Distribution and Enrichment of Nuclide Cs-137 in Typical Fishery of North Pacific High Seas

Fenghua Tang* and Xuesen Cui
Key Laboratory of East China Sea & Oceanic Fishery Resources Exploitation and Utilization, Ministry of Agriculture, East China Sea fisheries Research Institute,Chinese Academy of Fishery Sciences, Shanghai 20009, China.
Email: f-h-tang@163.com.

Keywords: North Pacific high seas, radionuclide, neon flying squid, risk assessment

Abstract: Typical biological samples were analysed in central fishing grounds in North Pacific high seas for three consecutive years from 2011-2013, to understand the impact of Japan's Fukushima nuclear leakage on the high seas fishery, in order to develop the natural fishery resources of the north Pacific Ocean. The sample was dominated by Neon flying squid. It was found that all samples contained radionuclide Cs-137 by a gamma ray spectrometer and cross-check analysis was carried out among different species, organs and tissues. The distribution of nuclide in Marine organisms in the northern Pacific Ocean and its risk assessment were carried out. There were a large number of samples collected from 2011 to 2012. A Cs-137 specific activity higher than the base one was detected for the entire sample. In 2011, the nuclide was 0.05 to 6.21 $Bq•kg^{-1}$, compared with the activity range of 0.02 to 0.46 $Bq•kg^{-1}$ in 2012. In 2013, there were only two types of samples collected, and the range of activity was the base value to 0.37 $Bq•kg^{-1}$. The quality activity of the nuclides of each nutrient-grade organism was averaged, and the average of the three years was 0.49 $Bq•kg^{-1}$. The mass activity of nuclide has inverted pyramid distribution. The quality activity level of the nuclide during the three years' survey did not exceed the concentration of radionuclide quality in Chinese food and the standard line of general level. At present, the risk assessment of radioactive element Cs-137 showed that the quality activity of nuclide was in a relatively safe range, but the follow-up monitoring was needed.

1 INTRODUCTION

In March 2011, the nuclear leakage accident at the Fukushima nuclear power plant in Japan released radioactive substances into the atmosphere or deposited on land or in the ocean, which not only caused serious impacts on the surrounding land environment (Thakur et al., 2013). After the Fukushima nuclear accident, the radioactive nuclide released by the nuclear power plant not only caused an increase in the external radiation dose rate, but also affected the fishing industry (Buesseler, 2014; Inoue et al., 2015). The squid fishing ground in the high seas of the north Pacific is one of the most important ocean fisheries in China. Among them, neon flying squid (*Ommastrephes bartramii)* is the main economic variety in the area (Yamamoto et al., 2002). The central fishing ground is located in the North Pacific Ocean, where the black tide and the pro-tidal mixing zone and the mixed water eastward extend the sea surface temperature distribution densely (Fan et al., 2009). Now, it is necessary to understand and master the concentration of Cs-137 nuclide in marine organisms and the degree of hazard risk. Therefore, this research in the three consecutive years from 2011 to 2013 in the area for collection mainly related to biological utilization gamma energy spectrum analysis method to determine the specific activity radioactive substances, in Cs-137 nuclide ecological environment monitoring and risk assessment, discusses the Fukushima nuclear leak to the north Pacific high seas neon flying squid fishing grounds and the diffusion trend influence on subsequent, resources reasonable development.

Tang, F. and Cui, X.
Distribution and Enrichment of Nuclide Cs-137 in Typical Fishery of North Pacific High Seas.
In *Proceedings of the International Workshop on Environment and Geoscience (IWEG 2018)*, pages 115-119
ISBN: 978-989-758-342-1

2 MATERIAL AND METHODS OF EXPERIMENT

2.1 Sampling Method

The sampling time was divided into 2011-2013 fishing season. The data were taken from the fishing grounds of the north Pacific high seas center, and the working vessels were "Zhouyu 1301" and "Zhouyu 901", and the operation mode was light squid fishing. The biological experiment samples were recorded, including sampling biological name, sampling date, space location, etc.

The sampling area and main fishing objects were shown in figure 1. A box is shown in the center of the north Pacific high seas fisheries sampling area, located at 135°E ~165°E and 39°N ~46°N. The specific position of samples taken along the 3 years were different but from the same sampling space. The samples were taken for neon flying squid and related Marine life. All samples were frozen and brought back to laboratory for processing and testing.

