ORIGINAL ARTICLE

Time variation of radon in tap water in locations of a Greek area with geological background for elevated radon-in-water concentrations and correlation study between radon, gross alpha, uranium, and radium concentrations

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Abstract

Background: Radon (^{222}Rn), a naturally occurring radioactive gas, dissolves in water, and it can be found in elevated concentrations in public water supplies when water originates from ground sources in areas rich in uranium. An area of great interest for measuring radon-in-water is the Migdonia basin in Northern Greece due to its geological background and because all of its villages are supplied with water from boreholes.

Objectives: The main aim of this paper was to study the time variation of radon in tap water activity concentration in nine villages of the Migdonia basin supplied with water from boreholes and to determine factors that may affect it. Radon in water correlation between the source (borehole) and the consumption point (tap) was studied for some villages. Also, the correlation among radon, gross alpha, beta, uranium (^{238}U), and radium (^{226}Ra) activity concentration in water was studied.

Design and methods: Water samples were collected and measured for their radon activity concentration from 66 villages in the Migdonia basin in order to find places with relatively high radon concentrations. The time variation of radon-in-water was studied for villages that showed relatively high radon concentrations for 3–4 years (2018–2022). All samples were measured for their ^{222}Rn activity concentration using gamma-ray spectrometry. Water samples were also analyzed for their gross alpha, beta, and uranium isotopes activity concentration.

Results and conclusions: Average radon in tap water activity concentrations measured in the area ranged from background concentrations to 185 Bq L^{-1}. The corresponding annual effective doses from waterborne radon inhalation using both UNSCEAR and ICRP dose conversion factors ranged from 0.01 to 0.466 mSv y^{-1} and from 0.02 to 0.868 mSv y^{-1}, respectively, while radon ingestion annual effective doses varied from 0.007 to 0.324 mSv y^{-1}. Time variation of radon activity concentration in tap water for villages supplied from one borehole or a constant number of boreholes showed relatively low standard deviations (<24%) at a coverage factor of k = 1. Those deviations are probably caused by the time variation of boreholes' radon concentration and water demand changes. A significant decline in radon concentration from the source (borehole) to the consumption point (tap water) was observed. Therefore, sampling should be performed at the consumption point. However, knowing the supplying borehole concentration is useful as it determines the potential for radon in drinking water. No apparent correlation was found among radon, gross alpha, beta, uranium, and radium concentrations in water. However, in some cases, remedial actions (withdrawal of boreholes) for uranium concentration also decreased radon concentration.

Keywords: *natural radioactivity; radon-in-water; time variation; borehole (source); tap water (consumption point); effective dose*

Drinking waters generally contain variable amounts of radioactivity from natural or anthropogenic sources. Many radionuclides, especially those belonging to the natural decay series of thorium and uranium, are transferred into the water from the aquifer rocks by erosion and dissolution mechanisms and are more commonly detected in supplies derived from groundwater sources (1). While surface waters are exposed

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to artificial radionuclide contamination caused by radioactive fallout, in groundwaters, usually only natural radionuclides can be detected.

Water ingestion is one of the pathways of incorporating radioactive substances into the human body. That is why in recent years, a great interest arose toward radioactivity in drinking water. The Council Directive 2013/51/EURATOM of 22 October 2013 (2) laying down requirements for the protection of the health of the general public concerning radioactive substances in water intended for human consumption, which has been transported in the Greek legislation in 2016, obliged public authorities to organize drinking water surveys throughout the country. The Directive lays down values for radon, tritium, and the indicative dose, which covers many other radionuclides (2).

Radon (^{222}Rn), a naturally occurring radioactive gas, is of great interest when considering radioactivity in drinking water, mainly when water derives from ground sources. It can be found in elevated concentrations when water municipality supplies derive from natural springs, boreholes, or wells (3). Radon dissolves in water, and, therefore, when the water used for human consumption comes from groundwater in areas where the rocks are rich in uranium (^{238}U), the radon concentrations in the water can be particularly significant (4). It is known that some types of rocks have higher uranium concentrations. These include volcanic rocks, granites, dark shales, sedimentary rocks that contain phosphate, and metamorphic rocks derived from these rocks (5).

Human exposure to radon from domestic water use can occur through ingestion and inhalation, with the latter being the most important (6). Radon becomes airborne from water during its different uses in homes, like showering, dishwashing, and cooking. Previous studies have estimated that 10 Bq L^{-1} of radon (^{222}Rn) in water contributes to an increase of 1 Bq m^{-3} of radon in indoor air (7).

An area of great interest for measuring radon-in-water due to its geological background and because most of its villages are supplied from boreholes is the Migdonia basin in Northern Greece. A previous study conducted in this area in 1999 showed that radon concentrations in drinking tap water ranged from background concentrations to 170 Bq L^{-1} (8).

This study aims to determine the radon-in-water activity concentration in most locations (villages) in the Migdonia basin, to find places with relatively high radon concentrations, and to assess the annual effective dose from inhalation and ingestion of waterborne radon. Also, the time variation of radon-in-water concentration was monitored (for 3–4 years) in nine locations that showed relatively high radon concentrations to point out factors that may affect it. Water samples from locations that showed relatively high radon concentrations were analyzed for their gross alpha, beta, and uranium isotopes activity concentrations. An attempt was made to correlate radon-in-water activity concentration with gross alpha, beta, uranium (^{238}U), and radium (^{226}Ra) concentrations in water.

Materials and methods

Study area and sampling method

Granitic and igneous granitoid bodies with elevated concentrations of natural radioactive elements exist around the Migdonia basin. The water supply for the population (50367 inhabitants) of this area derives from boreholes located in the basin sediments and the metamorphic rocks and granites. Some locations (villages) are supplied with water from more than one borehole. Drinking tap water from 66 locations in the Migdonia basin was collected and measured from 2018 to 2022. Nine locations that showed relatively high radon concentrations were measured more than once for 3–4 years (2018 to 2022) to study the time variation of their radon-in-water concentration. The radon-in-water concentration of supplying boreholes for some of these places was also measured.

All water samples were carefully collected with a sampling method previously checked by the authors for its reliability (9). The sampling method involves the connection of the tap to a water hose and immersing the water hose in the sampling container. Before collecting the sample, water was left to flow at a high rate for 10 min to obtain fresh water. Aluminum sampling containers were used as they are non-porous to radon (10–12). The containers used were previously checked for their radon tightness and showed no loss of radon within standard uncertainties as their fitting curves followed the literature radon decay curve (9). The containers were fully filled, and no space was left for radon to escape after closing the containers with their cap. Figure 1 shows the locations (villages) from which the water samples were collected.

