Referee #3 comments and responses:

The study investigates the radionuclides (both natural and artificial ones) in the neon flying squids from east Japan following the Fukushima disaster in 2011. It has merits and deserves publication but some points need to be modified before its acceptance. In particular, the calculations of internal doses and human exposure for polonium should be based on studies dedicated to squid as well to avoid biased estimations (see below). Also, the ms should have been prepared with more care as there are many mistakes all along `the text which should have been avoided by a careful reading.

Response: The authors thank the anonymous reviewer for their useful comments. We largely agree with the points raised and revised the manuscript accordingly.

Specific points: The title suggest a general approach on "squid" but only one single species is used in the study. I suggest to modify the title by including the name of the squid as follow "Artificial radionuclides in the neon flying suid Ommastrephes bartramii from. . .."

Response: The title of was changed into "Artificial radionuclides in neon flying squid from northwestern Pacific in 2011 following the Fukushima accident".

Line 10. The correct name of the species is Ommastrephes bartramii. This is to be changed consistently throughout the ms. Also specify here "neon flying squid"

Response: The text was changed throughout to "*Ommastrephes bartramii*". The common name of the species "neon flying squid" was also specified here.

Line 14. It should be easier for the readers to write 2.9 104. This should be used consistently throughout the ms.

Response: In line 15-16, the format of the figure was revised as 2.9×10^4. Similar changes were made in Table 2 and Line 121-122.

Line 22. I agree that cephalopods constitute an important commercially group but here you considered only one single species and it may be somewhat tricky to extrapolate the present *result to the whole group, especially to nectobenthic species (cuttlefishes) and to coastal benthic ones (octopuses). Do you believe that similar results are to be found for such Orders?*

Response: We are aware the ERICA-Tool has an option for using transfer parameters from similar species, and of Jeffree et al. 2013 that explores the similarity of transfer parameters among related species. However, this study did not produce sufficient data to test these topics. Therefore, we do not suggest our results be extrapolated to other species, especially those in highly different environments such as shallow coastal benthic octopus species. Upon review of the text, we found no such extrapolations, including line 22 which simply states that our results add to the scarce data on open-ocean organisms.

We compare tissue distribution data against those from another free swimming cephalopod cuttlefish, but take care to avoid a suggestion that squid transfer parameters should be extrapolated to cuttlefish.

Line 32 and Line 38. The years are missing for the references.

Response: Years of the citation were added. The text of references was also updated.

M&M. Where the sexes considered when grouping the individuals? Sexual dimorphism occurs in this species so it can results in grouping individuals of similar size/weight but with different ages. How did you manage this?

Response: (We assume here the question is “Were” (not “Where”)). The main purpose of the paper was to report dose rates (to seafood consumers and squid). The study found these doe rates to low relative to benchmarks, and therefore, it was not necessary to explore male/female

differences. Although not essential to this study, we agree it is an interesting topic, and could be investigated further in a future study.

Line 72. Gut tissues is very vague and seems to mainly refer to organs and tissues involved in the digestive processes. If this is true, it means that other tissues such as the gills, heart, gonads and associated glands were not considered. Can you please clarify?

Response: "Gut tissue "has now been clarified (lines 78-79).

Line 74. Define HPGe here and remove it at Line 78. Lines 79-80. Detection efficiencies for the other radionuclides should be also provided here. Line 112. "yr-1" . Line 118 and Line 119. Spaces are missing before and inside the references. Please prepare you ms with more care. Line 124. "activity of a radionuclide"

Response: For all of the above, the text was revised accordingly.

Line 127. As for CRWB:water, define CRWB:Tissue

Response: We improved and clarified both descriptions with more information.

Lines 130-140. This paragraph should move to the M&M section: it is not "results" but just a description of the sampling which was missing in the M&M section.

Response: While some of these lines could be moved to the methods section, most of this paragraph is interpretation of data and we prefer the entire paragraph to remain here as it includes discussion and begins a flow of logic that connects to subsequent discussion text.

Page 6. The table is a duplicate of Table 1 page 9. Remove it from page 6.

Response: The table appeared on page 6 by mistake and has been deleted.

Line 147. Do you mean independently of the size classes?

Response: The word "maximum" implies "for all size classes." However, we have added text to clarify.

Line 160. CF factor has been determined experimentally for cuttlefish by Bustamante et al. 2006 in JEMBE with lower values than reported here.

Response: As described above, we have made some tissue distribution comparisons with another cephalopod cuttlefish, but have not compared our open-ocean squid CR data with laboratory-derived cuttlefish CR data. They are two different species, with different diets. But also, laboratory data often under predict CR values due to relatively short exposure times compared with real world conditions, and due to the difficulty of replicating real-world diet pathways in the laboratory. There are multiple factors that can make open-ocean vs laboratory CR data different, as well as the CRs from two species different. While possible, and interesting, such a topic was not in our objectives, and therefore we have chosen to not add a lengthy discussion on an important, but tangential topic. It is a good idea for another paper.

