ON-SITE DETERMINATION OF THE RADON CONCENTRATION IN WATER: SAMPLING & ON-LINE METHODS

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ABSTRACT

Radon can dissolve and build up in water from underground sources. If water comes from a well, it may contain Radon. Radon in the water dissolves and escapes into the air during household water use, especially when it is heated. An easy-to-handle experimental setup for the on-site determination of the Radon concentration in water samples has been developed and tested. In contrast to laboratory techniques for Radon-in-water determination such as liquid scintillation counting or gamma-ray spectroscopy, the described equipment and method allow for a fast and uncomplicated determination of the Radon concentration in water samples immediately on site with satisfactory accuracy. The method is based on the water/air partitioning of Radon, which is known quantitatively over a wide range of temperatures. Another set-up has been designed for on-line Radon monitoring in water using the newest Radon monitor AlphaGUARD DF2000.

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/ INTRODUCTION

The first goal of this paper is to discuss the characteristics of Radon in water and the associated health risks. The second goal is to present the methodology and application of Radon in water measurement, either by sampling or continuous monitoring. The final goal of this paper is to show why Radon in water measurement is relevant before and after building.

Radon transfers into and out of the water phase, it is found in groundwater that flows through uranium rich rock formations. In buildings, if water comes from a well, it may contain Radon. Radon in the water dissolves and escapes into the air during household water use, especially when it is heated. In case of high Radon level found in buildings, finding Radon sources entries could be a challenge. Before construction, measuring assessing the risk could help to set adapted countermeasure to get low Radon levels in new build.

Within this paper, the Radon concentrations in the groundwater samples were measured using the continuous active Radon monitor ALPHAGUARD of Bertin Instruments - SAPHYMO, US-France-Germany. This detector was selected due to its proven calibration stability and fast response to concentration gradients, which have been confirmed in numerous studies.

There are many positive applications assessment of Radon concentrations (e.g. earthquakes predictions) since Radon concentration is highly related to the geologic formation (Virk & Singh, 1993). Radon concentration measurements are also used as environmental tracer or indicator for the localizations and evaluation of non-aqueous phase liquid contamination in soils and aquifers (Schubert et al., 2001; Schubert et al., 2011), estimating the ground water flow velocities (Hamada, 2000), and investigating the ground water/surface water interaction process (Schubert et al., 2008).

/ RADON IN WATER & ASSOCIATED HEALTH RISKS

Human beings are continually exposed to natural radiation. The exposure of human body from natural radiation sources is external and internal. Internal exposures originate from the intake of naturally occurring radioactive materials (NORM) by inhalation and ingestion. The major contribution to this human exposure comes from Radon (222Rn). Radon and its short-lived decay products constitute more than 50 % of the total dose originating from natural sources. Radon is the heaviest, colorless, odorless, tasteless and only radioactive member of noble gas group with a half-life of 3.82 days. It belongs to the 238U decay series.

Radon can get into the human body through inhalation Radon in the air and ingestion Radon in drinking water. Radon and its daughters emit ionizing alpha and beta radiation that may cause cancer in human organs. Radon gas is soluble in water, thus Radon in water generated by the decay of uranium in the rocks and soil. The contaminated underground water sources can contaminate drinking supplies such as wells, springs and boreholes. Geology is the primary influence on dissolved Radon levels (Loomis, 1987; Campbell, 2006 and 2008). Radon is similar to carbon dioxide in a soda bottle. The carbon dioxide is dissolve and then release when the soda bottle is open. Therefore, domestic water can make variable contributions to indoor Radon level. The health risks can be significant especially in regions where drinking water comes from underground sources with uranium-rich rocks.

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Concentrations of Radon in water can vary widely within a given rock formation, area, or county. Well owners in moderate to high susceptibility zones have very low Radon levels while some well owners in areas outside these zones have very high Radon levels (see Final Report of the NC Radon-in-Water Advisory Committee, March 9, 2011). This is problematic because a significant proportion of the American population relies on groundwater as its principal drinking supply.

