Levels of radionuclide concentrations in benthic invertebrate species from the Balearic Islands, Western Mediterranean, during the period 2012–2018

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Abstract

Baseline levels of radionuclides in the marine environment of the Balearic Islands in the Western Mediterranean are non-existent. Because of their ecological role and acknowledged sensitivity to pollutants, the activity concentrations of ^{210}Po, ^{40}K, ^{210}Pb, ^{90}Sr and ^{234}Th were measured in two types of benthic invertebrates species (mussels and sea urchins) sampled during the period 2012–2018.

The activity concentrations of ^{210}Po; ^{40}K; ^{210}Pb and ^{234}Th ranged from 38 ± 1 to 325 ± 11 Bq kg^{-1} dry weight (d.w.); 220 ± 10 and 996 ± 46 Bq kg^{-1} d.w.; from ND (lower than the limit of detection) to 55 ± 8 Bq kg^{-1} d.w.; and from ND to 70 ± 15 Bq kg^{-1} d.w., respectively. In all cases, no artificial ^{90}Sr activity was detected in the collected samples. The committed effective dose to humans was calculated to be in the range from 48 to 640 μSv year^{-1}.

Keywords: benthic invertebrates; 210Po; 210Pb; 90Sr; 234Th; Balearic Islands

There are two well-known sources of radioactivity in the marine environment: ^{40}K and natural decay series of ^{238}U and ^{232}Th; and man-made radionuclides (^{137}Cs, $^{(239+240)}Pu$ or ^{90}Sr) (Polykarpov, 1966; Shannon and Cherry, 1967). These chemically-analogue radionuclides of metabolically essential elements are bioaccumulated in tissues of the marine species through different mechanisms (Carvalho, 2018). Therefore, these radionuclides are introduced into the food chain via ingestion of marine food. In order to evaluate intake of radionuclides by human, an assessment of radionuclide concentrations in marine organisms must be carried out. The dose to the body (the energy absorbed from internal irradiation due to ingestion of radionuclides) produces biological effects in humans. An exhaustive radiation dose assessment establishes a baseline that is useful to control any increase due to nuclear accidents or industrial activities (Feroz Khan et al., 2014).

Artificial radionuclides are mainly released in the environment by Nuclear power plant (NPP) accidents (Chernobyl in 1986, or Fukushima Daiichi NPP in 2011), and nuclear weapon tests (during the 1960s) (Fathivand and Amidi, 2007). Among the artificial radionuclides, ^{90}Sr is considered a key radionuclide to be controlled (Castrillejo et al., 2016). ^{90}Sr is a primary fission product (Habibi et al., 2015) with a long biological (13 years) and nuclear half-life (28.9 years) that persists in the environment for a long time. Furthermore, ^{90}Sr is a chemical analogue of calcium metabolizing in tissues with high calcium content, such as bones, and consequently strontium is one of the most hazardous radionuclides.

Concerning natural radionuclides, ^{238}U decay chain includes ^{210}Po, ^{210}Pb, and ^{234}Th. The main sources of ^{210}Po ($T_{1/2}$ = 138 days) and ^{210}Pb ($T_{1/2}$ = 22.3 years) in the environment

