Effect of Fukushima Accident on Fishes and Fish Like other Foods Collected from the Bay of Bengal and Karnaphuli River, Bangladesh

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Abstract
An experiment was carried out to assess the radiological exposure due to the intake of fishes and fish like foods from some common estuary (Karnaphuli River) and Marine (Bay of Bengal) contaminated by Fukushima Reactor Accident, if any, by using gamma spectroscopy. The analysis was done to estimate the radioactivity concentrations due to natural radionuclides namely $^{238}$U, $^{232}$Th and $^{40}$K and artificial radionuclide $^{137}$Cs in Karnaphuli estuary and Chittagong city adjoining Bay of Bengal fish and fish-like samples. The activity concentration for $^{238}$U in all the samples ranged from 0.10615 ± 0.0000 to 2.4767 ± 0.0005 Bq.kg$^{-1}$ with the mean of 0.6109 ± 0.0001 Bq.kg$^{-1}$. The Activity of $^{232}$Th ranged from 0.0009 ± 0.0000 to 0.0273 ± 0.0000 Bq.kg$^{-1}$ with the mean of 0.0074 ± 0.0000 Bq.kg$^{-1}$. The activity concentration for $^{40}$K was found to be in the range from 1.5516 ± 0.0311 to 74.8658 ± 0.2532 Bq.kg$^{-1}$ with the mean of 24.9533 ± 0.0702 Bq.kg$^{-1}$. The artificial radionuclide $^{137}$Cs was not found in any of the samples. The specific activity values for most of the sample were within the permissible limits. The average effective dose and internal hazard index due to consumption of those fishes and fish-like
foods was found to be 0.1 mSv.y⁻¹ and 0.0085 ± 0.0000 mSv.y⁻¹ respectively. The research work showed that the consumers of concerned fishes and fish-like foods of the Chittagong area have no risk of radioactivity ingestion even though no amount of radiation is assumed to be totally safe.

**Keywords:** River Fishes; Sea Fishes; Ingestion; Radioactive Exposure.
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1. Introduction

Radioactivity up to a certain level has always been present in the environment since the formation of the earth [1]. The level of radioactivity in the environment has begun to increase continually with the use of radioisotopes in medicine, industry, testing of nuclear weapons, nuclear accidents and nuclear power plants for nuclear energy. Radiation damage is severe in the case of acute exposure, than in the case of chronic exposure. This means that the human body is able to recuperate and repair radiation damage which is kept within certain limits and below certain threshold. There is no way to prevent the natural radiation exposure completely and absolutely. The purpose of the radiation protection is to reduce the radiation doses received by general masses to below the threshold amount which would induce permanent injury [2]. The radioactive substances which enter into the body are among the most insidious threat to living tissue. Radionuclides present in the atmosphere reach the ground level, also in the sea and river water principally through rain and dry deposition. Finally they enter into the human body mainly through food and water, i.e. ingestion and inhalation [3].
Bangladesh is a country of enormous natural beauty having nationwide river, large coastal area, water lakes, etc. The climate condition in Bangladesh makes it possible to grow huge variety of fishes. Sources of fishes here are the sea, rivers, canals, lakes, open wetlands and ponds. Fish is a food of excellent nutritional value, providing high quality and easily digestible protein and a wide variety of vitamins and minerals. Fishes have a significant positive impact on improving the quality of dietary protein by complementing the essential amino acids that are often present in low quantities in vegetable-based diets. Fish oils in fatty fish are the richest source of a type of fat that is vital to normal brain development in unborn babies and infants. Without adequate amounts of these fatty acids, normal brain development does not take place [4 – 6]. Bangladeshi people usually like to keep fish in their daily menu of foods.

According to nutrition scientist an adult people should consume about 0.8g ∙ kg\(^{-1}\) (body mass) proteins daily [7, 8]. Among them 15.1g will be animal protein. For Bangladeshi people 80% of animal protein comes from fishes [9] Fish is popular accepted protein because it can widely preserve through drying, salting, freezing and canning. This makes fish a food of choice not only to the Bangladeshi people but many other people of the world. About 12% of Bangladeshi people are directly or indirectly engaged with fish and fish related jobs and business [9]. People earn their livelihood by catching fish from open stream, rivers or deep sea. The position of fish is in third for earning foreign currency to Bangladesh. About 10% of total export income comes from the fish and fish products [10].
Due to serious nuclear accident in Fukushima, Japan in March 2011 a large amount of radionuclides ($^{131}$I, $^{134}$Cs, $^{137}$Cs) went into the Pacific Ocean [11 – 13]. The present study is to investigate the effect of this huge radiation in the sea-foods of Bay of Bengal.

