined

29 effect of the great earthquake and subsequent tsunami in March 2011, resulted in increased levels





30 of artificial radioactivity in the marine environment to the east of Japan (IAEA 2015). The 31 radioactive releases, dominated by radiocesium, were transported and dispersed widely in the North 32 Pacific within a few years (Aoyama et al., ;Smith et al.), raising concerns about the potential impact 33 on the marine biota and human consumers of seafood products.

A large amount of research has been conducted to determine the level of artificial radionuclides in biota samples and to assess the relevant radiological impact to both human and marine species. However, most studies have focused on the concentration of radiocesium in fish (Johansen et al., 2015), and only a few publications have reported on radionuclides in other marine species (Buesseler et al., ;Yu et al.). Few data are available for open-ocean locations as compared with coastal areas, especially from 2011. Filling these data gaps will improve and expand understanding of the dynamics of cesium in the early months following the accident.

41 Ommastrephe bartrami (neon flying squid) is a migratory squid species that is commercially 42 important, consumed by humans, and common in the Pacific Ocean and circumglobally in temperate 43 and tropical waters. It feeds near the surface on small fish and is thus a potential accumulator of 44 radiocesium via diet and water pathways. Moreover, cephalopods have a strong capability to 45 accumulate silver in their bodies (Miramand and Bentley, 1992;Bustamante et al., 2004) and would potentially indicate uptake of the short-lived (0.70 year half-life) Ag-110m released from Fukushima 46 Daiichi NPP Accident. Similarly, the presence of Cs-134 (2.1 year half-life) in samples would also 47 indicate a pathway from Fukushima Daiichi NPP releases. Therefore, specimens captured at 48 locations in the North Pacific may serve as bio-indicators of the presence, strength, and movement 49 50 of the radioactive signal from Fukushima Daiichi Accident.

This study assesses samples of *O. bartrami* obtained from the northwestern Pacific in November 2011 for a range of artificial and natural radionuclides (Cs-134, Cs-137, Ag-110m, U-238, Ra-226 and K-40). The radiological dose rates and relevant risk levels were determined for the squid, as well as potential dose rates to human consumers of squid seafood. Consistent with international efforts to compile transfer data, Concentration Ratios (wholebody-to-water and wholebody-to-tissue) are calculated and reported, including those for different age classes of squid.





58 2. Materials and methods

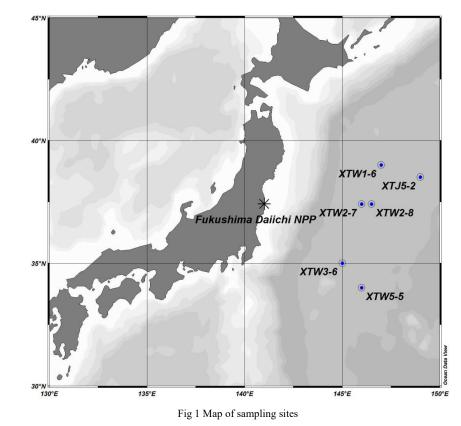
- 59 2.1. Sample collection and analytical procedure
- Thirteen composite samples of O. bartrami samples with a total weight of 126.2 kg were obtained 60 61 by bait fishing in open water in northwestern Pacific. Six sampling locations were selected in the area of 34°-39°N to 145°-149°E to investigate eastward deposition and oceanic migration pathways 62 of radionuclide releases from the Fukushima Daiichi NPP (Fig 1). To ensure sample mass was 63 64 sufficient to reach minimum detectable activity (MDA) levels for key radionuclides, composite samples were made with multiple specimens from the same sampling site. For those sites with 65 66 enough sample mass, the specimens were divided into different composite categories according to their body weight. Specimens with body mass less than 1 kg were categorized as "small", those 67 between 1 kg and 2 kg were categorized as "medium" and those more than 2 kg were categorized 68 as "large". The samples were -18 °C frozen on board for transport to the laboratory for the 69 70 subsequent analysis.

Squid samples were dissected into muscle and gut tissues after thawing, dried at 50 °C, and ashed at 450 °C. The fresh weight and ash weight of the composite samples were recorded. The ash was sealed in cylindrical 75 mm diameter containers, and then subjected to HPGe spectrometry for detection of gamma-emitting radionuclides.

75 Gamma rays from artificial radionuclides (Cs-134, Cs-137, Ag-110m, Co-58, Co-60, Mn-54 and 76 Zn-65) and natural radionuclides (K-40, Ra-226 and U-238) were analyzed using a planar highpurity germanium (HPGe) detector (Model BE6530 with Multi Channel Analyzer Lynx system; 77 Canberra, U.S.A.). Detection efficiencies for the geometry used were 2.7885 % and 2.4476 %, for 78 79 Cs-137 and Ag-110m respectively. The counting time for each sample was 24 hr. Genie 2000 80 software was used to analyze the respective peaks in the energy spectrum. The concentrations were corrected for decay to the initial date of the nuclear accident on 12 March 2011, when the first 81 82 hydrogen explosion occurred in Unit 1 of the FDNPP (Wakeford).