Radon in the water measurement method

All water samples were measured for their radon activity concentration using gamma-ray spectrometry with a high purity germanium detector (HPGe) of 50% relative efficiency at 1,332 keV. The samples were transferred carefully from the original (aluminum) sampling container into our standard sample counting container to measure radon-in-water activity concentration. No radon loss due to this transfer from the original container into our standard sample counting container has been found (9). Our standard sample counting container is a cylindrical container of 260 mL capacity. Radon (^{222}Rn) activity concentration was determined using the gamma-ray peaks of ^{214}Pb (352 keV) and ^{214}Bi (609 keV) after secular equilibrium between ^{222}Rn and its

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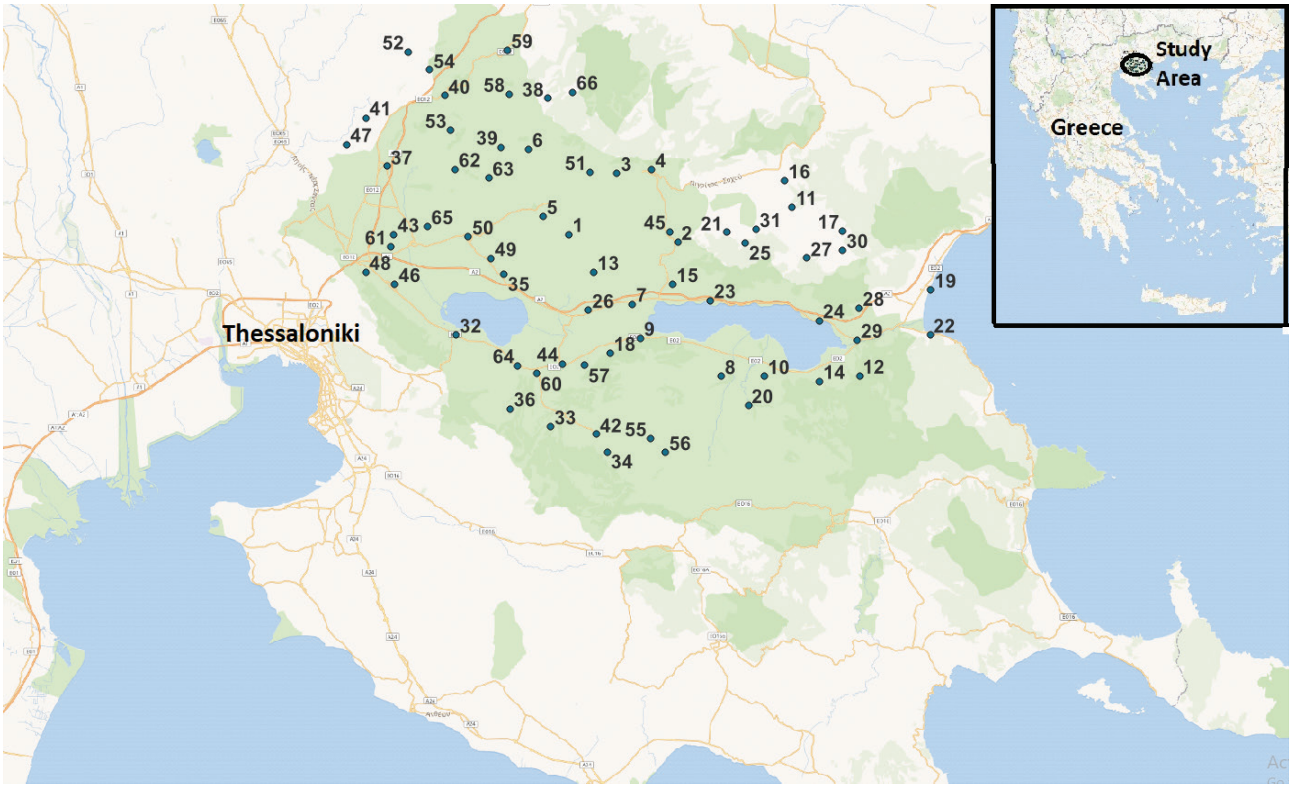

Fig. 1. The location of water sampling sites (villages) in the Migdonia basin in Northern Greece. The numbers are the identification (ID) numbers of the villages (1–66).

short-lived daughters (^{214}Pb and ^{214}Bi) was established. Gamma-ray spectrometry efficiency values were obtained experimentally using a multinuclide standard source. The counting time depends on the activity concentration of each sample. Our gamma-ray spectrometry method's detection limit (DL) is 2 Bq L^{-1}.

Annual effective dose due to radon-in-water inhalation and ingestion

Radon in drinking tap water leads to exposure from the ingestion of drinking water and from the inhalation of radon released into the air when water is used.

Annual effective dose due to ingestion of radon in water

The annual effective dose due to ingestion of radon-in-water was calculated using the relationship:

$$AED_{ing}(mSv\ \ y^{-1}) = DCF_{ing} \times C_{W,avg} \times C_{W,rate} \times 10^{-6}$$

where DCF_{ing} is the ingesting dose conversion factor of ^{222}Rn, $C_{W,avg}$ is the average radon-in-water concentration (Bq L^{-1}), and $C_{w,rate}$ is the water consumption rate consumed directly from the tap per year (L y^{-1}). According to UNSCEAR (7), DCF_{ing} and $C_{w,rate}$ for adults are equal to 3.5 nSv Bq^{-1} and 500 L y^{-1}, respectively. 10^{-6} is the unit conversion factor from nSv to mSv.

Annual effective dose due to inhalation of radon released from tap water

The annual effective dose due to inhalation of radon released from tap water was calculated using the relationship:

$$AED_{inh}(mSv\ \ y^{-1}) = DCF_{inh} \times C_{W,avg} \times 10^{3} \times T \times 10^{-4} \times 0.4 \times 10^{-6}$$

where DCF_{inh} is the dose conversion factor for inhalation of ^{222}Rn, $C_{W,avg}$ is the average radon in water concentration (Bq L^{-1}), T is the average indoor occupancy time per person (7,000 h y^{-1}), 10^{-4} is the ratio of radon in the air to water, and 0.4 is the equilibrium factor between radon and its daughters. According to UNSCEAR (13) and ICRP (14), the dose conversion factors for the inhalation of ^{222}Rn are 9 $\frac{nSv\ h^{-1}}{Bq\ m^{-3}}$ and 16.75 $\frac{nSv\ h^{-1}}{Bq\ m^{-3}}$, respectively. 10^{3} is the conversion factor from $\frac{Bq}{L}$ to $\frac{Bq}{m^{3}}$, and 10^{-6} is the unit conversion factor from nSv to mSv.

Gross alpha, beta, and uranium isotopes activity concentrations

All chemical reagents were of analytical grade. Three-liter water samples were collected by laboratory personnel in 2020. After filtering, the water samples were acidified with nitric acid to a pH of about 2.0 at the laboratory on the same day of reception and stored for later analysis. Alpha/beta activity and uranium isotopes analysis were performed in 10 samples, and ^{210}Po and ^{226}Ra analysis in the same.

Alpha/beta activity measurement

The samples were prepared according to the ISO 11704 (15) 'Water Quality-Gross alpha and gross beta activity-Test method using liquid scintillation counting'. The prepared samples, 6 mL sample mixed with 14 mL scintillation cocktail ULTIMA GOLD AB, were transferred in polyethylene scintillation vials with a 20 mL capacity and measured by a Quantulus scintillation counter for 600 min. The method's DLs are 0.04 and 0.1 Bq L^{-1} for alpha and beta activity, respectively.

