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Descriptive statistics and risk assessment for the control of seasonal pollutant effects of ^{210}Po and ^{210}Pb in coastal waters (Çanakkale, Turkey)

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Abstract
^{210}Po is absorbed into the human body by seafood intake. Especially, mollusks and mussels are known to have much higher ^{210}Po concentration than fish among various other types of seafood and are consumed in large quantities in Aegean Sea. ^{210}Po and ^{210}Pb radionuclide concentrations are obtained in the Mediterranean mussel (*Mytilus galloprovincialis*) and in the sediment samples collected from the Çanakkale. The activity concentrations of ^{210}Po and ^{210}Pb are counted using alpha spectrometry. Activity concentrations of ^{210}Po and ^{210}Pb in mussels are in the ranged of 227 ± 11–540 ± 38 and 17 ± 4–48 ± 5 Bq kg^{-1} dw (dry weight), for sediments the ranges are 23 ± 6–41 ± 3 and 15 ± 3–44 ± 1 Bq kg^{-1} dw, respectively. Additionally, annual committed effective dose are calculated due to consumption mussel in Çanakkale coastal region. The highest effective doses of ^{210}Po and ^{210}Po are found as 3187 and 56 µSv, respectively. Finally, risk analysis assessment is recommended to determine the pollutant effects of radionuclides. The risk fractions at the concentrations are easily determined with this evaluation process. This methodology has made a great contribution to risk assessments.

Keywords Risk analysis · ^{210}Po · ^{210}Pb · Descriptive statistics · Mussel · Sediment · Annual dose

Introduction

It is well-known that ^{210}Po is an alpha emitter with half-life of 138.4 days and ^{210}Pb is a beta emitter with a half-life of 22.17 years, which are members of the ^{238}U decay chain. They show very strong binding to particle surfaces, including organisms [1]. Especially, ^{210}Po is of major importance, because it is a large contributor to the internal dose in man due to natural background radiation [2–4].

Marine organisms are used as a kind of biological indicators of radioactive contamination effects and to locate the sources of pollutions [5, 6]. Marine mussels accumulate most of the contaminants at much higher levels than those found in seawater. Consequently, humans consuming the different species of the genus Mytilus, are mainly used for monitoring due to their settled, filter feeding habits [7, 8]. In addition, sediments are considered to be pollution sinks. Part of it is scavenged from the water by particles and is removed to the sediment [9, 10]. Radionuclides are easily transferred from the sediments to benthic species and bottom-feeding fish [11].

The objectives of this paper are to determine

(1) ^{210}Po and ^{210}Pb amounts in mussels and sediments collected from the area of the Northwest Aegean Sea region (Çanakkale) of Turkey,
(2) the total annual committed effective dose received from ^{210}Po and ^{210}Pb by mussel consumption for the population living in Çanakkale,
(3) the effects of pollutants brought by radionuclides that are important for human and environmental health.

In addition, the pollution control and the evaluation of the risk are very important for future studies. For this reason, the risk situations of the ^{210}Po and ^{210}Pb radionuclides are obtained and are evaluated in the study area using

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probability distribution functions (pdfs) and risk analysis methodologies.

Materials and methods

The mussel samples (*Mytilus galloprovincialis*) are collected from Çanakkale in the 2010–2012 (Fig. 1). The location of the study area is at 40°06′08.4″N and 26°23′09.9″E, which is a small city called Çanakkale (Dardanelles) located on the Aegean Sea coast in North-West Turkey on European (Thrace) side. The European part has Gelibolu peninsula, while the Anatolia part is largely neighbouring with the historic region of Troad in Asian. Water flows in both directions along the Çanakkale strait, from the Sea of Marmara to the Aegean via a surface current and in the opposite direction via an undercurrent. Çanakkale region has a typical Mediterranean climate [12, 13].

