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Radon measurements in well and spring water in Lebanon

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Abstract

The variation of dissolved radon (^{222}Rn) levels in water supplies remains of interest because of the radiation-induced public health hazards. A large part of the Lebanese population relies on springs and wells for their drinking water. ^{222}Rn measurements in spring and well water sources were conducted using the E-PERM method at sites ranging from sea level to 1200 m above sea level and across several geologic formations within Lebanon. The dissolved radon concentrations ranged from a low of 0.91 Bq L^{-1} in a coastal well source to a high of 49.6 Bq L^{-1} for a spring source in a mountainous region. Of the 20 sites sampled, only five had radon levels above 11 Bq L^{-1} and these mostly occurred in areas adjacent to well-known geological fault zones. A preliminary national average radon level was determined to be about 11.4 Bq L^{-1}. In general, as all determined concentrations were well below the 100 and 146 Bq L^{-1} revised reference levels proposed in the European Union and the United States, respectively, it is concluded that there is no reason to believe these water sources pose any radon-related hazard. On the other hand, at locations where water is collected directly from the springhead, it is advisable to have a settling/piping system installed allowing for further radon decay and radon loss into the air to alleviate any possible radon problem.

Keywords: Radon; Dissolved; Water; Lebanon; E-PERM

1. Introduction

While radon has existed since the formation of the earth elevated occupational radon exposure in uranium mines has not been well studied until the 1950s. On the other hand, not much attention was given to public exposure until the 1980s when dangerous radon (^{222}Rn) levels were reported inside homes and schools in the United States (USEPA, 1993). Human beings are exposed to radon through inhalation or ingestion. Dissolved radon is released to air upon usage of water, which adds to the dose received from inhalation of airborne radon emanating from the ground itself. In 1991, the United States Environmental Protection Agency (EPA) proposed a National Primary Drinking Water Regulation (NPDWR) for ^{222}Rn with a maximum contaminant level (MCL) of 11 Bq L^{-1} (300 pCi L^{-1}) (USEPA, 1991). The National Academy of Sciences (NAS) revised the MLC and established an alternative maximum contamination level (AMCL) (NAS, 1999). According to NAS (1999), the AMCL may be set higher than the MCL such that “the contribution of radon from drinking water to radon levels in indoor air is equivalent to the national average concentration in outdoor air”. For the United States, this leads to an AMCL of 4000 pCi L^{-1} (146 Bq L^{-1}). On the other hand, the European Union (EU) issued a non-binding recommendation in 2001 setting 100 Bq L^{-1} as a reference level; a concentration above this level warrants consideration of possible remedial actions. The EU recommendation also sets 1000 Bq L^{-1} as the upper bound above which remedial action is definitely required (EU, 2001).

Because of its potential public health hazard, surveys of radon in water sources have proliferated exponentially over the two past decades (Yu et al., 1994; Vásárhelyi et al., 1997; Al-Bataina et al., 1997; Otwoma and Mustapha, 1998; Tayyeb et al., 1998; Horvath et al., 2000; Al-Kazwini and Hasan, 2003; Segovia et al., 2003; Moussa and El Arabi, 2003; Erees et al., 2006; Schubert et al., 2006). Much of the attention was given

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to the dissolved radon concentration at the water source and how geologic formations (including faults) qualitatively impact the variability of dissolved radon (e.g. Al-Tamimi and Abumurad, 2001; Choubey et al., 2003; Baykara and Doğru, 2006). The composition of geological formations is known to critically affect radon concentration near water sources, although when water is transported from the source to the consumers' fetching point as free water surface flow, natural aeration is likely to reduce radon concentrations. Natural radon aeration is dependent on numerous processes, including atmospheric pressure and temperature, both known to vary with site elevation above mean sea level. In addition, radon natural decay contributes to the reduction of radon concentrations. As a result, piped water from springs or wells is likely to contain significantly reduced radon levels due to radon decay and radon loss into the air during transport to consumers.

Dissolved radon concentration data were collected at the water fetching point across a wide range of geological formations and site elevations in Lebanon. The geography and geology of Lebanon are ideal for this study because: (1) public water sources, used for service and drinking purposes, are primarily from springs and wells at high elevations; (2) the variability in geological formations is sufficiently large to induce an appreciable spread in dissolved radon concentrations.