88 are: dry and wet fallout of emanated ^{222}Rn from the earth's crust to the atmosphere
89 (Baskaran, 2011), radioactive decay of ^{226}Ra dissolved in seawater (Bacon et al., 1976;
90 Carvalho and Fowler, 1993), and waste discharges from industries processing
91 technologically enhanced naturally occurring radioactive materials (TENORM)
92 (Carvalho et al., 2010; Villa et al., 2009).
93
94 ^{210}Po is a toxic element that releases locally the radiation dose through its alpha-particle
95 decay of 5.3 MeV originating a fatal damage to different organs and tissues (Stewart et
96 al., 2008). It is the major contributor to radiation dose received by humans in sea food,
97 such as fish, crustaceans and molluscs (Dahlgaard, 1996; Wildgust et al., 2000). On the
98 other hand, ^{210}Pb is a beta emitter and the second highest radiotoxic radionuclide in the
99 natural decay chain (Štrok and Smodiš, 2011). Regarding ^{234}Th, it is a natural
100 radionuclide with a half-life of 24.1 days. The radioactive levels of ^{234}Th have been
101 published by previous studies in plankton (Marsh and Buddemeier, 1984; Paschoa et al.,
102 1981). However, few studies have been carried out on bioaccumulation of ^{234}Th in marine
103 organisms in the higher trophic levels (Ishikawa et al., 2004).
104
105 Finally, natural ^{40}K has a very long half-life ($T_{1/2}$ = 1.251×10^9 years) and the reported
106 activity concentrations represent the natural background levels for marine organisms
107 (Meli et al., 2008). The dose produced by ^{40}K is due to its beta emission and it is negligible
108 compared to the radiation dose associated to ^{210}Po (UNSCEAR, 2008).
109
110 Of all marine species, benthic invertebrates have been widely utilized as bioindicators for
111 assessing the contamination and ecotoxicology taking place in the marine environment
112 (Carvalho et al., 2010; Fonollosa et al., 2017; Francioni et al., 2007; Kiliç et al., 2014;
113 Rožmarić et al., 2012; Thébault et al., 2008; Topcuoğlu et al., 2003; Uğur et al., 2011).

As far as we known, there are no previous studies on radioactive pollution in the Balearic Islands in the Western Mediterranean. In view of this fact, the main objectives of this study are (1) to assess the activity concentration of ^{210}Po, ^{40}K, ^{210}Pb, ^{90}Sr and ^{234}Th in two types of benthic invertebrates distributed in the Balearic Islands, and (2) to estimate the annual effective ingestion dose to human population due to seafood consumption.

Samples were collected at two sampling locations around the Balearic Islands (Western Mediterranean), specifically at Menorca Island (BAL1/01 and BAL1/02) (see Fig. 1). The BAL 1/01 location was at the Port of Mahon, near to the city of approximately 28,000 inhabitants. The collection sites are aquaculture zones of marine invertebrates. Radioactivity levels were determined in two commercial invertebrate species regularly sold in the domestic markets. The studied species included one mollusc bivalve species (*Mytilus galloprovincialis*) and one sea-urchin ecninoderm (*Paracentrotus lividus*). The mussels were not native to Balearic Islands, they were transplanted in autumn from Ebro Delta or Adriatic Sea regions.

Fig. 1. Map showing the study area and sampling stations.

Samples were collected from 2012 to 2018 at the two locations. The life cycle of the invertebrate species and the commercial extraction periods established the sampling dates (Table 1).

Table 1. Sampling locations and period.

Location	Specie	Coordinates	Sampling period
Puerto de Mahón (BAL 1/01)	*Mytilus galloprovincialis*	39° 53.65′N–04° 16.10′E	2012–2018
Costa Este Menorca (BAL 1/02)	*Paracentrotus lividus*	39° 52.40′N–04° 18.38′E	2012–2017