Sediments and mussels are collected seasonally during 2010 and 2012 at Çanakkale, alongside the Aegean Sea. The Çanakkale Strait is a 74.1 km long and 1.3–7.5 km wide water channel, which attains a maximum water depth of 113 m. Dardanos region is chosen as the study area, which has a length of 10–12 km with sampling area about 140 km^2. Sediment samples are collected with a Van Veen grab sampler (5 L capacity). The samples used in the study were collected from the top layer (< 15 cm) of sediments.

Sediment samples have different physical, chemical, geological form and location characteristics. The geology of the region surrounding the Çanakkale Strait consists of pre-Miocene to Miocene bottom and upper Miocene and younger (Pliocene, Quaternary) sediments. Upper Miocene sediments consisting of siliciclastic and limestone particles deposited in fluvio-lacustrine, beach and fordable marine environments, as well as some volcanics crop out on both sides of the Çanakkale Strait. Pliocene sediments overlie the upper Miocene sediments, whereas the younger deposits around the Çanakkale Strait contain both marine and terrestrial (fluvial and alluvial) sediments of Quaternary age supplied by modern rivers discharging into the strait [14]. The structure of collected sediment is sand.

Each of the sediment samples has an approximate weight of 3 kg. They are placed in polyethylene bottles. Then each sediment samples is dried in an oven at 80 °C, and sieved to through a < 2 mm and homogenised [15].

Approximately 5 kg of mussels are placed into the mussel cages and then are transported to the laboratory with the external materials of the samples in a shell length of 3–5 cm then are washed and are cleaned. The mussel shells are opened and their soft tissues are extracted from the shells. All the samples are dried at 80 °C and then pulverized, homogenized, ^{210}Po and ^{210}Pb are quantitatively determined.

^{210}Po in mussels is measured using the standard techniques. 1 g of mussel tissues are digested with HCl, HNO_3 and H_2O_2 in a vessel after adding a known activity of ^{209}Po (0.2 Bq mL^{-1}) as a yield tracer. ^{210}Po is then spontaneously deposited onto a silver planchet. The activity of deposited polonium isotopes is counted using alpha spectrometry (Ortec, Alpha Duo, USA).

The standard deviation of the measurements has changed between 5 and 10%. The internal quality control was checked using certified reference material, IAEA-384, within ± 1 standard deviation (SD) of the reference value. The accuracy and precision of radiochemical determination of ^{210}Po is approximately < 5%.

After plating, the residual solution is stored for 6 months to allow ingrowth of ^{210}Po from ^{210}Pb, and then the ^{210}Po platting step is repeated and the ^{210}Po activity is measured [15–17].

The activity of ^{210}Pb was estimated through ^{210}Po by allowing the ^{210}Po-plated solution for a period of 6 months to build up the ^{210}Po from ^{210}Pb.

Fig. 1 Sampling area

Results and discussion

The activity of ^{210}Po and ^{210}Pb in mussels (*M. Galloprovincialis*) are shown in Figs. 2 and 3. During sampling for all periods of 2010 and 2012 at the Çanakkale coast.

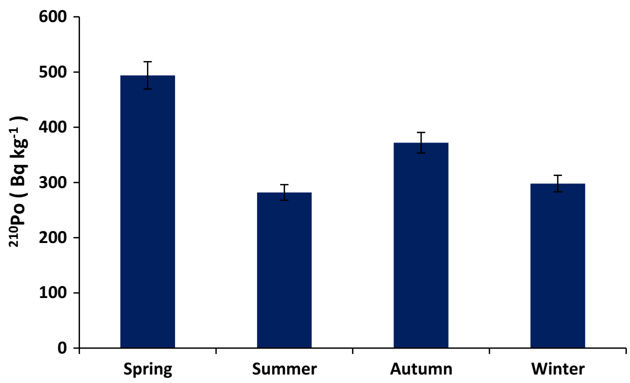

Fig. 2 Overview of ^{210}Po concentrations (Bq kg^{-1} dw) measured in mussel samples

