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An assessment of the external radiological impact in areas of Greece with elevated natural radioactivity

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Abstract

In the present study, the radiological impact assessment in three selected areas of elevated natural radioactivity in Greece is attempted, based on measurements, theoretical relations, and simple model application. These areas are Milos – an island of volcanic origin in Cyclades Archipelago, Ikaria – an island in the Eastern Aegean Sea and Loutraki – a coastal area in mainland Greece. These areas are characterized by their geothermal springs and vents, which emit fluids into the littoral and sublittoral zones. The results include: (a) the exposure dose rates assessed by a car-borne scintillation spectrometry system; (b) laboratory measurements of the activity concentrations of the gamma-emitters of ^{238}U and ^{232}Th series and ^{40}K in soil, spring water, seawater and sediments by gamma-spectrometry; (c) estimations of the effective dose rate equivalents and health risk assessment for humans and external dose rates for natural aquatic populations in relation to organism habitat; and (d) a radiological evaluation for the environmental quality, in terms of the discrete zones of impact of ionizing radiation.

Keywords: Natural radioactivity; Gamma-radiation; Dosimetry; Greece

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1. Introduction

The presence of naturally occurring radionuclides in the environment may result to an external and internal dose received by a population exposed to them directly and via the ingestion/inhalation pathways. The assessment of the radiological impact on a population, as result of the radiation emitted by these radionuclides, is particularly important since they may have a significant contribution to the collective dose of the population. This assessment is based on the terrestrial radionuclides ^{238}U and ^{232}Th as well and their decay products and ^{40}K.

The natural radiation exposure levels have been determined in three areas of Greece chosen on the basis of their characteristic geological features. These areas are the islands of Milos and Ikaria and the coastal city of Loutraki in mainland Greece (Fig. 1).

The island of Milos is part of the Hellenic volcanic arc located in the southern Aegean Sea parallel to the subduction zone of the lithospheric plates of the Eastern Mediterranean (Fytikas, 1975; Pippos, 1985). Besides, the island is characterized by the presence of geothermal vents used for energy production by the Public Power Corporation. Hence, the abiotic materials and organisms inhabiting the coastal areas of Milos are influenced directly and indirectly by the underground hydrothermal fluids emitted by the vents (Kritidis and Florou, 1989; Boisson et al., 2001). The lithospheric environment of the island is characterized by alumino silicate and iron sulphate ores and volcanic ash, whereas the soil is mainly texturally calcareous and clay. Since some volcanic areas are characterized by elevated concentrations of natural radionuclides (Sciocchetti et al., 1988), gamma-radiation measurements of abiotic materials from the terrestrial and marine environment are of particular interest.

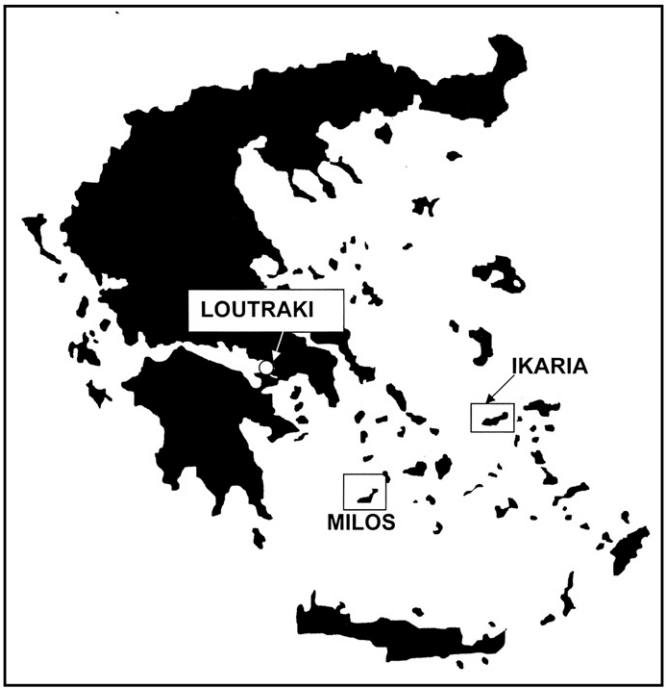

Fig. 1. Map of Greece indicating the areas studied.

