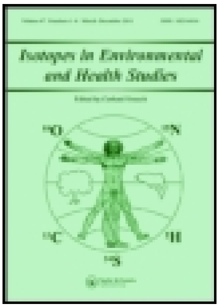

Isotopes in Environmental and Health Studies
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gieh20

Preliminary risk assessment of radon in groundwater: a case study from Eskisehir, Turkey

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Version of record first published: 04 Mar 2013.

Isotopes in Environmental and Health Studies, 2013
http://dx.doi.org/10.1080/10256016.2013.739562

Preliminary risk assessment of radon in groundwater: a case study from Eskisehir, Turkey

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(*Received 24 March 2012; final version received 14 September 2012*)

The aim of this study was to determine the radon concentrations in the water supplies of a residential area of central west Anatolia, Turkey. This research provides a preliminary risk assessment for inhabitants in the study area which can be applied for other regions. In 14 out of the 19 water supplies analysed, radon concentrations exceeded the maximum contaminant level (11.1 Bq l^{-1}). The total annual effective doses of 10 for the wet season and 14 for the dry season out of the 19 water supplies are greater than the values recommended by EPA [*Cancer Risk Coefficients for Environmental Exposure to Radionuclides*, Federal Guidance Report No. 13 (US Environmental Protection Agency, Washington, DC, 1999) <http://www.epa.gov/rpdweb00/docs/federal/402-r-99-001.pdf>] (0.1 mSv a^{-1}). The elevated radon concentrations in water resources are most probably linked with geological origin which contains significant amounts of radioactive minerals.

Keywords: cancer; drinking water, effective dose, radon; radon-222; risk assessment; Turkey

1. Introduction

Radon inhalation at high or even moderate levels is known to cause lung cancer [1–6], although it has been suggested by some researchers [7] that most of the lung cancer incidences are linked to smoking rather than environmental radon exposure. It has been reported that 12 % of lung cancer deaths (i.e. 19,000–22,000 of 160,000 deaths from lung cancer) in the USA each year are linked to exposure to ^{222}Rn and its short-lived decay products in indoor air [6,8–10]. Darby et al. [11] reported 7148 cases of lung cancer linked with radon-in-air values above 100 Bq m^{-3} in their review of residential radon and lung cancer across 10 European countries. Krewski et al. [12] also recorded 3662 cases of lung cancer linked to radon-in-air levels above 100 Bq m^{-3} in North America. Mowlavi et al. [13] presented the radiation risk for humans in the Ramsar region (Iran) because of high radon contents in drinking water. These studies highlight the direct relationship between residential radon and lung cancer risk.

Unlike radon in air, there is no conclusive evidence showing that cancer may result from chronic exposure to radon in water. Although the global dose from ingestion of radon in drinking water is relatively low, health risk from inhalation and ingestion might be almost equal in areas where the population depends on drinking water supplies that are naturally enriched in radon,

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and therefore the exposure to radon by ingestion as well as inhalation should be considered for a comprehensive risk assessment. According to the World Health Organization (WHO) [6], 95 % of exposure to environmental radon is from indoor air, with only about 1 % from drinking water. Much of the exposure from drinking water is through inhalation of radon gas from bathing, showering and cleaning. Even though the ingestion risk of radon from drinking water is lower than from inhalation [6], stomach cancer is of particular concern due to diffusion of waterborne radon [14,15]. Thus, the risk mostly depends on how much radon entered the blood system that carries it to other organs and tissues.

Radon is a noble gas and has three relatively short-lived natural isotopes: ^{222}Rn, ^{220}Rn and ^{219}Rn, with ^{222}Rn having the longest half-life (3.8 days). ^{222}Rn may occur in groundwater, and its concentration ranges from virtually zero in fully degassed groundwaters to thousands of Bq l^{-1}. The concentration of radon and radionuclides in general in drinking and domestic water might exceed the allowable limits because the aquifer lithologies are enriched in their parent elements U and/or Th [3,4,16–28] . The fate of ingested radon and its persistence in the human body were investigated in the 1960s by the National Research Council (NRC) [8]. According to the NRC, every year around 30,000 people in the USA die of lung cancer resulting from radon inhalation in dwellings located in areas with a high natural soil and groundwater radioactivity [5,29–31]. In Turkey, some studies have investigated radon concentrations in water and its relation with the environment [32–40].

This study focuses on the determination of radon concentrations in water supplies of some residential areas of central Turkey. These waters are derived from wells (between 10 and 300 m deep) or springs intersecting granite, dacite, rhyodacite, rhyolitic tuff, dacitic–andesitic tuff, and all these lithologies have high natural radioactivity. Our results show that radon concentrations in most of the analysed water supplies exceed maximum contaminant levels (MCLs), and our standard risk assessment analysis revealed that the local population is exposed to a potential health risk derived from the ingestion of these waters.

2. Geology of the study area

The study area has received a great deal of attention from earth scientists due to its complex tectonic history, variety of lithological units, and diverse mineralogical and geochemical features [41–47]. For the purposes of this study, geological information is summarised from the previous literature [48,49].

The study area includes igneous rocks (granite, subalkaline monzodiorite and gabbro), an ophiolitic suite, peridotite, volcanic (rhyodacite, dacite, rhyolitic tuff and dacite–andesite), metamorphic (marble, schist and talcschist) and sedimentary rocks (limestone, dolomitic limestone, clayey limestone, conglomerate, marl and gypsum) (Figure 1). The northeastern part mainly comprises Jurassic–Cretaceous igneous rocks, Paleogene and Neogene volcano-sedimentary units together with the Kaymaz Granite and Sivrihisar Monzodiorite. The Kaymaz Granite and the Topkaya Granite intrude the ophiolitic suite, and the hydrothermal alteration products of these granites and pegmatites outcrop near Karakaya village. The Kaymaz Granite, which has high silica contents (73–75 % SiO_2) [49], is composed of quartz, orthoclase, oligoclase, biotite, hornblende, zircon, apatite, tourmaline, sphene, pyrite and magnetite. Uranium and Thorium are relatively more enriched (average 16.6 and 49.9 ppm, respectively) in Kaymaz Granite than in the Sivrihisar Monzodiorite (3.49 and 15.8 ppm, respectively) [49]. The Sivrihisar Pluton is a monzodiorite derived from calc-alkaline magma, whereas the mineralogical and chemical composition of the Kaymaz Pluton is typical of granite. The Sivrihisar Monzodiorite contains the same mineral assemblage as the Kaymaz Granite, but it has relatively lower silica content (57–60 % SiO_2).