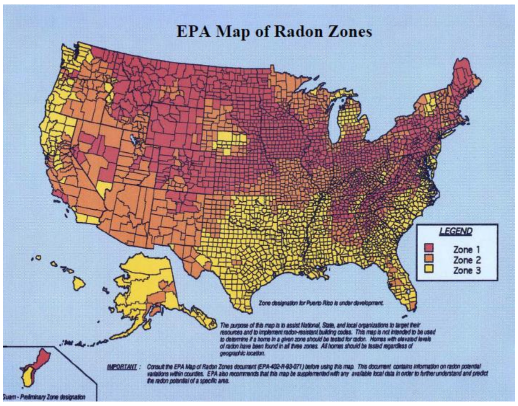

Figure (1): EPA Map of Radon Zones by U.S. County (epa.gov)

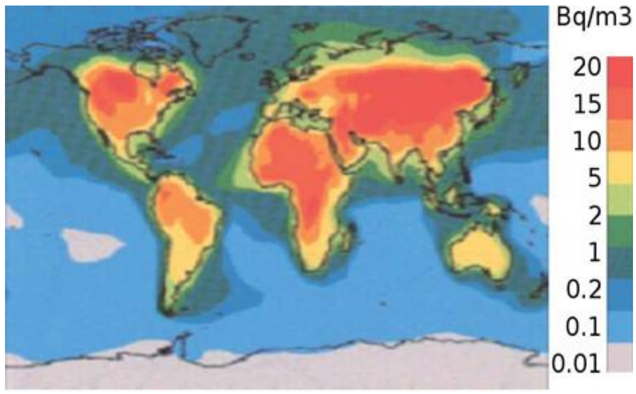

Figure (2): Atmospheric Radon Concentration (fr.academic.ru)

According to USEPA (2010), the primary health risks from Radon in drinking water are lung cancer, from inhaling Radon discharged from water used in the home, and stomach cancer, from ingesting Radon in drinking water. Radon is released into the air when water is used for showering and other household purposes. High level of Radon in domestic water can make major contribution to the indoor Radon exposure. When Radon is inhale or ingest, the alpha particles from its radioactive decay can interact with the biological tissues leading to DNA damage. Even very small amounts of airborne Radon may pose at least some risk (Field, 2010, 2000; Krewski and others, 2005; Darby and others, 2005).

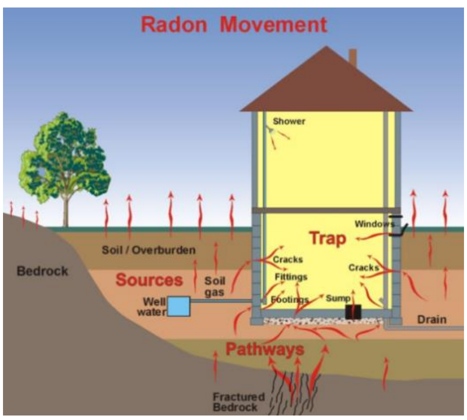

Figure (3): In house Radon infiltration (www.seacoastinspections.com)

A Radon concentration of 10,000 pCi/l in water contributes, on average, an additional 1 pCi/l to the overall indoor air Radon concentration (National Research Council, 1999). The primary risk associated with long term exposure to Radon in water is from inhalation (lung cancer) - about 90 percent of the total risk (National Research Council, 1999). A much lower secondary risk is from ingestion (stomach cancer). The lung cancer risks associated with long-term exposure to Radon are significant when compared to other environmental hazards. In fact, Radon is estimated to cause about 21,000 lung cancer each year in the United States (EPA, 2003). Cancer mortality risks associated with Radon in water are greater than with Radon in air. Because of all these reasons, the evaluation studies of the radiation dose from Radon ingestion and inhalation in drinking water are conducted continuously all over the world. All of these studies are improving the understanding of the environmental processes that effect Radon exposure.