142 Adult specimens of the same species per sampling year were pooled together in order to
143 obtain at least, 100 g of dry weight to carry out the analytical methods. At least, 1,000 g
144 of wet flesh of each species were treated for each sampling year.
145
146 In the laboratory, the invertebrate samples were washed with distilled water in order to
147 remove the attached sand/silt. Then, their soft tissues were dissected, weighted, frozen at
148 -20 ºC, and freeze-dried for 24 h. Finally the dehydrated samples were weighted, crushed
149 and homogenized.
150
151 To determine ^{210}Po activity concentration, a known activity of internal tracer ^{209}Po was
152 added to the lyophilized sample in order to calculate the analytical recovery. First,
153 samples were digested with concentrated HNO_3 and H_2O_2 during 24 h at low temperature
154 (<25 ºC) in order to avoid foam formation. The obtained solution was then digested at
155 high temperature (<80 ºC) and slowly evaporated to near dryness on a hot plate. The
156 obtained residue was dissolved again 2 or 3 times with concentrated HNO_3 and H_2O_2 to
157 destroy all the organic material. Finally, the residue was dissolved in 100 mL of 1 M HCl
158 and filtered through a 0.45 μm pore filter. About 0.1 g of ascorbic acid salt was added to
159 the solution as a reducing agent. A silver disc was introduced into the solution
160 continuously stirred for 4 h at 85-90 ºC and pH 1.5–2.0 being ^{210}Po spontaneously plated
161 onto the disc. The disc was then removed, washed with distilled water and acetone, and
162 dried. An alpha-spectrometry instrument (Alpha Analyst, Canberra) containing a
163 Passivated Implanted Planar Silicon (PIPS) detector was used for counting the disc. Alpha
164 spectrum analysis was performed using Alpha Analyst software taking into account the
165 decay corrections for ^{210}Po and ^{209}Po activities obtaining high chemical yields (>70%)
166 (Hurtado-Bermudez et al., 2017).
167

168 The radioanalytical method accuracy was validated by the analysis of certified reference
169 materials (CRM) provided by the International Atomic Energy Agency (IAEA-437,
170 IAEA-330 and IAEA-414). It was observed that obtained values were in good agreement
171 with the recommended values.
172
173 Regarding gamma-ray emitting radionuclides, lyophilized samples were sealed into 80
174 mL cylindrical beakers and were allowed to elapse at least 4 weeks before counting to
175 reach secular equilibrium between ^{226}Ra and their daughters (^{214}Pb and ^{214}Bi). Samples
176 were counted for 1–3 days using a low-background Canberra HPGe GR-6022 detector
177 with 60% relative efficiency, surrounded by a 10 cm thick high-purity lead shield. The
178 absolute efficiencies of the detectors were calculated using Canberra LabSOCS software
179 taking into account the effect of self-attenuation of low energy gamma-rays within the
180 sample (Hurtado and Villa, 2010). The spectra were analyzed using Canberra Genie 2000
181 gamma software v3.2. ^{210}Pb activity concentration was calculated by 46.5 keV gamma
182 emission, ^{226}Ra activity concentration was obtained through 295 and 351 keV gamma-
183 rays emitted by ^{214}Pb. ^{40}K was estimated using 1460.8 keV gamma emission. ^{234}Th
184 activity concentration was assessed by analyzing the 63.3 keV gamma-ray. Each
185 uncertainty of activity concentration covered one sigma counting statistics.
186
187 The validation of the gamma-ray spectrometry analyses was carried out through the
188 successful participation on several intercomparison exercises organized by ICRM on
189 efficiency calibration and coincidence-summing correction (Lépy et al., 2012; Vidmar et
190 al., 2008).
191
192 ^{90}Sr activity concentration was determined by measuring its daughter ^{90}Y in secular
193 equilibrium (Rivera-Silva et al., 2019). Samples were dried and ashed at 610ºC to remove

194 organic materials, and then 1 to 3 g of ashes were used for the analysis. About 20 mg of
195 Y carrier (Y_2O_3) was added to the ashes and then dissolved in 60 mL of 1 M HCl. Then,
196 pH value was adjusted to 1.2, by the addition of 2 g of citric and ascorbic acid and, finally,
197 it was transferred into a separation funnel containing 60 mL of 5% HDEHP in toluene.
198 After shaking for 5 minutes, the Sr/Y were separated whereas Y remained in the organic
199 phase. Y was back-extracted from HDEHP by shaking for 1 minute with 60 mL 0.08 M
200 HCl, and then 50 mL 3 M HCl. Yttrium was precipitated in the aqueous phase at pH to
201 9-10 as $Y(OH)_3$. Then, it was dissolved in 1 mL of 65% HNO_3, and transferred into a
202 plastic scintillation vial containing 19 mL of distilled water. The ^{90}Y activity was
203 immediately measured in an ultralow background liquid scintillation spectrometer,
204 Quantulus 1220^{TM} (PerkinElmer, Inc.) through Cerenkov counting. The spectra were
205 acquired and analyzed by WinQ and EASYView software. After the measurement, the
206 recovery yield was obtained by complexometric titration of Y with EDTA.
207
208 The radioanalytical method accuracy was validated by the analysis of certified reference
209 materials (CRM) provided by the International Atomic Energy Agency (IAEA-473,
210 IAEA-330 and IAEA-384). The obtained values were in good agreement with the
211 recommended values.
212
213 Activity concentrations of ^{90}Sr, ^{210}Po; ^{40}K; ^{210}Pb and ^{234}Th in benthic invertebrate
214 samples in the sampling sites and for each sampling campaign are presented in Table 2.
215
216
217
218
219
220