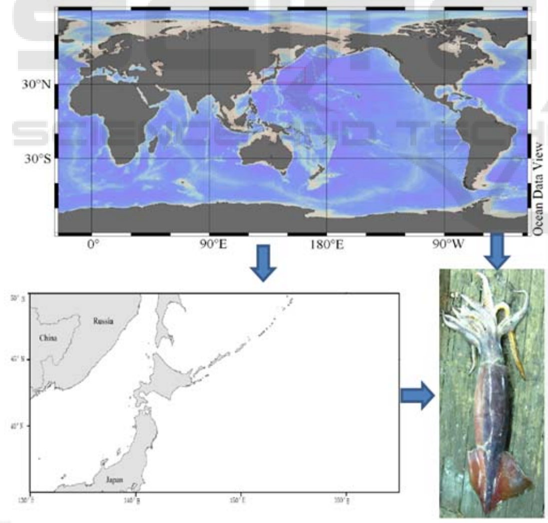

Figure 1: Sampling area in the North Pacific Ocean and an example of a sample.

2.2 Detection and Evaluation Methods

(1) Food Chain Nutrition Level

Through the marine biological sample stomach contained ingredients and feeding level analysis and biology identification, bait consists of cephalopod, fish and crustaceans. The bait of neon flying squids was found to consist of cephalopods, fish and crustaceans. And it is a prey for large fish (Wang and Pan, 2004). Studies to Neon flying squid is an intermediate nutrition class, which establishes a simple tertiary level of nutrition.

(2) Determination of Nuclide Ratio Activity

The pretreatment of the test was more complicated. The basic steps were: thawing, dissection, subdivision, and cutting, incineration, sample loading, and final weighing mark. The nuclide detection method was used to determine the sample Cs-137 ratio activity by using the High Purity Germanium passive efficiency scale. The measurement accuracy of radionuclides was as the standard reference material of IAEA-414 (IAEA, 2016). The calculation formula of activity concentration is below (Walling and Quine, 1993).

$$a = as \cdot W \cdot r / [\varepsilon(E) \cdot P \cdot m] \quad (1)$$

In the formula, a is the quality activity of Cs-137 in the sample (Bq·kg-1); as is standard source Cs-137 total peak net count rate (s-1); ε(E) is the standard source Cs-137 all-around peak detection efficiency; P is Cs-137 661.6 keV universal peak branching ratio; M for sample ash measurement (g); W is the gray fresh ratio (g·kg-1); R is the Cs-137 time decay correction coefficient.

(3) Risk Assessment Method

The Risk Assessment of the radioactive material of Fukushima nuclear accident on Marine life is based on the EU Assessment and Management of Environmental Risk from ionizing framework to assess the ecological Risk of ionizing radiation (Larsson, 2008).

3 RESULT

3.1 Detection Overview and Position Distribution of Nuclide

The detection situation of Cs-137 is shown in figure 2 in 2011-2013. The highest Cs-137 activity in 2011 was located at 154°51′E and 43°12′N, but the lowest position was the Sea of Japan Sea, in 132° 44′E and 37.85° N. The highest Cs-137 activity in 2012 was located in 160°37′and 45°23′N, but the lowest of them was the Sea of Japan, in 131°47′E and 36°50′N. The highest Cs-137 activity in 2013 was located in the vicinity of 42°N and 158°E of the fishery center.

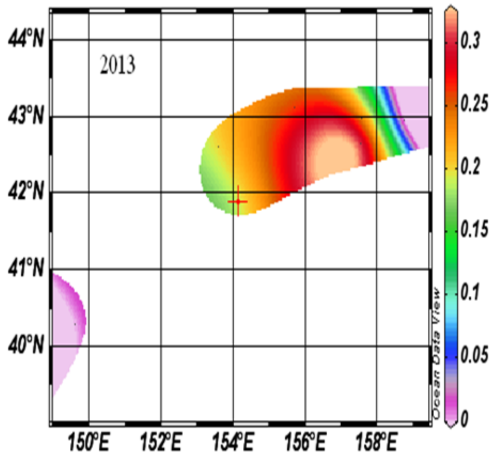

Figure 2: Spatial distribution of nuclide Cs-137 mass activity.

