RESEARCH ARTICLE

Radioactivity measurements and dose rate calculations using ERICA tool in the terrestrial environment of Greece

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Abstract In the present study, the radioactivity levels to which terrestrial non-human biota were exposed are examined. Organisms (grass and herbivore mammals) and abiotic components (soil) were collected during the period of 2010 to 2014 from grasslands where sheep and goats were free-range grazing. Natural background radionuclides (^{226}Ra, ^{228}Ra, ^{228}Th) and artificial radionuclides (^{137}Cs, ^{134}Cs, ^{131}I) were detected in the collected samples using gamma spectrometry. The actual measured activity concentrations and site-specific data of the studied organisms were imported in ERICA Assessment Tool (version 1.2.0) in order to provide an insight of the radiological dose rates. The highest activity concentrations were detected in samples collected from Lesvos island and the lowest in samples collected from Attiki and Etoloakarnania prefectures. The highest contribution to the total dose rate was clearly derived from the internal exposure and is closely related to the exposure to alpha emitters of natural background (^{226}Ra and ^{228}Th). The Fukushima-derived traces of ^{137}Cs, ^{134}Cs, and ^{131}I, along with the residual ^{137}Cs, resulted in quite low contribution to the total dose rate. The obtained results may strengthen the adaptation of software tools to a wider range of ecosystems and may be proved useful in further research regarding the possible impact of protracted low level ionizing radiation on non-human biota. This kind of studies may contribute to the effective incorporation of dosimetry tools in the development of integrated environmental and radiological impact assessment policies.

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Introduction

In recent years, more attention has been given to the assessment of radiological impact on non-human biota taking into account that human protection may be inadequate to ensure environmental protection (ICRP 2003, 2007). Significant consideration has been given to the estimation of non-human biota radiological dose rates, independently from dose rates calculations for humans. The quantification of non-human biota exposure to ionizing radiation may be performed applying specific methodologies and software that were developed over the last years (Beresford et al. 2007, 2008; Brown et al. 2008; IAEA 2010, 2014b; ICRP 2008, 2009; USDoE 2002, 2004; Wood et al. 2009).

Within the 6th Framework Program of the European Commission, the ERICA Integrated Approach (ERICA I.A.) and the ERICA Assessment Tool were developed for calculation of radiological dose rates, risk characterization and prediction of eventual effects of ionizing radiation to non-human biota (Beresford et al. 2007; Brown et al. 2008; Larsson 2008; ERICA 2014). The ERICA I.A. and ERICA Tool may provide a valuable methodology for the integrated radiological impact assessment of a region on the ecosystem level. Thus, over the last years, several research works have studied its application on specific populations and ecosystems using actual radioactivity measurements (e.g., Karimullina et al. 2013;

Nedveckaite et al. 2011; Vetikko and Saxén 2010; Wood et al. 2008, 2009).

Natural radioactivity levels in the terrestrial environment of Greece are generally known (mainly referring to members of ^{238}U and ^{232}Th natural series) (Florou et al. 2007; Kioupi et al. 2015; Probonas and Kritidis 1993). However, there is no information concerning the levels of internal and external exposure to which natural terrestrial organisms are subjected taking into consideration that terrestrial natural radioactivity may be a significant source (Probonas and Kritidis 1993).

Artificial radionuclides have been introduced in the terrestrial environment of the country mainly from the Chernobyl nuclear power plant accident and the global fallout with ^{137}Cs still detectable in the environment (Kritidis and Florou 1995, 2001). Additionally, in 2011 traces of artificial radionuclides were also introduced by the radioactive materials released from Fukushima Dai-Ichi nuclear power plant accident. Radioactive traces were also detected in air over Greece (Athens, Thessaloniki, and Ioannina) (Kritidis et al. 2012; Manolopoulou et al. 2011; Potiriadis et al. 2012) and it can be assumed that traces of ^{137}Cs that resulted from the Fukushima accident were added to the residual ones. Concerning ^{134}Cs and ^{131}I, they were detected again in 2011 and it is clearly assumed that they are originated from Fukushima, since there was a long time intervened from their last presence at the early period after the Chernobyl accident.

The aim of the present work was to characterize the baseline levels of natural and artificial radioactivity in semi-natural areas of the Greek rural territory. Actual radioactivity measurements were performed in organisms and abiotic components in regions of free-range grazing, since there is a general lack of information concerning the radiological exposure of natural organisms in the country. The objective was to identify the process that is related to the calculation of radiological dose rates to non-human biota, in order to contribute to the development of the radiological impact assessment procedure. The application of ERICA Tool towards this objective may reveal insights related to the extents or limitations of its application in similar environments.

Materials and methods

Sampling locations

During the period from 2010 to 2014, a total sum of 52 soil, grass, and mammals' tissue samples were collected from five randomly chosen sampling areas (SA) in Greece (Fig. 1). The specific case study sites were chosen because they are representative of the semi-natural environment of the country where uncultivated grasslands are used for free-range grazing of herbivore mammals. Furthermore, at these locations, we could gain accessibility to the remote grasslands and to the

shepherds and slaughter houses of the regions. From each case study site, samples of soil, grass, and mammals were collected.

The studied regions are mainly covered by phryganic ecosystem (similar to the Garrigue vegetation) which is one of the typical vegetation of the Mediterranean type ecosystem developed at the most arid and warm margins of the region. The vegetation at these regions consists mainly of woody dwarf shrubs (shorter than 1 m height) and from grasses of the Poaceae family (Margaris 1976, 1981; Margaris and Vokou 1982). A significant part of the rural territory in Greece is also covered by grasslands (Merou and Papanastasis 2009; Papanastasis 1981). The phryganic communities are important grazing lands for livestock, mainly sheep in the autumn–winter time and are extensively used for free range grazing of livestock, mainly sheep and goats (Arianoutsou-Faraggitaki 1985; Giourga et al. 1998). At the studied regions loam and silt clay loam soils are widespread (JRC 2001).

