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Pressure response of radon detecting devices placed at depth in aquifers

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RESUMEN

En muchos casos se necesita la medición del Rn en el agua debajo de diferentes profundidades en tiempo real, lo cual se hace con equipos electrónicos. El efecto de la presión puede tener un gran papel sobre el sensor electrónico y también sobre la geometría del dispositivo. Ese estudio proporciona informaciones sobre la sensibilidad de sistemas de este tipo en función de la profundidad/presión, y suministra una curva de corrección. También el estudio demuestra que la solubilidad del Rn en el agua puede ser investigada en función de la presión.

PALABRAS CLAVE: Radón, presión, agua subterránea, sensibilidad, profundidad, medición.

ABSTRACT

Real-time *in situ* Rn measurements under water at different depths are often required. Pressure effects on electronic equipment can play a role on the sensor and on the geometry of the device. We discuss the sensitivity of such systems depending on depth/pressure variation and provide a correction factor curve. We show that Rn solubility in water can be investigated as a function of pressure.

KEY WODS: Radon, pressure, groundwater, sensitivity, depth, measurement.

INTRODUCTION

The study of radon concentration variations in the water of aquifers provides an additional contribution to the instead of the physical characteristics of an aquifer (fracturation, permeability, porosity), on the water flow processes and possibly on the origin of the groundwaters. In spite of its relatively short half-life (3.82 d) radon, the time variation of its water concentration may allow to assess the transit time of the percolating waters originating from the rainfalls or from the water flows near by the considered aquifer (Hoehn and Von Gunten, 1989). Several groups have already started the investigation of aquifer functioning by this means (e.g. Monnin *et al*., 1994; Surbeck and Eisenlohr, 1994; Eisenlohr and Surbeck, 1995; Reddy and Sukhija, 1996; Cook *et al*., 1999 ; Hamada, 2000; Monnin and Seidel, 2001). In a different field, underwater radon measurements have been performed in order to assess underwater fresh water discharge on the continental shelf (e.g. Hussain *et al*., 1998; Cable *et al*., 1996). In such investigations, continuous recording of the radon concentration *in situ* is more useful than discrete sampling measurements. Such continuous recording can be achieved with electronic detecting devices where an inverted cup is immersed into the water. The radon concentration in the aerial phase above the water reflects the radon concentration in the water itself. However, it can be thought that this response can be strongly pressure dependent. Therefore, depending on the depth at which measurements are performed, the relationship between actual radon concentration and readings must be established, which is the purpose of this work.

EXPERIMENTAL TECHNIQUE

The aim of the experiment was to assess the peculiar response to a pressurised environment of the Clipperton probe. The Clipperton radon probe was devised and built in our laboratory (Monnin and Seidel, 1998). It is a field instrument designed for continuous long-term radon measurement. It is based on the detection of alpha-particles using a 1 cm^2 area solid-state electronic sensor. The probe is designed for the selective counting of radon decays with recorded count values over specified time intervals. The countings recorded during a space of time which can be selected at will among 1, 10, 20, 60, 120, 1440 and 2880 mn, are stored on RAM within the device. The RAM can store up to 3250 measurements and their identification labels. Both the alpha-particle sensor and the RAM are installed in a cylindrical stainless steel tube with an overall length of 50 cm, opened at one end in order to allow Rn to enter the probe, and a diameter of 5

cm. With this arrangement the probe responds within 12 minutes to any change of the Rn concentration occurring at the level of its opened end. The Clipperton probe allows the measurement of radon activity levels in a range between 100 $Bq.m^{-3}$ and 1000 $kBq.m^{-3}$, permitting an accuracy of about ±7.5%. Its sensitivity is: 1 $count.h^{-1}$ = 90 $Bq.m^{-3}$. The probe is operated on 4 R20 DC batteries. The data extraction is carried out on the field with a lap top computer. Data can be stored on disk as ASCII files and further treated by data base and spread sheets such as Excel, QuattroPro or Lotus 1-2-3.

In order to study the specific response of the probe, regardless of the pressure controlled dissolution of radon in water, a special vessel was built. It is shown in Figure 1, right side. It consists of a steel case (diameter: 50 mm, length: 60 mm). At the top of it is placed the active part of the probe, i.e. the 1 cm^2 solid state detector. The bottom part of the pressure vessel is closed by a mobile piston. When moving the piston towards the detector, one increases by compression the inner pressure inside the vessel, up to 15 bars. The pressure is measured by means of the pressure gauge placed on the side of the vessel. The vessel is connected to a high activity radon source (specific activity in the MegaBecquerel range). If and when needed, radon is made to enter the pressure vessel by means of a gas pump at a nominal rate of 1 $L.mn^{-1}$ in a closed circuit.

