

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 (*Ommastrephes bartramii*) 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  $10 \mu\text{Gy h}^{-1}$ . 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 eastwards in surface water across  
32 the mid-latitude North Pacific with a speed of 3-7 km day<sup>-1</sup> (3.5-8.0 cm s<sup>-1</sup>) and dispersed widely  
33 in the North Pacific within a few years (Aoyama et al., ;Smith et al.), raising concerns about the  
34 potential impact on the marine biota and human consumers of seafood products.

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35 A large amount of research has been conducted to determine the level of artificial radionuclides in  
36 biota samples and to assess the relevant radiological impact to both human and marine species.  
37 However, most studies have focused on the concentration of radiocesium in fish (Johansen et al.,  
38 2015), and only a few publications have reported on radionuclides in other marine species  
39 (Buesseler et al., ;Yu et al.). Few data are available for open-ocean locations as compared with  
40 coastal areas, especially from 2011. Filling these data gaps will improve and expand understanding  
41 of the dynamics of cesium in the early months following the accident.

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

52 This study assesses samples of *O. bartrami* obtained from the northwestern Pacific in November  
53 2011 for a range of artificial and natural radionuclides (Cs-134, Cs-137, Ag-110m, U-238, Ra-226  
54 and K-40). The radiological dose rates and relevant risk levels were determined for the squid, as  
55 well as potential dose rates to human consumers of squid seafood. Consistent with international  
56 efforts to compile transfer data, Concentration Ratios (wholebody-to-water and wholebody-to-tissue)  
57 are calculated and reported, including those for different age classes of squid.

58

## 59 **2. Materials and methods**

### 60 **2.1. Sample collection and analytical procedure**

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

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

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

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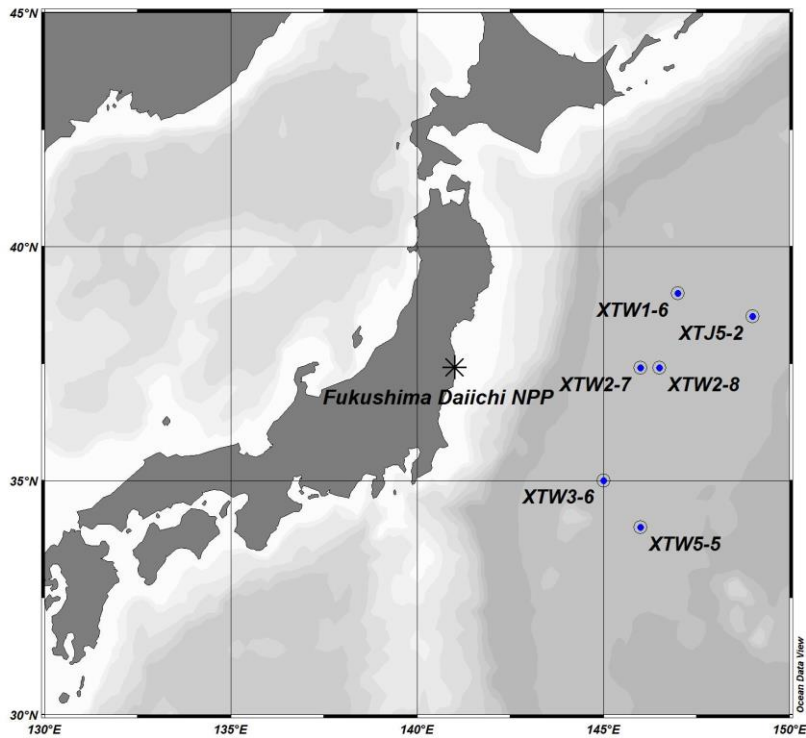


Fig 1 Map of sampling sites

## 2.2. Dose assessment for squid

The ERICA Assessment Tool (version 1.2) (Brown et al., 2008) was used with Tier 2 assessment 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 (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 to be very similar to the standard ERICA “pelagic fish” and therefore the dose rates are very similar as calculated by ERICA. The measured activity concentrations in the whole-body of  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{110\text{m}}\text{Ag}$ ,  $^{226}\text{Ra}$  and  $^{238}\text{U}$  in the samples were used as dose calculation input. The maximum tissue activity concentrations were used for a more conservative result. As *O. bartrami* are migratory, their radionuclide tissue levels represent an integrated accumulation from recently traversed areas in the open ocean area. The exact migratory routes are not known. Therefore, the external dose rates to the squid were calculated using the average of water radioactivity levels in the

99 study capture region (average of samples across all sampling locations). Use of the average is  
100 reasonable in this instance as the external dose rates for artificial radionuclides were much  
101 smaller than internal dose rates and therefore variable water activity concentrations had little  
102 influence on overall dose results. For internal dose rates to squid, the dose conversion  
103 coefficients (DCCs) were calculated within the ERICA tool (supplemental). The occupancy  
104 factors were 100% in water, and weighting factors of internal low beta, internal beta/gamma  
105 and internal alpha were set as 3, 1 and 10 respectively.

