A Concise Review Of Radon And Radon In Ground Waters In Jordan: A Case Stuy

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ABSTRACT

With the increasing attention towards Radon gas and its impact on health, a brief review of radon as a tracer, physical models, characterization and techniques, treatment and removal are addressed. Also, the current status of radon in Jordan was highlighted and a case study of radon concentrations in ground waters in fifteen wells, in the Amman-Zarqa basin, were investigated. Liquid-scintillation counting (LSC) technique was used. Samples were collected from 15 wells in different areas of Ras Al-Ain, Al-Rsaifeh and Al-Hashemite. The Al-Rsaifeh area has recorded the highest values for radon water concentrations reaching up to 14.8 and 30.7 Bq/l. The average value of all tested samples was 5.77 Bq/l.

1. INTRODUCTION

It's a naturally occurring radioactive gas which is invisible, odorless and tasteless. Relative to other isotopes, radon (^{222}Rn) has a long half-life estimated time of 3.825 days. It comes from the radioactive decay of radium (^{226}Rn) and two other natural isotopes of Actinon (^{219}Rn) and Thoron (^{220}Rn). Radon is an element found in most rocks, soils and mainly ground waters. Surface water has low concentration because it can easily emit the Radon. It is known that indoor Radon accumulates to levels orders of magnitude higher than the outdoor levels (**Liendo *et* al., 1997**). Radon (^{222}Rn) is the hidden killer, causing an increasing risk for lung cancer (**National Academy of Sciences, 1988; Jostes, 1996; Hofmann,1998**; **Torres-Duran et al, 2014).** Previously, radon concentrations above 400 Bq/m^3 was considered to constitute a health risk, however, recent epidemiological findings demonstrate lung cancer risk from exposure to indoor radon at levels in the order of 100 Bq/m^3 (**Pacheco-Torgal, 2012**; **NAS, 1999; European Commission, 2001; Health Canada, 2009).** Also, a link was established between radon exposure and employees working in drinking water supply systems, such as well houses, conditioning plants, reservoirs, pumping stations (**Schmitz et al, 2000; Schmitz and Nickels, 2001**). To support the previous link, **Trautmannsheimer et al, 2002**, looked at the whole of Bavaria, Germany, water supply facilities and investigated radon concentrations in groundwater and indoor air and as to the radon exposure levels to which the staff working. The authors found that in the east Bavarian crystalline region, indoor radon gas concentrations of up to 300 kBq/m^3 were observed. About 10% of the processing plant workers of this region get an annual effective dose of more than 20 mSv.

2. RADON AS A TRACER

There are various applications for radon as an important natural tracer especially in earth and geochemical sciences such as; studies of aquifer flow rates, groundwater-recharge rates, mixing, and residence times. Therefore radon concentrations in ground waters play an important role in detecting seismic activities (**Corbett et al., 1997; Dubinchuk, 1981; TURK et al, 1996; Hussain et al., 1999; Lehmann and Purtschert, 1997; Schmidt et al., 2010; Snow and Spalding, 1997; Tuccimei et al., 2005; Kim and Hwang, 2002; Burnett and Dulaiova, 2003; Hwang et al., 2005).** One of the earlier model review was carried out by **Monnin and Seidel, 1997**. They presented physical models related to radon emission in connection with dynamic manifestations in the upper terrestrial crust. In seismic activity studies, **THEODORSSON, 1996** described a single phototube liquid scintillation counting system for automatic determination of radon in ground water for earthquake prediction., **Singh et al, 1999**, established a radon earthquake link which is assumed to be related to the possible compression of the deep rocks due to different ground levels prior to the earthquake. In Japan, **Nishizawa et al, 1998,** detected an anomalous increase in radon concentration associated with the earthquake swarm off the E coast of the Izu Peninsula in 1995.

