

## Natural radioactivity as an impact factor in drinking water quality

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Received 31 March 2009; accepted 9 July 2009

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### ABSTRACT

This study has evaluated the levels of natural radionuclides in spring water and drilled water, which are used as potable water reservoirs and samples from the local water supply network, in the island of Ikaria in the Aegean Sea and potable spring water in Loutraki in the Korinthiakos Gulf, Greece. The activity concentrations of  $^{226}\text{Ra}$  and  $^{222}\text{Rn}$  were in the range of  $<0.1\text{--}0.7\text{ Bq l}^{-1}$  and  $<0.1\text{--}114\text{ Bq l}^{-1}$ , respectively. The radiological impact on the habitants, due to consumption of drinking water, was calculated taking into account the annual intake, through ingestion, of  $^{226}\text{Ra}$  and  $^{222}\text{Rn}$ . In order to estimate the radiological impact from  $^{222}\text{Rn}$  inhalation, due to its release from water, the mean contribution of the waterborne  $^{222}\text{Rn}$  to the indoor air concentrations was evaluated on the basis of the transfer coefficient of  $10^{-4}$  from water to air. The effective dose equivalents due to  $^{226}\text{Ra}$  ingestion were in the range of  $25\text{--}175\text{ }\mu\text{Sv y}^{-1}$ , with the maxima reaching the recommended limit of  $100\text{ }\mu\text{Sv y}^{-1}$ . The effective dose equivalents due to  $^{222}\text{Rn}$  ingestion were in the range of  $0.1\text{--}114\text{ }\mu\text{Sv y}^{-1}$ . For both radionuclides, the highest doses were corresponding to the ingestion of potable spring water, compared to the drilled and tap water. The resulting effective dose equivalent from the inhalation of the waterborne  $^{222}\text{Rn}$ , in equilibrium with its daughters, was in the range of  $0.36\text{--}85\text{ }\mu\text{Sv y}^{-1}$ . According to our results, the ingestion of  $^{226}\text{Ra}$  presents the higher impact factor of natural radioactivity in drinking water of the studied areas and appropriated remedies should be taken.

**Keywords:** Natural radioactivity; Drinking water;  $^{226}\text{Ra}$ ;  $^{222}\text{Rn}$ ; Dose rate

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### 1. Introduction

The radiological quality of water for domestic use is an issue of evaluation in case of areas of high radioactive background, where elevated concentrations of the natural radionuclides  $^{226}\text{Ra}$  and  $^{222}\text{Rn}$  have been measured in abiotic materials including water natural reservoirs.  $^{226}\text{Ra}$  is of significant radiological importance as its activity concentrations in drinking water are transformed to dose rates by a conversion factor of  $250\text{ }\mu\text{Sv y}^{-1}$  per  $\text{Bq l}^{-1}$ , which is relatively high

[1,2].  $^{222}\text{Rn}$ , as a mobile gas, escapes from the water, draining into indoor places and is related to the ambient air to a certain degree. Hence, it contributes to the radiological impact on human organism through the inhalation exposure, too.

The natural radiation status has been evaluated in the selected coastal areas of the island of Ikaria in the Aegean Sea and Loutraki in the Korinthiakos Gulf, Greece in previous studies [3,4]. The selected areas are characterized by elevated natural radioactivity levels in abiotic materials (rocks, soil, water) attributed to the special physicochemical characteristics of their local environment, which are related to the presence of

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geothermal springs and vents. The pathway exposure to humans i.e. internal dose rates due to the radionuclides transferred through ingestion and inhalation have been carried out. In terms of ingestion pathway, because local water reservoirs can be used for drinking and household water, especially in isolated areas as islands, the internal exposure due to potable water must be taken into account in areas of elevated natural radioactive background. The major apportionment to the dose received through inhalation is due to the  $^{222}\text{Rn}$  escaped from the water body.

In the present study the levels of  $^{226}\text{Ra}$  and  $^{222}\text{Rn}$  in potable spring water, tap and drilled wells water were measured in the island of Ikaria in the Eastern Aegean Sea and Loutraki in Korinthiakos gulf in Central Greece. The study summarizes the most important results in water samples of our 8 years investigation related to natural radioactivity measurements in the selected areas. Radionuclide levels were significantly elevated in potable spring water in the two studied areas. The results obtained were used for the evaluation of the radiological impact on human organisms.

