Natural radioactivity concentration measurements in shrimp and sea bass samples from the Mexican Pacific Ocean and the Gulf of Mexico

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The radioactivity concentration for shrimp and sea bass retrieved from the Mexican Pacific Ocean and the Gulf of Mexico is reported in this work. The activity of $^{40}$K and $^{208}$Tl was determined with gamma spectroscopy using a High Purity Germanium detector. Overall, the radioactivity concentration of samples from the Mexican Pacific Ocean is higher than from the Gulf of Mexico. The $^{40}$K concentration measured for shrimp is $575 \pm 14$ Bq/kg (Pacific) and $443 \pm 10$ Bq/kg (Gulf), while the activity for sea bass is $753 \pm 18$ Bq/kg (Pacific) and $502 \pm 14$ Bq/kg (Gulf). Similarly, the measured concentration of $^{208}$Tl for shrimp and sea bass is $0.75 \pm 0.03$ and $1.09 \pm 0.05$ Bq/kg (Pacific), respectively, and $0.51 \pm 0.02$ and $0.85 \pm 0.04$ Bq/kg (Gulf), respectively. No other natural radionuclides or radiation contaminants were observed with significant activity. This is the first comparison of radioactivity concentration in fish and shellfish between the two main Mexican marine ecosystems.

Keywords: Natural radionuclides; fish and shellfish radioactivity; pacific ocean seafood; Gulf of Mexico seafood.

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1. Introduction

Environmental radioactivity is originated from natural sources containing several radionuclides including radiogenic isotopes such as potassium ($^{40}$K), uranium ($^{238}$U and its decay series), and thorium ($^{232}$Th and its decay series). These isotopes are also typically found in products consumed by humans, which produces a transfer of the radionuclides into the human body. One example of such transfer is when fish and shellfish are ingested. The number of natural radionuclides present in seafood can be evaluated by identifying the gamma lines emitted using widely-known spectroscopy methods. These techniques are also able to identify radiation contaminants that would be ingested in addition to the natural radioisotopes. In addition, such measurements could also be relevant for the identification of native species and for the monitoring of nuclear accidents, as in Chernobyl or Fukushima [1].

Mexico is an important distributor and consumer country of seafood due to its geographical location between the Pacific Ocean and the Gulf of Mexico. This gives an opportunity to find several species of fish and shellfish, such as sea bass (robaló in Spanish), tuna, red snapper (huachinango in Spanish), shrimp, and octopus.

There are no reports comparing radioactivity concentration between fish and shellfish retrieved from the Mexican Pacific Ocean and the Gulf of Mexico that could provide information about differences in the seabed, food intake by fish and shellfish, or even nuclear accidents. Previous studies have focused on specific areas in different countries [2–7] without comparison between sea bodies. A comparison of the content of radionuclides among the same or similar species or even different family species could contribute to the assessment of many important differences between both oceans. Shrimp and sea bass are two species that are identified and captured in both, the Mexican Pacific Ocean and the Gulf of Mexico. On the other hand, the radioactivity concentration of the radioisotopes with higher natural activity usually observed in consumable products, such as $^{40}$K and $^{208}$Tl, can be measured using High Purity Germanium detectors.

In this work, samples of shrimp and sea bass from the Mexican Pacific Ocean and the Gulf of Mexico were obtained and their radioactivity content was determined using a High Purity Germanium detector at the Institute of Physics of the National Autonomous University of Mexico. The isotopes $^{40}$K and $^{208}$Tl were identified and measured, providing data about differences between both oceans or fish and shellfish species. This manuscript is organized as follows. First, the experimental setup is described, including the main characteristics of the Germanium detector used for the gamma spectrometry measurements. The analysis and Monte Carlo simulations performed are detailed in the following sections. Lastly, the results and conclusions are presented.

2. Experimental setup

A Broad Energy Germanium detector (BEGe) model BE2820 manufactured by Canberra was used for the gamma spectrometry measurements. The detector has a planar horizontal crystal configuration (30 mm radius and 20 mm height) and is cooled down with liquid nitrogen, in a 30 l cryogenic storage dewar. An operational bias of +3000 V is provided by an Ortec High Power voltage supply, model 456. The signal is processed by an Ortec amplifier, model 572A fol-
owed by a pocket MCA 8000 A from Amptek, using the ADMCA software. Data was collected from approximately 100 to 3000 keV over 4096 channels. The relative efficiency of the detector reported by Canberra is 13% for the 1332 keV $^{60}$Co gamma line. The energy uncertainty for this peak is reported at 0.016% (Full Width at Half Maximum over energy), additional information about the detector response, resolution, and efficiency are reported in Ref. [8]. The measured efficiency and resolution during the characterization of the detector at the Institute of Physics (UNAM) showed a discrepancy of approximately 25%. A Monte Carlo simulation of the detector was developed using the simulation toolkit GEANT4 [9], version 10.01.p03. Experimental efficiencies are in agreement with simulations within uncertainties, the details are also reported in Ref. [8].

