Technical note

Determination of uranium and radon in potable water samples

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Abstract

In this work, potable water samples collected from boreholes of the Migdonia valley, located NE of the city of Thessaloniki, were analyzed for the determination of uranium (^{238}U) and radon (^{222}Rn) concentrations. The objective of the present work is to examine if there is any correlation between radon and uranium concentrations in the water samples.

For the determination of traces of uranium in water samples, an analytical technique was developed based on the selective adsorption of uranium on the chelating resin, SRAFION NMRR, and the in situ determination of the retained uranium by instrumental neutron activation analysis (INAA). By the described procedure, it was possible to determine uranium amounts in the range of μg/l. For measuring radon in water, a liquid scintillation counting system, using the Packard protocol was employed. The measured ^{222}Rn activity concentrations are from background level up to $160 \, \text{Bq} \, \text{l}^{-1}$. © 2002 Published by Elsevier Science Ltd.

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1. Introduction

The area under investigation is the Migdonia Basin (Fig. 1) located NE of the city of Thessaloniki, Greece. The extent of the examined area is about $1100 \, \text{km}^2$ and encompasses an area of about $600 \, \text{km}^2$ large valley, with the Lakes of Koronia and Volvi. These Lakes are situated in two different geological formations, Koronia in the Circum-Rhodope Belt and Volvi in the Serbo-Macedonian Massif (Jacobshagen, Geologie von Griechenland, & Borntraeger, 1986). The Migdonia valley is an E–W

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graben. The E–W faults are large, some km in length, and are still active. The tectonic depression of the Migdonia basin is controlled by at least one active fault with recent strong seismic activity, the catastrophic earthquake of 1978. In Fig. 1, two types of faults are shown, the intensive faults that are the deep ones and the normal faults. The intensive faults are on the west part of the Lake Volvi and give earthquakes of magnitudes 4.5–5.9 on the Richter scale. Also, in this area there are thermal waters. It seems that the deep faults are down to the hot nucleus and so radon probably is transported through the faults.

Around the Migdonia basin granitic and/or igneous granitoid bodies exist, that contain elevated concentrations of natural radioelements, such as uranium, radium, radon, thoron, etc. In the cycle of the different natural weathering processes these elements are transferred into surface and subsurface water horizons.

Potable waters from areas with volcanic activity, seismicity, tectonism and the thermal waters (Crenshaw, Williams, & Stoiber, 1982; King, 1978; Hatzidimitriou, Papazachos, & Karakaisis, 1994) have elevated concentrations of radionuclides in the uranium and thorium series.

2. Experimental

Fig. 1 is a map of the area showing the sampling locations. The water samples were collected on 19th of December 1999. All the samples are from taps that are supplied directly from boreholes. From each village, two samples were collected simultaneously from the same tap, one for the determination of ^{238}U concentration and another one for the determination of ^{222}Rn.