2.2. Dose assessment for squid

The ERICA Assessment Tool (version 1.2) (Brown et al., 2008) was used with Tier 2 assessment 86 87 to evaluate the radiological risk to squid from the study areas in 2011. The ERICA Tool includes the capability to specify organism sizes, and in this study, average mass (1.3 kg) and dimensions 88 89 (ellipsoid equivalent of 0.3m, 0.1m, 0.085m length, width, height respectively) from the specimens were used to calculate dose rates. The dimensions of the average O. bartrami happen 90 to be very similar to the standard ERICA "pelagic fish" and therefore the dose rates are very similar 91 92 as calculated by ERICA. The measured activity concentrations in the whole-body of ¹³⁷Cs, ¹³⁴Cs, ^{110m}Ag, ²²⁶Ra and ²³⁸U in the samples were used as dose calculation input. The maximum tissue 93 activity concentrations were used for a more conservative result. As O. bartrami are migratory, 94 95 their radionuclide tissue levels represent an integrated accumulation from recently traversed 96 areas in the open ocean area. The exact migratory routes are not known. Therefore, the external 97 dose rates to the squid were calculated using the average of water radioactivity levels in the





98	study capture region (average of samples across all sampling locations). Use of the average is
99	reasonable in this instance as the external dose rates for artificial radionuclides were much
100	smaller than internal dose rates and therefore variable water activity concentrations had little
101	influence on overall dose results. For internal dose rates to squid, the dose conversion
102	coefficients (DCCs) were calculated within the ERICA tool (supplemental). The occupancy
103	factors were 100% in water, and weighting factors of internal low beta, internal beta/gamma
104	and internal alpha were set as 3, 1 and 10 respectively.
105	2.3. Dose from ingesting squid by human consumers
106	Committed effective doses (Sv) to human consumers of squid were estimated using standard
107	exposure-to-dose conversion factors for ingestion from ICRP Compendium of dose coefficients
108	based on ICRP Publication 60 (ICRP, 1999). Key DCFs are 1.30E-08 and 1.90E-08 Sv Bq ⁻¹ for Cs-
109	137 and Cs-134 (DCFs provided in the supplemental). The factors are multiplied by intake (e.g. kg
110	yr-1) to obtain committed effective doses to the consumer. In this study, the annual intake rate of
111	seafood by an adult consumer is assumed to be 20 kg yr $^{-1}$ (consistent with world per capita fish and
112	related seafood consumption (FAO, 2016). As a conservative assumption, the entire 20 kg yr $^{-1}$ for a

113 hypothetical consumer is assumed to be sourced from the squid of the study area east of the

114 Fukushima Daiichi NPP (in practice, only a small percentage of seafood diet would be sourced from

115 this region). As most dose to human consumers of seafood typically comes from the natural

116 radionuclide Po-210 (~89%;(Johansen et al., 2015)), the seafood ingestion dose rates here included

117 Po-210 using conservative generic data for marine seafood(Carvalho, 2011;Hosseini et al., 2010).

118 2.4. Whole-body concentration ratios

119 The water-to-organism whole-body Concentration Ratio CR_{WB} (in L/kg) used here is defined as:

120
$$CR_{WB:Water} = \frac{Whole-Body Activity Concentration (fresh mass) (Bq/kg-wet)}{Water Activity Concentration(Bq/L)}$$
 (1)

121 The whole-body activity of an radionuclide was estimated using a mass balance approach

122 (Yankovich et al., 2010) to reconstruct the amount of radionuclide in the whole-body of the squid.

- 123 The whole-body to tissue concentration ratio (dimensionless) was estimated as:
- 124 $CR_{WB:Tissue} = \frac{\sum [Tissue_t Activity Concentration (fresh mass) \cdot Tissue_t fresh mass fraction]}{Tissue_t Activity Concentration (fresh mass)}$ (2)





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126 3. Results and discussions

127 3.1. Description of O. bartrami specimens

128 In total, 98 specimens were obtained from 6 stations. The mass of the specimens ranged from 118 g 129 to 2551 g, on average 1347 g. Sixty percent of the specimens weighed 701 g to 1700 g. The trunk length of the specimens ranged from 115 mm to 440 mm, on average 333 mm. Seventy-five percent 130 131 of the specimens had a length greater than 290 mm (adult size) suggesting that the majority of the 132 specimens were hatched in winter of 2010 or spring in 2011 and had been living for 8 to 11 months 133 (Wang and Chen, 2005). Combining the estimated age of the squid, and assuming residence in the 134 general area east of Fukushima Prefecture, it can be inferred that most specimens had been accumulating radionuclides since Fukushima Daiichi NPP accident, while a minor proportion (the 135 136 small size category) were likely to have been hatched after the accident and had shorter exposure 137 times.