Since most of the geological background is cretaceous, the carefully selected sites tested should reflect similar radon results. However, by including some sites in the few but highly utilized water sources in geological fault zones, we expected to see a higher radon reading. In nearly all sampled water sources, the fetching point to spring mouth or well bottom was selected so as to be approximately equal. However, of the 20 sites, five had no or minimum piping, and the corresponding samples were taken at the natural source itself. These five sites represented virgin radon emanating from the ground and could be indicative of the mother rock radon precursor content (radium). In fact, these sites included some of the highest readings noted in this study.

In Lebanon, while some attention has been given to general water safety issues since the early 1990s, little has been done in relation to radiological aspects and a national average has not yet been established. In this study, the dissolved radon levels in a number of potable water sources covering the main administrative regions in Lebanon were determined. As far as we know this study is the first to determine the dissolved radon concentrations in ground water in Lebanon.

2. Background information

Lebanon is a country with an area of 10,452 km^2 and an estimated population of 3.5 million people of which 88% live in urban areas. The country lies around the conjuncture of three tectonic plates: the Arabian, Anatolian and African plates. A major geological feature in Lebanon is the northern extension of the great African/Red Sea/Dead Sea rift valley. This is manifested in a major fault line with many branched sub-faults. Currently, and in the recent past, the area has not been very active geologically, although mild earthquakes are not uncommon.

From north to south, the coastline in Lebanon extends over 217 km, while from east to west it measures 80 km at its widest point. Lebanon's landforms fall into four parallel belts that run from northeast to southwest: a narrow coastal plain along the Mediterranean shore; the massive Mount Lebanon rising steeply from the coastal plain to 3083 m to dominate the entire country before dropping eastward; a fertile between-mountains basin, with an approximately uniform altitude of 800–1000 m, called the Beqaa Valley; and the ridges of the Anti-Lebanon Mountains dominated in the south by the volcanic Mount Hermon at 2814 m.

In Lebanon, the potable water supplies are mostly spring-fed, although some river dependence also exists (MOE, 2001). As such the nature of the bedrock may be an important influence on the dissolved radon level in these waters. The geologic formations are summarized in Table 1 . While the winter season in Lebanon is wet and precipitation reaches 1500 mm in some places falling mostly from November to April, the summer is totally dry. It may thus be surmised in general that in the summer, when the demand for water is greatest, discharged water would tend to have had a longer residence time in the ground (notably towards mid to late summer) and this may contribute to higher levels of dissolved radon in ground water.

3. Materials and methods

3.1. Selection of the measurement points and duration

Dissolved radon levels in water largely depend on the extent to which the water is aerated. Hence, to measure the actual public exposure levels, water samples from spring and well sources were collected at consumers' fetching points.

The dissolved radon concentrations in water from 18 different points across Lebanon were measured in the spring of 2004. This was done while attempting to distribute the sampling locations amongst the six administrative districts in Lebanon corresponding to: Beirut, Mount Lebanon, North Lebanon, South Lebanon, Nabatiyeh and the Beqaa (Fig. 1). Additionally, for each district, it was attempted to have several different points selected based on some predetermined criteria such as: geological background, convenience of sampling, altitude, and popular water usage intensity. Table 1 provides information on the study sites.

3.2. Sampling method

The water samples were collected using 68 or 136 mL collection bottles. After ensuring the water has run for several minutes from the spring or well source, a bucket was filled to overflow and then raised so that the tap (or source) is below the surface. The collection bottle is then submerged and filled from the bottom of the bucket and capped underwater (Kotrappa, 1999). Two samples were taken at each location and the time and date of sampling were noted. The samples were transported to the laboratory while keeping track of the transportation time.