2.1. Uranium determination

For the uranium separation from water samples the chelate-forming anion-exchanger with thiourea groups, SRAFION NMRR from Ayalon, Haifa, Israel, was used (Srafion NMRR; Ochsenkühn-Petropulu & Parissakis, 1985; Ochsenkühn, Ochsenkühn-Petropulu, Papadopulos, & Parissakis, 1996). To 1.5 l of water samples 0.5 ml of the regenerated and swollen chelating ion exchanger, with a grain size of about 0.8 mm, were added. The mixture was stirred for 1 h. Then, the ion exchanger was separated quantitatively by filtration and dried for 10 min on the filter. The dried grains were then transferred into small polyethylene capsules of 4 mm ID and about 0.4 ml volume, which were afterwards heat-sealed. The heat-sealed capsules with the samples were irradiated together with reference capsules, containing 10, 20 or 35 µl of a 1.198 µg uranium per ml of standard solution, at the swimming pool reactor of the NCSR "Demokritos", Athens, for 15 min at a thermal neutron flux of 8×10^{13} cm^{-2} s^{-1}. After a cooling time of 1 day, the capsules were transferred into glass-tubes of 10 mm diameter and measured with an Accuspec 4096 channel multi-channel analyzer system (Nuclear Data). The counting time was 1 h, using a Canberra HP germanium well-type detector (well diameter 16 mm, depth 40 mm, FWHM = 1.9 keV for the Co-60 photopeak at 1332 keV, rel. efficiency = 20.9%). The
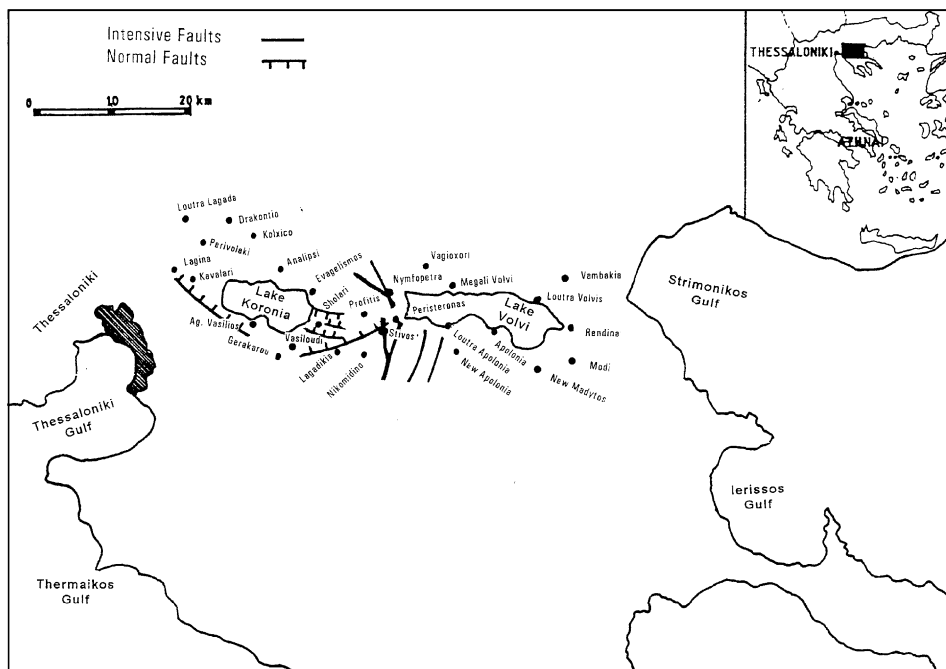
Fig. 1. Map of the investigation area with the sampling points.

gamma-spectra was obtained by the Cambera OS/2 program, "Spectroscopy Assistant V2.1".

2.2. Radon determination

Radon measurements were performed using the Packard Tri-Carb 2560 TRXL Liquid Scintillation Analyzer (LSA), interfaced to a PC containing the Packard Applications Management System (AMS) and spectrograph software packages. Samples were counted with a wide window setting of 25–900 keV as in the method described by Kappel, Keller, Kreienbrock, and Nickels (1993). The rest of the counting parameters and procedures, including equipment standardization and normalization, were set and carried out, respectively, as described in Packard Pico-Rad 5.9 Program for radon in water measurements (Packard, 1988). For the determination of radon concentrations, all the samples have been measured by the LSC system for 50 min, 30–50 h after sampling. The lower limit of detection varied with measured time after sampling but generally for the above mentioned parameters it is not higher than 3 Bq l^{-1}. The lower limit of detection is estimated by supposing that a sample counting rate is $(3\sqrt{B})$ per 50 min, where B are the background counts collected in 50 min. The background counts have been obtained by the analysis of a water sample free of radon.

For sampling, 40 ml glass vials sealed with a Teflon-rubber disk are used to prevent leakage of radon from water. The water sampling has been conducted with a minimum of aeration. The vials are filled to the brim without leaving any trapped air. The reason is that radon prefers to be in air rather than in water in the ration 4 : 1 (at room temperature $\sim 20^{\circ}$C). This could give an apparent radon in water concentration lower than the true concentration.