138 3.2. Activity concentrations and CRs in squid

The activity levels of radionuclides in Table 1 indicate that all *O. bartrami* size classes had accumulated radionuclides from Fukushima Daiichi NPP releases as indicated by Cs-134 and Ag-110m. The squid specimens had a strong capability to concentrate Ag in their bodies. The maximum activity of Ag-110m in the whole body of *O. bartrami* was up to 9 Bq/kg, as compared to that in water which was below the MDA of 0.22 Bq/m³, indicating a maximum concentration factor that is higher than 4×10^4 . The mean CRs for Ag-110m were calculated as $>2.95 \times 10^4 \pm 9.84 \times 10^3$ (Table 2), using the MDA as the activity of seawater in Equation (1).

146 Although this estimate is with large uncertainties because of using MDA of Ag-110m as the water 147 concentration, these Ag data provide new insights for international researchers and fill a gap as the (Wildlife 148 relevant international database Transfer Parameter Database: 149 www.wildliftransferdatabase.org) which has entrees for Ag uptake in the mollusk category, but none specifically for squid/cephalopods. 150

151 The mean CR_{WB} values for Cs-134 and Cs-137 in *O. bartrami* were 6.33 (\pm 2.80 S.D.) and 5.57 152 (\pm 2.59 S.D.) respectively. These values are similar to previously published mean concentration





153 factors for Cs in cephalopods ranging from 9 to 14 (IAEA, 1978;Ishii et al., 1978;Suzuki et al., 154 1978;IAEA, 2004). The slightly lower CR_{WB} in this study is well within the range of expected variation, which can be very high for water-to-organism CR values (e.g. reported CRs for Cs-137 155 156 in marine fish range over nearly an order of magnitude) (Beresford, 2010). The activity 157 concentration of ¹³⁷Cs/¹³⁴Cs in the research area reached a maximum of ~600 Bg m⁻³ in June 2011 and soon decreased to below 100 Bq m-3 (Aoyama et al., 2016). Considering the temporal change 158 159 of radiocesium in seawater and its relatively short biological half-life (~70 days) in marine organisms, in this study, the CR calculation used mean Cs-134 and Cs-137 seawater activity 160 161 concentrations (35.1 and 36.2 Bq m⁻³ respectively) from this study's November 2011 sampling, which were similar to the ~50 Bq m⁻³ reported for July-December timeframe from the same open 162 163 ocean area (Kaeriyama, 2017).

164 The results also showed that both Cs-134 and Cs-137 were concentrated mainly in the muscle of the 165 squid. Cesium behaves similarly to potassium in biota and it tends to be distributed to the muscle tissue. These results for the open ocean, real-world conditions are consistent with previous 166 laboratory results of 80+ % accumulation in the muscle and head of cuttlefish after only 8 hours of 167 168 exposure to water (Bustamante et al., 2004). In contrast, for Ag, the open ocean squid had 95 % Ag in the gut vs muscle. This result was also consistent with the laboratory cuttlefish which had 98% 169 170 Ag in the gut following a single spiked feeding and 29 d depuration (Bustamante et al., 2004). From 171 the same study, within the gut, accumulation of Ag is dominantly in the digestive gland.

172 The smallest squid samples had the highest concentration factors for Cs-134, Cs-137, Ag-110m and U-238 (Fig 3). The higher accumulation occurred in the smaller size class despite their inferred 173 shorter exposure times (shorter lifespan) as compared with the larger size class. These results are 174 175 consistent with observed Cs depuration rates in juveniles cephalopods (Sepia officinalis) being 176 ~four times slower than that of adults, with however, both being relatively fast (adult cuttlefish biological half-life of 16 days for Cs and 9 days for Ag (Bustamante et al., 2004)). This previous 177 178 study, suggests the radiocesium accumulation and depuration in O. bartrami is relatively rapid and 179 that our results therefore primarily reflect recent (~ several months) exposure rather than longer-180 term accumulation.





181 The levels of activity for ⁵⁸Co, ⁶⁰Co, ⁵⁴Mn and ⁶⁵Zn in the samples were all below the MDA (0.22

182 mBq/g-ash).

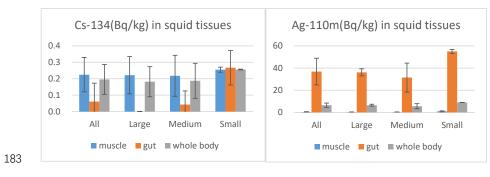




Fig 2 Activity concentrations of Cs-134 (left) and Ag-110m (right) in squid tissues.



