Radiocesium Activity of the Seawater and Fishes in the San Jose Coast of the Lagonoy Gulf

Ian Rey I. Gubat[1], Rujyla Claire P. Cariño[2], John Rogie R. Raymundo[2*], Ryan Joseph Aniago[3]
[1]Polytechnic University of the Philippines, Sta. Mesa, Manila 1016 Philippines
[2]De La Salle University, Taft Avenue, Manila 1004 Philippines
[3]Philippine Nuclear Research Institute, Diliman Quezon City, 1101 Philippines
**Corresponding Author: john_rogie_raymundo@dlsu.edu.ph*

Abstract: The Fukushima Daiichi Nuclear Power Plant accident occurred as a consequence of the Great Tohoku earthquake on 11 March 2011. The accident resulted to a partial meltdown of the power plant reactors and the subsequent release of radionuclides, including ^{134}Cs and ^{137}Cs, both to the atmosphere and the marine environment. In this paper, seawater and six fish samples from the Lagonoy Gulf, Bicol, Philippines were analyzed for radiocesium activity using high-photon germanium gamma spectrometry. Results showed that the average activity concentration for ^{137}Cs in seawater was 0.5901 ± 0.3916 Bq/m^3, which is below the baseline activity concentration of ^{137}Cs in the Asia-Pacific region. Among the fish samples, the forktail large-eye bream has the highest ^{137}Cs activity concentration at 0.8271 ± 0.4135 Bq/kg. Activity concentration of ^{134}Cs for both seawater and the fish samples was below the lower limit of detection. Concentration factors suggest that the common dolphin fish, forktail large-eye bream and skipjack tuna bioconcentrates ^{137}Cs.

Key Words: radiocesium activity; seawater; San Jose coast

1. INTRODUCTION

On March 2011, an accident occurred in the Fukushima Nuclear Daiichi Power Plant, where partial meltdown of nuclear reactors occurred. The accident resulted to the release of radionuclides, including iodine-131, tellurium-131, strontium-90, cesium-134, cesium-137, and plutonium isotopes, around the globe through natural processes. These radioactive contaminants can cause adverse effects to the environment and ultimately to human health.

The Philippine marine environment, located in the Asia-Pacific region, is no less than vulnerable to these radiological threats. As a response, the Health Physics Research Division of the Philippine Nuclear Research Institute (PNRI), in cooperation with the International Atomic Energy Agency, initiated a project to assess the impact of the radioactive release from the Fukushima Nuclear Accident on the Philippine marine environment. Several coastal areas on the Pacific Seaboard (east coast) and the west Philippine Seaboard (west coast) in the country were selected as study areas for the project. The Lagonoy Gulf in the Bicol region, which is a part of the Pacific Seaboard, was selected by the researchers of the study. Radioactive contaminants, when spread in the marine ecosystem, can affect marine life and consequently, the human population. Thus, this study aims to determine the activity concentration of radioactive anthropogenic ^{134}Cs and ^{137}Cs in seawater and selected biota from the Lagonoy Gulf. The concentration factors of these selected contaminants will also be determined, as well as if the selected marine organisms are possible indicators of the analyzed contaminants.

2. METHODOLOGY

2.1 Sample Collection of Seawater and Biota for Radiocesium Analysis

Seawater samples were taken from three (3) different sampling points in the San Jose Coast of the Lagonoy Gulf. The exact location of the sampling points were determined using a GPS device. In each sampling site, 175 liters of surface seawater was collected in plastic gallons, which were rinsed with seawater prior to sample collection.

Six (6) different species of fish samples, big-eye tuna (scientific name), brownstripe tuna (scientific name), common dolphin fish (scientific name), forktail large-eye bream (scientific name), narrow-barred Spanish mackerel (scientific name), skipjack tuna (scientific name) were bought from the wet market, ensuring that the fish came from the study site. Approximately 3 to 7 kilograms of each species were collected and kept in cold storage prior to analysis. The wet weights of the samples were obtained. The samples were authenticated by the Zoology Division of the Philippine National Museum.

2.2 Determination of ^{134}Cs and ^{137}Cs in Seawater

Collected seawater samples were transferred into two 75-L volume-calibrated plastic containers. Each sample was acidified to pH 2-3 with dilute hydrochloric acid (HCl). A stable Cs carrier (20 mg/mL) was added to the sample solution and was stirred manually for 30 minutes. The resulting solution was equilibrated for one hour. Seventy-five grams of ammonium phosphomolybdate (AMP) powder was added to each container and was stirred manually for 30 minutes before allowing to settle overnight. The resulting solution was decanted and the residue, the CsAMP resin, was washed with 1% HCl and was collected into a one-liter wide-mouthed plastic bottle.

Prepared standard samples with known activity of ^{134}Cs and ^{137}Cs and with the same geometry were used for analysis. A high-photon germanium (HPGe) gamma ray spectrometer was used to analyze the signals during calibration and sample analysis.

2.3 Determination of ^{134}Cs and ^{137}Cs in Biota

Edible portions of fish samples were separated and cut into small pieces. The wet weight of each portion was recorded. The samples were dried in an oven at 110° and the dry weight was recorded. The samples were ground using a mortar and pestle and were transferred into geometry-counting bottles prior to subjection in gamma counting using an HPGe-Gamma spectrometer.