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/ HOW TO MEASURE RADON IN WATER

The concentrations of Radon in the groundwater samples were measured using the continuous active Radon monitor ALPHAGUARD of Saphymo (formerly Genitron), Germany. This detector was selected due to its proven calibration stability and fast response to concentration gradients, which have been confirmed in numerous studies. It utilizes a pulse-counting ionization chamber of 0.56 liter active volume to detect Radon and it is suitable for long-term monitoring of Radon gas concentrations from 2 to 2,000,000 Bq/m3 (0.054 to 54,000 pCi/l). The measurement procedure is based on a sophisticated multilevel digital signal processing method (DSP) of the measured pulses caused by Radon that enters into the detection volume of the ionization chamber. In addition, the ALPHAGUARD registers the values of the main environmental parameters (temperature, humidity and atmospheric pressure) during the time of measurement

/ SAMPLING SYSTEM

Radon measurements of water samples by using AquaKIT are precise and correct. This is assured by the calibration of AlphaGUARD DF2000, which can be traced back to different national standards, as well as by the integrated quality assurance system.

The portable combination AquaKIT - AlphaGUARD DF2000 is adapted for operation in the field.

AlphaGUARD ionization chamber is part of the gas cycle. Radon is expelled from water samples (placed in emanation vessel) using the pump in a close gas cycle. The safety vessel is connected to the emanation vessel. All drops would deposit in it if they had got into the gas cycle during the degassing process. Therefore, the stress of the water vapor is minimized for the Radon monitor. Finally, the water is injected into the emanation vessel, and the AlphaGUARD is switched on. The glass vessels of the AquaKIT measurement equipment grant a hermetically sealed enclosure of the Radon expelled from the water samples as well as a fast change of samples, which prevents incorrect measurements due to leakages.

The flow rate of the pump was 0.3 l/min. After 10 min the pump was switched off and the AlphaGUARD remained switched on for another 20 min, so the Radon measurement was continued. The AlphaGUARD monitor worked in a ‘flow’ mode in a 1 minute measurement cycle. Before every water sample measurement, for a few minutes, the background of empty set-up was measured. Calibration of the measuring system has been carried out by Saphymo (formerly Genitron Instruments), Germany, with a guaranteed stability and accuracy for 5 years.

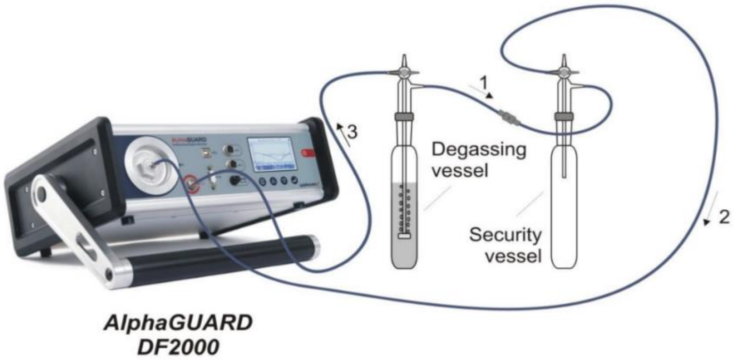

Figure (4): Measuring set-up with AlphaGUARD DF2000

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Figure (5): AquaKIT Components

AquaKIT Components:
1 x degassing vessel
1 x security vessel
1 x spare degassing vessel
4 x storage vessels for sampling (2 x 100 ml/ 2 x 500 ml)
4 x plastic injections for sampling (100 ml)
1 x mounting socket
7 x connecting tubes
1 x measuring cylinder
1 x thermometer
1 x active coal filter cartridge

/ DETERMINE IF RADON IS GENERATED FROM Ra-226

To determine if part of the Radon in the water sample is generated from Ra-226, which could be dissolved in the own sample, a part of each water sample has been stored in their glass containers in order to re-measure the Radon content after the time required for Ra-226 to reach secular equilibrium with its progeny.

/ PRECISION MEASUREMENT BY DIMINISHING THE BACKGROUND RADON LEVEL IN THE MEASURING SET-UP

Generally, only for high precision measurement it may be necessary to diminish the background Radon level in the measuring set-up before entering the sample. For this, the supplied active coal filter cartridge is bound into the gas cycle of the measuring set-up (see Figure 6). The Radon in the measuring set-up is absorbed by the active coal and therefore excluded of the gas cycle.