Uranium isotopes ^{226}Ra and ^{210}Po were measured by alpha-spectrometry. The equipment used is a fully automated and integrated alpha spectroscopic system (AAnalyst, Canberra), consisting of 24 Passivated Implanted Planar Silicon (PIPS) detectors with a 600 mm^2 active area. Detection counting efficiency is about 26%. The counting time depends on the activity concentration of each sample. The uncertainties were calculated according to ISO 11929-2 (16). The addition of the tracers ^{232}U, ^{229}Th, and ^{209}Po allows the determination of the activity concentration, and the chemical yield of the uranium isotopes ^{226}Ra and ^{210}Po. The chemical recovery for uranium was found to vary between 90 and 99%, for ^{226}Ra between 48 and 75%, and for ^{209}Po between 85 and 95%, respectively.

Results and discussion

Radon-in-water activity concentrations and effective doses in the Migdonia basin

A total of 268 tap water samples from 66 locations in the Migdonia basin were collected and analyzed for their radon activity concentration during a 4-year period (2018–2022). Each location in the Migdonia basin is supplied with water either from one or more boreholes. Table 1 presents the average radon in tap water activity concentrations measured in each location in the Migdonia basin, along with the coordinates, measurement period, number of measurements, and minimum and maximum radon concentrations observed over the measurement period. Also, the calculated annual effective doses resulting from inhalation and ingestion of waterborne radon are presented. As seen in Table 1, the average ^{222}Rn tap water activity concentrations in the studied area range from background concentrations up to 185 Bq L^{-1}. This wide range observed in tap water radon concentrations is also a sign of a significant wide range among the radon concentrations of boreholes supplying these locations. Two locations showed average radon concentrations higher than the parametric value of 100 Bq L^{-1} adopted by Greece in accordance to EURATOM (2). EURATOM sets a range of parametric values between 100 and 1,000 Bq L^{-1} and states that the 'Remedial action is deemed to be justified on radiological protection grounds, without further consideration, where radon concentrations exceed 1,000 Bq/L^{-1}'. Although only two of the 66 locations showed average radon-in-water concentrations higher than the parametric value, four other locations overcame the parametric value at least once through the measurement period. The annual effective doses from ingestion of radon ranged from 0.007 to 0.324 mSv y^{-1}. Regarding radon inhalation, the annual effective doses ranged from 0.01 to 0.466 mSv y^{-1} using the UNSCEAR dose conversion factor and from 0.02 to 0.868 mSv y^{-1} using the ICRP dose conversion factor.

Comparing this study's results with a previous study conducted in the same area in 1999 (8), the most significant differences are observed in locations 8, 9, and 18. Radon in tap water measured in Location 8 in 1999 was 101 Bq L^{-1}, while the average value of the concentration measured in the context of the present study is 25 Bq L^{-1}. This difference is due to a withdrawal of a borehole used to supply the village in 1999. Locations 9 and 18 also showed a significant decline in their radon concentrations as their concentrations dropped from 170 and 112 Bq L^{-1} to 13 and 38 Bq L^{-1}, respectively. Most probable boreholes with elevated radon concentrations used to supply these villages in the past have been withdrawn.

Time variation of radon-in-water activity concentrations in selected villages

The time variation (3–4 years) of radon-in-water activity concentration was studied in nine locations (supplied from boreholes) of the Migdonia basin and presented in Figures 2–4. For some of these locations, the radon concentration of their water-supplying boreholes was also measured and presented in Table 2. Table 2 also shows the locations' tap water radon concentrations as measured on the same day of their supplying boreholes measurement. For some locations supplied from more than one borehole, the water supply does not always derive from all the boreholes. Mainly, for these locations, the water demand determines the participation of boreholes in their water supply. Also, a change in the number of boreholes supplying a location can occur, e.g. due to a problem relating to the water supply system. Locations provided with water from more than one borehole receive a mixture of water from the boreholes as it mixes in tanks before reaching the tap water of the place.

As seen in Figures 2 and 3, locations 1 and 5, supplied by one (different for each location) borehole, showed standard deviations for their average ^{222}Rn

Table 1. Radon in tap water activity concentration of locations (villages) in the Migdonia basin along with the annual effective doses resulting from inhalation and ingestion of waterborne radon

Location ID	Latitude	Longitude	Measurement period	Number of measurements	Average ^{222}Rn concentration ($Bq\ L^{-1}$)	STDEV [a] (k = 1)	Min ($Bq\ L^{-1}$)	Max ($Bq\ L^{-1}$)	AED_{inh} [b] ($mSv\ y^{-1}$)	AED_{inh} [c] ($mSv\ y^{-1}$)	AED_{ing} ($mSv\ y^{-1}$)
1	40.75760500	23.25401500	Mar-2019 – Jun-2022	24	69	15	46	114	0.174	0.324	0.121
2	40.75021700	23.38737500	Jun-2020 – Jun-2022	24	64	14	31	89	0.160	0.298	0.111
3	40.81417200	23.31297400	Oct-2018 – Jun-2022	31	90	17	63	128	0.228	0.424	0.158
4	40.81755000	23.35532100	Jan-2019 – Jun-2022	33	48	15	23	104	0.121	0.225	0.084
5	40.77426000	23.22248600	Mar-2019 – Jun-2022	28	185	35	89	239	0.466	0.868	0.324
6	40.83646000	23.20453300	Jan-2020 – Jun-2022	10	83	28	31	123	0.209	0.388	0.145
7	40.69285000	23.33196800	Aug-2019 – Feb-2022	6	33	8	22	44	0.084	0.157	0.058
8	40.62575600	23.44049300	Apr-2018 – Jun-2022	18	25	24	2	97	0.063	0.117	0.044
9	40.66125800	23.34167600	Apr-2018 – Jun-2022	15	13	4	7	23	0.033	0.061	0.023
10	40.62578366	23.49315243	May-2021	1	38	4	38	38	0.095	0.176	0.066
11	40.78266390	23.52758880	Dec-2020	1	7	1	7	7	0.017	0.032	0.012
12	40.62608456	23.60979213	May-2021	1	15	2	15	15	0.038	0.070	0.026
13	40.72237900	23.28457500	Mar-2019 – Feb-2022	5	38	11	19	51	0.096	0.179	0.067
14	40.62117493	23.56104350	May-2021	1	6	1	6	6	0.016	0.029	0.011
15	40.71116600	23.38122100	Jan-2019 – Nov-2021	2	65	20	51	79	0.164	0.305	0.114
16	40.80728810	23.51833560	Dec-2020	1	9	1	9	9	0.023	0.043	0.016
17	40.76110630	23.58948490	Dec-2020	1	5	1	5	5	0.012	0.023	0.009
18	40.64731500	23.30457400	Feb-2019 – Sep-2020	3	38	10	29	52	0.096	0.179	0.067
19	40.70602180	23.69714660	Dec-2020	1	9	1	9	9	0.024	0.044	0.016
20	40.59865500	23.47380000	Mar-2019 – Sep-2020	2	15	2	15	15	0.038	0.071	0.026
21	40.75945000	23.44690800	Oct-2018 – Dec-2020	2	19	5	14	24	0.048	0.090	0.033
22	40.66417200	23.69626500	Sep-2017 – May-2021	5	13	2	11	15	0.032	0.060	0.022
23	40.69621900	23.42723600	Sep-2018	1	12	1	12	12	0.030	0.056	0.021
24	40.67769600	23.56053200	Mar-2019	1	10	1	10	10	0.026	0.048	0.018
25	40.74993400	23.47015300	Oct-2018	1	11	1	11	11	0.027	0.050	0.019
26	40.68712300	23.27806600	Jan-2019 – Feb-2022	4	11	1	9	13	0.027	0.050	0.019
27	40.73562300	23.54530300	Oct-2018	1	8	1	8	8	0.020	0.037	0.014
28	40.68907600	23.60916800	Mar-2019	1	12	1	12	12	0.030	0.056	0.021
29	40.65943200	23.60722300	Mar-2019	1	15	2	15	15	0.037	0.068	0.026
30	40.74282000	23.58918800	Oct-2018 – Dec-2020	2	7	1	6	8	0.019	0.035	0.013
31	40.76220080	23.48305040	Jan-2020 – Dec-2020	2	16	7	9	23	0.039	0.073	0.027
32	40.66419300	23.11641700	Sep-2020	1	16	2	16	16	0.041	0.077	0.029
33	40.57895396	23.23185700	Feb-2021	1	13	1	13	13	0.033	0.061	0.023
34	40.55570700	23.30102200	Mar-2019	1	8	1	8	8	0.020	0.038	0.014