In order to study the solubility of radon in water as a function of pressure (depth) another set-up was devised and built. It can be seen on Figure 1, left side. It consists of a steel cylinder whose lid is tightly secured on its upper part. The lid bears a Rn detector at its bottom side. The vessel is filled with water up to 5 cm below the detector so as to represent a comparable volume to the one available when measuring in the field. Rn is made to circulate from the Rn source into the vessel and to bubble inside the water by means of a glass gas bubbling device located at the bottom part of the vessel. After a while (namely one to two hours) bubbling is stopped and the vessel is completely closed. It is left so as to wait for radon to part between the aerial and the liquid phase. Then the inner pressure is increased by means of an external air compressor, to a maximum air pressure of 7 bars.

RESULTS AND DISCUSSION

The first experiment consists in measuring the response of the detector as a function of pressure, compared to what it is under normal atmospheric pressure, and in a geometry which is not pressure dependent. In order to achieve this, a flat ^{238}U metal foil (thickness 0.5 mm) was placed in close contact with the detector. The maximum energy of the alpha particles from the ^{238}U decay is 4.268 MeV, that of the alpha particle from the ^{222}Rn decay is 4.78 MeV. The kinetic energy of these particles are quite similar and hence the use of U to simulate Rn is legitimate. Furthermore, the U foil being thick with respect to the range of the alpha particles in U, the set-up will provide the full spectrum of alpha particles energies, just as it is when measuring Rn activities. The activity was measured first with the piston in its farther position. Then the piston was moved so as to reach successively 5, 10 and 15 bars (representing theoretical depth in the water of 50, 100 and 150 metres) and the activity was measured each time. Moving then the piston backward, similar measurements were carried out. The resulting data are exhibited on Figure 2. An excellent reproducibility is found whether the pressure increases or decreases (the experimental error being less than

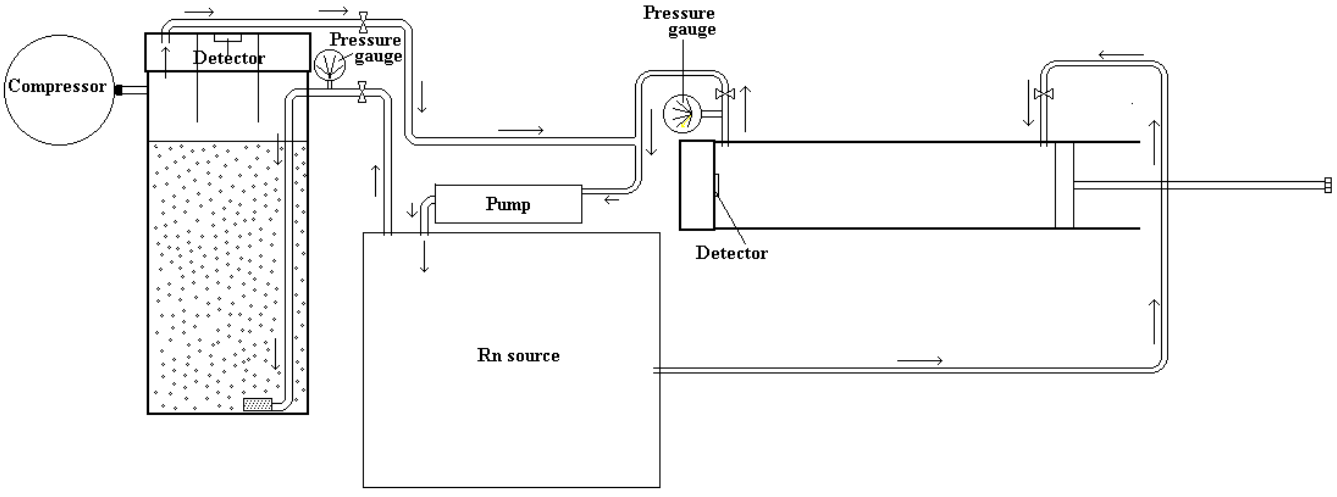

Fig. 1. Schematic diagram of the experimental set-up.

1%). With respect to the "normal" response, the sensitivity of the detector has decreased by a factor of 2 under 5 bars, by a factor of 6.7 under 10 bars and by a factor of 40 under 15 bars. The sensitivity of the detector Σ_p is exponentially decreasing with the pressure p according to the following formula: $\Sigma_p = \Sigma_1 \exp(-0.09\,[p - 1])$; where Σ_1 is the sensitivity under atmospheric pressure.