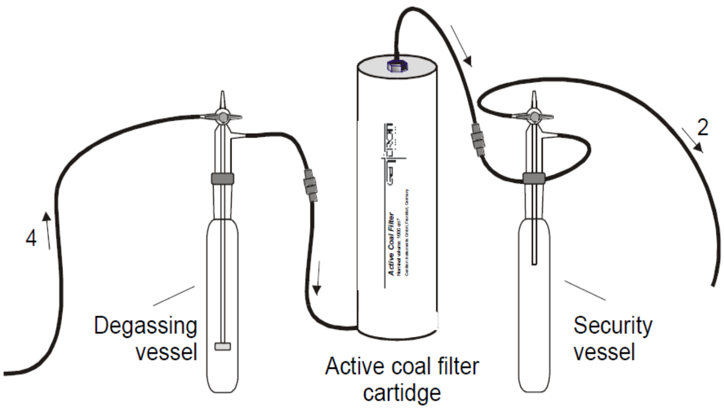

Figure (6): Measuring set-up: Active coal filter cartridge

Improvement of the detection efficiency can also be achieved by a lower dilution of the Radon in the system expelled of the water, e.g. by increasing the relation V_{sample} : V_{system}. By using the 500 ml lower glass parts instead of the 100 ml vessels the relation of the volume is elevated from 1 : 11 to approximately 1 : 2 and by this the dilution is lowered by the adjacent reciprocal value.

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/ RADON CONCENTRATION FORMULA IN WATER SAMPLES

c_{water}*[Bq/L]*

$$c_{water} = \frac{c_{air}\left(\frac{V_{system} - V_{sample}}{V_{sample}} + k\right) - c_0\left(\frac{V_{system} - V_{sample}}{V_{sample}}\right)}{1000}$$

c_{water}*[pCi/L]*

$$c_{water} = c_{air}\left(\frac{V_{system} - V_{sample}}{V_{sample}} + k\right) - c_0\left(\frac{V_{system} - V_{sample}}{V_{sample}}\right)$$

c_{water}	=	Radon concentration in water sample *[Bq/l]* or *[pCi/L]*
c_{air}	=	Radon concentration *[Bq/m³]* or *[pCi/L]* in the measuring set-up after expelling the Radon (indicated by AlphaGUARD)
c_0	=	Radon concentration in the measuring set-up before sampling (background) *[Bq/m³]* or *[pCi/L]*
V_{system}	=	Interior volume of the measurement set-up *[ml]*
V_{sample}	=	Volume of the water sample *[ml]*
k	=	Radon distribution coefficient water/air *[ ]*

The exactness and correctness of the AlphaGUARD indicated values and thus also for the determined Radon concentration of the water sample depends on the following boundary conditions:

- Relation of volume sample/measurement set-up
- Tightness of measurement set-up
- Temperature of sample
- Radon level in the set-up before sampling
- Background (intrinsic effect) of AlphaGUARD

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/ INTERIOR VOLUME OF THE SET-UP

The determination of the relation of sample and measurement set-up requires the accurate knowledge of the interior volume of the complete set-up. The interior volume of the set-up results from the sum of the interior volumes of all components integrated into the set-up.

The following table shows the AquaKIT components with their particular volumes. The volumes are distinguished between the measurement set-up for performing of so-called quick measurements and precision measurements:

Quick measurement (system components)	Volumes [ml]	Precision measurement (system components)	Volumes [ml]
Ionization chamber, inbuilt pump, muffler and tubing of *AlphaGUARD DF2000* :	750	→	750
100 ml degassing vessel	190	500 ml degassing vessel	618
Security vessel	191	→	191
Tube connection (1,5 m):	19	→	19
Total volume:	**1150**	→	**1578**

Figure (7): Volumes of AquaKIT system components and calculated total volumes of measurement set-up when combined with AlphaGUARD DF2000