Table 1. (Continues)

Location ID	Latitude	Longitude	Measurement period	Number of measurements	Average ^{222}Rn concentration (Bq L^{-1})	STDEV [a] (k = 1)	Min (Bq L^{-1})	Max (Bq L^{-1})	AED_{inh} [b] (mSv y^{-1})	AED_{inh} [c] (mSv y^{-1})	AED_{ing} (mSv y^{-1})
35	40.72102500	23.17467100	Mar-2019	1	9	1	9	9	0.021	0.040	0.015
36	40.59576100	23.18206500	Sep-2020	1	7	1	7	7	0.018	0.033	0.012
37	40.82070000	23.03160000	Feb-2020	1	24	2	24	24	0.060	0.111	0.041
38	40.88362900	23.22821600	Jan-2020	1	10	2	10	10	0.025	0.046	0.017
39	40.83849500	23.17146900	Feb-2020	1	34	5	34	34	0.086	0.160	0.060
40	40.88614800	23.10213400	Feb-2020	1	34	5	34	34	0.084	0.157	0.059
41	40.86569100	23.00558500	Feb-2020	1	12	2	12	12	0.031	0.057	0.021
42	40.57196400	23.28756500	Apr-2019 – Feb-2021	2	13	3	10	16	0.033	0.062	0.023
43	40.75754500	23.03982000	Feb-2020	1	6	2	6	6	0.015	0.029	0.011
44	40.63744200	23.24571000	Sep-2020	1	5	1	5	5	0.013	0.024	0.009
45	40.76009900	23.37732600	Sep-2018	1	119	8	119	119	0.300	0.558	0.208
46	40.71120000	23.04110000	Feb-2020	1	5	1	5	5	0.013	0.024	0.009
47	40.84100000	22.98200000	Feb-2020	1	8	2	8	8	0.021	0.038	0.014
48	40.72240000	23.00530000	Feb-2020	1	7	2	7	7	0.017	0.032	0.012
49	40.73519800	23.15900100	Nov-2019	1	6	1	6	6	0.014	0.027	0.010
50	40.81556400	23.28041900	Nov-2019	1	<DL	0	1	1	0.001	0.002	0.001
51	40.81556400	23.28041900	Nov-2019	1	19	2	19	19	0.048	0.089	0.033
52	40.92667900	23.05700400	Feb-2020 – Jan-2021	2	7	0.4	6	7	0.017	0.031	0.012
53	40.85412200	23.10916700	Feb-2020	1	29	6	29	29	0.073	0.135	0.050
54	40.90992506	23.08355372	Jan-2021	1	9	2	9	9	0.022	0.041	0.015
55	40.56795648	23.35385987	Feb-2021	1	73	5	73	73	0.183	0.341	0.127
56	40.55499300	23.37225000	Mar-2019 – Feb-2021	2	19	10	9	30	0.049	0.091	0.034
57	40.63611900	23.27316400	Dec-2018	1	22	3	22	22	0.054	0.101	0.038
58	40.88743800	23.18077100	Jan-2022	1	7	1	7	7	0.016	0.030	0.011
59	40.92803400	23.17883900	Jan-2020 – Jan-2022	2	39	22	17	61	0.098	0.182	0.068
60	40.62831000	23.21471500	Dec-2018	1	7	1	7	7	0.017	0.031	0.012
61	40.74594600	23.03567300	Jun-2021	1	6	1	6	6	0.015	0.028	0.011
62	40.81786800	23.11528900	Jan-2020	1	6	1	6	6	0.016	0.029	0.011
63	40.80978900	23.15592200	Jan-2020	1	6	1	6	6	0.015	0.028	0.011
64	40.63582500	23.19172500	Feb-2019	1	17	2	17	17	0.042	0.077	0.029
65	40.76482600	23.08108900	Jan-2020	1	11	2	11	11	0.028	0.053	0.020
66	40.88945900	23.25850000	Feb-2020	1	4	1	4	4	0.010	0.018	0.007

[a] STDEV is the standard deviation of the average value of measurements for locations measured more than once, while for locations measured only once is the combined measurement uncertainty at a coverage factor of k = 1.

[b] Using UNSCEAR dose conversion factor.

[c] Using ICRP dose conversion factor.