Erees et al., 2006 and 2007, showed that there is relationship exists among high radon values and high heat water flow, high seismicity in Western Anatolia, western Turkey. Also, **Yalım et al, 2012**, measured radon concentrations in water from 4 wells in the Aks ehir-Simav Fault System (ASFS) in Afyonkarahisar province, Turkey, from August 2009 to September 2010. The authors observed Anomalous decreases in radon concentrations in the wells to precede the earthquakes of magnitudes ranging from 2.6 M to 3.9 M. **Nevinsky et al, 2015**, showed the changes of radon maps in the territory of South Russia during earthquakes.

In China, water well and spring parameters data (water radon, gas radon, water level, water temperature) are collected from a large network of stations and managed by the China Earthquake Network Center (CENC). **Ye et al, 2015**, studied the behavior and characteristics of such data of water radon during the period 2002-2014. They concluded that for the two major earthquakes along Longmenshan fault, the data analysis showed a positive and negative relation of water radon and water level prior to these earthquakes, and it is difficult to find any trend of water radon and changes in water radon pattern with these two earthquakes that could prove as a reliable precursor of earthquakes.

3. RADON PHYSICAL MODELS

There are several models were presented during the last decade to illustrate radon migration and the exhalation rate from porous uranium ore or tailings and building materials (**Petropoulos et al., 2001; Righi and Bruzzi, 2006; Sahoo et al., 2010; Kumar et al., 2014; Fan et al., 2015; Sundar et al., 2015). Garcia-Vindas and Monnin, 2005**, assessed the time required for radon to dissolve in water and reciprocally to degas from it, and estimated the partition ratio between the two phases.

Recently, **Ye et al, 2016**, established a one-dimensional steady-state mathematical model of radon transport in fragmented uranium ore. The model was utilized to obtain an analytical solution for radon concentration in the air-water, two phase system under steady state conditions, as well as a corresponding radon exhalation rate calculation formula. Meanwhile, **Rovenská and Jiránek, 2012**, carried out a review of various methods used for Radon diffusion coefficient measurement in waterproofings.

On the other hand, **Ongori et al, 2015**, investigated the transport of radon at the water–air interface is investigated at very low turbulence and by using a mathematical model and an experiment the radon transfer velocity coefficient from the water–air interface was found to be $(1.4\pm0.2)\times10^{-6}$ m s^{-1}. By using statistical approach, **Fernandez-Cortes et al, 2013,** identified fluctuations in radon activity as a response to variations in weather and microclimate conditions by clustering entropy changes on the radon signal and parameterized to predict radon concentration anomalies.

4. RADON CHARACTERIZATION AND TECHNIQUES

Testing for radon concentrations in drinking water is not an easy task. This is due to the ease of radon release from water during handling, stirring and/or emptying and filling water from one container to another. All these actions will help in releasing dissolved radon and affect the accuracy of measured results. Also, heating up to boiling will completely release radon from the water into the air. **Jobbágy et al, 2017**, presented a brief overview on radon measurements in drinking water and information about currently used standard and routine methods for radon analysis.

During the last 25 years, researchers worldwide got heavily involved in developing different characterization techniques to analyze Radon (^{222}Rn) in drinking water. These techniques are divided into three categories; a) Liquid-scintillation counting (LSC) (**Prichard and Gesell, 1977; Vitz, 1991; Barnett et al., 1995; Hamanaka et al.,1998**), b) γ-Ray spectrometry (**Countess, 1976; Sanchez et al., 1995; Shizuma et al., 1998; Erlandsson et al., 2001; Povinec et al., 2006**), c) ∝-Particle instruments (**Burnett et al., 2001; Dulaiova et al.,2005; Lee and Kim, 2006**; **Dimitrova et al, 2011**) which include; Alpha scintillation detectors, Alpha track-etch detectors, and Alpha radiation spectrometers. These devices depend on whether the techniques measure radon or radon decay products, and the duration of the measurements. Also, field measurements applicability, portability, convenience and reliability are important design parameters for these instruments (**Papastefanou, 2002**). Reviews were conducted by **Durrani (1997),** and **Nikolaev and Ilic, 1999,** on passive radon radiometers, based on alpha particle etched track detectors which are used to assess radon exposure. The authors reviewed various devices used for measurement of the volume activity of radon isotopes and their daughters and determination of equilibrium coefficients. The visualization of tracks was highlighted by **Tommasino (1997).** Whereas, calibration, standardization, and sources of error in etched-track radon measurements were presented by **Miles (1997) and Ibrahimi et al, (2009).**

