ACTIVITY CONCENTRATIONS OF POLONIUM-210 AND LEAD-210 IN LEBANESE FISH

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ABSTRACT

As part of the marine environmental monitoring program, the activity concentrations of Po-210 and Pb-210 were determined in benthic and pelagic marine fish, collected from the fresh catch sold in the local markets in 4 different stations along the Lebanese coast. A total number of 11 samples were collected such that they represent the mostly consumed species by the majority of the Lebanese population. Po-210 was measured by alpha spectroscopy technique, after chemical separation consisted of drying, digestion, tracer addition and then spontaneous deposition on a silver disk. Pb-210 was analyzed directly by the gamma spectrometry method with extended range low-level High Purity Germanium detector of 50 % relative efficiency. The activity concentrations varied between 1.31 and 45 Bq/kg wet weights for Pb-210 and between 0.22 and 62.64 Bq/kg wet weights for Po-210. These variations are due to the differences in metabolism and food intake pattern. The highest concentrations were found in the pelagic species. Many factors may be the reasons of these concentrations such that, particulate stream run-off, particulate scavenging process and precipitation process.

Keywords: Po-210, Pb-210, Lebanese fish, alpha spectroscopy, gamma spectroscopy, marine monitoring

INTRODUCTION

Polonium-210 and Lead-210 are naturally occurring radionuclides that exist in the environment as decay products of the U-238 decay chain (Masque *et al*., 2002). They can enter the body *via* two principle ways: ingestion and inhalation of radon gas originated from U-238. That means eating and drinking of contaminated food or breathing contaminated air (Momoshima *et al*., 2002; Martin & Ryan, 2004).

Po-210 and Pb-210 are of radioecological interest as they are the most important representatives of the natural marine radioactivity, and they can be considered the major contributors to radiation doses received by humans, among natural radionuclides in the marine environment (IAEA TECDOC, 1995; Al Masri *et al*., 2000; Suriyanarayanan *et al*., 2008).

Marine biota such as mussels, fish and algae are capable of concentrating within their tissues various toxic elements including radionuclides, although the concentration of most of these elements or radionuclides in the medium occurs at ultra trace levels.

The concentration of most toxic elements and radionuclides occurs in marine environment at ultra trace levels. Marine biota such as mussels, fish and algae are capable to concentrate the toxic elements and radionuclides within their tissues.

Therefore many countries and international organizations have determined the concentration of these radionuclides in seafood from which the annual intake and the radiation doses due to the human consumption (Smith & Towler, 1993; Yamamoto *et al*., 1994; IAEA TECDOC, 1995; Yu *et al*., 1997; Al-Masri *et al*., 2000).

For these reasons a national monitoring program was implemented in Lebanon by the environmental radiation control department at the Lebanese Atomic Energy Commission (LAEC). Gamma radiation and radioactivity measurements in soil, water, air and the main constituents of the Lebanese diet have been evaluated (El Samad *et al*., 2007; Nasreddine *et al*., 2008). Marine environment is part of this program, and scientists at the LAEC started to study the activity concentrations of different radionuclides present in the marine environment including Po-210 and Pb-210 in different species of fish collected from different locations along the Lebanese coast.

MATERIALS AND METHODS

Sampling and sample preparation

In the present work, 11 samples of different species of benthic and pelagic marine fish were collected from the fresh catch sold in the local markets in 4 different stations along the Lebanese coastal cities, Tripoli, Batroun, Beirut and Saida. The chosen species were the mostly consumed by the majority of the Lebanese population. The locations of the catch were verified by the fisherman.

The total weight of fish sample used for analysis ranges from 1 kg to 2 kg. The samples were oven-dried at 85 ^{0}C for 48 hours and the dry/wet ratio is calculated for each one, followed by grinding and homogenizing them. Aliquots (5g) from the dried samples were subjected to radiochemical analysis of Po-210. The remaining homogenized samples were prepared in fixed counting geometry polyethylene containers for the measurement of Pb-210 by gamma spectrometry.

