ESTIMATION OF DOSE RATES TO HUMANS EXPOSED TO ELEVATED NATURAL RADIOACTIVITY THROUGH DIFFERENT PATHWAYS IN THE ISLAND OF IKARIA, GREECE

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Received January 19 2010, revised July 19 2010, accepted September 1 2010

A radiological survey has been carried out in the island of Ikaria based on the natural radionuclide inventory in abiotic environment and the consequent dose rate assessment for the critical groups of population. The island of Ikaria—Aegean Sea, Greece is characterised by the presence of mineral and thermo-mineral springs, which have an apparent influence on natural background radiation of the island. The levels of natural radionuclides in spring water (either for spa treatment and household use), potable water (local domestic network), and rock and soil samples were measured in this island. The concentrations of ²²²Rn and natural gamma emitters were found to be significantly elevated in spring water and some rock and soil samples. In terms of NORM and TENORM, the external and internal dose rates (mSv y⁻¹) were estimated in three groups of population selected on the basis of water use as: habitants of the island, working personnel and bathers in spa installations. According to the derived results, the working personnel in the thermal spa installations are exposed to significant radiological risk due to waterborne ²²²Rn with a maximum dose rate up to 35 mSv y⁻¹, which led to overexposure in terms of the 20 mSv y⁻¹ professional limits. Therefore, this group can be considered as the critical one for the radiological impact assessment in the island.

INTRODUCTION

The first references regarding the therapeutic properties of spring waters and their use in spa installations have been recorded since antiquity (Hippocrates 46–377 BC, Epicures 341–270 BC)^(1, 2).

On the basis of the staff studies (3-7), among the thermo-mineral springs spread throughout Greece, four of them (named Ikaria, Loutraki, Kamena Vourla and Edipsos) have been studied and are recognised as 'radioactive' due to their elevated concentrations of natural radionuclides. The reported radon concentrations in the wide areas of these springs are >100 Bq 1^{-1} . In all cases the springs bubble up in the littoral zone, in the sub-littoral zone (under the strata through the surface of the bottom to the seawater layer above) and in temperate locations in the inland parts of these areas. Hence, they have been established, through centuries, as places of 'therapeutical tourism', where medicine practices are combined with the usual vacations near sea. Nowadays, these springs are still used for balneotherapy, whereas, in some cases spring waters are used for limited water supply. This is the case of Ikaria, where spa and potable use of spring waters is carried out.

The island of Ikaria, with an area of 267 km² and a population of 7500, is located in the Eastern Aegean Sea, in Greece. A mountainous area dominates the island, which is located in the eastern edge of the Cycladic crystalline belt, called Atticocycladic

mass of Hellenides, which is considered as a transitional zone to the Menderes mass of the Anatolia (Asia Minor Peninsula). In the littoral zone around the island there are several mineral and/or thermomineral springs, whereas in the sub littoral zone some springs bubble up from the bottom as well. These are located in Therma (Springs of Apollo and Spilaio), Agios Kirikos (Aesculapius), Lefkada and Xylosirtis (Figure 1). They are divided into thermal bath spas (45–65°C water temperatures) and potable spring water (20°C water temperature). The springs in Therma, Agios Kirikos and Lefkada are used for balneotherapy. Xilosortis spring water, called locally 'immortal water', is used as a potable source.

Most of the Ikaria springs are contact springs between different rock types at the perimetric zone of the western Ikaria granite, usually bearing various dissolved ions, with NaCl as the most common among them followed by sulfosalts. The radioactive springs are related to the granitic body of the western tectonic unit, in which a high radioactive background is observed, while in the eastern unit, the rocks (granites, schists, marbles and limestone) do not present positive radiometric anomalies.

In the present study, the natural radiation status has been evaluated in the island of Ikaria. The radiological impact in areas of radioactive springs has been investigated for the habitants of the island, the thermal spa workers and the users of spa water.