For water-radon measurements, there are several international standards which can be used such as; ISO13164-3,2013, ISO13164-4,2015, and ASTM D5072-09,2016. The use of calibrated standard solution is vital. **Salonen, 2010**, demonstrated that ^{210}Pb, ^{210}Bi and ^{210}Po can be extracted from water to organic cocktails when radon measurement in drinking water is calibrated with a ^{226}Ra standard solution containing these nuclides. Radon loss from water during 15 days of storage was evaluated by **Lucchetti et al, 2016**, and found that Polyethylene terephthalate (PET) and polylactic acid (PLA) bottles lost 0.4-7.1% and 3.7% of initial radon respectively. Whereas, percent radon loss during storage in 1 L HDPE bottles was estimated at 0.0045 min^{-1} (**De Simone et al, 2015**).

Usually large water samples are needed to obtain a highly sensitive radon assay of water, and the results become questionable when using small low-level detection instruments. **Schubert et al, 2014**, presented sample volume optimization for LSC method. They showed that sample volumes smaller than about 500 ml lead to noticeably lower count-rates (poorer counting statistics), and volumes of about 500–900 ml should be chosen for LSC radon-in-water detection, if 20 ml vials are applied. To maximize the LS accuracy and sensitivity, correct selection of caps, scintillator and water to fluor ratio is highly important. **Hakl,et al, 1995 and 1996**, developed a cheap method, called track etch radon monitor in a glass vessel, for the simultaneous examination of a larger number of samples. The work was extended to address the temperature dependence of the calibration factor of radon and radium determination in water (**HUNYADI et al 1999**).

Wang et al 1999, described the design and application of an electrostatic radon detector to determine the amount of radon emanating from a column containing MnO_2 used to extract radium from the water. **Yasuoka et al, 2005,** measured the ^{222}Rn concentration in groundwater using the direct dpm method based on the integral counting method for a two-layer sample. Whereas, **Al-Azmi, 2004**, demonstrated the feasibility of using olive oil for radon gas measurements in water.

It is known that the radioactivity of ^{222}Rn in ground Water depends on the radioactivity of its parent ^{226}Ra and its grandparent ^{230}Th in the solid and water. **Sun and Semkow, 1998**, addressed and investigated successive Alpha recoils processes where a radioactive daughter is mobilized from its initial position by the energy of an $\propto$-decay. **Salih et al, 2004**, observed that ^{222}Rn has a positive correlation with fluoride and uranium and that emanation of ^{222}Rn from water as a function of fluoride, pH and carbonate. The authors observed that fluoride in water adheres or trap ^{222}Rn preferably in acidic water.