Line 171. Change Bustamante et al 2004 (dedicated to Ag and Co) by Bustamante et al. 2006 (dedicated to Cs and Am).

Response: The range of previous results of Cs in cephalopods was changed into 2–14, with the citation of Bustamante et al 2006.

Line 174. One important aspect is that the digestive gland is the storage tissue independently of the exposure pathway (food or seawater).

Response: We don't disagree. And the point the referee mentions shows a value of laboratory studies where diet vs water exposures can be controlled. But, in this open-ocean study we could not test this question. It would seem somewhat of a reach to include it as a conclusion in this

paper.

Line 175. Is this significant?

Response: Yes, it is ($P<0.05$, in t-test).

Line 179. Provide a reference. Line 180. Add Bustamante et al. 2006 as a reference for Cs.

Response: Citation of Bustamante et al., 2004 was changed into Bustamante et al., 2006. Same change in Line 195.

Page 9, Table 1. "Statistics" in the title is not appropriate here; there is no statistics in this table but activities of the radionuclides only. For "small individuals", means and standard deviation have been calculated with only 2 individuals, which is not fully correct.

Response: The title of Table 1 was changed into "Radionuclides levels in composite samples". The "n" numbers in this table is the number of composite samples.

*Page 11. *** is not applied to Cs, so it should be limited to Ag.*

Response: Line 226-227: Text was added to clarify the calculation for the values of WB-W for Cs-134, Cs-137 and Ag-110m.

Line 216. The value of 15Bq/kg seems a bit high compare to what it is found for muscle in squids. In the cited review (Carvalho 2011) , the value is 1.61 Bq/kg wwt, so I guess you took the wrong value in the table. See also for example Waska et al 2008 in STOTEN who reported 5.7 Bq/kg dry wt (so approx. 5 times less when expressed relatively to the fresh weight) in the squid Todarodes pacificus from the Japan Sea. Also, Heyraud et al. 1994 reported values of 15 to 21 Bq/kg dry wt (so between 3 to 4 in wet wt) in Loligo vulgaris from South Africa. Revise your dose calculation accordingly.

Response: The comment encouraged us to add text that clarifies our approach. The Po-210 value of 15 Bq/kg was selected purposefully. The astute reviewer is correct that it is higher than the average of the available data. As explained in the text, it is being used here as a conservative value

in dose calculations. By conservative, we mean it is representative of the upper portion of the available data. This approach is typical in dose assessments. If we used an average value, as suggested, it would ignore the upper 50% of potential dose rates, and could lead to an erroneous result when comparing with benchmarks. We could add dose rates for the average value Po-210, and a low value as well. However, Po-210 is not the focus of the study. It is being presented here simply to provide a context for the FDNPP-related radionuclides, and use of a conservative value is appropriate data for such context. We have clarified the text accordingly.

Line 231. Do you mean "0.010 mSv"?

Response: The figure of "0.01 mSv" was changed into "0.010 mSv" to make the significant digits constant.

Line 234-243. Calculations to be revised according to relevant Po values.

Response: See previous response (two above). For human dose rates, we also do not want to use an average Po-210 value as it is not conservative. Using the average under predicts 50% of potential dose rates. We use a higher value representative of the upper portion of the data as described above, which is appropriate given we are using the Po-210 dose rate simply for context here. The comment has encouraged us to improve the text on this topic.

References. The bibliographic references should be homogeneous. For example, Line 276, the *journal title is not in full as for the other references.*

Response: The bibliographic references were updated.

Artificial ~~Radionuclides~~radionuclides in ~~Squid~~neon flying squid from the northwestern Pacific in 2011 following the Fukushima accident

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Abstract:

In order to better understand the impact of the Fukushima Daiichi Nuclear Power Plant (~~NPP) Accident~~FDNPP) accident on a commercial marine species, neon flying squid (~~*Ommastrephe bartrami*)~~*Ommastrephes bartramii*), samples~~,~~ obtained from the northwestern Pacific in November 2011~~,~~ were analyzed for a range of artificial and natural radionuclides (Cs-134, Cs-137, Ag-110m, U-238, Ra-226 and K-40). Short-lived radionuclides Cs-134 and Ag-110m released from the Fukushima Nuclear Power Plant (NPP ~~Accident)~~) accident were found in the samples, with an extremely high water-to-organism concentration ratio for Ag-110m (> 2.~~9E+04). The radiological~~9 $\times 10^4$). While accident-derived radionuclides were present, their associated dose rates for the squid ~~from the radionuclides measured~~ were far lower than the relevant benchmark of 10 µGy h^{-1}. For human consumers ingesting these squid, the dose contribution from natural radionuclides ~~(>99.9%)~~, including Po-210, was far greater (>99.9%) than that of Fukushima-accident radionuclides (<0.1%). The whole-body to tissue and whole-body to gut concentration ratios were calculated and reported, providing a simple method to estimate the whole-body concentration in ~~the~~ environmental monitoring programs, and filling ~~the~~a data gap ~~of~~for concentration ratios in cephalopods. Our results help fill data gaps on uptake of NPP radionuclides in the commercially important Cephalopoda class and add to scarce data on open-ocean nekton in the northwestern Pacific ~~soon~~shortly after the Fukushima accident.