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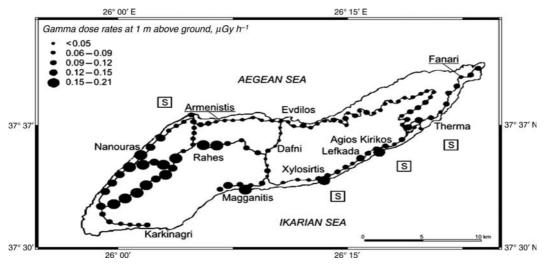


Figure 1. Gamma dose rates and the sampling points in the island of Ikaria.

MATERIALS AND METHODS

In order to evaluate the inventory of the environmental radiation levels in the island of Ikaria, samples of soil, rock, fault materials, spring water and domestic water were appropriately collected and analysed in the laboratory by gamma spectrometry. The samples were measured for 70 000 s in a highresolution gamma-spectrometry system, incorporating an HpGe detector of 20 % relative efficiency and a computerised multichannel analyzer of 4000 ch (in a spectrum extending to 2000 keV). ORTEC software was used for the analyses of the spectra obtained. The relative statistical error (1σ) does not exceed 10 %. The efficiency was determined by two ²²⁶Ra standard sources of simulating the counting geometry of the samples. An energy calibration correction was made using a 60Co point source of 3.7×10^4 Bq, in the 1173.2 and 1332.5 keV peaks.

Sampling was performed initially in the field of the springs (water, soil and rock) and drilled wells. In the wide area, samples were appropriately collected according to the systematic sampling procedure carried out, based on the geological characterisation of the area. The site-specific sampling was performed randomly, as to cover the geologically defined area.

Water samples were transferred in 11 Marinelli beakers (special shaped in aluminum for radon measurement by 'Vintantonio de Palma 20125, Milano, Viale Sacra 51, Italy') and in 51 plastic bottles with the pH adjusted to 1 by adding acid nitric of 60 % normality. The ²²²Rn determination was carried out with the beakers sealed and stored for 3 h prior to measurement to ensure that equilibrium between ²²²Rn and its daughters has been

achieved. The activity of ²²²Rn was derived from the analysis of the 295.2 and 352 keV gamma lines of ²¹⁴Pb and 609.4 keV gamma line of ²¹⁴Bi, taking into account the correction factor for the decay that occurred in the meantime between sampling and measurement. The estimated uncertainty of the measurements was 25 % due to gas losses during sampling. Concerning the ²²⁶Ra determination, the 5l samples were evaporated to 1l samples and sealed in the measurement pots after the removal of ²²²Rn by aeration of the samples. The samples were kept sealed for at least 20 d to ensure the secular equilibrium between ²²⁶Ra and its daughters. Activity concentrations of ²²⁸Ra, ²²²Rn, ²²⁸Ra, ²²⁸Th and ⁴⁰K in spring and domestic water were determined for the studied area.

The soil, rock and fault material samples were also measured after the equilibrium between ^{226}Ra and ^{222}Rn had been achieved, by keeping them in sealed pots of 68 mm diameter and 15 mm height for a period of 20 d. Activity concentrations of the natural radionuclides $^{238}U,\ ^{226}Ra,\ ^{228}Ra,\ ^{228}Th$ and ^{40}K were determined.

Energies used for the measurements of the above-mentioned radionuclides are as follows: 63.3 and 92.6 keV for ²³⁸U, 295.2, 352.0 and 609.4 keV for ²²⁶Ra, 338.4 and 911.1 keV for ²²⁸Ra, 238.6 and 583.1 keV for ²²⁸Th, 1460.7 keV for ⁴⁰K.

RESULTS AND DISCUSSION

The results obtained for the studied area are presented on the basis of: (1) activity concentrations of naturally occurring radionuclides in the abiotic materials of the island (Tables 1–3) and

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Table 1. Range of activity concentrations of natural radionuclides in soil, rocks and fault materials sampled in the island of Ikaria (Bq kg^{-1}).