The island of Ikaria is located in the eastern Aegean Sea. It is dominated by a mountainous area divided, from the geological point of view, into two petrologically distinct equal parts: (a) the eastern part consisting of largely metamorphosed sedimentary formations and (b) the western part mainly consisting of granitic formations (Ktenas and Marinos, 1969). In the littoral zone around the island, there are several geothermal springs. In the sublittoral zone, some springs emerge under the strata through the surface of the bottom to the seawater layers above (Trabidou et al., 1996). The major geological features of the abiotic materials sampled are described by Makris et al. (1965), Pippos (1985) and Trabidou (2004). Most of the springs are contact springs with different types of rocks at the limits of the granitic formations and they contain various ions, dominant with NaCl and in smaller amounts sulphate salts. The springs are characterized by the elevated concentrations of ^{226}Ra and ^{222}Rn if compared with respective spring waters' concentrations in Greece and worldwide. High disequilibria factors between ^{226}Ra and its decay product ^{222}Rn have been calculated (Anagnostakis et al., 2005).

The city of Loutraki is located in Korinthiakos gulf in central Greece. It is the south-western edge of the Hellenic volcanic arc (Papanicolaou, 1986). Several springs are found in the littoral and sublittoral zones and in the wider sampling area considered. The springs are characterized by the presence of ^{226}Ra and ^{222}Rn with concentrations lower than the respective concentrations in the springs of the island of Ikaria.

In this work, results from the determination of the activity of the terrestrial radionuclides and their decay products in soil, sediment, spring water and seawater are presented from the areas studied. Furthermore, an assessment of the radiological impact on the population due to the naturally occurring radionuclides is presented.

2. Methodology

2.1. In situ and laboratory measurements

Gamma-radiometry was carried out *in situ* in order to determine the dose rate due to the gamma-radiation emitted by the naturally occurring radionuclides. A car-borne system was used, comprising a $2'' \times 2''$ NaI (Tl) cylindrical detector with a sensitivity for ^{226}Ra of 1 cpm per 3.5×10^{-3} μR h^{-1} at 1 m above the ground (Florou and Kritidis, 1992; Trabidou, 2004).

Gamma-spectroscopy was carried out in laboratory on soil, sediment, spring water, and seawater samples from the areas studied by gamma-radiometry. The purpose of this analysis is twofold: firstly, the determination of the composition of these samples in the radionuclides ^{238}U, ^{226}Ra, ^{232}Th, ^{228}Ra, ^{228}Th, ^{222}Rn and ^{40}K; and secondly, the calculation of the dose rates because of these compositions. A high-resolution gamma-spectrometry system is used to measure gamma-ray spectra in an energy range between 50 and 2000 keV. The system comprises a high-purity germanium (HPGe) detector with an efficiency of 20% relative to a $3'' \times 3''$ NaI (Tl) scintillator. The multi-channel analyzer emulation software Maestro2 (ORTEC) allowed data acquisition, display, storage and analysis of the acquired spectra. The samples were measured for 70 000 s, whereas the relative statistical error (1σ) was kept always to within 10%. Hence, the activity concentrations of the radionuclides under consideration were determined from the analysis of gamma lines of their decay products (Florou, 1992; Trabidou, 2004). This required a parent–daughter secular equilibrium within the samples that was achieved through their sealing in containers and a time delay prior to gamma-spectroscopy.

The soil and sediment samples were placed in pots of 68 mm diameter and 15 mm height. The samples were kept for 20 d before gamma-spectroscopy was carried out, achieving the required equilibrium between ^{226}Ra, ^{222}Rn and its decay products. The activity of ^{232}Th was estimated from the mean activity of its decay product radionuclides ^{228}Ra and ^{228}Th. The mean value is considered to give more reliable result, since the equilibrium factor for ^{228}Ra/^{228}Th has been found to be different than one. In this sense,

equilibrium factors, in lithospheric materials for the island of Ikaria, in the range of 0.12–4.52 and 0.29–2.00 for $^{226}Ra/^{238}U$ and $^{228}Th/^{228}Ra$, respectively, have been previously reported (Trabidou, 2004). Furthermore, it should be noted that in the wide area of Greece, these equilibrium factors are in the range of 0.06–4.8 for $^{226}Ra/^{238}U$ (Anagnostakis et al., 2005).