The studied plants were grasses of the Poaceae family (formerly Gramineae, order of Monocotyledons of the Angiosperms class), with great abundance at the phrygana vegetation sites and at the grasslands, which are the main pasture of the grazing animals at these regions. The collected mammals (sheep and goats) were primary herbivore mammals, ruminants of the Bovidae family, mainly used for human consumption. The classification used in this study is at the taxonomic order of family, in accordance to the taxonomic class used in the Reference Organism (RO) module of the ERICA Tool (ERICA 2014) and to the Reference Animals and Plants (RAP) of ICRP Publication 108 (ICRP 2008). The choice of sheep and goats for the conduction of this work was due to the fact that the chosen organisms are representative species of the terrestrial semi-natural Mediterranean ecosystem and significant part of human food chain. Furthermore, the access to sufficient number of samples could be obtained through official slaughter houses.

Sampling and treatment procedures

Soil samples were collected in a polythene bag using a split-blade corer, covering an area of 1 m^2 where 4–6 sampling points resulted to a composite sample representative for each site. Samples were collected from 0 to 10 cm soil depth which is the extent of the rooting system of most species of the Poaceae family, and is also considered the standardized soil zone for the definition of soil-to-plant transfer factors (IAEA 2009). Extraneous materials like roots, leaves, pieces of gravel and stones were removed from the samples and samples were weighted to wet mass, dried at 100 °C for 14 h and weighted again to dry mass. Samples were homogenized, sieved through 2-mm sieve, separated to subsamples, and transferred to plastic pots of 70 mm diameter and 20 mm height to be weighted again.
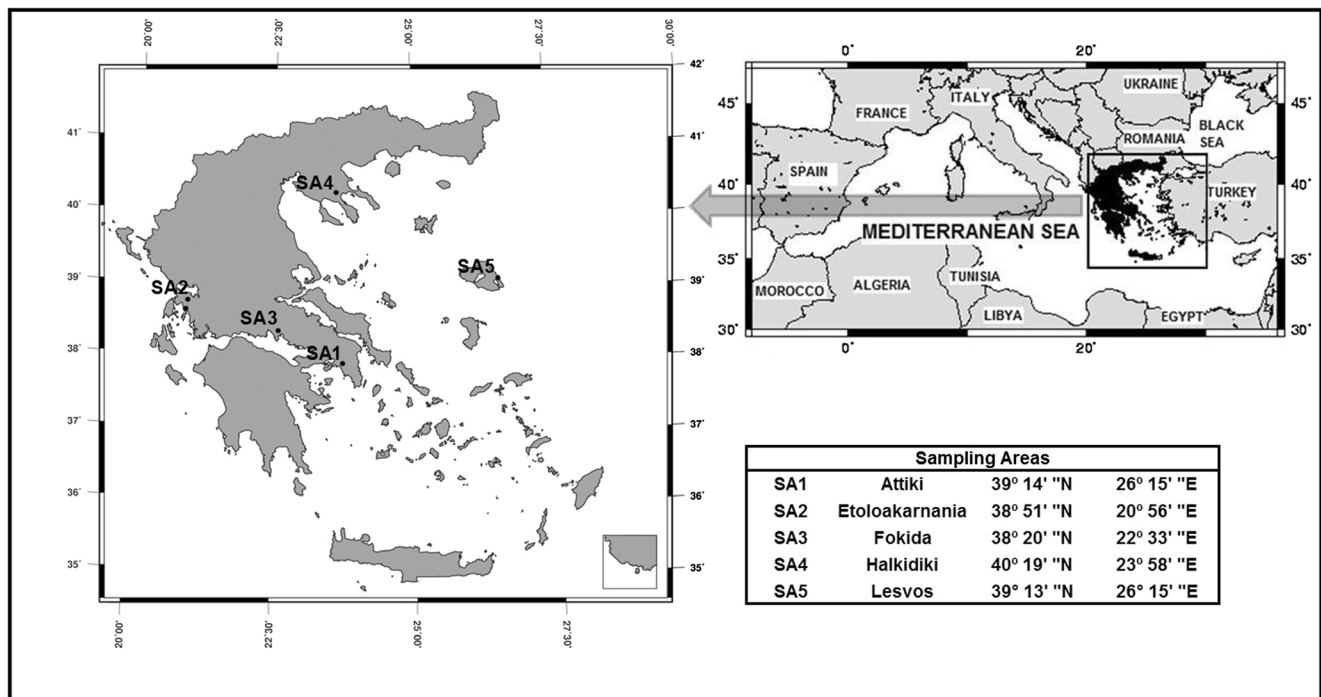
Fig. 1 Map of Greece indicating the sampling areas. Sampling areas with coordinates are numbered as showed in the adjacent table

Soil and grass samples were collected from the areas where herbivores were grazing, using the same coverage. The above ground plants' body was kept for measuring and roots were removed from the samples, as they are not taken into consideration in the dosimetric calculations. The collected grass samples were washed with distilled water, weighted to fresh mass, and transferred to porcelain dishes where they were dried at 200 °C for 5 h and at 250 °C for 7 h. Afterwards, the samples were weighted again to dry mass, homogenized, and transferred to the above-mentioned plastic pots.

The studied mammals (sheep, *Ovis aries*, and goats, *Capra aegagrus*) were collected from accredited slaughtering houses located close at the grazing areas (Fig. 1). Specific tissues were sampled (muscle, bones, and organs) as they are the major target tissues for specific radionuclides (e.g., muscle for ^{137}Cs, bone for ^{226}Ra, etc.). In order to ensure sufficient material for analysis, muscle and bone samples comprised of about 2 kg of tissue, while organ samples were composed from lungs, thyroid, kidneys, heart, spleen, and liver (gastrointestinal tract was excluded). Tissue samples were immediately frozen after sampling, when needed, in order to prevent tissues decomposition. It has to be noted that samples of grass and mammals' tissues that were collected during the first month of the Fukushima impact to Greece were sliced into small pieces, in order to fit into Marinelli bakers of 1 L, and measured immediately after sampling for the detection of the short-lived ^{131}I. Afterwards, tissue samples were dried in order to obtain the best counting geometry for gamma spectrometry. Sliced samples were weighted to fresh mass, dried to 300 °C to remove organic matter, weighed to dry mass,

grounded, and homogenized. Subsamples were transferred to the appropriate aforementioned plastic pots.

The Environmental Radioactivity Laboratory (ERL) of the National Centre for Scientific Research 'Demokritos' (Greece) has adopted protocols and methods (briefly described above) from the relevant international literature (IAEA 1989; Klement 1982; USDoE 1997).