Size Cx-137 Cx-134 Agr110m K-40 Ra-226 II-328 II-328 All Range Average Average <t< th=""><th></th><th></th><th></th><th></th><th>Table</th><th>1 Statistics o</th><th>f radionuclide</th><th>Table 1 Statistics of radionuclides' levels in composite samples (Bq/kg-fresh mass)</th><th>nposite sample</th><th>es (Bq/kg-fresh</th><th>ו mass)</th><th></th><th></th><th></th></t<>					Table	1 Statistics o	f radionuclide	Table 1 Statistics of radionuclides' levels in composite samples (Bq/kg-fresh mass)	nposite sample	es (Bq/kg-fresh	ו mass)			
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WB $0.08-0.38$ 0.23 ± 0.10 $0.05-0.31$ 0.20 ± 0.09 $1.70-9.04$ 6.49 ± 1.97 $53.57-88.09$ 72.13 ± 8.45 $nd-0.17$ 0.07 ± 0.05 $0.27-5.32$ M $0.13-0.46$ 0.2 ± 0.13 $0.09-0.39$ 0.22 ± 0.11 $0.06-0.36$ 0.24 ± 0.13 $6.762-94.80$ 80.55 ± 11.49 $nd-0.07$ 0.04 ± 0.03 $0.33-0.94$ GNDNDNDNDNDNDND $32.25-40.50$ 36.14 ± 3.19 $49.57-58.88$ 53.00 ± 3.66 0.24 ± 0.27 $nd-70.76$ 0.34 ± 0.27 $nd-789$ WB $0.11-0.38$ 0.21 ± 0.11 $0.08-0.31$ 0.18 ± 0.09 $5.53-7.78$ 6.54 ± 0.80 $5.55-88.09$ 75.76 ± 9.86 $nd-0.15$ 0.04 ± 0.06 $0.54-2.09$ M $0.11-0.38$ 0.21 ± 0.11 $0.08-0.31$ 0.18 ± 0.09 $5.53-7.78$ 6.54 ± 0.80 75.76 ± 9.86 72.76 ± 9.86 $nd-0.15$ 0.07 ± 0.06 M $0.11-0.38$ 0.21 ± 0.11 $0.08-0.31$ $0.08-0.34$ 0.22 ± 0.12 $0.06-0.45$ 6.54 ± 0.30 $5.52-78.89$ 72.30 ± 8.82 $nd-0.15$ $nd-0.16$ 0.24 ± 0.27 M $0.10-0.41$ 0.27 ± 0.14 $0.08-0.34$ 0.22 ± 0.12 $0.06-0.45$ 0.25 ± 0.13 $5.52-78.89$ 72.30 ± 8.82 72.30 ± 8.82 0.07 ± 0.06 $0.16-0.86$ M $0.10-0.41$ 0.02 ± 0.12 $0.06-0.34$ 0.22 ± 0.12 $0.06-0.45$ 8.10 ± 2.73 47.80 ± 2.118 $nd-0.53$ 0.16 ± 0.05 $0.16-0.86$ M $0.21-0.24$ 0.22 ± 0.12 0.09 ± 0.24 0.25 ± 0.26 0.25 ± 0.26 $0.$	ША ()	IJ	nd-0.33	0.05 ± 0.10		0.06 ± 0.11	8.10-56.27	36.85 ± 12.02	9.72-72.37	53.03±15.77	0-bu	0.28 ± 0.28	nd-26.89	5.40 ± 7.60
M 0.13-0.46 0.26±0.13 0.09-0.39 0.22±0.11 0.06-0.36 0.24±0.13 67.62-94.80 80.55±11.49 md-0.07 0.04±0.03 0.33-0.94 G ND ND ND ND ND ND 32.540.50 36.14±3.19 49.57-58.88 53.00±3.66 md-0.07 0.04±0.03 0.33-0.94 WB 0.11-0.38 0.21±0.11 0.08-0.31 0.18±0.09 5.53-7.78 6.54±0.83 64.55-88.09 75.76±9.86 md-0.15 0.07±0.06 0.54±0.20 M 0.11-0.38 0.21±0.11 0.06-0.34 0.25±0.19 5.53-73.88 75.76±9.86 md-0.15 0.07±0.06 0.54±2.09 M 0.10-0.41 0.27±0.14 0.06-0.34 0.25±0.19 56.29+78.78 72.30±8.12 md-0.15 0.07±0.02 0.16-0.86 0.54+2.09 M 0.10-0.41 0.27±0.12 0.04±0.03 8.10-45.05 72.30±8.12 md-0.15 0.07±0.02 0.16-0.36 0.16-0.36 M 0.21-0.31 0.35±0.22 0.24±0.13 5.61±2.31 5	(c1=11)	WB	0.08-0.38	0.23 ± 0.10		0.20 ± 0.09		6.49 ± 1.97	53.67-88.09		nd-0.17	0.07 ± 0.05	0.27-5.32	1.35 ± 1.40
