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Setting up a continuous monitor for the control of temporal variability of ^{222}Rn in groundwater: Application to samples from Tadla Basin, Morocco

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Abstract

Owing to its short period (t1/2= 3.8 d), the development of a technique allowing the analysis of the radon with very low content was a challenge. In this paper we shall describe a robust method and portable technique (RAD7) for measuring moderate levels of 222Rn in various matrices: soil, air, coastal water, river water and groundwater. By purging air through the sample, radon is emanated until a chemical equilibration is obtained between the two phases. The radon in the air loop is determined using the radon-in-air monitor. Since the distribution of radon at equilibrium between the air and water phases is governed by a well-known temperature and humidity dependence, to check that, several tests were performed in the laboratory. This technique is used for the first time in Morocco for monitoring radon in the atmosphere and spatial distribution in natural waters, to establish a database on radon activity and for a radiological surveillance either for hydrological applications. The obtained results showed that the sensitivity varies inversely proportional to the moisture in the measuring chamber. Also the water temperature dependence of the concentration of radon has been clearly shown. The atmospheric radon activity inside the laboratory was 0.326 ± 0.058 pCi/L.

1. Introduction

Considering its physico-chemical and radiological properties, the Radon-222, a noble gas with half-life of 3.82 days, is often requested to answer to several hydrological and environmental issues. These include tracing groundwater input to streams [1, 2, 3], lakes [4, 5], and coastal zones [6, 7, 4, 8] as well as rates of river water infiltration to banks [9, 10] and sediment-water exchange [11].

For measurements in the field, the methods used were based on the complementary use of proportional counter and nuclear track detectors. Also the measure of the radon in the water was based on the scintillation, such as Lucas cell scintillation counting [12, 13], liquid scintillation [14, 15, 16, 17, 18], and γ counting, as well as a number of automated methods [19]. Although these classical techniques have proven their high precision and their quality of analysis, it present some distrusts, it require cumbersome handwork and a relatively long time for extraction, transfer, and radioactive equilibration. Moreover, the complex instrumental setup is inconvenient in-situ on field sites.

Recently, several works were made in this sense. Burnett et al. [20] underlined a simple method for the continuous monitoring of ^{222}Rn, by working on two approaches: an "active" pumping system that will continuously strip radon out of a stream of water with subsequent detection of the gas phase by an atmospheric radon monitor, and a "passive" system that is based on diffusion of radon through a membrane into a submerged chamber with an on-board radon detection system. The results showed the importance of the active approach using the RAD7.

The RAD7 is a sophisticated measuring instrument widely used in laboratories, in the field and research work in several disciplines. This system determines ^{222}Rn in continuously circulating air (in a closed air loop), which is in equilibrium with a constant stream of water passing through an air-water exchanger. The concentration of ^{222}Rn in a water sample is calculated using the well-known temperature-dependent distribution factor of ^{222}Rn between air and water. Dulaiova et al. [21] improved the counting efficiency of this method by connecting several radon-in-air monitors in a parallel setup.

In this study, we inspected the parameters influencing the Radon counting by performing several laboratory tests. The experiments are attempted in the laboratory of the National Center for Nuclear Energy, Science and Nuclear Techniques (CNESTEN). Then we applied this air-water radon equilibrium technique to analyze radon in water samples. This will enable a rapid in situ assessment of moderate levels of ^{222}Rn activity in various types of samples (air, surface and ground water) in Tadla basin. We chose the Tadla basin for our survey since it is a compact and easily accessible region with a variety of geological formations [22]. Furthermore, there does not appear to have been a significant study of the natural radioactivity of water in this area. Some of this water is used to supply both the local population and some major conurbations.