Gamma spectrometry measurements

The determination of activity concentration of natural and artificial gamma emitting radionuclides, in soil, grass, and mammals' tissues was performed with gamma spectrometry. All samples were measured for natural and artificial gamma-emitting radionuclides using two high-resolution gamma ray spectrometry systems. The first system consists of a high-purity Germanium (hpGe) coaxial detector of 91.5 % relative efficiency and 1.99-keV resolution at the 1.33-MeV photopeak of ^{60}Co. The system is connected to an 8-k multi-channel analyzer (MCA) with an energy calibration of 0.25 keV/channel operated with Canberra Gennie 2000 software used for the spectrum analysis. The second system consists of a 20 % relative efficiency hpGe coaxial detector with an energy resolution of 1.9 keV at 1.33 MeV (of ^{60}Co) connected to an 4 k MCA with an energy calibration of 0.5 keV/channel operated with Ortec Maestro II software. Both systems are carefully shielded, in order to reduce the influence of background radiation.

For the determination of ^{226}Ra the photopeaks of ^{214}Pb (295.2 and 352.0 keV) and ^{214}Bi (609.4 keV) were used, for

^{228}Ra the photopeaks of ^{228}Ac (338.4 and 911.1 keV) and for ^{228}Th the photopeaks of ^{212}Pb (238.6 keV) and ^{208}Tl (583.1 keV). For the artificial radionuclides ^{137}Cs, ^{134}Cs, and ^{131}I, the photopeaks used were 661.6, 604.7, and 364.5 keV, respectively. All samples were kept sealed before measuring for at least 24 days in order to ensure radioactive equilibrium between ^{226}Ra and its daughter nuclides. The measuring duration was around $2 \cdot 10^5$ seconds. The activity concentrations are reported with 2σ confidence level propagated uncertainty and the relative statistical error is up to 30 %. Samples whose activity concentration was below the minimum detectable activity (MDA) were excluded from subsequent statistical analysis and further consideration.

The energy and efficiency calibration of both hpGe systems in ERL performed using standard sources of the same geometry and density as the samples. The standard sources used were appropriate to cover the full energy range of up to 2000 keV; a source of ^{226}Ra, prepared from an original standard solution of 3.7 kBq L^{-1} and a multi-nuclide source of ^{241}Am, ^{137}Cs, and ^{60}Co of 584 Bq total activity (produced by Institute for Nuclear Research and Nuclear Energy, Sofia, Bulgaria). Quality control of the results is constantly performed by participating in national and international interlaboratory comparison and calibration exercises and in proficiency tests (e.g., IRMM 426, IAEA 375, IAEA CRP1471-01, IAEA-ALMERA-TEL-2014-04, etc.).

Application of ERICA Assessment Tool

The model that is currently used is the ERICA Assessment Tool (version 1.2.0), which is the supporting software of the ERICA Integrated Approach (ERICA I.A.). The ERICA I.A. and the ERICA Tool have been developed under the 6th Framework Program of the EC for the assessment and management of environmental risks from ionizing radiation (Beresford et al. 2007; Brown et al. 2008; Larsson 2008; ERICA 2014).

At the present study, the second Tier of the ERICA Tool was implemented because it is site-specific dependent and user-defined data can be introduced. The site-specific data refer to the radioactivity measurements of soil and biota and to the dimensions and mass of the mammals that were sampled. Tier 1 was not used due to the fact that it is a screening Tier in which only the default parameter datasets can be used. Due to the absence of any interaction of animals with humans (i.e., animals were not domesticated or breed) it is assumed that the ERICA Tool may be applied at a semi-natural environment. As mentioned above, the studied radionuclides were ^{226}Ra of ^{238}U natural series and ^{228}Ra and ^{228}Th of ^{232}Th natural series detected in the case study sites. The studied artificial radionuclides were ^{137}Cs, which is from the Chernobyl and Fukushima accidents and from global fallout, and ^{134}Cs and ^{131}I that are Fukushima-derived.

Within ERICA Assessment Tool dose rates are calculated on the basis of external (D_{ext}), internal (D_{int}), and total (D_{tot}) absorbed dose rate (in μGy h^{-1}). The method of dose conversion coefficients (DCC in μGy h^{-1} per Bq kg^{-1}) is applied, which is based on the calculation of dose to an organism, based on the activity concentration in which the organism is exposed (Brown et al. 2003, 2008; ERICA 2014; Prohl et al. 2003; Ulanovsky et al. 2008). The calculation of D_{ext} is performed using the activity concentration in soil (C_{media}), assuming that organism is exposed in uniformly distributed radionuclides in soil and using the typical source–target relation of external exposure, multiplied by the respective dose conversion coefficient for external exposure (DCC$_{ext}$). The calculation of D_{int} is performed using organism activity concentration ($C_{organism}$) multiplied by dose conversion coefficient of internal exposure (DCC$_{int}$), assuming homogenous distribution of radionuclides within organism's body and that all relevant pathways of internal exposure have been taken into consideration (ingestion, inhalation, and water uptake for mammals and root and foliar uptake for grass). Additionally, the radiation weighting factors (wf) had to be defined, whereas in this study the default values of ERICA Tool were applied (10 for α, 3 for β, and 1 for γ radiation) (ERICA 2014; Prohl et al. 2003). The weighted total dose rate (D_{tot}) for each radionuclide considered (i) is the sum of D_{int} and D_{ext}, as it is shown in the following equation, while the $\sum D_{tot}$ is the total dose rate from all radionuclides considered.

$$\sum_i D_{tot} = \sum_i \left(D_{ext,i} + D_{int,i} \right)$$
$$= \sum_i \left(OFC_{media,i} \, wf DCC_{ext,i} + C_{organism,i} \, wf DCC_{int,i} \right)$$

For the representation of grass samples, the default reference organism (RO) "Grasses and Herbs" was used which is based on "Wild Grass" of the ICRP list of Reference Animals and Plants (RAP) (ICRP 2008). The DCC$_{ext}$ and DCC$_{int}$ that correspond to each organism–radionuclide combination were used in the calculation of D_{ext} and D_{int}, multiplied by the mean activity concentrations of soil and grass samples from each sampling area.