G ND ND </th <th>0.000</th> <th>W</th> <td>0.13-0.46</td> <td>0.26 ± 0.13</td> <td></td> <td>0.22 ± 0.11</td> <td>0.06-0.36</td> <td>0.24 ± 0.13</td> <td>67.62-94.80</td> <td>80.55 ± 11.49</td> <td>nd-0.07</td> <td>0.04 ± 0.03</td> <td>0.33-0.94</td> <td>0.63 ± 0.26</td>	0.000	W	0.13-0.46	0.26 ± 0.13		0.22 ± 0.11	0.06-0.36	0.24 ± 0.13	67.62-94.80	80.55 ± 11.49	nd-0.07	0.04 ± 0.03	0.33-0.94	0.63 ± 0.26
WB 0.11-0.38 0.21±0.11 0.08-0.31 0.18±0.09 5.53-7.78 6.54±0.83 64.55-88.09 75.76±9.86 md-0.15 0.07±0.06 0.542-09 M 0.10-0.41 0.27±0.14 0.06-0.34 0.25±0.12 0.06-0.46 0.25±0.19 56.29-78.78 72.30±8.12 md-0.15 0.016-0.66 0.16-0.86 M 0.10-0.41 0.27±0.12 0.06-0.46 0.25±0.19 56.29-78.78 72.30±8.12 md-0.53 0.16-0.86 0.16-0.86 M 0.10-0.41 0.27±0.12 0.06-0.45.85 31.40±13.05 9.72-67.73 47.80±21.18 md-0.53 0.16-0.86 0.16-0.86 WB 0.08-0.35 0.23±0.12 0.06+0.45.85 31.40±13.05 57.6-73.94 68.16±7.76 md-0.11 0.04±0.04 0.27-2.45 M 0.21-0.34 0.27±0.09 0.24+0.27 0.46-0.15 5.61±2.31 53.67-73.94 68.16±7.75 md-0.11 0.04±0.04 0.27-2.45 M 0.21-0.34 0.24+0.27 0.46-0.25 0.65+1.26 0.65-1.23 0.27-2.35 0.24	Large	IJ	ŊŊ	ND	ND	Ŋ	32.25-40.50	36.14 ± 3.19	49.57-58.88	53.00 ± 3.66	nd-0.68	0.24 ± 0.27	nd-7.89	2.61 ± 3.22
M 0.10-0.41 0.27 \pm 0.14 0.06-0.34 0.22 \pm 0.12 0.06-0.46 0.25 \pm 0.19 56.29-78.78 72.30 \pm 81.12 ind-0.05 0.02 \pm 0.02 0.16-0.86 G ind-0.13 0.03 \pm 0.05 ind-0.21 0.04 \pm 0.08 8.10-45.85 31.40 \pm 13.05 9.72-67.73 47.80 \pm 21.18 ind-0.05 0.18 \pm 0.19 ind-10.44 WB 0.08-0.35 0.23 \pm 0.12 0.04 \pm 0.01 1.70-8.09 5.61 \pm 2.31 53.67-73.94 68.16 \pm 7.56 ind-0.05 0.18 \pm 0.19 ind-10.44 MB 0.21-0.34 0.25+0.02 0.19\pm0.11 1.70-8.09 5.61 \pm 2.31 53.67-73.94 68.16 \pm 7.56 ind-0.05 0.04 \pm 0.04 0.27-2.45 M 0.21-0.34 0.27+0.09 0.24-0.27 0.25+0.02 0.65+1.26 0.97\pm0.45 75.05-78.88 76.07 \pm 3.97 ind-0.05 0.02\pm0.03 0.41-1.77 M 0.21-0.33 0.27\pm0.09 0.19-0.34 0.27\pm0.12 53.64-56.27 54.95 \pm 1.86 65.30-72.37 68.83 \pm 4.99 0.67\pm0.32 12.00-26.89 0.41-0.78 0.41-0.89	(c=II)	WB	0.11-0.38	0.21 ± 0.11		0.18 ± 0.09	5.53-7.78	6.54 ± 0.83	64.55-88.09		nd-0.15	0.07 ± 0.06	0.54-2.09	0.97 ± 0.63