Table 2.

Activity concentrations of radionuclides (Bq/kg d.w.), their corresponding uncertainties (u), and annual ingestion dose for invertebrate samples in the study areas during the sampling campaigns

Station	Date	Season	Specie	^{210}Po (Bq/kg)	u	^{210}Pb (Bq/kg)	u	^{234}Th (Bq/kg)	u	^{40}K (Bq/kg)	u	^{90}Sr (Bq/kg)	dose (µSv/year)
BAL1/01	26/7/12	summer	Mussel	206	7	13	2	10	3	357	18	< 1.9	201
BAL1/01	9/12/12	autumn	Mussel	123	4	29	3	20	5	427	22	< 1.9	133
BAL1/01	17/12/13	autumn	Mussel	108	4	12	2	42	9	332	12	<1.6	102
BAL1/01	28/12/14	winter	Mussel	208	11	12	5	< 9		262	17	< 3	640
BAL1/01	15/12/15	autumn	Mussel	254	9	35	10	31	12	438	24	< 9.1	233
BAL1/01	12/12/16	autumn	Mussel	146.4	4.9	< 10		< 8		395	32	< 2.7	145
BAL1/01	24/4/17	spring	Mussel	325	11	18	6	27	9	352	19	< 2	240
BAL1/01	12/12/18	autumn	Mussel	218	7.4	11.4	4.2	< 9		376	25	< 2	209
BAL1/02	26/7/12	summer	Sea urchin	38	1	6	1	27	5	364	16	< 1.7	48
BAL1/02	17/12/13	autumn	Sea urchin	50	2	21	1	60	12	220	10	< 1.7	93
BAL1/02	22/4/14	spring	Sea urchin	42	2	22	4	55	11	291	16	< 1.6	83
BAL1/02	6/4/15	spring	Sea urchin	56	2	< 10		70	15	272	12	< 1.9	85
BAL1/02	12/12/16	autumn	Sea urchin	56.4	1.9	55	8	48	10	996	46	< 2.1	111
BAL1/02	11/12/17	autumn	Sea urchin	61.5	5.1	15.4	4.2	41.2	12.7	396	42	< 1	51

The activity concentrations of ^{210}Po were in the range from 108 ± 4 to 325 ± 11 Bq kg^{-1} d.w. for mussels; and from 38 ± 1 to 61.5 ± 5.1 Bq kg^{-1} d.w. for sea urchin. The highest ^{210}Po activity concentration was found in mussels and sea urchin during 2017 sampling campaign. The lowest activities were determined in the samples from 2012 to 2013 for sea-urchins and mussels respectively.

The highest activity concentrations of ^{210}Po in the species were found in mussels (Table 2). All bivalves (*M. galloprovincialis*) in our study presented higher ^{210}Po contents than the sea-urchin (*P. lividus*), which presented the lowest ^{210}Po activity concentrations (Table 2). Statistically significant differences were found between the species (ANOVA,