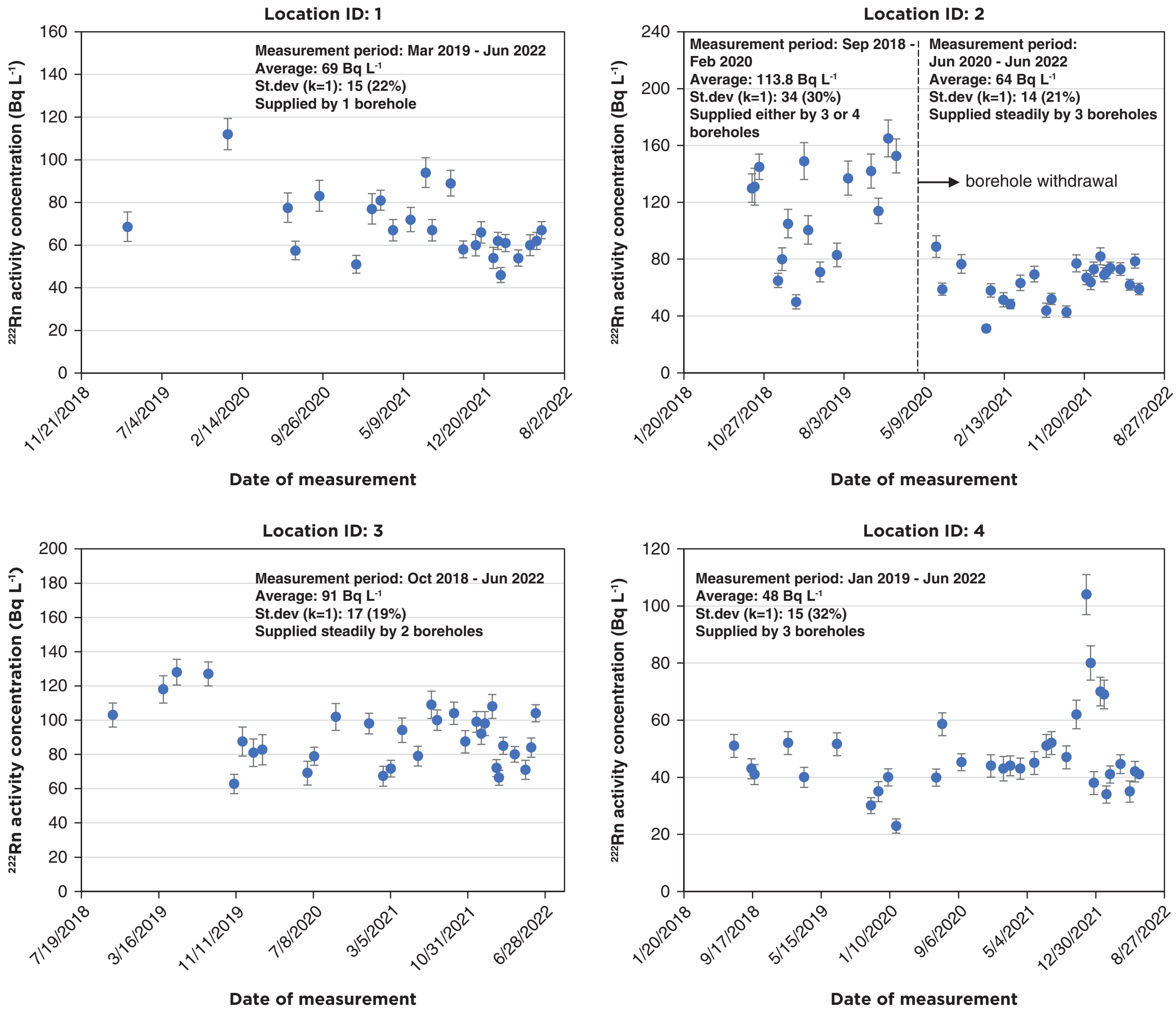

Fig. 2. Time variation of radon-in-water activity concentration for 3–4 years in Locations 1–4 in the Migdonia basin.

activity concentration measured over the 3–4-year period of 22 and 19%, respectively. Location 7 (Figure 3) was supplied from one borehole from December 2018 until August 2019. For this period, the average ^{222}Rn activity concentration measured in tap water was 244 Bq L^{-1} with a standard deviation of 12%. The borehole that provided the village with water was replaced by another borehole due to its ^{238}U concentration. After replacing the borehole, the average ^{222}Rn concentration from August 2019 until February 2022 was 33 Bq L^{-1}, with a standard deviation of 23%. From the above discussions, it can be said that when the water supply was derived from one borehole, the average radon concentrations in the drinking tap water showed relatively low standard deviations of less than 24% at a coverage factor of $k = 1$.

Location 2 (Figure 2) showed an overall (from September 2018 to June 2022) standard deviation for its average ^{222}Rn activity concentration of 41%. It was supplied either by three or four boreholes from September 2018 to February 2020, showing a standard deviation of 30%. Because of remedial actions performed for ^{238}U, one borehole was withdrawn. However, after the borehole's withdrawal and its supply steadily from three boreholes, it showed (from June 2020 to June 2022) a lower standard deviation for its average radon concentration (64 Bq L^{-1}) of 21%. Location 3 (Figure 2), supplied steadily by two boreholes, showed a standard deviation for its average radon concentration (91 Bq L^{-1}) of 19%. From the results discussed for Locations 2 and 3, it can be said that when the water supply was derived from a constant number of boreholes, the average radon concentration in the drinking water showed a standard deviation of less than 22% at