2. Materials and Methods

2.1 Counting Efficiencies of the HPGe Detector

The counting efficiency of the detector was calculated using the following formula [14, 15].

$$Efficiency = \frac{CPS}{Activity \times Intensity}$$

where,

$CPS =$ Net count per second (i.e. CPS from standard source – CPS for background)

$Activity (A) =$ Standard source activity for respective energy

$Intensity (I_\gamma) =$ Intensity of gamma energies.

2.2 Data acquisition and analysis for Activity calculation

The samples after pretreatment, preparation and packing in the air tight sealed pots have been stored for 4 weeks to reach secular equilibrium between the $^{238}$U and $^{232}$Th series and their respective progeny before their measurements [15, 16]. It is assumed that $^{222}$Rn and $^{220}$Rn could not escape from the sealed containers after closure. The gamma ray activities of the fish samples have been in High Purity Germanium Detector (HPGe) that has been coupled with Digital Spectrum Analyzer-1000 (DSA-1000). For calculating the counting efficiencies and hence the
activity of samples, the adjustment of necessary parameters of the detector such as resolution, peak to Compton ratio etc and measurement of minimum detectable activity of the detector have been done. Each of the standard source and collected samples have been placed on the top of the detector one by one in due courses within the shielding arrangement by following the standard procedure. The standard sources samples have been counted for 55,160 seconds and each of the collected samples have been counted for 10,000 seconds. The most prominent gamma ray energy peaks have been found to be [3, 15, 17] at 238.63 keV (due to $^{212}$Pb); 727.17 keV (due to $^{212}$Bi); 295.21 keV & 351.92 keV (due to $^{214}$Pb); 338.40 keV, 911.07 keV & 969.11 keV (due to $^{228}$Ac); 510.57 keV, 583.19 keV & 2614.53 keV (due to $^{208}$Tl); 609.31 keV & 1764.49 keV (due to $^{214}$Bi); 1460.75 keV (due to $^{40}$K) and 661.66 keV (due to $^{137}$Cs). Activities of the natural radionuclides presented in the fish samples have been calculated by using the following formula [18]:

$$A = \frac{CPS \times 1000}{\varepsilon(\text{abs}) \times I_\gamma \times W}$$

Where, $A$ is the activity in Bq.kg$^{-1}$,

- $CPS$ is the net count per second of the experimental sample,
- $W$ is the weight of the sample in gm,
- $\varepsilon(\text{abs})$ is the absolute counting efficiency of the detector, and
- $I_\gamma$ is the absolute gamma intensity of the corresponding gamma ray energy.

The error of the measurements was expressed in terms of standard deviation ($\pm \sigma$), when, $\sigma = \sqrt{\bar{x}}$; where, $x = \text{Activity of radionuclides.}$
2.3 Preparation of Samples
In this study, a total number of 14 samples have been collected from four locations of Chittagong City namely (1) FishariGhat (shipping fishing zone), (2) Reazuddin Bazar, (3) Chawk Bazar and (4) Directly from fisherman of “fifteen no. Naval Ghat” by ensuring that the fishing have been done on the Karnaphuli estuary or/and the Bay of Bengal. The samples were collected in a clean polyethylene bag and proper identification number was given. All the samples were divided into two parts like crab and flesh after that the samples were prepared for taking the counting [19, 20].