Table 1
Cities and villages chosen from each administrative Muhafaza

District	No.	Site	Elevation (m)	Source/description/geologic formation[a]
Beirut	B1	Ashrafiye	30	Well/high rise area/GF5
Mount Lebanon	ML1	Deir El Kamar	850	Spring/midtown/GF5
	ML2	Falougha	1200	Spring/used by locals and nearby villages filling containers for drinking and cooking—the springs on western slopes on mount Kneissi (2200 m)/GF6
	ML3	Barouk	900	Spring/western slopes of Mount Barouk—1942 m/GF6
Beqaa	BK1	Zahle	900	Spring/on the eastern drier side of Mount Sannine (2620 m). Famed Bardawni River runs through here and drains into Litany river/GF2
	BK2	Ain Bourday—Baalbeck	1000	Spring/eastern Lebanon mountains/GF4
	BK3	Ablah	850	Well/Litany river drainage basin/GF2
	BK4	Taalabaya	870	Well/Litany river drainage basin/GF2
	BK5	Ras el Ein	1100	Spring/eastern Lebanon mountains/GF5
South Lebanon	SL1	Tyre	S.L.	Well/on the coast/GF2
North Lebanon	NL1	Ehden	1300	Spring/Mar Sarkis. Very high misty summer resort/GF5
	NL2	Kousba El Koura	450	Spring/cave/GF3
	NL3	Barsa El Koura	150	Well/GF3
	NL4	Kobayat—Akkar	540	Spring/GF1/GF5
	NL5	Bire—Akkar	400	Spring/GF1
Nabatieh	NB1	Kfar Remman	500	Spring/GF6
	NB2	Wazzani	300	Spring/in the western rainfed side of volcanic Mount Hermon (2814 m)/GF1
	NB3	Khiam	550	Spring/pond./GF6
	NB4	Houmein	400	Spring/GF5
	NB5	Ain Ibil	650	Spring/in Jabal Aamel/GF4

[a] The Geologic Formation number is shown with GF1 being Upper Cenozoic (Basaltic volcanics), GF2 being Quaternary (Dunes and lake deposits), GF3 being Miocene (Limestone on coasts), GF4 being Upper Cretaceous (Chalks and limestones), GF5 being Lower Cretaceous (Fluvio-deltaic sandstone overlain by thick marine limestones), GF6 being Lower-Upper Jurassic (Thick shelf limestones).

Samples that showed the existence of air bubbles in the laboratory were discarded as these would result in unrepresentative figures.

3.3. Dissolved radon measurement device

In this study, the Electret Passive Environmental Radon Monitor (E-PERM) Electret Ion Chamber (EIC) system manufactured by Rad Elec Inc. (Kotrappa, 1999) was used to measure radon concentrations in water. The application of the E-PERM system for radon-in-water concentrations has been shown to be an easy method giving accurate results (Tai-Pow et al., 1992). E-PERMs have been also used in several other studies of airborne and water-borne radon and have been shown to give results comparable to the other techniques such as liquid scintillation counters and emanation methods (Tai-Pow et al., 1992; Hamlat et al., 2003). The E-PERM method is also cost-effective when repeated measurements are required.

The E-PERM system (Kotrappa et al., 1988, 1990; Kotrappa, 1999) consists of three components: an electret, which is an electrostatically charged Teflon disk that collects ions, a conductive plastic ion chamber into which the electret is loaded, and a voltage reader that records the voltage of the electret. When the electret is screwed into the chamber, an electrostatic field is established resulting in the formation of a passive ionization chamber. The chamber inlet is fitted with a filter that is permeable only to radon and not its daughters. After diffusing into the chamber, radon, and subsequently its daughters, decays by the emission of radiation that results in the ionization of the chamber. The generated negative ions are attracted to the positively charged electret resulting in the decrease of the electret voltage. The drop of the electret voltage is proportional to the radon concentration.

Since the E-PERM is an ionization chamber, the environmental gamma background radiation contributes to the ionization of the chamber resulting in an overestimation of the radon concentration if the background radiation is not accounted for. The E-PERM protocol accounts for the background radiation by subtracting a value that is proportional to the background gamma radiation from the measured value. The background gamma radiation level, found to be equal to 14.08 μR h^{-1}, was measured using a portable Eberline radiation monitor consisting of an E-600 meter and an SHP-270 compensated GM detector.