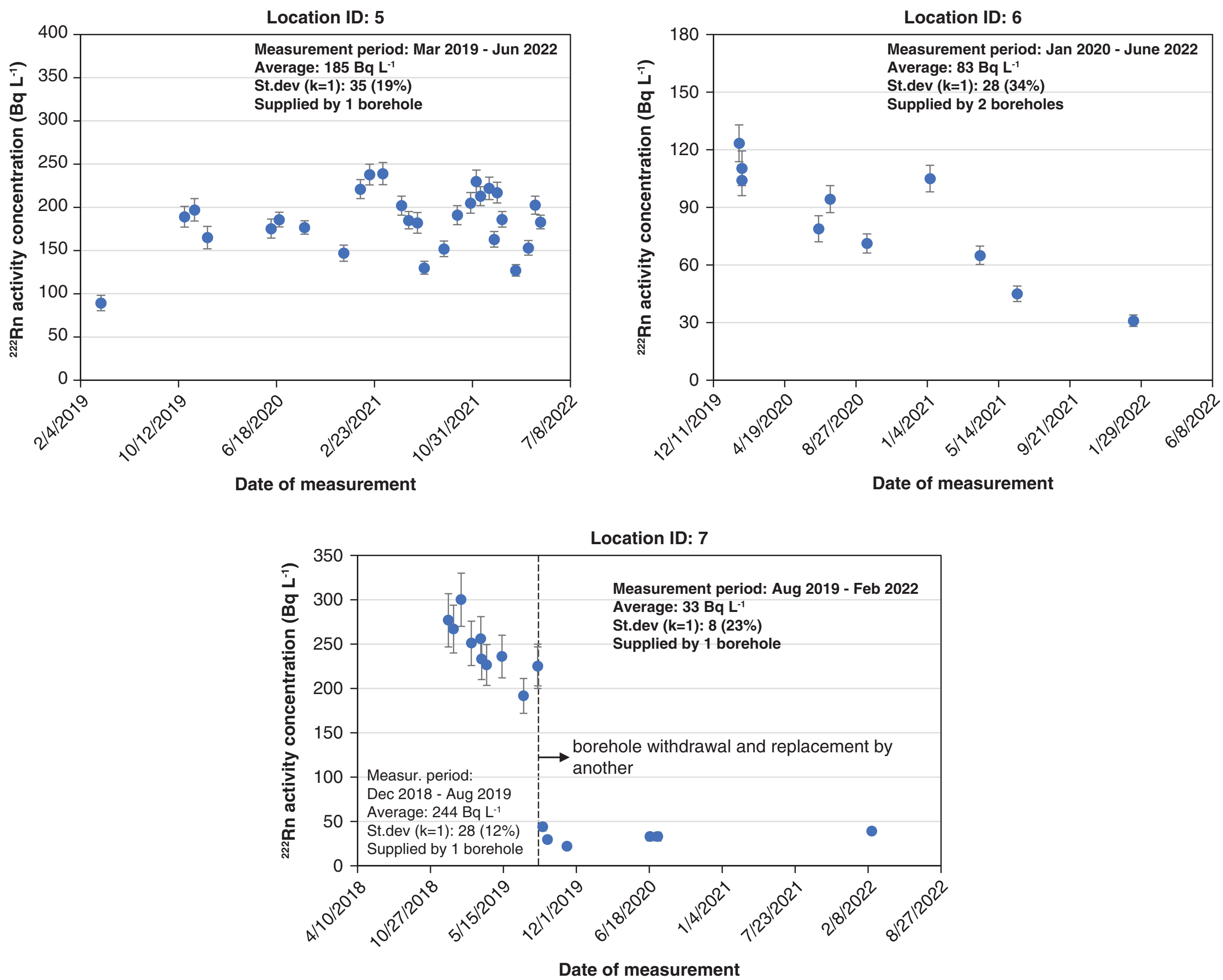

Fig. 3. Time variation of radon-in-water activity concentration for 3–4 years in Locations 5–7 in the Migdonia basin.

a

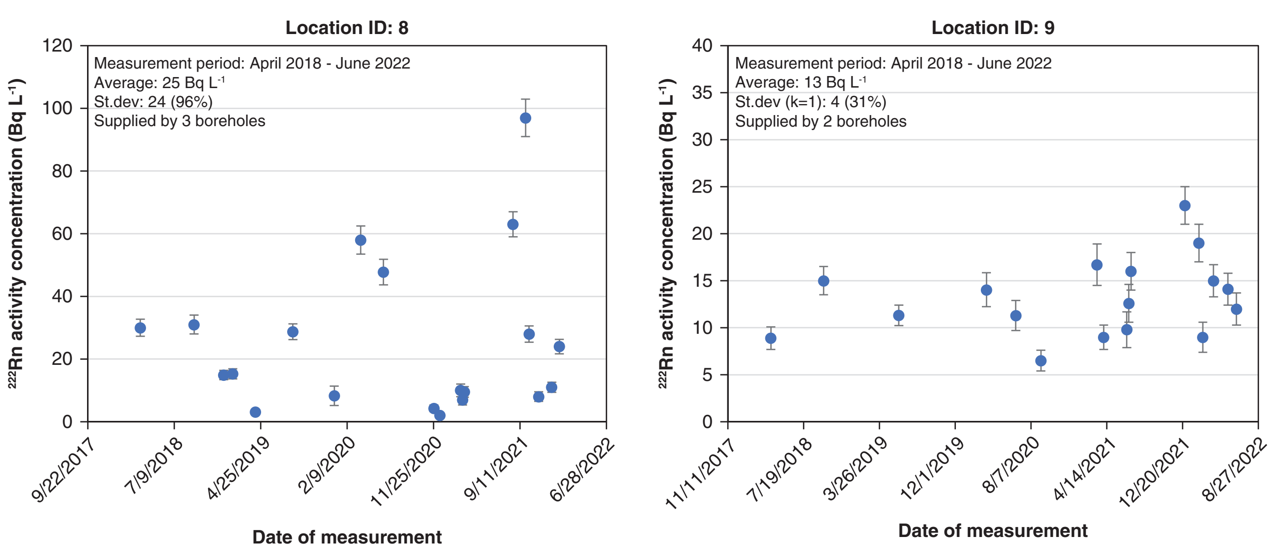

Fig. 4. Time variation of radon-in-water activity concentration for 3–4 years in Locations 8 and 9 in the Migdonia basin.

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Table 2. ^{222}Rn in water activity concentration measured in the tap water of locations (points of consumption) and in supplying boreholes (sources)

Location ID	Supplying borehole's ID	Supplying borehole's ^{222}Rn concentration (Bq L^{-1})	Drinking tap water ^{222}Rn concentration (Bq L^{-1})[d]	Average ^{222}Rn in location (Bq L^{-1})	St.dev (k = 1)	Min ^{222}Rn in location (Bq L^{-1})	Max ^{222}Rn in location (Bq L^{-1})
1	a	97 ± 6	62 ± 4	69	15	46 ± 3	112 ± 7
2	b	217 ± 21[a]	131 ± 13[b]	64[c]	14[c]	31 ± 2.4[c]	89 ± 8[c]
	c	62 ± 7					
	d	132 ± 13					
	e	83 ± 4					
3	f	157 ± 9	84 ± 5.3	91	17	63 ± 5	128 ± 8
	g	29 ± 2.4					
5	k	389 ± 19	203 ± 10	185	35	89 ± 8	239 ± 13

[a]Borehole was withdrawn before June 2020 due to its ^{238}U concentration.

[b]Before the withdrawal of borehole b.

[c]After the withdrawal of borehole b (from June 2020 to June 2022).

[d]Measured on the same day of supplying boreholes measurement.

coverage factor of $k = 1$. It is worth mentioning that the ratio of the contribution of each participating borehole to the total water supply must be relatively constant for the above discussion to be considered credible. This mention is due to the fact that the concentration of radon between the boreholes involved in the water supply of a location can vary considerably, as seen in Table 2.

Location 4 (Figure 2) is supplied with water from three boreholes. It showed an overall (from January 2019 to June 2022) average radon concentration of 48 Bq L^{-1} with a standard deviation of 32%. An abnormal increase (>2k) in its radon concentration was observed in November 2021. Several studies have correlated anomalies in groundwater radon concentrations with pre-seismic and post-seismic phases (17–20). This observation is possibly related to a change in the supplying ratios of the participating boreholes to the total supply, as no seismic activity was recorded during that period near Location 4.

Location 8 (Figure 4) has three boreholes available for its water supply, but it was not supplied continuously from all three boreholes. For this reason, its radon activity concentrations vary significantly, ranging from 2 to 97 Bq L^{-1}. This observation concludes that the radon concentrations of its supplying boreholes also differ significantly. Therefore, special attention must be taken considering radon-in-water measurements in places supplied (not steadily) from more than one borehole because the average (over the year) radon concentration may be underestimated or overestimated, leading to an underestimated or overestimated dose calculation. In these cases, it should be considered which combination of boreholes prevails during the year, and it will be beneficial to know the radon concentration of each borehole to determine the potential of radon in tap water.

Location 6 (Figure 3), supplied with a mixture of water from two boreholes, seems to follow a downward trend in its radon concentration. Most probable, this decline is due to changes in the contribution of each supplying borehole to the total water supply. Location 9 (Figure 4), supplied from two boreholes, showed a standard deviation for its average ^{222}Rn activity concentration (13 Bq L^{-1}) of 31%.

The following are some observations concerning the correlation between radon concentrations measured in borehole water (source) and tap water (consumption) at the locations, according to the results presented in Table 2.