2. Material and Methods

The multiple applications of radon have been hampered by the time consuming logistical requirements of collecting and analyzing samples in a conventional manner. Actually, the specialist look for more automated systems may be applied in order to increase the sampling resolution and efficiency of the process [23].The technique described in this paper is modified from the continuous radon monitor developed by Burnett et al. (2001). We are using a RAD7 (Durridge Co., serial number 3203) as a radon-in-air monitor that determines the concentration of ^{222}Rn by collection and measurement of the α-emitting daughters, ^{214}Po and ^{218}Po.
Endowed with several protocols, the RAD7 allows the measurement of various matrices:

i) The Sniff mode allows a scanning of the radon existing in the atmosphere, and that in 10 min instead of waiting 1 hour in the normal mode.

ii) The Grab mode is useful when it is not possible to take the RAD7 to the field, or when the system is pre-occupied with continuous monitoring and will not be available until later. The Grab functionality is also useful when many samples must be gathered from different places within a short timeframe.

iii) The Wat250/40 mode providing an effective counting for water samples. A glass bottle is connected to a radon-in-air monitor (Fig.1) in a closed air-loop mode, the degassing of the water sample is provided by a Glass Frit, before the air inlet of the counter, it passes through a desiccant column in order to prevent the entry of moisture to the counting chamber. The ^{222}Rn from the water sample continuously circulates through the desiccant column, RAD7, and then back to the water sample via a Check valve, so equilibrium between the water and the air is reached. Then, the activity of the ^{222}Rn is determined by counting its alpha-emitting daughters in the monitor.

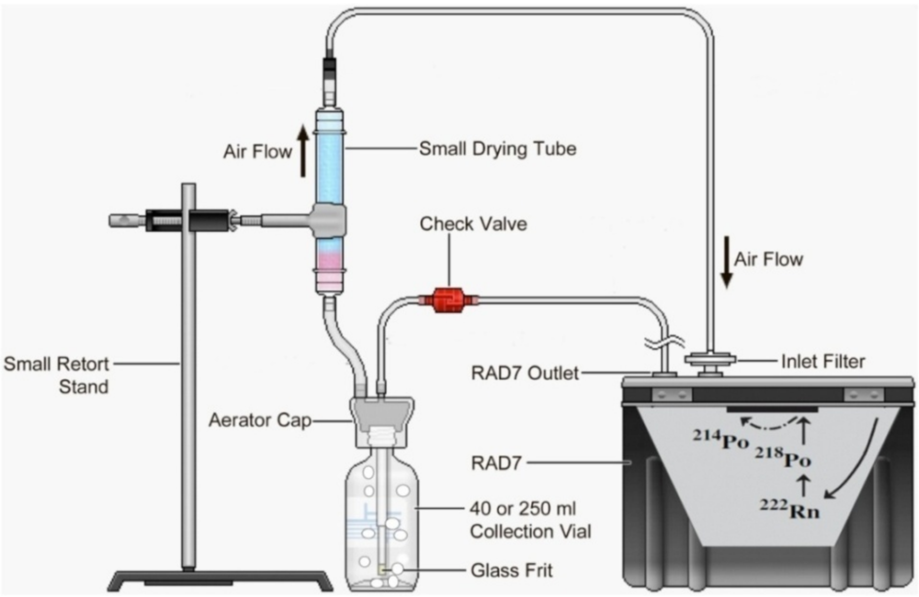


Figure 1: RAD H_2O Schematic: the components, as shown, automatically perform everything required to determine the radon concentration in the water. (Durridge Co., serial number 3203)

The RAD7 is equipped by an internal air pump, which re-circulates the air at a flow rate of about 1 L/min, purging radon in water to achieve a rapid equilibrium of radon between water and air. After the air-water equilibration is obtained, the radon in the air loop is measured by the RAD7. In the RAD7, a high electric field (2.0-2.5 keV) in the detection chamber propels the positively charged polonium daughters, ^{218}Po ($t_{1/2}$=3.05 min; Eα=6.00 MeV) and ^{214}Po ($t_{1/2}$=164 ms; 7.67 MeV), onto an alpha semiconductor detector at great potential. This

allows one to select either one or both isotopes for measuring ^{222}Rn. Then, the detector counts ^{218}Po and ^{214}Po, with energy discrimination of both isotopes.
The activity of ^{222}Rn in water is governed by the distribution factor of radon concentration between water and air. The actual activity of ^{222}Rn in a water sample is the sum of ^{222}Rn activities in the air loop and water in the sample bottle, where ^{222}Rn is partitioned by Weigel's [24] equation:

$$C_{water} = C_{air}\left(\frac{V_{air}}{V_{water}} + K\right) \quad (1)$$

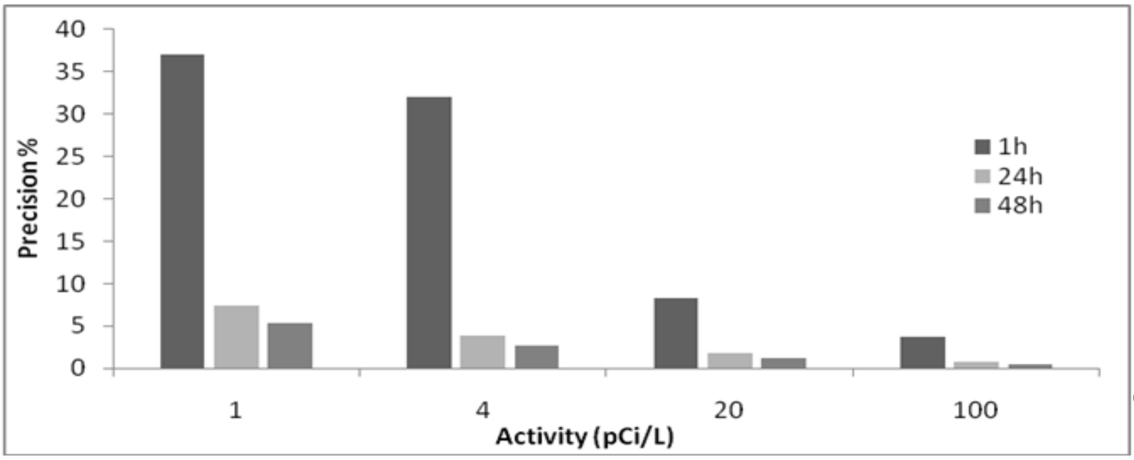
Where C_{air} (Bq/L) is the activity of ^{222}Rn in the air loop (after air water equilibrium), V_{water} and V_{air} are the water volume and the air volume in the loop (sample bottle, detection chamber, desiccant, and tubes), respectively.
k: is the radon concentration ratio of water to air, and T (°C) is the temperature of the water.

$$K = 0.105 + 0.405e^{-0.0502T} \quad (2)$$

3. Results and discussion

3.1. Precision & Accuracy

The precision of a measurement system, related to reproducibility and repeatability, is the degree to which repeated measurements under unchanged conditions show the same results. An accurate instrument is necessarily precise, but a precise instrument can be inaccurate. The precision can be impacted by the environmental factors, but in the normal ranges of operation of functioning, those are revealed to be much less important. As long as the operator follows consistent procedures, counting statistics will dominate the RAD7's precision.
DURRIDGE calibrates all instruments to a set of four "master" instruments with a calibration precision of about 2%. The master instruments have been calibrated by way of inter-comparison with secondary standard radon chambers designed by the U.S. EPA. They estimate the accuracy of the master instrument to be within 4%, based on inter-comparison results. DURRIDGE estimate the overall calibration accuracy of the RAD7 to be better than 5%.
The figure 2 summarizes the precision of the RAD7 relating to the contribution of counting statistics. Operating conditions and other factors may affect precision by as much as 2%. The uncertainty values reported by the RAD7 are estimates of precision based on counting statistics alone.



(1)

Figure 2: Typical RAD7 precision based on counting statistics only (DURRIDGE) (1)

3.2. Radon-222 water activity vs sample temperature

RAD7 system is equipped with a water-air exchanger, which is simply a clear plastic tube, has water flowing through it continuously with a provision for a stream of air which is pumped by an internal pump of the RAD7. An aerator stone provides a stream of fine bubbles to help equilibrate the radon-in-water with the radon-in-air. After some time, the radon concentration in the air reaches equilibrium with the radon in the water, the ratio at equilibrium being determined by the water temperature.
We have conducted some tests to verify the dependence of the activity of radon in water to temperature. The experiment is carried out on samples assumed same activity but with a different water temperature. We have also attempted to shake the temperature of the water sample before starting each experience.