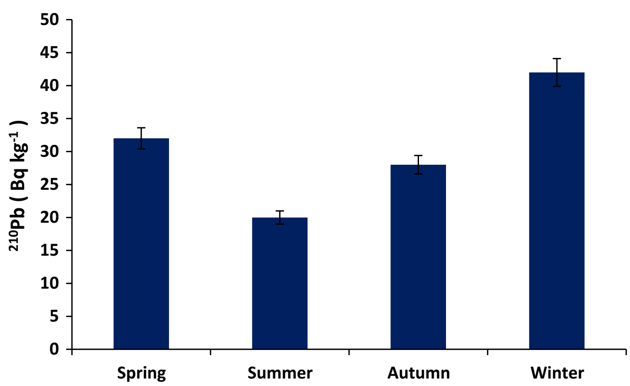

Fig. 3 Overview of of ^{210}Pb concentrations (Bq kg^{-1} dw) in mussel samples

In the mussel samples of Çanakkale ^{210}Po activity varied from 227 ± 11–540 ± 38 Bq kg^{-1} with mean value 362 ± 22 Bq kg^{-1} and the activity of ^{210}Pb as 17 ± 4–48 ± 5 Bq kg^{-1} with mean value 31 ± 4 Bq kg^{-1}.

The highest ^{210}Po concentration (540 ± 38 Bq kg^{-1}) is calculated in mussel for springtime. An analysis of variance showed that the differences are significant in ^{210}Po concentrations in mussel samples among the seasons (ANOVA, $p < 0.001$).

The values may indicate that obtained ^{210}Po activity concentrations in the mussel samples do not only occur from ^{238}U decay series. Therefore, it is obvious that ^{210}Po data presented in this study are little affected by the ^{210}Pb decay.

According to Kılıç et al. [17], ^{210}Po is derived little from settled uranium decay chain radionuclides, which show that the plurality of ^{210}Po accumulated by the mussel is unsupported. In addition, many researchers also reported that unsupported ^{210}Po is selectively accumulated in comparison with its precursor [17–19].

Higher concentrations in spring can be measured for ^{210}Po and ^{210}Pb due to both biological and environmental conditions, which violently affect the level of bioaccumulation in mussel tissue [20].

Our previous studies in mussels (*M. galloprovincialis*) showed values that ranged from 7.1 to 15.4 with the mean value of 12.5 for ^{210}Po/^{210}Pb ratio. There are no significant differences in ^{210}Po and ^{210}Pb concentrations in mussel samples as far as the sizes are concerned (Pearson correlation, $r = 0.018$, $p > 0.05$).

For the ^{210}Po and ^{210}Pb is then observed a behaviour previously defined in the literature with a common pattern: ^{210}Po > ^{210}Pb in marine organisms. It can be observed that the ^{210}Po has a higher bio-accumulative behaviour than ^{210}Pb along the marine trophic chain [21].

In the present study, these results are also in conformity with the published literature [1, 20, 22, 23] and they are given in Table 1.

The consumption is calculated using the ^{210}Po and ^{210}Pb results measured in investigated mussel samples in present research. The annual committed effective dose (E_{d}) is calculated as per the following formulations.

$$E_{\mathrm{dPo-210}} = A\left(^{210}\mathrm{Po}\right) \cdot m \cdot C_{\mathrm{f}}\left(^{210}\mathrm{Po}\right) \quad (1)$$

$$E_{\mathrm{dPo-210}} = A\left(^{210}\mathrm{Pb}\right) \cdot m \cdot C_{\mathrm{f}}\left(^{210}\mathrm{Pb}\right) \quad (2)$$

E_{d} is the annual committed effective dose (μSv y^{-1}), where E_{d} is the annual committed effective dose in μSv y^{-1}, A is the activity concentration (Bq kg^{-1}), m is the fresh weigh, (kg) and the annual intake of mussels estimate is 1 kg of fresh mussel tissue per year per capita. C_{f} is the effective dose conversion factor reported to be 1.2×10^{-6} Sv Bq^{-1}for ^{210}Po and 6.9×10^{-7} Sv Bq^{-1} for ^{210}Pb [15, 23, 25].