Gamma spectrometry analysis

For Pb-210 analysis a gamma spectrometer from Canberra was used, equipped with extended range low-level coaxial High Purity Germanium detector (HPGe) with beryllium window, and 50 % relative efficiency and high resolution FWHM 2.1 keV at 1332 keV.

The detector was surrounded with a 10 cm thick lead shield in order to reduce the background and by a 0.5 cm copper layer to attenuate X-rays emitted by the lead shield. The detector was connected to standard integerated data processor DSA1000 desktop inspector electronics and the spectra were accumulated in 8K MCA.

The energy calibration of the spectrum was done using a standard multigamma radioactive source from Isotope Products Laboratories (ISP) prepared in the same geometry as the samples to be analyzed, the efficiency calibration was performed. Furthermore, efficiency curves were corrected for attenuation and absorption. The linearity and the resolution of the detector were checked using a standard Eu-152 point source. The background spectra were measured frequently under the same conditions applied on the samples measurements and was used to correct the calculated sample activities.

Each sample was counted for a time that ranges between 36 to 48 hours, due to the low activity concentrations of natural radionuclides in fish samples. The spectra were analyzed using Genie 2000 software from Canberra Version V3.1a, including peak search, nuclide identification, activity and uncertainty calculation, and MDA calculation modules. Figure 1 illustrates a typical spectrum of fish sample showing the Pb-210 peak.

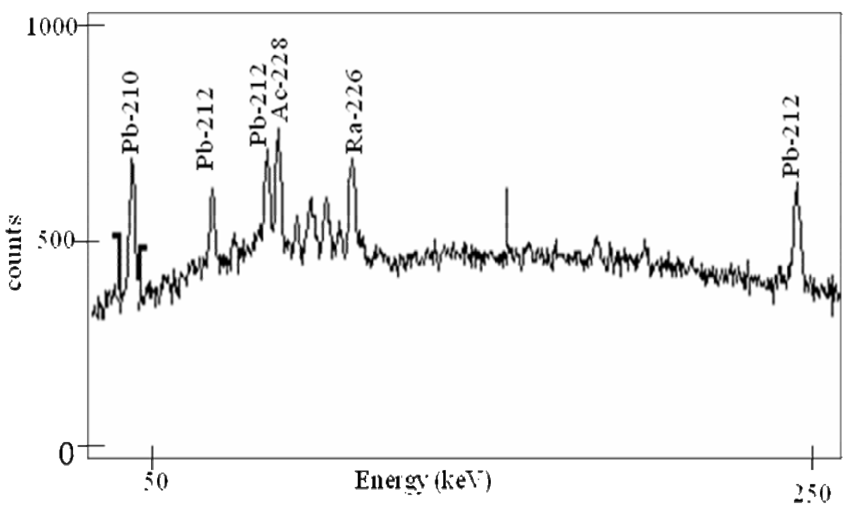

Figure 1. Gamma spectrum, Pb-210.

Alpha spectrometry analysis

For Po-210 analysis a chemical sample treatment was needed following a standard IAEA procedure. A known activity of Po-208 standard tracer was added to the sample which was dissolved by a mixture of concentrated acids (nitric and perchloric acid 10 ml to 1ml) for 24 hours. When the solution became clear, concentrated hydrochloric acid was added and gently evaporated to near-dryness. The moist residue was then dissolved again in 100 ml hydrochloric acid 0.5 mole.l^{-1}. Ascorbic acid was added in this stage to reduce iron (III) and eliminate its interference during the deposition process (Suriyanarayanan *et al*., 2008). The final solution was heated to 80^0C and the Po-210 was spontaneously deposited on a rotating silver disc (Matthews *et al*., 2007). The disc was counted using alpha spectrometer with Passivated Implanted Planar Silicon detector of resolution 10.5 keV at 5486 keV, active area