### 106 **2.3. Dose from ingesting squid by human consumers**

107 Committed effective doses (Sv) to human consumers of squid were estimated using standard  
108 exposure-to-dose conversion factors (DCFs) for ingestion from ICRP Compendium of dose  
109 coefficients based on ICRP Publication 60 (ICRP, 1999). Key DCFs are 1.30E-08 and 1.90E-08 Sv  
110 Bq<sup>-1</sup> for Cs-137 and Cs-134 (DCFs provided in the supplemental). The factors are multiplied by  
111 intake (e.g. kg yr<sup>-1</sup>) to obtain committed effective doses to the consumer. In this study, the annual  
112 intake rate of seafood by an adult consumer is assumed to be 20 kg yr<sup>-1</sup> (consistent with world per  
113 capita fish and related seafood consumption (FAO, 2016). As a conservative assumption, the entire  
114 20 kg yr<sup>-1</sup> for a hypothetical consumer is assumed to be sourced from the squid of the study area  
115 east of the Fukushima Daiichi NPP (in practice, only a small percentage of seafood diet would be  
116 sourced from this region). As most dose to human consumers of seafood typically comes from the  
117 natural radionuclide Po-210 (~89%;(Johansen et al., 2015)), the seafood ingestion dose rates here  
118 included Po-210 using conservative generic data for marine seafood(Carvalho, 2011;Hosseini et al.,  
119 2010).

### 120 **2.4. Whole-body concentration ratios**

121 The water-to-organism whole-body Concentration Ratio CR<sub>WB</sub> (in L/kg) used here is defined as:

$$122 \quad CR_{WB:Water} = \frac{\text{Whole-Body Activity Concentration (fresh mass) (Bq/kg-wet)}}{\text{Water Activity Concentration(Bq/L)}} \quad (1)$$

123 The whole-body activity of an radionuclide was estimated using a mass balance approach  
124 (Yankovich et al., 2010) to reconstruct the amount of radionuclide in the whole-body of the squid.

125 The whole-body to tissue concentration ratio (dimensionless) was estimated as:

$$CR_{WB:Tissue} = \frac{\Sigma[Tissue_t \text{ Activity Concentration (fresh mass)} \cdot Tissue_t \text{ fresh mass fraction}]}{Tissue_t \text{ Activity Concentration (fresh mass)}} \quad (2)$$

127

### 128 3. Results and discussions

#### 129 3.1. Description of *O. bartrami* specimens

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

#### 140 3.2. Activity concentrations and CRs in squid

141 The activity levels of radionuclides in Table 1 indicate that all *O. bartrami* size classes had  
 142 accumulated radionuclides from Fukushima Daiichi NPP releases as indicated by Cs-134 and Ag-  
 143 110m. The squid specimens had a strong capability to concentrate Ag in their bodies. The maximum  
 144 activity of Ag-110m in the whole body of *O. bartrami* was up to 9 Bq/kg, as compared to that in  
 145 water which was below the MDA of 0.22 Bq/m<sup>3</sup>, indicating a maximum concentration factor that is  
 146 higher than 4×10<sup>4</sup>. The mean CRs for Ag-110m were calculated as >2.95×10<sup>4</sup> ± 9.84×10<sup>3</sup> (Table 2),  
 147 using the MDA as the activity of seawater in Equation (1).

148 Although this estimate is with large uncertainties because of using MDA of Ag-110m as the water  
 149 concentration, these Ag data provide new insights for international researchers and fill a gap as the  
 150 relevant international database (Wildlife Transfer Parameter Database;  
 151 [www.wildlifetransferdatabase.org](http://www.wildlifetransferdatabase.org)) and IAEA Technical Reports Series No.422 which ~~has have~~  
 152 entrees for Ag uptake in the mollusk category (3.6×10<sup>4</sup> and 6×10<sup>4</sup>, respectively), but none  
 153 specifically for squid/cephalopods.