The liquid samples were placed in 1 l Marinelli beakers and the pH was adjusted to 1 by adding nitric acid. The ^{222}Rn determination was made 3 h after sealing the samples, in order to ensure the equilibrium between ^{222}Rn and its decay products. The activity of ^{222}Rn was derived from the analysis of the 295.2 keV line and 352 keV line of ^{214}Pb and the 609.4 keV line of ^{214}Bi. The uncertainty of the measurements is estimated at 25% due to gas losses during sampling. In the case of the ^{226}Ra, the samples were aerated and closed after the removal of ^{222}Rn. Then, measurements were performed 20 d after sealing the samples to ensure the secular equilibrium between ^{226}Ra and its decay products.

2.2. Determination of dose rates received by humans

The *external* dose rate to a population due to the naturally occurring radionuclides in the soil and sediment was calculated according to the following formula (Probonas and Kritidis, 1993):

$$H_{ext} = 0.061A(^{238}U) + 3.103A(^{226}Ra) + 1.741A(^{228}Ra) + 2.582A(^{228}Th) + 0.276A(^{40}K) \quad (1)$$

where H_{ext} ($\mu Sv\ y^{-1}$) is the exposure dose for terrestrial gamma-radiation and $A(i)$ is the activity concentration ($Bq\ kg^{-1}$) of the radionuclide i determined by gamma-spectroscopy in the soil and sediment samples analysed.

2.3. Determination of dose rates received by marine organisms

The radiological impact on marine organisms exposed to the radiation of the naturally occurring nuclides in the abiotic materials (seawater and sediment) is assessed through the external dose rates received per habitat of these organisms. The absorbed dose rates were calculated on the basis of two assumptions: (1) an infinitive space (4π geometry) for the population habitat marine organism in seawater and sediment; and (2) two semi-infinitive spaces (2π geometry) of different activity, with the organisms being considered to live in the seawater–sediment interface. The formulae used to calculate the required dose rates are in accordance with the respective equations cited in the literature (UNSCEAR, 1982, 1988; Nero, 1983; Papastefanou et al., 1984; Lin et al., 1987; Amiro, 1997) and are based on the general Eq. (2), derived by the equation reported by Henry (Henry, 1969).

$$D = \left(\frac{dE_a}{dt}\right)\left(\frac{1}{M}\right) \quad (2)$$

where D is the absorbed dose rate in an infinitive space ($Gy\ s^{-1}$); E_a is the absorbed energy (Joule); and M is the mass of elemental space (kg).

This equation is adapted for each case of the habitat considered as described below.

In the 4π geometry of the *seawater* and *sediment* phases, the dose rates D_{sea} ($Gy\ s^{-1}$) and D_{sed} at any point in these phases, respectively, are given by the formulae

$$D_{sea} = 1.6 \times 10^{-16} A_{sea}(i) \sum_j E_{\gamma j} I_{\gamma j} \quad (3)$$

and

$$D_{sed} = 1.6 \times 10^{-16} A_{sed}(i) \sum_j E_{\gamma j} I_{\gamma j} \quad (4)$$

where $A_{sea}(i)$ and $A_{sed}(i)$ is the activity concentration ($Bq\,l^{-1}$ and $Bq\ kg^{-1}$) of the radionuclide i determined in the seawater and sediment samples analysed, $E_{\gamma j}$ (keV) is the energy of the jth gamma-radiation

emitted by the radionuclide i and $I_{\gamma j}$ is the intensity of gamma-quanta of the energy $E_{\gamma j}$ per disintegration of the radionuclide i.

The dose rate $D_{\text{sea-air}}$ (Gy s^{-1}) at the *seawater–air* intermediate phase, in the case of the 2π semi-infinitive radioactive space (*seawater*) and 2π semi-infinitive non-radioactive space (*air*), is given by the formula

$$D_{\text{sea-air}} = 8 \times 10^{-17} A_{\text{sea}}(i) \sum_j E_{\gamma j} I_{\gamma j} \tag{5}$$

where $A_{\text{sea}}(i)$ is the activity concentration (Bq l^{-1}) of the radionuclide i determined in the seawater samples analysed, $E_{\gamma j}$ (keV) is the energy of the jth gamma-radiation emitted by the radionuclide i and $I_{\gamma j}$ is the intensity of gamma-quanta of the energy $E_{\gamma j}$ per disintegration of the radionuclide i.