Samples of shrimp and sea bass from the Pacific Ocean and the Gulf of Mexico were purchased in La Nueva Viga Market in Mexico City. The identification of the sample’s origin was determined by a combination of the vendors’ expertise and apparent physical differences. La Nueva Viga is the second largest fish market in the world, where 1,500 tonnes of seafood are received daily from the Pacific Ocean and the Gulf of Mexico [10]. The samples were washed with ultrapure water, dried by sun exposure, and placed in 1-liter Marinelli beakers for counting in the BEGe detector. Each sample was counted between 4 and 5 days. An example of the experimental setup, the Marinelli container with the sample inside the lead shielding, is shown in Fig. 1.

Table I shows the measured mass for the samples and the estimated volume in the Marinelli container used in the Monte Carlo simulation.

3. Analysis

The data analysis was performed with the ROOT toolkit [11]. Two photopeaks were clearly identified as corresponding to the $^{40}$K and $^{208}$Tl gamma lines emitted by the potassium decay and thorium chain, respectively. Figure 2 shows the energy spectrum for the sea bass sample from the Pacific Ocean.

The $^{40}$K $\gamma$-ray is emitted with an energy of 1.460 MeV with a branching ratio of 10.67%, while the $^{208}$Tl $\gamma$-ray has an energy of 2.615 MeV and a branching ratio of 100%. Each photopeak was fitted to a Gaussian plus a linear function using ROOT. The number of events above the background in each peak is determined by counting the number of events within a 3-σ region centered at the peak followed by subtraction of the integrated events below the linear function, in the same energy range. In addition, a background run was performed with an empty Marinelli beaker and the number of background events produced by the two photopeaks studied was obtained following the outlined procedure. The number of events obtained from the background run was subtracted from the events obtained from the shrimp and sea bass samples. The measured rates for the samples and empty Marinelli run are shown in Table II.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$^{40}$K (events/s)</th>
<th>$^{208}$Tl (events/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Empty Marinelli</td>
<td>$(4.85 \pm 0.06) \times 10^{-4}$</td>
<td>$(3.33 \pm 0.06) \times 10^{-4}$</td>
</tr>
<tr>
<td>Pacific Shrimp</td>
<td>$(3.37 \pm 0.08) \times 10^{-3}$</td>
<td>$(2.06 \pm 0.08) \times 10^{-4}$</td>
</tr>
<tr>
<td>Gulf Shrimp</td>
<td>$(1.70 \pm 0.04) \times 10^{-3}$</td>
<td>$(9.21 \pm 0.32) \times 10^{-5}$</td>
</tr>
<tr>
<td>Pacific Sea Bass</td>
<td>$(4.10 \pm 0.10) \times 10^{-3}$</td>
<td>$(2.73 \pm 0.10) \times 10^{-4}$</td>
</tr>
<tr>
<td>Gulf Sea Bass</td>
<td>$(5.80 \pm 0.14) \times 10^{-3}$</td>
<td>$(4.33 \pm 0.18) \times 10^{-4}$</td>
</tr>
</tbody>
</table>
Figure 2. Energy spectrum for the sea bass sample from the Pacific Ocean. The two photopeaks corresponding to the $^{40}\text{K}$ and $^{208}\text{Tl}$ gamma lines are indicated.