239 $p < 0.001$). These findings may be explained by four factors: (a) the relative position of
240 the species within the marine food chain, (b) the organism's polonium elimination
241 mechanisms, (c) the characteristics of the particles the organisms feed on, and (d) the
242 differences between the sampling sites BAL 1/01 and BAL 1/02. Therefore, the increase
243 of ^{210}Po activity contents at each trophic level seems to partly explain the differences
244 between *M. galloprovincialis* (filter feeder) and *P. lividus* (herbivore grazer and into a
245 lower position in the food chain) (Fowler, 2011). Moreover, mussels feed on suspended
246 material what is intrinsically more associated to polonium uptake. Additionally, the
247 behavior of ^{210}Po in both species was similar to that of persistent organic pollutants
248 (POPs) such as polychlorinated biphenyls (PCBs) in the sampling area (Deudero et al.,
249 2007). This fact may be due to the lipid solubility and the absence of adequate metabolic
250 pathways in organisms accumulating both ^{210}Po and PCBs in fatty tissues (Ambrosio et
251 al., 2018). Moreover, the nearest geographical area in the western Mediterranean with
252 available ^{210}Po data for mussels is the Ebro Delta (813.0 ± 72.9 Bq kg^{-1}). Our results in
253 *M. galloprovincialis* samples are lower than those reported in Ebro Delta probably due to
254 the fact that the Ebro Delta is a highly industrialized area (Fonollosa et al., 2017).
255
256 On the other hand, there was an overall average decrease of the ^{210}Po activity
257 concentrations for bivalve molluscs samples in 2013 and 2016 (see Fig. 2). Probably,
258 some environmental parameters changed, such as the amount of suspended particulate
259 matter or plankton what involved lower values for activity concentrations of ^{210}Po in
260 bivalve molluscs, either the bivalve molluscs were originated from different areas before
261 being transplanted. The values for activity concentrations of ^{210}Po in sea urchin *P. lividus*
262 present a constant and low value around 50 Bq kg^{-1} d.w. (see Fig. 3).
263
264

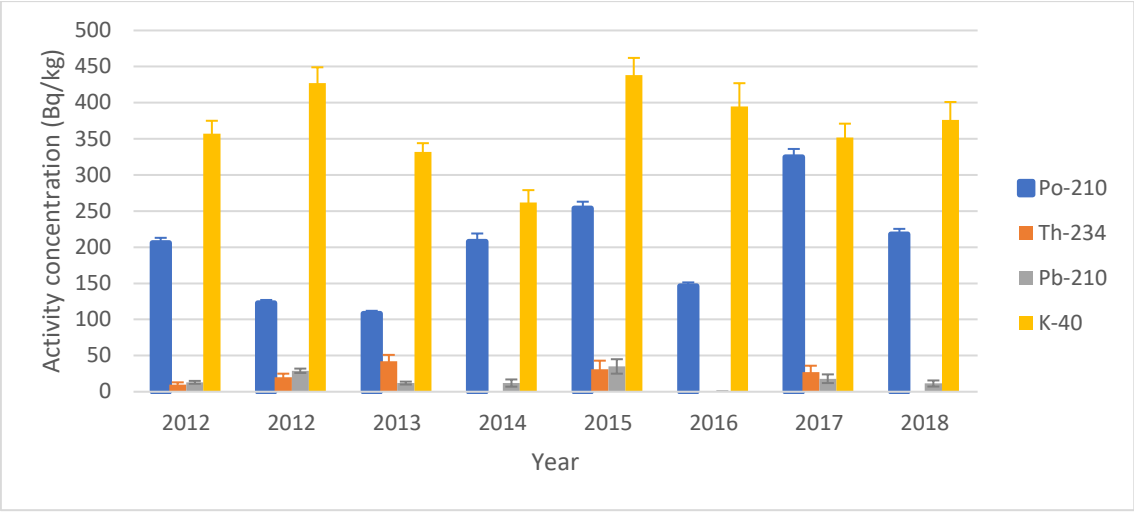

Fig. 2. Activity concentrations of different radionuclides (Bq kg^{-1} dw) in benthic invertebrates in BAL 1/01 sampling site from 2012 to 2018

The concentration of ^{210}Pb in mussels ranged from ND (not detected or below Minimum Detectable Activity) to 35 ± 10 Bq kg^{-1} d.w. in 2015, and from ND to 55 ± 8 Bq kg^{-1} d.w. in 2016 for sea urchin. No statistically significant difference was found between the species (ANOVA, $p > 0.05$). ^{210}Pb activity concentrations were relatively low probably due to the fact that ^{210}Po is mainly associated to the cytoplasm of the cells and it is subsequently assimilated into the internal tissues of the organisms ingesting these cells. On the other hand, particle reactive nuclides like ^{210}Pb and ^{234}Th are mainly bind to the surface membrane of the cell becoming poorly assimilated by the grazer organism (Stewart et al., 2008).