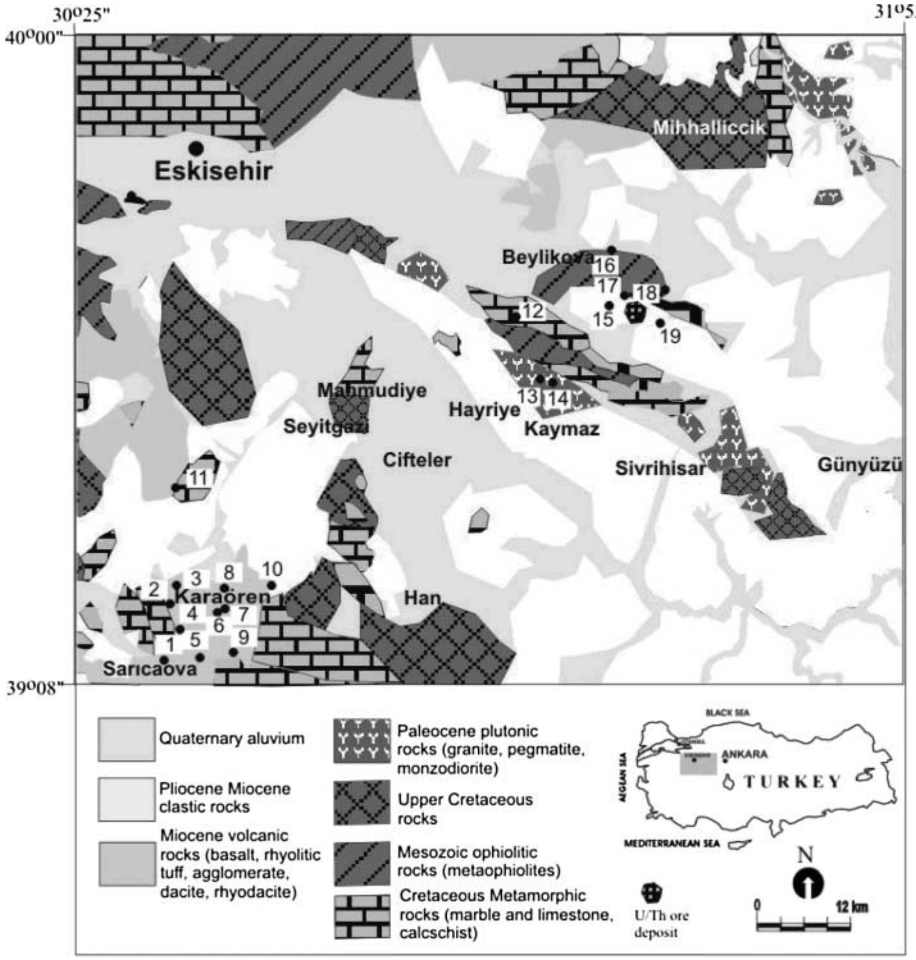

Figure 1. Geological map of the study area [50], showing the main lithological units and water sampling points for this study.

Phonolite, basalt, tuff, and agglomerate all resulting from young volcanism (Miocene–Pliocene), can be observed in the southern part of Kaymaz. Tuffs contain barite, sericite, calcite, opaque minerals and minerals highly enriched in Th, Mn and to a lesser extent Fe and Al. Early and Late Miocene volcanic rocks are also found in the southwest of the study area, around Phrygian valley, Yazilikaya, Han, Kirka and Karaören. Phonolites, basalt, tuff and agglomerate are products of young but extinct volcanoes (Miocene–Pliocene) in the southwestern parts of the study area.

3. Materials and methods

A total of 19 water samples (from six water bores, five fountains, four reservoir waters and a dam's reservoir water) were measured in the study area during the 2008 wet season (April and May) and again during the dry season in October 2011. Well water was allowed to run for at least 15 min before sampling. With the exception of Kizilcaoren well (No. 17), all other well waters are used for drinking and domestic purposes without any treatment. From time to time, the Karaoren

Table 1. Previously measured gross alpha and beta values [50], and measured radon values.

	No.	Location name	Lattitude	Longitude	Well depth (m)	Gross alpha (Bq l^{-1})	Gross beta (Bq l^{-1})	Measured radon (Bq l^{-1})[a] Wet season	Measured radon (Bq l^{-1})[a] Dry season	Lithology
Yazılıkaya–Phyrgian valley	1	S.ova-Çiftbaşıspring	39°09″10′	30°30″49′		0.06 (±0.005)	0.23 (±0.04)	5.7	**42.4**	Silicified Rhyolithic tuff
	2	Karaören shallow well	39°13″40′	30°34″35′	30			3.6	6.6	Rhyolithic tuff
	3	Karaören shallow well	39°13″47′	30°34″40′	10			1.6		Rhyolithic tuf
	4	Fethiye repository	39°13″52′	30°32″20′				8.1	**30**	Rhyolithic tuff
	5	S.ova–water repository	39°09″00′	30°30″32′				**78.9**	**96.5**	Silicified Rhyolithic tuff
	6	Hatap spring/Karaoren	39°13″44′	30°35″40′		**0.35** (±0.031)	0.32 (±0.04)	**146.0**	**155.7**	Rhyolithic tuff
	7	Hatap fountain/Karaoren	39°13″58′	30°34″57′				7.2	5.5	Rhyolithic tuff
	8	Karaören deep well	39°13″43′	30°34″32′	170	**1.42** (±0.058)	**1.02** (±0.05)	**230.0**	**251.0**	Rhyolithic tuff
	9	Gökbahçe repository	39°09″37′	30°35″14′				**29.7**	**14.3**	Rhyolithic tuf
	10	Yazılıkaya spring	39°12″03′	30°42″48′		**0.39** (±0.032)	0.65 (±0.04)	4.3	**22.2**	Rhyolithic tuff
	11	Kırka artesian well	39°16″37′	30°31″18′	187	0.03	0.4	**13.5**	**21.1**	Tuff and limestone
Beylikova–Kaymaz area	12	Nemani fountain	39°36″59′	31°10″52′				2.6	3.7	Schist + Calc-schist
	13	Karakaya well	39°30″10′	31°15″07′	178	**0.6** (±0.045)	0.72 (±0.05)	**50.0**	**30.5**	Alkaline granite
	14	Kaymaz repository	39°31″28′	31°11″27′				**57.3**	**68.0**	Alkaline granite
	15	Yayla fountain	39°36″51′	31°22″21′		**0.164** (±0.017)	0.34 (±0.04)	**79.4**	**22.7**	U+Th ore deposit
	16	Kızılcaören dam water	39°37″34′	31°21″55′		**0.104** (±0.009)	0.26 (±0.04)	7.79	8.0	Schist+Calc-schist+U
	17	Kızılcaören artesian well	39°36″03′	31°22″10′	300	**0.524** (±0.040)	**1.89** (±0.06)	**48.0**	**30.0**	U+Th ore deposit
	18	Adalet fountain	39°37″59′	31°21″56′		**0.35** (±0.013)	0.6 (±0.04)	**16.5**	**27.4**	U+Th ore deposit
	19	Karapınar fountain	39°36″06′	31°22″53′		0.053 (±0.010)	0.12 (±0.04)	**89.8**	**93.9**	U+Th ore deposit

[a]Bold numbers indicate radon values exceeding 11.1 Bq l^{-1} (equivalent to 300 pCi l^{-1} maximum contaminant limit, US EPA [1].