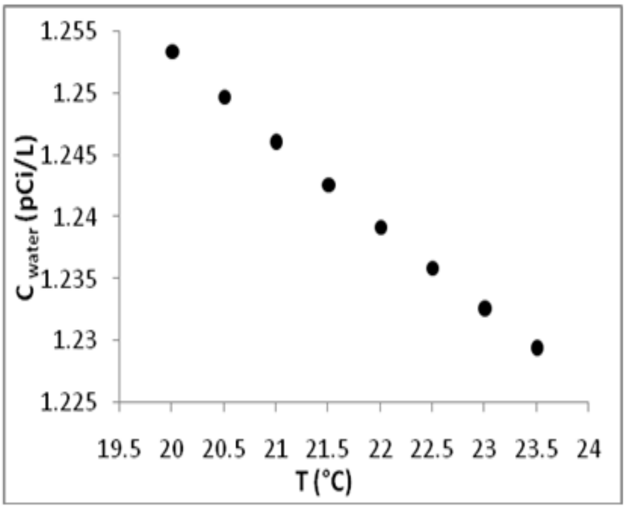


Figure 3:th eoretical correlation between temperature and radon water activity; obtained using the equation (1) and (2), C_{air}, V_{air} and V_{water} are taken constant, and we vary the sample temperature.

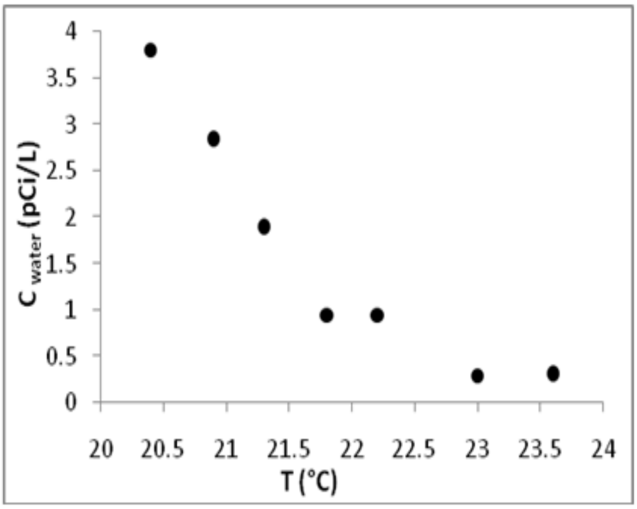


Figure 4:experimental correlation between temperature and radon water activity; obtained by analyzing samples with different temperatures, in the laboratory of CNESTEN.

We notice a perfect resemblance between the experimental results and theoretical ones established following equation Weigel. The figures show clearly that the water activity varies inversely proportional to the sample temperature.

3.3. Radon water activity vs humidity

The high sensitivity of the RAD7 is a result of the large collecting volume of the measurement chamber. In coastal areas or in a rainy day, radon measurement seems difficult because of the High rate of humidity. The affect is a function of the absolute humidity (Fig. 5).