/ TEMPERATURE DEPENDENCY OF THE DIFFUSION COEFFICIENT '*k*'

The diffusion coefficient '*k*' indicates which relation of concentration there is for Radon between a watery phase and the air volume existing above. This relation dependents on the temperature. With falling temperatures, the quantity of Radon soluble in water increases. E.g. the diffusion coefficient '*k*' grows when the temperature drops. Neglect the temperature dependency of the diffusion coefficient as recommended for quick measurements would lead to an unacceptable elevation of the measurement uncertainty. In Figure (8) the dependency for temperature of the diffusion coefficient for the transition of the phase water/air is presented:

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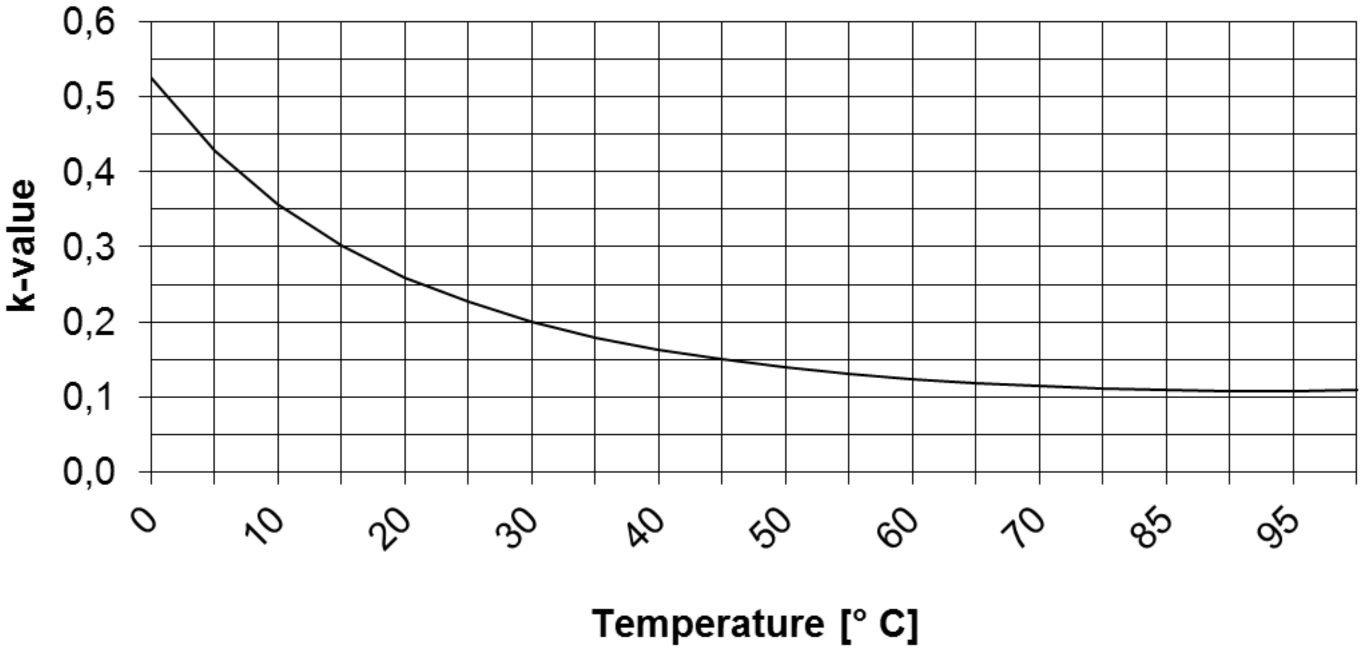

Figure (8): Temperature dependency of the diffusion coefficient 'k'

The influence of the Radon diffusion coefficient 'k' is only low in the temperature range between 10° C and 30° C. With quick measurements this is based on the given relation of volume between the air enclosed in the set-up and the watery measuring sample of approximately 10 : 1. Because of this it can be said that with quick measurements within the specified temperature range around a mean room temperature of 20° C a diffusion coefficient k of 0.26 can be applied.