The introduction of sampled mammals within the Tool was performed through the "Add Organism Wizard". Once all necessary information about the new organism (Sheep5) was imported in the Tool (taxonomic characteristics, dimensions, mass, etc.) the ellipsoid phantom was formed and the relevant DCC$_{ext}$ and DCC$_{int}$ were calculated (Table 1). The DCCs of any new organism are calculated based on the absorbed energy fraction which is estimated after the Monte Carlo calculations performed within the Tool. The DCCs include the contribution of the daughter radionuclides if their half-lives are shorter than 10 days, assuming that they are in secular

Table 1 Data imported within ERICA Tool for the terrestrial mammal and the respective calculated DCC_{int} and DCC_{ext} (in µGy h^{-1} per Bq kg^{-1})

Name	Sheep5		DCCext		DCCint	
			Beta-gamma	Beta-gamma	Low beta	Alpha
Ecosystem	Terrestrial	^{226}Ra	2.47E-4	8.68E-4	1.63E-6	1.38E-2
Wildlife group	Mammal	^{228}Ra	1.36E-4	4.60E-4	9.14E-6	
Specimen	Ground-living animal	^{228}Th	2.15E-4	7.72E-4	3.73E-6	1.85E-2
Mass (kg) Dimensions (m)	20 Height: 0.4; width: 0.4; lenght: 0.8	^{137}Cs	7.99E-5	2.57E-4	3.71E-7	
		^{134}Cs	2.21E-4	4.03E-4	3.59E-7	
Occupancy Factor	1 - on ground	^{131}I	5.27E-5	1.92E-4	2.82E-7	

equilibrium with the parent radionuclide (Taranenko et al. 2004; Ulanovsky et al. 2008).

For the calculation of D_{ext}, the DCC_{ext} was multiplied by the arithmetic mean of the activity concentration measured in soil samples of each sampling area. In order to calculate mammals internal exposure, the measured activity concentration in the sampled body parts (muscle, bones, and organs) was converted to whole-body activity concentration applying the appropriate conversion factors derived from the literature (IAEA 2014a; Yankovich et al. 2010). This is due to the fact that in the biota dose assessment models the parameters and benchmark values usually refer on whole-body basis (CR, DCC, etc.) and the available dose-effect data for radionuclides are mostly based on a whole-body exposure. The D_{int} was calculated through the DCC_{int} multiplied by the mean activity concentration in organism for each sampling area on whole-body basis. For both grass and mammals, the time that the organism spent at a specific location in its habitat (which is represented by the occupancy factor (OF) within the tool) was set to unity on the ground.

Results and discussion

Radioactivity measurements in the terrestrial environment

The measured activity concentrations of natural radionuclides in soil (Table 2) exhibited a good agreement with measurements performed earlier at the country. The mean value of ^{226}Ra activity concentration in soil samples calculated equal to 32 ± 18 Bq kg^{-1} ($n = 16$) which is within the range of 7–310 Bq kg^{-1} (mean value equal to 40 ± 17 Bq kg^{-1}) reported in previous studies performed in the country (Florou et al. 2007; Probonas and Kritidis 1993). The mean value of ^{228}Ra in soil was equal to 48 ± 28 Bq kg^{-1} ($n = 16$), which is also within the reported values from Probonas and Kritidis (1993) of 7–190 Bq kg^{-1}, who reported a mean of 47 ± 17 Bq kg^{-1}. They also reported a mean of 33 ± 12 Bq kg^{-1} for ^{228}Th (3–150 Bq kg^{-1}) which is close to the mean calculated for this study at 42 ± 25 Bq kg^{-1} ($n = 16$). As shown in Table 2, the

concentrations of naturally occurring radionuclides in soil (^{226}Ra, ^{228}Ra, and ^{228}Th) exhibited some variations which are generally correlated with the chemical composition of soils from region to region (Probonas and Kritidis 1993).

Natural radionuclides detected in plants due to the transfer from soil through root uptake and to foliar absorption of the resuspended radionuclides. The mean activities in grass samples from all sampling areas were equal to 1.9 ± 1.6 Bq kg^{-1} ($n = 11$) for ^{226}Ra, to 5.0 ± 7.0 Bq kg^{-1} ($n = 12$) for ^{228}Ra and to 1.0 ± 0.7 Bq kg^{-1} ($n = 11$) for ^{228}Th (Table 2). It should be noted that the higher activities in soil and grass were detected in samples collected from SA5 (Lesvos island) and the lower activities in samples from SA2 (Etoloakarnania).

In soil samples, the mean activity ratio of ^{228}Th/^{232}Th was 0.93 ($n = 16$) which implies secular equilibrium among the members of ^{232}Th decay series. The equilibrium is also supported by the activity ratio of ^{228}Th/^{228}Ra that was calculated equal to 0.87 (Fig. 2a). On the other hand, low degree of secular equilibrium was calculated for grass samples. The mean ^{228}Th/^{232}Th activity ratio was 0.49 ($n = 10$) and the ^{228}Th/^{228}Ra ratio was 0.36, furthermore, ^{228}Th activity concentration in grass was about four times lower than ^{228}Ra activity concentration (Fig. 2b). The low degree (or absence) of equilibrium in organisms can be attributed to radionuclides' behavior within organisms (transfer, uptake, accumulation, etc.) which may be affected, to a greater or lesser degree, by the biological processes of organisms, soil characteristics and elemental and isotopic properties of radionuclides (e.g., radiogenic profile of ^{228}Th) (Sheppard et al. 2008; Vera Tomé et al. 2002).

During the conduction of the sampling campaigns that had been already planned, the Fukushima Nuclear Power Plant accident occurred on March 2011. Thus, radionuclides released in the atmosphere were also detected in environmental samples (Manolopoulou et al. 2011; Kritidis et al. 2012; Potiriadis et al. 2012). The Fukushima-derived ^{134}Cs and ^{131}I were detected in grass samples with mean activity concentration of 0.2 ± 0.2 Bq kg^{-1} ($n = 6$) and 1.3 ± 0.8 Bq kg^{-1} ($n = 3$), respectively.