Key words:

Fukushima NPP Accident; squid; concentration ratios; radiological dose; Silver-110m.

1. Introduction

The Fukushima Daiichi Nuclear Power Plant (NPP) Accidentaccident, which was caused by the combined effect of the greatMarch 2011 earthquake and subsequent tsunami in March 2011, resulted in increased levels of artificial radioactivity in the marine environment to the east of Japan (IAEA 2015).(IAEA, 2015). The radioactive releases, dominated by radiocesium, were transported eastwards in surface water across the mid-latitude North Pacific withat a speed of 3-7 km day^{-1} (3.5-8.0 cm s^{-1}) and dispersed widely in the North Pacific within a few years (Aoyama et al., ;Smith et al.), raising(Aoyama et al., 2013; Smith et al., 2015), which raised concerns about the potential impact 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 toon both human and marine species. However, most studies have focused on the concentration of radiocesium in fish (Johansen et al., 2015),(Johansen, 2015; Wada, 2016), and only a few publications have reported on radionuclides in other marine species (Buesseler et al., ;Yu et al.).(Buesseler et al., 2012; Yu et al., 2015). Few data are available for open-ocean locations as compared withto 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.

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

This study assessesassessed samples of *O. bartrami**bartramii* 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~~for human consumers of squid~~ seafood~~. Consistent with international efforts to compile transfer data, ~~Concentration Ratios (wholebody to~~ concentration ratios (whole-body to water and ~~wholebody~~ whole-body to- tissue) ~~are~~were calculated and reported, including those for different age classes of squid.

2. Materials and methods

2.1. Sample collection and analytical procedure

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

Squid samples were dissected after thawing into muscle and gut tissues ~~after thawing,~~(other soft tissues, including the digestive tract, gills, heart, gonads and associated glands), dried at 50 °C, and then 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 was then subjected to ~~HPGe~~a planar HPGe (High Purity Germanium) spectrometry for detection of gamma-emitting radionuclides.

Gamma rays from artificial radionuclides (Cs-134, Cs-137, Ag-110m, Co-58, Co-60, Mn-54 and Zn-65) and natural radionuclides (K-40, Ra-226 and U-238) were analyzed using a planar ~~high-purity germanium (~~HPGe~~)~~ detector (Model BE6530 with Multi Channel Analyzer Lynx system;

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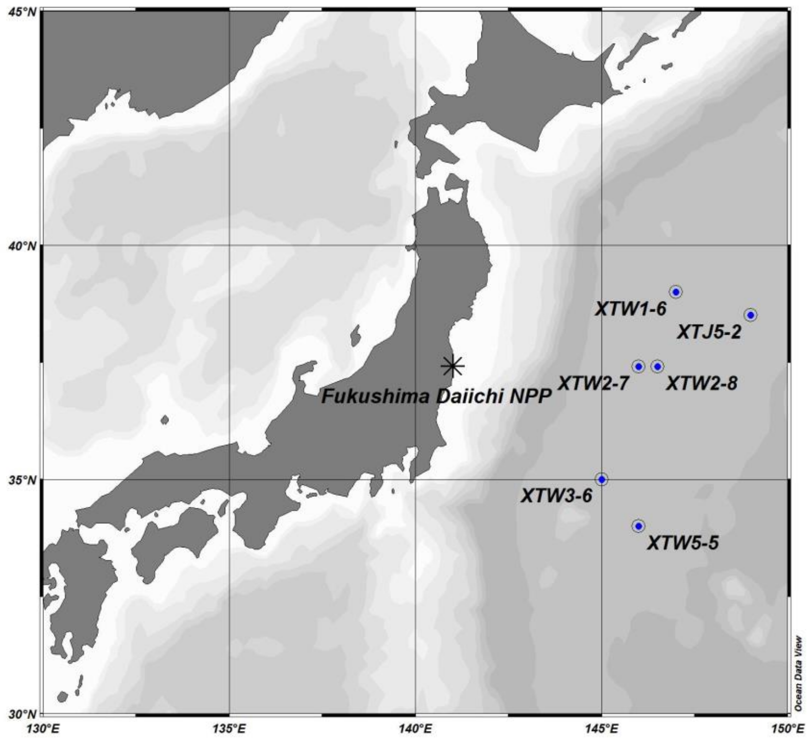
Canberra, U.S.A.). Detection efficiencies for the geometry used were 2.~~7885~~ 75%, 2.72%, 2.56%, 2.15%, 1.51%, 2.10%, 1.65%, 1.46%, 4.30% and ~~2.4476~~ 8.52%, for Cs-134, Cs-137 ~~and~~, Ag-110m, Co-58, Co-60, Mn-54, Zn-65, K-40, Ra-226 and U-238, respectively. The counting time for each sample was 24 hr. Genie 2000 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 hydrogen explosion occurred in Unit 1 of the FDNPP ~~(Wakeford).~~(Wakeford, 2011).