2.4 Samples collected

| Samples collected from the Karnaphuli Estuary | Samples collected from the sea (the Bay of Bengal) |
|------------------------------------------------|--------------------------------------------------|
| 1. Crab (*Epibolocera sinuatifrons*),         | 1. Surma (*Rastrelliger kanagurta*),             |
| 2. Chingri (*Macrobrachium carcinus*),         | 2. Baila (*Awaous guamensis*),                   |
| 3. Shing (*Gangata youssoufi*),               | 3. Chanda (*Pampus argenteus*),                   |
| 4. Hilsha (*Tenualosa ilisha*),               | 4. Powa (*Otolihes argentue*),                   |
| 5. Tengra (*Nemapteryx nenga*),               | 5. Loytta (*Harpodon nehereus*),                 |
| 6. Gura Chingri (*Palaemon styliferus*), and   | 6. Mola (*Amblypharyngodon mola*), and           |
| 7. Tailla (*Eleutheronema tetractylum*).      | 7. Chandan Ilish (*Hilsa kanagurta*).            |

2.5 Calculation of Radiological parameters

2.5.1 Dose estimations from annual intakes
The risk associated with an intake of radionuclides in the body is proportional to
the total dose delivered by the radionuclides while staying in the various organs. In general it is assumed that stochastic effects occur linearly with dose and usually the effective dose equivalent is used to define the risk. So, effective dose equivalent is a parameter for the biological effect. Intakes to effective dose equivalent conversion factors are needed in order to convert the intake into dose on ingestion of radionuclides into the body. The intake to dose conversion factors (50 years period) cited in the ICRP publication no. 51 for the members of the public (adults) was used [21]. Radioactivity levels in fish were used to estimate internal effective doses. The effective dose to an individual from an intake of a radionuclide via ingestion of fish is calculated by this formula [21, 22],

$$D_{ing} = C_R \times I_F \times E_D$$

where,

$D_{ing}$ is the annual effective dose to an individual due to ingestion of radionuclides (Sv.y$^{-1}$)

$C_R$ is the concentration of radionuclides in ingested fish (Bq.kg$^{-1}$)

$I_F$ is the annual intake of fish containing radionuclide’s (kg.y$^{-1}$) and the intake rates for Bangladeshi were taken from fish consumption statistics data [22, 23].

And, $E_D$ is the ingestion dose conversion factor for radionuclides (Sv.Bq$^{-1}$)

Therefore, the total dose via ingestion is calculated by this formula,

$$D_{ing} = \left[ C_R(\frac{238}{92}U) \times I_F \times E_D \right] + \left[ C_R(\frac{232}{90}Th) \times I_F \times E_D \right] + \left[ C_R(\frac{40}{19}K) \times I_F \times E_D \right]$$
2.5.2 Internal hazard index, $H_{\text{int}}$

The internal hazard index ($H_{\text{int}}$) gives the internal exposure to carcinogenic radon in the fish samples and is given by equation [22, 24].

$$H_{\text{int}} = \frac{C_{\text{U}}}{185} + \frac{C_{\text{Th}}}{259} + \frac{C_{\text{K}}}{4810}$$

The value of this index should be less than 1 mSv y$^{-1}$ in order for the radiation hazard to have negligible hazardous effects to the respiratory organs of the public [22, 24].

3. Results and Discussion

3.1 Activity of $^{238}\text{U}$

The activity concentration of $^{238}\text{U}$ has been found in the range from $0.10615 \pm 0$ to $2.4767 \pm 0.0005$ Bq kg$^{-1}$ with the mean of $0.6109 \pm 0.0001$ Bq kg$^{-1}$. The comparative data of the activity concentration of $^{238}\text{U}$ of all samples is shown in table 1 and their corresponding graphical comparison is shown in figure 1. The comparison of the results of the mean activity concentration of $^{238}\text{U}$ with the worldwide value and other study of the world is shown in table 2 and graphically in figure 2. The graphical comparison of the mean activity concentration of $^{238}\text{U}$ for river and sea samples is given in figure 3.

3.2 Activity of $^{232}\text{Th}$

In the fish samples, activity concentration of $^{232}\text{Th}$ was found to be in the range of $0.000858 \pm 0$ and $0.0273 \pm 0$ Bq kg$^{-1}$, with a mean value $0.0074 \pm 0$ Bq kg$^{-1}$. The activity concentration of $^{232}\text{Th}$ of all samples is shown in table 1 with a
comparative graphical scheme in figure 4. The variation of the mean activity concentration of $^{232}\text{Th}$ of present study with the worldwide value and other study of the world is shown in table 2. The graphical comparison of the mean activity of $^{232}\text{Th}$ of present study with the worldwide value and other study is shown in figure 2. The comparative pictorial representation of the mean activity concentration of $^{232}\text{Th}$ for river and sea samples is given in figure 5.