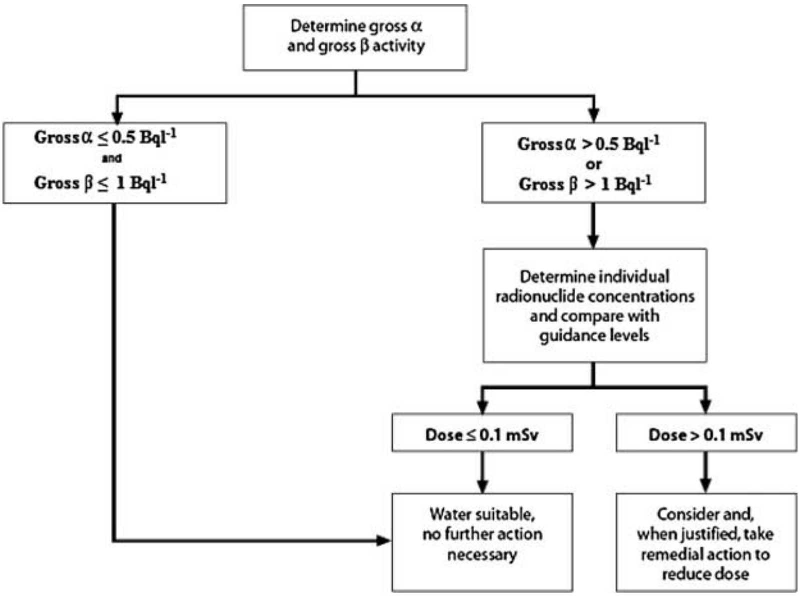

Figure 2. Screening scheme and guidance levels for radon in groundwater (from WHO [6]).

well (No 8) water is mixed with the Hatap spring water after aeration, and this process is likely to decrease the risk for the Karaoren well water. Tap water in the area is supplied through a pipeline from the main outlet/source (e.g. No 6 tap water derives from Hatap spring). Therefore, a part of radon content will be lost during transportation from the well to the end-user as tap water. So, in reality, the calculated dose rates depict a worstcase scenario.

The *in-situ* radon measurement in water was carried out in selected sites (Table 1) which were known to have high gross alpha values based on the previous survey [50]. The 'Hayriye and Mahmudiye' site [50] was not included in the present study because of contamination due to the existence of an old waste disposal area. Selection of the sampling sites specifically targeted the lithologies rich in U and Th identified by Orgun et al. [49] and which yielded gross alpha and gross beta values (Figure 2) above the threshold values (0.5 and 1 Bq l^{-1}, respectively; see [6]). Three of the 19 water supplies (sample Nos 8, 13, and 17) had gross alpha and gross beta values above the threshold values, and therefore this study was performed to determine their radon values. However, the sampling was extended to the other water supplies that gave gross alpha values above 0.1 Bq l^{-1} (MCL according to Turkish Standards Institution [51]).

Dissolved radon gas measurements were performed using a GEO-RTM 2128 instrument manufactured by SARAD

3.1. *Working principle of GEO-RTM 2128*

The measurement of the radon activity concentration of a water sample is based on the equilibrium state of radon between air and water, which takes place within a sealed system after a certain period of time. The working principle of the GEO-RTM, the alpha spectroscopy instrument used for this study, is based on information provided by the manufacturer. The GEO-RTM 2128 offers two ways of calculating the radon concentration: (i) fast mode, based only on ^{218}Po and (ii) slow mode, based on both ^{218}Po and ^{214}Po. The advantage of the 'fast' mode is the quick response to concentration changes, while the 'slow' mode gives double sensitivity as compared with the fast

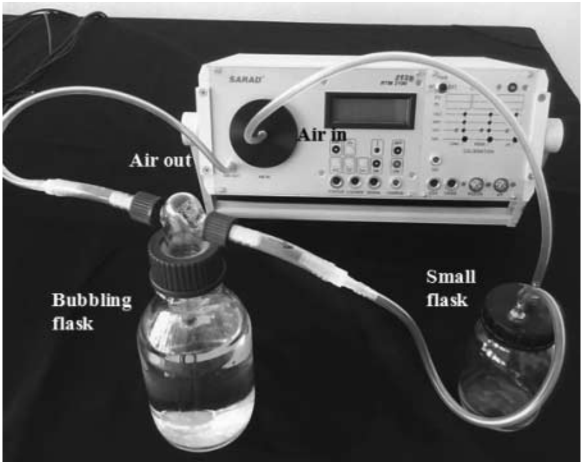

Figure 3. Equipment for radon measurement, and the closed-loop system used to transfer radon from water to air.

mode. The higher sensitivity reduces the statistical error of a measurement which depends on the number of counted decay events only.

The dissolved radon gas in the water samples is de-gassed using an air bubbling flask. The bubbling flask is connected to the GEO-RTM to provide a closed air loop. To protect the GEO-RTM radon monitor against the direct sucking of water from the bubbling flask, a small and tight glass flask is inserted between the air inlet of the instrument and the outlet of the bubbling flask (Figure 3).

The detection limit for this study was optimised by using a 500ml bubbling flask (total 380 ml air volume) in combination with the GEO-RTM 2128. The relative statistical error for radon activities ranged from over 10 % for low radon values ($\sim$2–10 Bq l^{-1}) to less than 5 % for values greater than 10 Bq l^{-1} The measurement time was chosen at a minimum of 1 h to obtain reliable results with a high confidence level (95 %) with respect to detection limits. The 'fast mode' was selected for the determination of the radon concentration, because a previous survey [50] had established that alpha activities in the area are generally high and that the 'fast mode' was therefore appropriate for this study.

To calculate radon in water concentrations from the displayed concentration on the GEO-RTM 2128 device, the software 'Radon in Water Calculator' was used [52]. The main principles of the software are based on the ratio between the activity concentrations and the temperature of the water sample. The dependency between activity concentration and water temperature can be expressed by the so-called Oswald coefficient, which indicates that the solubility of radon in water decreases for increasing water temperature. Thus, higher water temperatures result in higher radon diffusion in air.

After reading each average radon concentration in air, the software is used to calculate corrected radon concentration values in the water. Calibration by the manufacturer [52] provides the conversion of the count rates into radon activity. A correction factor was applied to take into consideration the fact that some radon is lost during sample preparation, and that some radon is adsorbed to the inner surfaces of the air loop. The correction factor was selected at 0.9, which indicates that 10 % of the radon is assumed to be lost due to these processes.

The risk calculation was performed based on the actual water usage by the population of each village or town. Information on water usage and on the number of inhabitants in each village was obtained from the local authority (mukhtar or district/council governor).