Nuclides sample	²³⁸ U	²²⁶ Ra	²²⁸ Ra	²²⁸ Th	²³² Th	⁴⁰ K
Soil (number of samples, 50)						
Soil (spring areas)	13 - 138	19 - 122	20 - 305	10 - 296	18 - 301	247 - 1149
Soil (wide area)	5 - 230	10 - 422	2 - 220	4 - 156	3 - 187	194-1531
Wide Greek environment and worldwide ^(8-12, 29-35)	0 - 690	0 - 900	5 - 109	3 - 150	1 - 360	0 - 3200
Rocks (number of samples, 50)						
Rocks (spring areas)	14 - 121	8 - 129	2 - 81	6 - 61	4 - 71	73 - 1396
Rocks (wide area)	3 - 258	3 - 162	0.5 - 136	2 - 102	0.8 - 112	12 - 1514
Fault materials (number of samples, 30)						
Fault materials (spring areas)	46 - 1049	52 - 764	69 - 101	23 - 69	46 - 85	1010-2664
Fault materials (wide area)	96 - 203	162 - 422	5 - 80	3 - 46	4-63	307 - 723

Table 2. Range of activity concentrations of natural radionuclides in spring water sampled in the island of Ikaria (Bq 1⁻¹).

Nuclides sample	²²⁶ Ra	²²⁸ Ra	²²² Rn	⁴⁰ K
Ikaria (number of samples, 20) Wide Greek environment and worldwide ^(3-7, 20-22, 25,26)	$0.3-5.0 \\ 0.01-6$	<0.1-5.5 0.03-0.5	25-2468 0.8-1780	4-24

Table 3. Range of activity concentrations of natural radionuclides in tap and drilled water sampled in the island of Ikaria (Bq l^{-1}).

Nuclides sample	²²⁶ Ra	²²⁸ Ra	²²² Rn
Tap water			
Îkaria (number of samples, 5)	< 0.1-0.1	< 0.1	0.1 - 2
Wide Greek environment and worldwide	0.004 - 0.089		1.2 - 600
Drilled water			
Ikaria (number of samples, 4)	< 0.1-0.2	< 0.1-1.3	9-24
Wide Greek environment and worldwide	< 0.003-1.8	0.175 - 0.440	0.815 - 346

(2) the derived dose rates for the several population groups exposed under various conditions (Table 4).

Concerning the soil, rock and fault materials samples (Table 1), elevated levels of natural radio-nuclides were detected in some of the measured samples in comparison with the respective background levels from a wide sampling network in Greece⁽⁸⁻¹²⁾. It is noteworthy that the highest concentrations were detected in the fault materials. These values range in the value spectrum reported in the international literature and are lower than the upper limits⁽⁵⁾.

Concerning the spring water samples (Tables 2 and 3), ²²⁶Ra and ²²²Rn show higher concentrations compared with other Greek areas⁽⁵⁾, whereas the measured domestic water samples show lower and/ or comparable concentrations of ²²²Rn and ²²⁶Ra, in comparison with the respective values of the international literature⁽⁵⁾. From the radiological point of view, the measurements for ²²²Rn were below the

limit of 100 Bq l⁻¹, recommended by the Commission of the European Communities⁽¹³⁾.

Dosimetry calculations

The assessment of the radiological impact on humans, in relation to the springs has been evaluated for three cases: (A) for the habitants of the wide area, (B) the working personnel in spa installations and (C) bathers. Considering the pathway exposure of humans and the above classification, the external and internal dose calculations were performed as follows (Table 4):

- (1) The exposure of the population to gammaradiation of terrestrial origin.
- Radiological impact on the population—user of the water.
- (3) Radiological impact on thermal spa working personnel.
- (4) Radiological impact on spa users—patients.

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Table 4. Summarised results of dose rates to habitants, thermal spa working personnel and users of spa in the island of Ikaria (mSv y^{-1}).

	$mSv y^{-1}$
Habitants	
Dose rate equivalent due to terrestrial gamma radiation	0.20 - 3.31
Effective dose equivalent due to ²²² Rn intake from potable water	0.0001 - 0.12
Effective dose equivalent due to ²²² Ra intake from potable water	0.025 - 0.18
Effective dose equivalent due to ²²² Rn intake from potable water Effective dose equivalent due to ²²² Ra intake from potable water Effective dose equivalent due to the inhalation of ²²² Rn released from potable water	0.0004 - 0.09
Thermal spa working personnel Effective dose equivalent due to the inhalation of ²²² Rn released from spa water	5-35
Spa users	
Dose rate due to gamma-radiation during the immersion into bath water	$0.0012 \times 10^{-3} - 0.012 \times 10^{-3}$
Dose rate due to gamma-radiation during the immersion into bath water Dose rate due to ²²² Rn radiation during the immersion into bath water	0.25 - 1.7
Effective dose equivalent due to the inhalation of ²²² Rn released from spa water	0.04 - 0.29