G nd-0.13 0.03±0.05 nd-0.21 0.04±0.08 8.10-45.85 31.40±13.05 9.72-67.73 47.80±21.18 nd-0.53 0.18±0.19 nd-10.44 WB 0.08-0.35 0.23±0.12 0.05-0.29 0.19±0.11 1.70-8.09 5.61±2.31 53.67-73.94 68.16±7.56 nd-0.11 0.04±0.04 0.27-2.45 M 0.21-0.34 0.27±0.09 0.24+0.27 0.25±0.02 0.65-1.29 0.97±0.45 73.26-78.88 76.07±3.97 nd-0.05 0.02±0.03 0.41-1.77 M 0.21-0.33 0.27±0.09 0.19-0.34 0.24-56.12 54.95±1.86 65.30-72.37 68.83±4.499 0.44-0.89 0.67±0.32 12.00-26.89 WB 0.21-0.34 0.27±0.09 0.19-0.34 0.27±0.11 53.64-56.27 54.95±1.86 65.30-72.37 68.83±4.499 0.67±0.32 12.00-26.89 WB 0.21-0.34 0.27±0.09 0.25±0.10 8.90-9.04 8.97±0.10 73.13-76.76 74.95±2.57 0.07-0.17 0.12±0.07 221-5.32		W	0.10 - 0.41	0.27 ± 0.14	0.06-0.34	0.22 ± 0.12	0.06 - 0.46	0.25 ± 0.19	56.29-78.78	72.30 ± 8.12	nd-0.05	0.02 ± 0.02	0.16 - 0.86	0.40 ± 0.28
WB 0.08-0.35 0.23±0.12 0.05+0.29 0.19±0.11 1.70-8.09 5.61±2.31 53.67-73.94 68.16±7.56 nd-0.11 0.04±0.04 0.27-2.45 M 0.21-0.34 0.27±0.09 0.24+0.27 0.25±0.02 0.65-1.29 0.97±0.45 73.26-78.88 76.07±3.97 nd-0.05 0.02±0.03 0.41-1.77 G 0.21-0.33 0.27±0.09 0.19-0.34 0.27±0.11 53.64-56.27 54.95±1.86 65.30-72.37 68.83±4.499 0.67±0.32 12.00-26.89 WB 0.21-0.34 0.27±0.09 0.19-0.34 0.27±0.11 53.64-56.27 54.95±1.86 65.30-72.37 68.83±4.499 0.67±0.32 12.00-26.89 WB 0.21-0.34 0.27±0.09 0.25±0.26 0.26±0.00 8.90-9.04 8.97±0.10 73.13-76.76 74.95±2.57 0.07-0.17 0.12±0.07 221-55.32		IJ	nd-0.13	0.03 ± 0.05			8.10-45.85	31.40 ± 13.05	9.72-67.73	47.80±21.18	nd-0.53	0.18 ± 0.19	nd-10.44	3.05 ± 3.84
M 0.21-0.34 0.27±0.09 0.24-0.27 0.25±0.02 0.65-1.29 0.97±0.45 73.26-78.88 76.07±3.97 md-0.05 0.02±0.03 0.41-1.77 G 0.20-0.33 0.27±0.09 0.19-0.34 0.27±0.11 53.64-56.27 54.95±1.86 65.30-72.37 68.83±4.99 0.44-0.89 0.67±0.32 12.00-26.89 WB 0.21-0.34 0.27±0.09 0.26±0.00 8.90-9.04 8.97±0.10 73.13-76.76 74.95±2.57 0.07±0.07 221-5.32	(0=U)	WB	0.08-0.35	0.23 ± 0.12		0.19 ± 0.11	1.70-8.09	5.61 ± 2.31	53.67-73.94	68.16±7.56	nd-0.11	0.04 ± 0.04	0.27-2.45	0.86 ± 0.80
G 0.20-0.33 0.27±0.09 0.19-0.34 0.27±0.11 53.64-56.27 54.95±1.86 65.30-72.37 68.83±4.99 0.44-0.89 0.67±0.32 12.00-26.89 WB 0.21-0.34 0.27±0.09 0.25-0.26 0.20+0.04 8.97±0.10 73.13-76.76 74.95±2.57 0.07-0.17 0.12±0.07 2.21-5.32	C mol	Μ	0.21-0.34	0.27 ± 0.09		0.25 ± 0.02	0.65-1.29	0.97 ± 0.45	73.26-78.88	76.07 ± 3.97	nd-0.05	0.02 ± 0.03	0.41-1.77	1.09 ± 0.97
WB 0.21-0.34 0.27±0.09 0.25-0.26 0.26±0.00 8.90-9.04 8.97±0.10 73.13-76.76 74.95±2.57 0.07-0.17 0.12±0.07 2.21-5.32		IJ	0.20-0.33	0.27 ± 0.09		0.27 ± 0.11		54.95 ± 1.86	65.30-72.37	68.83 ± 4.99	0.44 - 0.89	0.67 ± 0.32	12.00-26.89	19.45 ± 10.53
	(7-11)	WB	0.21-0.34	0.27 ± 0.09		0.26 ± 0.00	8.90-9.04	8.97 ± 0.10	73.13-76.76	74.95±2.57	0.07-0.17	0.12 ± 0.07	2.21-5.32	3.76 ± 2.19