3.2. *Effective dose estimation*

Although radon in water may be ingested through drinking water, to some extent, it is exposed and transferred to air. Therefore to calculate the total dose estimation, both ingestion and inhalation should be considered. Estimation of dose from ingestion and inhalation of radon dissolved in drinking water and the resulting health risks were performed in this study. Consumption of drinking water significantly changes with the climatic conditions and physical activity. Apart from drinking water consumption, tap water is also used for household purposes in the bath and kitchen where radon can be released/transferred to the air in the house as the so-called 'indoor radon' [1,6].

The conversion dose factor for ^{222}Rn is 0.1×10^{-4} mSv Bq^{-1}. By using conversion factor (mSv Bq^{-1}) and annual consumption of water in litres per year, the activity level of radon in water can be converted from Bq l^{-1} into mSva^{-1}. On the other hand, the calculated annual effective dose rates are still less than the recommended level of UNSCEAR [53] (2.5 mSv a^{-1} 100 m^{-3} radon) and International Commission on Radiological Protection (ICRP) [54] (1 mSv a^{-1}) for indoor radon values (via inhalation).

The guidance MCL for radon in drinking-water was calculated using the following equation [6]:

$$\mathrm{GL} = \mathrm{IDC}/(h_{\mathrm{ing}} \times q), \tag{1}$$

where GL is the guidance level of radionuclide in drinking-water (Bq l^{-1}), IDC the individual dose criterion (0.1 mSv a^{-1} for this calculation), h_{ing} the dose coefficient for ingestion by adults (mSv Bq^{-1}) and q the annual ingested volume of drinking-water, assumed to be 730 l a^{-1} [6].

The h_{ing} coefficient is calculated as $h_{\mathrm{ing}} = 1.23 \times 10^{-5}$ mSv Bq^{-1} by WHO [6], and 3.5×10^{-6} mSv Bq^{-1} by EPA [1]. If the latter is used for the calculation, then a GL of 39 Bq l^{-1} for radon in drinking-water is obtained. Using h_{ing} recommended by WHO [6], a GL of 11.1 Bq l^{-1} is obtained.

To calculate the annual effective dose rate (D_{ing}) for ingested radon, the above equation is transformed into the following equation [55–57]:

$$D_{\mathrm{ing}} = A_{\mathrm{w}} \times V_{\mathrm{w}} \times F_{\mathrm{ing}}, \tag{2}$$

where A_{w} is the radionuclide activity concentration in water (Bq l^{-1}), V_{w} the volume of water ingested annually, assumed to be 730 l a^{-1} for an adult person and F_{ing} the effective dose equivalent conversion factor for ingestion (mSv Bq^{-1})

Assuming an annual water consumption for drinking purposes equal to 730 l a^{-1} the calculated annual effective dose rates are given in Tables 2 and 3 by using both conversion factors (F_{ing}).

The annual effective dose rate for indoor radon (D_{inh} mSv a^{-1}) or inhaled radon in the course of various water-using activities (household activities) is given by the equation [58]:

$$D_{\mathrm{inh}} = A_{\mathrm{w}} \times \mathrm{TF} \times F \times F_{\mathrm{inh}} \times T, \tag{3}$$

where A_{w} is the radionuclide activity concentration in water (Bq l^{-1}), TF is the 10^{-4} transfer factor, which is the increase in radon concentration in indoor air per unit radon concentration in water, F 0.4 is the indoor equilibrium factor between radon and its progeny, F_{inh} the effective dose coefficient 9.0×10^{-6} mSv h^{-1}Bq^{-1}m^{-3} [53,59] and T the exposure time to this concentration in hours (assumed to be equal to 7000 h year^{-1}, an occupancy factor of 80 %, ($0.8 \times 24\,\mathrm{h} \times 365 = 7000$)) [53].

The radon dose limits recommended by WHO [6] and EPA [1] for public drinking water and release from water into air are 0.04 and 0.10 mSv a^{-1}, respectively. However, the US NRC [8] and the ICRP [54] recommend a dose limit of 1 mSv a^{-1} for the general public.

Table 2. Calculated effective doses for the Yazilikaya–Phrygian valley.

		Drinking water dose values (limit value: 0.04 mSv a^{-1}) $F_{ing} = 1.23 \times 10^{-5}$ mSv Bq^{-1} [6]		Drinking water dose values (limit value: 0.04 mSv a^{-1}) $F_{ing} = 3.5 \times 10^{-6}$ mSv Bq^{-1} [1]		Dose values for indoor radon transferred from water (Limit value: 0.1 mSv a^{-1} recommended by EPA [1]; limit value: 1 mSv a^{-1} recommended by ICRP [54]) $F_{inh} = 9.0 \times 10^{-6}$ mSv h^{-1} $Bq^{-1}m^{-3}$ [1]	
No.	Yazilikaya–Phrygian valley	Wet season	Dry season	Wet season	Dry season	Wet season	Dry season
1	S.ova-Çiftbasi spring	**0.051**	**0.381**	0.015	**0.108**	0.014	**0.107**
2	Karaören S. well	0.032	**0.059**	0.009	**0.017**	0.009	0.017
3	Karaören S. well	0.014		0.004		0.004	
4	Fethiye depository	**0.073**	**0.269**	0.021	**0.077**	0.020	**0.076**
5	S.ova depository	**0.708**	**0.866**	**0.202**	**0.3247**	**0.199**	**0.243**
6	Hatap spring	**1.311**	**1.398**	**0.373**	**0.398**	**0.368**	**0.392**
7	Hatap fountain	**0.065**	**0.049**	0.018	0.014	0.018	0.014
8	Karaören D. well	**2.065**	**2.254**	**0.588**	**0.641**	**0.580**	**0.633**
9	Gökbahçe depository	**0.267**	**0.128**	**0.076**	0.037	**0.075**	0.036
10	Yazilikaya spring	0.039	**0.199**	0.011	**0.057**	0.011	**0.056**
11	Kirka well	**0.121**	**0.189**	0.034	**0.054**	0.034	**0.053**

Table 3. Calculated effective doses for the Beylikova–Kaymaz area.

		Drinking water dose values (limit value: 0.04 mSv a^{-1}) $F_{ing} = 1.23 \times 10^{-5}$ mSv Bq^{-1} [6]		Drinking water dose values (limit value: 0.04 mSv a^{-1}) $F_{ing} = 3.5 \times 10^{-6}$ mSv Bq^{-1} [1]		Dose values for indoor radon transferred from water (limit value: 0.1 mSv a^{-1} recommended by EPA [1]; limit value: 1 mSv a^{-1} recommended by ICRP [54]) $F_{inh} = 9.0 \times 10^{-6}$ mSv $h^{-1}Bq^{-1}m^{-3}$[1]	
No.	Beylikova–Kaymaz area	Wet season	Dry season	Wet season	Dry season	Wet season	Dry season
12	Nemani fountain	0.023	0.033	0.007	0.009	0.007	0.009
13	Karakaya well	**0.449**	**0.274**	**0.128**	**0.078**	**0.126**	**0.077**
14	Kaymaz depository	**0.514**	**0.611**	**0.146**	**0.174**	**0.144**	**0.171**
15	Yayla fountain	**0.713**	**0.204**	**0.203**	**0.058**	**0.200**	**0.057**
16	Kizilcaören dam[a]	0.039	**0.072**	0.011	0.020	0.020	0.020
17	Kizilcaören well[a]	**0.431**	**0.269**	**0.123**	**0.077**	**0.121**	**0.076**
18	Adalet fountain	**0.148**	**0.246**	**0.042**	**0.070**	**0.042**	**0.069**
19	Karapinar fountain	**0.806**	**0.843**	**0.229**	**0.240**	**0.226**	**0.237**

[a]These water supplies are not being used for drinking purpose.