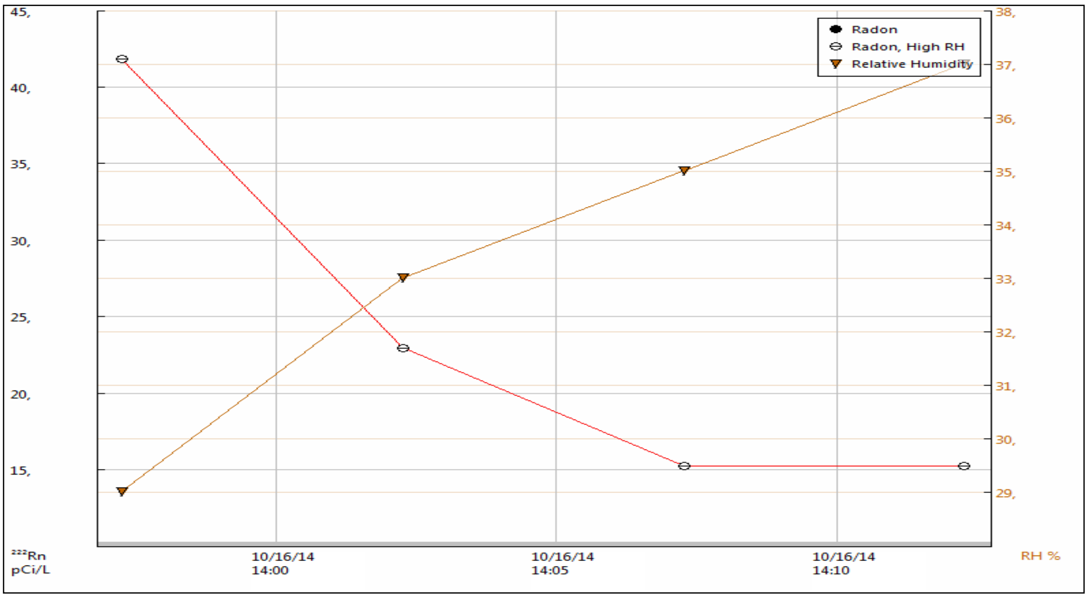


Figure 5: the influence of humidity in the counting chamber on the radon water activity, the figure shows that a high humidity reduces the sensitivity of the RAD7.

Some ions in the presence of water vapor will attract water molecules, as they are polar, until a group of a few water molecules gathers around each of them. These cluster molecules move more slowly in the electrostatic field and thus there is more time for the ^{218}Po atoms to become neutralized on the way to the detector surface, and therefore lost. So with high humidity the sensitivity of the instrument drops. Excessive humidity inside the chamber makes it more difficult to maintain the high insulation resistance necessary. In this state of mind, we performed an experiment with a low efficiency desiccant; the results obtained are mentioned in the figure below.

At normal room temperature and with good desiccant in the air sample path, the humidity in the measurement chamber at the start of a measurement must be below 10% RH and will eventually settle below 6%. In these conditions the collection has maximum efficiency and there is no humidity correction required. If the desiccant expires and/or if the operating temperature rises well above normal room temperature, the absolute humidity may become significant and a humidity correction may be required to compensate for the drop in sensitivity.

3.4. Radon concentration in air

The system existing in our laboratory allows a measurement of radon in the air, which will allow an ongoing assessment of radon levels. We conducted a test, which allowed us to obtain an average concentration of radon in the air in the order of 0.326 ± 0.058 pCi/L. During this test the relative humidity was 3.98% (<10%). For the first three readings that are in the "sniff" mode, the RAD7 count only the ^{218}Po for window A, and after 3 hours the mode automatically switches to "normal" mode.

In this experience, the system measures a sensitivity of 0.227 and 0.47 (cpm/pCi/L) in Sniff and Normal mode respectively, which is better than those indicated in the data sheet of the equipment.

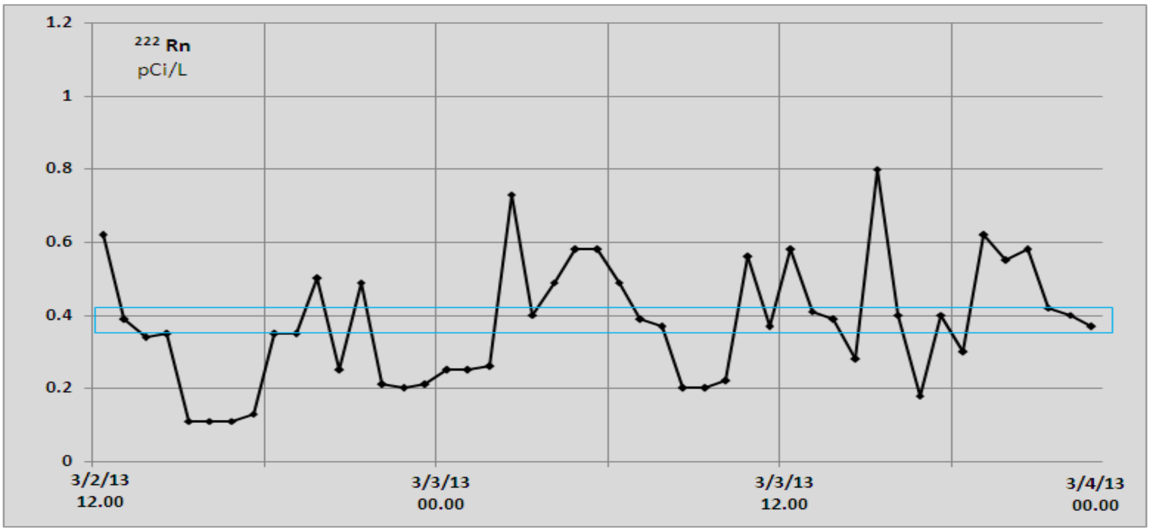


Figure 6: radon spectrum measured in the air by RAD7

3.5. Applications to natural waters

✓ Description of the study area

At an altitude of 400 m, the Tadla basin, with an area of over 10.000 km^2, is located in the center of Morocco (Fig.7). The Tadla region located on the northern edge of the Atlas Mountains of Beni Mellal, is a complex zone, where two important domains meet (the Atlas Mountains and the Tadla plain) through piedmont formations and the north Atlas faults. It consists of three geographical areas, the Tadla plain in the south and the Plateau of Phosphates in the north and Tassaout Aval in the west.