Case A: Radiological impact on the population-habitant of the island

Exposure of the population to gamma radiation of terrestrial origin. A major parameter in radiological impact due to natural radiation is the exposure to terrestrial gamma radiation. The dose equivalent rate (mSv y⁻¹) has been calculated using Eq. 1^(5, 8, 14) and the results of gamma spectrometry measurements in rock, soil and fault materials sampled from Ikaria areas.

$$H = 0.061 A_s(^{238}\text{U}) + 3.103 A_s(^{226}\text{Ra}) + 1.741 A_s(^{228}\text{Ra}) + 2.582 A_s(^{228}\text{Th}) + 0.276 A_s(^{40}\text{K}),$$
(1)

where H is the dose equivalent rate (μ Sv y⁻¹) and A_s is the activity concentration of the respective radionuclide (Bq kg⁻¹).

In the island of Ikaria, the annual dose equivalent ranged between 0.20 and 3.31 mSv y^{-1} (Table 4). The mean values in the areas of interest are 1.52 mSv y^{-1} in Lefkada, 0.29 $\mu Sv\ y^{-1}$ in Therma and Aghios Kirikos and 0.65 mSv y^{-1} in Xylosirtis. In comparison with the wide Greek area (0.1–1 mSv y^{-1} , mean value of 0.49 mSv $y^{1})^{(12)}$ the dose rates in Ikaria presented higher maxima. Nevertheless, in the international literature maxima >5 mSv y^{-1} have been reported $^{(15)}$

In the vicinity of the springs, the following were observed: in Lefkada, the range of the calculated dose equivalent rates was 71–360 nSv h⁻¹, resulting in average and maximum annual values of 1.52 and 3.31 mSv y⁻¹, respectively. The respective range in Therma was 20–33 nSv h⁻¹ and the respective annual average and maximum values were 0.28 and 0.29 mSv y⁻¹. In the vicinity of the spring 'immortal water', the dose rate values were in the range of 53–149 nSv h⁻¹, with an average and maximum dose equivalent of 0.65 and

1.30 mSv y^{-1} , respectively. Compared with the reported mean and maximum dose equivalent rates for other Greek insular areas⁽¹⁶⁾ of 0.43 and 0.74 mSv y^{-1} , the equivalent dose rates in Lefkada and immortal water were relatively higher. On the contrary, in areas around the radioactive springs in Therma the external dose rates were relatively low.

Radiological impact on the habitants due to water consumption. The significance of water in relation to the radiological impact on the population user becomes important in case of areas of high radioactive background, where the water contains high concentrations of natural radionuclides. The detected radionuclides in inland spring water that present interest for the radiological impact were the ²²⁶Ra and ²²²Rn.

Concerning the ²²²Rn contained in water draining into indoor places, it is released into the air of these places to a certain degree. Hence, it contributes to the radiological impact on human organism through the respiratory way also. A significant increase to the total indoor concentration of ²²²Rn, is observed in the cases of high radioactive background areas where the water contains high concentrations of ²²²Rn. This is of importance in bath-therapy installations, where the springs with high concentrations of ²²²Rn are widely used.

The radiological impact on the habitants, due to consumption of drinking water, was calculated taking into account the intake, through ingestion, of ²²⁶Ra and ²²²Rn. In this account, the concentrations of ²²⁶Ra and ²²²Rn in spring, tap and drilled water were considered:

²²⁶Ra: the activity concentrations of ²²⁶Ra were in the range of <0.1-0.7 Bq l⁻¹ (Tables 2 and 3). The effective dose equivalents due to ²²⁶Ra ingestion were calculated (Table 4) on the assumption that the water consumption is 0.5 l per day, per person and considering a conversion factor of 250 $\mu Sv\ y^{-1}$ Bq l^-1(17, 18). The calculated values were in the range of 0.025–0.18 mSv y^-1 with an average value of 0.1 mSv y^-1. The highest dose corresponds to the ingestion of potable spring water. The respective range in the international literature is 0.0018–1.3 mSv y^-1(18–21)