Table III. Monte Carlo efficiencies for the $^{40}\text{K}$ and $^{208}\text{Tl}$ gamma lines in each sample. Systematical and statistical errors are included, combined in quadrature.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Efficiency $^{40}\text{K}$ ($10^{-4}$)</th>
<th>Efficiency $^{208}\text{Tl}$ ($10^{-4}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pacific Shrimp</td>
<td>1.690 ± 0.007</td>
<td>8.50 ± 0.07</td>
</tr>
<tr>
<td>Gulf Shrimp</td>
<td>1.609 ± 0.009</td>
<td>8.05 ± 0.20</td>
</tr>
<tr>
<td>Pacific Sea Bass</td>
<td>1.5254 ± 0.0004</td>
<td>7.51 ± 0.18</td>
</tr>
<tr>
<td>Gulf Sea Bass</td>
<td>2.34 ± 0.03</td>
<td>11.0 ± 0.3</td>
</tr>
</tbody>
</table>

3.1. Monte Carlo simulation

Monte Carlo simulations for each sample were performed in order to determine the detector efficiency. The radioactive decays of the $^{40}\text{K}$ and $^{208}\text{Tl}$ radionuclides in the sample were simulated in GEANT4 following the geometrical configuration shown in Fig. 1. As for the chemical composition of the samples; 76% water, 19% carbon, and 5% phosphorus, with a density of 0.79 g/cm$^3$ was assumed for the shrimp [12]. 70% water, 20% carbon, 7% hydrogen, and 3% nitrogen with a density of 0.82 g/cm$^3$ was assumed for the sea bass [13]. Variations within 5% of the chemical composition and density have a negligible effect on the sample efficiency. The efficiencies obtained are presented in Table III. Errors reported include statistical and systematical effects from the chemical composition, density, volume, and branching ratio, combined in quadrature.

The activity ($A$) of the samples was determined using Eq. (1):

$$A = \frac{N}{\epsilon \times BR \times m},$$

where $N$ is the event rate in the sample after background subtraction, $\epsilon$ is the efficiency obtained from the Monte Carlo simulation, $BR$ is the branching ratio, and $m$ is the mass of the sample. Results are presented in Table IV.

Table IV. Activity determined for the $^{40}\text{K}$ and $^{208}\text{Tl}$ radionuclides in each sample. The error includes statistical and systematical contributions combined in quadrature.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$A_{^{40}\text{K}}$ (Bq/kg)</th>
<th>$A_{^{208}\text{Tl}}$ (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pacific Shrimp</td>
<td>575 ± 14</td>
<td>0.75 ± 0.03</td>
</tr>
<tr>
<td>Gulf Shrimp</td>
<td>443 ± 10</td>
<td>0.51 ± 0.02</td>
</tr>
<tr>
<td>Pacific Sea Bass</td>
<td>753 ± 18</td>
<td>1.09 ± 0.05</td>
</tr>
<tr>
<td>Gulf Sea Bass</td>
<td>502 ± 14</td>
<td>0.85 ± 0.04</td>
</tr>
</tbody>
</table>

4. Results and conclusions

The radioactivity concentration of shrimp and sea bass samples from the Mexican Pacific Ocean and the Gulf of Mexico was determined in this work. The samples were obtained from La Nueva Viga seafood market in Mexico City. Two natural radionuclides were clearly identified, namely $^{40}\text{K}$ and $^{208}\text{Tl}$. The samples from the Mexican Pacific Ocean showed higher activity for both radionuclides. No significant
amounts of other natural radionuclides or radiation contaminants were observed.

The $^{40}$K concentration measured for shrimp from the Mexican Pacific Ocean is $575 \pm 14$ Bq/kg while the shrimp from the Gulf of Mexico has an activity of $443 \pm 10$ Bq/kg (approximately 30% difference). The activity measured for the sea bass from the Mexican Pacific Ocean is $753 \pm 18$ Bq/kg, nearly 50% higher than for the sea bass from the Gulf of Mexico, which has an activity of $502 \pm 14$ Bq/kg. Similarly, the measured concentration of $^{208}$Tl for the shrimp and sea bass samples from the Pacific Ocean is higher than for the samples from the Gulf of Mexico.

The present study is the first to report measurements of fish and shellfish in both oceans surrounding Mexico. The findings of this work could be useful for the identification of similar species between both oceans or differences in the seabed or food intake by fish and shellfish. The concentration of salt in seawater from the Gulf of Mexico is higher than in the Pacific Ocean, which indicates that the radioactivity measured in this study is not caused by the potassium in the seawater.

The results presented aim to motivate experts in marine biology and other related science areas to determine the nature of the differences observed in the activity of the samples from both sea bodies. An extended study could be established by investigating other types of fish and shellfish, in addition, to broadening it to other radionuclides. The results reported indicate no health concern for both radiation contaminants and naturally occurring radionuclides, considering that no other radioisotopes were identified with activities higher than the potassium-40 activity observed.

Acknowledgments

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