Location 5 is supplied with water from one borehole. Radon concentration was measured the same day (May 2022) in both the borehole and the location's tap water. Radon concentration in the borehole (389 Bq L^{-1}) was 92% higher than in the location's tap water (203 Bq L^{-1}). In 28 tap water measurements in this location, none exceeded the radon concentration measured in its supplying borehole (389 ± 19) Bq L^{-1}.

In Location 1, also supplied from one borehole, the radon concentration measured in the borehole (97 Bq L^{-1}) was 56% higher than in the tap water (62 Bq L^{-1}). In 24 measurements performed in Location 1, the radon concentration exceeded the borehole's concentration (measured once in May 2022) only once. Therefore, radon concentration is expected to be lower at the point of consumption than at the source (borehole) when a location is supplied with water from one borehole.

Regarding the concentration of radon in tap water in areas supplied with a mixture of water from more than one borehole, the concentration in tap water is expected to be lower than the borehole concentration with the highest measured concentration. It is worth noting that if, for some reason, it is necessary, for example, to interrupt the supply of location 3 from borehole g (29 Bq L^{-1}), only borehole f (157 Bq L^{-1}) will supply water to this site. This will increase the radon concentration in the tap water of

Location 3 because the water from the two boreholes will no longer be mixed. Therefore, it is crucial to know the radon concentrations of the boreholes that supply a site, especially if a location is provided with water from more than one borehole. The lower radon concentrations at the point of consumption are caused by the procedures that follow water pumping from the boreholes and that precede its consumption, such as transportation, storage in tanks, and treatment through filtration systems, which can cause both radon loss and radioactive decay (due to delay).

Gross alpha, beta, and uranium isotopes activity concentration results

All the samples obtained have their origin in boreholes. Ten water samples have been examined regarding radioactive substances according to the Council Directive 2013/51/EURATOM. The measured alpha-activity concentration values lie between 0.11 and 5.00 Bq L^{-1} (Table 3). It is noticed that the gross alpha-activity in all samples exceeds 0.1 Bq L^{-1}; for this reason, analysis for specific radionuclides was required. Considering all relevant information about likely sources of radioactivity, the radionuclides of choice for further investigation were the uranium isotopes ^{238}U and ^{234}U. The determined activity concentrations for ^{238}U and ^{234}U lie between 6.5 to 1,800 mBq L^{-1} and 30.1 to 2,640 mBq L^{-1}, respectively. The determination of ^{210}Po was performed in all samples, except sample A. The reason for this determination was that the difference between the alpha-activity concentrations and the uranium isotopes concentrations exceeds the 0.1 Bq L^{-1}, which is the value that corresponds to the derived concentration of ^{210}Po stated in the European Directive. In all the measured samples, the ^{210}Po activity concentrations were less than 0.1 Bq L^{-1}. They varied between 3 to 12 mBq L^{-1}.

Furthermore, in sample F, the alpha-activity concentration was 0.5 Bq L^{-1} higher than the respective uranium isotopes concentrations. Therefore, according to the Council Directive, further investigation was required. The investigation refers to the radionuclide radium-226. The detected ^{226}Ra activity concentrations are less than the parametric value of 0.5 Bq L^{-1} set in the Council Directive and indicate the existence of an additional alpha-emitting radionuclide (Table 4). Also, in samples A and H, ^{226}Ra analysis was performed since elevated radon activity concentration was indicated in these samples.

Besides, in samples F and G, the parametric value is higher than the parametric value of 0.1 mSv y^{-1}, defined by the Council Directive 2013/51/Euratom. In these cases, although the water intended for human consumption from an individual supply served about 50 persons, remedial actions have been taken to comply with requirements for the protection of human health from a radiation protection point of view. The competent authorities and relevant bodies were informed, and they proceeded with remedial actions. For Location 7, the remedial actions concern the replacement of an existing borehole by another borehole, whereas for Location 13, the reduction of uranium through water filtration. The measurement results after remediation for samples F* and G* are presented in Table 3.

Correlation among radon, gross alpha, uranium, and radium concentrations

An attempt to correlate radon-in-water activity concentration with radium, uranium, gross alpha, and beta activity concentrations was made. The results are shown in Figure 5. Although no obvious correlation is observed

Table 4. Radium (^{226}Ra) activity concentration in selected water samples

Location ID	Sample code	^{226}Ra (mBq L^{-1})
4	A	90.7 ± 5.6
13	F	158 ± 9.4
5	H	175 ± 11.5

Table 3. Gross alpha, beta, and uranium isotopes activity concentration results

Location ID	Sample code	^{238}U (mBq L^{-1})	^{238}U (μg L^{-1})	^{234}U (mBq L^{-1})	$^{234}U/^{238}U$	α (Bq L^{-1})	β (Bq L^{-1})	^{222}Rn (Bq L^{-1})
4	A	6.5 ± 1.24	0.52 ± 0.1	30.1 ± 3.2	4.6	0.11 ± 0.01	0.4 ± 0.02	59 ± 4.2
2	B	240 ± 20.3	19.3 ± 1.64	409 ± 32.5	1.7	0.98 ± 0.05	0.66 ± 0.04	44 ± 3.7
3	D	405 ± 28	32.6 ± 2.2	556 ± 37	1.4	1.2 ± 0.07	0.63 ± 0.03	79 ± 5
6	E	92 ± 7.1	7.4 ± 0.57	146 ± 10.5	1.6	0.57 ± 0.04	0.57 ± 0.04	94 ± 5.3
13	F	1,800 ± 119	146 ± 9.6	2,640 ± 172	1.5	5.00 ± 0.18	1.85 ± 0.08	–
13*	F*	100 ± 7.6	8.1 ± 0.6	140 ± 7.2	1.4	0.28 ± 0.02	<0.01	47 ± 3.8
7	G	1,780 ± 82	143.5 ± 6.6	2,536 ± 132	1.4	4.4 ± 0.13	0.7 ± 0.03	244 ± 20
7*	G*	164 ± 11.2	13.2 ± 0.9	315 ± 20.3	1.9	0.77 ± 0.04	0.45 ± 0.02	33 ± 3.6
5	H	246 ± 17	19.8 ± 1.4	367 ± 25.3	1.5	0.79 ± 0.05	0.55 ± 0.03	186 ± 8.4
1	J	300 ± 21.2	24.2 ± 1.7	319 ± 22.3	1.1	0.85 ± 0.05	0.56 ± 0.03	58 ± 4.1

F* & G* samples: measurements after remediation.