3. RESULTS AND DISCUSSION

3.1 Radiocesium Analysis in Fish and Surface Seawater

Table 1. Activity Concentration of ^{134}Cs and ^{137}Cs in Fish

Sample Name	Activity Concentration (Bq/kg)	
	^{134}Cs (795 keV photon)	^{137}Cs (662 keV photon)
Big-eye Tuna	<LLD (lower limit of detection)	<LLD
Brownstripe Tuna	<LLD	<LLD
Common Dolphin Fish	<LLD	0.7256 ± 0.3029
Forktail Large-eye Bream	<LLD	0.8271 ± 0.4135
Narrow-barred	<LLD	03926 ±

Spanish Mackerel		0.0718
Skipjack Tuna	<LLD	0.7325 ± 0.3053

Table 1 above shows the activity concentration of ^{134}Cs and ^{137}Cs in the six different species of fish. All the six samples showed ^{134}Cs activity concentration lower than the limit of detection. This is because the half-life of ^{134}Cs is 2.0648 years, shorter than the time between the sampling date and the Fukushima incident.

It can be observed that the forktail large-eye bream has the highest activity concentration of ^{137}Cs at 0.8271 ± 0.4135 Bq/kg, while the lowest activity concentration of ^{137}Cs was observed in the narrow-barred Spanish mackerel at 0.3926 ± 0.0718 Bq/kg. The ^{137}Cs activity concentration in brownstripe tuna and big-eye tuna were assumed to be less than their respective minimum activity (MDA), hence the activity concentration is below the lower limit of detection (LLD). The ^{137}Cs activity concentration of forktail large-eye bream, skipjack tuna, and common dolphin fish are higher than the ^{137}Cs activity concentration in fish muscle reported in ASPAMARD (2000), which is 0.50 ± 0.53 Bq/kg for wet samples. The ^{137} activity concentration of narrow-barred Spanish mackerel is below the baseline data reported in literature.

Table 2 below shows the activity concentration of ^{134}Cs and ^{137}Cs in surface seawater of the Lagonoy Gulf determined using gamma spectrometry. The ^{134}Cs activity concentration in the three sites were all below the lower limit of detection which can be accounted for by the half-life of ^{134}Cs, which is shorter than the time interval between the sample collection and the Fukushima accident. Activity concentration of ^{137}Cs in site 3 was higher than that in site 4. Site 5 showed ^{137}Cs activity below the lower limit of detection. The activity concentration values in sites 3 and 4 are lower than the average activity concentration in the Asia-Pacific region from 1990-1999 which is 2.68 ± 0.84 Bq/m^3.

Table 2. Activity Concentration of ^{134}Cs and ^{137}Cs in Surface Seawater

Sample Name	Activity Concentration (Bq/kg)	
	^{134}Cs (796 keV photon)	^{137}Cs (662 keV photon)
Site 3	<LLD	0.80 ± 0.40
Site 4	<LLD	0.69 ± 0.40
Site 5	<LLD	<LLD

3.2 Concentration factor of ^{134}Cs and ^{137}Cs in Fish

Table 3. Concentration factor of ^{134}Cs and ^{137}Cs in Fish

Sample	Concentration Factor	
	^{134}Cs	^{137}Cs
Big-eye Tuna	---	---
Brownstripe Tuna	---	---
Common Dolphin Fish	---	1.2279 ± 0.9631
Forktail Large-eye Bream	---	1.4016 ± 1.1647
Narrow-barred Spanish Mackerel	---	0.6654 ± 0.4580
Skipjack Tuna	---	1.2413 ± 0.9728

Table 3 shows the calculated concentration factor of the fish samples collected in the Lagonoy Gulf. From the results, it can be observed that the common dolphin fish, forktail large-eye bream, and skipjack tuna have concentration factors for ^{137}Cs greater than unity, which indicate that the activity concentration of the fish samples is greater than that of the seawater. Consideration solely on the concentration factor possibly indicates that these three fish samples bioconcentrate ^{137}Cs into their bodies. The narrow-barred Spanish mackerel has a concentration factor lower than unity, which indicates that this species do not bioconcentrate ^{137}Cs. The remaining fish samples have no reported concentration factor values for ^{137}Cs. All fish samples have no reported concentration factor values for ^{134}Cs.

4. CONCLUSIONS

The activity concentration of ^{134}Cs and ^{137}Cs in seawater and fish in the Lagonoy Gulf were determined. The average ^{137}Cs activity concentration for seawater in the sampling site was 0.5901 $\pm$ 0.3916 Bq/m^3, which is below the 1990-1999 baseline in the Asia-Pacific region at 2.68 $\pm$ 0.84 Bq/m^3. Among the six selected fish samples, the forktail large-eye bream has the highest activity concentration of ^{137}Cs which is 0.8271 ± 0.4135 Bq/kg. The big-eye tuna and the brownstripe Tuna were both reported to have ^{137}Cs acitivity concentration that were below the lower limit of detection. Both seawater samples and the fish samples were observed to have ^{134}Cs activity concentration below the lower limit of detection. From the average ^{137}Cs activity of seawater and ^{137}Cs activities of the fish samples, the concentration factors were calculated. It was found that there were three fish samples that bioconcentrate ^{137}Cs, namely the common dolphin fish, forktail large-eye bream, and skipjack tuna. Results from this study can be used as current baseline data that can help track the changes in the levels of the radiocesium contaminants in the Lagonoy Gulf marine system. This study can also provide insights regarding the impact of environment-related incidents which occurred in the previous years on the marine organisms of the Lagonoy Gulf.

5. ACKNOWLEDGMENTS

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