This is suitable as long as the samples are measured under normal conditions. This stipulation of the diffusion coefficient leads - related to the final result - to an uncertainty of ± 2 % and is therefore negligible for the error resulting of sampling.

With precision measurements, the exact value of the diffusion coefficient should always be calculated. When the relation of the volume between air enclosed in the set-up and the watery sample is only about 2 : 1, the negligence of the temperature dependency of the diffusion coefficient as recommended for quick measurements would lead to an unacceptable elevation of the measurement uncertainty. Temperature deviations of ± 10° C would lead to a measuring error of up to 7 %.

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/ EXAMPLE OF MEASUREMENTS WITH AQUAKIT

Results of radium measurements in bottled waters in Marmara Region of Turkey

Sample code	Origin of the water	^{226}Ra concentration ± standard error [Bq·L^{-1}]
S1	Hendek	0.03 ± 0.01
S2	Hendek	0.09 ± 0.02
S3	Hendek	0.07 ± 0.02
S4	Hendek	0.43 ± 0.09
S5	Uludag	0.23 ± 0.06
S6	Uludag	0.20 ± 0.03
S7	Uludag	0.03 ± 0.01
S8	Uludag	0.08 ± 0.02
S9	Uludag	0.10 ± 0.03
S10	Uludag	0.15 ± 0.03
S11	Uludag	0.11 ± 0.03
S12	Uludag	0.16 ± 0.03

Figure (9): Results of radium measurements in bottled waters in Marmara Region (Turkey)

Sources: U. Akar, O. Gurler & others "Measurements of Radium Levels in bottled natural spring water of Marmara region (Turkey)" Physics Department, Faculty of Arts and Sciences, Uludag University (2009).

Country	Radon Activity (Bq l^{-1})	References
Jordan	2.5-4.7	Al-Bataina *et al.* (16)
Venezuela	0-2	Horvath *et al.* (17)
Brazil	0.39-0.47	Marques *et al.* (18)
Kenya	0.80–4.70	Otwoma and Mustapha (19)
Algeria	0.26–2.28	Amrani and Cherouati (20)
Cyprus	0.1–2.00	Sarrou and Pashalidis(21)
Egypt	0.007-2.33	Abbady *et al.* (22)
India	0.084-0.83	Kant *et al.* (23)

Figure (10): Rn levels in tap water: Int. J. Radiat. Res., Vol. 14 No. 3, July 2016

The concentration of radium has been assessed in the bottled natural spring water samples commercially available in Turkey. Measurements of Ra-226 concentrations in bottled natural spring water samples were carried out by AlphaGUARD and measuring the Radon in equilibrium with radium. The radium concentration in these samples is found to be between 0.03 and 0.43 Bq/l (or 0.81 and 11.6 pCi/l).

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/ AQUAKIT CHARACTERISTICS

Typ. operating range:	from 5 to 20,000 Bq/l or from 135 to 540,540 pCi/l (direct measurement)
Time of execution:	< 15 min / sample (sampling, degassing, measuring, analyzing)
Statistical error:	≤ 10 % at 5 Bq/l or 135 pCi/l (1-sigma error)

Larger sampling volumes (up to 500 ml) and longer measuring times (≥ 45 min) decrease the statistical error band and enlarge the lower limit of the measuring range up to 0.1 Bq/l (or 2.7 pCi/l).

/ CONTINUOUS MEASUREMENT OF RADON IN WATER

Two other devices have been developed for on-line monitoring of Radon in Water, mainly for research applications. The REM 1 module is used for short term monitoring, the air exchanger is embedded in a suitcase and can operate directly by connecting ducts, suitcase closed.

The REM 2 system is dedicated for long term monitoring. The exchanger module can be placed directly in the water to be monitored or placed inside a container.