3.3 Activity of $^{40}\text{K}$

The mean specific activity of $^{40}\text{K}$ in the fish samples has been found to be $24.9533 \pm 0.0702 \text{ Bq.kg}^{-1}$ having the range from $1.5516 \pm 0.0311$ to $74.8658 \pm 0.2532 \text{ Bq.kg}^{-1}$. The distribution of the activity concentrations of $^{40}\text{K}$ of all samples is shown in table 1 and their corresponding graphical comparison is shown in figure 6. The comparison of the mean activity concentration of $^{40}\text{K}$ with worldwide value and other study of the world has been shown in table 2. The variation of the mean activity of $^{40}\text{K}$ of the present study with the worldwide value and other study has been shown in figure 2. The comparative pictographic illustration of the mean activity concentration of $^{40}\text{K}$ for river and sea samples is given in figure 7.

3.4 Activity of $^{137}\text{Cs}$

The anthropogenic radionuclide $^{137}\text{Cs}$ was not found in any samples.

Table 1: The comparative data of the activity concentration of parent radionuclide’s, $^{40}\text{K}$ and $^{137}\text{Cs}$ of all fish samples
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| Sample ID | Activity Concentration of $^{238}$U (Bq.kg$^{-1}$) with (±1 σ) | Activity Concentration of $^{232}$Th (Bq.kg$^{-1}$) with (±1 σ) | Activity Concentration of $^{40}$K (Bq.kg$^{-1}$) with (±1 σ) | Activity Concentration of $^{137}$Cs (Bq.kg$^{-1}$) with (±1 σ) | Total activity concentration in (Bq.kg$^{-1}$) with (±1 σ) |
|-----------|---------------------------------------------------------------|---------------------------------------------------------------|---------------------------------------------------------------|---------------------------------------------------------------|---------------------------------------------------------------|
| R-01      | $0.2328 ± 0.0001$                                            | $0.0034 ± 0$                                                 | $1.7646 ± 0.0437$                                            | ND                                                            | $2.0001 ± 0.04$                                               |
| R-02      | $2.4767 ± 0.0005$                                            | $0.0273 ± 0$                                                 | $74.8658 ± 0.2532$                                           | ND                                                            | $77.3698 ± 0.25$                                             |
| R-03      | $0.7170 ± 0.0002$                                            | $0.0086 ± 0$                                                 | $55.0649 ± 0.0796$                                           | ND                                                            | $55.7905 ± 0.08$                                             |
| R-04      | $0.1706 ± 0.0000$                                            | $0.0019 ± 0$                                                 | $15.3436 ± 0.0233$                                           | ND                                                            | $15.5161 ± 0.02$                                             |
| R-05      | $1.5038 ± 0.0003$                                            | $0.0203 ± 0$                                                 | $29.3928 ± 0.1778$                                           | ND                                                            | $30.9169 ± 0.18$                                             |
| R-06      | $0.7274 ± 0.0002$                                            | $0.0101 ± 0$                                                 | $48.2191 ± 0.0882$                                           | ND                                                            | $48.9566 ± 0.09$                                             |
| R-07      | $0.8303 ± 0.0002$                                            | $0.0075 ± 0$                                                 | $47.3487 ± 0.1028$                                           | ND                                                            | $48.1865 ± 0.10$                                             |
| S-01      | $0.1062 ± 0$                                                 | $0.0010 ± 0$                                                 | $11.6636 ± 0.0156$                                           | ND                                                            | $11.7708 ± 0.02$                                             |
| S-02      | $0.1917 ± 0.0001$                                            | $0.0039 ± 0$                                                 | $1.5516 ± 0.0311$                                            | ND                                                            | $1.7472 ± 0.03$                                              |
| S-03      | $0.1653 ± 0$                                                 | $0.0009 ± 0$                                                 | $2.1360 ± 0$                                                 | ND                                                            | $2.3022 ± 0$                                                 |
| S-04      | $0.3022 ± 0.0001$                                            | $0.0033 ± 0$                                                 | $20.3025 ± 0.0287$                                           | ND                                                            | $20.6080 ± 0.03$                                             |
| S-05      | $0.5007 ± 0.0001$                                            | $0.0072 ± 0$                                                 | $15.5568 ± 0.0636$                                           | ND                                                            | $16.0647 ± 0.06$                                             |
| S-06      | $0.4495 ± 0.0001$                                            | $0.0058 ± 0$                                                 | $9.2995 ± 0.0493$                                            | ND                                                            | $9.7548 ± 0.05$                                              |
| S-07      | $0.1784 ± 0.0001$                                            | $0.0025 ± 0$                                                 | $16.8369 ± 0.0263$                                           | ND                                                            | $17.0178 ± 0.03$                                             |
| Mean      | $0.6109 ± 0.0001$                                            | $0.0074 ± 0$                                                 | $24.9533 ± 0.0702$                                           | ND                                                            | $25.5716 ± 0.07$                                             |