Traces of ^{137}Cs were detected in grass samples that were collected before and after the Fukushima impact in Greece. In 2 grass samples collected prior to the accident ^{137}Cs

Table 2 Mean activity concentrations of ^{226}Ra, ^{228}Ra, ^{228}Th, ^{137}Cs, ^{134}Cs, and ^{131}I in samples of soil and grass (in Bq kg^{-1}) for each case study site

	Nuclide	Sample	
		Soil	Grass
SA1	^{226}Ra	19 ± 0.4 (18–19) (2)	0.5 ± 0.2 (1)
	^{228}Ra	21 ± 3 (18–23) (2)	<0.07
	^{228}Th	18 ± 3 (16–20) (2)	0.2 ± 0.02 (1)
	^{137}Cs	3 ± 3 (1–6) (2)	0.6 ± 0.3 (0.3–0.8) (2)
	^{134}Cs	<0.02	0.3 ± 0.3 (0.1–0.5) (2)
	^{131}I	<0.04	1.7 ± 0.6 (1.2–2.1) (2)
SA2	^{226}Ra	21 ± 9 (15–27) (2)	<0.04
	^{228}Ra	20 ± 4 (16–23) (2)	0.3 ± 0.2 (1)
	^{228}Th	18 ± 3 (15–20) (2)	<0.04
	^{137}Cs	25 ± 10 (19–32) (2)	0.1 ± 0.03 (0.1–0.2) (2)
	^{134}Cs	<0.02	<0.01
	^{131}I	<0.04	<0.03
SA3	^{226}Ra	30 ± 12 (19–43) (4)	1.7 ± 0.6 (0.9–2.3) (4)
	^{228}Ra	37 ± 11 (25–52) (4)	2.3 ± 1.4 (1.3–4.3) (4)
	^{228}Th	32 ± 10 (22–45) (4)	0.9 ± 0.4 (0.6–1.5) (4)
	^{137}Cs	23 ± 24 (1–57) (4)	0.4 ± 0.2 (0.2–0.7) (4)
	^{134}Cs	<0.02	<0.01
	^{131}I	<0.04	<0.03
SA4	^{226}Ra	83 ± 1 (1)	<0.04
	^{228}Ra	45 ± 2 (1)	0.4 ± 0.1 (1)
	^{228}Th	36 ± 1 (1)	<0.04
	^{137}Cs	27 ± 1 (1)	0.4 ± 0.1 (1)
	^{134}Cs	<0.02	0.1 ± 0.04 (1)
	^{131}I	<0.04	<0.03
SA5	^{226}Ra	33 ± 14 (14–52) (7)	2.2 ± 2.1 (0.3–5.0) (6)
	^{228}Ra	72 ± 24 (37–96) (7)	8.4 ± 8.6 (0.7–20.2) (6)
	^{228}Th	63 ± 22 (29–87) (7)	1.2 ± 0.9 (0.4–2.8) (6)
	^{137}Cs	26 ± 21 (10–61) (6)	0.6 ± 0.5 (0.2–1.5) (6)
	^{134}Cs	<0.02	0.1 ± 0.1 (0.1–0.2) (3)
	^{131}I	<0.04	0.6 ± 0.2 (1)

Soil samples are reported in dry weight and organisms in fresh weight

Samples below the minimum detectable activity denoted by "≤ MDA"

The range of all samples measured and the number of the considered samples for each area are indicated in brackets

For $n=1$ the results are given as activity concentration ± uncertainty due to counting statistics

mean activity concentration was calculated equal to 0.3 ± 0.1 Bq kg^{-1}, while, in 13 samples collected after the arrival of the radioactive cloud ^{137}Cs mean activity concentration was calculated equal to 0.5 ± 0.4 Bq kg^{-1} ($n=15$). According to the measurements, it can be assumed that Fukushima-derived traces were added to the residual ones. This is also supported by the presence of detectable amounts of ^{134}Cs in undisturbed grass. Considering that the measured ^{137}Cs/^{134}Cs ratio in air in

Greece equaled to 1.1 ± 0.3 (Kritidis et al. 2012) it can be assumed that this ratio may be applicable for grass samples as well. Thus, a rough estimation of the Fukushima-derived ^{137}Cs in grass is provided and is equaled to 0.2 Bq kg^{-1}, which is in good agreement with the aforementioned calculated values. It has to be noted that according to the measurements, the ratio of ^{137}Cs/^{134}Cs in grass was equal to 3.4 indicating that ^{137}Cs has an additional origin except Fukushima which is the Chernobyl accident and the global fallout.

In soil samples mean activity concentration of ^{137}Cs equaled to 22 ± 19 Bq kg^{-1} ($n=15$) which is within the range that has been previously reported by Theocharopoulos et al. (2000) and Kioupi et al. (2015) of 17 to 572 Bq kg^{-1}. 134Cs and ^{131}I were not detected in soil indicating that the uptake by grass is attributed to the radioactive deposition on foliar surfaces.

For comparison reasons, studies dealing with the Fukushima impact in European region are indicatively mentioned. The Fukushima-derived ^{134}Cs activities concentrations in grass samples that reported by Baeza et al. (2012) exhibited a mean value of 0.2 Bq kg^{-1}, while Perrot et al. (2012) reported a range of 0.2–0.6 Bq kg^{-1} that were quite close to 0.2 ± 0.2 Bq kg^{-1} of this study. Activity concentrations reports on ^{131}I are also in very good agreement to the mean value of this study (1.3 ± 0.8 Bq kg^{-1}) where Baeza et al. (2012) exhibited a range of 0.4–9.6 Bq kg^{-1} and Perrot et al. (2012) calculated a mean of 1.5–15.4 Bq kg^{-1}. Taking into consideration the peculiarities of ^{137}Cs activities where newly imported traces are mixed with residual ^{137}Cs, the values reported by Baeza et al. (2012) (0.4 Bq kg^{-1}) and Perrot et al. (2012) (0.2-0.7 Bq kg^{-1}) are also in good agreement to the mean value calculated within this study (0.5 ± 0.4 Bq kg^{-1}). In a number of studies, Fukushima-derived traces were detected in a bulk of environmental samples such as air, rainwater, plants, animals and dairy products (e.g., Baeza et al. 2012; Kritidis et al. 2012; Manolopoulou et al. 2011; Perrot et al. 2012; etc.), while, results concerning soil measurements are not reported.