## 2. Materials and methods

In order to evaluate the inventory of the  $^{226}\text{Ra}$  and  $^{222}\text{Rn}$  radiation levels in water in the studied areas, samples of potable spring water and domestic water (tap and drilled wells) were appropriately collected and analyzed in the Laboratory by gamma-spectrometry.

The samples were collected and transferred in 1l Marinelli beakers (special shaped in aluminum for radon measurement by "Vintantonio de Palma – 20125, Milano – Viale Sacra 51, Italy") and 5l in plastic bottles with the pH adjusted to 1 by adding nitric acid of 60% normality.

- The  $^{222}\text{Rn}$  determination was carried out with the Marinelli beakers sealed and stored for 3 h prior to measurement to ensure that equilibrium between  $^{222}\text{Rn}$  and its daughters has been achieved. The activity of  $^{222}\text{Rn}$  was derived from the analysis of the 295.2 keV and 352 keV gamma lines of  $^{214}\text{Pb}$  and 609.4 keV gamma line of  $^{214}\text{Bi}$ , 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  $^{222}\text{Rn}$  gas losses during sampling, meaning gas losses during collection of water until the Marinelli beakers totally sealed.
- Concerning the  $^{226}\text{Ra}$  determination, the 5l samples were evaporated to 1l sample and sealed in the

Table 1

Range of activity concentrations of natural radionuclides in potable spring water, tap and drilled water in the island of Ikaria ( $\text{Bq l}^{-1}$ )

Ikaria		
Nuclides Sample	$^{226}\text{Ra}$	$^{222}\text{Rn}$
Potable spring water (No samples = 10)	MDA*–0.7	MDA–114
Tap water (No samples = 5)	MDA–0.1	0.1–2
Drilled water (No samples = 7)	MDA–0.2	9–24
Wide Greek environment and worldwide		
Potable spring water	<0.003–2	1–815
Tap water	0.004–0.089	1.2–600
Drilled water	<0.003–1.8	0.815–346

\*MDA has been estimated to  $0.1 \text{ Bq l}^{-1}$ .

measurement pots (Marinelli beakers) after the removal of  $^{222}\text{Rn}$  by aeration of the samples. The samples were kept sealed for at least 20 days to ensure the secular equilibrium between  $^{226}\text{Ra}$  and its daughters.

Afterwards, the samples were measured in a high-resolution gamma-spectrometry system, incorporating an HpGe detector of 20% relative efficiency and a computerized multichannel analyzer of 4096 ch (in a total spectrum area of 2,000 keV). ORTEC software was used for the analyses of the spectra obtained. The relative statistical error ( $1\sigma$ ) did not exceed 10%. The efficiency was determined by two  $^{226}\text{Ra}$  standard sources of simulating the counting geometry of the samples. An energy calibration correction was made using of a  $^{60}\text{Co}$  point source of  $3.7 \times 10^4 \text{ Bq}$ , in the 1,173.2 keV and 1,332.5 keV peaks.

## 3. Results and discussion

### 3.1. Gamma spectrometry

The results of gamma spectrometry measurements in spring water and domestic water are given in Tables 1 and 2.

The activity concentrations of  $^{226}\text{Ra}$  were in the range of  $<0.1\text{--}0.7 \text{ Bq l}^{-1}$ . Concerning the potable spring water samples (Tables 1 and 2), elevated values of  $^{226}\text{Ra}$  and  $^{222}\text{Rn}$  were detected in comparison with the respective values in other Greek areas as well as in areas quoted to in the international literature [4]. Finally, lower and/or comparable concentrations of  $^{222}\text{Rn}$  and  $^{226}\text{Ra}$ , with respect to values quoted in the international literature [4], occur in domestic water

Table 2  
Range of activity concentrations of natural radionuclides in potable spring water, tap and drilled water in Loutraki ( $\text{Bq l}^{-1}$ )

Nuclides Sample	$^{226}\text{Ra}$	$^{222}\text{Rn}$
Loutraki Potable spring water (No samples = 5)	0.7	–

samples. The measurements for  $^{222}\text{Rn}$  in tap and drilled wells water were below the limit of  $100 \text{ Bq l}^{-1}$ , recommended by the Commission of the European Communities [5].