The ingestion dose from the consumption of mussels in present study, due to ^{210}Po and is estimated to vary between 1434 ± 70 and 3187 ± 167 μSv y^{-1}, in Çanakkale coast. The ^{210}Pb ingestion dose received by the consumers is found as 56 ± 16 and 175 ± 18 μSv y^{-1}.

In the sediment samples of Çanakkale, ^{210}Po activity ranged from 23 ± 6 to 41 ± 3 Bq kg^{-1} and the activity of ^{210}Pb varied from 15 ± 3 to 44 ± 1 Bq kg^{-1}.

There are no significant differences in ^{210}Po activity concentrations in the sediment samples in regards to the sampling seasons (ANOVA, $p > 0.05$).

In the sediment samples, ^{210}Pb activity concentrations are relatively low during all periods. An analysis of variance showed that the observed differences are significant in ^{210}Pb concentrations in sediment samples among the seasons (ANOVA, $p < 0.001$).

Concentrations of ^{210}Po and ^{210}Pb for sediment samples are compared with those from different studies in Table 2. Activity of ^{210}Po and ^{210}Pb in the sediment samples are presented in Fig. 4.

Table 1 ^{210}Po and ^{210}Pb concentrations in mussel samples from different areas

Area	^{210}Po (Bq kg^{-1} dw)	^{210}Pb (Bq kg^{-1} dw)	^{210}Po/^{210}Pb	References
Slovenia	51.2–106.2 (fw)	2.71–3.03 (fw)	16.9–113.3	[1]
Turkey (Aegean Sea)	53–1960 (dw)	6–135 (dw)	3.6–137	[24]
Turkey (Aegean Sea)	34–1855 (dw)	ND–64 (dw)	1.0–106.8	[8]
Croatia (Adriatic Sea)	22.1–207 (fw)	2.8–9.3 (fw)	6.2–30.7	[23]
Candarlı Gulf (Aegean Sea)	52–109 (dw)	38–92 (dw)	19.4–32.7	[15]
Canakkale	282–494	20–42	7.1–15.4	Present study

fw Fresh weight

Table 2 ^{210}Po and ^{210}Pb activity concentrations in sediment samples from different areas

Area	^{210}Po (Bq kg^{-1} dw)	^{210}Pb (Bq kg^{-1} dw)	References
Ghazaouet Bay (Algeria)	52–118	60–131	[26]
Milos Island	60 ± 8–100 ± 10	10 ± 2–20 ± 2	[27]
Gökova Bay		50 ± 4–113 ± 8	[28]
Izmir Bay	43 ± 6–132 ± 12	27 ± 5–91 ± 9	[11]
Milos Island	20 ± 2–166 ± 8	14 ± 3–107 ± 3	[29]
Sudan, (Red Sea)	1.6–12.5	3.5–16.3	[30]
Aegean Sea	24 ± 5–126 ± 6	18 ± 3–59 ± 4	[31]
Candarlı Gulf (Aegean Sea)	52 ± 5–109 ± 8	38 ± 5–92 ± 9	[15]
Canakkale	25 ± 5–35 ± 6	18 ± 4–41 ± 4	Present study

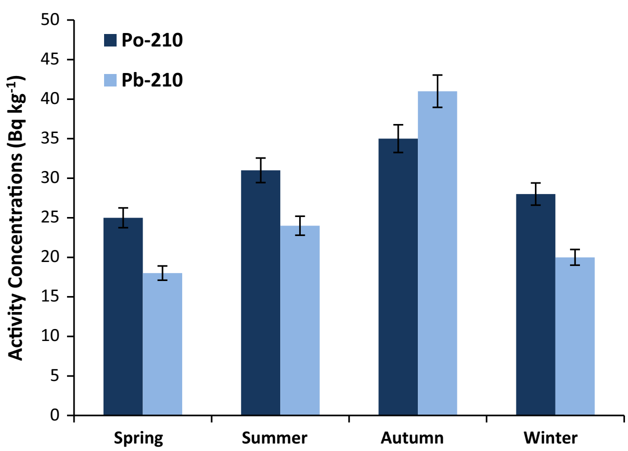

Fig. 4 Activity of ^{210}Po and ^{210}Pb (Bq kg^{-1} dry weight) in sediment samples for all periods

^{210}Pb activity concentrations are higher in sediments of Çanakkale in autumn and summer periods. The ^{210}Po/^{210}Pb ratio is concerned in sediment, and the ratios ranged from 0.85 to 1.40. This means that a significant fractionation occurred between ^{210}Po and ^{210}Pb during their removal from solution to particles as reported by other authors [32].