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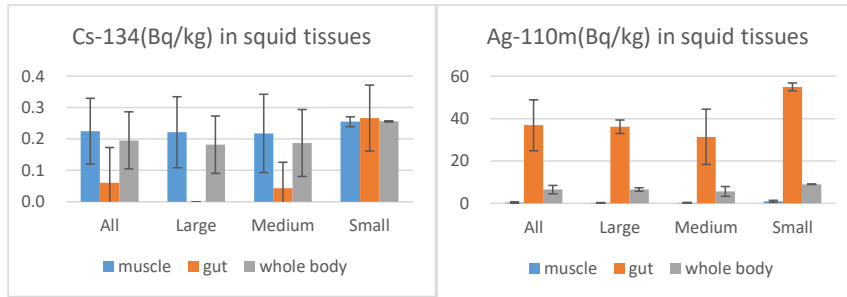
154 The mean  $CR_{WB}$  values for Cs-134 and Cs-137 in *O. bartrami* were 6.33 ( $\pm$  2.80 S.D.) and 5.57  
155 ( $\pm$ 2.59 S.D.) respectively. These values are similar to previously published mean concentration  
156 factors for Cs in cephalopods ranging from 9 to 14 (IAEA, 1978;Ishii et al., 1978;Suzuki et al.,  
157 1978;IAEA, 2004). The slightly lower  $CR_{WB}$  in this study is well within the range of expected  
158 variation, which can be very high for water-to-organism CR values (e.g. reported CRs for Cs-137  
159 in marine fish range over nearly an order of magnitude) (Beresford, 2010). The activity  
160 concentration of  $^{137}Cs/^{134}Cs$  in the research area reached a maximum of  $\sim$ 600 Bq  $m^{-3}$  in June 2011  
161 and soon decreased to below 100 Bq  $m^{-3}$  (Aoyama et al., 2016). Considering the temporal change  
162 of radiocesium in seawater and its relatively short biological half-life ( $\sim$ 70 days) in marine  
163 organisms, in this study, the CR calculation used mean Cs-134 and Cs-137 seawater activity  
164 concentrations (35.1 and 36.2 Bq  $m^{-3}$  respectively) from this study's November 2011 sampling,  
165 which were similar to the  $\sim$ 50 Bq  $m^{-3}$  reported for July-December timeframe from the same open  
166 ocean area (Kaeriyama, 2017).

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

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

183 term accumulation.

184 The levels of activity for  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{54}\text{Mn}$  and  $^{65}\text{Zn}$  in the samples were all below the MDA (0.22  
185 mBq/g-ash).



186

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



Table 1 Statistics of radionuclides' levels in composite samples (Bq/kg-fresh mass)

Size	Tissues	Cs-137		Cs-134		Ag-110m		K-40		Ra-226		U-238	
		Range	Average	Range	Average	Range	Average	Range	Average	Range	Average	Range	Average
All (n=13)	M	0.10-0.46	0.27±0.12	0.06-0.39	0.22±0.10	0.06-1.29	0.36±0.33	56.29-94.80	76.05±9.40	nd-0.07	0.03±0.03	0.16-1.77	0.59±0.44
	G	nd-0.33	0.05±0.10	nd-0.34	0.06±0.11	8.10-56.27	36.85±12.02	9.72-72.37	53.03±15.77	nd-0.89	0.28±0.28	nd-26.89	5.40±7.60
	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	53.67-88.09	72.13±8.45	nd-0.17	0.07±0.05	0.27-5.32	1.35±1.40
Large (n=5)	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	nd-0.07	0.04±0.03	0.33-0.94	0.63±0.26
	G	ND	ND	ND	ND	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
	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	nd-0.15	0.07±0.06	0.54-2.09	0.97±0.63
Medium (n=6)	M	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
	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	3.05±3.84
	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	0.86±0.80
Small (n=2)	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	1.09±0.97
	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	19.45±10.53
	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	3.76±2.19

\* Tissues: M – muscle, G – gut, WB – whole body. \*\*ND: level was below the minimum detectable activity.

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  
190 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).

195 The whole-body:muscle and whole-body:gut concentration ratios for radionuclides in squid  
196 samples are listed in Table 2. For many radionuclides, the tissue-specific concentration values  
197 for the small squids tend to be higher than those for large squids. The uncertainty of the whole-  
198 body:gut CRs for Cs-137 and Cs-134 are relatively high because of the relatively low level and  
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.

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Table 2 Concentration ratios for radionuclides in 2011 conditions following the accident (see text).

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

207 \* CR: WB-M is whole-body to muscle concentration ratios, WB-G is whole-body to gut concentration ratios, and WB-W is whole-body to water concentration ratios.

208 \*\* NA: Data is not available because radioactivity of specific radionuclides in at least one tissue was below MDA.

209 \*\*\* Values were calculated using mean Cs-134 and Cs-137 seawater activity concentrations of 35.1 and 36.2 Bq m<sup>-3</sup>, and the MDA of <sup>110m</sup>Ag in seawater (0.22 Bqm<sup>-3</sup>).