$R →$ River, $S →$ Sea
Fig. 1: Comparison of the activity concentrations of $^{238}$U of all the fish samples.

Fig. 2: Comparison of the mean activity concentration (Bq kg$^{-1}$) of parent radionuclides, and $^{40}$K of all fish samples with world average and other study.
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Fig. 3: Comparison of activity concentration of $^{238}$U between river (Karnaphuli estuary) and marine fish samples.

Fig. 4: Comparison of the activity concentrations of $^{232}$Th of all the fish samples.
Fig. 5: Comparison of activity concentration of $^{232}$Th between river (Karnaphuli estuary) and marine fish samples.

Figure. 6: Comparison of the activity concentrations of 40K of all the fish samples.
Table 2: Comparison of present results with corresponding world average values and other study

| Country       | Reference | $^{238}\text{U}$ (Bq.kg$^{-1}$) | $^{228}\text{Ra}$ (Bq.kg$^{-1}$) | $^{232}\text{Th}$ (Bq.kg$^{-1}$) | $^{40}\text{K}$ (Bq.kg$^{-1}$) |
|---------------|-----------|---------------------------------|---------------------------------|---------------------------------|---------------------------------|
| Bangladesh    | Present study | 0.6109                          | -                               | 0.0074                          | 24.95                           |
|               | [19]       | 1.05                            | 0.77                            | -                               | 61.5                            |
|               | [21]       | 0.26                            | -                               | 0.45                            | 109.11                          |
| Nigeria       | [25]       | 37.22 ± 4.31                    | -                               | 94.82 ± 3.82                    | 618.2 ± 26.81                   |
|               | [25]       | 25.6 ± 7.4                      | -                               | 52.4 ± 28.7                     | 426 ± 80                        |
| Pakistan      | [26]       | 1.3 ± 0.3                       | 1.0 ± 0.2                       | -                               | 90 ± 15                         |
|               | [27]       | 1.112                           | -                               | 0.029                           | 480                             |
| Monaco        | [28]       | 0.487±0.011                     | -                               | 0.0715±0.002                    | -                               |
|               | [29]       | -                               | -                               | -                               | 127                             |
| Greenland     | [30]       | 0.004                           | -                               | 0.001                           | -                               |
| Brazil        | [31]       | 0.055                           | -                               | 0.026                           | -                               |
| Worldwide     | [16]       | 0.008                           | -                               | 0.0007                          | 45                              |
Fig. 7: Comparison of activity concentration of $^{40}$K between river (Karnaphuli estuary) and marine fish samples.

### 3.5 Radiological Parameters or Radiation Hazard in the Fish samples:

#### 3.5.1 Effective dose rate for fish samples:

The mean value of the annual effective dose has been found to be 0.1 mSv.y$^{-1}$. The values of the effective dose for all samples obtained were lower than that of the world-wide average of 0.3 mSv.y$^{-1}$ [16]. The comparative data of the effective dose rate for all the samples are given in table 3.

#### 3.5.2 Internal hazard index, $H_{int}$:

The internal hazard index of these fish samples has been in the range from $0.0013 \pm 0$ to $0.0291 \pm 0.0001$ with an average value of $0.0085 \pm 0$, which is less than the permissible limit 1 (22,32). The results for all the samples are shown in the table 3.
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Table 3: The comparative data of the radiological parameters for the all fish samples