The Karst of the Tadla region is well developed in limestones and dolomites of the Lower and Middle Lias, limestone of Middle Jurassic and Turonian (synclinal Ait Attab, Ouaouizeght). These karstic systems and the associated aquifers contribute in a remarkable way to the sustainability and to the support of the low-water marks of Atlasic Rivers submitted to the Mediterranean climatic constraints. In the Tadla plain, the groundwater resources, constituted by a multilayer aquifer system with groundwater, Eocene and Turonian are estimated at 440 Million m^3. They are characterized by a good water quality.

✓ Natural water analysis

After installation and equipment efficiency testing, we applied the method to analyze radon activity in natural water samples collected from the Tadla basin of Morocco. First, we filled 250 ml sample bottle carefully with water, then we connect all the tubes between sample bottle cap, desiccant, and RAD7. After some time, the sample water volume was adjusted, the bottle was capped immediately, and the ^{222}Rn activity was measured for 30 min. Then, the count rates were corrected for the detection efficiency of our RAD7, and the activity of ^{222}Rn in water was calculated from Eqs. (1) and (2). Here, we used ^{218}Po counts alone after ^{222}Rn reached chemical equilibration with ^{218}Po and physical equilibration between water and the air, which took about 30 min. The measured specific activities of ^{222}Rn around Tadla basin ranged from 500 to 10500 Bq/m^3 (Table 1). The counting of one sample agreed within 10% uncertainty with the result based on the continuous radon measurement system.

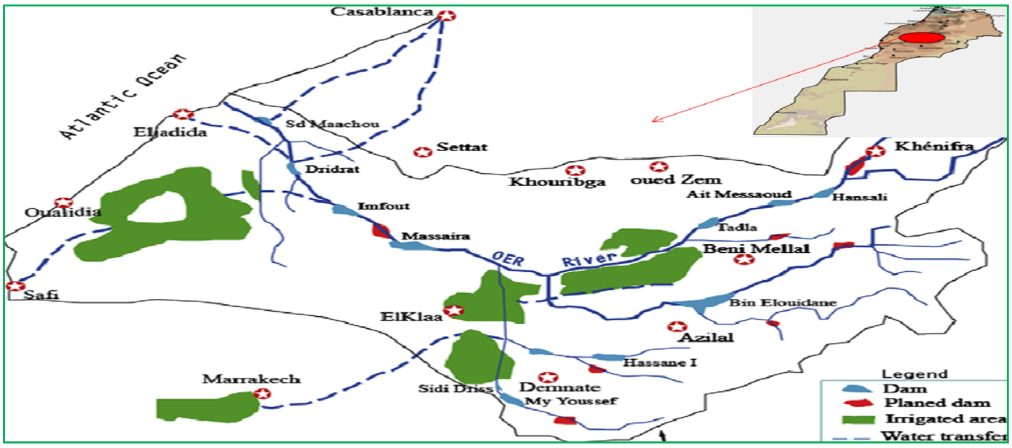


Figure 7: General and detailed maps of the Oum Er-RiBia (OER) river and Tadla basins in the central part of Morocco.