In the laboratory, a 20 ml syringe is filled with 10 ml of the appropriate elution cocktail (OPTI-FLUOR, blend of long chain alkylbenzenes with scintillators PPO and bis-MSB), and 10 ml of water from the bottom of the sampling vial is drawn into the syringe. The mixture is then transferred to a liquid scintillation vial for counting and is shaken for about 5 s before being set aside for equilibration to take place. After 3 h, the equilibrium between radon and its daughters is established and the elution is more than 95% complete. The counts per minute obtained in the Liquid Scintillation Counting system after at least 4 h yields the radon concentration.

Each decay of radon in water results in 5 detected counts: three alpha particles and two beta particles. The activity of radon in water is given by the following formula:

$$C = (N - N_{B}) \times 100 \times 0.017 \times Exp(\lambda t)/(5 \times 0.964), \tag{1}$$

where C is the radon concentration at the time of sampling (Bq l^{-1}), N the sample count rate (count min^{-1}), N_{B} the background count rate (count min^{-1}), t the elapsed time between sample collection and counting (min), λ the ^{222}Rn decay factor (1.3×10^{-4} min^{-1}), 100 the conversion factor from per 10 ml to per 1 l, 0.017 the conversion factor from disintegration per min to Bq, 5 the conversion factor from counting efficiency for 5 detected particles to ^{222}Rn disintegration and 0.964 the fraction of ^{222}Rn in the Opti-Fluor cocktail (At room temperature the proportional strengths of the partition functions of water, the scintillator Opti-Fluor and air is 1 : 48 : 4. So air has 4 times the partition function of water and Opti-fluor has 12 times the partition function of air. A 22 ml liquid scintillation vial for counting, contains 10 ml Opti-Fluor, 10 ml water and 2 ml air. At equilibrium, the Opti-Fluor contains 96.4% of the radon i.e. $480/(10 + 480 + 8)$.)

The background counts have been obtained using 10 ml of cocktail and 10 ml of water drawn from municipal tap several months before. The background rate is 35 counts min^{-1}.

3. Results and discussion

By using two different radioanalytical methods, radon and uranium have been determined in water samples from the Migdonia basin in Northern Greece. The analyses results for both the radionuclides are presented in Table 1. As it can be seen by the frequency distribution of uranium and radon, most of the data are lower than 7 µg l^{-1} for uranium (Fig. 2) and from 4 to 30 Bq/l for radon (Fig. 3).

An interesting observation can be made, when looking at the distribution of the two elements in the investigated area (Figs. 4 and 5). Using least-square smoothing for a trend surface calculation, uranium trend surface increases in western and

Table 1
Uranium and radon concentrations in water samples collected on the 19th of December from the Migdonia basin

| Sampling point	Uranium ($\mu g\,l^{-1}$)	Radon ($Bq\,l^{-1}$)
Vasiloudi	3.9	Below DL
Gerakarou	15.2	19.3
Lagadikia	0.94	9.3
Stivos	0.67	112
Peristeronas	5.52	161
Loutra Apolonias	1.62	15
Apolonia	4.15	19
Nea Madytos	3.51	14.5
Modi	6.78	10.2
Vambakia	0.26	8
Loutra Volvi	0.31	8
Vagioxori	48.9	40
Nymfopetra	4.84	62
Profitis	1.09	16
Evagelismos	2.25	14.3
Analipsi	9.06	13
Drakontio	2.13	112
Kolxico	0.03	14
Loutra Lagada	0.51	12
Aghios Vasilios	0.70	16
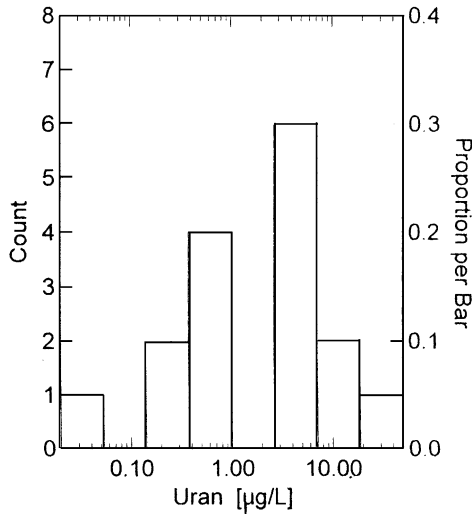
Fig. 2. Histogram of the log-normal frequency distribution of uranium.
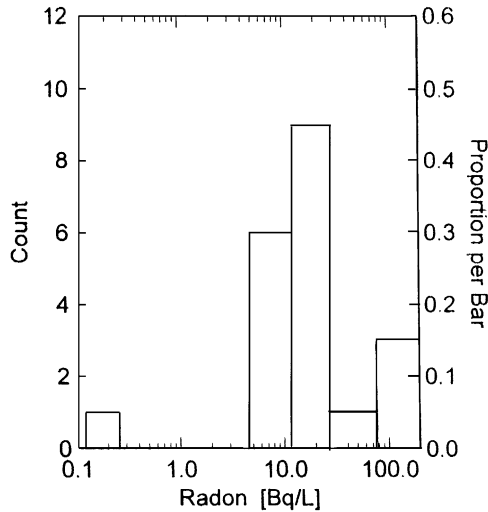
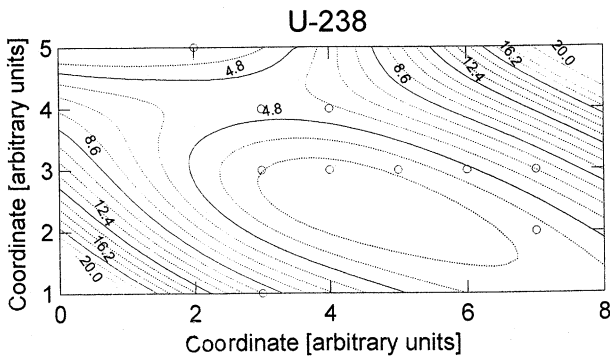Fig. 3. Histogram of the log-normal frequency distribution of radon.