The dose rate $D_{\text{sed-sea}}$ (Gy s^{-1}) at the *seawater–sediment* intermediate phase, in the case of the 2π semi-infinitive radioactive space (*seawater*) and 2π semi-infinitive radioactive space (*sediment*), is given by the formula

$$D_{\text{sed-sea}} = 8 \times 10^{-17} \left[A_{\text{sea}}(i) \sum_j E_{\gamma j} I_{\gamma j} + A_{\text{sed}}(i) \sum_j E_{\gamma j} I_{\gamma j} \right] \tag{6}$$

where $A_{\text{sea}}(i)$ and $A_{\text{sed}}(i)$ are the activity concentrations (Bq l^{-1} and Bq kg^{-1}) of the radionuclide i determined in the seawater and sediment samples, respectively; $E_{\gamma j}$ (keV) is the energy of the jth gamma-radiation emitted by the radionuclide i; and $I_{\gamma j}$ is the intensity of gamma-quanta of energy $E_{\gamma j}$ per disintegration of the radionuclide i.

3. Results and discussion

The results obtained for the three areas of interest in Greece are presented on the basis of: (1) the abiotic materials analysed and (2) the naturally occurring radionuclides and their decay products measured. Activity concentrations (Tables 1–3) and external doses to humans and marine biota (Tables 5 and 6) are included in the results. Activity concentrations in other Greek regions and internationally have been included in Table 4 for comparison purposes (ICRP, 1977; E.R.L., 1989; Probonas, 1992; Trabidou, 2004).

3.1. Activity concentrations of natural gamma-emitters in the abiotic materials sampled

The range of concentrations of the naturally occurring radionuclides measured in soil, sediment, spring water and seawater samples from the Milos, Ikaria and Loutraki sites are shown in Tables 1–3, respectively. It should be noted that the values for Ikaria refer to the wide area of the island as well as in the neighbourhood of the springs.

Table 1
Range of activity concentrations of the natural radionuclides in abiotic materials sampled in Milos island (Bq kg^{-1} and *Bq l^{-1})

Nuclides Sample	^{238}U	^{226}Ra	^{232}Th	^{40}K
Soil	≤1[a]–1420	21–101	26–94	458–1796
Sediment (coastal)	85–127	34–69	26–86	429–1352
Seawater*	32×10^{-3}–42×10^{-3}	1.5×10^{-3}–1.6×10^{-3}	0.2×10^{-4}–0.3×10^{-4}	9.8–11.8

[a] Low limit of detection.

Table 2
Range of activity concentrations of the natural radionuclides in abiotic materials sampled in Ikaria island (Bq kg^{-1} and *Bq l^{-1})

Nuclides Sample	^{238}U	^{226}Ra	^{228}Ra	^{232}Th	^{222}Rn	^{40}K
Soil	8–1049	17–422	–	5–296	–	238–1439
Sediment (coastal)	7–1044	10–760	–	10–80	–	194–2653
Spring water*	<0.1–13	<0.1–1.1	<0.1–2.4	–	114–2468	19–24
Seawater*	≤0.1	≤0.1–1.9	<0.1–1.1	–	13–35	19–20

Concerning the *soil* samples, elevated concentration values in comparison to other Greek regions have occurred for: ^{238}U in Milos and Ikaria, ^{226}Ra in Ikaria, ^{232}Th in Milos and Ikaria and ^{40}K in Milos. Among these samples, the highest concentration of ^{238}U is found in Milos and of ^{232}Th in Ikaria. It should be noted that in the international literature, higher maxima are reported for ^{226}Ra (Trabidou, 2004).

Concerning the s*ediment* samples, elevated concentrations of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K occurred in Milos and Ikaria, with respect to other Greek regions. Among these samples, the highest concentration values of ^{238}U, ^{226}Ra and ^{40}K were found in Ikaria, while the concentration of ^{232}Th is comparable in these two islands.