450 mm^2, mounted in a vacuum chamber, and connected to standard electronics to display spectra on the MCA. The detector was energy calibrated using standard multi-alpha source. Pulser test was applied to assess the performance of the spectrometer through the control of the resolution (FWHM) and the energy. The counting time ranges between 24 to 48 hours. The spectra showed two singlet peaks, at 5.15 and 5.3 MeV which are characteristics for Po-208 tracer and Po-210 respectively; the net peak areas were calculated by Genie 2000 for alpha acquisition and analysis version V2.0. The chemical yield, which characterizes the quality for the preceding radiochemical procedure, was calculated from the net peak area of the tracer, and the activity of Po-210 was calculated and corrected for recovery by comparison with the measured activity of the Po-208 yield tracer and for radioactive decay starting at the sampling time (Kanish, 2004). The minimum detectable activity was 8.9 x 10^{-5} Bq as calculated using Curie formula. Figure 2 illustrates a spectrum showing Po-208 and Po-210 peaks.

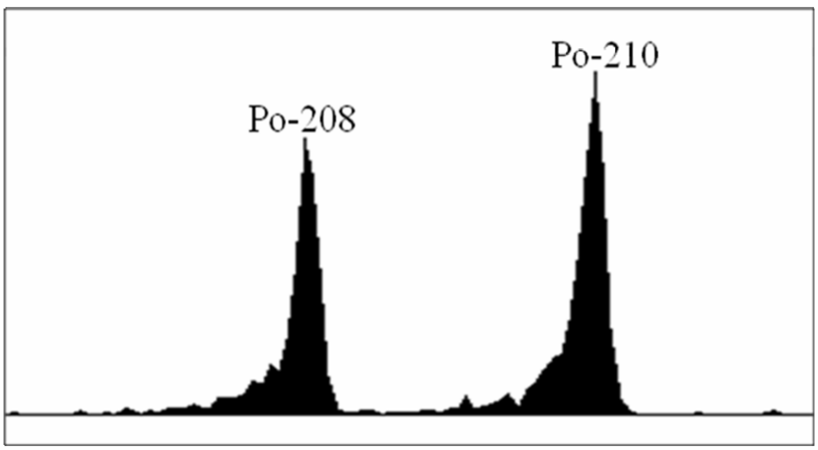

Figure 2. Alpha spectrum, Po-208 and Po-210.

The Po-210 and Pb-210 activity concentrations in the collected samples were calculated on a wet weight basis and the results were compared to those published in other countries (Noshkin *et al*., 1994; Saito & Cunha, 1997). Quality control procedure was applied using a fish standard reference material supplied by the IAEA, code IAEA-414, and the results are presented in Table 1.

TABLE 1

Results of Po-210 in Fish Reference Sample IAEA-414

Certified value (Bq/kg dry)	95% Confidence Interval (Bq/kg dry)	Measured value (Bq/kg dry)
2.1	1.8 - 2.5	2.19 ± 0.32
2.1	1.8 - 2.5	1.92 ± 0.36
2.1	1.8 - 2.5	2.17 ± 0.40

RESULTS AND DISCUSSIONS

The obtained results, from the radiochemical analysis of Po-210 and from the direct measurement of Pb-210 by gamma spectrometer in all of the collected samples are presented in Table 2. The dry/wet ratios in the collected samples varied between 20 and 26 %. The activity concentrations in wet mass were found to be 4 times less than that in dry mass. All results were discussed on the basis of fresh weight.

TABLE 2

Pb-210 and Po-210 Activity Concentrations in Lebanese Fish

Location	Species	Pb-210 (Bq/kg wet)	Po-210 (Bq/kg wet)
Tripoli	*Siganus rivulatus*	1.43 ± 0.35	31.23 ± 1.68
	Mugil sp.	1.79 ± 0.35	10.77 ± 0.81
	Sparus auratus	3.05 ± 0.47	21.51 ± 1.10
Batroun	*Pagellus erythrinus*	1.86 ± 0.29	47.82 ± 2.44
	Diplodus sargus	18.24 ± 0.79	34.51 ± 1.62
	Patella vulgata	45 ± 2	62.64 ± 2.68
Beirut	*Siganus rivulatus*	1.31 ± 0.40	4.37 ± 0.28
	Pagellus erythrinus	5.35 ± 0.41	7.94 ± 0.37
	Diplodus sargus	2.08 ± 0.21	10.15 ± 0.90
Saida	*Mugil* sp.	< MDA	4.10 ± 0.22
	Sparus auratus	1.33 ± 0.15	0.22 ± 0.03