3.2 Distribution of Nuclide Cs-137 in Marine Organisms

Distribution of Cs-137 mass activity in different marine organisms is shown in figure 3. Among them, the activity of the shark (*Mustelus griseus*) was the highest, and the lowest was Japanese flying squid (*Todarodes pacificus*) in the Sea of Japan in 2011. In 2012~2013, the quality activity of Cs-137 was basically reduced by an order of magnitude, and the highest in 2012 was the Dolphin fish (*Coryphaena hippurus*), the lowest in the Sea of Japan. In 2013, the average quality activity Cs-137 of shark was 0.31 Bq·kg^{-1}, and average quality activity Cs-137 of neon flying squid was 0.18 Bq·kg^{-1}. These marine life samples have been collected essentially within the same fishery area.

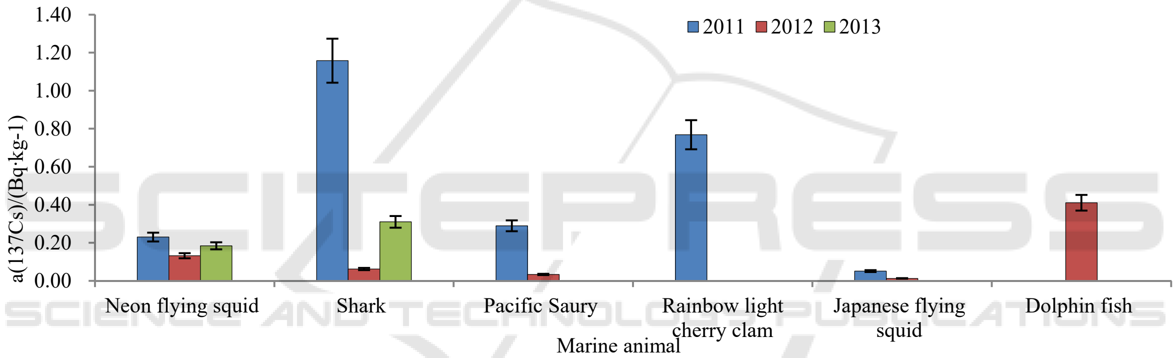

Figure 3: Distribution of nuclide Cs-137 activity in Marine organisms.

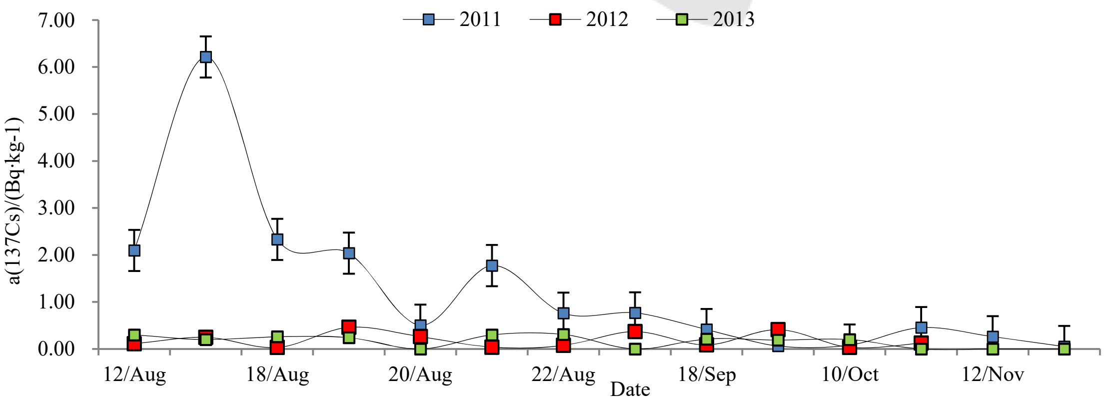

Figure 4: Distribution Change of nuclide Cs-137 with Sampling Time.

Table 1: Results of nuclide total radiation dose rates each year from ERICA tools evaluation (mGy·h^{-1}).