3.4. Measurement of dissolved radon in water

The water sample bottle was placed open at the bottom of a special jar of known volume (Kotrappa, 1999). The jar allows

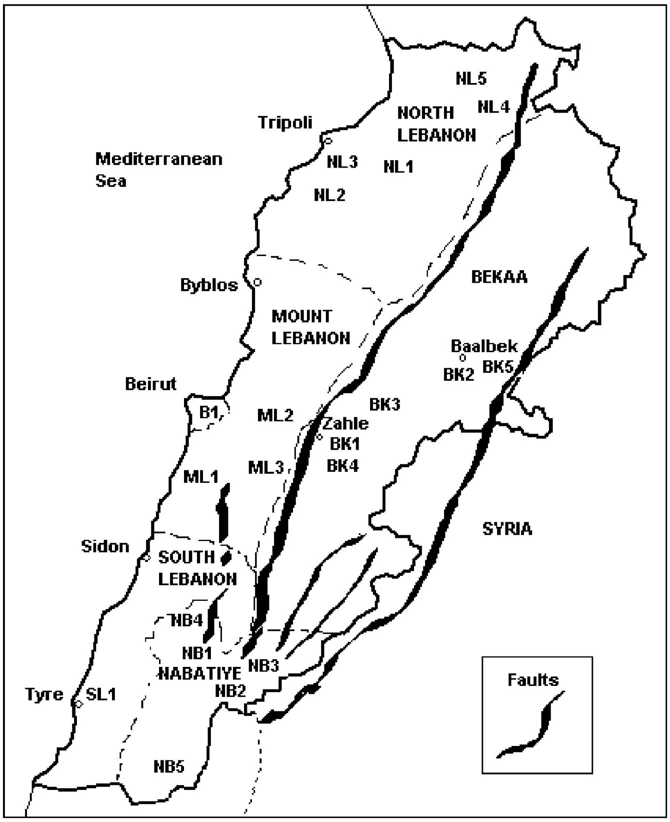

Fig. 1. Map of Lebanon showing administrative districts and sampled water sources (Table 1 gives the name of each location).

an unscrewed-top E-PERM chamber to be suspended from the top of the jar after which the jar is closed for exposure. Following exposure, the radon concentration in the water (R_W) was determined from the radon in the jar air value using the following equation (Kotrappa, 1999):

$$R_W = R_A C_1 C_2 C_3, \tag{1}$$

where

$$R_A = \frac{(V_i - V_f)}{C\tau} - R_\gamma \tag{2}$$

and

$$C_1 = \frac{1}{e^{-\lambda\tau_d}}, \quad C_2 = \frac{\lambda\tau_a}{(1 - e^{-\lambda\tau_a})}, \quad C_3 = \frac{V_{air}}{V_{water}} + c. \tag{3}$$

In the above equations, R_A is the average concentration in the air phase in the measuring jar; V_i and V_f are the initial and final voltage readings, respectively; τ is the exposure time in days; τ_d is the delay period from the time of sample collection to the time of inserting the sample into the measuring jar; τ_a is the analysis period from the time of inserting the sample into the measuring jar to the time the E-PERM is removed for voltage reading.

R_γ is the background gamma count in pCi L^{-1}; V_{air} and V_{water} are the volumes of air and water in the jar, respectively; γ is the radon decay constant (0.1814 d^{-1}); and C is a calibration factor that can be found for short-term electrets from Eq. (4) (Kotrappa et al., 1988):

$$C = 1.8864 + 0.000638\,\bar{V}, \tag{4}$$

where $\bar{V}$ is the mean of initial and final voltage readings. Thus C_1 corrects for radon decay during transport while C_2 accounts for the in-jar radon decay during analysis. The Ostwald coefficient c in Eq. (3) accounts for radon solubility in water and is defined as the ratio of radon concentration in the water phase to that in the gaseous phase. The Ostwald coefficient c is about 0.26 at 20 °C. For the RadElec 68 mL bottle–3720 mL jar system used, C_3 has a value of 55.5; this value is divided by two when using the 136 mL bottle (Kotrappa, 1999).