In order to measure the response of the set-up to radon under pressure, radon from the Rn source is admitted into the vessel. After the activity measured in the probe has reached a value comparable to that of the radon source S_1, the gas circulation is stopped. Then, by moving the piston, the pressure is increased successively from1 to 15 bars. The results are shown Table 1 (experimental error: %). Every time the pressure is increased, the signal S_p varies in intensity (starting from 70 c.p.m at atmospheric pressure for this particular case). It increases up to a pressure of 7 bars, then it decreases slightly. However, these "raw" data must be corrected by the equation established during the first experiment. The resulting $S_{p,cor}$ values are listed in the second column of the table. In the third column, one finds the ratio of $S_{p,cor}/S_1$. The last column shows the ratio C/C_1 of the Rn concentration under pressure to that under normal atmospheric pressure (which is of course identical to the pressure variation). On Figure 3 the corrected signal ratio $S_{p,cor}/S_1$ is plotted as a function of pressure and/or of relative Rn concentration. This curve is best fitted by the following equation $S_{p,cor}/S_1 = 0.44\,p + 0.74$. This variation in response results from increase of the detector's collection efficiency due to the fact that the range of the alpha particles decreases under pressure; thus the effective number of Rn atoms that are likely to emit alpha particles able to reach the detector also increase.

In order to measure the possible solubility variation of Rn in water under pressure, Rn was made to solubilize in the water contained in the second pressure vessel described above. The pressure was increased to 3.5 and to 7 bars. In the first case the counting was increased by a factor of 1,38 after correction, in the second case by a by a factor of 1.9 after correction (experimental error: 3% in both cases). The corresponding increase in the absence of water was 2.34 at 3.5 bars and 3.8 at 7 bars. The comparison of the two sets of values reflects the increased solubility of Rn with pressure

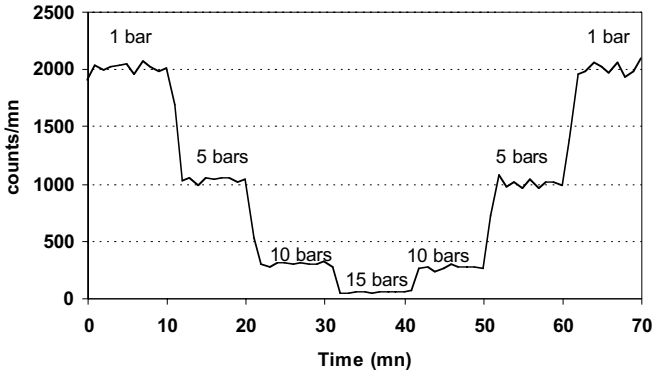

Fig. 2. Electronic sensor response to alpha particles as a function of pressure.

Table 1

Rn Signal S_p under, corrected signal $S_{p,cor.}$ and ratio $S_{p,cor.} / S_1$, as a function of the pressure p and/or of the concentration ratio C/C_1

P	S_p	$S_{p,cor.}$	$S_{p,cor.} / S_1$	C/C_1
1	70	70	1	1
2	108	118	1.69	2
3	125	150	2.14	3
4	135	178	2.54	4
5	135	193	2.76	5
6	165	257	3.67	6
7	155	266	3.80	7
10	150	340	4.85	10
15	145	517	7.38	15

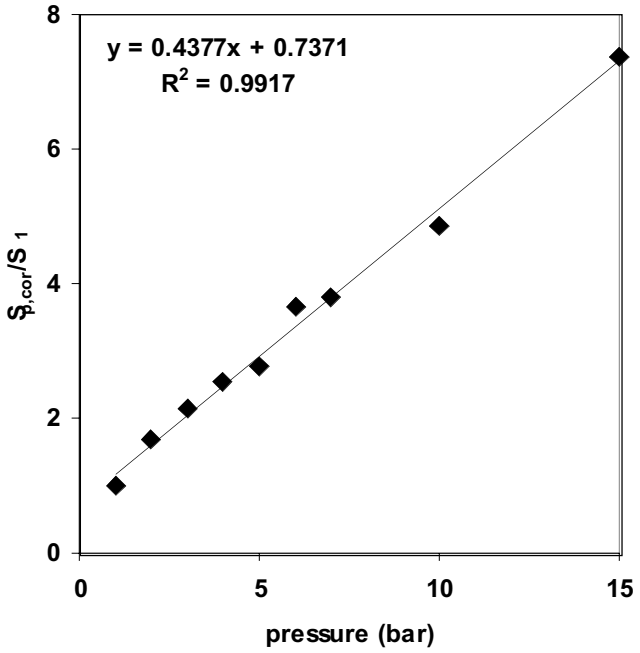

Fig. 3. Plot of the $S_{p,cor.} / S_1$ as a function of pressure.

since the Rn concentration in the aerial phase under 3.5 bar is 58% of what it was under atmospheric pressure and only of 50% under 7 bar. However, and generally speaking, these experiments demand to be improved, particularly by providing a good thermal insulation around the vessels so as to improve the statistics by lowering inner convection.

CONCLUSION

It has been shown that *in situ* deep measurements performed under water with silicon detectors such as those used in the Clipperton probes must be corrected. The specific sensitivity of the detector strongly decreases. However, in the mean time, the number of alpha particles which reach the detector for a given Rn concentration increases. The combination of the two sets of data provides the correction factors for deep water measurements. Similar experiments carried out in the presence of a substantial amount of water provides a way to experimentally determine the Rn solubility in water as a function of pressure (or depth).

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