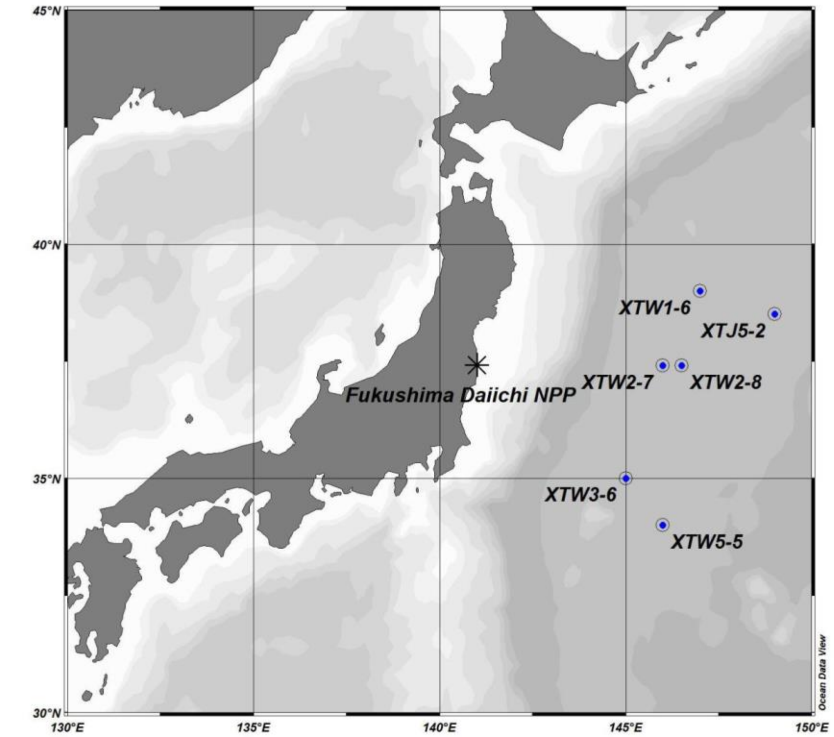


Fig 1 Map of sampling sites

2.2. Dose assessment for squid

The ERICA Assessment Tool (version 1.2) ~~(Brown et al., 2008)~~(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~~has the capability to specify organism sizes, and in this study, average mass (1.3 kg) and dimensions (ellipsoid equivalent of 0.~~3m~~3 m, 0.~~1m,~~1 m, and 0.~~085m~~085 m for length, width, and height, respectively) from the specimens were used to calculate dose rates. The dimensions of the average *O.* ~~*bartrami* happen to be~~*bartramii* are very similar to the standard ERICA "pelagic fish" and therefore, the dose rates ~~are very similar~~ , as calculated by ERICA, are also similar. The measured activity concentrations in the whole-body of ^{137}Cs, ^{134}Cs, ^{110m}Ag, ^{226}Ra and ^{238}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*~~*bartramii* are migratory, their radionuclide tissue levels represent an integrated accumulation from recently traversed areas in the open ocean~~ area. The~~. Since the exact migratory routes are ~~not known. Therefore~~unknown, the external dose rates to the squid were calculated using the average of water radioactivity levels in the study capture region (average of samples across all sampling locations). ~~Use of~~In this instance, using the average is reasonable ~~in this instance as~~because the external dose rates for artificial radionuclides were much ~~smaller~~lower than the internal dose rates~~ and therefore~~. As a result, variable water activity concentrations had little influence on overall ~~dose results~~dosages. For internal dose rates ~~to~~for squid, the dose conversion coefficients (DCCs) were calculated within the ERICA tool (supplemental). The occupancy factors were 100% in water, and weighting factors of internal low beta, internal beta/gamma and internal alpha were set as 3, 1 and 10, respectively.~~–~~

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2.3. Dose from ingesting squid ~~by human consumers~~