The Po-210 activity concentrations in the fish samples are due to the selective uptake from the surroundings areas and to the decay of Pb-210. This concentration varies between 0.22 and 62.64 Bq/kg fresh weight with an average of 21.38 Bq/kg. For the Pb-210, the concentration varies between 1.31 and 45 Bq/kg fresh weight with an average value of 7.40 Bq/kg.

These variations in activity concentrations are due to the differences in metabolism and food intake pattern for different species. Usually mussels like *Patella vulgate* concentrate high Po-210 and Pb-210 radionuclides in their tissues. Also these radioactivity levels are related to the sampling location and the anthropogenic activities on the surrounding areas, whose effluents may increase the concentrations of naturally occurring radionuclides in the environment specially Po-210 and Pb-210. This explains the highest concentrations found in Batroun region, where many industries are distributed in this area. The average activity concentration of Po-210 and Pb-210 in sea fish, in spite of some high levels were found to be relatively comparable to those reported in other countries as shown in Table 3.

The mean annual effective doses by intake from Po-210 and from Pb-210 due to the fish consumption were calculated, taking into account the consumption rate of fish for the Lebanese population (Nasreddine *et al*., 2008) and the conversion factor for ingestion of Po-210 and Pb-210 (Safety Series 115, 1996). The effective dose from Po-210 was found to vary between 6.5 µSv/year at Saida sampling fish to 110 µSv/year at Batroun sampling fish. While the effective dose from Pb-210 varied between 7 µSv/year at Saida sampling fish to 160 µSv/year at Batroun sampling fish. The total mean annual effective dose by intake from Po-210 and Pb-210 radionuclides varied between 14 and 270 µSv/year.

TABLE 3

Po-210 and Pb-210 Activity Concentrations in Fish from Different Areas in the World

Country	Po-210 (Bq/kg fresh wt.)	Pb-210 (Bq/kg fresh wt.)	Reference
Lebanon	0.22 – 47.82	1.31 - 45	This work
Syria	0.27 – 27.48	0.05 – 0.38	Al-Masri *et al*. (2000)
Japan	0.6 – 26	0.04 - 0.54	Yamamoto *et al*. (1994)
Hong Kong		0.047	Yu *et al*. (1997)
Portugal	0.2 – 11		Carvalho (1988)
Australia	0.9 – 44.1		Smith & Towler (1993)
Denmark	0.35 - 0.9		Dahlgaard, 1996
South Africa	2.2 - 20.3		Cherry *et al*. (1994)
Brazil	0.5 - 5.3		Saito & Cunha (1997)
America	0.4 - 153.3	0.1 - 7	Noshkin *et al*. (1994)
Poland	0.9 – 5		Skwarzec & Falkowski (1997)

CONCLUSION

Polonium-210 and lead -210 are widely studied due to their potential radiological health significance. Many studies in different countries have shown them to be the largest contributors of natural radiation dose to marine organisms, with resulting implications for human radiation exposure; particularly where sea foods are involved. Po-210 and Pb-210 activity concentrations have been determined in 11 benthic and pelagic species collected from the Lebanese local fish markets. Relatively higher concentrations were found in Batroun region near anthropogenic activities that increase the concentration of naturally occurring radionuclides. Po-210 and Pb-210 radioactivity levels were found nearly comparable to those reported in other countries. Therefore low annual effective doses will be received for Lebanese population through the consumption of the analyzed fish species. Further studies in the future will be done to cover more species and more regions.

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