As Radon partition coefficients are available for a range of pure substances, several on site radon testing systems were developed and available in the open literature. These methods use the effect of radon partitioning between water and air (**Monnin and Seidel, 1998; Freyer et al., 2003; Dulaiova et al., 2005)**. Hence, both **Lee and Kim, 2006 and Schubert et al, 2006**, 2007 and 2008 developed a simple and portable technique which allows for fast and uncomplicated measurement for moderately and high levels of ^{222}Rn in natural waters on-site. Whereas, **Bonotto and Mello, 2006**, developed a combined method of the emanation procedures and gamma spectrometry to determine the ^{222}Rn volumetric activity in waters, as well the direct measurement of their daughters ^{214}Bi and ^{214}Pb. Whereas, **Talha et al, 2008**, presented a model which uses the time evolution of radon concentration in borehole water to indicate the pumping time required for sampling an aquifer. **Talha et al, 2010**, presented a quasi in-situ approach of measuring radon in the field using γ-ray spectrometry whish is based on a MEDUSA detector inserted in a steel drum or in a plastic tank and the MDA of this method is 0.5 Bq/l radon in-water. Recently, **Voltaggio and Spadoni, 2013**, studied the use of passive gas accumulators made of polydimethylsiloxane (PDMS) mixed with activated Carbon (AC) to measure their efficiency for sampling Rn in water and a linear relationship between them. The detection limits for ^{222}Rn in water was lowered to 20 Bq m^{-3}.

5. REMOVAL OF RADON

It is clear that radon emanation is highly influenced by radon source (**MISDAQ and SATIF, 1996**). Several researchers conducted various techniques to remove and reduce radon concentrations in drinking and treated waters. In an enclosed room, the equilibrium of the radon concentration between water and the air is estimated by the solubility coefficient, which is highly dependent on temperature and air flow through the water **(Clever 1985; Reichelt,1996**). There are two main parameters which influence radon removal in groundwater; air-to-water ratio and detention time. **Alabdula'aly and Maghrawy, 1999**, concluded that the radon removal increases independently with the increase of the air-to-water ratio and the detention time in an aeration system and can give an effective radon removal up to (~90%). In a lab scale operation, **ABULFARAJ and MAMOON, 1995**, examined the effect of activated carbon, aeration and heating on the removal of radon from treated water. They concluded that ^{222}Rn concentration decreased when increasing contact time and mass of the added charcoal. Also, decreased with increasing air flow rate, duration of aeration and temperature increase.

There are various factors affecting radon emanating from water, such as: water temperature, flow rate, spraying techniques, delayed evaporation and mixing rates (**Raunemaa et al, 1995**; **Jalili-Majareshin et al, 2012**). Also, it was found that it is possible to reduce radon concentrations treating water using both physical and chemical methods such as filtration and reverse osmosis, flocculation/sedimentation, coagulation and disinfection. These methods can be efficient by ~30–95% **(Fonollosa et al., 2015; Gäfvert et al., 2002;Montaña et al., 2013; Nieto et al., 2013; Al-Jaseem, et al, 2016).**

6. RADON IN JORDAN

On the health impact of radon, **Khalidi and Taweel, 2016**, found an evident lack of knowledge of radon and its health hazards in Jordan, and an overall estimate of the level of awareness on radon information among Jordanians is 63.1%, which is quite low. **Al-Zubaidy, 2012** showed an increase in the probability of Lung Cancer Disease in some homes of Al-Mafraq City caused by Radon due to poor ventilation. The average radon concentration in Al-Mafraq city was (49.3 ± 17.4) $Bq.m^{-3}$ which is comparable to the nation radon concentration of 42 Bq/m^3.

Indoor radon concentrations (Bq/m^3) in selected cities in Jordan were previously recorded as; Ajloun 50, Amman 41, Irbid 30, Jerash 40, Karak 105, Madaba 74, Sault 46, Tafilah 41, Zarka 33 (**Khataibeh et al, 1997**). **Elzain, 2011**, found that the seasonal radon concentration levels in pharmacies and shops, are minimum in summer (18.2±6.7) Bq/m^3 in pharmacies, and (16.6±3.9) Bq/m^3 in shops, and maximum in winter season (50.0±4.1) Bq/m^3 in pharmacies, and (45.1±4.2) Bq/m^3 in shops at Al-Zarqa town. Also, **Al-Zubaidy and Mohammad, 2011 and 2012**, measured radon gas concentration inside homes in Hakama city which lies 80 km north of Jordan The average radon concentration was ranged from 18.8 Bq/m^3 to 37 Bq/m^3 which is below the Jordanian national level. In As-Salt area, **Ya'qoub et al, 2009**, recorded radon gas concentration levels in houses of As-Salt area to have an average value of 111 ± 4 Bq m^{-3} during April to July of the year 2004. The concentration of radon short-lived daughters was estimated to be 44 ± 2 Bq m^{-3}.