### 3.2. Dosimetry calculations

#### 3.2.1. Radiological impact on the habitants due to water consumption

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

#### 3.2.2. $^{226}\text{Ra}$

The activity concentrations of  $^{226}\text{Ra}$  were in the range of  $<0.1\text{--}0.7 \text{ Bq l}^{-1}$  (Table 1). The effective dose equivalents due to  $^{226}\text{Ra}$  ingestion were calculated (Table 3) on the assumption that the water consumption is 0.5 L per day, per person and considering a conversion factor of  $250 \mu\text{Sv y}^{-1}$  per  $\text{Bq l}^{-1}$  [1,2]. The calculated values were in the range of  $25\text{--}175 \mu\text{Sv y}^{-1}$ . The highest dose corresponds to the ingestion of potable spring water. The maximum dose exceeds the recommended limit of  $100 \mu\text{Sv y}^{-1}$ . From the measured water samples, only two main outflows of potable spring water present effective dose

Table 3  
Range of effective dose equivalent due to drinking water use ( $\mu\text{Sv y}^{-1}$ )

Effective dose equivalent due to $^{222}\text{Rn}$ intake from potable water	0.1–114
Effective dose equivalent due to $^{226}\text{Ra}$ intake from potable water	25–175
Effective dose equivalent due to the inhalation of $^{222}\text{Rn}$ released from potable water	0.36–85

equivalents higher than the limit of  $100 \mu\text{Sv y}^{-1}$ . The respective range in the international literature is  $1.8\text{--}1,300 \mu\text{Sv y}^{-1}$  [5–7].

#### 3.2.3. $^{222}\text{Rn}$

- The activity concentrations of  $^{222}\text{Rn}$  were in the range of  $<0.1\text{--}114 \text{ Bq l}^{-1}$  (Table 1). The effective dose equivalents due to  $^{222}\text{Rn}$  ingestion were calculated (Table 3) on the assumption that the water consumption is 0.5 L per day, per person and considering a conversion factor of  $1 \mu\text{Sv y}^{-1}$  per  $\text{Bq l}^{-1}$  [1,2]. The calculated values were in the range of  $0.1\text{--}114 \mu\text{Sv y}^{-1}$ . The highest doses were corresponding to the ingestion of potable spring water. In the case of drinking water the dose limit for  $^{226}\text{Ra}$  does not apply for  $^{222}\text{Rn}$  too. It should be noted that the respective range in the international literature is  $0.5\text{--}120 \mu\text{Sv y}^{-1}$  [6–9].
- In order to estimate the radiological impact from  $^{222}\text{Rn}$  inhalation, due to its releasing from water, the mean contribution of the waterborne  $^{222}\text{Rn}$  to the indoor air concentrations was evaluated on the basis of the transfer coefficient of  $10^{-4}$  from water to air [10]. The resulting effective dose equivalent from the inhalation of the waterborne  $^{222}\text{Rn}$ , in equilibrium with its daughters, was in the range of  $0.36\text{--}85 \mu\text{Sv y}^{-1}$  (Table 3).

According to UNSCEAR 1988 [10], the total effective dose equivalent from the radiological impact on natural origin through all the pathways of internal irradiation, in areas of normal radioactive background is estimated for  $^{222}\text{Rn}$  at  $850 \mu\text{Sv y}^{-1}$  and for  $^{226}\text{Ra}$  at  $7 \mu\text{Sv y}^{-1}$ . According to these values, the calculated maximum dose of  $175 \mu\text{Sv y}^{-1}$  for  $^{226}\text{Ra}$ , resulting from this study, is estimated as 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.

## 4. Conclusions

Elevated levels of the natural radionuclides  $^{226}\text{Ra}$  and  $^{222}\text{Rn}$  were detected in some of the measured water samples. In terms of the pathway exposure to humans internal dose rate calculations were carried out. The  $^{226}\text{Ra}$  ingestion resulted to high dose rates ( $25\text{--}175 \mu\text{Sv y}^{-1}$ ), with the maxima reaching the recommended limit of  $100 \mu\text{Sv y}^{-1}$ . As a final conclusion, natural radionuclide concentrations and the consequent dose rates to humans must be taken into account in

areas of elevated natural radioactivity background, especially in terms of domestic use of water.

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