Committed effective doses (Sv) ~~to~~for human consumers of squid were estimated using standard exposure-to-dose conversion factors (DCFs) for ingestion from the ICRP Compendium of dose coefficients based on ICRP Publication 60 ~~(ICRP, 1999).~~(ICRP, 1999). Key DCFs are 1.~~30E-08~~30 $\times 10^{-8}$ and 1.~~90E-08~~90 $\times 10^{-8}$ Sv Bq^{-1} for Cs-137 and Cs-134, respectively (DCFs provided in the supplemental material). The factors are multiplied by intake (e.g~~.~~., kg yr^{-1}) to obtain committed effective doses ~~to~~for the consumer. In this study, the annual intake rate of seafood by an adult consumer is assumed to be 20 kg yr^{-1} (consistent with world per capita fish and related seafood consumption ~~(FAO, 2016).~~(FAO, 2016)). As a conservative assumption, the entire 20 kg yr^{-1} for a hypothetical consumer is assumed to be sourced from the squid of the study area east of the Fukushima Daiichi NPP (in practice, only a small percentage of a seafood diet would be sourced from this region). As most dose to human consumers of seafood typically comes from the natural radionuclide Po-210 (~89~~%;(Johansen et al., 2015)),~~% (Johansen et al., 2015)), the seafood ingestion dose rates here ~~included~~were compared with and without Po-210 ~~using~~to provide a context of the relative influence of Fukushima NPP accident radionuclides. For this comparison, a generic Po-210 seafood value of 15 Bq kg^{-1} was used based on Hosseini et al. (2010) and consistent with the conservative ~~generic~~ (lognormal 95th percentile) based on the limited squid data ~~for marine seafood(Carvalho, 2011;Hosseini et al., 2010).~~in (Carvalho, 2011; Heyraud et al., 1994; Waska et al., 2008).

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2.4. Whole-body concentration ratios

The water-to-organism whole-body ~~Concentration Ratio~~ concentration ratio (CR_{WB} ~~(in L/kg~~$_{:Water}$) used here is defined as:~~.~~

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

The whole-body activity of ~~an~~a radionuclide was estimated using a mass balance approach ~~(Yankovich et al., 2010)~~(Yankovich et al., 2010) to reconstruct the amount of radionuclide in the whole-body of the squid. The whole-body to tissue concentration ratio (~~dimensionless~~$CR_{WB:Tissue}$) was estimated as:

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

3. Results and ~~discussions~~discussion

3.1. Description of *O.* ~~*bartrami*~~*bartramii* specimens

In total, 98 specimens were obtained from 6 stations. The mass of the specimens ranged from 118 g to 2551 g, ~~on~~with an average of 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 of the specimens had a length greater than 290 mm (adult size~~)~~), suggesting that the majority of the specimens ~~were~~ hatched in the winter of 2010 or spring in 2011 and had been living for 8 to 11 months ~~(Wang and Chen, 2005).~~(Wang and Chen, 2005). Combining the estimated age of the squid, and assuming residence in the general ~~area~~region east of Fukushima Prefecture, it can be inferred that most specimens had been accumulating radionuclides since the Fukushima Daiichi NPP accident~~, while~~. However, a minor proportion (the small size category) ~~were likely to~~may have ~~been~~ hatched after the accident and had shorter exposure times.

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3.2. Activity concentrations and CRs in squid

The activity levels of radionuclides in Table 1 indicate that all *O.* ~~*bartrami*~~*bartramii* 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~~*bartramii* reached 9 Bq/kg, as compared to that in water, which was below the MDA of 0.22 Bq/m^3, indicating a maximum concentration factor ~~that is~~ higher than 4×10^4~~.~~ for all size classes. The mean CRs for Ag-110m were calculated as > 2.95×10^4 ± 9.84×10^3 (Table 2), using the MDA as the activity of seawater in Equation (1).

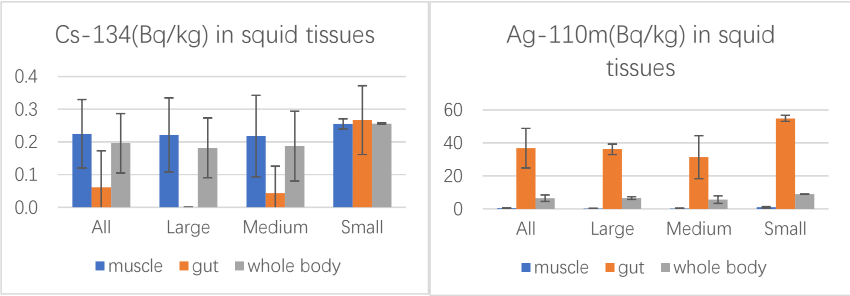
Although this estimate ~~is with~~contains large uncertainties because of using MDA of Ag-110m as the water concentration, these Ag data provide new insights for international researchers ~~and~~. Additionally, they fill a gap, as the relevant international database (Wildlife Transfer Parameter Database; ~~www.wildlifetransferdatabase.org)~~www.wildlifetransferdatabase.org) and IAEA Technical Reports Series No.422 ~~which~~ have entrees for Ag uptake in the mollusk category (3.6×10^4 and 6×10^4, respectively), but none specifically for squid/cephalopods.