188 **3.3.** Whole-body:muscle and whole-body:gut concentration ratios

- 189 Most of non-human biota radiation dose assessing models focus on estimation of dose rates
- using the *whole-body* activity concentrations of radionuclides (Brown et al., 2008;DOE, 2004).
- 191 However, muscle tissue (vs. whole-body) is measured in most monitoring programs which
- 192 typically focus on seafood tissues consumed by humans. Therefore, there exists a need for
- 193 whole-body:tissue concentration ratios that allow for estimation of whole-body concentrations
- 194 from commonly measured tissue data (Yankovich et al., 2010).
- The whole-body:muscle and whole-body:gut concentration ratios for radionuclides in squid 195 196 samples are listed in Table 2. For many radionuclides, the tissue-specific concentration values for the small squids tend to be higher than those for large squids. The uncertainty of the whole-197 body:gut CRs for Cs-137 and Cs-134 are relatively high because of the relatively low level and 198 199 large activity range of radiocesium in the gut samples. These CRs presented here are calculated 200 for the non-equilibrium conditions following the accident. This issue is somewhat compensated 201 for by focusing on radionuclides that are taken up relatively quickly, and by using the average 202 activity concentrations over their relatively short lifespan of the squid. Equilibrium conditions 203 are generally not achieved in natural systems, and our results, like all CRs should be considered 204 in context. Further research is necessary to obtain a better estimation the biokinetics of uptake 205 in squid and of the whole-body:gut CRs for Cs-137 and Cs-134.



CR*	Size	Cs-137	Cs-134	Ag-110m	K-40	Ra-226	U-238
	IJМ	0.93 ± 0.28	$0.94{\pm}0.30$	41.87 ± 39.49	1.04 ± 0.28	2.75 ± 1.60	2.36 ± 1.36
	Large	0.82 ± 0.01	0.82 ± 0.01	38.89 ± 30.21	0.94 ± 0.01	2.42 ± 1.69	1.64 ± 0.79
M-GW	Medium	0.85 ± 0.03	0.86 ± 0.06	47.90±50.29	0.96 ± 0.01	2.00 ± 1.53	2.35 ± 1.20
	Small	1.00 ± 0.00	1.01 ± 0.07	10.30 ± 4.67	0.99 ± 0.02	3.58	4.22 ± 1.73
	All	2.59 ± 2.50	2.29±2.44	0.18 ± 0.02	1.30 ± 0.18	0.33 ± 0.29	0.24 ± 0.03
	Large	NA^{**}	NA^{**}	0.18 ± 0.01	1.43 ± 0.18	0.50 ± 0.43	0.28 ± 0.01
רם ש-פוע	Medium	4.15 ± 4.15	3.54 ± 3.54	0.18 ± 0.02	1.25 ± 0.12	0.24 ± 0.09	0.24 ± 0.01
	Small	1.03 ± 0.01	1.04 ± 0.40	0.16 ± 0.00	1.09 ± 0.12	0.17 ± 0.02	0.19 ± 0.01
	All	6.33 ± 2.80	5.57±2.59	$>2.95E+4 \pm 8.94E+3$	6.17 ± 0.71	14.66 ± 11.92	37.97±39.39
4711 U.M	Large	5.90 ± 2.91	5.18 ± 2.60	$>2.97E+4 \pm 3.76E+3$	6.42 ± 0.84	16.35 ± 12.46	27.34±17.89
W.b-W	Medium	6.30 ± 3.17	5.33 ± 3.04	$>2.55E+4 \pm 1.05E+4$	5.89 ± 0.69	9.56 ± 9.40	24.11±22.43
	Small	7.52±2.52	7.29 ± 0.06	$>4.08E+4 \pm 4.50E+2$	6.35 ± 0.22	25.76±15.07	106.09 ± 61.85



*** Values were calculated using mean Cs-137 seawater activity concentrations of 35.1 and 36.2 Bq m⁻³, and the MDA of ^{110m}Ag in seawater (0.22 Bqm⁻³).



(i) (c)