210 **3.4. Dose assessment results**

211 **3.4.1. Dose rates for squid**

212 The internal radiological dose rates to squid from artificial radionuclides ( $^{110m}\text{Ag}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ )  
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  
215 surrounding seawater. The internal dose rates from FDNPP-associated artificial radionuclides were  
216 lower, by two orders of magnitude, than those from the natural radionuclides measured in this study.  
217 From these radionuclides, only approximately 1.4 % of the total dose rate is estimated to have come  
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  
219 radionuclides, and increases to approximately  $0.61 \mu\text{Gy}\cdot\text{h}^{-1}$  when adding Po-210 a natural  
220 radionuclide and significant dose contributor in marine organisms (using a conservative generic  
221 marine value of  $15 \text{ Bq kg}^{-1}$ -fresh mass and  $0.001 \text{ Bq L}^{-1}$  in squid and seawater, respectively based  
222 on (Carvalho, 2011) and (Hosseini et al., 2010). These dose rates are much lower than the most  
223 conservative screening benchmark dose rate of  $10 \mu\text{Gy}\cdot\text{h}^{-1}$ (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.

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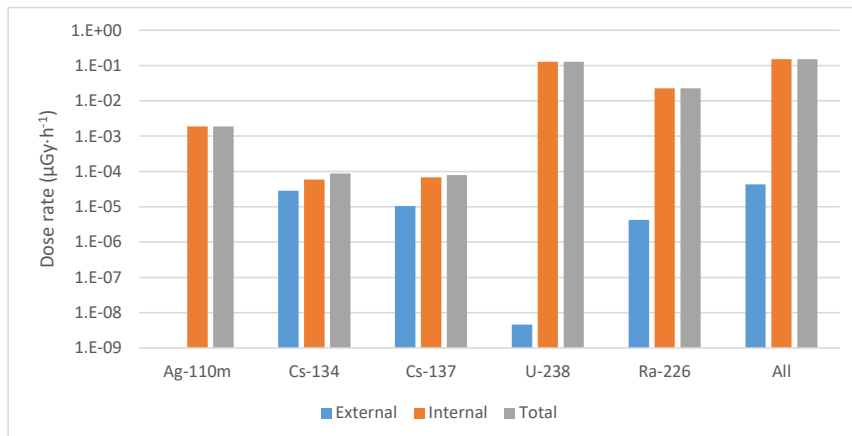


Fig 3 Dose rates ( $\mu\text{Gy}\cdot\text{h}^{-1}$ ) from measured radionuclides for squid samples

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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  
 236 dose of 0.01 mSv (median; minimum = 0.007 mSv, maximum = 0.014 mSv) would have occurred  
 237 to a hypothetical human consumer of 20 kg yr<sup>-1</sup> of squid from the study area (based on squid  
 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  
 243 Daiichi NPP. This is consistent with previous findings that natural radionuclides provided far greater  
 244 dose rates to potential consumers of Pacific tuna (Fisher et al., 2013), and even for seafood sourced  
 245 within a few kilometers of the Fukushima Daiichi NPP in 2013 (Johansen et al., 2015). The dose  
 246 contribution from the Fukushima Daiichi NPP releases for squid consumption of this study are far  
 247 below the 1 mSv per year recommended constraint for prolonged exposure by the public from  
 248 nuclear facility releases (ICRP, 1999).

249

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

minimum	median	maximum	% this study*
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<b>K-40</b>	6.98E-06	9.43E-06	1.18E-05	3.12%
<b>Ag-110m</b>	3.36E-09	2.02E-08	7.22E-08	0.01%
<b>Cs-134</b>	2.28E-08	8.36E-08	1.48E-07	0.03%
<b>Cs-137</b>	2.60E-08	7.02E-08	1.20E-07	0.02%
<b>Ra-226</b>		1.68E-07	3.92E-07	0.06%
<b>U-238</b>	1.44E-07	5.31E-07	1.59E-06	0.18%
<b>Po-210**</b>	1.44E-05	2.92E-04	1.08E-03	96.59%

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.*  
 257 *bartrami*) samples collected at NW Pacific in November 2011. This study filled a gap in  
 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  
 260 in squid was found as high as  $4 \times 10^4$  L/kg in the smallest samples, with a mean value of  $2.95 \times 10^4$   
 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

#### 267 **Acknowledgement**

268 This study was partially supported by the Scientific Research Foundation of Third Institute of  
 269 Oceanography, SOA (2015010), the Northwestern Pacific Marine Environmental Monitoring  
 270 Project, ~~the International Organizations and Conferences Project of the State Oceanic~~  
 271 ~~Administration of China~~, the Coordinated Research Project (CRP K41017) and Regional  
 272 Cooperative Agreement Project (IAEA/RCA RAS7028) of the International Atomic Energy Agency  
 273 (IAEA), ~~the National Key Scientific Instrument and Equipment Development Project~~  
 274 ~~(2016YFF0103905)~~, ~~the International Organizations and Conferences Project of the State Oceanic~~  
 275 ~~Administration of China~~, and the Public Science and Technology Research Funds Projects of Ocean  
 276 (201505005-1).

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