In this study, it is shown clearly that the sediment could not affect the concentration of ^{210}Po and ^{210}Pb. Positive correlation ($r = 0.684$, $p < 0.05$) is found between concentration of ^{210}Po and ^{210}Pb.

Descriptive statistics for in mussel and sediment

In this section, some statistical calculations are made to see the changes of ^{210}Po and ^{210}Pb concentrations in mussel and sediment samples.

A descriptive statistical analysis of the radioactivity is detected in the ^{210}Po and ^{210}Pb radionuclides to find possible radioactive pollution in the Mediterranean Mussels, which is given in Table 3. As can be seen in Table 3, the fluctuations in ^{210}Po are relatively larger than the ^{210}Pb in terms of standard deviations of the samples. Kurtosis describes a distribution with respect to its flatness or peakedness as compared to a normal distribution. In Table 3, kurtosis has a negative value, which characterizes a relatively flat situation. Its value is − 0.264 for ^{210}Po and − 0.478 for ^{210}Pb. On the other hand, skewness characterizes the asymmetry of a distribution. In our samples in Table 3, the Skewness coefficient is positive (it is 0.636 for ^{210}Po and is 0.369 for ^{210}Pb). Positive skew indicates the longer tail in the probability distribution function (pdf) in the direction of extremely high values.

Descriptive statistics of ^{210}Po and ^{210}Pb radionuclides contamination in sediment samples are given in Table 4. The average ^{210}Po pollution in mussel samples is approximately 10 times greater than sediment samples. This high rate is not observed in ^{210}Pb samples. This brought to mind the thesis that the mussels have accumulated ^{210}Po in their bodies. In addition, the kurtosis value of ^{210}Po in the sediment samples is positive (0.980), unlike the mussel's

Table 3 Descriptive statistics for in the mediterranean mussel

^{210}Po (Bq kg^{-1})		^{210}Pb (Bq kg^{-1})	
Sample	12	Sample	12
Mean	361.416	Mean	30.5
SE	26.622	SE	2.737
Median	339	Median	29.5
Mode	–	Mode	25
SD	92.223	SD	9.482
Sample variance	8505.174	Sample variance	89.909
Kurtosis	− 0.264	Kurtosis	− 0.478
Skewness	0.636	Skewness	0.369
Range	313	Range	31
Min	227	Min	17
Max	540	Max	48
Sum	4337	Sum	366
Confidence level (95.0%)	58.596	Confidence level (95.0%)	6.024

Table 4 Descriptive statistics for in the sediments

^{210}Po (Bq kg^{-1})		^{210}Pb (Bq kg^{-1})	
Sample	12	Sample	12
Mean	29.750	Mean	25.666
SE	1.451	SE	2.775
Median	28.5	Median	22
Mode	28	Mode	19
SD	5.030	SD	9.613
Sample variance	25.295	Sample variance	92.424
Kurtosis	0.980	Kurtosis	− 0.340
Skewness	0.886	Skewness	1.034
Range	18	Range	29
Min	23	Min	15
Max	41	Max	44
Sum	357	Sum	308
Confidence level (95.0%)	3.195	Confidence level (95.0%)	6.108

samples. The positive kurtosis value characterizes the peakness of the distribution relatively.

Risk analysis for ^{210}Po and ^{210}Pb pollution control in mussel

When Fig. 5 is examined, it is seen that the pollution of ^{210}Pb and ^{210}Po radionuclides in mussel samples are statistically correlated in the positive direction in summer and autumn. In the spring and winter months, there is no serious relationship.