210 3.4. Dose assessment results

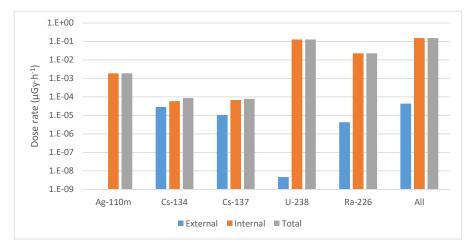
211 3.4.1. Dose rates for squid

The internal radiological dose rates to squid from artificial radionuclides (110mAg, 134Cs and 137Cs) 212 213 were collectively much higher than the external dose rates (Fig. 4). This is consistent with the 214 observed accumulation of radionuclides inside the squid body as compared with that in the surrounding seawater. The internal dose rates from FDNPP-associated artificial radionuclides were 215 216 lower, by two orders of magnitude, than those from the natural radionuclides measured in this study. From these radionuclides, only approximately 1.4 % of the total dose rate is estimated to have come 217 218 from the Fukushima Daiichi NPP releases. The total dose rate for squid is $0.15 \,\mu\text{Gy}\cdot\text{h}^{-1}$ from study radionuclides, and increases to approximately 0.61 µGy·h⁻¹ when adding Po-210 a natural 219 220 radionuclide and significant dose contributor in marine organisms (using a conservative generic 221 marine value of 15 Bq kg⁻¹-fresh mass and 0.001 Bq L⁻¹ in squid and seawater, respectively based on (Carvalho, 2011) and (Hosseini et al., 2010). These dose rates are much lower than the most 222 223 conservative screening benchmark dose rate of $10 \,\mu\text{Gy} \, h^{-1}$ (Garnier-Laplace et al., 2008). The dose 224 calculations used the measured activity concentrations in the squid (not CRs) and the calculated 225 dose rates represent a point in time (November 2011) with likely higher doses prior to, and lower 226 doses following the sampling date. However, the relatively low values indicate a more detailed (e.g. 227 pulse-dynamic uptake) dose calculation) is not necessary in this case. Overall, results indicate that 228 the radioactive releases from the Fukushima accident would not have a significant adverse effect on 229 O. bartrami individuals or populations living in the study area.

230







231 232

Fig 3 Dose rates $(\mu Gy \cdot h^{-1})$ from measured radionuclides for squid samples

233

234 **3.4.2.** Dose rates for human consumers of seafood

235 From the radionuclides measured in edible squid tissue (muscle), a committed effective ingestion dose of 0.01 mSv (median; minimum = 0.007 mSv, maximum = 0.014 mSv) would have occurred 236 to a hypothetical human consumer of 20 kg yr⁻¹ of squid from the study area (based on squid 237 238 captured in November 2011). The doses calculated here are hypothetical and are intended to be 239 conservative overestimates given the unrealistic assumption that all of the consumer's yearly 240 seafood came from the study area. If consumption of Po-210 (from natural background) is also 241 included, the total dose increases to 0.30 mSv, with almost all derived from Po-210 (Table 3). Of 242 this dose (including Po-210), less than 0.1 % is estimated to have been sourced from the Fukushima Daiichi NPP. This is consistent with previous findings that natural radionuclides provided far greater 243 244 dose rates to potential consumers of Pacific tuna (Fisher et al., 2013), and even for seafood sourced within a few kilometers of the Fukushima Daiichi NPP in 2013 (Johansen et al., 2015). The dose 245 contribution from the Fukushima Daiichi NPP releases for squid consumption of this study are far 246 247 below the 1 mSv per year recommended constraint for prolonged exposure by the public from 248 nuclear facility releases (ICRP, 1999).

250	Table 3. Ingestion dose estimates to human consumers of the squid in this study (Sv y ⁻¹ based on
251	20 kg consumption of study squid).

minimum	median	maximum	% this study*





6.98E-06	9.43E-06	1.18E-05	3.12%
3.36E-09	2.02E-08	7.22E-08	0.01%
2.28E-08	8.36E-08	1.48E-07	0.03%
2.60E-08	7.02E-08	1.20E-07	0.02%
	1.68E-07	3.92E-07	0.06%
1.44E-07	5.31E-07	1.59E-06	0.18%
1.44E-05	2.92E-04	1.08E-03	96.59%
	3.36E-09 2.28E-08 2.60E-08 1.44E-07	3.36E-09 2.02E-08 2.28E-08 8.36E-08 2.60E-08 7.02E-08 1.68E-07 1.44E-07 5.31E-07	3.36E-09 2.02E-08 7.22E-08 2.28E-08 8.36E-08 1.48E-07 2.60E-08 7.02E-08 1.20E-07 1.68E-07 3.92E-07 1.44E-07 5.31E-07 1.59E-06

252 * Based on median activity concentration values this study (Table 1 data, average of all sizes).

253 ** Po-210 from generic published data (Carvalho, 2011;Hosseini et al., 2010).

254

255 4. Conclusions

256 Elevated levels of Cs-134 and Ag-110m from Fukushima NPP Accident were found in the squid (O. bartrami) samples collected at NW Pacific in November 2011. This study filled a gap in 257 258 international transfer data by providing concentration ratios for several key NPP-associated 259 radionuclides in the whole-body and tissues of cephalopods. The Concentration Ratio for Ag-110m in squid was found as high as 4×10^4 L/kg in the smallest samples, with a mean value of 2.95×10^4 260 261 L/kg in all the samples, indicating that squid was a good bioindicator for Ag-110m from Fukushima 262 NPP Accident. The radiological dose contribution from the Fukushima Daiichi NPP releases for 263 squid living in the study area, and for human consumers of these squid, were both far below the 264 recommended dose limits. By comparison, natural radionuclides, particularly Po-210, provide 265 orders of magnitude greater dose rates.

266

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