The mean CR_{WB} values for Cs-134 and Cs-137 in *O. ~~bartrami~~bartramii* were 6.33 (± 2.80 S.D.) and 5.57 (±2.59 S.D~~.)~~.), respectively. These values are similar to previously published mean concentration factors for Cs ~~in cephalopods~~ ranging from ~~9~~2 to 14 ~~(IAEA, 1978;Ishii et al., 1978;Suzuki et al., 1978;IAEA, 2004).~~ in cephalopods (Bustamante P, 2006; IAEA, 1978; Ishii et al., 1978; Suzuki et al., 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 in marine fish range over nearly an order of magnitude) ~~(Beresford, 2010).~~(Beresford, 2010). The activity concentration of ^{137}Cs in the research area reached a maximum of ~600 Bq m^{-3} in June 2011 and soon decreased to below 100 Bq m^{-3} ~~(Aoyama et al., 2016).~~(Aoyama et al., 2016). Considering the temporal change 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 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 ocean area (Kaeriyama, 2017).~~ the same open ocean area July-December timeframe (Kaeriyama, 2017).

The results also showed that both Cs-134 and Cs-137 were concentrated mainly in the muscle of the 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 laboratory results of 80~~+ % accumulation in the muscle and head of cuttlefish after only 8 hours of exposure to water (Bustamante et al., 2004). In contrast, for Ag, the open ocean squid had 95 % Ag in the gut vs muscle.~~ +% accumulation in the muscle and head of cuttlefish after only 8 hours of exposure to water (Bustamante et al., 2004; Bustamante et al., 2006). In contrast, for Ag, the open ocean squid had 95% Ag in the gut versus muscle. This result was also consistent with the laboratory cuttlefish which had 98% Ag in the gut following a single spiked feeding and 29 d depuration ~~(Bustamante et al., 2004). From the same study, within the gut, accumulation of Ag is dominantly in the digestive gland.~~ (Bustamante et al., 2004). From the same study, within the gut, accumulation of Ag is dominant in the digestive gland.

The smallest squid samples had the highest concentration factors for Cs-134, Cs-137, Ag-110m and U-238 (Fig 3). ~~The~~Despite their inferred shorter exposure times (shorter lifespan), the higher accumulation occurred in the smaller size class ~~despite their inferred shorter exposure times (shorter lifespan) as~~ compared ~~with~~to the larger size class. These results are consistent with observed Cs depuration rates in ~~juveniles~~juvenile cephalopods (*Sepia officinalis)* being ~four times slower than that of adults, ~~with however,~~but both ~~being~~ relatively fast (adult cuttlefish have a biological half-life of 16 days for Cs and 9 days for Ag ~~(Bustamante et al., 2004)).~~(Bustamante P, 2006)). This previous study~~,~~ suggests the radiocesium accumulation and depuration in *O.* ~~*bartrami*~~*bartramii* is relatively rapid and that our results ~~therefore~~ primarily reflect recent (~ several months) exposure rather than longer-term accumulation.

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



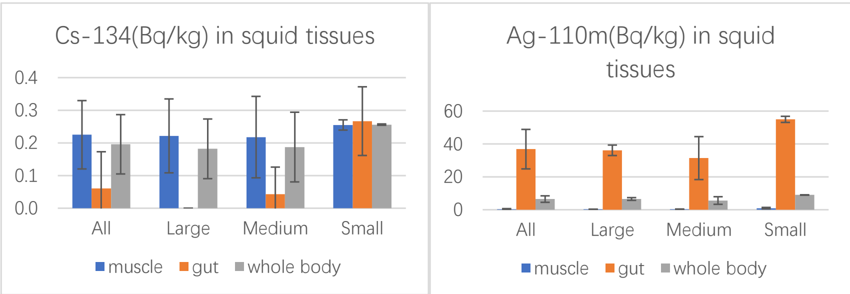


Fig 2 Activity concentrations of Cs-134 (left) and Ag-110m (right) in squid tissues.

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Table 11 Statistics of radionuclides' Radionuclide 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.

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3.3. Whole-body~~:~~ to muscle and whole-body~~:~~ to gut concentration ratios

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).~~(Brown et al., 2008; DOE, 2004). However, muscle tissue (vs. whole-body) is measured in most monitoring programs, which typically focus on seafood tissues consumed by humans. Therefore, there exists a need for whole-body~~:~~ to tissue concentration ratios that allow for estimation of whole-body concentrations from commonly measured tissue data ~~(Yankovich et al., 2010).~~ (Yankovich et al., 2010).

The whole-body~~:~~ to muscle and whole-body~~:~~ to gut concentration ratios for radionuclides in squid samples are listed in Table 2. For many radionuclides, the tissue-specific ~~concentration values~~concentrations for the small squids tend to be higher than those for large squids. The uncertainty of the whole-body~~:~~ to gut CRs for Cs-137 and Cs-134 are relatively high because of the ~~relatively~~comparatively low level and large activity range of radiocesium in ~~the~~ gut samples. ~~These~~The CRs presented here are calculated for the non-equilibrium conditions following the accident. This issue is somewhat compensated for by ~~focusing on radionuclides that are taken up relatively quickly, and by~~ using the average activity concentrations that have accumulated over ~~their~~ time, albeit over the relatively short ~~lifespan~~lifespans of the squid. Equilibrium conditions are generally not achieved in natural systems, and our results, ~~like~~ all CRs should be considered in context. Further research is necessary to obtain a better estimation the biokinetics of uptake in squid and of the whole-body~~:~~ to gut CRs for Cs-137 and Cs-134.~~–~~