In Soum area, **Abumurad and Al-Tamimi, 2005**, highlighted that radon levels in winter exceed that of fall or spring by about (10–12%). The average radon concentration in the study area was 144 Bq m^{-3}, which is about three times the national average of Jordan. In Tafila Province, **Abu-Haija et al, 2010**, found the overall average of radon concentration level inside the dwelling of Tafila province was 26.28 Bq/m^3. In the dwellings of Foara and Hawar villages, Mohammad and Abumurad, 2008, recorded concentration between (36.6 ± 7.5) Bq/m^3 and (50.8 ±5.4)Bq/m^3. **Abumurad, 2001 and Abumurad et al, 1997**, determined radon (^{222}Rn) levels were in Al-Mazar area, northern part of Jordan by passive diffusion dosimeters (using CR-39) used indoors in different seasons. The author found that the average radon concentration during the summer season was about 62 Bq/ while in winter season it was about 81 Bq/m^3. **Abdullah, 2012**, presented a study to measure natural radioactivity concentration in the dwellings of the Umm Gais village by using a solid state nuclear track detectors. The author showed that the concentration was relatively high (80 ± 16

Bq/m^3) in buildings made of stones while concentration was low (42 ± 11 Bq/m 3) in the buildings made of blocks. In addition, the concentration was different with increase in age of the building, the average were (56 ± 12 Bq/m^3), (44 ± 11 Bq/m^3), (30 ± 10 Bq/m^3) in more than 25 years, between 15 – 25 years and less than 15 years, respectively. In general the radon concentration in the Umm Gais village was found to be about (58 ± 14 Bq/m^3).

In radon concentrations in soils, at least 80% of the radon emitted into the atmosphere comes from the top layers of the ground around 1.5 m **(NCRP, 1984**). **Al-Shereideh et al, 2006 and Ershaidat et al, 2015**, measured soil radon concentration levels, at different depths, in a phosphate site in the city of Irbid, north of Jordan by using the CR-39 dosimeters for the depths 0, 20, 40, 60, 80 and 100 cm were 1390, 9682, 16778, 19817, 21720, and 24206 Bq/m^3 respectively. **Kullab, 2005**, conducted an assessment of Radon-222 concentrations in buildings, building materials, water and soil in various cities in Jordan.

. In Jordanian drinking water and hot springs, **Al-Kazwini and Hasan, 2003**, found that ten sources with radon activity have less than (11) BqL^{-1}, 12 sources with activity between 11 and 37 Bq L^{-1} and three sources with activity greater than (37) BqL^{-1}. The two sources of drinking water with the highest radon activity are those at Awajan (Zarka province, northern region) and at Umm- Al-O'rooq in Shafa Badran (Amman Suburb, central region). Fortunately, these water sources provide water to a very limited number of residents. The third source with radon activity larger than 37 Bq/l is at Al-Khaldiyah and Dleel. This source provides water to several small towns in Al-Mufraq province. **AL-Amairyeen, 2010**, measured radon in Wadi Bin Hammad cold and hot water springs used a portable Geiger-Muller counter and an NaI (Tl) detector. The measured absorbed dose rates in air ranged from 15-30 nGyh^{-1} in cold and 100 to 2740 nGyh^{-1} in hot water spring areas

6.1 AMMAN-ZARQA BASIN: CASE STUDY

Jordan has a Mediterranean climate, where it is hot and dry in summer, and cool and wet in winter. The rainfall occurs in winter mainly between the period of November to April, and with an average annual rainfall of less than 200mm in east south to more than 500mm in the northwest of Jordan. The recorded average temperature in the selected study area ranges from 4 C in winter and up to 35 C in summer (WAJ).