4. Results and discussion

For each site, two measurements were taken and then averaged (Table 2). Results in Table 2 showed that the dissolved radon concentrations ranged from 0.91 to 49.6 Bq L^{-1} for the sampled 20 sites. This spread in dissolved radon concentration (about a factor of 50) is comparable to the variations reported in Table 3 from recently published studies.

Our measurements showed that only five sites exceeded the 11 Bq L^{-1} EPA limit (Table 2). The four sites with the greatest concentration were: Ain Bourday in the eastern Beqaa valley near the ancient city of Baalbeck (33 Bq L^{-1}), Houmein in the central south of the country (20.8 Bq L^{-1}), and Ain Ibil (49.6 Bq L^{-1}) and Khiam (31.6 Bq L^{-1}) both in the extreme south of the country. Khiam lies near the Golan basaltic formations surrounding the flanks of Mount Hermon. The geological feature of this area is characterized by the near convergence of the Serghaya, the Rachaya, the Hasbaya and the Roum faults. The Beqaa site, at an elevation of about 1200 m lies on the western slopes of the Anti-Lebanon Range in an area that is close to the Serghaya fault. The site at Houmein in the central south near to the coast falls just west of the relatively active Roum fault, which was the site of the most recent major earthquake in the country in 1956 (Walley, 1998). The Ain Ibil result was not totally expected, the site lies in a gently folding calcareous hill area. The fifth site with relatively high radon levels (i.e. greater than 11 Bq L^{-1}) was at Ablah (BK3). This site was the only one out of five well sites that gave a high reading. This region lies in the Bekaa valley (a northern extension of the great rift valley) nearly midway between the Serghaya fault (near Baalbeck on the east) and the main Yamouna fault on the west (Fig. 1).

In fact, of the 15 spring sites sampled, seven may be considered to be somewhat adjacent to geologic fault zones. In four of these seven sites, the water is collected by the users at (or very near) the actual springhead and as a result had a relatively high radon level (with an average of 23.8 Bq L^{-1}). The other three sites had a piping supply system and gave low radon readings (with an average of 4.4 Bq L^{-1}). The remaining eight sites (out of the 15 spring sites) were at locations deemed to be sufficiently distant from the main fault zones. Seven out of these eight sites gave low readings (with an average of 4.8 Bq L^{-1}) and in actuality all these sites had the water piped from the actual springhead. The sample at Ain Ibil (NB5) was taken at the

Table 2
Dissolved radon concentration in the 20 sampled well and spring sources in Lebanon

Location	Initial voltage (V_i)	Final voltage (V_f)	Exposure time (days)	Delay time (days)	Average radon concentration (Bq L^{-1})
Zahle (BK1)	472.5	437	8.97	1.027	3.01
Taalabaya (BK4)	526.5	497.5	8.91	1.18	2.87
Falougha (ML2)	503	473	4.08	1.13	7.57
Houmein (NB4)	643.5	570.5	4.68	1.05	20.79
Deir El Kamar (ML1)	671	643.5	3.22	1.81	9.36
Barouk (ML3)	621	598	3.23	1.74	7.12
Tyre (SL1)	685.5	671	4.27	1.19	0.91
Achrafieh (B1)	698	632	5.03	0.68	7.90
Ras El Ein (BK5)	632	621	4.25	1.11	0.46
Barsa El Koura (NL3)	590.5	558	7.16	2.83	4.89
Kfar Rimman (NB1)	546	506.5	6.76	2.17	7.48
Ain Bourday—Baalbeck (BK2)	390	274	4.82	0.25	33.08
Ablah (BK3)	433.5	356.5	4.83	0.19	19.88
Wazzani (NB2)	607	565	7.04	0.13	5.29
Ain Ibil (NB5)	476	258.5	7.09	0.13	49.60
Ehden (NL1)	256	240	3.67	0.29	2.71
Kousba El Koura (NL2)	358	343	3.67	0.23	2.11
Khiam (NB3)	358	256.5	31.9	0.24	31.56
Kobayat—Akkar (NL4)	728	660	3.76	0.32	9.82
Bire—Akkar (NL5)	741	720	3.77	0.31	1.73

Table 3
Concentration ranges of dissolved radon from recent studies that included ground water and springs