Risk analysis is applied to the samples to find radioactive risk levels in mussel samples. For this purpose, the most suitable pdf is adapted for the examples in this research, which is the log-normal pdf. The level of risk for ^{210}Pb is shown in Fig. 6 and for ^{210}Po in Fig. 7.

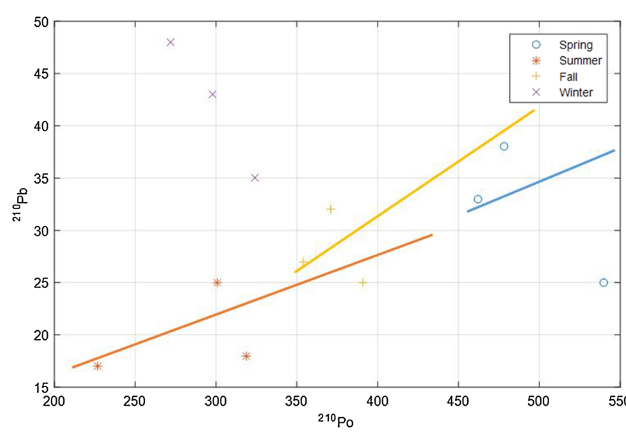

Fig. 5 Scatter graph showing the statistical tendency on of the ^{210}Pb and ^{210}Po radionuclides in mussels collected seasonally on the Çanakkale shores

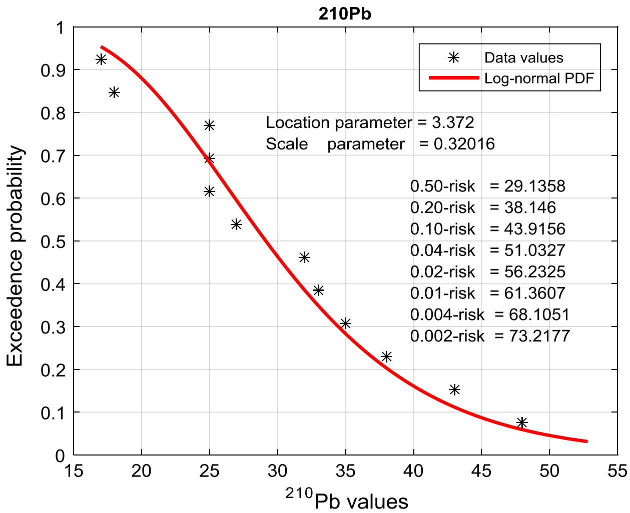

Fig. 6 Possibility of exceeding ^{210}Pb in mussel samples

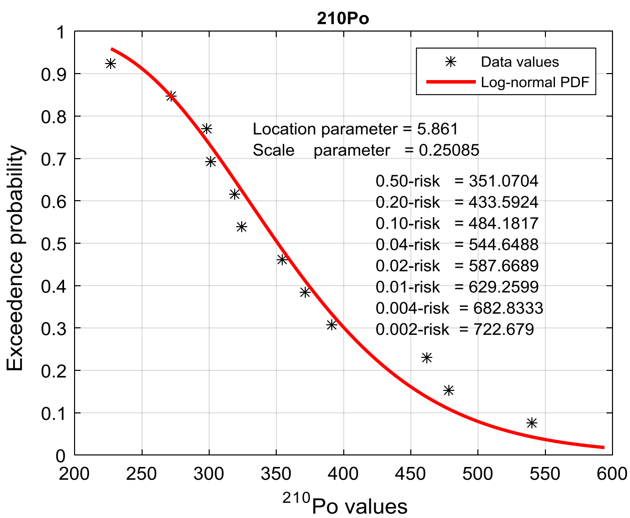

Fig. 7 Possibility of exceeding ^{210}Po in mussel samples

The risk levels on these graphs are given also as a legend. For example, in Fig. 6, the limit value for exceeding 50% risk level on the legend for ^{210}Pb is 29.13 Bq kg^{-1}. For ^{210}Po, the limit value for 50% risk level is approximately 351.07 Bq kg^{-1}. Knowing these values is crucial for the control of radionuclides pollution of the mussels.