The Amman–Zarqa basin is the selected area covered in this study, which is about 850 Km^2, where 70% of Jordan population live. The groundwater of this area is used to meet the domestic, irrigation and industrial demands. The study area contains hundreds of public and private wells in addition to many sprigs. Ras Al-Ain spring is the most famous one in this basin, which is the main source of water supply in the Amman–Zarqa basin. The average rainfall ranges from 400–500 ml in the west part of the basin, while declines to 150-200 ml in the east part of the basin (WAJ). The geological aspects of the area under study, consists of two groups, Balqa and Ajlun, which are formed during the Cretaceous period. The first group of developed areas are Al-Hasa, Amman and Wadi Ghudran. These areas, mainly contain phosphate, variable sequence of chert, limestone, dolomite and chalk, while the second group of developed areas; Wadi Sir, Shueib, Hummar, Fuhies and Naur, and they mainly contain limestone, chert, dolomitic-limestone, marl and chalk.

6.2 Collected Samples

Samples were collected from 15 wells in different areas of Ras Al-Ain, Al-Rsaifeh and Al-Hashemite which represent the main water sources though the Amman-Zarqa Basin. These samples were taken over the period from May to July 2017 during the season of summer. In collecting the samples attention was paid to minimize the loss of radon during collection and transport.

6.3 Measuring method

Liquid scintillation counting Tri- Carb 3110 with discriminator which is widely used for measurement of Radon. It has an advantage of the high solubility of radon in organic solvents, which are used in cocktails for LSC, and of high counting efficiency for alpha particles. Several chemicals and reagents were used such as; Scintillation cocktail Opti-fluor, Nitric acid HNO3 aristar 69%, ^{226}Ra standard certified solution with 0.2086µCi/l, and Distilled Water. The efficiency of the narrow alpha window is approximately 300% due to the presence of the three alpha emitters, ^{222}Rn, ^{218}Po, ^{214}Po .The discriminator setting of the used LSC is shown in Figure 1. The Alpha /Beta discrimination was tested by using standard of Sr-90 as Beta emitters and Am -241 as Alpha emitters.