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
WB-M	**All**	0.93±0.28	0.94±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
	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
WB-G	**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
	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
WB-W* **	**All**	6.33±2.80	5.57±2.59	>2.~~95E+4~~95×10^4 ± 8.~~94E+3~~94×10^3	6.17±0.71	14.66±11.92	37.97±39.39
	Large	5.90±2.91	5.18±2.60	>2.~~97E+4~~97×10^4 ± 3.~~76E+3~~76×10^3	6.42±0.84	16.35±12.46	27.34±17.89
	Medium	6.30±3.17	5.33±3.04	>2.~~55E+4~~55×10^4 ± 1.~~05E+4~~05×10^4	5.89±0.69	9.56±9.40	24.11±22.43
	Small	7.52±2.52	7.29±0.06	>4.~~08E+~~08×10^4 ± 4 ~~± 4.50E+2~~.50×10^2	6.35±0.22	25.76±15.07	106.09±61.85

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

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

*** Values for Cs-134 and Cs-137 were calculated using mean Cs-134 and Cs-137 seawater activity concentrations of 35.1 and 36.2 Bq m^{-3}~~.~~, and the values for Ag-110m were calculated using the MDA of ~~^{110m}Ag~~Ag-110m in seawater (0.22 Bqm^{-3}).

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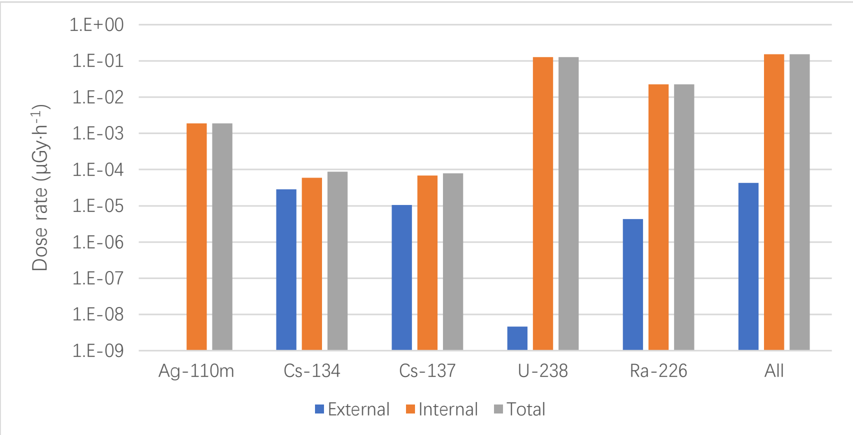
3.4. Dose assessment results

3.4.1. Dose rates for squid

The internal radiological dose rates ~~to~~in squid from artificial radionuclides (^{110m}Ag, ^{134}Cs and ^{137}Cs) were collectively much higher than the external dose rates (Fig. 4). This is consistent with the 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 lower, by two orders of magnitude, than those from the natural radionuclides measured in this study. ~~From these radionuclides, only~~Only approximately 1.4 % of the total dose rate is estimated to have come from the Fukushima Daiichi NPP releases. The total dose rate for squid is 0.15 μGy·h^{-1} from ~~study radionuclides, and increases to approximately 0.61 μGy·h^{-1} when adding Po-210 a natural radionuclide and significant dose contributor in marine organisms (using a conservative generic 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~~radionuclides measured in this study, and increases to approximately 0.61 μGy·h^{-1} when adding Po-210, a natural radionuclide significant dose contributor in marine organisms (assumes 0.001 Bq/L in seawater and a generic marine value of 15 Bq/kg- whole-body fresh mass which is consistent with a general value in Hosseini et al (2010) and with the lognormal 95th percentile of limited squid Po-210 data (Carvalho, 2011; Heyraud et al., 1994; Waska et al., 2008)). When median squid data are used (3 Bq/kg WB, FM), the total dose rate is 0.25 μGy·h^{-1}. Regardless of using the median or 95th percentile, these dose rates are much lower than the most conservative screening benchmark dose rate of 10 μGy·h^{-1}~~(Garnier-Laplace et al., 2008). The dose calculations used the measured activity concentrations in the squid (not CRs)~~ (Garnier-Laplace, 2008). The dose calculations used the measured activity concentrations in the squid (not CRs), and the calculated dose rates represent a point in time (November 2011) with likely higher doses prior to, and lower doses following, the sampling date. However, the relatively low values indicate that a more detailed (e.g.~~,~~., pulse-dynamic uptake) dose calculation~~)~~ is not necessary in this case. Overall, results indicate that the radioactive releases from the Fukushima accident would not have a significant adverse effect on *O.* ~~bartrami~~*bartramii* individuals or populations living in the study area.