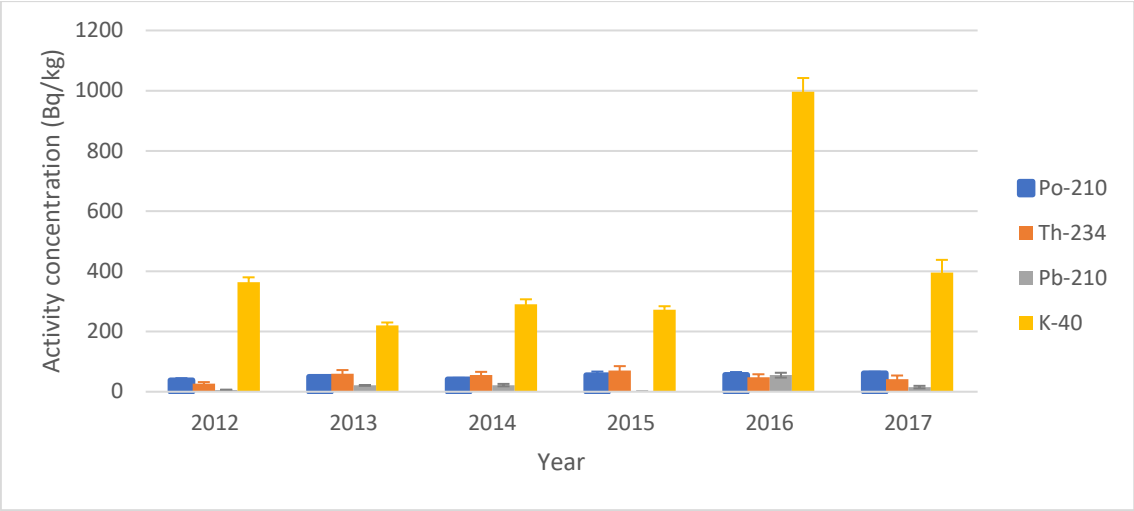

Fig. 3. Activity concentrations of different radionuclides (Bq kg^{-1} d.w.) in benthic invertebrates in BAL 1/02 sampling site from 2012 to 2017

^{210}Po and ^{210}Pb activity concentrations measured in this study are similar to the values reported in the literature (see Table 3). In this study, the ^{210}Po/^{210}Pb ratio was found to be in the range from 4 to 19 with a mean value of 13, being the ^{210}Po activity in the bivalves unsupported. The range of ^{210}Po/^{210}Pb ratio is similar to those reported by several studies (see Table 3). The ^{210}Po/^{210}Pb ratios in sea urchin samples were much lower ranging from 1 to 6 with a mean value of 3.

298 Table 3. ^{210}Po and ^{210}Pb activity concentrations, ^{210}Po/^{210}Pb ratio, and annual effective
299 ingestion dose (E_d) expressed in microsievert per year (μSv year^{-1}), in bivalves reported
300 by other authors.
301