Biota	2011	2012	2013	Trend
Pelagic fish	7.69E-03	4.34E-04	2.32E-04	↓
Benthic fish	8.55E-04	6.77E-03	1.92E-03	↓
Phytoplankton	6.55E-04	1.82E-04	1.14E-03	↓
Zooplankton	8.22E-03	4.36E-04	8.74E-03	↓
Crustaceans	1.74E-02	1.23E-05	5.56E-04	↓
Benthic mollusks	3.35E-03	3.31E-04	2.96E-04	↓
Polychaete worms	1.24E-02	3.44E-04	4.12E-03	↓
Reptiles	4.46E-03	8.99E-04	8.24E-04	↓
Sea anemones or true corals -colony	8.14E-03	3.44E-04	1.36E-03	↓
Mammals	9.98E-03	9.16E-04	2.11E-03	↓
Macroalgae	3.44E-04	3.43E-03	2.33E-04	↓

3.3 Distribution of Nuclide cs-137 Overtime

The distribution of the Cs-137 nuclide in specific time changes is shown in figure 4. The highest quality activity of Cs-137 in 2011 sampling date was Aug. 16, and the lowest was Dec. 3. The highest quality activity of Cs-137 was October 1st, and the lowest was December 7th in 2012. The highest quality activity of Cs-137 in 2013 was Aug. 22, the lowest of which was Aug. 20. There was no obvious change in the distribution of Cs-137 with sampling time every year.

3.4 Delivery and Enrichment of Nuclide Cs-137 in the Marine Food Chainss

According to the food chain relationship of neon flying squid, the simple three-level food chain relationship was proposed, and the quality activity of Cs-137 nuclides of various nutrition-grade organisms was normalized in 2011-2013. Among them, large fish include shark and dolphin. The average of 3 years was 0.49 Bq·kg^{-1}. The average of neon flying squid was 0.18 Bq·kg^{-1}. Small fish include pacific saury and Japanese flying squid, with an average of 0.10 Bq·kg^{-1}. After 3 years of nutritional level analysis, as shown in figure 5, the quality activity of the three nutrient-level Cs-137 nuclides has inverted pyramid distribution. There were many samples used to get the average activities every year.

3.5 Risk Assessment of Radionuclides

With ERICA secondary assessment system do the filter value choice 10 mGy·h^{-1} assessments of Cs-137 for 3 years, the output under the labels are listed in table 1. All of the organisms had total dose basic within 10^{-4}~10^{-2} orders of magnitude. From 2011 to 2013, the total dose of Cs-137 in each biological group was decreasing year by year, and the maximum value was lower than 1.0 mGy·h^{-1}. As the table shows, the overall assessment of all biological groups showed a downward trend in the relative safety limits.

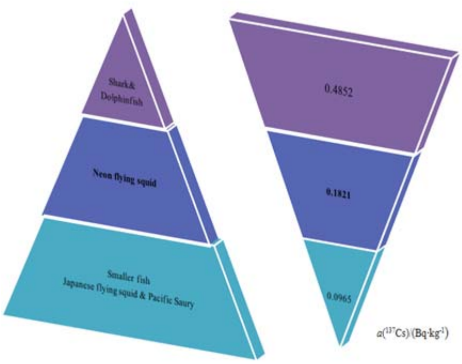

Figure 5: Distribution of activity in trophic level of average of three years in the fisheries.

4 CONCLUSIONS

Quality activities of Cs-137 in the sea area of the North Pacific Ocean were beyond the base value of the sample, according to the investigation in 2011-2013. In 2011, the average quality activity of the nuclide Cs-137 in the sample was 0.50 $Bq \cdot kg^{-1}$, and the average quality activity was 0.19 $Bq \cdot kg^{-1}$ in 2012~2013. Had not been measured quality of nuclide activity level more than China's food quality of radionuclide activity of 300 $Bq \cdot kg^{-1}$ limit concentration and the general level of 1000 $Bq \cdot kg^{-1}$ line. The test results contrast in prophase investigation showed a trend of decrease. Different intake routes, including water and food, radionuclide role of marine biological accumulation, and the marine life of radioactive nuclide concentration as well as the process of accumulation in different tissues and organs, are worthy of further research in the future.

ACKNOWLEDGMENTS

The study was Supported by National Nonprofit Institute Research Grant (2016Z01-03), Central Public-interest Scientific Institution Basal Research Fund, CAFS (NO.2018HY-XKQ0305), Open Research Fund of State Key Laboratory of Estuarine and Coastal Research (SKLEC201206).

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