Risk analysis for ^{210}Po and ^{210}Pb pollution control in sediments

The pollution rates of ^{210}Po and ^{210}Pb in seasonal sediment samples taken from Çanakkale coasts are statistically more significant than the samples of mussels. This result can be clearly seen in Fig. 8.

Especially in spring, winter and summer, there is a correlation of the relevant radionuclides in the positive

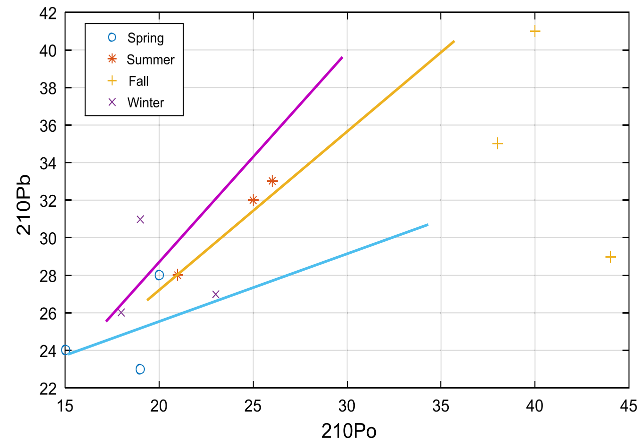

Fig. 8 Scatter graph showing the statistical tendency on of the ^{210}Pb and ^{210}Po radionuclides in sediments collected seasonally on the Çanakkale shores

direction. Only an inhomogeneous correlation is observed in autumn (Fig. 8). It is possible to attribute the cause to active autumn rains in the region. As a result of these precipitations, one can think that there is a dilution in the radionuclides concentration in the study area.

The change of ^{210}Po radionuclide in sediment samples is given in Fig. 9. The change of ^{210}Pb in the sediment samples is also given in Fig. 10. The statistical variation of these two radionuclides in the sediment has log-normal pdf. Risk analyses are also made using the pdf and they are shown as legends on both figures. Risk analyses give valuable results in terms of controlling the pollution rates that can bring the relevant radionuclide into a research zone.

These legends can be assessed as follows: For example, in Fig. 9, the value corresponding to 50% risk value is 29.5948 Bq kg^{-1}, which shows that 50% of the possibility in the level of ^{210}Po is about 29.5 Bq kg^{-1} in Çanakkale