3.3. *Cancer risk assessments for ingested and inhaled radon*

3.3.1. *Ingested*

Ingested radon diffuses into the tissues of the stomach and small intestine. From there it enters the bloodstream and is carried throughout the body. The majority of ingested radon is thought to be exhaled when the blood flow carries it to the lungs. The NAS [9,10] modelled the fate of ingested radon and its daughters in the body and estimated the associated cancer risk. It was determined that most of the radiation dose is delivered to the stomach as radon diffuses through the stomach wall. Most of the cancer risk from ingested radon is thus a stomach cancer risk, although there is a small additional risk of cancer in other tissues of the body [60].

3.3.2. *Inhaled*

The basic equation used to calculate inhalation uptake of radon gas is based on both the uncertainty and the variability of the unit dose factor. The risk factor per unit dose, however, is based on a single value. The unit dose factor for gas released from water depends on three factors according to the equation [61]:

$$\mathrm{UD} = (\mathrm{TF}) \times (\mathrm{BR}) \times (\mathrm{OF}) \times (365\ \mathrm{da}^{-1}), \qquad (4)$$

where UD is the unit dose (pCi inhaled per year per pCi l^{-1} of radon in water), TF the transfer factor, which is the increase in radon concentration in indoor air per unit radon concentration in water (pCi l^{-1} [air] per pCi l^{-1} [water]). The amount of radon from water degassing into the air is different throughout a dwelling but is higher in the areas such as bathrooms and kitchens where active water is in use. It also depends on the radon concentration in the water and specific household activities. The EPA recommends the value of 1.0×10^{-4} as the transfer coefficient [61]. BR the breathing rate ($\mathrm{l\ d}^{-1}$). On average, people take about 20,000 litres of air per day [61]. OF is the occupancy factor (fraction of time person spends indoors). This is typically 0.8 [61].

The mean population risk (PR) can be calculated by EPA [61] as follows:

$$\mathrm{PR} = (\mathrm{UD}) \times (\mathrm{RF}) \times (C_{\mathrm{mean}}) \times (N), \qquad (5)$$

where PR is the population risk of fatal cancer (cancers per year) posed by ingestion of radon gas in water, UD the unit dose (pCi inhaled per year per pCi l^{-1} of radon in water), RF is the risk factor, lifetime risk of cancer per person per pCi inhaled per year. EPA [61] has taken into consideration the coefficient of inhalation risk factor, RF of 1.1×10^{-12} cancer death $\mathrm{person}^{-1}\mathrm{pCi\ l}^{-1}$ of radon in water. N is the number of people in the population. C_{mean} is the population mean concentration of radon in water, pCi l^{-1}, calculated as

$$C_{\mathrm{mean}} = \frac{\Sigma A_i P_i}{\Sigma P_i}, \qquad (6)$$

where A_i is the activity concentration of radon (pCi l^{-1}) and P_i the population served by each water supply.

4. Results and discussion

Radon measurements of water supplies in this study indicate that high activity concentrations are consistent with aquifer lithology, because high radon values in water samples are associated with the distribution of U and Th-rich granites and rhyolitic tuffs. Such a relationship was also recorded in the literature [38,62]. Radon concentrations exceeded the MCL (11.1 Bq l^{-1}) in 11 and 14 of the 19 water supplies tested during the wet season and the dry season, respectively (Figure 4). The spatial distribution of radon levels shows two hot-spots: (I) the Phrygian valley (covered by volcanic rocks such as tuff, rhyolitic tuff, andesite and dacite) and (II) the Beylikova–Kaymaz area (covered by crystalline bedrock such as granite, phonolite, pegmatite rocks and thorium–uranium ore). Figure 5 and Table 1 show a strong correlation between high radon concentrations and lithology. The spatial pattern of geologic units with elevated radon-222 concentrations in groundwater is consistent with gross alpha pattern due to high U content [49]. Thus, elevated radon levels in groundwater are indicative of rocks rich in U- and Th-bearing minerals. The highest levels are usually found in uranium-bearing rhyolitic tuffs (up to 251 Bq l^{-1}, Table 1). However, the radon concentration was not sufficiently high to confirm a high-radon lithology for sample 17 (Kizilcaoren deep well). Waters that may have originally inherited high radon levels

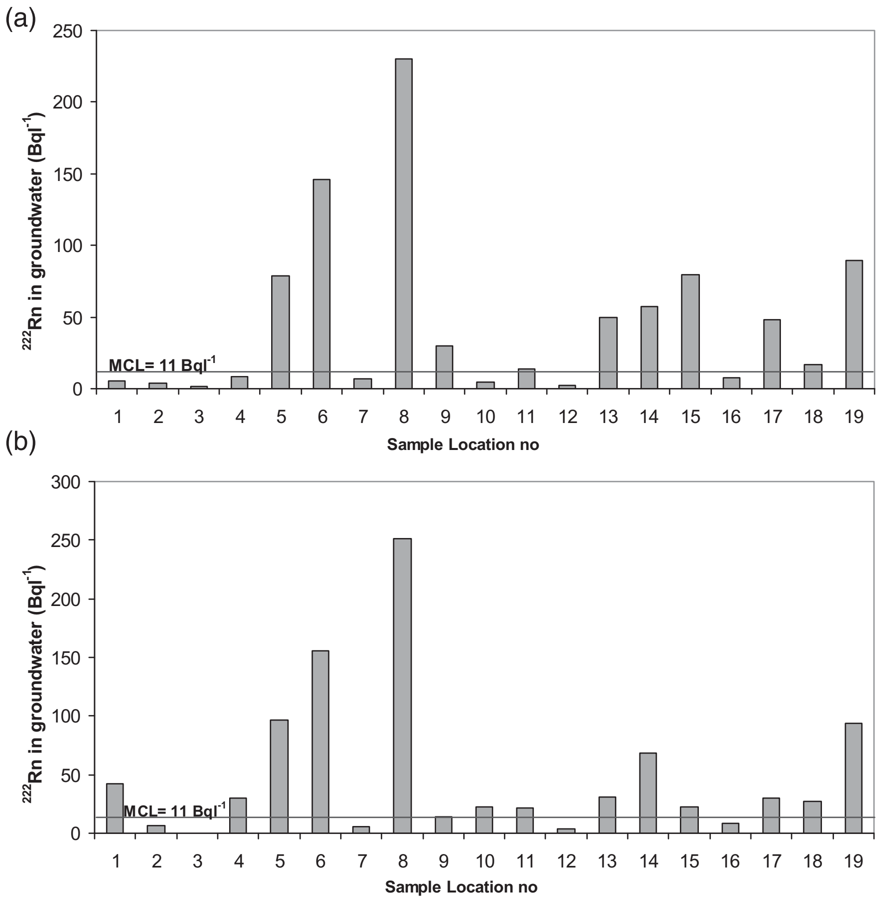

Figure 4. Radon concentrations (Bq l^{-1}) in water samples for the wet (a) and dry (b) season.