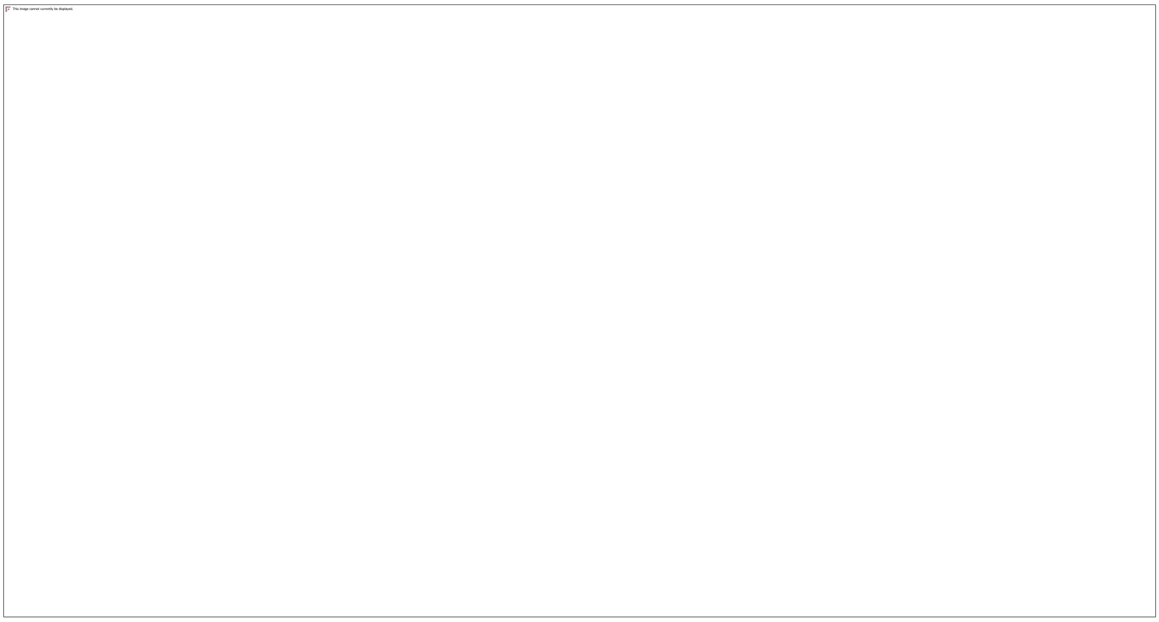

Figure 2. Discriminator setting of the used LSC

The main critical values of the operated system are the pulse decay discriminator (PDD) = 136, % Alpha spillover 3.84, and % Beta spillover 2.55. The LSC system was calibrated using three standards of Radium at different concentrations of 10, 5 and 2.5 Bq in 500 ml each solution. The calculated average efficiency is around 0.769 with average error of 0.0195.

6.4 RESULTS AND DISCUSSION

The international acceptable level oof radon concentration in drinking water is below 300 pCi/L (11 Bq/l). From all examined samples from different wells as shown in Tables 1-3, it is clear that radon water concentrations are at the acceptable levels except in two wells (8 and 9A) in the Al-Rsaifeh area having values reaching 14.8 and 30.7 Bq/l. From health and safety point view, these values do not impose much concern on health as these values are expected to drop dramatically during the handling and pumping of water to the housing facility. These values were expected due to presence of phosphate mines in the area where it contains noticeable amount of Uranium. Throught the Amman-Zarqa basin, the highest recorded value was 30.7 Bq/l and lowest was 0.60 Bq/l with an average value of 5.77 Bq/l. Figure 2 shows a schematic representation of radon levels in the study area.

Table 1 Calculated ^{222}Rn by using LSC for wells in Ras Al-Ain area.

Location name	Average count per minute(CPM)	Calculated Radon Concentration (Bq/l)	Estimated error
Ain Gazal Well	181	5.55	±0.5
Ras Al-Ain deep well	20	0.60	±0.1
Al-Mohagreen well	142	4.40	±0.4
Amman mills	24	0.74	±0.1

Table 2 Calculated ^{222}Rn by using LSC for well in Al-Rsaifeh area.

Location name	Average count per minute(CPM)	Calculated Radon Concentration (Bq/l)	Estimated error
Al-Rsaifeh Well-1A	199	6.09	±0.6
Al-Rsaifeh well-1	169	4.52	±0.5
Al-Rsaifeh well-11	256	6.85	±0.7
Al-Rsaifeh	115	14.79	±0.3

well-9A			
Al-Rsaifeh Well-8	263	30.70	±0.7

Table 3 Calculated ^{222}Rn by using LSC for wells in Al-Hashemite area.

Location name	Average count per minute(CPM)	Calculated Radon Concentration (Bq/l)	Estimated error
Auajan 21	175	5.37	±0.5
Auajan 23	66	1.76	±0.2
Al-Hashemite 2	29	0.77	±0.1
Al-Hashemite 5	37	1.00	±0.1
The spring of Qeneh	18	2.34	±0.1
Well Karam/ Qaneh	8	0.89	±0.1