²²²Rn: first, the activity concentrations of ²²²Rn were in the range of <0.1-114 Bq 1^{-1} (Tables 2 and 3). The effective dose equivalents due to ²²²Rn ingestion were calculated (Table 4) based on the assumption that the water consumption is 0.5 l per day, per person and considering a conversion factor of 1 µSv y^{-1} Bq $1^{-1(17-19)}$. The calculated values were in the range of 0.0001-0.12 mSv y⁻¹ with an average value of 0.07 mSv y⁻¹. The highest doses were corresponding to the ingestion of potable spa water. It should be noted that the respective range in the international literature is 0.0005–0.12 mSv y⁻¹⁽²⁰⁻ ²²⁾ and secondly in order to estimate the radiological impact from 222Rn inhalation, due to its release from water, the mean contribution of the waterborne ²²²Rn to the indoor air concentrations was evaluated. The waterborne ²²²Rn concentrations in the indoor air of Ikaria residences was calculated on the basis of the transfer coefficient of 10⁻⁴ from water to air^(11, 19). The waterborne ²²²Rn concentrations in the indoor air of Ikaria residences were in the range of 0.01-2.4 Bq m⁻¹, with an average value of 0.8 Bq m⁻¹. For comparison, it is mentioned that the indoor radon concentrations in the island of Ikaria are as high as 583 Bq $m^{-3(23)}$. It is obvious that the contribution of water as a source of ²²²Rn in the indoor air is insignificant.

The resulting effective dose equivalent from the ingestion of the waterborne 222 Rn, in equilibrium with its daughters, was in the range of 0.0004-0.09 mSv y^{-1} (Table 4).

According to UNSCEAR 1988⁽¹⁹⁾, the total effective dose equivalent from the radiological impact on natural origin through all the pathways of internal irradiation, in areas of normal background is estimated for ²²²Rn to be 0.85 mSv y⁻¹ and for ²²⁶Ra to be 0.007 mSv y⁻¹. According to these values, the calculated mean dose of 0.1 mSv y⁻¹ for ²²⁶Ra, resulting from this study, is estimated to be very high and taking into account the fact that it concerns only one way of internal irradiation (ingestion of water) while as the values of UNSCEAR report are referred to a total internal exposure.

Case B: Radiological impact on the working personnel exposed to waterborne ²²²Rn

The working personnel in spa installations are exposed to internal irradiation from waterborne

radon. It is noteworthy that according to the ICRP 65 report⁽¹⁷⁾, the workers in radon thermal spas belong to the professional groups, which may be, exposed to high radiation does, due to high concentrations of ²²²Rn in the indoor air of the installations.

The spa installations at Apollo, Spilaio and Asclepiad are widely used for the cure of many different diseases. In order to estimate the radiological impact on the workers due to waterborne ²²²Rn inhalation, the mean contribution of the water ²²²Rn in the indoor air concentrations was evaluated, based on a transfer coefficient from water to air of $2.7 \times 10^{-3(19, 24)}$. Hence, the ²²²Rn concentrations of the spa indoor installations were found to be 2611 Bq m⁻³ for Apollo, 362 Bq m⁻³ for Spilaio and 2746 Bq m⁻³ for Asclepiad. The annual effective dose equivalent due to inhalation was calculated using the conversion 1.5×10^{-1} mSv per Bq h m⁻¹. The calculations were performed on the assumption that the total working time was 900 h y^{-1} . The resulting effective dose equivalent rate to the working personnel, due to inhalation of 222Rn released from the water, was in the range of 5-35 mSv y⁻¹. The respective dose values found in the international and Greek literature for working personnel in other spa installations^(6, 7, 18, 25, 26) are in the range of 0.4-44 mSv y^{-1} . It should be noted that in most of the referred areas, the spas function 9 months per year and the dose calculations have been performed on a higher total working time than the total working time in Ikaria spas.