Concerning the *seawater* samples, ^{222}Rn was measured in both Ikaria and Loutraki, with the former containing higher amount. Both ^{226}Ra and ^{228}Ra concentrations are higher in Ikaria than Loutraki. In Tables 2 and 3, it is observed that the seawater sampled in Ikaria contained higher concentration of ^{226}Ra than ^{228}Ra, whereas in Loutraki these concentrations are comparable. On this basis, one can conclude that in Ikaria the ^{238}U series is most abundant than the ^{232}Th series, whereas in Loutraki the series are comparable. Finally, elevated activity concentrations of ^{226}Ra and ^{228}Ra, with respect to values quoted in the international literature (Trabidou, 2004), were found in *spring water* in the vicinity of geothermal springs in these two islands. The activity concentrations of ^{238}U, ^{222}Rn and ^{228}Ra in spring water are higher in the samples from Ikaria compared to Loutraki.

3.2. Dose rates to exposed humans

The external annual dose rates obtained from *in situ* gamma-radiometry carried out on soil in the three areas analysed are summarized in Table 5. The range of the annual dose rates in other regions of Greece is also included in Table 5 for comparison purposes (Kritidis and Florou, 1989; Florou and Kritidis, 1992; Anagnostakis et al., 1996). High external annual doses in the areas of Milos and Ikaria are found, in relation to other Greek regions, through the *in situ* gamma-radiometry.

Table 3
Range of activity concentrations of the natural radionuclides in abiotic materials sampled in Loutraki (Bq kg^{-1} and *Bq l^{-1})

Nuclides Sample	^{238}U	^{226}Ra	^{228}Ra	^{232}Th	^{222}Rn	^{40}K
Soil	17–29	18–22	–	15–25	–	230–310
Sediment (coastal)	17–25	11–16	–	2.5–13	–	159–277
Spring water*	≤0.1–5	0.1–0.8	≤0.1–0.5	–	42–96	4–13
Seawater*	≤0.1	0.1–0.5	<0.1–0.5	–	47–96	6–13

Table 4
Range of activity concentrations of the natural radionuclides in abiotic materials in other Greek regions and internationally (Bq kg^{-1} and *Bq l^{-1})

Nuclides Sample	^{238}U	^{226}Ra	^{228}Ra	^{232}Th	^{222}Rn	^{40}K
Soil	(5–190) [6.3–53]	(7–310) [10–3700]		(20–88) [6.3–221]		(30–1440) [48–1586]
Sediment (coastal)	(10–54) [0.43–67]	(6–50) [5–1720]		(4–47) [0.53–96]		(47–627) [10.98–653]
Spring water*		0.01–6.2[a]	0.03–0.07[a]		0.8–1780[a]	
Seawater*	(–) [48×10^{-3}–53×10^{-3}]	(–) [1.2×10^{-3}–1.6×10^{-3}]	(–) [3.7×10^{-5}–37×10^{-5}]			(–) [118]

( ): Greek regions other than those considered in this study; [ ]: international values.
[a] Greek and international literature (Trabidou, 2004).

Furthermore, the external annual gamma doses due to soil, seawater, and sediments, calculated according to Eq. (1) on the basis of laboratory gamma-spectroscopy, are given in Table 5. The annual dose rates in Greek regions are also included in Table 5 for comparison purposes (Kritidis and Florou, 1989; Florou and Kritidis, 1992). The doses, due to sediment and soil, are higher in the vicinity of the radioactive springs in the island of Ikaria, related to other Greek areas. The external gamma dose rates to humans exposed to *seawater* while swimming are higher in Ikaria and Loutraki than in Milos and the other areas of Greece.

3.3. Dose rates to exposed marine organisms

The dose rates to exposed natural populations in the marine ecosystem have been calculated using Eqs. (3)–(5), and are presented in Table 6. In this respect, dose rates due to the natural gamma-radiation from *seawater*, *sediments* and in the *intermediate phases in seawater–air* and *seawater–sediment* are given. The corresponding values for the Greek marine environment are included for comparison purpose (Florou, 1992).

Comparing the exposure dose rates due to *seawater* and *seawater–air intermediate phase* for the studied areas, it is noted that the values estimated in Ikaria and Loutraki are higher than those in Milos. Nevertheless, all values are within the background in the Greek marine environment. Considering the respective dose rates from *sediments* and *sediment–seawater*

Table 5
Annual gamma doses to humans due to external exposure

Area	*In situ* gamma-radiometry (μSv y^{-1})	Gamma-spectrometry		
		Soil (μSv y^{-1})	Sediment (μSv y^{-1})	Seawater[c] (μSv y^{-1})
Milos	245–1226	219–701	263–964	0.14–0.18
Ikaria	307–1410	622–3136[a] 175–289[b]	639–4318[a] 219–377	6.5–15.2
Loutraki	60–187	44–133	63–186	2.3–6.7
Other Greek areas	100–1000	201–797	447–736	0.12–0.14

[a] Values concerned the vicinity of the radioactive spring.
[b] Wider area in Ikaria.
[c] On the assumption of 35-h swimming per year.