from U-rich lithology may also have lost part of this radon due to degassing and water–rock interaction. In line with this observation, we note that the low radon-222 values measured in this study belong to either springs or shallow wells and repositories that are open to the atmosphere (Table 1). Geologic units with the lowest radon-222 concentrations include limestone, schist and calc-schist. Generally, the radon content in water-wells shows a slight increase with depth (Figure 6).

An increase in the activity concentration of radionuclides in water would result in high gross alpha (alpha emitters such as ^{210}Po, ^{226}Ra, ^{239}Pu, ^{222}Rn, ^{232}Th and ^{238}U) or gross beta (beta emitters such as ^{137}Cs, ^{60}Co, ^{90}Sr, ^{99}Tc, ^{204}Ta and ^{40}K) activity concentrations, and this would provoke to screen specific radionuclides. The presence of radon in groundwater is mainly due to the decay of radium-226 naturally found in local rocks and soils. The gross alpha (including radium-226 but excluding radon-222 and uranium-238) and gross beta values are compared with radon concentrations in Figure 7. In spite of the fact that there is a quite positive relationship between gross alpha and radon (Figure 7(a)) (because both are the members of ^{238}U decay series), this relation becomes weaker once dissolved radon concentration increases in water. There is no relationship between gross beta and radon due to the different decay series.

On the basis of on-site measurements, concentrations of dissolved radon in water analysed for this study range from 1.6 to 230 Bq l^{-1} for the wet season and from 3.7 to 251 Bq l^{-1} for the

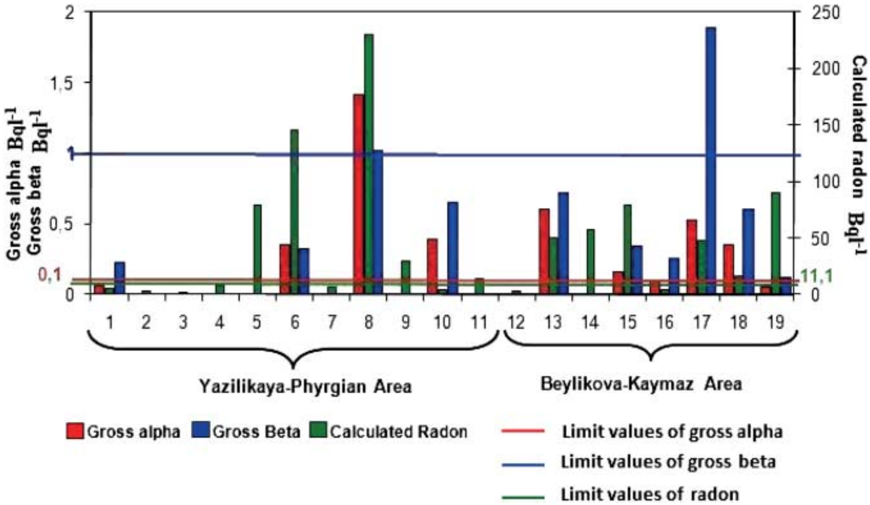

Figure 5. Summary of gross alpha, gross beta (from [50]) and radon values. Limit values for gross alpha and gross beta are 0.1 Bq l^{-1} (red horizontal line) and 1 Bq l^{-1} (blue horizontal line), respectively, and for radon in drinking water is 11.1 Bq l^{-1} equivalent to 300 pCi l^{-1} (green horizontal line), as recommended by US EPA [66].

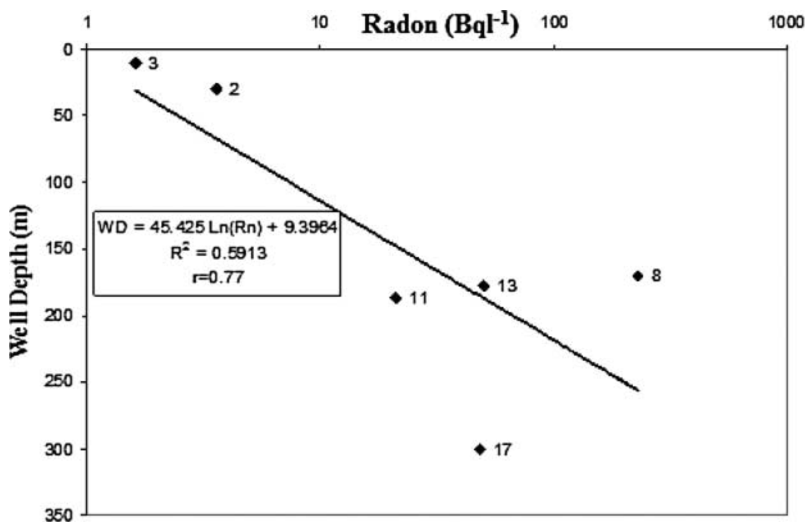

Figure 6. Correlation between dissolved radon content and well depth.

dry season (Table 1). The investigation area was divided into two sections: Yazilikaya–Phrygian valley and Beylikova–Kaymaz. As mentioned in Section 2, the first area is mostly covered by rhyolotic tuff and the second is overlaid by alkali granite and uranium–thorium ores as well as hydrothermal deposits (Table 1). Radon concentrations in 11 out of 19 measured water supplies were higher than the 11.1 Bq l^{-1} recommended by EPA [1] as the MCL.