Table 1: Activity of ^{222}Rn in surface and groundwater samples from Tadla Basin

Sample Id	Nature	Depth (m)	Sample temp °C	Radon (pCi/L)
T001	groundwater	604	40.3	213 ± 28
T002	groundwater	480	35.8	190 ± 27
T006	groundwater	180	26.6	15.2 ± 8
T010	groundwater	330	30	86.7 ± 18
T014	groundwater	236	23.5	283 ± 33
T020	groundwater	85	24.2	22.9 ± 9
T021	groundwater	135	37.9	103 ± 20
T023	groundwater	245	23.5	120 ± 18
T024	groundwater	236	24.2	233 ± 30
ES1	river	-	19	87.8 ± 18
T007	spring	-	23.1	107 ± 20
T013	spring	-	16.4	19.1 ± 9

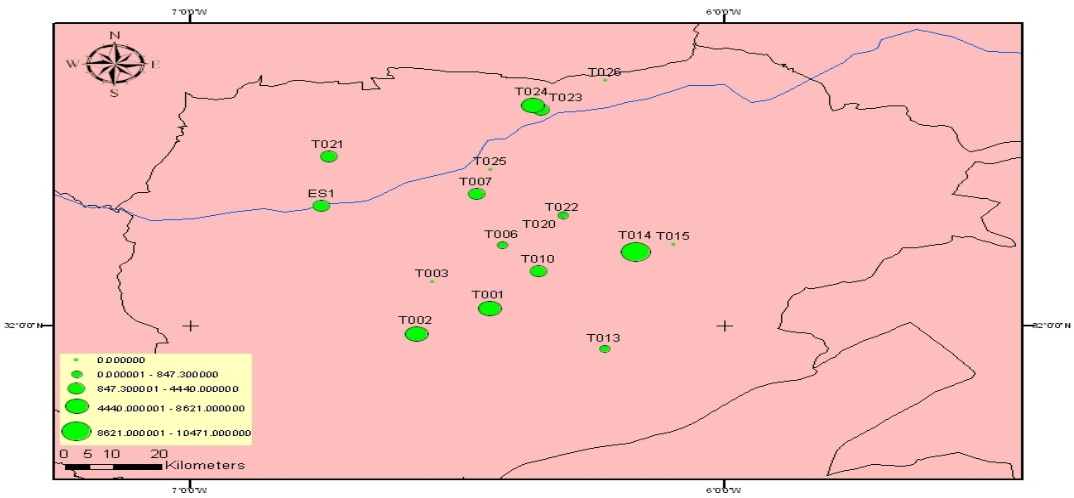


Figure 8: Spatial variation of ^{222}Rn in the Tadla basin

It is well known that radon activity is high in ground waters [25, 26], while for surface water the radon content is variable and low. Expected ranges of radon concentrations are 0-185 mBq/L in lakes and 185-3703 mBq/L in streams [27].

Springs: Ain Assardoun spring (T007) present a high radon concentration, we hypothesize that this water is composed of water flowing from the Atlas Mountains characterized by karstic limestone with higher permeability. Contrariwise the Ain Kaicher spring (T013) contains a low radon concentration, that there explained by the made that we took a sample of the holding tank and not directly of the inaccessible spring, so an equilibrium between water spring and air was enriched.

ES1: Oum Er-Ribia River, relatively high activity which supposed affected by several factors, viz. the geology of the area, bottom sediments, inputs from streams, degree of water turbulence and temperature. Also it can indicate a discharge area of groundwater.

For the other samples: correlation between depth and radon concentration it is not well established, because the radon activity depends on the nature of the bedrock or sediment, the presence of faulting, the degree of turbulence, and the supply of fresh water from tributaries and ground waters.

Conclusion

In this work, we quantified the dependence between the sensitivity and water temperature, this will allow us to make corrections for abnormal cases. Also the automatically correction of the results depending on the value of moisture can be manually checked.

The results founded are very encouraging in terms of having a system that can determine moderately high levels of ^{222}Rn in natural water samples. Since the measurement system is portable and simple, the method is particularly attractive for on-site measurements. Overall the master instruments have been calibrated by way of inter-comparison with secondary standard radon chambers counting. Tests conducted in the laboratory have shown good correlation between the water temperature and the radon concentration. We note that the low humidity in the measuring chamber increases the sensitivity. As well the system allows monitoring of radon in the atmosphere, an activity is determined inside the laboratory of about 0.326 ± 0.058 pCi/L. A measurement of certain points in the Tadla region provides a vision of space distribution radon in natural waters. The concentrations found in this region vary between 0.56 and 10.47 Bq/l, this value remains below that tolerated by The European Commission 100 Bq/l (IRSN 2012). However these results can be fully exploited in other applications such as hydrology.

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