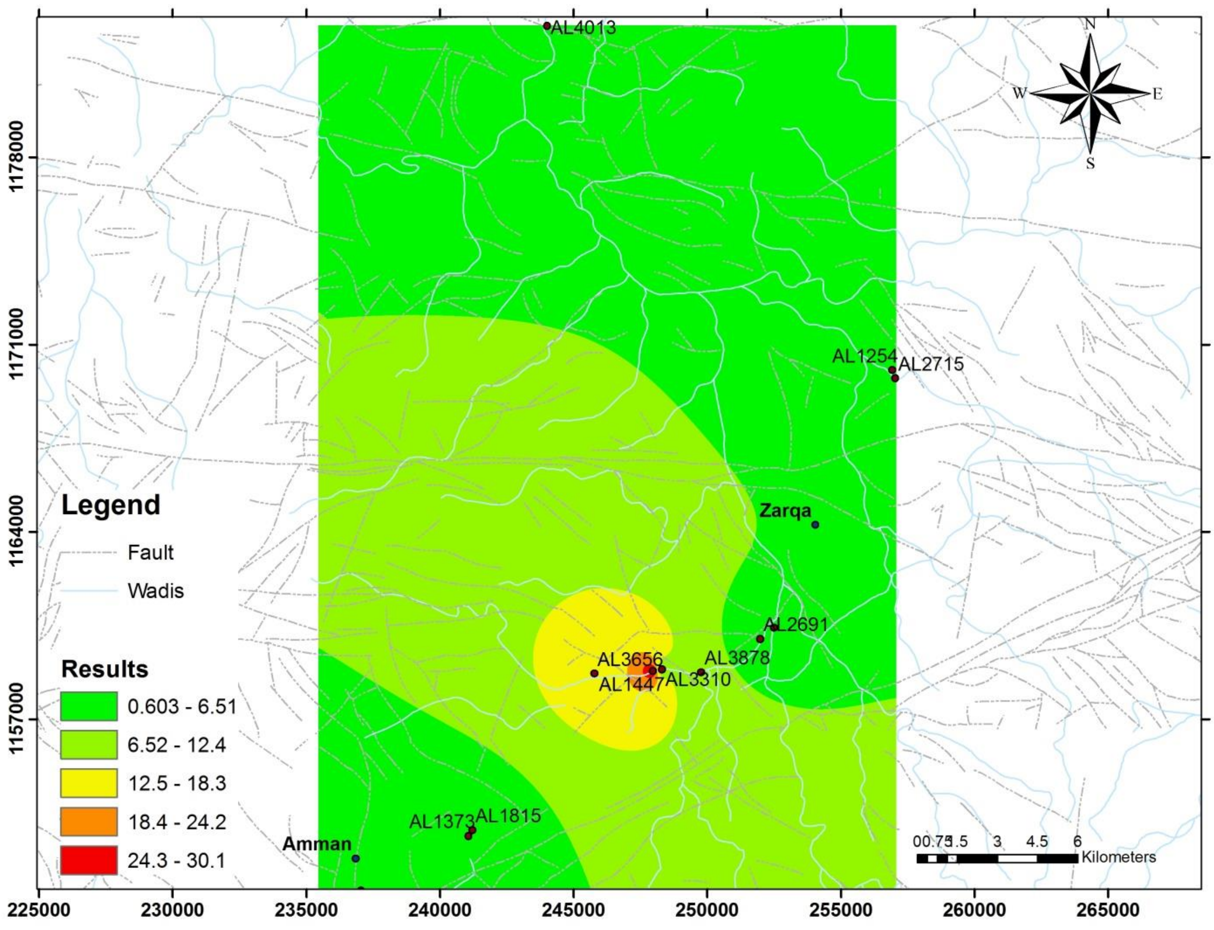

Figure 2 Schematic representation of radon concentration levels in the study area

7. CONCLUSIONS

During the last decade, radon has gained great attention at both national and international levels. Advances were made in various measuring methods, especially for the direct test measure on location. Based on this brief review on Jordan, it is clear that Radon health risk problem is not well addressed within the community, and there is a lack of knowledge of how such problem exists and can be minimized or treated. There is a need to develop national regulations and standards. The Al-Rsaifeh area has recorded the highest values for radon water concentrations reaching up to 14.8 and 30.7 Bq/l. The average value of all the tested samples was 5.77 Bq/l.

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