The calculated annual effective dose values based on radon concentrations in water for both study areas are given in Tables 2 and 3. The average doses from radon in drinking water have been calculated as 0.11 and 0.13 mSv a^{-1} via inhalation and 0.41 and 0.46 mSv a^{-1} via ingestion (for $F_{\text{ing}} = 1.23 \times 10^{-5}$ mSv Bq^{-1}) for the wet and dry season respectively, as compared with the background inhalation dose of 1.1 mSv a^{-1} from air [59]. Considering the value of 0.1 mSv as the

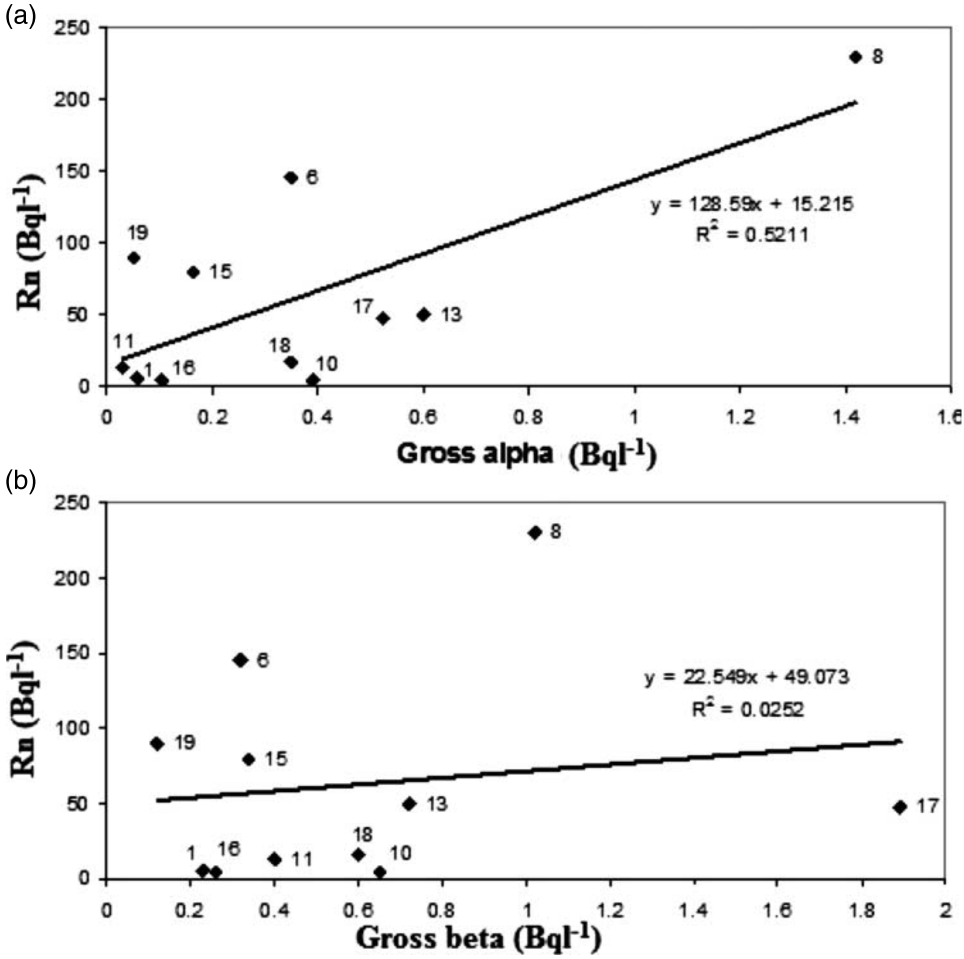

Figure 7. Radon vs. gross alpha (a) and gross beta (b) activities.

recommended limit of the effective dose from annual consumption of drinking water, values found in this study are significantly higher (2.065 mSv a^{-1} for the wet season and 2.254 mSv a^{-1} for the dry season) than the recommended value. The calculated annual effective dose values of 10 (wet season) and 14 (dry season) out of 19 measured water supplies exceed the recommended limits given by the WHO [6] and EPA [1]. Only two of the water supplies exceed the recommended limit value (1 mSv a^{-1}) set by NRC [8] and ICRP [54]. Consequently, it can be concluded that some of the waters (indicated in the bold style in Tables 2 and 3) measured in the studied area are not recommended as drinking water supplies for human consumption due to a remarkably higher indoor radon gas content.

The results of PR of fatal cancer related to high radon concentrations in water supplies are listed in Table 4. The population values were taken from the 2008 census as well as from data provided by local authorities. The total regional population was considered to be 16075 for Yazilikaya–Phrygian and 3992 for Beylikova–Kaymaz, respectively. As evident from Table 4, the values of population-weighted cancer risk are consistent with population increase and radon activity concentration. Therefore, cancer risk due to radon inhalation for Kaymaz (No 14) and Karaören villages (Nos 6 and 8) are 9 and 3 per ten thousand people, respectively, while this risk increased

Table 4. Population-weighted cancer risk.

	Radon					C_{mean}		Population risk (PR)	
	Wet season		Dry season						
Sample no.	Bq l^{-1}	(pCi l^{-1})	Bq l^{-1}	(pCi l^{-1})	Population (N)	Wet season	Dry season	Wet season	Dry season
Yazilikaya–Phrygian									
1	5.7	154.1	42.4	1145.9	150	1.44	10.69	1.3E-07	1.0E-06
2	3.6	97.3	6.6	178.4	1149	6.95	12.75	5.1E-06	9.4E-06
3	1.6	43.2			1149	3.09		2.2E-06	
4	8.1	218.9	30	810.8	83	1.13	4.19	6.0E-08	2.2E-07
5	78.9	2132.4	96.5	2608.1	150	19.90	24.34	1.9E-06	2.3E-06
6	146.0	3945.9	155.7	4208.1	1149	282.05	300.78	2.0E-04	2.2E-04
7	7.2	194.6	5.5	148.6	1149	13.91	10.62	1.0E-05	7.8E-06
8	230.0	6216.2	251.0	6783.8	1149	444.32	484.89	3.0E-04	3.5E-04
9	29.7	802.7	14.3	386.5	571	28.51	13.73	1.0E-05	5.0E-06
10	4.3	1791.9	22.2	600.0	82	9.14	3.06	4.8E-07	1.6E-07
11	13.5	364.9	21.1	570.3	9294	210.95	329.73	1.0E-03	1.9E-03
Beylikova–Kaymaz									
12	2.6	610.8	3.7	100.0	360	55.08	9.02	1.2E-05	2.0E-06
13	50.0	1351.4	30.5	824.3	141	47.73	29.11	4.3E-06	2.6E-06
14	57.3	1548.6	68.0	1837.8	1867	724.28	859.51	8.6E-04	1.0E-03
15	79.4	2145.9	22.7	613.5	360	193.52	55.33	4.4E-05	1.2E-05
16	4.3	10.5	8.0	216.2	360	18.99	19.50	4.3E-06	4.5E-06
17	48.0	1297.3	30.0	810.8	360	116.99	73.12	2.7E-05	1.6E-05
18	16.5	445.9	27.4	740.5	360	40.22	66.78	9.3E-06	1.5E-05
19	89.8	2427.0	93.9	2537.8	184	111.87	116.97	1.3E-05	1.3E-05

Table 5. Averaged annual total cancer mortality risk.