Table 6
Dose rates to marine organisms due to natural gamma-radiation in the marine ecosystem (μGy d^{-1})

Area	Seawater	Seawater–air intermediate phase	Sediment	Sediment–seawater intermediate phase
Milos	0.064×10^{-3}–0.0986×10^{-3}	0.0326×10^{-3}–0.05×10^{-3}	1.46–5.82	0.71–2.7
Ikaria	<0.045–0.104	<0.0224–0.052	1.76–26.8	0.9–13.5
Loutraki	<0.0156–0.0462	0.008–0.023	0.49–1.7	0.36–0.9
Greek marine environment	0.081–0.098	0.0406–0.05	0.38–7	0.19–3.5

intermediate phase, higher values are found in Ikaria in comparison to Milos and Loutraki, where the values are of the typical background for the Greek marine environment.

As it is known, the effects of the exposure dose rates received by marine biota depend on the radiosensivity of each exposed organism. It should be recalled that metabolic malfunctions in organisms occur due to a chronic radiation dose exposure of 1×10^4 μGy d^{-1} (Tsytsugina and Polikarpov, 2003). For a better estimation of the effects on the abiotic components in the studied areas due to exposure to the environmental radiation, the model by Polikarpov for the chronic effects of ionizing radiation is employed (Polikarpov and Tsytsugina, 1996).

According to this model, the range of the calculated dose rates in *seawater* and *seawater–air intermediate phase* are given in Table 6. The range is comparable to the values encountered in the marine environment of Greece (Florou, 1992) and within the 'uncertainty zone for dose rates' (2.7×10^2–1×10^1 μGy d^{-1}). In the case of *sediments* and *sediment–seawater intermediate phase* (Table 6), the range of calculated dose rates in Ikaria is higher than the values encountered in the marine environment of Greece (Florou, 1992; E.R.L., 2003). These values are within the 'physiological masking zone' (1.4×10^1–1.4×10^2 μGy d^{-1}). The dose rates were calculated on the basis of the measured radionuclides ^{238}U, ^{226}Ra, ^{228}Ra, ^{232}Th and ^{40}K. The nuclide ^{226}Ra is the main contributor to these dose rates, which is in accordance with its high concentrations in sediments and seawater encountered in Ikaria (Table 2). The dose rates contribution of the other radionuclides is within the 'uncertainty zone for dose rates' and the 'background zone of radiation well being' (<1.4×10^1 μGy d^{-1}). The dose rates are in the area of overlap between minor radiation effects and natural variability in physiological function. This zone probably covers a dose rate range in which it is possible to detect minor changes in function, morphology, and an increase in the incidence of diseases, e.g. decrease in the weight of the spleen of voles in regions of high natural radioactivity (Polikarpov, 1998).

4. Conclusions

The activity of the terrestrial radionuclides and their decay products has been measured in soil, sediment, spring water and seawater as well as through gamma-radiometry in three areas in Greece. Gamma-spectroscopy, both *in situ* and in laboratory, was employed for the measurements. Furthermore, an assessment of the external radiological impact on the population due to the naturally occurring radionuclides has been made.

The geological origin of the three studied areas, characterized by the presence of geothermal springs and vents seems to have an apparent influence on the concentrations of natural

radionuclides in the abiotic components resulting in elevated values compared to other areas of Greece and worldwide. Therefore, the metallic springs can be considered responsible for carrying natural radionuclides to the environmental abiotic components of the areas studied, especially for the island of Ikaria. Hence, the external dose rates to the exposed population in Milos and Ikaria were elevated.

The dose rates to the exposed marine organisms range in between the 'uncertainty zone for dose rates' and the 'physiological masking zone'. The maximum value of the dose rate observed in the case of sediment in Ikaria (26.8 μGy d^{-1}) does not reach the threshold reported for metabolic malfunctions in the exposed organisms. Nevertheless, these studies can be used as a baseline for research on the effects of chronic exposure in non-intervention radiation levels.

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