Activity concentrations measured in muscle and bone tissue and in organs are shown in Table 3. Natural radionuclides were detected in bone tissue, with mean activities equal to 2.2 ± 3.0 Bq kg^{-1} ($n=14$) for ^{226}Ra, 4.4 ± 7.3 Bq kg^{-1} ($n=16$) for ^{228}Ra, and 0.8 ± 0.9 Bq kg^{-1} ($n=12$) for ^{228}Th. Radium (^{226}Ra and ^{228}Ra) concentrations in soft tissues (muscle and organs) are lower than in bone. This is attributed to the behavior of radium in mammals that in general is similar to the behavior of calcium (IAEA 2014b). The radiogenic profile of ^{228}Th, assuming a possible decay of ^{228}Ra already present in tissues, may have led to ingrowth of ^{228}Th in tissues. This may cause a slight overestimation of its measured activities. These radionuclides were scarce in soft tissues, where ^{228}Th was detected in 3 samples of muscle (with a mean of 0.1 ± 0.01 Bq kg^{-1}), ^{228}Ra in 1 sample (0.2 ± 0.07 Bq kg^{-1}) and ^{226}Ra in 2 samples of muscle (0.1 ± 0.06 Bq kg^{-1}) and in 1 sample of organs (0.1 ± 0.04 Bq kg^{-1}) (Table 3).
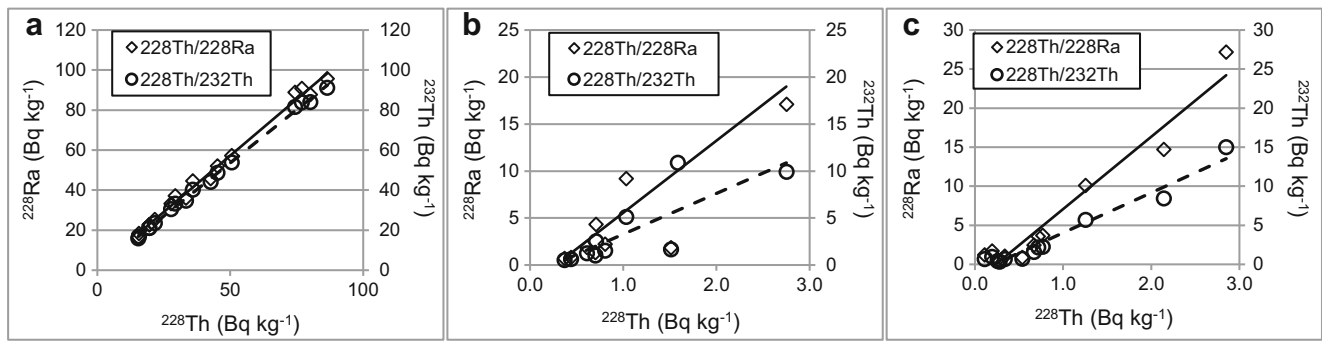
Fig. 2 Activity concentration equilibrium ratios of ^{228}Th/^{228}Ra and ^{228}Th/^{232}Th in **a** soil, **b** grass, and **c** bone tissue

Table 3 Mean activity concentrations of ^{226}Ra, ^{228}Ra, ^{228}Th, ^{137}Cs, ^{134}Cs, and ^{131}I in samples of muscle, bone, and organs (in Bq kg^{-1}) for each case study site

	Nuclide	Tissue sample		
		Muscle	Bones	Organs
SA1	^{226}Ra	<0.03	0.40 ± 0.13 (1)	<0.03
	^{228}Ra	<0.05	0.64 ± 0.07 (0.59–0.69) (2)	<0.05
	^{228}Th	<0.03	<0.03	<0.03
	^{137}Cs	0.26 ± 0.04 (0.23–0.28) (2)	<0.01	0.44 ± 0.12 (1)
	^{134}Cs	0.18 ± 0.06 (0.13–0.22) (2)	0.20 ± 0.08 (1)	0.19 ± 0.18 (0.06–0.32) (2)
	^{131}I	0.28 ± 0.08 (1)	<0.02	1.52 ± 0.25 (1)
SA2	^{226}Ra	0.17 ± 0.05 (1)	0.74 ± 0.03 (0.72–0.76) (2)	<0.03
	^{228}Ra	<0.05	0.79 ± 0.35 (0.54–1.04) (2)	<0.05
	^{228}Th	0.11 ± 0.01 (0.10–0.12) (2)	0.34 ± 0.09 (1)	<0.03
	^{137}Cs	0.18 ± 0.18 (0.05–0.30) (2)	<0.01	<0.01
	^{134}Cs	0.20 ± 0.07 (1)	<0.01	<0.01
	^{131}I	<0.02	<0.02	<0.02
SA3	^{226}Ra	<0.03	0.80 ± 0.11 (0.73–0.93) (3)	<0.03
	^{228}Ra	<0.05	0.66 ± 0.19 (0.44–0.89) (4)	<0.05
	^{228}Th	<0.03	0.36 ± 0.16 (0.26–0.54) (3)	<0.03
	^{137}Cs	0.33 ± 0.21 (0.18–0.48) (2)	<0.01	0.23 ± 0.07 (1)
	^{134}Cs	0.07 ± 0.01 (0.06–0.07) (2)	0.18 ± 0.05 (1)	0.08 ± 0.03 (1)
	^{131}I	<0.02	<0.02	<0.02
SA4	^{226}Ra	<0.03	1.68 ± 0.13 (1)	<0.03
	^{228}Ra	<0.05	1.73 ± 0.30 (1)	<0.05
	^{228}Th	<0.03	0.20 ± 0.07 (1)	<0.03
	^{137}Cs	0.40 ± 0.10 (1)	<0.01	<0.01
	^{134}Cs	<0.01	<0.01	<0.01
	^{131}I	<0.02	<0.02	<0.02
SA5	^{226}Ra	0.09 ± 0.03 (1)	3.44 ± 3.96 (0.60–11.59) (7)	0.12 ± 0.04 (1)
	^{228}Ra	0.20 ± 0.08 (1)	8.99 ± 9.35 (1.20–27.16) (7)	<0.05
	^{228}Th	0.10 ± 0.05 (1)	1.22 ± 0.96 (0.11–2.85) (7)	<0.03
	^{137}Cs	0.48 ± 0.40 (0.09–1.04) (6)	0.19 ± 0.17 (0.07–0.44) (4)	1.27 ± 0.18 (1)
	^{134}Cs	0.11 ± 0.02 (0.09–0.13) (3)	0.08 ± 0.02 (0.06–0.09) (2)	0.08 ± 0.02 (2)
	^{131}I	0.37 ± 0.11 (1)	<0.02	0.78 ± 0.24 (1)