				Averaged annual total cancer mortality risk coefficients					
				5.0×10^{-9} [8]		1.3×10^{-8} [8,13]		1.9×10^{-9} [1]	
		Radon (Bq l^{-1})		Averaged annual total cancer mortality risk values					
Sample no.	Population	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry
Yazilikaya–Phrygian Valley									
1	150	5.7	42.4	6.3E-04	4.6E-03	1.6E-03	1.2E-02	2.0E-04	1.7E-03
2	1149	3.6	6.6	2.3E-02	4.2E-02	6.0E-02	1.1E-01	8.0E-03	1.6E-02
3	1149	1.6		1.0E-02		2.0E-02		3.0E-03	
4	83	8.1	30	2.7E-04	1.0E-03	7.0E-04	2.6E-03	1.0E-04	3.8E-04
5	150	78.9	96.5	8.7E-03	1.0E-02	2.0E-02	2.7E-02	3.0E-03	4.0E-03
6	1149	146.0	155.7	9.4E-01	1.0	2.46	2.62	0.3	0.3
7	1149	7.2	5.5	4.6E-02	3.5E-02	0.12	9.2E-02	1.7E-02	1.3E-02
8	1149	230.0	251.0	1.49	1.62	3.8	4.2	0.5	0.6
9	571	29.7	14.3	4.7E-02	2.2E-02	0.1	5.9E-02	1.8E-02	8.6E-03
10	82	4.3	22.2	2.1E-03	7.3E-04	5.0E-03	1.9E-03	8.3E-04	2.7E-04
11	9294	13.5	21.1	5.72	8.94	14.8	23.2	2.1	3.4
Beylikova–Kaymaz									
12	360	2.6	3.7	5.7E-02	9.4E-03	0.1	2.4E-02	2.0E-02	3.6E-03
13	141	50.0	30.5	1.9E-02	1.1E-02	5.0E-02	3.1E-02	7.4E-03	4.5E-03
14	1867	57.3	68.0	3.94	4.68	10.2	12.1	1.5	1.7
15	360	79.4	22.7	0.2	5.8E-02	0.5	0.1	7.7E-02	2.2E-02
16	360	4.3	8.0	1.9E-02	2.0E-02	5.0E-02	5.3E-02	7.5E-03	7.7E-03
17	360	48.0	30.0	0.12	7.6E-02	0.3	0.1	4.6E-02	2.9E-02
18	360	16.5	27.4	4.2E-02	7.0E-02	0.1	0.1	1.6E-02	2.6E-02
19	184	89.8	93.9	6.0E-02	6.2E-02	0.1	0.1	2.2E-02	2.3E-02

by almost one order of magnitude in Kırka village (No. 11) due to the larger population. Risk increases in the dry season more than in the wet season.

In addition to the calculation of population-weighted cancer risk values, the averaged annual total cancer mortality risk values corresponding to each water supply were calculated using several risk coefficients for radon inhalation (Table 5). The risk of lung cancer posed by lifetime exposure to ^{222}Rn in water at 1 Bq m^{-3} was calculated to be 1.3×10^{-8} [14]. The similar risk for 1 Bq m^{-3} was stated as 5.0×10^{-9} by NRC [8]; 1.3×10^{-8} in [13,14] and 1.9×10^{-9} by EPA [1]. Results in Table 5 show that cancer incidence rates increase with the population, as expected.

The cancer risk resulting from radon exposure is related to age, gender, specific usage rates of tap water and smoking habits [6,63,64]. However, the calculation based on different risk coefficients in Table 5 can only give some rough estimation regarding cancer risk. To obtain more reliable results, it is necessary to make further analyses and detailed investigations. Considering one of the most recent reports (New Jersey Drinking Water Quality Institute Report [65]), lifetime (70 year) cancer risk resulting from radon in drinking water (ingestion only) was estimated to be 2.0E-05 for 11.1 Bq l^{-1} (limit value), 7.0E-05 for 37 Bq l^{-1} and 3.0E-04 for 148 Bq l^{-1}. These rates will increase by 10 times with the addition of radon inhalation. The calculated mortality risk values from radon in drinking water (ingestion only) in this study (Table 5) are reasonably compatible with the aforementioned report (particularly for risk coefficient 1.3×10^{-8} and calculated PR).

5. Conclusions

Radon concentration in the 19 water supplies considered for this study ranged between 1.6 and 230 Bq l^{-1} in the wet season and 3.7 and 251 Bq l^{-1} in the dry season. The higher radon concentrations are associated with higher gross alpha values. High concentrations of radon were found in water samples near uranium–thorium bearing minerals associated with granite rocks (Kaymaz–Beylikova area) and rhyolitic tuffs (Yazilikaya–Phrygian valley). There is a slight inverse correlation between pH and radon content, which may be due to the effect of acidic fluids on the enrichment of U in the system.

The results of the survey indicated that 11 (for the wet season) and 14 (for the dry season) out of the 19 water supplies exceeded the MCL of 11.1 Bq l^{-1}, recommended by EPA [1]. Controls should be considered if the radon concentration in drinking water for public water supplies exceeds 100 Bq l^{-1} [6]. Therefore, based on the restrictions recommended by the WHO [6] one of the Karaoren deep well waters (sample 8) and the Hatap spring water (sample 6) should be consumed in a controlled manner and/or aerated before drinking.

The annual effective doses of 14 out of 19 water supplies exceed the values (0.04 mSv a^{-1}) recommended by EPA [1] for drinking water while 13 out of 19 are above the limit recommended by ICRP [54] (1 mSv a^{-1}) for indoor radon values (via inhalation). However, the annual effective dose rates calculated from radon concentrations do not exceed the recommended limit by UNSCEAR [53] (2.5 mSv a^{-1}).

As expected, lung cancer risk increases proportionally with population. Accordingly, cancer risk calculations for Kaymaz and Karaören villages, each with population over 1000, indicate a risk of 9 and 3 per ten thousand people respectively. Moreover, this risk increases to 1‰ at Kirka, which has a population close to 10,000. Risk elevates more from the wet season to the dry season.

The results show a significant relationship between radon concentrations of water resources and their geological origin because the elevated radon concentrations occur when the water-bearing formations contain a significant amount of radioactive minerals or are fed from deep groundwater sources containing radionuclides as is the case in this study (the deep wells Nos 8, 13, and 17).

Finally, there are some risk areas and a positive correlation between high radon concentrations in wells drilled through granites, rhyolitic tuff and U/Th ore deposits. A weak positive correlation was also noted between elevated radon concentration and well depth.

The present study has revealed the need for further investigations in the study area for a better understanding and evaluation of the effects of exposure to high radon concentrations on human health. Although current data suggest that it is conceivable that ingested radon may increase cancer risk, it is not possible at this stage to quantify this risk based on currently available epidemiological observations. In reality, the local people currently do not continuously use some of the fountains (Nos 10, 15, 18 and 19) and the artesian well No 17 for drinking water which means no exposure to significant health risk, despite the high radon content measured in these waters.

Acknowledgements

The authors are grateful to Drs Heiko Woith and Vivek Walia, whose insightful comments and suggestions considerably improved this article

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