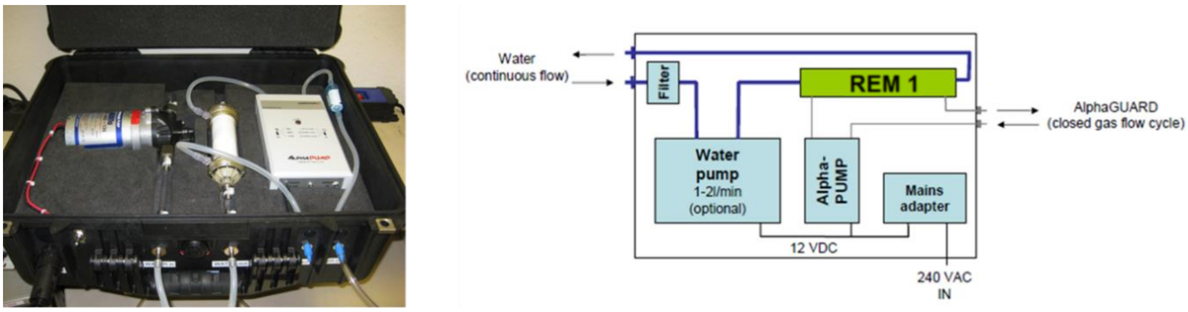

Figure (11): Exchanger module REM 1

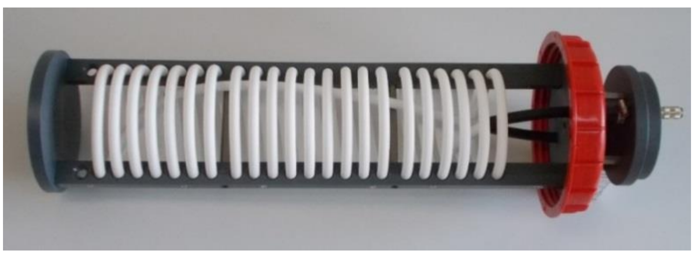

Figure (12): Exchanger module REM 2

Figure (13): Complete REM 2 system

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/ RADON IN WATER USED AS A TRACER: EARTHQUAKE STATION

Sudden changes (either increase or decrease, also called Radon anomalies) may be observed in the time series of Radon concentration. These have been found to be related to an increase in seismic (King, 1986; Zmazek et al., 2002) or volcanic (Cigolini et al., 2007; Gasparini and Mantovani, 1978) activity of a region, or activity change of a tectonic or geologic fault (Sebela et al., 2010).

On the other hand, Radon anomalies in the spatial distribution of Radon levels in a region have been observed to coincide with the locations of tectonic and geologic faults, either well expressed on the surface or still hidden (Burton et al., 2004; Swakon et al., 2005).

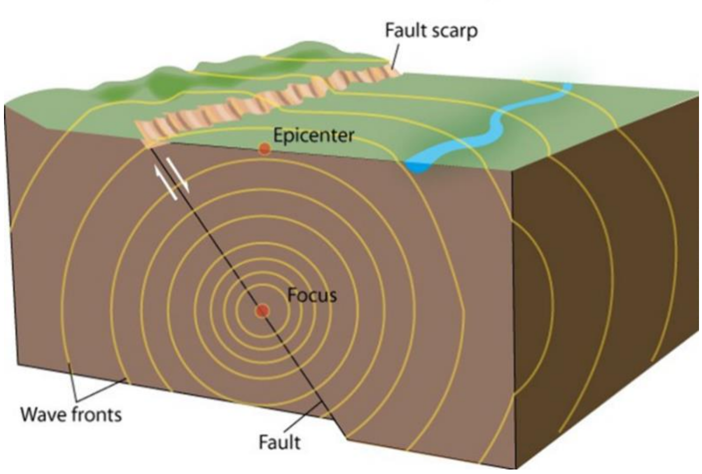

Figure (14): Seismic waves diagram (evergreenfacts.com)

/ RADON AS A NATURAL RADIOACTIVE TRACER TO STUDY AQUIFER SYSTEMS

The behavior of the four Ra isotopes (measured by gamma spectrometry) and Rn-222 (measured with an AlphaGUARD ionization chamber) has been investigated in NaCl-rich thermal waters of Balaruc-les-Bains on the Mediterranean coast (South of France). This study allows identification of the deep thermal water signature and reveals the influence of seawater or karst water inflows on Ra isotopes and Ra-222.