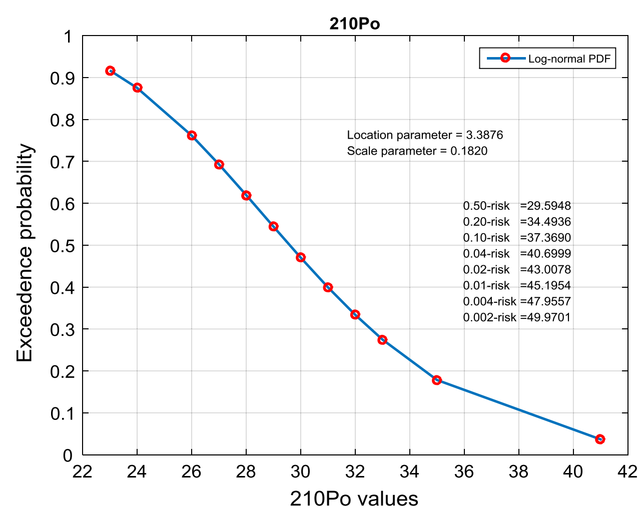

Fig. 9 Possibility of exceeding ^{210}Po in sediment samples

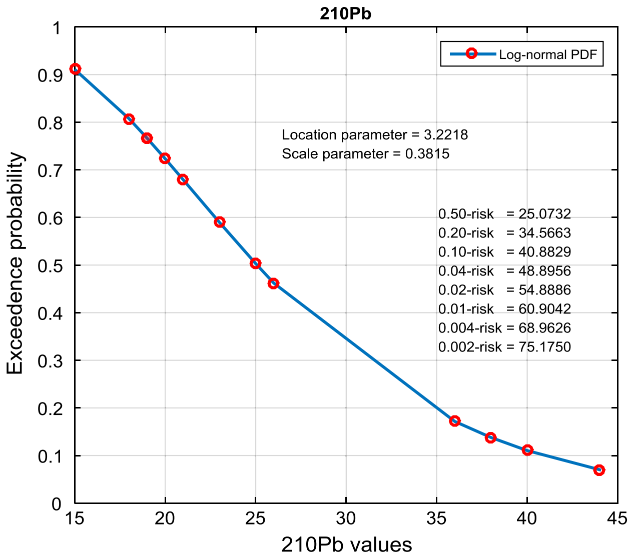

Fig. 10 Possibility of exceeding ^{210}Pb in sediment samples

shore sediments. Similarly, the likelihood of ^{210}Po reaching an approximate limit of 49.9 Bq kg^{-1} is around two per thousand when all the data are evaluated. Figure 10 shows that the concentration of ^{210}Pb reaches approximately 25.07 Bq kg^{-1}, and at 50% probability reaches to 48.9 Bq kg^{-1} as 4%. Furthermore, the likelihood of reaching 61 Bq kg^{-1} is 1% and the probability of 75 Bq kg^{-1} is two per thousand. Other risk values can also be calculated in a similar way.

Conclusions

This study provides ^{210}Po and ^{210}Pb concentrations in the mussels' soft tissue and sediments in the Çanakkale coast of the Aegean Sea. The activity ratio ^{210}Po/^{210}Pb (mean value is 12.5) shows that the radionuclides ^{210}Po and ^{210}Pb are not in equilibrium and the accumulation of ^{210}Po in mussel is greater than that of ^{210}Pb. Generally, the highest levels of ^{210}Pb and ^{210}Po concentrations in mussel tissues appear in spring period than in other periods. Results confirmed that the level to which a radionuclide is accumulated in an organism depends on a wide range of factors, such as its metabolic mutation, chemical and biological processes.

The disequilibrium between ^{210}Po and ^{210}Pb (mean value is 0.83) and the higher activity of ^{210}Pb shows the existence of unsupported ^{210}Pb in the sediments. A significant correlation is observed between the concentrations of these radionuclides in the sediment samples. The higher activity in Çanakkale could significantly affect agricultural or urban waste, industrial activities, and shipping in the Çanakkale strait. These results provide more accurate baseline levels of ^{210}Po and ^{210}Pb concentrations in the mussel and sediment from Çanakkale, the North-West Aegean Sea region (Turkey).

The Aegean Sea population has the seafood as an essential component of his diet. The result obtained in present study shows that for the Aegean Sea (Çanakkale) population heavily consuming seafood products, the annual committed effective dose via ingestion due only to ^{210}Po can even reach the value of 1 mSv. In general obtained values, due exclusively to ^{210}Po, are higher than the average value defined by UNSCEAR to the annual committed effective dose received by the worldwide population due to the ingestion of natural and anthropogenic radionuclides. The obtained all results highlights the importance of the course shown in this study related to the total dose received due to natural sources by the Aegean Sea (Çanakkale) population. This appeared to represent the highest ^{210}Po concentration in mussels found in Aegean Sea.

Descriptive statistical calculations for mussel and sediment samples provide meaningful information for similar investigations. It is possible to think that this type of calculations provides important ideas to the relevant researchers important for further observation and forecasting. Pollution control in environmental milieu is of great importance, whether it is pollutant in micro or macro scales. With the risk analysis procedures conducted in this study, important results are obtained for pollution control in terms of the relevant variables. The log-normal probability distribution function (pdf) for pollution control enabled to perform successfully risk analysis for all samples. It can be said that the risk analysis calculations used to monitor the pollutant effects of radioactive nuclei analysed as in this study can be used successfully for other pollutants.

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