It should be mentioned that the Chernobyl accident has caused to the mean Greek habitant—in an interval of 8 months (5/86–12/86)—additional dose of 0.51 mSv. The respective dose for the mean Greek habitant of a critical group reaches the 1.94 mSv (DEMO 86/3G and DEMO 86/10G)^(27, 28). As a critical group, the population of North Greece (Macedonia) is considered, as this region accepted the highest impact during the period of the radioactive cloud arrival.

Case C: Radiological impact on bathers

The dose rates for bathers depend on the type of therapy. Bathers using the spa waters, besides the external exposure from bath-therapy in the water, are subject to internal irradiation due to inhalation of ²²²Rn. In the ICRP report^(17, 18), an indoor air radon limit of 200–600 Bq m⁻³ for non-occupational exposure is recommended. Considering this limit, the indoor ²²²Rn concentrations in Apollo and Asclepiad spa installations were relatively high. More specific is the following:

External dose rates due to immersion in the water. The dose rates (Gy s⁻¹) due to natural

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gamma emitters in spa water were calculated on the basis of the following equations⁽⁵⁾, taking into account the natural radionuclides' concentrations in spa installation water:

$$D(^{238}\text{U}) = 0.533 \times 10^{-14} A_s(^{238}\text{U}),$$

$$D(^{226}\text{Ra}) = 28.10 \times 10^{-14} A_s(^{226}\text{Ra}),$$

$$D(^{228}\text{Ra}) = 15.76 \times 10^{-14} A_s(^{228}\text{Ra}),$$

$$D(^{228}\text{Th}) = 23.41 \times 10^{-14} A_s(^{228}\text{Th}),$$

$$D(^{40}\text{K}) = 2.50 \times 10^{-14} A_s(^{40}\text{K}).$$

The dose rates resulting from the ²²²Rn contained in water were calculated on the assumption of a 0.5 equilibrium factor between ²²²Rn and its daughters. The conversion factors were 0.286 and 1.95 Sv y⁻¹ per Bq cm⁻¹ for ²¹⁴Pb and ²¹⁴Bi, respectively. The results are given in Table 4.

Internal dose rates due to waterborne ^{222}Rn inhalation. The annual effective dose equivalent due to ^{222}Rn inhalation was calculated using the factor 1.4×10^{-5} mSv per Bq h m $^{-3}$, on the assumption of a mean total stay in the indoor place of the installations 45 min for a mean number of 10 therapies. The results are shown in Table 4.

CONCLUSIONS

Elevated levels of natural radionuclides were detected in some of the measured samples in comparison to the respective background levels in Greece. The existence of high concentrations of natural radionuclides in the abiotic environment of the island of Ikaria resulted in some cases to overdoses in relation to the typical background (2.5 mSv y^{-1}). The range of external and internal dose rates for the habitants, working personnel and spring water users was very broad (0.0012×10⁻³–35 mSv y^{-1}).

In comparison to the wide Greek territory and the reported literature, the dose equivalent rates due to terrestrial gamma radiation determined in the island of Ikaria were found to range in the upper limit of the value spectrum. Considering the consumption of potable water, the spring called 'immortal water' results to overdoses for the population, i.e. due to the ²²⁶Ra ingestion (0.025–0.18 mSv y⁻¹), with the maxima exceeding the recommended limit of 0.1 mSv y⁻¹. The impact on working personnel in spa installations was due to waterborne ²²²Rn with

a maximum dose rate up to 35 mSv y^{-1} , which led to overexposure in terms of the 20 mSv y^{-1} professional limits. In terms of the bathers, they are exposed mainly to 222 Rn exposure during the immersion into bath water, with a dose rate of 0.25–1.7 mSv y^{-1} .

ACKNOWLEDGEMENTS

The authors thank Assistant Professor G. Nicolaou (Faculty of Engineering, Department of Electrical and Computer Engineering, Laboratory of Nuclear Technology, Demokritus University of Greece) for his critical reading of the manuscript.

FUNDING

Self-funded program of third party services (provision of services), entitled "Measurements of radionuclides in goods and other products of public use – RP-802(GEL)". ERL/INT-RP/NCSRD, Athens, Greece.

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