References	Country	^{210}Po (Bq kg^{-1} d.w.)	^{210}Pb (Bq kg^{-1} d.w.)	^{210}Po/^{210}Pb	E_d (μSv year^{-1})
(McDonald et al., 1996)	UK	103–3124	-	-	-
(Uğur et al., 2002)	Turkey	52–1344	6–167	3-25	-
(Desideri et al., 2011)	Italy	75–223	2–25	4-62	96–466
(Carvalho et al., 2011)	Portugal	102-759	2.6-45	18-51	-
(Charmasson et al., 2011)	France	203	20	10-15	-
(Uğur et al., 2011)	Turkey	53-1960	6-135	4-137	-
(Štrok and Smodiš, 2011)	Slovenia	51-106	2.7-3.0	17-111	8.5
(Rožmarić et al., 2012)	Croatia	22-207	2.8-9.3	6-31	53-497
(Kiliç et al., 2014)	Turkey	26-280	1-23	8-24	0.2-3.3
(Kim et al., 2017)	Korea	240	-	-	19-189
(Fonollosa et al., 2017)	Spain	263-813	3.4-5.4	49-241	100.8
(Hurtado-Bermúdez et al., 2018)	Spain	40-506	17-92	4-16	41-479
This work	Spain	108-325	35	4-19	102-640

302
303
304 ^{40}K in bivalves ranged from 262 to 438 Bq kg^{-1} d.w., and they are similar than the global
305 average value (420 Bq kg^{-1}) (UNSCEAR, 2008). The activity concentration in sea urchin
306 for ^{40}K ranged from 220 to 996 Bq kg^{-1} d.w., achieving the highest value in 2016 due
307 probably due to port activity either fishermen or tourism. Regarding ^{234}Th radionuclide,
308 its activity concentrations ranged were in the range from 10 to 42 Bq kg^{-1} d.w., and from
309 27 to 70 Bq kg^{-1} d.w. for mussels and sea urchin, respectively. Statistically significant
310 differences were found between the species (ANOVA, $p < 0.005$). These values are
311 comparable to those reported in the only two published studies involving ^{234}Th values in
312 marine biota (Hurtado-Bermúdez et al., 2019; Ishikawa et al., 2004).

Finally, ^{90}Sr activity was not detected in any of the analyzed samples. Probably, ^{90}Sr has reached levels lower than limit of detection being unavailable for uptake into the food chain indicating that fallout ^{90}Sr may have bounded in soils or sediments. It may also occur that the detection limit of the technique (higher than 1 Bq kg^{-1}) is not low enough to measure the expected concentrations (0.8 Bq kg^{-1}) in the area (Nonova and Tosheva, 2016).

Many benthic invertebrates are consumed in the Mediterranean diet. Therefore, it is essential to evaluate the radiation dose due to the consumption of seafood. Annual ingestion doses, as a result of the consumption of benthic invertebrates, were calculated for adults using the following equation (IAEA, 2014):

$$E_d = A \times m \times C_f \quad (6)$$

where E_d (Sv) is the annual effective ingestion dose, A is the activity concentration (Bq kg^{-1} w.w.) in the sample, m (kg) is the estimation of annual intake of seafood, and C_f is the dose coefficient for adults (Sv Bq^{-1}). The annual intake of seafood was obtained from the database of food consumption maintained by the Spanish Ministry of Agriculture, Food and Environment (m = 4.5 kg of wet tissue per year per capita). The values of the dose coefficients used for ^{210}Po, ^{210}Pb, ^{40}K and ^{234}Th were $1.26 \cdot 10^{-6}$, $6.90 \cdot 10^{-7}$, $6.20 \cdot 10^{-9}$, $3.40 \cdot 10^{-9}$ Sv/Bq, respectively (IAEA, 2014).

The annual effective doses, resulting from the ingestion of ^{210}Po, ^{210}Pb, ^{40}K and ^{234}Th through the consumption of the seafood samples analyzed in this study, are shown in

Table 2. The obtained values were in range from 102-640 μSv/year to 48-111 μSv/year for mussels and sea urchin, respectively. The annual ingestion doses, calculated for the analyzed mussels, are similar to those reported in other studies (see Table 3), and are within the interval of ingestion dose due to natural radiation (200–1000 μSv/year) (UNSCEAR, 2011). However, the annual effective ingestion dose strongly depends on the annual intake of seafood via diet. This fact should be taken into account when comparing annual effective doses between different regions or countries.

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