Tissue samples are reported in fresh weight

Samples below the Minimum Detectable Activity denoted by "≤ MDA"

The range of all samples measured and the number of the considered samples for each area are indicated in brackets

For $n = 1$ the results are given as activity concentration ± uncertainty due to counting statistics

The significant difference of ^{228}Th and ^{228}Ra activities in bone tissue (^{228}Ra was five times higher than its progeny ^{228}Th) with a mean activity ratio of ^{228}Th/^{228}Ra at 0.27 and a mean ratio of ^{228}Th/^{232}Th at 0.40 indicated low degree of secular equilibrium in bones (Fig. 2c). The low degree of equilibrium in bones is attributed to the elemental and isotopic properties that affect radionuclides' transfer from the abiotic components to organism and to the accumulation by different type of tissues. The activity concentration of ^{228}Ra and ^{228}Th in grass and mammals were considered separately due to the low degree of equilibrium. Similarly, dose rates were calculated separately for these radionuclides.

Cesium is mainly accumulated in soft tissues in mammals and particularly in muscle due to their relatively large mass. The mean activity concentration of ^{137}Cs in muscle was calculated equal to 0.4 ± 0.3 Bq kg^{-1} ($n=13$) and of ^{134}Cs equal to 0.1 ± 0.1 Bq kg^{-1} ($n=8$) (Table 3). Since cesium behaves as potassium in mammals it can be uniformly distributed in body; hence, it was also detected in bones and organs. The mean activity concentration of ^{137}Cs in bones was equaled to 0.2 ± 0.2 Bq kg^{-1} ($n=4$) and in organs equaled to 0.7 ± 0.6 Bq kg^{-1} ($n=3$), while ^{134}Cs mean value respectively equaled to 0.1 ± 0.1 Bq kg^{-1} ($n=4$) and to 0.1 ± 0.1 Bq kg^{-1} ($n=5$). It should be noted, that the mean ratio of ^{137}Cs/^{134}Cs in muscle tissue was calculated equal to 3 and equal to 7 in organs indicating the existence of an additional source of ^{137}Cs besides Fukushima accident. It can be assumed that it is the residual ^{137}Cs (even though ^{137}Cs was not detected in the two samples of mammals that had been collected before the accident) since quite a small number of samples were measured.

The Fukushima-derived ^{131}I was detectable in mammals for a finite period after Fukushima accident. 131I is accumulated in soft tissues of mammals and mainly in thyroid. Thus, it was detected in organs (mean value equal to 1.2 ± 0.5 Bq kg^{-1}) and in muscle tissue (0.3 ± 0.1 Bq kg^{-1}) of 2 samples collected from SA1 (Attiki) and SA5 (Lesvos island).

The mean whole-body activity concentrations were calculated equal to 2.5 ± 2.3 Bq kg^{-1} ($n=14$) for ^{226}Ra, to 4.5 ± 7.3 Bq kg^{-1} ($n=16$) for ^{228}Ra and to 0.8 ± 0.9 Bq kg^{-1} ($n=13$) for ^{228}Th. Significantly lower were the activities calculated for ^{137}Cs at 0.4 ± 0.3 Bq kg^{-1} ($n=13$), for ^{134}Cs at 0.2

± 0.1 Bq kg^{-1} ($n=8$) and for ^{131}I at 0.7 ± 0.2 Bq kg^{-1} ($n=2$). The whole-body activity concentrations were imported in the ERICA Tool for the subsequent calculation of internal dose rates.

Radiological dose rates to biota

The mean total dose rate from all radionuclides considered ($\sum D_{tot}$) for each sampling area is illustrated in Fig. 3a for grass and Fig. 3b for mammals. The lowest $\sum D_{tot}$ was calculated for grass collected from SA2 (Etoloakarnania) ($1.8 \cdot 10^{-2}$ μGy h^{-1}) and for mammals from SA1 (Attiki) ($7.3 \cdot 10^{-2}$ μGy h^{-1}). The highest $\sum D_{tot}$ was calculated for samples collected from SA5 (Lesvos island) and equaled to $57 \cdot 10^{-2}$ μGy h^{-1} for grass and to $76 \cdot 10^{-2}$ μGy h^{-1} for mammals.

In Fig. 3a, b the calculated percentage contribution of each radionuclide to the $\sum D_{tot}$ is shown. The dose rates obtained through the application of ERICA Tool indicated that the higher contribution to $\sum D_{tot}$ derived from the exposure to ^{226}Ra and ^{228}Th with a joint contribution of 85 % for grass and 80 % for mammals. The contribution of ^{228}Ra corresponds to 9 % for grass and 3 % for mammals. The artificial radionuclides (^{137}Cs, ^{134}Cs, and ^{131}I) exhibited a relatively low contribution of 5 % for grass and 1 % for mammals with the highest contribution deriving from ^{137}Cs and equaled to 4 % and 0.8 %, respectively.