Site	Range (Bq L^{-1})	Reference
Jordan (many locations)	2.8–116	Al-Kazwini and Hasan (2003)
Tassili, Southeast Algeria	0.67–21.25	Amrani et al. (2000)
Eastern Doon Valley, outer Himalaya	20–95	Choubey et al. (2003)
Northern Venezuela	0.1–576	Horvath et al. (2000)
Sudety Mountains, South-Western Poland	1.7–376; the "medicinal spring water" > 74	Kozlowska et al. (2001)
Cyprus (many locations)	0.1–5 (ground water) 0.2–2 (tap water)	Sarrou and Pashalidis (2003)
Bavaria, Germany	Median: 3–50 Max: 16–1220	Trautmannsheimer et al. (2002)
Midgonia Basin, Greece	Below DL-161	Zouridakis et al. (2002)
Lebanon (many locations)	0.46–49.6	This study

actual springhead resulting in a high reading of 49.6 Bq L^{-1} (the highest reading in the study). This suggested that piped water had lower radon concentration than ground water.

Although dissolved radon levels were relatively elevated in five sampled sites, they remained lower than both the European Union reference level set at 100 Bq L^{-1} (EU, 2001), and the currently debated U.S. AMCL set at 146 Bq L^{-1} as an upper limit for drinking water in the United States (NAS, 1999). Based on our study, 11.4 Bq L^{-1} is a preliminary average of dissolved radon in domestically used spring and well water in Lebanon. We also compared the mean, μ, and standard deviation, σ, dissolved radon concentration in well water ($\mu = 7.29$ Bq L^{-1}; $\sigma = 7.5$ Bq L^{-1}) with those in spring water at the fetching point ($\mu = 13.86$ Bq L^{-1}; $\sigma = 15.2$ Bq L^{-1}) and found no significant difference in mean concentration between the two water resource types at the 99% confidence level.

The range of the Lebanese data was compared with that determined in several other countries (Table 3). These data sets included a data set from northern Venezuela that offers a broad range of elevations (360–1250 m), rock types (Granite, Volcanic, and Quartz), and dissolved radon concentrations and a data set from Jordan. Further compared were ensemble-averaged values of other data sets including the Klodska region in Poland, the Tassili region in Algeria, and a data set from 10 regions in Bavaria, Germany. The data set reported in Table 3 from Poland is an arithmetic average of all their 29 sites and across all the reported seasons and spas; the data set from Algeria is a spatial average across all their 15 sites; the data set from Germany reported in Table 3 is from the mean value of their reported median values for their 10 regions.

Geographically, Lebanon is centered upon the high north–south Lebanon range where most of the water sources are located. A large proportion of the population resides, most of the time (it is customary for many to spend one or two months in a mountain resort during summer), in the costal cities and towns and their water is piped from far up in the mountains passing through distribution stations and treatment facilities, resulting in low radon levels at use point. However, a smaller proportion of the population resides permanently in villages and towns and relies heavily on nearby springs especially for their drinking water. In some of these locations the springs are connected to the fetching point by a simple piping system and showed reduced radon levels in our study. On the other hand, water collected from springs directly at the springhead and without piping showed relatively higher radon levels. These sites require further scrutiny and recommendations may be made for water settlement and pipe installation. While results also show some correlation with geological mother-rock and geological structure, more research is needed to validate this result.

5. Conclusion

Ground water samples covering the main administrative districts of Lebanon have been analyzed for dissolved ^{222}Rn concentrations using E-PERM devices. Dissolved radon levels were found to be generally low with a maximum that is less than one half of the reference levels proposed in the European Union and the United States. The preliminary Lebanese national average was found to be $11.4\,\mathrm{Bq\,L^{-1}}$. In many towns where spring water is transported via simple steel pipes to the fetching point, usually in a town center, radon levels were found to be low as opposed to where spring water was used straight from the spring head. The transport mechanism including settling, piping and distribution (taking into account radon decay and radon aeration, type of water flow, travel distance and time, among other factors) of water from a source rich in radon to the consumer's fetching point should receive more attention when health information statistics about water supply and water quality are being compiled. With the various results obtained within such a small, but geologically interesting country, it is recommended that a more detailed study be made using additional data points.

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