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/ CONCLUSION

Although there are some exceptions, in general, the migration of Radon up from the soil contributes the largest percent of Radon found in the average home. Radon from a groundwater type water supply source, particularly a bedrock well (also known as artesian or drilled well), contributes the next largest percentage of Radon in the home.

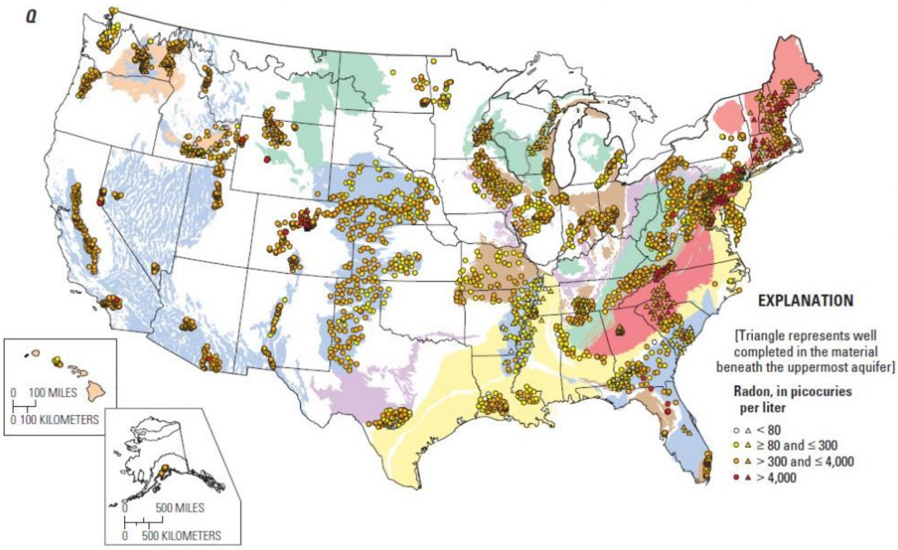

Figure (15): Radon in Groundwater across the United States

Testing home for Radon is easy. If Radon has been found, it may be of interests to check Radon in water, especially in Karst area or if the home is getting water from a well…

Furthermore, measuring Radon in water (tap water, from a well…) is recommended and even compulsory in many states, countries and levels have been defined.

Current standards for Water Radon contamination	Value
US EPA Maximum Contaminant Level (MCL)	11.1 Bq/l or 300 pCi/l
Recommandations of European Union	100 Bq/l or 2,700 pCi/l

Finally, the method described with AlphaGUARD and AquaKIT has been used for years and can be performed in a lab or on site with the reliability, reproducibility and quality measurement of AlphaGUARD.

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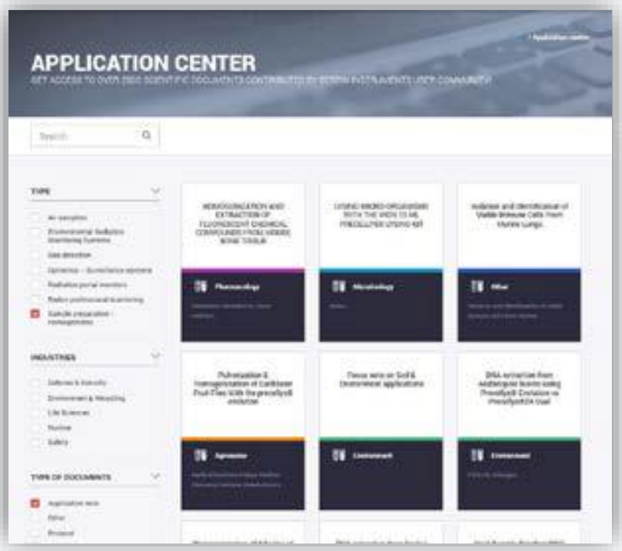

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