Fig. 4. Simplified area of investigation with trend structures for uranium obtained by least-squares smoothing (isolines $\mu g\, l^{-1}$)

northeastern direction, leaving a minimum surface in the middle and the southeastern area. In contrast to uranium, the radon shows a maximum in the middle part and falls to the borders of the investigated area. That means, that more or less the trend structures are both connected to the graben structure. Assuming that there are post volcanic activities from nuclei and leaching processes of these subsoil formations, the following model could be made for the investigated area. As intensive faults often cut aquifers, interrupting the water circulation from one side of the fault to the other, it looks as if this occurred also in the Migdonia basin. In between the two lakes, a series of faults can be found, lined up in north–south direction, which blocks any groundwater movements in east–west direction. So uprising ground waters are forced in the investigated area to move westward in the
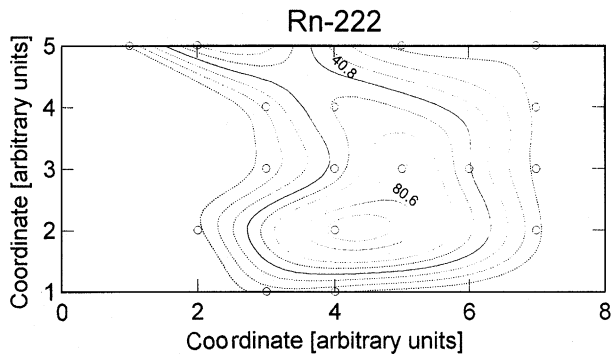
Fig. 5. Simplified area of investigation with trend structures for radon obtained by least-squares smoothing (isolines $Bq\,l^{-1}$).

western part and eastward in the eastern. Waters from the fault area, as very young geological formations, show less uranium. On the other hand for radon as a gaseous element, it is easier to leave the subsoil over small channels and clefts in the intensive fault areas as an effect of pressure reduction. This is a very probable explanation of why there is no correlation between the two elements.

4. Conclusion

Potable water samples from the Migdonia basin in Northern Greece have been analyzed for the determination of radon and uranium. Migdonia basin is an area with seismicity. Intensive faults as well as normal faults are in existence in the area. The results of analyses showed that uranium trend surface leaving a minimum surface in the middle and the southeastern area and radon shows a maximum in the middle part falling to the borders of the investigated area. The data are in the range from 0 to $50\,\mu g\,l^{-1}$ for uranium and from detection limit to $160\,Bq\,l^{-1}$ for radon.

Acknowledgements

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