| Sample no | Internal hazard index, $H_{int}$ | Effective dose (Sv.y$^{-1}$) |
|-----------|----------------------------------|------------------------------|
| R-01      | 0.0016 ± 0                       | $7.14 	imes 10^{-6}$        |
| R-02      | 0.0291 ± 0.0001                  | $3.03 	imes 10^{-4}$        |
| R-03      | 0.0154 ± 0                       | $2.23 	imes 10^{-4}$        |
| R-04      | 0.0041 ± 0                       | $6.20 	imes 10^{-5}$        |
| R-05      | 0.0143 ± 0                       | $1.19 	imes 10^{-4}$        |
| R-06      | 0.0140 ± 0                       | $1.95 	imes 10^{-4}$        |
| R-07      | 0.0144 ± 0                       | $1.91 	imes 10^{-4}$        |
| S-01      | 0.0030 ± 0                       | $4.72 	imes 10^{-5}$        |
| S-02      | 0.0014 ± 0                       | $6.28 	imes 10^{-6}$        |
| S-03      | 0.0013 ± 0                       | $8.64 	imes 10^{-6}$        |
| S-04      | 0.0059 ± 0                       | $8.21 	imes 10^{-5}$        |
| S-05      | 0.0060 ± 0                       | $6.29 	imes 10^{-5}$        |
| S-06      | 0.0044 ± 0                       | $3.76 	imes 10^{-5}$        |
| S-07      | 0.0045 ± 0                       | $6.81 	imes 10^{-5}$        |
| **Mean**  | **0.0085 ± 0**                   | **1.01 	imes 10^{-4}**      |

4. Discussion

The present study shows the concentrations of U and Th -series radionuclides’ ($^{232}$Th, $^{238}$U) and $^{40}$K in the River (Karnaphuli estuary) and Marine fishes and fish like samples. The specific activity concentrations of U and Th -series in all fish samples were found to be higher than that of the world average.

The mean activity concentration of $^{238}$U in all samples were higher than those of Brazil [31], Ireland [30], England [28], other study [21] and lower than those of Nigeria [22], Monaco [27]. Again the activity concentrations of $^{238}$U in River
samples are found to be higher than the activity concentration of $^{238}\text{U}$ in marine samples.

The mean specific activity of $^{232}\text{Th}$ in all samples were found to be higher than those of Ireland [30] and lower than those of England [28], Nigeria [22], Brazil [31], Monaco [27], other study [21]. Again the activity concentrations of $^{232}\text{Th}$ in River samples were found to be higher than that of the activity concentration of $^{232}\text{Th}$ in marine samples. The mean activity concentrations of $^{40}\text{K}$ in the all samples are found to be lower than those of Nigeria [22], Monaco [27], and other study [21]. Again the specific activities of $^{40}\text{K}$ in River samples are found to be higher than that of the specific activity of $^{40}\text{K}$ in marine samples.

However, the activity concentration of $^{232}\text{Th}$, $^{238}\text{U}$, $^{40}\text{K}$ in Crab was found to be higher than that of the activity concentrations of $^{232}\text{Th}$, $^{238}\text{U}$, $^{40}\text{K}$ in Flesh. All the values of effective dose due to intake of radionuclides ingestion in humans obtained are lower than the world-wide average annual effective dose [17]. Moreover, the recommended value of the internal radiation hazard index $H_{\text{int}}$ is one [3, 22] but in all samples the internal radiation hazard is less than one. The mean internal radiation hazard index is found to be 0.0085. So the radioactive exposure from Marine and River (Karnaphuli estuary) fishes and other fish like food due to Fukushima nuclear accident does not pose any significant radiological threat to the population who are the consumer of the fishes of the present study areas.
5. Conclusions

The following are the consequences of the present study.

- The activity concentration of natural radionuclides of Uranium series has been found to be higher than that of the Thorium series in the entire fish samples.
- The activity concentration of $^{238}\text{U}$ has been found to be high in river (Karnaphuli estuary) fish samples than Marine fish samples.
- A large variation (1.5516 to 74.8658 Bq.kg$^{-1}$) of the activity concentration of $^{40}\text{K}$ is found in these samples.
- The Radiological parameters viz. annual effective dose and internal hazard index in River (Karnaphuli estuary) fish sample is higher than the Marine fish samples.
- The observed average internal radiation hazard index in the entire fish sample is 0.008529, but the recommended value of the radiation hazard index is 1(one).

Since the radiation hazard index is less than that of the standard value, so the population who are the consumer of this food are under the safety limit of radiological threat due to the dumping of the Fukushima nuclear accidental wastes in the Pacific ocean.

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