For the integrated evaluation of each radionuclide's contribution to the total dose rate the examination of D_{int} and D_{ext} influence was performed. As it can be seen in Fig. 4a for grass and Fig. 4b for mammals the highest contribution derives from the exposure to ^{226}Ra and ^{228}Th with D_{int} higher than D_{ext} for about one order of magnitude. This is due to the fact that the organism is internally exposed by ^{226}Ra and ^{228}Th, which are alpha emitters and cannot reach the organism through external exposure. This is not the case for ^{228}Ra and ^{137}Cs that exhibited higher contribution to the total dose rate through external exposure. Concerning ^{134}Cs and ^{131}I, these two Fukushima-derived radionuclides were not detected in soil and hence, only D_{int} was calculated associated to the incorporated radionuclides. The mean internal dose rate for
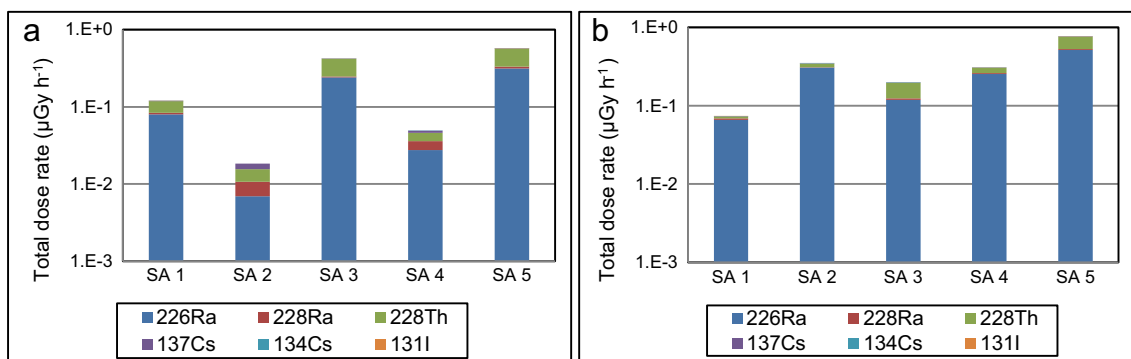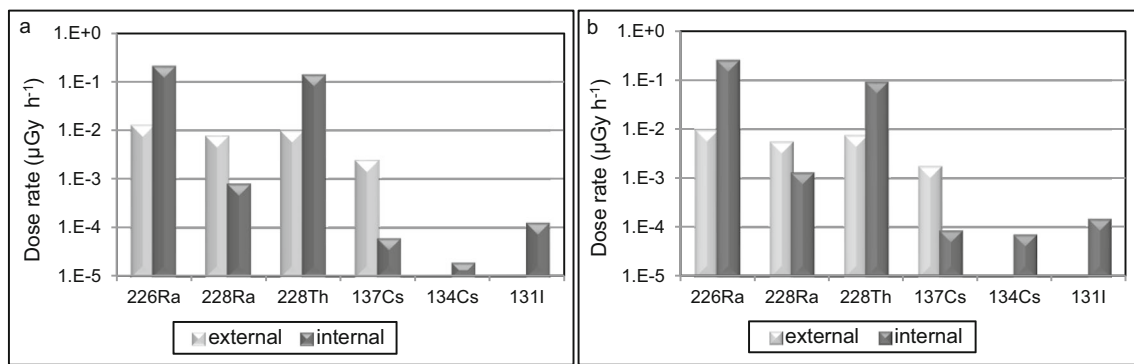

Fig. 3 Contribution from each radionuclide to the total dose rate ($\sum D_{tot}$) in samples of **a** grass and **b** mammals, for each case study site

Fig. 4 Internal and external dose rate in **a** grass and **b** mammals, from each radionuclide considered (mean values derived from all case study sites)

grass equaled to $20 \cdot 10^{-2}$ µGy h^{-1} and the mean external equaled to $3 \cdot 10^{-2}$ µGy h^{-1}. For mammals, the mean internal equaled to $31 \cdot 10^{-2}$ µGy h^{-1} and the mean external to $2 \cdot 10^{-2}$ µGy h^{-1}.

The calculated $\sum D_{tot}$ for grass and mammals is indicative of the exposure of non-human biota to natural and artificial radionuclides at the case study sites, since specific radionuclides were considered. The mean $\sum D_{tot}$ for grass samples was calculated equal to $23 \cdot 10^{-2}$ µGy h^{-1} and for mammals samples equal to $34 \cdot 10^{-2}$ µGy h^{-1}. In order to estimate the dose rate resulted from Fukushima impact, ^{134}Cs, ^{131}I, and ^{137}Cs were taken into account, whereas the contribution of the residual ^{137}Cs was excluded as it was described previously. In this case, the mean total dose rate was calculated equal to $1.7 \cdot 10^{-4}$ µGy h^{-1} for grass and to $2.7 \cdot 10^{-4}$ µGy h^{-1} for mammals.

The calculated dose rates were clearly below the screening criterion of ERICA Tool at 10 µGy h^{-1}, referred to the ecosystem level (Garnier-Laplace et al. 2008). The report of the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) about the effects of ionizing radiation on non-human biota (UNSCEAR 2008) proposed a chronic dose rate of 100 µGy h^{-1} for animals and 400 µGy h^{-1} for plants in order to indicate a threshold below of which it would be unlikely to have significant effects on population level. Furthermore, the ICRP (ICRP 2008) proposed numerical guidance with respect to RAP in the form of dose consideration reference level (DCRL), in order to prevent unnecessary or lack of effort concerning the environmental protection. The dose rates that were calculated within the framework of this study were below the lower band of 0.1 mGy day^{-1}. It should be noted however that the aforementioned comparison of the calculated dose rates values to the proposed benchmarks is indicative, seeking to provide an initial comparison of the current baseline levels of exposure at the studied ecosystems to the international levels of concern.

Conclusions

The aim of the present study was to analyze the recent radioactivity levels at the terrestrial environment in Greece and to estimate the radiological exposure of selected non-human biota. The application and adaptation of ERICA Tool in the specific ecosystems was performed using actual radioactivity measurements of organisms and abiotic components collected from the case study sites.

The results of gamma spectrometry showed that organisms are exposed to natural background radionuclides (^{226}Ra, ^{228}Ra, and ^{228}Th) and to artificial radionuclides of Fukushima origin (^{137}Cs, ^{134}Cs, ^{131}I) and from residual traces of the Chernobyl accident and global fallout (^{137}Cs). The dose rates calculated by ERICA Tool indicated that natural radionuclides exhibited significantly higher contribution to the total dose rate than the artificial radionuclides. Internal dose rate, which was significantly higher than external, is mainly attributed to alpha emitters (^{226}Ra and ^{228}Th) and to the incorporation of their daughter nuclides at the calculation of dose rates. On the other hand, the radiological exposure to Fukushima-derived radionuclides was quite low and owed to internal exposure from the incorporated radionuclides. The studied non-human biota is exposed to low-level ionizing radiation and no significant impact can be estimated. However, further consideration of the exposure levels could be considered taking into account the eventual effects of protracted low level ionizing radiation on the various levels of life organization.

The present work is the first effort in recent years to evaluate the level of exposure of non-human biota to naturally and artificially derived radionuclides. Taking into consideration the lack of relevant studies at the region, the availability of this kind of measurements may be valuable in future research in order to draw more reliable findings about the radiological exposure of terrestrial non-human biota. The measured site-specific data may be proved useful in studies relevant to the assessment of radiological exposure in areas dominated by similar ecosystems. In summary, the results obtained through the application of dose assessment models could contribute to the development of integrated decision making policies respecting the environmental and radiological impact assessment.

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