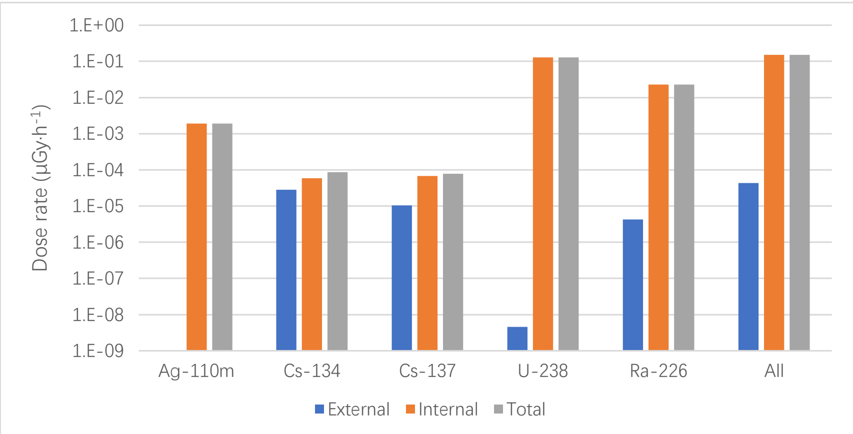


Fig 3~~3~~ Dose rates (µGy·h^{-1}) from measured radionuclides ~~for~~in squid samples

3.4.2. Dose rates for human consumers of seafood

From the radionuclides measured in edible squid tissue (muscle), a committed effective ingestion dose of 0.~~01~~010 mSv (median; minimum = 0.007 mSv, maximum = 0.014 mSv) would have occurred ~~to~~in a hypothetical human consumer of 20 kg yr^{-1} of squid from the study area (based on squid captured in November 2011). The doses calculated here are hypothetical and are intended to be conservative overestimates given the unrealistic assumption that all of the consumer's yearly seafood came from the study area. If consumption of Po-210 (from a natural background) is also included, the total dose increases to 0.30 mSv, with almost all derived from Po-210 using a conservative generic value as described above (Table 3). Of this dose (including Po-210), less than

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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 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).~~(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 contribution from the Fukushima Daiichi NPP releases for squid consumption of this study are far below the 1 mSv per year recommended constraint for prolonged exposure by the public from nuclear facility releases ~~(ICRP, 1999).~~ (ICRP, 1999).

Table 3~~.~~ Ingestion dose estimates ~~to~~for human consumers of the squid in this study (Sv y^{-1} based on 20 kg consumption of study squid).

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	minimum	median	maximum	% this study*
K-40	6.~~98E-06~~98 × 10^{-6}	9.~~43E-06~~43 × 10^{-6}	1.~~18E-05~~18 × 10^{-5}	3.12%
Ag-110m	3.~~36E-09~~36 × 10^{-9}	2.~~02E-08~~02 × 10^{-8}	7.~~22E-08~~22 × 10^{-8}	0.01%
Cs-134	2.~~28E-08~~28 × 10^{-8}	8.~~36E-08~~36 × 10^{-8}	1.~~48E-07~~48 × 10^{-7}	0.03%
Cs-137	2.~~60E-08~~60 × 10^{-8}	7.~~02E-08~~02 × 10^{-8}	1.~~20E-07~~20 × 10^{-7}	0.02%
Ra-226		1.~~68E-07~~68 × 10^{-7}	3.~~92E-07~~92 × 10^{-7}	0.06%
U-238	1.~~44E-07~~44 × 10^{-7}	5.~~31E-07~~31 × 10^{-7}	1.~~59E-06~~59 × 10^{-6}	0.18%
Po-210**	1.~~44E-05~~44 × 10^{-5}	2.~~92E-04~~92 × 10^{-4}	1.~~08E-03~~08 × 10^{-3}	96.59%

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

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

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

4. Conclusions

Elevated levels of Cs-134 and Ag-110m from the Fukushima NPP ~~Accident~~accident were found in the squid (*O. ~~bartrami~~bartramii*) samples collected ~~at~~from NW Pacific in November 2011. ~~This study filled a gap in international transfer data by providing concentration ratios for several key NPP-associated radionuclides in the~~ The whole-body ~~and tissues of cephalopods. The Concentration~~

~~Ratio~~to water CRs for Ag-110m in squid ~~was~~were found to be as high as 4×10^4 L/kg in the smallest samples, with a mean value of 2.95×10^4 L/kg in all the samples, indicating that squid was a good bioindicator for Ag-110m from the Fukushima NPP ~~Accident~~accident. The radiological dose contribution from the Fukushima Daiichi NPP releases for squid living in the study area in 2011, and for human consumers of these squid, were both far below the recommended dose limits. By comparison, natural radionuclides, particularly Po-210, ~~provide orders of magnitude greater dose rates~~provided greater dose rates by several orders of magnitude. This study filled a gap in international transfer data by providing concentration ratios for several key NPP-associated radionuclides in the whole-body and tissues of an open ocean cephalopod.

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