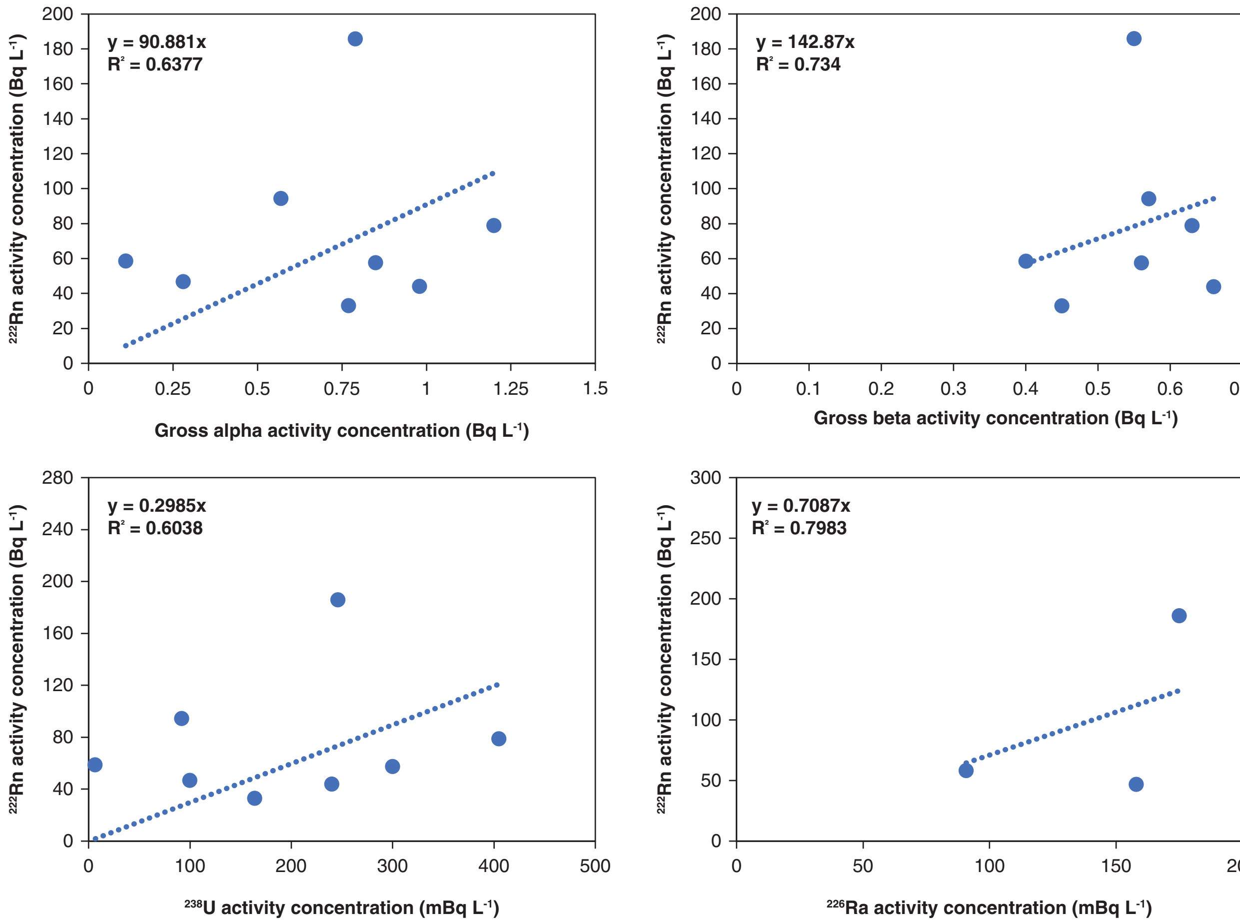

Fig. 5. Correlations between ^{222}Rn and gross alpha, beta, ^{238}U, and ^{226}Ra concentrations in water.

between radon concentration and alpha, beta, uranium, and radium concentrations, radon depletion was observed in two locations (2 and 7) after measures were taken to reduce uranium (^{238}U) concentration. In the first case involving Location 2, one of the four boreholes supplying the location was withdrawn due to its uranium concentration. After the borehole withdrawal, a significant reduction in radon concentration was also observed. Therefore, the borehole, which had a high uranium concentration, also had a high radon concentration. In the case of Location 7, the supplying borehole was replaced by another borehole. After this replacement, the radon-in-water activity concentrations measured in this location were also reduced. Several studies performed in the past (21–26) have not found any apparent correlation between radon with uranium and radium in groundwater samples.

Conclusions

This paper aimed to study a specific region in Northern Greece (Migdonia basin) regarding radon-in-water. This region's locations (villages) are supplied with water from boreholes located in the basin sediments and the metamorphic rocks and granites.

Radon-in-water average activity concentrations in the Migdonia basin collected from public water supplies ranged from background concentrations up to 185 Bq L^{-1}. The corresponding annual effective doses from inhalation of waterborne radon varied from 0.01 to 0.466 mSv y^{-1} using the UNSCEAR dose conversion factor and from 0.02 to 0.868 mSv y^{-1} using the ICRP dose conversion factor, while the annual effective dose due to radon ingestion ranged from 0.007 to 0.324 mSv y^{-1}. This wide range observed in tap water radon concentrations is also a sign of a significant wide range among the radon concentrations of boreholes supplying these locations, as each location is supplied from different boreholes. Therefore, the existence of a borehole inside a rich in uranium area does not necessarily mean an increased radon concentration in its water. Regarding average radon concentrations measured over 3–4 years, only two locations overcame the parametric value of 100 Bq L^{-1} set by EURATOM. Although only two of the 66 locations showed average radon-in-water concentration higher than the parametric value, four other locations overcame the parametric value at least once through the period of measurements.

The time variation (3–4 years) of radon in tap water activity concentration was studied for locations supplied by only one or more boreholes. The average radon concentration showed relatively low standard deviations (<24%) in places provided with water from only one borehole. Locations supplied with water from a constant number of boreholes also showed relatively low (<22%) standard deviations from their average radon concentrations. These standard deviations are probably due to radon time variation of supplying boreholes and water demand changes. In the case of more than one borehole supplying the location, it is worth mentioning that the ratio of the contribution of each participating borehole to the total water supply was constant over time. If this condition does not apply, the standard deviations are expected to be higher if the boreholes involved in the water supply show significant differences in their radon concentrations.

Regarding the radon-in-water concentrations measured in boreholes (source) and tap water (consumption), a significant decline in radon concentration was observed from the source to the consumption point. This observation is due to treatment processes that cause radon degassing and storage in tanks, leading to the radioactive decay of radon. Therefore, water sampling should be performed at the point of consumption. However, radon measurement at the borehole (source) could be useful to determine the potential for radon in drinking tap water. Also, as seen from the results, if a location is supplied from more than one borehole, the water supply on the day of sampling may not derive from the combination of boreholes that prevail during the year. Therefore, the radon concentration and dose calculation may be underestimated or overestimated.

The study of the correlation between radon with gross alpha and beta, uranium, and radium showed no obvious correlation. Although no apparent correlation was found between radon and uranium, a decline in their radon concentrations was observed in two locations after remedial actions were performed to decrease uranium concentrations. However, as the Member States of the European Union are obliged to measure the gross alpha radiation, if they find it relatively high, this would be a good starting point along with geology for also measuring the radon concentration. Still, it does not necessarily mean radon will be found in elevated concentrations.

Conflict of interest and funding

The authors declare no potential conflicts of interest.

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